An Investigation of Interference Lithography Applications using Evanescent Fields

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Abstract

Since the dawn of large scale integrated circuitry photolithography has been the primary means of pattern production. Over the following 60 years the size of these patterns has shrunk massively, along with the consequent increase in the complexity of the photolithography process. The demand for smaller, more powerful, and more energy efficient computational devices requires further shrinking of the patterns. The development of processes for further pattern size reduction however is not a simple one, thus a great deal of research and investments has been focused towards it. The research within this thesis is aimed at discovering new methods and techniques for photolithography pattern reduction by exploiting fields in an interference lithography setting.

Previously it was shown that dielectric resonant underlayers could be employed to enhance the depth of field for evanescent interference lithography. This however is limited by the availability of transparent high refractive index dielectric layers. To extend this to higher effective refractive indices an investigation into applying Herpin effective media within resonant underlayers was carried out. These underlayers were shown to be effective for combinations which have propagating fields within at least one layer; for combinations where all layers were evanescent however, the method broke down. Investigations into generic resonant underlayers also led to the development of a resonant overlayer method for increasing the evanescent field strength within a PR layer while allowing thicker and/or lower refractive index IMLs.

Further to this a new form of BARC for hyper-NA photolithography termed an evanescent-coupled ARC was developed. These ARCs rely on evanescently coupled dielectric or surface state polariton resonators to produce destructive interference within the PR. The properties and design constraints for each of these systems was explored and two experimental designs developed. Experiment verification of evanescent-coupled ARCs was successfully demonstrated for a SiO\textsubscript{2}|HfO\textsubscript{2} dielectric resonator based ARC. Demonstration of a MgF\textsubscript{2}|Cr surface state polariton resonator based ARC was partially demonstrated with resonance within the underlayer and the consequent alteration of the PR standing wave pattern observed.

The use of prism coupling for interference lithography is limited by the maximum refractive index of the coupling prism; above this refractive index all fields are evanescent and no energy will coupled into the PR. To overcome this limit grating coupled evanescent near-field interference lithography methods are employed. The higher order diffraction orders from the grating can have NAs far greater than the refractive index of naturally occurring materials, thus patterning with these diffraction orders produces far smaller interference pitches than prism coupled systems are capable of. Grating coupled systems involve the use of evanescent fields, plasmonic resonances, as well as coupled resonators all within subwavelength scales, consequently simulation and optimization of these systems is very computationally intensive. To improve this a genetic algorithm process was applied to reduce the computational time for optimization, and to allow the use of an inverse design process. Application of this method produced an order of magnitude improvement in optimization time compared to a full parameter sweep. Models including resonant overlayers, overlayers and underlayes, as
well as those employing extremely high NAs and/or higher $|m|$ diffraction orders were produced. Simulations showed that extremely high NAs up to 20 may theoretically be used for patterning of structures with a pitch of $\lambda/40$ equating to a full pitch of 10.1 nm with an exposing wavelength of 405 nm.
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Abbreviations

193i 193 nm Immersion Photolithography
AFM Atomic Force Microscope
ARC Antireflection Coating
ATR Attenuated Total Reflection
BARC Bottom Antireflection Coating
DOF Depth of Field
DR Dielectric Resonator
DSA Directed Self Assembly
EBL Direct Write Electron Beam Lithography
EIL Evanescent Interference Lithography
ENFOL Evanescent Near-Field Optical Lithography
EUV Extreme Ultraviolet Lithography
FEM Finite Element Method
FTIR Frustrate Total Internal Reflection
GA Genetic Algorithm
GLAD Glancing Angle Deposition
GRIN Gradient Refractive Index
IL Interference Lithography
IML Index Matching Liquid
ITRS International Technology Roadmap for Semiconductors
LELE Litho-Etch-Litho-Etch
MP Multiple Patterning (Lithography)
MSE Mean Square Error
NA Numerical Aperture
NIL Nanoimprint Lithography
PD Pseudo-Dose
PML Perfectly Matched Layer
PR Photoresist
SADP Self Aligned Double Patterning
SEM Scanning Electron Microscope
SEP Surface Exciton Polariton
SPP Surface Plasmon Polariton
SSR Surface State Polariton Resonator
TARC  Top Antireflection Coating
TE    Transverse Electric Polarisation
TIR   Total Internal Reflection
TM    Transverse Magnetic Polarisation
TMM   Transfer Matrix Method
Chapter 1

Introduction: Lithography

1.1 Background

The semiconductor industry is a major part of modern society; for the year of 2016 global semiconductor industry sales were a record high $338.9 billion USD [1]. The key drivers for this industry are consumer electronics, mobile devices, cloud computing, and the rise of internet of things, which is the presence and integration of computing elements into everyday objects and actions. The trend towards smaller more specialised devices looks likely to continue for the foreseeable future.

Since the construction of the first transistor and integrated circuit there has been a relentless push towards faster, denser, smaller, cheaper and recently more power efficient electronics. The ability to meet all these demands is embodied in the much referenced Moore’s Law which loosely states that the number of components per computer chip doubles every 18-24 months (Fig. 1.1) [2, 3]. Recently there has also been much attention focused on non-Moore’s Law type developments such as System on Chips (SoCs), analogue/RF, biochips, sensors and actuators which add value but do not scale in a Moore’s Law type fashion [2, 4]. This is referred to as ‘More than Moore’, a good example of which is the modern smart phone, which features traditional Moore’s Law components such as processors and RAM, but also non-Moore’s Law components such as SoC’s and sensors.

These developments however come at a cost, that is, increasingly large capital expenditure for each new manufacturing generation, as well as rapid obsolescence of newly acquired machinery and newly produced commodities. Thus a consortium of semiconductor industry experts were gathered to create the International Technology
Figure 1.1: Moore’s Law. (a) - The ‘classic’ Moore’s Law plot, showing the massive increase in transistor count with time [3]. (b) - An Intel 80386 die [6]. Year of production 1985, transistor count 275,000, die size 104mm\(^2\), manufacturing process 1.5\(\mu\)m. (c) - An Intel Core i7 6700K die [7]. Year of production 2015, transistor count 1,750,000,000, 122mm\(^2\) die size, 14nm manufacturing process. Note the massively greater transistor density in the modern Core i7 processor; the perfect example of Moore’s Law.
Roadmap for Semiconductors (ITRS) \[2,8,3\]. The role of the ITRS is to help ‘steer’ the industry so that the available resources can be well managed and utilised. So (ideally) when the limits of one fabrication generation are reached the next generation is ready for production. This is achieved by identifying the relevant research and development areas for the upcoming generation as well as for subsequent generations, out to a 15 year horizon. Traditionally this focussed on the continual shrinking of CMOS (Complementary Metal-Oxide-Semiconductor) circuits and the allied technologies. The ITRS has recently however been forced to change its outlook for two reasons as follows \[2,4\]. Firstly the recognition that Moore’s Law has slowed down (if not come to a halt), particularly where the cost of each generation concerned has altered the manufacturing landscape. This part of the industry is now controlled by only a handful of manufacturers who can afford the R&D costs for the next manufacturing generations. Consequently, they have their own in-house development roadmaps making this part of the ITRS roadmap redundant. Secondly, the major growth areas in the semiconductor industry are no longer mainframe or desktop computers, but in consumer electronics, mobile and wireless devices, and cloud computing. These new areas have a different set of development priorities such as connectivity, bandwidth, battery characteristics etc. With this in mind a new ‘ITRS 2.0’ has been produced to help facilitate development and integration in these areas. The 2015 ITRS 2.0 plan identified seven areas of focus \[8,9\]:

- **System Integration** - Examination of architectures, and the integration of heterogeneous blocks.

- **Heterogeneous Integration** - Optimisation of the integration of separately manufactured components into a single package.

- **Heterogeneous Components** - Research targets including different devices that form heterogeneous systems, such as MEMS, power generation, and sensing devices.

- **Outside System Connectivity** - Development of wireless technologies and their implementation.

- **More Moore** - Continued research into CMOS shrinking.

- **Beyond CMOS** - Alternatives to CMOS technology, such as spintronics, memristors, etc.
Factory Integration - Development of new tools and processes for heterogeneous integration.

At the heart of many of these areas, particularly ‘More Moore’ and ‘Beyond CMOS’, is photolithography. Photolithography is the production work-horse of the semiconductor industry, where a pattern is transferred into a photosensitive medium (Photoresist (PR)) via illumination of a mask containing the desired pattern (Fig. 1.2). Photolithography is used for the bulk of all the patterning required to produce a semiconductor device. As this thesis deals with several photolithography techniques and ideas it is worth first becoming familiar with the principles of photolithography.

1.2 Photolithography

As previously stated, photolithography is the process of producing patterns in a photosensitive medium via a light pattern. Photolithography has always been the most important aspect of Moore’s Law, with other contributions coming from processing factors such as improved light sources, and increasing wafer sizes which improve the manufacturing economics. As the push is towards producing the smallest features economically feasible it is natural to ask questions about the smallest theoretical feature sizes patternable, and what methods are the current industry standard? The pattern resolution in photolithography is often generalized as \[10\]
\[ Resolution = k_1 \frac{\lambda}{NA}, \text{ with } NA = n \sin \theta, \] (1.1)

where the resolution is the half width between two features, \( k_1 \) is a system dependent process factor, \( \lambda \) the exposure wavelength, and \( NA \) the numerical aperture (the lens refractive index \( n \) times the sine of half the maximum exit angle of the lens).

The system process factor, \( k_1 \), is a combination of factors such as: pattern type (i.e. regular parallel lines can be better resolved); illumination characteristics (bandwidth, coherence, angle, etc.); processing limitations (PR fidelity, photo-acid dispersion, etc.); as well as resolution enhancements techniques (phase-shifting masks, off-axis exposure, etc.) [10, 11]. The minimum (single exposure) theoretical value for the \( k_1 \) factor is 0.25 [10]. This is an asymptotic limit with a value of \( k_1 \approx 0.28 \) considered the practically achievable single exposure limit [12].

The exposure wavelength, \( \lambda \), has followed a clear trend towards progressively shorter wavelengths. Beginning with 1:1 exposures using 514 nm (Ar laser), then progressing to lamp based reduction-projection systems using 436 nm (Hg lamp, g-line), 405 nm (Hg lamp, h-line) and 365 nm (Hg lamp, i-line). The industry then shifted to excimer laser based systems which provide shorter wavelengths, higher intensities and hence faster throughput, beginning with 248 nm (KrF laser), before moving onto the current industry standard 193 nm (ArF laser) [13, 14]. Further reductions to 157 nm and 121.6 nm were researched but deemed infeasible and subsequently abandoned [13, 11]. The current state of the art remains the 193 nm ArF excimer laser, where great strides have been made in power, throughput, bandwidth control, and laser lifetime since its introduction over two decades ago.

The numerical aperture, \( NA \), being the denominator in Equation (1.1) naturally has followed an increasing trend. In the last 50 years the NA has increased from about 0.25 to 0.93 for in-air exposure systems [15]. As these values are in air (\( n_{air} = 1 \)), they represent angles \( \theta = 14.5^\circ \) and \( \theta = 68.4^\circ \) respectively. Since \( \sin \theta \) has a maximum value of 1, further gains from increasing \( \theta \) are not worth the considerable expense required to develop the optical system, thus we look to the gains available from increasing \( n \). At first glance one would think the refractive index of the lens can simply be increased (ignoring the great difficulty of developing the extremely high quality materials required). This however is not as simple as it sounds, if the NA exceeds the refractive index of any of the media within the system the light will suffer total internal reflection (TIR) at high-index to low-index interfaces. To mitigate this effect
and further increase the NA, immersion photolithography was developed. Immersion photolithography has its final imaging lens immersed in ultra-purified water; this allows higher NA’s to be achieved before TIR occurs. Immersion photolithography at 193 nm (193i) is the current industry standard for large scale production of lithographic features. At this wavelength the refractive index of water is 1.44 [16], thus allowing the NA to increase above 1. In practice reaching an NA of 1.4 is very challenging from a lens design standpoint, consequently the NA has peaked at 1.35 [13, 17, 18].

Using these extreme values ($k_1 = 0.28$, $\lambda = 193$ nm and $NA = 1.35$) we arrive at what is considered the single exposure half-pitch resolution limit for 193i of approximately 40 nm. So with improvements in $k_1$, $\lambda$, and the NA largely depleted how have recent gains been made? The key has been the fact that $k_1$ has a single exposure limit of 0.25. Shifting to multiple exposures has allowed (the effective) $k_1$ to further decrease. The latest Intel skylake processors are on the 14 nm manufacturing node, corresponding (using Eq. (1.1)) to a $k_1$ value of 0.098 with triple patterning used in one or more critical stages of production [19]. Further manufacturing nodes (10 nm and 7 nm) are also expected to be manufactured using multi-patterning methods [19, 18]. However as multiple patterning (MP) is further increased the number of processing steps increases as does the number of photomasks, both of which substantially impact the manufacturing economics. As such the ITRS has identified four ‘likely’ alternative systems for the manufacturing of future process nodes (i.e. 10 nm, 7 nm, 5 nm, etc.): direct-write electron beam lithography (EBL), directed self assembly (DSA), nanoimprint (NIL), and extreme ultraviolet lithography (EUV) [20]. In the next section a brief summary will be give of the most likely (MP), likely (EBL, DSA, NIL, and EUV), and a proposed alternative complementary lithography which is part of the justification for the research presented in this thesis.

1.3 Lithography for future manufacturing nodes

1.3.1 Multiple patterning lithography

Multiple patterning is a process whereby the final design layout is achieved by multiple patterning stages. This is the current industry standard and is expected to remain so for at least the next two process nodes [19, 18]. Generally there are three components to multiple patterning: the patterning itself, mask/pattern decomposition, and resolution enhancement techniques [21].
Patterning is carried out using a variety of base techniques including (but not limited to): exposure, development, PR addition/stripping, etching, hardmask layers, chemical PR ‘freezing’, as well as spacer layers [22]. These base techniques are used in various ways to produce the desired outcome. There are a large number of multiple patterning methods in the literature [23, 21], as this is not the focus of this research only two representative examples will be introduced here (Fig. 1.3) litho-etch-litho-etch (LELE) and self aligned double patterning (SADP).

LELE (Fig. 1.3(a)) begins with a standard film stack of PR/hardmask/silicon wafer. At manufacturing process nodes 45 nm or less the PR thickness is reduced so much that the PR can no longer act as an effective barrier to etchants. To mitigate this an intermediate layer called a hardmask is introduced which has a much faster etch rate than the PR and hence can be effectively patterned [22]. A first exposure (yellow areas) is transferred into the hardmask by an etching process. PR is spun on a second time and a second exposure carried out, with the second pattern transferred to the hardmask by another etch process. Each expose/develop step is termed a ‘litho’ step while the hardmask etching steps are termed ‘etch’ steps, thus the name litho-etch-litho-etch.

SADP (Fig. 1.3(b)) begins with the same PR/hardmask/silicon film stack. Again it begins with an exposure then a development step. This is followed by the application of a spacer layer. Etching the spacer layer leaves behind the vertical parts of the spacer layer and exposes the PR which is then stripped away leaving just the vertical spacer layer patterns. This pattern is then transferred into the hardmask by another etching step, then finally the remaining spacer layer material is stripped away. The remaining pattern is termed self aligned due to the fact that it creates its own (doubled) pitch from the original exposure pitch and duty cycle. This method can produce a reduction in pitch of approximately 30% which is roughly equivalent to one manufacturing process node step [24].

Mask/pattern decomposition is another major component of MP. Multiple patterning requires the design pattern to be decomposed into multiple component photomasks. This is a necessity as diffraction hotspot effects, especially around line termination points and corners, can lead to over exposing of neighbouring areas thus ruining pattern fidelity. This issue has caused a major shift in processor design, leading to the development of restricted and gridded design rules [25, 26]. The simplest patterns to produce with the highest fidelity are regularly spaced lines, thus these new design rules are aimed at exploiting this fact by placing features in regular locations and orientations. Gridded design rules take this idea one step further by forcing the pattern
Figure 1.3: Multiple patterning examples. (a) Litho-Etch-Litho-Etch. (b) Self aligned double patterning.
1.3. LITHOGRAPHY FOR FUTURE MANUFACTURING NODES

Figure 1.4: Mask Decomposition. (a) Base pattern showing allowed spacings (ticks) and disallowed spacing (crosses). (b) Dividing of feature $a$ into $a$ and $b$ allows for permissable spacings. (c) Decomposed ‘blue’ mask. (d) Decomposed ‘red’ mask.

locations to line up with regular areas on an underlying grid, i.e. not only a regular pitch but also minimal staggering of features to retain long range alignments [26]. This is often at the expense of added layout area, but with reduced manufacturing costs due to the comparative simplicity of the layout [25].

With these new design rules in mind the overall pattern needs to be decomposed into constituent photomasks. In some respects the key to this is the fact that the single exposure resolution limit is approximately 40 nm, this provides the minimum mask feature separation and provides the underlying grid dimension for the gridded design rules. The method of feature colouring is employed to decompose the mask (Fig. 1.4) [27, 28], the idea being that if any two adjacent features are within the single exposure resolution limit (40 nm) they must be placed on separate masks. In Figure 1.4(a) we have the total pattern in green, features $a$, $c$, and $d$ are all within the resolution limit of each other (marked by red crosses). It can be seen in the colour decomposed pattern (Fig. 1.4(b)) that feature $a$ must be split between two masks to allow the minimum distance between colour criteria to be met. In comparison the distance between $c$, $d$, and $e$ are all greater than the minimum distance thus $e$ remains unchanged. We then see an example of the resultant decomposed blue and red masks (Figs. 1.4(c) and 1.4(d)). This is a very simple example, when spread across a full processor design incorporating billions of transistors suitable colour separation can take a very long time to achieve.

Another aspect of MP methods are resolution enhancement techniques. Generally these fall into two categories: source based enhancements, and mask/pattern optimizations. Source based enhancements include off axis illumination, and dipole illumination.
Off-axis illumination tilts the illumination source and hence the mask transmission diffraction orders at an angle. Rather than the traditional lens collection of, for instance, the $0^{th}$ and $\pm 1^{st}$ diffraction orders, in off-axis illumination the lens will collect the $0^{th}$ and only one of the $1^{st}$ with the other being blocked. These orders recombine at half the angle compared to the angle between the two $1^{st}$ diffracted orders resulting in an improved depth of field [29, 30]. Dipole illumination allows for resolution enhancement perpendicular to the axis of illumination, but comparatively worse in the parallel direction [22]. Naturally this plays into mask decomposition considerations providing a natural split along the lines of separate X-axis and Y-axis illuminations, i.e. X and Y masks. Mask/pattern optimizations include, phase-shifting masks, optical proximity corrections, and sub-wavelength mask assist features. Phase shifting masks exploit interference effects to produce sharper line shapes [31, 32]. Optical proximity corrections alter the mask layout to minimize under/over-dosing of particular features [23, 33, 34]. Sub-wavelength mask assist features are mask additions which are too small to pattern but nevertheless add enough energy to improve PR feature definition [35].

The complexity of these techniques (patterning, mask/pattern decomposition, and resolution enhancement techniques) has inevitably led to the development of computational lithography, the full scale simulation of the lithography process [36]. With each manufacturing process node shrink the computational lithography needs increase massively, especially with the shift from planar gates to 2.5D/3D FinFet and TriGate transistors in which errors in all three spatial dimensions need to be considered.

**Future prospects of multiple patterning lithography**

**Pros:** A known quantity. Uses existing hardware. Design rules simplify photolithography stages.

**Cons:** Very expensive mask sets. Computational decomposition of design into several masks. Design rules require thorough simulation.

**Outlook:** Multiple patterning is the industry standard for large scale semiconductor production and is likely to remain so for the next few manufacturing nodes. The benefit of being a ‘known’ quantity means that current manufacturing methods can be adapted to the tighter constraints of future manufacturing nodes without the very large capital outlay required to retool for a new lithography method.
1.3.2 Extreme ultraviolet lithography

Extreme ultraviolet lithography is seen by many in the industry as the natural successor to 193i with MP [37, 38, 39]. EUV operates at a wavelength of 13.5 nm; considering Eq. (1.1) clearly the approximately 15 fold decrease in wavelength has massive potential to improve the resolution. This is particularly beneficial due to the fact that it allows smaller feature sizes without all the necessary complications associated with heavy use of MP.

EUV lithography in principle, is the same as 193 nm optical projection lithography i.e. light incident upon a mask produces a pattern in PR which is then used to transfer the pattern onto silicon. In practice however many factors are different. The 13.5 nm wavelength used is so short that it must be produced from an excited 4d electron in a multiply ionized tin plasma [40]. The tin is converted into a plasma using either a CO$_2$ laser or a spark gap discharge; the excited plasma then emits a broadband EUV burst [41]. A multi-layered molybdenum-silicon mirror collector is then used to filter and focus the light. EUV light is absorbed by all matter, which has two significant impacts on any EUV system. Firstly, the entire optical system must be within a vacuum, to prevent absorption by air. This means that any materials/parts used in the system must be manufactured and used (i.e. out-gassed) in a manner which prevents contaminants entering the system. Secondly, it means refractive lenses cannot be used in the system as they will reduce the power too much through absorption. Thus a complex array of multi-layered molybdenum-silicon mirrors are employed as the reflectance losses are far less than the transmittance losses. Each mirror has a theoretical maximum reflectance of 72%, the rest being absorbed [38]. Due to the short wavelength involved, phase effects from defects are greatly enhanced. For this reason the mirrors must be manufactured to extremely tight tolerances, accurate to less than 2 nm across a 30 cm mirror, which is equivalent to 1 mm across a 1500 km surface [38]. The compounding mirror losses coupled with the poor wall-output efficiencies of the laser or discharge plasma (i.e. a 40 kW CO$_2$ laser is needed to produce a 100 W EUV output) make the development of a suitable EUV light source one of the most pressing issues in the commercial establishment of EUV lithography [20].

The ITRS roadmap identifies two other significant short term challenges [20]. Firstly, production of defect free masks and mask manufacturing. EUV masks are mirror masks, they are made of the same multi-layered molybdenum-silicon structure as the beam transport mirrors in EUV systems. This multi-layered structure makes them very
prone to compounding defects i.e. the footprint of a dust grain on the bottom layer will get sequentially bigger as successive layers are added \cite{20}. Secondly, much work is also required on the PR formulation to meet the promise of EUV lithography. At EUV wavelengths, rather than just breaking chemical bonds as happens at visible/UV wavelengths, the EUV photons cause ionization and the production of secondary electrons which can migrate and effectively blur the image \cite{42}.

**Future prospects of extreme ultraviolet lithography**

Pros: Shorter wavelength provides direct resolution benefits. Compatible with many already developed multiple patterning techniques.

Cons: Massive intensity loss from the reflecting optics necessitates a far larger source power than 193i. Use of CO\textsubscript{2} laser plasma source massively impacts the wall efficiency of any source. Mask and mirror errors highly prone to defects due to the multi-layered nature.

**Outlook:** Although EUV has made great advances, it continues to be beset by light source problems. This coupled with the relative strength of MP keeps pushing EUV to later process nodes. One of the greatest benefits of EUV is that it doesn’t require multiple patterning, but if EUV continues being delayed it’s likely to require such methods upon its release.

### 1.3.3 Direct-write e-beam lithography

Direct write electron beam lithography (EBL) uses a narrow pencil beam of electrons (or an array of such beams) to directly write a pattern in PR. Very fine control of the e-beam(s) allows for regular sub-10 nm resolution as well as the patterning of arbitrary patterns \cite{43, 44}. For this reason EBL is already well established as the method by which photomasks are made, as well as for production of low-volume, test, and research semiconductor devices \cite{45, 46, 47, 44}. The use of individual e-beams however raises the biggest issue of EBL, that is, an inherently very low throughput. For this reason research is being focussed towards massively parallel e-beam systems \cite{47, 48, 49, 44}. Massively parallel systems employ tens to hundreds of thousands of individual beamlets to write large areas simultaneously. Producing, shaping, and targeting this many beamlets is a significant technical challenge which raises interesting theoretical issues considering the information bandwidth of such a system compared to that of a conventional large area 193i exposure \cite{50, 51}. The ability to directly produce sub-10
nm patterns without the development and use of a (very) expensive mask set is however a great benefit and thus development continues in this area. Sub-5 nm linewidths have been produced with single beam systems [52, 53], although producing dense features with such a linewidth are difficult considering the space charge and proximity effects that come from using multiple beams.

Future prospects of direct-write e-beam lithography


Cons: Low throughput, EBL is an inherently slow process considering the write volume of a single beam. Space charging and proximity effects limit the write density. Very high informational bandwidth required for massively parallel systems.

Outlook: The ITRS cites direct write EBL as a possible use for producing less dense features such as contact holes, and cut levels. Throughput of massively parallel e-beam systems is still too low for commercial semiconductor patterning.

1.3.4 Nanoimprint lithography

Nanoimprint lithography (NIL) uses a nano-patterned mold to imprint a pattern into a soft resist [54]. NIL in contrast to all other ITRS roadmap technologies is an inherently high throughput method due to its ability to patterning full wafers in a single step, and its potential for being employed in massively parallel configurations. The simplicity and low cost of NIL make it ideal for the manufacturing of high nanometer/low micrometer structures where it has found use in novel electronic developments (hybrid plastic and organic electronics), photonics (organic lasers, OLED pixels, diffractive elements, display elements, and broadband polarizers), structures for control of polymer growth, medical nanoparticles, and in biological applications such as DNA manipulation and protein patterning [55, 44, 56]. Clearly NIL is a technique of great promise, to the point where it was named one of the “Ten Breakthrough Technologies of the Year” by the MIT technology review in 2003 [57]. The transition to production scale volumes of feature sizes required for advanced semiconductor devices however has yet to be achieved [20].

The process of NIL is conceptually very simple, i.e. a mold is pressed into a soft deformable material which is then cured via heat or UV illumination. The mold is then released revealing the pattern for further processing. This 1:1 reproduction allows
for excellent resolution and low line-width roughness. As the feature size decreases however it begins to cause issues. Of particular importance are defect errors; these are particularly damaging in an NIL setting as they will be replicated in subsequent imprints, and the master mold can potentially pick up further debris. Reduction of defects is achieved via slowing down the imprint process which directly impacts NIL’s greatest benefit, that of natively high throughput. Also of considerable difficulty is the production (and infrastructure required) of such high resolution defect free 1:1 imprint molds using other lithography methods [20].

Future prospect of nanoimprint lithography

Pros: Natively high throughput. Very good resolution and line width roughness.

Cons: Prone to defect errors. Low throughput required to reduce defects. Mask production difficult at small feature sizes.

Outlook: NIL despite its potential has yet to break into the traditional semiconductor IC manufacturing processes with their much more stringent production constraints. The primary issues are defect reduction, and mask production.

1.3.5 Directed self assembly lithography

Directed self assembly (DSA) is the guided production of nanopatterns using the self aligning of block co-polymer phase domains. Generally a mixture of two immiscible block co-polymers are used. Thermodynamic incompatibility forces a separation of the two block co-polymers into domains [58]. The development of domains can be guided using traditional lithography methods which act as a template for self assembly [59]. The two general forms of which are epitaxial (chemically defined) and graphoepitaxial (topologically defined) self assembly [60, 59, 58]. The polymer phases have different etch profiles so can be used for pattern transfer. In many respects DSA is the ultimate for nanopatterning as it produces patterns literally at the molecular scale. Transferring this to the macroscopic length scales required to semiconductor fabrication however is proving to be the biggest road block, with one of the main problems being incomplete phase separation in both 2D and 3D [20].

The use of self-assembling materials in a patterned template offers several benefits, an example of which would be using a photolithographically produced regular pitch line pattern with block co-polymers in the pattern depressions. If the equilibrium block co-polymer period is commensurate with the line pitch, a very dense line period can
form between them. Generally block co-polymers form an equilibrium pitch of 3-50 nm [59]. Thus it can be seen the great potential which exists for pitch division [20]. Due to the phase domain nature of the pattern it can also have very good line edge roughness, and very good pattern reproduction considering the lack of proximity effects. Another benefit of this system is the chemical nature of it means it can potentially have very low processing costs.

Future prospects of directed self assembly lithography

Pros: Pattern multiplication (4x demonstrated [59]). Very good line edge roughness. Low processing costs. Benefits from heavy co-development in other industries [58].

Cons: Defects. Pattern placement (for future manufacturing nodes). Modification of designs to fit DSA properties.

Outlook: DSA has many potential benefits but these, particularly the development costs, need to be weighed up against conventional well understood photolithography techniques. Generally it is thought that its most likely application will be in highly regular structures such as memory modules [20].

1.3.6 Shortcomings of next generation lithography techniques

From the brief overview above it can be seen that the identified next generation lithography techniques suffer from some fairly major difficulties as summarised in Table 1.1. With the difficulties and limitations present in all these methods, several entities are investigating complementary lithography techniques designed to leverage the strengths of different methods to overcome their individual shortcomings [61, 36, 62].

1.4 Complementary lithography

Complementary lithography seeks to leverage the strengths of different lithography techniques. Generally one method is used for large scale but inflexible patterning, while a secondary slower but more flexible method is used to cut the finer scale patterns. An example of this is given in Fig. 1.5 where an interference lithography exposure is used to create a large area interference pattern and e-beam lithography is used to produce the more arbitrary cut lines. Complementary lithography has four primary
<table>
<thead>
<tr>
<th>Lithography technique</th>
<th>Difficulties and limitations</th>
</tr>
</thead>
<tbody>
<tr>
<td>193 nm immersion</td>
<td>Rayleigh’s criterion limits - $\lambda, k_1$, and NA.</td>
</tr>
<tr>
<td>Multiple patterning</td>
<td>Reduced throughput and greater manufacturing complexity for every extra step (cost and compatibility). Compounding alignment constraints with feature size reduction and increasing step number. Ever increasing number and cost of photomasks.</td>
</tr>
<tr>
<td>Direct-write e-beam</td>
<td>Difficulty controlling e-beam targeting (charge build up, outside fields, and beam-beam interactions). Small area of exposure. Low throughput.</td>
</tr>
<tr>
<td>Nanoimprinty</td>
<td>Low tolerance for defects. Sub 50 nm mask must be produced by other lithographic means reducing cost competitiveness. Throughput</td>
</tr>
<tr>
<td>Directed self assembly</td>
<td>Only suitable for regular patterns. Defects Requires new design rules.</td>
</tr>
<tr>
<td>Multiple methods</td>
<td>Increasing computational lithography costs. Low throughput. New design rules required.</td>
</tr>
</tbody>
</table>

**Table 1.1**: Difficulties and limitations of next-gen lithography technologies.

Strengths: 1) the use of a regular large area pattern fits nicely with the concept of gridded design rules, 2) off-loading the fine detail to a more suitable technique (say e-beam lithography) will massively reduce mask complexity and cost, 3) reducing mask complexity also leads to a reduction in computational lithography needs, 4) usage of multiple methods allows for better integration of ‘More than Moore’ technologies.
Figure 1.5: Complementary lithography schematic. (a) - Large scale patterning is produced rapidly using interference lithography. (b) - A slower secondary finer resolution cut pattern is carried out using e-beam lithography. (c) - The final combined pattern showing the cut patterns in the regular lined structure.

For instance hybrid electronic/sensor systems may require semiconductor, photonic, organic and even biological components and thus a variety of lithography techniques to produce them.

So far several different complimentary lithography combinations have been studied to some degree: 193i/EBL [50, 43], Optical/NIL [50, 36], and Interference lithography/193i [36]. Each of these uses the strengths of both lithography methods to achieve ends not (easily) manageable by either method alone.

193i/EBL

193i - Large area, rapid throughput, regular (straight line) structures
EBL - Small area, slow throughput, irregular (arbitrary) structures

Optical/NIL

Optical lithography - Simple micron sized patterning, positive tone PR
NIL - Simple nanometer sized patterning, same positive tone PR thermally set

Interference lithography/193i

Interference lithography - Large scale regular pattern, rapid throughput
193i - Large scale less regular pattern, rapid throughput

Interference lithography (also called holographic lithography) is a powerful tool for complementary lithography as it allows full wafer patterning of a very narrow pitch with $k_1$ capable of reaching its theoretical single exposure minimum of 0.25. The optical
nature of it makes it compatible with all existing manufacturing photolithography systems and capable of utilising the same materials and processing steps, and would be inherently compatible with gridded design rules [62].

One of the limits of complementary lithography is the minimum feature size of the large area patterning step as this is what ultimately determines the combined feature size. For this reason 193i and interference lithography methods are the most likely methods to be employed for this patterning stage. Another option however is the use of modern contact photolithography methods such as evanescent near-field optical lithography (ENFOL) and grating coupled near-field interference lithography which also potentially offer very small feature sizes over a large area.

Interference lithography is the ‘base’ technique for this thesis, in the following chapter we will delve further into the fundamentals of interference lithography and investigate methods of extending it to shorter pitches.

1.5 Thesis overview

In this body of research we choose to investigate various techniques to enhance interference lithography, particularly in the evanescent regime; not only because of its possible application to complementary lithography schemes, but also because it is useful in its own right for producing high quality nano-patterning for applications such as diffraction gratings, nanophotonics, directed self assembly templates, nanofluidic devices, nanomagnetic devices, nanoscale epitaxial growth, metrology, interferometry, optical lithography testing systems, etc. [50]. The layout and content of this thesis are as follows:

Chapter 1 - Introduction: Lithography - To set the scene for this research current lithography techniques are introduced with an emphasis on the developments required to meet the manufacturing criteria for future process nodes. Complementary lithography exploiting interference lithography is identified as a possible alternative to the ‘mainstream’ lithography techniques. The chapter concludes with a list of the research outputs for this body of research.

Chapter 2 - Interference lithography - The concepts and theory of 2 beam interference and interference lithography are explored. Numerical aperture regimes are introduced leading to the ultra high-numerical aperture regime defined by evanescent fields within the photoresist. The evanescent fields within the photoresist result in a poor depth of field, to improve this resonant underlayers are introduced. Finally
near-field grating coupled interference schemes are described as an introduction to the systems developed in chapter 6.

Chapter 3 - Herpin effective media resonant underlayers - The dielectric resonant underlayers introduced in chapter 2 have an upper numerical aperture limit dependent upon their constituent refractive indices. In this chapter we investigate the use of Herpin effective media as resonant underlayers. Herpin effective media are known to be able to theoretically produce effective refractive indices greater than the constituent materials. As such, the aim of this chapter is to investigate if these extremely high effective refractive index materials are capable of acting as resonators at extremely high numerical apertures. Transfer matrix method and Herpin effective media theory are developed to investigate this possibility. This chapter closes with an investigations of resonant underlayer parameter space which leads to the development of two new techniques, evanescent coupled antireflection coatings, and overlayer resonators.

Chapter 4 - Evanescent-coupled antireflection coatings - This chapter begins with a general introduction to antireflection coatings, describes the forms available, then covers how they are applied for photolithography purposes. Evanescent-coupled antireflection coatings are then introduced as an alternative to the ‘traditional’ quarter-wave type bottom antireflection coating for hyper-numerical aperture photolithography. The transfer matrix method is employed to design, optimize, and characterize the different forms of evanescent-coupled antireflection coatings. The chapter closes with several examples suitable for modern 193 nm immersion lithography methods.

Chapter 5 - Experimental verification of evanescent-coupled ARCs - A primer is given on the interference lithography experiment focussing on the experimental apparatus, film stacks, and photolithography processing. The factors influencing experimental design are then discussed, these include: the optimal development process, the optimal imaging method, and most importantly how successful reflection suppression will present experimentally. Two forms of evanescent-coupled antireflection coatings are investigated, a surface state resonator based antireflection coating employing a MgF$_2$|Cr resonator, and a dielectric resonator based antireflection coating employing a SiO$_2$|HfO$_2$ resonator. Experimental results and possible experimental errors are then discussed.

Chapter 6 - Genetic algorithm optimization of grating coupled near-field interference lithography - Grating coupled near-field interference lithography is introduced as a method of extending interference to extremely high numerical apertures. A brief literature review of the topic is covered to decide on goals for this research. The
finite element method is then covered as the means for modelling of these systems. To reduce the time consuming nature of system optimization using finite element methods a genetic algorithm optimization method is employed. Trial models are then developed for overlayer resonator systems and dual resonators systems. These models are then used as test-bed systems to reduce the interference pitch as much as possible.

Chapter 7 - Conclusions and future work - This chapter covers the general development of the thesis focussing on each of the body chapters (3, 4, 5, and 6). A brief background for each is given followed by the key original developments of this research. Suggestions for future research directions for each chapter are then given.

1.6 Research outputs

At the time of submission the research outputs of this body of work include 3 journal publications (with a further one submitted and another at the pre-submission stage), 1 provisional patent, 1 conference presentation, and 2 poster presentations.

Journal publications (published)


Journal publication (submitted)


Journal publication (pre-submission)

1.6. RESEARCH OUTPUTS


**Intellectual property submissions**

**Conference presentations (presenter in bold)**


**Poster presentations**

Chapter 2

Interference lithography

Prologue

This chapter provides a brief theoretical introduction to 2-beam interference. Interference lithography (IL) is introduced along with its two main variants, Lloyd’s mirror and Mach-Zehnder interferometer based systems. Low, high, and ultra-high NA exposure regimes are introduced along with the requirements and consequences of each regime. A theoretical description of the evanescent fields present in the ultra-high NA regime is produced. These evanescent fields result in a much diminished depth of field for ultra-high NA IL, and to counter this evanescent coupled resonant underlayers are introduced. The chapter closes with a description of contact lithography and near-field grating coupled interference lithography schemes.

The purpose of this chapter is for the reader to better understand the theoretical and experimental methodologies employed in later chapters. All of these employ interference lithography as a test bed system, evanescent fields, and resonant underlayers for depth of field enhancement.

2.1 2-beam interference

Interference of two coherent light beams is one of the classic experiments in physics, where it was used to confirm the wave nature of light [63]. Despite its age this effect is still very much at the forefront of modern technology, for example the LIGO experiment which recently announced the detection of gravity waves operates on the same principles (using a very large Michelson interferometer) [64]. For the purposes of this thesis a brief
2.1. 2-BEAM INTERFERENCE

Figure 2.1: 2-beam interference lithography schematic.

Overview of this topic will suffice. If the reader requires a more thorough treatment of this topic it is available in many optics textbooks [63, 65].

When two coherent plane waves from two mutually coherent sources with a fixed phase relationship overlap they form a complicated amplitude distribution called an interference pattern (as outlined in Figs. 2.1 and 2.2). The theoretical description of this effect begins with two in phase (i.e. \( \Delta \Phi = 0 \)) time independent monochromatic plane waves

\[
E_1(r) = E_1 e^{i k_1 \cdot r} \quad \text{and} \quad E_2(r) = E_2 e^{i k_2 \cdot r}.
\]

(2.1)

When these two waves overlap the intensity is given by the square of the sum of the electric fields

\[
I_{12} \propto |(E_1(r) + E_2(r))|^2.
\]

(2.2)

\[
I_{12} \propto E_1(r) \cdot E_1^*(r) + E_2(r) \cdot E_2^*(r) + [E_1(r) \cdot E_2^*(r) + E_2(r) \cdot E_1^*(r)]
\]

(2.3)

\[
I_{12} \propto I_1 + I_2 + 2E_1 \cdot E_2 \cos((k_1 - k_2) \cdot r).
\]

(2.4)

From this we can see that the resultant intensity \( I_{12} \) is not only the sum of the two incident intensities but is also modulated by a cosine term. Due to the nature of the cosine function there will be maxima and minima (collectively called fringes) when cosine is at its extremities. The variable inside the cosine \( (k_1 - k_2) \cdot r \) is called the spatial phase \( \Phi_s \) as it dictates the phase of the cosine relative to the spatial position \( r \).

To investigate the spacing of the maxima and minima we must further expand the \( k \) vectors. The difference in \( k \) vectors is often termed the fringe vector, and can be expanded in Cartesian coordinates as

\[
k_f = k_1 - k_2 = k_{fx} \hat{x} + k_{fy} \hat{y} + k_{fz} \hat{z}.
\]

(2.5)
2.1. 2-BEAM INTERFERENCE

Figure 2.2: General interference schema. Two coherent incident TE beams \( k_1 \) and \( k_2 \) produce an interference pattern in the \( x \) direction. The beams also interfere with their reflections (\( k_1 \) and \( k'_1 \)) from the \( x \) axis mirror to produce a standing wave interference pattern in the \( z \) direction.

Taking \( r \) in the direction of \( k_f \) the spatial phase becomes \( \Phi_s = |k_f|r \). Thus we can say the periodicity of the cosine and hence the fringe periodicity \( (P) \) will be

\[
P = \frac{2\pi}{|k_f|}. \tag{2.6}
\]

Assuming monochromatic light and equal angles of incidence (Fig. 2.2)

\[
|k_1| = |k_2|, \tag{2.7}
\]

and hence via expansion

\[
k_{1x} = -k_{2x}, \ k_{1y} = k_{2y}, \ \text{and} \ k_{1z} = k_{2z} \tag{2.8}
\]

thus

\[
k_f = 2k_{1x}\hat{x}. \tag{2.9}
\]

If we substitute Eq. (2.9) into Eq. (2.6) and use the wavenumber \( k = 2\pi n/\lambda \) we arrive at the equation for fringe pitch

\[
P = \frac{\lambda}{2n \sin \theta} \tag{2.10}
\]

where \( n \sin \theta \) is the NA with \( n \) being the refractive index and \( \theta \) the angle of incidence shown in Fig. 2.2. This is identical to the general resolution equation (Eq. (1.1)).
where $P$ is the full pitch (i.e. $2R$) and $k_1$ the resolution scaling factor is its theoretical maximum of 0.25.

If we now consider $k_1$ to be incident on a reflecting surface ($x$ axis in Fig. 2.2), then we have another interfering wave $k'_1$ from a virtual source. The $k_1$ for this interference will be called $k_{sw}$ as it represents the standing wave fringe vector. Now with the only asymmetry being in the $z$ direction we have

$$k_{sw} = |k_1 - k'_1| = 2k_{1z} \hat{z} \quad (2.11)$$

If we substitute Eq. (2.11) into Eq. (2.6) and use the wavenumber we arrive at the equation for standing wave fringe pitch

$$P_{sw} = \frac{\lambda}{2n \cos \theta} \quad (2.12)$$

The same 2-beam (and by extension multi-beam) interference effects are commonly employed as a lithographic patterning technique.

### 2.2 Interference lithography

If a layer of PR is placed at the mirror plane in Fig. 2.2 an interference pattern such as that delineated by the yellow line in Fig. 2.1 will be produced. Patterning by this method is termed interference lithography. Interference lithography is carried out in two standard configurations: Lloyd’s mirror, and Mach-Zehnder interferometers (Fig. 2.3).

Lloyd’s mirror interferometers (Fig. 2.3(a)) employ a corner type configuration where half the incident beam is reflected back onto the other half allowing the beams to interfere. The Lloyd’s mirror type system offers several benefits: it is simple, compact and relatively cheap requiring a minimal amount of optics; rotation about the interferometer corner allows easy change of incident angle and hence pitch; and the short path length between the incident and the reflected beams make this system more robust against air currents. The monoblock mirror/sample holder also makes it resistant to vibrations as the whole system moves in unison. In some respects the compactness of the Lloyd’s mirror system is its biggest drawback in that it is not (easily) flexible in terms of automation, as well as sample and mirror mounting configurations. This system can also not be used with lasers with poor beam transverse coherency (such as excimer lasers), where the two interfering halves will generally not be mutually coherent and thus produce poor interference [50]. Classically this is an open air exposure method with modern variants including solid immersion [66, 67] and liquid immersion.
2.2. INTERFERENCE LITHOGRAPHY

The method chosen for this research (Chapters 3-5) is a solid immersion Lloyd’s mirror system.

Mach-Zehnder interferometers act by splitting a single incident beam into two separate beams and recombining them to interfere (Fig. 2.3(b)). The beam is generally either split with a diffraction grating (as depicted) or a beam splitter. Steering mirrors are then used to recombine the beams on the sample. In the case of a diffraction grating splitter the +1 and -1 diffracted orders are recombined with the 0th order being blocked. The main benefit from using a Mach-Zehnder type system is its flexibility, with its ability to steer the beams at will, enough room to add other beams, and an unobstructed sample stage. In contrast to Lloyd’s mirror interferometers, the two overlapping beams have the same spatial relation, thus deep-UV excimer lasers (the type employed for commercial photolithography purposes) which have poor transverse coherency are able to be employed [50]. These features make this system a favourite for PR testing applications were a simple (compared to full patterning systems) very well controlled system is required. This flexibility (read complexity) is also its greatest drawback compared to Lloyd’s mirror interferometers in that it’s more expensive, and more prone to vibrations and air currents. Variants of this also system include solid immersion [69, 70] and liquid immersion [50, 71].

Figure 2.3: *IL methods.* (a) - Lloyd’s mirror interferometer. (b) - Mach-Zehnder interferometer. Numbers represent diffracted orders.
2.3 Interference lithography NA regimes

Interference lithography and photolithography in general can be split into three different exposure regimes based on the numerical aperture (NA) and its limiting factors (Fig. 2.4). The NA is usually defined as the refractive index multiplied by the half angle of acceptance of a lens. Lloyd’s mirror systems however have no lenses, so one may ask what exactly is the NA of an IL system without a lens? In this instance it refers to the angle of incidence to the PR surface normal. Low-NA (Fig. 2.4(a)) exposures occur in the air which limits the practically achievable NA to less than the refractive index of air ($n_{\text{air}} = 1$). Assuming $\lambda = 193\text{ nm}$ and an $\text{NA} = 0.9$ the minimum achievable pitch (by Eq. (2.10)) is 107.2 nm.

To further increase the resolution one must increase the refractive index of the medium above the PR (the PR coupling medium), this is the high NA regime (also termed hyper-NA within the industry). In the high NA (Fig. 2.4(b)) regime exposure occurs through a liquid (or infrequently a solid), thus the term immersion lithography. The semiconductor industry employs ultra-purified water for this purpose with a refractive index ($\lambda = 193\text{ nm}$) of 1.44 [72]. This increase in refractive index allows the NA to increase by a commensurate amount. Assuming $\lambda = 193\text{ nm}$, and an achievable $\text{NA}$ of 1.4, gives a single exposure minimum pitch in water of 68.9 nm. As there are many materials with a refractive index greater than the PR refractive index ($n_{\text{PR}}$) the upper NA limit for this regime is when the fields within the PR are no longer propagating, this occurs when the NA exceeds $n_{\text{PR}}$. The real part of $n_{\text{PR}}$ is approximately 1.71 [10], assuming an achievable $\text{NA}$ of 1.65 and $\lambda = 193\text{ nm}$, a single exposure minimum pitch of 58.5 nm is possible. The industry has thoroughly investigated high refractive index liquids (so called gen-2 and 3 liquids) to further exploit this regime [16, 22, 72]. Although low absorption liquids with higher refractive indices are available, the relatively small gains in resolution were eventually deemed not worth the substantial cost required to retool the exposure systems and overcome any material incompatibilities [72].

Further gains in resolution can be made by utilizing evanescent fields in the PR; this is termed the ultra-high NA regime. Although conventional wisdom would have it that no energy can be transferred via evanescent fields this is not the case, and indeed evanescent fields are an effective means of patterning [73, 74, 75, 76, 77, 78]. The cost of using evanescent fields however is the exponential decay of the fields, and its subsequent impact on the depth of field as can be seen in Figure 2.4(c). The NA in this
2.3. INTERFERENCE LITHOGRAPHY NA REGIMES

Figure 2.4: Interference lithography NA regimes. (a) Low NA regime - Open air exposure with a propagating field in the PR. (b) High NA regime - Coupling through a higher refractive index (than air) medium, typically water or an index matching liquid (IML). Propagating fields within the PR. Extra path length leads to greater absorption within the PR. (c) Ultra-high NA regime. Coupling through a medium with an index greater than $n_{PR}$. Evanescent fields within the PR. Much reduced depth of field within the PR due to evanescent fields and absorption.

regime has a practical limit imposed by the layers immediately above the PR. If any of the layers has a refractive index less than the NA the fields within it will be evanescent, and if this layer is too thick it will effectively prevent any energy from reaching the PR and thus prevent patterning. Further to this, in a prism coupled system if the NA exceeds the prism refractive index energy cannot couple into the optical stack making patterning practically impossible\(^1\) thus the NA is essentially constrained to less than 2.9 (at $\lambda = 193$ nm) with the highest refractive index available low-absorbing material being diamond ($n = 2.93 + 0.017i$ \(^79\)). The use of diamond in large scale optics is currently uneconomical; however the ultra-high NA regime (at 193 nm and other wavelengths) is readily achievable with high index glasses and crystals such as SF11, YAG and sapphire allowing NAs $\approx 1.8$ \(^77, 80, 78, 81\).

The exploitation of evanescent fields in photolithography is a major theme of this thesis, thus in the next section evanescent fields and the use of resonant underlayers to enhance their poor depth of field will be properly introduced.

\(^1\)The NA can however be further increased using diffraction grating coupling methods (see Chapter 6)
2.4 Evanescent fields

Evanescent fields are exponentially decaying EM fields that occur at interfaces upon total internal reflection (TIR) (Fig. 2.5). When light travels from one medium to another the angle of refraction is determined from Snell’s Law (Eq. (2.13)).

\[
n_1 \sin \theta_1 = n_2 \sin \theta_2 \quad (2.13)
\]

If \( n_1 > n_2 \) then \( \theta_2 > \theta_1 \); thus an angle (the critical angle) exists where \( \sin \theta_2 = 1 \). TIR occurs whenever \( n_1 \) is greater than \( n_2 \) and the critical angle is exceeded (Eq. (2.14)).

\[
\sin \theta_1 > \frac{n_2}{n_1} \quad (2.14)
\]

When TIR occurs an EM field travels along the interface (i.e. \( \theta_2 = 90^\circ \)) and no light propagates away from the interface into the second medium. This, however, isn’t the full story as it would require a discontinuity in the tangential \( E \) field interface conditions, which is forbidden [63]. If we examine the field in medium 2 we have

\[
E_2(r, t) = E_2 e^{i(k_2 \cdot r - \omega t)}. \quad (2.15)
\]

Expanding the dot product we have

\[
k_2 \cdot r = k_2 \hat{x} + k_2 \hat{z} = k_2 \sin \theta_2 \hat{x} + k_2 \cos \theta_2 \hat{z}. \quad (2.16)
\]

Using identities we have

\[
k_2 \cos \theta_2 \hat{z} = \pm ik_2 \sqrt{n_2^2 \sin^2 \theta_1 - 1} \hat{z}, \quad (2.17)
\]

substituting into Eq. (2.13) we get

\[
k_2 \hat{z} = \pm ik_2 \sqrt{n_2^2 \sin^2 \theta_1 - 1} \hat{z} = \pm i \beta \hat{z}. \quad (2.18)
\]

Substituting Eq. (2.18) back into the Eq. (2.15) and discarding the non-physical positive exponential we have

\[
E_2(r, t) = E_2 e^{-\beta \hat{z} \cdot x} e^{i(k_2 \sin \theta_2 \hat{x} - \omega t)}. \quad (2.19)
\]

Equation (2.19) indicates the field in medium 2 consists of two parts, an exponentially decaying component perpendicular to the interface called an evanescent field and a propagating field component parallel to the interface.

Note that Snell’s Law is equivalent to \( NA_1 = NA_2 \) i.e. the NA doesn’t change as the EM fields traverse a multilayered system. For this reason the NA is often referred to as \( \beta \) the Snell’s law invariant in thin-film optics.
2.4. EVANESCENT FIELDS

Figure 2.5: Evanescent field schemes, $n_1 > NA$ and $NA > n_2$ in all schemes. Dark blue - $NA > n$. Light blue - $NA < n$ and $\kappa = 0$. Red - $NA < n$ and $\kappa > 0$. (a) Total internal reflection. The evanescent field in $n_2$ is delineated by a dotted line. (b) Attenuated total reflectance. The evanescent field in $n_2$ also has an absorbing component, thus the faster decay of the field and reduced reflectance. (c) Frustrated total internal reflection. The incident energy is redistributed between the reflected and transmitted beams. A small amount of absorption may also be present in $n_2$.

The common understanding of evanescent fields is that no energy propagates across the interface. Assuming medium 2 is non-absorbing and many wavelengths thick this is indeed the case. If $z$ is much greater than the wavelength the exponentially decaying component will be effectively zero. As the propagating component is sinusoidal along the interface its field will alternately impinge on medium 1 then medium 2, thus the time average of the Poynting vector will be zero and no (total) energy will flow into medium 2, therefore all the energy will be reflected (hence TIR). There are however two exceptions to this where the fields are perturbed: 1) if medium 2 is absorbing attenuated total reflection (ATR) occurs, or 2) if a third medium with $n_3$ greater than the NA is present near ($d \approx \lambda$) to the medium 1/medium 2 interface frustrated TIR (FTIR) occurs [63, 82].

If medium 2 is absorbing an extra decaying component arises [83]. This can be seen to be coming from the fact that $n_2$ has a complex refractive index ($\tilde{n} = n + i\kappa$), thus when the complex version of the wavenumber is used ($k = 2\pi\tilde{n}/\lambda$) an absorbing component arises in the $x$ direction (from $i^2\kappa\hat{x}$) and a propagating component in the $z$ (from $i\kappa\hat{z}$). When a third medium is brought in to within $d \approx \lambda$ of the medium 1/medium 2 interface whatever amount of the $E_z$ component remains can couple across the interface become propagating again, provided $n_3$ is greater than the NA [82].
Both ATR and FTIR are integral to this research. ATR allows energy to be deposited in the PR and thus patterning to occur. FTIR allows energy to evanescently couple into resonant underlayers thus producing an improved depth of field for evanescent exposures.

2.5 Resonant underlayers in the ultra-high NA regime

Resonant underlayers (also called reflectors) were developed to improve the depth of field in the ultra-high NA regime for evanescent-field lithography [74, 83, 77]. In this regime the depth of field is severely reduced due to the dual exponential decays from the (dominant) evanescent field and from the PR absorption (Fig. 2.4(c)). It was recognised that the solution to this would be to introduce a source from underneath to provide a backward going evanescent field which would sum with the forward going evanescent field to produce a flatter PR intensity profile, and hence improve the depth of field (Fig. 2.6). As simultaneous front and backside illumination of the system is not practical (due to the general requirement of a silicon substrate which is strongly absorbing at patterning wavelengths) a resonant underlayer must be employed to build up the incoming energy from the top and re-emit it back into the PR. There are two types of suitable resonators available: dielectric, and surface-state resonators.

![Resonant underlayer intensity profiles](image)

**Figure 2.6:** Resonant underlayer intensity profiles. Blue dashed line - prism derived evanescent fields. Blue dotted line - resonator derived evanescent fields. Black solid line - Incident field in the prism, and the sum of the fields within the PR. (a) An under-resonant system with an asymmetric field profile which is underexposed at the bottom. (b) An optimally resonant system with a symmetric field profile. (c) An over-resonant system with an asymmetric field profile which is overexposed at the bottom. Note: in (a), (b), and (c) the resonator is a plasmonic resonator with $n < NA$ and $\kappa >> 0$ thus the exponential decay within this layer.
2.5.1 Dielectric resonators

Dielectric resonators (DR) are composed of a low-\(n\)|high-\(n\)|low-\(n\) type slab waveguide structure, where low is defined as less than the NA and high greater than the NA (Fig. 2.7). Energy is able to couple via FTIR through the top low-\(n\) (PR) layer \(n_1\) into the high-\(n\) layer \(n_2\) where it becomes propagating. The bottom low-\(n\) layer \(n_3\) is required to confine (via TIR) the fields to the high-\(n\) waveguiding layer and thus allowing energy to build up.

The condition for waveguiding occurs when the waves inside the high-\(n\) layer are in phase, that is, the round trip phase change is an integer multiple of \(2\pi\) \cite{1}. Implicit in this is the existence of waveguide modes which occur at angles (NAs) which satisfy the phase criteria, as well as the existence of a cutoff angle (NA) below which the phase criteria cannot possibly be satisfied \cite{1}. These two facts together mean a resonant thickness high-\(n\) layer can be designed to operate at any NA between \(n_{PR}\) \((n_2)\) and \(n_{high}\) \((n_2)\).

As the waves inside the high-\(n\) layer are confined by TIR, evanescent fields are present in both the bounding low-\(n\) layers. The thicknesses of these layers and the presence of any loss in them must be carefully balanced to allow the right amount of energy to build up in the resonator. Too much energy and the PR will be over exposed at the base; too little and there won’t be enough exposure at the base to flatten the intensity profile (Fig. 2.6). The decay characteristics of the evanescent field are entirely determined by the refractive index of the medium they are in and the NA producing them (Eqs. (2.18) and (2.19)). For this reason within the PR the forward and backward going evanescent fields will be the exact mirror image provided their source intensities are the same, and all the layers are linear and isotropic (of which all are in this thesis).

Another benefit of DRs is the fact they can resonate for both TE and TM polarizations. This is an important fact from a photolithography stand point where TE is the preferred polarization as it allows theoretically perfect interference minima at high NAs, whilst TM (although still usable) has a DC component preventing it from producing perfect interference minima except under normal or tangential illumination \cite{50}. This can be understood when one considers the dot product of the \(E\) vectors in the intensity equation (Eq. (2.4)). For TE polarization \(E_1\) and \(E_2\) are parallel thus perfect interference minima may result, while for TM polarization the value of the dot product ranges from 1 to 0 depending on the angle of incidence, thus the cosine term will be less than the constant (DC) component. The presence of a DC component
2.5. RESONANT UNDERLAYERS IN THE ULTRA-HIGH NA REGIME

Figure 2.7: Dielectric resonator schematic. Light (TE or TM) incident from \( n_0 \) couples evanescently into the dielectric resonator \((n_1|n_2|n_3)\). Energy builds up in layer \( n_2 \) where it is confined by TIR at its top and bottom interfaces. The forward and backward going evanescent fields in the red PR layer \((n_1)\) sum to a flatter intensity profile (dotted line), therefore improving the depth of field. Notes: \( n_0 \) and \( n_2 \) are greater than the NA. \( n_1 \) and \( n_3 \) are less than the NA.

reduces the fringe visibility and produces a background flood exposure, both of which reduce the process window making TM the less preferred polarization.

2.5.2 Surface state resonators

When a TM polarized wave of light is incident upon a surface with mobile charge carriers the electric field component can create a collective charge oscillation (Fig. 2.8). The charge oscillations are bound to the interface and emit evanescent fields into both bounding media. Once again these evanescent fields can be used to flatten the PR intensity profile and improve the depth of field in evanescent-field photolithography. Light incident on a surface with mobile charge carriers acts as a sinusoidally driven oscillator system with the charges suffering both a restoring force, and a damping force. As such, there is a resonance condition \((k\text{ vector (momentum) matching})\) that must be met for energy to couple in effectively.

The availability of mobile charge carriers is due to surface effects. These occur at the edges of materials where the charge carriers occupy an asymmetric field environment e.g. electrons at the surface of a metal only see the metal crystal field from one side and thus have greater mobility. For this reason the properties of surface state excitations often are vastly different from that of the bulk material. Surface polariton states
2.5. RESONANT UNDERLAYERS IN THE ULTRA-HIGH NA REGIME

Figure 2.8: Surface state resonator schematic. Light (TM only) incident from $n_0$ couples evanescently into the surface state resonator ($n_1|n_2$). Energy builds up at the interface where it is confined to the surface as a polariton (sinusoid along the $n_1|n_2$ interface). The forward and backward going evanescent fields in the red PR layer ($n_1$) sum to a flatter intensity profile (dotted line), therefore improving the depth of field. Notes: $n_0$ is greater than the NA. $n_1$ is less than the NA. $n_2$ has a negative real part of its dielectric constant.

occur when the exciting light couples to the surface wave field resulting in a hybrid field quasiparticle (a polariton). The polariton is strongly bound to the surface and will travel along it until it is absorbed into the surface or reradiated off the surface. In this research two types of surface polariton waves are employed, surface plasmon polaritons (SPP), and surface exciton polaritons (SEP). In SPPs the charge oscillations are composed of free electrons, whilst those in SEPs are bound charge pairs called excitons.

Despite the drawback of operating only with TM polarization, surface state resonators offer the benefits of a simpler resonator film stack and potentially greater field enhancements (Section 6.7). Both excitation types can be employed in planar (Chapters 4 and 5) and/or grating configurations (Chapter 6). Although SEP resonators remain largely unexploited, SPP resonators have found widespread usage in biosensors and lab on chip applications [86, 87].

Both dielectric and surface state resonators are investigated and employed in this research. Each has its own strengths which lends it to particular uses. Now that the concepts of interference lithography, evanescent fields, and resonant underlayers have been discussed, a further interference lithography configuration will now be introduced.
2.6 Near-field grating coupled interference lithography schemes

Up until this point IL has been discussed in terms of maskless prism (or open air) coupled interference systems which are the forms employed in Chapters 3 to 5. If the distance between the diffraction grating and PR in the Mach-Zehnder configuration (Fig. 2.3(b)) is reduced, another configuration becomes possible, that of near-field grating coupled interference lithography. The enhancement and optimization of near-field grating coupled interference lithography is investigated in Chapter 6. In this configuration the non-zero diffracted orders are all evanescent; at normal incidence the $0^{th}$ order is not capable of interfering with itself (along the interface direction) thus the interference produced results from the interference of evanescent diffraction orders. Patterning utilizing evanescent fields requires the distance between the grating and PR to be (much) less than $\lambda$, thus the term near-field. The close proximity to the PR and the lack of a projection lens means that these systems can be considered a form of contact photolithography. A brief background will now be given for this configuration beginning with contact photolithography, then progressing on to the two forms most relevant to this research evanescent near-field optical lithography (ENFOL) and evanescent interference lithography (EIL).

2.6.1 Contact photolithography as a near field lithography

Contact photolithography was the first method of ‘high volume’ processing employed in the semiconductor industry. It consisted of an exposure through a mask brought into close proximity to the PR (Fig. 2.9(a)). Exposure through such a setup results in a 1:1 pattern replication, but the requirement for close proximity led to rapid increases in patterning defects due to pick-ups of particulate matter, mask damage, and mask flatness problems [10]. For this reason the industry soon shifted to projection photolithography methods which had greatly improved yields. The rapid development of projection lithography over the next few decades led to the effective shelving of contact lithography. Perceived roadblocks in the continuing shrinkage of projection lithography however meant contact photolithography employing conformable masks [88] was resurrected in an updated form in the early 2000s.

The smaller patterns in modern day photolithography methods represent higher spatial frequencies (corresponding to higher diffraction orders). The capturing of these
spatial frequencies determines the patterning fidelity a system is capable of. For instance if the high spatial frequencies have been removed the fine details of the mask will not be resolved in the PR. This is an important detail as any system employing a projection lens is only capable of collecting a certain number of the diffraction orders from the mask. As such these systems are termed diffraction limited systems [Fig. 2.9(b)], where the pattern resolution is limited by the diffraction capturing ability of the (traditional) lens. This however is not the case with contact photolithography where all orders capable of reaching the PR can be used for patterning, thus contact photolithography has a superior theoretical resolution compared to projection photolithography.

To capture these spatial frequencies the mask has to be much closer to the imaging stack to allow the evanescent orders to pattern the PR. Several techniques have been developed exploiting this configuration including embedded amplitude masks [89, 90], light coupling masks [91, 92], and ENFOL [73, 93, 75]. As ENFOL is the direct precursor to the systems investigated in Chapter 6 the discussion will now focus on that.

### 2.6.2 ENFOL and enhanced ENFOL

ENFOL is a photolithographic patterning technique where a conformable subwavelength amplitude mask is brought into very close ($d << \lambda$) contact with the PR (Fig. 2.10). This very close proximity to the PR allows for patterning by evanescent fields which is an absolute necessity for (deep) subwavelength mask features. This
Figure 2.10: ENFOL - (a) An ENFOL exposure system schematic [74]. ENFOL example - (b) SEM image of a 64 nm a-Si mask pattern and (c) the corresponding 1:1 resist pattern produced via a trilayer resist process [94].

method has been employed to produce 64 nm full pitch lines with a 365 nm exposure wavelength, corresponding to a $\lambda/5.7$ feature size [94], while modelling suggests this may be further reduced to a half pitch of $\lambda/20$ [93]. The major strength of the ENFOL process is that it theoretically has no resolution limit, the caveat being that the fields must still be capable of reaching the PR through the mask. This becomes more and more of an issue as the mask feature sizes decrease with the consequent evanescent fields decaying faster and the transmission into the PR decreasing. The publication of papers by Ebbesen et al. [95] and Pendry [96] offered possible routes around this.

The effect of extraordinary optical transmission of TM polarized light through subwavelength hole arrays was first described by Ebbesen et al. in 1999. This paper detailed a massive enhancement in transmission through a metal sub-wavelength hole array. An early theory by Bethe [97] suggested the transmission through a sub-wavelength aperture should scale by $r/\lambda^4$, where $r$ is the aperture radius and $\lambda$ the wavelength, thus it was expected the transmission would be very small. The increase in transmission was attributed to plasmonic field enhancements. This effect allows ENFOL to be employed for deep sub-wavelength patterning where the field enhancement reduces exposure times.

In 2000 Pendry published a paper detailing the concept of a ‘Perfect’ lens (generally termed a superlens). Superlenses operate on the principles of negative refraction and plasmonic enhancement of evanescent fields. Negative refractive indices occur when both $\varepsilon$ and $\mu$ are simultaneously negative [98]. Negative refraction allows the refocu-

$^3$Although one would think if $n = \sqrt{\varepsilon\mu}$ we can just take the positive root, this is not the case, as both the wavevector and hence the phase velocity are reversed. Thus the negative root is taken to align the sign of $n$ with $k$ such that $k = 2\pi n / \lambda$ remains true.
sing of diverging rays to a point behind the planar lens. As no naturally occurring materials have a broadband negative refractive index, Pendry [96] suggested the use of silver which has a negative real refractive index at optical frequencies. Pendry recognized that at subwavelength scales one can neglect the $\mu$ component provided TM polarization is employed. When silver has a negative $\varepsilon$, $\mu$ is not negative thus the use of subwavelength structures allows one to bypass the double negative constraint. Interestingly this negative $\varepsilon$ is also a necessity for plasmon resonance, thus the fields transmitting through the silver planar lens are also enhanced by plasmon resonance which helps overcome the absorption within the lens.

Superlenses have been placed beneath an ENFOL type mask configuration to enhance the fields and transfer them to a PR layer. Superlens systems are limited by the absorption in the lens, but are none the less capable of patterning very small structures with experimentally achieved resolutions of $\approx \lambda/6$ [99, 100, 101, 102]. Both ENFOL and superlens enhanced ENFOL are capable of patterning very small features, but are still fundamentally 1:1 imaging systems. They were however the inspiration for evanescent interference lithography (EIL) [103], a method of employing interference of plasmonic fields to produce a pitch division effect in the PR.

EIL exploits the plasmonic response of metals to interfere evanescent diffraction orders which expose the PR (Fig. 2.11). Depending on the pitch and duty cycle of the grating these may occur at the corners of the gratings [103, 104] as well as under the solid part of the grating [104]. The benefit of these systems over standard ENFOL systems is that the interference effect produces a single exposure pitch division of the grating pitch without the need for any multiple patterning techniques.

The systems designed and optimized in Chapter 6 are a variant upon EIL consisting of a grating|filter|resonator|PR|resonator structure. Rather than the interference occurring at the grating, the system is designed to resonantly couple energy from a particular diffracted order into the resonators where interference occurs thus producing an interference pattern within the PR. As previously discussed a bottom resonator is also employed in this structure to improve the DOF. The top resonator ideally should allow for improved experimental performance as the interfering fields are in intimate contact with the PR, thus reducing any intensity variations due to mask/PR separation variations.
2.7. Summary

In this chapter the concepts of 2-beam interference and IL have been introduced. As a method IL is employed for producing extremely regular larger area gratings for applications such as photolithography testing, spectrometer diffracting elements, polymer growth templates etc. The different NA regimes were introduced highlighting the evanescent fields present within the PR in the ultra high-NA regime. These fields severely impact the DOF, for which resonant underlayers are employed to improve. The chapter closes with a description of contact photolithography and its application for interference of evanescent diffraction orders.

This was only a brief introduction to what is a highly refined technique. For an excellent and in depth coverage of non-evanescent IL the reader is suggested towards Walsh’s PhD thesis [105], and for evanescent IL and ENFOL the works of Blaikie, McNabb, Mehrorta, and Lowrey [103, 74, 78, 106, 75, 77].

Figure 2.11: EIL example - An incident TM polarized wave excites surface plasmon polaritons on the grating underside. Note the effective pitch division of the high intensity areas on the grating underside compared to the mask aperture spacing [104].
Chapter 3

Herpin effective media underlayers

3.1 Prologue

In the previous chapter the idea of resonant underlayers to improve the performance of ultra-high NA IL was introduced, where of particular importance to this chapter are dielectric resonators. Dielectric resonators have a fundamental limitation in that the field must be propagating in the high-$n$ layer, otherwise the entire structure will have evanescent fields and hence not resonate. For high resolution (high NA) this requires high index materials, so the effect is limited by the availability of high index dielectrics at the wavelength of interest. In this chapter we investigate a possible way around this, known as Herpin effective media, which is an effective medium method long employed in the antireflection coating industry where it is used to produce (effective) refractive indices that are not naturally or easily available. It is also known to be able to produce theoretically arbitrary, and arbitrarily high, effective refractive indices, thus it was seen as a possible means of extending dielectric resonant underlayers to very high NAs. Here we test the hypothesis that Herpin effective medium underlayers can be used as resonators to extend evanescent interference lithography to arbitrarily high NAs.

The Herpin effective media theory derives from the matrix treatment of thin-film optics, consequently this chapter begins with a brief introduction followed by a build-up of the transfer matrix method of thin-film optics modelling. The formalism for this will be developed and act as a lead-in to Herpin effective media theory. The use and limitations of Herpin effective media in ultra-high resolution lithography is explored, after which we investigate the possibility of other alternative resonator systems.
3.2 Introduction

The dielectric resonators in the previous chapter have a natural upper NA limit where the NA is equal to the real refractive index of the $n_{\text{high}}$ layer. If this is exceeded all the fields in the resonator system will be evanescent, thus it will no longer be able to build up energy. With this in mind, what are the highest NAs available? At 405 nm\textsuperscript{1} the highest refractive indices of low absorbing ($\kappa < 0.05$) materials tend to be wide band gap semiconductors and/or transition metal oxides such as GaN, diamond, ITO, HfO$_2$, SnO$_2$, etc. all with real refractive indices less than 2.6 \cite{79, 107, 108, 109, 110}. Higher NAs also lead to lower tolerances of absorption, as higher NAs equate to longer path lengths within the propagating layer. So how can the available NAs be increased for dielectric resonators considering $n \approx 2.6$ seems to be the physical limit? A possible answer lies in Herpin effective media. Herpin effective media is a concept commonly employed in the antireflection coating industry to produce multilayer combinations with an effective refractive index that is not available with existing materials \cite{111}. As it is generally employed, it is used to produce any (effective) refractive index between the refractive indices of the two media used to produce the effective medium. There is however a commonly known side-effect of this technique, which is, that it theoretically allows arbitrary effective refractive indices to be produced outside these bounds \cite{112, 111, 113}. The availability of these arbitrary effective refractive indices presents the possibility of replacing the NA limiting $n_{\text{high}}$ layer in a resonator with a Herpin effective medium trilayer or multilayer to access higher NAs. To fully appreciate Herpin effective media we must first look at the transfer matrix methods (TMM) treatment of thin-film optics, from which Herpin media are a natural result. The transfer matrix method is heavily employed in this thesis to model thin film structures. The following section details the mathematical foundations for TMM which was used for modelling the Herpin relations. To model more complex structures a custom *Birefringent Thin Films and Polarizing Elements* toolbox for Matlab was employed for the calculations \cite{113}. This toolbox is built upon the same TMM foundations but uses a more general 4x4 approach called the Berreman calculus which is designed for birefringent materials and the full range of polarizations \cite{114, 113}. In the case of isotropic materials and linear polarization the Berreman calculus reduces to the standard Heavens \cite{115} and Abelès \cite{116} matrices, and thus is perfectly suited for this work.

\textsuperscript{1}Unless otherwise stated all values in this thesis are at $\lambda = 405$ nm.
3.3 The transfer matrix method of thin-film optics

The transfer matrix method is a method of simulating the optical properties of thin-film stacks\(^2\). Here we paraphrase Hodgkinson’s derivation \([113]\). In Figure 3.1 we see a schematic for this method showing, the film layers, the 4 basis vectors, and the general progression of TMM calculations. The reader is advised to refer back to this figure as they progress through this section to better understand how the TMM is constructed.

\[ \begin{align*}
\text{Cover} & \quad \text{Medium} & \quad \text{Substrate} \\
\vec{a}_{\text{TE}}^{-} & \quad \vec{a}_{\text{TE}}^{-} & \quad \vec{a}_{\text{s}} \\
\vec{a}_{\text{TM}}^{-} & \quad \vec{a}_{\text{TM}}^{-} & \quad \vec{m}_{\text{s}} = \hat{F}_{\text{s}} \vec{a}_{\text{s}} \\
\vec{a}_{\text{C}}^{-} & \quad \vec{m}_{\text{M}} = \hat{F}_{\text{M}} \vec{a}_{\text{M}}(x) \\
\vec{m}_{\text{C}}^{-} = \hat{F}_{\text{C}}^{-1} \vec{m}_{\text{C}} & \quad \vec{a}_{\text{M}}(x-d) = \hat{A}_{d} \vec{a}_{\text{M}}(x) \\
\end{align*} \]

**Figure 3.1:** Transfer matrix method schematic - Red and blue arrows \((a_{\text{TE,TM}}^{+, -})\) are the basis vectors of which any linearly polarized fields within the thin-film stack are comprised. \( \hat{F}_{\text{C,M,S}} \) are the field matrices which carry the \( E \) and \( H \) fields of the basis vectors in the cover, medium and substrate layers respectively. \( \vec{m}_{\text{C,M,S}} \) are the total actual fields given by \( \vec{m} = \hat{F} \vec{a} \). \( \hat{A}_{d} \) is the propagation matrix used to transfer the fields through a layer. The definitions of TE and TM are relative to the given axes with the field components given in Eq. (3.1) and the red and blue arrows indicating the \( k \) vector directions. The equations, from top to bottom \((1)-(8)\), show the progress of the calculations through the system.

When a linearly polarized wave is incident upon an isotropic layered system at any point generally four waves may be present, i.e. forwards and backwards going TM and TE waves depending on the polarization (Fig. 3.1). As any linear polarization can be represented by a combination of these waves, we use them as an orthogonal basis.

\(^2\)Thin refers to the path length within the layers being less than the coherence length of the light, thus interference effects are present.
vector set. These can be grouped into a 4x4 field matrix

\[
F = \begin{bmatrix}
E_{yTM}^+ & E_{yTM}^- & 0 & 0 \\
H_{yTM}^+ & H_{yTM}^- & 0 & 0 \\
0 & 0 & E_{zTE}^+ & E_{zTE}^- \\
0 & 0 & H_{yTE}^+ & H_{yTE}^-
\end{bmatrix},
\]

where the subscripts \(y\) and \(z\) indicate the field directions (parallel to any interfaces), TM and TE indicate the orthogonal basis vectors, and + and − indicate the direction of the waves. This matrix only contains the tangential \(y\) and \(z\) field components as these are the ones conserved across and interface, and \(x\) component can be derived from these components and the angle of incidence. Each column of \(F\) contains only the fields for its particular basis vector, thus the zeros. The field ratios (optical admittance) between the tangential \(E\) and \(H\) fields are given by

\[
\gamma_{TM} = \frac{H_z^+}{E_y^+} = -\frac{H_z^-}{E_y^-} = \frac{n}{(z_0 \cos \theta)}
\]

\[
\gamma_{TE} = \frac{H_y^+}{E_z^+} = -\frac{H_y^-}{E_z^-} = -\frac{n \cos \theta}{z_0},
\]

where \(n\) is the refractive index, \(\theta\) the angle of incidence, and \(z_0\) the impedance of free space\(^3\). The optical admittance is required for oblique incidence calculations; thus for clarity \(F\) is recast using the field ratios as

\[
\hat{F} = \begin{bmatrix}
1 & 1 & 0 & 0 \\
\gamma_{TM} & -\gamma_{TM} & 0 & 0 \\
0 & 0 & 1 & 1 \\
0 & 0 & \gamma_{TE} & -\gamma_{TE}
\end{bmatrix}.
\]

The field matrix however only tells us the relations between the fields, it doesn’t tell us the proportions of each of the basis vector fields present, i.e. do we have TE, TM, or a mixture of both. For this we use a coefficient vector

\[
\vec{a} = \begin{bmatrix}
a_{TM}^+ \\
a_{TM}^- \\
a_{TE}^+ \\
a_{TE}^-
\end{bmatrix},
\]

\(^3\)For the TM example using Ampère’s Law for propagation in isotropic media \(\nabla \times \mathbf{H} = -\varepsilon_0 \varepsilon \frac{\partial \mathbf{E}}{\partial t}\) and the plane, harmonic wave operator relations \((\frac{\partial}{\partial t} \to -i\omega \text{ and } \nabla \to ik)\) we obtain the tangential components \(k_x H_z = \varepsilon_0 \varepsilon \omega E_y\). Using \(k_z = nk_0 \cos \theta\) and the relations between \(k, c, n, \varepsilon, \varepsilon_0, \) and \(z_0\) we arrive at the TM tangential field ratio \(\frac{H_z}{E_y} = \frac{n}{z_0 \cos \theta}\). The same method using the Maxwell-Faraday equation provides the tangential field ratio for TE polarization.
3.3. THE TRANSFER MATRIX METHOD OF THIN-FILM OPTICS

which specifies the proportions of each basis vector. When \( \vec{a} \) is combined with \( \hat{F} \) the total actual fields (\( \vec{m} \)) are given by

\[
\vec{m} = \hat{F} \vec{a} = \begin{bmatrix}
a_{TM}^+ + a_{TM}^-
\gamma_{TM} a_{TM}^+ + \gamma_{TM} a_{TM}^-
a_{TE}^+ + a_{TE}^-
\gamma_{TE} a_{TE}^+ + \gamma_{TE} a_{TE}^-
\end{bmatrix}.
\]

(3.5)

Conversely the field coefficients can be obtained from the total field using the inverse of \( \vec{F} \).

\[
\vec{a} = \hat{F}^{-1} \vec{m}
\]

(3.6)

The field coefficients in \( \vec{a} \) are complex valued allowing the phase of the basis vectors to be kept track of. For this a phase matrix \( \hat{A}_d \) is employed to transfer the fields within a medium, where the phase thickness is given by \( \phi_{TM,TE} = 2\pi n \cos \theta / \lambda \).

\[
\hat{A}_d = \begin{bmatrix}
e^{-i\phi_{TM}} & 0 & 0 & 0
0 & e^{-i\phi_{TM}} & 0 & 0
0 & 0 & e^{-i\phi_{TE}} & 0
0 & 0 & 0 & e^{-i\phi_{TE}}
\end{bmatrix}
\]

(3.7)

\( \hat{A}_d \) is used to propagate the coefficient vector (and thus the fields) such that the propagation from point \( x \) to \( x - d \) is given by

\[
\vec{a}_{x-d} = \hat{A}_d \vec{a}_x.
\]

(3.8)

For the sake of understanding, a simple example will now be given of a three layer system consisting of the same cover, intermediary layer, and substrate (C|L|S) system shown in Fig. 3.1.

Starting from a point just under the intermediary layer inside the substrate we have the field coefficients \( \vec{a}_S \) (Fig. 3.1 (1)). To match the \( E \) and \( H \) fields we must convert \( \vec{a}_S \) to the full field components \( \vec{m}_S = \hat{F}_S \vec{a}_S \) (Fig. 3.1 (2)). As \( \vec{m} \) is continuous across the interface we can say \( \vec{m}_S = \vec{m}_M \) (Fig. 3.1 (3)). The field coefficients within layer M can be obtained by \( \vec{a}_M = \hat{F}_M^{-1} \vec{m}_M \) (Fig. 3.1 (4)), and transferred across layer M by the phase matrix i.e. \( \vec{a}_M(x - d) = \hat{A}_d \vec{a}_M(x) \) (Fig. 3.1 (5)). Now that we’re at the M|C interface we need to convert back to the full field (\( \vec{m}_M = \hat{F}_M \vec{a}_M(x - d) \)) (Fig. 3.1 (6)). The full field is continuous across the boundary, therefore \( \vec{m}_C = \vec{m}_M \) (Fig. 3.1 (7)) and the field coefficients in the cover can be obtained by \( \vec{a}_C = \hat{F}_C^{-1} \vec{m}_C \) (Fig. 3.1 (8)).
With this in mind a nomenclature can be established to make the discussion a little less cumbersome. The full transfer across the C|M|S system can be written as

\[ \vec{a}_C = \hat{F}_C^{-1} \hat{F}_M \hat{A}_d \hat{F}_M^{-1} \hat{F}_S \vec{a}_S. \]  (3.9)

The central part of Eq. (3.9) \((\hat{F}_M \hat{A}_d \hat{F}_M^{-1})\) is often termed the characteristic matrix of the layer and is designated \(\hat{M}_i\). If multiple layers are present the total characteristic matrix \((\hat{M})\) is simply the product of the constituent characteristic matrices i.e. \(\hat{M} = \hat{M}_1 \hat{M}_2 \hat{M}_3 ... \hat{M}_N\). Thus Eq. (3.9) can be rewritten as

\[ \vec{a}_C = \hat{F}_C^{-1} \hat{M} \hat{F}_S \vec{a}_S. \]  (3.10)

The matrix \(\hat{F}_C^{-1} \hat{M} \hat{F}_S\) describes the full transfer from the substrate interface to the cover interface, thus it is called the system matrix \((\hat{A})\).

\[ \vec{a}_C = \hat{A} \vec{a}_S. \]  (3.11)

From the system matrix the reflectance, transmittance, and various other optical quantities can be obtained or derived. The necessity of the coefficient matrices in Eq. (3.11) is to allow us to specify any combination of basis vectors as the input fields.

Of particular interest for investigating optical systems is the form of \(\hat{M}\) as this dictates the optical properties of the stack independent of the cover and substrate properties. For a single layer the characteristic matrix is given by

\[ \hat{M} = \hat{F} \hat{A}_d \hat{F}^{-1} \]  (3.12a)

\[ = \frac{1}{2} \begin{bmatrix} 1 & 1 & 0 & 0 \\ \gamma_{TM} & -\gamma_{TM} & 0 & 0 \\ 0 & 0 & 1 & 1 \\ 0 & 0 & \gamma_{TE} & -\gamma_{TE} \end{bmatrix} \times \begin{bmatrix} e^{-i\phi_{TM}} & 0 & 0 & 0 \\ 0 & e^{-i\phi_{TM}} & 0 & 0 \\ 0 & 0 & e^{-i\phi_{TE}} & 0 \\ 0 & 0 & 0 & e^{-i\phi_{TE}} \end{bmatrix} \begin{bmatrix} 1 & 1/\gamma_{TM} & 0 & 0 \\ 1 & -1/\gamma_{TM} & 0 & 0 \\ 0 & 0 & 1 & 1/\gamma_{TE} \\ 0 & 0 & 1 & -1/\gamma_{TE} \end{bmatrix}, \]  (3.12b)

\[ = \begin{bmatrix} \cos \phi_{TM} & -i\gamma_{TM}^{-1} \sin \phi_{TM} & 0 & 0 \\ -i\gamma_{TM} \sin \phi_{TM} & \cos \phi_{TM} & 0 & 0 \\ 0 & 0 & \cos \phi_{TE} & -i\gamma_{TE}^{-1} \sin \phi_{TE} \\ 0 & 0 & -i\gamma_{TE} \sin \phi_{TE} & \cos \phi_{TE} \end{bmatrix}. \]  (3.12c)
3.4 Herpin effective media

The operating principle behind Herpin effective media is that any symmetric three layer stack can be represented by a single layer with an equivalent effective phase thickness ($\Phi$) and effective refractive index ($N$) (Fig. 3.2). What this means in practice is that, for instance, if we required a quarter wave antireflection layer to have a refractive index of 1.73 which (for the purposes of demonstration) may not exist, we can replicate the effect by the use of a symmetric 3 layer stack of 2 materials which do exist, which when combined together have the same optical properties as the single layer it is designed to replicate.

A symmetric three layer stack has the form $A|B|A$, and a matrix representation (where $M_i$ is either $M_{TM}$ or $M_{TE}$ from Eq. (3.13))

$$\hat{M} = \hat{M}_A \hat{M}_B \hat{M}_A.$$  

The components of $\hat{M}$ are (with $\gamma_{A,B}$ being the $\gamma_{TM}$ or $\gamma_{TE}$ for the material $A$ or $B$)

$$M_{11} = M_{22} = \cos 2\phi_A \cos \phi_B - \frac{1}{2}(\gamma_B/\gamma_A + \gamma_A/\gamma_B) \sin 2\phi_A \sin \phi_B,$$

$$M_{12} = (-i/\gamma_A)[\sin 2\phi_A \cos \phi_B + \frac{1}{2}(\gamma_A/\gamma_B + \gamma_B/\gamma_A) \cos 2\phi_A \sin \phi_B$$

$$+ \frac{1}{2}(\gamma_A/\gamma_B - \gamma_B/\gamma_A) \sin \phi_B],$$

$$M_{21} = -i\gamma_A[\sin 2\phi_A \cos \phi_B + \frac{1}{2}(\gamma_A/\gamma_B + \gamma_B/\gamma_A) \cos 2\phi_A \sin \phi_B$$

$$- \frac{1}{2}(\gamma_A/\gamma_B - \gamma_B/\gamma_A) \sin \phi_B].$$

Which can further be represented as a partitioned matrix

$$\hat{M} = \begin{bmatrix} \hat{M}_{TM} & \cdot \\ \cdot & \hat{M}_{TE} \end{bmatrix},$$

containing the single characteristic matrices for both TE and TM polarizations. As both forward and backwards components of the TE and TM waves have the same angle of incidence the components of $\hat{M}$ can be redefined using the exponential definitions of sine and cosine. With these matrices we now have the tools to develop Herpin effective media concepts.
### 3.4. HERPIN EFFECTIVE MEDIA

#### Figure 3.2: Herpin effective media schematic - A Herpin effective medium is a symmetric trilayer structure ($A|B|A$) with an effective refractive index ($N$) and an effective phase thickness ($\Phi$). The structure is designed such that the Herpin (H) effective medium parameters $N$ and $\Phi$ match $n_M$ and $\phi_M$ respectively of a desired single layer material (S). The values of $N$ and $\Phi$ are dependent on interference effects within the trilayer, thus $N$ is generally not the average of $n_A$ and $n_B$, nor is $\Phi$ the sum of the phases ($2\phi_A + \phi_B$).

As the components carry the same relations to the single film characteristic matrix (i.e. $M_{11} = M_{22}$ and $M_{12} = M_{21}/\gamma_i^2$), $\hat{M}$ can be restated as

$$
\hat{M} = \begin{bmatrix}
\cos \Phi & \frac{-\gamma_i^2}{\Gamma_i} \sin \Phi \\
-i\Gamma_i \sin \Phi & \cos \Phi 
\end{bmatrix},
$$

(3.18)

where $\Phi$ is the effective phase thickness, and $\Gamma_i$ is the effective optical admittance for the polarization $i$. $\Phi$ can be obtained from

$$
\Phi = \cos^{-1}(M_{11}) = \cos^{-1}(M_{22}),
$$

(3.19)

and $\Gamma_i$ from

$$
\Gamma_i = \sqrt{\frac{M_{21}}{M_{12}}},
$$

(3.20)

To obtain the effective refractive index we first need to consider $\Gamma_{TE}$ as

$$
\Gamma_{TE} = -N \cos \theta/z_0,
$$

(3.21)

where $N$ is the effective refractive index. If we then redefine $n \cos \theta$ as $\alpha$, such that $n^2 = n^2(\cos^2 \theta + \sin^2 \theta) = \alpha^2 + NA^2$, then $\Gamma_{TE} = -\alpha z_0$, and the effective refractive index can obtained from

$$
N = \sqrt{\Gamma_{TE}^2 z_0^2 + NA^2},
$$

(3.22)

without considering what the effective $\theta$ is for the system. For the sake of verification we stick to the simpler TE case, although a similar process can be used for TM. Now we have established the formalism behind the transfer matrix method and Herpin effective media we can look at several of its properties.
3.4. HERPIN EFFECTIVE MEDIA

Following the example of Epstein [112] we look at the Herpin properties of an A|B|A structure in both the $n_{\text{low}}|n_{\text{high}}|n_{\text{low}}$ (L|H|L) and $n_{\text{high}}|n_{\text{low}}|n_{\text{high}}$ (H|L|H) configurations, with $n_{\text{low}} < n_{\text{high}}$. The layers A and B are composed of MgF$_2$ ($n_{\text{low}} = 1.38$) and ZnS ($n_{\text{high}} = 2.30$) depending on the configuration, with a total phase thickness of $\phi = 2\phi_A + \phi_B$. If $\phi$ is varied between 0 and $\pi$ the effective phase thickness ($\Phi$) varies essentially linearly up to a stop band region where it diverges (Fig. 3.3(a), note both L|H|L and H|L|H have the same values). Over the same range of $\phi$, $N$ varies a lot more with the impact of the stop band being far greater (Fig. 3.3(b)). The stop band occurs when $M_{11} \geq -1$ (Fig. 3.4); thus the inverse cosine ($\Phi = \cos^{-1} M_{11}$) used to obtain $\Phi$ becomes imaginary. As $\Phi$ becomes imaginary so does $N$, which is indicative of a highly reflecting layer combination (i.e. a stop band). Looking at the low $\phi$ values ($\phi < 2$) it can be seen how Herpin effective media can be employed to produce intermediate index layer combinations. This region is commonly employed in antireflection coatings where the single homogeneous layer can be replaced be a symmetric trilayer combination where its $\Phi$ differs very little from $\phi$. Near the stop bands however $N$ has some interesting properties, that is, it can have a value that is less than or greater than its constituent components. This ability to exceed the refractive index of the components, and its potential exploitation in dielectric resonant underlayers is the focus of this chapter.

The values in Fig. 3.3 can be used to construct a Herpin trilayer replacement for a single layer with a phase thickness of $\phi = 0.25\pi$, a refractive index of $n = 1.82$, with a characteristic matrix (Eq. (3.13)) of

\[
M_S = \begin{bmatrix}
0.7071 + 0.0000i & 0.0000 + 0.3885i \\
0.0000 + 1.2869i & 0.7071 + 0.0000i
\end{bmatrix}.
\]

The Herpin equivalent values at $2\phi_A + \phi_B = 0.25\pi$ are $\Phi = 0.2529\pi$, $N = 1.8214$, with a characteristic matrix (Eq. (3.18)) of

\[
M_H = \begin{bmatrix}
0.6876 + 0.0000i & 0.0000 + 0.3987i \\
0.0000 + 1.3225i & 0.6876 + 0.0000i
\end{bmatrix}.
\]

The discrepancy between the two sets of values is due to the divergence of $\Phi$ from $2\phi_A + \phi_B$ as is visible in Fig. 3.3(a). This difference can be reduced by reducing the difference between the refractive indices of the two Herpin constituent materials, this reduces the size of the band gap and thus the phase divergence. From a manufacturing standpoint however, one cannot (simply) arbitrarily change the refractive indices as needed, consequently the physical phase thickness ($2\phi_A + \phi_B$) is often changed to match
3.4. HERPIN EFFECTIVE MEDIA

Figure 3.3: Herpin effective media characteristics. \( n_A = 1.38, n_B = 2.30, 2\phi_A/\phi_B = 1 \) and \( NA = 0 \) - (a) \( \phi \) vs. \( \Phi \). \( \Phi \) increases approximate linearly with \( \phi \) until a stop band (grey band) is approached. Both \( L|H|L \) and \( H|L|H \) have the same values. Black dashed line indicates a 1:1 phase relation. (b) \( \phi \) vs. \( N \). \( N \) changes slowly at low \( \phi \) but changes rapidly nearing the stop bands. Near the stop bands we see regions where \( N \) is less than and greater than the constituent refractive indices. Note the half-wave hole discontinuity at \( 2\pi \).

\( \Phi \) to that of the desired \( \phi \). This highlights an important point on the usage of Herpin effective media, that is, it is generally applied as a first design in the full knowledge that further refinements are needed. For the remaining uses of Herpin effective media in this chapter further simple refinements will be employed.

In traditional ARC applications Herpin media are used as a ‘straight’ replacement for a quarter-wave layer that is not materially feasible, or to simplify calculations by replacing larger multilayered structures with a far smaller number of Herpin layers. The key requirement for this is that the difference between \( \phi \) and \( \Phi \) is very small. If this is the case then a Herpin trilayer can ‘simply’ be substituted for a single layer material with a refractive index that is not available. If the difference is significant certain parameters can be altered to reduce this difference, notably the difference in refractive indices. Naturally if the two materials have similar refractive indices the medium will act more like a true homogeneous single layer, and the divergence near the stop bands will be reduced. If \( N \) is not suitable for the required \( \Phi \), there are three options available which minimally impact the effective phase [112, 117]. Firstly if the desired effective index is outside the LHL curve a HLH combination can be used (Fig. 3.3(b)), particularly if one requires lower refractive indices. Secondly \( N \) can be
Figure 3.4: $\phi$ vs. $M_{11}$. Stop band locations occur when $M_{11} = -1$. A secondary stop band known as a half-wave hole occurs at $M_{11} = 1$. Both $L|H|L$ and $H|L|H$ have the same values. Note: model parameters are the same as in Fig. 3.3.

‘fine tuned’ by altering the relative thicknesses of the two materials ($2\phi_A/\phi_B$), this does however exacerbate the effect of the half-wave hole (discussion of follow). Thirdly, the $n_A/n_B$ ratio may be increased, this increases the rate of change of $N$ before the first stop band and increases the separation of the LHL and HLH curves between the first and second stop bands. The draw back of increasing the $n_A/n_B$ ratio is that it opens the stop bands further which also increases the divergence of $\Phi$ from $\phi_{total}$.

Notable in the effective refractive index plot (Fig. 3.3(b)) is the discontinuity which occurs at $\phi_{total} = 2\pi$. The source of this discontinuity is the value of $M_{11}$ being equal to one (Fig. 3.4) and thus $\Phi = 0$. This means $M_{12}$ is equal to zero, as is the denominator in Eq. (3.20), thus the discontinuity occurring in $N$. If the phase ratio $2\phi_A/\phi_B$ isn’t equal to 1, a similar discontinuity opens up in the $\phi$ vs. $\Phi$ plot at the same location. This effect is well known in the thin-film coating industry and is sometimes referred to as a “half-wave hole”. This effect is important if one is producing a broadband transmission coating where at a particular angle or wavelength the effect of the half-wave hole will be to produce a peak in reflectance, thus limiting the performance of the coating and placing tight manufacturing constraints on any coatings operating in this region.

Now that the theory and characteristics of Herpin effective media have been discussed we turn towards testing the hypothesis that Herpin replacement layers can be employed as resonant underlayers for ultra high-NA lithography.
3.4. HERPIN EFFECTIVE MEDIA

3.4.1 “Traditional” Herpin resonators ($N < n_{low}$)

The minimal difference between $\Phi$ and $\phi$ as well as the large areas of slow changing $N$ (Fig. 3.3) suggest that provided Herpin effective media behave similarly at highly oblique incident angles, and the desired phase thickness is not within a stop band, Herpin replacement resonant underlayers should be possible. To prove this we attempt to replicate the performance of an Al$_2$O$_3$ single layer resonant underlayer with a Herpin trilayer resonant underlayer. Table 3.1 at the end of this subsection serves as a comparison between the two underlayers for a prism coupled evanescent IL system.

Traditional Herpin replacement layers are comprised only of materials supporting propagating fields, consequently we need only attempt to match the properties of the propagating layer within the single layer resonator which is the Al$_2$O$_3$ layer. The Al$_2$O$_3$ layer has a thickness of 141 nm, a refractive index of 1.7854, and a design NA of 1.7. These values equate to a phase thickness of $\phi = 0.3798\pi$. The phase condition for resonance (waveguiding) in a dielectric waveguide is a round trip phase change of $2\pi m$, hence one would expect the phase thickness to be a fairly ‘nice’ value such as $\phi = \pi/2$ or $\pi$ (assuming $\pi$ phase changes upon reflection); this is not the case as the system must be optimally off resonance to produce a symmetric intensity profile within the PR. The value of ‘optimally off’ varies depending on the cladding materials, but one can get an idea for it considering the phase thickness of the Al$_2$O$_3$ single layer is $\phi = 0.3798\pi$. Now that we have a design NA, $\phi$, and $n$ we can construct a Herpin replacement layer.

The requirement for propagating fields means the Herpin layers $n_{low}$ and $n_{high}$ must be greater than the NA. After several refinements of the Herpin parameters, fictitious refractive index values of $n_{low} = 1.75$ and $n_{high} = 1.83$ were arrived at. Fictitious materials are necessary for the Herpin resonator layers here as they allow $N$ to be altered to match the refractive index of Al$_2$O$_3$ without appreciably shifting $\Phi$. The Herpin phase relationships for this material combination (Fig. 3.5(a)) shows that once again $\Phi$ is approximately equal to $\phi$ across all physical thicknesses with the exception of the stop bands and adjacent areas. At the design phase thickness of $\phi = 0.3798\pi$ the effective phase thickness is $\Phi = 0.3792\pi$, thus the phase thickness matching criteria has been successfully achieved.

The Herpin effective refractive index relationships are notably different to the low NA (Fig. 3.3(b)) case in that the lines appear compressed. This is due to the effective refractive index being dependent upon the NA as well as the particular material combination. As the effective index is given by $N = \sqrt{\Gamma^2 z_0^2 + NA^2}$, the value for $N$...
3.4. HERPIN EFFECTIVE MEDIA

Figure 3.5: Herpin effective media characteristics - Herpin replacement resonator. $n_A = 1.75$, $n_B = 1.83$, $2\phi_A/\phi_B = 1$ and $NA = 1.7$ - (a) - $\phi$ vs. $\Phi$. This plot is approximately the same as the low NA example (Fig. 3.3(a)). Note: black dashed line indicates $\phi = \Phi$, and the grey regions bounded by red and green dashed lines indicate the location of stop bands. (b) - $\phi$ vs. $N$. This plot appears compressed compared to the low NA example (Fig. 3.3(b)). This is due to the influence of the NA on the effective refractive index. Note: black dashed lines indicate values of $n_{low}$ and $n_{high}$.

will increase as the NA increases; this also effectively places a limit on the minimum value of $N$ for a particular NA. At the design phase thickness of $\phi = 0.3798\pi$ the effective refractive index is $N = 1.7826$ which is very close to the design refractive index ($n_{Al_2O_3} = 1.7854$), thus the refractive index matching criteria has also been successfully achieved.

Considering both $\phi \approx \Phi$ and $n_{Al_2O_3} \approx N$, the characteristic matrices of the $Al_2O_3$ layer ($M_S$) and the Herpin replacement layers ($M_H$) should also be approximately the same, as is indeed the case with

$$M_S = \begin{bmatrix} 0.37461 & 0 + 1.6999i \\ 0 + 0.50573i & 0.37461 \end{bmatrix} \quad \text{and} \quad M_H = \begin{bmatrix} 0.37053 & 0 + 16664i \\ 0 + 0.51771i & 0.37503 \end{bmatrix}.$$ (3.25)

The discrepancy between the two characteristic matrices is due to the divergence of $\Phi$ from $\phi$. Although the characteristic matrices are very well matched they could be further refined using an iterative optimization process using either materials indices and/or thicknesses, for the sake of demonstration however we have only adjusted the refractive indices to closely approximate the desired refractive index, and reduced the physical phase thickness by 1% to better match $\Phi$ to the desired phase thickness.
### Table 3.1: Standard resonant underlayer vs. Herpin replacement layer resonant underlayer.

Herpin effective media methods have been employed to match the properties of the Al₂O₃ layer to those of the Herpin trilayer at the design NA of 1.7 and TE polarization. Both \( n_{\text{high}} \) and the phase thicknesses have been well matched. The 1D intensity trace shows an intensity profile within the PR (red layers) in both cases. Resonator intensity profiles (yellow layers) are similar with a slightly greater intensity in the Herpin case. The other layers are as follows: dark blue - LAF2, light blue - SiO₂, and dark grey - Si. The 2D IL intensity profile has a very similar intensity distribution with a symmetric intensity profile evident in the PR layer (bounded by blue lines).

4Refractive index sources - LAF2 glass [118], PR - AZMIR701 [119], Al₂O₃ [120], SiO₂ [121], Si [122]
3.4. HERPIN EFFECTIVE MEDIA

Considering the characteristic matrices are approximately the same, so should the 1D and 2D intensity traces (Table 3.1). In the 1D intensity trace both stacks show the desired symmetric intensity profile with the PR layer. The Herpin resonator however has a noticeably greater intensity which is due to the resonator maximum intensity occurring slightly further beneath the PR base. Ideally a Herpin replacement layer should have the same optical effect as a single layer of the same refractive index and phase thickness; if this is indeed the case then the intensity trace in the bounding layers of the resonators should also be the same, as is visible in the 1D trace plots. Likewise the 2D intensity traces in an interference lithography configuration also show an approximately identical intensity distribution with in the PR.

3.4.2 Extreme Herpin resonators - one evanescent material

\((n_{\text{low}} < NA < n_{\text{high}})\)

In Figs. 3.3(b) and 3.5(b) it was shown that for particular refractive index and thickness combinations \(N\) can be less than or greater than the constituent refractive indices \((n_{\text{low}} \text{ and } n_{\text{high}})\). This raises the possibility of utilizing NAs greater than are physically obtainable with a real single layer, but suggests the question of what happens to Herpin effective media when one or more of the layers contain evanescent fields? To investigate this a comparison between a standard Al\(_2\)O\(_3\) resonant underlayer and a Herpin trilayer resonant underlayer is produced with details given in Table 3.2.

The model used for this comparison employs a Herpin trilayer in the HLH configuration with \(n_{\text{low}} = 1.6 \text{ and } n_{\text{high}} = 2.3\); thus for the design NA of 1.7 the fields are evanescent within the \(n_{\text{low}}\) layer and propagating within the \(n_{\text{high}}\) layer. Previous models had been constructed by varying \(\phi\) without needing to consider the actual physical thickness (\(d\)) this implies, the presence of an evanescent layer however prevents this method from being used as the evanescent layer produces a complex phase thickness and consequently a complex physical thickness which has no real meaning (to the best of the author’s understanding). Instead the model takes a step back and deals with the real-valued physical thicknesses and uses their associated phases. This allows \(N\) to be tailored by altering the physical thickness ratio \(2d_H/d_L = 0.3815\) and thus the ratio of phase thicknesses. Via this a model was able to be constructed which matched very well \(n\) to \(N\) as well as \(\phi\) to \(\Phi\).

Comparisons of the 1D and 2D intensity traces in Table 3.2 show the desired symmetric intensity profiles within the resist in both cases. The intensity profile within the
### 3.4. HERPIN EFFECTIVE MEDIA

<table>
<thead>
<tr>
<th>NA</th>
<th>Standard Resonant Underlayer</th>
<th>Herpin Replacement Underlayer</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>LAF2 glass (n=1.773, d=bulk)</td>
<td>LAF2 glass (n=1.773, d=bulk)</td>
</tr>
<tr>
<td></td>
<td>PR (n=1.68+0.03i, d=200 nm)</td>
<td>PR (n=1.68+0.03i, d=200 nm)</td>
</tr>
<tr>
<td></td>
<td>Al(_2)O(_3) (n=1.7854, d=141 nm)</td>
<td>Fictitious (n=2.3, d=14.9 nm)</td>
</tr>
<tr>
<td></td>
<td>SiO(_2) (n=1.47, d=300 nm)</td>
<td>Fictitious (n=2.3, d=14.9 nm)</td>
</tr>
<tr>
<td></td>
<td>Si (n=5.44+0.34i, d=bulk)</td>
<td>SiO(_2) (n=1.47, d=300 nm)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Si (n=5.44+0.34i, d=bulk)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>(n_{\text{high}})</th>
<th>Al(_2)O(_3) (n=1.7854)</th>
<th>Fictitious trilayer (N = 1.7854)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Phase Thickness</td>
<td>Al(_2)O(_3) layer: (\phi = 0.3798\pi)</td>
<td>Fictitious trilayer: (\Phi = 0.3798\pi)</td>
</tr>
</tbody>
</table>

**Table 3.2:** Standard resonant underlayer vs. Herpin trilayer resonant underlayer - evanescent \(n_{\text{low}}\) layer. Herpin effective media matching of an Al\(_2\)O\(_3\) layer to a Herpin trilayer in the HLH configuration at a design NA of 1.7, \(n_{\text{low}} = 1.6, n_{\text{high}} = 2.3\), and TE polarization. The 1D intensity trace shows a symmetric intensity profile within the PR (in red) in both cases. Resonator intensity profiles (in yellow) are clearly different with the Herpin layer appearing to act as two coupled resonators. The other layers are as follows: dark blue - LAF2, light blue - SiO\(_2\), and dark grey - Si. The 2D IL intensity profile again has a symmetric intensity profile evident in the PR layer (bounded by blue lines), and the drop off in intensity within the Herpin trilayer clearly evident.

---

5Ref: refractive index sources - LAF2 glass [118], PR - AZMIR701 [119], Al\(_2\)O\(_3\) [120], SiO\(_2\) [121], Si [122]
3.4. HERPIN EFFECTIVE MEDIA

resonators however are markedly different, with the Al₂O₃ appearing as a normal resonator while the Herpin resonator acts a pair of coupled resonators with peaks occurring in the \( n_{\text{high}} \) layers.

3.4.3 Extreme Herpin resonators - two evanescent materials

\( (NA < n_{\text{high}}) \)

As was previously discussed when near the stop bands Herpin effective media are theoretically able to produce effective refractive indices greater than the constituent material refractive indices. The models in the previous section were further employed to see if it was possible to match the refractive indices for the same Al₂O₃ layer \( (NA = 1.7, \ n_{\text{Al}_2\text{O}_3}, \ d = 141 \ \text{nm}) \) with that of a Herpin trilayer with both \( n_{\text{low}} \) and \( n_{\text{high}} \) less than the NA. These trials failed to find an even remotely close match even though the Herpin theory at Fig. 3.5 show that such supra-index solutions are possible. To understand why we must take a closer look at the equations that define the characteristic matrix components, specifically the \( M(1,1) \) component.

The single layer characteristic matrix \( M(1,1) \) component is given by

\[
M_S(1,1) = \cos \phi, \quad (3.26)
\]

where \( \phi = k \alpha d \) and \( \alpha = n \cos \theta = \sqrt{n^2 - NA^2} \). The crucial fact here is that the NA is less than \( n_{\text{Al}_2\text{O}_3} \), hence \( \theta, \alpha, \) and \( \phi \) are all real numbers, and propagating fields are present within this layer.

For the Herpin trilayer the \( M(1,1) \) component is given by

\[
M_H(1,1) = \cos 2\phi_A \cos \phi_B - \frac{1}{2} (\gamma_B/\gamma_A + \gamma_A/\gamma_B) \sin 2\phi_A \sin \phi_B, \quad (3.27)
\]

where \( A \) and \( B \) are the material designations (i.e. \( n_{\text{low}} \) and \( n_{\text{high}} \)), \( \phi \) is as previously described, and \( \gamma_A,B \) are the optical admittances for the sublayers which are given by (for TE polarization) \( \gamma_{A,B} = -n_{A,B} \cos \theta_{A,B}/z_0 = -\alpha_{A,B}/z_0 \). In this case both \( n_{\text{low}} \) and \( n_{\text{high}} \) are less than the NA, consequently \( \alpha, \theta, \gamma, \) and \( \phi \) are all complex. As both \( n_{\text{low}} \) and \( n_{\text{high}} \) layers are evanescent the real part of \( \cos \theta \) is equal to 0. As such both terms on the RHS of Eq. (3.27) have a real component equal to zero; thus the matching of \( \phi \) to \( \Phi \) is not possible for this type of system as the single layer has a real non-zero value phase thickness while the Herpin trilayer has a complex phase thickness with the real part equal to zero. For this reason Herpin replacement trilayers with all mediums evanescent are not suitable for use as a resonant underlayer.
3.5 Alternative underlayer systems

The failure of Herpin effective media methods for two evanescent layers, although disappointing, led to further (fruitful) investigations into other alternative resonant underlayer systems.

3.5. ALTERNATIVE UNDERLAYER SYSTEMS

The use of Herpin replacement layers is predicated on the ability to produce a symmetric trilayer stack of materials with an effective refractive index and effective phase thickness equivalent to some desired single layer. This method however breaks down when evanescent fields are present in all three layers due to the lack of phase propagation in evanescent fields. Although the use of the Herpin effective media method simplifies the calculations, in many respects it artificially constrains our scope to symmetric propagating systems. What if we bypass these limitations, and with the goal of a symmetric PR intensity profile (Fig. 3.6(a)), instead look at the full thickness and refractive index combinations available? The transfer matrix method formalism established at the start of this chapter proves to be the perfect tool for this task allowing us to efficiently explore the full parameter space. The optimal solutions, where the PR profile is symmetric, are locations where (due to the forward and backward going evanescent fields having the same decay rates) the intensity at the top and bottom of the PR are equal (i.e. $\Delta I_{PR} = I_{\text{top}} - I_{\text{bottom}} = 0$). Due to how $\Delta I_{PR}$ is defined over-resonant areas present (in Figs. 3.6(b) to 3.6(d)) as blue toned areas while under-resonant areas present as dark red toned areas.

As an example, if we begin with a simple Prism|PR|single layer|Substrate system, taking an NA of 1.8 so the system is in the desired ultra high-NA regime and use an $n = 2$ prism material to allow coupling of evanescent fields in the PR. As a resonant underlayer is required to act as a backwards going source of evanescent fields in the PR to produce a symmetric intensity profile, naturally we look for sources of resonance. In the case of TE (Fig. 3.6(b), $NA = 1.8$, $d_{\text{under.}} = 150$ nm, $d_{PR} = 100$ nm, and $n_{\text{sub.}} = 1.4696$) the only resonant systems available are slab waveguide modes. Multiple refractive indices satisfy the criteria for waveguiding, all of which have $\kappa \approx 0$. As the layer thickness is fixed, these different refractive indices represent increasing multiples of the round trip phase condition $2\pi m$ where the lowest resonant refractive index is indicative of the cutoff thickness. If the single layer thickness is altered the resonant refractive indices will shift accordingly.

Changing to TM polarization we see a much richer resonant refractive index space
Figure 3.6: Resonant underlayer parameter space sweep. (a) - PR intensity profiles. Subsequent plots in this figure plot the front PR interface intensity minus the back interface PR intensity. Positive values indicate an under-resonant system, negative values an over-resonant system. The black contour indicates a symmetric PR intensity profile. Common parameters: $NA = 1.8, n_{prism} = 2, n_{PR} = 1.6844 + 0.0307i, n_{sub} = 1.4696, d_{prism} = \text{bulk}, d_{sub} = \text{bulk}, d_{PR} = 100 \text{ nm}$. (b) - TE refractive index space - $d_{underlayer} = 150 \text{ nm}$. For TE polarization only dielectric slab waveguide resonances are available, consequently there are no high $\kappa$ peaks, thus the compressed y-axis. (c) - TM refractive index space - $d_{underlayer} = 30 \text{ nm}$. Dielectric slab waveguide resonances are again present along the x-axis. Significantly, surface state polariton resonances are also present in the upper left-hand corner. (d) - TM refractive index space - $d_{underlayer} = 150 \text{ nm}$. Waveguide modes are present in the same locations as the TE case (Fig. 3.6(b)), except with larger $\kappa$ values. For comparison the y-axis is reduced to remove the high-$\kappa$ surface state resonances. Note: different y axes in (b) & (d), and (c).
3.5. ALTERNATIVE UNDERLAYER SYSTEMS

(Fig. 3.6(c). $NA = 1.8$, $d_{\text{under.}} = 30$ nm, $d_{PR} = 100$ nm, and $n_{\text{sub.}} = 1.4696$). The very thin underlayer shifts the dielectric waveguide resonance to a high refractive index ($n \approx 4.6$). More noticeably at low $n$ and high $\kappa$ we have two additional resonance peaks due to surface state resonances. The larger resonance occurs at the PR|single layer interface, whilst the smaller resonance occurs at the single layer|substrate interface. Generally, if $n < 1$ and $\kappa >> n$ this resonance is termed plasmonic, or if $n > 1$ and $\kappa > n$ the resonance is termed excitonic, although this is just semantics with there being no clear line of demarcation seen in Fig. 3.6(c) [123]. The strength of the larger resonance is not dependent on the single layer thickness as the resonance is confined to the PR|single layer interface. Provided the $n_{\text{sub.}}$ is less than that of the single layer and the single layer is thin enough ($\approx \lambda/16$ nm) the smaller surface state resonance is present. As the fields from this resonance have to back traverse the single layer they produce a smaller area resonance peak.

If the underlayer thickness is increased for comparison to that of the TE examples (d=150 nm), for TM polarization (Fig. 3.6(d)) similar dielectric slab waveguide resonances are present, although at different refractive index values due to the different resonance criteria. Note the resonance peaks for these resonance modes are broader than their TE analogues indicating a possibly useful greater tolerance to absorption.

Moving to more complicated multilayer underlayer systems we see the same resonances repeated, although in the form of coupled resonators. Similar behaviour was seen in the HLH Herpin trials where the two outer layers acted as weak coupled resonators (Table 3.2). Similar coupled thin-film resonators are employed elsewhere, with the most relevant example being that of metal-insulator-metal (MIM) structures used in superlensing applications [124, 125]. Generally speaking the greater the number of periods (coupled resonators) the narrower the peak transmission spatial frequency (NA) linewidth. This makes sense considering the transmitted field must be resonantly enhanced by each resonator to be transmitted and hence any non-resonant frequencies will be absorbed/damped. MIM superlenses are employed above the PR and hence can be an effective means of patterning well defined structures [124, 126, 125]. Coupled resonant underlayers however are not particularly useful in an IL setting as the upper resonator tends to dominate any lower ones. If any non-optimal spatial frequencies (NAs) are also appreciably resonant in the upper resonator they will be reradiated back into the PR reducing the contrast, regardless of any subsequent lower resonators. In addition the prism coupled source interference pattern will have its own NA linewidth thus further limiting the impact of multiple coupled resonant underlayers.
3.5.1 Applications

If we decide to look at non-standard applications however we come across a couple of interesting cases. One, which will be explored thoroughly in the next chapter, is the use of resonant underlayers in the high-NA regime as an antireflection coating. Another interesting example is the use of coupled resonators either side of the PR (Fig. 3.7(b)). Although, generally speaking, coupled resonators complicate the system as they must be well matched to allow adequate transmission, in this case however it is beneficial as the upper resonator acts to enhance (and potentially filter) the incident evanescent fields entering the PR. When exposures are carried out at NAs greater than 1 an index matching liquid (IML) is a necessity due to the evanescent fields present in the air-gap between the coupling prism and the sample. Highly transparent ($\kappa \leq 10^{-5}$) IMLs at $\lambda = 405$ nm are available up to an NA of 1.77 (i.e. $n_{IML} = 1.77$) [106], with the higher refractive index IMLs unfortunately tending to be composed of toxic chemicals. For NA exposures greater than this (ignoring $n > 1.77$ and $k \gg 10^{-5}$ IMLs), evanescent fields in the IML are an inevitability which must be tolerated. As the NA increases the decay constant of the evanescent field increases, thus as the NA increases the thickness of the IML must decrease to allow sufficient intensity to enter the PR. The ability to produce a uniform extremely thin layer of IML (<50 nm) however requires immense pressure to overcome the hydrodynamic forces of the IML [80, 106]. Application of such high forces can potentially damage the prism and often deform the sample resulting in large differences in the IML thickness across the sample. Thus a method of allowing exposures through thicker, non-toxic, lower refractive index IMLs would be beneficial. This can be achieved with the use of a matched top resonator.

In Fig. 3.7 we compare a standard resonant underlayer system and a matched top resonator dual resonator system, both operating at an NA of 1.95 (well above the high n IMLs available), a PR thickness of 100 nm, and a 50 nm thick water IML ($n = 1.34$) for the resonant underlayer system and 150 nm thick for the dual resonator system. In the case of the standard resonant underlayer (Fig. 3.7(a)) the incident evanescent fields traverse two successive mediums resulting in a much reduced intensity upon reaching the resonator. When a resonant overlayer is added (Fig. 3.7(b)) the incident evanescent fields only traverse a single layer allowing greater intensities to reach the resonator and thus stronger fields to be present in the PR. The key point in Figs. 3.7(a) and 3.7(b) is the roughly equivalent PR intensity profiles (Fig. 3.7(d)) even though the IML thickness in Fig. 3.7(b) is three times thicker. Thus the use of a dual resonator system allows
3.5. ALTERNATIVE UNDERLAYER SYSTEMS

Figure 3.7: Overlayer comparison - Common parameters: NA = 1.95, \( n_{\text{prism}} = 2 \), \( n_{\text{IML}} = 1.34 \), \( n_{\text{res.}} = 2.069 \), \( n_{\text{PR}} = 1.6844 + 0.0307i \), \( n_{\text{SiO}_2} = 1.4696 \), \( n_{\text{Si}} = 5.4375 + 0.3420i \), \( d_{\text{res.}} = 170 \text{ nm} \), \( d_{\text{PR}} = 100 \text{ nm} \), and \( d_{\text{SiO}_2} = 100 \text{ nm} \). (a) Resonant underlayer - \( d_{\text{IML}} = 50 \text{ nm} \). (b) Dual matched resonators - \( d_{\text{IML}} = 150 \text{ nm} \). (c) Dual matched resonators - \( d_{\text{IML}} = 50 \text{ nm} \). (d) - PR intensity trace for (a) and (b). Note: PR intensity trace of (c) is off the scale of figure (d).
equivalent exposures with 3 times the IML thickness. Such a large IML thickness is important as it allows exposures without the need for application of high pressures to the sample and prism, and consequently provides far greater IML thickness uniformity. Further gains can be made by using a higher refractive index IML as the decay rate of the evanescent field reduces in higher index mediums and thus even thicker IML layers employed. If the dual resonator system used a 50 nm IML thickness the fields within the PR are approximately 50x greater than those within the PR of the resonant underlayer system Fig. 3.7(c). If the IML thickness of the standard resonant underlayer system was 150 nm thick the PR minimum intensity would be reduced by a factor of 84x compared to the 50 nm IML standard resonant underlayer PR minimum intensity. A secondary benefit of using a resonant overlayer is it allows one to use lower refractive index IMLs such as water which are far ‘nicer’ to use from an experimental standpoint. The use of water is significant as it is the IML of choice in the semiconductor industry, from which they are not likely to shift [72].

The use of a dual resonator system however is limited by the availability of high $n$ materials capable of being deposited and stripped away without damaging the PR layer. If these materials are available it is conceivable to produce a planar multilayer superlens type structure overlayer to not only couple in the fields but also to act as a filtering system to improve the exposure characteristics in noisy systems.

### 3.6 Summary

The transfer matrix method for simulating thin film optical structures employed throughout this thesis has been described in this chapter. An effective media approach known as Herpin effective media exploits the transfer matrix method in the form of a symmetric trilayer replacement layer which can be used to manufacture or model intermediate effective refractive index values which may not be available for the desired purpose. This method is heavily employed in the thin film industry particularly for ARCs. A well known side-effect of Herpin effective media is the presence of effective refractive indices near the stop band far exceeding the refractive indices of the component materials. The application of these high effective refractive indices to dielectric resonant underlayers was considered as a possible method of bypassing the refractive index limit of naturally occurring transparent materials of approximately 2.5. For NAs lower than the constituent Herpin trilayer materials it was shown that it is possible to produce Herpin replacement layers for standard single layer type dielectric resonators. The goal
3.6. SUMMARY

of resonator NAs greater than the refractive index of naturally occurring transparent dielectrics requires the individual Herpin layers to be evanescent. To this end models were constructed with NAs that exceed the refractive indices of one or both of the Herpin layer materials. Herpin replacement resonators were able to be constructed for systems where one of the Herpin layer materials supported evanescent fields. For the Herpin systems where both Herpin materials supported evanescent fields it was not possible to find a Herpin replacement due to the single layer having a real non-zero phase thickness ($\phi$), while the Herpin effective medium systems had complex phase thicknesses ($\Phi$) with a real component of zero due to the presence of evanescent fields within all the Herpin layers. Due to this the phase thicknesses ($\phi$ and $\Phi$) were not able to be matched and Herpin replacement resonators were not able to be produced. Thus it fails in our goal to push the NAs higher than is achievable with naturally occurring material for evanescent interference lithography.

The exploration of Herpin effective media, although not a success in increasing the NA, lead to a better understanding of resonant underlayer systems and the discovery of two novel systems. Firstly a new form of ARC for the high-NA regime was discovered capable of employing non-standard materials such as metals, semiconductors and highly absorbing dielectrics. This system is thoroughly investigated in the following chapter. A second system was also devised which employs a dual resonator system with matched resonators either side of the PR layer. A common problem in the ultra high-NA regime is the lack of very high refractive index transparent IMLs. In this regime a very high pressure is applied to the sample to minimise the gap between the sample and prism, thus minimising the amount of field lost due to evanescent decay in the IML. The use of a top resonator however allows one to use a relatively low index IML such as water without the application of high pressure due to the resonant enhancement effect of the top resonator.
Chapter 4

Evanescent-coupled antireflection coatings

4.1 Prologue

The resonant underlayers so far discussed in this thesis have all been designed to operate in the ultra high-NA regime ($NA > n_{PR}$) to produce a symmetric intensity distribution within the PR layer. This is a relatively specialised area of application that involves evanescent fields in the PR and requires prism or grating-coupled of the exposing light source. These resonators are not suitable for use in the high-NA regime ($0.5 < NA < n_{PR}$) regime where the fields are propagating within the PR, and the requirement of a matching or reflecting underlayer ($n_{high}$) are different. As the fields are propagating, a reflection will occur at the PR|$n_{high}$ interface of an ultra high-NA resonant underlayer thus producing an undesirable standing wave pattern within the PR which reduces pattern (and consequently pattern transfer) fidelity. In the ultra high-NA regime the resonator acts as a bottom source of evanescent fields; as evanescent fields lack phase propagation, interference effects normal to the interfaces are not present within the PR. In the high-NA regime however, phase effects are present due to the propagating nature of the fields, and thus interference patterns (standing waves) occur in the direction normal to the interface. As the purpose of these underlayers is to improve the pattern characteristics (pattern fidelity and depth of field), the resonator ‘equivalent’ in the high-NA regime is one which removes the standing wave phase effects.

\[^{1}\text{Within the industry this is termed the hyper-NA regime, we stick to the name high-NA to avoid confusion between the ranking of the hyper and the ultra high-NA regimes.}\]
4.2. INTRODUCTION - ANTIREFLECTION COATINGS

i.e. an antireflection coating (ARC). This chapter is dedicated to the development of these new ARCs for the high-NA regime using the insights gained from studying the ultra-high-NA regime.

This chapter begins with a brief introduction to the general theory, concepts, and types of ARCs. As an alternative to these methods, evanescent-coupled ARCs are developed. The principles, theory, and types of evanescent-coupled ARCs are then explored with emphasis on the two different forms available. A brief discussion on applying evanescent-coupled ARCs to 193\textit{i} photolithography is then covered. The chapter ends with a recap of the design concepts and characteristics of evanescent-coupled ARCs.

4.2 Introduction - Antireflection coatings

The need to suppress reflections and/or enhance transmission is a common theme throughout many optical technologies. The increasingly widespread usage of optical and optoelectronic technologies demand novel ARCs to meet the specific needs of these new systems. So, although ARCs are a very well established technology, a great deal of development is still occurring \[127, 128\]. For instance new nanocomposite materials \[129\] have been developed to allow efficient reflection suppression for 193\textit{i} photolithography, without which the extremely complex catadioptric (lens/mirror) projection optics systems would have very high losses and a large amount of image ghosting, thus rendering them unusable.

ARCs fall into two general categories, destructive interference and refractive index matching ARCs (Fig. \[4.1\]). In either case the reflectance from a single interface is given by Fresnel’s equations,

\[
R_{\text{TE}} = \left| \frac{n_1 \cos \theta_i - n_2 \cos \theta_t}{n_1 \cos \theta_i + n_2 \cos \theta_t} \right|^2 \quad \text{and} \quad R_{\text{TM}} = \left| \frac{n_1 \cos \theta_i - n_2 \cos \theta_t}{n_1 \cos \theta_i + n_2 \cos \theta_t} \right|^2,
\]

where the reflectance \(R_i\) is dictated by the refractive indices \((n_1 \text{ and } n_2)\), angle of incidence \((\theta_i)\), angle of transmission \(\theta_t\), and the polarization. At normal incidence Eq. \((4.1)\) reduces to

\[
R = \left| \frac{n_1 - n_2}{n_1 + n_2} \right|^2,
\]

indicating that the simplest method of reflection reduction is to minimise the difference in refractive indices. Indeed, this is visible if one compares the transparency of an old (weathered) pane of glass to a new one. Old glass has an intermediate layer
4.2. INTRODUCTION - ANTIREFLECTION COATINGS

4.2.1 Destructive interference ARCs

When a wave is incident upon a three layered system (Fig. 4.1(a)), reflections occur at two interfaces ($n_0|n_1$ and $n_1|n_2$). Assuming layer $n_1$ is thin enough, thin film interference will occur between the reflections ($R_1$ and $R_2$). The degree of interference is determined by the relative phases and the amplitudes of the reflected waves. The phase is dictated by the thickness ($d$) and refractive index of the $n_1$ layer; assuming normal incidence, if $d = \lambda/4$ the round trip path length will be $\lambda/2$, therefore $R_2$ will have a phase difference of $\pi$ relative to $R_1$ and the two waves will destructively interfere. This is the classic example of an ARC called a quarter-wave ARC. Of course, having the reflectances $\pi$ out of phase is not enough, with the amplitudes of $R_1$ and $R_2$ also required to be equal to eliminate reflections. Matching the reflection amplitudes at low angles for non-absorbing materials requires $n_1 = \sqrt{n_0n_2}$. Assuming $n_0 = 1$, as it produced by weathering; this intermediate layer has a lower refractive index than the glass which acts as a refractive index step between the air and glass thereby reducing the reflectance. The presence of this intermediate layer however causes its own problems (and solutions), namely, thin film interference effects.
does for most applications, a suitable $n_1$ cannot be found for most common glasses. For instance if Crown glass ($n = 1.52$) is the substrate, the required $n_1$ will equal 1.22 for which there are no naturally occurring transparent materials. As was mentioned in the previous chapter, Herpin effective media are often used in this capacity to produce (an effective) $n_1$ when a suitably close refractive index is not available. The use of single layer antireflection coatings is visible in everyday life, where it is often noticeable as a blue tinge seen in the reflectance from things such as solar panels, and low cost sunglasses. The blue tinge however is indicative of poor broadband reflectance suppression.

Single layer $\lambda/4$ ARCs are non-optimal for broadband illumination due to the layer thickness requirement being equal to $\lambda/4$, thus if $\lambda$ changes the thickness will no longer satisfy the minimum reflectance criteria. Likewise changing the the angle of incidence alters the relative phase thickness of the layer, thus the reflectance increases as the angle diverges from the design angle. The use of a single layer fundamentally limits the reflectance characteristics due to the limited range of variables available. The requirement for stronger reflection suppression at a single wavelength, two wavelengths, or broadband suppression requires multiple layers to allow access to enough variables to find suitable solutions [111].

Multilayered ARCs often employ multiple $\lambda/4$ and $\lambda/2$ layers, where the former is used for destructive interference and the later is used as an absentee layer often to broaden the reflectance minimum [111]. The addition of multiple layers and materials provides greater design flexibility at the cost of rapidly increasing complexity. Even at three layers the number of permutations (thickness, refractive index, ordering) of parameters becomes considerable. Thus the design of ARCs from this point on becomes somewhat of an art form where experience, and trial and error become significant. For this reason multilayered systems will not be further examined here (except in relation to BARCs (Section 4.3.1)), although if the reader is interested, *Thin-Film Optical Filters* by MacLeod is generally considered a very good start [111].

Although destructive interference based ARCs perform very well in many applications, they are fundamentally limited by the refractive index step at the the top interface (Fig. 4.1(a)). This is important as whenever the system is not under optimal illumination conditions the reflectance from this step will be evident. To alleviate this, non-porous ultra-low refractive index nanocomposites and polymers have been developed and employed but are yet to overcome this difficulty completely [129, 130]. This points to a second avenue for ARCs, that is, removing the refractive index step all
4.2. INTRODUCTION - ANTIREFLECTION COATINGS

4.2.2 Refractive index matching ARCs

The production of a reflection at an interface is due to a mismatch in refractive indices (optical impedances), thus if one can remove this mismatch the accompanying reflection will also be removed. This can be carried out, for example, using subwavelength nano-textured structures, where the effective refractive index of the nano-textured structure changes as the wave propagates into the medium. The example in Fig. 4.1(b) is an inverted nanoparticle structure with a triangular cross-section [131]. At the top of the nanoparticle layer the light ‘sees’ an effective refractive index approximately equal to that of the cover medium ($n_0$). As the wave propagates deeper however, it progressively sees a greater proportion of the nanoparticle material refractive index ($n_2$), thus the effective refractive index steadily increases. Provided the nano-textured material is that of the substrate, index matching will also occur at the base of the nano-textured layer.

Refractive index matching ARCs fall into two classes: regular, and pseudo-random nanostructures. Regular nanostructures are frequently called ‘Moth-Eye’ ARCs after the structures found on the cornea of moths which were the inspiration for this class of ARCs [132, 133, 134, 135]. The structures on moths eyes are subwavelength (100 nm diameter and 170 nm pitch) cones, which not only allow moths to see better in very low light conditions and reduce reflections from their large corneal area, but also remove any difference in polarization sensitivity. A large amount of study has been carried out in this area with a study by Bernhard et al having examined the corneas of 361 different moth species [133]. Interestingly the structures on the moths corneas show a great deal of variation, with refractive index profiles (described by structure shape) such as nipple, conical, paraboloidal, and Gaussian-bell shaped.

Man-made structures have similarly been produced with novel shapes such as pyramids, cones, domes, gratings, pillars, as well as their inverse structures. These type

\[\text{Note this is not the same as the Herpin effective media refractive index discussed in the previous chapter. In this case it is more of an \textquoteleft average\textquoteright\ refractive index. There are several effective refractive index models, perhaps most notably the Maxwell-Garnett model for homogeneous layer pairs, and the Bruggeman homogeneous multilayer approximation for inhomogeneous layers [127]. These are however not necessary for understanding Fig. 4.1(b) where a simple in-plane averaging will suffice, i.e. the greater the proportion of } n_0 \text{ present the closer the effective refractive index will be to } n_0.\]
of structures are perfect for solar power type applications which require very low broadband reflectance across a wide range of angles and all polarizations. This type of ARC however is limited by the need to keep the nano-structure free of debris. Consequently for practical applications an ethylene-vinyl acetate encapsulating module is employed [136].

Pseudo-random nanostructured ARCs employ similar refractive index profiles to those of regular nanostructures except they are produced by pseudo-random processes such as etching (wet, dry, electrochemical), sol-gel, glancing angle deposition (GLAD), and chemical vapour deposition. All these methods can be used to produce porous structures which have pseudo-random packing arrangements, for instance GLAD produces randomly packed columnar structures which although randomly placed on the substrate, grow in a particular preferred direction [113]. As the columns get higher the packing density decreases thus a gradient refractive index (GRIN) profile is produced. If multiple layers or processing steps are employed a large range of GRIN profiles can be produced such as linear, parabolic, cubic, gaussian, quintic, exponential, exponential sine etc. with quintic and exponential sine generally considered the optimal profile [137, 128]. Generally these GRIN ARCs can be optimised for very good simultaneous broadband, wide angle, and low polarization sensitivity. The highly porous nature of the uppermost layer of these ARCs however severely reduces the mechanical strength and durability of these ARCs thus limiting their applications [127].

Refractive index matching ARCs have very low broadband reflectance, very high angle tolerance, and often polarization independent. All these strengths however are often out-weighed by manufacturing cost (compared to planar layer ARCs), poor durability, and the need to keep the nanostructures clean and thus maintain efficiency. For this reason these type of ARCs are employed in high value products often as an internal layer to reduce fouling of the ARC, although self-cleaning hydro-phobic nanostructured ARCs are also being developed [138, 139, 140, 141].

4.3 Antireflection coatings for photolithography film stacks

ARCs are heavily employed by the photolithography industry; as was previously mentioned, ARCs are an absolute necessity for the very complex beam transport and projection optics systems which can have upwards of 50 optical elements, without which the amount of reflections and ghost images would rapidly make this type of system
infeasible. There is another very important use of ARCs in photolithography, that is, to remove the reflections in the PR layer of the film stack to prevent the formation (and consequently patterning) of standing waves. The use of ARCs in the film stack is a very well established aspect of photolithography where it is required to reduce the reflectance from the substrate.

Airy’s equation [142] for the total amplitude reflection coefficient of a generic single layer stack (assuming infinite cover and substrate) is given by

\[
\tilde{r} = \frac{\tilde{r}_{12} + \tilde{r}_{23}e^{2i\tilde{\beta}}}{1 + \tilde{r}_{12}\tilde{r}_{23}e^{2i\tilde{\beta}}},
\]

(4.3)

where \(\tilde{r}_{mn} = (\tilde{p}_m - \tilde{p}_n)/(\tilde{p}_m + \tilde{p}_n)\), for TE polarization \(\tilde{p}_m = \tilde{n}_m\cos\tilde{\theta}_m\) or for TM polarization \(\tilde{p}_m = \cos\tilde{\theta}_m/\tilde{n}_m\), \(\tilde{n}_m\) is the complex refractive index in medium \(m\), and \(\tilde{\theta}_m\) is the angle of incidence. With the total reflectance given by

\[
R = |\tilde{r}|^2 = \frac{\tilde{r}_{12}^2 + \tilde{r}_{23}^2 + 2\tilde{r}_{12}\tilde{r}_{23}\cos 2\tilde{\beta}}{1 + \tilde{r}_{12}^2\tilde{r}_{23}^2 + 2\tilde{r}_{12}\tilde{r}_{23}\cos 2\tilde{\beta}}.
\]

(4.4)

In Fig. 4.2(a) we see a plot of the total reflectance vs. the PR thickness for a layer of PR (\(n = 1.6844 + 0.0307i\)) on a Si substrate (\(n = 5.4375 + 0.3420i\)) illuminated through a water immersion layer (\(n = 1.3431\)) at normal incidence with a wavelength of 405 nm. Two trends can be seen in this plot, firstly, a modulation is present due to the the cosine term in Eq. (4.4), secondly an overall downwards slope indicative of increasing absorption with thickness \(3\). This plot implies that the reflectance and therefore importantly the dose to the PR changes with PR thickness. This is referred to as a swing curve and is a very important consideration in photolithography as it dictates the optimum exposure time (dose) required for full pattern clearance \[10\]. Under-dosing may result in incomplete pattern clearance, while over-dosing can result in excess development and increased line edge roughness. The effect of the swing curve can be particularly bad for 3D morphologies where existing patterns are already present and thus create a great deal of PR thickness variation across the exposure field resulting in different doses across the sample. For these reasons, measures are taken to flatten the swing curve. If we consider Eq. (4.4) we see there are two options. Firstly,
4.3. ARCS FOR PHOTOLITHOGRAPHY FILM STACKS

![Reflectivity swing curve](Image)

Figure 4.2: (a) Reflectivity swing curve for a Water|PR|Si stack. Parameters in footnote\(^4\). (b) Single layer BARC schematic - if layer \( n_2 \) is of a \( \lambda/4 \) thickness destructive interference occurs in layer \( n_1 \) (PR) which prevents standing waves.

\( \tilde{r}_{23} \) may be set to zero by employing a bottom antireflection coating (BARC) under the PR. Secondly, and less obviously, \( \tilde{r}_{12} \) may be set to zero by using a top antireflection coating (TARC). Both BARCs and TARCAs flatten the swing curve, in practice however neither are perfectly equal to zero thus both are often employed.

### 4.3.1 BARCs

BARCs are ARCs placed below the PR to prevent standing waves within the PR (for instance layer \( n_2 \) in Fig. 4.2(b)). This application necessitates simple non-porous planar layers thus planar destructive interference type ARCs are employed. If we assume a simple trilayer PR|BARC|substrate stack, we can use Eq. (4.3) to minimize the reflectance. The condition for zero reflectance occurs when the numerator of Eq. (4.3) is equal to zero i.e.

\[ \tilde{r}_{12} + \tilde{r}_{23} e^{2i\beta} = 0. \]  

(4.5)

In the case of a classic non-absorbing \( \lambda/4 \) ARC with non-absorbing bounding media \( \tilde{r}_{12} = \tilde{r}_{23} \) and \( e^{2i\beta} = -1 \). As the BARC is applied between two absorbing bounding media (PR|BARC|Si) this simple balance fails due to a mismatch in impedance caused by the imaginary refractive index components. If the reflection coefficients are expressed as a magnitude and phase \( \tilde{r}_{ij} = |r_{ij}| e^{i\theta_{ij}} \), Eq. (4.5) becomes an equated pair of equalities where both the real and imaginary components must equal the same

---

\(^4\)Parameters: Prism/Water - \( n = 1.3431 \)  [144], \( d = \) bulk, PR AZMIR701 - \( n = 1.6844+0.0307i \)  [119], \( d = 0-500 \) nm, Si - \( n = 5.4375+0.3420i \)  [122], \( d = \) bulk, TE polarization, \( \lambda = 405 \) nm.
4.3. ARCS FOR PHOTOLITHOGRAPHY FILM STACKS

thickness $d$.

$$d = \frac{\lambda}{4\pi\kappa} \ln \left| \frac{r_{23}}{r_{21}} \right| = \frac{\lambda}{4\pi n} (\theta_{21} - \theta_{23}) \quad (4.6)$$

Although Eqs. (4.5) and (4.6) have a simple form they are often difficult to solve for anything other than the non-absorbing $\lambda/4$ and are typically solved numerically [10].

The reflectance minimum for single layer $\lambda/4$ ARCs narrows as the NA increases, thus they struggle to meet the needs of high-NA photolithography. To overcome this difficulty dual and trilayer BARCs have been developed [10, 22, 145]. Typically these utilize a low-$\kappa$ upper layer to allow high transmission into the BARC and better dose uniformity at the bottom of the PR. The subsequent BARC layers have higher $\kappa$ values and act as planarizing layers [22].

4.3.2 TARCs

Although not directly relevant to this thesis, the concept of top antireflection coatings (TARCs) will be briefly covered for completeness. TARCs are overlayers designed to reduce $\tilde{r}_{12}$ or equivalently to allow maximum transmission, thus they are the type employed on things such as camera lenses. The cosine term in Eq. (4.4) $(2\tilde{r}_{12}\tilde{r}_{23}\cos 2\tilde{\beta})$ gives rise to the reflectivity swing curve. One of the means of flattening the swing curve is to use a TARC to set $\tilde{r}_{12} = 0$, and thus remove reflections ($R_0$) from the top of the PR. (Fig. 4.2(b)). Suitable $n$, $\kappa$ and $d$ values can be obtained using Eq. (4.3) or Eq. (4.6) and if necessary by using an effective reflectance approach if the stack is comprised of further layers beneath the PR [146].

TARCs are frequently employed in 193i lithography (water|TARC|PR stack) where the required refractive index for a $\lambda/4$ ARC is $n_{\text{TARC}} \approx 1.56$ which is readily achieved using spin on polymers. As with BARCs though, the simple $\lambda/4$ assumptions of non-absorbing materials and low NAs are not true, and $n$, $\kappa$, and $d$ must be altered accordingly. TARCs however are not simply for reflection control, they also perform the crucial rolls of being a top coat to prevent water from entering the resist, and to modify the contact angle of the water immersion liquid allowing it to flow freely across the wafer. Layers performing multiple functions is a common practice in photolithography for the purpose of improving manufacturing economics. This is particularly important for the layers beneath the PR to the point where these techniques are now referred to as underlayer technologies [22].
4.4 Underlayer technology

Manufacturing using modern 193i multipatterning techniques is a very complex multi-staged process where the economics dictates usage as much as the performance. The photolithography process alone can employ over 50 different individual steps, each involving many different factors including reflection control, etch selectivity, adhesion, chemical compatibility, via filling, etc. Underlayer technology seeks to meet the various underlayer requirements in as few layers as economically possible. A good example of underlayer technology is the replacement of a separate BARC and hardmask (Section 1.3.1) layers with a single SiON BARC, which acts as both the BARC and hardmask [147]. With the very high cost of ownership of 193i lithography systems and a current lack of an high volume manufacturing successor, it becomes vital to identify and develop further underlayer techniques to extend the lifetime of the 193i process.

The evanescent-coupled ARCs which are the theme of this chapter serve as a possible extension to underlayer techniques. Conventional BARCs for 193i lithography employ weakly absorbing dielectrics \( n \approx 1.5 - 1.8 \) and \( \kappa < 1 \) which are typical of absorbing \( \lambda/4 \) ARCs. Evanescent-coupled ARCs however are capable of utilizing a far greater range of materials including previously excluded materials such as metals, semiconductors, high-\( \kappa \) dielectric, and low-\( n \) \( (n < NA) \) dielectrics. This great expansion of ARC materials may potentially be of benefit to the semiconductor industry, allowing new multi-use underlayer designs as well as new manufacturing permutations and processes.

4.5 Evanescent-coupled ARCs

Evanescent-coupled ARCs are a form of destructive interference-based ARC that operate under high NA illumination \( (1 < NA < n_{PR}) \), which utilize a resonant underlayer to provide the backward going field to destructively interfere with the primary reflection from the \( PR|ARC \) interface. Such a form of ARC has not been considered before for semiconductor photolithography, or for any other application, to the best of our knowledge.

Resonant structures available for evanescent-coupled ARCs include surface state polariton resonators (SSR) [148, 129] (Fig. 4.3(a) and Section 2.5.2) and dielectric resonators (DR) [149] (Fig. 4.3(b) and Section 2.5.1) systems. Both require the presence of low-\( n \) \( (n < NA) \) layers supporting evanescent fields to allow resonant confinement
4.5. EVANESCENT-COUPLED ARCS

**Figure 4.3:** Evanescent-coupled ARC schema. (a) Surface state based stack in the Otto configuration. (b) Dielectric resonator based stack.

of the fields. Energy couples into the resonator through this evanescent layer (thus the name) via frustrated total internal reflection. To act as an ARC, the intensity of the backward-going field must match that of the reflected TIR wave at the PR|underlayer interface. The thickness of the resonator layers and the NA must be carefully balanced to allow this condition. The accumulated phase difference arises from a combination of the evanescent layer thickness and the phase changes due to the absorbing and/or propagating components of the resonators [148, 63]. Although the phase is ‘locked’ in evanescent fields the phase of the TIR reflection is not and changes depending on the NA, the thickness of the evanescent layer, and the effective refractive index and reflectance of any further layers [63, 150, 151].

Examples of both classes of systems are now presented prior to a discussion on the analytical and modelling techniques needed for system design. These models are then further developed to highlight the key characteristics and principles of evanescent-coupled ARCs. Despite these models using 405 nm illumination (in an immersion interference lithography configuration), the concepts scale to any wavelength including 193 nm for 193i lithography which is explored later in this chapter (Section 4.9). Experimental investigation of this technique is presented in Chapter 5, which together with this chapter contribute two of the main original contributions of this thesis.

### 4.5.1 Surface state polariton resonator based systems

Surface states are electronic states that occur at material interfaces (such as A in Fig. 4.3(a)) provided certain refractive index combinations and illumination conditions.
are met [148, 123]. They present as charge oscillations at the interface and thus require absorbing media, either metals or lossy dielectrics, to provide the charge carriers. Photons can excite and couple into these oscillations as polaritons thereby producing a surface state resonator (SSR). Excitation of polaritons is achieved using TM illumination at an NA such that the wave vector component in the plane of the interface \( k_x \) is matched with that of the surface state polariton resonant condition wave vector \( \beta \) (discussed layer, Eq. (4.14)). When these wave vectors are matched energy can efficiently couple into the oscillations at the interface.

In the planar configuration SSRs require an interface between a low-\( n \) (\( n < NA \)) low-\( \kappa \) medium (referred to as an evanescent layer) and a high-\( \kappa \) medium (Fig. 4.3(a)). From a conceptual standpoint this makes perfect sense as not only is a source of charges required (high-\( \kappa \)), but also the field must be confined to the interface to allow resonance/waveguiding to occur hence the low-\( n \) low-\( \kappa \) layer. With this type of configuration the oscillating charges at the interface produce electric fields which decay either side of the interface due to evanescent fields and/or absorption. The decaying evanescent field(s) have the effect of confining the energy to the interface and hence allowing energy to build up. The requirement for evanescent fields determines the meaning of ‘low-n’ for the evanescent layer, that is, it must be less than the NA or else the energy will radiate away. Excitation of the oscillations is carried out using a prism/coupling medium with a refractive index greater than the NA; in the case of evanescent-coupled ARCs the coupling medium is the PR thus the evanescent layer must have \( n < n_{PR} \).

There are two options for this system i.e. high-\( \kappa \) as the bottom layer (Otto configuration Fig. 4.4(a)) or the top layer (Kretschmann configuration Fig. 4.4(b)) [148]. Both configurations are capable of acting as ARCs, although in practice the Otto configuration is superior as it does not suffer the heavy coupling absorption losses which the Kretschmann configuration does owing to fields being coupled through a strongly absorbing medium (see Fig. 4.4). The lower inherent coupling losses of the Otto configuration allows for greater design flexibility for intensity and phase matching.

### 4.5.2 Dielectric resonator based systems

Evanescent-coupled ARCs can also act in a dielectric resonator (DR) configuration, using the ‘classic’ trilayer configuration, have a \( n_{low} | n_{high} | n_{low} \) film stack (Fig. 4.3(b)). \( n_{low} \) must be less than the NA and \( n_{high} \) greater than the NA in order to support evanescent coupling and resonance. Provided the \( n_{high} \) layer is of a resonant thickness and illumination is at an appropriate NA, energy will couple into the resonator via frustra-
4.6 MODELLING OF EVANESCENT-COUPLED ARCS

![Diagram of evanescent-coupled ARC surface state resonator schema. Blue layer low refractive index low absorbing dielectric. Grey layer strongly absorbing metal or dielectric. Polaritons (arrow sign waves) occur at the interface between the two layers. (a) Otto configuration and (b) Kretschmann configuration.](image)

...ted TIR allowing energy to build up. TIR at the PR side of the \( n_{\text{high}} \) boundary provides the backward-going field to destructively interfere within the PR. Both TE and TM polarizations can resonate in dielectric resonators, although the different modal conditions mean different \( n_{\text{low}} \) and \( n_{\text{high}} \) thicknesses are required. For DR evanescent-coupled ARCs low-\( \kappa \) materials are used in all layers. For the \( n_{\text{low}} \) layer, \( \kappa \) must be sufficiently low to allow enough energy to couple in (and importantly out). For the \( n_{\text{high}} \) layer, \( \kappa \) must be low enough to prevent over suppression of the resonance. The \( n_{\text{low}}|n_{\text{high}}|n_{\text{low}} \) stack may also be symmetric or asymmetric, with the two \( n_{\text{low}} \) layers not necessarily the same materials or thicknesses. The modelling of evanescent-couple ARCs (both SSR and DR based) in practical photolithography conditions is now explored.

4.6 Modelling of Evanescent-coupled ARCs

The transfer matrix method is built upon the transfer of fields across interfaces using Maxwell’s equations, as such, it naturally incorporates both surface state polariton and dielectric resonator effects. Due to this and the multilayered thin film structure of evanescent-coupled ARCs, the transfer matrix method employed in Chapter 3 serves as the ideal method of modelling these systems particularly for strongly absorbing multilayered systems where analytical solutions become rapidly complex (as is the case...
using Airy’s reflectance equation Eq. (4.3)).

Once again (repeating for the sake of ease of reading) we have the optical effect of a thin film system given by

$$\vec{a}_C = \hat{F}_C^{-1}\hat{M}\hat{F}_S\vec{a}_S,$$

(4.7)

where $\vec{a}_{C,S}$ are the field coefficients in the cover and substrates respectively, $\hat{F}_C^{-1}$ is the inverse of the cover field matrix, $\hat{M}$ is the characteristic matrix of the thin film layer(s), and $\hat{F}_S$ is the field matrix in the substrate. If we use the 2x2 partitioned form of these matrices (see Eq. (3.13)) and expand it Eq. (4.7) becomes

$$\begin{bmatrix} a_C^+ \\ a_C^- \end{bmatrix} = \begin{bmatrix} 1 & 1/\gamma_C \\ 1 & -1/\gamma_C \end{bmatrix} \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix} \begin{bmatrix} 1 & 1 \\ \gamma_S & -\gamma_S \end{bmatrix} \begin{bmatrix} a_S^+ \\ a_S^- \end{bmatrix},$$

(4.8)

Where $a_C^\pm$ and $a_S^\pm$ are the field coefficients in the cover and substrate respectively, where $+$ represents the forward going fields and $-$ the backward going fields, $\gamma_{C,S}$ is the effective optical admittances in the cover and substrate, and $M_{ij}$ are the components of the characteristic matrix $\hat{M}$ given previously in Eq. (3.12). Equation (4.8) contains the basis vectors for a single polarization, thus the appropriate optical admittances for the cover, intermediate layers, and substrate must be employed. For TE polarization $\gamma = -n \cos \theta/z_0$, and for TM polarization $\gamma = n/(z_0 \cos \theta)$, where $n$ is the refractive index of the particular layer, $\theta$ the angle within the layer, and $z_0$ the impedance of free space. The characteristic matrix is the matrix product of the individual layer transfer matrices and thus contains the optical effect of the stack excluding the cover and substrate mediums i.e.

$$M_i = M_1M_2M_3...M_N.$$ 

(4.9)

The reflection coefficient ($r$) for the total system is given by the ratio of the output to the input fields in the cover. Thus using Eq. (4.8) the reflection coefficient is given by

$$r = \frac{a_C^-}{a_C^+} = \frac{\gamma_C M_{11} + \gamma_C \gamma_S M_{12} - M_{21} - \gamma_S M_{22}}{\gamma_C M_{11} + \gamma_C \gamma_S M_{12} + M_{21} + \gamma_S M_{22}},$$

(4.10)

and the total reflectance $R$ is given by

$$R = |r|^2.$$

(4.11)

In the following sections these equations will be used to design and optimize example SSR and DR based evanescent-couple ARC systems.


4.7 SSR based evanescent coupled ARCs

In Fig. 4.5 we have an example of a SSR evanescent-coupled ARC. This system is based on a MgF$_2$|Ru bilayer (Fig. 4.5(a)) operating at an NA of 1.4091 and TM polarization. The design principles for choosing these layers are outlined in Section 4.7.1 that follows. Figure 4.5(b) is an intensity trace through this system (blue line) with a comparison trace (red line) on bare silicon. There are several things of note in this plot. Firstly it can be seen that the underlayer is acting as an ARC with a smooth intensity within the PR layer, where the intensity profile is dictated by Beer-Lambert law absorption alone. Secondly there is an uptick in the trace at the MgF$_2$|Ru interface which is due to polariton resonance at the interface. This is the source of the backwards going waves for destructive interference. Thirdly, a prominent standing wave is visible in the non-ARC intensity trace, due to the strong reflection at the PR|Si interface. Finally it is worth noting that non-ARC intensity profile is also the swing curve for this particular system, and as one would expect, the application of an ARC flattens the swing curve.

4.7.1 $d$, $n$ and $\kappa$ characteristics

In the design and optimisation of both SSR and DR based evanescent-coupled ARCs the refractive index and the layer thicknesses go hand in hand. For instance the degree to which the fields can be enhanced by the resonator is dependent upon both the radiative and absorptive losses of the system, both of which depend on the refractive indices and the thickness of the individual layers (assuming a suitable NA). A good example is the $n_{low}$ coupling medium, which not only serves to couple energy in, but also to optimise the amount of energy coupling into the resonator. Thus the same amount of coupled energy can be achieved by either changing the thickness and/or the absorption of the $n_{low}$ layer. For this reason there exists a parameter space ($n_i$, $\kappa_i$, $d_i$, etc.) of optimal solutions rather than a single one. Although fully flexible models of these systems can be constructed, the results involve a 6-7D parameter space and thus are not very informative. Hence applicable models are constructed here employing several fixed parameters so that others may be explored, typically the refractive index of layer(s) or the layer thickness(es).

The $d$, $n$ and $\kappa$ characteristics of SSR evanescent coupled ARCs will now be investigated, beginning with defined layer materials to find the optimum thicknesses, then use these same thicknesses to back solve the systems to get an idea of uniqueness of these solutions in terms of refractive indices. For the sake of coherency we use this
4.7. SSR BASED EVANESCENT COUPLED ARCS

Figure 4.5: MgF$_2$|Ru based SSR evanescent-coupled ARC, NA=1.4091 and TM polarization.\(^5\) (a) - Film stack. (b) - Intensity trace through the film stack, blue line indicates the ARC case with the ARC layers highlighted in tea green, red line the no ARC case. Note the smooth Beer Law absorption trace within the PR when the ARC is employed. Surface state resonance is evidenced by the up tick in intensity at the MgF$_2$|Ru interface. The no ARC case shows prominent standing waves within the PR. The ARC has been replaced with Si thus the decaying fields beneath the PR.

Figure 4.6 shows an evanescent layer vs. high-κ layer thickness reflectance plot. This system is in the Otto configuration ($n_{low}$ on top), with a film stack PR|MgF$_2$|Ru|Si at a design NA of 1.4091 (Eq. (4.14)), and TM polarization. The minimum reflectance for this system ($R = 1.2 \times 10^{-6}$) occurs at an evanescent layer thickness of 115 nm and a high-κ layer thickness of 27 nm; which unsurprisingly is the system specified in Fig. 4.5(a). As was mentioned in Section 4.3.1 the reflectance minimum occurs when the numerator of the reflectance coefficient equation (Eq. (4.10)) is equated to zero, i.e.

$$\gamma_C M_{11} + \gamma_C \gamma_S M_{12} - M_{21} - \gamma_S M_{22} = 0. \quad (4.12)$$

As reflections arise from both the real and imaginary components of $r$, both must be simultaneously equal to zero to achieve full reflection suppression.

$$\text{Re}(r) = \text{Im}(r) = 0 \quad (4.13)$$

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\(^5\)Prism/SiO$_2$ - $n = 1.4696$ [121], $d = \text{bulk}$, PR AZMIR701 - $n = 1.6844+0.0307i$ [119], $d = 500$ nm, MgF$_2$ - $n = 1.38$ [152], $d = 115$ nm, Ru - $n = 2.2137+3.5864i$ [153], $d = 27$ nm, Si - $n = 5.4375+0.3420i$ [122], $d = \text{bulk}$, NA = 1.4091, TM polarization, $\lambda = 405$ nm.
4.7. SSR BASED EVANESCENT COUPLED ARCS

Figure 4.6: PR|MgF$_2$|Ru|Si SSR evanescent coupled ARC thickness space reflectance map. Reflectance map for differing evanescent and high-$\kappa$ layer thicknesses. Blue contours represent $\text{Re}(r)=0$ and the green contours $\text{Im}(r)=0$. The intersection of these contours is the location of the reflectance minimum ($R = 1.222 \times 10^{-6}$), indicated by a white cross. Yellow and cyan dashed lines indicate the thickness transects for Figs. 4.7(a) and 4.7(b) respectively.

In Fig. 4.6 the blue contours indicate $\text{Re}(r)=0$, and the green contours $\text{Im}(r)=0$; the intersection of these two contours should be the location of the minimum reflectance, as is indeed the case. Interestingly, the higher order real and imaginary contours do not intersect. This is due to the effect of the thickness of the high-$\kappa$ layer. If the Si substrate is replaced with Ru (i.e. a bulk thickness high-$\kappa$ layer) none of the real and imaginary contours intersect. Although the resonant intensity is similar for these two systems, the Ru substrate case is not able to provide the correct phase for destructive interference; for this reason none of the higher contours in Fig. 4.6 intersect.

The photolithography industry generally specifies a suitable ARC reflectance maximum of 0.5% [22], which is delineated by a white contour in Fig. 4.6. The area within this contour suggests there is a reasonable amount of leeway in the thickness of the layers for this particular ARC.

To get an indication of the effect of thickness change on standing wave production two plots have been produced by varying the evanescent layer and high-$\kappa$ layer

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$^6$PR AZMIR701 - $n = 1.6844+0.0307i$ [119], $d =$ bulk, MgF$_2$ - $n = 1.3957$ [152], $d =$ 0-200 nm, Ru - $n = 2.2137+3.5864i$ [153], $d =$ 0-200 nm Si - $n = 5.4375+0.3420i$ [122], $d =$ bulk, TM polarization, $\lambda = 405$ nm.
4.7. SSR BASED EVANESCENT COUPLED ARCS

Figure 4.7: SSR ARC layer thickness dependence Parameters and transects given in Fig. 4.6 (a) - Intensity traces along the evanescent layer thickness transect (yellow dashed line Fig. 4.6). As the evanescent layer thickness shifts from the optimal value the standing wave strength grows noticeably. Red and blue lines indicate thicknesses less and greater than the optimal value (black line) respectively. (b) - Intensity traces along the high-κ layer thickness transect (cyan dashed line Fig. 4.6). Thicker layers than the optimum value results in small standing waves whilst thinner layers produce far larger standing waves. Note the lower PR interface represented is that of the optimum value.

thicknesses along the yellow and cyan transects in Fig. 4.6 respectively. Varying the low-n evanescent layer thickness (Fig. 4.7(a)) produces a roughly symmetric standing wave response as one would expect considering the approximate symmetry about the optimum for this transect. The high-κ case (Fig. 4.7(b)) however indicates the system is more sensitive to thinner high-κ layer thicknesses but less sensitive to thicker layers. As was indicated previously, this is due to the thickness exceeding a certain value whereby no energy can couple out or reflect from the back surface and thus the amount of energy coupled to the interface is constant for larger high-κ layer thicknesses.

Now we can use the optimal thicknesses found in Fig. 4.6 to investigate the potential variation in refractive indices. Once again to keep plots in 2D space we have to assume certain parameter values, in this case these are the thicknesses of the evanescent and high-κ layers are those discovered in Fig. 4.6. As there is more variety in the high-κ

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3Figure 4.7(a) film stack as in Fig. 4.6 except MgF$_2$ d = 93.6, 97.6, 101.6, 105.6, 109.6, 113.6, 117.6, 121.6, 125.6, 129.6, 133.6 nm. Figure 4.7(b) film stack as in Fig. 4.6 except Ru d = 4.1, 8.1, 12.1, 16.1, 20.1, 24.1, 28.1, 32.1, 36.1, 40.1, 44.1 nm.
4.7. SSR BASED EVANESCENT COUPLED ARCS

Figure 4.8: SSR ARC reflectance maps - n-κ dependence. Stack: PR|low-n|high-κ|Si. (a) n-κ space for a 27 nm high-κ layer and 115 nm MgF\textsubscript{2} evanescent layer. The global reflectance minima is a SSR with value (\tilde{n} = 2.2 + 3.6i) is very similar to that of Ru (\tilde{n} = 2.21 + 3.59i). A secondary weak minima is present at approximately \tilde{n} = 2 + 0i, this is a TM DR solution. (b) n-κ space for a 115 nm evanescent layer and a 27 nm high-κ layer (\tilde{n} = 2.2 + 3.6i). A single minima is present at n = 1.38. Note: scales differ on this figure to remove non-relevant propagating λ/4 solutions.

For naturally occurring materials this is investigated first, Fig. 4.8(a) shows the high-κ layer reflection map for n-κ space. n-κ parameter space is large enough to cover the vast majority of all naturally occurring materials including metals, dielectrics, and semiconductors, thus we should find all suitable solutions. The discovered minimum (\tilde{n} = 2.2 + 3.6i) as one would expect is very similar to that of Ru (\tilde{n} = 2.2137 + 3.5864i) which was used to produce the layer thicknesses. The 0.5% reflectance contour is a circle around this minimum. As this minimum is somewhat broad and flat the standing wave sensitivity is comparatively low for n and κ variations for this layer. A secondary local minima is also present at approximately \tilde{n} = 2 + 0i, this is a weak DR mode where the layer thicknesses and cladding materials are not suitable to produce good reflection suppression.

If we now fix the \tilde{n} of the high-κ layer to that which we discovered in the previ-

\footnote{Fig. 4.8(a) parameters: PR AZMIR701 - n = 1.6844+0.0307i \cite{119}, d = bulk, MgF\textsubscript{2} - n = 1.38 \cite{152}, d = 115 nm, high-κ - n and \kappa varied, d = 27 nm, Si - n = 5.4375+0.3420i \cite{122}, d = bulk, NA = 1.4091, TM polarization, \lambda = 405 nm. Fig. 4.8(b) parameters: PR AZMIR701 - n = 1.6844+0.0307i \cite{119}, d = bulk, low-n - n and \kappa varied, d = 115 nm, high-κ - n = 2.2+3.6i, d = 27 nm, Si - n = 5.4375+0.3420i \cite{122}, d = bulk, NA = 1.4091, TM polarization, \lambda = 405 nm}
ous plot, and produce the \( n-\kappa \) space reflectance map of the evanescent coupling layer (Fig. 4.8(b)) we see a much narrower minimum. The value of this minimum (\( \tilde{n} = 1.378 \)) is very close to that of MgF\(_2\) (\( \tilde{n} = 1.38 \)). Note the axes of this plot are different to those of Fig. 4.8(a); primarily this is to remove all propagating (\( n > NA \) and \( d = \lambda/4 \)) solutions which are not of interest here. The \( n < 1 \) and \( \kappa > 1 \) regions have also been removed as there are no local minima in these regions. The 0.5% contour centred on \( \tilde{n} = 1.38 \) includes a small amount of absorption (\( \kappa \leq 0.02 \) is allowable); this is important as it indicates that weakly absorbing evanescent coupling layers can also be employed. As both the evanescent fields and the absorption are loss mechanisms an increase in absorption can be compensated by a reduction in the evanescent layer thickness and vice versa. The narrowness of this minimum indicates the standing wave effects for this layer is more sensitive to refractive index variations than the high-\( \kappa \) layer.

### 4.7.2 SSR NA response and constraints

The intensity matching requirement for destructive interference requires the underlayer to be at or near resonance to provide enough field enhancement to match the intensities. SSRs have specific NA and material requirements for resonance to occur; this fact places fundamental limits on the range of NAs and materials capable of being employed as SSR ARCs. The fields must be simultaneously propagating within the PR (\( NA < n_{PR} \)) and evanescent within the evanescent layer (\( NA > n_{low} \)); as such the available NA space for evanescent coupled ARCs, in general, is \( n_{low} < NA < n_{PR} \).

In practice for SSR ARCs however the design NA is approximately the peak resonant NA which is determined by the two layer SSR resonance condition given by

\[
NA = \frac{\beta}{k_0} = \text{Re} \left( \sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}} \right),
\]

(4.14)

where \( \beta \) is the polariton resonance wave vector, \( k_0 \) the free space wave vector, and \( \varepsilon_i \) the material dielectric constants. Provided the incident NA is equal to this NA energy will couple into the system. The fact that the NA in Eq. (4.14) is determined solely by the material dielectric constants (refractive indices) limits the available NAs to those combinations that are physically available.

The NA response of SSR based evanescent coupled ARCs is dependent on the resonance characteristics of the systems. Figure 4.9(a) shows the intensity at an MgF\(_2\)|Ru interface versus NA, where the resonance condition is clearly visible at an NA of 1.4091. This peak is typical of surface state resonators. The narrowness of this peak indicates
that these systems will not respond well across a broad range of NAs as is seen in Fig. 4.9(b) where standing waves become more prominent as the NA shifts away from the resonance NA. This fact is not necessarily a fundamental limit in terms of modern photolithography where narrow NA exposures are often employed for large area regular patterns.

Figure 4.9: SSR NA characteristics. (a) Resonator interface intensity as a function of NA. Simple Prism|MgF$_2$|Ru|Si stack to highlight the effect of the resonator alone. (b) NA response for an SiO$_2$|PR|MgF$_2$|Ru|Si SSR ARC. The standing waves increase as the NA shifts from the peak NA (1.4091) seen in (a).

4.7.3 SSR polarization characteristics

The polarization characteristics of SSR based ARCs are heavily influenced by the fact that SSR resonance only occurs for TM polarization. This is clearly evident in Fig. 4.10 where the TM and TM reflectance and standing wave characteristics of a SSR based ARC are compared. The reflectance across NA for both TE and TM polarizations of a MgF$_2$|Ru SSR ARC is shown in Fig. 4.10(a) TM polarization reflectance increases up to an NA of approximately 0.8 where it begins to decrease as it approaches Brewster’s angle and energy (off resonantly) starts coupling into the SSR. The reflectance reaches a minimum at the SSR condition (Eq. (4.14)), then rapidly increases after that. The

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Figure 4.9(a) Prism - n = 2.25, d = bulk, MgF$_2$ - n = 1.38, d = 115 nm, Ru - n = 2.2137+3.5864i, d = 27 nm, Si - n = 5.4375+0.3420i, d = bulk, NA = varied, TM polarization, $\lambda$ = 405 nm. Figure 4.9(b) SiO$_2$ - n = 1.4696, d = bulk, PR - n = 1.6844+0.0307i, MgF$_2$ - n = 1.38, d = 115 nm, Ru - n = 2.2137+3.5864i, d = 27 nm, Si - n = 5.4375+0.3420i, d = bulk, NA = varied, TM polarization, $\lambda$ = 405 nm.
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Figure 4.10: SSR based evanescent coupled ARC polarization characteristics\(^\text{10}\): (a) Reflectance across NAs for TE and TM polarization for a MgF\(_2\)|Ru SSR ARC system.

The red dashed line indicates \( NA = n_{\text{MgF}_2}\) and blue \( NA = n_{\text{PR}}\). Note the TM case where the reflectance dips to zero at the ARC design NA. The TE case shows strong reflectance across all angles. (b) SSR ARC polarization standing wave response. The standing wave is fully suppressed in the case of the design TM polarization. For TE polarization very prominent standing waves are present indicative of strong reflectance from the evanescent layer, metal layer, and the substrate.

TE polarization reflectance in contrast steadily increases with NA. As no resonance occurs when illuminated with TE polarized light, there is no resonance dip present, and thus all we see is the expected increasing reflectance with NA.

The effect of this difference for a given SSR ARC is shown in Fig. 4.10(b). The TM polarization is a smooth intensity trace indicative of reflection suppression. The TE polarized intensity trace has strong standing waves in the PR, as is expected considering the very high reflectance (\( R > 0.9\)) shown in Fig. 4.10(a). In the past when unpolarized light was employed for photolithography this difference would be a major issue heavily reducing the feasibility of this type of ARC. Today however, modern exposures are carried out using single polarizations, which are suitable for this type of ARC.

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\(^\text{10}\)Figure 4.10(a) parameters: Prism - \( n = 2.25\) (fictitious), \( d = \) bulk, MgF\(_2\) - \( n = 1.38\) \[152\], \( d = 115\) nm, Ru - \( n = 2.2137+3.5864i\) \[153\], \( d = 27\) nm, Si - \( n = 5.4375+0.3420i\) \[122\], \( d = \) bulk, NA = varied, TE and TM polarizations, \( \lambda = 405\) nm. Fig. 4.10(b) parameters: Prism/SiO\(_2\) - \( n = 1.4696\) \[121\], \( d = \) bulk, PR AZMR701 - \( n = 1.6844+0.0307i\) \[119\], \( d = 500\) nm, MgF\(_2\) - \( n = 1.38\) \[152\], \( d = 115\) nm, Ru - \( n = 2.2137+3.5864i\) \[153\], \( d = 27\) nm, Si - \( n = 5.4375+0.3420i\) \[122\], \( d = \) bulk, NA = 1.4091, TE and TM polarizations, \( \lambda = 405\) nm.
4.7.4 SSR ARCs substrate considerations

For an underlayer to act as a destructive interference type ARC it must produce a backwards going field with the following relations to the surface reflections: 1) it must be $\pi$ out of phase with the surface reflection and 2) it must have the same intensity. These are the conditions for destructive interference, and thus reduced reflectance; the fact that this results in increased transmission however cannot be ignored. The energy from the increased transmission must be able to leave the system (via absorption or transmission through the substrate) or else the matched intensity condition will likely not be able to be met.

As SSR based evanescent ARCs inherently require a high-$\kappa$ (i.e. absorbing) underlayer, the bulk of the energy is lost in this layer as evidenced by the sharp drop in intensity within the PR in Fig. 4.11. Importantly however, not all the energy is lost in this layer, with some of it having to be lost in the substrate. This has important implications for photolithographic purposes and types of substrates capable of being employed. In Fig. 4.11 an SRR based ARC optimized for an Si substrate is simulated with Si, SiO$_2$, Ru, and MgF$_2$ substrates. As expected the Si case shows a flat PR intensity profile, with the excess energy (for this discussion we exclude that lost in the SSR layers) being lost in Si substrate. The SiO$_2$ case has an approximately flat PR intensity profile, and importantly an outgoing propagating field within the SiO$_2$ substrate which allows the excess energy to leave the system. The Ru substrate (i.e. the high-$\kappa$ layer and substrate are continuous) results in over resonance at the SSR interface, consequently a standing wave is present in the PR. The use of an evanescent substrate (MgF$_2$) prevents the excess energy from leaving the system through the substrate. This results in the energy feeding back into the PR and large standing waves appearing.

These results indicate that SSR ARCs are capable of being employed on any type of substrate which can dissipate the excess energy at the base of the SSR layers. This allows common substrates such as Si, SiO$_2$, and Al$_2$O$_3$ to be employed as well as less common substrates such as metals provided they are not the same material as the high-$\kappa$ layer, although materials which support evanescent fields are not capable of being employed as substrates. If this type of substrate is required energy dissipating layers may be employed between the ARC and the substrate.

The use of SSR based evanescent coupled ARCs is limited to using high absorbing materials, TM polarization, and at a relatively narrow NA range determined by the
SSR based ARC substrate types. For a given SiO$_2$|PR|MgF$_2$|Ru|Substrate model. We now shift to DR based evanescent coupled ARCs which are more flexible and employ non-(or low)absorbing materials, TM or TE polarization, and can be designed to operate at a wide range of NAs by altering the layer thicknesses.

4.8 DR based evanescent coupled ARCs

In Fig. 4.12 we have an example of a DR evanescent-coupled ARC, with design principles discussed in Section 4.8.1 following. This system is based on a CaF$_2$|HfO$_2$|CaF$_2$ resonator illuminated with TE polarized light at an NA of 1.45 (Fig. 4.12(a)). Figure 4.12(b) shows an intensity trace of this system with a comparison non-ARC intensity trace on a SiO$_2$ substrate. A Beer-Lambert law absorption type curve is present within the PR. Resonance is also clearly present in the ARC underlayer, although this time not confined to the interface; this is indicative of the propagating nature of the fields within the HfO$_2$ layer. Once again a clear standing wave profile is present within the PR layer in the non-ARC case, this time with an even greater amplitude. In both cases approximately 52% of the intensity is seen to be exiting through the transparent substrate; this point is significant and will be revisited in Section 4.8.4.

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Prism/SiO$_2$ - n = 1.4696 [121], d = bulk, PR AZMIR701 - n = 1.6844+0.0307i [119], d = 500 nm, MgF$_2$ - n = 1.38 [152], d = 115 nm, Ru - n = 2.2137+3.5864i [153], d = 27 nm, Si - n = 5.4375+0.3420i [122], d = bulk, NA = 1.4091, TM polarization, $\lambda = 405$ nm.
4.8. DR BASED EVANESCENT COUPLED ARCS

Figure 4.12: CaF$_2$|HfO$_2$|CaF$_2$ based DR evanescent-coupled ARC, NA=1.45 and TE polarization. (a) - Film stack. (b) - Intensity trace through the film stack, blue line indicates the ARC case where the ARC layers are shaded tea green, red line the no ARC case. Note the smooth Beer Law absorption trace within the PR when the ARC is employed. Dielectric resonance is evidenced by the up tick in intensity centred in the middle of the HfO$_2$ layer. The no ARC case (ARC layers replaced with SiO$_2$) shows prominent standing waves within the resist. As the substrate (SiO$_2$) is transparent we see a flat outbound intensity.

4.8.1 $d$, $n$ and $\kappa$ characteristics

The design criteria for DR based evanescent-coupled ARCs are different from the SSR case based on the fact that an NA can be any value between that of the $n_{\text{low}}$ and whichever is lower of $n_{PR}$ and $n_{\text{high}}$. As one typically does not know what a suitable thickness combination is a priori, generally it is more practical to start by picking a suitable $n_{\text{low}}$|$n_{\text{high}}$ combination and optimising the system to find the optimal layer thicknesses.

Figure 4.13 is an evanescent layer vs. resonator layer thickness reflectance plot for a symmetric trilayer DR ARC, with a PR|CaF$_2$|HfO$_2$|CaF$_2$|SiO$_2$ film stack, at an NA of 1.45. The minimum reflectance for this system ($R = 1.7 \times 10^{-4}$) occurs at an $n_{\text{low}}$ thickness of 149 nm and an $n_{\text{high}}$ thickness of 18 nm. The Re$(r) = 0$ and Im$(r) = 0$ contours again intersect at the minimum reflectance. Notably, this time there are

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12 Prism SiO$_2$ n = 1.4696 [121], d = bulk, PR AZMIR701 - n = 1.6844+0.0307i [119], d = 500 nm, CaF$_2$ - n = 1.4415 [152], d = 149 nm, HfO$_2$ n = 1.9804 [154], d = 18 nm, NA = 1.45, TE polarization, $\lambda = 405$ nm.
4.8. DR BASED EVANESCENT COUPLED ARCS

Figure 4.13: DR ARC thickness space reflectance map. CaF$_2$|$\text{HfO}_2$|$\text{CaF}_2$|$\text{SiO}_2$ DR film stack$^{13}$. Evanescent versus high-\(n\) layer thicknesses. Blue contours represent \(\text{Re}(r)=0\) and green \(\text{Im}(r)=0\). The intersection of these contours is the location of the reflectance minimum \((R = 1.721 \times 10^{-4})\), indicated by a white cross. Green and cyan dashed lines indicate the thickness transects for Figs. 4.14(a) and 4.14(b).

multiple places where this occurs all with the same evanescent layer thickness. These extra solutions are the higher order waveguide modes. If \(n_{\text{low}}\) is absorbing (weakly) the evanescent layer thickness will decrease to allow more energy to couple in. The high-\(n\) layer thickness will change to compensate for the differing reflection phase changes from the now absorbing evanescent layer. If \(n_{\text{high}}\) is absorbing (weakly) the evanescent layer thickness will again decrease, this time to compensate for the loss in the \(n_{\text{high}}\) layer. As the higher order minima correspond to longer path lengths in the absorbing layer, extra energy needs to couple in to compensate for this. As such the higher the order the thinner the evanescent coupling layer, to the point where \(\text{Re}(r) = 0\) and \(\text{Im}(r) = 0\) no longer intersect thereby limiting the number of usable solutions. The 0.5\% reflectance contour (white) is far narrower in the DR ARC case when compared to SSR ARCs, because of the phase condition for DR waveguide resonance these systems are very sensitive to thickness variations.

If we look at the standing waves across the evanescent layer transect (Fig. 4.14(a)) we see this system isn’t very sensitive to thickness variations in the evanescent layer and only small standing waves result despite the layer thickness being 15 nm greater or less than the optimum value of 150 nm. Altering the high-\(n\) layer thickness by the same margins however produces far stronger standing waves, particularly as the layers

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$^{13}$Prism/PR AZMIR701 - \(n = 1.6844+0.0307i\) $^{119}$, \(d = \text{bulk}\), CaF$_2$ - \(n = 1.4415\) $^{152}$, \(d = 149\) nm, HfO$_2$ \(n = 1.9804\) $^{153}$, \(d = 18\) nm, NA = 1.45, TE polarization, \(\lambda = 405\) nm.
Figure 4.14: DR ARC layer standing wave thickness dependence, and reflectance maps - n-κ dependence\(^{14}\) (a) - Intensity trace of evanescent layer thickness transect locations (green dashed line Fig. 4.13). Red and blue lines indicate thicknesses less and greater than the optimal value (black line) respectively. (b) - Intensity trace of high-n layer thickness transect locations (cyan dashed line Fig. 4.13). Note: lower PR interface represented is that of the optimum value. (c) - n-κ space reflectance map for the high-n layer (low-n=1.4415). A DR reflectance minima occurs at \(\tilde{n} = 1.985\), which is very similar to that of HfO\(_2\) (\(\tilde{n} = 1.9804\)). (d) - n-κ space reflectance map for the low-n layer (high-n=1.985). A minima is present at \(n = 1.441\). Note: different scales on this figure to remove non-relevant propagating \(\lambda/4\) solutions.

\(^{14}\)Figure 4.14(a): film stack as in Fig. 4.13 except CaF\(_2\) \(d = 135, 138, 141, 144, 147, 150, 153, 156, 159, 162, 165\) nm. Figure 4.14(b): film stack as in Fig. 4.13 except HfO\(_2\) \(d = 3, 6, 9, 12, 15, 18, 21, 24, 27, 30, 33\) nm. Figure 4.14(c): Prism/PR AZMIR701 - \(n = 1.6844+0.0307i\) \[^{119}\], \(d = \text{bulk}, \text{CaF}_2\) - \(n = 1.4415\) \[^{152}\], \(d = 149\) nm, high-n - \(n = \text{varied}, d = 18\) nm, SiO\(_2\) - \(n = 1.4696\) \[^{121}\], \(d = \text{bulk}, \text{NA} = 1.45, \text{TE polarization}\). Figure 4.14(d): Prism/PR AZMIR701 - \(n = 1.6844+0.0307i\) \[^{119}\], \(d = \text{bulk}, \text{low-n - n = varied}, d = 149\) nm, high-n - \(n = 1.985\), \(d = 18\) nm, SiO\(_2\) - \(n = 1.4696\) \[^{121}\], \(d = \text{bulk}, \text{NA} = 1.45, \text{TE polarization}\).
get thinner (Fig. 4.14(b)). This is due to the high-$n$ layer changing both the resonator intensity and the phase of the backward going fields. It can be seen that the greatest resonator intensity occurs for a thinner than optimum high-$n$ layer thickness.

Now we use the optimal thicknesses discovered in Fig. 4.13 to investigate the potential solutions in refractive index space. For the sake of making these plots 2D we choose to fix the adjacent layer materials. If we fix the $n_{\text{low}}$ index to that of CaF$_2$ we get a reflectance minima at $n = 1.985$ (Fig. 4.14(c)), which is very close to that of the design material HfO$_2$ ($n = 1.9804$). The area within the 0.5% reflectance contour is very small in this case as the system requires a very specific phase thickness to allow destructive interference to occur. When we look at the $n_{\text{low}}$ refractive index space (Fig. 4.14(d)), the discovered minimum ($n = 1.441$) is essentially equal to that of CaF$_2$ ($n = 1.4415$). The area within the 0.5% reflectance contour is very small which is indicative of the tight phase constraints on DR systems. A secondary minima is present at approximately $\tilde{n} = 1.675 + 0.8i$, which is a propagating $\lambda/4$ ARC mode. There are further higher order minima but these are all propagating and thus not relevant to this discussion; the axes are limited to the region of the first and second minima.

### 4.8.2 DR NA response and constraints

The NA response of DR based evanescent coupled ARCs is determined by the make up of the resonator. The $n_{\text{low}}|n_{\text{high}}|n_{\text{low}}$ structure requires evanescent fields to couple energy into the propagating $n_{\text{high}}$ layer. As such the design NA must be greater than $n_{\text{low}}$, but lower than the lowest refractive index of either $n_{\text{high}}$ or $n_{\text{PR}}$ to allow propagating fields in both the PR and the $n_{\text{high}}$ layer.

The maximally resonant NA for DR systems is dependent upon the thickness of the $n_{\text{high}}$ layer. This is required to match a $2\pi m$ roundtrip phase condition for resonance to occur. As such, unlike SSR based evanescent-coupled ARCs, the NA for DR based evanescent-coupled ARCs can be tailored to any NA between the limits previously discussed simply by altering the $n_{\text{high}}$ layer thickness. DR resonance are by their nature sharp due to the phase requirement (Fig. 4.15(a)). As a consequence of this they are not very tolerant to variations in the NA (Fig. 4.15(b)).

### 4.8.3 DR polarization characteristics

Figure 4.16(a) is the reflectance for TE and TM polarized light of a CaF$_2$|HfO$_2$|CaF$_2$ DR based ARC. The system employed here is optimized for TE polarization at an
Figure 4.15: DR NA characteristics - (a) - Max resonator intensity as a function of NA. Simple Prism|CaF$_2$|HfO$_2$|CaF$_2$|SiO$_2$ stack to highlight the effect of the resonator alone. The red dashed line indicates the NA ($NA = n_{CaF_2}$) beneath which all fields are propagating, and the green dashed line indicates the NA ($NA = n_{PR}$) above which the PR fields are evanescent. (b) - NA response for an SiO$_2$|PR|CaF$_2$|HfO$_2$|CaF$_2$|SiO$_2$ DR ARC. The standing waves increase as the NA shifts from the peak NA seen in (a).

NA of 1.45. The TE polarization plot increases steadily in the low NA due to the reflectance at the CaF$_2$|HfO$_2$ interface until resonator coupling begins to dominate. After this the NA reaches the optimum resonance condition for the waveguide (phase thickness = $\pi$) and hence reflectance peaks. As the NA increases the enhancement from the resonator decreases as it is now off resonance and the PR absorption begins to dominate. The TM polarization line remains relatively flat up to the Brewster angle ($NA \approx 1.17$), then peaks at the TM waveguide resonance NA and tails off as the resonant enhancement decreases and the PR absorption dominates.

4.8.4 DR based ARC substrate types

The requirement for the removal of excess resonator energy is perhaps best represented by DR based ARCs. Figure 4.17(a) shows a CaF$_2$|HfO$_2$|CaF$_2$ based ARC designed to operate on a SiO$_2$ substrate. The SiO$_2$ substrate shows a nice smooth PR intensity pro-
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Figure 4.16: DR ARC polarization characteristics\(^\text{16}\) (a) - Reflectance vs. NA for TE and TM polarization for a CaF\(_2\)|HfO\(_2\)|CaF\(_2\) DR ARC system. The red dashed line indicates \(NA = n_{CaF_2}\) and green \(NA = n_{PR}\). TE reflectance increases as the NA increases until energy starts coupling into the resonator, with a reflectance minimum at the design NA of 1.45. A rapid increase is seen after this as the resonator goes out of phase and the high angles lead to very high reflectances. The TM polarization reflectance remains low as it approaches Brewster’s angle, then its reflectance increases. (b) - Comparing the TE and TM polarizations for the fully optimized stack, it’s obvious that the primary difference is the lack of resonance in the TM case, consequently prominent standing waves are visible.

file, resonance in the HfO\(_2\) layer, and outbound propagating fields within the substrate. If absorbing/reflecting/evanescent substrates (Si, Ag, and CaF\(_2\) for example) are employed the energy from the DR is not able to escape the system through the substrate. This causes the energy to feed back into the resonator and PR, as evidenced by greater resonator intensities and standing waves within the PR. Consequently the traditional trilayer DR is only suitable for transparent substrates which support propagating fields at the design NA. The task of designing DR based ARCs for absorbing/reflecting substrates led to the development of modified trilayer DR systems.

Modified trilayer DR systems replace the lower evanescent layer with a reflecting

\(^{16}\)Figure 4.16(a): Prism/PR AZMIR701 - \(n = 1.6844+0.0307 i\) [119], \(d = \text{bulk}\), CaF\(_2\) - \(n = 1.4415\) [152], \(d = 145.5 \text{ nm}\), HfO\(_2\) - \(n = 1.9804\) [154], \(d = 27 \text{ nm}\), SiO\(_2\) - \(n = 1.4696\) [121], \(d = \text{bulk}\), NA = varied, TE and TM polarization, \(\lambda = 405 \text{ nm}\). Figure 4.16(b): Prism/SiO\(_2\) - \(n = 1.4696\) [121], \(d = \text{bulk}\), PR AZMIR701 - \(n = 1.6844+0.0307 i\) [119], \(d = 500 \text{ nm}\), CaF\(_2\) - \(n = 1.4415\) [152], \(d = 145.5 \text{ nm}\), HfO\(_2\) - \(n = 1.9804\) [154], \(d = 27 \text{ nm}\), NA = 1.45, TE and TM polarization, \(\lambda = 405 \text{ nm}\).
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Figure 4.17: DR ARC substrate effects\textsuperscript{17} (a) - SiO\textsubscript{2}|PR|CaF\textsubscript{2}|HfO\textsubscript{2}|CaF\textsubscript{2}|Substrate model with SiO\textsubscript{2}, Si, Ag, and CaF\textsubscript{2} substrates. Resonance in the HfO\textsubscript{2} layer is strong in the non-SiO\textsubscript{2} cases resulting in prominent standing waves in the PR. (b) - Modified DR ARC with a Si substrate. Note the significantly different layer thicknesses compared to the SiO\textsubscript{2} optimized system in (a).

substrate, for example in Fig. 4.17(b) the lower CaF\textsubscript{2} layer from Fig. 4.17(a) has been removed and the substrate changed to Si. The role of the bottom evanescent layer is to confine the fields within the high-\(n\) layer to allow energy to build up. Similarly a strongly reflecting material can also be employed, consequently materials such as Si and Ag are capable of being employed as substrates. The use of a non-TIR reflecting layer changes the reflection phase relations within the high-\(n\) layer. To account for change in reflection phase the layer thicknesses change from \(d_{\text{CaF}_2} = 145.5\) nm and \(d_{\text{HfO}_2} = 18\) nm in Fig. 4.17(a) to \(d_{\text{CaF}_2} = 86\) nm and \(d_{\text{HfO}_2} = 91\) nm in Fig. 4.17(b). The thickness of the HfO\textsubscript{2} layer changes to satisfy the resonance condition, while the thickness of the CaF\textsubscript{2} layer reduces to couple more energy into the resonator to compensate for losses at the HfO\textsubscript{2}|Si interface.

From a manufacturing stand point a modified DR structure is beneficial as it uses one less layer and can be manufactured directly on Si. Indeed this is one of the forms we sought to experimentally verify in Chapter 5 using a SiO\textsubscript{2}|HfO\textsubscript{2}|Si DR based ARC.

\textsuperscript{17} Figure 4.17(a) Prism/SiO\textsubscript{2} - \(n = 1.4696\) \textsuperscript{121}, \(d = \text{bulk}\), PR AZMIR701 - \(n = 1.6844+0.0307i\) \textsuperscript{119}, \(d = 500\) nm, CaF\textsubscript{2} - \(n = 1.4415\) \textsuperscript{152}, \(d = 145.5\) nm, HfO\textsubscript{2} - \(n = 1.9804\) \textsuperscript{154}, \(d = 27\) nm, CaF\textsubscript{2} - \(n = 1.4415\), \(d = 145.5\) nm, SiO\textsubscript{2} - \(n = 1.4696\), \(d = \text{bulk}\), NA = 1.45, TE polarization, \(\lambda = 405\) nm.

Figure 4.17(b) Prism/SiO\textsubscript{2} - \(n = 1.4696\), \(d = \text{bulk}\), PR AZMIR701 - \(n = 1.6844+0.0307i\), \(d = 500\) nm, CaF\textsubscript{2} - \(n = 1.4415\), \(d = 145.5\) nm, HfO\textsubscript{2} - \(n = 1.9804\), \(d = 27\) nm, Si - \(n = 5.4375+0.3420i\) \textsuperscript{122}, \(d = \text{bulk}\), NA = 1.45, TE polarization, \(\lambda = 405\) nm.
4.9 Evanescent-coupled ARCs for 193i photolithography

Evanescent-coupled ARCs fundamentally require illumination by a polarized high-NA source. This severely limits the utility of these ARCs, but at the same time points to their most likely application, that of modern hyper-NA photolithography which operates within these constraints, thus we will now look at its potential application to 193i photolithography.

Evanescent-coupled ARCs are a potential BARC solution for 193i photolithography. The use of these evanescent-coupled ARCs is heavily dependent on the availability of suitable low refractive index materials. As the industry has already indicated that it is not likely to shift to next-gen immersion fluids, the $n_{\text{low}}$ coupling layer must have a refractive index less than $n_{\text{H}_2\text{O}} = 1.44$. This places severe limits on the available materials; with the lowest refractive index of a commonly available material at a wavelength of 193 nm being 1.385 for NaF [155]. As the highest NA used for 193i is approximately 1.35, NaF is clearly not suitable for use with 193i photolithography. As such we look to more ‘exotic’ alternatives. To meet the challenges of ARC coatings at 193 nm Nikon has developed a nanocomposite material with a refractive index of 1.18 at 193 nm [129]. This is employed on the projection lenses of IC steppers and scanners. If we use this same material we are able to formulate evanescent-coupled ARCs for 193i photolithography.

At this wavelength $\kappa$ for most metals decreases due to poor coupling of energy to charges when the driving frequency exceeds the plasma frequency of the material [63]. This not only limits the amount of resonance possible, but also reduces the energy loss, consequently the range of materials available for SSR based ARCs is severely limited at 193 nm. Aluminium however is known to be a suitable plasmonic material in the deep UV [156, 157]. Figure 4.18 shows intensity contours for exposure of two SSR ARCs examples in an IL configuration, Fig. 4.18(a) in the Otto configuration, and Fig. 4.18(b) in the Kretschmann configuration, operating at an NA of 1.3904 which is pushing the limits of modern 193i systems. The Otto configuration was used in the previous examples (Section 4.7) where the low-$n$ layer is above the high-$\kappa$ layer. The Kretschmann configuration is the reverse of this with the high-$\kappa$ layer above the low-$n$ layer. This is the reason for the inversion of the fields within the resonator layers. In both cases resonances are visible at the Nikon|Al interface, with the Kretschmann example showing a secondary resonance at the $n_{\text{low}}$|Si substrate interface.
4.9. EVANESCENT-COUPLED ARCS FOR 193I PHOTOLITHOGRAPHY

Figure 4.18: 193i SSR evanescent-coupled ARC examples\textsuperscript{18} (a) - Otto configuration. Film stack: PR|Nikon material|Al|Si. Note the good intensity profile within the PR and the presence of plasmon resonance at the Nikon|Al interface. (b) - Kretschmann configuration. Film stack: PR|Al|Nikon material|Si. Note the good intensity profile within the PR and the presence of plasmon resonance at the Nikon|Al interface as well as a secondary resonance as the Nikon|Si interface.

At a wavelength of 193 nm the refractive index of Si is $n_{Si} = 0.89 + 2.67i$ \textsuperscript{159}; this is essentially a metal from an optical standpoint, thus in Fig. 4.18(a) the substrate acts as a strong reflector. This strong reflectance coupled with insufficient loss in the SSR means too much energy is back-coupling into the PR, resulting in (weak) standing waves. Although the Si substrate itself cannot be used as an ARC an interesting variant of this system utilizes it by having a PR|Ge|n$_{low}$|Si type structure (Fig. 4.19(b)). This system has dual resonances occurring within a very thin Ge layer and the n$_{low}$|Si interface. Usefully this system operates and a certainly achievable NA of 1.26.

The same Nikon low index material can be employed to construct DR based ARCs at 193 nm as well. Energy loss can be achieved through an absorbing high-$n$ layer, an absorbing/reflecting substrate or a transparent substrate. ‘Complete’ suppression of standing waves caused by the metallic nature of a Si substrate is an issue at this wavelength. These standing waves however can be effectively suppressed by using weakly absorbing resonator materials.

\textsuperscript{18}Figure 4.18(a) Prism/PR - $n = 1.7 + 0.02i$ \textsuperscript{10}, d = bulk, Nikon material - $n = 1.18$ \textsuperscript{129}, d = 58.7 nm, Al - $n = 0.1143 + 2.2186i$ \textsuperscript{158}, d = 10.5 nm, Si - $n = 0.8900 + 2.6700i$ \textsuperscript{159}, d = bulk, NA = 1.3904 TM polarization, $\lambda = 193$ nm. Figure 4.18(b) Prism/PR - $n = 1.7 + 0.02i$ \textsuperscript{10}, d = bulk, Al - $n = 0.1143 + 2.2186i$ \textsuperscript{158}, d = 17.1 nm, Nikon material - $n = 1.18$ \textsuperscript{129}, d = 105.5 nm, Si - $n = 0.8900 + 2.6700i$ \textsuperscript{159}, d = bulk, NA = 1.3904 TM polarization, $\lambda = 193$ nm.
4.9. EVANESCENT-COUPLED ARCS FOR 193I PHOTOLITHOGRAPHY

Figure 4.19: Ge/Si 193i SSR evanescent-coupled ARC. Film stack: PR|Ge|Nikon material|Si. (a) - Intensity trace. Note the dual SSR resonances in the Ge layer and the Si interface. (b) - 2D field simulation for the film stack in (a). Again the resonance at the Si interface is obvious.

The reduction in $\kappa$ at this wavelength has some interesting implications for DR ARC design. That is, due to the decreased $\kappa$ some materials become transparent enough to act as evanescent coupling layers, for example Y ($n = 1.0369 + 0.0723i$) [160] and PbSe ($n = 0.68 + 0.78i$) [155]. Crucially these refractive indices are less than the refractive index of water at this wavelength so are capable of acting as evanescent coupling layers for the NAs regularly employed in 193i systems. Curiously PbSe is capable of operating in the low NA regime.

193i photolithography is in many respects a natural fit for evanescent-coupled ARCs. The required high-NA and single polarization exposure conditions are fundamental to both technologies. The narrow NA range of evanescent-coupled ARCs is applicable to regular grid type patterning which is a key component to modern gridded design rule photolithography layouts. Ultimately, the use of this ARC will come down to the benefits it offers relative to existing ARC methods.

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Figures 4.19(a) and 4.19(b): Prism/PR - n = 1.7+0.02i [10], d = bulk, Ge - n = 1.01 + 2.05i [155], d = 17.1 nm, Nikon material - n = 1.18 [129], d = 105.5 nm, Si - n = 0.8900+2.6700i [159], d = bulk nm, NA = 1.3904 TM polarization, $\lambda = 193$ nm.
4.10 Summary

In this chapter the concept of evanescent-coupled ARCs for high-NA photolithography applications was introduced. These ARCs are based on the concept of resonant underlayers which produce a backwards going field to destructively interfere with the incident fields within the PR, thus acting as an ARC. Two types of resonators are available, SSR and DR resonators. The design and characterization of example ARCs based on each of these resonator types was carried out using the transfer matrix method. The characterization process led to the discovery of general design factors for the application of each type of evanescent-coupled ARCs. These design factors are largely dependent on the type of resonator employed, polarization, the desired NA, and the type of substrate required.

- SRR ARCs design factors
  - Polarization - TM
  - NA range - Limited to resonant NA of available material combinations
  - Film stacks - Otto and Kretschmann configurations

Figure 4.20: 193i DR SSR ARC. Film stack: PR|PbSe|Al2O3|PbSe|Si[20] (a) - Intensity trace. Note the resonance in the DR layers. (b) - 2D field simulation for the film stack in (a). Again the resonance in the DR layers is obvious.

\[ \text{Figures 4.20 and 4.20(a): } \]

Prism/PR \( n = 1.7 + 0.02i \) [10], \( d = \text{bulk} \), PbSe \( n = 0.6800 + 0.7800i \) [155], \( d = 19 \text{ nm} \), Al2O3 \( n = 2.08 \) [77], \( d = 16 \text{ nm} \), Si \( n = 0.8900 + 2.6700i \) [159], \( d = \text{bulk} \), NA \( = 0.75 \) TE polarization, \( \lambda = 193 \text{ nm} \).
4.10. SUMMARY

- Materials - Lossy dielectrics, metals, and semiconductors
- Substrates - All possible substrates

- DR ARCs design factors
  - Polarization - TE and TM
  - NA range - All values between $n_{low}$ and $n_{PR}$
  - Film stacks - Slab waveguide - traditional trilayer or modified trilayer with a strongly reflecting lowest layer
  - Materials - Low loss dielectrics
  - Substrates - Traditional trilayer - transparent substrates. Modified trilayer - all possible substrates.

The greatest strength of evanescent-coupled ARCs is the ability to utilize a far broader range of materials than standard $\lambda/4$ ARCs (Fig. 4.21). This flexibility may potentially be of use for 193i lithography where an underlayer technology has been developed to improve processing economics by using layers for multiple purposes.

![Figure 4.21](image)

**Figure 4.21:** Planar layer ARC materials in refractive index space - Clearly evanescent-coupled ARCs massively increase the range of available ARC materials. Standard $\lambda/4$ ARC materials are within the blue circle, DR evanescent-coupled ARC materials within the green striped area, and SSR evanescent-coupled ARC materials within the grey striped area. Note: ‘x’ replaces overlapping material names. $n$ and $\kappa$ are wavelength dependent, thus they may not be exactly where the reader expects.
The requirement for an evanescent-coupling layer \((n < NA)\) in evanescent-coupled ARCs pushes the NA of operation into the high-NA regime \((NA > 1)\) due to the lack of transparent ultra-low refractive index materials. For this reason, and the requirement for underlayer flexibility, high-NA (hyper-NA) photolithography is seen as the natural application for these ARCs as exposures are typically carried out in this regime. To this end the applicability of evanescent-coupled ARCs for 193i lithography was also explored with candidate systems of both SSR and DR based evanescent-coupled ARCs developed.
Chapter 5

Experimental verification of Evanescent-coupled ARCs

In the previous chapter evanescent-coupled ARCs were introduced. Modelling and theory was carried out to investigate the types available and the operating characteristics of each. In this chapter we seek to experimentally verify this effect for the first time. Two example evanescent-coupled ARC systems were manufactured for this purpose. Both systems were tested in a Lloyd’s mirror solid immersion interference lithography apparatus. For the sake of ease of reading, the bulk of the details of the experimental setup and method are shifted to Appendix A. A brief but complete overview of the experimental process and experimental design considerations will be given. Experimental results will then be presented and the results discussed.

5.1 Experimental overview

In this section the fundamentals of the experiments are presented with an emphasis on the experimental apparatus, film stacks, and processing details. Whilst many of these techniques are standard, the resonant nature of the ARC system meant that care was needed with alignment and exposure procedures to ensure that repeatable and reliable results could be obtained.

5.1.1 Experimental apparatus

The experimental apparatus used for these experiments is a Lloyd’s mirror interference lithography system (Section 2.2). Figure 5.1 shows a schematic for the system utilized
5.1. EXPERIMENTAL OVERVIEW

**Figure 5.1:** Experimental setup - Light from the fibre coupled laser is projected onto the back corner of a cubic prism. One of the faces of the back corner is mirrored, the other has the sample affixed. The beam-half reflected from the mirror interferes with the other beam-half on the sample. Notes: 1) The fibre acts as a spatial filter to clean up the laser beam. 2) The mask is to reduce spurious reflections from the prism edges. 3) A half-wave plate is inserted between the collimating lens and the mask to change the polarization from TE to TM. 4) The prism is mounted on a rotation stage upon an x-translation stage to allow the correct NA to be dialled in and the beam to be centred on the back corner 3-4 of the prism.

in this work consisting of a laser source, beam transport system, and a prism and prism holder (not shown).

Lloyd’s mirror interference lithography systems require (for optimum performance) a coherent, narrow linewidth, and spatially filtered light source. For the purpose of these experiments a linewidth narrowed polarization maintaining single mode fibre coupled 405 nm wavelength diode laser was acquired \[161\]. For an interferometer to produce well defined interference fringes the coherence length must be greater than the path length difference between the beams \[105\]. The coherence length of this particular laser is specified as more than 1 m which is far greater than the approximately 25mm path length difference in our Lloyd’s mirror system. The coherence length is inversely proportional\(^1\) to the linewidth of the laser beam, thus by extension, a narrow linewidth laser is also required. A narrow linewidth is required to reduce the feature ‘blurring’ and beat patterns that arise from broadband laser sources. The laser employed here uses a volume Bragg grating to produce an output beam linewidth of \(\lambda_{FWHM} < 1 \text{ pm} \) (1.83 GHz) which is sufficiently narrow for our purposes.

To provide a nice clean interference pattern spatial filtering of the beam is required,

\(^1\)Typically this relation is given as \(\Delta \lambda \approx \frac{\lambda^2}{\Delta x}\) in the wavelength domain, or \(\Delta v \approx \frac{c}{\Delta x}\) in the frequency domain, where \(\Delta \lambda\) is the wavelength linewidth (FWHM), \(\lambda\) the central wavelength, \(\Delta x\) the coherence length, \(\Delta v\) the frequency linewidth (FWHM), and \(c\) the speed of light. As lineshape differs between different laser types these formulas are only approximations \[162\]-\[163\].
which removes noise from the system such as scatter from objects on the optics as well as from any other lasing spatial modes. The laser employed here takes its output through a fibre coupled single mode polarization maintaining fibre, which provides the spatial filtering. The waveguiding condition for the fibre is only satisfied for a narrow range of NAs, with all NAs outside this range strongly attenuated in the fibre cladding; as such the fibre acts as a very good spatial filter outputting a clean circular TEM00 beam for interference. As polarized light is required for these experiments a polarization maintaining fibre was used; provided the fibre is not bent too sharply the beam polarization exiting the fibre should be essentially the same as that coupled into the fibre. The output ferule of the fibre is affixed to a rotational mount allowing the output polarization (and polarization ratio) to be optimized.

The beam transport system employed included a collimating lens, an electronic shutter, an optional half-wave plate, and a beam mask. As the beam from the fibre is highly divergent a collimating lens is employed to allow the beam to be projected on a distant prism allowing other optical components to be placed in the intervening space. The electronic shutter allows precision exposure time control. The laser output is in the TE polarization orientation relative to the sample configuration. To produce TM polarization a half-wave plate is employed.

A square beam mask is used after the half-wave plate primarily to cut the beam shape to reduce spurious reflection and scatter from the prism edges. The back side of the beam mask is painted with a fluorescent material; this allows the centralization of the beam on the back corner of the prism to be ascertained. When an angle (NA) is dialled in for the prism, provided the prism is level, the beam will reflect back down its own path thus if the beam is slightly diverging the fluorescent paint around the mask aperture will glow showing the corner edge as a dark line in the middle of the back reflected beam. The prism translation stage can then be shifted across the path of the beam allowing the dark line to be centralized in the middle of the incident and reflected beams.

A soda-lime glass cubic prism was employed to construct the immersion Lloyd’s

\[2\text{Note this is generally considered a very bad thing and is well known for damaging lasers. In this case however it is unavoidable and indeed required for accurate pitch production. Ideally an optical isolator should be used to prevent the possibility of damaging feedback to the laser. In our case the chances of damaging the laser are deemed to be very slim (but not zero) due the extremely fine adjustments required to couple appreciable levels of power into the approximately 3 micron fibre aperture. To further reduce the likelihood of damage the half-wave plate is placed at a slight angle to deflect the beam away from the fibre head.}\]
mirror system for this work. The in-plane facets of the prism (see Fig. 5.1) are as such 1) absorbing, 2) polished, 3) polished, and 4) polished and mirrored; the incident beam should not touch the top and bottom facets, as such they can be finished as desired. In this case the non-critical facets (1, top, and bottom) were painted with black nail-polish to improve absorption of spurious reflections. Also as the sample size is much smaller (approximately a quarter) than the facet size, the non-sample areas of facet 3 were painted with fluorescing green apple colour nail-polish as an aid for locating the beam. The choice of prism glass is dictated by the range of NAs required for the experiment. In practice a cubic prism can only access NAs from $0 < NA < 1$ when illuminated through facet 1, and

$$n_{\text{prism}} \sin(90° - \sin^{-1} \left( \frac{1}{n_{\text{prism}}} \right)) < NA < n_{\text{prism}}$$

when illuminated through facet 2. The refractive index of soda-lime glass is 1.5366, which equates to an available NA range through facet 2 of $1.167 < NA < 1.537$. The two design ARCs for this chapter have design NAs of 1.4046 and 1.5 respectively, both of which are in the range of accessible NAs for this prism. Now that the experimental apparatus has been described (with full details given in Appendix A) the experiment ARC film stacks and processing considerations will be discussed.

5.1.2 Film stacks

Three different film stacks were used in these experiments: a control stack of PR on silicon; a MgF$_2$|Ru SSR based ARC; and a SiO$_2$|HfO$_2$|Si DR based ARC (Fig. 5.2). The design of these ARCs was carried out using the methods described in Chapter 4. The design NAs for these systems are 1.4046 and 1.5 for the SSR and DR based ARCs respectively. As both of these NAs are in the high-NA regime an IML must be used to prevent intensity loss through evanescent decay between the prism and the sample. The IML employed here is a Cargille Labs Series A IML with $n_{486nm} = 1.543$. Although the IML is not perfectly matched to the prism refractive index ($\Delta n \approx 0.01$), this is not an issue here as it does not impact the relative phases of the destructively interfering waves at the PR|ARC interface. This has been verified using the transfer matrix method where the ARC effect is shown to be effective regardless of whether or not an IML is present. Without an IML however an air gap between the prism and

\[3\text{Although one would prefer to have the design NAs closer to the centre of the NA range, after much experimental time and frustration with an incorrect prism material a soda-lime glass prism was accepted as a suitable option.}\]
5.1. EXPERIMENTAL OVERVIEW

Figure 5.2: Experimental film stacks\(^4\) (a) Control stack - suitable for both SSR and DR experimental comparisons. (b) MgF\(_2\)|Cr SSR ARC stack - design NA=1.4046, TM polarization. (c) SiO\(_2\)|HfO\(_2\) DR ARC stack - design NA=1.5, TE polarization.

film stack would be present (due to the difficulty of imposing intimate contact); very large evanescent coupling losses would occur within this air gap, thus in practice an IML is an absolute necessity for these experiments. The IML employed here is suitably well matched for these experiments resulting in a less than 1% Fresnel reflectance for both ARC experiments.

The IML strongly attacks the resist, thus an approximately 12 nm thick poly-vinyl alcohol (PVA) barrier layer is used to prevent this \(^{[109]}\). Both the IML and PVA layers are removed by a simple water rinse after exposure and before development. The IML serves a secondary purpose of adhering the sample to the prism by the surface tension of the IML against the sample and the prism. Experiments have shown that, provided a minimal amount of IML is applied, the sample will not move over the course of a standard exposure (\(t \sim 1\) min). An approximately 1.3 \(\mu\)m thick PR layer is used in these experiments. Provided a prebake of the wafer is carried out good PR adhesion is achieved for both the control and SSR samples, this however is not the case for the DR samples where SiO\(_2\) is known to suffer from poor PR adhesion. To fix this an adhesion promoter HMDS is spin coated on the sample before the PR is applied providing good PR adhesion.

\(^4\)Refractive indices: Soda-lime glass \(n = 1.5366^{[164]}\), IML \(n = 1.543^{[165]}\), PVA \(n \approx 1.5^{[166]}\), AZMiR701 PR \(n = 1.6844 + 0.0307i^{[119]}\), Si \(n = 5.4375 + 0.3420i^{[122]}\), MgF\(_2\) \(n = 1.38^{[167]}\), Cr \(n = 2.0356 + 2.8804i^{[168]}\), SiO\(_2\) \(n = 1.4696^{[121]}\), and HfO\(_2\) \(n = 1.9804^{[154]}\)
5.1.3 Photolithography processing

The lithography process for these experiments is a four stage process: spin coating, bake, exposure, and development. Spin coating is a process for applying thin films to a sample by dropping the coating liquid on a sample and spinning it until a uniform layer of material is coated on the wafer; this method was used to apply the PR and PVA layers. A pre-exposure softbake is performed to drive off the remaining solvent in the PR layer.

The exposure process is dictated by the NA and the dose. The NA is dialled in using the rotation stage on the prism mount. The optimum dose for these experiments is determined by performing a dose sweep and settling on a dose that provides (ideally) exposure to base of the PR, without over-exposing the PR and suffering heavy PR lift-off. As will be discussed later, this is difficult to achieve for the MgF$_2$|Cr SSR based ARC experiments. Dose in these experiments is given as a ‘Pseudo-Dosage’ (PD), which is defined as

$$PD = Power(mW) \cdot time(mins).$$

The power is typically measured using a power meter with a 9.5 mm diameter sensitive area at a point near where the beam intercepts the prism. As the beam is collimated the location of this measurement is not critical, and in practice the crucial aspect of this is that it is always measured in the same place for comparative purposes between experiments. As the beam strikes the PR within a solid medium through an IML it is not necessarily a simple task to determine the actual physical dose (mJ/cm$^2$) incident upon the PR considering the multiple reflections and coupling efficiencies at play. As the purpose of this research is to show an effect produced by a dose, rather than say by its dose characteristics, we bypass the need for an actual physical dose by using a PD which allows for a good simple comparison between experiments.

After exposure the IML and PVA layers are removed with a simple water wash. Development is then carried out using the ‘puddle’ method for 60 s using 2.38% dilute tetramethylammonium hydroxide (TMAH) developer at a ratio of 2:1 (TMAH:H$_2$O). Both weaker and stronger developer mixtures were trialled with a 2:1 ratio deemed the most suitable. This value allowed for a reasonably long development period without over-developing the sample as was seen with higher concentration trials. As will be discussed in the following section a, cleave-then-develop method was found to be the most suitable method for these experiments.

Often a further post-exposure bake is applied for photolithography processes, in
this case however this is forbidden as it is known to cause migration of photo-acids in
the PR which reduces the visibility of standing waves. Further details on the experi-
mental setup, IMLs, spin coating, baking, and development procedures are given in
Appendix A.

5.2 Experimental design considerations

To achieve experimental verification of an ARC’s effectiveness in a photolithography
setting one must (ideally) show upright structures with no standing waves present, in
comparison to a control sample which shows standing wave effects (Fig. 5.3). To do this
one must consider the standing wave period and how it compares to the PR thickness
and consequently the PR aspect ratio (height:pitch). The ARC designs investigated in
these experiments have NAs of 1.4046 and 1.5, which correspond to pattern (horizontal)
pitches of 144.2 nm and 135 nm, and (vertical) standing waves pitches of 217.8 nm and
264.3 nm respectively. To unambiguously show standing waves the PR thickness must
be at least twice the standing wave pitch. If the PR thickness were equal to the
standing wave pitch one may be able to show a bridging layer at the surface indicative
of a standing wave, but the same bridging layer may also be produced by processing
defects, thus a least twice the standing wave pitch is considered a suitable thickness.

Figure 5.3: Cross-section of PR lines without and with an ARC. Standing waves are
clearly evident in the case without an ARC. Note the standing wave pitch is far less
than the grating pitch. Source: http://www.brewerscience.com/products/arc/

A second but equally important factor when considering the PR thickness is the
aspect ratio (height:pitch). For pattern transfer purposes it is beneficial to have an
aspect ratio as high as possible to improve etch selectivity. At micron scale pitches
aspect ratios greater than 4:1 are readily achievable, at submicron scale pitches however
the aspect ratio steadily decreases to the point where 1:1 is considered good. This is due
to the balance of forces during the development, rinse and drying stages. As the water
dries a meniscus forms between neighbouring structures, the surface tension from the meniscus pulls the structures over producing resist collapse. At higher pitches this has less of an impact due to the greater surface contact area at the base of the structure. The pitches for these experiments (135 nm and 144 nm) are deeply submicron, thus the twice standing wave pitch PR thickness requirement results in aspect ratios of 3:1 and 4:1 respectively, both of which are very hard to achieve at this pitch without resist collapse. This is compounded by the presence of narrow points which also break due to strong standing waves. To bypass this issue we use a particular cleave-then-develop development process.

5.2.1 Cleave-and-develop process

Structures such as those in Fig. 5.3 are achieved using a develop-then-cleave process. In this process the sample is optimally exposed such that development proceeds through to the substrate and the aspect ratio is not so great that resist collapse occurs. Thus when the sample is cleaved after development to study the resist profile standing structures with full clearance to the substrate are produced. For the extremely challenging pitch and aspect ratios we desire this method is not suitable, so we employ the cleave-then-develop method.

These two development processes are illustrated in Fig. 5.4. Assuming full exposure to the base of the PR is achieved and strong standing waves are present, these two development processes will produce two different results. The presence of strong standing waves produces ‘bridging’ layers. In the case of the develop-then-cleave process the bridging layers prevent the development from proceeding to subsurface PR exposure structures, thus cross-section views of the resist show only shallow depressions at the surface (shown experimentally in Fig. 5.5(a)). In the cleave-then-develop process however these structures are exposed to the developer and consequently developed laterally from the cleaved surface. Thus the cross-section view of the resist will in addition to the shallow surface depressions show multiple exposed ellipses extending to the base of the PR (shown experimentally in Fig. 5.5(b)). The ability to show these subsurface exposure profiles is the strength of the cleave-then-develop process. Provided the development is balanced correctly in the cleave-then-develop process, development will only proceed laterally a short distance in from the side. This allows the remaining undeveloped PR to act to support and stabilise the developed standing wave structures, even at very high aspect ratios. For this method to be successful the PD must be carefully balanced so as to allow patterns to be developed to the base of the PR.
5.2. EXPERIMENTAL DESIGN CONSIDERATIONS

Figure 5.4: Development processes. The Develop-then-cleave process begins with an exposure (exposed areas in yellow) and an IML-PVA rinse. Development is then carried out (developed areas in blue). The development only proceeds a short distance into the PR. The sample is then cleaved and the side-on profile examined with an SEM. The observed final structure shows only shallow surface depressions. The Cleave-then-develop process likewise begins with an exposure and an IML-PVA rinse. The sample is then cleaved and then developed. Cleaving the sample before the development stage allows the subsurface exposed areas to be developed. These are observed in the side-on profile of the final PR structure as subsurface ellipses.
without removing too much PR, else the support structures suffer resist collapse.

Use of a cleave-then-develop process was crucial in developing the experimental method of this chapter. Early trials employed thin (\(\sim 500\) nm thick) PR layers and used the develop-then-cleave method, with the aim of producing images such as those in Fig. 5.3. Experimental results often showed either complete PR lift-off (over dosed and/or developed), shallow surface depressions (under dosed and/or developed), or ‘stubs’ indicative of vertical PR structures breaking off due to over dosing and/or over development. The prevalence of these three results made it very difficult to discern between processing errors and/or ARC manufacturing errors, and consequently the discovery of a usable process window. To improve the ability of successfully discerning between these two types of errors and consequently discovering a usable process window, a set of cleave-then-develop protocols were developed using thick (\(\sim 1300\) nm) PR films. This allowed for the successful imaging of the subsurface PR exposure profile, optimization of the experimental process, and assessment of ARC performance. The shift from a thin to thick PR layer also impacted the imaging methods by which the effectiveness of the ARC was assessed, thus at this point it is worth considering what is the best imaging modality to verify these experiments.

5.2.2 Imaging methods (AFM vs. SEM - surface vs. cross section imaging)

An effective ARC should remove the bridging layers produced by standing layers. For the initial thin PR layer (develop-then-cleave) experiments atomic force microscope (AFM) imaging was employed. Provided the PR thickness is greater than the standing wave pitch, the removal of bridging layers will result in an extra deep AFM profile. An example AFM transect is shown in Fig. 5.5(c), where a regular pitch grating is observed with several ultra deep troughs. Notably these ultra deep troughs terminate in a sharp point. This is indicative of the AFM cantilever being too broad to reach the bottom of the grating. In the 2D AFM scans of this sample these same features present as dark areas (Fig. 5.5(d)). In this figure it is notable that the grating lines appear to ‘waver’ due to partial grating collapse. This indicates another issue with the use of AFM imaging, that of the discernment between deep troughs caused by the removal of standing waves and those due to partial grating collapse. For these reasons AFM imaging was discarded as the imaging method for verifying ARC effectiveness. AFM imaging however remained useful to this research as a calibration method for verifying
5.2. EXPERIMENTAL DESIGN CONSIDERATIONS

Figure 5.5: Development method comparison and imaging modalities. (a) - Develop-cleave cross-section SEM image. Standing wave bridging layers prevent development from proceeding to deeper subsurface exposed areas. (b) - Cleave-develop cross-section SEM image. Development after cleaving the sample exposes the lower exposed areas to development, thus the subsurface exposure profile is visible. This is the optimum development and imaging method for these experiments. (c) - AFM transect. A regular grating structure is present with deeper troughs exhibiting sharp ends indicative of the AFM tips not reaching the bottom of the trough. (d) - In-plane AFM image. A regular grating structure is visible. Black regions are areas where the top bridging layer has been broken through. Grating lines are wavy due to resist collapse.
the grating pitch and hence the NA. To overcome the issues inherent in AFM imaging of these structures scanning electron microscope (SEM) imaging of the cleaved face of the sample was chosen.

SEM imaging of the cleaved face of the sample shows the subsurface exposure profile within the PR (Figs. 5.5(a) and 5.5(b)). Although this method is slightly more involved than AFM imaging, the images clearly show structures multiple standing wave pitches deep and thus are suitable for observing standing wave suppression and the PR footing.

5.2.3 Characteristics of standing wave suppression

A further but very important consideration is what standing wave suppression will actually look like. To investigate this the intensity within different film stacks is simulated to see how the PR intensity profile is affected (Fig. 5.6). For Figs. 5.6(b) to 5.6(d) the intensity has been reversed to produce an image with similar colour tones as the SEM figures in this chapter. A positive tone PR is used, thus white areas represent unexposed areas, while black areas represent exposed and thus developed away areas. For an optimum ARC (Figs. 5.6(a) and 5.6(b)), the intensity profile within the PR (bounded by blue lines in Fig. 5.6(b)) is uniform with no standing waves present. In contrast, for an overly resonant ARC standing waves are present with a high intensity spot at the base of the PR (Fig. 5.6(a)); this corresponds to a developed away semi-ellipse at the base of the PR (Fig. 5.6(c)). For the control stack strong standing waves are present with a reflection node present at the base of the PR (Fig. 5.6(a)); this is more clearly visible in Fig. 5.6(d) where the base of the PR remains undeveloped.

From Fig. 5.6 it is obvious that besides the removal of the standing wave pattern, there is a secondary measure available for resonant underlayer ARCs. If we consider the footing (bottom of the PR) of the PR structures we see that evanescent-coupled ARCs have a significantly different footing than that of the non-ARC case. This is due to the influence of the resonator on the phase and intensity at the base of the PR, which leads to a different PR profile compared to that of the non-ARC case. This indicates the resonator is resonating and in principle the standing wave is being altered. This is an important fact as it allows us to state that although the system may not be fully suppressing the standing wave, it is doing so albeit in a sub-optimal fashion. This leads us to the experimental goals of this chapter which are:

1. Show full standing wave suppression indicated by the full removal of horizontal bridging layers
5.2. EXPERIMENTAL DESIGN CONSIDERATIONS

Figure 5.6: ARC footing - MgF$_2$|Ru based SSR ARC intensity trace; TM polarization and NA=1.4091. Importantly the three cases have vastly different intensities at the base of the PR. The overly resonant trace is achieved by reducing the real component of the Ru refractive index from $n = 2.2137 + 3.5864i$ to $n = 1.2137 + 3.5864i$ to produce a stronger resonance. (b)-(d) - 2D intensity distribution for the plots in (a). For the sake of clarity white lines have been added to demonstrate the likely PR structures visible in SEM images indicated by the white/grey areas. These lines represent the $1/e$, $1/e^{0.5}$, and $1/e$ intensity contours. From these plots it is obvious the three cases have visibly different footing, which should allow discernment between the three possible outcomes. Note the PR is bounded by the blue lines.

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Parameters: Optimum ARC - Film stack SiO$_2$|PR|MgF$_2$|Ru|Si. SiO$_2$ - $n = 1.470$\cite{121} $d =$ bulk, AZMIR701 PR - $n = 1.684 + 0.031i$\cite{119} $d =$ 500 nm, MgF$_2$ - $n = 1.38$\cite{167} $d =$ 115 nm, Ru - $n = 2.214 + 3.568i$\cite{153} $d =$ 27 nm, Si - $n = 5.438 + 0.342i$\cite{122} $d =$ bulk. Overly resonant ARC - As in the optimum ARC example except the Ru refractive index has been reduced to $n = 1.214 + 3.568i$. No ARC - As in the optimum ARC example except the MgF$_2$ and Ru layers replaced with Si.
or if this is not possible

2. show an alteration of the PR footing indicative of the ARC resonator impacting the phase and intensity relations at the base of the PR.

Ideally both of these characteristics will be evident as a series of samples is swept through the resonance that determines the evanescent ARC phenomena that has been proposed.

In the following sections we seek to achieve these goals for both types of evanescent-coupled ARC examples. The general methodology for this will be to first carry out a PD sweep to find a PD (at the design NA) which produces well defined structures to the bottom of the PR. This PD will then be used to sweep the NA range about the design NA to locate the NA which produces the optimum PR structure and ideally produces sub-resonant and over-resonant structures also. The results of these two sweeps will be given first, followed by secondary data/information highlighting specific important factors relating to the particular experiment.

### 5.3 SSR based evanescent-coupled ARC results

The SSR based evanescent-coupled ARC employed in this section consists of a MgF$_2$|Cr resonant underlayer designed to operate at an NA of 1.4046 under TM illumination (Fig. 5.2(b)). The design NA for this system is that of the surface state resonance condition defined in Eq. (4.14), while the thicknesses of the MgF$_2$ and Cr layers were obtained by using the TMM as was employed in Sections 4.6 and 4.7.

Although the rotational stage can be tuned to the desired NA, this is of little use without using the correct PD. As such, initial trials focused upon finding a suitable PD, a representative example of which is given in Fig. 5.7. The PD for Figs. 5.7(a) to 5.7(d) are 1.0, 1.1, 1.2, and 1.3 mW-mins. What is perhaps most notable in these figures is the general degradation of the PR visible in Figs. 5.7(a) to 5.7(d). This is largely due to the DC component inherent in the interference of TM waves (see Section 5.4.1), determination of the degree to which the PR has been removed is somewhat difficult due to the tilt angle of the SEM image and the angle of the developed cleave face, although the number or standing wave periods compared to the pre-development thickness of approximately 1300 nm can be used as an estimation. The large scale roughness seen in these same figures is likely due to the surface roughness of the prism. This appears to be significantly worse for this NA and polarization compared to later TE trials at an NA of 1.5 (Section 5.5).
5.3. SSR BASED EVANESCENT-COUPLED ARC RESULTS

Figure 5.7: MgF$_2$|Cr SSR ARC - PD sweep, NA=1.4091, TM polarization. (a) - PD = 1.0 mW-mins. (b) - PD = 1.1 mW-mins. (c) - PD = 1.2 mW-mins. (d) - PD = 1.3 mW-mins. Note the general reduction in PR thickness with PD indicative of the DC exposure component from TM polarized illumination. The large scale roughness is due to the relatively poor polishing of the prism. The optimum exposure PD in this case is 1.2 mW-mins which has the best clearance to the PR bottom.
The removal of large amounts of overlying PR and the larger scale roughness make this system unfortunately very sensitive to the exposure conditions. Even so, the optimum PD for this particular experiment is clearly 1.2 mW-mins (Fig. 5.7(c)) where both exposure/development clearance to the PR base is present and the PR footing is visible, with the alteration of the PR footing compared to a control example (Fig. 5.5(b)) indicating resonant exposure. Whereas Figs. 5.7(a) and 5.7(b) are underexposed with clearance to the PR base not present, Fig. 5.7(d) conversely is overexposed with so much overlying PR removed that the large scale noise reaches the PR base in many areas resulting in ill defined structures, and often a large amount of ‘scum’ occluding the PR structures.

Once a suitable PD has been discovered an NA sweep can be carried out to search for both the experimental goals, that of, standing wave suppression and alteration of the PR footing. Figures 5.8(a) to 5.8(d) are a representative example of an NA sweep at the optimum PD with NAs of 1.38, 1.39, 1.4046, and 1.42, where the design NA is 1.4046. In all cases standing waves are clearly visible, thus failing the aim of complete standing wave suppression. Of note in these figures however is the reduction in standing wave prominence with increasing NA. This has one of two possible explanations: firstly, the ARC may be working, and secondly the reduced dose with NA (due to exterior reflections) is reducing the reflection intensity and therefore reducing the standing wave intensity. To help discern between these two possibilities a model was constructed of the 2D intensity distribution within the PR for the different NAs.

Figures 5.9(a) to 5.9(d) show the 2D intensity profile within the PR|MgF$_2$|Cr SSR ARC film stack for the NAs used in Fig. 5.8. These figures clearly indicate that assuming the ARC is working the PR structures should have significantly different profiles than those seen in Fig. 5.9, particularly when the PR footing is considered, as such the reduction in standing wave prominence is not due to the ARC. For these same angles (NA=1.38, 1.39, 1.4046, 1.42) the corresponding reflectances from the Air|Prism interface are 0.0133, 0.0154, 0.0185, and 0.0218. The increase in reflectance reduces the intensity reaching the PR. This reduced intensity coupled with the increased path length within the PR for increasing NAs results in a lower intensity reflectance from

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6 Note: For this particular experiment a different prism material was used hence due to different reflectances off the prism interfaces a different optimum PD of 0.8 was employed.

7 Note: Increasing the NA requires an angle closer to normal incidence at the Air|Prism interface. For TM polarization this results in an increased reflectance with increasing NA as the angle shifts away from the Brewster minimum. For TE polarization the Air|Prism interface reflectance decreases with increasing NA due to the lack of a Brewster minimum.
Figure 5.8: MgF₂|Cr SSR ARC - NA sweep, PD = 0.8, TM polarization

(a) \( NA = 1.38 \)  
(b) \( NA = 1.39 \)  
(c) \( NA \approx 1.4046 \)  
(d) \( NA = 1.42 \)

Note: Standing waves are present at all NAs, although reducing in prominence with increasing NA. This is from a separate set of trials thus the PD differs from the optimum found previously. The developed PR thickness is less than the approximately 1300 nm pre-exposure thickness due to the DC background exposure inherent in TM interference.
5.3. SSR BASED EVANESCENT-COUPLED ARC RESULTS

### Figure 5.9: MgF$_2$|Cr SSR ARC - NA sweep simulations$^8$ TM polarization

(a) $\text{NA} = 1.38$. (b) $\text{NA} = 1.39$. (c) $\text{NA} \approx 1.4046$. (d) $\text{NA} = 1.42$. Note: prominent standing waves are present for all NAs other than the optimum NA. Also the PR footing changes with the NA. The blue lines demarcate the PR layer. The white contour indicates the $1/e^{0.33}$ intensity contour, this value has no special significance other than to relatively closely match the SEM imaged structures. The bleached PR refractive index is employed here as it better represents the observed PR structures (see Section 5.4.2).

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$^8$ Parameters: Film stack Prism|PR|MgF$_2$|Cr|Si. Prism - Soda-lime glass - $n = 1.5366$[164] $d = \text{bulk}$, AZMIR701 PR bleached - $n = 1.684 + 0.031i$[119] $d = 1300 \text{ nm}$, MgF$_2$ - $n = 1.38$[167] $d = 102 \text{ nm}$, Cr - $n = 2.0356 + 2.8804i$[168] $d = 18 \text{ nm}$, Si - $n = 5.438 + 0.342i$[122] $d = \text{bulk}$. 

the PR|MgF$_2$ interface and thus a reduced standing wave intensity. Further to this the reduction in intensity should result in a reduction in the DC background exposure too, this is a trend seen in Fig. 5.8. In the authors opinion this is the effect causing the reduction in standing wave prominence observed in the NA sweep experimental results.

### 5.3.1 Standing wave suppression and footing

Many experiments were carried out with this ARC to attempt to verify full standing wave suppression; to date however this has not been achieved with the best result being an altered footing. In Fig. 5.10 this altered footing (Fig. 5.10(a)) is compared to a simulation of the design ARC (Fig. 5.10(b)), as well the experimental control (Fig. 5.10(c)) and control simulation (Fig. 5.10(d)). At this point it is worth noting that the simulation voids have a different orientation to those in the experiment due only to the scaling of the axes. The axes scaling remains however to better display the structures.

The altered footing in Fig. 5.10(a) can be attributed to the resonator resonating but in a sub-optimal fashion. This is evident when comparing it to the simulation of the optimal ARC case where only very weak standing waves are present (Fig. 5.10(b)). The presence of these very weak standing waves are discussed in Section 5.4.2. Compared to the control experiment (Fig. 5.10(c)) and simulation (Fig. 5.10(d)) however it is obvious the system is not acting as a simple reflector. If this were the case the PR footing would be a node point, thus no voids would occur at the base of the PR as they do in Fig. 5.10(a). Instead an intensity hot spot is present producing the voids. In the following section the reasons for this sub-optimal resonance will be discussed along with several important design and experimental considerations.

### 5.4 SSR based evanescent-coupled ARC experimental discussion

The system designed to verify SSR evanescent-coupled ARCs due to the design NA and the use of TM polarization unfortunately requires a very finely balanced set of system and experimental parameters which make it extremely difficult to verify. As such, in this section several of the factors leading to this sensitivity will be discussed.
Figure 5.10: MgF$_2$|Cr SSR ARC - footing and control results and simulations. **NA≈1.4046, TM polarization.**

- **(a)** - Footing experimental.
- **(b)** - Footing simulation.
- **(c)** - Control experimental.
- **(d)** - Control simulation.

Note: the footing in **(a)** compared to the control footing **(c)** indicates the resonator is resonating but not in the correct manner to produce full standing wave suppression. Also of note is the significant difference between the experimental footing example structure **(a)** and the optimal design ARC structure **(b)**, again indicating the system is sub-optimally resonant. The control experiment **(c)** matches the simulation **(d)** well. Simulation notes: Bleached PR refractive index was used (see Section 5.4.2). Blue lines demarcate the PR layer. White contours for **(b)** and **(d)** indicate the $1/e^0$ and $1/e^1$ intensity contours respectively.

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Parameters: Footing film stack Prism|PR|MgF$_2$|Cr|Si. Prism - Soda-lime glass - $n = 1.5366$ | $d = $ bulk, AZMIR701 PR - $n = 1.684 + 0.031i$ | $d = 1300$ nm, MgF$_2$ - $n = 1.38$ | $d = 102$ nm, Cr - $n = 2.0356 + 2.880i$ | $d = 18$ nm, Si - $n = 5.438 + 0.342i$ | $d = $ bulk. Control film stack Prism|PR|Si. Prism - Soda-lime glass - $n = 1.5366$ | $d = $ bulk, AZMIR701 PR - $n = 1.684 + 0.031i$ | $d = 1300$ nm, Si - $n = 5.438 + 0.342i$ | $d = $ bulk.
5.4.1 Polarization interference fringe visibility

In both the PD and NA sweep figures it was clearly visible that a considerable amount of PR had been removed from the surface of the PR. This is due to the use of TM polarized illumination and the consequent DC component of the E fields. This DC component acts as a flood exposure component exposing across the entire area of the PR.

The intensity pattern for the interference of two equal intensity beams (see Section 2.1) is given by

\[ I_{12} = 2I(1 + (\hat{e}_1 \cdot \hat{e}_2)) \cos((k_1 - k_2) \cdot r), \]  

(5.3)

where \( k_i \) are the wave vectors of the two beams. Of importance to this discussion is the dot product of electric field direction unit vectors \( \hat{e}_1 \cdot \hat{e}_2 \). For TE polarization \( \hat{e}_1 \) and \( \hat{e}_2 \) are parallel i.e. \( \hat{e}_1 \cdot \hat{e}_2 = 1 \) for all angles of incidence, thus when the cosine term is at its extremes the intensity ranges from \( 4I \) to 0 allowing optimal fringe contrast to be achieved. For TM polarization however this is not the case, where only a component of \( \hat{e}_1 \) and \( \hat{e}_2 \) are parallel and \( \hat{e}_1 \cdot \hat{e}_2 \) varies with angle (Fig. 5.11(a)).

If we consider this in terms of the fringe visibility (also called the Michelson contrast) given by

\[ V = \frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{max}} + I_{\text{min}}}, \]  

(5.4)

we see why the use of TM polarization is such an issue, particularly for this particular ARC. Substituting Eq. (5.3) into Eq. (5.4) and plotting for TM polarization versus the full angle \( (\theta_1 + \theta_2) \) between the beams (Fig. 5.11(b)). The fringe visibility for TM polarization follows an approximate ‘V’ shape with the minimum occurring at 90°, at this angle the \( \hat{e}_1 \) and \( \hat{e}_2 \) are orthogonal \( (\hat{e}_1 \cdot \hat{e}_2 = 0) \) thus the cosine term disappears and \( I_{\text{max}} \) is equal to \( I_{\text{min}} \), thus the visibility is equal to zero. For 0° and 180° the fields are parallel thus the fringe visibility is equal to one.

For this particular experiment the fringe visibility is 0.406 (Fig. 5.11(b), black line). This causes poor visibility and is responsible for the narrower PD process window for the TM polarization SSR based ARC compared to the TE polarization DR based ARC. The TM experiments also appear to be far more sensitive to noise likely because the intensity of the noise is larger compared to the process window of these exposures.

5.4.2 PR bleached vs unbleached

Throughout the course of these experiments it was noticed that the PR standing wave profile generally does not match the simulations for either the unbleached \( n = 1.6844+ \)
5.4. SSR BASED EVANESCENT-COUPLED ARC DISCUSSION

Figure 5.11: TM fringe visibility \((a)\) TM interference schematic. Note the \(E_i\) fields are generally not parallel, thus the fringe visibility changes with the NA. \((b)\) TM fringe visibility vs. the combined angle of incidence \((\theta_1 + \theta_2)\). The TM MgF\(_2\)/Cr SSR ARC with a design NA of 1.4046 has a visibility of 0.406.

0.0307i) or bleached \((n = 1.6751 + 0.0031i)\) PR refractive indices, and in fact appears to have a value somewhere closer to the middle of these two values. Unfortunately this partially undermines the ability of the two experimental ARC systems to suppress standing waves due to the fact they were designed using the unbleached PR refractive index.

In Fig. 5.12(a) we have an example SEM image of a TE exposure at an NA of 1.35. Things of note in this image include the resist collapse at the top, as well as the well defined equal sized holes beneath this. If we compare this image to a 2D intensity trace of this same experiment employing the unbleached (Fig. 5.12(b)) and the bleached (Fig. 5.12(c)) PR refractive indices we see that neither fully captures the features seen in Fig. 5.12(a). The hot spots in these figures represent high intensity; these are the exposed areas which are removed when the PR is developed. Comparing these (and other SEM) images we realize that although the resist collapse is indicative of the unbleached refractive index, the very regular holes beneath this are more representative of the bleached refractive index case.

As the PR is exposed its refractive index changes from the unbleached to bleached values, thus the ‘true’ value which we define as what is displayed in the experimental figure, lies somewhere between these two values. The intensity profiles for both SSR and DR based ARCs are shown in Fig. 5.13. In both cases we see the standing wave progressively increase as the PR becomes bleached\(^\text{10}\). The fact that the ARCs were

\(^{10}\text{Note: Accurate modelling of this effect would also take into account the fact that the bleaching}
5.4. SSR BASED EVANESCENT-COUPLLED ARC DISCUSSION

Figure 5.12: PR refractive index - bleached vs unbleached - parameters

(a) - A representative image of the standing waves produced with no ARC present. Note the resist collapse at the top, but also the well defined holes all the way to the bottom of the PR. 
(b) - 2D intensity profile using the unbleached PR refractive index \( n = 1.6844 + 0.0307i \). The PR (bounded by the blue lines) intensity profile tapers due to absorption. 
(c) - 2D intensity profile using the bleached PR refractive index \( n = 1.6751 + 0.0031i \) - Due to weak absorption there is little tapering of the intensity profile. Considering these figures it appears the ‘true’ value of \( n_{PR} \) is between the unbleached and bleached values. Note: the white lines represent the 1/e intensity contour.

designed to operate with the unbleached PR refractive index undermines their ability to suppress standing waves. If this same SSR based ARC is reoptimized for the bleached PR the layer thicknesses are

- Bleached PR thicknesses - MgF\(_2\) = 99 nm, Cr\(_2\) = 18 nm

compared to the unbleached PR layer thicknesses

- Unbleached PR thicknesses MgF\(_2\) = 102 nm, Cr\(_2\) = 18 nm.

This difference produces a slight (likely insignificant) standing wave; this however adds to the possible ARC layer errors present, the effects of these will be discussed in Section 5.4.3.

process does not occur equally throughout the entire depth of the resist, when in fact the upper region bleaches first with a bleaching front progressing downwards through the PR. For the sake of simplicity this effect is ignored, with Fig. 5.13 deemed sufficient to inform one of the effects of bleaching on these experiments.

Figure 5.12(b) - Prism - Soda-lime glass - \( n = 1.5366 \) \[164\], \( d = \) bulk, PR AZMIR701 unbleached - \( n = 1.6844+0.0307i \) \[119\], \( d = 1300 \) nm, Si - \( n = 5.4375+0.3420i \) \[122\], \( d = \) bulk. Figure 5.12(c) - Prism - Soda-lime glass - \( n = 1.5366 \) \[164\], \( d = \) bulk, PR AZMIR701 bleached - \( n = 1.6751+0.0031i \) \[119\], \( d = 1300 \) nm, Si - \( n = 5.4375+0.3420i \) \[122\], \( d = \) bulk. NA=1.35, TE polarization
Figure 5.13: Effect of PR refractive index bleaching\textsuperscript{12} (a) - MgF\textsubscript{2}|Cr SSR base ARC. (b) - SiO\textsubscript{2}|HfO\textsubscript{2} SSR base ARC. The PR refractive varies from unbleached \((n = 1.6844 + 0.0307i)\) to bleached \((n = 1.6751 + 0.0031i)\) in equal increments \((n_{PR(2)}, n_{PR(3)}, n_{PR(4)}, \text{and } n_{PR(5)}).\)

In Fig. 5.13 another fact is visible, that of the difference in intensity slope within the PR between the two ARC systems. The lower NA and the use of TM polarization for the SSR experiment results in a higher insertion intensity (i.e., lower reflectances at the Air|Prism and Prism|PR interfaces). The difference in the intensity slope explains some of the results seen in the SSR ARC PD sweep (Fig. 5.7), namely the large removal of PR at the top of the PR layer. Assuming the dose at the base of the PR is the optimal value, the surface of the PR receives an approximately 6x greater dose. This fact combined with the DC component of TM exposure and the noise from prism surface roughness likely leads to an appreciable non-periodic dose in the upper part of the PR, hence a large portion of the PR is developed away.

The faster intensity drop in Fig. 5.13 suggests a method of improving this particular experiment by altering the conditions to ‘flatten’ the intensity profile. This can be achieved by either reducing the insertion intensity by increasing the reflectance or absorption in the layers above the PR, or by reducing the PR thickness. Halving the PR thickness results in the surface dose being approximately three times that of the base dose. Further experimental trials will be required to verify if this dose difference is narrow enough to allow suitable base exposure without significant removal of the upper PR surface during development; or if further PR thickness reduction is required.

\textsuperscript{12}Figure 5.13(a) - Soda lime glass - \(n = 1.5366\) \[164\], \(d = \text{bulk}\), PR AZMIR701 - \(n = \) values in caption, \(d = 1300\) nm, MgF\textsubscript{2} - \(n = 1.38\) \[167\], \(d = 102\) nm, Cr - \(n = 2.0356+2.8804i\) \[168\], \(d = 18\) nm,
5.4.3 ARC layer thickness and refractive index errors

In these experiments both a suitable PD and the correct NA were able to be set reliably as evidenced by Figs. 5.8 and 5.10(a) which show suitable exposure and structure clearance to the base of the PR as well as having the pitch corresponding to the design and adjacent NAs. Despite this, standing waves are still clearly visible with the best result being evidence of resonance indicated by a shifted standing wave node at the base of the PR (Fig. 5.10(a)). The most likely reason for the lack of full standing wave suppression is a compounding of modelling and processing errors in the refractive indices and the ARC layer thicknesses; these errors take the system away from optimal resonance. To get a feel for the impact of these errors, in this section simulations are carried out using a range of likely errors (informed by ellipsometric wafer measurements and literature data) and a more appropriate intermediate PR refractive index \((n = 1.6797 + 0.0169i)\) with the aim of identifying which errors are most likely responsible for the observed results.

For these experiments four MgF\(_2\)Cr ARC coated wafers were manufactured (by the ANFF\[169\]) and ellipsometric characterisation was carried out using the CompleteEase (Version 5.10) software and alpha-SE multi-angle spectral ellipsometer by J.A. Woollam Co.\[170\]. Ellipsometers measure the change in polarization of reflected and transmitted beams from a sample. Models can be constructed to fit these measurements to ideally give very accurate values for the refractive indices and thicknesses of the individual film layers. The ability to produce very accurate results however requires intimate knowledge of the deposition process and conditions, film structure (nano, crystalline, amorphous, etc.), as well as secondary measurement validation methods. Due to this, ellipsometry is best employed in a processing feedback type arrangement where an ellipsometry fitting model can be constructed, optimized, and validated in a series of trials. For this research however neither the deposition process and conditions or the film structure are well known due to the films being deposited off site. Secondary validation methods were used, but with limited accuracy they were employed only as fitting parameter limits. The construction of ellipsometry fitting models is somewhat of an art form on which many textbooks have been written \[171, 172, 173\]. As the goal of this section is more to investigate the possible causes of experimental error

Si - \(n = 5.4375+0.3420i\) \[122\], \(d = \) bulk, NA = 1.4046, TM polarization, \(\lambda = 405\) nm. Figure 5.13(b)
- Soda lime glass - \(n = 1.5366\) \[164\], \(d = \) bulk, PR AZMIR701 - \(n = \) values in caption, \(d = 1300\) nm, SiO\(_2\) - \(n = 1.4696\) \[121\], \(d = 88\) nm, HfO\(_2\) - \(n = 1.9804\) \[154\], \(d = 95\) nm, Si - \(n = 5.4375+0.3420i\) \[122\], \(d = \) bulk, NA = 1.5, TE polarization, \(\lambda = 405\) nm
rather than to fully optimize the fitting models (for which not enough information is available), simple but representative models have been constructed to produce values for the ARC layer refractive indices and thicknesses.

Ellipsometers measure the change in TE and TM components of the reflected (and transmitted when possible) beam. The measured quantities are $\Delta$ the phase shift between the TE and TM components, and $\Phi$ the amplitude ratio of the TE and TM components; from these the complex refractive index ($\tilde{n} = n + ik$) and the thickness can be obtained. For transparent materials this is relatively simple process as $k$ is equal to zero. For absorbing materials however this is not the case where we now have two measured values and three unknowns; for this reason regressive analysis of fitting models (as well as informed model limits) are required to uniquely identify the refractive indices and thicknesses. In absorbing materials $n$ and $k$ are related in an often complex manner related to the properties of the particular material; to fit the myriad range of different materials various oscillator models have been developed [173]. The quality of the fit is specified by the mean square error (MSE) which is the average of the square error between the measured and model curves. Transparent films generally have lower MSEs (<5) corresponding to better fits, while absorbing (particularly thin metal) films have higher MSEs (<10), however the suitability of a particular fit is up to the users best judgement.

The MgF$_2$|Cr wafer fitting model comprised of a Cauchy model for the MgF$_2$ layer, a general oscillator model for the Cr layer consisting of a Drude(NMu) and three Lorentz oscillators, and the Si wafer represented by standard inbuilt silicon wafer refractive indices [173]. Cauchy models are designed for non-absorbing dielectrics, thus this is deemed an appropriate model for the MgF$_2$ layer. As silicon wafers are very homogeneous the use of a reference refractive index is also deemed appropriate. The construction of the general oscillator model however is more difficult, as metals, particularly as components of multilayer stacks, are well known for being difficult to fit accurately [174, 173, 175]. The literature mentions two different methods of configuring a general oscillator model for Cr. Firstly, Tompkins et al. [176] report a method using a combination of four Lorentz oscillators for 5-30 nm thick layers. Secondly, Hilfiker et al. [174] employ a combination of a Drude and two Lorentz oscillators to effectively fit Cr layers 10-40 nm thick. They also add further Lorentz oscillators to better fit the finer details of the measurements, this however results in a degradation of the

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$^1$Note: Surface roughness was not included in these models as it had very little impact on the fitting results.
fitness uniqueness. The fitting model constructed for this research utilized Hilfiker’s oscillator configuration in its simplest form (one Drude and two Lorentz oscillators) with an extra Lorentz oscillator as this produced the best fits without adding further oscillators which had little impact on the fit quality.

Ellipsometry measurements were carried out at five standard locations (Fig. 5.14) on each of the four wafers (designated blue, red, black, and green) to ascertain the refractive indices and thicknesses variations across individual wafers and between different wafers. The MSE for each of these measurements is shown in Fig. 5.15(a) indicated by coloured crosses for each of the wafers. The mean of the mean MSE across the wafers is 7.6297 which is considered reasonable for this type of film [173]. The blue, red, and black wafers all fit in a somewhat similar fashion with the green wafer appearing to be an outlier. Across individual wafers the fitting is fairly uniform especially for the blue wafer with the green wafer again being a clear outlier.

The fit for the MgF$_2$ refractive index ($\kappa = 0$) is shown in Fig. 5.15(b). The refractive index values are between 1.3931 and 1.4166 with a total mean value of 1.4049. Once again the blue, red, and black wafers fit fairly regularly with the green wafer being an outlier. The model value for the MgF$_2$ refractive index is 1.38. Values in the literature range from 1.38 to 1.3957 [167, 177, 152] with the value 1.38 considered the most relevant here as it is for a sputter coated film (as are the films in this experiment) while the others are for single crystals which display birefringence. The fact that (non-annealed) sputtered films are non-birefringent bypasses the issue of birefringence, but also raises another issue, that is, that sputter coated fluorides are typically porous and therefore should have a refractive index less than that of the crystalline material.

These wafers were checked for the presence of Ca by EDS on the off chance that CaF$_2$ ($n = 1.44$)
Although substrate heating and ion-assisted deposition methods \cite{178} can be employed to make the films less porous, the films used in these experiments likely have a refractive index less than the values in the literature indicating the fitting model is not very accurate for this parameter and the results should only be taken as a guide.

The fits for thickness of the MgF$_2$ layer are shown in Fig. 5.15(c) with a minimum value of 122.2 nm, a maximum of 133.7 nm, and an overall mean of 127.9 nm. The values across all wafers are fairly similar despite the variations seen in the previous refractive index plot. The values however are significantly different (approximately 26 nm) from the model design value of 102 nm. The impact of this likely error will be further investigated later in this section.

The fitting of thin metal films (particularly within a multilayer stack) is significantly more difficult than for a transparent dielectric such as MgF$_2$. The fits for the Cr layer reflect this difficulty showing a far greater variation in parameters than the MgF$_2$ fits. The difficulty lies in the requirement to fit the extra parameter \( \kappa \) as well as the complex relationship between \( n \) and \( \kappa \). An equally difficult issue is the fact that metal thin films often form islands which coalesce into a ‘bulk’ film as the thickness increases. The design thickness for these experiments however is thin enough that islands are most likely present resulting in a granular film of lower density than the bulk film as well as a reduced conductance due to poor conduction between islands. Both of these issues impact the refractive index thus there is a large amount of variation in the literature for Cr \cite{168, 179, 180, 153}. The fit for the real component of the Cr refractive index is shown in Fig. 5.15(d) with a minimum of 1.841, a maximum of 2.2781, and a mean value of 2.1375. Although the values are mostly higher than the model value of 2.0356, this value appears to be an appropriate choice for the Cr layers in these experiments. The imaginary component of the Cr refractive index is shown in Fig. 5.15(e) with a minimum of 2.2672, a maximum of 3.3548, and a mean value of 2.8052. Although greater variation is seen in these fit values, the model value once again appears to be appropriate.

The fits for the thickness of the Cr layer are shown in Fig. 5.15(f) with a minimum of 15.96 nm, a maximum of 24.18 nm, and a mean value of 20.31 nm. Most notable in this plot is the large range of errors in these value indicating the difficulty of fitting such a material (general-oscillator model). The values are all above the model thickness, although as with all these fit values, they should only be used as a guide unless the fit model can be properly validated. Nonetheless the values of these fits will along with

\footnote{was mistakenly used but no Ca was detected}
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**Figure 5.15:** MgF$_2$|Cr ARC wafer ellipsometry measurements and fitting. (a) - MSE. (b) - MgF$_2$ $n$. (c) - MgF$_2$ thickness. (d) - Cr $n$. (e) - Cr $\kappa$. (f) - Cr thickness. Note: Johnson et al., Rakic et al., Weaver et al., and Lozanova et al. are Cr refractive index values from the literature.
the literature values (where appropriate) be used to inform the investigation of the impact of possible processing errors.

The effect of variation in the thicknesses of both the MgF$_2$ and Cr is displayed in Figs. 5.16(a) and 5.16(b). Figure 5.16(a) shows the intensity trace for a ±10% thickness variation of the MgF$_2$ layer as well as the minimum and maximum values from the ellipsometry fitting, these values correspond to thicknesses of 91.8, 102, 112.2, 122.2, and 133.7 nm, where 102 nm is the design thickness. The standing wave variation for differing MgF$_2$ thicknesses produces large standing wave amplitudes for the likely film stack errors. Although the likelihood of MgF$_2$ thickness having such a large error is slim due to the very slow deposition rate (~1 nm/min), the ellipsometry fit (along with other alternative fits) suggests that the thickness is significantly greater than the design thickness of 102 nm. Figure 5.16(b) shows the intensity trace for a ±10% thickness variation of the Cr layer as well as the maximum Cr thickness value from the ellipsometry fit, these values correspond to thicknesses of 16.2, 18, 19.8, and 24.18 nm, where 18 nm is the design thickness. Variation of the Cr layer thickness has less of an impact on the standing wave magnitude with errors of approximately 20% required to produce similar standing wave magnitude as the MgF$_2$ thickness variation. The relative thinness of the Cr layer however does increase the likelihood of such an error.

For variations in the refractive index we must consider what values are actually feasible, thus each plot has a slightly different range of variations to account for this. Generally speaking for non-absorbing dielectric materials there is little difference in the refractive index of bulk films, with variations arising from different crystalline structures and/or stoichiometric differences. Fluorides however are known to commonly form porous structures, as such refractive index variations of ±2.5% are plotted corresponding to $n=1.359$, 1.38, and 1.415. The ellipsometry fit values for $n_{MgF_2}$ ($n = 1.3931$ and 1.4166) are not plotted as they are close to or within the plotted errors. This ARC design is clearly sensitive to errors in this parameter. The reader might wonder why we would even consider refractive index variations greater than that of the bulk material? The answer being that if this layer is in fact porous, the PR ($n = 1.6844 + 0.0307i$) may permeate this layer producing a composite refractive index greater than that of MgF$_2$ alone. Although MgF$_2$ is not absorbing at this wavelength, the possibility of PR permeating into MgF$_2$ pores also raises the possibility of the $\kappa$ of this layer affecting the ARC properties. The values used in this model ($n=1.38$, 1.38+0.0154i, and 1.38+0.0307i) correspond to zero $\kappa$, $\kappa_{PR}/2$, and $\kappa_{PR}$. Variations of this size are considered not very likely as they correspond to high proportions of PR which contradicts
5.4. SSR BASED EVANESCENT-COUPLED ARC DISCUSSION

Figure 5.16: MgF₂|Cr ARC parameter variations. Design values in bold. (a) - MgF₂ thickness variation ±10%. (b) - Cr thickness variation ±10%. (c) - ±2.5% variation of the real component of the MgF₂ refractive index. (d) - Variation of the imaginary component of the MgF₂ refractive index. (e) - ±20% variation of the real component of the Cr refractive index. (f) - Variation of the imaginary component of the Cr refractive index.
5.4. SSR BASED EVANESCENT-COUPLED ARC DISCUSSION

the relatively dense films observed with the SEM and AFM.

Ellipsometry fit data and literature data suggest there is potentially a significant difference in the experimental and model refractive indices for the Cr layer [168, 179, 180, 153]. To account for this the $n$ and $\kappa$ variation plots (Figs. 5.16(e) and 5.16(f)) both employ large variation percentages of $\pm 20\%$. In Fig. 5.16(e) we see the standing wave response if the real refractive index component of Cr is altered by $\pm 20\%$, this corresponds to $n$ values of 1.6285, 2.0365, and 2.4428, where 2.036 is the optimal value and the imaginary component has been kept fixed. The ellipsometry values ($n_{\text{min}} = 1.841$ and $n_{\text{max}} = 2.2781$) are not plotted as they are within the 20% interval. In Fig. 5.16(f) we see the standing wave response if the imaginary refractive index component is altered by $\pm 20\%$, this corresponds to $\kappa$ values of 2.3043i, 2.8804i, and 3.4565i. The ellipsometry values ($n_{\text{min}} = 2.2672$ and $n_{\text{max}} = 3.3548$) are not plotted as they are close to or within the 20% interval. The effect of both $n$ and $\kappa$ variation is of approximately the same magnitude. Although interestingly variation of $n$ impacts the resonance strength by a greater amount than $\kappa$ variation.

The manufacturing of these ARCs was carried out as a first attempt at these experiments, consequently the manufacturing methods and constraints were not performed to exacting standards, indeed, at the stage of manufacturing the exact requirements were not very well refined. As such, the ARC refractive indices and thicknesses are likely different to those of the model values. Considering the ellipsometry fit values and the literature values, it is most likely that the greatest error present is in the Cr layer refractive indices and thicknesses. Also the porous nature of the MgF$_2$ will likely change the refractive index of this layer, particularly if the PR is able to permeate the structure. The ellipsometry data does suggest the MgF$_2$ is significantly thicker than the design value, but this must be approached with caution as the fit model also suggests the refractive index is greater than the bulk refractive index for this material which is not possible without a higher refractive index material (such as PR) filling the voids.

The best results for this ARC showed hot spots at the base of the PR (Fig. 5.10(a)). The error modelling in this section suggests the origin of these hot spots is due to either: too thick a Cr layer, too great an MgF$_2$ refractive index, or too low a Cr imaginary refractive index value. From the data at hand it is not possible to positively identify which factor is causing the lack of total standing wave suppression with the most likely reason being a combination of a differing PR refractive index in conjunction with manufacturing errors in the ARC layer parameters.
5.5 DR based evanescent-coupled ARC results

The DR based evanescent-coupled ARC employed in this section is a modified trilayer type system composed of a SiO$_2$|HfO$_2$|Si(substrate) stack designed to operate at an NA of 1.5 under TE polarization (Fig. 5.2(c)). As with SSR based ARC, the design of this system was also developed using TMM.

![SiO$_2$|HfO$_2$ DR ARC - PD Sweep - NA = 1.5, TE polarization](image)

(a) (b)

(c) (d)

**Figure 5.17:** SiO$_2$|HfO$_2$ DR ARC - PD Sweep - NA = 1.5, TE polarization. (a) - PD = 0.8 mW-mins. (b) - PD = 1.0 mW-mins. (c) - PD = 1.2 mW-mins. (d) - PD = 1.4 mW-mins. Note the depth of the development front increases with PD. The optimum PD is clearly 1.4 mW-mins, as evidenced by development to the base of the PR.

Figure 5.17 shows a representative example of a PD sweep for this system at an NA of 1.5. For Figs. 5.17(a) to 5.17(d) the PDs are 0.8, 1.0, 1.2, and 1.4, with the
optimum clearly occurring at a PD of 1.4 where full clearance to the PR base has been achieved and thus both the standing wave suppression and footing can be investigated. Most notably is that fact that Fig. 5.17(d) appears to be functioning as an ARC, which will be further discussed shortly. Standing waves are present in all examples (but minimized in Fig. 5.17(d)) getting progressively deeper with PD. Also evident in these figures is the largely constant PR thickness across PDs indicative of the lack of a DC component in the interference, highlighting why TE polarization is the preferred form of illumination.

Figure 5.18: SiO$_2$|HfO$_2$ DR ARC - NA Sweep - PD = 1.5, TE polarization. (a) - NA = 1.48. (b) - NA = 1.49. (c) - NA = 1.50. (d) - NA = 1.51. Design NA = 1.5.

The NA sweep for this system was performed for NAs 1.48, 1.49, 1.50, and 1.51, where the design NA is 1.5. Unfortunately clear standing waves are not present in each of these figures, despite a higher PD of 1.5 being used on account of the thicker
PR layer\textsuperscript{15,16} What is visible however is the presence of plateaus corresponding to standing wave node points (i.e. bridging layers), particularly in Figs. 5.18(a) to 5.18(c). Interestingly the plateaus are less prominent in the $NA = 1.51$, despite it being at an NA greater than the design NA. This sample also shows (faint) hot spots in the PR footing (left-hand side).

NA sweep simulations (Fig. 5.19) show reducing standing wave intensity as the design NA is approached (Figs. 5.19(a) and 5.19(b)). Minimal standing waves occur at the design NA (Fig. 5.19(c)), ideally they should not be present, but as the design was developed using the unbleached PR refractive index they remain. Above the design NA the strength of the standing waves increase (Fig. 5.19(d)). The PR footings are notable in that for NAs less than the optimum a node is present, while for NAs greater than the optimum the system is over-resonant and a hot spot is present. Although it’s tempting to assign the simulated footing hot spot to that of the experimental footing hot spot, the experimental figures are unfortunately not clear enough to positively assign all the simulations, thus assigning this result must be done with caution as the experimental results could equally be due manufacturing errors as discussed in Section 5.6.

### 5.5.1 Standing wave suppression and footing

Figure 5.20 shows the main results of this chapter. In Fig. 5.20(a) suppression of standing waves is clearly visible with the presence of extremely high aspect ratio (collapsed) upright structures with minimal standing waves present on the edges. Crucially these upright structures have clearance to the base of the PR. This indicates that the dose at the base of the PR is strong enough to produce standing waves but they have been suppressed. Adjacent areas still feature a standing wave likely due to noise in the exposing beam (PD) as well as in the ARC thicknesses and refractive indices. These structures compare favourably with the simulation of this system (Fig. 5.20(b)). The weak standing waves along the upright edges of the collapsed structures also validate (approximately) the usage of the bleached PR refractive index in the simulations.

\textsuperscript{15}The PR thickness spin curve is thick at low rpms but quickly thins and flattens out as the rpms increase \textsuperscript{15}. For this PR recipe the PR is spun at 2500rpm which lies on the steepest part of the curve. As such, any change in spinning conditions such as PR temperature, room temperature, pressure, humidity etc. may result in seemingly disproportionate changes in PR thickness, this is however typical of a non-climate controlled environment.

\textsuperscript{16}This also indicates one of the drawbacks of using a PD, if the system is variant the PDs are often more of a ballpark figure rather than an exact comparison. As such, PDs serve as more of short term guide which may change over time if the experimental conditions alter.
Figure 5.19: SiO$_2$|HfO$_2$ DR ARC - NA sweep simulations$^{17}$. TE polarization. (a) NA = 1.48. (b) NA = 1.49. (c) NA = 1.50. (d) NA = 1.51. Note: standing waves are present for all NAs with the smallest ones occurring at the design NA of 1.5. Also the PR footing changes with the NA. The blue lines demarcate the PR layer. The white contour indicates the 1/e intensity contour. The bleached PR refractive index is employed here as it better represents the observed PR structures (see Section 5.4.2).

$^{17}$ Parameters: Film stack Prism|PR|SiO$_2$|HfO$_2$|Si. Prism - Soda-lime glass - $n = 1.5366$$^{[164]}$ $d = \text{bulk}$, AZMIR701 bleached PR - $n = 1.684 + 0.031i$$^{[119]}$ $d = 1700 \text{ nm}$, SiO$_2$ - $n = 1.4696$$^{[121]}$ $d = 88 \text{ nm}$, HfO$_2$ - $n = 1.9804$$^{[154]}$ $d = 95 \text{ nm}$, Si - $n = 5.438 + 0.342i$$^{[122]}$ $d = \text{bulk}$. 
5.5. DR BASED EVANESCENT-COUPLED ARC RESULTS

Figure 5.20: $\text{SiO}_2|\text{HfO}_2$ DR ARC - standing wave suppression and control results and simulation$^{18}$, $\text{NA} = 1.50$, TE polarization. (a) - Standing wave suppression experimental. (b) - Standing wave suppression simulation. (c) - Control experimental. (d) - Control simulation. Note: both the standing wave suppression and control experiments compare well with their respective simulations. Resist collapse is present in Fig. 5.20(a) due to the lack of support from the removal of neighbouring bridging structures. The control experiment (Fig. 5.20(c) is a very good example of why a cleave-then-develop process was used, where despite a (mainly) closed PR surface full development to the bottom the PR is still possible. Simulation notes: Bleached PR refractive index was used (see Section 5.4.2). Blue lines demarcate the PR layer. White contours for (b) and (d) indicate the $1/e$ intensity contours.

$^{18}$ Parameters: Footing film stack Prism$|$PR$|$SiO$_2$|HfO$_2$|Si. Prism - Soda-lime glass - $n = 1.5366$$^{164}$ $d = \text{bulk}$, AZMIR701 PR bleached - $n = 1.6751 + 0.0031i$$^{119}$ $d = 1300$ nm, SiO$_2$ - $n = 1.4696$$^{121}$ $d = 88$ nm, HfO$_2$ - $n = 1.9804$$^{157}$ $d = 95$ nm, Si - $n = 5.438 + 0.342i$$^{122}$ $d = \text{bulk}$. Control film stack Prism$|$PR$|$Si. Prism - Soda-lime glass - $n = 1.5366$$^{164}$ $d = \text{bulk}$, AZMIR701 PR - $n = 1.684 + 0.031i$$^{119}$ $d = 1300$ nm, Si - $n = 5.438 + 0.342i$$^{122}$ $d = \text{bulk}$. 
The control figures for this experiment (Figs. 5.20(c) and 5.20(d)) are also very well matched with clear standing waves present to the base of the PR. Comparing these figures to the ARC figures it is clear that a very different PR exposure profile is present indicating the ARC is performing as it was designed. Interestingly the control experiment has a (mainly) closed PR surface highlighting exactly why a cleave-then-develop process was employed, where despite the closed surface development is allowed to proceed to the base of the PR thereby showing the sub-surface PR exposure profile.

To further bolster our claims that the ARC is indeed performing as intended in Fig. 5.20(a) we present Figs. 5.21(a) and 5.21(b) which are from the same set of trials. Both samples are from the same wafer (also area on the wafer) and have an NA of 1.5. Fig. 5.21(a) however has a PD of 1.2 mW-mins while Fig. 5.21(b) has a PD of 1.4 mW-mins. The extra dose in Fig. 5.21(b) implies the resonator is receiving more than enough intensity to produce standing waves but its ARC nature is suppressing them. As both these samples had the same exposure NA it is interesting to note that there must be enough variation in the ARC refractive indices and thicknesses to cause Fig. 5.21(a) to be in an over-resonant state, while Fig. 5.21(b) is close to optimal. This fact will be further discussed in the section to follow.

Figure 5.21: Standing wave suppression and footing. (a) - Over-resonant footing. (b) - Near optimal resonance footing. Note the differences in the footing of these two samples, the optimal resonance footing has a weak node point at the base, while the over-resonant footing has a clear semi-ellipse void at the base corresponding to a standing wave anti-node.

Finally, to properly validate these set of experiments one must not only consider
the experimental results versus those of the control, but also consider the possibility of other explanations. In this case there is the possibility that the experiment has either an incorrect NA or SiO$_2$ refractive index and the hot-spot at the base of the PR is in fact due to the NA (incorrectly) being less than the refractive index of the SiO$_2$, hence propagating fields will be present within the SiO$_2$ which will shift the standing wave node from the base of the PR to the base of the SiO$_2$ layer. As the SiO$_2$ layer is approximately 1/3 of the standing wave pitch a hot-spot should indeed be present at the base of the PR. To investigate this possibility simulations across a broad range of NAs (1.1, 1.2, 1.3, 1.4, 1.5, and 1.51) were carried out to compare the footing and general structure to those of the experimental results (Fig. 5.22).

For NAs just less than that of the refractive index of SiO$_2$ ($n=1.4696$) the angle of incidence should still be very high and thus produce very high reflectances, as such, one would expect standing waves nodes to still be present at the base of the PR where the majority of the reflectance originates from. This idea is confirmed for the NA = 1.4 example (Fig. 5.22(d)) where a node is present at the base of the PR despite the presence of propagating waves in the SiO$_2$ layer. As the NA is further decreased the node shifts further beneath the PR base as the resonator progressively loses resonance and a greater proportion of the reflectance originates from beneath the PR layer. To show similar PR footing hot-spots as those of the over-resonant case (Fig. 5.22(f)) the NA must be reduced to approximately 1.1 (Fig. 5.22(a)). At this point however the horizontal pitch has increased significantly from 134 nm to 184 nm and the standing wave pitch consequently decreased from 270 nm to 160 nm. With these numbers in mind we can positively state the structures seen in Figs. 5.20(a), 5.21(a) and 5.21(b) are not mistakenly exposed at a low NA. This is most simply verified by counting the number of anti-nodes developed away; for the approximately 1400 nm thick PR layer and an NA of 1.1 approximately nine anti-nodes should be present, while for an NA of 1.5 five anti-nodes are present, which is indeed the case.

Although this ARC has been confirmed to be able to suppress standing waves, for completeness, it is also worth investigating the homogeneity of the coated wafers used in these experiments along with the effects of any manufacturing errors upon the ARC system.
Figure 5.22: SiO$_2$|HfO$_2$ ARC - alternative footing simulations - (a) - NA = 1.1. (b) - NA = 1.2. (c) - NA = 1.3. (d) - NA = 1.4. (e) - NA = 1.5. (f) - NA = 1.51.

Parameters: Film stack Prism|PR|SiO$_2$|HfO$_2$|Si. Prism - Soda-lime glass - $n = 1.5366^{[164]} \quad d = \text{bulk},$ AZMIR701 bleached PR - $n = 1.684 + 0.031i^{[119]} \quad d = 1700 \text{ nm}$, SiO$_2$ - $n = 1.4696^{[121]} \quad d = 88 \text{ nm}$, HfO$_2$ - $n = 1.9804^{[154]} \quad d = 95 \text{ nm}$, Si - $n = 5.438 + 0.342i^{[122]} \quad d = \text{bulk},$ TE polarization.
5.6 DR based evanescent-coupled ARC experimental discussion

In these experiments both a suitable PD and the correct NAs were able to be dialled in, with standing wave suppression successfully achieved. The reader may note that the NA sweep example failed to achieve these goals, this is likely due to the effects of variations in the thicknesses and refractive indices of the ARC layers. In this section we will explore the influence of these factors by considering ellipsometry measurements as well as comparing the intensity transects for variations in the ARC parameters.

The SSR based evanescent-coupled ARC experimental discussion raised several issues relating to these experiments. The discussion also covered (for comparison) this DR based evanescent-coupled ARC experiment, thus we shall briefly restate the relevant factors. Firstly, the use of TE polarization allows for perfect fringe visibility which allows better patterning without the DC component inherent in TM polarization exposing the top of the PR layer. Secondly, the resist thickness for this set of experiments is suitable as it allows development to the base of the resist without producing too great a difference in dose between the top and bottom of the PR, as was the case in the SSR based ARC experiments. The true value of the refractive index for ARC design optimization however is also an issue here resulting in a different optimal layer thickness, with the different thickness for the layers being

- Bleached thicknesses - SiO₂ = 82 nm, HfO₂ = 95 nm

compared to the original design thicknesses

- Design thicknesses - SiO₂ = 88 nm, HfO₂ = 95 nm.

Although the PR refractive index is in error, the largest source of error is likely to be variations in the manufactured ARC parameters. Ellipsometry results for the ARC wafers along with intensity trace models for the film stack with different parameter errors will now be discussed.

5.6.1 ARC layer thickness and refractive index errors

The SiO₂|HfO₂ wafer ellipsometry fitting model consisted of a Cauchy model for the SiO₂ layer, a Cauchy model for the HfO₂ layer, and the Si wafer represented by standard inbuilt silicon wafer refractive indices. The designed thickness along with SEM
thickness measurements provided appropriate fitting limits for the two layers. Ellipsometry measurements were carried out at five standard locations (Fig. 5.14) on each of the four wafers (designated blue, red, black, and green) to ascertain the refractive indices and thicknesses variations across individual wafers and between different wafers. The MSE for each of these measurements is shown in Fig. 5.23(a) indicated by coloured crosses for each of the wafers. The mean of the mean MSE across the wafers is 2.8298 which is considered good for this type of film [173]. All wafers fit in a roughly similar fashion, while the intra-wafer fits follow a similar pattern likely indicative of the deposition geometry.

Fitting for the SiO$_2$ refractive index ($\kappa = 0$) is shown in Fig. 5.23(b). The refractive index values are between 1.4008 and 1.4665 with a total mean value of 1.4225. All the wafers fit in approximately the same fashion with the top of the wafer consistently showing a higher refractive index closest to the model value of 1.4696 [121]. Literature values for amorphous/polycrystalline SiO$_2$ range from 1.4696 to 1.4892 [121, 181]. The fitted values are lower than either of these values likely due to them being slightly porous from a non-optimal sputter coating process. Fitting for the thickness of the SiO$_2$ layer is shown in Fig. 5.23(c) with a minimum value of 71.67 nm, a maximum of 97.35 nm, and an overall mean of 91.20 nm. The values across all wafers are fairly similar with the exception of the wafer bottom location measurement which averaged a thickness of 78.37 nm. The optimum value for this layer is 88 nm (or 82 nm for a bleached PR), thus the fitted thicknesses lie either side of the design thickness(es).

Fitting of the HfO$_2$ layer refractive index ($\kappa = 0$) followed a similar trend to the SiO$_2$ fit with a peak in refractive index occurring at the top wafer measurement location (Fig. 5.23(d)). The fitted refractive index values are between 1.8305 and 2.0325 with a total mean value of 1.8970. The value used in the ARC model is 1.9804 which lies within the range of the fitted values. The literature reports values ranging from 1.9-2.2 [154, 109], thus the fitted values are considered appropriate for this material, with the variations displayed most likely due to density differences from the deposition geometry and process. The fitted thicknesses for the HfO$_2$ layer is shown in Fig. 5.23(e) with a minimum value of 91.63 nm, a maximum value of 118.74 nm, and a mean value of 105.71 nm. Once again the bottom wafer location is thinner than the left, centre, and right locations, also present though is an additional thinner area at the top of the wafer. The fitted values at the bottom and top wafer locations most closely match the design thickness of 95 nm. Overall the SiO$_2$|HfO$_2$ wafers are a better match for the design ARC specifications (in comparison to the MgF$_2$|Cr ARC) and thus are more likely to
5.6. DR BASED EVANESCENT-COUPLED ARC EXPERIMENTAL DISCUSSION

Figure 5.23: $\text{SiO}_2|\text{HfO}_2$ ARC wafer ellipsometry fitting data. (a) - MSE. (b) - $\text{SiO}_2$ $n$. (c) - $\text{SiO}_2$ thickness. (d) - $\text{HfO}_2$ $n$. (e) - $\text{HfO}_2$ thickness.
successfully act as an ARC, which was indeed the case. Nevertheless intensity trace models of the ARC system for error of the ARC parameters will now be covered.

Figure 5.24(a) shows the intensity trace for a \( \pm 10\% \) thickness variation of the SiO\(_2\) layer as well as the minimum value from the ellipsometry fitting (the maximum value is within the \( \pm 10\% \) range), these values correspond to thicknesses of 71.7, 79.2, 88, and 96.8 nm, where 88 nm is the design thickness. The standing wave magnitude across this range is minimal indicating that error in this parameter is likely to impact the ARC performance minimally.

Figure 5.24(b) shows the intensity trace for a \( \pm 10\% \) thickness variation of the HfO\(_2\) layer as well as the maximum HfO\(_2\) thickness value from the ellipsometry fit (the minimum value is within the \( \pm 10\% \) range), these values correspond to thicknesses of 85.5, 95, 104.5, and 118.7 nm, where 95 nm is the design thickness. Variation of the HfO\(_2\) layer thickness has a more significant impact on the standing wave magnitude than the SiO\(_2\) thickness variation. This is understandable considering the HfO\(_2\) layer thickness largely determines the waveguiding (resonance) properties of the system, thus any change in thickness directly changes the amount of energy feeding back into the PR.

For variations of the SiO\(_2\) refractive index a range of \( \pm 5\% \) was considered feasible as this layer will likely be slightly porous thus a lower refractive index should be considered, also the pores may be filled with PR in the actual experiment thus a slightly higher refractive index is also worth considering. Figure 5.24(c) shows the intensity traces for a \( \pm 5\% \) variation in the real component of the SiO\(_2\) refractive index, these values correspond to refractive index values of 1.3961, 1.4696, and 1.5431, where 1.4696 is the design value. The standing wave magnitude in this plot indicates that the system is relatively sensitive to variation in this parameter. Variations in \( \kappa \) for the SiO\(_2\) was also considered with refractive index values of 1.4696+0i, 1.4696+0.001i, and 1.4696+0.01i Fig. 5.24(d). The weak standing waves for relatively strong (\( \kappa = 0.01i \)) absorption indicates the system is not very sensitive to errors in this parameter.

The literature suggests that the HfO\(_2\) refractive index can have a fairly large range from 1.9 to 2.04 \([109]\), as such a refractive index error of \( \pm 5\% \) has been chosen for this layer, also the minimum value from the ellipsometry will also be used (the maximum value is within the \( \pm 5\% \) range). Figure 5.24(e) shows the effect of this variation for real refractive index (\( \kappa = 0 \)) values of 1.8305, 1.8814, 1.9804, and 2.0794, where 1.9804 is the design value. As with changing the HfO\(_2\) physical thickness, changing the refractive index changes the phase thickness of the layer, consequently the ARC is sensitive to
Figure 5.24: SiO\textsubscript{2}|HfO\textsubscript{2} ARC parameter variations. Design values in bold. (a) - SiO\textsubscript{2} thickness variation ±10%. (b) - HfO\textsubscript{2} thickness variation ±10%. (c) - ±5% variation of the real component of the SiO\textsubscript{2} refractive index. (d) - Variation of the imaginary component of the SiO\textsubscript{2} refractive index. (e) - ±5% variation of the real component of the HfO\textsubscript{2} refractive index. (f) - Variation of the imaginary component of the HfO\textsubscript{2} refractive index.
errors in this parameter as evidenced by the presence of strong standing waves. Adding absorption ($\kappa = 0i, 0.001i, \text{and} 0.01i$) to the HfO$_2$ has little impact on the standing wave magnitude (Fig. 5.24(e)).

This particular ARC was able to fully suppress standing waves within the PR layer. If one considers the manufacturing error plots for this ARC (Fig. 5.24) it can be concluded that the most sensitive parameters (HfO$_2$ thickness, HfO$_2$ real refractive index, and the SiO$_2$ real refractive index) must all have values close to the design values. The ellipsometry fit data backs up this hypothesis with the wafer ‘top’ location data being a good match for the design values. Also, as there is inter- and intra-wafer variation in parameter values the optimum values are likely to exist on this set of wafers, thus the success of this particular set of ARC experiments.

5.7 Summary

This chapter was dedicated to the experimental verification of the ARC effect of evanescent-coupled ARCs. An experimental primer was given detailing the Lloyd’s mirror interference lithography system employed in these experiments. Three film stacks were produced, a control of PR on silicon, a MgF$_2$|Cr SSR based ARC, and a SiO$_2$|HfO$_2$|Si modified trilayer DR based ARC. The photolithography process consisted of spin coating of the PR, PVA, and HMDS. A pre-bake stage was required to drive off the remaining PR solvent. The use of a post-exposure bake was precluded from these experiments as it is known for promote photoacid migration which reduces standing wave prominence. Although a post-bake may have improved the structural stability of the PR structures, the confounding effect of the reduced standing waves was deemed too much of a potential issue for its use. Both dose (PD) and NA sweeps were carried out to locate the best PR patterns. Finally the samples were developed and imaged using an SEM.

Typically an ARC will be demonstrated by showing free standing structures with no standing waves present. This requires at least two standing wave periods to unambiguously show that the ARC is operating, rather than say if the PR was thinner, being in a flat spot of a standing wave. For the pitch and standing wave pitch of the experimental ARCs two standing wave heights correspond to aspect ratios (height:full pitch) of 3:1 and 4:1, both of which are extremely difficult to achieve without massive resist collapse. To bypass this problem a thick film ($\sim 1.3\mu m$) PR cleave-develop method was employed. This method allows development from the side of the PR which allows
any free standing structures to be anchored at the back to the main body of the resist thereby reducing resist collapse.

The primary experimental proof for an ARCs efficacy is full standing wave suppression, but when one considers the resonant nature of these ARCs a second verification method presents itself. As these ARCs designs are resonant in nature they must have energy building up within the resonator. If this energy exceeds the intensity matching condition a hot spot will occur at the base of the PR. In contrast the control case of PR on Si has strong reflectance from the PR|Si interface, thus a node point will exist at the base of the PR. The strong contrast between these two effects is evidence to prove that the system is indeed resonating, but in a sub-optimal fashion.

PD sweeps and NA sweeps were performed on the MgF$_2$|Cr SSR based ARC to find the best PR structures. Although the correct pitch and full pattern clearance to the base of the PR was achieved, full standing wave suppression was not observed. Hot spots in the PR footing however were observed; this alteration of the footing indicates the ARC resonator was resonating, albeit in a sub-optimal fashion. Ellipsometry fits for the ARC coated wafers in this experiment indicated that there was likely significant errors in the manufactured ARC parameters, thus alteration of the PR footing was considered a good outcome for the first iteration of this experiment. To improve this experiment for further iterations the author suggests designing the ARC parameters around the manufactured values by building up a process of verification of the ellipsometry model. Further to this the PR thickness should be reduced to prevent the top of PR being over-exposed relative to the base of the PR. Alternatively a new SSR based ARC design may be necessary to shift the design NA to an NA value which has greater fringe visibility than the 0.406 fringe visibility value of this ARC design.

PD sweeps and NA sweeps of the SiO$_2$|HfO$_2$|Si system show successful production of both standing wave suppression and PR footing hot spots. Ellipsometry fit data and manufacturing error analysis suggest that the ARC coated wafers for this particular ARC design closely matched the design values in particular regions of the wafer. The manufacturing error analysis suggested that the error in the PR refractive index in the design process was not an issue for this system as the ARC design was shown to be fairly tolerant to the SiO$_2$ thickness reduction required to accommodate the more accurate PR refractive index value.

At this point we believe we have experimentally verified the concept of evanescent-coupled ARCs. The experimental aspects have been largely ironed out with a suitable apparatus, photolithography process, and experimental verification methods all well
developed. As such, further iterations of these designs/experiments would focus on the ARC manufacturing and design aspects, ideally with a manufacturing led design process to produce wafers with equal ARC parameters across the entire wafer which would allow for more consistent experimental verification.
Chapter 6

Genetic algorithm optimization of grating coupled near-field interference lithography systems

6.1 Prologue

In Chapter 2 the use of evanescent fields for patterning in interference lithography was discussed. Further to that, the concept of resonant underlayers to enhance the depth of field was introduced (Section 2.5), as well as the types of resonators available and the limits for coupling energy into them. There is, however, another more fundamental limit to these systems, namely, that energy must be able to couple into the system in the first place. Traditional near-field IL systems employ a prism as a coupling medium (Fig. 6.1(a)). If the NA exceeds the refractive index of the prism material the fields within it will be evanescent and hence not be able to couple into the system. This places a practical NA upper limit (at 405 nm) on the system of approximately 2.45 by using diamond [79] as the prism medium; this equates to a full pitch of 86.2 nm assuming a maximum accessible NA of 2.35. To push the NA further we need to discard the prism and start using diffractive coupling via a diffraction grating (Fig. 6.1(b)). The theme of this chapter is the modelling and optimization of grating coupled interference lithography systems with an aim of reducing the interference pitch as much as possible.

We will begin with an introduction to diffraction gratings and the concept of evanescent diffracted orders. We will then discuss the fact that to use these evanescent orders one must consider it as a near-field interaction, as such there are no suitable
6.1. PROLOGUE

Figure 6.1: IL coupling method comparison. (a) - Prism coupled IL. Incident beams refract with the prism and interfere where they overlap. (b) - Grating coupled IL. An incident plane wave strikes a grating producing diffracted orders \((0, \pm 1, \pm 2, \ldots)\) which may interfere. The presence of multiple diffracted orders requires this system to be designed to resonate with a particular diffracted order i.e. \(|m| = 1\).

simple and flexible analytical models to describe this system. Instead a full field simulation must be performed, in our case using the COMSOL Multiphysics® finite element method software, of which a brief primer will be given. The finite element method is a very computationally intensive process which is not particularly well suited for optimization within a large parameter space. For this reason an inverse design method using a genetic algorithm is employed. This method provides at least an order of magnitude improvement in optimization time over a full parameter sweep, and thus allows the exploration of new near-field IL systems.

With the MATLAB genetic algorithm toolbox integrated into COMSOL the ability to optimize complex grating coupled near-field interference lithography systems was realised. Initial trial models employing SPP or dielectric resonators are constructed first followed by dual top and bottom SPP and dielectric resonator systems. Once these base models have been established the possibilities for reducing the interference pitch by reducing the grating pitch or using the higher \(m\) diffraction orders was explored. The use of a genetic algorithm approach to the optimization of such a complex coupled multi-resonator near-field IL scheme has not previously been reported. This therefore represents another of the significant original research contributions of this thesis.
6.2 Diffraction gratings

Diffraction gratings are a fundamental component of the modern optics tool kit where they are employed as a highly compact light weight dispersive element critical to spectroscopic applications such as UV-Vis-NIR spectroscopy, fluorescence microscopy, or space telescopes. Rather than for wavelength-selection purposes here they are required for spatial frequency generation in near-field IL. Diffraction gratings are a research area unto themselves, as such in this section a brief introduction to diffraction gratings and their properties will be given whilst highlighting the most pertinent details in regards to this chapter.

When a monochromatic EM plane wave is incident upon a diffraction grating (Fig. 6.2) the transmitted wave splits into multiple diffraction orders which correspond to regions of constructive interference maxima. The angles of the diffraction orders ($\theta_m$) are given by

$$n_2 \sin \theta_m = n_1 \sin \theta_i + \frac{m \lambda}{p},$$

(6.1)

where $\theta_i$ is the angle of incidence in medium $n_1$, $\theta_m$ is the angle of the $m$th diffraction order in medium $n_2$, $m$ is the diffraction order value, $\lambda$ is the wavelength, and $p$ is diffraction grating pitch as shown in Fig. 6.2. One may note that this equation is in essence a modified version of Snell’s law ($NA_1 = NA_2$), thus the additional term ($m\lambda/p$) indicates that the $m \neq 0$ diffracted orders gain NA from interacting with the diffraction grating. It is this extra NA that is of interest to us in this research as, theoretically, $m$ has no upper limit therefore neither does the NA.

For this work we exclusively use normal incidence illumination ($\theta_i = 0$) thus under normal incident conditions Eq. (6.1) reduces to

$$\sin \theta_m = \frac{m \lambda}{p n_2}.$$  \hspace{1cm} (6.2)

The type of fields present on the $n_2$ side of the grating are determined by the $k$ vectors of the diffracted orders. The $k$ vector can be decomposed into its $x$ and $y$ components (assuming the classical diffraction mounting $k_z = 0$) as

$$k^2 = \left(\frac{2\pi n}{\lambda}\right)^2 = k_x^2 + k_y^2,$$  \hspace{1cm} (6.3)

As $k_x$ is the sine component of $k$, from Eqs. (6.1) and (6.3) we can obtain $k_x$ as

$$k_{x,m} = k n_2 \sin \theta_m = \frac{2\pi m}{p},$$  \hspace{1cm} (6.4)
thus each diffracted order generates travelling waves with an $x$-directed spatial frequency. For each order $k_{y,m}$ is given by

$$k_{y,m} = 2\pi \sqrt{\left(\frac{n_2}{\lambda}\right)^2 - \left(\frac{m}{p}\right)^2}.$$  \hspace{1cm} (6.5)

If we now consider Eqs. (6.2) and (6.5) we see that a cutoff $m$ value exist below which $\theta_m$ and $k_{y,m}$ is real and above which they are imaginary. These real and imaginary values correspond to propagating and evanescent fields respectively. Thus the transmitted fields will be some combination of these fields for the $2m + 1$ diffraction orders ($\pm m$ and $0^{th}$ orders). The way in which the diffraction orders combine is dependent upon the distance from the grating.

As we move away from the $n_1|n_2$ grating interface (Fig. 6.2), the total $\mathbf{E}$ field evolves through several regions. The propagating regions are generally referred to as the Fresnel near field and the Fraunhofer far field, both of which correspond to certain approximations about the shape of the interfering wavefronts \[182\]. The Fraunhofer far field regime is far enough from the apertures that the wavefronts can be considered planar; this region is where we see the ‘classic’ diffraction pattern. In contrast the Fresnel near field region is close enough to the apertures that the wavefronts must be considered to be spherical, thus producing a diffraction pattern where the maxima and minima shift depending on the distance normal to the aperture, and further secondary maxima and minima occur between the primary ones. Both of these regimes have

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{diffraction_grating.png}
\caption{Schematic representation of a transmission diffraction grating in the classical mounting configuration ($z = 0$).}
\end{figure}
well documented analytical solutions[63, 183]. Inside the Fresnel near field however is another region, which we’ll call the evanescent region, in this region evanescent fields are an appreciable component of the total electric field. The models in this chapter are designed to operate in this regime.

The analytical solutions in the propagating (Fresnel and Fraunhofer) regions make simplifying assumptions about the grating such as: it is infinitely thin; composed of a perfect conductor; and the fields are propagating with non-absorbing bounding media [63, 65, 75]. In the models we are constructing however none of these conditions are valid. The finite thickness of the grating produces diffraction and reflections as the fields transmit through the aperture. The grating and bounding materials are all real materials which act as both absorbers and sources in the case of plasmonic responses. For these reasons the evanescent region has no closed form analytical solutions for gratings of a finite thickness composed of real materials, and thus only full field solutions employing Maxwell’s equations are suitable for modelling this region. There are several different full field modelling techniques employed in the literature, in our case we use the finite element method which will be discussed later in the chapter.

The models developed in this chapter operate in the evanescent region and are all designed to operate with evanescent diffraction orders. If the pitch is small enough \( p < m\lambda/n \), all orders greater than or equal to \( m \) will be evanescent with only the \( 0^{th} \) order propagating. The intensity in each of the diffraction orders is dependent upon the grating design and angle of incidence, but generally speaking the higher the \(|m|\) the lower the amount of energy. This has important implications for this research where we attempt to employ these higher \( m \) diffraction orders.

We will reiterate several of these points as this chapter progresses, but at this point it is instructive to look at an idealised example system, discuss its main features, and then look at the progress so far reported in the literature.

### 6.3 Idealised grating coupled near-field interference lithography

Figure 6.3 shows an idealised version of the type of systems we seek to design. This system works as follows, a monochromatic plane wave incident from the top strikes the diffraction grating producing a multitude of diffraction orders. The coupling/filtering layer serves to both couple the fields and filter (ideally) the diffracted orders \((\pm m)\) so that only the resonant ones remain. The resonator is designed to resonate with
Figure 6.3: Grating coupled near-field interference lithography schematic - A normal incidence TM plane wave strikes the diffraction grating producing diffraction orders. A pair of coupled resonators are designed to preferentially resonate with one of these orders and produce an interference pattern within the PR. The coupling/filter layer is designed (ideally) to minimize the amount of non-resonant diffraction orders from reaching the resonator.

...
area so far and use this to settle on design goals for our systems.

Interference of evanescent diffraction orders for lithography was proposed and simulated by Blaikie and McNab [103], and subsequently experimentally verified by Luo and Ishihara [104]. To improve the field uniformity and enhance the DOF plasmonic underlayers were proposed [74, 84]. Initial papers employed the \( m = \pm 1 \) diffraction orders; simulation of higher diffraction orders and the subsequent decreases in interference period were first explored by Xu et al. [99] \((m = 1, 2)\) and Bezus et al. [184] \((m = 3, 4, 5)\). The first experimental use of a resonant underlayer in a grating coupled interference lithography system was published by Xu et al. who employed a Ag|PR|Ag structure to produce a \( \lambda/7.12 \) interference pitch of the \( m = \pm 1 \) diffraction orders [101].

The desire for smaller absolute interference pitches has seen the shift towards deeper UV wavelengths [185, 186, 187, 188]. As such they have shifted to 193 nm exposures utilizing aluminium as a plasmonic material, and often employ metamaterial type structures to filter the diffraction orders [185, 186]. Practical production of metamaterial type structures however is very challenging at this wavelength due to compounding thickness and roughness errors which severely impact field transfer through the structure [189, 125]. The difficulty of experimental verification of this type of structure (especially for 193 nm illumination) has led to the employment of simpler PMMA spacer (filter) Al superlens overlayer structures illuminated at 405 nm to produce 122 nm full pitch interference patterns over large areas (square centimetre areas) [190]. One of the goals of this chapter is to explore the possibilities of these simpler style of structures, with the aim of producing sub-40 nm feature sizes using 405 nm illumination.

The classic design methodology for these types of structures is somewhat of a guided search, i.e. equations are employed to match the \( k \) vectors of the diffracted orders to those of the resonant structures beneath, estimates of the layer thicknesses can be simulated using methods such as the transfer matrix method, but require further fine tuning once the full systems is modelled. Naturally as the complexity of the full system increases the accuracy of these estimates decreases while the fine tuning time increases. Here we seek to bypass these issues by employing a genetic algorithm strategy to automatically optimize these thicknesses and materials. The strength of this is that it allows us to use a so called inverse design process, that is, we ask what features we want (pitch, resonance type, materials, etc.) and the GA finds (ideally) a particular structure which fits our design criteria. Inverse design processes have not been applied to the full grating coupled near-field interference lithography, although they have been applied to components of the system. Macías, Vial et al. [194, 192] have
### Table 6.1: Grating coupled near-field IL literature development. Note: E. = experimental, S. = simulation, and all feature sizes specified are full pitch.
investigated surface relief grating parameters towards the optimization of interference fringe visibility at a plane 15 nm above the exit plane of an Ag grating. Several evolutionary algorithms were employed with the result being that it is possible to use these techniques for optimization although the uniqueness of the solution is poor. The optimal coupling of higher diffracted orders (±3) has been investigated by Bezus and Doskolovich using a merit function designed to optimize the SPP intensity at the wave vector corresponding to the excitation diffraction order [193]. The exact optimization procedure in this paper is not discussed.

Both Macías and Bezus’s methods are designed to find the grating parameters which produce the optimal surface plasmon interference pattern in terms of difference from an ideal surface plasmon interference pattern at a given line/plane beneath the grating. The use of a cut plane such as this is suitable for lithography systems where the fields only come from one direction. If however resonant underlayers are employed fields come from two directions, as such a single cut plane may not give a suitable representation of the fields present within the PR. For instance if one was to compare a PR midline cross-section where only the top resonator is resonating versus both the top and bottom resonating, depending on the resonance quality the cross-sections may be identical. For this reason we have chosen to do a 2D optimization process of the fields within the PR layer. Also as these optimization examples are at 532 and 550 nm respectively we aim to transfer the ideas to a more photolithographically appropriate wavelength of 405 nm.

As the utilization of a GA for grating coupled near-field IL has not been achieved or reported previously the aims of this chapter fall into two categories, firstly those related to the integration of the GA into system design, and secondly those related to optimization goals.

Grating optimization aims

- Integrate GA optimization into optical stack design

---

1Evolutionary algorithms are essentially a tailored form of GA, quoting Michalewicz “Evolution programs borrow heavily from genetic algorithms. However, they incorporate problem-specific knowledge by using ‘natural’ data structures and problem-sensitive ‘genetic’ operators. The basic difference between GAs and EPs is that the former are classified as weak, problem-independent methods, which is not the case for the latter.” [193][p.289] In practice this means evolutionary algorithms (or programs) are tailored using knowledge about the system to speed up the optimization process. This tailoring however makes it less flexible, consequently GAs are slower but better suited for ‘open’ type problems.
- Produce both top and bottom resonator systems
- Increase the NA as high as possible (minimize pitch)
- Use the highest $m$ orders possible
- Investigate optimum layer materials

6.4 Finite element method - COMSOL

Simulations of classical electromagnetic systems such as those in this section, are entirely explained by Maxwell’s equations, its constitutive relations, and the Lorentz force law. Although these equations are capable of completely describing any macroscopic electromagnetic system, closed form analytical solutions are often only available for specific simple symmetric systems generally without internal sources. The systems we seek to simulate however are unfortunately not simple, with plasmon resonances, evanescent diffracted orders, and coupled resonators. There are many numerical strategies for solving these type of problems including (but not limited to): finite element method (FEM), finite-difference time-domain, rigorous coupled-wave analysis, and finite integration method [75, 104, 196, 197, 100, 101]. The method chosen for this research was FEM as it has a good track record for modelling systems such as ours, and COMSOL (the software employed) has a tight integration with MATLAB allowing the customization and optimization of the design process from the MATLAB front end.

FEM is a commonly employed technique for simulation of systems best described by partial differential equations (PDEs). Its greatest strength is its ability to operate over complex domains where the required precision may vary over the domain. The heart of the method is the discretization of the continuous functions which describe the system. This discretization is carried out over many individual subdomains, which are easier to analyse than the full system. The production of these subdomains is termed meshing, and in Fig. 6.4(a) we see an example of this. In this case we are using triangular mesh elements which are most suitable for a 2D structure such as ours. Dynamic meshing is employed to produce less dense meshing in low detail areas such as the cover and substrates, and high density meshing within thin layers and regions of high field gradients such as at grating corners. This provides an improvement in calculation time without sacrificing accuracy.

Each mesh element is described by an unknown field $\phi_i(x, y)$ that must satisfy Maxwell’s equations. The field $\phi_i(x, y)$ within the element $i$ can be approximated by
6.4. FINITE ELEMENT METHOD - COMSOL

Figure 6.4: COMSOL model mesh and layout. (a) An example of COMSOL’s dynamic meshing, note the greater mesh density at the grating corners. The simulations in this work actually employ a much denser but less visually distinguishable ‘extremely fine’ mesh. (b) Example COMSOL model system showing the key components including the geometry, PMLs, an excitation port, and Floquet boundaries.

a function \( \tilde{\phi}_i(x, y) \) which is determined by the geometry of the mesh element and the node values. The fields at the element boundaries must satisfy Maxwell’s equations boundary conditions. The full simulation function is the sum of the individual element functions

\[
\Phi(x, y) \approx \sum_{i=1}^{n} \tilde{\phi}_i(x, y),
\]

which is substituted into the wave equation

\[
\nabla \cdot \nabla \Phi + k^2 \Phi = 0,
\]

where \( k \) is the wave vector. As Eq. (6.6) is just an approximation, Eq. (6.7) will not equal zero and a residual will remain. The software will select coefficients and forms of \( \phi_i \) to minimise the residual, produce the nodal values, and thus simulate the full fields.

6.4.1 Model setup

The RF module of COMSOL was used to construct a model of a 405 nm wavelength TM plane wave at normal incidence upon a grating/dual resonator structure as shown in Fig. 6.4(b). The incident wave is produced from an excitation port at the upper
PML|SiO$_2$ interface (blue line). The excitation port is a forward oriented domain-backed port. The top and bottom layers are perfectly matched layers (PML). A PML is a fictitious material designed to absorb any waves entering it without producing reflections. PMLs act essentially as infinite bounding media. As the model is symmetric and periodic about the $y$-axis Floquet boundary conditions are employed on the sides. Floquet boundary conditions apply a phase shift to the tangential components of $\mathbf{E}$ and $\mathbf{H}$ which is determined by $k$ and the point to source distance. This effectively creates a model of an infinite grating in the $x$ direction. An automatic extremely fine mesh is applied. An example run of the model in Fig. 6.4 using the extremely fine mesh produces a mesh with 13244 mesh elements with a minimum area of $2.661 \times 10^{-17}$ m$^2$ and a maximum area of $8.168 \times 10^{-17}$ m$^2$, although naturally the mesh statistics change as the model alters the model thicknesses.

The models employed in this chapter were systematically optimized using the MATLAB genetic algorithm global optimization toolbox. It will now be discussed what exactly a genetic algorithm is and how it has been implemented for optimization of grating coupled near-field IL systems.

### 6.5 Genetic algorithm - MATLAB

As was previously mentioned, modelling of grating coupled near-field interference lithography systems is a very computationally intensive process. Increasing the size of the parameter space therefore leads to large increases in computation times, as such the need for an efficient optimization strategy is obvious. In this section we will discuss the chosen optimization strategy for this research, that of, genetic algorithms (GA). The choice of GA optimization was made because GAs are known to be suitable for optimization of systems with large parameter spaces where the function describing the system likely contains multiple local minima and is not well known [198].

The relationship(s) between the diffraction grating (forward and back) coupling as well as the resonator couplings is not a simple one, thus the necessity for finite element simulation. Because of this, the function describing the fitness (sometimes referred to as the penalty) across the entire parameter space is not known and may contain multiple minima; as such gradient optimization methods are not suitable for these systems and a stochastic optimization method is applied.

Genetic algorithms are an optimization technique designed originally to mimic the process of natural selection through a selection process (i.e. survival of the fittest) and
6.5. GENETIC ALGORITHM - MATLAB

<table>
<thead>
<tr>
<th>Term</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fitness</td>
<td>A measure of an individuals suitability</td>
</tr>
<tr>
<td>Individual</td>
<td>A single member of the population</td>
</tr>
<tr>
<td>Population</td>
<td>The number of individuals in a single generation</td>
</tr>
<tr>
<td>Generations</td>
<td>Number of population iterations</td>
</tr>
<tr>
<td>Parents</td>
<td>Members of the previous generation selected to produce children</td>
</tr>
<tr>
<td>Children</td>
<td>Offspring of the previous generation</td>
</tr>
<tr>
<td>Elite children</td>
<td>Clones of previous generations fittest individual</td>
</tr>
<tr>
<td>Crossover children</td>
<td>A combination of two parent’s parameters</td>
</tr>
<tr>
<td>Mutation children</td>
<td>Children produced by random changes to a parent’s parameters</td>
</tr>
<tr>
<td>Selection</td>
<td>The selection of parents for the next generation.</td>
</tr>
<tr>
<td>Stop criteria</td>
<td>A set of conditions which if any are met the GA terminates</td>
</tr>
</tbody>
</table>

Table 6.2: Glossary of GA terms

population evolution. As such there are three components to any genetic algorithm, firstly a method of determining the fitness of the individuals within a generation, secondly a means by which the population may reproduce and evolve, and thirdly constraints for which the algorithm will stop. In this section we will introduce GAs, the methodology of their implementation in general and specifically how they are employed in this context.

For the sake of familiarity a simple description of the GA optimization process will now be given, with the details and terminology further explained in the sections to follow. Figure 6.5 is a flow chart of how the GA is utilized in this work; a glossary of GA terms is also given in Table 6.2. The process starts by generating an initial (ze-roth generation) population of starting grating/resonator/PR parameter values. These individuals are then simulated in COMSOL and their fitnesses determined. If a stop criteria has been met the optimization process will terminate; if a stop criteria has not been met the optimization process will go into a loop where subsequent generations are produced from the previous generation by varying grating, resonator, and/or PR parameters until a stop criteria has been met.

6.5.1 Fitness

The fitness of an individual is an objective ‘valuation’ of the individual based on some predetermined set of desired properties. For instance in a living organism this may
Figure 6.5: GA flow chart - The process begins with the random generation of an initial $0^{th}$ generation. The $0^{th}$ generation is simulated in COMSOL and the fitnesses of the individuals determined in MATLAB. If a stop criteria has been met the process terminates. If not, the process goes into a loop producing subsequent generations which are simulated and their fitnesses calculated until a stop criteria has been met. Orange objects represent those stages carried out in MATLAB, blue objects represent those carried out in COMSOL.

represent individual growth rate, reproductive success, group success, etc. The key to the use of a fitness metric is that it must be an unambiguous measurable quantity to allow correct weighting of the population reproduction. In the case of grating coupled near-field IL the fitness is determined by the electric field distribution within the PR layer.

The ideal field distribution within the PR is a tapered exponential or catenary interference pattern originating from the resonator layer(s) as in Fig. 6.3. This can be modelled as the product of two functions, a cosine function in the $x$ direction representing the interference pattern, and evanescent fields originating for the resonator(s). This ideal PR field distribution is the optimal field distribution for the systems we seek to design, as such, we define the fitness of an individual in relation to this. For the purpose of optimization the fitness of an individual is defined as the sum of the square
of the error (SSE)

\[ SSE = \sum_{i=1}^{n} (y_i - f(x_i))^2, \]

between the ideal PR intensity distribution \( (y_i) \) and PR intensity distribution of the individual \( (f(x_i)) \) where \( i \) designates the particular pixel within the PR layer.

### 6.5.2 Population parameters

The GA population parameters heavily influence the speed and accuracy of the optimization process. These parameters (and GAs in general) are capable of a great deal of customization \[195, 198, 199\] for the purposes of this research however we prefer the simpler approach provided by the MATLAB GA toolbox so we may concentrate on exploring grating coupled interference systems rather than the intricacies of GA customization. In this section we will cover the population parameters of the GA giving a general description of each and how they have been applied in this body of work.

#### Population size and generation number

The first step in any GA is the production of the initial population (0\(^{th}\) generation). The MATLAB GA by default generates a randomly distributed population spanning the parameter space, although if one knows generally the locus of the optimum the parameter space can be tightened up. With the use of TMM modelling (Section 3.3) we are able to predict suitable parameter ranges, particularly for the resonator layers.

The population size of each generation is also an important factor in GA optimization. A suitable population size is inherently dependent on the type of problem being optimized and the parameters by which the next generation is produced (i.e. Elite, Crossover, and Mutation children proportions). A small population generally requires a greater number of generations to find the optimum, and vice versa. The primary consideration here is that the parameter space must be adequately covered to be able to locate the optimum. Increasing the population size improves the parameter space coverage, but as the population size approaches that of the parameter space the method becomes largely pointless. Increasing the number of generations allows more mutations to occur thereby providing improved coverage of the parameter space with a smaller population size. Although as a minimum the population size must be at least that of the number of dimensions of the parameter space.

If the problem has a well defined and known value extrema an optimum population size can be ascertained by running the optimization process many times and locating
the population size where the number of false optimums is suitably low for the problem at hand [200]. For problems with large simulation costs, the very process of proving a false optimum can be so time consuming as to render this method of determining the optimum population size infeasible. As such, a trial and error approach is often applied. A useful method of determining this is by monitoring the progress in fitness improvement as the optimization process runs. If the improvements stall for several generations then one can assume either the optimum has been located or the optimization process is stuck in a local minima. This touches on a key point of GAs: as this method is fundamentally stochastic the optimization process must be run multiple times to verify that the (or a suitable) optimum has been located, although if necessary a broad starting range can be narrowed for subsequent optimization trials.

The optimum of the systems in this work appear to be regions of low curvature connected by multiple arms corresponding to the local optimum for each parameter. The low curvature means the optimization process slows down as it approaches the global optimum. This fact combined with the lack of a known (or quickly provable) optimum value means highly optimized population and generation numbers are not available. Population values ranged from 10-25 and Generation values from 10-15, with the higher values being applied to larger parameters spaces. These values depend heavily on the proportions of Elite, Crossover, and Mutation children.

**Parent selection and the production of children**

Once the fitness of the initial population members has been analysed suitable parents must be chosen for the next generation. A stochastic uniform selection method was employed here. This method produces a line where each member of the previous generation is represented by a length scaled by its fitness value, thus fitter individuals occupy a greater proportion of the total line. Starting from a random location (less than the step size) the algorithm moves along the line in equal spaced steps selecting parents until the required number of parents have been selected. The following generation is then produced by three means of Elite, Crossover, and Mutation reproduction which must be judiciously balanced.

Elite children are those which are exact replicas of the fittest individuals in the parent population. Their role is to prevent the GA fitness from worsening as the optimization proceeds. The value for the elite count depends on the population size and the function space being optimized. If the number of elite children is too high in proportion to the population size the elite children may cluster in the same minima,
thereby making the optimization process slower and more likely to get stuck in a local minima. The population size for these simulations is low (generally between 10-25), therefore we use a single elite child to reduce this clustering effect.

Crossover children are a random combination of the two parents gene sets (parameter values). As one of the parents is often an elite member, the role of crossover children is (effectively) to scan the parameter space near the elite children. For this work a crossover fraction of 0.8 was used. This means that for a population size of 10 with one elite child the number of crossover children will equal 7, i.e. \((10 - 1) \times 0.8 = 7.2\), bearing in mind that we can only have an integer number of individuals. The remaining two individuals are mutation children.

Mutation children are produced by randomly altering the genes of an individual parent. Mutation children are a crucial component of GAs as they allow the optimization process to effectively cover a greater region of the parameter space and reduce the chances of the process getting stuck in a local minima. The number of mutation children is important as too little and the GA will not adequately explore the parameter space, but too many and it will not have enough crossover children to quickly explore the parameter space adjacent to the optimum values and thus will likely slow down the optimization process.

Once the next generation has been generated the system calculates the fitness of these individuals and produces subsequent generations; this process continues until a stop criteria has been met.

### 6.5.3 Stop criteria

Stop criteria are a set of stopping conditions which if met at any stage during the optimization process will stop the algorithm; examples include fitness value, generation limit, number of stall generations, calculation time limit, and function tolerance constraints. As the optimum fitness value for our systems are not known generally we set the fitness tolerance to be an order of magnitude less than what we expect the algorithm to reach, thereby preventing the system from stopping prematurely. Due to this the fitness tolerance stop criteria is never met, thus we use a more appropriate stop criteria, that of, a generation limit. To decide if a global optimum has been found the GA must be run several times, as such an idea of a suitable generation limit can be ascertained from this provided multiple GA trials produce very similar results. The use of a stall generations criteria would be useful, but we do not know enough about the optimum fitness value to specify a suitable minimum average fitness change.
6.6 System development

In this section a series of base models will be built up to satisfy the initial design goals of integrating GA optimization into grating coupled near-field IL optical stack design, and producing models employing top, and dual top and bottom resonators for both SPP and dielectric resonators. These simulations were built up sequentially with various goals needing to be met along the way. As such, this section will introduce different models, building up complexity, pointing out the most important features of each and reiterating these points where relevant later on.

6.6.1 SPP overlayer resonator

The initial trial models constructed were for SPP overlayer resonator systems where the coupling/filter, and SPP layers were optimized. SPP systems were chosen first as the broader resonance profile of these systems would likely lead to a more forgiving design profile in terms of resonator thicknesses thus the GA should easily be able to find a reasonable solution. Figure 6.6(a) is a schematic for this system consisting of a Cr grating embedded within an SiO$_2$ layer, a Mg layer as an overlayer resonator, and a PR bottom layer. The lower PML, although present in all these models, is only shown in the plots for the SPP and dielectric overlayer models to illustrate how the fields taper off in this medium. All models in this chapter employ normal incident TM illumination at a wavelength of 405 nm. The details for this model are given in Table 6.3.

The grating pitch for SPP resonator based systems is determined by the need for the $k_{x,m}$ component of the diffracted fields to match that of the plasmon resonance condition ($\beta$), i.e.

$$Re(\beta) = k_{x,m}$$  \hspace{1cm} (6.9)

and

$$Re(k\sqrt{\frac{\varepsilon_1\varepsilon_2}{\varepsilon_1 + \varepsilon_2}}) = \frac{2\pi|m|}{p},$$  \hspace{1cm} (6.10)

where $m$ is the diffraction order, $p$ is the grating pitch, $k$ is the wave vector and $\varepsilon_{1,2}$ are the dielectric constants for materials 1 and 2. When this condition is met the chosen diffraction order will optimally couple into the plasmon resonance mode. This resonance can occur at (depending on the design) either the grating interfaces or as in this case in a separate interface beneath the grating. This configuration was chosen as having the resonance at the PR interface results in greater fields within the PR.
Table 6.3: SPP overlayer resonator COMSOL model details. cyan highlighted rows are optimization parameters. Material references: SiO$_2$[121], Cr[168], Mg[158], and PR[119].

For this system resonance occurs at the Mg|PR interface at an NA of 1.9056+0.0718i, which equates to a grating pitch of 212.5 nm to optimally excite the SPP resonance with the $m = \pm 1$ diffraction orders resulting in a 106.3 nm interference pitch. This is already better than can be achieved with conventional prism-coupled EIL without resorting to exotic prism materials such as diamond. Alternatively this system could be optimized to resonate at the SiO$_2$|Mg interface, but the lower NA (1.6111+0.0164i) is not useful to us as the fields it produces are in the propagating regime within the PR, as well as being heavily diminished from having to traverse the Mg layer.

In Fig. 6.6(b) we see the norm of the total E field ($\text{normE} = \sqrt{E_x^2 + E_y^2 + E_z^2}$) for an optimized model of this system (parameter optimization plots are shown in Figs. 6.7(b) to 6.7(d)). There are two things of particular note in this figure. Firstly, dark protrusions are visible extending from the bottom corners of the Cr grating down to the Mg layer. These are due to localised plasmon resonances [201]. The TM polarized light has an in-plane $E_x$ field which drives the free electrons in the Cr to the corners of the grating, this is evident by hot spots at the corners of the gratings. This effect can be reduced by the use of ‘filleting’ which is the rounding of the corners to reduce the build up of charge [75]. It does however have the downside of further increasing the

Note: for clarity the author has tried to be careful to refer to the pitch within the PR as the interference pitch, and the diffraction grating pitch as the grating pitch.
Figure 6.6: Plasmon overlayer resonator - Optimized COMSOL simulation. (a) - System schematic. (b) - Normalized $E$ field. (c) - $E_x$ component. (d) - $E_y$ component. Note: all simulation plots have been windowed to highlight the fields within the PR.

calculation time due to the increased complexity of the meshing on these corners. Also it is difficult to ascertain what a suitable filleting radius of curvature is \cite{115}, thus we have decided not to employ filleting in this work. Secondly, SPP resonance is visible as hot spots at the Mg|PR interface, and most importantly we can see the fields from this extend in to the PR producing the desired interference pattern.

Figures 6.6(c) and 6.6(d) show the $x$ and $y$ components of the $E$ field respectively. The $E_x$ component maximum occurs in the SiO$_2$ spaces within the grating due to gap SPP modes \cite{202}. Directly beneath this maximum a small amount of this field is seen to penetrate into the PR. The $E_y$ component, being the dominant (greatest magnitude) component beneath the grating, closely mirrors the normalized $E$ field (Fig. 6.6(b)). It is also noticeable that the fields above and below the Mg layer have an antiphase relationship; this is referred to as an antisymmetric surface plasmon polariton mode (explained further shortly).
Figure 6.7: Plasmon resonant overlayer model - PR fields, GA population distribution, and parameter evolution (a) - PR fields - (top) simulated E fields, and (bottom) ideal E fields. (b) GA population distribution. Population = 10, Generations = 10. (c) - SiO$_2$ population evolution. Note the rapid discovery of the 39 nm optimum. (d) - Mg population evolution. Note the slower discovery of the 56 nm optimum.

Figure 6.7(a) shows (top) the optimized field distribution within the PR, and (bottom) the ideal field distribution where SiO$_2$ and Mg layer thicknesses have been used as the variable for optimization. We can see that a good match has been found between the optimized model field distribution and that of the ideal case of an evanescently decaying interference pattern of equal amplitude consisting of only the $m = \pm 1$ diffraction orders with zero amplitude for all other diffracted orders. The primary difference is the broadening of the interference peaks especially beneath the grating solid areas, due to the effect of the $E_x$ component and some residual $0^{th}$ order propagation.
In Fig. 6.7(b) we see the GA population distribution for the entire optimization process. This type of plot is important to note when one is performing a GA optimization. Although running the GA multiple times is always advisable to verify the optimum has been found, it is also important to verify that the particular optimization process adequately covers the parameter space. In this plot we see the initial population is dispersed well across the full parameter space; this generally allows the GA to find the optimum faster as it means the GA is not relying so much on the mutation children to cover the parameter space. The ‘roles’ of each of the different children types is also visible here; the elite children maintain the optimum points for each generation, the crossover children search the neighbouring space for better solutions, and the Mutation children randomly search the parameter space. The relative clustering in the $x$ axis versus the $y$ is indicative of the system being more sensitive to the SiO$_2$ thickness. This is better illustrated in Fig. 6.7(c) where the SiO$_2$ population evolution shows a strong affinity towards the 39 nm mark. The ‘hotness’ of each point on this plot indicates the frequency of this particular value within that generation. In contrast the Mg population evolution (Fig. 6.7(d)) takes a greater number of generations to settle around the 55 nm thickness mark. The broader spread of the Mg population across all generations indicates the system is less sensitive to this parameter. This is likely to be due to the fact that plasmonic resonances are an interface effect, and thus less sensitive to the individual layer thicknesses.

**Symmetric/Antisymmetric SPP modes**

Looking at the $E_y$ fields either side of the Mg layer in the optimum solution Fig. 6.6(d), it is noticeable that the fields are of opposite signs for the same $x$ coordinate. This is indicative of the fields being in an antisymmetric configuration. The standard surface plasmon polariton relation (Eq. (6.10)) is for an interface between two effectively infinite media. This is not the case in these systems which consist of one (or two) metal layers sandwiched between two dielectrics. Provided the metal layer is thin enough the plasmon resonance fields at the top and bottom interfaces of the metal layer can couple producing symmetric and antisymmetric SPP modes. The physics of multilayered resonant modes is a rich one which is too extensive to explore here; instead we shall discuss the SPP modes present in a symmetric dielectric system (i.e. $\varepsilon_d|\varepsilon_m|\varepsilon_d$). Although we don’t have a symmetric dielectric system, this model is suitable for explaining the fields seen in these simulations.
The equations for these two modes are 

\[ k_{z,m}t = 2 \text{atanh} \left( \frac{-\varepsilon_m k_{z,d}}{\varepsilon_d k_{z,m}} \right), \]  
(6.11)

\[ k_{z,m}t = 2 \text{atanh} \left( \frac{-\varepsilon_d k_{z,m}}{\varepsilon_m k_{z,d}} \right), \]  
(6.12)

where \( t \) is the thickness of the metal layer, \( \varepsilon_{m,d} \) is dielectric constant of the particular medium, and \( k_{z,m,d} \) is the \( z \) wave vector in each medium given by \( k_{z,m,d} = (k_x - \varepsilon_{m,d} k_0)^{1/2} \). The \( H_y \) field distributions for these two modes is shown in Fig. 6.8. Often the antisymmetric mode is of interest as for a fixed metal layer thickness the \( k_x \) vector for the SPPs is larger than for the symmetric mode; thus the antisymmetric mode produces a shorter interference pitch. In our case however the metal layer is free to change to the value which optimizes the field profile within the PR, as such, over the course of GA optimization the thickness will change to suit whichever of these two modes produces the best field profile.

Throughout the course of these simulations for the SPP based resonator systems, the GA optimized towards antisymmetric SPP mode configurations. This can be understood by considering the field profiles within the metal layer a schematic of which is shown in Fig. 6.8. The key thing to note is that the antisymmetric mode has less fields (area) within the metal layer. As a consequence of this the antisymmetric mode suffers less loss within the metal layer compared to the symmetric mode. Due to this
a greater proportion of the available fields reaches the PR. As this field produces the interference, the stronger it is compared to the background fields the more the GA will optimize towards it, therefore the GA selects towards antisymmetric solutions.

**Grating thickness and duty cycle values**

For this system the grating thickness was set to 50 nm and the duty cycle to 50%. The primary reason for this was to simplify and speed up the GA process while the testbed overlayer and dual overlayer and underlayer systems were developed (Sections 6.6.1 to 6.6.4). In the sections following the testbed systems the grating thickness and duty cycles are employed as optimization parameters, although the duty cycle is limited to 0.25 – 0.75% to allow the gratings to be manufacturable.

Generally speaking optimizing over these two parameters produces one of two results, each with a duty cycle less than 50%, examples of which are shown in Fig. 6.9: an intermediate duty cycle thin-layer result (Figs. 6.9(a), 6.9(c) and 6.9(e), duty cycle = 25%), and a small duty cycle thick-layer result (Figs. 6.9(b), 6.9(d) and 6.9(f), duty cycle = 12%). Firstly if the duty cycle has an intermediate value (20 – 80% say) the optimum grating thickness is that which produces a resonant mode within the grating aperture with 1 antinode in the middle (Fig. 6.9(c)). If the duty cycle is allowed to vary across a larger range (5 – 95%) the optimization process will settle upon a solution with a narrow grating aperture and a grating thickness which produces a higher order resonant mode (Fig. 6.9(f)). As expected these plots show that the larger duty cycle produces stronger PR fields. But if one looks at the E fields within the PR, particularly the \( E_y \) components it is evident that the narrow grating aperture solutions produce better refined PR fields, as is born out by the optimization process where the larger duty cycle solution has a fitness value (lower is better) of 2230.6, and the lower duty cycle solution fitness value of 1335.4, which is significantly better in this context. This result is also somewhat expected as a thicker grating coupled with a narrower aperture results in less \( 0^{th} \) order transmission, thus the \( 1^{st} \) order interference pattern should be proportionately greater than in in the thinner grating and broader aperture simulation.

Although the smaller duty cycle example has a better fitness value, it clearly has weaker fields within the PR (compare Fig. 6.9(c) and Fig. 6.9(d)). The maximum PR field in the small duty cycle example is 21% of the maximum field in the cover, while for the larger duty cycle example the maximum PR field is 40% of the maximum field in the cover. Thus if one is going to use such a system in practice one must consider the trade off between pattern fidelity, dose, exposure time, and throughput.
Figure 6.9: Grating thickness and duty cycle optimization effects. (a) - Intermediate duty cycle (25%) system schematic. (b) - Low duty cycle (12%) system schematic. (c) - Intermediate duty cycle norm $E$ fields. (d) - Low duty cycle norm $E$ fields. Note the weaker fields beneath the grating compared to (c). (e) - Intermediate duty cycle $E_y$ fields. (f) - Low duty cycle $E_y$ fields. Note the different mode order and cleaner fields within the PR compared to those in (e).
6.6.2 Dielectric overlayer resonator system

Dielectric overlayer resonator systems employ a high-$n$ waveguiding medium above the PR as an alternative to SPP overlayers; the fields are propagating within this layer but evanescent in the bounding layers thus allowing the system to resonate provided the waveguiding phase condition has been met. The resonance condition must be satisfied for energy to build up in the resonating overlayer, and consequently for (an appreciable amount of) energy to enter the PR. In previous chapters it was mentioned that dielectric resonators were of interest as they can be designed to resonate in TE as well as TM configurations; this is not of use here as only the TM components can appreciably pass through a sub-wavelength grating \[75\]. For this reason all models in this chapter employing dielectric resonators also do so under TM polarized illumination conditions.

In this example we have a dielectric overlayer resonator composed of an SiO\textsubscript{2} cover medium, Cr grating, SiO\textsubscript{2} filter/coupling layer, HfO\textsubscript{2} high-$n$ resonator layer, PR, and a PML (Fig. 6.10), the details of which are given in Table 6.4. As in the plasmonic example, the grating period must be designed to produce the same $k_{x,m}$ vector as that of the design $k_x$ (NA) of the resonator. In this case the design NA is 1.75 which equates to a $k_x$ value of $2.715 \times 10^7 \text{ m}^{-1}$ ($k_x = k \text{NA}$), and hence a grating period of 231 nm ($|m|2\pi/k_x$) for the first diffraction order. The dielectric overlayer resonator is

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<th>Details</th>
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<tr>
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<td>m</td>
<td>$, $</td>
</tr>
<tr>
<td>NA</td>
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<td></td>
</tr>
<tr>
<td>Interference pitch</td>
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<td></td>
</tr>
<tr>
<td>Grating pitch</td>
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<td>231 nm</td>
</tr>
<tr>
<td>Grating thickness</td>
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<td></td>
</tr>
<tr>
<td>Grating duty cycle</td>
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<tr>
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Table 6.4: Dielectric overlayer resonator COMSOL model details. GA optimized parameters highlighted in cyan. Material references: SiO\textsubscript{2}[121], Cr[168], HfO\textsubscript{2}[154], and PR[119].
Figure 6.10: Dielectric overlayer resonator - Optimized COMSOL simulation. (a) - System schematic. (b) - Normalized $E$ field. (c) - $E_x$ component. (d) - $E_y$ component. Note: all simulation plots have been windowed to highlight the fields within the PR.

an SiO$_2$|HfO$_2$|PR trilayer. The optimization process is designed to optimize the SiO$_2$ and HfO$_2$ layer thicknesses with grating thickness set to 50 nm and the duty cycle to 50%.

Figure 6.10(b) is an optimized dielectric resonator overlayer system. The fields around the grating layer are very similar to the previous SPP overlayer example (Fig. 6.6(b)) as the exposure conditions are the very similar, with the only difference being the increased grating pitch of 231 nm (vs 212.5). At this point it’s worth noting that the COMSOL plots for different systems all have different scales so visual comparisons between systems are just that. Within the high-$n$ HfO$_2$ layer we see the hallmarks of waveguiding (periodic hotspots in the $x$ direction), indicating the system is operating at or near resonance. Beneath this we see the desired interference pattern extending into the PR layer.
The $E_x$ fields (Fig. 6.10(c)) show the same bright spot within the grating apertures as the plasmon overlayer system, although more noticeable due to windowing [202]. The $E_y$ fields (Fig. 6.10(d)) show strong waveguiding within the HfO$_2$ layer. This is markedly different from the interface bound resonances of the plasmon example (Fig. 6.6(d)). Within the HfO$_2$ layer waveguiding fields in the $x$ direction are clearly visible. In the bounding layers we see the effect of the refractive index upon the evanescent field decay length. The higher index of the PR ($n=1.684 \pm 0.031i$) has a greater decay length, thus the evanescent fields from the dielectric waveguide extend further into this layer than the upper bounding SiO$_2$ ($n = 1.47$) layer. Furthermore,
when we consider the $E_x$ and $E_y$ components together, we can see why the resonator fields and the PR fields in the normalized total $E$ field appear to ‘arch’ in a little due to the influence of the $E_x$ hot spots at the edges of the HfO$_2$ layer on the dominant $E_y$ component.

If the optimized PR field distribution is compared to the ideal field distribution (Fig. 6.11(a)) we see a poorer optimization than that of the plasmon resonator overlayer system. This is likely due to two reasons: firstly as was previously mentioned the increased $E_x$ component extending into the PR will impact the PR field distributions; and secondly the increased grating aperture (231 nm vs. 212.5 nm) will result in greater $0^{th}$ order transmission.

The population evolution of the optimization parameters (SiO$_2$ (Fig. 6.11(c)) and HfO$_2$ (Fig. 6.11(d))) indicate the system is most sensitive to the high-$n$ HfO$_2$ layer thickness. This is entirely expected as dielectric waveguides have a well defined thickness required for the waveguiding phase condition to be met. The optimum value for this (118 nm) is found rapidly in approximately 5 generations. The optimum thickness for the SiO$_2$ layer is less well refined, taking approximately 10 generations to strongly settle on a value of 75 nm.

Now that we have shown we can optimize overlayer resonator systems we shift our attention to dual overlayer/underlayer systems.

### 6.6.3 SPP-SPP dual resonator systems

We now extend the GA to SPP resonators above and below the PR to provide improved depth of field. The more complex (refined) ideal PR distribution should in theory make the GA more efficient at solving this problem as the solution should be more unique.

The design for this model consists of a SiO$_2$ cover, a Cr grating, a SiO$_2$ filter/coupling layer, a Mg top SPP layer, PR, a Mg bottom SPP layer, and a Si substrate Fig. 6.12(a), with the optimized parameters given in Table 6.5. The optimization process is carried out on three parameters: the filter/coupling layer and the two SPP resonators; once again the grating thickness and duty cycle are fixed for simplicity. This system is designed to operate at an NA of 1.9056 which corresponds to the SPP resonance for the Mg|PR interfaces. A schematic and the electric field plot for an optimized example are shown in Fig. 6.12(b). As for all SPP based systems the grating pitch is designed to produce a $k_{x,m}$ matched to the plasmon resonance $\beta$. This model is very similar to the overlayer plasmon example (Fig. 6.6(b)), except with the addition of the thin Mg plasmon bottom layer. An interference pattern is visible within the
Table 6.5: Dual plasmon resonator COMSOL model details. GA optimized parameters highlighted in cyan. Material references: SiO$_2$[121], Cr[168], Mg[158], PR[119], and Si [122].

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<tr>
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<tr>
<td>Grating duty cycle</td>
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PR layer. Both the $x$ and $y$ components of $E$ (Figs. 6.12(c) and 6.12(d)) have been scaled to highlight the fields within the PR and adjacent layers. This is required as the fields around the gratings far exceed those within the PR layer and hence saturate the scale colourbar. The $E_x$ component shows periodic features within the PR. The $E_y$ component has antisymmetric SPP resonances at the Mg|PR interfaces. The fields from these resonances stretch into the PR producing the interference pattern. The magnitude of the $x$ and $y$ fields within the PR are approximately the same, thus the broader $E_x$ components are responsible for reducing the interference minima within the PR (Figs. 6.12(b) and 6.13(a)).

The comparison of the optimized and ideal field distributions (Fig. 6.13(a)) shows the resonance in the bottom layer is underpowered resulting in a slightly asymmetric field distribution within the PR. The bottom resonator is underpowered due to the Si substrate reducing the resonance ability of the SPPs. If the substrate is changed to SiO$_2$ greater resonance can be produced in the bottom resonator. The influence of the aperture and the solid areas of the grating is once again apparent in the optimized field distribution with deeper nulls beneath the apertures.

Looking at the population evolution of the parameters (Figs. 6.13(b) to 6.13(d))
we see that the optimization of this system is most sensitive to the thickness of the bottom Mg plasmon resonator layer with an optimized value of 7 nm. This likely has a very fine balance due to the need to resonate but not bleed too much energy into the Si substrate. The SiO$_2$ thickness narrows down to 38 nm in approximately 7 generations, while the top Mg resonator takes approximately 10 generations to settle down to 46 nm. The length of time for the SiO$_2$ and top Mg parameters to settle down is likely due to their being strongly dependent on each other with a greater sensitivity to the Mg thickness.

The optimization process for this model employed a population size of 15 and 15 generations. A larger population and generation size is required to account for the greater parameter space. The ‘traditional’ GA optimization progress plot is shown in Fig. 6.13(e). This plot tends not to display useful information for simple examples.
Figure 6.13: Plasmon-Plasmon resonator system - Optimized PR intensity distribution, and optimization parameter population evolution. 
(a) - Optimized PR intensity distribution. (b) - SiO$_2$ population evolution, optimum value = 38 nm. (c) - Upper Mg layer population evolution, optimum value = 46 nm. (d) - Lower Mg layer population evolution, optimum value = 7 nm. (e) - Traditional GA optimization process. (f) - GA optimization process population distribution.
like the plasmon and dielectric overlayer models as they typically converge too rapidly producing, for instance, one large step in the best fitness (penalty) followed by a series of barely noticeable improvements. As such this plot was neglected to be shown for the earlier model optimizations. Although this plot is suitable for tracking the progress of the optimization process of more complex models (such as those to follow) it gives no details of the coverage of the parameter space, thus this plot we not be further shown, instead a GA population distribution plot is used.

The GA population distribution plot for the optimization of this system is shown in Fig. 6.13(f). Note the well distributed population and the shifting optima indicated by generation number (within the plot space) as the GA progresses. The $x$ and $y$ axis of this plot are pseudo-parameters constructed to allow the representation of greater than two parameters in a 2D plot. The pseudo-space is constructed by taking the three parameter values and combining them into a single number i.e parameter values 10, 20, 30 concatenate to give 010020030, where the extra padding zeros are to accommodate the respective range of each of the parameters. This allows each population member to be represented by a unique location for that particular parameter combination within the pseudo-parameter space. Although this makes the location within the plot (Fig. 6.13(f)) less intuitive, it aids in confirming that the GA has adequately covered the parameter space. Even so, trends are still clearly visible with the central band being preferential for this particular optimization.

This particularly model was used as a benchmark system to verify the ability of the GA to find the global optimum, and to get an idea of the computational gains produced by using a GA. To this end a full parameter space sweep was carried out to find the global optimum; the GA was then run several times to verify that it found the same optimum, as was indeed the case. The full parameter space sweep took 6hr 16m 4s and included 1600 individuals; in comparison the GA optimization process took 26m 14s and included 240 individuals to find the same global optimum. Considering these times it is obvious that the use of GA allows for a massive reduction in computation time.

6.6.4 Dielectric-dielectric dual resonator systems

A dual top and bottom dielectric resonator system designed to produce interference at an NA of 1.8 was also constructed consisting of an SiO$_2$ cover, a Cr grating, SiO$_2$ filter/coupler, a HfO$_2$ top high-$n$ resonator, PR, a HfO$_2$ top high-$n$ resonator, and an SiO$_2$ substrate, schematic for which is given in Fig. 6.14(a). The optimized details
for this system are given in Table 6.6, a notable difference being the SiO$_2$ substrate employed here to show a little bit of the flexibility of this system.

The optimized model for this system (Fig. 6.14(b)) is similar to the previous dielectric top resonator example except for the presence of waveguiding in the bottom HfO$_2$ layer. It can also be seen here that the lower resonator is under powered (compared to the upper resonator) leading to an asymmetry in the PR field distribution. From the $E_x$ component (Fig. 6.14(c)) it is evident that the hot spots in $E_x$ are again causing the ‘arching’ of the resonator fields. The $E_y$ field distribution (Fig. 6.14(d)) is dominated by the presence of the waveguiding within the high-$n$ HfO$_2$ layers without the arching seen in the normalized E field.

The comparison plots for this optimization (Fig. 6.15(a)) show a very good agreement between the optimized fields and the ideal fields. The weakened minima along the top are directly over the locations of the maxima in the $E_x$ plots. The GA population distribution (Fig. 6.15(b)) for this system covers the parameter space well with a clear band of near optimal solutions present.

The GA parameter population evolution plots (Fig. 6.15) show the bottom dielectric resonator layer to be the most sensitive with rapid location of the optimum in

### Table 6.6: Dual dielectric resonator COMSOL model details. GA optimized parameters highlighted in cyan. Material references: SiO$_2$[121], Cr[168], HfO$_2$[154], and PR[119].

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6.7. REDUCTION OF INTERFERENCE PITCH

Figure 6.14: Dielectric-dielectric resonator system - Optimized COMSOL simulation. (a) - System schematic. (b) - Normalized $E$ field. (c) - $E_x$ component. (d) - $E_y$ component. Note: all simulation plots have been windowed to highlight the fields within the PR.

approximately 5 generations (Fig. 6.15(e)). The upper resonator layers (Figs. 6.15(c) and 6.15(d)) again show a slower convergence; this is likely due to the bottom resonator being optimized to maximum intensity and subsequently the upper resonator being optimized to match it. Now that these base models have been established, we look at extending them to higher NAs.

6.7 Reduction of interference pitch

The aim of this section is to produce extremely high NA interference patterns that would not be possible at all using prism-coupled EIL. There are two strategies for
For grating coupled interference systems the grating pitch, NA, and therefore the interference pitch go hand in hand. For a given $m$, reducing the grating pitch increases the numerical aperture ($NA = \frac{|m| \lambda}{p}$), thus reducing the interference pitch ($P_x = \frac{\lambda}{2NA}$). These relations are plotted in Fig. 6.16(a) for different $|m|$ values. As the grating pitch decreases the NA increases asymptotically, whilst the interference pitch approaches zero. Increasing $|m|$ results in a higher NA contour with a faster increase in the NA.
with decreasing grating pitch. These higher NA contours consequently result in smaller interference pitches. Although these methods appear to offer a method of reducing the interference pitch greatly they have two opposing factors, that of increasing decay rates of evanescent fields with increasing NA, and the very low grating efficiencies of the higher $|m|$ orders.

If we take either of these routes we run into an issue, that is, the NA exceeds the refractive indices of all naturally available materials. As was discussed earlier this limits the use of dielectric resonators to an NA of approximately 2.45 ($n_{\text{diamond}} = 2.4582$, $\lambda = 405$ nm). As the fields in plasmon resonators are bound to an interface and evanescent either side, they are not required to be propagating, as such they offer a way to bypass this NA limit. The plasmon resonance equation shown previously ($NA = \sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}}$) only specifies the NA of optimum resonance; if we plot Fresnel’s equation across NA for a PR|Metal interface (Fig. 6.16(b)) we get an idea of the maximum reflectance (resonance) available for several common plasmonic metals as well as two interesting 405 nm examples (Mg and In). It can be seen that for all these materials other than Ag there is a strong reflectance peak just below an NA of 2, the a rapid drop off in reflectance as the NA increase. Noticeably the Ag reflectance curve doesn’t drop off as fast and has a high NA tail of a significant magnitude. The fact that this is still much greater than unity reflectance implies Ag is capable of resonance and

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3The standard Fresnel’s equation for TM reflectance, $R = \left| \frac{n_1 \cos \theta_2 - n_2 \cos \theta_1}{n_1 \cos \theta_2 + n_2 \cos \theta_1} \right|^2$
consequently field enhancements at extremely high NAs. The aim of this section is to exploit these extremely high NA plasmon resonances to compensate for the increasing evanescent decay rates at high NAs to reduce the interference pitch as much as possible.

6.7. REDUCTION OF INTERFERENCE PITCH

6.7.1 \( m = \pm 1 \) Extreme NA

Interference of the \( m = \pm 1 \) diffraction orders using a very small pitch grating is perhaps the most obvious method of reducing the interference pitch. Use of the this diffraction order is preferred as it has the greatest diffraction efficiency, however it places more stringent resolution criteria on the manufacturing of the grating, particularly if small duty cycles are required.

With the aim of reducing the interference pitch a generic dual top and bottom SPP based system was modelled. The system was designed to be as flexible as possible consisting of a SiO\(_2\) cover, Cr grating, SiO\(_2\) filter layer, Ag top SPP resonator, PR layer, Ag bottom resonator, and a Si substrate. As was previously the case, the SiO\(_2\) filter layer, and both Ag resonator layers are optimization parameters. In addition to this, the grating thickness and duty cycle have been added as optimization parameters to improve the chances of finding a good solution, although the duty cycle range has been restricted to 25 – 75\% to allow reasonably manufacturable gratings. Both the grating pitch and interference pitch follow the relations shown in Fig. 6.16(a). The PR thickness is varied to produce an aspect ratio (height:full pitch) of 1/2. The SPP layers (top and bottom) have been changed from Mg to Ag. Although the Mg has a very high field enhancement at an NA of approximately 2, it is not suitable for higher NAs as the field enhancement drops off rapidly. Instead Ag is employed here as it displays strong field enhancements, even for very high NAs (Fig. 6.16(b)).

GA models were constructed for sequentially higher NAs (\( NA = 2.5, 5, 7.5, 10 \)), the PR field plots and transects of which can be seen in Fig. 6.17 with the optimized model details given in Table 6.7. In the PR field plots it can be seen that, with the use of GA optimization, design of a system displaying interference and dual resonator resonance is possible up to very high NAs, exceeding those available through prism coupling by more than a factor of 4. However, as the NA increases the ‘cleanness’ of the PR interference pattern gets progressively worse (Figs. 6.17(a), 6.17(c), 6.17(e) and 6.17(g)). It is also notable that the interference patterns diverge at the bottom the PR as the NA increases. Similar patterns are visible in the PR transect plots (Figs. 6.17(b), 6.17(d), 6.17(f) and 6.17(h)). Notable in these plots is the roughness of the PR transect at the top of the PR due to the influence of the hot spots at the corners of the grating,
Figure 6.17: $m=\pm 1$ - extreme NA models and PR transects. Figs. 6.17(a) and 6.17(b) - NA=2.5. Figs. 6.17(c) and 6.17(d) - NA=5. Figs. 6.17(e) and 6.17(f) - NA=7.5. Figs. 6.17(g) and 6.17(h) - NA=10.
and the mesh size as the NA increases. This effect increases as the filter and top SPP resonator layer thicknesses decrease. The reduction of the influence of these fields is theoretically achieved by using a multilayered structure beneath the grating to remove these fields while selectively transmitting the desired diffracted fields [102, 189].

In terms of patterning, the minimum required contrast \( V = \frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{max}} + I_{\text{min}}} \) value is generally accepted as 0.2 [203]. Looking at the transect plots in Fig. 6.17 we see that as expected, the contrast decreases as the NA increases. Contrast values for each of the transect lines are given in Table 6.8 noting that the contrast given is the minimum contrast for each transect. The \( NA = 2.5, 5, 7.5 \) models are all suitable for patterning within contrasts greater than 0.2. The \( NA = 10 \) model although it has a suitable contrast is not suitable for patterning as the bottom of the interference patterns merge together. Generally speaking the contrast decreases as the as distance increases from the grating. It be seen in Fig. 6.17 that the midline PR fields have a better interference null, whilst the top and bottom PR fields suffer from a broadening of the interference maxima. The effects of this broadening may potentially be mitigated by placing index matched materials either side of a central PR layer thus allowing these low contrast field regions to be in non-patterning layers, thus potentially allowing patterning of \( \lambda/20 \) full pitch interference patterns with this system.

### Table 6.7: \( m=\pm 1 \) - extreme NA model parameters. Material references: \( \text{SiO}_2 [121], \ Cr [168], \ Ag [155], \) and \( \text{PR} [119], \) and \( \text{Si} [122]. \)

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### 6.7. REDUCTION OF INTERFERENCE PITCH

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<td>0.63</td>
<td>0.64</td>
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<td>5</td>
<td>0.71</td>
<td>0.69</td>
<td>0.38</td>
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<tr>
<td>7.5</td>
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<td>0.68</td>
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<tr>
<td>10</td>
<td>0.65</td>
<td>0.43</td>
<td>0.78*</td>
<td>0.62</td>
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</table>

**Table 6.8:** $m=\pm 1$ - extreme NA model contrast values.

The 40.5 nm pitch grating required for producing the NA=10 interference pattern, although very narrow, is achievable with modern 193i photolithography processes. Pattern transfer of similar sized interference pitch and PR thickness has been shown to be capable using a trilayer resist stack, and ion beam and $O_2$ reactive ion etching [204]. Thus these systems appear producible from a manufacturing standpoint.

A (non-physical) issue visible in these plots are interpolation and mesh size errors of the COMSOL data, which is most noticeable as the sharp points seen in the interference minima in the PR comparison plots. The script currently extracts the COMSOL model data and reconstructs the PR fields so that it can compare them against the ideal PR fields. This is unfortunately a necessity as the GA requires this data to assign fitness values. The issue occurs when the triangular COMSOL mesh is converted to a rectangular mesh and the field values interpolated to the mesh points via the MATLAB scatteredInterpolant function are not as smooth as the parent data. Occasionally this interpolation will fail at the edges as well, particularly when the parameter values are small, this produces a horizontal bar along the top and bottom edges with a low field strength. Although this effect skews the GA, provided a ‘strong’ interference pattern is present however this is not an issue as the individuals with a strong interference pattern will have a superior fitness value.

For the sake of exploration two further models were produced with NAs of 15 and 20, which equate to interference pitches of 13.5 nm and 10.125 nm ($\lambda/30$ and $\lambda/40$ full pitch) respectively. These systems consisted of a $SiO_2$ cover, Ag grating, $SiO_2$ coupling/filter layer, Ag top resonator, PR, Ag bottom resonator, and a Si substrate, the details for each optimized system are given in Table 6.9. Interestingly these systems produce a reasonably well defined interference pattern (Figs. 6.18(a) and 6.18(c)). Looking at the PR transects (Figs. 6.18(b) and 6.18(d)) however it is noticeable that the contrast at the bottom of the PR in both cases is very low. The $NA=15$ example has a minimum contrast at the base of the PR of $V=0.21$, while the $NA=20$ example
Table 6.9: $m=\pm 1$ - $NA = 15$ and $20$ model parameters. Material references: $SiO_2$[121], $Cr$[168], $Ag$[155], and $PR$[119], and $Si$[122].

has a minimum contrast at the base of the PR of $V = 0.44$. Both of these patterns are therefore technically patternable; although the $NA = 15$ example is just barely, and the $NA = 20$ example has a fairly rough and irregular pattern. Theoretically these two issues should be able to be resolved. For the $NA = 15$ example it may be possible for the bottom resonator fields to be strengthened. The $NA = 20$ example appears to be resolution limited judging by the ‘spikiness’ of the PR transects; improved simulation protocols should improve this.

At this point we chose to stop trying to increase the NA further. Although COMSOL is able to simulate subnanometer layer thicknesses, the values for the $NA = 20$ optimization are considered extremely challenging to manufacture and further process. Also for such thin layers the refractive index of any manufactured layer will likely differ significantly from the values employed in the model. However, we have clearly shown that simple systems for extreme-NA interference lithography are possible, albeit with challenging requirements for fabricating the grating coupler.
6.7. REDUCTION OF INTERFERENCE PITCH

Figure 6.18: \( m=\pm 1 \) - extreme NA models and PR transects 2. Figs. 6.18(a) and 6.18(b) - NA=15. Figs. 6.18(c) and 6.18(d) - NA=20.

6.7.2 \( |m| > 1 \) Extreme NA

To relax the fabrication tolerances for the grating coupler in extreme-NA EIL, higher diffraction orders (\( |m| > 1 \)) can be employed. The use of higher \( m \) orders can be divided into two different configurations, those with a fixed grating pitch, and those with a fixed interference pitch.

Fixed grating pitch

The use of a fixed grating pitch provides smaller interference pitches with increasing \( m \) values (Fig. 6.16(a)). This method is of potential interest as it would allow different interference pitches to be produced from the same grating, either in the same PR
to produce a pseudo-random grating type structure, or as individual exposures in a reusable mask type setting. The use of a higher $m$ value however has the drawback of lower intensities associated with the lower diffraction efficiency of the higher diffraction orders from a simple planar grating. To explore the usefulness of these higher diffraction orders for interference lithography in a simple filter/resonator system such as ours several models were constructed.

These models consist of a SiO$_2$ cover, a Cr grating, SiO$_2$ coupler/filter, Ag SPP resonators, PR, and a Si substrate, the optimized details of which are given in Table 6.10. The grating pitch is fixed at 200 nm and optimization occurs over the grating thickness, duty cycle, coupler/filter layer, and the SPP layers for each of the desired $|m|=1$, 2, or 3 diffraction order. As the corresponding NAs are greater than 2 the resonator material is Ag to allow field enhancements at such high NAs. Figure 6.19 shows the PR field distributions for the optimized systems with a PR thickness required for an aspect ratio of 1/2. The $|m|=1$ plot of the PR fields (Fig. 6.19(a)) shows a clear interference pattern, with the top resonator slightly over-resonant. The PR transects (Fig. 6.19(b)) for this system show that it clearly suitable for patterning with a minimum contrast of $V \approx 0.53$ at the top of the PR. The $|m|=2$ plot of the PR field (Fig. 6.19(c)) shows an interference pattern strongly impacted by the 0\textsuperscript{th} order

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Table 6.10: Extreme NA model parameters - Fixed grating pitch, $m = 1, 2, 3$. Material references: SiO$_2$[121], Cr[168], Ag[155], PR[119], and Si [122].
transmission between the apertures, although the interference pattern is still clearly visible. The PR transects for this system (Fig. 6.19(d)) further highlight the impact of the 0\(^{th}\) order transmission. In this figure it is evident that the contrast decreases with depth into the PR, with the bottom transect showing no evidence of the interference pattern other than the hot-spot interference maxima beneath the grating corners. The impact of the 0\(^{th}\) order transmission is even more evident for the $|m| = 3$ case. The PR field plots (Fig. 6.19(e)) show strong interference maxima originating from the grating corners; while those beneath the aperture are barely distinguishable and do not visibly reach the bottom of the PR. This distribution is further confirmed in the PR transects (Fig. 6.19(f)) with the bottom PR transect being approximately flat beneath the grating apertures.

The norm $E$ optimized system plots are shown in Fig. 6.20. From these plots it is evident that only the $|m| = 1$ system, which equates to an NA of 2, has a well defined interference pattern (Fig. 6.19(a)). Both the $|m| = 2$, and $|m| = 3$ examples show only the strong interference maxima present beneath the grating corners. As the NA increases for both these two examples ($NA = 4$ and $NA = 6$ respectively), as expected, the optimized layer thicknesses decrease (Figs. 6.19(c) and 6.19(e)).

An interesting trend is visible in the optimized system parameters Table 6.10; that is, that the duty cycle increases with NA. The increasing duty cycle has the effect of ‘tightening’ up the high intensity interference maxima originating from the grating corners. This improves the fitness by reducing the error caused by these high intensity maxima; as they have a greater intensity than those under the apertures they improve the fitness despite the increased 0\(^{th}\) order component.

**Fixed NA (interference pitch)**

Although the general direction of this chapter is towards ever shorter interference pitch lengths, the production of subwavelength scale interference pitches from superwavelength grating pitches is also a desirable outcome as the required mask is simpler to produce. This may be achieved by using a large pitch grating and higher $m$ diffraction order. The use of these higher diffraction orders however requires a filter layer(s) to remove the other diffraction orders. This results in the use of multilayered metal dielectric structures to filter and enhance the desired diffraction order [102, 189]. In this work however, we have not yet configured the GA to operate on such multilayered structures, instead we see what is achievable using only a single coupling/filter layer.

These models consist of a SiO\(_2\) cover, a Cr grating, SiO\(_2\) coupler/filter, Mg SPP
Figure 6.19: Extreme NA - |m| > 1, fixed 200 nm grating pitch, optimized systems. Fig. 6.19(a) - |m| = 1 PR fields comparison. Fig. 6.19(b) - |m| = 1 system PR transects. Fig. 6.19(c) - |m| = 2 PR fields comparison. Fig. 6.19(d) - |m| = 2 system PR transects. Fig. 6.19(e) - |m| = 3 PR fields comparison. Fig. 6.19(f) - |m| = 3 system PR transects.
Figure 6.20: Extreme NA - $|m| > 1$, fixed 200 nm grating pitch, norm $E$ of optimized systems. Fig. 6.20(a) - $|m| = 1$. Fig. 6.20(b) - $|m| = 2$. Fig. 6.20(c) - $|m| = 3$. 
### Table 6.11: Extreme NA model parameters - Fixed NA, \( m = 1, 2, 3 \). Material references: SiO\(_2\)\[^{[121]}\], Cr\[^{[168]}\], Mg\[^{[158]}\], PR\[^{[119]}\], and Si \[^{[122]}\].

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<td>Si</td>
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</table>

As expected optimization of the \(|m| = 1\) system produces a well defined interference pattern (Fig. 6.21(a)). The PR transects for this system (Fig. 6.21(b)) show that the interference is suitable for patterning with a minimum contrast of \( V \approx 0.64 \). Optimization of the \(|m| = 2\) system also produces a well defined interference pattern (Fig. 6.21(c)). The PR transects for this system (Fig. 6.21(d)) also show well defined interference suitable for patterning with a minimum contrast of \( V \approx 0.46 \). The \( x \) axis for the contrast plots has been left at its full value to give a better feel for the relative density of the interference patterns for the different \(|m|\) values. Optimization of the \(|m| = 3\) system produced similarly well defined interference patterns (Fig. 6.21(e)) to the \(|m| = 2\) system. The PR transect for this system (Fig. 6.21(f)) show an interference pattern that is suitable for patterning with a minimum contrast of \( V \approx 0.46 \).
Figure 6.21: Extreme NA - $|m| > 1$, fixed NA = 1.9056, optimized systems. Fig. 6.21(a) - $|m| = 1$ PR fields comparison. Fig. 6.21(b) - $|m| = 1$ system PR transects. Fig. 6.21(c) - $|m| = 2$ PR fields comparison. Fig. 6.21(d) - $|m| = 2$ system PR transects. Fig. 6.21(e) - $|m| = 3$ PR fields comparison. Fig. 6.21(f) - $|m| = 3$ system PR transects.
Figure 6.22: Extreme NA - $|m| > 1$, fixed NA = 1.9056, norm $E$ of optimized systems.
Fig. 6.22(a) - $|m| = 1$. Fig. 6.22(b) - $|m| = 2$. Fig. 6.22(c) - $|m| = 3$. 
Notably in the $|m| = 2$, and $|m| = 3$ systems the interference pattern contrast decreased beneath the grating apertures (Figs. 6.21(c) and 6.21(e)). This is due to the influence of an increasing grating aperture as can be seen in the norm E plots for these systems (Fig. 6.22), the values for which are given in Table 6.11. In all cases an interference pattern is clearly visible within the PR layer. Despite the slightly poor contrast of the $m = 2$ and $m = 3$ interference patterns, the ability to use relatively large period gratings is of great benefit as both the 425 nm and 638 nm pitch gratings can easily be made using conventional IL methods.

6.8 A note on GA optimization of layer materials

The previous sections discussed the likely need for a multilayered filter/resonator system to produce better interference patterns using higher NAs and $m$. One of goals of the GA system was to see if it was possible to achieve similar characteristics to a multilayered structure with a simple filter/resonator system. To this end, a cursory study of the optimal layer materials was carried out.

Initial trials used the GA to optimize $n$, $\kappa$ and $d$ of one or more layers, this however proved to be ineffective with the process not producing a reasonable optimization. There are two reasons for this; firstly, as the resonance relies on the phase thickness, varying both the $n$, $\kappa$, and $d$ results in a body of optimal solutions rather than a single one; and secondly, if $d$ is constrained the difference in parameter optimization speed causes an issue. For example for a given layer of an arbitrary thickness the optical properties of it (depending on its role) may change by a very small amount if $n$ and $\kappa$ are altered; this is particularly bad for metal layers. This low sensitivity has the effect of flattening out the global optimum which slows the optimization process to the point where it becomes infeasible to employ due to the very large time required to run the COMSOL models. The use of fixed parameters for all the other layers can be employed, however this somewhat defeats the purpose of using the GA. An alternative method was thus employed, that of using a database of real material refractive indices$^4$.

Optimization of both the filter/coupler and resonator layers proved to be somewhat anticlimactic as the resonance condition is dependent upon the thickness of the layers.

$^4$If a database of values is to be employed in a GA setting there must be some physical logic to the ordering of the database, else the concept of crossover children becomes meaningless as the nearby neighbours no longer represent close parameter values, thus the search for the optimum in the neighbouring parameter space generally fails. For this optimization the database was organised in order of increasing real component of the refractive index.
and the refractive index combination. Consequently if the optimization process was run with a fixed grating pitch and layer thickness it would discover the material combination required to produce SPP resonance for the $m$ diffraction order, which would be the selfsame materials required to discover the thicknesses in the first place. Optimization of the grating material however proved more fruitful with the GA generally settling on good or plasmonic metals such as Cr, Mg, Ni, Al, Ag, and In. The use of this parameter in the GA however slowed the system optimization process down significantly and often showed little difference in the optimum fitness values between different optimal metals. There was however one observable trend which was at lower NAs ($NA < 2$) Cr, Ni, and Mg produced the best interference patterns, while for NAs greater than 2 Ag produced superior interference patterns. Due to this the models in this chapter use Cr gratings for NAs less than 2, and Ag gratings for NAs greater than 2.

6.9 Summary

In this chapter we have introduced the concept of grating coupled near-field interference lithography, and discussed the literature build up of the concepts involved leading to the current state of the literature. Grating coupled near-field interference lithography uses a diffraction grating to allow coupling of high NA diffraction orders into resonators that are not possible using conventional prism coupling due to the presence of evanescent fields within the prism. The complexity of these systems however requires that the modelling be carried out in a full field fashion, in our case using the COMSOL finite element software. Finite element is a very time intensive optimization process, thus a genetic algorithm was employed to speed up this optimization process by an order of magnitude.

The GA optimization process was employed to construct several test models (plasmon overlayer, dielectric overlayer, plasmon-plasmon, and dielectric-dielectric), and eventually to construct extremely high NA interference systems. NAs as high as 20 were simulated with interference patterns clearly visible. NAs of this size have not previously been described. The pattern fidelity is however not optimal, this can be improved with the use of the buffer layers to remove the low contrast upper and lower PR field regions.

The use of higher diffraction orders for interference was also investigated. Two configurations were tested, firstly a system employing a fixed grating pitch, and secondly a system designed to produce a fixed interference pitch. The fixed grating pitch system
resulted in increasing NAs as the $m$ value increased. The resultant interference pitches, although still visible, got progressively washed out as $m$ increased due to the low grating efficiency of the higher $m$ orders and the presence of the $0^{th}$ order component. Fixing the interference pitch required the grating pitch to increase as $m$ increased to allow the diffracted order to have the correct NA. This produced very good diffraction patterns for the higher $m$ diffraction orders using gratings that could be manufactured using conventional IL ($p = 425$ nm and $p = 638$ nm for $|m| = 2$ and $|m| = 3$ respectively), although the areas under the grating apertures still had relatively poor interference fringe contrast.

Ultimately GAs were shown to be a suitable method for the optimization of grating coupled evanescent near-field IL systems. As in all optimization processes however, to get the best results out one must use informed parameter ranges. When these were considered the primary design goal of increasing the NA to extremely high values was achieved.
Chapter 7

Conclusions and future work

The common theme throughout this research has been the search for methods of enhancing IL by utilizing evanescent fields. Three methods were explored as part of this thesis: Herpin effective media resonant underlayers, evanescent-coupled ARCs, and GA optimization of evanescent near-field IL. In this chapter the contents of this thesis relating to each of these methods will be briefly discussed leading on to potential extensions of this work.

7.1 Herpin effective media resonant underlayers

The highly efficient use of single layer $\lambda/4$ ARCs is often hindered by the lack of a suitable refractive index material. To bypass this Herpin effective medium symmetric trilayers which have the same optical properties as the desired single layer are often employed. These trilayers consist of transparent dielectrics in either a $n_{\text{low}}|n_{\text{high}}|n_{\text{low}}$ or $n_{\text{high}}|n_{\text{low}}|n_{\text{high}}$ arrangement, depending on the thicknesses of the individual layers, the trilayers can be constructed to have an effective refractive index that is not (easily) otherwise available in naturally occurring materials. A well known side effect of this method is the generation of theoretically arbitrarily high effective refractive indices. We sought to exploit these arbitrarily high effective refractive indices for use as extremely high NA resonant underlayers for EIL.

Simulations of Herpin effective media resonant underlayers were produced for ultra high-NA (i.e. evanescent fields within the PR layer) IL systems. The aim was to develop Herpin trilayer replacements for ‘standard’ single layer dielectric resonant underlayers. This was successfully achieved firstly in a configuration where all the Herpin layers
contained propagating fields, and secondly where the $n_{\text{low}}$ layer contained evanescent fields ($NA > n_{\text{low}}$). Extension to combinations where all three Herpin layers were evanescent however resulted in the breakdown of this particular method. The effective refractive index derived from the Herpin effective media is dependent upon the phase thickness of the layers; provided the phase thicknesses are real the effective refractive index will also be real. When all the Herpin layers are evanescent however the phase thickness of all the Herpin layers is complex and the effective refractive index has a large imaginary component, thus the system is not able to perform as a resonator as the losses are too high. This is not unexpected as ultimately Herpin effective media theory must take into account the real physical properties of the layers composing the trilayer, thus if they are all evanescent layers one would expect the full system to also be evanescent.

Although the use of Herpin effective media was shown to be capable of acting as a resonant underlayer, the requirement for a symmetric trilayer in many respects artificially constrains the search for effective underlayer solutions. As such, studies were carried out investigating refractive index space to see if other possible resonator solutions are possible. In the course of this search, not only were the standard surface state polariton and dielectric resonator solutions identified, two other configurations were discovered, that of evanescent-coupled ARCs (discussed in the following section), and resonant overlayer systems.

Resonant overlayer systems employ a resonator above the PR in the film stack to pre-enhance the magnitude of the fields before they enter the PR for ultra high-NA IL systems. For the ultra high-NA regime an IML is employed to remove the air-gap between the prism and the PR. Above an NA of approximately 1.77 however, IMLs become strongly absorbing, thus requiring very thin layers (achieved by very high pressures) to couple an appreciable amount of energy into the PR. The use of a resonant overlayer offers a remedy to relax the IML thickness requirement by resonantly enhancing the fields entering the PR. This allows the use of relatively thick and/or low index IMLs such as water to be employed instead. The use of low index IMLs is a peripheral but important fact as high refractive index IML often contain toxic materials, many of which are being phased out, thus it is beneficial from an experimental and manufacturing standpoint to be able to use safe non-toxic IMLs.

In this body of work experimental verification of resonant overlayers was not attempted, hence the logical next step for this technique would be to experimentally verify this effect. For processing reasons the resonant overlayer needs to be able to be
removed before development can be performed. Consequently a ‘release’ layer such as a very thin PVA layer may be required between the overlayer and the PR. For this reason a process compatible stack will need to be developed such that the IML will not attack the release layer or the PR. Although similarly, the stack edges could be sealed with a lacquer which may then be removed after exposure before development proceeds. The use of multilayered overlayer structures has been experimentally applied for superlens and grating coupled near-field IL systems (Chapter 6), thus this experiment should be achievable. Verification of the effectiveness of this technique is possible by comparing the patterning dose for systems with and without a resonant overlayer. The ability to produce comparable IML thicknesses however may be a challenge. In the absence of an accurate gapping (IML thickness) measurement, IML surface tension may be able to be exploited if the samples are adhered using surface tension. Provided the samples are the same size and the IML procedures are the same the surface tension and thus the IML thickness should be approximately the same. This concept needs to be further explored, although the experimental developments will represent a significant effort (and hence were out of the scope of this thesis).

7.2 Evanescent-coupled ARCs

The search for alternative resonant underlayer systems in the Herpin effective media work led to the application of resonant underlayers in the high-NA (propagating fields within the PR) regime as evanescent-coupled ARCs. Both surface state polariton and dielectric resonator underlayers are suitable for use as evanescent-coupled ARC. Development and characterization of these ARCs at a wavelength of 405 nm using the transfer matrix method revealed that they are suitable for high-NA, narrow NA range, TM or TE polarized exposures. Evanescent-coupled ARC designs were successfully extended to a wavelength of 193 nm indicated that this form of ARC can potentially be employed in modern semiconductor 193i photolithography methods.

For experimental verification of evanescent-coupled ARCs two system were manufactured, a MgF$_2$|Cr based SSR ARC, and a SiO$_2$|HfO$_2$ based DR ARC. Verification of AR effects required the development of a thick (∼ 1300 nm) PR film cleave-then-develop process. This allowed structures with aspect ratios as high as 10:1 to be achieved which allowed multiple standing wave pitches to be observed. With this method the MgF$_2$|Cr ARC resonator was shown to be resonating, albeit in a sub-optimal fashion, as evidenced by the presence of hot spots in the PR footing. Full standing
wave suppression however was not achieved with this particular set of manufactured coatings. The SiO$_2$|HfO$_2$ ARC system was shown to be a suitable ARC with full standing wave suppression demonstrated.

The experimental component of this section revealed several design/experimental issues that will ideally be improved upon for the next iteration of these experiments. The largest problem lay in refractive index and thickness errors in the manufactured coatings. Both SiO$_2$ and HfO$_2$ are fairly well behaved materials for coating as was born out in the experiments. MgF$_2$ and Cr however are relatively difficult materials to accurately produce a desired refractive index, and consequently optical thickness. MgF$_2$ is known to be porous and the thinness of the Cr layer likely led to the formation of islands and agglomerations which can massively alter the imaginary component of the refractive index (and by extension the real component). Ellipsometric measurements and ARC experimental results suggest that both the MgF$_2$ and Cr layers have refractive indices and thicknesses different enough from the design values, which prevented the MgF$_2$|Cr ARC from achieving full standing wave suppression. Accordingly, a second iteration of these experiments would most greatly benefit from a tighter integration of the ARC manufacturing and ARC design procedures to allow the correct layer thicknesses to be employed for the particular manufactured material refractive indices. Further to this, it was discovered that the unbleached PR refractive index used in the ARC design models was incorrect based on the experimental results and that a value closer to the bleached PR refractive index was more accurate. To accommodate this the thicknesses of the ARC layers should be altered slightly ($\sim 2$ nm). Finally the MgF$_2$|Cr ARC was designed to operate with TM polarization at an NA of 1.4046. This polarization and NA unfortunately corresponds to a poor fringe visibility of 0.406 which reduces the experimental process window making experimental verification more difficult. For this reason it would be beneficial to attempt to verify SSR based ARCs with a different resonant combination with a better fringe visibility.

7.3 GA optimization of grating coupled near-field IL systems

For prism coupled IL systems the maximum possible NA is limited by the coupling prisms ability to sustain propagating fields, consequently the maximum NA for prism coupled IL systems at a wavelength of 405 nm is approximately 2.5. To circumvent this limit diffraction gratings can be employed. The crucial aspect here is that with
normal incidence upon the grating NAs greater than 2.5 are produced as evanescent
diffraction orders. These diffraction orders can then be resonantly enhanced by SPP
or dielectric resonators and interfered to produce extremely small interference pitches.

The use of evanescent diffraction orders however requires the thin resonator layers
within close proximity to the grating. As a consequence of this there are no closed
form analytical solutions for such a system, and finite element method simulations are
needed. Finite element simulations are extremely computationally intensive, thus to
improve the optimization time a GA optimization process was successfully employed.
The application of a GA allowed for very large improvements in optimization times, for
instance in one case a full parameter sweep of 1600 individuals was carried out taking 6
hr 16 mins 4 s, in comparison applying the GA to the same model and parameter space
discovered the same global optimum in 26 min 14 s by simulating only 240 individuals.

Top, and dual top and bottom resonator test bed systems were constructed to
validate the use of the GA and its ability to find suitable optimization solutions. Once
this was established the systems were then used to explore the possibilities of increasing
the NA as much as possible. To this end extreme NA examples were generated using the
$|m| = 1$ diffraction orders and Ag SPP resonators to NAs up to 15, which is much higher
than anything previously reported in the literature. The use of higher $m$ diffraction
orders was also investigated and shown to be capable of producing interference patterns,
although the suitability of these interference patterns is limited by the presence of the
lower diffraction orders.

Throughout the GA optimization chapter it was noted that the interference pat-
tern fidelity was negatively impacted by the proximity of the PR to the grating. To
improve this situation multilayered resonator/filter (superlens) type structures would
theoretically provide improved interference patterns by enhancing the desired $k_x$ values
while dissipating the undesired values. Work is currently ongoing to modify the GA
towards discovering multilayer resonator/filter type solutions.

All the simulations in the GA optimization chapter were at an illumination wa-
velength of 405 nm. To both reduce the interference pitch and to employ modern
photolithography methods, it is also worth considering extending the GA optimization
and models to an illumination wavelength of 193 nm; Fig. 7.1 shows an example of this.
The use of 193 nm illumination requires the use of Al SPP resonator layers. The $|m|$
diffraction orders are being interfered, this coupled with the 74.4 nm grating results
in an interference pitch of 37.2 nm. The minimum contrast occurs at the base of the
PR with a value of $V \approx 0.4$, thus this system produces an interference pattern suitable
Figure 7.1: 193i example. (a) - Schematic. Note the use of Al as the SPP resonator material. (b) - PR field comparison.

for patterning. The development system shows that GA optimization of evanescent near-field IL is possible at 193 nm. Further development is required to see if the gains provided from using this method at $\lambda = 405$ nm are available at $\lambda = 193$ nm.
Appendix A

Experimental Methods, materials, and notes

Although a great deal of experiments were carried out in the process of completing this research, many of the details are consider extraneous to the main body of this text. For the this reason and ease of reading, many of the experimental details occur here rather than in the main body.

A.1 Lloyd’s mirror interference lithography

hardware and materials list

Lasers

Primary - Integrated Optics - 405 NM SLM MatchBox 2 (VBG Diode, PM Fibre) 15 mW CW laser.

Notes: Very nice clean beam shape out of the fibre head. A rotational mount for the output ferrule was used to align the axis of polarization of the output beam. The quoted 15 mW output is likely that of the diode output as the power output from the fibre is typically less than half this value.


Notes: Required either a pinhole (Thorlabs - KT310/M) or SM/PM fibre (Tholabs - P1-405BPM-FC-2) spatial filter. Fibre spatial filter had a superior beam compared to the pinhole spatial filter but with a lower power output. Fibre spatial filter however
displayed ‘breathing’, where the output power cycled at regular intervals.

Collimating lens

Primary - Thorlabs - LA1951-A - N-BK7 Plano-Convex Lens, 1” diameter, f = 25.4 mm, AR Coating: 350-700 nm

Electronic timing shutter

Primary - Thorlabs - SH05 - Optical Beam Shutter with 10’ Long Cable, 1/2” Aperture, 8-32 Taps. Thorlabs - SC10 - Optical Beam Shutter Controller.

Half-wave plate

Primary - Thorlabs - WPMH10M-405, 1” diameter, Multi-Order Half-Wave Plate, SM1-Threaded Mount, 405 nm

Prism mount

Primary - Thorlabs - K6XS 6-Axis Locking Kinematic Optic Mount.
Notes: This mount was mounted in a horizontal fashion with a custom made threaded prism holder which screwed into the mount aperture. This mount allowed for both rotation and levelling of the prism. Further to this, the entire prism mount was mounted on an x-translation stage to allow the mount to be shifted in the direction perpendicular to the incident laser beam.

Prism

Primary - glass-sphere.com - Soda-lime glass prism 30x30x30 mm. A mirror coating of 120 nm was applied to one of the prism faces.

A.2 Spin coating recipes

PR


<table>
<thead>
<tr>
<th>Spin recipe</th>
<th>Ramp time</th>
<th>Main spin</th>
<th>Main time</th>
<th>Thickness</th>
<th>Soft bake</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>5 s</td>
<td>2250 rpm</td>
<td>60 s</td>
<td>~1300 nm</td>
<td>60 s at 110°C</td>
</tr>
</tbody>
</table>
Notes: This recipe uses undiluted photoresist.

**PVA**

*Material* - Generic - Poly-vinyl alcohol (PVA)

<table>
<thead>
<tr>
<th>Spin recipe -</th>
<th>Ramp time</th>
<th>Main spin</th>
<th>Main time</th>
<th>Thickness</th>
<th>Soft bake</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>10 s</td>
<td>4000 rpm</td>
<td>60 s</td>
<td>~20 nm</td>
<td>N/A</td>
</tr>
</tbody>
</table>

Notes: PVA: Water solution, PVA 11.4 mg/mL.

**HMDS**

*Material* - Generic - Hexamethyldisilazane (HMDS)

<table>
<thead>
<tr>
<th>Spin recipe -</th>
<th>Ramp time</th>
<th>Main spin</th>
<th>Main time</th>
<th>Thickness</th>
<th>Soft bake</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>10 s</td>
<td>4000 rpm</td>
<td>60 s</td>
<td>monolayer*</td>
<td>N/A</td>
</tr>
</tbody>
</table>

Notes: HMDS is used as an adhesion promoter for PR on SiO$_2$. Ideally vapour priming should be used for application. However, in the absence of a vapour priming system, we have developed a recipe for spin coating of HMDS. For adhesion purposes the HMDS is allowed to ‘sit’ for 1 minute on the bare Si before PR is applied. The HMDS reacts with the Si wafer thus only the chemically attached HMDS should remain after the spinning process and an approximate monolayer should result. After HMDS spinning PR is applied and left to sit for approximately 5 seconds before PR spin coating to allow the PR to adhere to the HMDS. This method produced good quality PR films without any major visible defects before or after PR exposure and development.

**A.3 Other photolithography processes**

**Si wafer OH-bake**

The Si samples were all baked at 200°C to drive off the OH groups from the Si surface. This improves the adhesion of the PR to the Si.
Development

The developer used for these experiments was Tetramethylammonium hydroxide (TMAH) in water (2.38%). The development process employed this developer diluted 2:1 (TMAH:Water) for 60 seconds in a puddle, followed by a water rinse.

Initially AZ electronic materials AZ 726 MIF TMAH developer was used. Later we switched to Sigma-Aldrich TMAH 25% aqueous solution 331635-1L. Once diluted down this proved to be a considerably cheaper alternative with similar development properties. If the reader wishes to use this particular developer I would caution that the TMAH must be diluted using deionized water else the strength of the developer will be dulled.
Appendix B

Example MATLAB ARC scripts

The use of this script requires the use of a birefringent thin films toolbox for Matlab. This toolbox is available with the book *Birefringent thin films and polarizing elements* 2nd edition, by McCall, Hodgkinson, and Wu [113].

**Listing B.1:** Example Matlab ARC script for a MgF\(_2\)/Cr SSR based evanescent-coupled ARC.

```matlab
% this is the full matrix equation one, zero finesse

clear all; close all; clc

w=405; k=2*pi/w;

AZMiR701=1.6844 + 0.0307i; % unbleached PR refractive index
AZMiR701_bleached=1.6751 + 0.0031i; % bleached PR
MgF2=1.38;
Cr=2.0356 + 2.8804i;
Si_c=5.4375 + 0.3420i; % Crystalline Silicon

nC=AZMiR701;
nC=AZMiR701_bleached;
PR=nC; PR_thickness=2000;
n1=MgF2;
n2=Cr;
nS=Si_c;

beta=real(sqrt((n1^2+n2^2)/(n1^2+n2^2)));
alphaC=sqrt(nC^2-beta^2);
alpha1=sqrt(n1^2-beta^2);
alpha2=sqrt(n2^2-beta^2);
alphaS=sqrt(nS^2-beta^2);

alpha=[alphaC alpha1 alpha2 alphaS];
gCp=nC^2/(z0+alphaC);
g1p=n1^2/(z0+alpha1);
g2p=n2^2/(z0+alpha2);
gSp=nS^2/(z0+alphaS);
```
gp=[gCp g1p g2p gSp];
dx1m=0;dx1M=201;divx1=201;
dx2m=0;dx2M=201;divx2=201;
for id1=1:divx1
dx1(id1)=dx1m+(dx1M−dx1m)*(id1−1)/(divx1−1);
x1=dx1(id1);
for id2=1:divx2
dx2(id2)=dx2m+(dx2M−dx2m)*(id2−1)/(divx2−1);
x2=dx2(id2);
d1=k∗x1∗alpha1;
d2=k∗x2∗alpha2;

%%% Characteristic Matrix Components %%%
M11=cos(d1)∗cos(d2)−g2p/g1p∗sin(d1)∗sin(d2);
M12=−i/g2p∗cos(d1)∗sin(d2)−i/g1p∗sin(d1)∗cos(d2);
M21=−i∗g1p∗sin(d1)∗cos(d2)−i∗g2p∗cos(d1)∗sin(d2);
M22=cos(d1)∗cos(d2)−g1p/g2p∗sin(d1)∗sin(d2);
M1s(id1,id2)=M11;
M12s(id1,id2)=M12;
M21s(id1,id2)=M21;
M22s(id1,id2)=M22;
Ms=[M11 M12 M21 M22];

rp=(gCp∗M11+gCp∗gSp∗M12−M21−gSp∗M22)/(gCp∗M11+gCp∗gSp∗M12+M21+gSp∗M22);
rtop(id1,id2)=(gCp∗M11+gCp∗gSp∗M12−M21−gSp∗M22);

end

end

r_top_R=real(r_top);
r_top_I=imag(r_top);
r_top_abs=abs(r_top);

figure
contourf(dx1,dx2,Rp,'','linestyle','none')
xlabel('Evanescent Layer Thickness (nm)')
ylabel('High−\kappa Thickness (nm)')
% title('Reflectance')
contourcbar
colormap(hot)
[RA,RB]=find(Rp==min(min(Rp)));
hold on
%adding contour
axis([min(min(Rp)) max(max(Rp))]);
% contour(dx1,dx2,Rp,'',[0.005,0.005],','linewidth',2);
contour(dx1,dx2,Rp,'',[0.01,0.01],','linewidth',2);
contour(dx1,dx2,r_top_R,'',[0,0],',b',',linewidth',2);
contour(dx1,dx2,r_top_I,'',[0,0],',g',',linewidth',2);
%adding text
plot(dx1(RA),dx2(RB),'x','Color','w')
text(dx1(RA)-35,dx2(RB)+divx1/16,['R=',num2str(min(min(Rp)))],'Color','w','fontsize',12);
dc=20;
line([dx1(RA)-dc,dx1(RA)+dc],[dx2(RB),dx2(RB)],'Color','y','LineStyle',':','LineWidth',1.5);
hold off

nC=1.5366;
NA=beta;
t=0:pi/2; ct=cos(t); st=sin(t); %ct = TM, st = TE i.e. for TM ct =1 st = 0
N=1000; % number of divisions in tracesystem, i.e. look at je
hp=w/(4*NA);
% xpr=1.5*hp;
%
nL1=n1;
nA=n2;

% Ideal %
dL1=dx1(RA);
dH=dx2(RB);

system=[nC nC nC 0 0 0 Inf % cover
nC nC nC 0 0 0 100/w % prism
nPR nPR nPR 0 0 0 PR_thickness/w
nL1 nL1 nL1 0 0 0 dL1/w % low index layer required for TIR
nA nA nA 0 0 0 dH/w % Monolayer this is a place holder, thus zero thickness set.
nS nS nS 0 0 0 100/w % Si
nS nS nS 0 0 0 Inf]; % substrate

% Design System %

beta=NA;

[ex1,ey1]=tracesystem(system,beta,w,N,ct,st);
[ex2,ey2]=tracesystem(system,-beta,w,N,ct,st);
ex=ex1+ex2;ey=ey1+ey2;
Ip=abs(ex).*abs(ey)+abs(ey).*abs(ex).abs(ex);
Ip=Ip/abs(Ip(1:1:je(2))); % normalize against max cover field

dL_range=linspace(dx1(RA)-dc,dx1(RA)+dc,11); % must be odd to include value
[~,sss]=find(dL_range==dx1(RA));
dL_low=dL_range(1:sss-1);
dL_high=dL_range(sss+1:end);
for i1=1:length(dL_range)
system(4, 7) = dL_range(i1)/w;
[xp np je eh] = tracesystem(system, beta, w, N, ct, st);
ex1 = eh(:, 1); ey1 = eh(:, 2); ez1 = eh(:, 3);
ex2 = eh(:, 1); ey2 = eh(:, 2); ez2 = eh(:, 3);
ex = ex1 + ex2; ey = ey1 + ey2; ez = ez1 + ez2;
lp = abs(ex) .* abs(ex) + abs(ey) .* abs(ey) + abs(ez) .* abs(ez);
lp_range(:, i1) = lp / max([lp(je(1): je(2))]); % normalize against max cover field
% lp_range(:, i1) = lp; % normalize against max cover field
if i1 == sss;
je = je;
end
figure
hold on
for i2 = 1:length(dL_range)
if i2 < sss
% plot(xp, lp_range(:, i2), 'Color', [(length(dL_low) - i2)/length(dL_low)], 0, 0', 'LineWidth', 1.5)
plot(xp, lp_range(:, i2), 'r')
elseif i2 == sss
plot(xp, lp_range(:, i2), 'k', 'LineWidth', 2);
elseif i2 > sss
plot(xp, lp_range(:, i2), 'b')
end
end
hold off
set(gca, 'layer', 'top')
for i4 = 2:length(je)
line([xp(jes(i4)), xp(jes(i4))], [0, max(max(lp_range))], 'Color', 'k')
end
xlabel('Distance (nm)', 'FontSize', 12)
ylabel('Normalized intensity', 'FontSize', 12)
axis tight

% % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % % %
Appendix C

Example GA MATLAB and COMSOL scripts

This code was run using the following software COMSOL version 5.0, LiveLink for COMSOL version 5.0, MATLAB version 8.4.0.150421 (R2014b), Windows 10.

Listing C.1: Matlab parent script to run the GA optimization process.

```matlab
% this GA includes a lot of improvements in the matlab side of things

clear all; close all; clc;

global g_count oG

g_count=-1; % counter to sort out my ga extra generation problem

tic

SiO2=1.4696; % refractive index
Cr=2.0356 + 2.8804i; 
Mg=0.1794 + 3.5695i;
AZMiR701=1.6844 + 0.0307i; % photoresist
Si_c=5.4375 + 0.3420i; % crystalline silicon

w=405; % wavelength
m=1; % interfering diffraction orders
gr=1;
nPR=AZMiR701; % Photoresist

system_type=['plasmon']; % options = 'fixed pitch', 'plasmon', 'dielectric', 'no resonator'...

...Second cell pitch size for fixed pitch i.e. 200, dielectric NA

dC_index=SiO2; % cover material
dG_index=Cr; % grating material
d2_index=SiO2; % filter(ish) layer
d3_index=Mg; % high-n dielectric layer
dPR_index=nPR; % photoresist
```

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d5_index=Mg; % substrate material
d5_index=Si,c; % substrate material
materials=[dC_index dO_index d2_index d3_index dPR_index d5_index dS_index];
sp={"d2","d3","d5"};
    "SiO_2","Mg","Mg");
d2m=1; d2inc=1; d2M=100; % filter layer thickness values
d3m=1; d3inc=1; d3M=100; % plasmon layer thickness values
d5m=1; d5inc=1; d5M=100; % plasmon layer thickness values
mincM=[d2m,d3m,d5m];
d2inc,d3inc,d5inc;
d2M,d3M,d5M]; % component mins, increments and maxs
LB = ones(1, size(mincM,2)); % Lower bounds
for i1 = 1:size(mincM,2)
    UB(1,i1)=length(mincM(1,i1):mincM(2,i1):mincM(3,i1));
end
fpopdist=@(options,state,flag)gapopdist(options,state,flag,mincM); % anonymous
function for gapopdist, make sure you use the correct gapopdist 1,2,etc.
optsgaoptimset(...
    'PopulationSize',3,... % 25
    'Generations',1,... % 10
    'EliteCount',1,...
    'CrossoverFraction',0.8,... % default 0.8
    'CrossoverFcn',@crossoversinglepoint,... % GA options, crossover options. This
    should work but it doesn’t, maybe it’s the matlab version
    'TolFun',1e-8,...
    'FitnessLimit',1*10^-2,... % if it ends early for no reason this probably needs to be
    reduced.
    'Display','iter',...
    'Vectorize','on',...
    'PlotFcns',fpopdist,...
    'OutputFcns',fpop); % GA options
oG=opts.Generations; % to count generations.
pops=zeros(ops.PopulationSize,oG,length(LB)); % preloading d3
rng('shuffle'); % prevent it starting at the same point upon restart of matlab
f=@(x)fitness_20160127(x,mincM,w,m,gr,nPR,materials,system_type,sp);
[x, fval, exitflag] = ga(f,...
    length(LB),[],[],[],[],[],UB,[],1:size(mincM,2),opts);
Parameter_values=mapvariables.vector_ga(x,mincM);
display(Parameter_values);
display(fval);
display(exitflag);

population_evolution(pops,mincM,sp)
toc

load chirp
sound(y,Fs)

---

**Listing C.2:** Matlab function to run COMSOL and perform the fitness evaluation.

```matlab
function [sse1] = fitness_20160127(x,mincM,w,m,gr,nPR,materials,system_type,sp)
    g_count = g_count + 1;
    numbers=mapvariables_vector_ga(x,mincM); % population values

    % Parameter Sweep Values
    d2n=numbers(:,1);
d3n=numbers(:,2);
d5n=numbers(:,3);
sq=char(39); % single quotation mark
c=char(44); % comma

    if g_count<cG
        t2 = []; t3 = []; t5 = [];
        for i1=1:size(numbers,1) % sorting population into a COMSOL suitable format
            if i1==0
                d2s=strcat(num2str(d2n(i1)),c);
d3s=strcat(num2str(d3n(i1)),c);
d5s=strcat(num2str(d5n(i1)),c);
            elseif i1==size(numbers,1)
                d2s=strcat(num2str(d2n(i1)))
d3s=strcat(num2str(d3n(i1)))
d5s=strcat(num2str(d5n(i1)))
            else
                d2s=strcat(num2str(d2n(i1)),c);
d3s=strcat(num2str(d3n(i1)),c);
d5s=strcat(num2str(d5n(i1)),c);
            end
            t2=strcat(t2,d2s);
t3=strcat(t3,d3s);
t5=strcat(t5,d5s);
    end

    fp=strcat(t2,sq,{' '},sq,t3,sq,{' '},sq,t5); % full parameters
    else % this is a fudge to allow the final GA values to run the same COMSOL code
        t2=strcat(num2str(d2n),c,num2str(mincM(3,1)));
t3=strcat(num2str(d3n),c,num2str(mincM(3,2)));
t5=strcat(num2str(d5n),c,num2str(mincM(3,3)));
        fp=strcat(num2str(d2n),c,num2str(mincM(3,1)),sq,{' '},...
            sq,num2str(d3n),c,num2str(mincM(3,2)),sq,{' '},...
            sq,num2str(d5n),c,num2str(mincM(3,3)))
    end
```

---
if oG==0
    tic
end
import com.comsol.model.*
import com.comsol.model.util.*
model = ModelUtil.create('Model');
model.modelNode.create('comp1');
model.geom.create('geom1', 2);
model.mesh.create('mesh1', 'geom1');
model.physics.create('emw', 'ElectromagneticWaves', 'geom1');
model.study.create('std1');
model.study('std1').create('freq', 'Frequency');
model.study('std1').feature('freq').activate('emw', true);
model.param.set('lambda', '405[nm]', 'wavelength');
model.param.set('f0', 'c/const/lambda', 'frequency');
model.param.set('NA', '0', 'numerical aperture');
alpha_c = [asin(NA/num2str(materials(1)))];
model.param.set('dC_index', num2str(materials(1)), 'cover');
model.param.set('dG_index', num2str(materials(2)), 'grating');
model.param.set('d2_index', num2str(materials(3)), 'dielectric filter layer');
model.param.set('d3_index', num2str(materials(4)), 'plasmon layer');
model.param.set('dPR_index', num2str(materials(5)), 'photoresist');
model.param.set('d5_index', num2str(materials(6)), 'substrate');
model.param.set('dS_index', num2str(materials(7)), 'substrate');
alpha_c = [asin(NA/num2str(materials(1)))];
model.param.set('alpha', alpha_c, 'angle of incidence');
model.param.set('d1', '200[nm]', 'Cover');
model.param.set('d2', '50[nm]', 'grating');
model.param.set('d3', '50[nm]', 'filter layer');
model.param.set('d4', '50[nm]', 'plasmon layer');
model.param.set('d5', '100[nm]', 'PR');
model.param.set('d6', '50[nm]', 'plasmon layer');
model.param.set('d7', '200[nm]', 'Substrate');
model.param.set('m', num2str(m), 'diffraction order');
% Params for mphplot lines :
line_d1 = model.param().evaluate('d1');
line_d2 = model.param().evaluate('d2');
line_d3 = model.param().evaluate('d3');
line_d4 = model.param().evaluate('d4');
line_d5 = model.param().evaluate('d5');
line_d6 = model.param().evaluate('d6');
line_d7 = model.param().evaluate('d7');
if strcmpi(system_type{1}, 'plasmon')
  % Producing right grating pitch for plasmons
  e_m=(materials(4))ˆ2; % metal dielectric constant
  e_pr=(materials(5))ˆ2; % PR dielectric constant
  kx2=2*pi/(w*10^-9)*sqrt((e_m+e_pr)/(e_m*e_pr));
  model.param.set('kx', num2str(real(kx2)), 'beta'); % k number in the x direction
else
  elseif strcmpi(system_type{1}, 'fixed pitch')
    model.param.set('Px', [num2str(system_type{2}), '[nm]', 'Grating Pitch']);
    elseif strcmpi(system_type{1}, 'dielectric')
      kx=2*pi/w*system_type{2};
      Px=round(m*2*pi/kx);
    model.param.set('kx', [num2str(2*pi/kx)], 'beta'); % k number in the x direction. Note: beta not NA in this case:
    else
      error('No system type defined')
end

Pxv=model.param().evaluate('Px');
model.param.set('w', '3*Px', 'Width');
model.param.set('dc', '0.5', 'duty cycle');
model.param.set('gs', 'Px(1-dc)', 'grating sold');
model.param.set('gg', 'Px(dc)', 'grating gap');
line_w=model.param().evaluate('w');
line_dc=model.param().evaluate('dc');
line_gs=model.param().evaluate('gs');
line_gg=model.param().evaluate('gg');
model.variable.create('var1');
model.variable('var1').model('compl');
model.geom('geom1').run;
model.variable('var1').set('k1', 'emw.k0');
model.variable('var1').set('k1x', 'k1*sin(alpha)');
model.variable('var1').set('kly', 'k1*cos(alpha)');
model.geom('geom1').create('r1', 'Rectangle');
model.geom('geom1').feature('r1').set('size', {'w', '2*d1+d2+d3+d4+d5+d6+2*d7'});
model.geom('geom1').feature('r1').setIndex('layer', 'd7', 0);
model.geom('geom1').feature('r1').setIndex('layer', 'd7', 1);
model.geom('geom1').feature('r1').setIndex('layer', 'd6', 2);
model.geom('geom1').feature('r1').setIndex('layer', 'd5', 3);
model.geom('geom1').feature('r1').setIndex('layer', 'd4', 4);
model.geom('geom1').feature('r1').setIndex('layer', 'd3', 5);
model.geom('geom1').feature('r1').setIndex('layer', 'd2', 6);
model.geom('geom1').feature('r1').setIndex('layer', 'd1', 7);
model.geom('geom1').runPre('fin');
model.geom('geom1').run('r1');
model.geom('geom1').create('r2', 'Rectangle');
model.geom('geom1').feature('r2').label('Grating');
model.geom('geom1').feature('r2').set('size', {'w': 'd2'});
model.geom('geom1').feature('r2').set('pos', {'0': 'd3+d4+d5+d6+2d7'});
model.geom('geom1').feature('r2').setIndex('layer', 'gs/2', 0);
model.geom('geom1').feature('r2').setIndex('layer', 'gg', 1);
model.geom('geom1').feature('r2').setIndex('layer', 'gs', 2);
model.geom('geom1').feature('r2').setIndex('layer', 'gg', 3);
model.geom('geom1').feature('r2').set('layerbottom', 'off');
model.geom('geom1').feature('r2').set('layerleft', 'on');
model.geom('geom1').feature('r2').setIndex('layer', 'gs', 4);
model.geom('geom1').feature('r2').setIndex('layer', 'gg', 5);
model.geom('geom1').runPre('fin');
model.geom('geom1').run;

%% Plot of domain numbers use to check geometry
% figure
% axis tight
% error('stop')
% figurek
% mfiggeom(model, 'geom1', 'Facelabels', 'on')
% axis tight
% error('stop')

model.physics('emw').feature('weel').setIndex('materialType', 'solid', 0);
model.physics('emw').feature('weel').set('DisplacementFieldModel', 'RefractiveIndex');
model.material.create('mat1', 'Common', 'compl');
model.material('mat1').label('dC_index');
model.material('mat1').propertyGroup.create('RefractiveIndex', 'Refractive index');
model.material('mat1').propertyGroup('RefractiveIndex').set('n', {'real(dC_index)'});
model.material('mat1').propertyGroup('RefractiveIndex').set('ki', {'imag(dC_index)'});
model.material.create('mat2', 'Common', 'compl');
model.material('mat2').label('dG_index');
model.material('mat2').propertyGroup.create('RefractiveIndex', 'Refractive index');
model.material('mat2').propertyGroup('RefractiveIndex').set('n', {'real(dG_index)'});
model.material('mat2').propertyGroup('RefractiveIndex').set('ki', {'imag(dG_index)'});
model.material('mat2').selection.set([7 11 13 15]); Manually Change
model.material.create('mat3', 'Common', 'compl');
model.material('mat3').label('d2_index');
model.material('mat3').propertyGroup.create('RefractiveIndex', 'Refractive index');
model.material('mat3').propertyGroup('RefractiveIndex').set('n', {'real(d2_index)'});
model.material('mat3').propertyGroup('RefractiveIndex').set('ki', {'imag(d2_index)'});
model.material('mat3').selection.set([6]); Manually Change
model.material.create('mat4', 'Common', 'compl');
model.material('mat4').label('d3_index');
model.material('mat4').propertyGroup.create('RefractiveIndex', 'Refractive index');
model.material('mat4').propertyGroup('RefractiveIndex').set('n', {'real(d3_index)'});
model.material('mat4').propertyGroup('RefractiveIndex').set('ki', {'imag(d3_index)'});
model.material('mat4').selection.set([5]); Manually Change
model.material.create('mat5', 'Common', 'compl1');
model.material('mat5').label('dPR_index');
model.material('mat5').propertyGroup.create('RefractiveIndex', 'Refractive index');
model.material('mat5').propertyGroup('RefractiveIndex').set('n', {real(dPR_index)});
model.material('mat5').propertyGroup('RefractiveIndex').set('k', {imag(dPR_index)});

PR_domain=4; % Manually Change
model.material('mat5').selection.set([PR_domain]);
model.material('mat6', 'Common', 'compl1');
model.material('mat6').label('d5_index');
model.material('mat6').propertyGroup.create('RefractiveIndex', 'Refractive index');
model.material('mat6').propertyGroup('RefractiveIndex').set('n', {real(d5_index)});
model.material('mat6').propertyGroup('RefractiveIndex').set('k', {imag(d5_index)});
model.material('mat6').selection.set([3]);
model.material('mat7', 'Common', 'compl1');
model.material('mat7').label('dS_index');
model.material('mat7').propertyGroup.create('RefractiveIndex', 'Refractive index');
model.material('mat7').propertyGroup('RefractiveIndex').set('n', {real(dS_index)});
model.material('mat7').propertyGroup('RefractiveIndex').set('k', {imag(dS_index)});
model.material('mat7').selection.set([1 2]);

model.coordSystem.create('pml1', 'geom1', 'PML'); % top PML
model.coordSystem('pml1').selection.set([length(materials)+2]);
model.coordSystem.create('pml2', 'geom1', 'PML'); % bottom PML
model.coordSystem('pml2').selection.set([1]);

model.physics('emw').feature.create('pc1', 'PeriodicCondition', 1);
model.physics('emw').feature('pc1').set('PeriodicType', 'Floquet');</code>

RFBN=1:2:((length(materials)+2)*2–1); % depends heavily on the number of grating periods
RFBN_end=((length(materials)+2)*2+1+6*3+1);
RFBN=RFBN_begin+1:RFBN_end;

model.physics('emw').feature('pc1').selection.set([RFBN RFBN_end]);
model.physics('emw').feature('pc1').selection.set([LFBN RFBN]);
model.physics('emw').feature('pc1').selection.set([length(materials)+2]*2);
model.physics('emw').feature('pc1').set('PortExcitation', 'on');
model.physics('emw').feature('port1').setIndex('PortSlit', '0');
model.physics('emw').feature('port1').set('InputType', 'H');
model.physics('emw').feature('port1').set('H0', {'0' '0' 'exp(-i*k1*x)'}, 'TM polarization');
model.physics('emw').feature('port1').set('beta', 'abs(k1)');

model.mesh('mesh1').autoMeshSize(1); % Verify that this is extremely fine mesh
model.study('std1').feature('freq').set('plist', 'f0');
model.label('dual_resonator_plasmons_1.mph');
model.study('std1').create('param', 'Parametric');
model.study('std1').feature('param').setIndex('pname', 'lambda', 0);
model.study('std1').feature('param').setIndex('plistarr', '', 0);
model.study('std1').feature('param').setIndex('punit', '', 0);
model.study('std1').feature('param').setIndex('pname', 'd3', 0);
model.study('std1').feature('param').setIndex('plistarr', t2, 0);  # altered
model.study('std1').feature('param').setIndex('punit', 'nm', 0);
model.study('std1').feature('param').setIndex('pname', 'd4', 1);
model.study('std1').feature('param').setIndex('plistarr', t3, 1);  # altered
model.study('std1').feature('param').setIndex('punit', 'nm', 1);
model.study('std1').feature('param').setIndex('pname', 'd6', 2);
model.study('std1').feature('param').setIndex('plistarr', t5, 2);  # altered
model.study('std1').feature('param').setIndex('punit', 'nm', 2);

model.sol.create('sol1');
model.sol('sol1').study('std1');

model.study('std1').feature('freq').set('notlistsolnum', 1);
model.study('std1').feature('freq').set('notsolnum', 1);
model.study('std1').feature('freq').set('listsolnum', 1);

model.study('std1').feature('freq').set('solnum', 1);

model.sol('sol1').create('st1', 'StudyStep');
model.sol('sol1').feature('st1').set('study', 'std1');
model.sol('sol1').feature('st1').set('studystep', 'freq');
model.sol('sol1').create('v1', 'Variables');
model.sol('sol1').feature('v1').set('control', 'freq');
model.sol('sol1').create('s1', 'Stationary');
model.sol('sol1').feature('s1').create('pl1', 'Parametric');
model.sol('sol1').feature('s1').feature('pl1').feature('fDef');
model.sol('sol1').feature('s1').feature('pl1').set('pname', { 'freq' });
model.sol('sol1').feature('s1').feature('pl1').set('plistarr', { 'f0' });
model.sol('sol1').feature('s1').feature('pl1').set('punit', { 'Hz' });
model.sol('sol1').feature('s1').feature('pl1').set('pcontinutionmode', 'no');
model.sol('sol1').feature('s1').feature('pl1').set('preusesol', 'auto');
model.sol('sol1').feature('s1').feature('pl1').set('plot', 'off');
model.sol('sol1').feature('s1').feature('pl1').set('plotgroup', 'Default');
model.sol('sol1').feature('s1').feature('pl1').set('probesel', 'all');
model.sol('sol1').feature('s1').feature('pl1').set('probes', {});
model.sol('sol1').feature('s1').feature('pl1').set('control', 'freq');
model.sol('sol1').feature('s1').set('control', 'freq');
model.sol('sol1').feature('aDef').set('complexfun', true);
model.sol('sol1').create('fc1', 'FullyCoupled');
model.sol('sol1').feature('s1').create('fc1').set('linsolver', 'dDef');
model.sol('sol1').feature('s1').feature('s1').feature('fcDef');
model.sol('sol1').attach('std1');

model.batch.create('pl1', 'Parametric');
model.batch('pl1').study('std1');
model.batch('pl1').create('sol', 'Solutionseq');
model.batch('pl1').feature('sol').set('seq', 'sol1');
model.batch('pl1').feature('sol').set('store', 'on');
model.batch('pl1').feature('sol').set('clear', 'on');
model.batch('pl1').feature('sol').set('psol', 'none');
model.batch('pl').set('pname', sp(1:));
% model.batch('pl').set('plistarr', {'50,150' , '5.50' , '5.50' , '5.50' , '0.1,0.5'});
model.batch('pl').set('plistarr', fp);
model.batch('pl').set('sweepertype', 'sparse');
model.batch('pl').set('probesel', 'all');
model.batch('pl').set('proben', {});
model.batch('pl').set('plot', 'off');
model.batch('pl').set('err', 'on');
model.batch('pl').set('pdistrib', 'off');
model.batch('pl').attach('std1');
model.batch('pl').set('control', 'param');
model.sol.create('sol2');
model.sol('sol2').study('std1');
model.sol('sol2').label('Parametric Solutions 1');
model.batch('pl').feature('sol1').set('psol', 'sol2');
model.result.create('pgl1', 'PlotGroup2D');
model.result('pgl1').label('Electric Field (emw)');
model.result('pgl1').set('data', 'dset2');
model.result('pgl1').set('oldanalysistype', 'noneavailable');
model.result('pgl1').set('frametype', 'spatial');
model.result('pgl1').set('data', 'dset2');
model.result('pgl1').feature.create('surf1', 'Surface');
model.result('pgl1').feature('surf1').set('oldanalysistype', 'noneavailable');
model.result('pgl1').feature('surf1').set('data', 'parent');
model.result.numeral.create('gev1', 'EvalGlobal');
model.result.numeral('gev1').set('data', 'dset2');
model.result.numeral('gev1').set('expr', 'emw.S11dB');
model.batch('pl').run;
model.result('pgl1').run;

Number_of_mesh_elements=model.mesh('mesh1').getNumElm('tri');
stats = mphmeshstats(model);
if aG==0
toc
end
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%% Fishing with data %%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%% P=xv;
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%% for i2=1:size(numbers,1)
solu = mpheval(model, 'emw.normE', 'dataset', 'dset2', 'selection', PR_domain,'outersolnum',i2); % extracting solution data
v = solu.dl'; % normE values
x = solu.p(1,:); % x coords
y = solu.p(2,:); % y coords
mx=min(x);Mx=max(x);my=min(y);My=max(y);
xqa=mx;gr*10^−9:Mx;yqa=my;gr*10^−9:My;
\% F = scatteredInterpolant(x, y, v, 'nearest', 'nearest'); \% pick the one that 'looks' the best
F = scatteredInterpolant(x, y, v, 'linear', 'nearest');
\% F = scatteredInterpolant(x, y, v, 'linear', 'linear');
[xqb, yqb] = meshgrid(xqa, yqa);
vq = F(xqb, yqb);
xq = xqb(1, :) \* 10^-9; yq = yqb(:, 1) \* 10^-9;
top = max(max(vq));

ic = GA_ideal_curve_catenary(w, nPR, Px, m, xq, yq, mx, my, Mx, My); \% ideal curve function.
ic = real(ic);

\%sse1(i2) = sum(sum((vq/top - ic)^2));
end

%%%%%%%%%%%%%%%%%%%%%%%%%% Best fitness plots each generation %%%%%%%%%%%%%%%%%%%%%%%%%%%
[i, f2] = min(sse1);

solut = mpeval(model, 'emw.normE', 'dataset', 'dset2', 'selection', PR_domain, 'outersolnum', f2); \% extracting solution data
v = solut.d1; \% normE values
x = solut.p(1, :) \% x coords
y = solut.p(2, :) \% y coords
mx = min(x); Mx = max(x); my = min(y); My = max(y);
xqa = mx: gr \* 10^-9:Mx; yqa = my: gr \* 10^-9:My;
\% F = scatteredInterpolant(x, y, v, 'nearest', 'nearest'); \% pick the one that 'looks' the best
F = scatteredInterpolant(x, y, v, 'linear', 'nearest');
\% F = scatteredInterpolant(x, y, v, 'linear', 'linear');
[xqb, yqb] = meshgrid(xqa, yqa);
vq = F(xqb, yqb);
xq = xqb(1, :) \* 10^-9; yq = yqb(:, 1) \* 10^-9;
top = max(max(vq));

ic = GA_ideal_curve_catenary(w, nPR, Px, m, xq, yq, mx, my, Mx, My); \% ideal curve function
ic = real(ic);

\%sse2 = (vq/top - ic)\^2;

%%%%%%%%%%%%%%%%%%%%%%%%%% Plot best individual of each generation %%%%%%%%%%%%%%%%%%%%%%%%%%%
yq = yq - min(yq); \% simplify the axes
d2b = numbers(f2, 1);
d3b = numbers(f2, 2);
d5b = numbers(f2, 3);
figure
subplot(3, 1, 1)
contourf(xq, yq, vq/top, 100, 'LineColor', 'none');
colorbar
caxis([0 1]);
title(['Best Fitness PR E-field (Normalized) - d2=' num2str(d2b) ' d3=' num2str(d3b) ' d5=' num2str(d5b)])
xlabel('Width (nm)')
ylabel('PR Thickness (nm)')
 subplot(3,1,2)
 contourf(xq,yq,real(ic),100,'LineColor','none')
colorbar
caxis([0 1]);
title('Ideal PR E-field (Normalized)')
xlabel('Width (nm)')
ylabel('PR Thickness (nm)')
 subplot(3,1,3)
 contourf(xq,yq,sse2,100,'LineColor','none')
colorbar
caxis([0 1]);
title('Square of Error')
xlabel('Width (nm)')
ylabel('PR Thickness (nm)')
 g_count=g_count+1;
 if g_count<10
 print('-dtiffn',[pwd,'\Generation 0',num2str(g_count)],'-r70'); % 70 is perhaps not too giant. Consider changing to eps for latex compatibility.
 else
 print('-dtiffn',[pwd,'\Generation ',num2str(g_count)],'-r70'); % 70 is perhaps not too giant. Consider changing to eps for latex compatibility.
 end
 set(gcf,'visible','off')
 % % % % % % % % % % % % % % % % %
 if g_count==oG+1
 best_fit = mpheval(model,'emw.normE','dataset','dset2','outersolnum',f2); % extracting solution data
 figure
 mphplot(best_fit,'Mesh','off');
 axis tight
 print('-dtiffn',[pwd,'\Best_fit'],'-r70'); % 70 is perhaps not too giant. Consider changing to eps for latex compatibility.
 best_fit = mpheval(model,'emw.normE','dataset','dset2','selection',[2:1:length(materials)+1:length(materials)+3]:1:(length(materials)+8),'outersolnum',f2); % extracting solution data
 figure
 mphplot(best_fit,'Mesh','off');
 axis tight
 ax = gca;
 ax.XTickLabel = {'0','100','200','300','400','500','600','700','800'};
 ax.YTickLabel = {'0','100','200','300','400','500','600','700','800'};
 xlabel('Width (nm)')
ylabel('Height (nm)')
 % % % % lines (bottom to top) % % % %
 ld1=2*line_d7;
 ld2=ld1+d5b*10^-9;
 ld3=ld2+line_d5;
 ld4=ld3+d3b*10^-9;
ld5=ld4+d2b*10^-9;
ld6=ld5+line_d2;
lw=0.5;

line ([mx Mx],[ld1 ld1], 'LineWidth',lw,'Color','k')
line ([mx Mx],[ld2 ld2], 'LineWidth',lw,'Color','k')
line ([mx Mx],[ld3 ld3], 'LineWidth',lw,'Color','k')
line ([mx Mx],[ld4 ld4], 'LineWidth',lw,'Color','k')
line ([mx Mx],[ld5 ld5], 'LineWidth',lw,'Color','k')

line ([mx line_gs/2],[ld6 ld6], 'LineWidth',lw,'Color','k')
line ([mx+line_gs/2+line_gg ,mx+line_gs/2+line_gg+line_ggs], [ld6 ld6], 'LineWidth',lw,'Color','k')
line ([mx+line_gs/2+line_gg+line_ggs+line_ggs+line_ggs+line_ggs+line_ggs], [ld6 ld6], 'LineWidth',lw,'Color','k')
line ([mx+line_gs/2+line_gg+line_ggs+line_ggs+line_ggs+line_ggs+line_ggs+line_ggs+line_ggs], [ld6 ld6], 'LineWidth',lw,'Color','k')

figure
subplot(2,1,1)
contourf(xq,yq,vq/top,100,'LineColor','none');
h = colorbar;
ylabel(h,'I{\texttt{\{normalized\}}}','FontSize',16);
caxis([0 1]);
title('Best Fitness PR E-field ')
xlabel('Width (nm)')
ylabel('PR Thickness (nm)')

subplot(2,1,2)
contourf(xq,yq,real(ic),100,'LineColor','none')
h = colorbar;
ylabel(h,'I{\texttt{\{normalized\}}}','FontSize',16);
caxis([0 1]);
title('Ideal PR E-field ')
xlabel('Width (nm)')

\begin{verbatim}
print(’--diffn’, [pwd,’/Best_fit_zoom’],’--r300’); % 70 is perhaps not too giant.

Consider changing to eps for latex compatibility.
\end{verbatim}
Listing C.3: Matlab function to allocate the GA parameter values.

```matlab
function [x] = mapvariables_vector_ga(x, mincM)

% MAPVARIABLES Map the GA parameters to more ‘suitable’ ones
% Naturally suitable considers time to run. But also there is little
% point in having sub-nanometer thick layers nor high resolution on
% factors that have little impact. Also COMSOL will die if you put a zero
% thickness here.

for i1 = 1:size(mincM, 2)
    v1 = mincM(1, i1) : mincM(2, i1) : mincM(3, i1);
    x(:, i1) = v1(x(:, i1));
end
```

Listing C.4: Matlab function file for the PR evanescent fields.

```matlab
function [C]= GA_ideal_curve_exponential(w,nPR,Px,xq,yq,mx,Mx,my,My)

% all dimension in nm
% trying to produce the ideal curve to compare the comsol stuff against

k=2*pi/w;
nm=10^-9; % nm conversion

PR_thick=(My-my);
P=Px/nm; % convert to nm

Px=Px/abs(2*m); % pitch
kx=abs(2*pi*m/Px);
NA=kx/k;

x=xq';
A=(-cos(2*pi/P*x)+1)/2; %
y=yq;
y=y-my*nm; % offset back to zero
b=nPR*k*sqrt(NA^2/nPR^2-1);
B=exp(-b*(y-PR_thick));

figure
% subplot(2,1,1) % checking forms
% plot(x,A)
% axis([0,Mx*nm,0,1])
% subplot(2,1,2)
% plot(y,B)
% axis([0,PR_thick,0,1])
```
Listing C.5: Matlab function GA population distribution plots.

```matlab
function state = gapopdist1(options, state, flag, mincM)
% GAPOPDIST Plots the location of the parameters in a 1D space

persistent data; % may not be in use anymore?

x2=mincM(1):mincM(2):mincM(3); lx2=length(x2);

nEliteKids = options.EliteCount; % number of elite children.

nXoverKids = round(options.CrossoverFraction * (options.PopulationSize - nEliteKids));
% number of crossover kids

nMutateKids = options.PopulationSize - nEliteKids - nXoverKids; % number of mutants, ha

nK=nEliteKids+nXoverKids+nMutateKids; % population size, I don't know the state handle
for this, so this will do.

gen = state.Generation; %
x=state.Population;
x=x2(mincM(1):mincM(2):mincM(3)); % numbers

x_record=x*[ms(3);ms(2);ms(1)]; % concatenated numbers

data(:,gen+1)=x_record; % numbers storage

for i0=1:size(x,1)
    [aa]=find(x2==nx(i0));
    xrx(i0)=aa; xry(i0)=1; % coordinates of numbers. I artificially places it on a line
    xrx(i0)=nx(i0); xry(i0)=1; % coordinates of numbers. I artificially places it on a line
end

switch flag
    case 'init'
        xlim([1,mincM(3)])
        ylim([0,2])
        plot(xrx,xry,'xk')
        hold on
        title('GA population distribution')
    case 'iter'
        if gen>=2

        end
end
```

C=B^*A;
C=flipud(C);

Listing C.6: Matlab function GA population parameters evolution plots.

```matlab
function [] = population_evolution(pops, mincM, sp)
    % this plots the evolution of the population. Consider changing the plot
    % type, perhaps it's not as clear as it should be. Also colormap.

    for i1 = 1:size(mincM, 2)
        edges = [(mincM(1, i1) - mincM(2, i1)/2):mincM(2, i1):(mincM(3, i1)+mincM(2, i1)/2)];
        p(:, i2) = (histcounts(pops(:, :, i1), edges))';
    end
    pX = 0:size(pops(:, :, i1), 2) - 1;
    pY = mincM(1, i1):mincM(2, i1):mincM(3, i1);
    figure
    pcolor(pX, pY, p)
    title([('Population Evolution'), FontSize', 16])
    xlabel('Generation', 'FontSize', 16)
    ylabel('Parameter Value', 'FontSize', 16)
    colormap jet
    h = colorbar;
    ylabel(h, 'Individual Count', 'FontSize', 16);
    ax = gca;
    ax.FontSize = 16;
    shading interp
    print('-dtiffn', [pwd, '\Population Evolution - Parameter ', num2str(i1)], '-r70'); %
    70 is perhaps not too giant. Consider changing to eps for latex compatibility
end
```
References


Accessed Online; 09 February 2017.


REFERENCES


