Light-matter entanglement between telecom photons and solid-state quantum memories

Jelena Velibor Rakonjac

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Light-matter entanglement between telecom photons and solid-state quantum memories

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Abstract

Future networks for quantum communication will require a new form of technology in order to operate over long distances. The no-cloning theorem precludes the use of amplification, so quantum repeater schemes have been proposed to achieve long-distance quantum communication through the distribution of entanglement. A key device for this setup is a quantum memory, which is required in order to store entanglement and synchronise its distribution.

Among the many physical systems than can be used as a quantum memory, rare-earth ion-doped crystals stand out due to their long coherence times and potential for multiplexing and integration. This thesis reports the use of one such system, Pr$^{3+}$:Y$_2$SiO$_5$, as a quantum memory using the atomic frequency comb (AFC) protocol. Building on previous work, we improved the performance of the memory to be able to demonstrate light-matter entanglement with a telecommunications wavelength photon and the quantum memory, for both a bulk memory and an integrated version.

To measure light-matter entanglement, we use photon pairs generated through cavity-enhanced spontaneous parametric down conversion, where one photon (the signal) is stored in the memory and the other (the idler) is at a telecommunications wavelength. These photon pairs exhibit energy-time entanglement, which we analyse in the time basis using fibre- and AFC-based Mach-Zehnder interferometers.

In the first experiment, we demonstrate light-matter entanglement where the bulk quantum memory is used with on-demand retrieval via storage in a spin-wave. We measure interference fringes with average visibilities of 89(2)% for AFC storage, and 70(3)% for spin-wave storage, sufficient to demonstrate entanglement. This is the first such demonstration using an on-demand multimode solid-state memory and telecom photon. We show that entanglement is maintained for up to 47.7 $\mu$s of storage. These results were made possible through improvements in
the AFC efficiency and the noise filtering. Furthermore, we perform on-demand storage and retrieval of up to 30 temporal modes.

We take the next step towards implementing our system in a real world scenario by measuring entanglement over a distance. To do so, we send the idler photons through both optical fibre spools and deployed fibre in the metropolitan area of Barcelona, Spain, while the photon detection still occurs in the same lab. The resulting visibilities (80% or higher) demonstrate that the photonic qubit does not decohere during transmission through the optical fibre. We then decouple the photon creation and detection by measuring non-classical correlations between signal photons detected in the lab, and idler photons detected 16 km away (44 km in fibre) in a different location.

The final experiment uses a fibre-integrated memory for light-matter entanglement. We use a Pr$^{3+}$:Y$_2$SiO$_5$ crystal containing a Type I laser-written waveguide, interfaced directly with optical fibre. The fibre-integration allows for improved transmission and AFC efficiency compared to a free-space coupled waveguide memory. To analyse the performance of the memory, we demonstrate non-classical correlations for storage times up to 28 $\mu$s. We perform qubit tomography in the time basis for storage times of 3 and 10 $\mu$s, measuring two-qubit fidelities of 86(2)\% and 86(4)\%, respectively, demonstrating storage of entanglement.

These experiments demonstrate the potential of our system for use as part of a quantum repeater, with many opportunities for further improvement to the storage time, efficiency, and multiplexing capability.
**Resumen**

Las futuras redes de comunicación cuántica requerirán una nueva forma de tecnología para operar a larga distancia. El teorema de no clonación excluye el uso de la amplificación, por lo que se han propuesto esquemas de repetidores cuánticos para lograr comunicación cuántica a larga distancia mediante la distribución del entrelazamiento. Un dispositivo clave para esta configuración es una memoria cuántica (MC), necesaria para almacenar el entrelazamiento y sincronizar su distribución.

Entre los muchos sistemas físicos que pueden utilizarse como MC, destacan los cristales dopados con iones de tierras raras, debido a sus largos tiempos de coherencia y a su potencial para la multiplexación y la integración. En esta tesis se describe el uso de uno de estos sistemas, Pr$_3^+:Y_2$SiO$_5$, como MC utilizando el protocolo de peine de frecuencia atómica (AFC). Basándonos en trabajos anteriores, mejoramos el rendimiento de la memoria para poder demostrar entrelazamiento entre luz y materia (ELM) con un fotón de longitud de onda de telecomunicaciones y la MC, tanto para una memoria en bruto como para una versión integrada.

Para medir el ELM, usamos pares de fotones generados mediante conversión paramétrica descendente espontánea mejorada por cavidad, en la que un fotón (la señal) se almacena en la MC y el otro (el anunciador) está en una longitud de onda en el rango de las telecomunicaciones. Estos pares de fotones presentan entrelazamiento energía-tiempo, que analizamos en la base temporal utilizando interferómetros Mach-Zehnder basados en fibra y AFC.

En el primer experimento demostramos el ELM cuando se usa la MC en bruto con lectura bajo demanda a través del almacenamiento en una onda de espín. Medimos franjas de interferencia con visibilidades medias del $89(2)\%$ para el almacenamiento en AFC y del $70(3)\%$ para el almacenamiento en ondas de espín, suficientes para demostrar entrelazamiento. Se trata de la primera demostración
de este tipo en la que se utiliza una MC multimodo de estado sólido bajo demanda y un fotón de telecomunicación. Demostramos almacenamiento del entrelazamiento durante un máximo de 47.7 µs. Estos resultados fueron posibles gracias a mejoras en la eficiencia del AFC y del filtrado de ruido. Además, realizamos almacenamiento y lectura bajo demanda de hasta 30 modos temporales.

El siguiente paso hacia la implementación de nuestro sistema en un escenario del mundo real consiste en medir el ELM a distancia. Para ello enviamos los fotones anunciadores a través de carretes de fibra óptica y de fibra desplegada en el área metropolitana de Barcelona mientras que la detección de fotones sigue teniendo lugar en el mismo laboratorio. Las visibilidades resultantes (80% o más) demuestran que el qubit fotónico no pierde coherencia durante la transmisión a través de la fibra. A continuación, desacoplamos la creación y detección de fotones midiendo correlaciones no clásicas entre fotones de señal detectados en el laboratorio y fotones anunciadores detectados a 16 km de distancia (44 km en fibra) en una localización diferente.

El experimento final utiliza una MC integrada en fibra para el ELM. Usamos un cristal de Pr$^{3+}$:Y$_2$SiO$_5$ que contiene una guía de ondas de tipo I escrita con láser, acoplada directamente a la fibra óptica. La integración en fibra permite mejorar la transmisión y la eficiencia del AFC en comparación con una MC de guía de ondas acoplada al espacio libre. Para analizar el rendimiento de la MC, demostramos correlaciones no clásicas para tiempos de almacenamiento de hasta 28 µs. Realizamos tomografía de qubits en la base temporal para tiempos de almacenamiento de 3 y 10 µs y medimos fidelidades de dos qubits del 86(2)% y 86(4)%, respectivamente, demostrando el almacenamiento de entrelazamiento.

Estos experimentos demuestran el potencial de nuestro sistema para su uso como parte de un repetidor cuántico, con varias oportunidades para mejorar aún más el tiempo de almacenamiento, la eficiencia y la capacidad de multiplexación.
**Resum**

Les futures xarxes per a la comunicació quàntica requeriran una nova forma de tecnologia per operar a llargues distàncies. El teorema de no-clonació impedeix l’ús de l’amplificació, per tant s’han proposat esquemes de repetidors quàntics per aconseguir una comunicació quàntica a llarga distància mitjançant la distribució de l’entrellaçament. Un dispositiu clau per a aquesta configuració és una memòria quàntica (MQ), que es requereix per emmagatzemar l’entrellaçament i sincronitzar-ne la distribució.

Entre els molts sistemes físics que es poden utilitzar com a MQ, destaquen els cristalls dopats amb ions de terres rares pels seus llargs temps de coherència i el seu potencial de multiplexació i integració. Aquesta tesi descriu l’ús d’un d’aquests sistemes, Pr$^{3+}$:Y$_2$SiO$_5$, com a memòria quàntica utilitzant el protocol AFC (Atomic Frequency Comb). A partir de treballs anteriors, hem millorat el rendiment de la memòria per poder demostrar l’entrellaçament llum-matèria amb un fotó de longitud d’ona de telecomunicacions i la MQ, tant per a una memòria massiva com per a una versió integrada.

Per mesurar l’entrellaçament llum-matèria, utilitzem parells de fotons generats mitjançant una conversió paramètrica a la baixa espontànies millorada per una cavitat, on un fotó (el senyal) s’emmagatzema a la MQ i l’altre (l’anunciador) es troba a una longitud d’ona de telecomunicacions. Aquests parells de fotons presenten un entrellaçament d’energia-temps, que analitzem en la base del temps mitjançant interferòmetres Mach-Zehnder basats en fibres i AFC.

En el primer experiment, demostrem l’entrellaçament llum-matèria on la MQ massiva s’utilitza amb la recuperació sota demanda mitjançant l’emmagatzematge en una ona d’espín. Mesurem les franges d’interferència amb visibilitats mitjanes del 89(2)% per a l’emmagatzematge AFC i del 70(3)% per a l’emmagatzematge d’ona d’espín, suficient per demostrar l’entrellaçament. Aquesta és la primera demostració d’aquest tipus que utilitza una MQ d’estat sòlid multimode sota
demanda i un fotó de telecomunicacions. Mostrem que l’entrellaçament es manté fins a 47.7 µs d’emmagatzematge. Aquests resultats van ser possibles gràcies a millores en l’eficiència de l’AFC i el filtratge del soroll. A més, realitzem emmagatzematge i recuperació sota demanda de fins a 30 modes temporals.

Donem el següent pas per implementar el nostre sistema en un escenari del món real mesurant l’entrellaçament a distància. Per fer-ho, enviem els fotons anunciadors tant a través de bobines de fibra òptica com de fibra desplegada a l’àrea metropolitana de Barcelona, Espanya, mentre que la detecció de fotons encara es produeix al mateix laboratori. Les visibilitats resultants (80% o més) demostren que el qubit fotònic no decohereix durant la transmissió a través de la fibra òptica. A continuació, desacoblem la creació i detecció de fotons mesurant les correlacions no clàssiques entre els fotons de senyal detectats al laboratori i els fotons anunciadors detectats a 16 km de distància (44 km en fibra) en una ubicació diferent.

L’experiment final utilitza una MQ integrada en fibra per a l’entrellaçament llum-matèria. Utilitzem un cristall Pr$^{3+}$:Y$_2$SiO$_5$ que conté una guia d’ona tipus I escrita amb làser, connectada directament amb fibra òptica. La integració en fibres permet una transmissió i una eficiència AFC millorada en comparació amb una memòria de guia d’ona acoblada a espai lliure. Per analitzar el rendiment de la MQ, demostrem correlacions no clàssiques per a temps d’emmagatzematge de fins a 28 µs. Realitzem una tomografia de qubit en la base de temps per a temps d’emmagatzematge de 3 i 10 µs, mesurant fidelitats de dos qubits del 86(2)% i del 86(4)%, respectivament, demostrant l’emmagatzematge de l’entrellaçament.

Aquestes experiments demostren el potencial del nostre sistema per utilitzar-lo com a part d’un repetidor quàntic, amb moltes oportunitats per millorar encara més el temps d’emmagatzematge, l’eficiència i la capacitat de multiplexació.
List of Publications


**Publications not included in this thesis:**


*Contributed equally to this work*
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Chapter 1

Introduction

1.1 Quantum communication

The transfer of information is an incredibly vital part of our current society. Thanks to the internet, we can easily communicate with others or acquire information no matter where we are. Key services in our lives, such as banking, medical facilities, and any kind of store make use of local or global networks in order to function. A scientist can contact their colleagues across the world or read decades-old manuscripts from their office. While the development of the internet began more than 50 years ago for a more humble purpose of utilising computing resources more efficiently [1, 2], now it is so ubiquitous it is difficult to imagine going back to life without it.

Over the last few decades many new research areas and applications have arisen which employ quantum phenomena [3–9], including a new type of processing and communication [10]. Quantum bits or qubits are a key ingredient for these applications [3]. Unlike a bit which can only be in the states 0 or 1, qubits can be in the state $|0\rangle$, $|1\rangle$, or superposition thereof. The superposition has the form $\alpha |0\rangle + e^{i\phi} \beta |1\rangle$, where $|\alpha|^2 + |\beta|^2 = 1$, and measuring the state in the appropriate basis collapses the superposition to $|0\rangle$ or $|1\rangle$. The unique properties of qubits necessitate a different method of operation compared with the classical internet, in order to build a quantum network for quantum communication.

The nature of qubits allows for the use of phenomena unique to quantum mechanics. In particular, entanglement is a powerful tool that is necessary for any quantum network [11]. When two (or more) particles are entangled, the states of the individual particles cannot be described independently of the other particles.
For example, one such entangled state for two particles $A$ and $B$ is:

$$|\psi\rangle = \frac{1}{\sqrt{2}} (|0\rangle_A |0\rangle_B + |1\rangle_A |1\rangle_B),$$

(1.1)

where $|0\rangle$ and $|1\rangle$ are basis vectors, and the subscripts indicate the particle. As this is maintained over a distance, a network can be established through the distribution of entanglement [11].

There are various applications for such a network, also referred to as the quantum internet [12, 13]. In a similar theme to the early internet, a quantum network could allow for distributed quantum computing [14]. Blind cloud quantum computing is another potential application [15], where computation can be performed using a distant quantum computer in such a way that it is private for a client and thus unknown to the computer. A quantum network could also be used for more secure communication, which is a particularly hot topic [9, 16, 17], as well as connecting optical clocks or astronomical telescopes to improve their accuracy [18–20].

In the following sections, I will go into further detail about the requirements for creating a quantum network.

### 1.2 Quantum repeaters

Light transmitted through optical fibres serves as the backbone of the current global network. Similarly, light is also the natural choice for flying qubits (those that must be sent over a distance), but in this case, as single photons, with which qubits can be encoded using many different degrees of freedom [21]. Light experiences exponential attenuation when transmitted through optical fibres, which can be around 0.2 dB/km\(^1\) at 1550 nm; for light at other wavelengths, the attenuation is even higher. Wavelengths in the region between 1260 nm and 1625 nm are often referred to as “telecommunications wavelengths” or “telecom wavelengths” since they have the least attenuation, where 1550 nm (in the telecom C-band) has the best transmission.

Amplifiers are placed periodically throughout a classical network to maintain signals that are sent over a distance. For qubits however, the no-cloning theorem

\(^1\)The most recent ultra low loss fibre from Corning can reach 0.15 dB/km [22], but this is not widely deployed. In practice, the point-to-point loss will be higher due to fibre-to-fibre connections and other losses.
1.2 Quantum repeaters

Figure 1.1: (a) Entanglement distribution from node A to node D. First, A + B and C + D are entangled, then entanglement swapping between B and C entangles A and D. (b) Scheme for the repeater setup with four nodes and two links using photon-pair sources (PPS) and absorptive quantum memories (QM). Bell-state measurements are performed using a beam splitter (BS) and detectors (Det.).

prevents amplification (copying a quantum state without adding noise) from being used [23]. Simply sending a large number of photons would not work either – even if 1 billion photons were sent per second, for a distance of 1000 km, only one photon would reach its destination roughly every 3000 years. A different method is required in order to achieve communication over a long distance.

The quantum repeater has been proposed for this purpose [24]. Rather than sending individual photons across an entire distance, a communication channel can be broken down into smaller segments called nodes, and entanglement can be propagated through the channel instead.

If we consider a channel divided in half, such as in Fig. 1.1(a), then we would need four nodes, A to D. Here the distances between A and B (as well as C and D) should be short enough to have sufficient photon transmission between nodes. If A and B are entangled, and likewise for C and D, by performing a Bell-state measurement (BSM), which is a joint measurement on photons from B and C that allows them to be entangled (also called “entanglement swapping”), then the entangled state (and thus quantum information) can be shared between A and D. Once the entanglement has been established over the full distance, it can be used to teleport qubits [25]. Thus information can be distributed over longer and longer distances by using more nodes.
While the precise method of generating entanglement and propagating it depends on the scheme, there are some common elements required. A source of photons is required at every node. Entanglement between the two nodes should be established using these photons, which is generally done in a measurement induced fashion by performing a Bell-state measurement at a central station between the two nodes. Since many of the photon sources and thus methods of generating entanglement are probabilistic, and photons will be lost regardless due to fibre losses, some way of storing entanglement at the nodes themselves is needed for synchronisation and scalability. The device required for this is known as a quantum memory.

A quantum memory is a physical system used to store quantum bits. Many of the systems are atomic so the qubits are stored in atomic coherences. In the case where the entangled photons are generated by the memory protocol itself, then the memory is called emissive, or read-only [26]. One of the most well-known schemes for this (and the first practical proposal utilising a quantum memory) is the Duan-Lukin-Cirac-Zoller (DLCZ) protocol [27]. If the memory is only used to store photons generated from an external source, then it is called absorptive, or read-write [26]. In either case, one photon is sent over a distance through the channel (optical fibre) to where the BSM occurs. This photon is called the herald. It should be at a telecom wavelength in order to minimise loss when transmitted through a fibre. Unless an emissive memory already operates at this wavelength, quantum frequency conversion is required for the herald, which introduces losses to the system. An overview and analysis of several different protocols can be found in Ref. [11].

To obtain a telecom herald while storing a photon compatible with a memory, another option is to use an absorptive memory with a non-degenerate source of photon pairs [28]. This combination can be realised by using a parametric down conversion source, which can generate entangled photon-pairs at whatever wavelengths are required by selecting a particular pump laser wavelength and non-linear crystal for conversion [11]. In this way, a telecom photon can be generated together with a memory-compatible photon without the need for frequency conversion. When one of these photons is stored, this results in light-matter entanglement, the fundamental building block of quantum repeater nodes which will be the focus of the experiments presented in this thesis.
One way to implement the basic repeater scheme using an absorptive memory and a photon-pair source is as follows (see Fig. 1.1). Two sources, one at A and one at B, generate photon pairs. One photon (the signal) is stored in the memory, while the other (the herald) is sent through optical fibre to an intermediate point between the two nodes called the central station. At the central station, the photonic modes are sent to a beam splitter (BS) with single photon detectors at both outputs.

The entanglement can be generated through a one-photon or two-photon BSM [11]. For the one-photon case, the BS erases the information about the origin of the photon (the which-path information), and a photon detection after the BS heralds the entangled state $\frac{1}{\sqrt{2}} (|1\rangle_A |0\rangle_B + e^{i\Delta \phi} |0\rangle_A |1\rangle_B )$, where $|0\rangle$ represents an empty memory and $|1\rangle$ the storage of an excitation, with the subscript denoting the memory at A or B, and $\Delta \phi$ the phase difference between the two paths of the heralding photon. This method has the advantage that only one photon has to be created, transmitted, and detected, which leads to high heralding rates. However, it requires phase stability over the entirety of the link, which is experimentally challenging. A two-photon BSM works similarly, but does not have this phase stability requirement, and instead makes use of a different degree of freedom like polarisation or time-bins (the latter of which is particularly relevant to the experimental work in this thesis). The disadvantage with this method is that the rate is lower due to requiring the creation, transmission, and detection of two photons.

Once entanglement has been established in the same way between nodes C and D, the excitations of the memories at B and C can be read-out to retrieve the single photons. These photons are also sent to a BS, where once again a BSM erases the which-path information, thus indicating the generation of entanglement between A and D. As mentioned previously, more nodes can be used to extended to longer distances.

Note that in practice this type of quantum repeater cannot be scaled to arbitrary long distances [29]. The practical limitation is in range of 1000-2000 km [11, 29], due to an inevitable drop in rates from transmission losses, non-unit memory and detection efficiencies, and similar. Ref. [11] for example requires ambitious but achievable specifications such as 90% memory and detector efficiencies, 90% fidelity for the full repeater chain (and thus even higher fidelities for the individual links), high entangled pair generation rates, massive multiplexing,
1. Introduction

and more. Other proposals, such as those using satellites, can potentially reach longer distances since transmission loss is significantly reduced at high altitudes, particularly above the atmosphere \([30, 31]\). They have other limitations however, such as only being operational for very short periods of time due to orbits and weather conditions, requiring receivers in isolated locations, and the optical beam path (communication channel) is affected by divergence (due to finite receiver sizes), diffraction, and atmospheric turbulence. To obtain a truly global quantum network, it is likely that both fibre-based and free-space transmission of photons will need to be used, leveraging on the pros of each scheme \([29]\).

Note that there are other recent repeater schemes relying on qubit processing and error correction at the nodes which do not require memories, nor classical communication between nodes \([32, 33]\), albeit with much greater technological complexity and requirements, such as the use of cluster states of hundreds of photons, more frequent repeater stations every few km and less than 50% total loss between them \([33, 34]\). Specific requirements on gate speeds, coherence times and detection efficiencies are additionally required in order to outperform memory-based repeater protocols \([34]\).

### 1.3 Quantum memories: requirements

In the following section, I will introduce the requirements for a quantum memory to be used in a quantum repeater, as well as some of the physical systems investigated thus far.

A quantum memory should be able to store photons for the entire time required for entanglement to be established across the full repeater link. The time depends on the particular scheme used to implement the repeater, the distances involved, and the rate of entanglement distribution between links, but can be on the order of seconds for distances greater than 600 km \([11]\). Increasing the entanglement generation rate for individual links, as described in the following paragraphs, can reduce the storage time required.

A few of the memory parameters relate to the entanglement generation rate, which is ultimately limited by the photon generation rate of the source. The memory should thus preserve this rate as much as possible. Both the efficiency and multiplexability of the memory are important in this regard. Efficient readout is needed in order to maintain the rate given by the photon source, so the closer
to unit efficiency, the better, and likewise for the components used to couple to the memory, detect photons, etc. A reduction in efficiency thus necessitates an increase in storage time to maintain the same entanglement generation rate [35].

The memory should not simply store only one photonic mode at a time, but should be able to simultaneously store many modes, which is known as multiplexing. It is also very favourable for obtaining a high rate [11]. The main degrees of freedom used for multiplexing are time [28], frequency [36], and space [37]. Time and frequency are also related to the bandwidth of the memory, i.e., the frequency range of photons that a memory can store. Spatial multiplexing can be implemented using different physical areas of a single memory, multiple memories, or different orbital angular momentum (OAM) modes in the same memory. Not all types of physical systems or protocols used for quantum memories can support all of the types of multiplexing, and the photon source must be able to support it too. While different temporal modes can be distinguished by arrival time with a single detector, this is not the case for spectral modes or OAM modes in the same spatial region. For maximum parallelisation, a method of demultiplexing the stored modes is required [38], as well as a detector for each mode.

The bandwidth of a memory is also relevant to the rate, as wider bandwidth memories can store photons which are temporally shorter. It is also harder to generate narrowband photons, so wider bandwidths relax the photon source engineering requirements. For long distance applications however, the number of modes that can be stored is more relevant to the rate than the bandwidth.

The fidelity of the quantum memory is also an important factor [11]. To retrieve the initial photon state stored by the memory without degrading it, the memory protocol (or other parts of the repeater system) should not introduce much noise to the system. Note that a high retrieval efficiency does not imply a high fidelity.

Another feature is the ability to retrieve stored photons on-demand, which is useful for synchronising the readout of quantum memories. This property is not strictly necessary as proposals exist for which it is sufficient to have extensive multiplexing [36], but it is nonetheless very desirable for a memory.

Many different physical systems are being investigated for use as a quantum memory. There is no clear winning candidate for the best memory thus far, as

\[\text{In an ensemble-based memory, these two values are not linked. The important fidelity to consider here is the one measured conditioned on the readout or reemission of a photon, called the conditional fidelity.}\]
different physical systems and protocols all have advantages and disadvantages, and no single system or setup has achieved all the above requirements to an ideal level. In general, most memories investigated so far are based on atomic systems. These include ensembles of atomic gases [39, 40], single atoms [41] and ions [42], and solid-state spins (both singular [43] and as ensembles [44, 45]), as well as other materials such as nanofabricated devices [46]. In this thesis I will focus on a quantum memory based on a solid-state spin ensemble, namely, rare-earth ion-doped crystals (REICs).

Many different memory protocols have been proposed and implemented. Some of the more well-known ensemble-based memory protocols include the DLCZ scheme [27], electromagnetically induced transparency (EIT) [47], off-resonant Raman [48], Autler-Townes splitting [49], and off-resonant cascaded absorption [39] have been used primarily with atomic gases, while the atomic frequency comb (AFC) [50], controlled reversible inhomogenous broadening [51], gradient echo memory (GEM) [52], rephased amplified spontaneous emission (RASE) [53], and the revival of silenced echo [54] schemes have been used primarily with rare-earth ion-doped crystals. While I will not cover any of the memory protocols in detail except for the AFC protocol in Chap. 3, recent reviews such as Refs. [55, 56] cover many of the protocols and state-of-the-art of different kinds of memory systems, and Ref. [57] cover these aspects more specifically for rare-earth based quantum memories.

Solid-state spins in particular offer many advantages as a quantum memory. Unlike cold atomic gases or single atoms or ions, no trapping is required as the spins are naturally fixed in place by their host. These materials are also useful for scalability, as it is easier to concentrate more memories in one space, either by addressing different parts of the same solid-state sample, placing many samples close together or by fabricating integrated structures (i.e., waveguides). REICs in particular possess some of the longest coherence times (both optical [58] and spin [59]) of any material, which is necessary in order to have long storage time for a memory. They also have a large capacity for multiplexing in many degrees of freedom when used as an ensemble [60] compared to other solid-state spins which are typically single emitters that can only be multiplexed by addressing more spins individually.

There are other aspects of a quantum memory that are also important to consider if a quantum repeater would be deployed in a real-world setting. In
particular, the footprint of the experimental system should be minimised. As typical experimental setups can span an optical table or more, miniaturising the setup would eventually be required. Like many other quantum technologies, companies aiming to commercialise quantum network and/or memory devices are starting to make progress in this area [61], though a truly complete and practical device has yet to be realised. Implementing multiplexing in a way that does not increase the setup complexity very much (i.e., not duplicating an entire setup) would be required. Integration can be of use by allowing for the memories to interface with other on-chip components, or with optical fibre directly, to simplify the experimental setup.

1.4 State of the art

While a variety of the systems used as memories were highlighted in the previous section, here I will mainly focus on the performance of rare-earth based memories. In particular, I will discuss the state-of-the-art in terms of storage time, efficiency, and multimodality, as well as the implementations of integrated memories that have achieved single photon level storage thus far. While there has also been significant investigation on single rare-earth ions for communication and processing such as in Refs. [62–69], here I will only focus on ensemble-based REIC memories. Additionally, I will briefly mention some of the best achievements for memory performance amongst all of the physical systems, and highlight the systems where light-matter entanglement with a telecom photon has been achieved, as well as experiments with heralded matter-matter entanglement, i.e., the basis of an elementary repeater link, which is what we aim to use our quantum memories for.

REIC-based memories have achieved long storage times and are continuously improving. Classical light has been stored for around one hour [70] as well as up to 25 hours [71], and for 100 ms at the single photon level [72], all using Eu\(^{3+}\):Y\(_2\)SiO\(_5\). Note that Refs. [70, 72] use the AFC protocol. In Pr\(^{3+}\):Y\(_2\)SiO\(_5\), the material used in this thesis, classical light pulses have been stored for around a minute using EIT [73].

The highest storage efficiency achieved in the quantum regime was 69% using weak coherent states as an input in Pr\(^{3+}\):Y\(_2\)SiO\(_5\) with the gradient echo memory (GEM) protocol [74]. In comparison with the results reported for the GEM
protocol, the highest reported AFC efficiencies were obtained using impedance-matched cavities, reaching 56% for classical pulses in Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} [75], and 27.5% for single photons using Tm:YAG [76], and 12% with spin-wave storage and classical pulses [77]. Recently, an AFC efficiency of 62% was reached in our group using Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} with weak coherent states [78]. Without a cavity, the high AFC efficiencies achieved were ∼40% using weak coherent states and Eu\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} [72], and 30% for a single photon in Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} [79].

Spin-wave storage offers the possibility for longer storage times than purely optical such as Ref. [74] and allows for on-demand read-out. In this regime the highest storage efficiency achieved was 31%, obtained using the spectral hole memory protocol with Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} and weak coherent pulses with a 3 μs full width at half maximum [80]. The highest SW efficiency measured with single photons as an input was around 4% using the AFC protocol with spin-wave storage in Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} [44], while the longest storage time of around 1 ms was measured with Eu\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} using the AFC-DLCZ scheme [45].

As mentioned in Sec. 1.3, multiplexing is important for increasing the entanglement generation rate. All of the experiments with REICs noted in this paragraph implemented the AFC protocol, which has a very favourable scaling for temporal multimodality in particular. In terms of temporal multimodality, 1250 modes have been stored with a single-photon input in \textsuperscript{171}Yb\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} with excited state storage [81], while 50 classical pulses were stored in Eu\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} using spin-wave storage [82]. Regarding frequency multiplexing, time-bin qubits were stored in up to 26 spectral modes at the single-photon level [36], and heralded single photons were stored in 15 spectral modes using a waveguide in Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} [83]. The highest number of total modes stored was 1650 in erbium-doped silica fibre, using five frequency modes with 330 temporal modes each, albeit with a storage time for 200 ns [84]. Spatial multiplexing has been demonstrated using orbital angular momentum states, with up to 51 modes at the single photon level [85]. A combination of temporal, spectral and spatial multiplexing was also demonstrated at the single photon level in Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} [86].

The solid-state nature of these materials makes them a natural choice for implementing integrated memories, which I will discuss in the following paragraphs. There are two main approaches to creating integrated memories: doping rare-earth ions into waveguiding structures, and fabricating waveguides in already doped crystals. In terms of single photon storage, using the first approach heralded single
photons have been stored in Tm$^{3+}$ and Er$^{3+}$ ions in LiNbO$_3$ waveguides [87, 88]. Er-doped silica glass fiber has also been used to store temporally and spectrally multiplexed single photons [84, 89].

The second approach of fabricating waveguides in the material has also been used for storage of single photons. Our group has followed the approach of using femtosecond laser machined waveguides [90] in Pr$^{3+}$:Y$_2$SiO$_5$. Single photon storage and frequency multiplexing were both demonstrated with this type of integrated memory [83, 91]. Other waveguides have been made using ion beam milling to shape the crystal itself. Storage at the single photon level with this type of waveguide was demonstrated in Nd$^{3+}$:YVO$_4$ [92] and in $^{167}$Er$^{3+}$:Y$_2$SiO$_5$ [93]. While spin-wave storage at the single photon level has yet to be demonstrated using any type of integrated memory, an alternative method to obtain on-demand readout using excited state storage has been demonstrated with both femtosecond laser machined and ion beam milled waveguides using the Stark effect together with AFC storage in Eu- and Er-doped Y$_2$SiO$_5$ systems [94, 95], albeit with a reduced potential storage time.

Looking at the best performance across all systems, the highest classical efficiency achieved was 92% using EIT with a cold atomic ensemble of Cs atoms [96], and up to 87% for single photon storage in cold atomic ensembles of $^{85}$Rb [40] and Cs atoms [97], also using EIT. The longest storage of classical light was the aforementioned 25 hours achieved in Eu$^{3+}$:Y$_2$SiO$_5$ [71], while the longest demonstration of single photon storage was 500 ms using the DLCZ protocol with a cold atomic ensemble of $^{87}$Rb atoms [98]. The highest number of modes stored was 1650 in Er-doped silica fibre with the AFC, as mentioned previously, which was demonstrated using single photons [84]. Finally, the largest bandwidth achieved was 40 THz using the vibrational modes of diamond with Raman excitation [99], but the storage time of this system ($\sim$ ps) is not suitable for long-distance communication.

As mentioned in the previous section, entanglement between a quantum memory and a telecom photon is a key requirement for a quantum repeater node. This type of entanglement has been demonstrated with several different REIC-based memories combined with photon-pair sources [87, 89, 100, 101], but only with storage in an optically excited state. It has also been demonstrated in many other kinds of memories: other solid state systems like quantum dots [102] and colour centres in diamond [103], as well as cold atomic ensembles [104, 105],
single atoms [106] and single ions [42, 107]. These other systems all utilised quantum frequency conversion in order to obtain a telecom heralding photon.

Taking it a step further, many systems have also been used to demonstrate matter-matter entanglement in a heralded manner. Among these systems are atomic ensembles (both cold [108–111] and room temperature [112]), single atoms [113, 114], trapped ions [115–117], nitrogen vacancies in diamond [118–122], quantum dots [123], and REICs [124–126]. To highlight some experiments in a particular, only a few of these experiments also feature telecom heralding [111, 114, 125], one of which was from our group, using two Pr$^{3+}$:Y$_2$SiO$_5$ memories [125]. The longest distance achieved was 33 km in optical fibre in Ref. [114] using single atoms and importantly also using independent experimental setups for the nodes, with a rate of 0.012 entanglement events per second. A longer distance of 50 km was achieved in Ref. [111] with a heralded rate of 1.5 entanglement events per second, though the memories did not store the photons for the entire photon travel time. The heralded entanglement experiment with the largest physical separation between memories (1.3 km) was Ref. [119] using nitrogen-vacancy centres with an entanglement rate of one signal per hour, though the heralding was not telecom. While not heralded, post-selected entanglement between two memories separated 12.5 km has also been demonstrated [127]. The highest matter-matter entanglement rates were achieved using quantum dots, with 2300 entanglement events per second [123], or 7300 events per second in a post-selected entanglement experiment [128]. With telecom heralding included, the highest heralding rate was 1400 counts per second with photon-pair sources and Pr$^{3+}$:Y$_2$SiO$_5$ memories in Ref. [125].

1.5 Thesis outline and contributions

In this thesis, I will present our work on solid-state absorptive quantum memories using the rare-earth doped crystal Pr$^{3+}$:Y$_2$SiO$_5$. In the following, I will outline the content of my thesis and my contribution to the work presented here.

In Chapter 2, I will introduce the properties of rare-earth ion-doped crystals and how they are suitable as a quantum memory material, with a particular focus on Pr$^{3+}$:Y$_2$SiO$_5$.

In Chapter 3, I will present the theoretical background that is particularly related to the experiments in this thesis. This includes basic light-matter interaction
relevant to our system, as well as the AFC protocol which is used to implement the quantum memory. I will describe the photon statistics that we consider for our experiments. As all of the experiments in this thesis deal with entanglement, I will describe the type of entanglement we use: energy time entanglement, as well as the relevant metrics for analysing entanglement.

Chapter 4 details the important components of the quantum memory setup, how we prepare an AFC structure and how we use the memory, as well as other aspects of the experiments like the photon-pair source, and the interferometers that we use in order to analyse entanglement.

Chapter 5 deals with experiments that use Pr$^{3+}$:Y$_2$SiO$_5$ as an on-demand quantum memory. Building on previous work in the group, we demonstrate the memory’s utility for light-matter entanglement with a telecom heralding photon. The main results are reported in Physical Review Letters [129], with results from an additional experiment I performed making use of temporal multimodality, published in Quantum Science and Technology [60].

Chapter 6 follows a similar theme to the previous, where we investigate light-matter entanglement in Pr$^{3+}$:Y$_2$SiO$_5$ over a longer distance. Here we send the telecom photons from the photon-pair source through optical fibres, both through spools in the lab and deployed optical fibres in a metropolitan setting, while the other photon is stored as an optical excitation in the memory. A manuscript for this experiment is currently in preparation.

Chapter 7 describes our experiment demonstrating a fibre-integrated memory for light-matter entanglement. This experiment used an integrated memory fabricated by our collaborator Dr. Giacomo Corrielli from the group of Dr. Roberto Osellame. We performed a full time-bin qubit tomography on photon-pairs, with one photon stored as an optical excitation in the memory. The results of this experiment were published in Science Advances [130].

Finally, Chapter 8 concludes the thesis with a final summary and discussion of the results, and the next objectives for our quantum memory system.

My contribution to this work was to rebuild, improve and operate the Pr$^{3+}$:Y$_2$SiO$_5$ quantum memory setup that already existed in our group, as well as implementing the AFC-based analysis interferometer, alongside Dr. Alessandro Seri and Dr. Samuele Grandi. I was primarily operating the memory for the experiments presented here, which involved adapting the setup or experimental control as required. The photon-pair source as well as the telecom wavelength fibre
interferometer were built and operated by Dr. Dario Lago-Rivera, with assistance from Dr. Samuele Grandi. I was involved in all the data acquisition and analysis here, along with Dr. Dario Lago-Rivera, Dr. Samuele Grandi, Dr. Alessandro Seri, Dr. Sören Wengerowsky and Dr. Félicien Appas, as noted in the individual chapters.

The experiments presented here demonstrate advances in the use of REICs as quantum memories. All of the experiments show the utility of the memory for light-matter entanglement, particularly with on-demand retrieval for the first time, and as an integrated device. We also demonstrated an improvement in the number of stored temporal modes for an on-demand memory at the single photon level. In my capacity to operate the memory, I also participated in the experiments presented in Refs. [125, 131] which are not included in this thesis.
Chapter 2

Properties of rare-earth ion-doped crystals

This chapter covers basic properties of rare-earth ion-doped crystals, starting from the ions themselves and when doped in a crystal. As the experiments in this thesis use Pr$^{3+}$:Y$_2$SiO$_5$ exclusively, that particular REIC will be introduced in more detail, finishing with a small overview of other ions used for quantum information purposes. Much of the information on the general properties of REICs comes from Refs. [132–134].

2.1 Rare earths

The rare earths are categorised as the group of elements from lanthanum (57) to lutetium (71), also known as the lanthanides\(^1\). In terms of electronic structure, the 4f orbital is filled along the row of lanthanides, where lanthanum has an empty 4f shell and ytterbium and lutetium both have filled 4f shells. The 5s, 5p, and 6s shells are all filled. As ions, they are typically in the 3+ oxidation state, where both 6s electrons are lost as well as a 4f electron (or a 5d electron in some cases). In this state, only cerium (58) to ytterbium (70) have partially filled 4f shells, and so it is this group of ions that is spectroscopically relevant to us.

Rare-earth ions can be divided into two categories depending on the number of 4f electrons they have. These are known as Kramers ions and non-Kramers

\(^1\)Scandium (21) and yttrium (39) are sometimes included due to their chemical similarity to the lanthanides, but generally not in the community utilising rare-earths for quantum technologies.
ions, where Kramers ions have an odd number of 4f electrons and non-Kramers ions have an even number. This distinction has various implications for the energy level structure and other spectroscopic properties. The ion used in the experimental work of this thesis is praseodymium (Pr), a non-Kramers ion.

2.2 Rare-earth ion-doped crystals

REICs have been the subject of a plethora of research in past, including as a laser medium [134], and optical storage medium [135]. In the last decades, they gained particular interest for quantum technologies due to their long coherence times at cryogenic temperatures (around 3 K or below), starting from proposals for quantum computing with rare-earth ions [136, 137], and now focusing more on quantum communication, as detailed in Chap. 1.

Even when doped in a crystal, transitions within the 4f shell exhibit very narrow linewidths, similar to atomic spectra. This is due to the shielding effect of the filled 5s and 5p orbitals, which have a larger radius than the 4f orbital. Thus, only 4f to 4f transitions are generally used in this research field. These transitions are electric dipole forbidden for free ions due to parity selection rules, but the crystal field mixes the parity of the states, allowing for these transitions to occur with the caveat that their strengths are orders of magnitude weaker than transitions between different orbitals like 4f to 5d [133, 134]. Generally, rare-earth ions have lower transition dipole moments than the alkali atoms used for cold atomic or warm vapour based memories. For Rb and Cs, the value is on the order of $10^{-29}$ C·m [138–140], while for the rare-earths ions typically used, most are on the order of $10^{-32}$ C·m [141].

The energy levels of a rare-earth ion doped in a crystal can be described by the following Hamiltonian [132]:

$$H = [H_{\text{FI}} + H_{\text{CF}}] + [H_{\text{HF}} + H_{Q} + H_{\text{EZ}} + H_{\text{NZ}}], \quad (2.1)$$

where $H_{\text{FI}}$ is the free-ion Hamiltonian, $H_{\text{CF}}$ is the crystal field Hamiltonian, $H_{\text{HF}}$ is the hyperfine Hamiltonian describing the interaction between the electron and the nucleus of the ion, $H_{Q}$ is the nuclear quadrupole interaction, and $H_{\text{EZ}}$ and $H_{\text{NZ}}$ are the electronic and nuclear Zeeman interactions, respectively.

Let us consider each term one at a time to see their effects on the level structure. In the first instance, the free-ion term represents the levels of an ion on
2.2 Rare-earth ion-doped crystals

Figure 2.1: Energy levels of Pr$^{3+}$:Y$_2$SiO$_5$, starting from the free-ion levels of the 4f to 4f transitions of a Pr$^{3+}$ ion [142], the crystal field levels of the $^3H_4$ and $^1D_2$ levels in site 1 [143], and the hyperfine levels (with zero applied magnetic field) of their lowest energy crystal field levels [144]. The optical transition of interest, $^3H_4(0)$ to $^1D_2(0)$, is indicated on the diagram. The energy levels are roughly to scale, but only within each manifold of a level.

its own. These energy levels arise from Coulomb interactions of the 4f electrons and spin-orbit coupling. These levels are labelled using the term symbols $^{2S+1}L_J$, where $S$, $L$ and $J$ are the spin, orbital and total angular momenta numbers of the state, and $J = L + S$. Letters are used to refer to $L$, where $S, P, D, F, G, ...$ are used to represent $L = 0, 1, 2, 3, 4, ...$ and so on, continuing alphabetically but omitting $J$.

$S$ and $L$ are good quantum numbers if there is no or very small spin-orbit interaction compared to the Coulomb interaction. However, the Coulomb interaction and spin-orbit coupling are of a similar strength in the lanthanides [133]. Thus $S$ and $L$ are coupled and are no longer good quantum numbers, but $J$ is. The term symbols are still used as identifiers, but depending on the state, they can be
a combination of different terms [134]. The resulting separation between free-ion energy levels is on the order of $10^3$–$10^4$ cm$^{-1}$ (or 10–100 THz) [134].

The levels split further when the ion is doped in a crystal. The electric field of the crystal host lifts the degeneracy of the $J$ manifold, resulting in a number of levels that depend upon the symmetry of the crystal and whether the ion is Kramers or non-Kramers. If the ion is located at a site of low symmetry in the crystal, then for non-Kramers ions, the degeneracy is lifted completely to give $2J + 1$ electronic singlets with an effective $S = 0$. For Kramer ions, the state splits into $J + 1/2$ electronic doublets. In higher symmetry sites, both Kramers and non-Kramers ions can have more degenerate crystal field levels [134]. Here the separation between crystal field levels in the same manifold is around $10^1$–$10^2$ cm$^{-1}$ (or 100–1000 GHz) [134].

The other four terms determine the potential energy levels of individual crystal field levels. Hyperfine interaction comes from the interaction between nuclear and electron spins, and thus generally occurs for Kramers ions which also have non-zero nuclear spin. The nuclear quadrupole interaction arises from interaction between the nuclear spins (where the nuclear spin $I \geq 1$) and the host lattice, f-electrons and distortions of the electric field of the lattice [132]. Both of these terms are responsible for the energy level structure when there is no applied magnetic field, where the splitting between levels can be on the order of 1 MHz to 1 GHz [134].

The electronic and nuclear Zeeman interactions are present if a magnetic field is applied, and require non-zero electronic and nuclear spins, respectively, in the host ion. The strength of these interactions depend on the ion, where Kramers ions experience a larger shift in energy levels compared to non-Kramers ions due to the electron spin degree of freedom. Similarly, the electronic Zeeman term is dominant due to the larger magnitude of the Bohr magneton compared to the nuclear magneton. For example, the shift in Er$^{3+}$:Y$_2$SiO$_5$ is on the order of 10 GHz/T, while for Pr$^{3+}$:Y$_2$SiO$_5$ it is 10–100 MHz/T, depending on the direction of the applied magnetic field.\(^2\)

Figure 2.1 shows the resulting energy level structure from these interactions for Pr$^{3+}$:Y$_2$SiO$_5$ in particular. The next section will cover the structure and properties of this system.

\(^2\)Values calculated using the spin Hamiltonians from Ref. [145] and Ref. [146].
2.3 Praseodymium-doped yttrium orthosilicate

Our REIC of choice is Pr$^{3+}$:Y$_2$SiO$_5$: praseodymium-doped yttrium orthosilicate. Much like there is no ideal physical system to use as a quantum memory, no specific ion is clearly superior among the rare earths and many possess interesting properties. We use praseodymium as it features an appropriate balance of desirable properties for the type of memory we wish to implement, allowing for long storage times, high storage efficiency and a large multiplexing capability. The choice of host crystal is also important as it determines aspects of the level structure and coherence properties of the ions, which will be detailed later in this section.

There are several basic requirements for an ion to be used as a quantum memory, particularly when using the AFC protocol. The transition between crystal field levels should be accessible with a laser. A minimum of three hyperfine levels are required for performing on-demand storage with the AFC, which is only possible (with no applied magnetic field) if the ion has non-zero nuclear spin. The coherence properties must be good enough to allow for sufficiently long storage of photons. The transition should be inhomogeneously broadened, and it must be possible to efficiently optically pump the system to spectrally tailor the absorption of the photon. This ability depends on the optical transition strength and thus the Rabi frequency of the transition, as well as the lifetimes of the optical and spin transitions. The transition strength also affects the efficiency of population transfer of any stored light if it is required by the protocol. Praseodymium fulfills all of these requirements, and in particular without an applied magnetic field, which makes the implementation of a quantum memory much simpler. Additionally, the frequency separation between the hyperfine levels is small, which reduces the bandwidth of a single mode and is thus an advantage in terms of spectral multiplexing as more spectral modes can fit within the inhomogeneous line [60], but makes it more difficult to interface with a compatible source of photons.

The choice of host crystal is important because dopant ions are shielded from the external environment by the crystal, so perturbations from the host itself (i.e., the spins) are a major source of dephasing, and thus should be minimised. The host we use, Y$_2$SiO$_5$, has been extensively investigated and used with many rare-earth dopants [70, 81, 93, 124, 125]. This is because it is considered a “low noise” host as its constituent ions posses a low concentration of nuclear spins: only yttrium has nuclear spin, aside from uncommon isotopes of oxygen and silicon.
The orientation of the crystal can be described using the coordinate system of the principle axes of polarisation: $D_1$, $D_2$ and $b$, where $D_1$ and $D_2$ correspond to the directions where crossed polarisers extinguish light propagated along the crystallographic $b$ axis (i.e., the ⟨010⟩ direction) [147].

In Y$_2$SiO$_5$, yttrium ions are substituted for the dopant ions. The yttrium ions are located at two crystallographically inequivalent sites, both with $C_1$ symmetry (i.e., no rotational symmetry). As the structure around these sites is different, the energy levels of the dopant ions will also differ between the two sites.

Praseodymium has one naturally occurring isotope, $^{141}$Pr with nuclear spin $I = 5/2$. We only address Pr$^{3+}$ ions in the so-called crystallographic site 1 [144]. Figure 2.1 shows the level structure of Pr$^{3+}$:Y$_2$SiO$_5$ at this particular site. As these sites have low symmetry and Pr is a non-Kramers ion, the crystal field splits the free-ion levels into electronic singlets. At around 3 K and below, only the lowest crystal field level of the ground state, $^3H_4(0)$, is populated. We specifically use the optical transition between the lowest energy levels in the $^3H_4$ and $^1D_2$ manifolds, which has a wavelength in the visible region of 605.977 nm [144]. The absorption coefficient of a transition depends on the direction of polarisation, and for site 1, it is the highest for light polarised along $D_2$ [134]. We typically measure a value around 15 cm$^{-1}$ for a 0.05% Pr$^{3+}$-doped sample.

The $^3H_4(0)$ and $^1D_2(0)$ levels have $J = 0$ and thus no electronic spin degree of freedom, so the hyperfine splitting arises from nuclear electronic quadrupole interaction and second-order magnetic hyperfine interaction (also know as pseudo-quadrupole interaction) [144]. Each crystal field level splits into three pairs of degenerate hyperfine states. These levels are conventionally labelled using notation for angular momentum eigenstates of the nuclear spin: $\pm n/2$, where $n = 1, 3, 5$. However, the actual eigenstates do not completely correspond to these labels, and all of the transitions are allowed with varying strengths [144, 148], which were measured experimentally in Ref. [149]. For memory protocols which require a Λ scheme, three levels which are connected with two strong optical transitions are needed. This is best fulfilled with the $\pm 1/2_g \rightarrow \pm 3/2_e$ and $\pm 3/2_g \rightarrow \pm 3/2_e$ transitions, which have relative oscillator strengths of 0.4 and 0.6, respectively [149]. The degeneracy of the hyperfine levels is lifted when a magnetic field is applied due to the nuclear Zeeman effect. Due to the anisotropic nature of the substitutional sites, the shift in energy levels depends on the direction of the magnetic field [148].
In terms of coherence properties of Pr$^{3+}$:Y$_2$SiO$_5$, the optical transition we use has a coherence time of 114 $\mu$s \cite{143}, and the spin transition between $\pm 1/2_g$ and $\pm 3/2_g$ has a coherence time of 500 $\mu$s at zero magnetic field \cite{150}. The latter is sufficient for experiments involving memories separated by tens of km. The coherence time of the spin transition can be increased further to around 1 s by using transitions insensitive to fluctuating magnetic fields, but this requires applying very specific magnetic fields \cite{151}. As a result, this increases the complexity of the hyperfine level structure, but has allowed for storage of classical light for around one minute \cite{73}. The population lifetimes of these transitions are around 164 $\mu$s for the optical transition \cite{143}, and on the order of 10 to 100 s for the spin transitions \cite{144}. Since the spin lifetimes are much longer than the optical lifetime, the absorption line can be spectrally tailored into a structure that can persist for hundreds of milliseconds and longer, which we will make use of in our implementation of a quantum memory, as will be discussed in Chap. 3. Other rare-earth ions will be discussed in Sec. 2.5.

2.4 Spectral broadening

Like any other atom, rare-earth ion dopants have finite linewidths due to sources of broadening. These can be divided into two types of mechanisms: those that affect all ions equally and those that affect each ion individually. The first case results in homogeneous broadening, arising from lattice phonons or fluctuating nuclear or electron spins in the host \cite{132}. Cooling the crystals to around 3 K

\footnote{When extrapolated to zero excitation intensity with no applied magnetic field.}
significantly reduces the effect of phonons on the homogeneous linewidth. The second case results in inhomogeneous broadening. This type of broadening is generally from the crystal itself, where dislocations (irregularities in the crystal structure) and defects (such as impurities or even the dopants themselves, which inevitably distort the crystal structure due to ionic size differences) result in a varying electric field within the crystal, thus shifting the transition frequencies of the ions.

Figure 2.2 shows the effect of the broadening mechanisms on the spectrum with which we work. The individual ions have narrow homogeneous linewidths, which have varying central transition frequencies due to inhomogeneous effects. As a result, if we look at the absorption line of an ensemble of ions, we only see the overall distribution of these frequencies. For Pr\(^{3+}:Y_2SiO_5\) at a concentration of 0.05\%, for the optical transition, we typically measure an inhomogeneous linewidth of 10 GHz which has a Gaussian profile \([91]\), while the homogeneous linewidth can be as low as a few kHz \([143]\). Chapter 3 will go further into the mechanisms affecting the homogeneous linewidth, and explain how we can tailor the absorption line for use as a quantum memory.

### 2.5 Other rare-earth ions for quantum memories

As mentioned in Chap. 1, many different rare-earth ions have been investigated for use as quantum memories. In this section I will review some of their advantages and disadvantages.

Europium in particular is similar to praseodymium in terms of its level structure (a non-Kramers ion with \(I = 5/2\)), and also features much longer optical and hyperfine coherence times than Pr \([152, 153]\). However, it has some disadvantages. Its population lifetime is also longer (around 2 ms) \([152]\), which limits the rate of photon storage in certain protocols\(^4\), thereby also reducing the entanglement generation rate. The ratio of its inhomogeneous linewidth to hyperfine splitting is smaller than Pr \([154, 155]\), which reduces its spectral multiplexing capacity. Since it has a longer optical coherence time, it has a high temporal multiplexing capacity for optical storage, but it has a lower Rabi frequency due to having a notably lower transition dipole moment compared other rare-earth ions.

\(^4\)Namely when performing spin-wave storage with the AFC protocol, where the time between storage trials needs to be at least as long as the lifetime to minimise background noise from fluorescence from previous storage trials.
mentioned in this section. The transition dipole moment of $^{153}\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ is $5.0 \cdot 10^{-33} \text{ C} \cdot \text{m}$ [156], while it is $2.6 \cdot 10^{-32} \text{ C} \cdot \text{m}$ for $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$ [157] and the same order of magnitude for the other ions when doped in $\text{Y}_2\text{SiO}_5$ [141, 158].

A lower Rabi frequency limits population transfer to the hyperfine levels, and reduces its temporal multiplexing capability when used for spin-wave storage [60]. The only other non-Kramers ion used, thulium, does not allow for on-demand storage, but does feature long optical coherence times [159], and can be used for extensive multiplexing [36].

Kramers ions generally have larger transition strengths, and a larger splitting between hyperfine levels (which increases the memory bandwidth). However, they tend to have shorter spin lifetimes due to the magnetic moments of the dopant ions increasing dephasing in the system. As a result, efficient optical pumping challenging as the spin lifetime needs be much longer than the optical lifetime.

Amongst the Kramers ions, erbium stands out due to its telecom C-band optical transition, which has been shown to have a very long optical coherence time [58]. It has one isotope with non-zero nuclear spin where $I = 7/2$ (thus allowing for on-demand storage in principle, although no demonstration has been shown yet) which further complicates optical pumping due to the large number of resulting hyperfine levels (16), but has been shown to have a long spin coherence time [160]. Large magnetic fields are required in order to obtain good optical and spin coherence properties. Neodymium has also been used, but it has similar characteristics as erbium regarding its level structure. There are strategies for improving the optical pumping performance for both ions [161, 162]. Ytterbium is the most recent ion to be explored as a quantum memory, which appears very promising. Unlike the other Kramers ions, it can have the most simple hyperfine structure (4 levels) while also possessing nuclear spin, as the $^{171}\text{Yb}$ isotope has $I = 1/2$. It has been shown to have good optical and spin coherence properties without applying a magnetic field [163], and its level structure allows for the storage of wider bandwidth photons than non-Kramers ions like Pr or Eu [81], though its performance at the single photon level with on-demand storage has yet to be investigated.

One final point of comparison between the different ions is the aforementioned transition dipole moment. It is proportional to the square root of the oscillator strength, which is in turn related to the optical depth (OD) of the transition such that $f \propto \frac{1}{N} \int k(v)dv$ where $f$ is the oscillator strength, $N$ is the population of ions,
$k$ is the absorption coefficient, and $\nu$ is the frequency [164]. The OD depends on the oscillator strength (via the absorption coefficient) of the transition as well as the concentration of ions in the crystal. It is thus beneficial to have a higher transition dipole moment not just for ease of optical pumping, but also for higher OD. Increasing the doping can increase the OD to a point, but the inhomogeneous broadening will also increase, so the peak OD of the transition will not simply increase linearly. For cold atomic gases, the OD is not limited in this way and can reach values of up to 500 [97].
Chapter 3

Theoretical background

This chapter contains the theoretical background relevant to this thesis. The first part concerns light-matter interaction, introducing the basics of semiclassical radiation theory and the optical Bloch equations. The population lifetime and coherence time are introduced, as well as how they are measured, and the sources of dephasing in Pr\(^{3+}\):Y\(_2\)SiO\(_5\). The subsequent sections describe spectral hole-burning and our memory protocol, the atomic frequency comb, which are also aspects of light-matter interaction.

The final two sections present an overview of the single photon statistics that we make use of in our experiments, and an introduction to entanglement and how we measure it in the context of the experimental work in this thesis. The experimental implementation of the memory, and method of entanglement analysis will be introduced in the follow chapter, Chap. 4.

3.1 Light-matter interaction

This section will briefly introduce concepts related to optical resonance, to understand some of the coherent phenomena that arise in atomic systems like REICs when interacting with an applied electric field. The content of the first three subsections follows and summarises Ref. [165],\(^1\) unless stated otherwise.

\(^1\)Other treatments can be found in Refs. [166, 167].
3.1.1 Semiclassical radiation theory

In the following, we will consider a two-level system coupled to an external electric field (i.e., a laser). This description is analogous to that of a particle, or ensemble of particles, with a magnetic dipole moment and thus non-zero spin, coupled to a magnetic field in the theory of magnetic resonance [168]. In this case, we take an (artificial) two-level atom, that is similarly treated as a pseudo “spin”.

The applied field has the form

\[ E \propto \mathcal{E} e^{i\omega t}, \]  

(3.1)

where \( \mathcal{E} \) is the amplitude of the field, and \( \omega \) is its frequency, which is nearly resonant with the transition frequency of the two-level system, \( \omega_0 \). The resulting Hamiltonian for this interaction has the form

\[ \hat{\mathcal{H}} = \hat{H}_\text{A} - \hat{d} \cdot \hat{E}(r_0), \]  

(3.2)

where \( \hat{H}_\text{A} \) is the unperturbed Hamiltonian of the two-level system, \( \hat{d} \) is the dipole moment operator of the atom and \( \hat{E}(r_0) \) is the electric field operator evaluated at the position \( r_0 \).

Equation 3.2 can be rewritten in terms of the Pauli matrices \( \sigma_x, \sigma_y, \) and \( \sigma_z \), and the identity matrix \( I \) as

\[ \hat{\mathcal{H}} = \frac{1}{2} (W_+ + W_-) \hat{I} + \frac{1}{2} (W_+ - W_-) \hat{\sigma}_z - (\hat{d}_r \cdot \hat{E}) \hat{\sigma}_x + (\hat{d}_i \cdot \hat{E}) \hat{\sigma}_y, \]  

(3.3)

where \( W_+ \) and \( W_- \) are the upper and lower energy levels of the atom (i.e., the eigenvalues of \( \hat{H}_\text{A} \)), such that \( \omega_0 \equiv (W_+ - W_-)/\hbar \), and \( \hat{d}_r \) and \( \hat{d}_i \) are the real and imaginary components of \( \hat{d} \), respectively.

We are interested in how this system evolves with time. The time dependence for an operator \( \hat{O} \) can be calculated using \( i\hbar \dot{\hat{O}} = [\hat{O}, \hat{\mathcal{H}}] \). From this, we can obtain a set of simultaneous differential equations in terms of the Pauli matrix operators,
as well as the dipole and electric field operators:

\[
\begin{align*}
\dot{\sigma}_x(t) &= -\omega_0 \hat{\sigma}_y(t) + \frac{2}{\hbar} [\mathbf{d}_r \cdot \hat{\mathbf{E}}(t)] \hat{\sigma}_z(t), \\
\dot{\sigma}_y(t) &= \omega_0 \hat{\sigma}_x(t) + \frac{2}{\hbar} [\mathbf{d}_r \cdot \hat{\mathbf{E}}(t)] \hat{\sigma}_z(t), \\
\dot{\sigma}_z(t) &= -\frac{2}{\hbar} [\mathbf{d}_r \cdot \hat{\mathbf{E}}(t)] \hat{\sigma}_y(t) - \frac{2}{\hbar} [\mathbf{d}_i \cdot \hat{\mathbf{E}}(t)] \hat{\sigma}_x(t). 
\end{align*}
\]

(3.4a)

While these equations do not have very general solutions, with certain assumptions they can be recast in a more usable form.

The first such assumption is that quantum correlations between the field and the atom are not important. Then, by taking expectation values, the products of operators can be factored such that

\[
\langle \hat{\mathbf{E}}(t) \hat{\sigma}_z(t) \rangle = \langle \hat{\mathbf{E}}(t) \rangle \langle \hat{\sigma}_z(t) \rangle.
\]

(3.5)

Then, in semiclassical radiation theory, \( \langle \hat{\mathbf{E}}(t, \mathbf{r}_0) \rangle \) is taken to be a classical electric field. This assumption is sufficient for the phenomena we are interested for this thesis.

The expectation values of the Pauli matrix operators can be written as

\[
s_i(t) \equiv \langle \hat{\sigma}_i(t) \rangle, \quad i \in \{x, y, z\}.
\]

(3.6)

Note that the state remains normalised in time, i.e., \( s_x^2(t) + s_y^2(t) + s_z^2(t) = 1 \), so \( s(t) \) is a unit vector.

If we assume that the transition between the states of the two-level atom is \( \Delta m = 0 \), then we can discard the imaginary components of the equations for \( s(t) \). As a result, we obtain this set of semiclassical equations:

\[
\begin{align*}
\dot{s}_x(t) &= -\omega_0 s_y(t), \\
\dot{s}_y(t) &= \omega_0 s_x(t) + \kappa E(t, \mathbf{r}_0) s_z(t), \\
\dot{s}_z(t) &= -\kappa E(t, \mathbf{r}_0) s_y(t),
\end{align*}
\]

(3.7a)

(3.7b)

(3.7c)

where \( E \) is the component of \( \mathbf{E} \) along the unit vector in the direction of \( \mathbf{d}_r \), and \( \kappa \) is defined as

\[
\frac{\hbar \kappa}{2} \equiv d,
\]

(3.8)

where \( d \) is the magnitude of the dipole matrix element of the transition.
These equations for electric dipoles are analogous to those governing spin precession in magnetic resonance, so $s(t)$ is referred to as the electric-dipole “pseudospin” vector [165]. Thus Eq. 3.7 can be rewritten as if a torque $\Omega^F$ was acting on a solid body $s(t)$, taking the form

$$\frac{d}{dt}s(t) = \Omega^F (t) \times s(t),$$

(3.9)

where

$$\Omega^F (t) = (-\kappa E, 0, \omega_0).$$

(3.10)

The applied field $E(t)$ is oscillating at a frequency $\omega$, which is close to $\omega_0$, so $\Omega^F$ is changing rapidly with respect to $s(t)$. In this situation, it is difficult to analyse the behaviour of the pseudospin vector. For simplification, we can change the reference frame to one that rotates at the frequency $\omega$. To do so, we first rewrite $\Omega^F$ as the sum of three components:

$$\Omega^F = \Omega^+(t) + \Omega^-(t) + \Omega^0,$$

(3.11)

where

$$\Omega^0 = (0, 0, \omega_0),$$

(3.12a)

$$\Omega^+ = (-\kappa E \cos(\omega t), -\kappa E \sin(\omega t), 0),$$

(3.12b)

$$\Omega^- = (-\kappa E \cos(\omega t), +\kappa E \sin(\omega t), 0).$$

(3.12c)

Here $\Omega^+(t)$ and $\Omega^-(t)$ rotate circularly in the $x - y$ plane, in right-handed and left-handed directions, respectively, and $\Omega^0$ is a static component in the $z$ direction. We apply the rotating wave approximation, where we move to a reference frame where $\Omega^-(t)$ is essentially negligible\(^2\), so we can use $\Omega^F = \Omega^+(t) + \Omega^0$. This frame follows $s(t)$, which is rotating around the $z$ axis in a right-hand direction with angular velocity $\omega$, while oscillating sinusoidally.

We define a new pseudo-spin vector $\rho = (u, v, w)$, which is a unit vector stationary in the rotating frame, which relates to $s$ with the appropriate rotation

\(^2\)See Sec. 2.6 in Ref. [165] for the effect of $\Omega^-(t)$ on the resonance frequency.
3.1 Light-matter interaction

matrix. It follows this set of equations of motion:

\[
\begin{align*}
\dot{u} &= -(\omega_0 - \omega)v, \\
\dot{v} &= (\omega_0 - \omega)u + \kappa E w, \\
\dot{w} &= -\kappa E v.
\end{align*}
\]

(3.13)

Under this approximation, all variables change slowly over time, and the frequency of the field is near resonance, so \(|\omega_0 - \omega| \ll \omega_0\).

The components of \(\rho\) have a particular physical significance: \(w\) is the population difference of the atom, known as the inversion, while \(u\) and \(v\) are related to the dipole moment of the transition as the components in-phase and in-quadrature with the field \(E\) (or the dispersive and absorptive components of the dipole moment), respectively.

Note that compared with the theory governing a classical oscillator, the inversion is unique to the quantum (or in this case, semiclassical) theory.

3.1.2 Rabi frequency

The simplest solution to Eq. 3.13 occurs when the field is on resonance with the atoms, where \(\Delta = \omega_0 - \omega\).

We define a dimensionless quantity \(\theta(t)\) such that

\[
\theta(t) = \int_{-\infty}^{t} \kappa E(t') dt',
\]

(3.14)

which leads to the following solution of Eq. 3.13:

\[
\begin{align*}
u(t;0) &= u_0, \\
v(t;0) &= w_0 \sin \theta(t) + v_0 \cos \theta(t), \\
w(t;0) &= -v_0 \sin \theta(t) + w_0 \cos \theta(t),
\end{align*}
\]

(3.15)

where \(u_0 = u(t = 0; \Delta = 0)\), etc.

Physically, \(\theta(t)\) is the angle of \(\rho\) from \(w\), as indicated on the Bloch sphere shown in Fig. 3.1. If \(\rho\) is at \(w = -1\), the atom is in the lower energy level, or if \(w = +1\), the atom is in the higher energy level. From Eq. 3.15, we can see that \(\rho\) will rotate around \(u\), so that the atom cycles between the two energy levels. Note that the Bloch sphere is often used not just to depict the state of an atom.
but also qubits, where in both cases the north and south poles are the states $|1\rangle$ and $|0\rangle$, and the equator is an equal superposition between the two of the form $(|0\rangle + e^{i\phi} |1\rangle)/\sqrt{2}$.

If the applied field has a constant value $\mathcal{E}_0$ in the interval of time between $t_1$ and $t_2$, then integrating Eq. 3.14 gives

$$\theta(t) = \kappa \mathcal{E}_0 (t_2 - t_1) = \Omega(0)(t_2 - t_1),$$

(3.16)

where $\Omega(0)$ is the frequency at which the atom cycles between the two energy levels with the applied field, commonly referred to as the Rabi frequency on resonance. If the field is not on resonance, the solution becomes more complicated. As part of the solution (see Ref. [165]), we can define the generalised Rabi frequency as

$$\Omega(\Delta) = \sqrt{\Delta^2 + (\kappa \mathcal{E}_0)^2}.$$  (3.17)

If the atom is initially in the ground state, then after some time $\delta t$, where $\kappa \mathcal{E}_0 \delta t = \pi$, then the atom will be in the excited state. A pulse that fulfills this condition is called a $\pi$-pulse. Similarly, a $\pi/2$-pulse will place the atom in a superposition state between the two levels.

Experimentally, we measure the Rabi frequency using optical nutation. By sending a sufficiently long square pulse of many multiples of $\pi$, the light after the atom (or atoms in our case) will be modulated in amplitude at a frequency related to the Rabi frequency. For inhomogeneously broadened systems, the modulation follows $\Omega t_\pi \sim 5.1$, where $t_\pi$ is the length of the $\pi$-pulse, i.e., the length of time required to achieve an inversion of population with the pulse (see Ref. [169], in relation to Ref. [170]).
3.1 Light-matter interaction

3.1.3 Optical Bloch equations

Damping mechanisms will affect both the occupation of the atomic state and its coherence. To account for this, we introduce the constants $T_1$ and $T_2$, where $T_1$ is the lifetime of the higher energy level and $T_2$ is the coherence time of the two-level state. These constants can be incorporated into Eq. 3.13 to give:

$$
\dot{u} = -\Delta v - \frac{u}{T_2},
$$

(3.18a)

$$
\dot{v} = \Delta u - \frac{v}{T_2} + \kappa \mathcal{E} w,
$$

(3.18b)

$$
\dot{w} = -\frac{w - w_{eq}}{T_1} - \kappa \mathcal{E} v,
$$

(3.18c)

where $w_{eq}$ is the value the inversion $w$ relaxes to when $\mathcal{E} = 0$. This set of equations is known as the optical Bloch equations, analogous to the Bloch equations originally derived for magnetic resonance\(^3\).

These equations can be solved explicitly for a steady field, i.e., $\mathcal{E}$ is constant. The solutions are not included here, but can be found in Ref. [165], or in Ref. [171] for the original derivation for magnetic resonance.

The following subsections will go into further detail about these constants, how we measure them, and their significance for quantum memories.

3.1.4 Population lifetime: $T_1$

The population lifetime $T_1$ refers to a measure of time that an ion remains in a particular state. For optical transitions, this can be measured by exciting population to the excited state, and observing the emission of fluorescence from the transition, which decays exponentially. The lifetime is given by the decay constant of the fluorescence counts. For the $^1D_2(0)$ level of site 1 of Pr$^{3+}$:Y$_2$SiO$_5$, this value is $164 \pm 5 \mu$s, which comes primarily from radiative decay [143].

For the hyperfine levels, $T_1$ can be measured using spectral holeburning (see the following section), where a spectral feature decays according to the lifetime. This often not a single exponential decay, with the decay process depending on different types of interactions. For example, in Ref. [172] a double exponential decay was observed in Pr$^{3+}$:Y$_2$SiO$_5$, with decay times of 2 s and 35 s, and the different decays were attributed to Pr-Y and Pr-Pr interactions for the short decay,

\(^3\)Note that in Ref. [165], $T_2'$ is used instead of $T_2$. We use $T_2$ to be consistent with the typical terminology for the coherence time measured with spin echoes.
3. Theoretical background

Figure 3.2: The two-pulse photon echo, with the upper diagram indicating the temporal sequence of pulses, and the lower diagram indicating the behaviour of the ion ensemble during the process, represented on the Bloch sphere. The numbers on both diagrams indicate equivalent parts of the sequence.

and an Orbach process (a two-phonon transition to other crystal field levels [173]) for the long decay. A longer decay time on the order of hundreds of seconds has also been observed in Pr$^{3+}:Y_2SiO_5$ [144, 174]. For the implementation of the quantum memory in this thesis, the initial fast decay is the most critical one due to the duty cycle of the experiment.

3.1.5 Coherence time: $T_2$

The coherence time, $T_2$, of a transition is a measure of how long the state retains a well-known phase [167]. It is related to the homogeneous linewidth of a transition according to:

$$\Gamma_h = \frac{1}{\pi T_2}.$$  (3.19)

The homogeneous linewidth of a transition can be probed within an inhomogeneously broadened line through spectral hole burning. However, the resulting lineshape will be broadened by other factors such as the laser linewidth as well as power broadening, which is broadening of a spectral line that occurs with increasing laser intensity due to saturation effects [165]. These factors make it difficult to determine the homogeneous linewidth precisely.

To avoid these effects, the coherence time can be directly measured using the two-pulse echo technique, as shown in Fig. 3.2. Let us consider two levels $|0\rangle$ and $|1\rangle$, which correspond to the north and south poles of the Bloch sphere, respectively. The ensemble of ions is initially in the $|0\rangle$ state (step 1). By applying
a $\pi/2$ pulse, the ensemble of ions is shifted to a superposition state between $|0\rangle$ and $|1\rangle$, i.e., to the equator of the Bloch sphere (step 2). The ions start to dephase at different rates due to inhomogeneous broadening (step 3), which also results in coherent emission called free induction decay (FID). If a $\pi$ pulse is applied after some time $t$, then the ensemble of Bloch vectors will be rotated by $\pi$ radians across the equatorial plane such that is mirrored, but instead of the ions dephasing away from each other, they are now rephasing (step 4). At a time $t$ after the $\pi$ pulse, the ensemble will have rephased and will emit an echo (step 5).

This technique works for both optical and hyperfine transitions. It is referred to as the photon echo for an optical transition, and the spin echo for a hyperfine transition. The only difference between the two is the type of field used for pulsing: either optical, or radio-frequency. Spin echoes in magnetic resonance were the first to be observed [175], and photon echoes followed after [176]. We only perform photon echo measurements in this thesis.

Increasing the time $t$ between the two pulses will result in a decay of the echo intensity. The intensity $I$ of the echo decays exponentially (unless there are interactions with host ions or spectral diffusion), following the formula

$$I(t) = I_0 e^{-4t/T_2},$$

where $I_0$ is the echo intensity at $t = 0$. From this, the $T_2$ of the transition can be extracted. If the amplitude is measured instead, then the decay is proportional to $e^{-2t/T_2}$ [132]. Note that the protocol is also robust with pulse area, as the original measurements performed in Ref. [175] used pulses of equal area.

The coherence time and population lifetime are related according to [165, 167]

$$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{1}{T_2'},$$

(3.21)

where $T_2'$ is the pure dephasing, accounting for the different processes which can limit $T_2$. In the absence of pure dephasing, the coherence time is purely limited by the population lifetime such that $T_2 = 2T_1$.

Equation 3.21 can be written in terms of the homogeneous linewidth, with the different contributions to the pure dephasing written explicitly, as [143]

$$\Gamma_h = \Gamma_{\text{pop}} + \Gamma_{\text{ion-spin}} + \Gamma_{\text{ion-ion}} + \Gamma_{\text{phonon}},$$

(3.22)
where $\Gamma_{\text{pop}}$ comes from the population lifetime$^4$, $\Gamma_{\text{ion-spin}}$ comes from spin fluctuations of the host lattice ions, $\Gamma_{\text{ion-ion}}$ comes from ion-ion interactions (where the local environment changes due to excitation or relaxation of nearby ions), and $\Gamma_{\text{phonon}}$ comes from temperature-dependent phonon scattering.

The term $\Gamma_{\text{phonon}}$ is negligible at 1.4 K, where phonon scattering is frozen out$^5$. The next term, $\Gamma_{\text{ion-spin}}$, depends on the constituents of the host matrix. As mentioned in Chap. 2, $Y_2SiO_5$ has a particularly low (or no) magnetic moments in its constituent ions, which is important for minimising this term.

In the case of $Pr^{3+}:Y_2SiO_5$, the $\Gamma_{\text{ion-ion}}$ term is particularly significant, which comes from instantaneous spectral diffusion$^5$. Laser pulses can cause frequency shifts in the ions due to dipole-dipole coupling, which disrupts the rephasing of the echo. The strength of this dephasing depends on the number of ions excited, i.e., the intensity of the pulse, so the intensity must be minimised in order to measure long values of $T_2$. For site 1, $T_2 = 114 \, \mu s$ when extrapolated to zero excitation intensity and at 1.4 K$^4$, and in the lab we have measured a value of $116 \pm 6 \, \mu s$. Applying a magnetic field can also improve the $T_2$ slightly by suppressing spin flips ($\Gamma_{\text{ion-spin}}$) from the host yttrium ions$^4$.

While not measured here, the spin coherence time of the $\pm 1/2_g \rightarrow \pm 3/2_g$ transition is $500 \, \mu s$ $^5$. This is limited by magnetic field fluctuations at the Pr ions. To increase the coherence time, specific magnetic fields can be applied to the crystal such that the energy level structure contains transitions which have frequencies that do not have a first-order dependence on magnetic field $^6$. These are called zero-first order Zeeman shift (ZEFOZ) transitions.

### 3.2 Spectral holeburning

Spectral holeburning is a technique that can be used to shape the absorption profile of a transition by transferring ions to different energy levels. Various quantum memory protocols rely on creating spectral features in this way, including the atomic frequency comb which is used in this thesis and described in Sec. 3.3.

$^4\Gamma_{\text{pop}} = 1/(2\pi T_1)$

$^5$It should also be negligible, or close to it, at 3 K, which we work at. Otherwise, there is a strong dependence of linewidth to the temperature $T$ proportional to $T^7$ due to a Raman scattering phonon process$^1$ for equilibrium phonons. However, Ref. $^1$ suggests that non-equilibrium phonons may still contribute to $\Gamma_h$ for site 1 of $Pr^{3+}:Y_2SiO_5$ specifically.
3.2 Spectral holeburning

Figure 3.3: Illustration of spectral holeburning for a fictitious 3-level system where the inhomogeneous linewidth is wider than the homogeneous linewidth. Levels 1 and 2 are separated in energy by $\Delta g$, and levels 2 and 3 by $\Delta f$. A laser with frequency $\nu$ optically pumps ions out of one level, which relax into the other hyperfine level. Afterwards, if the laser is resonant with $\Delta f$, a hole is formed in the absorption line due to the removal of population. There are antiholes at $\Delta f \pm \Delta g$, where the laser is resonant to the transitions where there is population, which differs depending on the ion class (I or II).

A laser resonant with an inhomogeneously broadened transition can redistribute the population at that particular frequency to other energy levels if the laser is narrower than the transition. Figure 3.3 illustrates this process for a fictitious system which has two ground-state (e.g. hyperfine) levels 1 and 2 and an optically excited state 3. If the laser frequency $\nu$ is equal to the splitting between levels 2 and 3 ($\Delta f$), then the population in 2 will be excited to 3, and will decay to 1 or 2. With sufficient pumping, the population can all be transferred to level 1. Then, when $\nu$ is swept along the absorption line, there will be a drop in absorption at $\Delta f$, which is called a spectral hole. There will be an increase in absorption at $+\Delta g$ (which we call a spectral antihole) due to the increase in population of this level\(^6\).

If the inhomogeneous linewidth of the optical transition is wider than the the hyperfine splitting $\Delta g$, there will be another class of ions where the transition

\(^6\)A simpler system without multiple ground-state levels could use the optically excited state or a different electronic state for shelving the population instead [132], if their lifetimes are sufficiently long. In Pr\(^{3+}\):Y\(_2\)SiO\(_5\), this is not useful since we want to keep the population in the ground-state levels, and the optical lifetime is orders of magnitude shorter than the ground-state lifetime.
frequency between 2 and 3 is different, as illustrated in Fig. 3.3. For this class labelled II (class I corresponds to the first two level diagram), level 1 has been emptied instead, while level 2 experienced an increase in population. The transition between 2 and 3 is now $\Delta f - \Delta g$, which means there will be another antihole at this frequency, as shown in the absorption diagram. To measure the resulting structure, a probe beam is swept across the absorption line with very low power such that it does not redistribute the population of the ions\(^7\).

The hole and antihole structure will decay, following the lifetime of the ground-state levels. In order to efficiently transfer population in levels in such a way that one level is completely emptied and/or another contains all of the ion population, the lifetime of the hyperfine levels must be much longer than that of the optically excited state. While this is the case in $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$, it is not true for all REICs.

Similarly, the level structure of REICs is not as simple as that given in this explanation. At zero magnetic field, $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$ has 3 hyperfine levels in both the ground and optically excited states, and a much wider inhomogeneous linewidth than hyperfine splitting, so a laser probing the optical transition will be resonant to all 9 possible transitions, resulting in 9 classes of ions as shown in

\(^7\)We typically use around 60 nW with our samples of $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$.  

*Figure 3.4: Level diagram of the 9 ion classes for site 1 of Pr$^{3+}$:Y$_2$SiO$_5$. 
3.3 Atomic frequency comb

Throughout all of the experimental work presented here, we use the atomic frequency comb (AFC) protocol to store photons [50]. To do so, we prepare a particular spectral feature in the inhomogeneous absorption line, which takes the form of a periodic structure of alternating high to no absorption which resembles a comb. The shape of the structure and how it is implemented in Pr$^{3+}$:Y$_2$SiO$_5$ is shown in Fig. 3.5. Using spectral holeburning, the comb is prepared in the

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Fig. 3.4. The holeburning spectrum will be more complicated, and thus polarising all the ions in one level or emptying a level will require a carefully chosen pulse sequence to redistribute the population. One such method is described in Ref. [149], and the procedure we follow can be found in App. A. As a result, we transfer most of the population to the $\pm 1/2_g$ level, and the $\pm 3/2_g$ level is emptied.

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Figure 3.5: AFC and SW storage using Pr$^{3+}$:Y$_2$SiO$_5$. (a) Level diagram of Pr$^{3+}$:Y$_2$SiO$_5$ with the relevant transitions and energy levels labelled. (b) Spectral structure of a single class AFC that is also usable for SW storage. (c) Temporal diagram of AFC storage, indicating the positions of the input and echo. (d) Temporal diagram of SW storage, indicating the positions of the input, control pulses and echo. Acronyms and labels are defined in the main text.

8See Sec. 5.3.1 for an example of an AFC structure implemented in our experiments.
±\(1/2g\) level labelled \(|g\rangle\), which consists of peaks separated by a spacing of \(\Delta\). If a single photon is sent to the crystal resonant with the \(±1/2g \rightarrow ±3/2e\) transition (labelled \(|g\rangle \rightarrow |e\rangle\), this results in an excitation that is a superposition of each individual ion \(k\) in the ensemble of \(N\) ions excited to \(|e\rangle\) with the rest of the ensemble in the ground state \(|g\rangle\). This excitation can be represented by the Dicke state:

\[
\sum_{k=1}^{N} e^{-i2\pi \delta_k t}c_k |g_1 ... e_k ... g_N\rangle ,
\] (3.23)

where \(\delta_k\) is the frequency detuning of an ion \(k\) from the central frequency of the input photon and \(c_k\) are the amplitudes which depend on the absorption frequency and spatial position of the ion \(k\). The term \(\delta_k\) is an integer multiple of the comb tooth spacing \(\Delta\) such that each atom dephases according to \(\delta_k = m_k \Delta\). The absorption of the single photon occurs at \(t = 0\), and though the ions dephase, they will rephase at \(t = 1/\Delta = \tau\), which results in a coherent emission of the photon as what is called the AFC echo [Fig. 3.5(c)]. The efficiency of this emission (i.e., the storage efficiency) is at most 54% in the forward direction due to reabsorption of the photon by the ions [50]. By using an impedance-matched cavity, the efficiency can approach 100% [178]. No noise should be generated by this process.

With this method of storage, the AFC only functions as a delay line for single photons. To make the storage and retrieval on-demand, the excitation can be transferred from an optical excitation to a hyperfine transition in the ground state by applying a strong control pulse before the AFC echo is emitted at \(t = \tau\). In our implementation, the control pulse is applied on the \(±3/2g \rightarrow ±3/2e\) transition (\(|e\rangle \rightarrow |s\rangle\)). The \(|s\rangle\) level must be emptied of population, and in this case any excess population is moved to the \(±5/2g\) level labelled \(|aux\rangle\). The dephasing of the Dicke state from Eq. 3.23 is then suspended, and becomes a collective excitation (a spin-wave) between the spin state \(|s\rangle\) and \(|g\rangle\) of a similar form:

\[
\sum_{k=1}^{N} c_k |g_1 ... s_k ... g_N\rangle ,
\] (3.24)

The excitation can be retrieved on-demand back to \(|e\rangle\) by applying another control pulse on the \(|e\rangle \rightarrow |s\rangle\) transition. The evolution of the phases in Eq. 3.23 resumes, and leads to the emission of what we call the spin-wave (SW) echo. The total storage time is then \(T_s + \tau\), where \(T_s\) is the time between the applied control pulses and thus the time spent in the spin-wave excitation, as shown in Fig. 3.5(d). If
backward retrieval is implemented, the efficiency of this protocol can theoretically reach 100% [50].

Unlike optical storage alone, significant noise is generated by SW storage with the application of the control pulses. As the control pulses are often very close in frequency to the input photons (in Pr$^{3+}$:Y$_2$SiO$_5$, the separation is 10.2 MHz), so leakage from the control field must be minimised with filtering. The $|s\rangle$ level must be empty, as any residual population can be excited with the control pulses, which leads to fluorescence when the ions decay to other crystal field levels in the ground-state manifold manifold, or to any of the hyperfine levels in the ground state, and is thus a major source of noise for the protocol at the single photon level. Filtering this noise is a particularly challenging aspect of implementing SW storage, as will be addressed in the following chapters. FID from the control pulses can also contribute to the noise, but it decays on a shorter timescale than the fluorescence and if it is present in the measurements presented in Chap. 5, it is likely already filtered by the setup. We attribute the remaining noise to be fluorescence.

The approximate spectral structure we need to create in Pr$^{3+}$:Y$_2$SiO$_5$ follows Fig. 3.5. We are only concerned with the final structure in the spectral region containing the three transitions highlighted in the figure; the rest of the spectrum will have some combination of holes and antiholes as a result of optical pumping. We use the levels $\pm 1/2_g$, $\pm 3/2_g$, and $\pm 3/2_e$, as they form a Λ scheme with good transition strengths as previously noted in Chap. 2, with the stronger transition $\pm 3/2_g \rightarrow \pm 3/2_e$ used for the control pulses. An AFC is prepared in $|g\rangle$, so a comb structure is visible for both the $\pm 1/2_g \rightarrow \pm 3/2_e$ and $\pm 1/2_g \rightarrow \pm 5/2_e$ transitions, albeit with a lower optical depth for the second transition as it has a weaker transition strength. We use the $\pm 3/2_g$ level as $|s\rangle$, and the $\pm 3/2_g \rightarrow \pm 3/2_e$ transition for the control pulses, so this spectral region should not contain any population, including from other ion classes. From this picture we can also see that the maximum width of the comb is bounded by the frequency separation of the excited states $\pm 1/2_e$ and $\pm 5/2_e$ from $\pm 3/2_e$, which is around 4 MHz for Pr$^{3+}$:Y$_2$SiO$_5$.

The expected efficiency for a particular AFC structure depends on the OD, background absorption and finesse of the comb. Deviations from the ideal comb structure due to imperfect comb preparation can lead to a reduced AFC efficiency. The expected efficiency for an AFC echo retrieved in the forward direction is
given by \([179, 180]\)

\[
\eta_{AFC} = \tilde{d}^2 e^{-\tilde{d} d} e^{-d_0} \eta_{\text{deph}},
\]  

(3.25)

where \(\tilde{d}\) is the effective optical depth given by \(d/F\), \(d\) is the peak OD of the comb teeth, \(F\) is the finesse of the comb given by \(F = \Delta/\gamma\) where \(\gamma\) is the linewidth of a tooth, and \(d_0\) is the absorption background which can come from imperfect optical pumping, or is intrinsic to the crystal. The first two terms give the probability that the input photon is absorbed, stored and re-emitted by the atoms of the comb, while the third term accounts for the loss of photons due to the absorption background. The term \(\eta_{\text{deph}}\) is the intrinsic dephasing factor of the comb teeth that depends on their shape, which is \(e^{-7/F^2}\) for Gaussian teeth \([50]\), and \(\text{sinc}^2(\pi/F)\) for square teeth \([181]\). The latter gives a higher efficiency, but in practice it is difficult to create perfectly square spectral structures.

If SW storage is used, backward retrieval of the echo is possible through the use of counterpropagating control beams, and the resulting efficiency is \([50, 180]\)

\[
\eta_{SW} = (1 - e^{-\tilde{d}})^2 e^{-d_0} \eta_{\text{deph}},
\]  

(3.26)

which can approach 100% efficiency. A very high \(d\) (greater than \(\sim 20\)) combined with a large finesse \((\sim 10)\) is required to achieve high efficiencies \([50]\). Such optical depths are not common REICs, so the experiments performed thus far have used retrieval in the forward direction. In that case, the efficiency is given by

\[
\eta_{SW} = \eta_{AFC} \eta_T^2 \eta_C,
\]  

(3.27)

where \(\eta_{AFC}\) is the efficiency of the AFC from Eq. 3.25, \(\eta_T\) is the transfer efficiency of a single control pulse, and \(\eta_C\) denotes the proportion of the signal remaining after decoherence from the inhomogeneous broadening of the hyperfine levels. The decay will follow the Fourier transform of the line shape of the spin transition, which in the case of \(\text{Pr}^{3+}:Y_2\text{SiO}_5\) (and \(\text{Eu}^{3+}:Y_2\text{SiO}_5\)) is Gaussian and is thus is given by \([182]\)

\[
\eta_C = \exp \left( - \frac{(T \gamma_{\text{inhom}} \pi)^2}{2 \ln(2)} \right),
\]  

(3.28)

where \(\gamma_{\text{inhom}}\) is the inhomogeneous linewidth of the transition used for SW storage. In our experimental setup, we typically measure a \(\gamma_{\text{inhom}}\) of around 15 kHz for the \(\pm 1/2_g \rightarrow \pm 3/2_g\) transition (see Chap. 5). This limits storage in the hyperfine levels to tens of microseconds, but spin echoes can be used to overcome the
dephasing due to the inhomogeneous broadening and instead follow the limit imposed by the coherence time, or it can be increased further with dynamical decoupling techniques [45, 183].

There is an alternative to SW storage that also offers on-demand readout [184]. This uses the Stark effect to suppress the emission of the AFC echo using electric fields in the form of storage and retrieval pulses. This has the advantage of maintaining the low-noise echo emission of the AFC. The storage time is limited to the optical coherence time however, which even for the longest case (4 ms for Er\(^{3+}:\text{Y}_2\text{SiO}_5\) [58]) is much shorter than the longest spin coherence time in an REIC (6 hours for Eu\(^{3+}:\text{Y}_2\text{SiO}_5\) [59]).

The AFC has properties which offer advantages over other protocols. It is an intrinsically temporally multimode protocol, though this is not unique to the AFC. If a series of inputs (photons) are sent to an AFC, as long as the total time of the input modes is shorter than the AFC storage time \(\tau\), all of the photons will be stored and later emitted in the same order in time. The limit of the number of temporal modes, \(N_t\), depends essentially on the storage time of the AFC, its bandwidth, and the mode size, which for optical AFC storage is [60]

\[
N_t = \frac{1}{\Delta T_m},
\]

where \(T_m\) is the duration of one mode. There is a minimum mode duration imposed by the AFC bandwidth \(\Gamma\), related to the Nyquist-Shannon sampling theorem. The minimum mode size should therefore be [60]

\[
T_m = \frac{2.5}{\Gamma},
\]

The expression for \(N_t\) can then be expressed in terms of the number of AFC teeth \(N_{\text{tooth}}\), so \(N_t = N_{\text{tooth}}/2.5\), and thus each mode requires around 2.5 peaks in the AFC [60]. The temporal mode capacity with spin-wave storage will be discussed in Chap. 5.

In other protocols like electromagnetically induced transparency (EIT), controlled reversible inhomogeneous broadening (CRIB) and Raman, the temporal mode capacity depends on the OD instead [185], a parameter which is not trivial to increase. This scaling arises due to a dependence on absorption linewidth, and if the linewidth is increased (as in CRIB), the OD must also be increased to compensate. For the AFC, the OD does not need to be increased, only the
number of peaks in the comb as noted in the previous paragraph. For this reason, the best performance in terms of the number of modes stored has come from implementations of the AFC. Additionally, for applications where a fixed storage time is also acceptable, optical storage alone is essentially noiseless, allowing for high fidelity storage.

Aside from temporal multimodality, it can also be used for multiplexing using other degrees of freedom, such as frequency multiplexing by preparing multiple AFCs in the same optical absorption line [83], spatial multiplexing using orbital angular momentum modes [85], or by addressing different physical regions in crystal\textsuperscript{9}.

One could also imagine using the two-pulse photon echo described earlier as a storage protocol, where the first pulse is the input to be stored. However, the population inversion established by the π-pulse generates too much noise (through free induction decay and spontaneous emission) to be viable for quantum storage [186]. The application of additional pulses, or other modifications, can be used to reduce or eliminate this noise [187–189], but single photon storage has not yet been demonstrated. Ref. [189] in particular achieved the best signal-to-noise ratio, but it did not exceed the performance of SW storage with the AFC (in terms of signal-to-noise, storage time, or multimodality), such as that demonstrated in Chap. 5 of this thesis. An alternative protocol using optical rephasing is based on rephased amplified spontaneous emission [53] has also been modified similarly to reduce free induction decay noise [190], and has been used to demonstrate storage of nonclassical states [191].

### 3.4 Single photon statistics

The experiments here concern photons generated through spontaneous parametric down conversion (SPDC). The following section will cover the different metrics used to analyse the statistics of these photons, which in general should remain the same after storage in a quantum memory, i.e., their single-photon nature should be preserved, which should be the case with sufficient fidelity\textsuperscript{10}.

\textsuperscript{9}See Sec. 1.4 for a highlight of the state of the art of multimodal performance of REIC-based memories.

\textsuperscript{10}The required fidelity of the memory will depend on the number repeater links and therefore memories in repeater chain as well as the type of memory and protocol used, however the total fidelity of the repeater should be at least 90\% [11].
These measures are used throughout the experimental work in this thesis. Some parts of this section and Sec. 3.5 are based on Ref. [167].

### 3.4.1 Photon-pair source

Photons are generated through SPDC by sending a pump laser through a nonlinear crystal. The resulting output is a two mode squeezed state between two fields which we refer to as the signal $s$ and idler $i$. Energy and momentum must be conserved between the pump field and the signal and idler fields, and the condition for which this is satisfied is referred to as phase-matching. At low pump powers, the state can be written as [180]

$$|\Psi\rangle_{s,i} \propto |0\rangle_s |0\rangle_i + p^{\frac{1}{2}} |1\rangle_s |1\rangle_i + p |2\rangle_s |2\rangle_i + O(p^3).$$  \hspace{1cm} (3.31)$$

where $p$ is the probability of generating a photon pair which is low ($p \ll 1$) and proportional to the power of the pump beam, and $|n\rangle_x$ is the $n$ photon Fock state in the signal ($x = s$) or idler ($x = i$) modes.

Individually, these fields are thermal states, but are highly correlated with each other, as noted below.

### 3.4.2 Cross-correlation

To quantify the correlations between photon states, we use the normalised second order cross-correlation function, which is defined as [192]

$$g^{(2)}_{s,i}(\tau) = \frac{\langle E_s^\dagger(t) E_i^\dagger(t+\tau) E_i(t+\tau) E_s(t) \rangle}{\langle E_i^\dagger(t+\tau) E_i(t+\tau) \rangle \langle E_s^\dagger(t) E_s(t) \rangle},$$  \hspace{1cm} (3.32)$$

where $E_x^\dagger(t)$ ($E_x(t)$) for $x \in \{s,i\}$ is the creation (annihilation) operator for one of the mode fields, and $\tau$ is the delay from a time $t$.

In practice, we can measure this by looking at a coincidence histogram between the two fields, and observe the number of counts in a finite window $\Delta \tau$. Then we can calculate the cross-correlation using

$$g^{(2)}_{s,i}(\Delta \tau) = \frac{P_{s,i}}{P_s P_i},$$  \hspace{1cm} (3.33)$$

where $P_{s,i}$ is the probability of measuring a coincidence between the signal and idler fields, and $P_x$ is the probability of measuring an uncorrelated count from one
of the fields, both measured in a finite window of time \( \Delta \tau \). In an experiment, the two photons are sent to separate detectors, and we use the counts from a window in time with a peak in correlations for the numerator, while the denominator is obtained from accidental counts in a temporal region outside of the correlation peak. The minimum cross-correlation required to violate a Bell inequality\(^{11}\), introduced in the following section, is \( \approx 6 \).

### 3.4.3 Autocorrelation

The nature of a light source can be quantified using the second order autocorrelation function. This equation has a similar form to Eq. 3.32:\(^{167}\)

\[
g_{x,x}^{(2)}(\tau) = \frac{\langle E_x^\dagger(t)E_x^\dagger(t+\tau)E_x(t+\tau)E_x(t)\rangle}{\langle E_x^\dagger(t+\tau)E_x(t+\tau)\rangle\langle E_x^\dagger(t)E_x(t)\rangle},
\]

where, like for the cross-correlation, \( x \in \{s,i\} \). This value can be referred to as the unconditional autocorrelation.

This is measured using a Hanbury Brown-Twiss setup\(^{194}\), where the light field is split into two paths, each with its own detector, so the correlation of the field with itself can be observed. By checking the coincidences between the two at a time \( \tau = 0 \), the value of \( g^{(2)}(0) \) will differ depending on the type of light used\(^{167}\) (here the \( x,x \) subscript has been omitted for clarity). For a quantum state of light (antibunched), \( g^{(2)}(0) \leq 1 \). For a single photon in particular, ideally \( g^{(2)}(0) = 0 \) while a two-photon state gives \( g^{(2)}(0) = 0.5 \) and thus gives a bound for the maximum value of \( g^{(2)}(0) \) for single photons\(^{13}\). The individual modes produced by SPDC are a thermal state, so \( 1 < g^{(2)}(0) \leq 2 \), depending on the number of modes produced\(^{195}\).

To verify the single-photon nature of SPDC photon pairs, the heralded autocorrelation, \( g_{i,s,s}^{(2)} \), can be measured instead\(^{196}\). This still uses a Hanbury Brown-Twiss setup for one mode (e.g., the signal), but now a third detector is used to record heralding (idler) counts. A histogram is built by considering the number of idler counts measured between signal counts from the two signal detectors, for

\(^{11}\)Calculated by assuming the visibility \( V \) (see Sec. 3.5) is only limited by photon statistics, so \( V = (g_{s,i}^{(2)} - 1)/(g_{s,i}^{(2)} + 1) \)\(^{193}\). The memory must also maintain the coherence of the two stored modes required to demonstrate a Bell inequality violation.

\(^{12}\)For bunched light, \( g^{(2)}(0) \geq 1 \) and for coherent light, \( g^{(2)}(0) = 1 \).

\(^{13}\)This can be seen by considering the outcome of such a measurement for one-photon state versus a two-photon state, such as that given by Eqn. 8.64 in Ref. [167].
3.5 Entanglement

a particular binning size. In this way, similar to the unconditional autocorrelation, the \( g_{s,s}^{(2)} \) at the 0th bin exhibits the relevant single photon statistics following the same bounds as those noted in the previous paragraph.\(^{14}\)

\[ g_{s,i}^{(2)} : \]

3.4.4 Cauchy-Schwarz inequality

To verify the nature of the correlations between the signal and idler fields, we can use measurements of the cross-correlation and unconditional correlation with the Cauchy-Schwarz inequality, defined as \([197]\)

\[
g_{s,i}^{(2)} \leq \sqrt{g_{s,s}^{(2)} g_{i,i}^{(2)}}. \tag{3.35}\]

If the fields violate this inequality, the correlations between the signal and idler can be considered non-classical. If both the signal and idler fields have the highest autocorrelation possible, 2, then the classical bound that \( g_{s,i}^{(2)} \) must exceed is 2.

3.5 Entanglement

As discussed in Chap. 1, entanglement is the key to implementing a quantum repeater. Starting from the famous Einstein-Podolsky-Rosen (EPR) paper \([198]\), the concept of entanglement has fascinated both physicists and the general public. For a pair of entangled particles, the state of one particle can never be described without the other.

Entangled pairs are often considered in terms of the maximally entangled states, otherwise known as Bell states. These four states are

\[
|\Phi^{\pm}\rangle = \frac{1}{\sqrt{2}} (|0_A0_B\rangle \pm |1_A1_B\rangle), \tag{3.36}\]

\[
|\Psi^{\pm}\rangle = \frac{1}{\sqrt{2}} (|0_A1_B\rangle \pm |1_A0_B\rangle), \tag{3.37}\]

where the subscript \( A \) and \( B \) indicate which particle the state belongs to. The states denoted by 0 and 1 can represent any basis, such as orthogonal polarisations, number states, or time-bins. For the experiments in this thesis, we use the temporal basis, generally denoted as \(|e\rangle\) (‘‘early’’) and \(|l\rangle\) (‘‘late’’) time-bins.

\(^{14}\)See Fig. 7.6 for an example of this type of measurement.
3. Theoretical background

3.5.1 Metrics and verification of entanglement

The EPR paper ignited a debate over the workings of entanglement, particularly that measurements made on one particle of an entangled pair would be correlated with the other, even when physically separated. To explain this, the existence of local hidden variables were proposed, which would predetermine the outcome of a measurement. Bell later showed that the local hidden variable theory would not be able to account for the outcomes of such measurements in general [199]. The predictions from quantum mechanics instead suggest the violation of the inequality proposed by Bell. There have been other such inequalities constructed, and the Clauser-Horne-Shimony-Holt (CHSH) inequality is particularly prevalent due to its utility for experimental demonstrations [200]. If we consider an experiment where we have a source of entangled particles such as photon-pairs with some type of measurement device in the path of each photon, then this inequality can be written in the form [201]

\[ S = \left| E(a, b) + E(a, b') + E(a', b') - E(a', b) \right| \leq 2, \quad (3.38) \]

where \( a \) and \( b \) are related to the measurement settings for the two different paths of the entangled photons, and \( E(a, b) \) is correlation coefficient between them. Measuring an \( S \) parameter greater than 2 indicates that the two photons are entangled. The correlation coefficient comes from measuring the different possible detection events, while choosing values \( a \) and \( b \) which would maximise the value of \( S \). Here we use the sign convention for the correlation coefficients given in Ref. [202] which, like us, uses time-bins as the basis for the experiment.

Rather than measuring \( S \), we can also infer a violation of the CHSH inequality using interference fringes measured by observing the correlations between the two detectors, with one in each of the two paths. Following Ref. [202], the correlation coefficient can be written as

\[ E(a, b) = V \cos(a + b), \quad (3.39) \]

where \( V \) is the visibility of the interference fringe. Using this expression for the correlation coefficient and the settings of \( a \) and \( b \) required to maximise \( S \), the CHSH inequality can be rewritten as [202]

\[ S = 2\sqrt{2}V \leq 2, \quad (3.40) \]
so measuring $V \geq 1/\sqrt{2} \approx 70.7\%$ is required to violate the CHSH inequality. In practice, measuring $V$ involves fixing one setting, i.e., $a$, and sweeping the setting of $b$, then repeating the measurement for a different setting of $a$, i.e., $a'$, to show that the interference depends on both interferometer phases. For the scheme presented in the following section, we need $a' = a + \pi/2$ to be equivalent to the settings needed for a maximum value of $S$.

Even if this bound for the visibility is not violated, there are other implications for exceeding lower visibility metrics. In particular, 33% is the bound for separable states [203], which implies that the particles are entangled (i.e., not separable) as long as their state can be described using a Werner state, which is the maximally entangled state plus white noise [204]. Aside from this, there are many more metrics and witnesses for verifying entanglement depending on the type of system [204], but these are not needed or used for the experiments in this thesis.

The visibility is also related to the fidelity of a quantum state. Taking the result of one of the visibility measured as described above, the resulting two-qubit fidelity with respect to a maximally entangled two-photon state can be calculated using $(3V + 1)/4$, where $V$ is the visibility of an interference fringe [205]. We assume the noise is white noise.

Qubit tomography can also be used to analyse entangled states like those produced through SPDC. The purpose of tomography is to reconstruct the density matrix of the qubit state, and we can do so by performing measurements on the state in different bases [206]. Here the classical bound for the fidelity is 1/2, corresponding to the aforementioned 33% visibility bound [205]. We use both interference fringes and qubit tomography to characterise our entangled states in the experiments of this thesis, and the experimental implementation of how we measure this will be introduced in the following chapter.

### 3.5.2 Energy-time entanglement

The photon pairs generated through SPDC possess intrinsic energy-time entanglement. The photon pairs are generated at the same time within the coherence time $\tau_{\text{pump}}$ of the pump laser, as long as it is larger than the coherence time of the photons $\tau_{\text{biphoton}}$, i.e., $\tau_{\text{pump}} \gg \tau_{\text{biphoton}}$.

All of the experiments in this thesis deal with measurements of energy-time entanglement. To analyse this type of entanglement, we use the scheme
3. Theoretical background

![Figure 3.6](image)

**Figure 3.6:** Simplified setup of the Franson scheme which we use to measure energy-time entanglement. Det.: detector.

![Figure 3.7](image)

**Figure 3.7:** The three peaks which are visible when observing coincidences between the signal and idlers photons in the Franson scheme from Fig. 3.6. The central peak corresponds to the energy-time entangled state that we wish to analyse, which depends on the phases of the two interferometers.

proposed by Franson [207], where unbalanced Mach-Zehnder interferometers are placed in the paths of each photon from the source. A simplified version of this setup is shown in Fig. 3.6. Each interferometer requires phase control, which is labelled here as $\Delta \phi_i$ and $\Delta \phi_s$ for the idler and signal interferometers, respectively. The temporal path length difference of the arms, $\tau_{MZ}$, must fulfill $\tau_{pump} > \tau_{MZ} > \tau_{biphoton}$ in order to perform this analysis. For a quantum memory to be included in this scheme, it must be temporally multimode, which is indeed the case for the AFC protocol.

Although the energy-time entangled pairs are created over an infinite spread of times within $\tau_{biphoton}$, by detecting the photons in a particular time window we can postselect for an entangled state between two particular time bins. When the photons are sent through the interferometers, if we look at the coincidences between the signal and idlers photons, the resulting temporal shape will consist of three peaks, as shown in Fig. 3.7. Selecting the temporal window of the central peak postselects the state $(|e_i e_s\rangle + \exp(i\Delta \phi) |l_i l_s\rangle) / \sqrt{2}$, where $|e\rangle$ and $|l\rangle$ are the times bins associated with short and long arms of the interferometers, respectively, and $\Delta \phi$ depends on the relative phase between the two interferometers, such that
$\Delta \phi = \Delta \phi_s + \Delta \phi_t$. These phases would correspond to $a$ and $b$ in the previous section, so we can verify that the photons are entangled by measuring the $S$ parameter or interference fringes, as the number of coincidences in the central peak are proportional to $1 + V \cos(\Delta \phi_s + \Delta \phi_t)$. In the experimental work in this thesis, we do the latter in Chap. 5 and Chap. 6, as well as perform qubit tomography in Chap. 7 with the same type of experimental setup.
Chapter 4

Experimental setup and techniques

In the following chapter, I will introduce the key components and common elements for the experiments described in the subsequent chapters. I will discuss the key aspects of implementing an AFC-based quantum memory, which was introduced in the previous chapter, starting from the laser system used to address the Pr$^{3+}$ ions, then the optical setup dedicated to the memory, and finally the optical pumping sequence for preparing an AFC. Next I will cover the basic operation of the photon-pair source, and conclude with the characterisation and operating procedure of the two types of interferometers (fibre- and AFC-based) used to analyse energy-time entanglement.

Some of the text and figures here come from the Supplemental Material of Ref. [129], in particular some of the details and operation of the quantum memory and photon-pair source, as well as the operation and characterisation of the interferometers. Regarding the photon-pair source and idler interferometer, additional information comes from Ref. [208].

4.1 Laser system and locking

To address Pr$^{3+}$ ions at site 1, we use a frequency-doubled external cavity diode laser from 1212 nm to 606 nm (Toptica TA-SHG). The free-running linewidth of the 606 nm light is approximately 200 kHz. While this is sufficient
for AFCs with short storage times of a few microseconds, to achieve longer storage times, the laser linewidth must be reduced.

To reduce the linewidth, the laser is stabilised using the Pound-Drever-Hall (PDH) technique [209, 210]. To do so, a small portion of the laser light is modulated to produce sidebands, and is sent to a Fabry-Perot reference cavity. Using a photodetector and electronics including filters, an error signal can be generated which indicates if the laser is not resonant with the cavity as well as the sign in which the frequency has deviated from resonance, and this signal can be fed back to the laser to adjust its current and thus frequency.

We use a homebuilt Fabry-Perot reference cavity and the seed laser wavelength of 1212 nm. The cavity consists of two mirrors glued to a 22.5 cm long Invar rod held under vacuum without temperature stabilisation. It has a finesse of approximately 2000 and a free spectral range of approximately 660 MHz. After frequency doubling of the light, the available modes at 606 nm are separated by 1.3 GHz. We estimate the laser linewidth to be no greater than 10 kHz based on the linewidth of the spectral holes we can burn in Pr$^{3+}$:Y$_2$SiO$_5$.

4.2 Quantum memory setup

The setup for the Pr$^{3+}$:Y$_2$SiO$_5$-based quantum memory varies depending on the specific experiment, in particular whether a bulk Pr$^{3+}$:Y$_2$SiO$_5$ crystal or the fibre-integrated memory is used. The common elements for all of these experiments will be introduced in this section.

A cryostat is used to cool the Pr$^{3+}$:Y$_2$SiO$_5$ crystals to around 3 K. To minimise the effects of vibrations from the cryostat on the optical beam path, the optical setup is divided into two parts on different optical tables. The laser system and optics used for pulse shaping with acousto-optic modulators (AOMs) are on one table, while the cryostat which holds the Pr$^{3+}$:Y$_2$SiO$_5$ crystals and the optical elements required to address them are on a separate table, and are consequently affected by vibrations.

To prepare an AFC in the absorption line of Pr$^{3+}$:Y$_2$SiO$_5$, we use laser pulses with particular amplitude, frequency, and phase profiles. In our case, we utilise three AOMs, each in a double-pass configuration [211], as shown in Fig. 4.1. The outputs of the AOMs are fibre-coupled to deliver the light to the memory setup on the other optical table. The output of the AOMs follow one or more of the
4.2 Quantum memory setup

The Pr$_3^+$:Y$_2$SiO$_5$ crystals (grown by Scientific Materials) are used with a 0.05 % Pr$_3^+$ doping concentration. The dimensions of the bulk crystal are $2 \times 3 \times 5$ mm, and for the fibre-integrated crystal they are $5 \times 4 \times 3.7$ mm, corresponding to the $D_1, D_2$, and $b$ axes respectively. In the experiments using a bulk memory crystal, we also use an additional, identical crystal as an analyser, which is detailed in Sec. 4.5.2.

The cryostat we use for the experiments is a closed-cycle pulse-tube cryocooler (Optistat AC-V14, Oxford Instruments). It can reach 3 K at the sample.

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1 Another double-pass AOM could be used instead of the Pockels cell, but in this way we can conserve laser power and use one channel of the arbitrary waveform generator instead of two.
mount, typically even a few hundred mK lower. A layer of mu-metal lines the inside of the radiation shield to reduce the effect of static magnetic fields on the crystals. In this case, the shielding reduces the inhomogeneous broadening of the spin transitions. For experiments with bulk crystals, we can simply access the ions by sending lasers through windows already installed in the cryostat, but for the fibre-integrated experiments, we use a fibre feedthrough (designed in-house) to send light directly to the crystal. As a result, there are very few further optics required for the fibre-integrated memory.

The major limitation of the cryostat we use is that it imparts strong vibrations to any samples attached it, due to the periodic compression of helium gas required for cooling. The vibrations impart a strain on the crystal, which causes small frequency shifts and broadening of the transitions and thus reduces the coherence of the Pr$^{3+}$ ions [212]. This reduces the efficiency of the AFC, either by reducing the quality of the optical pumping when preparing an AFC, or destroying the structure of the AFC if it has already been prepared. In this cryostat it appears that the strain is more detrimental to the hole linewidth if it is present when creating a structure compared to when the structure has already been created.

To mitigate the effects of the vibrations, we used sample mounts designed to isolate the crystals. Two different mounts were used for the bulk and fibre-integrated experiments. Both follow a similar type of design – the mounts are made up of two pieces, where one piece is attached to the cold finger of the cryostat, and the other piece contains the crystals and is attached to the first piece with springs for vibration damping and a few thin copper foils to create a thermal connection. The specifics of the bulk crystal mount will be shown in Chap. 5, and in Chap. 7 for the fibre-integrated memory.

Figure 4.2 shows the general simplified optical setup for the bulk experiments. Two crystals are used: one as the quantum memory, and another as a spectral filter and/or time-bin qubit analyser (referred to as the analysis crystal). One continuous path is used for the input light to the crystal. Depending on the experiment, up to

Figure 4.2: Simplified, generic setup for the experiments utilising the bulk Pr$^{3+}$:Y$_2$SiO$_5$ crystals.
three other optical paths are required for the crystals. Each crystal has a separate preparation path (“Preparation” in the setup diagram), at an angle to the input, used for optical pumping in order to create some kind of spectral structure in the crystal – generally an AFC or a spectral pit. If spin-wave storage is required for the experiment, then another path (“Control” in the setup diagram) is used for the memory crystal, also at a angle to the input beam, and counterpropagating to it to reduce noise.

The input path (which can be classical pulses, weak coherent states or single photons, depending on the required measurement) arrives at the optical table via a single mode optical fibre. Here the polarisation optics have been omitted for clarity, but we polarise the beam along the \( D_2 \) axis of the crystal, ensuring maximum absorption of the light. All beam paths incident on the crystal are sent with this polarisation. The input first passes through the memory crystal, then via single mode optical fiber again to a separate input path through the analysis crystal. After the analysis crystal, the input passes through a band pass filter (BPF) (central wavelength 600 nm, linewidth 10 nm), then to a single photon detector (SPD) or a photodiode for measurements with bright pulses as the input. The specific detector used depends on the experiment, however all were Count-series silicon avalanche photodiode modules from Laser Components, with detection efficiencies around 60% and dark count rates from 10 to 50 counts/second. Counts from the detectors were recorded using a time-to-digital converter (SD TDC-H3344, Signadyne). All of the optical fibres used for the input path have black jackets in order to minimise noise counts from any stray light in the laboratory reaching the fibres.

The beam waists inside the crystals differ depending on the path. In general, the input mode should be smaller than the preparation modes so that the input mode experiences a uniform spectral structure. While the control mode should also be larger than the input, it is more optimal to use a smaller (and thus separate) mode to the preparation in order to have a larger Rabi frequency. The input mode has a waist\(^2\) of 40 \( \mu \text{m} \) in both crystals. In the memory, the preparation and control modes have waists of approximately 250 \( \mu \text{m}^3 \) and 100 \( \mu \text{m} \), respectively, and the preparation mode has a waist of 200 \( \mu \text{m} \) in the analysis crystal.

Shutters are placed in all of the beam paths in order to prevent any classical preparation light from entering the SPD. For the two preparation paths and the

\(^2\)The beam waists given here are all defined as the \( 1/e^2 \) radius.
\(^3\)This value was incorrectly given as 180 \( \mu \text{m} \) in Ref. [129].
4. Experimental setup and techniques

control path, these are placed in the beam paths before they reach the cryostat. Since the Pockels cell in the memory preparation/control path has a finite polarisation extinction ratio, the shutters block stray light in one path while the other path is in operation. The input path has two shutters: one placed after the memory, and one placed after the analysis crystal. The preparation path shutters are open during AFC preparation while the control and input path shutters are closed, and vice versa during the measurement period when single photon detection occurs.

4.3 AFC preparation

To obtain a single-class AFC, we use a preparation method similar to that described in Ref. [44]. First, we prepare a \( \sim 20 \) MHz wide spectral pit, then bring back population to this region by applying a burn-back pulse on the \( 5/2_g \rightarrow 5/2_e \) transition (the \( \pm \) prefixes have been dropped for clarity). We apply a cleaning pulse close to the \( 3/2_g \rightarrow 3/2_e \) transition to obtain a single class absorption feature corresponding to the \( 1/2_g \rightarrow 3/2_e \) transition.

To prepare the AFC structure itself, we proceed in one of two possible ways. We can either simply burn holes one at a time to create each tooth of the comb separately, often known as the serial method, or we can use a parallel preparation method similar to Ref. [82]. With the serial method, we have better control over the finesse of the comb, but the preparation time scales poorly with storage time as more comb teeth are required for the same bandwidth (roughly 30 ms per microsecond of storage time), so we only use this method for short storage times up to a few microseconds. In comparison, it only takes an additional \( \sim 80 \) ms to prepare a 25 \( \mu \)s AFC instead of a 10 \( \mu \)s one using our implementation of the parallel method.

For the parallel method, unlike Ref. [82], we simulate our desired comb structure, take its Fourier transform and apply this temporal waveform to the single-class spectral feature at four different frequencies, 1 MHz apart, in order to prepare a uniform structure over the 4 MHz comb bandwidth. To ensure that the \( 3/2_g \) level is kept empty, we alternate between applying the temporal waveform to burn the AFC and applying cleaning pulses on the \( 3/2_g \rightarrow 3/2_e \) transition. Finally, we apply stronger cleaning pulses on the \( 3/2_g \rightarrow 3/2_e \) transition which have the same power, waveshape, frequency span and central frequency as the CPs for further cleaning. For the CPs and the final cleaning pulses, we use pulses
with a Gaussian temporal profile, which have a full width half maximum of 2.5 $\mu$s, and a hyperbolic tangent frequency chirp which spans 4.2 MHz. These pulse parameters were chosen in order to maximise the signal-to-noise ratio of the retrieved spin-wave echo. A graphical depiction of the spectrum, the temporal waveform for burning the AFC, and the detailed pulse sequence that we use can be found in App. A.

The preparation time of the AFC depends on the storage time, where longer storage times require more preparation time. As an example, the 10 $\mu$s AFC commonly used in this thesis takes a total of 366 ms to prepare. We use the rest of the cryostat vibration period (707 ms) to measure storage with single photons, and repeat the preparation and measurement procedure each vibration period.

### 4.4 Photon-pair source

The source of entangled photon pairs is based on cavity-enhanced spontaneous parametric down-conversion (cSPDC), as previously described in [83, 91, 125], and in particular in detail in the PhD thesis of Dr. Dario Lago-Rivera [208], who developed and operated the source. The pump laser at 426 nm, generated through frequency doubling from an 852 nm seed laser (TOPTICA TA-SHG 110), is sent to a cavity containing a periodically-poled lithium niobate crystal (PPLN),
generating a photon with a wavelength of 1436 nm (the idler), which is in the telecom E-band, and another at 606 nm (the signal). A homebuilt Fabry-Perot reference cavity (Locking Cavity) is used to lock the seed laser to reduce the laser linewidth such that the coherence time of the laser is much longer than the coherence time of the resulting biphoton ($\tau_{\text{pump}} \gg \tau_{\text{biphoton}}$).

To ensure that the signal photon is resonant with the Pr transition used for the memory, the length of the cavity is locked to a 606 nm reference beam (resonant with the AFC) with a piezoelectric actuated mirror (PZ). The idler photon is kept resonant to the cavity by using feedback from a classical beam at 1436 nm generated in the same PPLN crystal through difference frequency generation (DFG) between the pump beam and the reference 606 nm beam. The feedback acts on the pump laser wavelength by adjusting the position of one of the reference cavity mirrors with a piezo-actuator to change the cavity length, and since the seed is locked to the cavity, the wavelength will follow the change. The resulting pump laser linewidth is 380(20) kHz, corresponding to $\tau_{\text{pump}} \approx 1\mu$s, determined using the idler interferometer in Sec. 4.5.1, and the biphoton linewidth is 1.8 MHz, with $\tau_{\text{biphoton}} = 120$ ns.

After the output of the source cavity, the signal and idler paths are separated with a dichroic mirror. As the spectrum of the photons is composed of 15 frequency modes [83], the idler photon is first sent to a filter cavity (FCav, free spectral range of 16.8 GHz, finesse of 210) to ensure single-mode operation (the inhomogeneous broadening of the memory crystal filters the signal modes). This cavity is locked to the maximum of its transmission by tuning the mirror attached to a PZ, which is monitored using the back reflection of the next optical component. Filtering of broadband noise is provided by a bandpass filter (BPF, central wavelength at 1440 nm, linewidth of 14 nm). The signal photon passes through a fused silica etalon filter (free spectral range of 100 GHz, finesse of 23.5) then a bandpass filter (central wavelength at 600 nm, bandwidth of 10 nm) for broadband filtering before being sent to the memory.

There is classical light used for locking in the same optical paths as both the signal and idler photons, specifically the 606 nm reference for the cavity length, and the DFG reference to lock the 426 nm laser and FCav. This necessitates some way of preventing the classical light from reaching the single photon detectors. A mechanical chopper at the output of the SPDC cavity in both photon paths is used.
for this purpose by alternating between locking the setup and measuring single photons. The cycle has a period of 33 ms, with a measurement duty cycle of 55%.

For the experiments presented here, the idler photons were detected using a superconducting nanowire single photon detector (ID281, ID Quantique), which has a detection efficiency of 80% and 10 counts/second of dark counts. In Chap. 5, under typical experimental conditions, spurious light increased the background rate to 120 counts/second, but this was reduced to close to the dark count rate in Chap. 6.

### 4.5 Time-bin interferometers

As mentioned in the previous chapter, the experiments presented here all deal with energy-time entanglement, which we characterise using the Franson scheme \[207\]. This requires an unbalanced Mach-Zehnder interferometer for both (idler and signal) of the photon paths.

We implemented these interferometers in two ways: using a fibre-based interferometer, or an AFC-based interferometer. The following sections will introduce both of these types of interferometers, and how we implemented them in the experiments.

#### 4.5.1 Fibre-based interferometers

For the idler photon in all of the experiments described in the following chapters, and for the signal photon in the experiment of Chap. 7, we used a fibre-based interferometer. Figure 4.4 shows the setup for the interferometer for the idler photons. It consists of a short arm with a polarisation controller (PC) and a long arm with a home-made fibre stretcher consisting of a cylindrical piezo-actuator with optical fibre wrapped around it, and an additional 90 m of optical fibre. The corresponding path length difference in time, \( \tau_{MZ} \), is 424 ns. This particular time was chosen as it needed to be large enough so that the photons retrieved from the memory would be well separated in time (and consequently longer than \( \tau_{biphoton} \)), and also so that the total span of the two time bins would be within the 1 \( \mu \)s coherence time (\( \tau_{pump} \)) of the source pump laser. In other words, \( \tau_{pump} > \tau_{MZ} > \tau_{biphoton} \).
The phase of the interferometer is controlled by the fibre stretcher. Applying a voltage to the piezo-actuator stretches the fiber, thus changing its length and the phase difference between the short and long paths.

To lock the interferometer to a specific phase, we use the classical DFG signal, which is already used to lock other parts of the source, as the input, and scan the phase of the interferometer to observe an interference fringe. We lock to a particular point using a side-of-fringe lock. We use a set point in the lower half of the fringe as it is less sensitive to power fluctuations. To shift to another interferometer phase, a constant voltage is added to the applied signal during the measurement phase. The interferometer is locked at the same time as the cSPDC source, but now the chopper wheel in the idler path must be placed after the interferometer since classical light passes through it.

To determine the visibility of the idler interferometer, we used a 1535 nm laser with a less than 10 kHz linewidth (Toptica CTL) as the input and scanned the interferometer phase. From the minimum and maximum values of the output, we estimated the visibility to be larger than 99%, limited by the reduced transmission of the long arm of the interferometer, which is 84% of the short arm. We repeated this measurement using the 1436 nm classical DFG light and obtained a visibility of 91.3(7)% by fitting a sinusoid to the transmission measured with the PD. As mentioned previously, the DFG light is generated using the 606 nm and 426 nm lasers, and thus its linewidth is dependent on both. Since the 606 nm light has a narrow linewidth of less than 10 kHz, a measurement with the DFG light will be limited by the 426 nm laser, which explains the observed reduction in visibility. Using the measured value for the visibility and a Gaussian model for the phase noise, we estimate the laser linewidth of the 426 nm pump to be 380(20) kHz [213].

The setup for fibre-based interferometer for the signal photons is shown in Fig. 4.5. It follows the same design as in Fig. 4.4. Here, the signal photons and classical reference light which is used for locking are sent to the interferometer.
4.5 Time-bin interferometers

![Diagram of fibre-based interferometer](image)

**Figure 4.5:** Schematic of the fibre-based interferometer for the signal photons.

through different inputs. This interferometer was exclusively used for the signal photons that were delivered directly via fibre from the integrated memory.

The locking method is the same as the idler interferometer, however the configuration of choppers used to block the reference light is different. One chopper before the interferometer controls whether or not reference light is sent through the device. Both of the outputs of the interferometer are used to measure coincidences, so a chopper after the interferometer is used to block light as required in both paths simultaneously. A back-reflection off of a glass window in one of the free space paths is used to monitor the reference light. There are also shutters in the both of the free space paths after the interferometer (not shown here) which are used to block the paths when classical AFC preparation light is sent through the signal path.

To characterise this interferometer, we sent classical 606 nm light through the device and measured an interference fringe by locking the interferometer for a range of phases, and fitting a sinusoid to those points. The resulting visibility was 98%. However, this interferometer is more sensitive to polarisation drifts than the idler interferometer\(^4\), potentially reducing the visibility by more than 10% over the course of a several-hour long measurement. Such a large drop in visibility was not observed with the telecom-wavelength idler photon interferometer. Additionally, the 90 m fibre spool in the long arm of the interferometer has 49% transmission compared to the short arm, which is much lower than for the idler interferometer due to the shorter wavelength used. This loss reduces the end-to-end transmission to around 25%.

\(^4\)The reason for this is uncertain, but one possibility is that temperature fluctuations affect the spooled (and thus slightly stressed) portion of the interferometers more at shorter wavelengths than longer wavelengths.
Figure 4.6: The input time-bin state which has a phase difference of $\phi$. If light is stored by the AFC (represented as a delay line with a loop on the right), it will gain an additional phase factor $\phi_{\text{AFC}}$, otherwise it does not change phase.

4.5.2 AFC-based interferometers

As mentioned in the previous section, fibre-based interferometers at visible wavelengths like 606 nm have drawbacks regarding transmission and stability. We can instead use temporal delays introduced by the AFC prepared in a second crystal (the analysis crystal in Fig. 4.2) as the “arms” of the interferometer [100, 214, 215]. An input photon can be split between two time bins either with a spectral structure containing two superimposed AFCs that each produce their own echo, referred to as a “double AFC interferometer”, or from a single AFC echo and the transmitted light through the AFC, referred to as a “single AFC interferometer”. This type of interferometer has the advantage of not requiring any locking scheme. For the experiment in Chap. 5, a second crystal aside from the memory is required, so using a fibre interferometer instead of an AFC interferometer would introduce more loss to the setup.

The phases of these interferometers can be controlled by shifting the frequency of the single AFC, or one of the two AFCs comprising the double AFC, relative to the input light [50]. To see how this works, let us consider the single AFC case which was introduced by Jobez in Ref. [215], and the scenario depicted in Fig. 4.6. If we consider the time-bin state $(|e\rangle + e^{i\phi} |l\rangle)/\sqrt{2}$ and send it to the AFC, the photons will either be transmitted or stored. The photons which are stored and reemitted by the AFC gain additional phase $\phi_{\text{AFC}} = 2\pi\Delta_0/\Delta$, where $\Delta_0$ is the detuning between the carrier frequency of the photon and the centre of the AFC [50]. The photons in the central peak of interference (see Fig. 3.7) will either have been from the late-bin and transmitted, thus retaining the phase $\phi$, or were from the early-bin and stored by the AFC, thus gaining the phase $\phi_{\text{AFC}}$. Assuming the transmitted and stored components of the AFC are equal as well as the early and late components of the input time-bin state, then the intensity of the
interference peak will be

\[ I_{\text{peak}} \propto |1 + e^{i(\phi - \phi_{AFC})}|^2 \propto 1 + \cos(\phi - \phi_{AFC}) \propto 1 + \cos(\phi - 2\pi \Delta_0/\Delta). \] (4.1)

Thus there is a sinusoidal dependence of the interference on the detuning of the AFC. If \( \Delta_0 \) is shifted by the comb tooth spacing \( \Delta \), it is equivalent to a \( 2\pi \) shift in phase. The interference between the two echoes of the double AFC interferometer works in the same way, where the phase depends on the frequency detuning of both combs relative to the input photon. We shift only one of the two combs to construct an interference fringe.

To prepare the AFC for either of the interferometers, we use a more simplified sequence compared to the memory as no class cleaning is required. In both cases, we first prepare a 6 MHz wide spectral pit centered at the QM AFC frequency. We then burn back population using the \( 5/2_g \) to \( 5/2_e \) transition. Then we prepare a comb using the serial holeburning method.

For the single AFC, we simply burn one comb. The storage time was chosen such that the echo is separated from the transmitted light by as close to 424 ns (the path-length different in time of the idler interferometer) as possible. In this case, the separation between the teeth, \( \Delta \), is 2 MHz, corresponding to a 500 ns storage time and due to slow light effects, the delay between transmitted light and echo is close to 424 ns. The storage efficiency is optimised such that light can be transmitted or stored with equal probability. Figure 4.7 shows the structure

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**Figure 4.7:** (a) OD vs frequency for the single AFC structure, used as an interferometer when performing spin-wave storage. Zero frequency detuning corresponds approximately to the input photon frequency. (b) Temporal spectrum of the AFC from (a) with a single classical pulse as the input. The darker region of the trace indicates the peaks corresponding to the transmitted and stored light, used as the short and long interferometer paths respectively.
of one such comb, as well as temporal spectrum of the transmitted and stored light that comprise the two time bins of the interferometer. This AFC has a width of $\sim$10 MHz, and it is not centered with the QM AFC, but instead at a lower frequency in order to avoid getting too close to the control pulse frequency (and thus noise) when performing SW storage, while taking advantage from an increase in efficiency from the comb bandwidth.

For the double AFC, we alternate repeatedly between burning a 2 $\mu$s and 2.4 $\mu$s comb. The preparation of this comb is optimised such that a photon can be stored for either time with equal probability. The final structure is then centered at the frequency of the AFC of the QM. The structure of one of the phases is shown in Fig. 4.8, as well as the resulting temporal spectrum with a single pulse as an input.

To scan the phase of these interferometers, we shift the position in frequency where the 2.4 $\mu$s comb is burnt for the double AFC, and similarly we shift the position of the comb for the single AFC interferometer. For each phase, we reoptimise the AFC sequence to equalise the probability of the photon traveling along either the short or long path, and maintain similar storage efficiencies across all the phases used.

Figure 4.8(c) shows the characterisation of the double AFC interferometer, with interference fringes measured with classical pulses for storage with and
without the memory. We used two classical light pulses, separated by 424 ns, through a transparency window in the QM. We measured the interference as a function of the change of the double AFC phase, $\Delta \phi_s$, and the resulting visibility was 91(2)%. Using pulses that had been stored in an AFC, the visibility was 86(2)%. A similar interference measurement was performed using the single AFC interferometer and a classical AFC echo as the input, but the phase of the second input pulse was varied while the AFC phase remained fixed. The resulting visibility was 98(1)%.

The AFC interferometer used (single or double) depends on whether AFC or SW storage is performed, due to the difference in background noise in the measurement. Figure 4.9 shows a visual representation of this difference.

For the double AFC interferometer and SW storage [see Fig. 4.9(d)], the signal we consider for our interference measurements will only be stored and emitted by the 2 $\mu$s and 2.42 $\mu$s AFCs, but the noise from the CPs can either be transmitted through the double AFC, stored and emitted for the two aforementioned storage times or at one of the secondary echoes, all of which can be seen experimentally in Fig. 4.8(b). The efficiencies of the 2 $\mu$s and 2.42 $\mu$s combs are 10% each, while
the echoes, secondary echoes and transmission through the double AFC total to 60% of the input. So for SW storage 60% of the noise (mostly due to the control pulses) will pass through the analysis crystal compared to 20% of the signal, therefore reducing the maximum $g_{SW,i}^{(2)}$ and thus the visibility of the interference. Thus for SW storage we use the single AFC instead. Unlike the double AFC interferometer, there are minimal secondary echoes, and so the signal photons are transmitted with a similar efficiency as the noise photons [see Fig. 4.9(b)]. The storage efficiency of the single AFC is optimised to be similar to that of the transmitted light through the comb, which is approximately 25% for both and thus 50% of the input light (or noise), a much better ratio of transmitted signal to transmitted noise than with the double AFC. As a result, the $g_{SW,i}^{(2)}$ at the phase of maximum interference is similar to that of the $g_{SW,i}^{(2)}$ measured without interferometers.

Despite the larger storage efficiency, the single AFC interferometer is less optimal for AFC storage alone as the interference window is in a temporal position where there is still stored noise from the pump laser, which will limit the visibility due to a reduction in $g_{AFC,i}^{(2)}$ [see Fig. 4.9(a)]. In the case of SW storage, the CP noise dominates all other sources of noise and so the stored pump noise in that temporal window is negligible. There is still some advantage in using the single AFC interferometer even for excited state storage however, since the efficiency is higher, the count rate will also be higher, thus reducing the measuring time of an experiment.
Chapter 5

Spin-wave multimode quantum memory for light-matter entanglement

In this chapter, I will present our experiment demonstrating entanglement between a telecom photon and an on-demand multimode solid-state quantum memory. We utilise the energy-time entanglement that results from photon-pairs generated by SPDC, and measure it in the temporal basis. Before doing so, we characterised the performance of the memory, as various improvements to the experimental setup were required in order to perform this measurement. We also investigated the temporal multimode capacity of the memory.

The main contents and results of this chapter are from the paper of Ref. [129] and its Supplemental Material. I am a co-first author along with Dr. Dario Lago-Rivera. In terms of the experimental work, I worked on assembling, developing and operating the memory setup, along with Dr. Alessandro Seri and Dr. Samuele Grandi. Dr. Dario Lago-Rivera developed and operated the photon-pair source, as well as the idler interferometer, with assistance from Dr. Samuele Grandi. All four of us acquired the data and contributed to the data analysis.

The results presented in Sec. 5.6 are from Ref. [60]. Our experimental contribution was measured and analysed by me (using the same memory setup), and some calculations were performed using the theory developed by our collaborators at the University of Geneva that is presented in the same paper.
Additionally, a measurement of the AFC efficiency versus storage time is from the Supplemental Material of Ref. [125].

5.1 Introduction

A key component of the quantum repeater scheme as described in Chap. 1 is the establishment of entanglement between a photon and a quantum memory. In the case of many memory systems, this would result in light-matter entanglement. The photon should be at a telecommunications wavelength (used for heralding the entanglement), and the memory needs a sufficiently long storage time as determined by the network size, amongst other requirements introduced in Chap. 1.

Many other systems have been explored for this purpose as noted in Chap. 1, but more specifically to our system, light-matter entanglement has already been demonstrated in other REICs. It was demonstrated using the AFC protocol with external photon-pair sources in Refs. [87, 100, 101]. However, in all of these cases the storage time was limited to less than 1 $\mu$s, and on-demand retrieval was not used. Light-matter entanglement was demonstrated with Pr$^{3+}$:Y$_2$SiO$_5$ used as an on-demand emissive memory [191, 214]. However, neither experiment utilised quantum frequency conversion and thus lacked a telecom herald.

In this chapter, we demonstrate light-matter entanglement between a telecom heralding photon and a photon stored in the spin state in an REIC. We do this by utilising the energy-time entanglement that results from the photon-pair source, store one of the photons in the memory using the AFC and SW storage, and measure the entanglement in the temporal basis using the scheme proposed by Franson in Ref. [207].

5.2 Experimental setup

Figure 5.1 shows a simplified version of the experimental setup used to establish and measure light-matter entanglement. Photon pairs are generated by pumping a PPLN crystal inside a cavity with a laser at 426 nm. The resulting photons are sent through filtering elements, then the idler photons are sent to a fibre-based interferometer and the signal photons are sent to the memory setup. A
5.2 Experimental setup

Figure 5.1: Experimental setup used to measure energy-time entanglement. The individual components used are: periodically-poled lithium niobate (PPLN), dichroic mirror (DM), bandpass filter (BPF), filter cavity (FCav), piezoelectric actuators (PZ), polarisation controller (PC), polarising beam splitter (PBS), and single photon detector (SPD).

detailed description of the photon-pair source and its locking scheme, the quantum memory and the interferometers can be found in Chap. 4.

The idler photons pass through a fibre-based Mach-Zehnder interferometer (see Chap. 4) before arriving at the SPD. A click at this detector heralds a single photon in the signal mode. When an idler photon is detected, the pump laser is switched off for tens of microseconds to prevent the creation of additional pairs and broadband noise during the retrieval of the stored photons. The signal photon is sent to the memory crystal via an optical fibre, then the photon is sent to the analysis crystal via another optical fibre. The analysis crystal is used as an interferometer, either in a single AFC or double AFC configuration (see Sec. 4.5.2 for details). The signal photons are then sent to a SPD. We use COUNT-10C-FC for the entanglement with AFC measurements and temporal multimode measurements and COUNT-50C-FC for the correlation measurements and entanglement with SW storage measurements.

For SW storage in particular, we moved the etalon that is normally directly after the source in the signal photon path to a position that is after the memory. This allowed us to slightly increase the heralding efficiency of the source since the etalon has a transmission of 90%, and at the same time filter some of the noise generated by the CPs.
Figure 5.2: The sample holder used to hold the bulk Pr$^{3+}$:Y$_2$SiO$_5$ crystals inside the cryostat, as viewed (a) from above and (b) from the front, mounted on the cold finger of the cryostat with the crystals attached to it.

Spectral filtering of the CPs is performed using three main components: the analysis crystal, the BPF, and the etalon. Both Pr$^{3+}$:Y$_2$SiO$_5$ crystals have an inhomogeneous linewidth of 10 GHz [91], and aside from its purpose as an interferometer, the analysis crystal is used to filter the light from the CP field which is 10.2 MHz away from the signal photons, as well as fluorescence from the $3/2_e$ level to the $3/2_g$ and $5/2_g$ levels. Fluorescence to the $1/2_g$ level cannot be filtered since it coincides with the frequency of the signal photons. The BPF is used to filter broadband light at other wavelengths, including fluorescence induced from the CPs from the $^1D_2(0)$ level to other crystal field levels in the $^3H_4$ manifold. The etalon can filter light in the region between the cutoff of the inhomogenous line of the analysis crystal and the BPF, which is again likely to come from fluorescence decay to other crystal field levels in $^3H_4$ manifold, and broadband noise from the source. Additionally, a PBS (which is also part of the polarisation control of the signal path) filters unpolarised fluorescence noise from the CPs.\footnote{In a previous iteration of the setup where the signal photon path between the QM and analysis crystal was exclusively in free space, the addition of a PBS reduced the noise floor by roughly 40%}

A key element of this setup compared to the one used previously in Ref. [44] is the use of a vibration isolating sample mount inside the cryostat, as shown in Fig. 5.2. This holder was designed by Dr. Alessandro Seri. It consists of a central block which the crystals are attached to with GE Varnish, and the block sits within a frame with six springs (four below the block and one on either side) that is bolted to the cold finger of the cryostat. The block and spring dimensions were chosen such that the resonant frequency of the block would be 0.6 Hz.
Figure 5.3: Storage efficiency of classical light pulses with a 10 µs AFC, measured under three different conditions to characterise the performance of the sample holder.

Four strips of copper foil are used to thermalise the block. Two are at the front and connect to the frame. Another two are at the back of the holder, and are attached directly to the cold finger. We verified that the block and crystals were well-thermalised using a temperature sensor attached to the block, which reached the same temperature as the cold finger. More importantly, the coherence properties of the Pr$^{3+}$ ions were still good, as we measured an optical coherence time of 116(6) µs, which is within error of the value reported in literature measured at 1.4 K [143]. Although these foils affect the resonant frequency of the sample holder, we saw that the resulting frequency worked in our favour to damp the vibrations.

We tested how well the holder works by comparing the AFC efficiency obtained in three different situations: the usual experimental sequence where the preparation of the AFC is synchronised to the vibration period of the cryostat (i.e., triggered the cryostat), preparing an AFC at a random time in the vibration period (not triggered), and immediately after turning off the cryostat so the crystals remain cold but are not vibrating. This comparison is shown in Fig. 5.3. Here the input to the AFC was classical light pulses, 50 measurement trials were attempted for each scenario, and the photons were stored for 10 µs. This storage time is long enough to be quite sensitive to vibrations due to the close spacing between comb teeth and narrow linewidth of the teeth.
The efficiency was clearly higher and more stable with the cryostat off, with an average efficiency of 19.7(6)%, though it decays due to the increasing temperature of the cold finger which broadens the optical transition. The average efficiency for the triggered and untriggered cases was very similar: 19.0(6)% versus 18.7(6)%. The largest difference between the maximum efficiencies of the three cases was less than 1%. The agreement between the triggered and untriggered measurements indicates that we could decouple our experimental sequence from the cryostat vibration cycle. While we do not do this in the experiments presented in this thesis, we made use of this stability in an experiment requiring simultaneous use of two cryostats in Ref. [125].

5.3 Memory performance

This section will cover the performance of the memory without measuring entanglement, i.e., without the interferometers. The process for storing photons with the AFC and with SW storage is described along with relevant pulse timing in Sec. 3.3. For most of the measurements in this section and Sec. 5.4 which follows, we use an AFC with a storage time of $\tau = 10 \, \mu s$, and if SW storage is also used, then we store photons in the spin state for $T_s = 6.9 \, \mu s$.

The following two subsections will go into further detail regarding the performance of both AFC and SW storage in this system. Section 5.3.1 will cover the AFC performance, first analysing the expected efficiency of the 10 $\mu s$ AFC, then assessing the efficiency as a function of storage time. In Sec. 5.3.2 which covers SW storage, we will first examine the control pulses, including their efficiency and the effect of the control pulse power on different metrics of SW storage. The final subsection, Sec. 5.3.3 will cover the performance of the memory when storing single photons from the source. We will analyse the storage performance in terms of the second-order cross-correlation, efficiencies, and coincidence rates, and compare our current results with those of the previous experiment in our group.

5.3.1 AFC characterisation

Figure 5.4 shows the structure of the AFC used to store photons for 10 $\mu s$, as well the spectrum of the analysis crystal when used as a filter only. The inset shows the structure of the individual comb teeth. The key values that can be extracted
are \( d = 4.1(3) \), \( F = 3.4(1) \), and \( d_0 = 0.18(11) \). From this, we can estimate the expected storage efficiency of the AFC. Using Eq. 3.25 and approximating the comb teeth as Gaussian, the expected efficiency is 20(3)\%, which is within error of the 19.7(6)\% efficiency measured with the photon-pair source as an input.

We can calculate the maximum possible efficiency for this storage time assuming that the optical pumping is perfect, i.e., maintain the same \( d \) and comb period \( \Delta \), but reduce \( d_0 \) to zero and shape the comb teeth with square tops and a finesse that maximizes the efficiency. For our system, \( F = 3.9 \) maximizes the efficiency with a value of 33\%. Due to bandwidth mismatch, 25\% of the input photon will be lost, thus limiting the maximum storage efficiency (for 10 \( \mu \)s) measurable with our setup to about 25\%.

We also characterised the AFC performance as a function of storage time. Ideally, the AFC efficiency should only be limited by the homogeneous linewidth of the \( \text{Pr}^{3+} \) ions. However, imperfections in the optical pumping procedure, an insufficiently narrow laser linewidth, and other effects such as vibrations can prevent the preparation of an ideal AFC structure. Thus we can define the decay of \( \eta_{\text{AFC}} \) as: [82]

\[
\eta_{\text{AFC}} = e^{-4\tau/T_{2\text{eff}}^2},
\]

(5.1)

where \( T_{2\text{eff}} \) is the effective coherence time of the AFC storage. By comparing \( T_{2\text{eff}} \) to the intrinsic \( T_2 \), we can get a measure of how imperfect our AFC preparation is compared to the ideal case. This value is obtained by fitting to the measured \( \eta_{\text{AFC}} \) versus \( \tau \).
Figure 5.5: A comparison between the memory setup used in this thesis and Ref. [44] of the AFC efficiency as a function of storage time. In both cases, single photons from a photon-pair source are used as the input.

Figure 5.5 shows $\eta_{\text{AFC}}$ versus $\tau$ for our current setup as well as the previous setup from Ref. [44]. For the memory setup presented here, $T_2^{\text{eff}} = 92(9)$ $\mu$s. This value is approaching the extrapolated value given in literature for the $T_2$ of this transition of Pr$^{3+}$:Y$_2$SiO$_5$, which is 114 $\mu$s [143], as well as our measured value of 116(6) $\mu$s. For the previous setup, $T_2^{\text{eff}} = 33(4)$ $\mu$s [44].

We attribute the improvement in AFC efficiencies and storage times to a few factors, with the most significant first: the vibration isolation sample holder, an improved laser lock, having separate preparation and control paths, and less bandwidth mismatch between the photons and comb as a result of using a new photon-pair source. Previously, a single path was used for both the AFC preparation and the CPs, which prevented this optical path from being very large, otherwise the Rabi frequency would be too low to send efficient CPs. However, the preparation path should be quite wide in order for the Rabi frequency to be homogeneous in the region of the input mode, so that the input interacts with a uniform AFC structure. With separate optical paths, we reached a higher AFC efficiency while maintaining a high Rabi frequency for the control pulses.

To further improve the AFC preparation, it could be beneficial to reduce the laser linewidth further, particularly for longer storage times, by using a more stable reference cavity. We also could improve our preparation routine, as with the parallel preparation method described in Chap. 4, we do not have sufficient control of the comb finesse or tooth shape to prepare an AFC with the ideal shape.
5.3 Memory performance

Figure 5.6: Amplitude and frequency chirp profile of the waveform used for the control pulses.

Increasing the AFC preparation beam waist even further could also improve the shape of the AFC.

5.3.2 Spin-wave storage characterisation

We now turn to the characterisation of the on-demand memory. As described in Sec. 3.3, spin-wave storage requires coherent transfer of the optical excitation to an excitation between the $1/2_g$ and $3/2_g$ levels. To do so, we use control pulses with a Gaussian amplitude profile with a 2.5 $\mu$s full width at half maximum, and a hyperbolic tangent frequency chirp that spans a total of 4.2 MHz. We use this type of frequency chirped pulse, which should be adiabatic, rather than a $\pi$-pulse as it can achieve a higher transfer efficiency at weaker Rabi frequencies [216]. Note that the duration of the control pulse will affect the number of temporal modes that can be stored by the memory, as discussed in Sec. 5.6. Figure 5.6 shows the normalised profile of the pulse that is sent to the AOM. The actual amplitude profile of the light before it is sent to the crystal can be seen in Fig. 5.8.

We can calculate $\eta_T$, the transfer efficiency of one control pulse, by sending a single control pulse and observing the decrease in the AFC echo efficiency:

$$\eta_T = 1 - \frac{\eta'_{AFC}}{\eta_{AFC}}$$

where $\eta'_{AFC}$ is the AFC efficiency after applying a single control pulse. For the control pulses used in our experiments, $\eta_T = 77(1)\%$. 

The SW efficiency, which directly affects the second-order cross-correlation $g^{(2)}_{SW,i}$, is related to the AFC efficiency $\eta_{AFC}$ according to

$$\eta_{SW} = \eta_{AFC} \eta_T^2 \eta_C$$

(5.3)

where $\eta_T$ is the transfer efficiency of a single control pulse, and $\eta_C$ denotes the proportion of the signal remaining after decoherence from the Gaussian inhomogeneous broadening of the hyperfine levels. The decay is given by Eq. 3.28, which for our storage time of $T_s = 6.9 \ \mu s$ results in $\eta_C = 0.93$. Note that $\eta_T$ can also be calculated using this equation, either by assuming both pulses have the same efficiency or that the transfer efficiencies differ between the two pulses. Assuming the pulses are equal, then the transfer efficiency is 56(2)%. In the latter case, $\eta_T^2$ can be replaced with $\eta_{P1} \eta_{P2}$, where efficiency of the first pulse, $\eta_{P1}$, is the value given by Eq. 5.2 and can be used in Eq. 5.3 to calculate the efficiency of the second pulse, $\eta_{P2}$. Then, the efficiency of the second control pulse is 40(3)%. See App. B for further details on how the transfer efficiency was calculated. From these measurements, we see that the second control pulse has a much lower efficiency than the first. The reason for this is unclear. There could be some other dephasing mechanism at play which we do not understand, or the first control pulse does not actually transfer all of the population removed from the AFC to the spin state. Regardless of if we assume the control pulses have equal efficiencies or not, in our experiments $\eta_{SW}$ is limited by $\eta_T$ rather than $\eta_C$.

We can easily increase $\eta_T$ by using more power for our control pulses. However, this will result in the generation of more noise unless we have perfectly emptied the population of the $3/2_g$ level. Our control pulse power was chosen specifically to maximise the signal-to-noise ratio (SNR) for experiments using weak coherent pulses as an input, which should correspond to a maximum in $g^{(2)}_{SW,i}$. To investigate the effect of the control pulse power, we performed a characterisation using weak coherent pulses as an input. We used input pulses with an amplitude profile matching that of the photons produced by the photon-pair source ($\sim 120$ ns full width at half maximum), with around 0.36 photons per pulse. Figure 5.7 shows how both the noise floor and SW echo efficiency increase with respect to control pulse power. In terms of $\mu_1$, defined as the ratio between the number of input photons per pulse and the signal to noise ratio, $\mu_1 = n_{input}/\text{SNR}$,
5.3 Memory performance

Figure 5.7: Measurements of (a) $\mu_1$, (b) spin-wave storage efficiency $\eta_{SW}$ and (c) noise floor (in a 500 ns window) versus applied control pulse power using weak coherent pulses as an input. For these measurements, the number of input photons per pulse was approximately 0.36 and $T_b = 6.9 \mu s$. The points corresponding to the power used for the measurements with single photons as an input are indicated with a box (8.5 mW).

The minimum occurs for a power of 8.5 mW, corresponding to $\mu_1 = 0.0110(5)^2$, $\eta_{SW} = 7.3(3)\%$, and $4.9(2) \cdot 10^{-4}$ photons per trial (in a 500 ns window). We used this power for the experiments presented here. For the maximum power investigated, $\mu_1 = 0.014(1)$, $\eta_{SW} = 11.5(7)\%$ and the noise floor was $9.8(6) \cdot 10^{-4}$

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2The error given for this value comes from the Poissonian statistics of the measurement only, and does not include the uncertainty of the input photon number. The latter was not measured but was likely greater than the photon statistics in most cases.
5. Spin-wave multimode quantum memory

Figure 5.8: Coincidence histograms showing the time difference between the detection of idler and signal photons. Three different histograms are shown here, corresponding to the signal photons passing through a transparency window in the QM (“Input”, brown), after storage in an AFC for 10 µs (“AFC echo”, red), and after storage in the AFC and spin-state for 10 and 6.9 µs respectively (“SW echo”, orange). The darker region of each histogram indicates the portion of counts considered for correlation and efficiency analyses. CP1 and CP2 (indicated in grey with an arbitrary maximum amplitude) are the two control pulses, measured on a photodiode before being sent to the QM.

photons per trial. Further optimisation of the temporal and spectral shape of the CPs is required in order to obtain a higher $\eta_T$ without increasing the noise floor, which would then result in a larger $g^{(2)}_{SW,i}$.

5.3.3 Single photon storage performance

Using the 10 µs AFC and the control pulses characterised in the preceding subsections, we perform AFC and SW storage using single photons from the source as the input for the memory. Figure 5.8 shows a combination of three different coincidence histograms, corresponding to the cases when the signal photon passes through a transparency window in the memory, when the photon is stored in the AFC only, and when the photon also undergoes SW storage. We store photons in the spin state for $T_z = 6.9$ µs with the two control pulses labelled CP1 and CP2. The storage efficiencies were 19.7(6)% and 6.2(3)% for AFC and SW storage, respectively.

Note that here the analysis crystal was only used as a filter. We prepared a 6 MHz wide transparency window centered at the frequency of the AFC to allow the signal photons to pass through. The optical depth outside of this region was around 6. This crystal is a critical component for noise filtering in SW storage, but not necessary for measuring the “Input” photon or AFC echo.
We also calculated the second-order cross-correlation for all three measurements. For the photons which passed through a transparency window in the memory crystal, \( g_{s,i}^{(2)} = 23.3(2) \) for a 280 ns window of time. This is the window size used for all of the cross-correlation measurements presented in this chapter. When stored for 10 \( \mu \)s in the AFC, it was \( g_{AFC,i}^{(2)} = 91(4) \). The increase in \( g_{AFC,i}^{(2)} \) with respect to \( g_{s,i}^{(2)} \) is due to the echo being retrieved in a temporal window where the pump laser is off and thus has a lower noise level, as well as spectral filtering from the inhomogeneous broadening of the Pr\(^{3+} \) ions [44, 217].

To perform SW storage, we send the control pulses conditioned on the detection of an idler photon. In order to measure the second-order cross-correlation after SW storage, \( g_{SW,i}^{(2)} \), we also need to measure the noise level in the same temporal region as the echo signal counts. To do this, we perform semiconditional spin-wave storage [44]. The main source of noise in this region is generated by the CPs, which in our experiment appears to be fluorescence\(^3 \) resulting from excitation of residual population in the \( 3/2_g \), so we can estimate the noise by sending CPs without any input photon. After performing the storage attempt, we subsequently sent \( N \) pairs of CPs every 400 \( \mu \)s, and calculated the noise level based on the average noise counts in the same temporal position after the CPs as where the echo would be. The value of \( N \) is chosen to minimise the measuring time require to obtain a \( g_{SW,i}^{(2)} \) with sufficient error (for example, \( N = 10 \) for these coincidence measurements), while the 400 \( \mu \)s repetition period was chosen to minimise the residual fluorescence from the CPs after every storage trial. This resulted in \( g_{SW,i}^{(2)} = 9.8(6) \), with a noise floor of \( 8.3(4) \cdot 10^{-4} \) photons per trial. This noise floor is higher than the one measured with weak coherent states in the previous section, which is likely because the single photon storage measurement was performed over a much longer time and was thus subject to more experimental instabilities that affect the noise.

The coincidence rates measured at the signal detector in the window of the echo were 3.8 counts/second and 0.062 counts/second for AFC and SW storage, respectively. The SW coincidence rate was much lower than that of the AFC due to the detection dead time imposed by the semiconditional SW storage sequence (4.4 ms for every idler detected). If we instead consider the

\(^3\)In Ref. [218] the noise was attributed to FID from the CPs. Here we believe the source is fluorescence instead, as we have a larger temporal separation between the second pulse and the SW echo, plus the timescale of the decay of the noise is longer than that of the signal from the control pulses.
number of coincidences per idler detected, we measured $3.8 \cdot 10^{-3}$ and $1.2 \cdot 10^{-3}$ coincidences per idler for AFC and SW storage, respectively. The AFC echo coincidence rate was nearly 60 times higher than previous work from our group [44], in part due to using a detector for the idler that is eight times more efficient, but otherwise due to improvements in the experiment, while the SW echo rate was only about three times higher, again limited by the dead time imposed by the experiment.

To summarise the performance of our system, the main parameters are shown in Tab. 5.1, with the same parameters from the previous work of our group, Ref. [44], to highlight the improvements we made in our system which allowed us to perform the following experiment measuring entanglement between a spin-wave and a telecom photon. Note that here the values for the noise floor and $g^{(2)}_{SW,i}$ from Ref. [44] correspond to the measurement made using a semiconditional sequence, like that presented in our experiment here, and our results were recalculated for a 320 ns window. The best value reported in Ref. [44] came from an unconditional measurement, which gave $g^{(2)}_{SW,i(320ns)} = 6.1(7)$, and a noise floor of $1.3(1) \cdot 10^{-3}$ photons per storage trial, where the lower noise floor (and thus higher $g^{(2)}_{SW,i}$) was attributed to a higher number of CPs sent per measurement period.

### Table 5.1: Comparison with our previous work [44].

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Ref. [44]</th>
<th>This work</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta \nu_{\text{pump}}$</td>
<td>&gt; 1 MHz</td>
<td>380(20) kHz</td>
</tr>
<tr>
<td>$\Delta \nu_{\text{bi-ph}}$</td>
<td>2.8 MHz</td>
<td>1.8 MHz</td>
</tr>
<tr>
<td>$T_2^\text{eff}$</td>
<td>33(4) $\mu$s</td>
<td>92(9) $\mu$s</td>
</tr>
<tr>
<td>$\eta_{\text{AFC}}$ ($\tau_{\text{AFC}}$)</td>
<td>11.0(5)% (7.3 $\mu$s)</td>
<td>19.7(6)% (10 $\mu$s)</td>
</tr>
<tr>
<td>$\eta_T$ (^a)</td>
<td>72.5%</td>
<td>77%</td>
</tr>
<tr>
<td>$\gamma_{\text{inhom}}$</td>
<td>20(3) kHz</td>
<td>16.1(7) kHz</td>
</tr>
<tr>
<td>$T_S$</td>
<td>6 $\mu$s</td>
<td>6.9 $\mu$s</td>
</tr>
<tr>
<td>Noise floor (320 ns)</td>
<td>2.0(1) $\cdot 10^{-3}$</td>
<td>0.99(4) $\cdot 10^{-3}$</td>
</tr>
<tr>
<td>$g^{(2)}_{SW,i(320ns)}$</td>
<td>5.0(3)</td>
<td>9.1(5)</td>
</tr>
</tbody>
</table>

\(^a\) Calculated using Eq. 5.2.
5.4 Measuring light-matter entanglement

Figure 5.9: Visibility fringes and relevant coincidence histograms for the maximum and minimum values of interference (the darker regions represent the coincidence windows used). The fringes correspond to (a) the signal photon not stored in the memory, (b) AFC storage and (c) semiconditional SW storage. In (a) and (b), the two fringes correspond to two different idler interferometer phases that differ by \( \pi/2 \). In (c) the two fringes correspond to two different signal interferometer phases that differ by \( \pi/2 \). The error bars correspond to one standard deviation. The shaded area represents the maximum \( g^{(2)}_{SW,i} (\pm 1\sigma) \) we expect in the ideal scenario, while the dashed gray line is the minimum value.
5.4 Measuring light-matter entanglement

To measure the quality of the entanglement, we first verified the combined performance of our two analysers by measuring energy-time entanglement of the photon-pair alone, without storage in the QM. To do this, we prepared a 16 MHz wide spectral pit in the QM, and a double AFC in the analysis crystal for use as an interferometer, as described in Chap. 4. We fixed the phase difference between the idler interferometer paths, and swept the phase of the double AFC to measure a fringe of interference. We repeated this for another idler interferometer phase that was shifted by $\pi/2$ with respect to the first fringe. The resulting interference fringes are shown in Fig. 5.9(a). Note that here the y-axis is given in terms of coincidences normalised to a particular noise window as an approximation for $g^{(2)}_{s,i}$ and $g^{(2)}_{\text{AFC},i}$ which will be detailed later in this section. We measured visibilities of 91(6)% and 89(3)% by fitting sinusoids to the data. This is well above the threshold of 70.7% required to violate a Clauser-Horne-Shimony-Holt (CHSH) inequality [200], showing the photons are indeed entangled as expected. Note that here we are not concerned with demonstrating non-locality with a CHSH violation, but to do so we would need to assume a sinusoidal dependence of the interference on the phase shift of the interferometer.

Next, we analysed the storage of the entanglement in the excited state of the QM. The signal photon was stored for 10 $\mu$s in the AFC, and again we used a double AFC in the analysis crystal. We swept the phases in the same way as for the previous measurement and obtained the fringes shown in Fig. 5.9(b). The resulting visibilities were 90(3)% and 88(3)%, 6 and 5 standard deviations above the 70.7% threshold, respectively. These visibilities, $V$ resulted in an average two-qubit conditional fidelity of 92(2)% for the light-matter entanglement [200], which is calculated as $(3V + 1)/4$, assuming white noise [205].

One consequence of using the double AFC as an interferometer is that we do not have a temporal window of noise (equivalent to the noise where we measure interference) with which we can calculate $g^{(2)}_{s,i}$ or $g^{(2)}_{\text{AFC},i}$. To calculate $g^{(2)}_{s,i}$ we use a temporal window where the pump is turned on, before the transmitted photon is detected. To calculate $g^{(2)}_{\text{AFC},i}$, we consider a temporal window after the pump has been turned off but before the AFC echo has been emitted, where there is only stored pump noise. If only the double AFC is used for example, then we should consider a noise window after the pump is switched off but before the first echo of the double AFC, but due to the non-instantaneous switching off of the
pump, there is no such window. This is also the case if there is also an AFC in the memory. Hence, we must use a noise window before the transmitted signal (if there is a spectral pit prepared in the memory) or before the AFC echo (if there is an AFC preparation in the memory) that would have more noise counts than the temporal window in which we measure interference. Using this window of noise would underestimate $g^{(2)}_{s,i}$ and $g^{(2)}_{AFC,i}$, so instead we refer to this value as the normalised coincidences in Fig. 5.9(a,b). This is also why the normalised coincidences are below one at the minimum of the interference fringes.

Additionally, for these interference fringes measured with a transparency window or AFC storage alone, the experimental conditions were slightly different to that depicted in Fig. 5.1. The etalon was in its original position directly after the source (not after the memory), and the signal photon traveled from the QM to the analysis crystal in free space through two single-pass AOMs instead of an optical fibre. Under these experimental conditions, we also measured coincidences without the interferometers, where the $g^{(2)}_{s,i}$ through a transparency window in the QM was 24(1), and $g^{(2)}_{AFC,i}$ was 61(7). The decrease of $g^{(2)}_{AFC,i}$ with respect to the value quoted Sec. 5.3 was due to the AFC not yet being well-optimised (thus being less efficient) at the time of the interference fringe measurements.

For the case of SW storage, we stored the photons for 10 $\mu$s in the AFC, and 6.9 $\mu$s in the spin state. This time we used a single AFC in the analysis crystal as an interferometer (see Sec. 4.5.2), and fixed its phase while sweeping the idler interferometer phase instead. We measured $g^{(2)}_{SW,i}$ in a semiconditional manner as described in Sec. 5.3.3. The resulting fringes are shown in Fig. 5.9(c), with visibilities of 71(3)% and 68(5)%, leading to a two-qubit fidelity of 77(2)%\(^4\). These values do not exceed the CHSH limit, but they do exceed the 33% bound for separable states by 12 and 7 standard deviations, respectively [203, 204]. This measurement thus demonstrates entanglement between a telecom photon and a solid-state on-demand multimode quantum memory.

We then varied the time $T_s$ between the CPs to investigate the effect of the storage time on the quality of the correlations and entanglement. We first measured the efficiency $\eta_{SW}$ as well as $g^{(2)}_{SW,i}$ as a function of $T_s$ from 0 to 37.7 $\mu$s. Figure 5.10(a) shows the storage efficiency as a function of $T_s$. The efficiency decays with increasing $T_s$, which is expected due to $\gamma_{inhom}$, the inhomogeneous

\(^4\)We note that we do not observe any interference in the noise at the temporal window of the SW echo used to calculate $g^{(2)}_{SW,i}$. 
broadening of the spin transition (see Sec. 3.3). The efficiency decays as [182]

$$\eta_{SW} = a_\eta e^{-(T_s \gamma_{inhom} \pi)^2/[2 \ln(2)]},$$

(5.4)

where $a_\eta$ is $\eta_{SW}$ for $T_s = 0$ (obtained from the fit). By fitting the data to this function, we obtained $\gamma_{inhom} = 16.1(7)$ kHz.

The cross-correlation versus $T_s$ is shown in Fig. 5.10(b), which also decays with increasing time due to the spin inhomogeneous broadening. Similarly, we
can fit the second-order cross-correlation as

\[ g^{(2)}_{SW,i} = a_g^{(2)} e^{- \left( T_s \gamma_{inhom} \pi \right)^2 / \left[ 2 \ln(2) \right]} + 1, \]  

(5.5)

where \( a_g^{(2)} \) is obtained from the fit, and obtained \( \gamma_{inhom} = 14.8(7) \) kHz. The reduction in \( \gamma_{inhom} \) compared to Ref. [44] is due to the addition of mu-metal shielding inside the cryostat chamber.

Finally, we measured the visibility as a function of \( T_s \). To do so, we measured a single interference fringe (where \( \Delta \phi_i \) is varied but \( \Delta \phi_s \) is fixed) for each value of \( T_s \) and obtained the visibility from a sinusoidal fit. The results are shown in Fig. 5.10(c), along with dashed lines corresponding to the upper and lower bound of the expected visibility. We see that for all of the storage times measured, the visibility was higher than the 33% bound for separable states, and thus we have stored entanglement in our QM for a total time of 47.7 \( \mu s \), including the AFC storage time.

Regarding the bounds for the expected visibility, if the visibility of the entanglement is only limited by photon statistics, the maximum of interference is given by \( g^{(2)}_{SW,i} \) and the minimum is 1. Then, \( V = (g^{(2)}_{SW,i} - 1) / (g^{(2)}_{SW,i} + 1) \) [193]. Figure 5.10(c) shows the upper and lower bounds in that scenario as dashed lines, calculated using the values of \( g^{(2)}_{SW,i} \) measured in Fig. 5.10(b). We see that this does not correspond to our measured \( V \), in particular for shorter values of \( T_s \).

The formula above is based on the assumption of having an ideal setup, e.g., having perfect analysers. As we know that this is not the case in our setup, and the initial entangled state is not perfect (see Sec. 5.5), we need to account for these imperfections. By considering that both the maximum and minimum \( g^{(2)}_{SW,i} \) will decay with \( T_s \) following a Gaussian curve, we obtain a new expression for the visibility as a function of \( T_s \):

\[ V = \frac{(a_{g^{(2)}} - b) e^{- \left( T_s \gamma_{inhom} \pi \right)^2 / \left[ 2 \ln(2) \right]}}{(a_{g^{(2)}} + b) e^{- \left( T_s \gamma_{inhom} \pi \right)^2 / \left[ 2 \ln(2) \right]} + 2}, \]  

(5.6)

where \( a_{g^{(2)}} \) and \( \gamma_{inhom} \) are the values that come from the fit to \( g^{(2)}_{SW,i} \) to \( T_s \), and \( b \) is a free parameter that we fit to using the data from Fig. 5.10(c). At \( T_s = 0 \), the minimum of interference should be \( b + 1 \). The fit corresponds to the shaded region in Fig. 5.10(c), from which we obtain \( b = 0.8(3) \).
Note that for $T_s = 37.7 \mu s$, the value of $g_{SW,i}^{(2)}$ is 1.8(6), which is below 2. The corresponding measured visibility of 44(8)% ensures the non-classicality of the correlations between the idler and retrieved spin-wave. Additionally, the more precise classical limit for $g_{SW,i}^{(2)}$ is given by the Cauchy-Schwarz inequality (Eq. 3.35), where $g_{SW,i}^{(2)}$ should exceed $\sqrt{g_{SW,SW}^{(2)}g_{i,i}^{(2)}}$. While we did not measure these particular autocorrelations, for comparison, in Ref. [44], they measured $g_{SW,SW}^{(2)} = 1.0(4)$ and $g_{i,i}^{(2)} = 1.32(4)$, placing the limit at $\approx 1.15$. Thus it is possible to still exceed the classical limit set by the Cauchy-Schwartz inequality even with $g_{SW,i}^{(2)} < 2$.

5.5 Limitations to measurable visibilities

In the following section, we will discuss the factors which limit the measurable visibility of entanglement with spin-wave storage. The main limitations are the quality of the correlations ($g_{SW,i}^{(2)}$ measured without interferometers), the pump laser linewidth, and the interferometers. With perfect analysers we should be able to measure an interference fringe with a visibility of 81% given the measured $g_{SW,i}^{(2)}$ of 9.8(6) (see the preceeding section). From Sec. 4.5.1, we know the idler interferometer visibility for 1436 nm light generated with the 426 nm pump laser is 91(3)%, primarily due to the pump laser linewidth, and that the estimated visibility of the single AFC interferometer is 98%. By multiplying these three values together, we would expect to measure a visibility of 73%, which is within the error bars of the visibilities we measured.

Even with a perfect signal analyser, our expected visibility (due to the source pump laser and the value of $g_{SW,i}^{(2)}$) would be 74%, which would make it difficult to exceed the 71% threshold for violating a CHSH inequality within experimental error. An improved pump laser linewidth and/or $g_{SW,i}^{(2)}$ would thus be required. A new laser or better locking cavity would be needed to improve the linewidth, and would already lead to a substantial improvement in visibility. Strategies to improve $g_{SW,i}^{(2)}$ will be discussed in Sec. 5.7.

The visibility is also affected by the temporal window we use to calculate $g_{SW,i}^{(2)}$. Figure 5.11 shows the visibility versus window size (for a fixed central time bin) for the two fringes measured, as well as the corresponding number of coincidences at the phase of maximum interference. We chose to use a window size of 280 ns as a balance between visibility and count rate, since we can increase
the visibility slightly by using a smaller window at the expense of decreasing the count rate.

5.6 Temporal multimode SW storage of weak coherent states

One advantage of using Pr\(^{3+}\):Y\(_2\)SiO\(_5\) for SW storage is that the Rabi frequencies for its optical transitions are higher than that of other dopants which otherwise possess longer coherence times (and thus storage times), like Eu for example. For that purpose, we further investigated the temporal multimode performance of SW storage in Pr\(^{3+}\):Y\(_2\)SiO\(_5\).

In Ref. [129], we note that in the experiment presented in the preceding sections, given the CP width of 3 \(\mu\)s and AFC storage time of 10 \(\mu\)s, we can store 17 independent temporal modes with a width of 420 ns. However, Ref. [60] develops theoretical limits to actual temporal multimode capacity based on the Nyquist-Shannon sampling theorem, due to limits imposed by the bandwidth of the AFC used in an experiment. The following equations presented in this section come from this paper.

As noted in Chap. 3, the minimum temporal mode size, independent of any specific profile, is given by

\[
T_m = \frac{2.5}{\Gamma},
\]  
(5.7)
where $\Gamma$ is the bandwidth of the AFC structure. If SW storage is involved then the number of temporal modes that can be stored $N_{SW}^t$ is

$$N_{SW}^t = \frac{1}{\Delta - T_C} T_m, \quad (5.8)$$

where $T_C$ is the total length of time taken by the control pulses. Therefore, the number of the temporal modes that can be stored will decrease if the control pulse length increases.

By taking into account the decay of AFC efficiency with storage time, and the control pulse efficiency in the most ideal case, Eq. 5.8 can be rewritten as

$$N_{SW}^t = \frac{1}{2.5} \left( \frac{\ln(1/\eta r_2)}{4} \Gamma r_2 - \chi \frac{\xi}{\pi^2} \frac{\Gamma^2}{\Omega^2} \right), \quad (5.9)$$

where $\eta r_2$ is equivalent to Eq. 5.1, $\chi$ is factor related to the control pulse shape and $\xi$ to its efficiency (see Ref. [60]), and $\Omega$ is the Rabi frequency of the pulse. The first term has been rewritten such that it depends on the coherence time of the transition and comb bandwidth, while the second term gives the required control pulse time given the Rabi frequency, comb bandwidth, and parameters of the pulse profile. Thus a specific control pulse profile is required in this scenario.

To observe the temporal multimode capacity of our memory, we performed storage of weak coherent states with our memory. We used the memory setup as described in Sec. 5.2, but instead of using signal photons as an input, we used laser pulses attenuated with neutral density filters such that we had an average of 0.90 ± 0.05 photons per mode. We used temporal modes with a Gaussian intensity profile, where the full-width at half maximum was 180 ns and the input mode size was $T_m = 620$ ns, based on the limits set by Eq. 5.7. To maximise the number of stored modes, we used the longest storage time we had previously utilised with single-photon storage, which was 25 $\mu$s (see Fig. 5.5), which had an efficiency of 7.7(3)% in the measurements performed in this section. Like the experiment described in the rest of this chapter, the AFC bandwidth was approximately 4 MHz, and the control pulses used were the same as described in Sec. 5.3.2. In this case, the transfer efficiency was 71(1)%, calculated using Eq. 5.3 which assumes both pulses have the same transfer efficiency. Otherwise, for the first pulse, the transfer efficiency calculated using Eq. 5.2 is 84(1)% and
5.6 Temporal multimode SW storage of weak coherent states

Figure 5.12: Input modes and retrieved spin-wave modes for (a) 20 input modes and (b) 30 input modes. The input modes measured through a transparency window in the memory are shown in red, while the retrieved spin-wave modes are shown in orange, and the equivalent noise without storage is shown in light orange. The counts were normalised to the number of triggers sent per measurement, where one trigger corresponds to one trial of sending a train of input modes.

for the second pulse the efficiency is 60(1)%, calculated using the result for the first pulse and Eq. 5.35.

Figure 5.12 shows the retrieved temporal modes after SW storage, where we looked at two cases: storage of 20 modes, and storage of 30 modes. For 20 input modes, the average SNR was 17.4(4), with $\eta_{SW} = 1.88(3)\%$. For 30 modes, the average SNR was 6.7(1), with $\eta_{SW} = 0.63(1)\%$. The SNR and $\eta_{SW}$ decreased for a larger number of modes because different $T_S$ were used: 14.1 $\mu$s and 20.7 $\mu$s for 20 and 30 modes, respectively. There is a residual signal from the AFC echoes due to the finite efficiency of the controls pulses, so $T_s$ has a minimum duration required to avoid overlap between the SW output modes and the residual AFC echoes, which depends of the number of modes stored. The

---

5These values were calculated using a measurement with 20 input modes, and averaged across all the modes except when calculating the transfer efficiency of the first pulse. In that case, some AFC modes overlapped with the control pulse so only 13 modes were used in the calculation.
residual signal from the AFC echoes can be reduced significantly by improving the control pulse efficiency, but not entirely, so this minimum duration will still be necessary. Due to dephasing in the spin state, increasing \( T_s \) will reduce the SW efficiency (see Sec. 3.3), thus the input mode number will affect the efficiency. In this experiment, the loss due to dephasing was also higher than that in Sec. 5.4 due to a larger \( \gamma_{\text{inhom}} \) (26.3 kHz) from imperfect shielding of stray magnetic fields. If we consider the \( \gamma_{\text{inhom}} \) measured for the previous SW experiments (16.1 kHz), the efficiencies would increase to approximately 3.5% and 2.4% for 20 and 30 modes, respectively.

Given the \( T_C \) of 5 \( \mu \)s, the ideal \( T_m \), and the AFC storage time of 25 \( \mu \)s, from Eq. 5.8 it should be possible to store 32 modes, which is close to the 30 modes stored here. However, the control pulses we used have neither the ideal profile or optimum transfer efficiency. Using Eq. 5.9, which assumes a higher transfer efficiency (taking \( \xi = 4 \) and \( \chi = 1.36 \) like the experiments performed with Eu in Ref. [60]), using \( T_{2e}^{\text{eff}} \) as \( T_2 \) and an estimated Rabi frequency of 666 kHz, the maximum number of possible modes is 32\(^6\). As with the result using Eq. 5.8, this is close to the number of modes we actually stored in the experiment. Although the transfer efficiency of our control pulses is low, given the Rabi frequency of the transition, we should be able to improve this to achieve higher SW efficiencies while maintaining the same number of temporal modes. Nonetheless, the measurements reported here have the highest number of temporal modes stored with on-demand retrieval at the single-photon level, showing the benefit of using materials allowing strong Rabi frequencies for temporally multimode spin-wave storage.

5.7 Discussion

The experiment presented in Sec. 5.4 demonstrated entanglement between a telecom photon and an on-demand multimode quantum memory for the first time, and for a total storage time of nearly 50 \( \mu \)s. If we consider a scenario when the photon would need to be sent over some distance, this storage time allows for

\(^6\)The value of the Rabi frequency reported in Ref. [60] was 410 kHz, which gave the maximum number of modes as 19. However, this value was measured with an incorrect transition that has a lower Rabi frequency than the \( 3/2_g \rightarrow 3/2_e \) transition, and the value was also mistakenly written at 410 kHz instead of the measured 416 kHz. Using Ref. [149], we estimate the Rabi frequency of the \( 3/2_g \rightarrow 3/2_e \) transition to be 666 kHz.
propagation through 10 km of fibre. Further developments and improvements are still required in order for the memory to be useful in a more practical scenario (i.e., for actual deployment over 10s of kilometres).

The first point would be that the storage time need to be long enough for the communication time required by an experiment. It is possible to extend the storage time in the spin state using spin-echo and dynamical decoupling techniques \cite{45}, which requires the use of RF pulses applied to the crystal. With no applied magnetic field to the system, the coherence time of this transition is around 500 $\mu$s \cite{150}, which means that storage times of 100s of $\mu$s are quite feasible without adding significant complexity to the setup or a large decrease in the efficiency. More complex RF sequences could increase the storage time to a few milliseconds.

To increase the storage time further, we would need to apply external magnetic fields. Some improvement could be expected in the low-field regime, which has been achieved with europium dopants \cite{183}. The longest storage times can be achieved with higher fields through the use of magnetic-field insensitive transitions \cite{59, 70, 151}, where storage of classical light pulses for up to one minute has been achieved in Pr$^{3+}$:Y$_2$SiO$_5$ \cite{73}, as well as storage of weak coherent states (with around 10s of photons per pulse) for over one second \cite{219}. However, the spin levels of Pr$^{3+}$:Y$_2$SiO$_5$ are no longer degenerate in this scenario, which makes optical pumping and class cleaning more complicated, and the splitting between adjacent levels reduces, which would require a narrower linewidth photon-pair source for efficient storage.

One issue with applying RF pulses is that they will never be 100% efficient, and thus will inevitably introduce noise to the measurement. It will therefore be of great importance to be able to maintain the same $g_{SW,i}^{(2)}$ as reported here, either by increasing signal counts, or reducing noise counts. It would also be of benefit for this type of entanglement experiment, where $g_{SW,i}^{(2)}$ also limited the visibility of the SW storage experiment.

There are several ways in which $g_{SW,i}^{(2)}$ could be improved. On the memory side, Sec. 5.3 goes through the current memory performance in terms of AFC and SW storage in detail. Any improvements in $\eta_{AFC}$ would also lead to an improvement in $\eta_{SW}$ and thus $g_{SW,i}^{(2)}$. Improvements in the AFC preparation could increase this value by at least a few percent. A larger increase could be achieved by using an impedance matched cavity \cite{178}, but it is still unknown how the noise
generated by SW storage is affected by such a cavity. Current experiments in the
group are currently working towards realising this type of memory.

Increasing $\eta_{\text{SW}}$ can be easily done by increasing the CP power, but as Fig. 5.7 shows, this also increases the noise. Using more efficient CPs could allow us
to increase the efficiency and potentially do so without increasing the noise. The hyperbolic-square-hyperbolic pulses from Ref. [220], which should give the
highest efficiency, have been successfully utilised in experiments using other
dopants [45, 60, 72]. This pulse type is the one considered in Eq. 5.9, and thus
may be needed in order to achieve more efficient temporal multimode SW storage.

Regarding reduction in noise from the CPs, as the experimental sequence
(the class cleaning sequence, for example) has been optimised to minimise the
noise floor, it is not so trivial to decrease it further, but is still necessary. Aside
from improvements in the optical pumping sequence, sending the signal photons
through the analysis crystal a second time could reduce the noise further (a
“double-pass” arrangement), depending on the frequency of the noise counts in
the current configuration.

Finally, $g_{\text{SW},i}^{(2)}$ can also be increased by improving the heralding efficiency of
the photon-pair source, which is currently 25%. This value essentially means
we have 0.25 signal photons per idler click, and any improvement in this value
would directly increase $g_{\text{SW},i}^{(2)}$ without increasing the noise floor. A major change
in the source that could be done without sacrificing other important characteristics
would be to replace the PPLN crystal with another crystal that has less losses. Currently a new generation of photon-pair sources is being built partially for this
purpose.

The multiplexability of the system is also critical for obtaining good count
rates in long-distance entanglement experiments. We investigated temporal mul-
timodality in this chapter, but other degrees of freedom, i.e., spectral [83] and
spatial [86] are also open to this system, which we could utilise to boost the count
rate of any kind of experiment we would like to perform.

Overall, our current setup achieves many of the requirements for a quantum
repeater system, as shown in these experiments. There are still many improve-
ments needed in order for it to actually be practical, but there are plenty of feasible
though challenging routes for improvement.
Chapter 6

Transmission of light-matter entanglement over the metropolitan area of Barcelona

In this chapter, I will present experiments demonstrating non-classical correlations and light-matter entanglement between telecom photons transmitted through deployed optical fibres across the metropolitan area of Barcelona and a Pr$^{3+}$:Y$_2$SiO$_5$-based quantum memory. Here we use the AFC protocol for the memory with solely optical storage. We measured non-classical correlations and entanglement with all of the setup and photon detection in the same laboratory (similar to Chap. 5), however the idler photons additionally travel through an extra distance in a fibre spool, or through deployed optical fibre. The last experiment decouples the idler photon detection from the rest of the experiment, and instead travels through 44 km of deployed fibre in the metropolitan area of Barcelona, and is detected at a different location, where we measure non-classical correlations between the stored signal photons and the idler photons.

The main contents of this chapter come from a manuscript currently in preparation. I am a co-first author, along with Dr. Samuele Grandi and Dr. Sören Wengerowsky. I operated the memory setup (as described and developed in Chap. 4 and Chap. 5), acquired the data along with Dr. Samuele Grandi, Dr. Sören Wengerowsky, Dr. Dario Lago-Rivera, and Dr. Félicien Appas, and analysed the data along with Dr. Samuele Grandi and Dr. Dario Lago-Rivera. The logistics and setup for the long-distance aspects of the experiment were done by Dr. Samuele
Grandi and Dr. Sören Wengerowsky. The source was operated by Dr. Samuele Grandi, Dr. Sören Wengerowsky, and Dr. Dario Lago-Rivera.

6.1 Introduction

In Chap. 5, we demonstrated light-matter entanglement between a quantum memory and a telecom heralding photon in a laboratory. If this setup were to be implemented as part of a quantum repeater, the heralding photon would need to be sent to and detected at another location via optical fibre. Several of the experiments mentioned in Chap. 1 measure light-matter (or matter-matter) entanglement with a telecom heralding photon which travels through a fibre spool \([105–107, 111, 114]\), and some also detect the telecom photon in a different location to where it is generated \([111, 114]\).

In this direction, we test the performance of our system in a similar, practical scenario, to verify that the entanglement between the telecom photon and the memory is not degraded by the telecom (idler) photon travelling through deployed fibre. The idler photons will not only experience exponential loss proportional to the fibre length (as discussed in Chap. 1), but also any deployed fibre will have additional loss from connections or splices in its path. It is thus of interest to characterise deployed fibres and see that they work as intended, especially given that we work in the non-standard telecom E-band (1436 nm). As we have access to deployed fibre in the area, we perform entanglement measurements in this chapter, as well a coincidence measurement with the telecom photon detected in another building located 16 km away (44 km in fibre).

6.2 Light-matter entanglement

6.2.1 Experimental setup

The setup used to measure entanglement is shown in Fig. 6.1. For these measurements, all of the photon detection still occurs in the same lab, but in some cases we send the idler photons through a 50 km loop of deployed optical fibre. This setup is generally very similar to that of Chap. 5, using the Franson scheme to measure energy-time entanglement. The photon-pair source and quantum memory setup also follow the more detailed description in Chap. 4. The main
differences occur in the idler photon optical and detection paths, as a consequence of the longer travel time.

The idler photons pass through some fibre of length $d_{\text{idler}}$, and so an additional bandpass filter (BPF, central wavelength of 1440 nm, linewidth of 14 nm)\(^1\) must be placed after the fibre to filter out noise counts from any stray light coupled to it\(^2\). The photon detection clicks also pass through electrical-to-optical transducers (ETO, AFL-4800 from AA Lab Systems) and a 50 km fibre spool before being sent to the TDC, in order to mimic a decoupled long-distance measurement where the detector would not be located in the lab. If $d_{\text{idler}}$ is only a few metres (i.e., no added fibre distance, referred to as 0 km), the ETOs and 50 km fibre spool are omitted\(^3\).

On the signal photon side, unlike Chap. 5 we do not perform spin-wave storage, so there is no control beam and the arrangement of the filtering elements follows the setup described in Chap. 4. We also use a single-AFC interferometer in the analysis crystal as the signal photon analyser. This type of interferometer is likely to introduce more noise to the measurement compared with the double-AFC interferometer (see Sec. 4.5.2), but the overall transmission is higher, which is

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\(^1\)This BPF is identical to the BPF located before the interferometer.

\(^2\)The deployed fibre passes through several rooms at ICFO, as well as other rooms in the other buildings noted in Fig. 6.2, which can contribute to the background counts.

\(^3\)We used the 50 km spool to send the classical signal for both $d_{\text{idler}} = 11$ and 50 km.
beneficial for long-distance experiments where the count rate is reduced due to fibre losses.

The entanglement analysis measurements are performed as follows. Photon pairs are generated by the source. The idler photons pass through filtering optics, then through optical fibre of some length \(d_{\text{idler}}\), then are sent directly to the idler interferometer. The photons are then coupled into a free-space path with the aforementioned additional BPF, before being coupled back to optical fibre to be detected by an SPD (see Sec. 4.4). If \(d_{\text{idler}} \neq 0 \text{ km}\), then the TTL signal from the detection click is sent to an ETO, which converts the electronic signal to an optical signal which is modulated such that the TTL signal can be propagated optically. This optical signal is sent to a 50 km fibre spool, introducing a \(\sim 250 \mu s\) delay in the detection time. The detection signal is sent to another ETO, which demodulates the optical signal to output an electronic TTL. This TTL is finally sent to the TDC module to record the photon detection timestamp.

The signal photon passes through filtering optics before being sent to the QM in the cryostat via optical fibre, then free space. The QM is prepared with a 10 \(\mu s\) AFC, with the same characteristics as the one detailed in Chap. 5. The photon then passes through optical fibre again before being sent to the analysis crystal, then to an SPD (COUNT-10C-FC) which sends detection clicks to the TDC.

We also perform coincidence measurements without measuring entanglement. For those measurements, we bypassed the idler interferometer, and we used QM storage times of 10 \(\mu s\) and 25 \(\mu s\). A transparency window was prepared in the analysis crystal instead of an AFC. Note that given these storage times, the signal photon will already have been emitted by the time the idler photon is detected for any measurements (of correlations or entanglement) with added distance (i.e., \(d_{\text{idler}} = 11 \text{ km and 50 km}\)).

We used three values of \(d_{\text{idler}}\) in this experiment: approximately 0, 11 and 50 km. For the 11 km case, we used a fibre spool (SMF-28 Ultra) placed in the same laboratory as the rest of the experiment, with a length of approximately 11.4 km. The measured loss through the spool was 2.33 dB at 1535 nm, where some of loss comes from splices in the fibre. For the 50 km case, we used two deployed fibres. These fibres connect our laboratory at ICFO in the municipality of Castelldefels, to the Centre for Telecommunication and Technologies of Information (CTTI) building located in the municipality of L’Hospitalet de Llobregrat. The locations are marked on the map in Fig. 6.2. We connected the two fibres
6.2 Light-matter entanglement

Figure 6.2: Map of the metropolitan area of Barcelona marked with the relevant locations and fibre links. The black line indicates the approximate path and length of the fibres, while the dashed line and corresponding marked length indicates the direct distance between locations. Map data taken from Google Earth (DATA SIO, NOAA, U.S. Navy, NGA, GEBCO. Image ©2023 TerraMetrics). Fibre path information courtesy of Xarxa Oberta de Catalunya and i2CAT.

at CTTI such that the idler photons traveled to and from the building for a total distance of approximately 50.2 km, and were ultimately detected at ICFO. We measured 13.3 dB of loss at 1535 nm, and 15.7 dB of loss at 1436 nm for the 50.2 km round-trip.

There is another pair of fibres which connect CTTI to a third location: the i2CAT Foundation, in the municipality of Barcelona. The length of these fibres is 18.9 km, and the loss is 12.4 dB at 1535 nm from ICFO to i2CAT (approximately 44 km). We do not use this fibre link for entanglement measurements, but we do use it for the correlation measurement reported in Sec. 6.3.

When $d_{\text{idler}} = 0$ km, we gate the source pump laser off if an idler count is detected$^4$. This gating significantly reduces the background noise in the temporal window of the AFC echo. However, given these short storage times, if there is substantial distance added such that the memory cannot store the signal photon for the idler photon travel time$^5$, idler-triggered gating is not possible. For $d_{\text{idler}} = 11,50$ km, this is indeed the case, so instead we periodically gated the pump laser on and off for an equal time such that the gating period was twice the AFC storage time. Using this time period allowed for the storage of the maximum number of temporal modes and their re-emission in a noise-free temporal window, which maximised the coincidence rate. Additionally, we gated the TDC with the same period, synchronised with the pump laser gating but with a constant

$^4$We do the same for the measurements in Chap. 5 and Chap. 7.

$^5$Approximately 5 μs of storage time per 1 km of distance.
temporal offset. We calibrated the offset such that the recording window of the TDC coincided with the detection of idler photons generated by the source. In this way, both the coincidence rate and $g^{(2)}_{s,i}$ were maximised. Note that we can keep the pump gate open for this extended period of time thanks to the temporal multimodality of the AFC. Given a 400 ns mode size and the fact that the pump is on for the full AFC storage time, the memory can store 25 or 62 temporal modes for a 10 $\mu$s or 25 $\mu$s storage time, respectively.

6.2.2 Results

In the first instance, we performed coincidence measurements for the three values of $d_{\text{idler}}$. Figure 6.3 shows coincidence histograms for the three values of $d_{\text{idler}}$ investigate and a 10 $\mu$s storage time. Here we can see the difference in the noise pattern between the 0 km and the other two distances. When there is no added distance [Fig. 6.3(a)], there is a drop in background noise from closing the pump gate for around 20 $\mu$s. This drop occurs after detecting an idler photon\(^6\). We use a window after the pump is gated off, but before the echo, as our estimation for the noise at the echo. For the 11 and 50 km measurements [Fig. 6.3(b,c)], the noise has a triangular pattern which arises from the convolution of two square waves (the pump and TDC gates). The photons transmitted through the QM are temporally located at this peak, while the photons absorbed and emitted as an AFC echo are at the minimum of the pump noise background. To estimate the noise level at the echo, we use the background counts that occur with a period equal to the pump/TDC gating period, which is twice the AFC storage time (e.g., 20 $\mu$s for the measurement in Fig. 6.3), with the same window size that we consider for the correlation counts of the echo (400 ns).

Note that the delay is negative in Fig. 6.3(b,c) because the coincidence histograms are built by determining when signal counts are detected relative to an idler count. The signal photon of a pair will be detected before the idler for the measurements where $d_{\text{idler}} = 11$ km or 50 km, hence the delay is negative.

We calculated $g^{(2)}_{\text{AFC},i}$ for all of these measurements, with the results summarised in Fig. 6.4 and Tab. 6.1. For $d_{\text{idler}} = 0$ km and 11 km, the values of $g^{(2)}_{\text{AFC},i}$ are consistent, but for $d_{\text{idler}} = 50$ km, $g^{(2)}_{\text{AFC},i}$ is significantly lower. This is due to the decrease in the detected idler rate, $R^i$, resulting from losses in the

\(^6\)Transmitted photons which were not absorbed by the QM are visible as the peak at the 0 $\mu$s delay.
6.2 Light-matter entanglement

Figure 6.3: Coincidence histograms between the signal and idler photons, where the signal photons are stored in the QM for 10 µs, and the idler photons are sent through (a) 0 km, (b) 11 km, and (b) 50 km of optical fibre. The red region [at (a) 10 µs, (b) -282 µs, and (c) -471 µs] corresponds to the bins considered as part of the AFC echo, the black region (with a grey background) corresponds to the bins considered as the noise [we use 70 additional regions to calculate the noise in (b), and 80 for (c), each 20 µs apart], and all the other coincidences are orange. The data was acquired in (a) 6.2 minutes, (b) 12 minutes, and (c) 42 minutes.

Fibre connection, which means that the background counts have a larger relative contribution and thus decrease the signal-to-noise of the measurement. In this situation, the idler rate is just over 10 counts/second (see Tab. 6.1) while the dark count rate is approximately 2 counts/second in the same measurement window.

The AFC efficiency at these storage times was 21(1)% and 7.5(6)% for 10 µs and 25 µs, respectively\(^7\), which is consistent within error with the efficiencies reported in Chap. 5. The decrease in \(g^{(2)}_{AFC,i}\) for 25 µs storage compared to 10 µs

\(^7\)Calculated using the measurements for \(d_{\text{idler}} = 11\) km.
Figure 6.4: \( g^{(2)}_{\text{AFC},i} \) versus \( d_{\text{idler}} \) for AFC storage times of 10 \( \mu s \) and 25 \( \mu s \). A 400 ns window within the AFC echo was used to calculate \( g^{(2)}_{\text{AFC},i} \).

Table 6.1: Summary of the values of \( g^{(2)}_{\text{AFC},i} \), the idler count rate \( R^i \), and coincidences per idler count in a 400 ns window for the different values of \( d_{\text{idler}} \) and AFC storage time \( \tau \) investigated.

<table>
<thead>
<tr>
<th></th>
<th>0 km</th>
<th>11 km</th>
<th>50 km</th>
</tr>
</thead>
<tbody>
<tr>
<td>( g^{(2)}_{\text{AFC},i} (\tau=10 \mu s) )</td>
<td>99(7)</td>
<td>96(8)</td>
<td>41(5)</td>
</tr>
<tr>
<td>( g^{(2)}_{\text{AFC},i} (\tau=25 \mu s) )</td>
<td>74(5)</td>
<td>86(9)</td>
<td>38(6)</td>
</tr>
<tr>
<td>( R^i_{\tau=10 \mu s} ) (counts/second)</td>
<td>1126</td>
<td>169</td>
<td>15</td>
</tr>
<tr>
<td>( R^i_{\tau=25 \mu s} ) (counts/second)</td>
<td>775</td>
<td>111</td>
<td>12</td>
</tr>
<tr>
<td>Coincidences/idler( \tau=10 \mu s ) ( \times 10^{-3} )</td>
<td>2.9</td>
<td>3.1</td>
<td>2.6</td>
</tr>
<tr>
<td>Coincidences/idler( \tau=25 \mu s ) ( \times 10^{-3} )</td>
<td>1.3</td>
<td>1.2</td>
<td>1.2</td>
</tr>
<tr>
<td>Loss at 1535 nm (dB)</td>
<td>0</td>
<td>2.33</td>
<td>13.3</td>
</tr>
</tbody>
</table>

can be attributed to its lower efficiency, and the lower \( R^i \) is due to the longer AFC preparation time required, reducing the duty cycle of the measurement.

We then added the interferometers to measure interference fringes for different values of \( d_{\text{idler}} \). For all three distances, we used an AFC storage time of 10 \( \mu s \). For \( d_{\text{idler}} = 0 \) km and 11 km, we measured interference fringes using two different values of \( \Delta \phi_s \) (corresponding to two different AFCs in the analysis crystal), with a \( \pi/2 \) difference of phase between them, and swept the value of \( \Delta \phi_t \) for each. For \( d_{\text{idler}} = 50 \) km, we only measured the maximum and minimum of the interference fringe for each value of \( \phi_s \) due to the lower \( R^i \).
Figure 6.5: Measurements of light-matter entanglement. Interference fringes for (a) $d_{\text{idler}} = 0$ km and (b) $d_{\text{idler}} = 11$ km, where $g^{(2)}_{AFC,i}$ is analysed as a function of $\Delta \phi_i$, both for two different values of $\Delta \phi_s$. For $d_{\text{idler}} = 50$ km, coincidence histograms for (c) a maximum and (d) a minimum of interference are shown, where $\Delta \phi_s$ is the same for both but $\Delta \phi_i$ was shifted. The red region indicates the window used for the $g^{(2)}_{AFC,j}$ analysis (window size: 400 ns). The weighted average visibility for all three values of $d_{\text{idler}}$ are shown in (e), with total measurement integration times of 440, 1254, and 1429 minutes, respectively.

The results are summarised in Fig. 6.5. The interference fringes measured for 0 km and 11 km are shown in Fig. 6.5(a,b), along with the sinusoidal fits used to calculate the visibility. For the 50 km measurement, only the histograms for one value of $\Delta \phi_s$ are shown [Fig. 6.5(c,d)]. The weighted average visibility for each value of $d_{\text{idler}}$ is shown in Fig. 6.5(e), with values of 83(4)%, 81(3)%, and 84(4) for increasing values of $d_{\text{idler}}$, respectively. All three values are above 80%, and exceed the 70.7% limit for violating the CHSH inequality by three standard deviations. The corresponding two-qubit fidelities are 87(3)%, 86(3)%, and 88(3)%, respectively. For $d_{\text{idler}} = 50$ km, this measurement is the first
Figure 6.6: Experimental setup used to measure coincidences between signal photons detected at ICFO and idler photons detected at i2CAT. The filtering components and memory setup details at ICFO have been omitted for clarity. The individual components used are: periodically poled lithium niobate (PPLN), dichroic mirror (DM), band pass filter (BPF), piezo actuator (PZ), single photon detector (SPD), time-to-digital converter (TDC), and electrical-to-optical transducer (ETO).

These visibilities are all consistent in value within error despite the different values of $g^{(2)}_{\text{AFC},i}$. This is because the limitation of visibility based on $g^{(2)}_{\text{AFC},i}$ follows $(g^{(2)}_{\text{AFC},i} - 1)/(g^{(2)}_{\text{AFC},i} + 1)$ [193], which is over 95% even for the longest distance. The more significant limitations to the visibility are those which affect all distances equally, i.e., the coherence time of the pump laser or imperfections in the interferometers (see Sec. 5.5).

Notably, these visibilities are lower than the one reported in Chap. 5, which had a weighted average of 89(2)%. Some of this reduction is due to using a larger window to analyse coincidences (400 ns vs 280 ns). For comparison, in a 280 ns window the visibilities are 87(4)%, 85(4)%, and 87(4)% for 0, 11, and 50 km, respectively. The measurement in Chap. 5 also used the double-AFC interferometer, which should have less noise at the coincidence window (see Sec. 4.5.2).
6.3 Correlations over a distance

For the last measurement, we decoupled the locations of the signal and idler photon detection and measured coincidences between them. As noted in the introduction, spatial separation of the detectors would be required for any realistic application of our combined photon-pair source and quantum memory setup.

The experimental setup for this measurement is given in Fig. 6.6. Most of the setup of Fig. 6.1 remained at ICFO, but we moved the idler detection to i2CAT (indicated in Fig. 6.2). The idler photons were thus sent through one of the deployed fibres across a total distance of 44 km (16 km of direct distance). We used an InGaAs detector (ID230, ID Quantique) for the idler detection as the previously-used superconducting nanowire detector setup could not easily be moved. This detector had a lower detection efficiency of 10% (compared to 80%), and a similar dark count rate of 10 counts/second. Thus $R_i$ was also reduced, to a rate that prohibited the measurement of entanglement. The SPD sent TTL signals to an ETO whenever a photon was detected, and the resulting classical signal was sent back to ICFO optically using the other fibre between the two locations. The optical signal was converted back to a TTL with another ETO and then recorded using the TDC module, which also recorded the signal photon counts. The memory setup remained identical to Fig. 6.1. Since we only measured coincidences, a spectral pit was prepared in the analysis crystal.

While not strictly necessary for this experiment, we moved the chopper wheel used in the idler path to the free-space portion of the setup at i2CAT\textsuperscript{8}. We wished to test the synchronisation of the choppers across a distance, which would be required for a measurement using the idler interferometer, for example. We used a function generator at i2CAT to generate a square wave which was used to trigger and synchronise the rotation of both chopper wheels. This signal was sent to ICFO via another channel of the ETO (and thus the same optical fibre as the photon detection clicks) to be used for the chopper in the signal photon path.

Figure 6.7 shows the resulting histogram from the measurement, where a storage time of 10 $\mu$s was used. The noise follows the same pattern as Fig. 6.4 because we again gate the source pump and TDC detection. The resulting $g^{(2)}_{\text{AFC},i}$ is 23(2). This decrease with respect to the measurements in the previous section is due to the decrease in the idler count rate from the reduced detection efficiency.

\textsuperscript{8}See Chap. 4 for the typical placement and usage of the chopper wheels.
thus increasing the relative contribution to the noise from background counts. Nonetheless, this value of $g^{(2)}_{\text{AFC},i}$ confirms that non-classical correlations between the idler and signal photons are maintained.

6.4 Discussion

The experiments in this chapter demonstrated light-matter entanglement, with light (idler photons) that were distributed over distances up to 50 km, as well as that non-classical correlations were maintained for the photons pairs, even if the detection of the idler photons was decoupled from that of the signal (and thus QM). We could not measure entanglement with the idler detection at i2CAT, but given the success of the entanglement measurement for $d_{\text{idler}} = 50 \text{ km}$, we are confident that this would succeed if we had used the higher efficiency detector.

Even so, given the short storage times that were used (10 and 25 $\mu$s), the signal photon was emitted and detected long before the idler photon was. This does not affect our ability to measure quantum correlations because all of the measurements are done with coincidences. However, for an implementation of a quantum repeater, the storage time of the quantum memory would need to be increased in order to maintain entanglement for the entire time required to detect an idler photon. This time would amount to approximately 5 $\mu$s of storage per
1 km of distance. For distances over 10 km, the use of spin-wave storage and spin echo techniques [45] would be required, as previously discussed in Chap. 5.

Although the decrease in idler rate across longer distances did not degrade the entanglement visibility, it significantly increased the data acquisition time. The idler rate can be increased by pumping the source with more power, but not infinitely so, as $g_{AFC,i}^{(2)}$ (and thus the visibility) decreases with pump power according to $1 + 1/p$, where $p$ is proportional to the power [11]. Using photons in the telecom C-band ($\sim$1550 nm) would also increase the detection rate, as less photons would be lost from attenuation in optical fibre. In order to achieve practical coincidence rates, the signal-side of the setup would need to be improved, particularly regarding the QM. Increasing the storage efficiency with an impedance-matched cavity [178], and increasing the multiplexing capability of the memory, as previously discussed in Chap. 5, are likely to result in the largest gains in coincidence rate. For an experiment in a long-distance quantum repeater scenario, we will not be able to pump the source as frequently as in the experiments presented here. We would need to perform SW storage unconditionally, where the pump is open for a finite time, then we send control pulses timed so that the signal photon is retrieved after we know an idler was detected (i.e., the signal is stored for the entire communication time), and only pump the source on again after the signal has been retrieved. While we will be able to store multiple temporal modes (depending on the AFC storage time and control pulse length, see Eq. 5.8), other forms of multiplexing, such as frequency or spatial, would need to be implemented in order to have a good entanglement distribution rate.
Chapter 7

A fibre-integrated laser-written quantum memory for light-matter entanglement

In this chapter I will present our experiments with a fibre-integrated quantum memory. In this case, the memory is a laser-written waveguide inside a Pr$^{3+}$:Y$_2$SiO$_5$ crystal with optical fibres pigtailed to it. We characterise the performance of the memory in terms of AFC storage time, efficiency, and preservation of single photon statistics. We also demonstrate the utility of this memory for establishing light-matter entanglement by storing a photon from a source of entangled photon-pairs. The entanglement is verified using quantum state tomography.

The main contents of this chapter come from Ref. [130]. I am a co-first author, along with Dr. Giacomo Corrielli and Dr. Dario Lago-Rivera. Dr. Giacomo Corrielli fabricated the waveguide and carried out the fiber pigtailing. Dr. Giacomo Corrielli, Dr. Samuele Grandi, Dr. Margherita Mazzera, Dr. Alessandro Seri and I all contributed to mounting the sample in the cryostat. I optimised and operated the memory setup, and Dr. Dario Lago-Rivera operated the photon-pair source. Dr. Dario Lago-Rivera and Dr. Samuele Grandi designed and operated the interferometers. Dr. Dario Lago-Rivera and I acquired all of the data, and analysed the data except for the tomography measurements, which were analysed by Dr. Samuele Grandi.
7. Fibre-integrated laser-written quantum memory

7.1 Introduction

In the previous two chapters, we demonstrated the utility of Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} as a quantum memory in terms of its ability to preserve entanglement. For use in future quantum networks, it is also important to consider the practicality of the memory. To this end, integration of quantum memories has become a significant area of research. In this case, the integrated memory could be a device that facilitates miniaturisation of the experimental setup [221], or interfacing with other integrated components and optical circuits [221, 222], and could also feature enhanced light-matter interaction [223–225].

There are two approaches for integrating REIC-based memories: either taking an existing waveguiding structure and doping ions in it, or fabricating a waveguide with or within a doped crystal itself. Storage of single photons, or of coherent states at the single photon level has been demonstrated with both of these approaches [83, 87–89, 91–93, 226].

An integrated memory with Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} has already been demonstrated in our group using a laser-written waveguide. My predecessors characterised its performance storing single photons [91], as well as demonstrated frequency multiplexing, which was facilitated by the enhanced light-matter interaction inside of the waveguide, allowing many AFCs to be created simultaneously while minimising the optical power used [83]. However, the only integrated component of the experiments was the crystal itself, so there was no significant difference in the optics used for this memory versus a bulk Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} memory. Additionally, the vibrations of the cryostat meant that the waveguide was periodically uncoupled from the light. If the memory could be integrated with optical fibre, it would both simplify the setup and improve the transmission through the device. The only previously demonstrated fibre-integrated rare-earth ion based memory used erbium ions doped in optical fibre and was limited in performance as the amorphous host (the optical fibre) degraded the coherence properties of the ions [89, 226]. Thus a fibre-integrated system with good coherence properties has yet to be demonstrated.

In the following chapter, I will introduce our memory that features fibre integration, which simplifies the experimental setup and allows for an improved performance of the device.
7.2 Fibre-pigtailed device and mounting

For the experiments presented in this chapter, we use a type I waveguide fabricated in Pr$^{3+}$:Y$_2$SiO$_5$ using femtosecond laser micromachining. This particular waveguide was the same one used in previous experiments from our group where single photon storage [91] and frequency multiplexing [83] were demonstrated. As such, a detailed characterisation of this waveguide and the fabrication procedure can be found in Ref. [91].

Light propagates through the waveguide along the $b$ axis in the area where the material has been modified. In this area, there is a positive refractive index change with respect to the rest of the crystal matrix. The dimensions of the waveguide (in the transverse plane) are $3.1 \times 6.0 \, \mu\text{m}$ ($e^{-2}$ full widths). The waveguide only supports single mode propagation for polarisation along the $D_2$ axis.

To fibre-pigtail the device, fibre patch cables (630HP) were inserted in 3 mm long glass ferules with an external diameter of 1 mm and butt-coupled at the input and output of the waveguide. They were glued with a ultraviolet-curing transparent resin (DELO Photobond GB345). The $e^{-2}$ diameter of the fibre mode intensity profile is $2.9 \, \mu\text{m}$ at 606 nm. Based on the mode profiles of the waveguide and fibre, the theoretical maximum transmission efficiency of the device is 45%. The actual transmission of the device (outside of the cryostat) was 25%, with additional losses coming from waveguide propagation loss, imperfect waveguide facets and splices in the fibres.

While bulk Pr$^{3+}$:Y$_2$SiO$_5$ crystals can be efficiently cooled in our cryostat, it is more difficult with a fibre-pigtailed sample. The fibres will eventually have to
leave the cryostat to the room temperature laboratory environment, so the fibres must be well-thermalised and distant from the crystal by the time they reach the fibre feedthroughs. The sample holder should also feature vibration damping in order to maximise the AFC efficiency of the system.

We tried several iterations of sample holders and mounting options, with the final result visible in Fig. 7.2, designed by Dr. Alessandro Seri. The fiber-pigtailed waveguide sits on the upper block of the mount, attached with GE Varnish. The fibres were then wrapped a few turns around the circumference of the block and secured with aluminium tape in order to thermalise the parts closest to the crystal, and to prevent the fibres from moving independently from the crystal which could induce strain on it. We also found that using short ferrules was crucial for reducing strain on the crystal.

The upper block rests on four springs that are supported by the lower block. Both blocks have a large gap in their center so the holder can slide onto the cold finger from below. The lower block is then attached to the cold finger. Two copper foils are used to thermalise the upper block to the cold finger. With respect to Fig. 7.2(a), the two foils are bent into the gap inside the block, and then are attached directly to the cold finger.

With the sample holder in place, we also wrapped the fibres for a few loops around the radiation shield, which is typically at a temperature of tens of kelvin, and secured them again with aluminium tape before sending them outside of the
cryostat via a fibre feed through. This process was done in order to reduce any possible temperature gradient near the crystal.

After mounting the device, cooling it to 3 K, and splicing the input to a 90:10 fibre beam splitter, the transmission reduced from 25% to 20%. The initial transmission was recovered when the device was returned to room temperature and unmounted from the cryostat, and maintained this transmission through more than one cooling cycle.

We characterised the performance of the sample holder in a similar way to the holder described in Chap. 5. We measured the efficiency of a 10 $\mu$s AFC that was prepared at different times during the cryostat vibration period; see Fig. 7.3. There was a clear variation in efficiency depending on the time at which the AFC was prepared, and the efficiency was comparatively stable when the cryostat was turned off\(^1\), which indicates that the vibrations had not been completely damped. Once the AFC was prepared however, the efficiency did not drop as significantly during the rest of the vibration period. Based on a comparison with a holder without vibration damping (Sec. 7.4), we believe that the vibration isolation, although imperfect, improved the overall performance of this integrated memory.

To verify that crystal was well-thermalised, we measured the optical coherence time using photon echoes. We measured a value of $43(2)$ $\mu$s, compared to

\(^{1}\)The efficiency varied with a standard deviation of $\pm0.5\%$ from the mean efficiency, which could be due to residual oscillations of the sample holder after the cryostat was turned off, or an effect of the AFC sequence not being optimised at the time of performing the measurement.
57(10) µs measured in Ref. [91] with the same waveguide. This lower value indicates that the crystal may not have been perfectly thermalised. The difference in measured values may also be due to the difficulty in measuring longer coherence times due to instantaneous spectral diffusion which depends on the excitation pulse intensity [143]. It is particularly challenging to reduce this effect in these waveguides, as they have very small modes, so the pulse intensity must be very small and the resulting echo signal is also weak. Since we could obtain better AFCs than without fibre-pigtailing [169] (see Sec. 7.4), we assumed the crystal was sufficiently thermalised.

7.3 Experimental setup

To characterise the performance of our integrated memory, we used the signal photons from the photon-pair source as the input. The setup is shown in Fig. 7.4. The photon-pair source, including the locking setup, is operated as described in Chap. 4, except different choppers were used such that the measurement duty cycle was 70%. The pump power used for all of the measurements presented here was 4.0 mW, unless stated otherwise. The signal photons are sent to the 90% input of a 90:10 fibre beam splitter, which is spliced directly to the fibre-pigtailed waveguide. The light used for optical pumping (AFC preparation) is sent to the 10% input port of the beam splitter. The total transmission through the 90% BS
input path was 20% when cold. All of the optical fibres used for 606 nm light were 630HP.

The fibre-based idler and signal interferometers were operated as described in Chap. 4. Unlike the idler interferometer, we connect both outputs of the signal interferometer to SPDs. For measurements without the interferometers, the idler photons go directly to the SPD, while the signal photons are sent to free space before being fibre coupled again, as a shutter is required to prevent the classical preparation light from reaching the SPD. To measure the heralded autocorrelation, the setup was in the same configuration as without the interferometers, and a 50:50 fibre beam splitter was used after the free-space section. Both outputs were connected to an SPD².

7.4 Single photon measurements

We first characterised the performance of our system without the interferometers. We measured coincidences between the idler photons and signal photons stored in the AFC from 2 µs up to 28 µs³. The resulting efficiencies are shown in Fig. 7.5(a). The maximum internal efficiency was 18% at 2 µs, corresponding to a

²A COUNT-10C-FC was used for all of these measurements, and when an additional SPD was required, a COUNT-20C-FC was used for the tomography measurements while a COUNT-50C-FC was used for the autocorrelation measurements.
³The AFCs prepared here are not single-class features.
fibre-to-fibre efficiency of 3.6%. The efficiency decreases with increasing storage time. We can extract an effective coherence time $T_{2}^{\text{eff}}$ for the AFC storage using Eq. 5.1, to obtain $T_{2}^{\text{eff}} = 27(3) \mu s$. In the same waveguide without fibre coupling, this value was 16(1) $\mu$s [169]. We attributed this improvement to the vibration isolation, which would not have been possible to implement in the same way without the fibre-pigtailing. However, the effective coherence time is still lower than the coherence time measured using photon echoes, 43(2) $\mu$s, indicating that there is still room for improvement, for example by implementing better vibration isolation.

We also calculated $g_{i}^{(2)}$ from the same coincidence measurements, with the results shown in Fig. 7.5(b). Here $g_{AFC,i}^{(2)}$ also decreases with increasing storage times, because a reduced efficiency results in fewer signal counts which will eventually be limited by background counts. While the $g_{AFC,i}^{(2)}$ is not limited by background counts in the bulk memory, the effect of the background is greater for the fibre-integrated memory since there is more loss of signal photons before reaching the crystal itself due to imperfect transmission from the glued fibre. We measured $g_{AFC,i}^{(2)} > 2$ for all of the storage times measured, thus showing that we maintained nonclassical correlations.

We also measured the heralded autocorrelation of the signal photons ($g_{i,s,s}^{(2)}$) using the technique introduced in Ref. [196]. Since we are using an asynchronous and heralded source of photons, we looked at the number of heralds that were recorded in between two signal detections, each from different detectors, rather than the time between two signal detections as is typical for antibunching measurements. We measured under two different conditions: the signal photons passing through a transparency window in the memory and after storage for 3 $\mu$s in an AFC. Both measurements are shown in Fig. 7.6. The resulting values of $g_{i,s,s}^{(2)}$ were 0.10(3) and 0.05(2), respectively, showing that the memory preserves the single photon statistics as expected. The reduction of $g_{i,s,s}^{(2)}$ is due to the temporal filtering effect of AFC storage [44, 217].

Finally, we investigated the effect of the pump power on the correlations between the signal photons stored in an AFC and the idler photons. Figure 7.7 shows that $g_{AFC,i}^{(2)}$ decreased with increased pump power. This reduction is due to the increase in multi-pair generation events. While in this work we use a pump power of 4 mW, we could still increase $g_{AFC,i}^{(2)}$ by using a lower pump power, at the expense of reducing the count rate of the experiments.
7.5 Time-bin qubit tomography

We performed a full tomography of the entangled state of the telecom idler photon and the stored signal photon. To do so, as in Chap. 5 and Chap. 6, we used the Franson scheme for the analysis [207].

Unlike the previous chapters, we used a fibre interferometer as the signal analyser, which is a convenient choice as the output of the memory could be connected directly to the interferometer which also minimises the optics needed for the experiment. We can also simultaneously measure both outputs of the signal interferometer to decrease the measuring time. Our method of analysis is also different: we perform qubit tomography instead of measuring interference fringes. The idea here is that by selecting particular phases with the interferometers, we can project the state of our qubit onto different bases of the idler and signal photons. The resulting coincidence measurements can then be used to reconstruct the original two-qubit state of the entangled photon-pair [206].

Figure 7.6: Heralded autocorrelation measurements, corresponding to the signal photons (a) propagating through a transparency window in the quantum memory or (b) stored in a 3 μs AFC. The solid line corresponds to the average number of coincidences between two signal coincidences across the entire measurement (except when the number of heralds is zero), while the dashed lines indicate one standard deviation of variation from this average. The bin size was 400 ns.
7. Fibre-integrated laser-written quantum memory

Figure 7.7: $g_{\text{AFC}, i}^{(2)}$ as function of the pump power of the SPDC source, measured using a 2 $\mu$s AFC. The error bars correspond to one standard deviation.

Figure 7.8: Bloch sphere representation of the time bases used in the tomography measurements.

We could have measured interference fringes instead as in the previous chapters, but we chose to characterise the fidelity with an alternative technique. Performing this type of tomography requires more measurements than the interference fringes, particularly if an AFC is used as the analyser instead of a fibre interferometer as we can only measure one signal interferometer phase at a time. Thus for the experiments with lower rates (the SW entanglement measurement of Chap. 5 and the long-distance measurements of Chap. 6), it was more time-efficient to measure interference fringes. In this experiment, the count rates were all sufficiently high for the measuring time to not be a limitation.

Figure 7.8 depicts the bases we considered for the measurements. Here, $|\pm\rangle = (|e\rangle \pm |l\rangle)/\sqrt{2}$ and $|R/L\rangle = (|e\rangle \pm i|l\rangle)/\sqrt{2}$. We would first select a basis to project the idler to: $|+\rangle_i$, $|L\rangle_i$, $|e\rangle_i$, or $|l\rangle_i$. Here the subscript $i$ and later $s$ indicates that the state corresponds to the idler or signal photon, respectively. Projections onto $|+\rangle_i$ and $|L\rangle_i$ required a particular phase setting for the idler
Figure 7.9: Example of a histogram of signal-idler coincidences where the signal photons are stored in an AFC for 10 μs. The dark brown region corresponds to the window of counts for a projection on $|+/-\rangle_s$ or $|R/L\rangle_s$, while the red regions on either side are the windows of counts for projections on $|l\rangle_s$ (left) and $|e\rangle_s$ (right). The black region (shaded) is the region considered for dark counts, and the orange area corresponds to all other coincidences from the measurement not considered in the calculation.

interferometer, while projections onto $|e\rangle_i$ and $|l\rangle_i$ were done by disconnecting one arm of the idler interferometer, such that the photons would only pass through the short or long arms, respectively. For each basis selected for the idler, we would project the signal photon onto $|+/-\rangle_s$, $|R/L\rangle_s$, $|e/l\rangle_s$. As for the idler, we carefully selected the phase required to project onto $|+/-\rangle_s$ and $|R/L\rangle_s$. Since we detected both outputs of the signal interferometer, we could measure each basis with a $\pi$ phase shift simultaneously. We also utilised the coincidences in the side peaks from these projection measurements, as they corresponded to projections onto the $|e/l\rangle_s$ basis (see Fig. 7.9).

For measurements involving a particular choice of phase for the interferometers, we calibrated the phase settings in the following way. We locked the phase of the idler interferometer to a value close to the bottom of its interference fringe to minimise the effect of amplitude oscillations. Then we prepared a transparency window in the memory and measured idler-signal coincidences for a range of signal interferometer phases. From this measurement, we were able to extract the voltage settings for locking the signal interferometer that would yield measurements in the $|+/-\rangle_s$ or $|R/L\rangle_s$ bases. With this setup, this required locking the signal interferometer to a minimum/maximum, depending on the interferometer output considered, or to a point shifted by $\pi/2$ corresponding to the other set of bases, respectively.
To obtain the full set of measurements required for the tomography, we started with an initial idler projection, determined the signal interferometer phase required to measure with $|+/−\rangle_s$, locked to it, and measured signal-idler coincidences. We shifted the signal interferometer phase to project onto $|R/L\rangle_s$ and once again measured signal-idler coincidences. We then changed the locking point of the idler interferometer to shift its phase by $\pi/2$, projecting the idler photon on a different basis, and repeated the calibration phase scan. With this, we could determine the exact phase shift imparted by the new locking point of the idler interferometer. We acquired signal-idler coincidences again, using the same locking points for the signal interferometer used in the previous projection. We concluded by projecting onto $|e\rangle_i$ and $|l\rangle_i$ and measuring using the same signal interferometer locking settings used previously.

We performed this measurement procedure for three different cases: with a transparency window in the memory (no storage), 3 $\mu$s of storage, and 10 $\mu$s of storage. The efficiency of the storage was 13.7(4)\% and 4.2(2)\% for the 3 and 10 $\mu$s AFCs, respectively.

With measurements we acquired, we could perform a complete tomography of the entangled state of the idler and signal [206], as well as compute the 1-qubit tomographies for the states prepared after the detection of an idler. For each experimental setting, we normalised the raw coincidences to the number of idler counts (normalising for the measuring time), and calculated $m_j = \frac{N_j}{N_j + N'_j}$. Here $N_j$ and $N'_j$ are the normalised coincidences measured for the same $j$-th experimental settings and for two orthogonal basis elements; for projections onto $|+/−\rangle_s$, $|R/L\rangle_s$ these are coincidences at opposite outputs of the signal interferometer. Values of coincidences for projections on $|e\rangle_s$ and $|l\rangle_s$ are summed over all those collected for every signal interferometer setting, and from both outputs of the signal interferometer.

To calculate the fidelities, we ran a maximum-likelihood tomography using the Quantum Tomography Python package\textsuperscript{4}. This tomography produces the required density matrices from which the fidelities can be calculated using [227]:

\begin{equation}
    F(\rho, \sigma) = \left( \text{tr} \sqrt{\sqrt{\rho} \sigma \sqrt{\rho}} \right)^2,
\end{equation}

\textsuperscript{4}https://quantumtomo.web.illinois.edu/
Table 7.1: Fidelities with respect to the ideal state for the input state and for the two storage times. Each column represents a different analysis setting, which is applied additionally to the previous ones (e.g. the last column has all the previous corrections). Raw: raw data. $\tilde{M}(\phi)$: data analysed with exact projective measurements. Analyser: data corrected for the imperfect analyser. DC: data corrected for dark counts. 280 ns: the coincidences are collected in a 280 ns window, instead of 400 ns. 1-qubit: 1-qubit fidelities, with all of the same corrections applied, in a 400 ns window.

<table>
<thead>
<tr>
<th></th>
<th>Raw</th>
<th>$\tilde{M}(\phi)$</th>
<th>Analyser</th>
<th>DC</th>
<th>280 ns</th>
<th>1-qubit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Input</td>
<td>75.4(2.1)</td>
<td>75.5(2.1)</td>
<td>80.6(2.3)</td>
<td>80.8(2.4)</td>
<td>83.7(2.7)</td>
<td>91.1(7)</td>
</tr>
<tr>
<td>3 µs</td>
<td>79.2(1.8)</td>
<td>79.6(1.8)</td>
<td>84.8(1.9)</td>
<td>85.9(2.0)</td>
<td>87.3(2.0)</td>
<td>93.8(7)</td>
</tr>
<tr>
<td>10 µs</td>
<td>76.9(3.1)</td>
<td>77.8(3.1)</td>
<td>82.2(3.3)</td>
<td>86.3(3.5)</td>
<td>87.8(3.7)</td>
<td>94(2)</td>
</tr>
</tbody>
</table>

where $\rho$ and $\sigma$ are density matrices. In this case, one is the ideal two qubit state, and the other is the reconstructed density matrix determined from the measurements. The results of the tomography are reported in Tab. 7.1, where the errors are calculated by simulating 1000 datasets.

For all three cases measured (input and the two storage times), the 2-qubit fidelities were below 80%, which are labelled as the “raw” fidelities in Tab. 7.1. The fidelities were affected by certain technical aspects of our measurement and setup, which we can correct for. These were:

- Incorrect measurement settings: the phases of the interferometer must be set precisely in order to project onto the exact bases that we with to measure with. We only have a finite precision with which we can lock the interferometers, so by using the interference fringes measured for calibration, we could determine the exact projective measurement we made.

- Analyser imperfection: since a long fibre spool is used to make the long arm of the interferometers, it will have less transmission than the short arm. Only the signal interferometer is significantly affected by this difference since visible wavelengths experience more loss in optical fibre. As a result, there will be an imbalance of counts for projective measurements onto $|e\rangle$ or $|l\rangle$. We corrected the counts measured in the right-most coincidence peak of the measurements (see the histogram in Fig. 7.9) by renormalising them to the ratio between the transmission through the late and early arm (49%) of the signal interferometer. The difference in transmission also reduces the visibility to 95%. For coincidences in the central peak (see Fig. 7.9), the normalised coincidences can also be written as $m_j = \frac{1+V_j}{2}$, where $V_j = (N_j - N_j')/(N_j + N_j')$ is the visibility of the $j$-th measurement.
Table 7.2: Density matrices for the three cases depicted in Fig. 7.10, with full corrections applied in a 400 ns window.

<table>
<thead>
<tr>
<th></th>
<th>(\langle ee\rangle)</th>
<th>(\langle el\rangle)</th>
<th>(\langle le\rangle)</th>
<th>(\langle ll\rangle)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\langle ee\rangle)</td>
<td>0.447 - 0.009 i 10.035</td>
<td>0.005 + 0.002 i 0.002</td>
<td>0.351 + 0.056 i 0.002</td>
<td></td>
</tr>
<tr>
<td>(\langle el\rangle)</td>
<td>-0.009 + 10.035 i 0.035</td>
<td>0.03 + 10.001 i 0.002</td>
<td>0.004 - 10.002 i 0.002</td>
<td></td>
</tr>
<tr>
<td>(\langle le\rangle)</td>
<td>0.005 - 10.002 i 0.035</td>
<td>0.03 - 10.001 i 0.003</td>
<td>0.048 - 10.037 i 0.002</td>
<td></td>
</tr>
<tr>
<td>(\langle ll\rangle)</td>
<td>0.351 + 10.056 i 0.002</td>
<td>0.048 + 10.002 i 0.037</td>
<td>0.468</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>(\langle ee\rangle)</th>
<th>(\langle el\rangle)</th>
<th>(\langle le\rangle)</th>
<th>(\langle ll\rangle)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\langle ee\rangle)</td>
<td>0.464 - 0.004 i 10.023</td>
<td>-0.005 + 10.016 i 0.002</td>
<td>0.382 + 10.031 i 0.002</td>
<td></td>
</tr>
<tr>
<td>(\langle el\rangle)</td>
<td>-0.004 + 10.023 i 0.035</td>
<td>0.03 - 10.003 i 0.003</td>
<td>0.009 - 10.005 i 0.002</td>
<td></td>
</tr>
<tr>
<td>(\langle le\rangle)</td>
<td>-0.005 - 10.016 i 0.003</td>
<td>-0.004 + 10.003 i 0.017</td>
<td>0.045 - 10.005 i 0.005</td>
<td></td>
</tr>
<tr>
<td>(\langle ll\rangle)</td>
<td>0.382 - 10.031 i 0.009</td>
<td>0.009 + 10.005 i 0.045</td>
<td>0.489</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>(\langle ee\rangle)</th>
<th>(\langle el\rangle)</th>
<th>(\langle le\rangle)</th>
<th>(\langle ll\rangle)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\langle ee\rangle)</td>
<td>0.466 0.028 - 10.008 i 0.014 - 10.003 i 0.391 - 10.088</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(\langle el\rangle)</td>
<td>0.028 + 10.008 i 0.032</td>
<td>0.008 - 10.016 i 0.019 - 10.01</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(\langle le\rangle)</td>
<td>-0.014 + 10.003 i 0.008 + 10.016 i 0.024</td>
<td>0.033 + 10.003 i 0.003</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(\langle ll\rangle)</td>
<td>0.391 + 10.088 i 0.019 + 10.001 i 0.033 - 10.003 i 0.477</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Then for projections onto the equatorial states, the coincidences can be recalculated as \(m'_j = \frac{1+V'_j}{2}\), where \(V'_j = V_j/0.95\).

- Dark counts: the overall transmission of signal photons through the setup is low when the fibre interferometer is included, so dark counts limited the quality of the correlations. To estimate the number of dark counts in the temporal window where we measure coincidences, we calculated the dark counts in a region which should have no other background counts. For the measurement through a transparency window, this region is where the pump is off, and in the case of AFC storage, it is also in a temporal region after the echo. These darks counts are normalised to the same temporal window size as the coincidences, then subtracted from the coincidence counts used in the tomography. This correction is particularly beneficial for measurements with a lower count rate, i.e., for 10 \(\mu\) s AFC storage.

By including these corrections, we see a significant improvement in the fidelities to 80% for the input measurement, and 86% for both of the AFC storage measurements. The fidelities measured for storage are similar, which is to be expected when not limited by dark counts or noise. The fidelity increases further if the window used for analysing coincidences is reduced from 400 ns (containing 92% of the photon wavefunction) to 280 ns (containing 78% of the photon wavefunction), at the expense of a reduced count rate. The reconstructed density
7.6 Discussion

Based on the measurements from this chapter, we have demonstrated storage of time-bin entanglement in a fibre-integrated quantum memory, which preserves entanglement for a predetermined storage time of up to 10 μs. This storage time is three orders of magnitude longer than previous realisations of fibre-integrated

![Figure 7.10: Reconstructed density matrixes from the entangled state tomography. (a) Density matrix of the input state, measured with the signal photon passing through a transparency window in the memory. The data were acquired over 30 minutes. (b) and (c) Density matrixes for the retrieved AFC echo for storage times of 3 and 10 μs, respectively. Data acquisition was completed in 120 and 130 minutes, respectively. Only the real part of the matrices are included.](image-url)
quantum memories\textsuperscript{5}, and an improvement of at least 20 times in efficiency with comparable values of fidelities [87, 226].

Further improvements are still required for practical use for quantum communication. The performance of AFC storage in this memory lags its bulk counterpart (see Chap. 5). It is unclear if the AFC burning procedure is limited by the coherence properties of the ions inside the waveguide. A shorter coherence time would result in wider spectral holes and thus a worse efficiency at longer storage times. We could not measure a coherence time as long as in the bulk, which could be due to the measuring procedure, or intrinsic properties of the ions themselves.

Improving the vibration isolation may also improve the efficiency. We do note that in other experiments performed with this type of holder and a similar fibre-pigtailed device, the vibration isolation performed more poorly. The poor performance of the vibration isolation was likely due to strain induced by the clamping. As there are many points where strain can be induced on the fibres, such as any place where the fibres have been taped, or clamped at the fibre feedthrough, the mounting procedure is also critical for vibration isolation. The arrangement of copper foils for thermalisation is also quite different compared to the bulk crystal holder, and thus may not have an ideal mechanical resonance for damping the vibrations of this cryostat.

The next step for this device would be to implement on-demand readout of the memory through spin-wave storage, as was done in Chap. 5. This has only been demonstrated classically, using a type I [169] and type II [228] waveguide in Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} and in a type IV waveguide in Eu\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} [229]. The main challenge with performing spin-wave storage comes from filtering noise from the control pulses. Unlike the experiments in Chap. 5, the signal and control modes would have to coincide, removing one of the aspects of filtering we implemented in that experiment. To combat this, thanks to the integrated nature of the memory, we could make use of narrow laser-written filters [230] or directional couplers [231]. Alternatively, electrodes could be used to implement noise-free on-demand storage using the Stark effect [94, 95, 184], however the storage time will be much more limited due to only utilising the optical transition of our ions. We measured a spin inhomogenous broadening of around 80 kHz in this

\textsuperscript{5}This was true at the time these results were published in Ref. [130], but with recent results from Ref. [84], our longest storage time is only two orders of magnitude longer.
waveguide which is larger than the bulk and would thus complicate SW storage without RF pulses. Further investigated is needed to see if this is the case for other waveguide samples and if it depends on the fabrication technique.

Additionally, any loss of photons from the source to the memory (i.e., a non-unit efficiency coupling of light between the fiber and waveguide, splices) would reduce the heralding efficiency and thus \( g^{(2)}_{SW,i} \) of a retrieved spin-wave echo. In this regard, our collaborators at Politecnico di Milano are developing new waveguides which have more Gaussian-shaped modes that are very promising for improving the transmission through the device, and may additionally offer more improved coherence properties.

Despite the challenges presented by this system, thanks to improvements in integrated memories we presented here, there are many possibilities and advantages that we can make use of in the future. The prospects for multiplexing are particularly noteworthy. We can spatially multiplex the device by fabricating an array of waveguides inside the crystal. Combined with the temporal and frequency multimodality already demonstrated in the system [44, 83], this would allow for a massively multiplexed memory with a small physical footprint. In the same theme, we could make use of fibre-arrays to connect to such a device, and we could even place superconducting single photon detectors in the same cryostat. Type I waveguides also have lower bending losses compared to other types [91, 94, 228], which would allow for more complicated waveguide geometries to be written, and for the memory itself to couple to other laser-written type I components.
Chapter 8

Conclusion and outlook

8.1 Summary

The work I have presented in this thesis demonstrated the use of Pr\(^{3+}:Y_2SiO_5\) as a quantum memory, in experiments demonstrating light-matter entanglement between photons generated through spontaneous parametric down conversion, where the signal photon is stored in the memory and the idler photon is in the telecom band at 1436 nm. In Chap. 5, the memory is operated as both a delay-line with optical AFC storage, and in a on-demand fashion using SW storage. We show an improvement in the memory performance compared to previous work from our group [44], achieving a \(g^{(2)}_{SW,i}\) of 9.8(6), thanks to an increase in the single photon storage efficiency to 6.2(3)\% and a reduction of the noise floor to 8.3(4)\times10^{-4} photons per trial. We demonstrated entanglement using the Franson scheme [207], measuring an average visibility of 89(2)\% using AFC storage alone for 10 \(\mu\)s, which is enough to violate the CHSH inequality by a few standard deviations. With SW storage, we stored the photon for an additional 7 \(\mu\)s in the memory and measured an average visibility of 70(3)\%. This value much greater than the 33\% limit for separable states, and the result constitutes the first demonstration of entanglement between a on-demand solid-state multimode quantum memory and a telecom photon. Using the on-demand nature of our memory, we also measured the visibility for storage times up to 37.7 \(\mu\)s in the spin levels and thus a total of 47.7 \(\mu\)s of storage in the Pr\(^{3+}:Y_2SiO_5\) crystal, remaining above the 33\% limit for all of the storage times investigated.

In an additional measurement in Chapter 5, we also investigated the temporal multiplexing capability of the quantum memory. Using weak coherent states with
0.90(5) photons per pulse as an input, we stored 20 and 30 temporal modes in a spin-wave, using a 25 µs AFC. The average SNR across the modes was 17.4(4) [with $\eta_{SW} = 1.88(3)$%] and 6.7(1) [with $\eta_{SW} = 0.63(1)$%], for SW storage times of 14.1 µs and 20.7 µs, and 20 and 30 modes, respectively.

Chapter 6 presented an experiment moving towards a more practical application of the bulk quantum memory. Here we measured entanglement similarly to Chap. 5, now with the telecom photon distributed through optical fibre in the lab or through deployed optical fibre in the metropolitan area of Barcelona. Using optical AFC storage for 10 µs, we measured visibilities of over 80% for all of the fibre distances investigated, the longest being a loop of 50 km of deployed fibre. These results show that the coherence of the telecom photon is maintained during transmission through the fibre, and the measurement at 50 km is the first demonstration of distribution of light-matter entanglement with telecom photons and solid state quantum memories using deployed optical fibre. In another experiment, we moved the idler photon detection setup to a different building, separated from the rest of the experimental setup by 44 km of deployed fibre. We measured non-classical correlations between the idler photons and the signal photons stored for 10 µs in the AFC, resulting in a $g^{(2)}_{\text{AFC}}$ of 23(2).

The final experiments used a fibre-integrated Pr$^{3+}$:Y$_2$SiO$_5$ memory as an interface for light-matter entanglement. We demonstrated an improved performance using the fibre-integrated device compared to the same waveguide fabricated in the Pr$^{3+}$:Y$_2$SiO$_5$ crystal addressed with free-space optics [169], reaching higher efficiencies and longer storage times of up to 28 µs. Unlike the other experiments reported in this thesis, we perform a qubit tomography to analyse the time-bin entangled state of the idler photon and stored signal photon. Using storage times of 3 µs and 10 µs, we measured two-qubit fidelities of 86(2)% and 86(4)%, respectively, well above the classical limit.

### 8.2 Outlook

The experiments summarised here follow a common theme of using Pr$^{3+}$:Y$_2$SiO$_5$ as an absorptive quantum memory to store photons generated from SPDC, and demonstrating that the entanglement of the photon pairs is maintained after storage in the memory. This system constitutes the basic requirements for a quantum repeater node. However, there are still several requirements for the
quantum memory that were introduced in Chap. 1 that need improvement before this system can be used for demonstrations of aspects of a quantum repeater over long distances.

One of the critical requirements currently lacking in the memory is a sufficiently long storage time. In Chap. 5, the maximum storage time we could achieve was close to 50 $\mu$s, which is long enough for a photon to travel 10 km through optical fibre. This is not enough for the lengths of fibres used in the experiments in Chap. 6, for example. Using spin echo techniques will allow us to reach storage times of 100s of microseconds, but this introduces a major challenge of maintaining a low noise floor despite the application of RF pulses. Unlike optical AFC storage which does not introduce noise, the imperfections of any transfer pulses (optical or RF) as well as imperfect class cleaning will introduce noise at the temporal window of the retrieved echo, thus lowering the fidelity of any measurement. It is likely that some combination of better noise filtering, an improved AFC/SW efficiency and higher heralding efficiency of the source will be needed in order to achieve a high fidelity with SW storage. Longer storage times can be achieved using weak magnetic fields [183], and even longer with magnetic field insensitive transitions [73], where storage of classical light for close to one minute was demonstrated. However, as discussed in Chap. 5, the latter configuration reduces the bandwidth of the memory and complicates the generation of memory-compatible single photons.

The storage efficiency and multiplexing capability, which both affect the entanglement generation rate, also require improvement. Along these lines, various ongoing research efforts in our group are very promising, but have yet to be implemented all together in one quantum memory. Regarding the storage efficiency, recent results from our group have demonstrated an AFC storage efficiency of 62% using an impedance-matched cavity [78]. Preliminary results show an improvement in the SW efficiency, and work towards using the enhanced SW memory at the single photon level is ongoing.

Multiplexing of frequency modes was previously demonstrated in an integrated Pr$^{3+}$:Y$_2$SiO$_5$ memory [83], while the storage of many temporal modes was demonstrated in Chap. 5 of this thesis. Another member of the group is currently investigating spatial multiplexing in Pr$^{3+}$:Y$_2$SiO$_5$ by addressing different spatial regions in one crystal. To achieved a beneficial enhancement of the entanglement rates, all of these degrees of freedom need to be combined. While challenging, a
combination of several different types of multiplexing has been demonstrated in other experiments [86]. As we wish to use a much greater number of modes, we will need to consider how to best utilise them, and how to demultiplex the modes, such as in Ref. [38].

While on-demand retrieval using Pr$^{3+}$:Y$_2$SiO$_5$ has been demonstrated here and in previous work [44] using SW storage with the bulk memory, it has yet to be achieved at the single photon level in an integrated memory. Previous attempts with the type I waveguide used for the fibre-integrated memory could not reach a sufficiently low $\mu_1$ for single photon storage [169]. Alternatives schemes with on-demand retrieval have been demonstrated using the Stark effect [94, 95], but as a result, the storage time is limited by the optical coherence time. For Pr$^{3+}$:Y$_2$SiO$_5$ with an optical coherence time of $\approx 120$ $\mu$s, it would not be possible to store photons long enough and with a high enough efficiency for measurements involving distances of tens of kilometres. Further investigation and engineering of the waveguides are required in order achieve SW storage. Otherwise, there is the option of using extensive multiplexing with only optical AFC storage [36].

There are still many convenient aspects of the fibre-integrated memory that we can make use of. We can envision reducing the footprint of the experiment by replacing the free space components of the AOM setup described in Chap. 4 with fibre-coupled components. There are a wide range of integrated components that can be fabricated using femtosecond laser micromachining [90], including photon sources, phase shifters and interferometers. The type I waveguide architecture used here is compatible with such devices, which could lead to new experiments or new paths to miniaturisation of the setup.

In the context of our node architecture, besides all of the aforementioned improvements to the memory, there are various aspects of the photon-pair source that can be improved. Some of these would simply improve the coincidence rate, such as by generating idler photons in the telecom C-band instead of at 1436 nm, which as noted in Chap. 6 is particularly beneficial when transmitting photons over long distances. Increasing the brightness is also useful in situations where we are limited by pump power. Increasing the heralding efficiency would not only increase the coincidence rate, but also improve $g_{s,i}^{(2)}$ in measurements limited by noise in the signal detection, particularly for SW storage in Chap. 5 and to a lesser extent the qubit tomography in Chap. 7. A new generation of photon-pair
sources is currently in development in the group, with the aim of realising these improvements.

Even with the current performance of our system, we have been able to demonstrate other elementary experiments related to quantum communication. In Ref. [125], we demonstrated telecom-heralded matter-matter entanglement with two Pr$^{3+}$:Y$_2$SiO$_5$ memories. Additionally, we used the memory for a demonstration of quantum teleportation from a telecom photon to the memory [131]. In both cases the memories were used with optical AFC storage only. It should already be possible to perform matter-matter entanglement using SW storage at short distances, which is indeed one of the next objectives of our group. Subsequent experiments would require more memories, such as demonstrating functional quantum repeater links or entanglement swapping between repeater links [11]. Extending any of these experiments to longer distances would require the previously detailed improvements.

Although there is a breadth of research investigating systems or protocols to be used for quantum memories, a much smaller quantity of systems have reached the performance required for single photon operation, let alone light-matter entanglement. The experiments presented here, demonstrating entanglement between a telecom photon and a quantum memory, already belong among the state-of-the-art for such systems. The road to improvement described here will be challenging, but holds great promise for using our system as a node for a quantum repeater.
Appendix A

Preparation of spectral structures

A.1 Preparing a single-class absorption feature

In order to perform SW storage, we need to create an AFC where the ground state is only populated by a single frequency class, and there must also be another ground state level which is unpopulated. We can obtain this configuration in Pr$^{3+}$:Y$_2$SiO$_5$ by addressing the ions in a specific sequence, which will be introduced in this section. We also follow this type of so-called class cleaning procedure before probing a transition with photon echoes or optical nutation.

Figure A.1 shows the transitions of the nine frequency classes of Pr$^{3+}$:Y$_2$SiO$_5$, arranged along a frequency axis with their corresponding shifts in frequency with respect to class 1. The frequencies of the first class are given such that the lowest frequency transition, $1/2_g \rightarrow 1/2_e$, has a detuning of 0 MHz. The dashed lines on Fig. A.1 indicate the frequency range of interest we consider when preparing the single-class absorption feature. We prepare the AFC on $1/2_g \rightarrow 3/2_e$ transition at a detuning of 4.6 MHz and apply the control pulses for SW storage using the $3/2_g \rightarrow 3/2_e$ transition at a detuning of 14.8 MHz. Thus, we need a single-class absorption feature at the $1/2_g \rightarrow 3/2_e$ transition frequency, and for the $3/2_g \rightarrow 3/2_e$ transition of the same class to be empty of population, as well as any other coincident transitions from other classes.

$^{1}$The ± prefixes have been dropped for clarity.
The procedure for preparing this feature is shown in Fig. A.2. It follows three steps, which are: spectral pit preparation, burning-back, and cleaning. Before applying any laser pulses, the absorption line in the small region we are looking at (≈ 20 MHz) is uniform, and contains all of the frequency classes. In the first step, we create a spectral pit where we remove all of the population in the 20 MHz region. This spectral feature is also called a transparency window, as any light incident in this window should pass through the crystal without being absorbed.

Next, we send a narrow laser pulse resonant to the $5/2_g \rightarrow 5/2_e$ transition of class 1, at a detuning of 36.9 MHz and a frequency chirp of ≈ 4 MHz. This pulse

Figure A.1: Transitions of the nine frequency classes of Pr$^{3+}$:Y$_2$SiO$_5$, arranged by the frequency shift (in MHz) relative to class 1. The length of each bar indicates the relative oscillator strength for each transition, with values taken from Ref. [149]. The 20 MHz range indicated on the chart indicates the frequency range we utilise for the AFC and SW storage. Figure adapted from Ref. [174].
Figure A.2: The three steps of preparing a single-class feature: (a) creating a spectral pit, (b) burning-back population, and (c) cleaning. At each step, the range of transitions affected by the laser pulse are indicated in the top-left and the bottom image, the resulting spectrum is shown in terms of optical depth (OD) in the top-right image (and the laser frequency if within range), and the population distribution after the laser pulse is shown at the bottom, represented by circles.
empties out the population in the $5/2_g$ level, and creates antiholes at transitions from the $1/2_g$ and $3/2_g$ levels which burns back population in the region of the spectral pit. The pulse is resonant with the other 8 transitions in the 8 other classes, but only antiholes from transitions of the first three classes will be present in the region of the pit. Thus, after this point we only need to consider these three classes.

The final pulse is used to remove population from (or “clean”) the $3/2_g$ level of class 1, and remove population from classes 2 and 3. The pulse is sent at a detuning of 14.8 MHz with a frequency chirp of $\approx 4$ MHz. From the Fig A.2(c), we can see this pulse is resonant with four transitions across the classes. As a result, the population is removed from the $3/2_g$ level of class 1, the $1/2_g$ and $3/2_g$ levels of class 2, and the $1/2_g$ level of class 3. The population should be redistributed to the $5/2_g$ level in all of the classes, which is not in the spectrum of the spectral pit, except class 1 where it is also transferred to the $1/2_g$ level, which is acceptable because we want to prepare the AFC in that particular level and class.

### A.2 Preparing an AFC

To prepare a single-class AFC, we first follow the steps to obtain a single-class feature as described in the previous section. From this point we can prepare an AFC using the serial or parallel method (see Chap. 5). The explanation that follows will be for the parallel method, which is what we used in the experiments with SW storage.
A.2 Preparing an AFC

The waveform we use to burn the AFC in the parallel method is obtained by simulating the ideal AFC shape for a given storage time, then taking the Fourier transform of this structure. The resulting temporal waveform consists of a series of peaks clustered into lobes which decay in amplitude with the respect to the central lobe, and alternate between positive and negative values. As this waveform is quite long, we only use a few of the clusters around the peak. An example is shown in Fig. A.3, which corresponds to the waveform used to create a 10 µs AFC. Here we selected the central lobe plus the two side lobes, and the amplitude has been corrected for the response of the AOM with respect to the voltage of the driving signal. Longer storage times will generally require a longer waveform, as the time between consecutive peaks is equal to the AFC storage time.

Based on the Fourier transform, the side lobes should have a negative amplitude, which can be achieved with double-pass AOMs by applying an appropriate phase shift to the RF signal used for modulation [82]. We do attempt to implement this in our setup, but due to hardware limitations it is likely that the AWG does not implement the phase shift precisely. With our implementation, the individual pulses likely do not have the same phase, but there is a constant phase difference between consecutive pulses. Better control of the phase could improve the AFC efficiency, while conversely if the phases are random, the efficiency may be reduced. The coherence time of the laser could also affect the optical pumping achieved with this waveform, i.e., if the coherence time is not long enough, the AFC structure could be worse. The duration of the waveform is close in value to the laser coherence time, so it is unclear if this could be a limitation to our AFC efficiency.

We apply the waveform described above at the frequency of the $1/2_g \rightarrow 3/2_e$ transition. We send this burning pulse at four points, each 1 MHz apart, in order to prepare the structure uniformly over the comb bandwidth, as well as use less optical power per pulse which reduces the effect of power broadening. The population that has been removed during this process will be redistributed amongst the hyperfine levels, so we applying cleaning pulses to the $3/2_g \rightarrow 3/2_e$ transition again to ensure that the $3/2_g$ level remains empty. We alternate between applying an AFC burning pulse and a cleaning pulse in the order indicated in Fig. A.4(a). Each cleaning pulse is detuned by 10.2 MHz from one of the AFC burning pulses.
For the final step we apply additional cleaning pulses to the $3/2_g \rightarrow 3/2_e$ transition (indicated in Fig. A.4) as an extra step to remove any excess population. For these cleaning pulses in particular we use the same amplitude profile and frequency chirp range and profile as the control pulses used for SW storage, described in Sec. 5.3.2.

For the 10 $\mu$s AFC used in the experiments in this thesis, the sequence of pulses we use, including the class cleaning, is as follows:

- **Spectral pit**: square pulses with an amplitude of $0.17P_{\text{max}}$, a duration of 100 $\mu$s, a linear frequency chirp of 14.4 MHz centred at 9.4 MHz, and repeated 500 times.

- **Burn-back**: square pulses with an amplitude of $0.1P_{\text{max}}$, a duration of 100 $\mu$s, a linear frequency chirp of 3.8 MHz centred at 36.9 MHz, and repeated 100 times.

- **Clean**: square pulses with an amplitude of $0.88P_{\text{max}}$, a duration of 2 ms, a linear frequency chirp of 5.6 MHz centred at 14.8 MHz, and repeated 15 times.
• AFC burning: pulses with the waveform shown in Fig. A.3, with a peak amplitude of $0.29P_{\text{max}}$, no frequency chirp and centred at 3.1 MHz.

• Clean: a square pulse with an amplitude of $0.26P_{\text{max}}$, a duration of 10 µs and a linear frequency chirp from 1.8 MHz centred at 13.3 MHz.

The AFC burning and cleaning steps are repeated four times with the centre frequency shifted by 1 MHz each time, then repeated 100 times.

• Extra cleaning pulses: Gaussian pulses with an amplitude and frequency profile as shown in Fig. 5.6, a duration of 5 µs, a peak amplitude of $P_{\text{max}}$, a frequency chirp of 4.2 MHz and a centre frequency of 14.8 MHz. These are first repeated 100 times with a period of 40 µs, then 200 times with a period of 200 µs.

Note that the maximum power used here is approximately 29 mW for the extra cleaning pulses, which is denoted as $P_{\text{max}}$ in the list above, and the frequencies are given in terms of the frequency detuning in Fig. A.1
Appendix B

Further details for Chapter 5

B.1 Analysis of coincidence histograms

Figure B.1 shows a larger temporal range of the measurements used in Fig. 5.8. Here we show the counts used as the noise when calculating $g_s^{(2)}$ using Eq. 3.32. For the input, we use a long region of about 1.6 ms, all before the coincidence counts of the photon-pair. For the AFC, we use a region after the source has been switched off but before the AFC echo coincidences arrive. For the SW measurement, we use a semiconditional sequence as described in the main text so we can obtain noise counts at the same temporal position after the control pulses, but with no input and thus no SW echo. The SW echo histogram and the noise estimation histogram are shown in Fig. B.1. The large peaks between the transmitted signal photons (at 0 μs) and the SW echo are counts originating from leakage of the control pulses. Their shape differs from the profile shown in Fig. 5.8, which can be due to dispersion affecting their profiles through slow light effects, or a short FID modulating their profiles.

To calculate the efficiency, we compare the number of echo counts to the number of input counts. The input counts in Fig. B.1 are not representative of the actual number of input counts before the memory crystal as the spectral pit does not have 100% transmission. We measured the transmission through the crystal by sending classical pulses (with a temporal shape matching the signal photon) through a spectral pit with the usual polarisation along the $D_1$, and repeated the measurement with the pulses polarised along $D_2$ and detuned from the centre of the inhomogeneous absorption line by ~20 GHz so that the pulses should not interact with any Pr$^{3+}$ ions. By comparing these pulse areas, we estimated the
transmission through the spectral pit to be 86(2)%, and apply this correction to
B.2 Calculating the transfer efficiency of the control pulses

We can use the semiconditional SW storage measurements to calculate the transfer efficiency of the control pulses. We measured $\eta_{SW}$ as a function of $T_s$, with the results reported in Fig. 5.10 within Sec. 5.4. Here we use the same measurements, but only analyse the transfer efficiency.

For all of the measurements except $T_s = 6.9 \mu s$, we can observe the depleted AFC echo signal which is temporally located between the control pulses. Using this signal and comparing it with a standard AFC echo, we can calculate the transfer efficiency using Eq. 5.2, which is shown in Fig. B.2. The efficiencies are consistent within statistical uncertainties across the different $T_s$, with an average of 77(1)%. Using this value for the first control pulse, we can estimate the value for the second control pulse using Eq. 5.3 which is again consistent for different $T_s$. 

The input counts. Some of the loss in the pit may be due to the edge of the input mode overlapping with the wall of the pit, as well the delay between creating the pit and sending an input through it (a few hundred ms) during which population outside the pit can decay into it.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figB2.png}
\caption{Transfer efficiencies of the control pulses measured for different values of $T_s$. The values for the first pulse are calculated using Eq. 5.2 which are then used with Eq. 5.3 to calculate the efficiency of the second pulse, while the value assuming equal efficiency only use Eq. 5.3. The data originates from the same measurements used in Fig. 5.10.}
\end{figure}
with an average value of 40(3)%. Finally, assuming both control pulses have the same efficiency gives a transfers efficiency that is consistent for different $T_s$ with an average value of 56(2)%. The statistical uncertainty increases with increasing $T_s$ for the second pulse and equal efficiency calculations due to the decrease in SW counts at longer $T_s$. 
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