Investigation of Monolithic Erbium-Doped Resonators for Application in Cavity Quantum Electrodynamics

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Abstract

In this thesis we investigate the potential of erbium ions coupled to crystalline whispering-gallery mode resonators as hardware for quantum information processing. Achieving strong coupling between the erbium ions and the cavity would be a significant breakthrough for multiple quantum computing applications. To this end, we fabricate millimetre-sized resonators made from 0.001\% erbium-doped yttrium orthosilicate (Er$^{3+}$:Y$_2$SiO$_5$). We show that the mechanical fabrication procedure used in making the resonators does not significantly impact the coherence properties of the erbium dopants.

We find that strong coupling should be achievable using currently available technologies, so long as significant absorption in the Y$_2$SiO$_5$ crystals is not present at the wavelength of the erbium transition.
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Chapter 1

Introduction

An important piece of technology for the future of quantum information science is a coherent, reversible interface between single photons and stationary qubits \[1\]. Such a component would allow the creation of quantum networks \[2\], essential for large-scale distributed quantum computing. An interconnect of this type could also function as a deterministic single photon source \[3, 4\], which would enhance the prospects of the promising linear optics quantum computing scheme \[5\], as well as improving the functionality of quantum key distribution \[6, 7\]. Currently the standard tool for creating single photons is using spontaneous parametric down-conversion (SPDC) \[8\], however such photons are created at random time intervals and cannot be produced on demand.

The difficulty is that most matter qubits do not couple anywhere near strongly enough to the electromagnetic field to have a coherent, reversible interaction with a single photon. The main tactic in overcoming this has been to use cavities to enhance the interaction. Coupling qubits to low loss cavities with small mode volumes, it is possible to reach a regime in which coherent light-matter interactions dominate the incoherent dissipative processes in the system. This regime is called the strong coupling regime of cavity quantum electrodynamics (QED) \[9, 10, 11\].

The best quantum light-matter interfaces to date have been made using laser-cooled atoms trapped in microscopic Fabry-Perot resonators. Jeff Kimble’s group at Caltech used this method to reversibly map a coherent state with average photon number of \(\bar{n} = 1.1\) onto the hyperfine states of a trapped cesium atom and back \[12\]. Interference between the retrieved state and a reference field phase coherent with the input state showed that the mapping was indeed coherent \[1\].

Similar experiments have been done using rubidium \[3, 13\]. In fact, an elementary quantum network consisting of two rubidium atom nodes has been created, and entanglement between the nodes demonstrated \[14\]. In that experiment the atom-cavity system is in the intermediate-coupling regime, and results would be improved in the strong-coupling regime, although this proves the intermediate-coupling regime is still useful.

Deterministic single-photon sources have also been created using quantum dots strongly coupled to pillar microcavities \[15, 16\]. These pillar microcavities are made of semiconductor substrate with

\[1\] A coherent state was used because of its well-defined phase which was necessary to observe interference fringes, and their experiment also works in principle with a true single photon.
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distributed Bragg reflector mirrors at each end, forming a monolithic Fabry-Perot type cavity with the quantum dots inside. Strong coupling has also been achieved between a single quantum dot and a photonic crystal cavity [17]. In that work, the authors identify a single quantum dot using atomic force microscopy (AFM), allowing them to choose a lone quantum dot with the desired emission characteristics. The photonic crystal cavity is then built around the chosen quantum dot, with cavity resonance frequency tailored to the dot. This novel technique holds much promise for future scalable quantum networks.

In this work, we investigate the possibility of using rare-earth ions strongly coupled to monolithic resonators in order to create a coherent light-matter interface. In addition to the possibilities outlined above, this would pave the way for scalable rare-earth ion quantum computing [18]. Rare-earth ions are promising candidates for qubits in a large-scale quantum computer, due to their long optical and hyperfine coherence times, which are maintained in a solid state environment [19]. The most well-developed proposals for rare-earth ion quantum computing to date use rare-earth ion doped crystals, the computer being a collection of ions located close enough together for electric dipole-dipole interactions to occur [20]. For ensemble-based schemes multiple identical collections of ions must occur throughout the crystal to provide enough interaction cross-section with the laser light used for manipulating the computer. The number of identical computer “instances” decreases exponentially with increasing numbers of qubits due to the random nature of the doping, hence such schemes are not scalable. In the strong coupling regime single dopants can be addressed [21] which would allow single-instance quantum computing using only one collection of ions, which has far better scalability prospects.

Instead of using Fabry-Perot resonators, we fabricated whispering gallery mode (WGM) resonators out of disks of yttrium orthosilicate doped with erbium (Er$^{3+}$:Y$_2$SiO$_5$). This scheme has much in common with those outlined above for quantum dots, and has several advantages over using trapped atoms in Fabry-Perot resonators. Firstly, miniaturisation of WGM resonators is more straightforward, as being monolithic vibrational instabilities are less of an issue [22]. Fabrication of a WGM resonator that has both small mode volume and large quality factor (both desirable properties in enhancing atom-cavity coupling) is relatively straightforward compared to making an Fabry-Perot resonator with the same properties. Also, using monolithic resonators with solid state optically active elements eliminates the need for complex atom traps. In our scheme, we do not need to trap our ions in the cavity field; they are already held in place by the crystal structure. This is a huge benefit, shared by the quantum dot schemes, as trapping single atoms is a technically non-trivial task.

Strong coupling has been achieved using WGM resonators by dropping laser cooled cesium atoms past a resonator, such that a few of them fall through the evanescent field and are strongly coupled during their transit [23]. Our scheme has benefits over this method, in that the field intensity will always be larger inside the cavity (where our ions are located) than in the evanescent field, and the fact that our ions are locked in place in the crystal removes the need for trapping atoms in the evanescent field.
Rare-earth ions strongly coupled to cavities also have some advantages over similar solid-state schemes using quantum dots. Manufacturing processes for quantum dots usually result in a variety of non-identical quantum dots with different optical properties. Also, transition frequencies in quantum dots are generally quite sensitive to temperature [15]. These factors could be problematic in a quantum network, in which different nodes should create identical single photons. Indistinguishable photons from distinct quantum dot sources have been demonstrated [24], however using rare-earth ions would simplify things as all ions in the same environment have identical optical properties, and transition frequencies are insensitive to temperature.

The material Er$^{3+}$:Y$_2$SiO$_5$ was chosen because the erbium ions have a long-lived transition with the potential for long coherence times which occurs at 1536 nm, in the very important telecommunications band where silica optical fibers have their lowest loss. A quantum network constructed from erbium ions strongly coupled to cavities could utilise the extensive current technologies that have been developed for optical data transmission around 1500 nm, and the long lifetime of the excited state (11.4 ms [25]) would allow storage at the nodes. This provides our motivation for investigating the plausibility of reaching the strong coupling regime using Er$^{3+}$:Y$_2$SiO$_5$ WGM resonators.

1.1 Previous Work

This work carries on directly from previous research done in the lab of Dr Jevon Longdell, in which the suitability of praseodymium-doped yttrium orthosilicate (Pr$^{3+}$:Y$_2$SiO$_5$) WGM resonators for quantum computing applications was investigated [26]. There are two reasons for switching material to Er$^{3+}$:Y$_2$SiO$_5$. The first is that the $^4I_{15/2} \rightarrow ^4I_{13/2}$ transition at 1536 nm in erbium ions has the potential for longer coherence times than the $^3H_4 \rightarrow ^1D_2$ at 606 nm in praseodymium ions investigated in previous work [27], an advantageous property for quantum computing applications. The second is that it was suspected higher quality factor resonators with less dissipation might be possible using light at 1536 nm inside the resonator rather than 606 nm. Because WGM resonators are monolithic, modes propagate inside the actual solid material rather than in free space like Fabry-Perot resonators. This means that the upper limit on quality factor is determined by absorption in the resonator material, which causes attenuation of the propagating light. It is common for transparent materials to have a window of minimum absorption in the vicinity of 1500 nm [28], as often this wavelength lies between absorption from electronic transitions at lower wavelengths and vibrational transitions at higher wavelengths. If yttrium orthosilicate had a transparency window in this region then the upper limit on the quality factor of a WGM resonator should be higher for light at 1536 nm than 606 nm.

It should be noted that there is a virtual absence of absorption data in the literature for many transparent crystals, due to the difficulty of measuring a tiny decrease in amplitude upon passing through a sample of small path length. Absorption in silica fibers is well known because the signal amplitude can be measured at either end of a very long fiber, however this is in general not possible for crystals due to the impracticality of growing a crystal which extends over several kilometres. Because of this it is unknown how absorption in yttrium orthosilicate actually compares at 1536 nm to 606 nm, and our earlier assertion that the absorption might be less is based on trends observed in
other crystals.

Also, scattering from surface defects (another factor that can diminish quality factor) is expected to be smaller for longer wavelengths. Intuitively, this can be understood because a defect of the same dimension is smaller relative to a wavelength at 1536 nm than 606 nm. This implies less polishing should be required to obtain the same quality factor when working at the longer wavelength.

In order to obtain the long optical coherence times that Er$^{3+}$ is capable of, it is necessary to apply strong magnetic fields ($\sim 7$ T) and have low temperatures ($\sim 1.5$ K). Temperatures of $\sim 2$ K are possible in our lab, however we have no capability to make such high magnetic fields, so experiments done in this thesis should be thought of as initial exploratory work in determining the suitability of Er$^{3+}$:Y$_2$SiO$_5$ in quantum computing applications rather than a serious attempt to reach the strong coupling regime. Resonators of $\sim 3$ mm diameter were fabricated as it is possible to make these by hand and infer the properties of smaller resonators based on results from the millimeter-sized resonators. Micrometer-sized resonators fabricated by automated single-point diamond turning will likely be necessary to achieve strong coupling in the future.

1.2 Thesis Outline

This thesis begins with an overview of background material necessary to place the work done in context and interpret results. Chapter 2 is a brief overview of basic quantum computing ideas, as well as a comparison of current experimental efforts at making one. Chapter 3 describes properties of rare-earth ion doped crystals in more detail, with a special focus on Er$^{3+}$:Y$_2$SiO$_5$. Chapter 4 introduces concepts from cavity QED important to this research. Chapter 5 discusses basic properties of WGM resonators, and also details methods for manufacturing and coupling light into them. Chapter 6 presents results, which are a characterisation of properties we observed in our Er$^{3+}$:Y$_2$SiO$_5$ samples as well as experiments determining how close our atom-cavity systems were to the strong coupling regime.
Chapter 2

Quantum Computing Overview

2.1 The Birth of Quantum Computing

In 1837, Charles Babbage came up with a design for what he called an “Analytical Engine,” which today is widely recognised as the first programmable computer [29]. Unfortunately for Babbage his design was never realised during his lifetime, and it was over a century until the world saw its first general-purpose computers during World War II [30]. In 1936, a young man by the name of Alan Turing laid the theoretical foundations for much of modern computer science, when he introduced the idea of the universal Turing machine [31]. A Turing machine can be thought of as representing a computer that can perform some algorithm. The scheme Turing proposed for his machine had an infinite line of ticker-tape that could be read and written upon by a mechanical head, which had access to a set of program instructions. The actual details of a Turing machine are less important than its usage as a model for computation, where it is still widely used in computational complexity theory. A universal Turing machine can simulate any other Turing machine, hence can simulate any algorithmic process. A machine that can compute the same range of functions as a universal Turing machine is called Turing complete. In this language, Babbage’s Analytical Engine was the first design for a Turing complete computer. Modern digital computers are Turing complete, and any program (even vastly complicated ones) can in theory be simulated efficiently on a universal Turing machine. Because of this, study of Turing machines has provided a great deal of understanding of the ultimate limits of computer power.

Surprisingly at the time, Turing also showed that certain processes do not lend themselves well to computation on a Turing machine [31]. Stochastic algorithms for certain problems have been shown to be far more efficient than any deterministic algorithm that could be simulated on a Turing machine [33]. The innate probabilistic nature of quantum mechanics led the great physicist Richard Feynman to contemplate the prospects for simulating quantum systems on computers. In 1982 he introduced the idea of a quantum computer when considering whether it would be possible for a computer to exactly simulate nature by behaving in exactly the same way [34]. Feynman went on to suggest that perhaps a

\[\text{In computer science, “efficient” simulation means that the amount of resources required must be bounded by a polynomial in the problem size [32]. If the resources required increase exponentially with the size of the problem then an algorithm is inefficient. Resources here could refers to things like the amount of memory required, or the number of steps in the algorithm.}\]
certain class of quantum computer could efficiently simulate another quantum system, alluding to what David Deutsch called a universal quantum computer [35]. This is important because quantum systems cannot be simulated efficiently on a conventional computer, in which the exponentially increasing size of the Hilbert space is problematic, and simulations are limited to quantum systems with a handful of constituents [36]. Other important problems currently intractable on a conventional computer have been shown to have efficient solutions on a universal quantum computer [37, 38].

The most common scheme for quantum computation uses two-level systems as the base elements of information [39]. These two-level computational units are called “qubits” (quantum bits) in analogy to their binary counterpart [40]. Other schemes that use more complicated quantum objects as the computational base elements also exist (for example using three-level qutrits [41]). These schemes may offer some advantages over qubits, however they have their own difficulties and are beyond the scope of this discussion.

Before quantum computing is discussed in more detail, it is useful to elaborate on what a qubit is, and how it differs from a bit.

### 2.2 Visualising Qubits

A qubit is a two-level quantum system, the simplest possible quantum object that shows non-trivial behaviour. For example, a qubit could be encoded in the ground and excited states of an atom, or the polarisation states of a photon, or the spin of an electron. Although a qubit can have many different physical implementations, mathematically and conceptually it is the same object across all of these systems [42].

The major difference between a qubit and a bit is that a bit can only be in one of its logical states at a time, whereas due to the peculiarities of quantum mechanics a qubit can be in a superposition of both [43]. The clearest visual representation of a qubit is the Bloch sphere, named after Swiss physicist Felix Bloch in honour of his contributions to nuclear magnetic resonance. The Bloch sphere is a unit sphere that can be used to express the state of any two-level quantum system. The rules for using this useful construct are simple. The line connecting the origin of the sphere and a point on the surface is called a Bloch vector, and carries all the information about the system state. Taking $|0\rangle$ and $|1\rangle$ to be orthogonal basis states spanning the two-dimensional qubit Hilbert space (called the computational basis states, and usually chosen to be eigenstates of the qubit), an arbitrary qubit state

$$|\psi\rangle = \alpha|0\rangle + \beta|1\rangle, \quad (2.1)$$

may be written equivalently as

$$|\psi\rangle = \sin \left( \frac{\theta}{2} \right) |0\rangle + e^{i\phi} \cos \left( \frac{\theta}{2} \right) |1\rangle, \quad (2.2)$$

due to the fact that sine and cosine form a linearly independent basis for the set $[0,1]$, so the (arbitrary) coefficients $\alpha$ and $\beta$ can be parameterised by $\theta$. The complex exponential allows for a phase difference of $\beta$ relative to $\alpha$. Such a state can be represented on the Bloch sphere as shown in Figure 2.1 below.
2.2 Visualising Qubits

Figure 2.1: Bloch sphere representation of the qubit given in equation 2.2.

It should be noted that a common convention inverts the $|0\rangle$ and $|1\rangle$ basis vectors relative to that shown here.

The Bloch sphere was introduced in the context of nuclear magnetic resonance, however due to the equivalence of a nuclear spin in a magnetic field to any other two-level system we can represent any qubit using this formalism. For example, a qubit defined by the ground and excited states of an atom is usually represented on the Bloch sphere by defining a “pseudospin” vector that traces out a path which can be mapped back to the time evolution of the atomic state [44].

Examining a few special cases is useful in illustrating how the Bloch sphere works. Firstly, binary bits can be represented with Bloch vectors either parallel (state $|1\rangle$ using the above convention) or anti-parallel (state $|0\rangle$) to the $\hat{z}$ axis. This is not surprising, as the state space of a quantum computer subsumes that of a classical computer of the same size [45], so it is expected that a qubit should be able to represent a bit. The superposition

$$|\psi\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |1\rangle),$$  \hspace{1cm} (2.3)

has a Bloch vector pointing to the equator of the Bloch sphere, directly along the $\hat{x}$ axis, and the superposition

$$|\psi\rangle = \frac{1}{\sqrt{2}}(|0\rangle + i|1\rangle),$$  \hspace{1cm} (2.4)

also has a Bloch vector on the equator, this time pointing directly along the $\hat{y}$ axis. This makes sense,\textsuperscript{2}

\textsuperscript{2}Physicists seemingly feel more comfortable with the $|1\rangle$ state at the top of the Bloch sphere, while computer scientists like it the other way around.
as from equation 2.2 it can be seen that for these superpositions \( \sin \left( \frac{\theta}{2} \right) = \cos \left( \frac{\theta}{2} \right) = \frac{1}{\sqrt{2}} \), so the polar angle \( \theta = \frac{\pi}{2} \) and the Bloch vector is along the equator. The convention implied in equation 2.2 is that the \( \hat{x} \) axis is the phase reference point, as shown in equation 2.3 which has zero phase offset. From equation 2.4 it is seen that the Bloch vector along the \( \hat{y} \) axis has a \( \pi/2 \) phase offset. It is thus seen that the projection of the Bloch vector along the \( \hat{z} \) axis gives information about the composition of the superposition, for example what would be called the inversion for an atomic level qubit, or the magnetic polarisation for a nuclear spin qubit. The projection onto the \( \hat{x} - \hat{y} \) plane tells us about the phase of the superposition. The state of any qubit can be represented as a point on the surface of the Bloch sphere in this way. Qubit dynamics are modeled by the path the Bloch vector traces out on the sphere.

### 2.2.1 Characterisation of Qubits

Certain parameters are common to all qubits irrespective of their physical implementation. A brief introduction to these parameters will be useful when it comes to comparing experimental quantum computing schemes in section 2.6. The first is the coherence time, which is how long phase information is preserved in a qubit before decoherence occurs. Using the Bloch sphere, this can be thought about as follows. If the state of a qubit is engineered to be an equal superposition of the computational basis states, the Bloch vector will point along the equator and precess around at a rate given by the frequency difference between the \( |0 \rangle \) and \( |1 \rangle \) basis states. If the qubit is not perfectly isolated from its environment, then random perturbations from uncontrollable external influences will cause stochastic alterations in the energy levels of the computational basis states. This means that the rotation rate of the Bloch vector is not precisely fixed and will have some uncertainty. How long it takes before the location of the Bloch vector can no longer be predicted from the initial rotation rate is an intuitive description of the coherence time. For optical transitions, the coherence time is equal to one over the homogeneous linewidth, \( T_2 = 1/\Gamma_h \) (\( T_2 \) is common notation for the coherence time).

Another way in which decoherence can occur is due to population decay. Instead of losing knowledge of the orientation of the Bloch vector due to variations in rotation rate, this causes the Bloch vector to decay back to state \( |0 \rangle \), which has an undefined phase. The rate at which this happens is the called the population lifetime in the context of atomic energy level qubits. The population lifetime puts an upper limit on the coherence time through the relation \( T_2 \leq 2T_1 \) [46], where \( T_1 \) is the population lifetime. It is common to have \( T_2 \ll T_1 \). Like much of the study of two-level systems, these quantities were first introduced in the context of nuclear magnetic resonance, in which \( T_1 \) is the spin-lattice relaxation rate and \( T_2 \) is the spin-spin relaxation rate [47].

Most quantum computing schemes involve a number of entangled qubits. Without yet referring to a physical form this can be thought of as a collection of Bloch spheres each one representing a qubit, and some sort of coupling between Bloch spheres such that the motion the Bloch vector on one sphere is strongly correlated with the motion on another sphere, representing entanglement. Decoherence destroys these correlations by randomising the phases of each qubit, destroying the computational power. This means that long coherence times are desired for qubits in quantum computing applications. Long in this context means long compared to gate operation times which change the orientation
of the Bloch vectors though some external influence. This will be discussed further in section 2.4.

2.3 Applications and Limitations of a Quantum Computer

It is often stated that quantum computers function in a fundamentally different way to conventional digital computers. In this section an attempt is made to give the reader a feel for the manner in which the computations differ. Are quantum computers inherently more efficient at solving problems than their classical counterparts? If so, why? Is there a reason for this based in physical laws? At this point in time it is impossible to say [48], as it is not even known what the problem-solving limits of classical computation are [49]. What is known is that there are certain problems for which quantum algorithms exist that are far more efficient than current algorithms for solving the same problem on a classical computer. In what follows an attempt will be made to give some insight into the mechanism of efficiency increase for certain quantum algorithms.

As a simple example consider a classical computer with a three-bit binary register, which can represent \(2^3 = 8\) different pieces of information. In order to perform a computation an algorithm might be performed in which the machine is initialised to some input state, then several bit operations performed to give an output state representing the result of the computation. The details of the algorithm are irrelevant for this discussion. Now consider a quantum three-qubit register in which the system state has been engineered to be a superposition of each possible classical option. We can write this (normalised) state out in Dirac notation (on the computational basis states) as:

\[
|\psi\rangle = \frac{1}{\sqrt{8}}(|000\rangle + |001\rangle + |010\rangle + |011\rangle + |100\rangle + |101\rangle + |110\rangle + |111\rangle).
\]

(2.5)

If an equivalent calculation to that performed on the classical register is now done using unitary transformations on the quantum register, some of the subtleties of quantum computing are apparent. A word of caution is necessary here; quantum computation by unitary evolution is inherently reversible, whereas classical computation may not be (for example, any circuit with an OR gate in it destroys information, as the input cannot be deduced from the output). Fortunately it turns out that an irreversible classical algorithm can always be represented as a reversible one with at most polynomial overhead [50], meaning that a universal quantum computer can efficiently simulate any classical algorithm [48]. In other words, a universal quantum computer is Turing complete, and our above example is valid. This does not mean that an arbitrary algorithm will run faster on a quantum computer; in fact, chances are there will be some overhead in satisfying the reversibility requirement. T.D. Ladd et al. draw a parallel between computers and light sources [45]. A laser can be thought of as a coherent quantum light source, just as a quantum computer is a coherent computer. In the same way that lasers have not replaced incoherent light sources (e.g. incandescent lightbulbs), quantum computers are not expected to replace incoherent (classical) computers for many applications. There are certain tasks that quantum computers are far more suited to, but chances are if a successful quantum computer is ever implemented it will work alongside classical computers rather than replacing them.

Because the quantum computer can be in a superposition of all the possible states the classical computer could be in at once, it can perform operations on all of these states simultaneously during
the same computation. The initial state comprises a superposition of all the classical register states, and running the appropriate algorithm gives an output state which contains information about solutions for all the classical input states. This quantum parallelism [35] is fundamentally different from conventional parallel computing, in that the computations are being carried out at the same time on the same piece of hardware. Hence quantum computation is parallel in a way classical computation can never be [51]. Extending our three-bit register to an N-bit register,

$$|\psi\rangle = \frac{1}{\sqrt{N}} \sum_{i_1, i_2, \ldots, i_N} |i_1, i_2, \ldots, i_N\rangle.$$  \hspace{1cm} (2.6)

Quantum computation then proceeds through a sequence of gate operations on the qubits, which mathematically involves unitary operations evolving the state forward in time. Take one such operation,

$$U|\psi\rangle = U \frac{1}{\sqrt{N}} \sum_{i_1, i_2, \ldots, i_N} |i_1, i_2, \ldots, i_N\rangle = \frac{1}{\sqrt{N}} \sum_{i_1, i_2, \ldots, i_N} U|i_1, i_2, \ldots, i_N\rangle,$$  \hspace{1cm} (2.7)

where the unitary transformation $U$ acts linearly on the quantum register state. Hence this gate operation acts on all the possible classical register states simultaneously, and a sequence of such operations linked together to make a computation will have carried out the same algorithm on all the possible classical input states at the same time. This enormous parallelism is what gives various quantum algorithms their power. However this is not the final story. It is well known that measurements on quantum systems project the system into an eigenstate of the observable being measured [52]. If the state of the quantum register is measured, one of the classical register states will be the result, and all information about the others will be lost. The quantum parallelism then has no observable effect. The system effectively picks one of the classical computational paths corresponding to a classical input. Where the quantum algorithm can out-perform the classical is by utilising interference between different solution paths [51]. This can give information about general properties of solutions, which in some cases can be used to efficiently find a specific solution. This is the basic idea behind Shor’s famous algorithm for factorising prime numbers, although the precise details are more complicated [37]. Shor’s algorithm is exponentially faster at factoring primes than the best known classical algorithm, which is important because a great deal of current encrypted data exchange is presumed to be secure based on the fact that it is very hard to factor large prime numbers using current algorithms on digital computers [55]. Shor’s algorithm was the first demonstration of a truly important problem that could be solved efficiently on a quantum computer for which a tractable classical algorithm is currently unknown (or possibly unknowable). Since then other quantum algorithms have been developed that are more efficient than their classical counterparts (such as Grover’s algorithm for searching an unsorted database [38, 56]), but it was Shor’s algorithm that generated the most interest in quantum computing.

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3Banks, intelligence agencies [53], government departments and all manner of people who wish to keep communications secret use encryption which would be made vulnerable by a sufficiently powerful quantum computer. However those concerned about their privacy need not worry just yet; current state of the art demonstrations of Shor’s algorithm have recently factorised the number 21 [54], a long way from posing a security threat.
After the excitement generated by Shor’s algorithm, a multitude of different approaches were proposed to experimentally realise a working quantum computer. The different systems considered spanned much of modern physics. As David DiVincenzo noted “This amazing variety of approaches has arisen because, the principles of quantum computing are posed using the most fundamental ideas of quantum mechanics, ones whose embodiment can be contemplated in virtually any branch of quantum physics [57].” Because at the most elementary level our world is a quantum one, any system displaying quantum behaviour that can be sufficiently controlled is a potential candidate. In an attempt to focus experimental research to plausible areas, in 2000 DiVincenzo introduced five requirements a quantum system should comply with in order to be a good candidate for a scalable quantum computer [57]. These have become known as the DiVincenzo criteria.

2.4 The DiVincenzo Criteria

All systems are thought to be quantum-mechanical at the most fundamental level, but not all systems are suitable for quantum computation. The DiVincenzo criteria give a checklist that a quantum system should fulfil to be considered as a viable candidate for the experimental implementation of a quantum computer. Different schemes struggle with different DiVincenzo criteria, and no current scheme fulfils all the DiVincenzo criteria completely satisfactorily [57]. They provide a useful means of comparison of experimental schemes as will be seen in section 2.6.

1. **A scalable physical system with well characterised qubits**
   A potential candidate should be scalable in that it should be possible to add more and more qubits to the computer without an exponential increase in the amount of resources necessary to control them. The meaning of the term “well characterised qubit” is that the essential parameters of the qubit Hamiltonian should be well known [57]. As well as the internal qubit Hamiltonian, this also includes couplings to unwanted states not part of the qubit, interactions between qubits that are important in entangling gate operations, and interaction with external fields used to manipulate the qubit state. For example for a qubit encoded in two energy levels of an atom, couplings between these states and other energy levels which are not part of the qubit should be well understood, atom-atom interactions should be known, as should the transition dipole moment for manipulating the qubit with laser light.

2. **The ability to initialise the qubits to a well-defined state**
   This requirement comes from the fact that to begin a computation, a known initial input state must be used, otherwise we do not know what we will be calculating\(^4\) (often the initial state is taken to be \(|000\ldots\rangle\)). DiVincenzo also pointed out that quantum error correction requires a steady stream of fresh qubits in some desired state (for example \(|0\rangle\)) [57], which puts some constraints on how the quantum system generates such states, as the qubits to be used for error correction must be generated fast enough to correct the error accumulating. For example in nuclear magnetic resonance (NMR) quantum computing fresh qubits for error correction

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\(^4\)A common phrase in computer science summarising this is garbage in, garbage out.
cannot be obtained simply by waiting for qubits to return to their ground state, as the coherence time is always shorter than the thermalisation time. This is one reason why NMR quantum computing is currently not a good candidate for a large-scale quantum computer.

3. **Long relevant decoherence times, much longer than the gate operation time**

Reducing decoherence in quantum systems is a major experimental challenge. In a qubit, decoherence occurs when the phase relationship between the $|0\rangle$ and $|1\rangle$ states is disrupted by interaction with the environment. The phase information is incredibly fragile, so extreme isolation from coupling to environmental degrees of freedom is required [45]. Decoherence is the mechanism by which the classical world emerges from the quantum, so to utilise quantum effects slow decoherence is a must. If the decoherence time is comparable to the gate operation time, then significant dephasing can occur during the operation, potentially giving large errors in the output. It can be shown that such errors can be corrected if the decoherence time is $10^4 - 10^5$ times the gate operation time (assuming the gate operation itself does not introduce any errors, which may not be a valid assumption) [57]. More generally, fault-tolerant quantum computing can be performed if cumulative errors are kept below a certain level (this is known as the threshold theorem) [58].

4. **A universal set of quantum gates**

A universal set of quantum gates is a set from which any other gate operation can be constructed. Any point in the Hilbert space spanned by the quantum computer can be reached applying only gates in the universal set. This means that if the quantum computer Hilbert space has a one-to-one mapping to another quantum system, a universal set of gates allows the quantum computer to completely simulate this other quantum system, hence acting like a universal quantum computer [48].

It turns out that a universal set of quantum gates can be implemented using only single-qubit arbitrary phase rotation gates and almost any two-qubit gate, often taken to be the controlled-not (CNOT) gate [48]. A single-qubit arbitrary phase rotation gate allows the state of a single qubit to be put anywhere on the Bloch sphere. The CNOT gate changes the state of a target qubit dependent on the state of a control qubit. The two-qubit gate provides an entangling operation. More complicated gates can be implemented by applying a sequence of these gates, for example a three-qubit gate operation could work using CNOT between two qubits, some single-qubit arbitrary phase rotations if desired then CNOT again between one of those qubits and a third qubit. The result is entanglement between all three qubits.

5. **A qubit-specific measurement capability**

As with any computation, a mechanism for reading out the output is necessary. To get the output, some way of measuring the state of individual qubits in the computer must exist. There are two distinct types of measurement which are performed for different quantum computing schemes; ensemble-based measurements and projective measurements. These will be discussed further in section 2.6.
DiVincenzo also introduced two “communication criteria” outlining the requirements for quantum networks. These are the ability to convert between stationary and flying qubits, and the ability to faithfully transmit flying qubits between specified locations [57]. An erbium ion strongly coupled to a cavity would be a significant step towards both of these requirements; conversion between stationary and flying qubits would in theory be possible [21] and the creation of flying qubits would be in the form of 1536 nm photons, which can be transmitted with minimum loss between locations using silica fibers.

Building a functioning quantum computer is a formidable technical challenge. Quantum superposition states are extremely fragile, and a quantum computer able to out-perform its classical counterpart will require a large multi-qubit superposition [48]. This requires a high level of isolation from the environment to preserve quantum information. On the other hand, controlled interaction with an external source and interactions between qubits are necessary in current schemes for single-qubit operations and multiple-qubit entangling operations. These two seemingly contradictory requirements are major reasons for the difficulties encountered in putting together a functioning quantum computer.

However, due to the remarkable ingenuity and perseverance of both theoretical and experimental physicists in the last 20 years, significant progress has been made in several systems. In what follows we discuss quantum computing using cryogenic rare-earth ion doped crystals, which is most pertinent to this research, and then briefly review other promising implementations.

### 2.5 Rare-Earth Ion Quantum Computing

Rare-earth ions are promising candidates for quantum information processing, due to their potential for long optical and hyperfine coherence times in a solid state host [25]. Several proposals have been put forward for rare-earth ion quantum computing [59, 60, 61, 18]. Initial efforts focused on ensemble-based schemes. As the scalability issues of ensemble-based proposals are a strong motivating factor for this research, one such scheme will be briefly outlined here to illustrate the issues. The scheme we discuss is based on work by Wesenberg et al. [62, 18], and is conceptually similar to other ensemble-based proposals. Our coverage here is by no means comprehensive, and is merely intended to highlight the difficulties with scalability.

The scheme is implemented using rare-earth ions doped into inorganic crystals, with qubits encoded in the energy levels of an optical transition. The ions in the crystal experience a variety of different environments depending on their location due to variations in crystal strain. This slight variation in local ion environment over the ensemble leads to a slight variation in transition frequencies, giving rise to inhomogeneous broadening. The range of transition frequencies means that individual qubits can be defined by their frequency. Gate operations on qubits are done with controlling laser beams to provide arbitrary single-qubit phase rotation. A CNOT gate can also be performed which gives a universal set of quantum gates, satisfying DiVincenzo criterion number four. CNOT is performed using static dipole-dipole couplings between ions in the crystal [63]. This works because the excited state of a rare-earth ion in a crystal can have a different static electric dipole moment to the ground state [64]. This means that if an ion is in the excited state it will have a different interaction
with ions in its vicinity than if it were in the ground state. A CNOT gate is where the operation on a target qubit is dependent on the state of a control qubit; it is hence seen that the ion-ion electric dipole interaction satisfies this requirement. The process is similar to the dipole blockade effect in Rydberg atoms \[65\] \[66\]. A viable concern here is that the dipole-dipole interaction depends on the distance between two ions, hence when trying to put together a multi-qubit quantum computer interaction strengths will be different between all the qubits. Fortunately it has been shown that as long as it exceeds a certain threshold the exact interaction strength does not matter \[18\].

Multiple different quantum computer “instances” will occur inside the crystal, made up of collections of ions close enough together for CNOT gates to be applied. Due to the random nature of the doping not all of these computer instances will be the same. Tailoring of the inhomogeneous line-shape can be used to define a quantum computer based on frequency through a process that has been called instance identification \[62\]. Instance identification of a two-qubit register in a rare-earth ion doped crystal is shown graphically in Figure 2.2. First narrow anti-holes are burned into the inhomogeneous linewidth. Tailoring of the inhomogeneous linewidth by spectral hole burning is a common technique for rare-earth ion doped media, as hyperfine levels can have lifetimes of the order hours. A narrow linewidth laser will cause pumping of resonant ions into other hyperfine states, hence there will be a decrease in absorption at the laser frequency. This is called a spectral hole. An anti-hole is a spectral region of increased absorption. Figure 2.2 shows two narrow anti-holes in the inhomogeneous linewidth created by burning away ions with nearby resonant frequencies. It should be noted that often in a crystal the inhomogeneous linewidth will have a Gaussian shape, and the flat profile shown in Figure 2.2 indicates the slow variation at this frequency resolution. The narrow anti-holes serve as qubits \[59\]. Ideally the anti-hole would be equal to the homogeneous linewidth of the ions. Many ions will have a frequency within the anti-hole and our qubit is really multiple individual qubits which will respond in the same way to laser light and should all be in the same state. This makes qubit readout straightforward, and means that this is an ensemble-based scheme.

To find the ions which have dipole-dipole interaction strengths above the threshold required for CNOT operation a $\pi$-pulse is applied at the transition frequency of qubit 1. This puts qubit 1 into the excited state and will hence cause a shift in frequency of nearby ions due to the different Stark shift from the excited state static electric dipole moment. In particular ions making up qubit 2 will be shifted in frequency by an amount dependent on their interaction strength with qubit 1 ions as shown in Figure 2.2b with a smaller shift meaning a smaller interaction strength. Ions which have not shifted enough to be above the interaction threshold for CNOT operation can then be pumped to a different hyperfine state, removing them from the qubit. This is shown in Figure 2.2c. The process can then be repeated with the roles of qubit 1 and 2 reversed. The result of all this is two qubits which can display a universal set of quantum gates \[64\]. A computer with more qubits could be made using the same instance identification procedures with more anti-hole qubits burned into the inhomogeneous line.

\[5\] Unless they happen to be equally spaced throughout the crystal, an exceedingly unlikely prospect for random doping
2.5 Rare-Earth Ion Quantum Computing

(a) Two qubits defined by burning a narrow anti-hole into the inhomogeneous linewidth. Ideally the anti-hole should be the same width as the homogeneous linewidth. Many ions in the crystal have transition frequencies inside the anti-hole which means larger signals for qubit readout, hence why this is an ensemble-based scheme.

(b) The ions of qubit 1 are excited, causing a frequency shift for atoms nearby due to the change in static dipole moment.

(c) All the ions in qubit 2 that have not shifted in frequency enough for CNOT operation to be possible are burned away, leaving only the desired ions. The process is then repeated with the roles of qubit 1 and 2 reversed.

Figure 2.2: Instance identification of two qubits in ensemble-based rare-earth ion quantum computing.
Rare-earth ions can have inhomogeneous linewidths $\sim 10^8$ times larger than homogeneous linewidths [67], so in theory the inhomogeneous linewidth is wide enough for a vast number of qubits. The problem with this scheme is that more and more ions have to be burned away to be left with the desired quantum computer instances as the number of qubits is increased. The number of available computer instances decreases exponentially with the number of qubits for weakly doped crystal [18]. This means the scheme is not scalable. One way of getting around this is to use stoichiometrically doped materials in which the ions become a majority species rather than a dopant [68]. In this thesis a different direction is pursued, which is moving away from ensemble-based computing methods and instead computing using single quantum computer instances. Achieving strong coupling between a rare-earth ions and a resonator would allow detection of single dopants [21] and hence qubit-specific readout in a single quantum computer instance. If this were to be achieved then all five DiVincenzo criteria could be satisfied using rare-earth ions.

2.6 Other Experimental Schemes

2.6.1 Liquid State Nuclear Magnetic Resonance

By the time quantum computing ideas had been developed far enough for practical implementation to become a real possibility, the field of nuclear magnetic resonance (NMR) spectroscopy was already very well established. Due to the significant body of knowledge that already existed regarding the control and interpretation of NMR results, some of the earliest demonstrations of non-trivial quantum algorithms were done using NMR quantum computers [69]. Qubits are encoded in the nuclear spin states of atoms in a molecule, which can have coherence times of the order seconds at room temperature [48] (for a liquid, coherence times are orders of magnitude less in a solid). A large static magnetic field breaks any nuclear spin degeneracy, providing the necessary two levels. Arbitrary phase rotation gates can be done by applying appropriate radio-frequency (RF) pulses, and two-qubit entangling operations required for universality are based on inter-nuclear interactions. The size of the molecule determines the number of qubits in the computer.

Because the energy difference between nuclear spin up and spin down states is very small, a large number of molecules ($10^{18}$ or more [32]) is required to give enough signal for read-out, which is done by measuring magnetic induction in a set of RF coils [48]. NMR quantum computing is hence an ensemble-based scheme. In many respects NMR quantum computing is similar to ensemble-based rare-earth ion schemes, where a collection of ions close enough to interact forms the “molecule.”

An NMR quantum computer with 12 qubits has been demonstrated [70], which is currently a very respectable number. However, the current consensus is that NMR-based schemes are not scalable beyond a handful of qubits [48]. NMR quantum computing is thus regarded as having provided a valuable learning and testing ground in the past, and not a viable candidate for the future.

2.6.2 Ion-trap

Some of the most impressive experiments to date have been done using ion-trap quantum computers. Ion-trap quantum computing was first proposed by Juan Ignacio Cirac and Peter Zoller in 1995 [71]...
and the essential components were swiftly demonstrated experimentally [72]. Qubits are encoded in hyperfine states of the trapped ion, which can have exceptionally long coherence times [73]. Single-qubit gate operations can be performed by addressing the ions with laser beams, and multi-qubit entangling operations are done using vibrational normal modes of ions in the trap [71]. Readout of a single qubit can be done by measuring fluorescence [74].

At the time of writing 14-qubit entanglement has been demonstrated in an ion trap [75], currently the state of the art in terms of controlled entanglement. The only DiVincenzo criterion that ion-trap quantum computing will face major problems with is the scalability requirement, as in the scheme described above the more ions in the trap the closer the vibrational energy levels are spaced, which causes problems for large ion numbers. However, physicists do not give up easily, and several proposals to make scalable ion-trap quantum computing a reality are currently being investigated, including the possibility of joining several ion-trap “nodes” via quantum channels, and the fabrication of ion-trap microchips [2, 76, 77, 78].

2.6.3 Photonic

In photonic quantum computing, the state of a photon itself is used as a qubit, rather than some sort of atomic degree of freedom as in the previous sections. This is very appealing for several reasons, firstly due to the fact that photons interact very weakly with their surroundings compared to electrons or atomic nuclei, meaning that controlling decoherence is relatively straightforward and photons are very stable carriers of quantum information. Also, photonic qubits can be manipulated easily using standard laboratory equipment. For example if a qubit was encoded in the orthogonal polarisation states of a photon, single-qubit gate operations could be performed using ordinary waveplates. Problems arise however when attempting the two-qubit operations that are essential for a universal quantum computer. The fact that photons are weakly interacting, which is beneficial for preventing decoherence, also means that arranging the necessary interaction for a two-qubit operation is difficult. Photons can be made to interact with each other in nonlinear materials, however performing an entangling operation on two specific qubits using a nonlinear medium is difficult, as the effect of the nonlinearity depends on the intensity of light in the crystal, and in general having only two photons does not give large enough interactions. Optical cavities can in principle be used to overcome this, however in 2001 Knill, LaFlamme and Milburn proposed a remarkable scheme in which two-qubit operations between photons can be done using only linear optical elements [5]. Their method is rather resource-intensive, and the details are not as important for this discussion as the conclusion that photonic quantum computing can be done efficiently using only linear optical elements. This scheme would benefit greatly from new hardware, such as deterministic single-photon sources (currently single photons are usually created non-deterministically using spontaneous parametric down-conversion).

2.6.4 Superconducting Qubits

It is well known from classical physics that identical equations describe an LC resonator circuit and a harmonic oscillator [79]. The same correspondence applies between a superconducting LC circuit
and a quantum harmonic oscillator. At very low temperatures quantisation of the energy levels can be observed. Quantum harmonic oscillator energy levels are however unsuitable qubits as they are all equally spaced in energy, so the two levels to be used as a potential qubit cannot be distinguished in frequency from all the other levels. For this reason a non-linear circuit element is needed to provide anharmonicity to ensure that the energy levels are unequally spaced and a unique qubit can be defined in frequency. For a superconducting circuit this element is the Josephson junction, which is a thin layer of insulating material sandwiched between superconductors. Cooper pairs can tunnel through this layer and a Josephson junction is a lossless circuit element, which is necessary to observe quantum effects.

Different types of superconducting qubits can be defined, for example flux qubits in which quantum superpositions can be defined by counter-propagating currents yielding a superposition in magnetic flux. Also charge qubits can be defined, in which a small superconducting “Cooper pair box” is coupled to a superconducting reservoir by a Josephson junction, and the qubit is defined by the location either in the reservoir or Cooper pair box of an excess Cooper pair. Phase qubits are defined by the phase difference of a superconducting wavefunction across a Josephson junction [80]. Superconducting qubits are promising in that they are based on electrons, which have very strong interactions. The possibility of extremely rapid electrical control of qubits is very promising for a high-speed quantum computer. The decoherence mechanisms for superconducting qubits are complex and not very well understood, however some important demonstrations have been carried out [81, 82]. Superconducting qubits are not the most established technology, but they do have great promise for the future. It is perhaps telling that computer giant IBM (which has been heavily involved in quantum computing from its inception) has chosen superconducting qubits for its bid to make a quantum computer [80].

2.6.5 Alternative Quantum Computing

Most quantum computing schemes involve a register of qubits in which computation is carried out by unitary evolution and the result measured at the end. There are alternatives to this, such as cluster quantum computing in which the computer is prepared in a highly entangled state, and the computation proceeds via projective single-qubit measurements [83]. An algorithm would then be defined by the sequence of single-qubit measurements. Cluster-state quantum computing is irreversible, however it has been shown to still be capable of universal quantum computation [84, 85]. The main difficulty is then creating the initial highly entangled state. Quantum computing without entanglement has also been investigated [86].

2.7 Short-Term Goals for Quantum Information

Although the prospect of a large quantum computer powerful enough to run Shor’s algorithm for a large number is very exciting, it is a long term goal. Quantum information applications with more immediate prospects are important in spurring on research, and will briefly be discussed here.

An interesting scheme that is already being used commercially is quantum key distribution (QKD) [6].

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[6] In fact, QKD was used in 2007 to secure data transmission in a Swiss election [87].
QKD solves a major problem in symmetric key encryption, where two parties both have access to a key that must be kept secret for secure communication. The issue is how to distribute the secret key to the two parties without it being intercepted by a nefarious character along the way. QKD ensures that the secret key is guaranteed to be secure by the laws of physics [88]. The secret key is distributed by sending qubits between parties along a quantum channel. A well-established protocol for constructing a secret key from the exchanged qubits is detailed in [89]. Due to the nature of measurement in quantum mechanics, if an eavesdropper attempts to intercept qubits in the channel the act of measurement will introduce anomalies that are detectable by the secure parties. If an eavesdropper is detected, the secret key is discarded.

Currently commercial QKD schemes use weak coherent pulses with photon number \( \bar{n} < 1 \) to transmit qubits. This is convenient as such states can be easily created by attenuating a laser pulse. However the Poissonian photon number distribution of a coherent state means that there is a finite possibility of having more than one photon in a pulse, leaving QKD open to a number splitting attack in which an eavesdropper can split off and observe extra photons without disturbing the other transmitted photon. Things get worse for higher loss transmission lines as multi-photon states transmitted will be overrepresented at the receiving end [88]. Using true single photons such as those from single emitters removes this problem, and it has been shown that QKD with single photons can have higher secure bit rates over lossy channels [6]. This is important as a major goal for the future is a global QKD network using communication satellites [90]. In 2007 QKD over a 40 dB channel loss was demonstrated using single photons [91]. This was an important milestone, as it is above the threshold thought to be necessary for satellite uplink, potentially paving the way for a high data rate global QKD network.

Another prospect for quantum computing which is drawing closer is the simulation of other quantum systems. This was the problem Feynman was considering when he first introduced the idea of a quantum computer [34]. Simulating quantum systems on classical computers is difficult as the size of the Hilbert space grows exponentially with the system size. For example simulation of \( \sim 50 \) qubits would be impractical on a conventional computer [92]. A 50 qubit quantum computer could hence simulate problems intractable for current computing technology. 50 qubits is an achievable number based on current progress in quantum computing, and far more reasonable than the thousands of qubits suspected to be required for a computer powerful enough factor large prime numbers. Quantum simulators would be immensely helpful in the study of many-body physics. A quantum computer with well-known and well-characterised interactions could be engineered to simulate a given Hamiltonian, then results from the simulator compared to whatever physical system that Hamiltonian was thought to represent. An example is the important problem of the mechanism for high-temperature superconductivity, for which a quantum simulator would be eminently useful [92].

A third area which is currently stimulated research efforts is the design and creation of devices and protocols which will be important for the future of quantum computing. Such things as improving quantum teleportation protocols for future quantum networks [69, 93], creating quantum memories for reliable storage at nodes without corrupting the information [94, 95, 96, 97, 98] and schemes for
long-range distribution of entanglement [1, 99]. Also creation of efficient deterministic single photon sources [16, 100, 13, 101, 102] and methods for creating entanglement [103, 104]. In summary, there is much to be done in the short and mid term for both experimental and theoretical physicists. Achievable goals that are interesting in their own right along the way to a large quantum computer are important in encouraging and stimulating research.

2.8 Summary

In this chapter a brief overview has been given of the field of quantum computing. A full survey of this vast and rapidly expanding field would take up many more pages than are available to this author; the interested reader is referred to [34, 48, 57, 51, 45, 80] for more detailed information. An attempt has been made to place this work in context as a step towards single instance rare-earth ion quantum computing, as well as progress towards a coherent interface between stationary qubits and flying qubits and single photon source.

In the next chapter the properties of rare-earth ions are investigated in more detail, with a special focus on the material Er\(^{3+}\cdot Y_2SiO_5\) from which resonators were fabricated out of in this work.
Chapter 3

Properties of Rare Earth Ions

3.1 Introduction

The lanthanide series (often referred to as the rare earth elements) is a particularly interesting area of the periodic table, located as shown in Figure 3.1. Contrary to their name rare earth elements are not particularly rare, the most common of them (cerium) having about the same abundance on Earth as copper, and the rarest stable rare earth (thulium) having a similar abundance to iodine. The first rare earths were discovered near the town of Ytterby in Sweden, hence several of them derive their names from this area or surrounds. For optical applications rare-earth elements are dominated by the $3^+$ oxidation state, and oxidised rare-earth elements are referred to as rare-earth ions.

Rare earth ions have been studied by scientists from various fields for a long time, due to their unique and useful optical properties. Solid-state lasers with rare-earth ion doped gain media are common in research and industrial applications around the world, the most ubiquitous being the well-known Nd:YAG laser. Erbium-doped fiber lasers are also used for a wide variety of applications, as well as erbium-doped fiber amplifiers which are crucial for long-range fiber communication in the 1.5-$\mu$m band where silica telecom fibers have minimum attenuation. Rare-earth ions are also prominent as phosphors in gas discharge lighting.

Rare-earth ions are characterised by partially filled 4f orbitals, located spatially inside full 5s and 5p orbitals, as shown in Figure 3.2. The interesting optical properties of rare-earth ions come from transitions between 4f orbitals. The 5s and 5p orbitals provide very effective shielding for electrons in the 4f orbitals, resulting in 4f-4f transitions that are relatively insensitive to the material environment the ion is placed in. Many of these transitions lie in the visible region of the spectrum or nearby, making rare-earth ions useful in numerous lighting applications.

Rare-earth ion doped materials have also been studied for use in all-optical data processing and storage, for which the natural shielding from the environment allows narrow homogeneous linewidths at cryogenic temperatures, which could provide high density data storage using extensive frequency multiplexing at one spatial location.

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1The name rare-earth elements comes from the fact that rare-earth minerals are typically widely dispersed rather than being found in concentrated mineral deposits.
Figure 3.1: Periodic table showing position of the Lanthanide series, commonly called the rare earth elements.

Figure 3.2: Radial probability distribution functions for the rare earth ion gadolinium (Gd$^{3+}$). The partially full 4f orbitals are located spatially inside the full 5s and 5p orbitals, meaning that electrons in the 4f orbitals are shielded from their environment. This is a property common to all optically interesting rare-earth ions. Image reproduced from [106].
More recently such materials have come under scrutiny for use in quantum computing hardware [25]. Memories which store the quantum state of light are essential for some quantum networking protocols [99], and memories based on ensembles of cryogenic rare-earth ions in a crystal have proved to be some of the most useful yet demonstrated [95, 97, 94, 112].

3.2 Rare-Earth Ion Doped Crystals

Rare-earth ions are commonly doped into crystals which have yttrium as a major constituent. In this work rare-earth ion doped crystals at cryogenic temperatures were investigated, so an overview of properties in a crystalline environment is appropriate. The ordered structure of a crystal allows for far longer optical and hyperfine coherence times than would be possible in an amorphous solid, which is beneficial for our purposes.

Due to the effective shielding by 5s and 5p orbitals mentioned earlier, the 4f-4f energy level spectrum for an ion in a crystal is similar to the free ion spectrum. The effect of a crystalline environment is usually treated as a perturbation (called the crystal-field interaction), with the eigenstates of the free ion Hamiltonian used as basis states for the mathematical machinery of perturbation theory [19]. This makes rare-earth ions in solids unique in that energy level calculations are done using methods from atomic spectroscopy rather than band structure calculations [115]. The weak coupling to the crystal lattice also means that the spectra of 4f-4f transitions are usually dominated by narrow zero-phonon lines [19].

The effect of the crystal however does produce some important differences. Firstly electric dipole transitions between the 4f states are forbidden for a free ion. Putting an ion into a crystalline environment breaks the rotational symmetry of the ion, meaning that total angular momentum eigenvalues are no longer good quantum numbers. The new eigenstates of the ion in the crystal will be linear combinations of the free ion states, meaning that some mixing of wavefunctions with opposite parity occurs (unless the substitution site is centrosymmetric [19], which was not the case for the crystals used in this work) and the 4f-4f transitions become weakly allowed. The fact that 4f-4f electric dipole transitions are only weakly allowed means that such transitions can have very long radiative lifetimes (of the order 10 milliseconds in Er$^{3+}:Y_2SiO_5$ [27]). As the radiative lifetime puts the upper limit on the coherence time this also presents the possibility of long coherence times.

3.2.1 Energy Level Structure

In the 1960s Dieke and Crosswhite complied rare-earth ion energy level data into a single figure, which is consequently called the Dieke diagram [116]. The Dieke diagram is shown in Figure 3.3 for trivalent rare-earth ions doped in LaCl$_3$, with the $^4I_{15/2} \rightarrow ^4I_{13/2}$ transition in erbium used in this work highlighted. Because of the efficient shielding of 4f electrons, the Dieke diagram is similar for different host environments.

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2Yttrium is a transition metal, with many similar properties to the lanthanides. It is common for rare-earth ions in the 3$^+$ oxidation to substitute for Y$^{3+}$ ions, for example in yttrium aluminium garnet (YAG).

3For comparison, optical coherence times of $\sim 1$ $\mu$s are considered exceptionally long in an erbium-doped glass [113], compared to the record of 4.2 ms in an erbium-doped crystal [114].
Figure 3.3: Dieke diagram showing the energy level structure of rare-earth ions, with the arrow indicating the $^4I_{15/2} \rightarrow ^4I_{13/2}$ transition in Er$^{3+}$ studied in this thesis. Modified from [116].
The Hamiltonian describing electronic energy levels in an isolated rare-earth ion can be separated into a spherically symmetric (or central field) and a non-symmetric part. Energy level splittings between the 4f configurations are the most important for this thesis, and are determined by the non-symmetric part. The non-symmetric part comes from spin-orbit coupling and the non-central component of electron-electron repulsion. Spin-orbit coupling has interaction energies of the order $10^3 \text{ cm}^{-1}$, and non-central electron-electron repulsion $10^4 \text{ cm}^{-1}$ [19].

For a free ion the fact that the Hamiltonian has a non-symmetric component means that spin and orbital angular momenta are not separately conserved, hence are not good quantum numbers with which to label a state. The total angular momentum $J = L + S$ is conserved for the free ion and hence is a good quantum number along with $m_J$ (the projection onto the quantisation axis). For a free ion the energy eigenstates do not depend on $m_J$ and hence have a $2J + 1$ fold degeneracy. However when the ion is placed in a crystalline environment the energy eigenstates may depend on the shape of their corresponding position eigenfunctions. This partially lifts the $2J + 1$ fold degeneracy to form crystal-field split levels. Crystal-field split levels with the same $J$ (but different $m_J$) are referred to as $J$-manifolds, the magnitude of the splitting typically being of the order a few hundred $\text{cm}^{-1}$ [25]. The thick bands observed for some levels in Figure 3.3 are $J$-manifolds of states, and would be seen as discrete at a higher frequency resolution. The number of non-degenerate states in a $J$-manifold depends on the crystal site symmetry. The lower the site symmetry the higher number of inequivalent energy states due to reduced degeneracy [19].

The degree of crystal field splitting in a $J$-manifold also depends upon the number of electrons in the 4f shell. Ions with an odd number of electrons (thus having an unpaired electron spin) are called Kramers ions, and can split into at most $J+1/2$ levels. The eigenstates of these ions are required to be doubly degenerate by Kramers theorem [42], and this degeneracy can only be split by applying a magnetic field; the crystal field interaction cannot break it. The maximum number of levels with distinct crystal field energy levels is hence half the total number of states. Ions with an even number of 4f electrons are called non-Kramers ions and the crystal field interaction can cause splitting into the full $2J+1$ levels for these ions.

Hyperfine splitting of the crystal field levels is also possible for ions with non-zero nuclear spin, as well as nuclear electric quadrupole splitting for ions with nuclear spin greater than 1. Hyperfine levels can be very long lived (of the order hours in Eu$^{3+}$:Y$_2$SiO$_5$ [32]) and can be very useful in providing long-term population storage for spectral hole burning applications. Long hyperfine coherence times have also been demonstrated [117], allowing for the possibility of storing coherence in these levels. Hyperfine levels also allow access to three level systems such as lambda levels, which are important for many quantum information protocols.

The energy level splitting for Er$^{3+}$:Y$_2$SiO$_5$ is shown in Figure 3.4. Erbium is a Kramers ion, hence each crystal field level is a degenerate Kramers doublet that can only be split by application of a magnetic field. Only one stable isotope of erbium has nuclear spin ($^{167}$Er, 22.87% natural abundance and $I = 7/2$) and hence hyperfine structure. As optical pumping to hyperfine levels was not important for this work hyperfine structure is not shown. The crystal field levels are roughly to scale, with data on level structure taken from [27].
Figure 3.4: Schematic of crystal-field splitting of ground state in Er$^{3+}$. The spacing of the crystal field levels is roughly to scale with the actual splittings. The Zeeman splitting shown here is not to scale, and would be much smaller relative to the crystal field splitting in reality, of the order 5 GHz ($\sim$0.2 cm$^{-1}$) for the magnetic field strengths used in our experiments. Hyperfine splitting is not shown as only one stable isotope of erbium has nuclear spin. The total crystal field splitting is $\sim$500 cm$^{-1}$ which is equal to $\sim$15 terahertz.
3.2 Rare-Earth Ion Doped Crystals

At cryogenic temperatures only the lowest crystal field level will be significantly populated, as the thermal energy $k_B T$ is much smaller than the level splitting (for Er$^{3+}$:Y$_2$SiO$_5$ at 4 K the fraction in the second crystal field level is $\sim 7 \times 10^{-7}$, versus around $\sim 0.2$ at room temperature). The electronic excited state will also be affected by the crystal field interaction. The lowest excited state crystal field level will have the longest lifetime as it can only decay radiatively; there are no energy levels close enough for rapid non-radiative decay. Transitions from the lowest crystal field level of the electronic ground state to the lowest crystal field level of the excited state can be extremely narrow. Exciting an atom to a higher excited state crystal field level results in a rapid non-radiative decay followed by a radiative decay to the lowest excited state crystal field level, which will be a much broader transition. Lowest to lowest transitions are desired for quantum computing applications, and also have very high quantum efficiencies in erbium [19]. In this thesis the notation $^4I_{15/2}(k)$ is used to denote the crystal field energy level in a $J$-manifold where $k$ is the crystal field level, so for example $^4I_{15/2}(1) \rightarrow ^4I_{13/2}(1)$ denotes a lowest to lowest transition in erbium.

To summarise, rare-earth ions can have very complicated energy level structure. 4f-4f transitions in free ions are split into total angular momentum eigenstates, with splittings of the order $10^3 - 10^4$ cm$^{-1}$. When placed in a crystal, these states split into $J$-manifolds, with splittings of the order $10^2$ cm$^{-1}$. Hyperfine splitting can also occur for ions with nuclear spin, and is of the order $10^{-3} - 10^{-1}$ cm$^{-1}$ [19]. Zeeman splitting in Kramers ions is also possible, the magnitude of which depends on the applied magnetic field, and was $\sim 0.2$ cm$^{-1}$ in our experiments.

3.2.2 Inhomogeneous and Homogeneous Broadening

Inhomogeneous broadening for ions in a crystal results from variations in crystal strain altering the local environment across the ensemble of ions. Homogeneous broadening results from dynamic interactions between the ions and their complex solid-state surroundings, and is generally assumed to affect all ions in the crystal equally. In rare-earth ion doped crystals the distinction between the two is that inhomogeneous broadening results from static interactions with the crystal, whereas homogeneous broadening results from dynamic interactions with species in the surroundings. At room temperature the homogeneous linewidth is dominated by phonon broadening, however this is negligible at cryogenic temperatures for materials which do not have low-lying crystal field levels. In general at cryogenic temperatures the inhomogeneous linewidth is significantly larger than the homogeneous linewidth, as shown in Figure 3.5. There is however a large degree of potential variation and the exact ratio depends upon the material used. For many applications it is desirable to have a large ratio of inhomogeneous to homogeneous broadening [118].
Figure 3.5: Relationship between homogeneous and inhomogeneous broadening. It should be noted that in reality the homogeneous linewidths would be much more closely spaced, giving a continuous inhomogeneous linewidth. Only a representative sample is shown here to illustrate the point. The height of the $\Gamma_h$ frequency bins represents the number of ions with that transition frequency, so more ions gives more absorption.

3.2.3 Spectral Diffusion

Spectral diffusion is a dynamic process due to perturbation of ions’ transition frequencies by the local environment [119]. This is an important issue for many practical applications, as spectral diffusion introduces a decoherence mechanism that should be minimised for device applications [120]. For example if a narrow frequency channel within the inhomogeneous linewidth is considered, dynamic perturbations to the transition frequency will cause each ion in the channel will undergo a random walk in frequency (in other words it will “diffuse” through the spectrum) [120]. Some time later, the group of ions that was originally within the narrow frequency channel will have a wider frequency distribution. This is spectral diffusion, and is illustrated in Figure 3.6.

Spectral diffusion was important for our work, as it introduces an extra decoherence mechanism which must be accounted for. This is discussed in more detail in section 3.3.3.
3.3 Methods for Probing the Homogeneous Linewidth

Rare-earth ions can have very narrow homogeneous linewidths (less than 100 Hz in some cases [19]), meaning that frequency-domain methods for probing them would require ultra-stable lasers. Fortunately well-developed time-domain techniques exist which yield direct information about the material coherence time. These techniques also naturally account for the issue of probing a single homogeneous linewidth in an inhomogeneously broadened sample.

Two-pulse photon echoes can be used to extract the coherence time (and hence the homogeneous linewidth), and three-pulse photon echoes (also called stimulated photon echoes) can be used to investigate spectral diffusion. In this section both these types of photon echoes are discussed, making extensive use of the Bloch sphere picture introduced in section 2.2.

3.3.1 Two-pulse Photon Echo

Photon echoes are the optical analogue of spin echoes observed in nuclear magnetic resonance experiments by Hahn in the 1950’s [121]. In rare-earth ions two-pulse photon echoes provide a straightforward and sensitive method for probing the coherence time, and hence the homogeneous linewidth. First a $\pi/2$ pulse is applied to the sample, putting a selection of ions into a superposition with Bloch vectors pointing along the equator. The phase of the $\pi/2$ pulse determines which axis the Bloch vectors are rotated around onto the equator. The pulse by nature has a variety of frequency components, which excite the range of ions with the corresponding transition frequencies in the inhomogeneous profile. Because the excited ions have different transition frequencies, they will rotate at different rates around the Bloch sphere and dephase, after a time having zero macroscopic$^4$ polarisation density oscillation, and hence zero emission in the direction of the exciting pulse. However zero macroscopic polarisation oscillation does not mean the microscopic ions have stopped oscillating, only that they

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$^4$Macroscopic polarisation density oscillation only occurs when the ions are oscillating in phase.
are oscillating out of phase with each other. Applying a $\pi$-pulse at a later time results in a superposition with Bloch vectors precessing in the opposite direction, which all come back into phase at a time $\tau$ after the $\pi$-pulse, where $\tau$ is the time between the applied pulses. This rephasing of coherence results in an intense pulse of light as all the ions are oscillating in phase again. Decoherence occurs for the ions between pulses, resulting in a decay in echo amplitude as fewer of the ions come back into phase to create the photon echo. By measuring echo amplitude as a function of $\tau$ the coherence time can be measured, from which the homogeneous linewidth of ions in an inhomogeneously broadened ensemble can be inferred. This treatment of photon echoes is loosely based on that from [44].

It should be noted that the phases of the exciting pulses do not need to be kept coherent. On the Bloch sphere the phase of the incident pulses corresponds to the axis about which the Bloch vectors are rotated during the pulse. The phase of the first pulse is arbitrary as any rotation axis will put the Bloch vectors in the desired superposition. The rate of dephasing depends on the bandwidth of the exciting pulse; a large bandwidth pulse will excite a range of ions from the inhomogeneous linewidth with a broad spread in transition frequencies, hence a broad range in Bloch vector rotation rates which leads to a rapid initial decay in polarisation oscillation. After this decay time the Bloch vectors are spread evenly about the equator of the Bloch sphere, hence it does not matter what axis the $\pi$-pulse rotates them around to restore coherence. This means that the phase of the second pulse is not important, which removes the technically difficult prospect of keeping a well-defined phase relationship between the two pulses. In Figure 3.7 the rotation axis is chosen to be the $\hat{x}$ axis for both pulses, however this is arbitrary.

Detection of photon echoes is simplified by the fact that the echo is temporally separated from the excitation pulses, giving the detector time to recover. Photon echoes are also robust against the effects of imperfect $\pi/2$ and $\pi$-pulses.

One of the major positives of photon echo techniques is that they remove the need for ultra-stable lasers to probe the homogeneous line. The laser only needs to be stabilised to less than the spectral width of a $\pi$ pulse over the time scale $\tau$. Otherwise if the laser drifts too much, a different population of ions will be addressed by the second pulse than the first. This requirement is not particularly difficult to achieve, and even if it is not quite satisfied some echo will be produced. So-called “lucky shot” detection takes the biggest echo from a series of pulse sequences with the same parameters, as that is the echo that corresponds to the minimum laser frequency drift over the sequence. In our work $\pi$ pulses were typically around 1 $\mu$s in length so a laser linewidth of 1 MHz over $\sim$ 10 $\mu$s would have been acceptable. In fact, the laser used was much better than this.

A graphical summary of the two-pulse photon echo sequence is shown in Figure 3.8.

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5The word phase has been used in this section to mean several different things. When referring to laser pulses the phase is used to mean the point in the electromagnetic wave oscillation cycle, and when referring to transitions in ions it is used to mean the phase relationship between the ground and excited state, and it is also used to refer to dephasing of polarisation density caused by Bloch vectors (which are still phase coherent) starting to point in different directions. Hopefully it is clear from the context what is meant.
3.3 Methods for Probing the Homogeneous Linewidth

(a) Initially the ensemble of ions is in the ground state.

(b) $\pi/2$-pulse rotates around the $\hat{x}$ axis, creating a superposition state.

(c) Bloch vectors spread out due to variation in excitation frequencies.

(d) $\pi$-pulse inverts the Bloch vectors, causing them to precess in the opposite direction.

(e) Rephasing occurs after some time $\tau$, resulting in the emission of a photon echo.

**Figure 3.7:** Two-pulse photon echo represented on the Bloch sphere.
3.3.2 Three-pulse Photon Echo

Three-pulse photon echoes (also called stimulated photon echoes) can be used to examine spectral diffusion of the homogeneous linewidth, as well as determine the time scale over which excited ions return to their ground state. Three $\pi/2$ pulses are used to create an echo. Understanding a three-pulse photon echo sequence is perhaps best viewed by picturing the effect of the echo sequence on the inhomogeneous line in the frequency domain. The Fourier transform of the first two pulses has a modulation $e^{-i2\omega \tau_{12}}$ (where $\tau_{12}$ is the time between the first two pulses), due to the transform property that a displacement in the time domain corresponds to a phase shift in the frequency domain. This sets up a population grating which can be viewed as a modulation in absorption profile of the inhomogeneous line with spacing $1/\tau_{12}$. A third $\pi/2$ pulse at some later time $T$ causes an echo to be emitted. The modulation of the inhomogeneous absorption profile has a spacing $1/\tau_{12}$ which means that a time $\tau_{12}$ after the third $\pi/2$ pulse the phase of oscillation of ions in all of the different frequency
fringes will be in phase and an echo will be emitted. The emission mechanism is similar to that for an atomic frequency comb (AFC) if this is familiar to the reader [122].

The three-pulse photon echo sequence is shown in Figure 3.9.

\[ A(\tau) = A_0 \exp \left( -\frac{2\tau}{T_M} \right)^x. \]  

(3.1)

Here \( x \) is a parameter which describes the deviation from pure exponential, and \( T_M \) is called the effective phase memory time. The effective phase memory time is a more appropriate measure of coherence than the analogous pure exponential decay constant in the presence of spectral diffusion [123]. An effective homogeneous linewidth can also be defined as \( \Gamma_{\text{eff}} = 1/T_M \).

With no spectral diffusion, echo amplitude decay for a three-pulse photon echo has the form

\[ A(\tau_{12}, T) = A_0 \exp \left( -\frac{T}{T_1} \right) \exp \left( -2\tau_{12} \Gamma_h \right), \]  

(3.2)

where \( \tau_{12} \) and \( T \) are as shown in Figure 3.9 and \( T_1 \) is the population lifetime. The \( e^{-2\tau_{12} \Gamma_h} \) term (equivalent to \( e^{-2\tau_{12}/T_2} \)) describes decoherence occurring during the time \( \tau_{12} \) between the first two applied pulses, and also between the final pulse and when the echo occurs. The \( e^{-T/T_1} \) term describes population decay during the hold time \( T \).

Spectral diffusion has been successfully incorporated by modelling the frequency drift of each ion as a Lorentzian diffusion process [120]. Echo amplitude decay is then given by

\[ A(\tau_{12}, T) = A_0 \exp \left( -\frac{T}{T_1} \right) \exp \left( -2\tau_{12} \Gamma_{\text{eff}}(\tau_{12}, T) \right), \]  

(3.3)
where $\Gamma_{\text{eff}}(\tau_{12}, T)$ is a time-dependent effective linewidth. For a fixed value of $\tau_{12}$, this is given by

$$\Gamma_{\text{eff}}(T) = \Gamma_0 + \frac{1}{2} \Gamma_{SD} \left(1 - e^{-RT}\right), \quad (3.4)$$

where $\Gamma_0$ is the initial linewidth at times too short for spectral diffusion to be important, $\Gamma_{SD}$ is the linewidth the Lorentzian diffusion kernel, and $R$ is a rate associated with the underlying spectral diffusion mechanism \[120\]. If multiple spectral diffusion mechanisms are important, another term should be added with appropriate values of $\Gamma_{SD}$ and $R$.

It is thus seen that three-pulse photon echoes can be used to probe spectral diffusion mechanisms. Fitting experimental echo amplitude decay curves to equation (3.3), $\Gamma_{SD}$ and $R$ can be used as parameters for the fit. The values of these parameters and how they vary with temperature and magnetic field strength gives insight into the underlying mechanism causing the spectral diffusion. Extensive studies in Er$^{3+}$:Y$_2$SiO$_5$ have shown that for temperatures less than $\sim$ 2 K and magnetic fields greater than $\sim$ 0.8 T, local magnetic field fluctuations caused by erbium ion spin flips are the dominant cause of spectral diffusion \[120\]. This will be discussed further in section 3.4. We were working at higher temperatures and lower magnetic fields, and in addition to this observed spectral diffusion due to some other mechanism, most probably yttrium nuclear spin flips.

### 3.4 Suppressing Decoherence in Er$^{3+}$:Y$_2$SiO$_5$

It has been shown that erbium ions in Y$_2$SiO$_5$ can have exceptionally long coherence times \[124\], and spectral diffusion can be completely suppressed under certain conditions. The coherence time is related to the homogeneous linewidth by $T_2 = 1/\Gamma_h$, hence broadening or frequency drift of the homogeneous linewidth can be thought of as the frequency domain manifestation of decoherence. The homogeneous linewidth for Er$^{3+}$ can be written as the sum of several contributions \[27\]

$$\Gamma_h = \Gamma_{\text{pop}} + \Gamma_{\text{Er-Er}} + \Gamma_{\text{phonon}} + \Gamma_{\text{Er-Host}} + \Gamma_{\text{ISD}}, \quad (3.5)$$

where $\Gamma_{\text{pop}}$ is the population decay rate, which puts a fundamental limit on how narrow the homogeneous linewidth can be, and is related to the excited state lifetime by $T_1 = 1/(2\Gamma_{\text{pop}})$. $\Gamma_{\text{Er-Er}}$ is due to interactions between nearby Er$^{3+}$ spins mentioned above, $\Gamma_{\text{phonon}}$ comes from phonon scattering which distorts the crystal lattice and perturbs optical centers, $\Gamma_{\text{Er-Host}}$ is due to electronic and nuclear spin fluctuations of the host lattice and $\Gamma_{\text{ISD}}$ is due to instantaneous spectral diffusion (ISD). $\Gamma_{\text{ISD}}$ is an intensity-dependent phenomenon, as a higher intensity exciting laser beam will raise more ions into the excited state, which as mentioned in section \[27\] has a different static electric dipole moment to the ground state. The electric field fluctuations in the local environment of an ion as surrounding ions are excited and de-excited causes dynamic perturbations of the transition frequency, hence broadening the homogeneous linewidth. This instantaneous spectral diffusion can be minimised by using weak exciting laser pulses or lightly doped samples, which create low excitation.

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6The effective linewidth defined as $\Gamma_{\text{eff}} = 1/T_M$ above is strictly only relevant for small amounts of spectral diffusion; however $T_M$ is still useful when characterising decoherence in the presence of spectral diffusion to a first approximation \[120\].
3.4 Suppressing Decoherence in Er\(^{3+}:Y_2\text{SiO}_5\)

density.

\(\Gamma_{\text{phonon}}\) can be reduced by working at liquid helium temperatures, and \(\Gamma_{\text{Er-Host}}\) can be decreased by using a “quiet” host, meaning that magnetic field fluctuations inside the crystal caused by magnetic dipoles of the host species are small. The material \(Y_2\text{SiO}_5\) is a particularly good choice as none of the constituents are paramagnetic, and either have small nuclear magnetic moments or low natural abundance of isotopes which have a sizeable magnetic moment \([14]\).

At liquid helium temperatures with zero external magnetic field across the sample \(\Gamma_{\text{Er-Er}}\) gives the dominant contribution. With no magnetic field applied, the unpaired electron spin characteristic of a Kramers ion such as erbium is prone to flipping, as the spin-up and spin-down (with respect to some quantisation axis) states have the same energy. Applying a magnetic field breaks this degeneracy, but if the thermal energy \(k_B T\) is of a similar magnitude to the splitting \(\Delta E = g\mu_B B\) (\(g\) is the electron \(g\)-factor, \(\mu_B\) is the Bohr magneton and \(B\) is the applied field) then spin flips of \(\text{Er}^{3+}\) ions cause rapid decoherence.

This phonon-induced spin-flipping of erbium ions is the main source of decoherence, hence the main contribution to the homogeneous linewidth for \(\text{Er}^{3+}:Y_2\text{SiO}_5\). In order to reduce decoherence from phonon-induced spin flips it is necessary to maximise the energy level splitting of the Kramers doublet, and if possible decrease the temperature. Maximising the level splitting is complicated by the presence of two crystallographically inequivalent sites at which \(\text{Er}^{3+}\) ions can substitute into, and the low site symmetry of \(Y_2\text{SiO}_5\). The level splitting depends critically on the orientation of the applied magnetic field, and finding the right magnetic field direction to optimise the coherence time is a non-trivial task that has been carried out by Boettger et al. \([14]\). An illustration of Kramers degeneracy splitting for both the ground and excited state lowest crystal field levels is given in Figure 3.10 showing the new transitions that are then possible.

\[ \begin{align*}
4\text{I}_{13/2}^{(1)} & \quad \text{Δ}E = g_e\mu_B B \\
4\text{I}_{15/2}^{(1)} & \quad \text{Δ}E = g_\gamma\mu_B B
\end{align*} \]

Figure 3.10: Zeeman splitting to form Kramers doublets in ground and excited state lowest crystal field level, showing the four transitions between the states. The labelling convention (a, b, c, d) used here will be referred to later. This figure was created by the author, based on a figure from [27].

\(^7\)The only stable isotope of yttrium is \(^{89}\text{Y}\) which has a small nuclear magnetic moment of \(-0.137\mu_N\) (where \(\mu_N\) is the nuclear magneton, which is \(~\times 2000\) times smaller than the Bohr magneton). \(^{29}\text{Si}\) is the only stable isotope of silicon with nuclear spin and has \(-0.554\mu_N\) with \(4.7\%\) natural abundance and \(^{17}\text{O}\) has \(-1.89\mu_N\) nuclear magnetic moment but only \(0.04\%\) natural abundance \([125]\).

\(^8\)The decomposition of the homogeneous linewidth into distinct components is not as straightforward as indicated in equation 3.5; \(\Gamma_{\text{Er-Er}}\) is driven by phonon processes so could conceivably be included in \(\Gamma_{\text{phonon}}\). In this work we exclude phonon-induced erbium spin flips from \(\Gamma_{\text{phonon}}\) instead isolating them in \(\Gamma_{\text{Er-Er}}\).
3.4.1 Choosing a Magnetic Field Orientation

To best suppress decoherence from erbium ion spin flips the energy of the Zeeman splitting $\Delta E = g\mu_B B$ should be made as large as possible for a given magnetic field. To do this the magnetic field should be applied along the direction which gives the largest g-factor. The size of the g-factor as a function of the applied magnetic field direction can be described by a rank two tensor, which has been fully mapped out by Sun et al. [126].

The g-factor tensor is highly anisotropic, due to the low site symmetry in $Y_2SiO_5$. Erbium ions can substitute into two crystallographically distinct sites, both of which have $C_1$ (no) site symmetry [127]. The $Y_2SiO_5$ crystal has a monoclinic unit cell and belongs to the space group $C_{2h}^6$ (in Schönflies notation), with the $C_2$ axis (axis of two-fold rotation symmetry) often labelled as $b$ [126]. The other crystal axis are labelled $a$ and $c$, and are located in the mirror plane perpendicular to the $b$ axis. There are three optical extinction axes (axes along which the polarisation of light will be unchanged during propagation, called extinction axes because no light will be transmitted when observed through crossed polarisers), the first being the $b$ crystal axis, and two others called $D_1$ and $D_2$ which lie in the a-c plane, as shown in Figure 3.11. The right-handed coordinate system defined by $b$-$D_1$-$D_2$ is the natural choice in describing magnetic field orientation.

![Diagram](image)

**Figure 3.11:** Relationship between optical extinction axes and crystal axes. The optical extinction axes are mutually perpendicular, and make a natural choice of coordinates when describing magnetic field orientation. The $b$ axis (coming out of the page in 3.11b) is the third crystal and optical axis.

$Er^{3+}$ substitutes for $Y^{3+}$ directly without charge compensation, and the ions are roughly the same size which leads to narrow inhomogeneous linewidths, of the order $\sim 500 \text{ MHz}$ for $0.001\%$ atomic doping concentration. In a crystal the main inhomogeneous broadening mechanism is variations in strain at different optical centers, hence why a dopant ion of similar size to the original leads to relatively small inhomogeneous linewidths. For several applications, a larger inhomogeneous linewidth is desirable which can be achieved by co-doping erbium and magnetically inert europium. The addi-
tion of Eu\(^{3+}\) has been shown to significantly broaden the inhomogeneous linewidth (from \(\sim 3\) GHz in 0.02\%Er:Y\(_2\)SiO\(_5\) to 11 GHz in 0.02\%Er:1\%Eu:Y\(_2\)SiO\(_5\)) without negatively affecting the coherence properties [118].

The two crystallographically distinct sites have different optical transition frequencies and crystal field splittings. The \(^4I_{15/2}(1) \rightarrow ^4I_{13/2}(1)\) transition for site 1 has a longer population lifetime, and occurs at 1536.478 nm in zero magnetic field, whereas the transition for site 2 occurs at 1538.903 nm [126]. Site 1 has advantageous properties for information processing, hence this site shall be our focus.

Choosing the magnetic field direction that gives the longest coherence times is difficult for the following reasons. As stated above, single-phonon induced Er\(^{3+}\) ion spin flips are the main cause of decoherence in Er\(^{3+}\):Y\(_2\)SiO\(_5\) at cryogenic temperatures, and to minimise them for a fixed temperature the Zeeman splitting of the Kramers doublet should be made as large as possible. But there are two different crystallographic sites for the Er\(^{3+}\) ions, both of which can contribute to decoherence via spin flips [27]. To reiterate, even though the optical transition of site 2 is well away from the resonance of site 1, when ions in site 1 are excited spin flips of off-resonance Er\(^{3+}\) ions at site 2 affect the coherence time. Because of the low site symmetry, the magnetic g-tensors for site 1 and site 2 are different, and there is no direction that simultaneously maximises the g-value for both sites. Also, if Er\(^{3+}\) ions are excited, spin flips in excited state Kramers doublets also occur, and the g-tensor for the excited state is different again. Only the excited state of site 1 needs to be considered, as the transitions for site 2 are not resonant so the excited state is not populated.

So there are three different Zeeman splittings to consider, and there is no direction that maximises all of them simultaneously. Boettger et al. have done full orientational Zeeman spectroscopy to find the magnetic g-tensors for the ground and excited states of site 1 and site 2 [120, 125]. They found a preferred magnetic field direction, and managed to extend the coherence time in Er\(^{3+}\):Y\(_2\)SiO\(_5\) to 4.2 ms, corresponding to a 73 Hz homogeneous linewidth, the narrowest ever measured in a solid [27].

There are also two magnetically inequivalent sites that Er\(^{3+}\) ions can occupy on crystallographically equivalent sites, related by the crystals C\(_2\) (180°) rotation. This means that for some magnetic field orientations, the Kramers doublet for an ensemble of ions splits into four states (two from each magnetically inequivalent state), meaning that 8 transitions are possible between members of the ground state lowest crystal field level and the excited state lowest crystal field level. Fortunately, these two sites become magnetically equivalent when the magnetic field is applied in the D\(_1\)-D\(_2\) plane, which is advantageous as then more ions will participate in a given transition. Lower doping concentrations are then possible, which decreases \(\Gamma_{Er-Er}\). The variation in g-factor as a function of magnetic field direction in the D\(_1\)-D\(_2\) plane is shown in Figure 3.12 for site 2, and Figure 3.13 for site 1. Also shown are the transition frequencies between ground and excited state Kramers doublets, which were used to find the g-factors. The labelling convention (a line, b line, c line, d line) is the same as in Figure 3.10.
Figure 3.12: Variation in g-factor for the ground and excited state of crystallographic site 2 in Er$^{3+}$:Y$_2$SiO$_5$ as a function of angle in the D$_1$ D$_2$ plane. The frequency of different optical transitions between ground state and excited state Zeeman-split levels is also shown, from which the g-values were calculated. The same labelling convention for the transitions is used as in Figure 3.10. Figure taken from [27] with the permission of the author.
3.4 Suppressing Decoherence in Er$^{3+}$:Y$_2$SiO$_5$

Figure 3.13: Variations in g-factor for site 1, along with corresponding transition frequency data. If the effect of Er$^{3+}$ ions at site 2 were neglected, the optimal magnetic field orientation to extend coherence time would be where the maximum g-factor is for the site 1 ground and excited states, around 120°. The actual optimum range including the effects of site 2 is shown in the shaded band. Figure taken from [27] with the permission of the author.
From Figure 3.13 we can see that the optimal magnetic field orientation is in the D$_1$-D$_2$ plane, at $\sim$140-160° measured counterclockwise from the D$_1$ axis if you are looking down b. By applying a magnetic field of 7 T to a sample of 0.001% Er$^{3+}$:Y$_2$SiO$_5$ at 1.5 K, the optical homogeneous linewidth of 73 Hz mentioned above was observed [114], currently the narrowest in any solid state material.

3.5 Summary

In this section we have given a brief introduction to the optical properties of rare-earth ion doped crystals. Having been used for many years in such things as phosphors and gain media for solid-state lasers, rare-earth ions are emerging as a species with significant potential in quantum computing applications. Er$^{3+}$:Y$_2$SiO$_5$ in particular is a promising candidate, due to its potential for ultra-narrow linewidths and long coherence times. The $^4$I$_{15/2} \rightarrow ^4$I$_{13/2}$ transition in erbium also falls in the telecom wavelength band, which has extremely well developed technology for low-loss transmission. This would be beneficial in the construction of future quantum networks [2].

We have also discussed two-pulse and three-pulse photon echo spectroscopy, which are powerful methods for studying coherence in inhomogeneously broadened samples. We then covered techniques for optimising the coherence time in Er$^{3+}$:Y$_2$SiO$_5$, the main conclusion being that a preferential magnetic field direction exists for suppressing erbium environment ion spins flips, which are the major source of decoherence at liquid helium temperatures.

The study of rare-earth ions in solid state hosts is a vast and active research area; for a more thorough introduction the interested reader is referred to [19,115].
Chapter 4

Cavity Quantum Electrodynamics

Cavity quantum electrodynamics is a thriving research area, both from the perspective of practical device applications and the pursuit of new knowledge. Quantum electrodynamics (QED) describes the radiation field as an infinite set of modes, and interactions with matter are described by coupling between modes. Cavity QED allows the possibility of altering the properties of the field modes, which in turn can alter interactions with atoms and produce some counter-intuitive effects. For example, spontaneous emission is usually considered to be an irreversible process. The only reason it appears irreversible however is due to the fact that there is an infinite number of radiation modes a decaying atom can release a photon into. Reversing spontaneous emission would require a photon incident on the atom that was in the same field mode as a time-reversed version of the photon emitted by the atom. Because there are so many field modes it is highly unlikely an attempt to reverse the process will pick the correct radiation mode. It is in this sense that spontaneous emission is irreversible.

In cavity QED, spontaneous emission can be reversible. A cavity only allows certain modes inside it, which can have some curious consequences. If an atom is inside the cavity, and has a transition frequency that is not resonant with the cavity, the lifetime of the atom can be increased because it cannot emit into the cavity mode. The frequency that the atom “wants” to emit at is not allowed in the cavity. Conversely, if the atom were resonant with a cavity mode the spontaneous emission rate could be enhanced. This begs the question; how does the atom “know” it is not allowed to emit into certain modes before it actually emits? The answer lies in the mechanism that causes spontaneous emission \[9\]. In QED electric and magnetic field amplitude are quantum observables, with a given uncertainty relation. This means that in each field mode, electric and magnetic fields are not simultaneously zero, as this would violate the uncertainty relation. “Vacuum fluctuations” are variations of the electric and magnetic fields in each field mode \[128\]. These vacuum fluctuations are what stimulate an atom in its excited state to emit a photon. A cavity can enhance or inhibit vacuum fluctuations in certain modes. This in turn leads to enhanced or inhibited spontaneous emission rates for atoms in the cavity. Herein lies the magic of cavity QED; the interaction between atoms and different field modes can be manipulated, and the quantisation of the radiation field can be readily observed.
4.1 Strong Coupling

A simple model of a cavity QED system is given by the Jaynes-Cummings Hamiltonian [129]. This describes one two-state atom interacting with a single cavity mode, neglecting any dissipative effects such as cavity decay or spontaneous emission. Such dissipative effects are important in real cavity QED systems [130], however the Jaynes-Cummings model provides a good starting point from which to define important cavity QED parameters. The Jaynes-Cummings Hamiltonian is given by

\[ H = \hbar \omega_A \left( a^\dagger a + \frac{1}{2} \right) + \frac{1}{2} \hbar \omega_C \sigma_z + \hbar g(r) \left( a^\dagger \sigma + a \sigma^\dagger \right), \tag{4.1} \]

where \((\omega_A, \omega_C)\) are the atom and cavity resonance frequencies respectively, \(a^\dagger\) and \(a\) are creation and annihilation operators for the radiation field mode, \(\sigma_z\) is the Pauli operator and \(\sigma, \sigma^\dagger\) are atomic raising and lowering operators. The terms \(a^\dagger \sigma\) and \(a \sigma^\dagger\) act to lower the energy state of the atom and create a photon in the cavity or raise the atomic energy level destroying a photon in the cavity respectively. From equation 4.1 \(\hbar g(r)\) gives the energy of this atom-cavity coupling, hence \(g(r)\) is the atom-cavity coupling rate, given by

\[ g(r) = \left( \frac{\mu^2 \omega_C}{2 \hbar \epsilon_0 V_M} \right)^{\frac{1}{2}} U(r) \equiv g_0 U(r) \tag{4.2} \]

where \(\mu\) is the atomic dipole moment, \(V_M\) the cavity mode volume and \(U(r)\) describes the cavity mode function. The cavity mode volume is defined such that \(V_M = \int |U(r)|^2 d^3x\). The single-photon Rabi frequency is an important parameter defined as \(2g_0\), and gives the maximum rate at which one quantum of excitation oscillates between the atom and the cavity field [10].

If the cavity mode is resonant with the atom transition frequency, the eigenstates of the Jaynes-Cummings Hamiltonian are given by

\[ |\pm\rangle = \frac{1}{\sqrt{2}} \left( |g, n\rangle \pm |e, n-1\rangle \right). \tag{4.3} \]

where \(|g, n\rangle\) represents a state with the atom in the ground state and \(n\) photons in the cavity mode, and \(|e, n-1\rangle\) the atom in the excited state and \(n-1\) photons in the cavity mode. These eigenstates are interesting in that they represent entanglement between the atom and cavity field mode. Energy eigenvalues of this Hamiltonian are

\[ E_\pm = n \hbar \omega_C \pm \sqrt{n} \hbar g(r). \tag{4.4} \]

Of course in a real system dissipation will play a crucial role. The main sources of dissipation in cavity QED are loss of photons due to leakage from the cavity and spontaneous emission by the atom into radiation modes other than the cavity mode. Photon leakage from the cavity is parameterised by

\[^1\text{The atom-cavity coupling rate depends on the cavity mode function, a fact that was used by Jeff Kimble’s group to determine the path of a cesium atom dropped through a cavity. They monitored the coupling rate and using knowledge of the mode function were able to figure out the trajectory of an atom in the cavity, a system dubbed the “atom-cavity microscope” [131].}\]
the cavity decay rate, $\kappa = \frac{\pi c}{MQ}$, and spontaneous emission by the spontaneous emission rate $\gamma = 1/T_1$, where $T_1$ is the lifetime of the atomic transition.

Using high-Q cavities with small mode volumes, a regime can be reached in which the coherent atom-cavity coupling rate $g_0$ exceeds the dissipative rates in the system. This is the strong coupling regime, and allows coherent interaction between a single atom and a single photon. In this regime a wide variety of important quantum information processing operations become possible [3][4][132]. A schematic of the relevant rates involved is shown in Figure 4.1.

![Figure 4.1: Important rates involved for strong coupling cavity QED.](image)

In the strong coupling regime the eigenstates of the system resemble the Jaynes-Cummings eigenstates (equation 4.3). For a single quantum of excitation, resonance frequencies are given by

$$\omega = \omega_C \pm g(r)$$

from equation 4.4 with $n = 1$, corresponding to $|\pm\rangle_{n=1}$ eigenstates. This shows a two-peaked resonance for the coupled atom-cavity system, in contrast to the singly peaked resonance of the empty cavity.

For a strongly coupled cavity, the presence of a single atom changes the spectrum from the empty cavity to a double peak for weak excitation energies. Weak excitation energies are necessary because a single photon is enough to saturate an atom in the strong coupling regime, and applying intense excitation will wash out the peaked structure [133]. This double resonance peak is called vacuum Rabi splitting, and serves as a marker of strong coupling.

The criteria for strong coupling can be cast into a more useful form using two dimensionless parameters, describing how much of an effect single quanta of excitation have. The first is the saturation
photon number, which is the number of intra-cavity photons required to saturate a single intracavity atom. This is derived starting from the saturation intensity for an atomic transition \([134]\):

\[
I_{\text{sat}} = \frac{4\pi^2 \hbar c \gamma}{3\lambda^3} \tag{4.5}
\]

and the intensity for \(n\) photons inside a cavity mode volume \(V_M\)

\[
I_{\text{cavity}} = \frac{n\hbar \omega c}{V_M}. \tag{4.6}
\]

Equating these two and simplifying yields the saturation photon number \(n_0\),

\[
n_0 = \frac{2\pi \gamma V_M}{3c\lambda^2}. \tag{4.7}
\]

This can be simplified using equation \([4.2]\) for the cavity decay rate, and the fact that \(\gamma\) is equivalent to the Einstein \(A\) coefficient given by \([135]\)

\[
\gamma = A = \frac{\mu^2 \omega^3}{3\pi \varepsilon_0 \hbar c^2} \tag{4.8}
\]

which can be used to write

\[
g_0 = \sqrt{\frac{\mu^2 \omega}{2\varepsilon_0 \hbar V_M}} \tag{4.9}
\]

\[
= \sqrt{\frac{3c\gamma \lambda^2}{4\pi V_M}}. \tag{4.10}
\]

The saturation photon number can now be expressed compactly in terms of the atom-cavity coupling and the spontaneous decay rate as

\[
n_0 = \frac{\gamma^2}{g_0^2}, \tag{4.11}
\]

which is the desired result. For strong coupling \(n_0 < 1\), meaning that a single photon is enough to saturate an intracavity atom. This paves the way for study of non-linear optics at the single quantum level \([11]\).

The second dimensionless parameter for concisely expressing strong coupling is the critical atom number \(N_0\), which is the number of atoms that must be strongly coupled to the cavity to have an appreciable affect on the intracavity field, and is given by

\[
N_0 = \frac{2\kappa \gamma}{g_0^2}. \tag{4.12}
\]

The criterion for strong coupling in terms of these parameters is \((N_0, n_0) < 1\).
4.2 Strong Coupling to Rare-Earth Ions

The preceding formalism requires a few modifications for rare-earth ion dopants coupled to monolithic cavities. Achieving strong coupling would be a significant milestone for rare-earth ion quantum computing. It would allow the detection of single-dopants and single-state read-out, allowing single instance rare-earth quantum computing that is free from the scalability issues that plague ensemble based rare-earth quantum computing [18]. Strong coupling requires that the coherent atom-cavity coupling rate should dominate the dissipative rates in the system. This is problematic for rare-earth ions, which have low oscillator strengths and hence do not couple very strongly to the electromagnetic field [19], meaning that the atom-cavity coupling rate is not expected to be very high. Fortunately, weak coupling to the electromagnetic field also means long lifetimes for atomic transitions, hence low spontaneous emission rates. By looking at equations (4.8) and (4.9) it can be seen that the atom-cavity coupling rate scales linearly with the transition dipole moment, whereas the spontaneous emission rate scales as the transition dipole moment squared, so for small oscillator strengths (hence small transition dipole moments) the atom-cavity coupling can dominate the spontaneous emission rate.

The strong coupling regime is defined by \( N_0 = \frac{2\gamma}{\kappa g_0}, n_0 = \frac{\gamma^2}{\kappa g_0} \) < 1, which is possible to achieve with \( \kappa > g \gg \gamma \). In this case the atom-cavity coupling rate is smaller than the cavity decay rate \( \kappa \), but it is still possible to obtain \( (N_0, n_0) < 1 \), due to the rapid cavity leakage being compensated by very slow irreversible spontaneous emission. This is called the bad-cavity strong coupling regime (due to the dominance of the cavity decay rate) [26] and it is likely where a rare-earth ion strongly coupled to a resonator would operate. It has been shown that important quantum information processing operations are still possible in the bad-cavity strong coupling regime [21]. For a rare-earth ion coupled to a monolithic resonator, it is also necessary to adjust the expressions for the critical atom number and saturation photon number, as equations (4.11) and (4.12) assume that the coherence is lifetime limited, which may not be the case for a rare-earth ion in a solid surrounded by all sorts of other species that could cause dephasing.

For rare-earth ions in monolithic resonators, expressions for the saturation photon number and critical atom number become [26]

\[
\begin{align*}
n_0 &= \frac{\gamma h}{\kappa g_0}, \\
N_0 &= \frac{2\gamma h}{g_0},
\end{align*}
\]

(4.13) (4.14)

where \( \gamma_h \) is the homogeneous linewidth, and other parameters are as defined earlier. It is informative to re-write the critical atom number in terms of parameters associated with the resonator,

\[
N_0 \propto \frac{2h\gamma h V_M}{\mu^2 Q} \propto \frac{V_M}{Q}
\]

(4.15)
where we have used equation\ref{eq:4.9} to re-write $g_0$, and substituted in $\kappa = \frac{\pi c}{\lambda Q}$. $N_0$ is hence proportional to the cavity mode volume over the quality factor. The saturation photon number can be re-written in a similar manner, and it is seen that

$$n_0 \propto \frac{V_M}{Q_{atom}},$$  

(4.16)

where $Q_{atom}$ is the inverse of the population decay rate $\gamma$, and can be thought of as the “quality factor” of the optical transition. Both $N_0$ and $n_0$ are directly proportional to the homogeneous linewidth $\gamma_h$. Writing these expressions out illustrates most clearly what is required for strong coupling with rare-earth ions; cavities should have high quality factors and small mode volumes, and ions should have long population lifetimes and narrow homogeneous linewidths.

### 4.3 Summary

In this chapter we have introduced the concepts from cavity quantum electrodynamics necessary for this work. A definition of strong coupling has been given in terms of two parameters, the critical atom number $N_0$ and the saturation photon number $n_0$. The relationship of these parameters to other cavity QED parameters (such as cavity quality factor, atomic homogeneous linewidth) was outlined in section 4.2. In the next section fabrication procedures for manufacturing crystalline resonators are described, which were used to make the erbium-doped resonators studied in this work.
Chapter 5

Properties and Fabrication of Whispering-Gallery Mode Resonators

5.1 Introduction

Whispering-gallery mode resonators are named after the famous Whispering Gallery in Saint Paul’s cathedral, London, which allows a whisper spoken on one side of the gallery to be clearly audible on the other side. The correct explanation of this phenomenon was deduced by Lord Rayleigh using a whistle and his observations of a flickering candle across the room [136], and is due to acoustic waves travelling around the inside of the dome-shaped roof. The analogous optical effect is responsible for keeping light confined in a whispering-gallery mode (WGM) resonator. Dielectric WGM resonators are circularly symmetric about some axis, and the light propagates around the equator of the resonator confined by total internal reflection at the boundary [137]. In order to confine light the resonator must be made of a material that has a higher refractive index than its surroundings.

The first direct observation of optical WGM resonances was in liquid microdroplets suspended by optical levitation (a technique in which small particles can be suspended using radiation pressure from photons to counteract gravity) [138,139]. Sharp resonances in the levitation force were attributed to light coupling into surface waves in the dielectric microdroplets. Since then structures that can support WGM resonances have been the subject of much study, due in a large part to the development of efficient coupling techniques.

WGM resonators have many applications in optics and photonics, due to their potential for high quality (Q) factors, small mode volumes, and relative ease of fabrication [22]. They are also easily amendable to miniaturisation, something that poses significant technical problems with Fabry-Perot resonators. Because total internal reflection is to a large extent independent of wavelength (wavelength dependence is only through the dispersive properties of the mediums of either side of the boundary), WGM resonators are also broadband [22], unlike the best Fabry-Perot resonators which use Bragg reflector mirrors coated for a specified wavelength range.
5.2 Mode Structure

The mode structure for real WGM resonators is in general very complex. Exact solutions are known for the case of perfectly spherical resonators, however crystalline resonators often have a spheroidal shape\(^1\). That being said, it is beneficial to study spherical resonators as they provide a foundation for the study of real resonators.

Solutions for the electric and magnetic fields in a spherical dielectric structure have been known for many years now [140]. Modes can be described using three mode numbers \((n, l, m)\), which are loosely analogous to the radial, angular and magnetic quantum numbers that may be familiar to the reader from the study of the hydrogen atom in elementary quantum mechanics. The mode number \(n\) determines how many zeros occur in the radial direction, and \(l\) is the number of wavelengths occurring in a round trip of the resonator. For modes propagating near the surface of the dielectric, the resonance condition is \(d = 2\pi R/\lambda \approx \ell/n_r\), where \(n_r\) is the refractive index of the dielectric supporting the WGMs. Taking the expression \(p = \hbar k = \hbar 2\pi n_r/\lambda\) for photon momentum, for WGM modes propagating tangentially to the surface the photon angular momentum is given by \(L \approx pR = 2\pi R\hbar n_r/\lambda = \hbar \ell\), hence \(l\) is identified as the photon angular momentum, and \(m\) its projection onto the polar axis. This is shown in Figure 5.1. Modes with large angular momentum \(l\) are a long way from the center of the sphere, confined near the surface of the dielectric. There are a zoo of modes propagating at or near the surface of the dielectric which could reasonably be called WG modes, and in this thesis we take WGMs to be modes with large \(l \sim m\) and small \(n \sim 1\).

**Figure 5.1:** Whispering gallery modes in a sphere. (a) shows the confinement mechanism, with the angle \((i)\) being the total internal reflection angle, \(i \approx \pi/2\) for WG modes. (b) shows the angular momentum \(L\) associated with a WG modes, as well as \(M\) its projection onto the polar axis (\(\ell\) and \(m\) in our notation). Image reproduced from [141].

\(^1\) A spheroid is simply what you would get if you compressed a sphere in one direction; an oblate spheroid is compressed in the vertical direction, and a prolate in the horizontal.
The normalised electric field mode function for a transverse magnetic (TM) mode inside the sphere for large $\ell$ and $\ell = m$ is given by [142]

$$\vec{\Psi}_{in}(r, \theta, \phi) = N(\ell + 1) \frac{j_\ell(kr)}{kr} \sin \ell \theta e^{i\ell \phi} + NF(r) \cos \theta \sin^{\ell - 1} \theta e^{i\ell \phi}$$

$$+ iNF(r) \sin^{\ell - 1} \theta e^{i\ell \phi}$$

(5.1)

inside the resonator and

$$\vec{\Psi}_{out}(r, \theta, \phi) = NB(\ell + 1) \frac{h_\ell^{(1)}(kr)}{kr} \sin \ell \theta e^{i\ell \phi} + NBH(r) \cos \theta \sin^{\ell - 1} \theta e^{i\ell \phi}$$

$$+ iNBH(r) \sin^{\ell - 1} \theta e^{i\ell \phi}$$

(5.2)

outside, where

$$F(r) = \frac{j_\ell(kr)}{kr} + \frac{\ell}{2\ell + 1} j_\ell(kr) - \frac{\ell + 1}{2\ell + 1} j_{\ell + 1}(kr)$$

(5.3)

$$H(r) = \frac{h_\ell^{(1)}(kr)}{kr} + \frac{\ell}{2\ell + 1} h_\ell^{(1)}(kr) - \frac{\ell + 1}{2\ell + 1} h_{\ell + 1}^{(1)}(kr)$$

(5.4)

$$B = \frac{F(R)}{H(R)}$$

(5.5)

In these equations $N$ is the normalisation factor required to give the mode function a maximum value of one, $j_\ell$ are spherical Bessel function of the first kind, $h_\ell^{(1)}$ are spherical Hankel function of the first kind, $k$ is the wavevector inside a sphere of refractive index $n_r$ and $\ell$ and $m$ are the polar and azimuthal mode numbers respectively. The dependence of these modes on the principle mode number $n$ is implicit in $k$. The transverse electric (TE) equations have a similar form [142].

These equations for the mode function are particularly un-illuminating written out like this, so it is helpful to plot important features to gain a more intuitive feel for them. Figure 5.2 shows radial mode structure for different values of $n$. In these diagrams, $\ell = m$ and $\theta, \phi$ are held fixed at $\pi/2$ and 0 respectively. We see that for $n = 1$ the electric field is concentrated very close to the surface of the resonator, and for higher $n$ there are more lobes and the “centre of mass” of the mode is pulled inwards. The evanescent field is negligible for millimetre-sized resonators, only making a significant contribution to the mode volume for $R \approx 10\, \mu m$. The evanescent field is however still of vital importance for coupling light into the resonator.

Modes with $\ell = m$ have a single lobe in the polar direction, whereas modes with $\ell \neq m$ have multiple lobes. Multi-lobe $\ell \neq m$ modes can be thought of as precessing around with a slight inclination to the horizontal plane [143].

For a perfectly spherical resonator, modes with different values of $\ell - m$ are degenerate. This degeneracy is broken in a spheroidal resonator. If the resonator deviates only slightly from a perfect sphere the degeneracy is weakly broken and a very dense mode spectrum results due to the slight
frequency spreading of a vast number of modes. This is desirable for some applications, for example a white-light resonator has been manufactured in which the mode spectrum is so dense that is looks continuous \[144\]. In the pursuit of strong coupling it is beneficial to couple into a fundamental \((n = 1, l = m)\) mode as this has the smallest mode volume. Resonators have been created which are truly single-mode, in that they support only one mode for each polarisation per free spectral range \[145, 146\]. Such engineering of the resonator spectrum was not possible using our fabrication procedure, however it bodes well for future applications that it has been shown to be possible.

A useful approximation for the mode volume of \(n = 1\) modes is given by

\[
V_M \simeq 3.4\pi^{3/2} \left( \frac{\lambda}{2\pi n_e} \right)^3 l^{11/6} \sqrt{2(l - m) + 1},
\]

which is accurate for resonators larger than \(\sim 100 \mu\text{m}\) in diameter \[146\]. This approximation is very useful and circumvents the need for numerical integration to accurately find mode volumes.


5.3 Quality Factor

Quality factor (commonly just referred to as Q) is a dimensionless parameter useful in a wide range of oscillator systems. It is defined as the energy stored divided by the amount of energy dissipated per cycle, given by

\[ Q = \frac{2\pi E_{\text{stored}}}{E_{\text{dissipated}}}. \]

For optical resonators a cycle refers to one electromagnetic field oscillation rather than a round-trip cycle. In a high-Q resonator many oscillations will occur before the electromagnetic field dies away. Equivalently a photon in a high-Q resonator will circulate in the cavity for a relatively long time. This means that high Q-factors are essential in trying to reach the strong coupling regime of cavity QED, as a long photon lifetime increases the effective interaction cross-section with ions in the resonator.

High-Q resonators should thus dissipate as little light circulating in the cavity field mode as possible. Multiple different factors effect the Q-factor in WGM resonators, which add like resistors in parallel to give the overall Q-factor,

\[ \frac{1}{Q} = \frac{1}{Q_{\text{abs}}} + \frac{1}{Q_{\text{rad}}} + \frac{1}{Q_{\text{surf}}} + \frac{1}{Q_{\text{water}}}. \tag{5.7} \]

\( Q_{\text{abs}} \) is due to absorption in the material from which the monolithic WGM resonator is made out of. \( Q_{\text{rad}} \) is radiative decay due to imperfect total internal reflection from a curved surface, \( Q_{\text{surf}} \) is due to scattering from imperfections on the resonator surface and \( Q_{\text{water}} \) is due to water absorption from atmospheric water adsorbing to the resonator surface or diffusing into the material.

\( Q_{\text{rad}} \) depends upon the radius of the resonator and the refractive index contrast between the resonator material and its surroundings. It increases exponentially for increasing resonator size, and for \( D/\lambda \geq 15 \) (where \( D \) is the resonator diameter and \( \lambda \) is the wavelength of circulating light) \( Q_{\text{rad}} > 10^{11} \) \[147\]. A Q of \( 10^8 \) would be very respectable for our purposes, so for resonators larger than \( \sim 15 \mu m \) the Q-factor is not limited by \( Q_{\text{rad}} \). For millimetre-sized resonators dissipation from imperfect total internal reflection is utterly negligible.\[2\]

\( Q_{\text{water}} \) is important in amorphous solids where atmospheric water can diffuse into the sample and act as an extra absorber \[147\]. However this diffusion does not occur in our crystals, and \( Q_{\text{water}} \) is not an issue \[26\]. \( Q_{\text{surf}} \) is due to light scattering from scratches and other defects on the surface of the resonator. An approximate expression for the surface scattering has been derived, given by \[134\]

\[ Q_{\text{surf}} \sim \frac{3\varepsilon(\varepsilon + 2)^2}{(4\pi)^3(\varepsilon - 1)^{5/2}} \frac{\lambda^{7/2}(2R)^{1/2}}{(\sigma B)^2}, \tag{5.8} \]

where \( \varepsilon = n^2 \) is the dielectric constant of the resonator material, \( R \) is the resonator radius and \( \sigma B \) is determined by the size and correlation length of surface inhomogeneities. Polishing of the resonator surface will decrease both the size and correlation length of surface defects, and manufacturing WGM resonators focuses on improving the surface quality. It is also seen that \( Q_{\text{surf}} \) is proportional

\[ ^2\text{For a } 100 \mu m \text{ fused silica microsphere resonator } Q_{\text{rad}} = 2.51 \times 10^{57} \tag{141}, \text{ and for a } Y_2SiO_5 \text{ resonator of } 2 \text{ mm diameter would be far, far larger than this.} \]
to \( \lambda_0^{7/2} \) so should be larger for longer wavelengths, so working with the \( ^4I_{15/2}(1) \rightarrow ^4I_{13/2}(1) \) telecom wavelength transition at 1536 nm in \( \text{Er}^{3+}:Y_2\text{SiO}_5 \) should have benefits over using the 606 nm transition in \( \text{Pr}^{3+}:Y_2\text{SiO}_5 \).

\( Q_{\text{abs}} \) puts an upper limit on resonator quality factor for a given material, as the absorption is an intrinsic property. It is given by

\[
Q_{\text{abs}} = \frac{2\pi n_r}{\alpha(\lambda)\lambda_0},
\]

where \( \alpha \) is the absorption coefficient for the material at wavelength \( \lambda_0 \). Projections for maximum possible Q-factor in several transparent crystals have been made by Savchenkov et al., calculated from models and based on what little experimental data is available in these materials \[28\]. They find a common trend of higher possible Q-factor at 1536 nm over 606 nm. This one major motivation for us in using \( \text{Er}^{3+}:Y_2\text{SiO}_5 \) to improve on studies done in \( \text{Pr}^{3+}:Y_2\text{SiO}_5 \)\[148\], as it was suspected that \( Y_2\text{SiO}_5 \) might share this trend. However, we observed similar Q-factors ranging from \( 1 \times 10^6 \) to \( 1 \times 10^7 \) in our resonators at both these wavelengths. The same resonator should have less surface scattering at 1536 nm, so this implies that perhaps \( Y_2\text{SiO}_5 \) is more opaque than we thought at 1536 nm. Undoped \( Y_2\text{SiO}_5 \) WGM resonators are in the process of being fabricated, the study of which should answer questions about absorption as a function of wavelength in \( Y_2\text{SiO}_5 \).

### 5.4 Fabrication of Crystalline Resonators

WGM mode resonators have been made from a variety of different materials. Early WGM resonators were fabricated by melting fused silica, and allowing surface tension forces to shape a smooth optical surface. This is appealing in that close to atomically smooth surfaces can be made with a relatively straightforward melting and cooling procedure. However many potential applications of WGM resonators benefit greatly from using a crystalline resonator \[149, 150, 151\], for which melting would disrupt the crystal lattice, causing scattering from different crystal domains and low Q-factors. For our purposes, crystals are essential due to the comparatively poor coherence properties of rare-earth ions in amorphous solids, which was mentioned in chapter 5. Because of this, crystalline WGM resonators are fabricated using mechanical grinding and polishing techniques. These techniques have progressed to the point that the highest Q resonators are crystalline resonators rather than amorphous ones created by surface tension reshaping. The current record is a CaF\(_2\) resonator with a Q of \( \sim 10^{11} \) \[152\].

Recent fabrication methods in crystalline resonators have actually used a procedure which melts a thin layer on the outer surface of the resonator, then gradually decreases the temperature such that the outer layer re-forms with the same crystal structure, seeded by the bulk of the inner crystal \[153, 154, 155\]. These methods are not very well developed, but show significant promise.

In our experiments, resonators were fabricated from monolithic disks of rare-earth ion doped crystals. The majority of experiments were performed on \( \text{Er}^{3+}:Y_2\text{SiO}_5 \) resonators, with Q factors of the order \( 10^6 \). Resonators were fabricated using a multi-stage procedure, with the \( \text{Er}^{3+}:Y_2\text{SiO}_5 \) disk first being cut to the correct approximate shape in a lathe with a fixed diamond-tip tool bit,
then a finer shaping stage using diamond lapping film, before being polished with a sequence of diamond suspensions of decreasing grit size. The resonator was checked intermittently for scratches and abrasions with an optical microscope.

To begin the first stage, 4 mm diameter disks of Er$^{3+}$:Y$_2$SiO$_5$ purchased from Scientific Materials Corporation were glued onto 2 mm diameter aluminium post using Selleys Araldite two-part epoxy, and left to cure until maximum bond strength was achieved. Initially a thermally conductive epoxy was considered to provide heat transfer for when the resonator was placed in the cryostat, however this was found to be too weak to keep the crystal disk in place during the manufacturing procedure. The glued sample was then held by the aluminium post in a bench-top mini-lathe with home-made vibration isolation rotating at speeds of up to 1500 rpm, and cut using a stationary diamond-tip tool bit to the shape (2) shown in Figure 5.3. The sample was then shaped to give the desired curvature ((3) in Figure 5.3) using diamond lapping film, with grit sizes decreasing from 30 $\mu$m to 6 $\mu$m to 1 $\mu$m to give progressively finer shaping. The sample was then transferred to an air tool for polishing, which provided rotation speeds of up to 60,000 rpm. Polishing was done with diamond suspensions of decreasing particle size, in the order 3000, 1000, 500, 250, 100, 50 nm. Lint free tissue paper was covered in the suspension and used to polish the resonator. After polishing thoroughly with each grit size the resonator should be left only with abrasions smaller than the diamond particle size. The resonator was cleaned using methanol and distilled water between each polishing step of decreasing particle size. This was especially important, as if contaminant particles of larger grit size are still present they will limit the possible surface quality.

The resonator was periodically checked with an optical microscope to ensure surface quality and the absence of obvious fractures in the crystal.

An example of a completed resonator is shown in Figure 5.4.
Figure 5.3: Steps involved in manufacturing a crystalline resonator. The first step is done using a fixed diamond drill bit to cut the disk to approximately the right shape, then finer shaping is done with diamond lapping film. Diagram based on a figure from [26].
5.4 Fabrication of Crystalline Resonators

Figure 5.4: Completed resonator, fabricated using the techniques described in the text.

It should be noted that after polishing below \( \sim 250 \) nm diamond grit size abrasions will no longer be resolvable optically, as they will be smaller than diffraction limited microscopy can resolve. Electron microscopy must then be used if one wishes to check the surface quality of a resonator, something not done in this work, but should be done in the future.

5.4.1 Determining Resonator Shape

For efficient coupling to WGM resonators, it is important to know the radius of resonator curvature in both the \( \hat{x} \) and \( \hat{z} \) directions. The radius of curvature in the \( \hat{x} \) direction is simply the radius of the resonator (\( R_x \) in Figure 5.3), and can be measured directly using calipers. The radius of curvature in the \( \hat{z} \) direction \( R_z \) is slightly more complicated and is measured using a Newton’s rings type diffraction experiment. \( R_z \) is the minor axis of an ellipse drawn in the vertical plane through the resonator, as shown in Figure 5.3, and determines the shape of the curved edge of the resonator. For example, a perfect sphere would have \( R_x = R_z \). Decreasing \( R_z \) to form an oblate spheroid breaks the frequency degeneracy of modes with different values of \( m \) which is important when trying to couple into a fundamental mode.

In order to find the ratio of \( R_z \) to \( R_x \), a microscope slide is placed on top of a resonator, which is illuminated from below by a monochromatic sodium lamp as shown in Figure 5.5. This setup gives interference rings in the transmitted light, which is focused by a microscope objective and viewed by imaging onto a CCD camera. The interference rings are caused by constructive and destructive interference between the surface of the resonator and the microscope cover slip. If \( d \) is an integer number of illumination wavelengths, then light bouncing backwards and forwards between the cover slip and the resonator surface by Fresnel reflection will constructively interfere, and destructively if \( d \) is a half integer multiple. This results in rings of constant resonator-cover slip distance \( d \) in the transmitted light, a contour ring interference pattern. When viewed in two dimensions, the contours may be elliptical if \( R_x \neq R_z \) which can be used to determine the ratio \( \frac{R_z}{R_x} \).
The radius of an interference ring for some \( d \) is given by

\[
X_\beta = R_\beta - (R_\beta - d)^2 \\
\approx 2R_\beta d \quad (R_\beta \gg d),
\]

(5.9)

where \( R_\beta \) denotes either \( R_x \) or \( R_z \). Using equation 5.9 for both and equating for the same \( d \) gives

\[
R_z = R_x \left( \frac{X_z}{X_x} \right)^2,
\]

(5.10)

which is used to find \( R_z \) after measuring \( R_x \). An example of such an interference pattern is shown in Figure 5.6. By taking the ratio of \( X_x \) to \( X_z \) on the same contour line the desired curvatures can be obtained using equation 5.10.

**Figure 5.5:** Setup for diffraction experiment to determine resonator shape. Based on a figure from [26].
5.5 Coupling to Resonators

Optical whispering gallery mode resonators have been known about for close to 50 years, however the study of their properties has only become an active research area in the last 15 years. This is due in large part to the development of efficient coupling techniques. For high-Q resonators of more than a few micrometers in size, the amount of radiative decay is so tiny that the reciprocal process of coupling light in using free space optics is prohibitively inefficient [141]. Instead near-field techniques based on evanescent field coupling have been developed which can give very efficient coupling. Efficiencies of greater than 99.97% have been achieved using tapered fibers [23] [156]. In this thesis, frustrated total internal reflection from the face of a prism was used for coupling. This is because the refractive index of the near-field coupler must be greater than that of the resonator for effective coupling, and our Er\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} resonators had a refractive index of \(\sim 1.8\), which is larger than conventional silica optical fibers (\(n_r \sim 1.5\)). High refractive index prisms (\(n_r \sim 2.2\)) were already available in the lab. Another reason is that it is much easier to put a prism in our cryostat than to try and arrange an optical fiber feed-through.

Coupling is done by reflecting laser light off the inner surface of the prism (in this work a cubic zirconia prism with a refractive index of 2.17 was used). If the surface of the coupling prism is brought close to the resonator (close being within a few wavelengths of the coupling beam), the

Figure 5.6: Interference pattern created as outlined above. Each ring (of either constructive or destructive interference) defines a contour of constant distance \(d\) between the resonator and transparent cover slip, which is equivalent to contour lines on the resonator as the cover slip is placed flat. The ratios of minor to major axis on the elliptical interference rings are used to find \(R_x\) and \(R_z\) in Figure 5.3(3).
evanescent field of the prism can couple light into resonator modes. This is called frustrated total internal reflection, shown graphically in Figure 5.7.

![Figure 5.7: Prism coupling schematic. The diagram as shown can be thought of as a slice through the coupling plane containing the prism and the equator of the resonator.](image)

There are three important parameters that can be altered to obtain optimal coupling [157]. The first is the angle of incidence on the prism surface, shown by $\Phi_0$ in Figure 5.7. The second is the location of the beam waist, which should occur at the reflection surface of the prism. The third is the spot size at the beam profile.

To couple into a fundamental $(n = 1, l = m)$ WGM mode the incidence angle in the coupling plane should be [157]

$$\Phi_0 = \sin^{-1}\left(\frac{l}{n_p k R_x}\right),$$

(5.11)

and the angle in the perpendicular plane should be $\Theta = 0$. In Figure 5.7 $\Theta$ would be the angle in the plane perpendicular to that shown.

Ideally, the angular spot size at the face of the prism should be [157]

$$\Delta \Phi^2 = \frac{\sqrt{n_p^2 - 1}}{n_p^2 k R_x \cos^2 \Phi_0},$$

(5.12)

\footnote{It should be noted that $n_p$ in these equations is the refractive index of the coupling prism, whereas $n_r$ is the refractive index of the resonator. These subscripts serve to distinguish these quantities from the mode number $n$.}
in the plane shown in Figure 5.7, and

\[ \Delta \Theta^2 = \frac{n_p + \sqrt{n_p^2 - 1}}{n_p^2 k R_z} \]  

(5.13)

in the plane perpendicular. This implies an elliptical beam profile for a spheroidal resonator, with different waists in the \( R_x \) and \( R_z \) directions,

\[ w_x = \frac{2\lambda}{\pi \Delta \Phi} \]  

(5.14)

\[ w_z = \frac{2\lambda}{\pi \Delta \Theta} \]  

(5.15)

Intuitively the coupling beam can be thought of as matching the shape of the resonator surface (thus having the same shape as the interference rings in Figure 5.6), however in this work we simply used Gaussian beams due to their natural occurrence in Fabry-Perot laser cavities. The Gaussian beam waist was chosen to be the average of \( w_x \) and \( w_z \). For the \( \text{Er}^{3+}:\text{Y}_2\text{SiO}_5 \) resonator used to obtain the results of this thesis, \( w_x = 84.8 \mu m \) and \( w_z = 93.1 \mu m \) and a Gaussian beam waist of \( w_r \sim 90 \mu m \) was used for coupling. A lens pair was used to get the correct beam waist at the surface of the prism. Matlab code kindly provided by previous student David McAuslan calculated correct lens spacings by propagating a Gaussian beam within the paraxial approximation. The beam waist evolution as a function of propagation distance calculated using this code is shown in Figure 5.8. Using the initial beam waist, desired beam waist and focal lengths of the lens pair as inputs, the lens spacing required to give the optimal spot size can be found.

To observe coupling once all the elements are in place the laser wavelength is scanned in frequency. For our Koheras Adjustik this was done using a built-in piezo element allowing scans of \( \sim \)2.5 GHz. The piezo scan was calibrated by putting side-bands on the laser frequency with an electro-optic modulator (EOM). The laser was then passed through an \( \text{Er}^{3+}:\text{Y}_2\text{SiO}_5 \) crystal at 3.5 K. The laser frequency was held on the erbium resonance for 10 \( \mu s \), then scanned rapidly using the piezo. This frequency sweep was done at 1 kHz, which due to the long optical lifetime of erbium (\( \sim 11.4 \) ms) meant that ions excited during the initial 10 \( \mu s \) remained in the upper state during the sweep. Because of this, regions of decreased absorption (spectral holes) will be observed at the initial laser frequencies (centre frequency plus side-bands). This is shown in Figure 5.10. The spacing between centre frequency and side-bands is very well known, as it is simply the input frequency to the EOM. This thus provides a method for putting known frequency markers on the unknown piezo scan width, and the piezo scan can be calibrated.

As the laser scans across a cavity resonant frequency, a dip in transmission is observed in the outcoupled light. This is shown in Figure 5.9 data which was taken on an inverting photodetector so the dips in transmitted intensity corresponding to modes are upwards “dips”.

5.5 Coupling to Resonators
Figure 5.8: Evolution of Gaussian beam width as a function of propagating distance, calculated using code provided by Dr David McAuslan. In this example a lens pair with $f = 50, 100$ mm is used to give a $90 \, \mu$m beam waist. Changing the focal lengths or initial beam width can move the beam waist of interest to a convenient location. Because our coupling prism was inside a cryostat, it was important to have a distance of 20-30 mm between the second lens and the beam waist.

Figure 5.9: Mode structure observed in transmitted light intensity as laser frequency is scanned.
Much like coupling light into an optical fiber, the most difficult part of prism coupling is getting light in initially. Once this is achieved, coupling can be tweaked and optimised. Obtaining this first coupling is a non-trivial challenge using infrared (IR) light at 1536 nm, outside the range of human vision and beyond silicon detectors. Placing the resonator inside an evacuated cryostat also means that fluorescent IR cards cannot be used for alignment. For this reason, red light from a Coherent 699 tunable dye laser was spatially overlapped with the 1536 nm light for alignment purposes. The bench setup used for coupling is shown in Figure 5.11.
Figure 5.11: Setup used for infrared (1536 nm) coupling to a resonator inside the cryostat. A beam from a tunable Coherent 699 dye laser operating at 606 nm is overlapped with the IR beam for alignment purposes. The overlap optic reflects most of the light at 1536 nm and transmits most at 606 nm. Neither the IR CCD camera nor the IR photodetector registered the presence of the visible light. The lens pairs are mounted on translation stages so their spacing can be adjusted. LP - linear polariser, HWP - half-wave plate, SM fiber - single-mode fiber.
The visible beam is passed through its own lenses as the usual procedure was to find visible coupling first, then tweak the parameters of the IR coupling setup. This was done because visible coupling is relatively easy to achieve compared to IR. The beam path into the cryostat required for coupling is similar for both visible and IR, hence coupling in the visible first meant a much smaller parameter space to explore when it came to finding coupling in the IR. Coupling was usually detected in the first instance as flickering in the outcoupled light on an Edmund Optics phosphor-coated CCD camera for 1460-1600 nm viewing. Optimisation was then done whilst monitoring the signal from an InGaAs photodiode, which gave information about the coupling efficiency. Maximum efficiencies of ~25% were obtained using this method. Piezoelectric nanopositioning devices were used to bring the prism within evanescent coupling distance of the resonator. The full prism-resonator mount is shown in Figure 6.1. A long working distance microscope was used to check how close the prism was to the resonator if their proximity was unknown. Crashing the prism into the resonator at times was unavoidable, but care was taken where possible to avoid it due to the potential for scratching the resonator.

It is possible to determine if you are coupled into a fundamental mode by observing the shape of outcoupled light. This can be done by placing a second prism in close proximity to the resonator, which introduces another out-coupling path. This spatially separates the out-coupled light from the light simply reflected from the first prism without coupling into the resonator, which for our coupling efficiencies was always significantly more intense. The outcoupled shape can then be observed directly without the intense background caused by reflected light. Examples for visible coupling are shown in Figure 5.12.

(a) Single lobe structure indicating a fundamental mode.

(b) Double lobe means we are coupled to some higher order mode.

Figure 5.12: Outcoupled beam profiles for visible coupling.
Unfortunately observing outcoupled light for IR coupling was more difficult due to the lack of a beam profilometer operating around 1536 nm. Attempts were made to determine the beam profile by imaging on the IR CCD camera, however it proved difficult to determine the beam shape with any sort of confidence. Hence experiments carried out in this thesis were done on modes of unknown order, not necessarily the fundamental. This is something that will definitely need to be improved upon in future work.

5.6 Summary

In this chapter we have provided an introduction to the whispering-gallery mode resonators manufactured in this thesis. Modes structure and prism coupling methods have also been briefly discussed.

We manufactured crystalline whispering-gallery mode resonators, due to the superior coherence properties of rare-earth ions in a crystalline environment over a disordered one. Procedures for manufacturing crystalline WGM resonators are now at the point where quality factors can exceed those of amorphous resonators made by surface tension reshaping [152]. The upper limit on quality factor is determined by bulk absorption in the resonator material. We thought that this might be less at 1536 nm than at the 606 nm used for Pr$^{3+}$:Y$_2$SiO$_5$ resonators in previous work [26], due to a common trend among other crystals. However, some of our observations suggest that this might not be the case. Un-doped Y$_2$SiO$_5$ are currently being fabricated for a more comprehensive investigation.
Chapter 6

Experiments and Results

In this chapter we describe experiments done on millimeter-sized $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$ resonators. Experiments were done on a 0.001% spheroidal resonator of radius 1.6 mm manufactured using the techniques described in chapter 5. Two-pulse photon echo spectroscopy was done in order to probe the coherence time of ions in the resonator, and data was also taken in a cubic sample of $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$ that was not subjected to any grinding or polishing (which we refer to as the bulk sample). The bulk sample provided a reference to check that the machining process had not ruined the coherence properties of the resonator ions. Three-pulse photon echo experiments were carried out with the initial intention of measuring the lifetime of the $^4I_{15/2} \rightarrow ^4I_{13/2}$ transition, however it was discovered that this was not possible due to significant spectral diffusion in our sample. We instead suggest possible spectral diffusion mechanisms based on our observations. Finally experiments were done to measure the length of a $\pi$-pulse for ions in the resonator, which was used to determine the atom-cavity coupling and calculate cavity QED parameters. The feasibility of reaching the strong coupling regime with $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$ WGM resonators is then discussed based on these results.

Data was taken on the $^4I_{15/2}(1) \rightarrow ^4I_{13/2}(1)$ transition between lowest crystal field levels in $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$, which occurs at 1536.478 nm in vacuum at zero magnetic field. This transition happens for ions in crystallographic site 1 using the convention of chapter 3 [127].

6.1 Experimental Setup

6.1.1 Resonator Mount

The resonator was mounted in a vibration-isolated S2 Corporation Cryomech PT405 two-stage cryostat with customised sample space, which was evacuated using a Pfeiffer TurboCube vacuum pump and cooled to 3.5 K. Four windows coated for optimal transmission around 1500 nm provided optical access to the sample space. Temperature was monitored using a Lakeshore 331 temperature controller reading from two silicon diode sensors mounted at the second stage cold finger and the sample mount. The operating temperature of 3.5 K is taken from the diode at the sample mount. Also mounted was a $5 \times 5 \times 5$ mm 0.001% $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$ crystal, a prism for near-field coupling to resonators and a neodymium rare-earth permanent magnet to provide Zeeman splitting of the Kramers degeneracy. The cubic crystal was mounted so that properties of erbium ions could be compared with
ions in the resonator, as there was some concern that the resonator manufacturing process might have damaged the crystal structure near the surface, which is exactly where the WGM modes propagate. The magnetic field strength was measured as 0.3 T and varied by $\sim 20\%$ spatially across the resonator.

The full mount that went in the cryostat is shown in Figure 6.1. Multiple factors were considered when designing this setup. Firstly, in order for the ions to have appreciable coherence times the samples needed to be close to liquid helium temperatures. This meant that the thermal connection between the cryostat cold head and the rare-earth samples was very important. The mount was constructed from aluminium, which has reasonable thermal conductivity at liquid helium temperatures [158] as well as being easy to machine. Apezion N vacuum grease was smeared between connecting surfaces to improve thermal contact. Protruding cold fingers from the aluminium served the dual purpose of holding the resonator and bulk sample in place, as well as increasing thermal contact area. This is illustrated in Figure 6.2, which shows a stripped down mount without the coupling prism obscuring the view.

The magnet was mounted vertically as shown partly due to tighter space constraints in the cryostat sample space in the horizontal direction to the vertical. This mounting also causes the magnetic field to differ across the resonator, being larger on the side of the resonator closest to the magnet and marginally smaller on the far side. This causes a sort of spatial inhomogeneous broadening, as ions on one side of the resonator will experience a different Zeeman splitting to those on the other. If the magnet was mounted such that its rotation axis coincided with that of the resonator, then ions sampled by the light as it circulated around the equator of the resonator would all have the same Zeeman shift, and this form of inhomogeneous broadening would not occur, as shown in Figure 6.2.

The reason this spatial inhomogeneous broadening is desirable is due to the narrow inhomogeneous linewidth of the $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$ samples used at cryogenic temperatures, of the order $\sim 500$ MHz. The free spectral range of the resonator is $\sim 15$ GHz, so with this narrow inhomogeneous linewidth and the lack of control over mode structure in our manufacturing procedure it is possible that there might be no cavity modes which overlap with the $\text{Er}^{3+}$ resonance. This would be troublesome for cavity QED experiments. Mounting the magnet as described increased the effective inhomogeneous linewidth from $\sim 500$ MHz to $\sim 2.5$ GHz. This increases the fraction of a free spectral range the $\text{Er}^{3+}$ resonance covers from $\sim 3\%$ to $\sim 17\%$, a worthwhile improvement given that this arrangement also made best use of the available space.
Figure 6.1: Mount that was placed in cryostat. The view in this image is along the axis a laser beam would propagate during experiments. For experiments in the bulk sample the laser beam would propagate straight through whereas for resonator experiments the beam would come out at right angles after reflecting from the out-coupling mirror. The resonator is just visible behind the coupling prism. Inside the cryostat this mount would be hanging down with the top part attached to the cold head. Most of the mount is made from aluminium (with the exception of the Attocubes) due to ease of machining and reasonably good thermal conductivity. Also using the same material for connected components alleviates issues of unequal thermal contraction when cooling down to cryogenic temperatures. Apezion N vacuum grease was smeared between surfaces joined together to increase thermal transport across joins.
Figure 6.2: Partially constructed mount to illustrate the position of the resonator and bulk sample. The resonator is held in place by a protruding aluminium finger, which also provides a thermal path for cooling the sample. The bulk sample is held in place with a nylon screw holding it against a cold finger in a similar fashion.
6.1 Experimental Setup

(a) Orientation used in resonator mount. The magnetic field for ions on the right of the resonator will be larger for those on the left, so there will be a gradient in Zeeman shifts in the ions the light samples as it propagates in WGM modes around the equator.

(b) Magnet positioned with rotation axis parallel to that of the resonator. Light propagating in WGM modes around the resonator equator will interact with ions experiencing the same Zeeman shift, as all these ions are in a plane the same distance from the magnet.

Figure 6.3: Illustration of spatial inhomogeneous broadening for resonator ions, using the magnetic field orientation described in the text.

The near-field coupling prism was made from cubic zirconia \( (n_r = 2.18) \). It was mounted on three Attocube piezoelectric nano-positioning devices capable of operating in vacuum at cryogenic temperatures. This allowed precise control over three spatial degrees of freedom for effective coupling to the resonator. The ANR101 rotator was placed at the bottom of the stack as it was the heaviest, allowing 360 degree rotation of the prism. Care was taken to relieve stress on the electrical connections to the rotator as it was observed that the rotator could not provide enough torque to function correctly if the stress from the connecting wires was too great. An ANGt101 goniometer was placed in the middle providing 6.6 degrees of angular variation (in the polar direction for our mount) and an ANPx101 translator with a 5 mm travel range and \( \sim 10 \) nm minimum step size at 4 K was placed on top.

The nano-positioning devices were driven using an ANC150 piezo step controller from Attocube Systems via an electrical feed-through to the cryostat sample space. The copper leads connecting to the nano-positioners were wrapped around the first-stage heat exchanger, which has a maximum cooling power of 40 W compared to 0.25 W for the second-stage cold head at 3.5 K. This was important as one end of the copper leads was at room temperature, so to stop them heating up the rest of the mount the wires needed to be cooled as much as possible before the Attocube terminals. The voltages and frequencies of the driving signals to the Attocubes were adjustable with the ANC150 and both were kept as low as practically possibly to avoid overheating, as the vacuum inside the cryostat provided little opportunity for heat dissipation. Coarse positioning was partially automated using
software provided by Attocube Systems and fine positioning was done by hand. This was because the
Attocube drive mechanism is based on friction and the step size is not precisely repeatable so fully
automated positioning would not be reliable. The approximate step size also varies by a significant
amount with temperature. This setup allowed positioning of the prism with the control necessary for
coupling to the resonator. A mirror was also fixed in the mount to direct light out-coupled from the
resonator out of the cryostat window for detection. Coupling was done as described in chapter 5.

As discussed in chapter 5, magnetic field orientation can have a significant impact on the coher-
ence properties of erbium ions. The bulk sample was orientated such that the magnetic field was
in the D$_1$-D$_2$ plane along the D$_1$ axis. This orientation is chosen as the two possible magnetically
inequivalent sites become magnetically equivalent when the magnetic field is applied in the D$_1$-D$_2$
plane [27] which has advantages outlined in chapter 3. Although Bottger et al. found optimal coher-
ence times for a magnetic field orientation 120-140° from the D$_1$ axis, it was far simpler to mount
the bulk sample such that the field was either directly along the D$_1$ or D$_2$ axis given that we had no
mechanism to rotate the sample holder and the sample was cut along these axes. The magnetic field
was chosen to be along the D$_1$ axis as this gives longer coherence times than along the D$_2$ axis [27].

Compared to the bulk sample there is some ambiguity in orientating the resonator to the optimal
magnetic field direction, as due to its rotational symmetry it is not immediately obvious where the D$_1$
and D$_2$ optical extinction axes are. The resonator was mounted without knowledge of the orientation
of these axis, however it was known that the magnetic field was being applied in the D$_1$-D$_2$ plane, giv-
ing magnetic equivalence of sites. This means that coherence times in the bulk sample and resonator
are not expected to be exactly the same. Potential temperature differences between the bulk sample
and resonator could also cause a disparity in coherence times. In future work x-ray crystallography
could be used to orientate the resonator.

It should also be noted that light travels along the b optical axis in the bulk sample, whereas in the
resonator it is coupled in and circulates in the D$_1$-D$_2$ plane. The impact of this warrants further study,
as Y$_2$SiO$_5$ is birefringent and so could alter the polarisation as light propagates in the resonator.

A Koheras Adjustik E15 fiber laser purchased from NKT Photonics was used to excite the Er$^{3+}$
ions. This single-mode laser had a linewidth of less than 1 kHz over a 120 µs integration time,
and thermal tuning allowed frequency scanning over the range 1535.809-1536.899 nm (≈140 GHz).
Piezo tuning allowed fast frequency scanning over ≈2.5 GHz, which proved useful when examining
resonator mode structure. Passive laser stabilisation was done entirely inside the commercial laser
module.

The optical path for the experimental setup is shown in Figure 6.4.
Figure 6.4: Ray diagram of optical bench setup. Acronyms used are polarising beam splitter (PBS), acousto-optic modulator (AOM), quarter-wave plate (QWP), linear polariser (LP) and polarisation maintaining (PM). The lenses have focal lengths $L_1 = 200$ mm, $L_2, L_4, L_5 = 50$ mm and $L_3 = 100$ mm. Lenses $L_2$ and $L_3$ are mounted on a track that allows adjustment of the spacing between them in order to position the beam waist at the reflection surface of the prism for coupling. The flip mirror allows the beam to pass through either the bulk sample or be coupled into the resonator. The Koheras laser module provided a separate low-power fiber-coupled output port for monitoring frequency, which was done using a Bristol 621 Wavemeter.
Most of our experiments required the creation of variable length pulses, which was done using a double-pass acousto-optic modulator (AOM). An acoustic wave forms inside an AOM when a radio-frequency (RF) electrical signal is applied to it, which acts as a diffraction grating for incident light. Light in the first diffracted order is only present when the RF signal is present, so by rapidly switching the electrical signal optical pulses can be obtained from a continuous wave (CW) laser by collecting the first order diffracted light. The frequency of light in the first diffracted order is shifted from the zeroth order by the RF drive frequency.

A double-pass AOM was used because the angle of the first diffracted order is dependent on the RF drive frequency, as this changes the spacing of the acoustic wave diffraction grating inside. Passing the first diffracted order back through the AOM in a double-pass configuration means the AOM drive frequency can be changed without steering the beam. This was important for us as we used the same double pass AOM to create a local oscillator shifted in frequency for heterodyne detection of photon echoes and wanted maximum overlap with the probe beam.

AOMs were driven by a signal from a SpinCore PulseBlaster DDS-II-300-AWG programmable RF pulse generator board which was amplified with a Minicircuits ZHL-1-2W high power amplifier. This function generator employed Direct Digital Synthesis (DDS) allowing zero-latency frequency shifting and frequency resolution of 0.28 Hz. Digital transistor-transistor logic (TTL) outputs were also available from the PulseBlaster core, and were used to trigger the Tektronix TDS2024B oscilloscope used for data acquisition.

6.1.2 Heterodyne Detection of Photon Echoes

Nonlinear spectroscopy using photon echoes was the most important technique in getting results for this work, and as such it was important to have sensitive methods for detecting echoes. Photon echoes were created using pulses from the double-pass AOM with the apparatus described in the previous section, and detected using heterodyne detection. Different types of photon echo were discussed in chapter 3.

In optical heterodyne detection, a local oscillator signal is mixed with a probe beam on some sort of nonlinear circuit element (in our case a photodetector which measured intensity) and a beat signal at the difference frequency is observed. Generally the probe beam is a weak signal that might be difficult to detect directly. Heterodyne detection makes it possible to detect these weak signals by combining them with a strong local oscillator. This can be seen as follows. The signal from the probe is given by $E_1 e^{i\omega_1 t}$ and the local oscillator is $E_2 e^{i\omega_2 t}$, where $E_1$ and $E_2$ are (potentially complex) electric field amplitudes. These signals superimpose on the photodetector which detects the intensity, proportional to the square of the electric field amplitude:

$$|E|^2 = (E_1^* e^{-i\omega_1 t} + E_2^* e^{-i\omega_2 t})(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t})$$

$$= |E_1|^2 + |E_2|^2 + E_1^* E_2 e^{i(\omega_2 - \omega_1) t} + E_2^* E_1 e^{-i(\omega_2 - \omega_1) t}. \quad (6.1)$$

If the electric field amplitudes $E_1$ and $E_2$ are assumed to be real (if this were not the case it would simply mean a phase shift in the beat signal) then it is seen from equation (6.1) that the signal
from the photodetector will have a DC component plus a component at the difference frequency given by $2E_1 E_2 \cos (\omega_2 - \omega_1) t$. The DC component can be filtered away leaving only the beat note, the amplitude of which depends on the product of both $E_1$ and $E_2$. The important point here is that a signal with small amplitude $E_1$ can be enhanced by using a strong local oscillator for more sensitive detection. Shot-noise limited heterodyne detection is sensitive enough to detect single photons [159]. The beat signal of a photon echo detected using heterodyne detection is shown in Figure 6.5.

A comparison of echoes detected using heterodyne detection with direct detection is given in Figure 6.6. It is seen from this figure that heterodyne detection gives echo amplitudes much further above the noise floor, which was important when detecting small echoes from the resonator.

In this work the double-pass AOM was run at its 80 MHz center frequency to create pulses, and when an echo was expected the local oscillator was switched on by driving the double-pass AOM at 74.7 MHz resulting in a 10.6 MHz beat frequency (although the drive frequency of the AOM is only changed by 5.3 MHz, the two passes give 10.6 MHz). Both the probe and local oscillator beams were sent through a single-mode polarisation maintaining fiber, which cleans the beam and allows good mode overlap between the two giving strong beat signals. A second AOM was used to gate the photodetector such that light could only reach it when an echo was expected.

![Figure 6.5: Beat signal from a two-pulse photon echo detected using heterodyne techniques described in the text. The large feature immediately before the echo is called a free induction decay (FID), which is unavoidable as it is produced by similar coherent transient phenomena to the photon echo. The other smaller features before the echo are due to frequency components around 10.6 MHz in the applied pulse edges getting through the band-pass filter.](image)

Optical signals were detected using an InGaAs G8376 photodiode with peak sensitivity at 1550 nm installed in a homemade detector circuit (based on a design from [160]). The photodetector
signal was filtered using a Mini-circuits BBP-10.7+ band-pass filter with 9.5-11.5 MHz passband to clean the signal around the beat frequency, then amplified with a Mini-circuits ZFL-500LN-BNC+ providing 24 dB of gain before being sent to a Tektronix TDS2024B digital storage oscilloscope. Data was then downloaded from the oscilloscope and saved for analysis. Experiment control and data acquisition was usually automated and run from Matlab using Mex files to interface with application programming interfaces (APIs) of lab equipment. A Matlab control GUI was created by the author and ran routines developed by himself and other group members.

![Figure 6.6: Comparison of echo signals from photodetector using heterodyne detection and direct detection. Different scales on the y axes are due to different amounts of amplification. Directly detected signals are smaller than heterodyne signals and as such it is more difficult to resolve the echo from the noise floor. The two echoes displayed here are from different data sets with comparable parameters (the delay between pulses to create the echo is the same, and the laser intensity is comparable). Noise is close to 20% of the signal amplitude for the direct signal, whereas it is around 3% for the heterodyne signal, making heterodyne detection superior when detecting small echoes such as those from the resonator.](image)

Data acquisition routines repeated echo amplitude measurements four times for each set of parameter values and averaged. Acquiring a data set could take a few hours for some of the three-pulse photon echo measurements, and as such there was the potential of long-term drift in laser frequency or intensity occurring. To monitor this the amplitude of a “reference echo” with fixed parameters was
recorded periodically as the experiment was run, and interleaved with the data. Fluctuations in the amplitude of the reference echo over the data acquisition time would indicate the laser intensity or frequency drifting, so this data was recorded to monitor this. The laser wavelength was also recorded over the data acquisition time using the Bristol 621 Wavemeter and was found to drift by no more than 30 MHz over a few hours. In our Er\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} samples the inhomogeneous linewidth is broadened by the spatial variation of the applied magnetic field to \(\sim 2.5\) GHz in the resonator, so the laser drift was about 1.2\% of the inhomogeneous linewidth, which was acceptable for experiments in the bulk sample. For three-pulse photon echo experiments in the resonator another concern was that the laser frequency might drift away from the cavity resonance. Experiments were done on modes with \(Q \sim 10^6\), which corresponds to a FWHM of \(\sim 200\) MHz. The laser drift was up to 15\% of this, and careful attention was paid to the size of the reference echoes, which varied up to 20\% in amplitude over the course of the data acquisition time in some cases. Three-pulse photon echo data in the resonator was thus treated with caution, and not used to make any definitive claims. For higher Q cavities this laser drift could be even more problematic. One solution for the future might be to lock the laser to the cavity mode.

It should be noted that for two-pulse photon echo experiments in the resonator the acquisition time was much shorter and laser drift was not an issue.

### 6.1.3 Processing of Raw Data

For photon echo spectroscopy experiments, the echo amplitude was the desired quantity to be measured. In order to accurately extract this value from the raw data traces, a software filtering routine was run on all such traces. Raw data of heterodyne signals from the scope was numerically Fourier transformed and filtered around 10.6 MHz (the heterodyne beat frequency) using a Gaussian filter. The filter was chosen to have approximately 1 MHz FWHM (full-width half-maximum), which was narrow enough in the frequency domain to remove unwanted noise at other frequencies, but still narrow enough in the time domain so as not to smear out features when the filtered signal was inverse Fourier transformed\footnote{Multiplying by a Gaussian in the frequency domain is equivalent to convolving with the inverse Fourier transform of the Gaussian filter (itself a Gaussian) with the original signal in the time domain. A narrower filter in the frequency domain means convolving with a wider Gaussian in the time domain which would serve to smear out sharp features. A balance must be struck in order to have clean photon echoes without too much smearing, and 1 MHz FWHM yielded good results.}. The width of the Gaussian filter compared to the spectral density of an echo signal is illustrated in Figure \ref{fig:6.7}.

After filtering the absolute value was taken, and the maximum of this function over the time period in which the echo was expected was defined as the echo amplitude for that data trace. This process is shown graphically in Figure \ref{fig:6.8} from which the benefits of this simple filtering process are clear.

Initially we attempted to fit each echo envelope to a Gaussian, and use the amplitude of the Gaussian as the echo magnitude, however better results were obtained by selecting the maximum of the echo data vector and averaging over multiple repetitions. The Gaussian fit proved unreliable for echoes approaching the noise floor.
Figure 6.7: Frequency domain Gaussian filter with \( \sim 1 \) MHz FWHM centered at 10.6 MHz. Also shown is the normalised spectral density of a heterodyne-detected echo signal, obtained by taking the absolute value of the Fourier transform. This filter was narrow enough in the frequency domain to clean the echo signal, but not so narrow as to smear out important features in the time domain.
6.1 Experimental Setup

(a) Example of echo data downloaded directly from oscilloscope.

(b) Data after filtering about 10.6 MHz with the filter shown in Figure 6.7.

(c) Absolute value of unfiltered data. The noise spikes make it difficult to precisely determine the echo amplitude.

(d) Absolute value of filtered data. The echo is now much cleaner than in (c), and the amplitude can be measured more accurately.

Figure 6.8: Determining photon echo amplitude from the raw data. Clearly, a simple filtering routine significantly improves resolution of the echo peak.
6.2 Results

6.2.1 Coherence Time

The coherence time of the $^4I_{15/2}(1) \rightarrow ^4I_{13/2}(1)$ transition in Er$^{3+}$:Y$_2$SiO$_5$ at 3.5 K and 0.3 T magnetic field strength was measured using two-pulse photon echoes, as described in chapter 3. Echoes were created, detected and measured using the techniques described earlier in sections 6.1.2 and 6.1.3.

Experimental data of echo amplitude decay is fitted to both an exponential and the Mims expression, given by

$$I(\tau) = I_0 \exp\left(\frac{-2\tau}{T_M}\right)^x,$$

(6.2)

Where $T_M$ is the effective phase memory time, $\tau$ is the pause between pulses and $x$ is a parameter describing how the decay deviates from a pure exponential, as explained in chapter 3.

Two-pulse photon echo results in the bulk sample are shown in Figure 6.9. The spacing between pulses ($\tau$) in the two-pulse echo sequence is varied from 1 to 30 $\mu$s in increments of 0.1 $\mu$s. Before the experiment commences pulse lengths are adjusted to give the maximum echo amplitude for a fixed $\tau$, meaning that they correspond to $\pi/2$ and $\pi$-pulses. The beam waist is $\sim 90 \mu$m as this was a good size for coupling to our resonator, as explained in chapter 5. The same beam was used for experiments in the resonator and bulk sample (with a flip mirror to switch between the two), so the 90 $\mu$m spot size applies for the bulk sample too.

Fitting to an exponential $A e^{-2\tau/T_2}$ the echo decay time is $T_2 = 7.4 \mu$s. However the Mims expression clearly gives a better fit, with effective phase memory time $T_M = 10.5 \mu$s. This gives an effective homogeneous linewidth of $\Gamma_{\text{eff}} \simeq 100$ kHz.

Coherence time results for light coupled into the resonator are given in Figure 6.10. Again pulse lengths in the echo sequence are adjusted to give maximum echo amplitude for otherwise fixed parameters. The pause between pulses is increased from 1 to 20 $\mu$s in increments of 0.1 $\mu$s, and echo amplitude recorded at each step. Exponential fit $A e^{-2\tau/T_2}$ give $T_2 = 5.6 \mu$s, and again the Mims expression provides a better fit, with effective phase memory time $T_M = 8.2 \mu$s. This gives an effective homogeneous linewidth of $\Gamma_{\text{eff}} \simeq 120$ kHz.

---

2While the cryostat cold head was at 3.5 K, there was no temperature sensor actually on the Er$^{3+}$:Y$_2$SiO$_5$ samples, so their precise temperature was unknown, but should have been close to this with a good thermal connection.

3It should be noted that it is not actually necessary for coherence time measurements to use $\pi/2$ and $\pi$-pulses, but it makes sense as this gives the most intense echoes.
6.2 Results

Figure 6.9: Two-pulse photon echo amplitude as a function of time interval between the applied pulses ($\tau$). Data taken in bulk sample. To collect experimental data $\tau$ is increased from 1 to 30 $\mu$s in increments of 0.1 $\mu$s. Each data point is the average of four experimental traces. Exponential and Mims expression fits to the data also shown. Clearly the Mims expression is a better fit, indicating the presence of spectral diffusion over the time scale $2\tau$. Data is normalised to the largest echo amplitude in the data set. The plateau observed in the experimental data corresponds to a noise floor, and these values have not been used in the fit.
Figure 6.10: Two-pulse photon echo amplitude as a function of interval between applied pulses ($\tau$), this time in the resonator. $\tau$ is increased from 1 to 20 $\mu$s in increments of 0.1 $\mu$s. Each data point is the average of four experimental traces. Exponential and Mims fits to the data also shown, with Mims fit again more closely matching the data indicating spectral diffusion. The apparently higher noise level in this plot compared to Figure 6.9 is an artefact of normalising smaller echoes. The plateau observed in the experimental data corresponds to a noise floor, and these values have not been used in the fit.
6.2 Results

6.2.2 Spectral Diffusion

Initially we attempted to use three-pulse photon echoes to measure the population lifetime of the \( ^{4}I_{13/2} (1) \) excited state. However it was discovered that at a temperature of 3.5 K and magnetic field strength of 0.3 T, significant spectral diffusion smears out the population grating much faster than the population lifetime, which has been measured at over 10 ms \( [27] \). This meant that we could not measure the lifetime using this technique, so instead we made an attempt to fit the shape of our echo decay curves into known models for spectral diffusion. We then tentatively suggest some potential causes of spectral diffusion in our sample. However it should be noted that we did not have access to the parameter variation in temperature and magnetic field strength necessary for a detailed study of spectral diffusion, which can result from a myriad of complex interactions in a solid \( [120] \).

![Graph of three-pulse photon echo decay curve](image)

Figure 6.11: Decay in three-pulse photon echo amplitude as a function of \( T \), the hold time between second and third \( \pi/2 \) pulses. Inset shows the echo sequence with this data set, for which the pause between pulses 1 and 2 was \( \tau_{12} = 1 \mu s \), giving a population grating with frequency spacing \( 1/\tau_{12} = 1 \) MHz. There are clearly two different decay time-scales, which have each been fitted to an exponential. The exponential decay time for the rapid initial decay is \( \sim 50 \mu s \), and for the slower later decay is \( \sim 500 \mu s \).

Figure 6.11 shows an example of a three-pulse echo decay curve for a 1 \( \mu s \) pause between applied pulses 1 and 2 \( (\tau_{12}) \) as a function of \( T \) (the pause between applied pulses 2 and 3). There is obviously two distinct decay times. The rapid initial decay has a \( 1/e \) time constant of \( t_{1} \simeq 50 \mu s \), and the slower
decay beginning after \( T \approx 200 \, \mu s \) has a time constant of \( t_2 \approx 500 \, \mu s \).

In the work of Bottger et al., the second decay time was identified with the population lifetime, which was corroborated with fluorescence lifetime measurements \([127]\). They found a population lifetime of 11.4 ms based on these results, which is significantly longer than our 500 \( \mu s \) decay rate observed above. This suggests that the population gratings created by the first two applied pulses smear out due to spectral diffusion faster than the population lifetime. Figure 6.12 shows the same data as Figure 6.11 with a fit to the expression for three-pulse echo amplitude decay in the presence of spectral diffusion:

\[
A(\tau_{12}, T) = A_0 \exp \left( \frac{T}{T_1} \right) \exp (-2\tau_{12} \Gamma_{\text{eff}}(\tau_{12}, T)),
\]

(6.3)

where \( \Gamma_{\text{eff}} \) is given by

\[
\Gamma_{\text{eff}}(T) = \Gamma_0 + \frac{1}{2} \Gamma_{SD} \left( 1 - e^{-RT} \right),
\]

(6.4)
which was introduced in chapter 3. In Figure 6.12, $\tau_{12}$ is fixed at $1 \mu s$. When fitting to equation 6.3, only $\Gamma_{SD}$, $A$ and $R$ are fitted parameters. $T_1$ is taken from the literature to be $11.4$ ms [127], and $\Gamma_0$ is the initial effective linewidth calculated from two-pulse echo decays (100 kHz in the bulk sample and 120 kHz in the resonator). It is important to note that our fit in Figure 6.12 only includes the effect of one spectral diffusion mechanism. More terms would need to be added to equation 6.4 to account for other mechanisms. From our fit we find that $\Gamma_{SD} = 1.4$ MHz and $R = 8.9$ kHz. Figure 6.13 shows similar data for different values of $\tau_{12}$, with values of fitted parameters inset.

![Graphs showing three-pulse echo amplitude decays for different values of $\tau_{12}$](image)

**Figure 6.13:** Three-pulse echo amplitude decays for different values of $\tau_{12}$, along with fits to the data using equation 6.3. Fitted parameters $\Gamma_{SD}$ and $R$ are inset. Data is less reliable for longer values of $\tau_{12}$ due to rapid dephasing during this time.
In a comprehensive study of spectral diffusion in Er$^{3+}$:Y$_2$SiO$_5$, Bottger et al. observe values of $R$ and $\Gamma_{SD}$ comparable with these [120]. Exploring the behaviour of these parameters as a function of temperature, magnetic field strength and erbium concentration, they demonstrate that erbium environment ion spin flips are the cause of this spectral diffusion. They also found that yttrium spin flips can cause spectral diffusion above temperatures of $\sim 4.2$ K, and we suspect this is what is causing our slower secondary decay rate, however more experiments would need to be done to confirm this assertion.

### 6.2.3 Atom-Cavity Coupling

The atom-cavity coupling rate in the resonator was measured using two-pulse photon echoes. By varying the applied pulse lengths over a range of values, the length of a $\pi$-pulse can be determined by selecting the pulse length that gives the largest echo amplitude for fixed power. The Rabi frequency ($\Omega$) is given by [26]

$$\Omega = \frac{\mu E}{\hbar} = \frac{\Theta}{\tau},$$  \hspace{1cm} (6.5)

where $\mu$ is the transition dipole moment, $E$ is the electric field amplitude, $\hbar$ the reduced Planck’s constant, $\Theta$ is the pulse area and $\tau$ is the pulse duration.

Photon echo amplitude as a function of applied pulse duration is shown in Figure 6.14. In this data set the laser power was 1.8 mW into the cryostat, coupled to a resonator mode of $Q \sim 1 \times 10^6$ with efficiency $\sim 8\%$. The duration of the second pulse in the two-pulse echo sequence was varied from 0.1 $\mu$s to 5 $\mu$s in increments of 0.02 $\mu$s. The first pulse was set to be half the duration of the second pulse in each case. Each data point is the average of five echo amplitude traces. Maximum echo amplitude should occur when the applied pulses have an area of $\pi/2$ and $\pi$ respectively. From this we can see that the length of a $\pi$-pulse is $1.30 \pm 0.05$ $\mu$s. This gives a Rabi frequency of $\Omega = 2\pi \times 385$ kHz. To calculate the coupling between a single photon and an ion in the resonator the Rabi frequency is divided by the square root of the number of photons in the cavity,

$$g = \frac{\Omega}{2\sqrt{n_{ph}}},$$ \hspace{1cm} (6.6)

The number of intra-cavity photons can be calculated starting from a quantum Langevin equation, given by [161]

$$\dot{a} = -\left(\frac{\gamma_p + \gamma_i}{2}\right)a - \sqrt{\gamma_p} a_{in} - \sqrt{\gamma_i} b_{in},$$ \hspace{1cm} (6.7)

where $a$ is the bosonic annihilation operator for the cavity mode, $\gamma_p$ is the coupling rate into and out of the cavity through prism, $\gamma_i$ is the intrinsic loss rate of the cavity, $a_{in}$ is the input field into the prism coupling mode and $b_{in}$ is input from other modes. The dot above $a$ on the left of equation 6.8 denotes a time derivative. The input field to the cavity from the laser can be modelled as a coherent state (and hence so can the output), and so taking expectation values in equation 6.8 we can replace
6.2 Results

Figure 6.14: Amplitude of two-pulse photon echo versus applied pulse duration in the resonator. Duration of second pulse in the two-pulse photon echo sequence shown on the x axis. The first pulse was chosen to be half this duration in each case. Second pulse varies from 0.1 to 5 µs in increments of 0.02 µs. The maximum echo amplitude occurs for a second pulse duration corresponding to a pulse area of π. The length of a π-pulse is hence 1.30 ± 0.05 µs.

the quantum operators with classical fields. The Langevin equation then becomes

\[ \dot{\alpha} = - \left( \frac{\gamma_p + \gamma_i}{2} \right) \alpha - \sqrt{\gamma_p} \alpha_{in} - \sqrt{\gamma_i} \beta_{in}, \]  

(6.8)

where \( \alpha \) is the classical field defined by \( a |\alpha\rangle = \alpha |\alpha\rangle \), and \( \alpha_{in}, \beta_{in} \) are the classical fields corresponding to \( \alpha_{in} \) and \( b_{in} \). In our experiments, the prism coupling mode was the only excitation mode, so \( \beta_{in} \) was equal to zero.

The steady-state cavity photon number is found by setting \( \dot{\alpha} = 0 \), from which we find

\[ \alpha = - \frac{2 \sqrt{\gamma_p}}{\gamma_p + \gamma_i} \alpha_{in}. \]  

(6.9)

The input-output relation for the prism coupling mode is given by

\[ \alpha_{out} = \alpha_{in} + \sqrt{\gamma_p} \alpha, \]  

(6.10)
and substituting in the expression for $\alpha$ in equation 6.9 we find that

$$\alpha_{out} = \left(1 - \frac{2\gamma_p}{\gamma_p + \gamma_i}\right) \alpha_{in}. \quad (6.11)$$

This equation determines how much of the light incident on the coupling prism is not coupled into the resonator, instead being reflected off the face of the prism and into the output mode. We can hence define a reflection coefficient as

$$R = \left(1 - \frac{2\gamma_p}{\gamma_p + \gamma_i}\right)^2, \quad (6.12)$$

and a corresponding transmission coefficient (which determines the fraction of light coupled or “transmitted” into the resonator)

$$T = 1 - R = \frac{4\gamma_p}{\gamma_p + \gamma_i} - \left(\frac{2\gamma_p}{\gamma_p + \gamma_i}\right)^2. \quad (6.13)$$

As our coupling efficiency was relatively low ($\sim 8\%$), the $R$ coefficient is much larger than the $T$ coefficient. This implies that $\gamma_p \ll \gamma_i$, and we can write an approximate expression for $T$ as

$$T = \frac{4\gamma_p}{\gamma_i}. \quad$$

Using this expression, and the fact that the coupling rates are related to the cavity quality factor by $\gamma_p + \gamma_i = \omega_0/Q$ (where $\omega_0$ is the cavity resonance frequency), we can solve equation 6.9. The photon number in the cavity mode is given by

$$n_{ph} = |\alpha|^2 = \frac{4\gamma_p}{(\gamma_i + \gamma_p)^2} |\alpha_{in}|^2, \quad (6.14)$$

and using our cavity parameters with an input power of 1.8 mW we find that $n_{ph} = 9.1 \times 10^5$.

From equation 6.6 the atom-cavity coupling rate is then $g = 2\pi \times 202$ Hz. We compare this experimental value to the theoretical atom-cavity coupling rate, given by

$$g = \frac{\mu}{n_r} \sqrt{\frac{\omega_a}{2\hbar \epsilon_0 V_M}}, \quad (6.15)$$

where $\mu$ is the transition dipole moment, $n_r$ is the refractive index of the cavity material, $\omega_a$ is the angular frequency of the atomic transition, $\hbar$ and $\epsilon_0$ are the usual physical constants and $V_M$ is the cavity mode volume. The cavity mode volume for the $n = 1$ modes of a WGM resonator can be approximated using the expression:

$$V_M \simeq 3.4 \pi^{3/2} \left(\frac{\lambda}{2\pi n_r}\right)^3 l^{11/6} \sqrt{2(l-m)+1}, \quad (6.16)$$

as was mentioned in chapter 5. For the resonator used in these experiments, the radius was 1.6 mm, giving an approximate fundamental mode volume of $V_M \simeq 1.38 \times 10^{-12}$ m$^3$. Substituting this back into equation 6.15 we find a theoretical atom-cavity coupling rate of $g = 2\pi \times 1.44$ kHz. This
result does not agree particularly well with our measured result, yielding an answer almost an order of magnitude larger. Potential causes of this discrepancy are discussed in section 6.3.3.

6.2.4 Cavity QED Parameters

The preceding results can be used to calculate the experimentally determined critical atom number

\[ N_0 = \frac{2\gamma h\kappa}{g^2}, \tag{6.17} \]

and the saturation photon number

\[ n_0 = \frac{\gamma^2 h}{4g^2}, \tag{6.18} \]

where \( \gamma = \frac{1}{T_1}, \gamma_h = \frac{1}{T_2}, \kappa = \frac{\pi c}{MQ} \) and \( g \) is the atom-cavity coupling from section 6.2.3. Using these definitions we find

\[ N_0 = 9.3 \times 10^7 \]
\[ n_0 = 1.7 \]

The criterion for strong coupling is \((N_0, n_0) < 1\), so clearly we are not in the strong coupling regime. The criterion for the saturation photon number \( n_0 \) is within reach, and should be straightforward to obtain with minimal optimisation. However the critical atom number \( N_0 \) is almost 8 orders of magnitude too large, and will prove to be more difficult. Prospects for achieving \( N_0 < 1 \) are discussed in section 6.3.4.

6.3 Discussion

6.3.1 Coherence Time Results

Our measurements show an effective phase memory time of \( T_M = 10.5 \mu s \) in the bulk \( \text{Er}^{3+}: \text{Y}_2\text{SiO}_5 \) sample and \( T_M = 8.2 \mu s \) in the resonator. Effective phase memory time is a more appropriate measure of coherence than the echo amplitude exponential decay time constant in the presence of significant spectral diffusion [120]. This leads to an effective homogeneous linewidth \( \Gamma_{eff} = 1/T_M \) of \( \approx 100 \) kHz in the bulk sample and \( \approx 120 \) kHz in the resonator. This implies that the mechanical grinding and polishing procedures used to fabricate the resonator did not ruin the coherence properties of the ions located around the perimeter of the resonator, which is where the WG modes propagate. Similar results were obtained in previous work by David McAuslan using \( \text{Pr}^{3+}: \text{Y}_2\text{SiO}_5 \) [26]. McAuslan also observed slightly shorter coherence times in the resonator, which were attributed to a probable temperature difference between resonator and bulk sample.

However, some care is necessary when interpreting our results. Because our resonator was mounted without knowledge of the magnetic field orientation relative to the optical extinction axes, coherence times are in general not directly comparable to the bulk sample.4 Unless we were lucky enough to get the orientations the same.
Figure 6.15: (a) Variation of site 1 and site 2 ground state and excited state g-factors as a function of applied magnetic field angle in the $D_1$-$D_2$ plane. (b) Homogeneous linewidth (measured using two-pulse photon echoes) as a function of applied magnetic field angle in the $D_1$-$D_2$ plane. It is seen that the homogeneous linewidth is relatively constant except for a spike around $35^\circ$ caused by small g-factors at site 1 around this angle. Unless we were very unfortunate, the orientation of the resonator would not fall into this angular window. The fact that photon echoes could be observed at all means that we were outside this window. Figure taken from [27], with permission from the author.

...
6.3 Discussion

6.3.2 Spectral Diffusion Mechanisms

It is seen from section 6.2.2 that our three-pulse echo decay curves have two distinct time scales. The second decay time is too fast to be attributed to population decay. It is also seen that equation 6.3 does not provide a particularly good fit to the data for longer values of $T$. This is due to the fact that we have only accounted for one spectral diffusion mechanism. Previous studies on spectral diffusion in 0.001% Er$^{3+}$:Y$_2$SiO$_5$ suggest that the initial rapid decay is caused by erbium environment ion spin flips. The spreading of the effective linewidth due to erbium spin flips saturates at some time, after which another spectral diffusion mechanism appears to take over. Bottger et al. observed yttrium nuclear spin flips as a contributor above 4.2 K $^{[120]}$, and we suspect this is happening in our samples.

The data presented here was all taken in the bulk sample, as resonator data proved too unreliable, as discussed at the end of section 6.1.2.

6.3.3 Atom-Cavity Coupling Results

We found an experimental atom-cavity coupling rate of $g = 2\pi \times 202$ Hz, compared to a theoretically calculated rate of $g = 2\pi \times 1.44$ kHz. The most obvious contributing factor in this discrepancy is that the theoretical value was calculated using the fundamental mode volume. During experiments it was unknown what type of mode the experimental data was taken in. In future work, it will be important to more carefully control which resonator mode is being excited.

However it is unlikely this is the only cause. The theoretical atom-cavity coupling rate is proportional to one over the square root of the cavity mode volume, $g \propto \frac{1}{\sqrt{V}}$. This means that to fully account for the observed difference between experiment and theory, the volume of the mode experiments were done it would need to be $\sim 70$ times larger than the theoretical fundamental mode volume. The theoretical mode volume expression assumes a perfect sphere, so it is unlikely to be exactly accurate for our spheroidal resonator, however it should be a reasonable first approximation. Calculating the theoretical atom-cavity coupling for higher-order modes, we find that for $\ell - m = 1$, $g = 2\pi \times 1.09$ kHz, for $\ell - m = 10$, $g = 2\pi \times 670$ Hz, and for $\ell - m = 100$, $g = 2\pi \times 382$ Hz. Higher values of the principal mode number $n$ also give larger mode volumes, although these are more difficult to calculate as simple approximations do not exist. This shows that the mode order can have a significant impact on the atom-cavity coupling rate.

Another possible reason for the disparity between theory and experiment is the fact that Y$_2$SiO$_5$ is birefringent. As light coupled into the resonator circulates in the D$_1$-D$_2$ plane (rather than directly along an optical extinction axis), the polarisation will be altered as the light propagates. The transition dipole moment of erbium ions in Er$^{3+}$:Y$_2$SiO$_5$ is polarisation-dependent, which was apparent as echoes in the bulk sample were twice as large for light polarised along the D$_2$ extinction axis compared to the D$_1$. Lack of a fixed polarisation direction for light in the resonator could lead to a lower effective transition dipole moment, which would mean a lower atom-cavity coupling, as $g \propto \mu$. More study as to the impact this might have would be a good thing to do in the future. It might be the case that light circulating in the D$_1$-D$_2$ plane (with polarisation in the same plane) is analogous to circularly polarised light passing through the bulk sample, with laser $k$-vector along the $b$ axis.

In summary, there are numerous reasons why our experimentally determined coupling rate differs
Experiments and Results

from the theoretical. In future work methods for reliably coupling into the fundamental mode at 1536 nm will be important. If such a large discrepancy is observed when experiments are done in the fundamental mode, the impact of other issues should be explored.

6.3.4 Feasibility of Reaching the Strong Coupling Regime

In our experiments we found a critical atom number of \( N_0 = 9.3 \times 10^7 \) and a saturation photon number of \( n_0 = 1.7 \). The criterion for strong coupling is \((N_0, n_0) < 1\). Our saturation photon number is only marginally larger than the requirement, and \( n_0 < 1 \) should be easily attainable with minor improvements. The critical atom number is nearly 8 orders of magnitude too large, and getting \( N_0 < 1 \) will be the more pressing task. There is hope however, as large improvements can be made quite straightforwardly.

Firstly, the coherence times of \( \text{Er}^{3+} \) ions can be enhanced by three orders of magnitude \[127\], by using higher magnetic fields and lower temperatures. This will decrease both \( N_0 \) and \( n_0 \) by three orders of magnitude, due to their linear dependence on \( \gamma_h \), the homogeneous linewidth. We have the capacity to cool down to 2 K in our lab, although new equipment will be required to achieve the required magnetic field strengths.

It is also expected that there is much room for improvement in our resonator fabrication procedures. Polishing was done using an air tool rotating at 60,000 rpm, instead of a lathe rotating at 1500 rpm or less. The logic behind this was that faster rotation should mean an equivalent amount of polishing could be done in a much shorter time, speeding up the manufacturing procedure. Our modest quality factors suggest that perhaps the rotation rate is too rapid for a thorough polish. Cleaning procedures have also been shown to be vitally important, with the best resonators undergoing cleaning in a clean room environment and using a thermal annealing procedure to remove surface impurities \[152\]. More investigation into fabrication procedures would likely be beneficial into achieving bulk absorption limited Q-factors.

Much smaller resonators can also be fabricated using automated single-point diamond turning \[162\]. Crystalline micro-resonators of 80 \( \mu \text{m} \) diameter have been fabricated using this technique \[146\]. A smaller resonator has a smaller mode volume, which would increase the atom-cavity coupling \( g_0 \) hence decrease the cavity QED parameters.

With this in mind we calculate \( N_0 \) and \( n_0 \) for a resonator of 100 \( \mu \text{m} \) radius and \( Q = 1 \times 10^8 \). A resonator of this size could be made using single-point diamond turning. The quality factor of \( 1 \times 10^8 \) was chosen because studies of absorption in bulk \( \text{Y}_2\text{SiO}_5 \) at 606 nm indicate that the upper limit on Q-factor is at least this high \[163\]. Using this resonator, along with the narrow erbium homogeneous linewidth values from the literature we find \[
N_0 = 0.22 \\
n_0 = 3.96 \times 10^{-7},
\]

\[8\]In this paper, the authors made a monolithic Fabry-Perot resonator from a sample with Bragg reflector mirrors at either end. Many round-trips in the cavity then provide the optical path length necessary to measure absorption in a transparent sample. Unfortunately absorption near 1536 nm was not measured.
which is well inside the strong coupling regime. Reducing the atom-cavity coupling from the theoretical value in the fundamental mode by a factor of $\sim 7$ (to model the disparity observed in our experiments) we still find

$$N_0 = 11.28$$
$$n_0 = 2.02 \times 10^{-5},$$

which is still close to strong coupling. It has been shown that interesting quantum information operations can be done close to strong coupling [14].

The major potential obstacle to achieving these cavity QED parameters is absorption in $Y_2SiO_5$ at 1536 nm. We did not observe the increase in Q-factor we expected at 1536 nm over 606 nm, which implies that perhaps $Y_2SiO_5$ is not less transparent in the infra-red. Future research should definitively answer this question.

### 6.4 Summary

We have investigated the coherence properties of rare-earth ions in resonators, and found that the manufacturing procedure does not seem to ruin coherence times. We have also observed spectral diffusion in our samples, and said what we can about the causes of this spectral diffusion based on our limited data. We also found the cavity QED parameters of erbium ions in home-made WGM resonators. Although we are currently a long way from the strong coupling regime, we discuss how significant gains can be made using currently available technology.

We conclude that if $Y_2SiO_5$ is as transparent at 1536 nm as it is at 606 nm, it should be possible to reach the strong coupling regime using monolithic $Er^{3+}:Y_2SiO_5$ resonators.
Chapter 7

Conclusion

7.1 Summary

In this thesis we have investigated the possibility of reaching the strong coupling regime of cavity QED using erbium-doped whispering gallery mode resonators. Strong coupling would allow creation of a coherent interface between light and matter [2], which could also function as a deterministic single photon source [3]. The state of a single dopant could also be measured, which would enhance the scalability prospects of promising rare-earth ion quantum computing schemes [18].

To this end, we fabricated millimetre-sized crystalline WGM resonators from erbium-doped yttrium orthosilicate, using mechanical grinding and polishing techniques. We used photon echo spectroscopy to ensure that the fabrication procedure did not damage the coherence properties of the ions. Significant spectral diffusion was observed in our Er$^{3+}$:Y$_2$SiO$_5$ samples, and we briefly discuss possible causes, chief among them being erbium environment ion spin flips. Finally, we measured the atom-cavity coupling rate and calculated the critical atom number ($N_0$) and the saturation photon number ($n_0$), two dimensionless parameters useful in characterising the strong coupling regime. We observe a saturation photon number of $n_0 = 1.7$, and the requirement that $n_0 < 1$ should be easily achieved with marginal increases in Er$^{3+}$ coherence times, or marginally smaller cavities. Our critical atom number is $N_0 = 9.3 \times 10^7$, which is almost 8 orders of magnitude larger than the requirement $N_0 < 1$. However, we are not really as far away as this number makes it appear; by optimising the erbium ion coherence properties the critical atom number can be decreased by three orders of magnitude right away. We calculate that it should be possible to achieve critical parameters well inside the strong coupling regime using existing resonator fabrication technology and ions with optimal coherence, provided that Y$_2$SiO$_5$ does not absorb more at 1536 nm than 606 nm.

7.2 Future Work

There are several areas that suggest themselves for further study from this thesis. The first is an investigation of absorption in Y$_2$SiO$_5$ at 1536 nm. Un-doped samples of pure Y$_2$SiO$_5$ have been purchased from Scientific Materials Corporation for this purpose. Manufacturing whispering-gallery mode resonators from these blank samples should allow direct comparison of Q-factors at 1536 nm
versus 606 nm. Using doped samples complicates matters as rare-earth ions have multiple transition frequencies. For example, early on in this work an attempt was made to compare Q-factors in a Pr\(^{3+}\):YAG resonator at 1536 nm and 606 nm. This was complicated by significant absorption by the praseodymium at both 1536 nm and 606 nm. Using an un-doped sample should give only information about absorption in Y\(_2\)SiO\(_5\).

Research into improving resonator manufacturing techniques should also be carried out. Practising on Y\(_2\)SiO\(_5\) resonators would combine both of these areas and be a very worthwhile thing to do.

Also, methods should be developed for deterministically coupling into the fundamental mode, something that was not done in this work.

All in all the future of Er\(^{3+}\):Y\(_2\)SiO\(_5\) resonators in quantum computing applications has the potential to be bright, conditional on acceptably low absorption in Y\(_2\)SiO\(_5\) crystals.
Bibliography


