Coherent control, quantum memories and quantum repeaters

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In the domain of quantum communication, one of the main challenges is the distribution of entanglement over long distances. A scheme, the so-called quantum repeater, has been proposed in order to overcome the problem of channel losses. In this thesis we address different problems related to quantum repeaters, both theoretically and experimentally. We present an alternative scheme of quantum repeater based on atomic ensembles and linear optics. It is based on the use of a qubit amplifier and two photon interference. The main features of the protocol are that the distributed entangled state does not need post selection and the entanglement distribution rates approach the optimal rates one might expect from quantum repeaters using probabilistic swapping. An important concern of the whole class of quantum repeaters based on one photon interference is the phase stability in the communication channels. We performed an experiment evaluating the realistic phase noise in installed optical fibers in the Geneva urban area. We found fluctuations of order of 0.1 rad on the time scale of order of 100 $\mu$s. The feasibility of the phase [...]
Coherent control, Quantum memories and Quantum repeaters

THÈSE

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par

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Abstract

In the domain of quantum communication, one of the main challenges is the distribution of entanglement over long distances. A scheme, the so-called quantum repeater, has been proposed in order to overcome the problem of channel losses. In this thesis we address different problems related to quantum repeaters, both theoretically and experimentally.

We present an alternative scheme of quantum repeater based on atomic ensembles and linear optics. It is based on the use of a qubit amplifier and two photon interference. The main features of the protocol are that the distributed entangled state does not need post selection and the entanglement distribution rates approach the optimal rates one might expect from quantum repeaters using probabilistic swapping.

An important concern of the whole class of quantum repeaters based on one photon interference is the phase stability in the communication channels. We performed an experiment evaluating the realistic phase noise in installed optical fibers in the Geneva urban area. We found fluctuations of order of 0.1 rad on the time scale of order of 100 μs. The feasibility of the phase stabilization is discussed.

One of the essential building blocks of quantum repeater is a quantum memory. We focus our attention on quantum memories based on solid state atomic ensembles and photon echo like techniques. In particular, we show, that the atomic coherence in Er$^{3+}$:Y$_2$SiO$_5$ can be controlled to a very high degree using an external electric field. This allowed us to implement the CRIB protocol in this material and realize the first storage of light at the single photon level at telecommunication wavelength.

A necessary step for the implementation of CRIB and AFC storage protocols, in order to achieve long storage times and unit efficiencies, is the use of control light pulses. We developed a numerical Maxwell-Bloch simulator allowing us to study a coherent control of atomic excitations. In particular, we investigate the use of adiabatic chirped pulses in the context of AFC protocol. We study the relationship between the pulse characteristics and the echo properties. The comparison with the conventional π pulses and the impacts on the multimode capacity of the AFC protocol are also discussed.
Résumé de la thèse

L’intrication et une caractéristique fascinante de la Nature et elle est toujours un sujet de recherche excitant. En plus de son intérêt fondamental, elle a déjà trouvé des applications intéressantes, par exemple pour la distribution de clés quantiques [1, 2]. La distribution de l’intrication dans des fibres optiques a été démontrée jusqu’à la distance de 100 km [3], mais la distribution sur des distances arbitrairement grandes avec des taux raisonnables est toujours un problème.

La distribution de l’intrication sur des longues distances est un grand défi à cause des pertes dans les canaux de communication. Pour résoudre ce problème, Briegel et al. [4] ont proposé le répétiteur quantique. C’est un schéma où la distance totale est coupée en segments plus courts (appellés des liens élémentaires), où l’intrication est distribuée dans chaque lien élémentaire avec des pertes acceptables, en suite stockée dans les mémoires quantiques et finalement elle est permutée entre des mémoires voisines pour être distribuée sur la distance désirée.

Pour illustrer le problème des pertes, considérons une distance de 1000 km et des fibres telecom standards avec des pertes de 0.2 dB/km. La transmission directe est de l’ordre de 10^{-20} ce qui donnerait le taux de détection (en utilisant une source de 10 GHz) de 10^{-10} Hz, ce qui correspond à une détection tous les trois cents ans en moyenne - une échelle du temps pas très pratique. Ce problème peut être résolu en utilisant le répéteur quantique, dont les ingrédients essentiels sont: création, stockage et permutation d’intrication.

Dans cette thèse, on adresse divers problèmes liés aux répéteurs quantiques, à la fois théoriques et expérimentaux.

On présente un schéma alternatif du répéteur quantique basé sur des ensembles atomiques et de l’optique linéaire. Le répéteur est basé sur l’utilisation d’un amplificateur de qubits et sur l’interférence à deux photons. Les caractéristiques remarquables de ce répéteur sont que l’état final distribué n’a pas besoin d’être post sélectionné et que les taux de distribution d’intrication approchent les taux optimaux qu’on peut espérer dans le cas de répéteurs utilisant la permutation probablistique.
La stabilité de phase dans les canaux de communication est un sujet important concernant toute une classe des répéteurs basés sur l'interférence à un photon. On a réalisé une expérience où on évalue le bruit de phase dans des fibres optiques installées dans la zone urbaine de la ville de Genève. On a trouvé des fluctuations d'ordre 0.1 rad sur des échelles du temps d'ordre 100 µs. La possibilité d'une stabilisation active de la phase est discutée.

Un des éléments essentiels dans les répéteurs quantiques est la mémoire quantique. Ici, on concentre notre attention sur des mémoires quantiques basées sur des ensembles atomiques dans les solides et les techniques basées sur l'écho de photon. En particulier, on a démontré que la cohérence atomique dans $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$ peut être contrôlée très précisément en utilisant un champ électrique externe. Cette capacité nous a permis de réaliser le protocole CRIB dans ce matériau et de réaliser le premier stockage de la lumière au niveau d'un photon unique à la longueur d'onde télécom.

Une étape nécessaire dans la réalisation des protocoles CRIB et AFC, pour atteindre de longs temps de stockage et des efficacités proches de un, consiste à utiliser des impulsions lumineuses de contrôle. On a développé un simulateur numérique de Maxwell-Bloch qui nous a permis d'étudier le contrôle cohérent des excitations atomiques. En particulier, on examine l'usage des impulsions adiabatiques chirpiées dans le cadre du protocole AFC. On étudie la relation entre les caractéristiques des impulsions et les propriétés de l'écho. La comparaison avec l'impulsion $\pi$ conventionnelle et les conséquences sur la capacité multimode du protocole AFC sont également discutés.
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Introduction

With the important technological progress in last years and decades, the so-called quantum technologies are of a great interest both for the scientific community and the potential industrial applications. Based on the laws of quantum mechanics, people discovered a completely new way to process information. One of the main principles behind the quantum information science is that the information carriers can be in superposition of both logical 0 and logical 1, in strong contrast to classical bits. Among several seminal papers in quantum information, we can cite e.g. [5] where Feynman discussed the possibility of constructing a device called quantum computer, using tiny quantum systems as basic building blocks, or more recently [6], where the authors presented an experimental proposal of such device using trapped ions which can be individually addressed and manipulated with laser beams. Another example is the experiment performed by Aspect and colleagues [7]. They showed, by measuring a Bell inequality, that two photons coming out from a cascade in a calcium atom are polarization entangled. Entanglement is a fascinating property of Nature and is still an exciting subject of research. Besides the fundamental interest of better understanding entanglement and its demonstrations, such as the non-local correlations, it can produce, it has already found some interesting applications, more specifically a quantum key distribution, originally proposed by Wiesner [8] and developed by Benett and Brassard in 1984 [9]. Although the protocol of Benett and Brassard do not rely on entanglement, some other protocols proposed more recently do so, for example [1, 2, 10]. The entanglement distribution in optical fibers has been demonstrated over distances up to 100 km [3], but the distribution over arbitrarily long distances with reasonable rates is still an experimentally unsolved problem.

When tackling the task of distributing the entanglement over long distances, the essential problem are the channel losses. In order to overcome this, Briegel et al. [4] came up with the idea of a quantum repeater. This is a scheme which consists of cutting the total distance into smaller pieces (called elementary links), distribute the entanglement in each elementary link in which the losses are now acceptable, store the entanglement using quantum memories and finally swap the entanglement [11] between adjacent quantum memories in order to extend the entanglement over the whole distance.
To illustrate the problem of losses let us consider e.g. a distance of 1000 km and standard telecom fibers with losses of 0.2 dB/km. The direct transmission is of order of $10^{-20}$ which would yield the detection rate (using 10 GHz source) of $10^{-10}$ Hz, i.e. one detection every more than three hundred years, a time scale not very suitable for designing an experiment. The problem can be overcome when using a quantum repeater. The key ingredients of the quantum repeater are: creation, storage and swapping of the entanglement.

There are several methods of creating the entanglement used in quantum repeater. They are based either on the use of entangled photon pair sources or single photon sources. The realization of an optimal source is a crucial task for quantum repeater, but remains a big challenge for physicists. By optimal we mean an on-demand source with unit efficiency (works all the time) and fidelity (emitted state does not contain multipairs/multiphoton elements).

Storage of a quantum state can be implemented in many ways. Here, we focus on the quantum memories in solid state atomic ensembles using photon echo based techniques.

Next, we can distinguish two kinds of swapping, either probabilistic swapping [11], which can be realized with elements of linear optics or a deterministic swapping, which requires CNOT like operation (e.g. using Rydberg atoms, [12, 13] or individual ions, [14]). Even if the deterministic swapping enables one to speed up the entanglement distribution rate by orders of magnitude, it is very challenging from the experimental point of view. Furthermore, quantum repeaters based on atomic ensembles with probabilistic swapping are more developed and in the present work we thus limit our attention to this class of repeaters.

An overview of quantum repeaters based on linear optics and atomic ensembles (used as quantum memories) is given in [15]. In principle, we can distinguish the repeaters based on one or two photon interferences\(^1\). An important requirement for protocols based on one photon interference is the phase stability between the two arms of the interferometer. The other class of repeaters uses two photon coincidence measurements\(^2\) and is known to yield higher entanglement distribution rates (if we compare the performances without multiplexing). An important remark is, that some architectures require quantum memories at the telecommunication wavelength.

In our work, we address different subjects related directly to quantum repeaters, both theoretically and experimentally. We measure and analyze the phase noise in optical fibers, which is relevant for the one photon interference based quantum repeaters (Chapter 1). Then we propose an alternative scheme of a quantum repeater based on qubit amplifier (Chapter 2). Next, we give a brief overview of quantum

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\(^1\)Several architectures have been proposed, such as the DLCZ protocol [16], SPS protocol [1] or a protocol based on photon pair sources and multimode memories [17].

\(^2\)for example an IPP protocol [18].
memories in atomic ensembles based on photon echo like techniques modified for quantum storage. In particular we show, why standard photon echo techniques are not good protocols for quantum storage (Chapter 3). In Chapter 4 we present an experiment demonstrating our ability to control the atomic coherence in a solid state ensemble to a high degree using an external electric field. This is a crucial ability for some quantum storage protocols. Finally, in Chapter 5, we present theoretical work concerning the coherent control of atomic excitations, an essential step in the implementation of quantum memories with long storage times.

Every chapter has a Summary section for easier and faster orientation.
Chapter 1

Phase noise in realistic fiber based quantum networks

1.1 Motivation

As it was stated in the Introduction, one important step in the quantum repeater is the entanglement swapping. The principle behind the entanglement swapping is the indistinguishability of the incoming particles, photons in our case. Imagine one photon interference experiment, where the phase difference, say \( \phi \), between the two arms of the interferometer is constant. When scanning the phase over \( 2\pi \), one can obtain visibility fringes with, in principle, perfect visibility \( V = 1 \). If now, due to the noise in the arms of the interferometer, the constant phase \( \phi \) is "blurred", the visibility will decrease. This is a detrimental effect which is of special importance for quantum repeaters based on one photon interference. Lets suppose that we have one atomic excitation, labeled as \( |1\rangle \), delocalized in two memories \( A \) and \( B \), so that we can write for the state of the system [17]

\[
|\psi_{\text{wf}}\rangle = \frac{1}{\sqrt{2}} (|0_A1_B\rangle + e^{i\phi} |1_A0_B\rangle)
\]  

(1.1)

Consider now a phase noise such that the constant phase \( \phi \) will be changed by some amount \( \delta \phi \)

\[
|\psi(\delta \phi)\rangle = \frac{1}{\sqrt{2}} (|0_A1_B\rangle + e^{i(\phi+\delta \phi)} |1_A0_B\rangle)
\]  

(1.2)

If the phase fluctuation \( \delta \phi \) obeys some statistical law, i.e. if it is distributed with some distribution probability, the density matrix describing the system can be calculated. Lets assume a gaussian distribution of \( \delta \phi \)
\[ p(\delta \varphi) = \frac{1}{\sigma \sqrt{2\pi}} e^{-\frac{\delta \varphi^2}{2\sigma^2}}, \]  

(1.3)

where \( \sigma \) is the width of the gaussian distribution. The density matrix is then

\[ \hat{\rho}_{\text{real}} = \int d\delta \varphi \rho(\delta \varphi) |\psi(\delta \varphi)\rangle \langle \psi(\delta \varphi)| \]  

(1.4)

and fidelity of the disturbed state with respect to the ideal one then reads

\[ F = \langle \psi_{id} | \hat{\rho}_{\text{real}} | \psi_{id} \rangle = \frac{1}{2} \left( 1 + e^{-\frac{\sigma^2}{2}} \right). \]  

(1.5)

This is an illustrative example showing the direct relationship between the fidelity of the shared state and the phase noise in the interferometer.

In order to evaluate quantitative and qualitative properties of the phase noise in telecom fibers, we used two complementary interference techniques, namely the Sagnac and the Mach-Zehnder (MZ) interferometers. The first technique shows that due to its geometry it is relatively robust against phase fluctuations. One can infer the phase noise directly from the interference fringes. The investigation of the phase noise in the Sagnac setup is however limited to the time scales up to the propagation time of the light. The Mach-Zehnder setup provides us with the information about the phase noise on time scales longer than the propagation time as well as the distribution of the phase noise. The price to pay is that the analysis of the interference pattern is more complicated than those of the Sagnac setup.

### 1.2 Sagnac interferometry

The experimental configuration for the Sagnac setup is shown in Fig. 1.1. A laser pulse created by the acousto-optical modulator (AM) was sent to the 50/50 fiber beam splitter. The two pulses coming from the beam splitter then counterpropagate in the Sagnac interferometer. A phase is applied to one of the pulses using a pulsed phase modulator (PM). A complete phase scan (over 2\( \pi \)) can be realized yielding an interference fringe at the output from the interferometer. Polarization controllers were used to align the polarization of the interfering light pulses. More experimental details can be found in [H].

An example of an interference fringe for a 36.5 km Sagnac interferometer is shown in Fig. 1.2. The advantage of this method is that a visibility of the fringe can be directly related to the phase noise. Assuming a gaussian distribution Eq. (1.3) of the noise, it can be shown by a simple calculation [H], that
Figure 1.1: Sagnac interferometer: experimental set-up. A rectangular laser pulse created by amplitude modulator AM is split at the coupler and phase is applied to one of the pulses by the phase modulator PM. The resulting interference signal is detected by detector D and oscilloscope OSC. PC1 denote polarization controllers.

\[ V = e^{-\sigma^2/2}. \]  \hspace{1cm} (1.6)

Since the phase was scanned linearly, a visibility can be obtained using a sinusoidal fit of the data.

A set of measurements was carried out with different fiber lengths ranging from 6.5 to 75 km. The measurements were performed both in laboratory (temperature stabilized fiber spools) and commercial Swisscom fiber networks installed in the urban Geneva city area. The results are shown in Fig. 1.3. It can be seen, that for short distances, the visibility is almost perfect. Furthermore, even for the longest fiber lengths, the visibility remains relatively high. For example the visibility for the longest installed telecom fiber (L=71.5 km) is \( V = 93.6 \% \) during the day and 98 \% during the night, corresponding to \( \sigma = 0.36 \) rad and 0.2 rad for the propagation time of 360 \( \mu \text{s} \). A remark is, that the visibilities for laboratory measurements are slightly lower than those of installed fibers of comparable length. A reason for that might be, that when a disturbance affects the fiber (e.g. mechanical vibration), this is applied to the whole length of the fiber in the laboratory, while in the case of installed fibers it applies only locally.

1.3 Mach-Zehnder interferometry

Another method we used was a Mach-Zehnder interferometry. In this experiment we used two installed fiber links of 35 and 36.5 km. The difference was compensated by laboratory fibers such that the length difference between the interferometer arms was of order of few centimeters during the measurement which is several orders of magnitude less than the coherence length of the laser (66 m). An interference pattern was recorded using a cw laser light sent into the interferometer. Value of phase change (due to the noise) \( \Delta \varphi \), for each time interval \( \tau \) could be calculated. Like that, phase change distribution could be obtained for each time interval \( \tau \).
Figure 1.2: (Color in electronic form) Example of interference fringe for a Sagnac interferometer of 36.5 km in the telecom network. The solid curve is a sinusoidal fit which gives the visibility 99.2%. The time to scan a fringe is about 10 minutes, which shows that fast phase fluctuations do not degrade the visibility significantly in this case.

Figure 1.3: (Color in electronic form) Visibility as a function of fiber length for the Sagnac setup. The decrease of visibility with increasing fiber length is obvious as well as the qualitative agreement between laboratory and telecom measurements. High visibilities confirm the robustness of the Sagnac interferometer setup. For the length 71.5 km, the difference between the day (more phase noise) and night (less phase noise) values of visibilities in the telecom network is worth noting (the night measurements have been performed between 11 P.M. and 1 A.M.). Each point in the graph is the average of a set of interference fringes and the errors are the standard deviations.

ranging from few \( \mu s \) (no phase change) up to hundreds of \( \mu s \). More details can be found in [H].

Lets now have a closer look at how the phase noise can be simulated in terms of a random walk [19, p. 73]. We will first derive the probability distribution for a discrete random walk and show that this corresponds to the gaussian distribution assumed for the phase noise. Next, we will show the direct link between this result and the measured quantity, which is a mean phase change \( \Delta \varphi \).

Consider a discrete one dimensional random walk with equal probability 0.5 of a step in the positive and negative directions with the starting position zero. The probability to be at some distance \( r \) from the center of coordinates after \( n \) steps is

\[
P_{r,n} = \frac{1}{2^n} \binom{n}{\frac{n+r}{2}},
\]

(1.7)

In order to perform the calculation, consider \( r \) and \( n \) even, \( r = 2k, n = 2N \). For a given \( N \) we can then find a recursive formula for probabilities \( P_{k,N} \)
\[ P_{0,N} = \frac{1}{22N} \left( \frac{2N}{N} \right) = \frac{1}{22N} a_0, \quad (1.8) \]

\[ a_k = a_0 \frac{N(N-1)...(N-k+1)}{(N+1)(N+2)...(N+k)} = a_0 \frac{1 \cdot (1 - 1/N)...(1 - (k-1)/N)}{(1+1/N)(1+2/N)...(1+k/N)}. \quad (1.9) \]

Considering \( N \to \infty \) and \( k \ll N \) one can use the approximation

\[ e^x \approx 1 + x \quad (1.10) \]

such that Eq. (1.9) becomes

\[ a_k = a_0 e^{-k^2/N} \quad (1.11) \]

We can now approximate \( a_0 \) using the Stirling’s formula in the form

\[ N! \approx \sqrt{2\pi} N^{N+1/2} e^{-N} \quad (1.12) \]

\[ P_{0,N} = \frac{1}{22N} a_0 \approx \frac{1}{\sqrt{\pi} N}. \quad (1.13) \]

The final distribution probability is then

\[ P_{k,N} = \frac{1}{\sqrt{2\pi \sigma}} e^{-\frac{k^2}{2\sigma^2}} \quad (1.14) \]

with

\[ \sigma = \sqrt{N/2}. \quad (1.15) \]

This is a known result, that the random walk yields a gaussian distribution, which we assumed in Eq. (1.3). The gaussian distribution Eq. (1.3) depends on the time interval \( \tau \). More precisely, with increasing \( \tau \), the phase noise distribution becomes larger. In the model of discrete random walk we presented above, the time interval \( \tau \) is proportional to the number \( N \) of steps, \( \tau \sim N \), and \( \delta \varphi \sim k \). Equipped with the knowledge of the phase noise distribution, we can calculate the mean phase change \( \Delta \varphi \) during the time period \( \tau \)

\[ \Delta \varphi \equiv \langle |\delta \varphi| \rangle = \sqrt{\frac{2}{\pi} \sigma} \quad (1.16) \]
The mean phase change $\Delta \varphi$ can be calculated for each $\tau$. Let us now present and discuss some experimental results. The dependence of $\Delta \varphi$ on $\tau$ is shown in Fig. 1.4 (see [H] for details). Using Eq. (1.15) and Eq. (1.16) we should have $\Delta \varphi \sim \tau^{1/2}$. What we observe is rather $\Delta \varphi \sim \tau^{x}$ with $x = 0.7 - 0.9$. One hypothesis is, that the time period $\tau$ is not linearly proportional to the number of steps $N$ in the random walk theory. On the other hand, it should not affect the gaussian distribution of the form Eq. (1.3). Indeed, despite the discrepancy of the dependence of $\Delta \varphi$, we observed that the phase noise distribution is in a very good agreement with the gaussian distribution Eq. (1.3). One such example for a time of propagation through the interferometer $\tau = 182 \, \mu$s is shown in Fig. 1.5.

In order to compare the results obtained with the two interferometric techniques, we will use the relations Eq. (1.6) and Eq. (1.16). In order to compare the two different geometries, we have to consider the cases where $L_{\text{Sagnac}} = 2L_{\text{MZ}}$. The reason is, that the phase difference between the two counterpropagating pulses in the Sagnac interferometer does not increase over the whole trajectory. Indeed, when the traveled distance is $L_{\text{Sagnac}}/2$, the two pulses overlap each other and thus the phase difference between them is zero. We thus compared the 36.5 km Mach-Zehnder interferometer with the 71.5 km Sagnac interferometer, both installed telecom fibers. The visibilities obtained for the Sagnac interferometer were $V = 93.6 \pm 2.1\%$ during the day and $V = 98.0 \pm 1.1\%$ during the night. For the Mach-Zehnder interferometer, these values are $V = 97.1 \pm 2.4\%$ during the day and $V = 99.0 \pm 0.7\%$ during the night. The two methods thus yield the same results within the error.
Let us finally discuss the relevance of the obtained results with respect to possible phase stabilization. In [H] we considered an example of entangling the total distance of 1000 km with 16 elementary links and we showed that in order to have a fidelity of the final state $F=0.9$, the acceptable phase noise corresponds to $\Delta \varphi = 0.1$ rad over the distance 36.5 km. The crucial question is how long is the time period needed for entanglement creation in the two parallel chains, used in the one photon interference protocols. This time period might range from very short times of order of $\mu$s [17] up to seconds [20]. In our experiment, we showed that the phase remains stable (with $\Delta \varphi = 0.1$ rad) over a time interval of order of 100 $\mu$s. This indicates that if one requires the phase to remain stable over longer time periods (depending on the used protocol), the active stabilization might be needed. The active phase stabilization experiments over time scales of thousands of seconds have been carried out [21] with precision at the limit of what might be required for quantum repeaters, which is a very promising step towards possible future implementations.

1.4 Summary

In this work we investigated the phase noise on the time scales up to hundreds of $\mu$s in installed telecom fibers. We used two different interferometric techniques - Sagnac and Mach-Zehnder interferometry. The first method showed, that high visibility interference fringes are achievable, even for the fibers of 75 km ($V > 93\%$). The second method provided us with the structure of the phase noise (gaussian distribution) and its value on the time scales longer than the propagation time in the interferometer. The found phase change is of order of 0.1 rad on the time scale of order of 100 $\mu$s. The results obtained with both methods are compatible and we could also compare the results from installed fibers to those obtained in laboratory conditions. Finally, we discuss the need for active phase stabilization.
Chapter 2

Quantum repeater based on qubit amplifier

2.1 Repeater performance

Let us briefly recall some basic quantities characterizing quantum repeaters. A crucial quantity used to compare the performance of different quantum repeaters’ (QR) architectures is the entanglement distribution time over a given distance $L$. The formula for the entanglement distribution time for the repeater with $n$ nesting levels is [15]

$$T_{\text{tot}} = \left(\frac{3}{2}\right)^n \frac{L_0}{c} \frac{1}{P_0 P_1 \ldots P_n},$$

(2.1)

where $L_0$ is the length of the elementary link ($L_0 = L/2^n$), $P_0$ the probability of successful entanglement creation between the quantum memories in the elementary link and $P_k$ with $k = 1, 2, \ldots$ is the success probability of the $k$–th swapping.

The communication time $L_0/c$ is already an approximation of a more general formula, where it is replaced by $T_0 = T_s + L_0/c$. Here, $T_s$ is the source preparation time and is usually supposed to be much smaller than $L_0/c$. Let’s denote the probability of the successful preparation of the source $P_0$. We can then write

$$T_s = (P_0^{\gamma_{\text{rep}}})^{-1},$$

(2.2)

where $\gamma_{\text{rep}}$ is the repetition rate of the preparation of the source.

The legitimate question is what is the best possible scenario one can hope for quantum repeaters based on quantum memories and linear optics. A scenario which approaches this limit (in the two photon coincidence regime) consists of a perfect pair entanglement source (i.e. on demand source with unit efficiency and fidelity) at each site and with a Bell measurement (BM) in the middle between the two distant sites. Consider detection, quantum memory and channel transmission efficiencies
\( \eta_D, \eta_M \) and \( \eta \), where \( \eta = \exp(-L_0/(2L_{att})) \) is the transmission efficiency between one site and the BM. The probability of a successful entanglement creation in the elementary link is \( P_0 = 1/2(\eta_D \eta)^2 \), where the factor 1/2 comes from the fact, that we are performing a probabilistic BM (based on linear optics). The probabilities of the \( k \)-th successful swapping read \( P_k = 1/2(\eta_D \eta_M)^2 \) for \( k = 1, 2, \ldots \). The resulting formula for the entanglement distribution time in this ideal case is then

\[
T_{tot, id} = 2 \cdot 3^n \frac{L_0}{c} \frac{1}{(\eta_D \eta_M)^2(\eta_D \eta)^2}.
\] (2.3)

In order to have an idea of what one might expect in this ideal case, we consider a standard example with the total distance to be entangled \( L=1000 \) km, memory and detector efficiencies \( \eta_M = \eta_D = 0.9 \) and a channel (fiber) attenuation length \( L_{att} = 22 \) km (i.e. 0.2 dB/km). The nesting level was set to \( n = 4 \) (in order to compare with the results presented in 2.3.3). For these parameters, the optimal achievable entanglement distribution time is \( T_{tot, id} = 5.8 \) s.

2.2 Principle of qubit amplifier

First, let us remind the principle of qubit amplifier [1], which is shown in Fig. 2.1. Consider first the state coming from the pair source to be a polarization entangled state (modes \( g \) and \( in \) are polarization entangled) and a state coming to a tunable beam splitter BS with reflectivity (in intensity) \( R \) to be a product 2-photon state \( |H1V \rangle \). If the modes \( in \) and \( c \) are indistinguishable, the Bell measurement BM teleports the mode \( in \) to the mode \( out \) leading to the entanglement between the modes \( g \) and \( out \). The qubit amplifier thus serve as a heralded entangled pair source. Following the approach of [20], our BM consists of a PBS in the \( \pm 45^\circ \) basis followed by PBS in H/V basis in each output (see Fig. 2.1). This setup automatically eliminates the vacuum in the memories. This can be understood as follows. The only situation yielding vacuum in both memories is when no pair is emitted and both photons coming from the single photon sources are reflected at the BS. But such event, due to the design of BM, will never produce a coincidence detection between the two modes under consideration. As we will see, the success probability of the entanglement generation is rather small, but when the desired coincidence is detected, we obtain a state with high fidelity.

We present a scheme, how to use the qubit amplifier in order to obtain a near-perfect local source of entangled photon pairs and we show that it approaches the optimal entanglement distribution rates one can expect with repeaters based on linear optics and probabilistic swapping. In our protocol, thanks to the use of qubit amplifier, the density matrix does not contain the vacuum component and the post selection is thus not needed. This leads to an additional speed up of the entanglement distribution.
2.3 Quantum repeater based on local heralded sources of entangled pairs

![Diagram of quantum repeater system](image)

**Figure 2.1:** (Color in electronic form) Quantum bit amplifier, QM - quantum memories, BM - Bell measurement. A pair source (star) produces a polarization entangled state $\rho_{\text{pair}} = (1-p) |0\rangle \langle 0| + p/2 |gHin_H + gV in_V \rangle \langle h.c.|$ and two single photon sources (triangles) produce a product state of two photons in H and V polarization in mode $a$: $|H a V\rangle$. Mode $a$ is split at the beam splitter BS with tunable reflectivity $R$ (in intensity) and the two photon coincidence measurement teleports the mode $in$ to the mode $out$, so that the measurement results in the entanglement between the modes $g$ and $out$ which are then stored in the memories. The BM here is inspired by [20] and consists of one PBS in the $\pm 45^\circ$ basis followed by a PBS in H/V basis at each output.

2.3.1 Local entanglement generation

As discussed in the introduction, the possibility to realize a near perfect source (i.e. with unit efficiency and fidelity) locally is essential. This can be actually done when taking the two QMs storing modes $g$ and $out$ both sitting at the same site. In this case we consider the transmissions of channels $in$ and $c$ to be equal to 1.

So far we have assumed ideal sources. In order to have a more realistic model, let us consider imperfect sources. With analogy to [15] we consider a SPDC like source which emits the polarization entangled photon pair with probability $p \rho_{\text{pair}}$ and a double pair with probability proportional to $p^2$. The photon pair state is $\rho_{\text{pair}} = (1-p) |0\rangle \langle 0| + p/2 |gHin_H + gV in_V \rangle \langle h.c.| + \frac{p^2}{16} (|gHin_H + gV in_V\rangle)^2 \langle h.c.|$. We will denote by $q$ the probability of a photon emission from the single photon source. The emitted state is thus $\rho_{\text{single}} = (1-q) |0\rangle \langle 0| + q |H V\rangle \langle h.c.|$ for the H,V polarization. Let's denote the density matrix after a $d - \tilde{d}$ coincidence detection (see Fig. 2.1) $\rho_0$

\[\rho_{\text{0}} = (1-q) |0\rangle \langle 0| + q |H V\rangle \langle h.c.| + \frac{p^2}{16} (|gHin_H + gV in_V\rangle)^2 \langle h.c.|.

\[\rho_{\text{0}} = |gHin_H + gV in_V\rangle \langle gHin_H + gV in_V|.

---

1 In this section we use the notation $(gHin_H + gV in_V) |0\rangle \equiv |gHin_H + gV in_V\rangle$
(s for site or source of entanglement). In general, the density matrix has now the elements containing multiple excitations in the memories. However, one can get rid of some of them by considering some reasonable assumptions, concretely

\[
1 \gg R \gg p \quad (2.4a)
\]

\[
1 \gg 1 - q, \quad (2.4b)
\]

where \(R\) is the intensity reflectivity of the beam splitter BS. This corresponds to a photon pair source with a low double pair production and near perfect single photon sources. With these assumptions the non-normalized density matrix (up to the unitary transformation) in the site is

\[
\rho'_0 = \alpha'_0 \text{ent} + \beta'_0 g_+ + \gamma'_0 \text{out}_2 \cdot g_{2+} + \delta'_0 \text{out}_+ g_{2+}, \quad (2.5)
\]

where

\[
\text{ent} = |\theta_H \text{out}_H + g_V \text{out}_V \rangle \langle h.c. | \quad (2.6a)
\]

\[
g_+ = |\theta_H \rangle \langle \theta_H | + |g_V \rangle \langle g_V | \quad (2.6b)
\]

\[
\text{out}_2 = |\text{out}_H \text{out}_V \rangle \langle h.c. | \quad (2.6c)
\]

\[
\text{out}_+ = |\text{out}_H \rangle \langle \text{out}_H | + |\text{out}_V \rangle \langle \text{out}_V | \quad (2.6d)
\]

\[
g_{2+} = |g^2_H - g^2_V \rangle \langle h.c. |. \quad (2.6e)
\]

The coefficients then depend on parameters \(\eta_D, R, p, q, (T = 1 - R)\) as follows

\[
\alpha'_0 = \eta^2_D / 8pRT q^2 \quad (2.7a)
\]

\[
\beta'_0 = \eta^2_D / 8pR (1 - q + (1 - \eta_D)Rq) \quad (2.7b)
\]

\[
\gamma'_0 = (\eta D / 8)^2 (pT q)^2 \quad (2.7c)
\]

\[
\delta'_0 = (\eta D / 8)^2 p^2 T q (1 - q). \quad (2.7d)
\]

One can see, that the weight of the entangled state can be changed with respect to the undesired components by tuning the beam splitter reflectivity. The entanglement generation in the elementary link is created when two \(\text{out}\) memories are read out and the photons are sent to the central station where a BM is performed. The BM at the central station contains first a PBS in H/V basis and then a ±45° PBS in each output of the H/V PBS. The state in the elementary link after the coincidence detection is

\[
\rho_0 = \alpha_0 \text{ent} + \beta_0 (g_+ g_{2+}^\dagger + g_{2+}^\dagger g_+^\dagger) + O(g_{2+} g_{2+}^\dagger), \quad (2.8)
\]

where the modes at one site are labeled as \(g\) and at the other (distant) site as \(g^\dagger\). It can be shown, that when respecting the considered approximations Eq. (2.4), the terms containing two excitations in each memory become negligible.
2.3.2 Swapping operation

In order to extend the entanglement between two neighboring elementary links, the entanglement swapping has to be performed. The state in each elementary link is described by Eq. (2.8). Let’s denote the memory modes of the first (second) elementary link by $f$, $f'$, ($g$, $g'$). We will swap, say, between the primed modes. The swapping operation consists in performing the same BM as at the central station in the elementary link. This means, that the prime modes enter the two inputs of a PBS in H/V basis, which is followed by $\pm 45^\circ$ PBS in each output, and are measured in the coincidence detection. The explicit form after the swapping between two neighboring elementary links is of the form

$$\rho_i = \alpha_i \text{ent} + \beta_i (f + g_2 + g_2 + f_2) + \gamma_i f + g + O(g_2 + f_2),$$

(2.9)

where $\alpha_i$, $\beta_i$ and $\gamma_i$ can be found (for the sake of simplicity, we omit to write down their explicit form). As before, the term containing two excitations in each memory becomes negligible and from now on, each next swapping yields the density matrix of the form Eq. (2.9). The explicit knowledge of the density matrix in each step of the protocol allows us for the calculation of the probabilities of successful entanglement creation and swapping. In order to make some quantitative analysis, we use standard quantities, i.e. fidelity of the final state $F$, source repetition rate $\gamma_{rep}$ and entanglement distribution time $T_{ld}$.

The fidelity is defined as $F = \langle \psi_{id} | \rho_i | \psi_{id} \rangle$, where $\rho_i$ is the final density matrix (after $n$-th swapping) and $\psi_{id} = 1/\sqrt{2} | f_H g_H + f_H g_V \rangle$ is the desired state shared at the end by the two distant quantum memories $f$ and $g$. A standard approach in the calculation is to fix the fidelity one requires at the end, which in turn imposes the limitations on the quality of sources $p$, $q$ and then defines the maximum achievable entanglement distribution rate.

The calculation of the entanglement distribution time, fidelity and the repetition rate of the source is straightforward, treatable analytically, but providing us with rather large analytical expressions. The main results and comparison to the idealized case are the matter of the next subsection.

2.3.3 Results

For the numerical simulations, we considered a standard example with the total distance to be entangled $L = 1000$ km, memory and detector efficiencies $\eta_M = \eta_D = 0.9$ and a channel (fiber) attenuation length $L_{att} = 22$ km (i.e. 0.2 dB/km). The maximum allowed nesting level was fixed to $n = 4$ in order to avoid the eventual purification due to swapping imperfections [15]. It also allows us in practice to make a comparison with different QR architectures [15].
For the realistic model we proceeded as follows. First, we assumed the single photon sources as perfect sources \((q = 1)\). We then looked in the range of pair source probability \(p \in [10^{-4}, 0.2]\) and beam splitter reflectivity \(R \in [0.001, 0.4]\). \(^2\) We are interested only in results with final fidelity \(F \geq 0.9\). The results are shown in Table 2.1.

<table>
<thead>
<tr>
<th>(T_{tot} [s])</th>
<th>(p)</th>
<th>(R)</th>
<th>(F)</th>
<th>(\gamma_{rep} [MHz])</th>
<th>(\text{optimization})</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.2</td>
<td>(10^{-4})</td>
<td>0.037</td>
<td>0.98</td>
<td>4430</td>
<td>(T_{tot})</td>
</tr>
<tr>
<td>8.4</td>
<td>0.0081</td>
<td>0.4</td>
<td>0.9</td>
<td>7.6</td>
<td>(\gamma_{rep})</td>
</tr>
</tbody>
</table>

\(\textbf{Table 2.1:}\) Results for the qubit amplifier based quantum repeater. The results were calculated for total distance \(L = 1000\) km and four nesting levels \((n = 4)\). The table contains the total entanglement distribution time \(T_{tot}\), the quality of the source of pairs of photons \(p\), beam splitter reflectivity \(R\), final fidelity \(F\) of the shared state (lower bounded by 0.9), repetition rate of the source given by Eq. (2.2), for which \(T_s = L_0/c\) and the chosen optimization (whether \(T_{tot}\) or \(\gamma_{rep}\) was minimized).

From the conceptual point of view, we were interested in the minimal entanglement distribution time one can achieve (optimization of \(T_{tot}\)). On the other hand, the repetition rate of the source might be an important practical limitation, we thus looked for the minimal achievable source repetition rate \(\gamma_{rep}\) and the corresponding \(T_{tot}\) (optimization of \(\gamma_{rep}\)). The best result, in terms of achieved entanglement distribution rate and fidelity, is logically obtained for the lowest \(p\), since it produces a state with smallest errors. On the other hand, it requires very high source repetition rate\(^3\), which is of order of GHz. The smallest entanglement distribution time is \(T_{tot} = 7.2\) s. This value is very close to the ideal value given by Eq. (2.3), \(T_{tot, id} = 5.8\) s.

When minimizing the repetition rate, one can observe, that a little increase in \(T_{tot}\) (8.4 s instead of 7.2 s) permits a significantly lower \(\gamma_{rep}\) (decrease by three orders of magnitude!). The difference between the two times is caused by more errors in the case with higher \(p\) (lower fidelity).

In order to make a fair comparison with the IPP protocol \([18]\), one has to estimate the required resources with respect to the entanglement distribution time. Probably the most technologically demanding part of the repeater is the quantum memory. We thus find adequate to restrict our comparison to these resources. In the case of the IPP protocol one has to be able to store four modes in each site. In the case of our qubit amplifier based protocol, one has to be able to store four modes. However,

\(^2\)An obvious remark is, that the used approximations Eq. (2.4) are not fully respected (e.g. we use values of \(R\) up to 0.4 and of \(p\) up to 0.2). However, we verified the complete density matrix at each site \(\rho_0\) and in the elementary link \(\rho_0\) and for the results shown in Table 2.1 the weight of the neglected components is at least \(10^3\) times smaller than the weight of the entangled state and we thus consider the approximation valid. Nevertheless, such a check has to be done every time the conditions Eq. (2.4) are not fully respected.

\(^3\)Given by the condition \(T_s = L_0/c\) (see Eq. (2.2)).
there are in addition the two on demand single photon sources. In order to realize these, two more modes have to be stored, which demands the total storage capacity of 6 modes per site. This gives a factor 1.5 in favor of the IPP protocol from the point of view of resources and a factor roughly 2 in favor of the qubit amplifier based protocol from the point of view of the distribution times. Even in this comparison, the qubit amplifier based QR gives better results.

Finally, we would like to mention three more points. First, an additional speed up could be in principle achieved when considering multimode memories [17]. Unfortu-
natelly, with given repetition rates $\gamma_{\text{rep}}$ of order of 10 MHz, this would require QMs with very large bandwidth and multimode capacitities, which is at the limit of current technologies and we do not analyze this situation here. Second, the configuration of the on-demand source of entangled pairs (Fig. 2.1) requires that the out memory is at the telecom wavelength. Third, while talking about the fidelity in the case of the IPP protocol, we mean the fidelity of the state after the post-selection, during which we get rid of the vacuum and one photon components. In the case of qubit amplifier based protocol, the post-selection is not needed. This is an important point for quantum repeaters, because one way how to achieve high fidelity is using the post-selection, but this further increases $T_{\text{tot}}$.

2.4 Summary

In this work we presented a new quantum repeater architecture based on qubit amplifier, linear optics and quantum memories. We proposed a possible realization of an on demand source of entangled pairs of photons with a high fidelity (provided that the repetition rate of the preparation of the source is high enough). This is a main building block sitting at each site of our repeater. With the new protocol one can approach optimal entanglement distribution rates achievable with quantum repeaters based on linear optics and atomic ensembles. Furthermore, there is no need for post-selection. This might be useful for specific use of QR, e.g. in the frame of device independent quantum key distribution.
Chapter 3

Quantum memories in atomic ensembles

3.1 Overview

One of the building blocks of a quantum repeater is a quantum memory. It is a device enabling the storage of light during a controlled time and preserving the quantum properties of the light. In practice it means that one has to convert the quanta of light to some stationary excitation. This is done by letting the light interact in a controlled manner with some physical system at rest, i.e. with atoms, or atom-like systems. A lot of work has been done in this direction and quantum memory is still a very active field of research. A detailed description of different approaches is given in the literature, e.g. [22, 23].

Let's however make a quick overview in order to illustrate the potential of possible physical implementations. Physical systems under research are either single atom like systems or atomic ensembles. The latter are either gases (e.g. Rb or Cs atoms) or solid state ensembles (typically crystals doped with rare earths). Some representatives of the first group are single atoms, NV defects in diamond or semiconductor quantum dots. The other important thing when implementing a quantum memory is the choice of an appropriate protocol, which allows for efficient storage and retrieval of the light. Among others we can cite e.g. photon echo like, Raman scattering or electromagnetically induced transparency based techniques, which are common in the case of atomic ensembles.

All the mentioned physical systems together with the storage protocols yield a large choice in terms of available wavelengths, storage times, operating temperatures, complexity of the experiment, etc. In the following we focus our attention (for reasons mentioned below) to the solid state atomic ensembles and photon echo like based protocols.
3.2 Solid state quantum memories based on photon echo techniques

In our case, the solid state system under consideration is a host crystal doped by rare earth (RE) atoms. The host crystal we usually deal with is YVO₄ or Y₂SiO₅ matrix. Commonly used rare earths dopants are: Pr, Nd, Eu, Er and Tm. This choice was motivated by several reasons. RE atoms of interest have optical transitions in the visible and IR region. They replace Y atoms in the host crystal and interact only weakly with the environment. Since we are talking about solid state ensembles with a crystalline structure, the atoms do not move and the time of experiment, contrary to the gaseous atomic ensembles, is not limited by the time the atoms spend in the laser beam. In addition, there is no need for complex atomic trapping. The operating temperatures are in the range of few K, which is in the reach of standard closed cycle coolers, e.g. pulse tube coolers. A nice feature of RE atoms is that due to the weak interaction with the environment, they have long optical (up to 6 ms for Er³⁺:Y₂SiO₅ [24]) and spin (of order of s for Pr³⁺:Y₂SiO₅ [25]) coherence times. Since we dispose of different atoms, several different optical wavelengths are also available, ranging from 580 nm for Eu up to 1530 nm for Er.

The drawback of the RE atoms is that they have only small dipole moments [26] and thus interact weakly with the electromagnetic field. This is, fortunately, compensated by the fact, that a large number of atoms is interacting with the light. This is a very important point, which was in more detail investigated by Dicke in 1954 [27]. The light interacting with a lot of atoms creates a coherent superposition of atomic states and this yields genuine interference effects, where the emission of light is collectively enhanced. This means, that the collective state of N atoms yields different dynamics than N atoms acting individually, leading to emission intensities \( I \sim N^2 \). Among such phenomena we can mention optical free induction decay (FID) [28] or photon echoes [29], which will be of further interest. Photon echoes result from the action of a large number of atoms which are inhomogeneously broadened, but which have a homogeneous widths significantly smaller than the inhomogeneous one. This allows for creating specific absorption shapes, which are at the heart of a CRIB and AFC protocols discussed in this chapter. The desired absorption profile can be created using methods of optical pumping [30, 31].

3.2.1 Why are classical photon echo techniques not suited for quantum storage

The technique of photon echoes (PE) consists basically in manipulating the optical coherence using other optical pulses. The most common techniques are the so-called 2 and 3 pulse PE, see e.g. [32]. While proven very successful for storage of classical
pulses of light [33], they are not suitable for quantum storage. For the 2 pulse PE, a unit efficiency \(^1\) can be achieved, at least in principle [34]. It requires bright optical pulses, which are used to rephase the optical coherence (areas of rephasing pulses of order of \(\pi\)). This leads to highly populated excited level, which is the crucial problem and which in turn intrinsically limits the fidelity of the protocol due to spontaneous emission. The case of 3 pulse PE is potentially very attractive, since the information about the signal field is stored in the form of population grating and the storage time is thus limited only by the population lifetime, which can be very high, up to days for Eu\(^{3+}\):Y\(_2\)SiO\(_5\) [35].

Let us sketch the evaluation of the efficiency and of the fidelity of the 3 pulse PE in the case of two level atoms. Lets start with a two level atom (with ground and excited states labeled \(|g\rangle\) and \(|e\rangle\)) in a general state

\[
|\psi(t)\rangle = a(t) |g\rangle + b(t) |e\rangle. \tag{3.1}
\]

The action of a pulse with Rabi frequency \(\Omega_i(t)\) is given by the propagator (provided the spectrum of the pulse is much larger than the atomic detunings \(\Delta\))

\[
U^{\theta_i}(\tau) = \begin{pmatrix} \cos \big( \theta_i/2 \big) & -i \sin \big( \theta_i/2 \big) \\ -i \sin \big( \theta_i/2 \big) & \cos \big( \theta_i/2 \big) \end{pmatrix}, \tag{3.2}
\]

\[
\theta_i = \int_{-\infty}^{\infty} d\Omega_i(s) \approx \Omega_{i}^{\text{max}} \tau
\]

where \(\tau\) is the temporal duration of the pulse and \(\Omega_{i}^{\text{max}}\) is the maximum value of the Rabi frequency. Atoms, which are in the excited level acquire a phase given by a free evolution propagator (in the rotating wave approximation, see e.g. [36])

\[
U^\Delta(t_f, t_i) = \begin{pmatrix} 1 & 0 \\ 0 & e^{-i\Delta(t_f-t_i)} \end{pmatrix}. \tag{3.3}
\]

The 3 pulse PE pulse sequence is sketched in Fig. 3.1. After the action of pulses \(\Omega_1\) and \(\Omega_2\), the coefficients \(a\) and \(b\) of the state vector Eq. (3.1) read (at time \(t_2\))

\[
a(t_2) = \cos \big( \theta_1/2 \big) \cos \big( \theta_2/2 \big) \\
- \sin \big( \theta_1/2 \big) \sin \big( \theta_2/2 \big) e^{-i\Delta(t_2-t_1)}, \tag{3.4}
\]

\[
b(t_2) = -i \cos \big( \theta_1/2 \big) \sin \big( \theta_2/2 \big) \\
- i \sin \big( \theta_1/2 \big) \cos \big( \theta_2/2 \big) e^{-i\Delta(t_2-t_1)}. \tag{3.5}
\]

\(^1\)We define the efficiency of the storage as the ratio between the intensity area of the echo and of the incident (stored) pulse.
Next pulse \((\Omega_3)\) is applied at a time \(t_3\), such that the time interval \(t_3 - t_2\) is considered much longer than the coherence time of the transition. This means, that all the information is stored only in population grating and the density matrix of the system, before the application of pulse \(\Omega_3\), has only diagonal elements

\[
\rho(t_3) = |a(t_2)|^2 |g\rangle \langle g| + |b(t_2)|^2 |e\rangle \langle e|.
\] (3.6)

Then the pulse \(\Omega_3\) is applied and using again the propagators Eq. (3.2) and Eq. (3.3), we can calculate the density matrix at time \(t_4 = t_3 + t_2 - t_1\) when we expect the echo.

In order to evaluate the echo efficiency and signal to noise ratio (which is directly connected to the fidelity), we consider an inhomogeneously broadened atomic ensemble, such that the spectral distribution function \(g(\Delta)\) is a Gaussian with characteristic width \(\Delta_0\) and normalized as \(\int_{-\infty}^{\infty} d\Delta g(\Delta) = N\), where \(N\) is the number of atoms.

Another assumption we make is that the first absorbed pulse (the pulse to be stored) is a weak pulse, such that we can make the approximation \(\theta_1 \approx 2\epsilon\). It implies, that in the case of single photon excitation, \(N\epsilon^2 = 1\) (there is only one excitation delocalized in the atomic ensemble). We can now evaluate the polarization of the atomic ensemble at arbitrary time:

\[
P(t) = \int_{-\infty}^{\infty} d\Delta g(\Delta) \varphi \langle g| \rho(t) |e\rangle,
\] (3.7)

where \(\varphi\) is the dipole moment of the \(|g\rangle - |e\rangle\) transition. The intensity of the radiation associated with the atomic polarization is directly proportional to \(|P(t)|^2\). We can thus evaluate the intensities related to \(P\) at times \(t_1\) (original polarization we want to restore) and \(t_4\) (polarization giving the echo intensity). This comparison leads us to the result, that in the case of single photon excitation, the echo efficiency \(|P(t_4)/P(t_1)|^2\) is upper bounded by \(1/4\).

Our description is semiclassical, and thus does not involve the spontaneous emission. This one, however, can be estimated by considering no initial excitation \((\epsilon = 0)\) and by evaluating the population in the excited level. The probability of each atom being in the excited state at time \(t_4\) is \(\langle e| \rho(t_4) |e\rangle\). The noise is directly proportional to the population in the excited state and together with the echo intensity, the signal to noise ratio and the fidelity can be calculated. In our case of two level system and 3 pulse PE we obtain the maximum achievable fidelity \(F_{max}\text{-level} = 3/5\).

We actually assumed an atomic sample whose dimensions were small compared to the wavelength. This is why we omitted the spatial dependence. Our analysis can be however generalized to the case, where the spatial dependences are taken into account and the emission in a given direction \(k\) is evaluated. A further generalization is to consider a three level system with two photon resonance defining “effective”
**Figure 3.1:** Pulse sequence associated with the 3 pulse photon echo in two level atoms. The first pulse corresponding to $\Omega_1$ interacts with the atoms at time $t_1$ in order to be stored. The two next pulses, $\Omega_2$ and $\Omega_3$ applied at time $t_2$ and $t_3$ respectively, correspond to rephasing pulses. The echo signal is expected at time $t_4 = t_3 + t_2 - t_1$.

**Figure 3.2:** (Color in electronic form) The principles of the atomic frequency comb (AFC) quantum memory. (a) An inhomogeneously broadened optical transition $|g\rangle - |e\rangle$ is shaped into an AFC by frequency-selective optical pumping to the $|\text{aux}\rangle$ level. The peaks in the AFC are separated by $\Delta$. (b) The input mode is completely absorbed and coherently excites the AFC modes, which will dephase and then rephase after a time $2\pi/\Delta$, resulting in a photon-echo type coherent emission. A pair of control fields on $|e\rangle - |s\rangle$ allows for long-time storage as a collective spin wave in $|s\rangle$, and on-demand read-out after a storage time $T_3$.

two level system. Analogous calculation can be performed yielding similar results concerning fundamental limitation of the storage efficiency and achievable fidelity. More details can be found in [D].

### 3.2.2 Controlled Reversible Inhomogeneous Broadening - CRIB

The photon echo like protocols are based on the fact, that manipulating correctly the atomic coherence, one can force the atoms to rephase in order that a collective emission of light follows. The crucial difference compared to the photon echoes is, that in the protocols for quantum storage, a single excitation is manipulated without exciting other atoms. This prevents the protocols from the detrimental noise from spontaneous emission and a perfect fidelity can be in principle achieved.
Let us briefly recall a quantum storage protocol based on controled reversible inhomogeneous broadening (CRIB) [37, 38]. This protocol employs the fact, that when using an external field (magnetic or electric), the detuning of $j$-th atom $\Delta_j$ can be reversed with respect to the central frequency (zero detuning), schematically $\Delta_j \rightarrow -\Delta_j$. When a single photon is absorbed by a sample of $N$ atoms, the state of the system after a time $\tau$ reads (omitting the spatial effects)

$$\sum_{j=1}^{N} e^{-i\Delta_j \tau} |g_1...e_j...g_N\rangle.$$  \hspace{1cm} (3.8)

The state after the phase flip which occured at time $\tau$ becomes ($t > \tau$)

$$\sum_{j=1}^{N} e^{i\Delta_j (t-2\tau)} |g_1...e_j...g_N\rangle.$$ \hspace{1cm} (3.9)

The atoms in the excited state continue to acquire the phase (but with the opposite sign), such that at time $t = 2\tau$ all atoms oscillate in phase. This is at the origin of a collective reemission of light.

In practice, the usual technique for a detuning flip is to apply a gradient (along the axis of propagation of light, say $z$) of a magnetic [39] or electric field [40, E], which requires the use of atoms possessing a Zeeman degeneracy or a linear Stark effect.

### 3.2.3 Atomic Frequency Comb - AFC

Another approach consists in shaping the absorption profile of the inhomogeneously broadened transition $|g\rangle - |e\rangle$ in a way, that the atoms make a comb with the comb peaks equally spaced in frequency by $\Delta$ [41], see Fig. 3.2. The detuning of the $j$-th atom can be written as $\Delta_j = m_j \Delta$, where $m_j \in \mathbb{Z}$.

As for the CRIB protocol, the state of the atomic ensemble after the absorption of a photon is of the form Eq. (3.8). It can be readily seen, that if the argument of the exponential $\Delta_j \tau$ is a multiple of $2\pi$, then all atoms oscillate in phase leading to the emission of light. A first time when this occurs is for $\tau = 2\pi / \Delta$, so the time of echo emission is given by the comb structure. This means that, contrary to the CRIB protocol, the storage time in the AFC protocol is preprogrammed. In order to achieve on-demand readout, an additional state, say $|s\rangle$, to which the population from the excited state $|e\rangle$ is coherently transferred is needed (Fig. 3.2). But there are other reasons, because of which this transfer is required not only for AFC, but also for the CRIB protocol. One reason is, that the spin transitions usually have much longer coherence times, than the optical transitions and thus the storage time is significantly increased. Another reason is, that a unit efficiency can be obtained.
(for both protocols) only with backward echo emission [38, 41] (except the so-called longitudinal CRIB protocol [42]). Using the phase matching condition, the backward readout can be achieved only with two additional counterpropagating control pulses.

### 3.3 Summary

We presented a short overview of possible experimental implementations of quantum memories. We then explained, why solid state crystals doped with rare-earths are a good choice of a physical system. We also explained that in materials possessing an inhomogeneous broadening, photon echo based techniques can be used for the light storage. However, the classical photon echoes are not a good method for quantum state storage. We showed the limitations for the efficiency and the fidelity by explicit calculation in the case of 3 pulse photon echo. Finally, we made a short description of two methods based on the photon echo technique, but allowing for a quantum storage. These are controlled reversible inhomogeneous broadening (CRIB) and atomic frequency comb (AFC) protocols.
Chapter 4

Control of collective atomic coherence using an external electric field and its application to the CRIB protocol

4.1 Optical Free Induction Decay and the Stark effect

The goal of this experiment was to characterize (qualitatively and quantitatively) our ability to manipulate atomic coherences. When an atomic sample gets excited by an optical pulse, all atoms are initially in phase giving rise to a macroscopic dipole moment, which is at the origin of an emission of light from the sample. This emission, however, quickly decreases as the individual dipoles dephase with respect to each other. The dephasing is governed by the spectrum of the excited atoms. The atoms, just after their excitation, are in a collective coherent superposition giving rise to the above described effect, known as optical free induction decay (FID). More detailed analytical treatment can be found (for limited optical depths) e.g. in [43], chapter "Coherent transient Infrared Spectroscopy" written by R. L. Shoemaker.

If one is able to manipulate the spectrum of the excited atoms, it enables in turn the manipulation of the decay of the light. In practice, this can be achieved for example using the DC Stark effect [44], which requires the use of atoms possessing a permanent electric dipole moment. The detuning $\Delta$ of the atom due to the linear Stark effect can be described as [45]

$$\Delta = \frac{\chi}{\hbar} \bar{u} \cdot \vec{E}$$  \hspace{1cm} (4.1)
where $\vec{\mu}$ is the difference of the permanent dipole moments of the two states of optical transition, $\vec{E}$ is the applied electric field amplitude and $\chi$ is the Lorentz correction factor. A simplified intuitive picture describing the FID manipulated via the DC Stark effect is as follows. The phase evolution of the $j$-th excited atom is given by $e^{-i\omega_j^t}$, where $\omega_j^t$ is the frequency of the optical transition unperturbed by the external electric field. When the field is applied, the transition frequency is changed by $\Delta_j$. Suppose that the electric field is turned on at time $t=0$ and turned off at time $t = \tau$. The phase acquired by the atom is then $e^{-i(\omega_j^t + \Delta_j)\tau}$. If the $\Delta_j$ depends, say linearly, on the coordinate of the atom $z_j$, the atomic transition is broadened and the dephasing increases (thus, the FID signal decays faster). If the electric field is now switched to the opposite polarity at time $t = \tau$, the frequency shift $\Delta_j$ becomes $-\Delta_j$ and the phase of the atom at time $t$ is given by

$$e^{-i(\omega_j^t + \Delta_j)\tau} e^{-i(\omega_j^t - \Delta_j)(t-\tau)}.$$ (4.2)

It can be seen that at a time $t = 2\tau$ the externally introduced phase shifts $\Delta_j$ cancel for all atoms and the atomic phase oscillates again at the frequency $\omega_j^t$ of the optical transition. If the optical transition has a natural broadening, the atomic evolution due to the controlled dephasing and rephasing is superposed with the natural evolution due to the broadening. Hence, in the case of FID, the intensity of the light after the rephasing should reach the intensity of the unperturbed FID signal.

### 4.2 Setup

In the experiment we used a $Y_2\text{SiO}_5$ crystal doped with erbium ions Er$^{3+}$ (with 10 ppm concentration). The atoms were excited on the transition $^4I_{15/2} \rightarrow ^4I_{13/2}$ at the telecom wavelength of 1536 nm [46]. The $Y_2\text{SiO}_5$ crystal has three mutually perpendicular optical extinction axes labelled D$_1$, D$_2$, and b. The direction of light propagation $\vec{k}$ is along the b axis. The dimensions of the Er$^{3+}:Y_2\text{SiO}_5$ crystal are 6mm x 3.5mm x 4 mm along the b, D$_1$, D$_2$ axis, respectively. The crystal was cooled to 2.6 K in a pulse tube cooler (Oxford Instruments). The optical absorption depth of the crystal is $\alpha L = 2$. In order to create the electric field gradient, we implemented a quadrupole scheme [40] using four electrodes attached directly to the crystal, perpendicular to the D$_1$ axis (see inset of Fig. 4.1). Such a configuration creates an electric field in the D$_1$ direction which changes linearly along the axis parallel to the light propagation. The electrodes were thin aluminium stripes, each of 1 mm width and spaced by 1.5 mm. To switch the electric field we used a fast electrical switch with a switching time of 10 ns and minimal/maximal voltage -100/100 V.
The experimental setup is shown in Fig. 4.1. The light source was a free running external cavity diode laser (Toptica) at 1536 nm. The light was amplitude modulated by an acousto-optic modulator in order to create the excitation pulses at a repetition rate of 10 kHz, with duration 3 $\mu$s. The light was then coupled to a single mode optical fiber and passed through a variable fiber attenuator, before being focused in free space through the crystal in the cryostat. After the crystal, the light was again coupled into a single mode fiber and sent through a fiber coupled AOM that served as optical gate in front of the detector to block the excitation pulses. The measurements were made in the low excitation regime (with an excitation pulse area $\ll \pi$, typically with about $10^6$ photons in the excitation pulses). As the amplitude of the FID signal is strongly non linear with respect to the excitation intensity [28], the FID signal was extremely weak (about 50 photons) and was detected with a superconducting single photon detector (SSPD) [47].

It is worth to mention some details concerning the electric field. The electrodes were attached directly to the crystal and the fast electric signal was sent through a thin coaxial cable with metal shielding installed inside the cryostat. A good thermalization of the cable within the three stages of the used cryostat (at respective temperatures 300, 70 and 4 K) is essential in order to prevent the crystal from heating. We used a thermally conducting varnish and mechanical fixation to assure a good contact of the coaxial cable with the cooling elements. Another detail worth mentioning is the profile of the electric field inside the crystal. We evaluated the electric field inside the crystal (along the light propagation axis) using a numerical simulation involving the dimensions of the electrodes, which are comparable to the distances between the electrodes. The field profile is shown in Fig. 4.2. For our parameters of the experiment, the field gradient is nonlinear through the crystal, but remains still symmetric with respect to the polarity, which still allows for the rephasing of the atoms after switching the electric field.

4.3 Experimental collapses and revivals of the Free Induction Decay

In order to evaluate the effect of the electric field gradient on the atomic coherence, we use several sequences of electric Stark pulses. The first Stark pulse has a duration $\tau$ and the field is then switched to the opposite polarity. Two different sequences are shown in Fig. 4.3. If the electric field is kept constant after the switching, the atoms are in phase again after a time $2\tau$ and we observe a temporary revival of the collective emission at this time (Fig. 4.3a). However, if the field is switched off to zero at the time $2\tau$, the controlled phase evolution is frozen and the natural inhomogeneous dephasing governs the evolution. In that case, the revived signal follows the unperturbed FID signal (Fig. 4.3b). The dashed curve represents the
**Figure 4.1:** Experimental scheme used to demonstrate the electric control of collective atomic coherences. Light pulses created with an acousto-optic modulator (AOM1) are attenuated with a fiber variable attenuator (VAR ATT) and focused through an Er\(^{3+}:\text{Y}_{2}\text{SiO}_{5}\) crystal cooled at 2.6 K in a pulse tube cooler. The excitation pulses are then blocked by an optical gate implemented with a fiber acousto-optic modulator (AOM2) and the weak FID signal at the single photon level is detected with a superconducting single photon detector (SSPD). The inset shows the crystal with the quadrupole configuration of electrodes that produce the electric field gradient along the light propagation direction.

**Figure 4.2:** Numerical simulation of the electric field in the quadrupole configuration. The b (dashed line) and D\(_1\) (solid line) components of the electric field along the light propagation axis (in the middle between the four electrodes, i.e. for D\(_2\) = 2 mm and D\(_1\) = 1.75 mm) are shown. Due to symmetry considerations, the E\(_b\) field should be zero. The small deviation is because of the numerical resolution of the simulation.

original unperturbed FID signal. Other pulse sequences can be used, e.g. to realize a multiple revival.

It can be clearly seen, that the quality of the FID revival is almost perfect. It is indeed possible to quantify the quality of the revival, yielding visibilities (defined as the ratio between the revival and unperturbed FID intensities) above 95\% for all measured revival times. The details of the measurement, together with other experimental results are presented in [G].

### 4.4 Application to the CRIB protocol

In this chapter we have shown that the dephasing and rephasing of the collective atomic coherence can be controlled to a very high degree using the DC Stark effect
Figure 4.3: Collapse and revival of collective emission for different electric field sequences. The unperturbed FID signal is represented by the dashed line. The dashed-dotted curve represents the damped FID signal when the electric field is not switched. The voltage applied on the electrodes is ± 95 V. (a) Temporary revival obtained when the polarity of the electric field is reversed at time $\tau$ and the field remains constant afterwards. (b) Revival obtained when the polarity of the electric field is reversed at time $\tau$ and the field is turned off at time $2\tau$. The revived signal then follows the unperturbed FID signal.

and an external electric field. This is a crucial capability for applications in photonic quantum storage based on controlled reversible inhomogeneous broadening. Using the transition of Er$^{3+}$:Y$_2$SiO$_5$ at 1536 nm, we could realize the CRIB storage protocol at single photon level (storage of weak coherent pulses) and telecom wavelength. The details of the experiment are reported in [E]. An example of a CRIB echo with an average photon number $\bar{n} = 0.6$ is shown in Fig. 4.4.

More generally, the ability to switch on and off at will the collective emission of light from the sample is an interesting resource for quantum state engineering and quantum state manipulation. In the case of CRIB protocol, if the first Stark pulse applied to the absorption peak has different amplitude than the second Stark pulse, it will modify the reemission time as well as the shape of the echo. Indeed, the temporal shape of the CRIB echo is given by the spectrum of the atoms in the absorption peak. When the amplitude of the second Stark pulse is smaller, the atoms take more time to rephase and the absorption peak is less broadened (in frequency) leading to a longer echo (in time). Like that one can change the retrieved light at will using the external field. Several examples of such manipulation for different combinations of amplitudes of the first and second Stark pulses are shown in Fig. 4.5.
Figure 4.4: CRIB echo for average photon number in the incident pulse $\overline{n} = 0.6$ (integration time 25000ns). Dark counts have been subtracted from the data.

Figure 4.5: (Color in electronic form) Full width at half maximum (FWHM) of the CRIB echo for three different voltages $U_2$ applied to the sample after switching the polarity of the electrical field as sketched in the inset. Before switching a voltage of $U_1 = 95\text{V}$ (green triangles) and $U_1 = 65\text{V}$ (blue circles), respectively, were applied. One can see that using this method, the FWHM of the echo can be changed.

4.5 Summary

We have characterized our ability to manipulate the atomic coherence using external electric field. We have used a material (Er$^{3+}$:Y$_2$SiO$_5$) possessing a DC Stark effect and we implemented a quadrupole configuration of electrodes producing an electric field gradient along the light propagation axis. We then used a simple optical setup in order to detect a free induction decay (FID) coming from the sample illuminated beforehand by a pulse of light. The highly controlled decays and revivals of the FID were demonstrated. The control of atomic coherences using external electric field allowed us for the realization of the CRIB protocol, where we stored weak coherent pulses of light at a single photon level and at the telecommunication wavelength. Furthermore we could manipulate the CRIB echo, i.e. to change its temporal shape and the time of emission.
Chapter 5

Coherent population transfer in the context of AFC quantum memories

5.1 Constraints on the control pulses in AFC protocol

It has been said in 3.2.3, that in order to achieve long storage times and unit efficiency, two control pulses are necessary to transfer back and forth the optical coherence to the spin coherence. In the proposal of AFC [41], pulses performing such a transfer with a unit efficiencies are assumed (typically π pulses), but their properties are not discussed in detail. This is the matter of this chapter.

In order to motivate this work, we will briefly discuss two points, which are the multimode aspect and the phases in AFC protocol. A very interesting feature of the AFC protocol is that, without the need of increasing the optical depth, it allows for the storage of a large number of temporal modes. Lets assume an AFC with absorption peaks spaced by Δ in frequency and with total width of Γ (thus allowing for storage of light with total spectral width ≤ Γ.) The time duration of one mode is approximately \( \tau \approx 12\pi/\Gamma \) (the pre-factor comes from the condition that the overlap of two adjacent temporal modes has to be negligible [41]). The total duration of the pulse train is limited by the AFC storage time \( 2\pi/\Delta \). The number of modes that can be stored is thus given by the ratio between the pulse train duration and the duration of one mode, i.e. \( \Gamma/(6\Delta) \). This roughly corresponds to the number of peaks in the comb structure \( N_{\text{peak}} = \Gamma/\Delta \). In rare-earth-doped materials, the ratio between inhomogeneous broadening and homogeneous linewidth is inherently large, making possible to prepare combs with hundreds of peaks and thus to store many modes. In practice this means that one has to transfer efficiently atoms between the
excited and the spin level over the whole frequency range $\Gamma$. This in turn imposes some conditions on the control pulses (duration, Rabi frequency). At the same time, the phase relation between individual absorption peaks (see Eq. (3.8)) has to be preserved in order to reemit the AFC echo with perfect fidelity. The design and comparison of different transfer pulses (namely the $\pi$ and chirped adiabatic pulse) is discussed in what follows.

### 5.2 Qualitative comparison of chirped adiabatic pulses and $\pi$ pulses

Let us assume a chirped control pulse with Rabi frequency $\Omega_c$, duration $\tau_c$ and frequency $\omega_c(t)$ acting on the $|s\rangle - |e\rangle$ transition. The Hamiltonian of the system in the $\{|s\rangle, |e\rangle\}$ basis is

$$H(t) = \hbar \left(-\frac{\Omega_c(t)}{2} e^{-i\phi} - \frac{\Omega_c(t)}{2} e^{i\phi}\right),$$

where $\Delta_j(t) = \omega_{se}^j - \omega_c(t)$ is the time dependent detuning of the $j$-th atom and $\omega_{se}^j$ is the atomic transition frequency. An explicit propagator can be derived under specific assumptions. Let us first consider the case, where the Rabi frequency is larger than the maximal detuning (in the AFC this condition means $\Omega_c > \Gamma$) and that the detuning is not a function of time. The propagator is given by Eq. (3.2). Optimal population transfer is achieved for the pulse area $\theta = \pi$ which corresponds to a $\pi$-pulse.

If $\Omega_c(t)$ and $\Delta_j(t)$ are slowly varying in time, the adiabatic propagator can be derived. The adiabatic process means, that the state of the system is an eigenstate of the hamiltonian at every time (i.e. the system has time to change its configuration so that its state corresponds to the eigenstate). Assuming the detunings going from the negative to positive values, this propagator takes the form

$$U(\tau_c, 0) = \begin{pmatrix} 0 & -u_+ \\ u_- & 0 \end{pmatrix},$$

where $u_\pm = \exp \left(-i \int_0^{\tau_c} \frac{1}{2} (\Delta_j(s) \pm \sqrt{\Omega^2(s)^2 + \Delta_j(s)^2}) ds \right)$. The two propagators Eq. (3.2) and Eq. (5.2) drive the atomic state Eq. (3.8). It has been shown by explicit calculations [C], that for the sequence of two control pulses applied to the $|s\rangle - |e\rangle$ transition (see also Fig. 5.1), the collectivity of the state is preserved and thus the AFC echo is not disturbed by the use of the control pulses.
**Figure 5.1:** (Color in electronic form) A schema showing the complete storage sequence. A pair of chirped control pulses is applied on the transition $|e\rangle \leftrightarrow |s\rangle$. The signal, first and second control pulses and echo are centered at times $t_0, t_1, t_2$, and $t_3$ respectively. Both control pulses have the same chirp $\Delta(t)$ going from $-\Delta_{max}$ to $\Delta_{max}$. In the simplified intuitive picture $t^j_1$ ($t^j_2$) is the time when the $j$–th atom is transferred by the first (second) control pulse (see text for details). Idem for $(t^k_1)$ and $(t^k_2)$ for the $k$–th atom. The dashed line represents the inverse chirp - the case analyzed in section 5.4.1.

The fact that both adiabatic control pulses must have the same chirp in order to preserve the collectivity of the state can be intuitively understood considering the following picture (Fig. 5.1). The control pulses are applied at times $t_1$ and $t_2$. The chirp goes from $-\Delta_{max}$ to $\Delta_{max}$. Let’s see what happens from the point of view of the $j$–th atom which has a detuning $\Delta_j$. The atom is first excited by the signal pulse at time $t_0$ and then its phase evolves freely until it is transferred by the first control pulse at time $t^j_1$. The phase evolution is "freezed" until it is re-transferred to the excited state $|e\rangle$ by the second control pulse at time $t^j_2$ and then the phase evolves freely again until the emission of the echo. The overall time of the free evolution for each atom $j$ is thus

$$\tau = t_3 - t_0 - (t^j_2 - t^j_1). \quad (5.3)$$

It can be seen that the time difference $t^j_2 - t^j_1$ is independent of the atom’s detuning if the chirps of the two control pulses are the same. In consequence, it leads to the rephasing of all atoms at time $t_3$ and to the emission of AFC echo.

**5.3 Numerical Maxwell-Bloch simulator**

In order to simulate the complex interactions of light with an atomic ensemble, we developed a Maxwell-Bloch (MB) simulator for three level systems. In its current version, the model accounts for propagation effects (field modification due to absorp-
tion and stimulated emission), detuning of the excited and spin levels and decays of coherences and populations. On the other hand, the fields are propagating only in the forward direction.

In this section we will present the equations used to model the atom + light system. The derivation of semiclassical MB equations for two level atoms under the rotating wave approximation (RWA) and under the slowly varying envelope approximation can be found in the literature, e.g. [36, chapter 5] or [48, 49]. Following the same derivation, it can be generalized to three level system. The state of the atomic part is described by the density matrix, while the fields are described by their time dependent Rabi frequencies. In order to stick to the notation used in the Matlab code, the levels $|\psi\rangle, |e\rangle, |s\rangle$ are labeled $|1\rangle, |2\rangle, |3\rangle$ respectively (see also Fig. 5.2). We consider the fields only on the $|1\rangle - |2\rangle$ transition, called signal field with corresponding Rabi frequency $\Omega^S$ and on the $|3\rangle - |2\rangle$ transition called control field ($\Omega^C$). The density matrix elements are expressed in terms of Bloch vector: $r_{ij}^R = \Re \rho_{ij}$ is the real part of the coherence with $i,j = 1,2,3$ and $i \neq j$. Similarly we have $r_{ij}^I = \Im \rho_{ij}$ for the imaginary part. The populations are described by two components, expressing actually the population inversions: $r^{21}_P = \rho_{22} - \rho_{11}$ and $r^{31}_P = \rho_{33} - \rho_{11}$. The total population is conserved, i.e. $\sum_i \rho_{ii} = 1$. The complete set of Bloch equations reads

$$\frac{d}{dt}\vec{r} = A \cdot \vec{r} + \vec{b},$$

where

$$\vec{r} = (r^{12}_R, r^{21}_R, r^{13}_R, r^{23}_R, r^{21}_I, r^{31}_I)^T,$$

$$A = \begin{pmatrix} -\gamma_{12}^{12} & -\delta & \Omega^C/2 & \Omega^R/2 & 0 & 0 & -\Omega^I_{12} & 0 \\ \delta & -\gamma_{12}^{12} & -\Omega^C/2 & \Omega^R/2 & 0 & 0 & \Omega^I_{12} & 0 \\ -\Omega^C/2 & \Omega^R/2 & -\gamma_{13}^{13} & \Delta & -\Omega^S/2 & -\Omega^R/2 & 0 & 0 \\ -\Omega^C/2 & \Omega^R/2 & -\gamma_{13}^{13} & \Delta & -\Omega^S/2 & -\Omega^R/2 & 0 & 0 \\ 0 & 0 & \Omega^S_R/2 & \Omega^S_I/2 & -\gamma_{23}^{23} & \delta + \Delta & -\Omega^F_L & \Omega^F_R \\ 0 & 0 & \Omega^S_R/2 & \Omega^S_I/2 & -\gamma_{23}^{23} & \delta + \Delta & -\Omega^F_L & \Omega^F_R \\ \Omega^S_I/2 & -\Omega^S_R/2 & 0 & 0 & \Omega^I_C/2 & \Omega^C_R/2 & -2p & p - \gamma_{31}^{31} \\ \Omega^S_I/2 & -\Omega^S_R/2 & 0 & 0 & \Omega^I_C/2 & \Omega^C_R/2 & 2q & -(q + 2\gamma_{31}^{31}) \end{pmatrix}$$

and

$$\vec{b} = (0, 0, 0, 0, 0, -p, q)^T.$$

$\delta$ is the detuning of signal field and $\delta + \Delta$ is the detuning of the control field (see Fig. 5.2). $\gamma^{ij}_{ij}$ are the coherence decay rates and $\gamma^{ij}_{ii}$ are the population decay rates from level $i$ to level $j$. In the equations, we assumed a thermal coupling of the ground state $|1\rangle$ and the spin state $|3\rangle$, i.e. $\gamma^{31}_1 = \gamma^{13}_1$. Finally, we have $p = (2\gamma^{21}_1 + \gamma^{23}_1)/3$.
and \( q = (\gamma_{13}^2 - \gamma_{21}^2)/3 \). In our description, the decay rates \( \gamma_{1,2} \) are phenomenological and their values are introduced by hand.

The evolution of the fields along the \( z \) axis is given by the following set of equations

\[
\frac{\partial \Omega_R^S}{\partial z} = \frac{\alpha^{12}}{2\pi} \int d\delta g^{12}(\delta) r_I^{12} \\
\frac{\partial \Omega_I^S}{\partial z} = -\frac{\alpha^{12}}{2\pi} \int d\delta g^{12}(\delta) r_R^{12} \\
\frac{\partial \Omega_R^C}{\partial z} = -\frac{\alpha^{23}}{2\pi} \int d\Delta g^{23}(\Delta) r_I^{23} \\
\frac{\partial \Omega_I^C}{\partial z} = -\frac{\alpha^{23}}{2\pi} \int d\Delta g^{23}(\Delta) r_R^{23}.
\]  

\( \alpha^{ij} \) and \( g^{ij} \) stand for the absorption coefficient and normalized absorption profile (i.e. \( \max(g^{ij}) = 1 \)) of the transition \( |i\rangle - |j\rangle \). \( \Omega_{R,I} \) are the real and imaginary parts of the Rabi frequencies.

### 5.4 Numerical results

The discussion presented in section 5.2 is valid for a general adiabatic pulse. We have considered a secent hyperbolic chirped pulse, for which

\[
\Omega_c(t) = \Omega_c^{\text{max}} \text{sech}(t/\tau_p), \\
\Delta_j(t) = \Delta^{\text{max}} \tanh(t/\tau_p) + \Delta_j,
\]  

Based on the work presented in [50], the set of adiabatic criteria is found to be

\[
2\Delta^{\text{max}} \sim \Gamma, \\
\Delta^{\text{max}} \tau_p \geq 2, \\
\Omega_c^{\text{max}} \sim \Delta^{\text{max}} \sqrt{1 - \left(\frac{\log(1 - \eta)}{\pi \Delta^{\text{max}} \tau_p} + 1\right)^2}.
\]

\footnote{Technical remark: The absorption coefficient \( \alpha \) is proportional to the number of atoms and dipole moment squared. In the code we have considered the case, where the dipole moments of both optical transitions \( |1\rangle - |2\rangle, |3\rangle - |2\rangle \) are the same. The effect of the number of atoms is included in the matrices representing the Bloch vector (see code for details).}
The first equation guarantees that the pulse frequencies overlap all the atomic spectrum. The second and third inequalities insure that the chirp and the Rabi frequency vary slowly in time. These equations offer a systematic method to realize an efficient population transfer over the set of atomic spectral components \( \Gamma \). The relationship between the transfer efficiency \( \eta \) and the value of the Rabi frequency amplitude \( \Omega_{\text{max}} \) (with the pulse duration \( \tau_c \)) is given by the last equation. In principle, for longer pulses we can afford weaker Rabi frequencies and still perform a perfect transfer (provided, that the adiabatic criteria are respected). On the other hand, the multimode capacity of the AFC memory is reduced, so we have a trade-off between minimizing the Rabi frequency and the multimode storage capacity.

In order to perform a quantitative comparison using our MB simulator, we considered the following example, inspired by recent experiments in praseodymium-doped solids [F]. We take an AFC composed of Gaussian peaks with a width \( \gamma = 2\pi \times 25 \) kHz. The optical depth per peak is set to \( \alpha L = 4 \). This leads to an optimal finesse close to 4 [41], corresponding to a peak separation of \( \Delta = 2\pi \times 100 \) kHz. The comb is composed of \( N_{\text{peak}} = 40 \) peaks and thus spans a frequency range \( \Gamma = 40 \times \Delta = 2\pi \times 4 \) MHz.

Let denote \( \eta \) the transfer efficiency between the levels \( |s\rangle \) and \( |e\rangle \). We can extract the overall transfer efficiency \( \eta^2 \) (back and forth) from the AFC echo efficiency. The dependance of \( \eta^2 \) on Rabi frequency is shown in Fig. 5.3 for \( \pi \) and chirped sech pulses. The chirp amplitude of the sech pulses was set to \( \Delta_{\text{max}}^\text{sech} = \Gamma/2 = 2 \) MHz.

We performed the simulation for two different durations of sech pulses, \( \Delta_{\text{max}} \tau_c = 2 \) and \( \Delta_{\text{max}} \tau_c = 15.7 \) allowing for storage of 7 and 1 mode(s) respectively (the total number of modes which can be stored is 8). Note that the number of lost modes when using the adiabatic pulses can be made negligible with respect to the number of modes that can be stored by narrowing the individual peaks of the comb.

One see from the Fig. 5.3 that both techniques (\( \pi \) and chirped adiabatic pulses) can realize close to unit transfer efficiency. However, the adiabatic pulses require Rabi frequencies 2 to 5 times smaller than \( \pi \)-pulses for \( \eta^2 = 0.9 \), leading to a gain between 4 and 25 in intensity. Furthermore, the chirped adiabatic pulse preserves the fidelity of the storage independently of the Rabi frequency. More details can be found in [C].

### 5.4.1 Effect of inverse chirp

The choice of same chirps for both control pulses was explained in section 5.2. An interesting question is what happens, if the two chirps are not the same but opposite. Here, we study the effect of the opposite chirp on the echo efficiency and fidelity. The simulation was performed with the AFC parameters mentioned before and with \( \Delta_{\text{max}} = 2\pi \times 2 \) MHz and \( \Delta_{\text{max}} \tau_C = 5.7 \). Fig. 5.4 shows the example of echoes, where the two control pulses had the same chirp (dashed line) and opposite chirps (dotted
Figure 5.3: (Color in electronic form) Transfer efficiency with \(\pi\) - (dotted blue line) and with chirped (red lines) pulses. The dashed line corresponds to chirped pulses of minimal temporal durations (satisfying the adiabatic criteria) for which the multimode capacity decreases of one mode. The full line is associated to chirped pulses of maximal temporal durations such that only one mode can be stored. The gain in term of the required intensity depends both on the desired efficiency and on the number of modes that can be sacrificed. The circles and crosses are the values obtained using the relation (5.14c). One can see a very good agreement with the numerical simulations.

Figure 5.4: AFC echoes obtained when using a pair of control pulses, with identical chirps (dashed line) or opposite chirps (dotted line). A two level AFC echo (without the transfer to the spin state) is shown for comparison (solid curve). The parameters of the AFC and of the control pulses are explained in the text (see subsection 5.4.1).
repeaters, the photon stored in the memory and the retrieved photon do not need to be the same, only the two photons used in the entanglement swapping need to be the same.

5.4.2 Comparison to the experiment

We used also our MB simulator in order to analyze the transfer in the experiment presented in [F]. In the experiment the AFC was prepared in the Pr	extsuperscript{3+}:Y	extsubscript{2}SiO	extsubscript{5} crystal with the distance between peaks $\Delta = 2\pi \times 250$ kHz and the echo was observed with the efficiency $\eta_{echo} = 4 - 5\%$. We first optimized the optical depth $\alpha L$ and the finesse $F$ of the AFC. We could reproduce the experimental results for $F = 1.65$ and $\alpha L = 3$ which was in agreement with the experimentally estimated values. We then performed the simulation with the control pulses used in the experiment, i.e. chirped sech pulses with $\Delta_{max} = 2\pi \times 1$ MHz and $\tau_C$ corresponding to the intensity FWHM of 500 ns. In the experiment, because of practical considerations, the control pulses were truncated such that the cutoff of one pulse was 600 ns. The simulation yielded the echo with efficiency $\eta_{echo} = 2.8\%$ which is significantly higher than the observed value of 0.5-1\%. A possible explication is that the spatial overlap between the signal and control fields was not optimized, leading to an echo with poor efficiency. Another remark is, that because of the short cutoff of the control pulse, the adiabaticity conditions are not fulfilled. An increase of this cutoff, allowing for an adiabatic pulse, would increase the echo efficiency close to its original value. Indeed, the simulation with the same parameters, but with the cutoff increased to 1200 ns yielded an echo with $\eta_{echo} = 4.5\%$. Note also, that with a signal field of duration of 900 ns, control fields with the cutoff up to 3.1 $\mu$s could be used.

5.5 Summary

We discussed the problems related to the need for the phase preservation in the case of the AFC protocol with the transfer to the spin state. We gave a qualitative description of the $\pi$ and chirped adiabatic pulses and we compared their performance using our Maxwell-Bloch simulator for the three level system. We showed that a significant reduction of the Rabi frequency amplitude can be achieved in the case of the chirped adiabatic pulses, but the price to pay is lower multimode storage capacity. Next, we discussed the effect of the inverse chirp on the echo shape. Finally, we presented the comparison of our simulations with the experiment.
Conclusions

An important branch of quantum information science is the fastly growing domain of quantum communication. It is not only interesting from the point of view of fundamental physics, but it has already found impressive applications, for example in the form of quantum cryptography. Despite of the progress which has been made in the last decade, there are still unsolved problems and technological challenges. In our work we focused on the tasks related to the distribution of entanglement over large distances. We addressed different problems related to the entanglement distribution, both experimentally and theoretically.

Some of the proposed quantum repeater architectures, i.e. the schemes allowing for the entanglement distribution over large distances, require interferometric phase stability of the communication channels. We evaluated to which point these phase fluctuations are important in real-world telecom fibers (paper [H]). Our results suggest, that for some configurations a phase stabilization at the limit of current technology might be required, remaining thus a challenging project.

As it has been said, several schemes of quantum repeaters have been proposed up to date with the goal of achieving the best performances with minimal resources, which are the trends going usually in the opposite sens. Assuming realistic imperfections, such as limited efficiencies of the components, we have proposed two different approaches, namely quantum repeater based on single photon sources (paper [J]) and quantum repeater based on qubit amplifiers (paper [B]) showing alternative ways for long distance fiber based entanglement distribution. In the case of the latter protocol, we showed that it approaches the optimal entanglement distribution rates one might expect from quantum repeaters based on linear optics. In addition, the post-selection of the final state is not needed, which allows for an additional speedup and also for a larger use of quantum repeaters, e.g. for device independent quantum key distribution [1, 51].

It is known that the key points allowing for a significant speed up of the entanglement distribution are multiplexing and deterministic swapping. Both these points are pursued actively, but there is still a lot of space for new proposals and experimental realizations.
One of the building blocks of a quantum repeater is a quantum memory. Also here, the field of research is very flourishing. Our work concerned quantum memories based on solid state atomic ensembles and photon echo like techniques. It was rather accepted, although not formally formulated, that classical photon echoes are not well suited protocols for the quantum storage. We showed by explicit calculation the fundamental limitations in terms of efficiency and fidelity of the standard photon echo technique (paper [D]). On the other hand, the protocols designed for the quantum storage have been proposed, such as Controlled Reversible Inhomogeneous Broadening (CRIB) or Atomic Frequency Comb (AFC) protocols. We performed an experiment showing, that the atomic coherence can be well controlled using external electric field (paper [G]) and it enabled us to realize the CRIB protocol in Er$^{3+}$:Y$_2$SiO$_5$, the first demonstration of storage at telecommunication wavelength at the single photon level (paper [E]). Although this was a successful proof of principle experiment, we obtained relatively poor storage times and efficiencies. This could be a subject to further improvements, ranging from laser stabilization and more efficient optical pumping to obtaining higher optical depths.

When theoretically dealing with quantum memories, the usual problem is a general dynamics of light propagation in the atomic ensemble. Since these processes are in general hard to treat analytically, we developed a Maxwell-Bloch simulator in order to simulate them numerically. The use of the simulator is a powerful tool in solving some essential questions, e.g. to study the use of adiabatic pulses as control pulses in the photon echo like quantum storage protocols (paper [C]). There are almost permanently new questions arising concerning spatial dependences of the excitations in the ensemble, use of control pulses in the presence of a cavity, etc. The search for solutions might require, besides the analytical treatment, further development of a corresponding numerical support.

So far, we have presented a series of experimental and theoretical works showing the feasibility of quantum repeaters, which arise from the need to overcome the losses in the communication channels. In the following, we sketch possible research directions and domains, which have to be pursued in order to outreach laboratory boundaries and to realize a useful realistic quantum repeater. An obvious direction which comes on one’s mind is the reduction of the transmission losses. When considering silica based optical fibers, even if there is a hope to further reduce the losses, a fundamental limit (in this material) is imposed by Rayleigh scattering and is estimated to be 0.13 dB/km [52]. It is easy to see, that the time needed for a successful entanglement distribution (using a 10 GHz ideal entanglement source with noise-free unit efficiency detections) is 1000 s for 1000 km and thus, although it helps, is not a final solution. Hence we have to still focus on quantum repeaters. We should recognize and emphasize the gap between what has already been done and the eventual practical realization. Despite this gap, current results and progress in this field are very encouraging. The ongoing experiments are performed basically
in physics laboratories, but some problems are rather of an "engineering" nature. We remind that main building blocks of a quantum repeater are entanglement sources, storage and swapping of entanglement. All these building blocks are in themselves complex and active subjects of research and our goal is not a detailed analysis, but rather to point out the principal problems. The resolution of these problems, which is imaginable in the near or mid-term future, would bring us significantly closer to the realization of a true long-distance quantum repeater.

There are few general problems concerning sources of the entangled photons, such as coupling to the fibers (in the fiber based experiments), achieving high production rate with a good fidelity of the produced state and spectral matching with quantum memories. Depending on protocol, the source might be either probabilistic or on demand. Probabilistic sources are commonly realized with the method of parametric down-conversion, where a pump photon spontaneously decays into two entangled photons due to nonlinear process in the medium. The source can be implemented using a bulk crystal or a waveguide with a corresponding $\chi^{(2)}$ nonlinearity. It was shown [53] that the waveguide sources require several orders of magnitude less pump power in order to achieve comparable entangled pairs production rate as the bulk sources. With respect to the higher production rate and to the coupling, the waveguide source seems to be a natural candidate in order to be implemented, together with a pump laser, as an integrated circuit. In principle, the coupling efficiencies close to one can be achieved for specific configurations of the sources [54]. Other interesting approaches for the entanglement sources are based on the use of single ions [55] or quantum dots [56-58]. The use of the latter could significantly reduce the complexity and size of the optical setups. This holds in particular for the heralded single photon sources, where a detector (and potentially a quantum memory for the on demand operation) is traditionally used in addition to the source of the entangled pair. On the other hand, the bandwidth of the produced photons is usually large and does not match the bandwidth of the quantum memory. An important filtering is thus needed, reducing significantly the useful photon production rate. In this respect, schemes using directly quantum memories (e.g. [18]) as the entanglement sources are more attractive. As quantum memories are anyway a neccessary part of quantum repeaters, the entanglement production using memories should be investigated in more detail. At the end, it will be the further development which will show, which method of entanglement generation is the optimal one.

A very important step in the quantum repeater is a detection. In most cases it means a single photon detection, but for some specific operations (e.g. swapping, [13]), a detection of excited atoms rather than photons can be used. Concerning the photon detection process, a very promising direction is the domain of single photon superconductor detectors (SSPD). An ideal detector should be photon number resolving, fast, efficient and without noise. A realization of a so-called transition edge detector in the infrared domain with almost unit efficiency and no noise was
reported recently (see e.g. [59]). The drawback is a relatively slow detection rate limited to hundreds of kHz, since the detector is based on the bolometric principle. On the other hand, so called nanowire SSPDs can achieve a very high detection rate of order of GHz [60]. The progress in the fabrication methods is a good promise for further improvements in terms of efficiency and dark counts. The common denominator of the mentioned detection methods is that the operation temperatures are in the range of few Kelvins (of order of 100 mK for the transition edge detectors), so that the use of complex cooling is required. An interesting way to be explored is probably the use of high temperature superconductors, which are operating in the regime of tens of K, i.e. temperatures which can be achieved much easier.

We have already mentioned that a significant speedup can be achieved when using deterministic swapping and multiplexing. Deterministic swapping requires gate operations at the single photon level. Despite some existing proposals [61–63] this is a far from trivial task and a convincing experimental realization is still awaited, so that an important theoretical, but especially experimental effort in this direction is needed.

Multiplexing can be implemented in different ways, namely spatial, frequency or time multiplexing. From this point of view, the atomic ensembles become attractive, rather than individual atom like systems, because they possess many degrees of freedom allowing for multiplexing. While theoretical proposals for spatial or frequency multiplexing exist [64], the temporal multiplexing seems to be a step further since experiments showing the storage of tens and hundreds of modes have been carried out [65, 66]. In addition, we could point out the advantage of the AFC protocol which does not require the increase in optical depth in order to increase the storage capacity. As we said in chapter 3, a large number of possible implementations is under investigation. We should recall, that a lot of quantum memories operate at cryogenic temperatures, which might be a strong practical limitation for the complexity and scalability of the experiment. The search for solutions either in the direction of the memories operating at other than cryogenic temperatures or for more compact schemes of cooling is necessary. The required storage time can be significantly reduced by the multiplexing and deterministic swapping, bringing it within the reach of memories operating at room temperature [67]. This is one of the interesting options, which deserve our attention. Finally, it is difficult to pick up a particular realization of a quantum memory, each having its advantages and drawbacks, so that more research in different directions is needed to show the optimal way.

In our work we addressed only a few of a large number of elements which constitute the world of quantum communication and we showed that even in these restricted domains, there is still a lot of work to do, a work, which will, hopefully, continue to stimulate and occupy the minds and hands of physicists.
Bibliography


Publication list


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Published articles
Approaches for a quantum memory at telecommunication wavelengths

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We report experimental storage and retrieval of weak coherent states of light at telecommunication wavelengths using erbium ions doped into a solid. We use two photon echo based quantum storage protocols. The first one is based on controlled reversible inhomogeneous broadening (CRIB). It allows the retrieval of the light on demand by controlling the collective atomic coherence with an external electric field, via the linear Stark effect. We study how atoms in the excited state affect the signal to noise ratio of the CRIB memory. Additionally we show how CRIB can be used to modify the temporal width of the retrieved light pulse. The second protocol is based on atomic frequency combs (AFC). Using this protocol we also verify that the reversible mapping is phase preserving by performing an interference experiment with a local oscillator. These measurements are enabling steps towards solid state quantum memories at telecommunication wavelengths. We also give an outlook on possible improvements.

I. INTRODUCTION

Light-matter interfaces in general, and quantum memories for single photons in particular, are of great interest for the field of quantum information [1]. Quantum memories could be used to synchronize probabilistic quantum processes. These are essential ingredients in, for instance, quantum repeater architectures which would allow us to extend fiber-based quantum communication schemes beyond today’s distance limit set by attenuation in optical fibers [2–4]. Another potential application is to transform a probabilistic, but heralded, single photon source into a close to deterministic source of single photons, which is of interest for a variety of experiments and applications [5].

A photonic quantum memory requires reversible mapping of light, at the single photon level, onto long-lived coherent atomic excitations. Important progress has been made during the last years towards the realization of an efficient and coherent quantum memory [6–12]. Almost all of these experiments have been performed with atomic transitions in the visible range. However, in the context of quantum communication, it would be of great interest to have the ability to store and retrieve photons at telecommunication wavelengths (around 1550 nanometers), which can be transmitted with low loss in optical fibers. Such a quantum memory could be easily integrated into fiber optical networks. It would also be useful to realize a narrow band deterministic source of single photons at telecommunication wavelengths for quantum communication. Finally, a quantum memory capable of storing photons in this range is also required for some efficient quantum repeater architectures [4,13,14].

One possibility to interface photons at telecommunication wavelengths and quantum memories is to use frequency conversion techniques, i.e. to convert the wavelength of the telecom photon to the resonance frequency of existing solid state or atomic gases quantum memories in the visible range or vice versa [15–17]. But such a conversion will induce additional loss and increase the complexity of the experiment. Moreover, the application of a strong pump laser applied to a optically nonlinear medium, a commonly used method for such a conversion, can lead to noise which may blur the single photon signal [18]. Another possibility is to directly use atomic systems with an optical transition in the telecommunication range. A promising candidate in this context is erbium doped crystals, since erbium has a well-known transition around 1530 nm. In particular Er$^{3+}$ ions doped into Y$_2$SiO$_5$ crystals [19] are highly interesting since an extremely long optical coherence time of around 6 milliseconds has been obtained in this solid-state system [20,21].

Quantum storage in erbium doped solids is challenging because of limitations in the required memory preparation as described in [22] for Er$^{3+}$:Y$_2$SiO$_5$. Here we will report on another difficulty when implementing a quantum memory with this material. Residual population in the excited state due to imperfect memory preparation together with a low memory efficiency causes a low signal-to-noise ratio when performing storage experiments at the single photon level. We will discuss this issue in more detail and present solutions to this problem.

Despite these challenges, we report proof of principle demonstrations of photon echo based storage at the single photon level in an erbium doped crystal. The first one is based on controlled reversible inhomogeneous broadening (CRIB) [23,24]. The main results of this work have already been published elsewhere [25]. Here we give a more detailed study on how the weak CRIB signal is affected by the fluorescence noise from excited atoms. We also show how this technique can be used to control the temporal profile of the retrieved pulses [26]. The second method is based on an atomic frequency comb (AFC) [27] written into the inhomogeneously broadened absorption profile of the rare earth ion doped solid. It has already
been implemented in a range of different materials at different wavelengths \[10, 30–37\]. Here we present the first results at telecommunication wavelengths. Besides the novelty of the material and the wavelength, this experiment also includes previously unexplored aspects of the AFC protocol, such as an alternative way of creating the atomic frequency comb using standard hole burning techniques. We also study the coherence of the storage process by performing an interference experiment with a local oscillator. Even if the efficiencies obtained for both protocols are still low, it shows the feasibility of a memory at telecommunication wavelengths and it hopefully will stimulate further research on the spectral properties of erbium doped solids.

The article is structured as follows. In section II we briefly review the theoretical basis and the experimental state of the art for the two quantum storage protocols used in this paper. Section III describes the experimental procedure used. In section IV and V we present the results obtained for the AFC and CRIB protocols, respectively. Finally, in section VI we propose to combine the two methods and show first experimental results on that.

II. PHOTON ECHO BASED QUANTUM STORAGE

In this section we discuss the underlying process of both photon echo based protocols we explored in this work. For this let us consider an ensemble of atoms forming an inhomogeneously broadened atomic absorption. If a single photon spectrally matching this broadened line gets absorbed, the atoms will be in a superposition of states (collective excitation):

\[
|\psi\rangle = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{i2\pi \delta_j t} e^{ikz_j} |g_1 \ldots e_j \ldots g_N\rangle.
\]

\(g_j\) denotes atom \(j\) in the ground, \(e_j\) in the excited state. A dephasing will take place since atoms absorbing at different frequencies within this line will acquire different phases \(\delta_j t\), where \(\delta_j\) denotes the detuning from the central absorption line. This dephasing inhibits the collective emission of light by the ensemble. However, if one finds a way to undo this dephasing, i.e. to obtain an overall phase factor of one after a time \(T\) for all atoms, such an emission will take place. In the classical two and three pulse photon echo, this is done by the application of bright optical pulses on the optical transition. These methods are interesting since they enable the storage and retrieval of large trains of classical optical pulses \[38\]. However, conventional photon echo techniques have strong limitations regarding the storage of single photons \[39, 40\]. The main reason is the application of the strong pulses that are necessary to induce the rephasing. They will transfer population from the ground- to the excited state and therefore cause an intrinsically low fidelity due to noise from incoherent de-excitation (fluorescence).

In order to use photon echo techniques as quantum storage protocols, it is thus necessary to devise methods where the rephasing mechanism does not involve a population of the excited state. In this article we present measurements on photon echoes at the single photon level using two different schemes fulfilling this requirement: controlled reversible inhomogeneous broadening (CRIB) and atomic frequency combs (AFC), which we are both going to describe in more detail now.

A. Controlled Reversible Inhomogeneous Broadening

The underlying rephasing mechanism of this scheme is based on a reversal of the inhomogeneous broadening in a controlled way (therefore it is known as controlled reversible inhomogeneous broadening, CRIB). This protocol was first proposed for hot atomic gases, where the Doppler shift of atoms depends on the direction of laser beams \[24\]. CRIB was later extended to rare-earth doped solids by three different groups \[24–26\]. The idea is the following \[24, 41\]: First, a narrow absorption line is created within a large transparency window. This can be done using optical pumping techniques as will be described in section III B. This initially narrow absorption line is then artificially broadened. In our case this is done by the application of an electric field gradient in the direction of light propagation (\(z\)). Since the ions possess a permanent electric dipole moment, their optical resonance frequency will shift due to the linear Stark effect by an amount \(\delta_j\) that depends on the position \(z_j\) of ion \(j\). After the absorption of a photon the atoms will be in a state as given by equation I and the atomic dipoles will dephase accordingly. This dephasing can be reversed by changing the sign of the detuning \(\delta_j \rightarrow -\delta_j\) at a time \(\tau\), which can be done by simply flipping the polarity of the electric field. Note that \(\tau\) can be chosen after the absorption ("on demand" readout of the memory). The state of the system at a time \(t\) after the flip becomes

\[
|\psi\rangle = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{i2\pi \delta_j \tau} e^{-i2\pi \delta_j t} e^{ikz_j} |g_1 \ldots e_j \ldots g_N\rangle.
\]

(2)

It is obvious that at the time \(t = \tau\), i.e. a total time \(2\tau\), the spectral phases will have canceled. The atomic dipoles are back in phase and the ensemble will collectively emit an echo of the incident light in the forward direction.

The forward retrieval efficiency is given by \[42\]

\[
\eta_{CRIB} = d_{br}^2 e^{-d_{br}} e^{-d_{br}} e^{-i2\tilde{\gamma}z^2},
\]

(3)

where \(d_{br}\) is the optical depth of the broadened peak, and \(\tilde{\gamma} = 2\pi\sigma\) the spectral width of the initial peak (standard deviation). The first term gives the raw efficiency for the photon to be absorbed and re-emitted. The second
term accounts for the possibility for the photon to be re-absorbed by the medium. In case of inefficient optical pumping, (re-)absorption of the light by an absorbing background with optical depth $d_0$ occurs, which is taken into account here as well. The last term describes the decoherence due to the finite width of the initial peak. Note that a unit efficiency memory can be achieved using a backward readout [42], provided that $d_0 = 0$.

CRIB was first realized in europium doped Y$_2$SiO$_5$ crystals [26]. Since the initial demonstration, a lot of progress has been made concerning its efficiency using praseodymium as dopant, approaching 70% in the quantum regime [11]. We have recently demonstrated the feasibility of the protocol at the single photon level in the telecommunication band using an erbium doped sample [27]. In this article we present an analysis of the noise occurring in this experiment and the methods we applied to reduce it which allowed us to work in this regime. Furthermore we present measurements showing pulse-compression and stretching at the few photon level. Note that CRIB has also been realized with controlled broadening of spin transitions in a rubidium vapor [43]. Its capability to serve as a pulse sequencer and pulse shaper also was shown experimentally [44].

### B. Atomic Frequency Combs

The rephasing mechanism in this scheme is based on a periodicity in the absorption profile [29]. We start with a number of equidistant absorption peaks with a separation of $\Delta$ (atomic frequency comb, AFC). Let us consider an incident photon with a central frequency $\omega_0$ and a spectral width that is smaller than the total width of the comb. The light will be absorbed as a single excitation distributed over the atoms forming the absorption peaks.

We can write the state of the atoms in the same manner as in Eq. 1. In the ideal case of very sharp comb peaks $\delta_j$ is given by $\delta_j = n_j \Delta$, where $n_j \in \mathbb{Z}$ is the number of the peak the ion belongs to. It is easy to see that, due to this periodicity, for times

$$T_m = \frac{m}{\Delta} \quad (m \in \mathbb{N})$$

the phase factors all are equal to one, i.e. the atomic dipoles are back in phase and a collective emission will take place (note that here the unit of $\Delta$ is Hertz). Unlike in the CRIB protocol the rephasing mechanism is an intrinsic property of the spectral shape and no external manipulation is required. However, for this reason the moment of rephasing can not be changed after the absorption. An additional coherent transfer of the excitation to a ground state spin level and back is required [29] in order to improve the scheme from a delay to an on demand memory.

The efficiency of the AFC scheme in the forward direction for the first echo is given by [10, 29]:

$$\eta_{AFC} = \frac{d^2}{F^2} e^{-\frac{d}{\gamma}} e^{-d_0 \frac{1}{\gamma}} \frac{1}{\pi^2} \frac{\Delta^2}{\gamma^2}.$$  \hspace{1cm} (5)

The finesses $F$ is the ratio between the width of the peaks $\gamma$ and their separation $\Delta$. $d$ is the optical depth of the peaks. Note that this formula is similar to the one given for the CRIB scheme (eq. 3). The optical depth of the broadened peak is simply replaced by the effective optical depth of the AFC, $d/F$. If one writes the finesses $F$ in terms of the storage time $T_1 = 1/\Delta$ and linewidth $\gamma$, one will find that the last term corresponds to the decoherence term in equation 3. Also here an absorbing background is taken into account. Note that, as for CRIB, unit efficiency is possible in backward configuration [29].

The AFC protocol has been used to demonstrate the first solid state light matter interface at the single photon level [10] and more recently to demonstrate entanglement between a photonic qubit and an atomic excitation in a solid [30, 31]. The extension of the scheme by spin wave storage was recently reported [31]. Further work has shown improved storage efficiencies [32, 34]. The most important feature of this protocol, that one can efficiently store and retrieve multiple temporal modes (see sec. II C 1), was also shown [10, 33, 45]. In this work we demonstrate AFC experiments at telecommunication wavelengths in an erbium doped crystal.

### C. Comparison of the two protocols

At this point we would like to give a brief comparison of the two protocols.

#### 1. Multimode Capacity

The possibility to store a train of incident photons (multiple temporal modes) in a quantum memory is of great interest in the context of quantum repeater architectures [40]. It has been shown that the number of temporal modes $N$ that can be efficiently stored in a CRIB memory grows linearly with the optical depth $d$ of the initial unbroadened absorption line [16, 47]. This can be understood by the following arguments:

The number of temporal modes that can be stored in the memory is roughly given by the ratio of the time $\tau$ (from the first input pulse to the moment of switching) to the duration of the each input mode $T_{CRIB} = N \cong \frac{\tau}{T_{CRIB}}$. The storage time of the CRIB protocol is of the order of
the inverse of the width of the initial peak $\tau \propto \frac{1}{\Delta \nu_{\text{crim}}}$. On the other hand, the broadened absorption profile should be at least as large as the linewidth of the incident light (see section IV). The number of temporal modes that can be stored is thus obviously given by the factor with which the initial line was broadened. The optical depth $d_{br}$ of the broadened line, however, decreases linearly with this factor and this diminishes the storage efficiency (see eq. 3). In short, to double the number of temporal modes that can be stored while keeping the storage efficiency constant, one has to double the initial optical depth $d$.

The situation is different for the AFC protocol. In order to increase the spectral width of the comb one can add peaks to it. In fact, the number of modes is limited by the number of peaks [29]. The optical depth and the finesse are not affected and the efficiency does not change (eq. 5). The limitations for the spectral width (and thus the number of peaks) of a comb, are material properties. These are for example the inhomogeneous broadening as well as hyperfine level spacings. The number of temporal modes that can be stored does not depend on the available optical depth, contrary to all other QM proposals. Furthermore is the efficiency for all modes the same. This is in contrast to the CRIB protocol where the efficiency decays exponentially.

An interesting aspect in the context of the multimode capacity of the two protocols is that AFC is a First In - First Out (FIFO) memory [16, 45], while CRIB is a First In - Last Out (FILO) memory [44]. This may be of interest for future applications of quantum memories.

2. Spin Wave Storage

The transfer of the excitation to a long-lived ground state level using control pulses ($\pi$-pulses) is a required step for an AFC memory in order to achieve an on-demand readout. However, it can also be used in the CRIB scheme to increase the storage time [24, 25]. In the case of a multimode memory discussed before, the bandwidths of the control pulses have to be sufficiently large in order to transfer all ions of the collective ensemble efficiently. In order to achieve this, one may use chirped pulses such as complex hyperbolic secant pulses [31, 48, 51]. These require a sufficiently long time between input mode and echo - in order to be able to apply them over the entire bandwidth - as well as a high oscillator strength of the transition. To realize an efficient multimode AFC-memory with spin-wave storage can thus be difficult. Since in CRIB the width of the spectrum can be controlled externally, one can narrow the required bandwidth to the width of the initial peak. Note that in this case the electrical control field is off during the transfer. The ability of being able to fully control the excitation spectrum with the electric field is a great advantage here, since it considerably relaxes the constraints on the control pulses to obtain an efficient transfer. Thus an efficient transfer is more easily achievable for the CRIB scheme. Note that both protocols require a third ground state level for spin wave storage.

3. Material Aspects

The CRIB protocol requires the existence of a linear Stark or Zeeman effect in the medium which allows to induce and control the inhomogeneous broadening. In thulium doped YAG for example, the Zeeman effect is very weak since Tm$^{3+}$ is not a Kramers ion and the magnetic field only acts on the nuclear spin. A fast switching of high magnetic fields, as they would be required in this material, is not possible. Moreover, a linear Stark does not exist in Tm$^{3+}$:YAG due to the crystal symmetry. While it is thus impossible to use Tm$^{3+}$:YAG in CRIB experiments, AFC experiments do not require either of the two and have already successfully been achieved in this material [30, 33, 35]. AFC can thus be implemented in a larger range of materials.

To sum up: the main advantage of the AFC is the multimode capacity. It can be performed in a higher variety of materials since it does not depend on the existence and the strength of the interaction with an external electric or magnetic control field. However, the transfer of excitations to a long-lived ground state level required for an on-demand readout in a AFC memory is more challenging. Both protocols have their advantages and are worth being investigated.

III. EXPERIMENTAL SETUP AND NOISE REDUCTION

In this section we describe the experimental setup as well as the optical pumping techniques and the experimental cycle common to both experiments presented in this work. We also study the main source of noise present in the system, which could be identified as fluorescence due to imperfect memory preparation. We show how this noise can be reduced.

A. Setup

The sample used is an Er$^{3+}$:Y$_2$SiO$_5$ crystal (10ppm) placed on the cold finger of a pulse tube cooler (Oxford Instruments), cooled down to 2.6K. The Y$_2$SiO$_5$ crystal has three mutually perpendicular optical-extinction axes labeled D1, D2, and b. Its dimensions are $3.5 \times 4 \times 6$mm along these axes. The Er$^{3+}$ can be found at two crystallographic inequivalent sites (sites 1 and 2). All measurements in this work were carried out on ions at site 1. A magnetic field was used to induce a Zeeman splitting necessary for state preparation. It was provided by a permanent magnet outside the cryostat and was applied in the $D_1-D_2$ plane at an angle of $\theta = 135^\circ$ with respect to the $D_1$-axis [52] (Figure 1 inset). For all measurements...
it was set to 1.5 mT in order to allow for an optimal optical pumping efficiency and not to decrease the optical depth by separating the inhomogeneously broadened Zeeman transitions completely. The light was traveling in a direction parallel to b and its polarization was aligned to maximize the absorption [19]. The optical depth of the sample was 2. The electric quadrupole field for the CRIB experiment was applied with the use of four electrodes placed on the crystal as shown in the inset of figure 1 and described in [53].

The experimental setup is shown in figure 1b. The preparation of the memory and the storage and retrieval of the weak pulses. For state preparation an external cavity diode laser at 1536 nanometers (Toptica) was used. It also served as light source for the weak input pulses. The timing of the experimental sequence is shown in Fig. 2. The preparation of the memory consisted in preparing the specific absorption structure required for each protocol (a single absorption line for CRIB and a comb for the AFC). The preparation was done with optical pumping techniques (see section III B) and lasted 120 milliseconds. After the preparation, we waited a time $T_{\text{wait}}$ to allow the atoms in the excited state to decay to the ground state before the actual storage experiment. For measurements at the single photon level, two optical paths were necessary: one for the intense pulses used in the optical pumping (strong path) and one for the weak pulses at the single photon level to be stored and retrieved in the crystal (weak path). In the weak path, the light was attenuated to the single photon level with a variable fiber attenuator. The two paths were recombined with a fiber coupled optical switch. The weak output mode was detected using a superconducting single photon detector (SSPD) [54] run in constant voltage mode with an efficiency of $\sim 7\%$ and a dark count rate of $10\pm 5$ Hz. During state preparation the SPD was blocked with a mechanical chopper in order to avoid it from being blinded by the relatively strong preparation pulses. During the storage sequence, the leakage from the bright arm was further blocked with another mechanical chopper in order to prevent noise.

### B. Spectral Tailoring

Both protocols require spectral tailoring of the inhomogeneous absorption profile. This can be done by optically pumping ions from one to another ground state level via the excited state. For this reason the degeneracy of the ground state levels, as it is present in Er$^{3+}$:Y$_2$SiO$_5$, has to be lifted by the application of an external magnetic field making use of the Zeeman effect.

The optical pumping efficiency depends on the ratio between the ground state relaxation time $T_Z$ and the excited state relaxation time $T_1$, and on the branching ratio $\beta$ of the two transitions connecting the ground state levels to the excited state. In erbium doped Y$_2$SiO$_5$, $T_1$ is about 11 milliseconds [19] and $T_Z$ has been measured to reach up to 130 milliseconds for similar experimental conditions [55]. $T_Z$ and $\beta$ are strongly dependent on the direction of the magnetic field with respect to the crystal axes. $T_Z$ also strongly depends on the temperature of the sample. The branching ratio for the optimal $T_Z \approx 130 \text{ ms}$ we found is of order of $\beta=0.1$. This means $\sim 90\%$ of the population de-excites down to the initial state. Under these conditions, the number of optical pumping cycles that can be performed is low, and it is very difficult to efficiently transfer population from one ground state level to the other. The optical pumping efficiency can be enhanced by artificially decreasing $T_1$ using stimulated emission to a short lived auxiliary level that quickly decays to the ground state via a non-radiative decay [22]. Hence, two lasers are used for the optical pumping, a pumping laser at 1536 nanometers, and a stimulation laser at 1545 nanometers.

The experimental cycle is illustrated in figure 2. Each preparation sequence takes 120 milliseconds of optical pumping during which both lasers are sent into the sam-
The frequency of the pump laser is repeatedly swept. In this way a wide pit is burned into the inhomogeneously broadened absorption line at 1536 nanometers. If the light is amplitude modulated using an acousto-optical modulator (AOM) and turned off each time the laser passes a frequency $\omega_{\text{gate}}$, a narrow absorption feature will be left at this frequency (fig. 3b). For preparation of the frequency comb the laser has to be periodically modulated as indicated in figure 3).

C. Storage and retrieval experiments

The time available to perform the actual storage experiments is limited by the Zeeman lifetime of $T_Z \lesssim 130$ milliseconds of the material. Population left in the excited state after preparation will lead to noise from fluorescence during the storage experiments. In order to deplete the excited state, the laser at 1545 nanometers is left on for an additional time $T_{\text{extra}}$ after the AOM of the pump laser has been closed completely (figs. 11 and 16). During the storage part the preparation path is closed and the chopper of the detection path is opened at the same time. At a rate of 200 kHz a sequence of $N$ strongly attenuated pulses is sent into the sample beginning at a time $T_{\text{wait}}$ after the pump pulse. At the beginning of each of these trials a start signal is sent to a time to analog converter (TAC). A detection with the SSPD provides the stop signal. The incident pulses are weak coherent states of light $|\alpha\rangle_L$ with a mean number of photons $\overline{n} = |\alpha|^2$. We determined $\overline{n}$ by tuning the frequency of the laser out of the spectral region of the inhomogeneously broadened absorption line and measuring the number of photons arriving at the SSPD. By measuring the losses between the input of the cryostat and the detector (transmission $\approx 15\%$) and the efficiency of the latter, we could trace back the average number of photons per pulse arriving at the crystal. The results were checked with a photo diode by measuring the intensity of the same pulses without attenuation. Measurements were performed at $T_{\text{wait}} = 86$ ms after state preparation in order to reduce noise from fluorescence as explained in section IV.

D. Noise reduction

An experimental issue arising when input pulses are at the single photon level is the fluorescence from atoms left in the excited state due to an imperfect preparation of the memory. As described above, in order to create the initial narrow absorption line, a population transfer between two ground states (in our case Zeeman states) using optical pumping via the excited state is used. In case of an incomplete transfer there will be population remaining in the excited state after the preparation sequence. If the depletion of this level is slow, which is the case for rare-earth ions, this can lead to a high noise level from fluorescence that will blur the weak echo pulse. This problem is especially important for Erbium doped $Y_2SiO_5$, were the optical relaxation time is very long ($T_1 = 11$ milliseconds [19]). In order to investigate this issue, we performed an experimental characterization of the noise in our system.

For this experiment we performed the state preparation as described above, leaving a single absorption line
at the center of the spectral pit. We then measured the number of counts in the same temporal mode as we would do it for a storage experiment, but with \( n = 0 \), i.e., no photons at the input. We carried out experiments for different powers of the stimulation laser at 1545 nanometers. For all cases, the stimulation laser was left on for a time \( T_{\text{extra}} = 10 \text{ms} \) after the pump laser was switched off. The result is presented in figure 4. One can see that the noise counts diminish with a decay time of 11 milliseconds. This confirms that this noise is coming from fluorescence due to ions left in the excited state. Application of the stimulation laser helps to reduce the noise significantly. However, with the limited stimulation power available, we did not manage to quench it completely. In order to perform experiments at the single photon level, we did not manage to quench it completely. In order to perform experiments at the single photon level it is thus advisable to go to higher waiting times also with stimulation laser applied. A drawback will be, that the memory efficiency goes down since the created absorption structure decays with \( T_Z \), which is only one order of magnitude longer than the excited state lifetime \( T_1 \). One thus has to find a compromise between storage efficiency and noise level. In section IV we show an analysis of the CRIB memory with respect to the efficiency and the signal to noise ratio for different delays. Note that this analysis also holds for the AFC measurements since they have been performed under the same conditions.

IV. CONTROLLED REVERSIBLE INHOMOGENEOUS BROADENING

In this section we report on the CRIB experiments. Each of the weak pulses arrives at the sample within the time of an electrical pulse applied to the electrodes of the sample (see the inset of figure 8), i.e., when the central peak is broadened.

![FIG. 4. (COLOR ONLINE) Noise measurement: Number of counts as a function of the delay after state preparation for different power of the stimulation laser (logarithmic plot). An exponential fit to the data gave a decay time of 11 milliseconds which confirms that the noise is due to ions left in the excited state. Application of the stimulation laser yields a strong reduction of this noise. Integration time was 100 seconds and \( N = 8000 \) for each data point. The power of the stimulation laser was measured before the cryostat.](image)

![FIG. 5. (COLOR ONLINE) Efficiency of the memory and signal to noise ratio as a function of waiting time after state preparation with and without stimulation laser. For these measurements \( N = 2000 \) consecutive pulses with a temporal width of 200 nanoseconds were sent into the sample. The distance between the pulses was 5 microseconds. The number of incident photons per pulse was \( \bar{n} = 27 \). Note that the signal to noise ratio is a linear function of \( \bar{n} \) and the efficiency is independent of the number of input photons in this regime [27]. The gray box labeled MEASUREMENT indicates the time interval in which storage experiments were carried out in the rest of this work.](image)

We first studied the effect of the fluorescence noise. In order to do this, we measured the signal to noise ratio as well as the efficiency of the storage, as a function of the time \( T_{\text{wait}} \) between the memory preparation and the beginning of the storage sequence (see fig. 5). The same measurements were also performed without the stimulation laser applied.

To calculate the signal to noise ratio we considered two time windows in the histograms recorded by the TAC (see fig. 6). The first one (A) contains the echo including the noise floor. The second one (B) has the same number of bins and is placed outside the region where the echo is expected, i.e., it only contains the noise floor. From the number of counts \( N_{A,B} \) in these windows, the signal to noise ratio can be calculated as

\[
\frac{S}{N} = \frac{N_A - N_B}{N_B}. \tag{6}
\]

Due to a better performance of state preparation when the stimulation laser is applied [22], the efficiency of...
the memory is significantly higher compared to the case where it is not applied. At the same time also the noise is reduced, as explained in section III.D. However, we noticed a heat input for higher stimulation power. The limited power we could apply to the sample lead to a lower efficiency of the memory. The use of a cryostat with a higher cooling power might thus lead to better results. Nevertheless, in this work we had to make use of its spontaneous de-excitation in order to lower the noise rate.

Measurements of storage and retrieval of weak pulses with a mean number of photons $\bar{n} < 1$ using this system have been reported in [27]. Fig. 6 shows another example with $\bar{n} = 0.9$. Here the input pulses have a duration of 100 nanoseconds and the initial line is broadened by applying a voltage of 70 volts to the electrodes which corresponds roughly to a broadening of the central peak by a factor of 3 [53]. In that case, the storage efficiency is of the order of 0.1%. Higher storage and retrieval efficiencies have been reported in [26] for smaller broadenings and longer pulse durations.

Remarkably, we observe that the efficiency does not decay as a function of $T_{\text{wait}}$ to the extent expected from the ground state Zeeman lifetime. This can be explained by a non-negligible contribution of long lived holes [55] to the spectrally tailored absorption profile. In order to confirm this we measured the lifetime of a spectral hole created with stimulation laser applied. Besides contributions from the excited state decay ($T_1 = 11\text{ ms}$) and the second ground state Zeeman level decay ($T_Z = 130\text{ ms}$) we observed a third decay time of about 15 minutes (a spectral hole could still be observed 1 hour after optical pumping). The origin as well as its dependence on experimental conditions (such as pump and stimulation power) of the occurrence of these long lived holes so far remain unclear and further investigations have to be carried out. This would be of great interest since the results presented in this work only could be obtained because the persistent hole allowed us to wait ($> 85\text{ ms}$) for the ions to de-excite without losing too much in retrieval efficiency. Another interesting feature of the long-lived holes is that they in principle allow to work at temperatures above the limit due to spin-lattice relaxation between the ground-state Zeeman levels presented in [53].

![FIG. 6. (COLOR ONLINE) CRIB echoes for two different switching times of the electric field. The number of photons per incident pulse was $\bar{n} = 0.9$. Input pulse duration was 100 nanoseconds. The integration time was $2.5 \times 10^5$ seconds. Dark counts of 10 Hertz have been subtracted from the data. The voltage applied on the electrodes was $\pm 70\text{ V}$. The enlargement of the echo with respect to the incident pulse can be explained by a spectral mismatch between pulse and absorption line. A and B indicate the time windows used to calculate the signal to noise ratio (see text).]

![FIG. 7. Efficiency of the CRIB memory as a function of temperature. At temperatures significantly above 3 kelvin there can be no contribution to the tailored absorption profile from population transferred between ground state Zeeman levels [53]. The existence of very long-lived (persistent) holes and the contribution to the absorption profile allows the observation of a CRIB echo at temperatures up to 4.5 Kelvin. The number of incident photons per pulse was $\bar{n} = 10$, integration time for each measurement was 200s. For the positive and the negative electrical pulses a voltage of $\pm 50\text{ V}$ was applied to the electrodes.]

Let us assume an initial artificially broadened line with a width $\Gamma_1$ and an incident light pulse that spectrally matches this line. The temporal width of this light pulse is proportional to the reciprocal of the spectral width...
(t₁ ∝ 1/Γ₁) since it is simply given by the Fourier transform. If we now mirror the broadening by reversing the polarity of the electric field, as described above, δj → −δj (Γ₂ = Γ₁), the echo coming out will have the same spectral and thus also temporal shape as the incident pulse. If we choose the electric fields to be asymmetric, i.e. δj → −αδj, the spectral width of the echo will be changed by a factor α (Γ₂ = αΓ₁) and so the temporal width will change as well. This method was studied in detail by Moiseev and Tittel [28]. It could serve as a tool for bandwidth matching of broadband photons to narrowband quantum memories or to increase bitrates in quantum communication networks. Its feasibility was experimentally demonstrated in a rubidium vapor using a switchable magnetic field gradient [44].

Figure 8 shows the temporal width (full width at half maximum) for six different combinations of the first and second electrical field (U₁ and U₂) used to induce and reverse the broadening. One can clearly see how the echo can be compressed or stretched with respect to the input pulse. Note that for this measurement we were limited by the width of the initial peak causing a fast decay of the efficiency with storage time. Temporally long pulses occur truncated by the envelope given by this decay curve. Nevertheless, we still can observe the expected effect without correcting for the decay.

**FIG. 8.** (COLOR ONLINE) Full width at half maximum (FWHM) of the CRIB echo for three different voltages U₂ applied to the sample after switching the polarity of the electrical field as sketched in the inset. Before switching a voltage of U₁ = 95V (green triangles) and U₁ = 65V (blue circles), respectively, were applied. One can see that using this method, the FWHM of the echo can be changed.

![FWHM vs Voltage](image)

### V. STORAGE OF WEAK COHERENT PULSES USING THE ATOMIC FREQUENCY COMB PROTOCOL

We now describe the measurements of storage and retrieval using the AFC protocol. Using the spectral hole burning technique described in Section III B we created an AFC with by turning off the pump laser 14 times during one frequency sweep. We then sent in weak pulses of light of 100 nanoseconds to be mapped on the crystal.

Figure 9 shows the result of a measurement with an input pulse with a mean number of $\bar{n} = 0.5$ photons. The comb had a finesse of $F ≈ 2.6$, an absorbing background of $d₀ = 1.5 ± 0.3$ and an optical depth of the peaks of the comb of $d = 0.5 ± 0.2$. One can clearly see the first echo at a delay of $\frac{\Delta}{2} = 360$ns. The efficiency for this echo is $\eta₁ = 0.7%$. As expected from equation 4, a second echo occurs at $\frac{\Delta}{2} = 720$ns since here the atomic dipoles get back into phase for a second time. Since the storage time is twice as long and due to the finite width of the peaks [29], this echo is much weaker and almost not visible over the noise level ($\eta₂ ≈ 2 × 10^{-4}$). Note that the finesse of the comb was chosen to reach the maximal efficiency one can expect for the values $d$ and $d₀$ (see Eq. 5). Measurements at different values of finesse confirmed this expectation. Note that other experiments have already demonstrated much higher efficiencies at other wavelengths in different materials [30, 32, 34, 36].

**FIG. 9.** (COLOR ONLINE) AFC echoes at the single photon level. The graph shows a histogram of $8.64 × 10^6$ trials (10h integration time) with a mean photon number of $\bar{n} = 0.5$ per incident pulse. The peak on the left is the transmitted part of the incident photons. On the right one can see the echoes at the first and the second time (360ns and 720ns) the atomic dipoles get back into phase.

In order to show that the coherence of the light is preserved during the storage, we performed an interference experiment using a local oscillator. To observe an interference fringe, we use the fact that the phase of the AFC echo can be controlled with the position in frequency of the AFC. As described in detail in [29], the occurrence and decay of the re-emitted light field is given by the Fourier transform $\hat{\theta}(t)$ of the periodic atomic distribution $\theta(\delta)$. At times $t = m\Delta$ (with $m ∈ \mathbb{N}$) after the absorption an echo will be emitted due to the periodical rephasing of the atomic dipoles. If the central frequency of the AFC is shifted by $\Delta₀$ with respect to the incident light, i.e.
\[ \theta(\delta) \rightarrow \theta(\delta + \Delta_0), \] an additional phase factor occurs in the Fourier transform: \( \exp(-i2\pi\Delta_0 t)\theta(t) \). The echoes will acquire a phase

\[ \phi_m = m2\pi \frac{\Delta_0}{\Delta} = m\phi_1. \tag{7} \]

with respect to the case where \( \Delta_0 = 0 \).

\[ \phi_m = m2\pi \frac{\Delta_0 + \delta\Delta_0}{\Delta} = m(\phi_1 + \delta\phi_1). \tag{8} \]

Assuming a Gaussian distribution with a width \( \sigma \) for the phase noise, the visibility for \( m = 1 \) is given by \[ \frac{9}{10} \] and \[ \frac{9}{10} \approx \frac{9}{10}. \]

In our case \( V_1 = 0.9 \). The value of 0.66 we found experimentally for \( V_2 \) fits well with this explanation.

The results of this experiment confirm that the coherence of the absorbed and re-emitted light is well preserved in an AFC-type light-matter interface. Moreover we see that one can use the AFC itself to shift the phase in a controlled way. This capability provides an interesting tool for interference experiments involving an AFC-storage. One can consider the output of this device as one of the outputs of an unbalanced Mach-Zender interferometer. In this picture the transmitted part of the incident light has taken the short, the re-emitted part the long (phase shifted) arm.

VI. COMBINED AFC-CRIB STORAGE SCHEME

We have here studied two storage schemes having different properties in a two-level configuration. The main advantage of CRIB is the on-demand read-out while for AFC it is the high multimode potential. Yet, the drawback of CRIB is its lower multimode potential whereas for AFC it is the lack of on-demand read-out (predetermined storage time). Note that, in order to turn the AFC into an on-demand memory one can use so-called spin-wave storage \[ \frac{21}{21} \] but this would require a three-level configuration with two ground-state spin levels and an optically excited state.

We here propose a modified scheme where we combine the two methods. The applied inhomogeneous broadening in CRIB can be used as a more general tool for controlling the collective emission rate, as we have shown in \[ \frac{53}{53} \]. In particular one can inhibit the revival of the interference fringes but for the second AFC-echo. In order to obtain a signal clearly above the noise the number of incident photons per pulse was set to \( \tilde{n} \approx 9 \). According to equation \ref{eq:vis} the acquired phase \( \phi_2 \) is twice as large for a fixed \( \Delta_0 \). This corresponds well to what we observe. With \( 66 \pm 3\% \) the visibility is significantly lower than for the first echo. This can be explained by the phase noise induced by the jitter of the laser used as local oscillator. Let us assume that during the time between state preparation and the measurements the laser has shifted by \( \delta\Delta_0 \) due to the jitter. Introducing it into equation \( \ref{eq:vis} \) we get:

\[ V_m = e^{-(m\sigma)^2/2} \]

For \( m > 1 \) the phase noise is \( m \) times larger. Therefore

\[ V_m = e^{-(m\sigma)^2/2} = V_1^{m^2}. \]
To summarize, by combining the two techniques one can create an AFC memory where the emission time can be controlled in discrete steps, a digital quantum memory. This while retaining it’s high multimode potential. Note that experimental results similar to those presented here have been obtained at the University of Calgary [57].

VII. POSSIBLE IMPROVEMENTS

The main issue that occurs when working with an erbium doped crystal as material for a quantum memory lies in the inefficient optical pumping. As described in sections [III and IV] this causes an absorbing background, limits the achievable optical depth of the prepared absorption feature and leads to fluorescence noise. As shown in [22], the pumping efficiency can be further increased by mixing the spins of the excited state using radio-frequency (RF) waves. In this way one improves the effective branching ratio and thus the probability for a population transfer. However, the application of the RF wave causes a heat input into the sample which can lower the lifetime of the ground state [22]. This technical problem can be easily solved with a sufficient cooling power of the cryostat used for the experiments. However, for the measurements presented in this work, such a cryostat was not at our disposal. The limited cooling power also put a bound to the stimulation power we could apply to our sample. A higher stimulation power would reduce the fluorescence noise even more and increase the efficiency of the population transfer at the same time. However, we noticed a measurable heat input by the stimulation laser into the sample at high power. Also here a higher cooling power as well as a better thermalization of the sample would probably allow a higher efficiency and a lower noise level and thus to a much better signal to noise ratio. It is likely that the fluorescence noise has its origin also in decay channels involving other ground state (crystal field) levels than the ones used for the experiments. The signal to noise ratio could thus be further improved by spectral and polarization filtering after the sample.

The most promising feature that played a role are the persistent holes mentioned in [IV]. As already discussed, their origin so far remains unclear. We thus suggest to perform a detailed analysis of their origin. If they are due to another level not being either of the two ground state Zeeman levels used in the experiment, which can be populated in a controlled way, one might use this level as shelving state. A third ground state level with a long life- and coherence time would allow one to extend both protocols with a spin wave storage. This would be the enabling feature for AFC- or CRIB-memory with spin wave storage at telecommunication wavelengths. Additionally we suggest to also explore different host materials [20] and to study the influence of the direction and strength of the external magnetic field in more detail. Storage time, both for the AFC and the CRIB, can be

FIG. 11. (COLOR ONLINE) Here we show control of AFC echoes using the CRIB effect. The dashed line (in blue) is a reference measurement where no CRIB effect was used. The short-dashed line (red) shows how the first echo can be suppressed by applying an electric field gradient to the sample. In practice a small echo can still be seen since we could not apply a large enough broadening with respect to the comb peak separation. Note that since the applied field is not reversed here, also the second echo is suppressed. The solid line (black) show the results if the broadening is reversed at $t = 1/\Delta$, such that the initial coherence is recovered at $t = 2/\Delta$, resulting in a recovery of the second echo. Here $\bar{n} \approx 8$ and dark counts have been subtracted from the data.

The first AFC echo by applying a large enough broadening after the absorption of the input pulse. Let us here remind that the AFC will rephase periodically due to the periodic comb structure. Hence, by reversing the applied inhomogeneous broadening between, for instance, the first and second echo, one can choose to only allow the material to emit the second echo. In Figure 11 we show an example of such a manipulation of AFC echoes using the CRIB technique.

In Figure 11 (and also in Fig. 9) we can also observe that the echo efficiency decreases with the echo number $m$. This can be understood within the AFC theory [29], which says that the comb finesse causes an intrinsic dephasing during the time spent in the memory. In the case where all previous $m - 1$ echoes before the $m$-th echo have been suppressed using the CRIB effect, the efficiency of the $m$-th echo can be calculated using eq. (5), with the last dephasing factor replaced by $e(-m^2/F^2)(\pi/4\ln 2)$. Otherwise energy will be emitted in the previous echoes and the energy emitted in the $m$-th echo is not trivial to calculate. Let’s also note that this result holds if the retrieval efficiency is low, such that a small amount of energy is emitted in each echo (a reasonable assumption in our experiment). The above arguments lead to the conclusion that in order for higher order echoes to be efficient, the comb finesse should be increased. Then this technique can be used to bias the retrieval efficiency towards higher order echoes.
increased by creating a narrower initial spectral feature. This would in turn require a frequency stabilized laser.

VIII. CONCLUSIONS

We have presented experimental results obtained with two different approaches for a light-matter interface at the single photon level. By using an erbium doped doped into an yttrium ortho silicate crystal (Er$^{3+}$:Y$_2$SiO$_5$), we were able to carry out these measurements at telecommunication wavelengths. The main difficulty when working with Er$^{3+}$:Y$_2$SiO$_5$ is the inefficient optical pumping. Fluorescence noise from ions left in the excited state after state preparation is an issue when working at the single photon level in this material. However, we have shown how to minimize this source of noise. We have presented results concerning the possibility of pulse compression and stretching with the CRIB scheme. We have also shown that the presence of persistent holes not only allows one to perform measurements at the single photon level at a reasonable noise level, but also to observe CRIB echoes significantly above the temperature expected from measurements of the Zeeman level lifetime in this material [3]. However, the efficiency of the optical pumping in this material remains low and further research on this is required in order to make the device a useful tool for future experiments in quantum information science.

Furthermore we have presented AFC experiments at the single photon level in the same material. We have demonstrated the preservation of coherence during storage by an interference experiment using a local oscillator. These measurements also show how the phase of the echo can be changed with respect to the input pulse by a frequency shift of the AFC. The preservation of coherence for the second AFC echo was also shown. We observe that its phase is shifted twice as much as the phase of the first echo. The limited visibility of the interference fringes of both measurements could be explained by the phase noise of the local oscillator.

Overall we believe that our experiments show the potential of erbium doped solids for quantum memory applications and we hope that it will motivate some further material research.

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Quantum repeaters based on heralded qubit amplifiers

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Qubit amplifiers were recently proposed to herald the remote entanglement creation. When combined with quantum memories, we show how the qubit amplifier provides on-demand entangled pair source. This is a useful resource in quantum repeaters using entangled photon photon pairs. In this context we show that the remote entanglement between two atomic ensembles can be created and further swapped towards long distances with a "perfect" fidelity, i.e. without vacuum components and multiphoton errors. The resulting quantum repeater thus do not need final postselection and achieves entanglement distribution rates approaching the optimal quantum repeater based atomic ensembles and linear optics.

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I. INTRODUCTION

A. Principle of quantum repeater

When tackling the task of distributing the entanglement over long distances, the essential problem are the channel losses. In order to overcome this, Briegel et al. [1] came with the idea of quantum repeater. This is a scheme which consists of cutting the total distance smaller pieces (called elementary links), distribute the entanglement in each elementary link in which the losses are now smaller, store the entanglement using quantum memories and finally swap the entanglement between adjacent quantum memories in order to distribute the entanglement over the whole distance. The crucial operations of the quantum repeater are: creation, storage and swapping of the entanglement. The creation, however, needs to be heralded.

In order to emphasize the importance of heralding the remote entanglement creation, we give a short counter example. Suppose that we have two distant memories with a probabilistic source of entangled photon pairs (typically a polarization entangled photon pair produced by spontaneous parametric down-conversion) localized half way between the memories. We assume that the photons propagate through lossy channels, typically optical fibres and that they are stored in the memories, but without any announcement whether the storage was successful or not. Next, two adjacent memories, each from another elementary link, can be read and the entanglement can be swapped through the two photon coincidence detection. For such protocol, the weight of the vacuum component (for the case with channel transmission typically small compared to one) is large compared to the entangled state. This is the principal cause of the failure of a potential quantum repeater based on this scheme and implies, that the final post-selection is needed and is, indeed, essential. This would yield the entanglement distribution rates of two orders of magnitude lower (~ 10⁻⁵ Hz) than the originally proposed DLCZ protocol [2].

The heralding of the entanglement creation can be actually realized if we introduce another source (which will be specified) and a measurement device between the two memories. Conditioned on the successful measurement by our device (typically a coincidence detection), we know that the entanglement between the memories has been created. Such a setup is called a qubit amplifier and a detailed description is given in the next subsection.

B. Principle of qubit amplifier

First, let us remind the principle of qubit amplifier [3], shown in Fig. 1. Consider first the state coming from the pair source to be a polarization entangled state and a state coming to a tunable beam splitter BS with reflectivity (in intensity) R to be a product 2-photon state |11⟩. If the modes in and c coming to the Bell measure (BM) are indistinguishable, then the coincidence detection entangles the modes g and out. This is true for general degrees of freedom (with a corresponding measure), but for our purposes we consider the polarization degrees of freedom. The qubit amplifier thus serve as an announced source of successful entanglement distribution. Furthermore, the weight of the entangled state can be tuned with respect to the undesired components, coming from the losses in communication channels (modes in and c) by the beam splitter reflectivity.

We show, how to use the qubit amplifier scheme in order to obtain a near-perfect local source of entangled photon pairs and we show that it approaches the optimal entanglement distribution rates one can expect with repeaters based on linear optics and quantum memories.

The requirement of the perfect source of entanglement is very natural and the fact, that it can be realized locally was already pointed out in the proposal of IPP protocol [4]. However, contrary to the IPP protocol, we show, that due to the use of qubit amplifier, the total density matrix does not contain the vacuum component and the post selection is thus not needed! More detailed description is given in the section II.
II. QUANTUM REPEATER BASED ON LOCAL HERALDED SOURCES OF ENTANGLED PAIRS

A. Entanglement distribution time

Let us remind the basic formula for the entanglement distribution time for the repeater with $n$ nesting levels [5]

$$T_{tot} = \left(\frac{3}{2}\right)^n \frac{L_0}{c} \frac{1}{P_0 P_1 \ldots P_n},$$

where $L_0$ is the length of the elementary link, $P_0$ the probability of successful entanglement creation between the quantum memories in the elementary link and $P_k$ with $k = 1, 2, \ldots$ is the probability of $k$-th swapping.

The legitimate question is what is the best possible scenario one can hope for quantum repeaters based on quantum memories and linear optics elements. A scenario which approaches this limit (in the two photon coincidence regime) consists of a perfect entanglement source (without errors) at each site and with a BM in the middle (since the situation is now symmetric). Consider detection, quantum memory and transmission efficiencies $\eta_D$, $\eta_M$ and $\eta_t$, where $\eta_t = \exp(-L_0/(2L_{\text{opt}}))$ is the transmission efficiency between one site and the BM. The probability of a successful swapping in the elementary link is then $P_k = 1/2(\eta_D \eta_M)^k$, where the factor 1/2 comes from the fact, that we are performing a probabilistic BM. The probabilities of the $k$-th swapping read $P_k = 1/2(\eta_D \eta_M)^k$ for $k = 1, 2, \ldots$. The resulting formula for the entanglement distribution time in this ideal case is then

$$T_{tot,id} = 2 \cdot 3^n \frac{L_0}{c} \frac{1}{(\eta_D \eta_M)^{2n+2} \eta_t}.$$

B. Local entanglement generation

As discussed in introduction, the possibility to realize a near perfect source locally is essential. This can be actually done when taking the two QMs storing modes $g$ and $out$ both sitting at the same site. In this case the transmissions of channels $in$ and $c$ are equal to 1. Following the approach of [Chen], our BM consists of a PBS in the $\pm 45^\circ$ basis followed by PBS in each output (see Fig. 1). This setup automatically eliminates the vacuum in the memories.

In order to have more realistic model, lets consider imperfect sources. With analogy to [5] we consider an SPDC like source which emits the polarization entangled photon pair with probability $p$ and a double pair with probability proportional to $p^2$. The incoming state is thus $\rho_{\text{pair}} = (1 - p) |0\rangle \langle 0| + p/2 |g_{\text{in}} h_{\text{in}} + g_{\text{in}} v_{\text{in}}\rangle \langle h_{\text{in}} | + p^2/8 (|g_{\text{in}} h_{\text{in}} + g_{\text{in}} v_{\text{in}}\rangle \langle h_{\text{in}} |)$. We will denote by $q$ the probability of a photon emission from the single photon source. The emitted state is thus $\rho_{\text{single}} = (1 - q) |0\rangle \langle 0| + q |\text{H}_{\text{in}}\rangle \langle \text{H}_{\text{in}} |$ for the H/V polarization. Lets denote the density matrix after a H-V coincidence detection $\rho^0_s$ (for site or source of entanglement). In general, the density matrix have now the elements containing multiple excitations in the memories. However, one can get rid of some of them by considering some reasonable assumptions, concretely

$$1 \gg R \gg p \quad (3a)$$

$$1 \gg 1 - q. \quad (3b)$$

This corresponds to a photon pair source with a low double pair production and a near perfect single photon sources. As we will see later, these conditions can be further relaxed to some extent. With these assumptions the resulting non-normalized density matrix (up to the unitary transformation) is

$$\rho^0 = \alpha^0_{\text{ent}} + \beta^0_{\text{g+}} + \gamma^0_{\text{out2, g+}} + \delta^0_{\text{out, g+}},$$

where

$$\text{ent} = |g_{\text{out}} h_{\text{in}} + g_{\text{out}} v_{\text{in}}\rangle \langle h_{\text{in}} |, \quad (5a)$$

$$g_{\text{+}} = |g_{\text{in}}\rangle \langle g_{\text{in}}| + |g_{\text{in}}\rangle \langle g_{\text{in}}|, \quad (5b)$$

$$\text{out2} = |\text{out}_{\text{H}}\rangle \langle \text{out}_{\text{H}}|, \quad (5c)$$

$$\text{out}_{\text{+}} = |\text{out}_{\text{H}}\rangle \langle \text{out}_{\text{H}}| + |\text{out}_{\text{V}}\rangle \langle \text{out}_{\text{V}}|, \quad (5d)$$

$$g_{\text{2+}} = |g_{\text{H}}|^2 - |g_{\text{V}}|^2 \langle h_{\text{in}} |. \quad (5e)$$

The coefficients then depend on parameters $\eta_D, R, p, q, (T = 1 - R)$ as follows

$$\alpha^0_{\text{ent}} = \eta_D^2/8RRT \eta_t^2 \quad (6a)$$

$$\beta^0_{\text{g+}} = \eta_D^2/8Rq(1 - q + (1 - \eta_D)Rq) \quad (6b)$$

$$\gamma^0_{\text{out2, g+}} = (\eta_D/8)^2 (pTq)^2 \quad (6c)$$

$$\delta^0_{\text{out, g+}} = (\eta_D/8)^2 p^2 Tq(1 - q). \quad (6d)$$
The swapping in the elementary link is performed between two out memories. The reason is that this configuration cancels the undesired component $g_+$ in Eq. (4) and thus increases the fidelity. The BM now contains first a PBS and then a PBS in ±45° basis in each output of the PBS. The state after the coincidence detection yields the density matrix in the elementary link
\[ \rho_0 = \alpha_0 |\psi_0\rangle \langle \psi_0 | + \beta_0 (g_+ g_2^+ + g_+ g_2^+) + O(g_2^+ g_2^+). \] (7)

It can be shown, that when respecting the considered approximations Eq. (3), the terms containing two excitations in each memory become negligible. We use the primed notation for one of the $g$ sites. From the point of view of the BM, the two sources are equivalent and the BM is thus symmetric (in the middle).

C. Swapping operation

The form of Eq. (4) and Eq. (7) is not the same. We have to calculate explicitly the form after the swapping between the two neighboring elementary links. Let denote the memory modes of the first (second) elementary link by $f$ ($g$). We will swap, say, between the primed modes. The result is
\[ \rho_1 = \alpha_1 |\psi_1\rangle \langle \psi_1 | + \beta_1 (f_+ g_2^+ + g_+ f_2^+) + \gamma_1 f_+ g_+ + O(g_2^+ f_2^+). \] (8)

As before, the term containing two excitations in each memory becomes negligible and from now on, each next swapping yields the density matrix of the form Eq. (8). In order to make some quantitative analysis, we use standard quantities, i.e. fidelity of the final state $F$, source repetition rate $\gamma_{\text{rep}}$ and entanglement distribution time $T_{\text{tot}}$.

Fidelity is defined as usually $F = \langle \psi_0 | \rho_n | \psi_0 \rangle$, where $\rho_n$ is the final density matrix (after $n$-th swapping) and $|\psi_0\rangle = 1/\sqrt{2} (|f_0\rangle + |g_0\rangle)$ is the desired state shared at the end by the two distant quantum memories $f$ and $g$.

The factor $L_0/c$ in Eq. (1) stands for the communication time, where the photon is sent from the source over a distance $L_0/2$, it is then measured in the BM and the result is then communicated back to the source. This is already an approximation of a more general formula, where $L_0/c$ is replaced by $T_0 = T_s + L_0/c$. Here, $T_s$ is the source preparation time and is usually supposed to be much smaller than $L_0/c$. In our case, the probability of the successful preparation of the source is $P_0^s = \text{Tr}(\rho_0^s)$ and thus
\[ T_s = (P_0^s)^{-1}, \] (9)
where $\gamma_{\text{rep}}$ is the repetition rate of the source.

The calculation of the entanglement distribution time, fidelity and the repetition rate of the source is straightforward, treatable analytically, but providing us with rather large analytical expressions. The main results and comparison to the idealized case are the matter of the next subsection.

D. Results

For the numerical simulations, we considered a standard example with the total distance to be entangled to be $L = 1000\text{km}$, memory and detector efficiencies $\eta_M = \eta_D = 0.9$ and a channel (fiber) attenuation length $L_{\text{att}} = 20 \text{ km}$. The maximum allowed nesting level was fixed to $n = 4$. For these parameters, the optimal achievable entanglement distribution time is (from Eq. (2))
\[ T_{\text{tot, id}} \approx 7.134 \text{ s}. \]

For the realistic model we proceeded as follows. First, we assumed the single photon sources as perfect sources ($q = 1$). We then looked in the range of pair source probability $p \in (10^{-4}, 0.2)$ and beam splitter reflectivity $R \in (0.001, 0.4)$. We are interested only in results with final fidelity $F \geq 0.9$. The results are shown in Table I

<table>
<thead>
<tr>
<th>$T_{\text{tot}}$ [s]</th>
<th>$p$ (q)</th>
<th>$R$</th>
<th>$F$</th>
<th>$\gamma_{\text{rep}}$ [MHz]</th>
<th>$\text{optimization}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.242 $10^{-4}$</td>
<td>0.03691 0.978</td>
<td>4426</td>
<td>$\text{optimization}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8.408 0.008096</td>
<td>0.4</td>
<td>0.9</td>
<td>7.603</td>
<td>$\text{optimization}$</td>
<td></td>
</tr>
</tbody>
</table>

TABLE I: Results for the quantum repeater using pair entanglement source at each site. The results were calculated for total distance $L = 1000 \text{ km}$ and four nesting levels ($n = 4$). The table contains the total entanglement distribution time $T_{\text{tot}}$, the quality of the source (if $q$ is fixed in the first column, the found optimal value of $p$ is shown in the third column and vice versa), beam splitter reflectivity $R$, final fidelity $F$ of the shared state (lower bounded by 0.9), repetition rate of the source given by Eq. (9), for which $T_s = L_0/c$ and the chosen optimization (whether $T_{\text{tot}}$ or $\gamma_{\text{rep}}$ was minimized).

From the conceptual point of view, we were interested in the minimal entanglement distribution time one can achieve (optimization of $T_{\text{tot}}$). On the other hand, the repetition rate of the source might be an important practical limitation, thus we also looked for the minimal achievable source repetition rate $\gamma_{\text{rep}}$ and the corresponding $T_{\text{tot}}$ (optimization of $\gamma_{\text{rep}}$). The best result is logically obtained for the lowest $p$, since it produces a state with smallest errors. On the other hand, smaller is the $p$, higher is the source repetition rate. As it can be seen from Table I, a little increase in $T_{\text{tot}}$ permits a significantly lower $\gamma_{\text{rep}}$ (decrease by three orders of magnitude!). Moreover the times $T_{\text{tot}}$ are very close to the optimal value $T_{\text{tot, id}}$.

An obvious remark is, that the used approximations Eq. (3) were not fully respected (e.g. we used values of $R$ up to 0.4 and of $p$ up to 0.2). However, we verified the complete density matrix at each site $\rho_0^s$ and in the elementary link $\rho_0$ and for the results shown in Table I the weight of the neglected components is at least $10^5$ times smaller than of the entangled state and we thus consider the approximation valid. Nevertheless, such a check has to be done every time the condition Eq.(3) is not fully respected.
III. CONCLUSION

* On demand entangled pair source
* Close to one fidelities achievable in principle

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Spin-wave storage using chirped control fields in atomic frequency comb-based quantum memory

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It has been shown that an inhomogeneously broadened optical transition shaped into an atomic frequency comb can store a large number of temporal modes of the electromagnetic field at the single-photon level without the need to increase the optical depth of the storage material. The readout of light modes is made efficient thanks to the rephasing of the optical-wavelength coherence similar to photon-echo-type techniques, and the reemission time is given by the comb structure. For on-demand readout and long storage times, two control fields are used to transfer the optical coherence back and forth into a spin wave. Here, we present a detailed analysis of the spin-wave storage based on chirped adiabatic control fields. In particular, we verify that chirped fields require significantly weaker intensities than π pulses. The price to pay is a reduction of the multimode storage capacity that we quantify for realistic material parameters associated with solids doped with rare-earth-metal ions.

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I. INTRODUCTION

In future quantum networks, quantum information may be exchanged between the network nodes using trains of single photons (i.e., photons prepared in temporally distinguishable modes) similar to today’s telecommunication networks. What we know with certitude is that temporal multiplexing greatly speeds up the distribution rate of entanglement over long distances in quantum networks relying on quantum repeaters [1,2]. This requires quantum memories enabling the storage of many temporal modes. However, the multimode capacity of most quantum memories based on atomic ensembles strongly depends on achievable absorption depths. Currently, only a few modes could efficiently be stored in available atomic ensembles using electromagnetically induced transparency, controlled and reversible inhomogeneous broadening, or Raman-type memories [3]. Time-multiplexed quantum storage could thus be the bottleneck of future quantum networks.

However, tens of temporal modes at the single-photon level have been stored recently in a rare-earth-ions-doped solid with a rather limited optical depth [4]. The key feature of this multimode storage has been presented in Ref. [5] and consists of shaping the inhomogeneous broadening of an optical transition into an atomic frequency comb (AFC). When a photon enters the solid, the comb modes are excited. They dephase, then rephase at a time given by the comb structure leading to a photon-echo-like reemission [6] in a well-defined mode. For both on-demand readout and long storage times, a pair of control pulses is used to transfer the optical coherence back and forth into a spin wave. This storage technique has already motivated a large number of proof-of-principle experiments [4,7–13] because its multimode capacity only depends on the comb structure provided that the optical depth is high enough to store a single mode efficiently. However, the requirements on the control pulses have never been studied in detail. So far, it has been suggested that π pulses could realize the desired population transfer over large frequency bandwidths. Here, we show that this necessitates high Rabi frequencies that are very challenging to achieve in rare-earth-doped materials where the dipole moments are typically 3 orders of magnitude weaker than in the alkali gases usually used in quantum optics experiments. Since adiabatic chirped pulses are commonly used for the coherent control of rare-earth-doped solids [14–18] and are known to need weaker intensities than π pulses [19], we study the possibility to use them in AFC protocols. In particular, we study the advantages of chirped pulses for light storage in solids in detail, and for limited Rabi frequencies, we highlight a trade-off between efficient population transfer and high time multiplexing.

The paper is organized as follows. In Sec. II, we recall the principle of AFC memories. We then present a specific sequence of two chirped pulses that offers an efficient population transfer from the optical transition to the spin transition and preserves the collectivity that is at the heart of the readout process in Sec. III. In Sec. IV, we give explicit examples relevant to rare-earth-doped solids to quantify the intensity gain and the multimode capacity losses when chirped pulses are used instead of conventional π pulses. Section V contains our conclusions and an outlook toward future works.

II. AFC PRINCIPLE

Let us briefly recall the principle of light storage based on AFC. Consider an ensemble of atoms with an optical transition formed by two levels |g⟩ and |e⟩, which is inhomogeneously broadened, see Fig. 1. The |g⟩-|e⟩ transition is spectrally shaped such that the atomic distribution consists of a frequency comb made of narrow peaks with a characteristic width γ, separated by Δ, and spanning a large frequency range Γ. When a signal photon enters the crystal, with a spectral profile centered on the frequency of the |g⟩-|e⟩ transition and with the bandwidth γp satisfying Δ ≪ γp ≪ Γ, it is completely absorbed provided that the atomic density is high enough. After the absorption, the photon is stored in a single atomic excitation delocalized over all the atoms, corresponding to the Dicke-type state [20],

$$\sum_{j=1}^{N} e^{-i\Delta_j t} e^{-i k_j z_j} |g_1 \cdots e_j \cdots g_N\rangle,$$

where N is the number of atoms, Δ_j is the detuning of the jth atom with respect to the laser frequency, k_j is the wave number of the absorbed signal field, and z_j is the position of the atom j. (Note that, in practice, the amplitudes of the
different terms may vary, depending, for example, on the laser profile.) For an AFC with very sharp peaks, the detunings $\Delta_j$ are approximately multiples of $\Delta$ (i.e., $\Delta_j = m_j \Delta$, $m_j \in \mathbb{Z}$). After a time $2\pi/\Delta$, the components $|g_1 \ldots e_j \ldots g_N\rangle$ are in phase and lead to the echo-type reemission of a photon [6] in a well-defined mode, which has the same wave number as the absorbed photon (forward reemission). This is a remarkable feature of collective excitations: For an atomic ensemble that contains sufficiently many atoms, the emission in one mode completely dominates all other modes thanks to a collective interference. This allows for a very efficient retrieval of the stored photon.

The excellent multimode capacity of quantum memory based on AFC can be understood as follows. The shortest duration $\tau$ of one mode that can be efficiently stored in the memory is limited by the frequency bandwidth of the comb (i.e., $\tau \approx 12\pi/\Gamma$) (the prefactor comes from the condition that the overlap of two adjacent temporal modes has to be negligible, see Ref. [5] for a detailed discussion). The total duration of the pulse train is limited by the echo time [i.e., the time it takes for the first mode to be reemitted (i.e., $2\pi/\Delta$)].

The number of modes that can be stored is given by the ratio between the pulse train duration and the duration of one mode [i.e., $\Gamma/(6\Delta)$]. This corresponds to the number of peaks in the comb structure $N_{\text{peak}} = \Gamma/\Delta$. In rare-earth-doped materials, the ratio between inhomogeneous broadening and homogeneous linewidth is inherently large making possible to prepare combs with hundreds of peaks and thus to store many modes without having to increase the absorption depth.

The technique based on AFC described so far only implements a memory with a predetermined storage time. For on-demand readout of the stored electromagnetic field, the single collective excitation (1) has to be transferred from the optical-wavelength transition to a spin transition involving the state $|g\rangle$ and an ancillary state, say the state $|s\rangle$. The spin transition does not have a comb structure so that the temporal evolution of the phase associated with each component of the collective state (1) is frozen. The resulting spin wave is thus described by
\[
\sum_{j=1}^{N} e^{-i\Delta_j (2\pi/\Delta - T_0)} e^{-i(k_j-k_s)\tau_s} |g_1 \ldots e_j \ldots g_N\rangle, \tag{2}
\]
leading to the desired reemission at time $t = 2\pi/\Delta + T_s$.

On-demand readout is thus achieved by controlling the delay $T_s$ between the two control pulses. Note that the use of counterpropagating control fields forces the reemitted light to propagate in the backward direction. This suppresses the potential reabsorption in the forward reemission and leads, in principle, to quantum memories with unit efficiency [5,21]. (This is not necessary if the atomic ensemble is embedded in an asymmetric cavity operated in the impedance-matching condition where unit efficiency can be obtained without reversing the propagation direction of control fields [22].) Furthermore, note that the spin wave allows for long-lived storage, since spin coherence lifetimes are generally longer than the optical coherence lifetimes.

So far, it has been considered that $\pi$ pulses (i.e., resonant monochromatic fields shaped by an envelop with the characteristic duration $\tau$ and associated with the Rabi frequency $\Omega_{\text{max}} = \max \Omega(t)$) such that $\Omega_{\text{max}} \tau \sim \pi$) could realize the desired population transfers. However, for an efficient conversion of the collective excitation from the optical transition to the spin transition, the field envelop has to be short in time such that its Fourier transform is larger than the comb bandwidth $1/\tau_s \gg \Gamma$. Together with the condition that the control pulse is a $\pi$ pulse, this leads to the requirement $\Omega_{\text{max}}^2 \gg \Gamma$. High Rabi frequencies are thus required to ensure efficient on-demand readouts of AFC memories with $\pi$ pulses. As mentioned in Sec. I, the use of control pulses with weaker intensities would greatly simplify the experiments.

Motivated by this last consideration, in what follows, we analyze on-demand readout of AFC-type memories with chirped adiabatic pulses [i.e., slowly varying fields with a time-dependent frequency, which satisfies $\Delta_{\text{max}}^\chi > 1$ and $\Omega_{\text{max}}^\chi \tau_c > 1$ where $2\Delta_{\text{max}}^\chi$ is the frequency range associated with the detunings (cf. later for the exact adiabatic criteria and Eq. (5) for the definition of $\Delta_{\text{max}}^\chi$)]. Intuitively, these chirped fields are attractive with respect to $\pi$ pulses. For chirped pulses with a Fourier transform dominated by the chirp, efficient transfer from the optical transition to the spin transition can be achieved provided $\Delta_{\text{max}}^\chi \gg \Gamma$. This allows one to use longer pulse durations than $\pi$ pulses (i.e., $\tau_c \gg 1/\Gamma$) so that the adiabatic requirements can be fulfilled for weaker Rabi frequencies $\Omega_{\text{max}}^\chi \lesssim \Gamma$. However, the total duration of the input pulses, including the train of pulses to store and one control pulse, is limited by the echo time $2\pi/\Delta$. Consequently, there is thus a trade-off between weak Rabi frequencies and high multimode capacity.
III. AFC PROTOCOL WITH CHIRPED PULSES

In order to use adiabatic chirped pulses in memories based on AFC, we have first to check that they preserve the collectivity [i.e., the phases of each of the components involved in the Dicke state (1)]. This is a priori not obvious, since the adiabatic manipulation of atoms is usually accompanied with nontrivial phase evolutions including, for example, dynamical and geometrical phases [23].

A. Notations

Let us recall the well-known dynamics of a single two-level atom, with levels $s$ and $e$, under a pulsed excitation. Consider that the system starts out in an arbitrary coherent superposition of states $s$ and $e$. It is excited with the Rabi frequency $\Omega_{s,t}(t) = \Omega_{c}^{\text{max}}(t)$ (all $g \in [0,1]$) by a control pulse propagating parallel to the $z$ axis with the wave number $k_c$ and corresponding to the field,

$$E_c(t) = \frac{\hbar}{\mu_{es}} \Omega_{c}(t) \cos \omega_{e} (t) t - k_c z + \phi_c.$$  \hfill (4)

$\mu_{es}$ is the dipole moment of the $|e\rangle$-$|s\rangle$ transition. The frequency of the control pulse is made time dependent to describe the chirp. This leads to a time-dependent detuning with respect to the atomic resonance $\omega_{re}$,

$$\Delta_{j}(t) \equiv \omega_{re}^j(t) - \omega_{e} - \omega_{c,}\Delta_{j} = \Delta_{j}^{\text{max}} f(t) + \Delta_{j} \quad (f \in [-1,1] \forall t).$$  \hfill (5)

Under the rotating-wave approximation, the corresponding Hamiltonian in the basis $|s,e\rangle$ is given by

$$H(t) = \hbar \begin{pmatrix} 0 & -\frac{\Omega_{c}(t)}{2} e^{-i\phi} \\ -\frac{\Omega_{c}(t)}{2} e^{i\phi} & \Delta_{j}(t) \end{pmatrix}.$$  \hfill (6)

The dynamics is fully determined by the propagator $U(t,t_\tau) = \exp(-i \int_{t_\tau}^{t} ds H(s))$.

B. $\pi$ pulses

Let us recall briefly the case of a $\pi$ pulse. Under the assumptions that the laser frequency is time independent [$f(t) = 0$] and the Rabi frequency is larger than the detuning $\Omega_{c}^{\text{max}} > \Delta_j$, the propagator takes the explicit form

$$U_{A}(\tau_c,0) = \begin{pmatrix} \cos \frac{\Delta_{c}}{2} & -i \sin \frac{\Delta_{c}}{2} \\ -i \sin \frac{\Delta_{c}}{2} & \cos \frac{\Delta_{c}}{2} \end{pmatrix},$$  \hfill (7)

where $A = \int_{0}^{\tau_c} ds \Omega_{c}(s) \approx \Omega_{c}^{\text{max}} \tau_c$ is the pulse area. Optimal population transfer is achieved when $A = \pi$, which corresponds to a $\pi$ pulse.

Let us check that a pair of $\pi$ pulses can preserve the collectivity. At time $2\pi/\Delta - T_0$ after the photon absorption, a $\pi$ pulse transfers the atom from $e$ to $s$ and the resulting state is obtained by applying $U_{A=\pi}(\tau_c,0)$ to the state (1) and corresponds to the state (2) up to an irrelevant global phase factor. A second $\pi$ pulse delayed by $T_s$ with respect to the first one, converts the spin wave back into an optical atomic excitation. The state is given by Eq. (3), and it leads to an echo-type reemission at time $2\pi/\Delta + T_s$. This confirms that $\pi$ pulses can be used to control the readout time of AFC memories. However, it highlights the requirement that the Rabi frequencies have to be larger than all the detunings $\Omega_{c}^{\text{max}} > \Delta_j \forall j$ (i.e., larger than the overall comb spectrum).

C. Chirped adiabatic pulses

Let us now focus on chirped adiabatic pulses. The Hamiltonian associated with the interaction between a chirped pulse and one atom now depends on time not only through the Rabi frequency, but also through the detuning [see Eq. (6)]. In the adiabatic regime where $\Omega_{c}(t)$ and $\Delta_j(t)$ are slowly varying in time, one can get an explicit expression for the propagator,

$$U(\tau_c,0) = \begin{pmatrix} c_0 c^\tau_{s} u_{-} + s_0 s_c^\tau_{s} u_{-} - c_0 s_c^\tau_{s} u_{+} & s_0 c^\tau_{s} u_{-} - c_0 s_c^\tau_{s} u_{+} \\ c_0 s^\tau_{s} u_{-} - c_0 s_c^\tau_{s} u_{+} \end{pmatrix}. \hfill (8)

$$c_0, s_0$ \begin{tabular}{l}$c_{s}$ $s_c$ \end{tabular}$ stand for cos $\theta(t)$ and sin $\theta(t)$ evaluated at initial ($t = 0$) and final ($t = \tau_c$) time where $\theta$ is defined as $\tan 2\theta(t) = -\Omega_{c}(t)/\Delta_j(t)$. $u_{\pm} = \exp \left[-i \int_{\tau_c,0}^{\tau} (\Delta_j(s) \pm \sqrt{\Delta_j(s)^2 + \Delta_{j}(s)^2}) ds \right]$. If the detuning starts with a negative value and ends up to be positive, the propagator reduces to

$$U(\tau_c,0) = \begin{pmatrix} 0 & -u_{-} \\ u_{-} & 0 \end{pmatrix}, \hfill (9)$$

and describes a complete atomic transfer.

Let us check that the collectivity is preserved when the conversion of the optical excitation into a spin-wave excitation is realized with chirped pulses. We focus on the case where the two control pulses have the same chirp. The reason can be understood intuitively as follows: Consider that the first control pulse has a chirp going from $\Delta_{j}^{\max}$ to $\Delta_{j}^{\max} - \Delta j$. An atom with a negative detuning will be transferred on the spin state sooner than an atom with a positive detuning. However, the latter is brought back to the excited state before the atom associated with a negative detuning if the chirp of the second control pulse ramps also from $-\Delta_{j}^{\max}$ to $\Delta_{j}^{\max}$. If the two control fields have exactly the same chirp shape, atoms associated with different detunings will spend the same time in the excited state during the duration $2\pi/\Delta - T_s$ leading to a high collectivity provided that the dynamical and geometrical phases canceled out. Following the scenario for the $\pi$ pulses, we start with the state (1). (Note that the characteristic duration of the chirped pulses is no more negligible with respect to the storage time in opposition to the $\pi$ pulses). After $2\pi/\Delta - T_0$, two identical chirped pulses delayed by $T_s$ interact with the atomic ensemble leading to the state,

$$\sum_{j=1}^{N} e^{-i \Delta_{j}(T_s - T_0)} U(\tau_c,0) U(\tau_c,0) |g_1 \cdots e_j \cdots g_N\rangle = \sum_{j=1}^{N} e^{-i \Delta_{j}(T_s - T_0)} U(t_\tau,0) U(t_\tau,0) |g_1 \cdots e_j \cdots g_N\rangle$$

$$= \sum_{j=1}^{N} e^{-i \Delta_{j}(T_s - T_0)} U(t_\tau,0) U(t_\tau,0) |g_1 \cdots e_j \cdots g_N\rangle$$

$$= \sum_{j=1}^{N} e^{-i \Delta_{j}(T_s - T_0)} U(t_\tau,0) U(t_\tau,0) |g_1 \cdots e_j \cdots g_N\rangle$$

$$= \sum_{j=1}^{N} e^{-i \Delta_{j}(T_s - T_0)} U(t_\tau,0) U(t_\tau,0) |g_1 \cdots e_j \cdots g_N\rangle.$$
Omitting the global phase factor, which is identical for each atom, one clearly sees that the collectivity is completely restored at time $t = 2\pi/\Delta + T$. Efficient readout of AFC memories can thus be achieved with chirped pulses provided that they are adiabatic and that they have the same chirp.

**IV. NUMERICAL EXAMPLES**

**A. Population transfer with chirped adiabatic pulses**

We now consider concrete examples for atomic inversion with amplitude and frequency modulated control fields. We choose the model proposed in the 1970s by Allen and Eberly [24], which involves control pulses with a hyperbolic secant temporal shape and a hyperbolic tangent chirp. This choice is motivated both by existing experiments in rare-earth-doped solids [17] and by the simplicity of the adiabatic criteria. Indeed, for

$$\Omega_c(t) = \Omega_c^{\text{max}} \text{sech}(t/\tau_c),$$  \hspace{2cm} (10)

$$\Delta_j(t) = \Delta_j^{\text{max}} \tanh(t/\tau_c) + \Delta_j,$$  \hspace{2cm} (11)

atomic inversion over the frequency range $\Gamma$ with finite efficiency $\eta$ is achieved provided [25]

$$2\Delta_j^{\text{max}} \sim \Gamma,$$  \hspace{2cm} (12a)

$$\Delta_j^{\text{max}} \tau_c \gtrsim 2,$$  \hspace{2cm} (12b)

$$\Omega_c^{\text{max}} \sim \Delta_j^{\text{max}} \left(1 - \frac{\log(1 - \eta)}{\pi \Delta_j^{\text{max}} \tau_c} + 1\right)^2.$$  \hspace{2cm} (12c)

The first equation guarantees that the pulse frequencies overlap all the atomic spectra. The second and third inequalities ensure that the chirp and the Rabi frequency vary slowly in time. These equations offer a systematic method to realize an efficient population transfer over the set of atomic spectral components $\Gamma$:

(i) First, one has to choose the chirp $\Delta_j^{\text{max}}$ so that $2\Delta_j^{\text{max}} \sim \Gamma$.

(ii) Then, the pulse duration $\tau_c$ has to be at least equal to $4/\Gamma$. The longer the pulse duration, the better the selectivity of the population transfer. In the limit of very long control pulse duration $\tau_c \gg 4/\Gamma$, the transfer follows a square function centered at $\Delta_j = 0$ and with full width half maximum (FWHM) spectral bandwidth $\Gamma$. We recall that the total duration of the input pulses, including the train of signal modes and the control pulse is limited by the echo time $2\pi/\Delta$. More precisely, we will see in the following that $\tau_c$ is limited by $(2\pi/\Delta - 12\pi/\Gamma)/\gamma$.

(iii) Finally, the transfer efficiency $\eta$ (for the resonant frequency $\Delta_j = 0$) is given by Eq. (12c) and depends on the achievable Rabi frequency. This last equation shows that the use of long pulse durations decreases the required Rabi frequencies. The price to pay is a reduction of the number of modes that can be stored in the AFC memory, since the total duration of input fields is limited by $2\pi/\Delta$. Hence, there is a trade-off on the control pulse duration $\tau_c$: On-demand readouts with weak control fields favor $\tau_c \rightarrow (2\pi/\Delta - 12\pi/\Gamma)/\gamma$, whereas a high multimode capacity favors $\tau_c \rightarrow 4/\Gamma$.

It is instructive to compare the population transfer with chirped pulses and with $\pi$ pulses. A spin transfer over $\Gamma$ with efficiency $\eta$ (minimum efficiency for each frequency) using $\pi$ pulses with hyperbolic secant temporal shape requires [25]

$$\Omega_c^{\text{max}} \sim \frac{\pi}{4} \frac{\Gamma}{\arctan(\eta)}.$$  \hspace{2cm} (13)

To efficiently transfer atoms spanning a given frequency domain $\Gamma$ using a $\pi$ pulse, a minimum Rabi frequency is required (see Eq. (13)). This leads to a maximum temporal duration (since $\Omega_c^{\text{max}} \tau_c$ is fixed), which can be compared to the minimum duration of a chirped pulse ($\tau_c \sim 4/\Gamma$). Furthermore, the minimum duration of a chirped pulse maximizes its Rabi frequency for fixed $\Gamma$ and $\eta$. Let us take an example. To achieve a population inversion with $\eta = 95$% efficiency, hyperbolic secant $\pi$ pulses are at least 8 times shorter but need to be 16 times more intense than chirped pulses corresponding to the Allen and Eberly model, independent of the frequency range $\Gamma$. Note that this comparison is approximative, since it does not take the transfer details into account [the formula (13) corresponds to the minimum transfer efficiency, while Eq. (12c) gives the transfer efficiency for the resonant frequency]. For more accuracy, we performed numerical simulations of a classical light storage using the AFC technique.

**B. Numerical simulations**

For numerical simulations, we choose an atomic comb inspired by recent experiments in praseodymium-doped solids [8]. We take an AFC composed of Gaussian peaks with a width $\gamma = 2\pi \times 25$ kHz. The optical depth per peak is taken equal to $\alpha L = 4$. This leads to an optimal finesse close to 4 [5], corresponding to a peak separation of $\Delta = 2\pi \times 100$ kHz. The comb is composed of $N_{\text{peak}} = 40$ peaks and thus spans a frequency range $\Gamma = 40 \times \Delta = 2\pi \times 4$ MHz. The storage efficiency is $\eta_{\text{echo}} = 25\%$ (forward readout without control fields [5]). This comb allows to absorb and retrieve roughly seven Gaussian temporal modes with a duration $12\pi/\Delta = 1.5 \mu s$ each and with negligible overlaps [1] within the time interval $2\pi/\Delta = 10 \mu s$. Furthermore, to control the multimode storage capacity, the control pulses are gated with a square function parametrized by its FWHM $T_{\text{cut}}$. For truncated hyperbolic secant pulses, choosing $T_{\text{cut}} \sim 7\tau_c$ is sufficient for the nonadiabatic corrections to be negligible. We chose either a cutoff $T_{\text{cut}} \sim 7\times 4/\Gamma = 1.2 \mu s$ corresponding to the minimal cutoff that fulfilled the adiabatic condition (12b) and reducing the multimode capacity by one mode or $T_{\text{cut}} \sim (2\pi/\Delta - 12\pi/\Gamma) = 8.8 \mu s$, the longer cutoff allowing to store a single mode.

The storage efficiency $\eta_{\text{tot}}$ is defined as the ratio of the echo intensity over the input intensity. It is the product of the absorption and reemission efficiency $\eta_{\text{echo}}$ with the transfer efficiency from the optical atomic excitation to the spin-wave squared $\eta^2$. Since one wants to characterize the transfer efficiency back and forth $\eta^2$, we divide the overall storage efficiency $\eta_{\text{tot}}$ (with the spin-wave transfer) by the echo efficiency without control fields $\eta_{\text{echo}}$. Figure 2 shows $\eta^2$ for $\pi$ pulses and for chirped adiabatic pulses with $\Delta_j^{\text{max}} \tau_c = 2$ ($T_{\text{cut}} = 1.2 \mu s$) and $\Delta_j^{\text{max}} \tau_c = 15.7$ ($T_{\text{cut}} = 8.8 \mu s$). One sees that both techniques can realize close to unit transfer efficiency. However, the adiabatic pulses require Rabi frequencies two to
FIG. 2. (Color online) Transfer efficiency with \( \pi \) (dotted blue line) and with chirped (red lines) pulses. The dashed line corresponds to chirped pulses of minimal temporal durations (satisfying the adiabatic criteria) for which the multimode capacity decreases by one mode. The full line is associated with chirped pulses of maximal temporal durations such that only one mode can be stored. The gain, in terms of the required intensity, depends both on the desired efficiency and on the number of modes that can be sacrificed. The circles and crosses are the values obtained using the relation (12c). One can see very good agreement with the numerical simulations.

Adiabatic methods are known to offer several advantages in practice over \( \pi \)-pulse-based techniques. As detailed before, \( \pi \) pulses are harder to implement due to laser power limitations. Furthermore, slowly varying chirped pulses allow for a population transfer with the same efficiency over a large frequency range leading to high storage fidelities. Figure 3 shows the overlap between the input signal field and the output field as a function of the Rabi frequency. This permits one to see that only chirped pulses preserve the shape of the stored field for weak Rabi frequencies. This is essential in many applications involving interference effects where the retrieved photon needs to be indistinguishable from the input signal photon. Note also that the high selectivity of chirped pulses avoids unwanted transitions to nearby levels. Finally, let us recall that population inversion with \( \pi \) pulses is not robust (e.g., with respect to Rabi frequency variations). For example, a small variation \( \epsilon \) in the intensity of one of the control fields induces an error on the transfer efficiency \( \eta^2 \) on the order of \( \epsilon^2/2 \) using \( \pi \) pulses, while the transfer efficiency with chirped pulses remain unchanged provided that the adiabatic criteria are satisfied.

**V. CONCLUSION**

We showed how adiabatic methods can be used for on-demand readout of AFC memories. In particular, for weak Rabi frequencies, we highlighted a trade-off between an efficient readout and a high multimode capacity. To get both an efficient readout and a multimode storage with achievable Rabi frequencies, the creation of a periodic narrow structure is crucial. In its ideal version, an AFC memory would be made of individual peaks as narrow as possible. Note that the width of individual peaks is limited ultimately by the homogeneous linewidth, which can be of a few kilohertz in rare-earth-doped solids. The peak separation is chosen to optimize the storage efficiency, which can be very high provided that the absorption per peak is high enough. The multimode capacity is then mainly determined by the achievable Rabi frequency. For Rabi frequencies on the order of megahertz, hundreds of modes could be efficiently stored in rare-earth-doped solids.

For concreteness, consider a detailed example based on europium-doped \( Y_2SiO_5 \). Europium absorbs at 580 nm and has the appropriate \( \lambda \)-energy structure. Since its homogeneous linewidth is very narrow (2\( \pi \times 122 \) Hz), one could realistically create a comb with \( \gamma = 2 \pi \times 2 \) kHz and \( \Delta = 2 \pi \times 20 \) kHz over the spectral range \( \Gamma = 2 \pi \times 12 \) MHz (i.e., a comb with \( N_{\text{peak}} = 600 \) peaks). The absorption coefficient is about 3 to 4 cm\(^{-1} \) [26] so that if an optical depth per peak of 40 is reached, the resulting echo efficiency is \( \eta_{\text{echo}} = 90\% \) (backward readout). The storage of a single mode with a spin storage based on chirped pulses with the overall efficiency \( \eta_{\text{echo}} \eta^2 = 80\% \) requires \( \Omega_c^{\text{max}} = 2 \pi \times 0.5 \) MHz. For
\[ \Omega_{\text{c}}^{\text{max}} = 2\pi \times 1 \text{ MHz}, \]
75 modes could be stored with the same efficiency. To fully profit from the multimode capacity (i.e., to store nearly 100 modes), \[ \Omega_{\text{c}}^{\text{max}} = 2\pi \times 5 \text{ MHz} \] is necessary.

In this paper, we focused on AFC memories assisted by chirped adiabatic pulses corresponding to the Allen and Eberly model to keep the experimental implementation simple. However, it would be of interest to consider alternative pulse shapes. For example, it has been shown, in the frame of nuclear magnetic resonance, that the use of more sophisticated pulses requires Rabi frequencies up to two times smaller than the Allen and Eberly pulses [27]. Looking further ahead, it is an interesting question whether chirped pulses are optimal with respect to both the laser intensity and the multimode capacity.

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Impossibility of faithfully storing single photons with the three-pulse photon echo

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The three-pulse photon echo is a well-known technique to store intense light pulses in an inhomogeneously broadened atomic ensemble. This protocol is attractive because it is relatively simple and it is well suited for the storage of multiple temporal modes. Furthermore, it offers very long storage times, greater than the phase relaxation time. Here, we consider the three-pulse photon echo in both two- and three-level systems as a potential technique for the storage of light at the single-photon level. By explicit calculations, we show that the ratio between the echo signal corresponding to a single-photon input and the noise is smaller than one. This severely limits the achievable fidelity of the quantum state storage, making the three-pulse photon echo unsuitable for single-photon quantum memory.

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I. INTRODUCTION

The distribution of entanglement over long distances is at the heart of future quantum networks. It may rely on quantum repeaters, which require photonic quantum memories enabling the storage of a large number of temporal modes for very long times [1]. Light storage based on photon-echo techniques in inhomogeneously broadened atomic ensembles has been widely studied in the classical regime [2]. These techniques are naturally suited to store intense light pulses in multiple temporal modes. Storage and retrieval of up to 1760-pulse sequences has been demonstrated [3]. Furthermore, when the photon echo is generated with a specific sequence of three pulses, the so-called three-pulse photon echo, it further offers very long storage times, greater than the phase relaxation time, limited by the population relaxation time only [4]. This technique could thus be very attractive, a priori, in the context of quantum repeaters, and it is natural to wonder whether it can be extended to the storage of single photons.

It has been shown already that the simplest form of the photon echo, namely the two-pulse photon echo, is not suited for quantum storage [5]. However, a recent paper [6] might let us believe, at first sight, that the photon echo in its three-pulse version could be used for the storage of light at the single-photon level. We thus analyze this last technique in detail and we show in this paper that it does not preserve the fidelity of the stored quantum state. The main reason is that the rephasing process, induced by intense pulses and leading to the collective emission of an echo, transfers a large number of atoms to an excited state. The excitation of the medium is followed by spontaneous emission of many photons in random spectral, temporal, spatial, and polarization modes, including those in which the stored photon may originally have been prepared. This makes complete filtering of fluorescence impossible, and it strongly degrades the fidelity of the echo signal associated with a few-photon input. Furthermore, the rephasing process is only partial, which intrinsically limits the storage efficiency. These fundamental limitations, which also apply to the protocol of Ref. [6], make it impossible to implement a high-fidelity and high-efficiency quantum memory based on the three-pulse photon echo in the few-photon regime.

To evaluate the response of an inhomogeneously broadened atomic ensemble to a sequence of many resonant pulses, we follow the semiclassical approach that originated in Refs. [7,8]. First, we focus on the temporal evolution of a single atom. Then, we sum over all individual responses to get (i) the macroscopic polarization of the sample and thus the intensity of the echo and (ii) the average population on the excited state and thus the noise. We then deduce the signal-to-noise ratio for the specific case of single-photon storage. This approach also allows us to bound the storage efficiency. However, its does not give any information about additional limitations caused by the propagation of the light fields, which could be obtained by solving the Maxwell-Bloch equations. Yet, it is interesting to note that a model as simple as the one based on the dynamics of a single atom clearly shows the impossibility of faithfully storing individual photons with the three-pulse photon echo. A complete comparison between this semiclassical approach and the quantum theory will be treated elsewhere [9].

In the next section, we present our analysis of the three-pulse photon echo in two-level systems. In Sec. III, we extend our study to three-level systems. The last section is devoted to our conclusion.

II. TWO-LEVEL SYSTEMS

We start with an ensemble of two-level systems with a ground state |1⟩ and an excited state |2⟩ as depicted in Fig. 1. We suppose that, initially, all N atoms are prepared in the state |1⟩. We first look at the temporal evolution of a single atom which interacts with several laser pulses.

A. Response of a single atom

Let us recall the expression of propagators associated with a two-level atom under a pulsed excitation detuned from the resonance by Δ and with the Rabi frequency Ωs(t). If the light
pulses are used to halt and to reverse the dephasing due to the amplitudes at $t_{i/Delta1}$. Since the atom is initially prepared in the ground state, we have respectively. Since the atom is initially prepared in the ground state, we have respectively.

The spectral-band-matching condition required that is, by a mixture of $N\epsilon$. At time $t_4$ the atom is finally described by

$$U^\Delta(t_4, t_3)U^{\theta_{b}}(t_3)|1\rangle = \cos (\theta_{3}/2)|1\rangle - i \sin (\theta_{3}/2)e^{-i\Delta(t_4-t_3)}|2\rangle$$

with weights $|a(t_2)|^2$ and $|b(t_2)|^2$, respectively.

**B. Intensity of the echo signal**

Consider the case where the first pulse corresponds to a few-photon pulse $\theta_1 \approx 2\epsilon$. In this case, the number of atoms transferred into the state $|2\rangle$ is given by $N \sin (\theta_{1}/2)^2 \approx N\epsilon^2$, which is, in the single-photon case, equal to 1. When absorbed by the sample, this few-photon pulse induces a weak macroscopic polarization

$$P(t_1) = \int_{-\infty}^{\infty} d\Delta g(\Delta) \varphi(1|\rho(t_1)|2)$$

$$= \int_{-\infty}^{\infty} d\Delta g(\Delta) \varphi a(t_1)b(t_1)^* = i \varphi N\epsilon, \quad (6)$$

where $\varphi$ is the electric dipole moment and $g(\Delta)$ is the atomic spectral distribution, which satisfies $\int_{-\infty}^{\infty} d\Delta g(\Delta) = N$. The light pulses in our model, in particular the pulse one wants to store, have spectral widths that are much larger than the width of the atomic distribution $g(\Delta)$, which we assume to have a Gaussian shape with the characteristic width $\Delta_0$. The spectral-band-matching condition required for efficient absorption is thus not met. However, one can still bound the readout efficiency by comparing the initial polarization (6) and the one at the echo time (cf. the following).

Within the context of a three-pulse photon echo, the rephasing pulses that maximize the echo intensity are $\pi/2$.
pulses (i.e., $\theta_2 = \theta_3 = \pi/2$). The electric dipoles radiate in phase at time $t_3 = t_1 + t_2 - t_1$, leading to a macroscopic polarization given by

$$P(t_3 + t_2 - t_1) = \int_{-\infty}^{\infty} d\Delta g(\Delta) \rho(1|\rho(t_4 = t_3 + t_2 - t_1)|2)$$

$$= -\int_{-\infty}^{\infty} d\Delta g(\Delta) \frac{i\phi}{2} (1 + e^{i2\Delta(t_2 - t_1)})$$

$$\to \frac{i\phi N\epsilon}{2} \text{ for } (t_2 - t_1) \gg 1/\Delta_0. \quad (7)$$

Hence, the initial polarization [see Eq. (6)] is not completely restituted. The efficiency of the storage protocol is thus intrinsically limited. Indeed, the polarization (7) of the atomic ensemble serves as a source for the collective emission of an echo that has an intensity given by

$$I_{\text{echo}} = I_0 \left| \int_{-\infty}^{\infty} d\Delta g(\Delta) \rho(1|\rho(t_4 = t_3 + t_2 - t_1)|2) \right|^2$$

$$= \frac{1}{4} N^2 \epsilon^2 I_0. \quad (8)$$

where $I_0$ is the radiation intensity from a single isolated atom prepared in the excited state in the spatiotemporal mode associated with the echo. Note that this mode is completely defined by the combination of excitation pulses. The quantity $I_{\text{echo}}$ is to be compared with the intensity related to the absorbed part of the input pulse, which can be deduced from the initial polarization using

$$I_1 = I_0 \left| \int_{-\infty}^{\infty} d\Delta g(\Delta) a(t_4) b(t_4)^* \right|^2 = N^2 \epsilon^2 I_0. \quad (9)$$

The readout efficiency $I_{\text{echo}}/I_1$ (and thus the overall storage efficiency) is upper bounded by 1/4. Note that other detrimental effects due to the propagation within the atomic ensemble (free-induction decay resulting in the propagation of intense light pulses in the atomic medium or the finite value of the optical depth) further limit the efficiency [5]. Let us now compare $I_{\text{echo}}$ with the incoherent emission from the same sample.

### C. Fluorescence

To determine the amount of fluorescence (i.e., the spontaneous emission), we evaluate the population in the excited state at the echo time $t_3 + t_2 - t_1$ without having exposed the atoms to the first excitation (i.e. for $\epsilon = 0$). We easily obtain that $\langle 2|\rho(t_3 + t_2 - t_1)|2 \rangle = 1/2$, meaning that the intensity associated with the spontaneous fluorescence is given by

$$I_{\text{noise}} = \frac{1}{2} N I_0. \quad (10)$$

Equations (8) and (9) remind us that the incoherent radiation as emanating from an ensemble of dipoles that are oscillating with random phases has an intensity proportional to $N$ whereas the collective emission coming from an ensemble of dipoles in phase gives rise to an echo with an intensity proportional to $N^2$ [10].

The ratio between the echo signal corresponding to a weak input and the noise is given by

$$\frac{I_{\text{echo}}}{I_{\text{noise}}} = \frac{1}{2} N \epsilon^2, \quad (10)$$

which reduces to 1/2 in the particular case of a single-photon input where $N \epsilon^2 = 1$. This calculation shows that, due to the spontaneous emission that severely limits the achievable fidelity of the quantum state storage, the conventional three-pulse photon echo cannot be used to implement a quantum memory in two-level systems. Indeed, the fidelity for storage and recall of a time-bin qubit, that is, a single photon in a superposition of two different emission times, $F = \text{tr}(\rho_{\text{in}} \rho_{\text{out}})$ with $\rho_{\text{out}} = (2F - 1)\rho_{\text{in}} + (1 - F) 1$, is limited to $1 - F = I_{\text{noise}}/(I_{\text{echo}} + 2I_{\text{noise}})$, that is, $F_{\text{2-level}} = 3/5 = 0.6$. The value for the fidelity is smaller than the limit $F_{\text{classical}} = 2/3$ imposed by optimum classical storage [11, 12]. Moreover, for storage times shorter than the phase relaxation time where the mixed-state description is removed, formulas (8), (9), and (10) still hold [in the limit where $(t_3 - t_2) > (t_5 - t_1) > 1/\Delta_0$], meaning that independently of the storage time, the fidelity and the efficiency of the three-pulse photon echo are fundamentally limited at the single-photon level.

Note that this calculation concerns a sample of $N$ atoms which is small compared with $\lambda^3$. For larger samples (i.e., for sample sizes large compared with $\lambda^3$), the spatial dependence of the electromagnetic fields must be taken into account. In this case, the polarization of the atomic ensemble at time $t_4 = t_3 + t_2 - t_1$ for the specific mode associated with the wave vector $\mathbf{k}$ is found to be

$$P(t_3 + t_2 - t_1, \mathbf{k})$$

$$= -\int_{-\infty}^{\infty} d\Gamma d\mathbf{r} \frac{i\phi}{2} (e^{i(k \cdot \mathbf{r} - \omega \tau)} + e^{i(k \cdot \mathbf{r} + \omega \tau)} e^{i2\Delta(t_2 - t_1)}),$$

where $\mathbf{k}_i, i = [1, 3]$, refers the $i$th pulse and the atomic density now depends on the spatial coordinate $\mathbf{r}$. The exponential terms average to zero unless the phase-matching condition is fulfilled: $\mathbf{k} \approx -\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3$. When the sample is small compared with $\lambda^3$, the collective emission is isotropic and the signal-to-noise ratio (10) holds for any direction. For sample sizes larger than $\lambda^3$, the collective emission is highly directional. For the specific direction associated with the wave vector $\mathbf{s}$ satisfying the phase-matching condition, the echo intensity is related to the spontaneous radiation intensity $I_0(\mathbf{k})$ from a single atom by $I_{\text{echo}}(\mathbf{s}) = \frac{1}{2} N^2 \epsilon^2 I_0(\mathbf{k})$. The noise intensity is given by $I_{\text{noise}}(\mathbf{s}) = \frac{1}{2} N I_0(\mathbf{k})$ and, as before, the signal-to-noise ratio reduces to 1/2 in the case of a single-photon storage. For another direction, the signal field is obtained by squaring each individual dipole and then summing. This leads to a much worse signal-to-noise ratio of order $\epsilon^2$. The conclusion is that a spatial filtering cannot be used to circumvent the problem of noise. Moreover, the simpler calculation where the spatial dependence of the fields is neglected, gives an upper bound for the signal-to-noise ratio. Now, we extend our analysis to three-level systems, omitting the spatial dependence of the light.
III. THREE-LEVEL SYSTEMS

A recent paper [6] suggests that the extension of the three-pulse photon echo to systems with additional ground states would allow us to implement a quantum memory. In the following we apply the approach just described to quantify the fluorescence in three-level systems and we show that, even in these systems, the photon echo is inappropriate for the storage of single photons.

The basic scheme, motivated by the analysis of the proposal of Ref. [6], is as follows. We start with an ensemble of three-level systems with an excited state \([2]\) and two ground states \([1]\) and \([3]\). We consider that the optical transition \([1]\)–\([2]\) is homogeneous and that the spin transition \([1]\)–\([3]\) exhibits an inhomogeneous broadening (see Fig. 3). As before, we first focus on the dynamics of a single atom interacting with a sequence of resonant pulses. To evaluate the response of the atomic ensemble, we sum over all individual responses to quantify the intensity of the echo and the fluorescence.

A. Response of a single atom

Consider a three-level atom under a pulsed Raman excitation corresponding to the effective Rabi frequency \(\Omega'_1(t) = \sqrt{\Omega_{1}'(t)^2 + \Omega_{2}'(t)^2}\), where \(\Omega_1(t)\) \([\Omega_2'(t)]\) is associated with the transition \([1]\)–\([2]\) \([\{2]\)–\([3]\)] and detuned from the two-photon resonance by \(\Delta_r\). Further suppose that the Rabi frequencies satisfy \(\Omega_1^\text{max} \approx \Omega_2'^\text{max}\) and that the pulse durations \(\tau\) are short enough so that \(\tau \ll 1/\Delta_0\), as before. In the basis \([1], [2], [3]\), the propagator is given by

\[
U_{\Omega_1'}(\tau) = \begin{pmatrix}
\frac{1}{\sqrt{2}} (1 + \cos \frac{\theta_1'}{2}) & i \sqrt{\frac{1}{4}} \sin \frac{\theta_1'}{2} & \frac{1}{\sqrt{2}} \left(1 - \cos \frac{\theta_1'}{2}\right) \\
\frac{1}{\sqrt{2}} \sin \frac{\theta_1'}{2} & \cos \frac{\theta_1'}{2} & \frac{1}{\sqrt{2}} \sin \frac{\theta_1'}{2} \\
\frac{1}{\sqrt{2}} (1 - \cos \frac{\theta_1'}{2}) & -i \sqrt{\frac{1}{4}} \sin \frac{\theta_1'}{2} & \frac{1}{\sqrt{2}} \left(1 + \cos \frac{\theta_1'}{2}\right)
\end{pmatrix}
\]

with \(\theta_1' = \Omega_1'^\text{max} \tau\). If the laser pulses are off, the propagator reduces to

\[
U_{\Delta_r}(t_f, t_i) = \begin{pmatrix}
1 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & e^{-i \Delta(t_f - t_i)}
\end{pmatrix}
\]

(11)

![FIG. 3. Basic scheme of three-level systems with an excited state \([2]\) and two ground states \([1]\) and \([3]\) that exhibits an inhomogeneous broadening on the spin transition. As in the two-level scheme, the atomic spectral distribution is supposed to be Gaussian with characteristic width \(\Delta_0\). The interaction of the atomic ensemble with a Raman laser pulse is parametrized by the effective Rabi frequency \(\Omega'_1 = \sqrt{\Omega_{1}'^2 + \Omega_{2}'^2}\) and by the two-photon detuning \(\Delta_r\).](image)

A possible procedure to transfer properly a weak-light excitation into a spin coherence requires first the preparation of atoms in one of the two ground states, say, the state \([1]\). The weak pulse that one wants to store is resonant with the transition \([1]\)–\([2]\) and generates a weak coherence \(\epsilon\) between the states \([1]\) and \([2]\) (see Fig. 4). By applying immediately a \(\pi\) pulse on the transition \([2]\)–\([3]\), the optical coherence is transferred to a coherence between the state \([1]\) and the state \([3]\). At time \(t_1\) after this sequence of pulses, the probability amplitudes \(a, b, c\) associated with the states \([1], [2], [3]\), respectively, are given by

\[
a(t_1) = 1, \quad b(t_1) = 0, \quad c(t_1) = -\epsilon;
\]

(12)

that is, the weak pulse information is correctly stored into a spin coherence.

Note that at this stage that if one would use rephasing pulses that are off-resonant with the \([1]\)–\([2]\) and \([2]\)–\([3]\) transitions, the protocol would be identical with the three-pulse echo in two-level systems already discussed. Nonresonant light could indeed compensate for the dephasing associated with the inhomogeneously broadened spin transition without population transfer to the excited state \([2]\), which could thus be eliminated adiabatically. The two spin states \([1]\) and \([3]\) would be equally populated at the rephasing time. As a consequence, the last \(\pi\) pulse used to transfer the spin coherence to an optical coherence would transfer the population of one of ground states into the excited state. The resulting spontaneous emission would lead to a signal-to-noise ratio equal to the one given in Eq. (10).

But let us consider resonant rephasing light pulses applied simultaneously on the transitions \([1]\)–\([2]\) and \([2]\)–\([3]\), as in Ref. [6]. More precisely, two Raman pulses corresponding to \(\Omega_2''\) and \(\Omega_2'^\prime\) are sent through the medium at times \(t_2\) and \(t_3\), respectively, to compensate for the inhomogeneous dephasing. Further, consider the case where the delay \(t_1 - t_2\) between these Raman pulses is much longer than the phase relaxation time of the \([1]\)–\([3]\) transition, as before. Finally, at time \(t_4 > t_1\), a \(\pi\) pulse resonant with the transition \([2]\)–\([3]\) transfers back the spin coherence into an optical coherence. At time \(t_4\) just

![FIG. 4. Pulse sequence for the three-pulse photon echo in three-level systems. The first pulse, resonant with the transition \([1]\)–\([2]\), is the one to store. It is immediately followed by a \(\pi\) pulse at time \(t_1\) on the transition \([2]\)–\([3]\) to transfer the optical coherence into a spin coherence. The next two pulses, corresponding to \(\Omega_2''\) and \(\Omega_2'^\prime\), which are Raman pulses, force the ensemble of dipoles to rephase. The resulting coherences are read out at time \(t_4 = t_1 + t_2 - t_1\) by a \(\pi\) pulse on the transition \([2]\)–\([3]\). The echo is emitted immediately after this final \(\pi\) pulse.](image)
after the interaction with this last \( \pi \) pulse, the state of the atom is described by a mixture of three states,

\[
|\psi_1\rangle = \frac{1}{2} \left( 1 + \cos \frac{\theta_2^r}{2} \right) |1\rangle \\
- \frac{i}{2} \left( -1 + \cos \frac{\theta_2^r}{2} \right) e^{-i\Delta_0(t_3-t_1)} |2\rangle - \frac{1}{\sqrt{2}} \sin \frac{\theta_2^r}{2} |\overline{3}\rangle,
\]

\[
|\psi_2\rangle = -\frac{i}{\sqrt{2}} \sin \frac{\theta_2^r}{2} |1\rangle \\
- \frac{1}{\sqrt{2}} \sin \frac{\theta_2^r}{2} e^{-i\Delta_0(t_3-t_1)} |2\rangle - i \cos \frac{\theta_2^r}{2} |\overline{3}\rangle,
\]

\[
|\psi_3\rangle = \frac{1}{2} \left( -1 + \cos \frac{\theta_2^r}{2} \right) |1\rangle \\
- \frac{i}{2} \left( 1 + \cos \frac{\theta_2^r}{2} \right) e^{-i\Delta_0(t_3-t_1)} |2\rangle - \frac{1}{\sqrt{2}} \sin \frac{\theta_2^r}{2} |\overline{3}\rangle,
\]

with respective weights \( a(t_2)^2 \), \( b(t_2)^2 \), and \( c(t_2)^2 \) given by

\[
a(t_2) = \frac{1}{2} \left( 1 + \cos \frac{\theta_2^r}{2} \right) - \frac{\epsilon}{2} \left( 1 + \cos \frac{\theta_2^r}{2} \right) e^{-i\Delta_0(t_2-t_1)},
\]

\[
b(t_2) = -\frac{i}{\sqrt{2}} \sin \frac{\theta_2^r}{2} + \frac{i \epsilon}{\sqrt{2}} \sin \frac{\theta_2^r}{2} e^{-i\Delta_0(t_2-t_1)},
\]

\[
c(t_2) = \frac{1}{2} \left( -1 + \cos \frac{\theta_2^r}{2} \right) - \frac{\epsilon}{2} \left( 1 + \cos \frac{\theta_2^r}{2} \right) e^{-i\Delta_0(t_2-t_1)}.
\]

B. Intensity of the echo signal

When absorbed by the sample, the first weak pulse induces a weak macroscopic polarization

\[ P(t_1) = i \varphi N \epsilon. \]  

If the two Raman pulses are Raman \( \pi \) pulses, that is, \( \theta_2^r = \theta_2^r = \pi \) as suggested in Ref. [6], they lead to a macroscopic polarization at time \( t_4 = t_3 + t_2 - t_1 \) given by

\[ P(t_4 = t_3 + t_2 - t_1) \rightarrow -i \varphi N \epsilon \]

where the signal-to-noise ratio is given by

\[ \frac{I_{\text{echo}}}{I_{\text{noise}}} = \frac{9}{8} N \epsilon^2. \]  

Since the intensity related to the absorbed part of the input pulse is equal to \( N^2 \epsilon^2 I_0 \), the storage efficiency is upper bounded by 9/64 (roughly 15%).

C. Fluorescence

To determine the amount of noise, we evaluate the population in the excited state at the echo time \( t_4 = t_3 + t_2 - t_1 \) without excitation, that is, for \( \epsilon = 0 \), as in two-level systems. We find

\[ I_{\text{noise}} = \frac{3}{8} N I_0 \]  

so that the signal-to-noise ratio is given by

\[ \frac{I_{\text{echo}}}{I_{\text{noise}}} = \frac{3}{8} N \epsilon^2, \]  

which reduces to 3/8 in the single-photon case corresponding to \( N \epsilon^2 = 1 \). Again, due to the spontaneous emission induced by the rephasing pulses, the fidelity of the quantum state storage is degraded beyond the classical limit: \( F_{3\text{-level}} = \frac{11}{12} \approx 0.917 \). Note that \( F_{3\text{-level}} < F_{2\text{-level}} \).

Before concluding, let us focus on the analysis presented in Ref. [6]. The author uses four-level systems with three ground states \( |1\rangle \), \( |3\rangle \), and \( |a\rangle \) and one excited state \( |2\rangle \). The studied protocol is similar to the one discussed here, except that the excited atoms are reversibly transferred to the auxiliary state \( |a\rangle \) during storage (i.e., between the times \( t_2 \) and \( t_3 \)). This population transfer to \( |a\rangle \) avoids the loss by spontaneous emission between times \( t_2 \) and \( t_3 \). However, the upper level population is restored at time \( t_3 \), before applying the second Raman \( \pi \) pulse. Hence, one exactly recovers the situation we considered here. As a consequence, the problem of spontaneous emission is not circumvented and the signal is equally dominated by the noise.

IV. CONCLUSION

In this paper, we proved that the conventional three-pulse photon echo cannot be used for quantum storage. A fundamental limitation is related to the medium excitation induced by the rephasing pulses. This excitation induces spontaneous emission, which produces a noise comparable to the retrieved signal and thus deteriorates the storage fidelity beyond the classical limit. This is a major difference compared to genuine quantum memory protocols such as stopped light based on electromagnetically induced transparency (EIT) [13], controlled reversible inhomogeneous broadening (CRIB) [14–17], or atomic frequency combs (AFC) [18], where the control pulses transfer a negligible number of atoms to excited states. Note finally that additional detrimental effects due to propagation [5] could further limit the fidelity and the efficiency of quantum storage based on three-pulse photon echo.

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[12] It is interesting to note that the classical limit is saturated using a two-pulse photon-echo storage approach [5]. The decay of the excited coherences in the three-pulse echo approach discussed here is at the origin of the additionally reduced fidelity.

Telecommunication-Wavelength Solid-State Memory at the Single Photon Level

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We demonstrate experimentally the storage and retrieval of weak coherent light fields at telecommunication wavelengths in a solid. Light pulses at the single photon level are stored for a time up to 600 ns in an erbium-doped Y$_2$SiO$_5$ crystal at 2.6 K and retrieved on demand. The memory is based on photon echoes with controlled reversible inhomogeneous broadening, which is realized here for the first time at the single photon level. This is implemented with an external field gradient using the linear Stark effect. This experiment demonstrates the feasibility of a solid-state quantum memory for single photons at telecommunication wavelengths, which would represent an important resource in quantum information science.

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Quantum memories (QMs) allowing the reversible transfer of quantum states between light and matter are an essential requirement in quantum information science[1]. They are, for example, a crucial resource for the implementation of quantum repeaters [2–5], which are a potential solution to overcome the limited distance of quantum communication schemes due to losses in optical fibers. Several schemes have been proposed to implement photonic quantum memories [6–12]. Important progress has been made during the last few years, with proof-of-principle demonstrations in atomic gases [13–20], single atoms in a cavity [21], and solid-state systems [22]. For all these experiments the wavelength of the stored light was close to the visible range and thus not suited for direct use in optical telecom fibers. The ability to store and retrieve photons at telecommunication wavelengths (around 1550 nm) in a QM would provide an important resource for long distance quantum communication. Such a QM could easily be integrated in fiber communication networks. In combination with a photon pair source, it could provide a narrow band triggered single photon source adapted to long distance transmission. Moreover, QMs at telecommunication wavelengths are required for certain efficient quantum repeater architectures [5,23,24].

A telecom QM requires an atomic medium with an optical transition in the telecom range, involving a long lived atomic state. The only candidate proposed so far is based on erbium-doped solids, which have a transition around 1550 nm between the ground state $^4I_{15/2}$ and the excited state $^4I_{13/2}$. These systems have been studied for spectroscopic properties [25,26] and classical light storage [27–29]. Photonic quantum storage in these materials is extremely challenging, because of the difficulties in the memory preparation using optical pumping techniques [30]. Yet in this Letter, we report an experiment of storage and retrieval of weak light fields at the single photon level in an erbium-doped solid.

Rare-earth doped solids have an inhomogeneously broadened absorption line. Single photons can be mapped onto this optical transition, leading to single collective optical excitations [22]. During the storage, inhomogeneous dephasing takes place, preventing an efficient collective reemission of the photon. This dephasing can be compensated for using photon echo techniques. The storage of quantum light (e.g., single photons) is however not possible using traditional photon echo techniques, such as two pulse photon echoes [31]. The main issue is that the application of the strong optical pulse ($\pi$-pulse) to induce the rephasing mechanism leads to amplified spontaneous emission and reduces the fidelity of the storage to an unacceptable level [32]. A way to overcome this problem is to induce the rephasing of the atomic dipoles by generating and reversing an artificial inhomogeneous broadening. This scheme is known as controlled reversible inhomogeneous broadening (CRIB) [8,9,33,34]. The CRIB scheme was first demonstrated with bright optical pulses, in a Eu$^{3+}$ : Y$_2$SiO$_5$ crystal at 580 nm [33]. The phase of the stored light pulses was shown to be well preserved [35]. For these experiments, the storage and retrieval efficiency was of the order of $10^{-6}$. It has been dramatically improved in more recent experiments at 606 nm in Pr$^{3+}$ : Y$_2$SiO$_5$ [36]. CRIB has also been demonstrated on a spin transition in a rubidium vapor [37] at 780 nm. Here, we report an experiment at telecommunication wavelength. Moreover, we also report the first experimental demonstration of CRIB at the single photon level, opening the road to the quantum regime.

In order to realize a CRIB experiment in a rare-earth doped solid, one first has to prepare a narrow absorption line within a large transparency window. The spectrum of this line is then broadened by an electric field gradient using the linear Stark effect to match the bandwidth of the photon to be stored. The incident photon is absorbed by the ions in the broadened line, and mapped into a single collective atomic excitation. During a time $t$ each excited...
ion $i$ will acquire a phase $\Delta_i t$ due to its shift in the absorption frequency $\omega_i = \omega_i^0 + \Delta_i$ from the central frequency $\omega_i^0$. Switching the polarity of the field after a time $t = \tau$ will reverse the broadening ($\omega_i = \omega_i^0 - \Delta_i$) and after another time $\tau$ the ions will be in phase again and reemit the photon. In order to create the initial narrow absorption line, a population transfer between two ground states (in our case Zeeman states) using optical pumping via the excited state is used [30]. In case of imperfect optical pumping, some atoms will remain in the excited state after the preparation. An experimental issue arising when input pulses are at the single photon level is the fluorescence from these excited atoms. If the depletion of this level is slow (as in rare-earth ions, with optical relaxation times $T_1$ usually in the range of 0.1 to 10 ms), this can lead to a high noise level that may blur the weak echo pulse. The problem is especially important for erbium-doped solids, where $T_1$ is very long ($\approx 11$ ms [25] in Er$^{3+}$:Y$_2$SiO$_5$). In our experiment, the population transfer is enhanced by stimulating ions from the excited state down to the short lived second ground state crystal field level using a second laser at 1545 nm [Fig. 1(a)] [30]. The application of this laser enhances the rate of depletion of the excited state and thus reduces the noise from fluorescence. Together with a suitable waiting time between the preparation and the light storage, it allows the realization of the scheme at the single photon level.

Our memory consists of an Y$_2$SiO$_5$ crystal doped with erbium (10 ppm) cooled to 2.6 K in a pulse tube cooler (Oxford Instruments). The crystal has three mutually perpendicular optical-extinction axes labeled $D_1$, $D_2$, and $b$. Its dimensions are $3.5 \times 4 \times 6$ mm along these axes. The magnetic field of $B = 1.5$ mT used to induce the Zeeman splitting necessary for the memory preparation is provided by a permanent magnet [Fig. 1(b)]. The light is traveling along $b$. The electrical field gradient for the Stark broadening is applied with four electrodes placed on the crystal in quadrupole configuration, as shown in Fig. 1(b) and described in [39]. The induced broadening is proportional to the voltage $U$ applied on the electrodes [39].

The experiment is divided into two parts: the preparation of the memory and the storage of the weak pulses [see Fig. 1(b)]. Each preparation sequence takes 120 ms of optical pumping during which both the pump and the stimulation lasers are sent into the sample. The frequency of the pump laser is repeatedly swept to create a large transparency window into the inhomogeneously broadened absorption line. If the laser is blocked for a short time at the center of each sweep using an acousto-optical modulator (AOM), a narrow absorption feature is left at the center of the pit [30]. The time available to perform the memory protocol is given by the Zeeman lifetime of $T_2 = 130$ ms [38] of the material. In order to deplete the excited state the laser at 1545 nm is left on for 23.5 ms after the pump pulses. Then the preparation path is closed and the detection path is opened. The storage sequence begins 86 ms after the pump pulse, in order to avoid fluorescence from the excited atoms. It is composed of 8000 independent trials separated by 5 $\mu$s. In each trial, a weak pulse of duration $\delta \tau$ is stored and retrieved. The initial peak is broadened with an electrical pulse before the absorption of each pulse. The polarity of the field is then inverted at a programable time after the storage, allowing for on demand readout. The whole sequence is repeated at a rate of 3 Hz. The weak output mode is detected using a superconducting single photon detector (SSPD) [40] with an efficiency of 7% and a low dark count rate of 10 $\pm$ 5 Hz. The incident pulses are weak coherent states of light $|\alpha|^L$ with a mean number of photons $\bar{n} = |\alpha|^2$. We determined $\bar{n}$ at the input of the cryostat by measuring the number of photons arriving at the SSPD (with the laser out of resonance), compensating for the transmission (16%) and detection efficiency.

We now describe the observation of CRIB photon echoes of weak pulses. As a first experiment, we sent pulses with $\bar{n} = 10$ and $\delta \tau = 200$ ns into the sample. Figure 2 shows a time histogram of the photon counts detected after the crystal. The first peak corresponds to the input photons transmitted through the crystal. The second peak is the CRIB echo. It is clearly visible above the noise floor. Only a small fraction of the incident light is reemitted in the CRIB echo (about 0.25%). The reasons for this low storage and retrieval efficiency and ways to improve it will be discussed in more detail below. As a consistency check, we verified that the echo disappears when the narrow absorption peak is not present (see blue open circles in Fig. 2).
lowered $\bar{n}$ by increasing the attenuation, for input pulses with $\delta \tau = 200$ ns. The result is shown in Fig. 4(a). Both the number of photons in the CRIB echo and the signal to noise ratio depend linearly on $\bar{n}$. This means that the efficiency and the noise are independent of $\bar{n}$. Figure 4(b) shows the result of a measurement with—in quantum key distribution terminology—pseudosingle photons ($\bar{n} = 0.6$). In that case, we still obtain a signal to noise ratio of $\sim 3$. The remaining noise floor may be due to residual fluorescence and leakage through the AOMs.

In the following we analyze the efficiency and storage time performances of our memory in more detail. Ref. [11] gives a simplified model for the CRIB memory. In this model, the storage and retrieval efficiency if the echo is emitted in the forward direction is given by

$$\eta_{\text{CRIB}}(t) = d^2 e^{-d} e^{-\gamma t^2},$$

where $\gamma = 2 \pi \gamma_p$ is the spectral width (standard deviation) of the initial Gaussian absorption peak, and $d$ is the optical depth of the broadened absorption peak. The main assumption here is that the spectral width of the absorption peak is much wider than the spectral bandwidth of the photon to be stored. By fitting the decay curve of Fig. 3 with Eq. (1), we find a full width at half maximum linewidth of the central peak of 1 MHz. This corresponds well to the results obtained by a measurement of the transmission spectrum. The minimal width is limited by the linewidth of our unstabilized laser diode and power broadening during the preparation of the peak. The optical coherence time of the transition under our experimental conditions has been measured independently by photon echo spectroscopy. It was found to be $T_2 = 2 \mu s$, corresponding to a homogeneous linewidth of 160 kHz. Note that the optical coherence in Er$^{3+}$:Y$_2$SiO$_5$ could be drastically increased using lower temperatures and higher magnetic fields [26].

In our experiment, imperfect optical pumping results in a large absorbing background with optical depth $d_0$, which acts as a passive loss, such that the experimental storage and retrieval efficiency is given by: $\eta(t) = \eta_{\text{CRIB}}(t) \times \exp(-d_0)$ [22]. The values of $d$ and $d_0$ can be measured
by recording the absorption spectra. This yields an optical depth of the unbroadered peak $d' = 0.5 \pm 0.2$ and an absorbing background of $d_0 = 1.6 \pm 0.1$. A voltage of 50 V on the electrodes corresponds to a broadening of a factor of $\sim 3$ [39], leading to $d = 0.17 \pm 0.07$. In our experiment, the photon bandwidth is of the same order as the broadened peak, so that the assumptions of the simplified model are not fulfilled. In order to have a more accurate description, we have solved numerically the Maxwell-Bloch equations with the measured $d'$, using a Gaussian initial peak. This gives storage and retrieval efficiencies of order $10^{-3}$ (including the passive loss $d_0$) for a storage time of 300 ns, in reasonable agreement with the measured values (see Fig. 3).

This study shows that the main reason of the low storage and retrieval efficiency in the present experiment is the small absorption in the broadened peak and the large absorbing background, due to imperfect optical pumping. About 80% of the retrieved photons are lost in the absorbing background. The limited optical pumping efficiency is due to the small branching ratio in the $\Lambda$ system and to the small ratio between the relaxation life times of the optical and the ground state Zeeman transitions [30]. This could be improved in several ways. First technical improvements can be implemented, such as using lower temperatures, higher stimulation laser intensities and spin mixing in the excited state using RF fields, as demonstrated in [30]. Second, the branching ratio and Zeeman lifetime strongly depend on the applied magnetic field angle and intensity. A full characterization of the optical pumping efficiency with respect to these parameters has not been carried out yet.

Finding optimal conditions may lead to significant improvements. It would also be interesting to investigate hyperfine states. Finally, other crystals might be explored, e.g., $Y_2O_3$, to search for longer Zeeman lifetimes.

In summary, we have presented a proof-of-principle of quantum memory for photons at telecommunication wavelengths. Pulses of light at the single photon level have been stored and retrieved in an erbium-doped crystal, using the CRIB protocol. Continuing efforts to increase the efficiency and the storage time will be required in order to build a useful device for applications in quantum information science. Our experiment is nevertheless an enabling step towards the demonstration of a fiber network compatible quantum light matter interface. It also confirms the feasibility of the CRIB protocol at the single photon level.

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Demonstration of Atomic Frequency Comb Memory for Light with Spin-Wave Storage

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We present a light-storage experiment in a praseodymium-doped crystal where the light is mapped onto an inhomogeneously broadened optical transition shaped into an atomic frequency comb. After absorption of the light, the optical excitation is converted into a spin-wave excitation by a control pulse. A second control pulse reads the memory (on-demand) by reconverting the spin-wave excitation to an optical one, where the comb structure causes a photon-echo-type rephasing of the dipole moments and directional retrieval of the light. This combination of photon-echo and spin-wave storage allows us to store submicrosecond (450 ns) pulses for up to 20 μs. The scheme has a high potential for storing multiple temporal modes in the single-photon regime, which is an important resource for future long-distance quantum communication based on quantum repeaters.

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A quantum memory (QM) for photons is a light-matter interface that can achieve a coherent and reversible transfer of quantum information between a light field and a material system [1]. A QM should enable efficient, high-fidelity storage of nonclassical states of light, which is a key resource for future quantum networks, particularly in quantum repeaters [2–6]. In order to achieve reasonable entanglement distribution rates, it has been shown that some type of multiplexing is required [4,5], using for instance independent frequency, spatial or temporal modes (multimode QM).

Several types of light-matter interactions have been proposed for building a QM, for instance electromagnetically induced transparency [7–10], Raman interactions [11–14], or photon-echo techniques [15–21]. Photon-echo techniques in rare-earth-ion doped crystals have an especially high multimode capacity for storing classical light [22]. Classical photon echoes are not useful, however, for single-photon storage due to inherent noise problems [23]. The photon-echo QM based on controlled reversible inhomogeneous broadening [15–18] is free of these noise problems. But this technique has a lower time-multiplexing capacity than classical photon echoes, for a given optical depth, due to loss of storage efficiency as the controlled frequency bandwidth is increased [19,24]. Some of us recently proposed a photon-echo type QM based on an atomic frequency comb (AFC) [19] that has a storage efficiency independent of the bandwidth, allowing optimal use of the inhomogeneous broadening of rare-earth-doped crystals. An AFC memory has the potential for providing multimode storage capacity [19,24] crucial to quantum repeaters. The few reported AFC experiments [20,21] have been investigating the physics of the optical AFC echo, where the memory storage time is predetermined by the periodicity of the comb. For quantum repeaters it is crucial to be able to choose the time of the memory readout (on-demand readout). Here we present the first light-storage experiment where an AFC is used in combination with reversible transfer of the excitation to a spin state [19], resulting in on-demand readout and storage times longer than 20 μs.

The underlying idea of the AFC QM is to shape an inhomogeneously broadened optical transition |g⟩→|e⟩ into a periodic series of narrow and highly absorbing peaks with periodicity Δ, see Fig. 1. A photon with a bandwidth that is matched to the width of the AFC structure is then stored as an optical excitation delocalized over the peaks, which we can write as |ψ⟩ = ∑Nj=1 cj e^iδj e^{-ikzj} |g_1 ... e_j ... g_N⟩ where N is the number of atoms in the AFC, δ_j is the detuning of atom j with respect to the laser frequency, z_j is the position, k is the wave number of the light field, and the amplitudes c_j depend on the frequency and on the spatial position of the particular atom j. The terms in this large superposition state accumulate different phases due to the inhomogeneous distribution of atomic resonance frequencies, resulting in a loss of the initially strong collective coupling to the light mode. But the periodic AFC peak separation Δ leads to a rephasing of the terms after a time 1/Δ, which restores the strong collective coupling, leading to a photon-echo type reemission [25], the AFC echo. The narrow and highly absorbing peaks can theoretically absorb all the light and completely emit the energy in the AFC echo [19]. A large number of peaks leads to a high multimode capacity [19,24].

In the original proposal [19] on-demand readout and longer storage times rely on reversible transfer of the optical excitation to a long-lived spin state by strong control pulses (see Fig. 1). If we imagine a perfect π pulse applied at time T', each term in the superposition state becomes |g_1 ... s_j ... g_N⟩; thus, we have a single collec-
ensemble of atoms whose different hyperfine transitions can be unambiguously excited. This distillation technique, which we will summarize here, has been the subject of several papers [27–29]. A laser beam whose frequency is swept pumps atoms from ground levels $|g\rangle$ and $|s\rangle$ to the auxiliary storage level $|\text{aux}\rangle$; see Fig. 1, which creates a wide transmission window within the inhomogeneous profile. In the next step a narrow absorption peak is created in the hole by coherently transferring back atoms, within a narrow frequency range, from $|\text{aux}\rangle$ to $|g\rangle$ [28]. In this experiment we extended this method by transferring back atoms at different frequencies to create a frequency comb. In Fig. 1(b) we show the absorption spectrum recorded after the preparation sequence. The AFC created on the $|g\rangle \leftrightarrow |e\rangle$ transition is clearly visible. The $|s\rangle$ level is used for the spin-wave storage, which means that the control pulses will be applied on the $|s\rangle \leftrightarrow |e\rangle$ transition displaced by 10.2 MHz with respect to the $|g\rangle \leftrightarrow |e\rangle$ transition. Note that the absorption spectrum in Fig. 1(b) is shown only for visualization purposes, since the fast frequency scan method [29] we used leads to distortions for absorption depths $d \geq 2$. The experimental setup is shown in Fig. 2. The control pulses were both counterpropagating with respect to the input pulse. By phase matching condition the output pulse then copropagates with the input signal [19]. Using this configuration we could reduce noise due to off-resonant free-induction decay emission produced by the strong control pulses.

In a preliminary experiment we investigate the AFC echo on the $|g\rangle \leftrightarrow |e\rangle$ transition without applying the control pulses, see inset in Fig. 3. This allows us to optimize the relevant comb parameters in order to obtain a strong echo. The input pulse duration was set to 450 ns so that the bandwidth is entirely contained in the 2 MHz wide AFC. The efficiency of this AFC echo, which we define as the

![Fig. 1](color online). (a) The experiment was performed on the $^3\text{H}_4 \rightarrow ^1\text{D}_2$ transition in Pr$^{3+}$. The ground and excited state manifolds both have three hyperfine levels denoted $M_I = \pm 1/2$, $\pm 3/2$, $\pm 5/2$. The three-level lambda system was formed by the levels labeled $|g\rangle$, $|s\rangle$ and $|e\rangle$, following the notation in [19]. (b) Experimental absorption spectrum showing the AFC on the $|g\rangle \leftrightarrow |e\rangle$ transition created within a 18 MHz wide transmission hole using the spectral holeburning sequence described in the text. The comb consists of 9 peaks with spacing $\Delta = 250$ kHz. The holeburning sequence also empties the $|s\rangle$ level, whereas $|\text{aux}\rangle$ is used for population storage. Note that the second AFC in the center of the spectrum is due to the weaker transition from the ground state $|s\rangle$ to the excited state $|e\rangle$. (c) The pulse sequence showing the input pulse to store, the two control fields for the back-and-forth transfer to the spin state, and the retrieved output pulse.

![Fig. 2](color online). The experimental setup. The spectral hole burning and storage pulse sequences were created using a frequency-stabilized laser and acousto-optic modulators similar to the setup in [28] (not shown). A beam splitter (BS) split the light into a strong and a weak beam, whose two modes were overlapped in the crystal cooled to 2.1 K. The detected output pulse propagating to the right originated from the combination of a weak input pulse incident on the crystal from the left, and two strong control pulses incident from the right. Note that the signal disappeared when the control pulses were blocked directly after the BS. An AOM was used to direct only the transmitted input pulse and the output pulse onto the photodiode (PD), effectively working as a detector gate.
ratio of the AFC echo area to the input pulse area, depend on the shape of the AFC [19]. Two critical parameters are the peak absorption depth \( d \) and the finesse defined as \( F = \Delta / \gamma \) where \( \gamma \) is the full-width at half maximum of a peak. For instance, a high finesse leads to low decoherence during the storage time \( 1/\Delta \), but also to a lower effective absorption \( \sim d/F \) of the input pulse [19]. The peak absorption \( d \) could be controlled by the power of the laser beam creating the peaks in the transmission hole, but a high power also had an impact on the finesse by causing power broadening of the peaks. The peak width was also limited by laser frequency stability, resulting in typical widths of \( \gamma = 100 \) kHz. For a periodicity of \( \Delta = 1 \) MHz, the optimized efficiency was about 15%. The delay of 1 \( \mu \)s was not sufficiently long, however, for applying the control pulses before the emission of the AFC echo. We therefore set the periodicity to \( \Delta = 250 \) kHz giving us 4 \( \mu \)s to apply the control pulses. The closer spacing of the peaks lowered the finesse of the comb, thus lowering the efficiency to \( \sim 5\% \) (see inset in Fig. 3). This efficiency is in reasonable agreement with numerical simulations using the experimentally estimated peak absorption depth and finesse of the comb.

In Fig. 3 we show the main result where two control pulses are applied on \( |s\rangle \leftrightarrow |e\rangle \) to transfer the excitation to the \( |s\rangle \) hyperfine level. The retrieved pulse is clearly observed above the noise level. This realizes a true storage of the input pulses, with on-demand readout. Thus, the control pulses provide a mechanism for momentarily interrupting the predetermined AFC evolution [19]. We tested this mechanism in detail by varying the time at which the first control pulse was applied \( T' = (1.17, 1.63, 2.23) \) \( \mu \)s [cf. Fig. 1(c) for notation]. This resulted in different measured durations \( T'' = (2.84, 2.41, 1.85) \) \( \mu \)s. The total time spent in the excited state \( |e\rangle \), however, is constant (within the measurement error), \( T' + T'' = (4.01, 4.04, 4.08) \) \( \mu \)s, corresponding to the expected \( 1/\Delta \).

In Fig. 4 we show storage experiments where the spin-wave storage time \( T_s \) is varied. The output signal is clearly visible up to 20 \( \mu \)s of total storage time \( (1/\Delta + T_s) \). The exponential decay of the output signal as a function of \( T_s \) can be attributed to inhomogeneous spin dephasing, corresponding to an inhomogeneous broadening of 26 kHz consistent with previous measurements [30]. We point out that this can be compensated for by spin echo techniques. With such techniques Longdell et al. [31] stopped light during >1 sec in a Pr\(^3+\):Y\(_2\)SiO\(_5\) crystal using electromagnetically induced transparency.

We now discuss the total storage efficiency \( \eta \). Clearly \( \eta \) is bounded by the AFC echo efficiency \( \eta_e = 4\%\text{--}5\% \) for \( 1/\Delta = 4 \) \( \mu \)s (cf. Fig. 3 inset). This can be improved by increasing the finesse and optical depth (see discussion above). The spin-wave storage further decreases the efficiency. By extrapolating the experimentally measured \( \eta \) to the limit \( T_s \rightarrow 0 \) we find that \( \eta = 0.5\%\text{--}1\% \), which is a value independent of spin dephasing. The effect of the control pulses can now be understood by a simple model. We assume that one pulse has a single-atom \( |s\rangle \leftrightarrow |e\rangle \) transfer efficiency \( \eta_T \), which is constant as a function of detuning. Then the application of the two control pulses reduce the efficiency to \( \eta = \eta_e \eta_T \). Numerical simulations using 3-level Maxwell-Bloch equations show that this simple model is correct if the control pulses do not induce any additional phase decoherence. From the experimental values given above we thus find \( \eta_T = 0.30\text{--}0.45 \).

To better understand this value we have performed numerical calculations. The control pulses were two identical complex hyperbolic secant pulses, which can achieve

**FIG. 3** (color online). Storage in the spin state using two control pulses. Shown are (from left) the partially transmitted input pulse, control pulses (strongly attenuated by the closed optical gate), and the output pulse (magnified by a factor 10 for clarity). Here \( 1/\Delta = 4 \) \( \mu \)s and \( T_s = 7.6 \) \( \mu \)s, resulting in a total storage time \( 1/\Delta + T_s = 11.6 \) \( \mu \)s. Inset: The AFC echo observed at \( 1/\Delta = 4 \) \( \mu \)s when the control pulses are not applied. Note that the vertical scales have been normalized to 100 with respect to the input pulse before the crystal; thus, these yield (in percent) the efficiency for the echo and transmission coefficient for the input pulse.

**FIG. 4** (color online). Experimental traces for spin-storage times \( T_s = 5.6, 7.6, 10.6, \) and 15.6 \( \mu \)s. All other parameters are the same as those in Fig. 3. The input pulses are superimposed to the left (truncated) and the output pulses for different \( T_s \) are seen to the right. The leakage of the control pulses through the optical gate is not shown. For clarity there is also a break in the horizontal scale. The decay of the signal (see inset) as a function of \( T_s \) is due to inhomogeneous spin dephasing. The solid curve is a fitted Gaussian function corresponding to 26 kHz (full-width at half maximum) spin broadening.
efficient, broadband transfer of population [28] and coherence [29]. Our pulses had duration 600 ns and Rabi frequency $\sim$1.2 MHz (close to the maximal value in this experiment), the frequency chirp being 2 MHz. These values were found by empirically optimizing the size of the output pulse. Using the numerical model we find $\eta_T = 0.75$, also averaged over the bandwidth of the AFC. Based on this we would expect a total efficiency $\eta = 5\% \times 0.75^2 = 2.8\%$ using the simplified model above, which is significantly higher than what we observe. The most probable explanation for the discrepancy is imperfect spatial mode overlap between the counterpropagating input and control pulses (see Fig. 2). A larger control beam would make a more spatially uniform Rabi frequency and would facilitate mode alignment. Note that the theoretical $\eta_T$ can be improved by increasing the Rabi frequency (by a factor of 2) and adapting the duration and chirp of the pulse in order to achieve an efficient ($\eta_T \approx 95\%$) transfer.

We finally show an example of storage of two temporal modes; see Fig. 5. Note that both modes are stored with the same efficiency, which is a particular feature of the AFC. Based on this we would expect a total efficiency $\eta = 5\% \times 0.75^2 = 2.8\%$ using the simplified model above, which is significantly higher than what we observe. The most probable explanation for the discrepancy is imperfect spatial mode overlap between the counterpropagating input and control pulses (see Fig. 2). A larger control beam would make a more spatially uniform Rabi frequency and would facilitate mode alignment. Note that the theoretical $\eta_T$ can be improved by increasing the Rabi frequency (by a factor of 2) and adapting the duration and chirp of the pulse in order to achieve an efficient ($\eta_T \approx 95\%$) transfer.

In conclusion, we have demonstrated the first light-storage experiment combining an atomic frequency comb and spin-wave storage. Using this method we stored optical submicrosecond (450 ns) pulses for up to 20 $\mu$s as a spin-wave in Pr$^{3+}$:Y$_2$SiO$_5$. This optical bandwidth is more than 1 order of magnitude higher than previous stopped-light experiments demonstrated in rare-earth crystals [31]. The spin-storage time could be greatly extended by spin echo techniques [31].

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Electric control of collective atomic coherence in an erbium-doped solid

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Electric control of collective atomic coherence in an erbium-doped solid

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Abstract. We demonstrate the fast and accurate control of the evolution of collective atomic coherences in an erbium-doped solid using external electric fields. This is achieved by controlling the inhomogeneous broadening of erbium ions emitting at 1536 nm using an electric field gradient, thanks to the linear Stark effect. The manipulation of atomic coherence is characterized with the collective spontaneous emission (optical free induction decay (FID)) emitted by the sample after an optical excitation, which does not require any previous preparation of the atoms. We show that controlled dephasing and rephasing of the atoms by the electric field result in collapses and revivals of the optical FID. Our results show that the use of external electric fields does not introduce any substantial decoherence and enables the manipulation of collective atomic coherence with a very high degree of precision on the timescale of tens of nanoseconds. This provides an interesting resource for photonic quantum state storage and quantum state manipulation.

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1. Introduction

The coherent control of quantum systems plays a central role in quantum information science and in quantum technology in general. In particular, the coherent manipulation of collective atomic coherences in material systems is crucial for applications in photonic quantum storage [1, 2] and in ensemble based quantum computing [3, 4]. A promising route towards these applications is to use solid state atomic ensembles implemented with rare-earth (RE) ion-doped solids.

In a solid state environment, the optical atomic lines of the RE impurities are inhomogeneously broadened [5]. When atoms are excited, for example following the absorption of a light pulse, the atomic dipoles oscillate at different frequencies, leading to inhomogeneous dephasing. In order to enable a constructive interference between all the emitters that will lead to a collective re-emission of the stored light, this dephasing must be controlled. The rephasing of the dipoles can be triggered by optical pulses, as in traditional photon echo techniques. These techniques, while very successful to store classical light [6, 7] and as a tool for high resolution spectroscopy [5, 8], suffer from strong limitations for the storage of single photons [9]. Another possibility is to exploit the fact that some materials exhibit permanent dipole moments, which give rise to a linear Stark effect [10]. The frequency of the atoms can then be controlled with a moderate external electric field. This effect has been used for the demonstration of Stark modulated photon echoes [11]–[15] as a tool for high resolution spectroscopy. The electric control of the resonance frequency of the atoms is also the key resource of a recently proposed modified photon echo protocol based on controlled and reversible inhomogeneous broadening (CRIB) [16]–[19].

The great advantage of using electric fields to manipulate atomic coherences is that it does not change the population distribution in the ground and excited states, contrary to optical rephasing pulses. In combination with an optical transfer of population to a long lived ground state, this enables, in principle, the long-term storage and retrieval of single photon fields with unit efficiency and fidelity [17, 19]. Quantum storage with unit efficiency can also be achieved without the transfer to the long lived ground state, using only electric fields for the light retrieval [20, 21]. The first proof of principle experiment of the CRIB protocol was performed in a Eu-doped crystal [22, 23] followed by another demonstration in a Pr-doped crystal [20]. The maximal efficiency of the storage and retrieval is directly proportional to the quality of the manipulation of the atomic coherences. It is thus extremely important to have a good characterization of the rephasing of the dipoles.

A CRIB experiment requires sophisticated optical pumping techniques in order to first isolate a narrow absorption peak within a large transparency window. Moreover, it is difficult to characterize the quality of the rephasing directly from a CRIB experiment, since the efficiency of the storage also depends on other parameters, such as available optical depth or quality of the optical pumping for the preparation of the memory. In this paper, we use a much simpler method to characterize the electric manipulation of the atomic coherence. We propose to infer the dynamics of the atomic coherences by studying the collective spontaneous emission of light from the atoms (a phenomenon known as optical free induction decay (FID) [24]). In particular, we show that controlled dephasings and rephasings of the atoms via the electric field result in collapses and revivals of the FID. The observation of FID does not require any optical preparation of the sample and can thus be done with a relatively simple experimental setup. In practice it is sufficient to excite the atoms with a single optical pulse in resonance with the
atomic transition and to measure the collective emission of light after the pulse. The FID is, however, strongly nonlinear with respect to the excitation pulse intensity and vanishes in the limit of weak excitation pulses [24, 25].

In this paper, we use erbium ions doped into a Y$_2$SiO$_3$ crystal. This is an interesting system since erbium ions have an optical transition at the telecommunication wavelength of 1536 nm. It could thus, in principle, enable the realization of a light matter quantum interface between photons that can be transmitted with low loss in optical fibres and stationary atoms in a solid. Such a quantum memory at telecommunication wavelength would be useful in the context of quantum repeaters [26]–[28]. The spectroscopic properties of Er$^{3+}$ : Y$_2$SiO$_3$ have been extensively studied, including optical coherence [29]–[32], spectral diffusion [30, 33], hyperfine structure [34], Zeeman relaxation lifetimes [35], Zeeman $g$ factors [36] and erbium host interactions [37]. Slow light has also been achieved in this material using coherent population oscillation [38]. Optical pumping techniques have also been developed [39], and a proof of principle experiment of CRIB with weak light pulses at the single photon level has been recently demonstrated in an Er$^{3+}$ : Y$_2$SiO$_3$ crystal [40].

2. Control of collective atomic coherences using the linear Stark effect

We now explain in more detail how the dephasing and rephasing of atomic dipoles can be controlled via the linear Stark effect, and how this affects the collective emission of light from the sample. In the presence of an external dc electric field, the energy levels of an atom with a permanent dipole moment are shifted by an amount proportional to the electric field. This phenomenon is known as the linear dc Stark effect [10]. If the dipole moments are different for different electronic levels, this shift leads to a shift in the associated optical transition frequency. The linear dc Stark effect can be observed in RE-doped solids, since the ions possess a permanent dipole moment induced by local electric fields due to the crystal environment. The detuning of the atom transition $\Delta$ due to the linear Stark effect can be described as [35]

$$\Delta = \frac{\Delta\mu e \chi \cos \theta}{\hbar} E,$$

where $\Delta\mu e$ is the difference between the permanent dipole moments for the two states of the optical transition, $E$ is the applied electric field amplitude, $\chi = (\epsilon + 2)/3$ is the Lorentz correction factor, $\epsilon$ is the dielectric constant of the sample and $\theta$ is the angle between $\hat{\Delta}\mu e$ and $\hat{E}$. Since the crystal used in this experiment (Y$_2$SiO$_3$) has an inversion symmetry, there are two classes of erbium ions with dipole moments pointing in opposite directions [10]. This leads to the splitting of the resonance frequency of the atoms when a homogeneous electric field is applied. Each frequency is shifted by $\Delta$ or $-\Delta$ with respect to the unperturbed absorption frequency $\omega_0$. If the electric field intensity varies with the position in the sample, each atom experiences a different Stark shift, which leads to an additional inhomogeneous broadening.

When a light pulse is absorbed in an inhomogeneously broadened sample, the atoms in resonance with the light will be excited. While the excited atoms are in phase after the pulse is turned off, they are in a superradiant state, and a strong collective emission takes place in the forward mode defined by the input pulse. This emission will then decay when the atoms dephase due to inhomogeneous dephasing. The decay rate of the FID depends on the spectral distribution of the excited atoms. In the real experiment, the spectral distribution is basically given by the spectrum of the excitation pulse and the spectral width of the laser (assuming that...
the homogeneous line width is smaller than the pulse and laser width). It is thus possible to accelerate the decay in a controlled way by broadening the spectral distribution of the atoms with the linear Stark effect, using an electric field gradient. The atoms are then no more in a superradiant state and the collective emission is inhibited. However, if no random phase is acquired during the process, it is possible to undo the inhomogeneous dephasing due to the electric field and to obtain a revival of the emission [16]–[18]. This can be realized by reversing the polarity of the electric field, which will reverse the detuning of each atom. The phase evolution will now be reversed and after a given time, all the atoms will be in phase again, leading to a collective emission of light.

More formally, this process can be written as follows: suppose that the phase of each atom evolves in time with frequency \( \omega \), i.e. we can write for the phase evolution of the \( j \)th atom: \( e^{-i\omega t_j} \). An external electric field will shift the frequency of the \( j \)th atom by \( \Delta_j \). Suppose that electric field is turned on at time \( t = 0 \) and turned off at time \( t = \tau \). The phase acquired by the atom is then \( e^{-i(\omega \Delta_{j1} + \Delta_j)\tau} \). Now, suppose that instead of turning off the electric field, it is switched to the opposite polarity at time \( t = \tau \). The frequency shift \( \Delta_j \) then becomes \( -\Delta_j \) and the phase of the atom at time \( t \) is given by

\[
e^{-i(\omega \Delta_{j1} + \Delta_j)\tau} e^{-i(\omega \Delta_{j1} - \Delta_j)(t-\tau)}. \tag{2}
\]

It can be seen that, at a time \( t = 2\tau \), the externally introduced phase shifts \( \Delta_j \) cancel out. If the optical transition has a natural inhomogeneous broadening, the atomic evolution due to the controlled dephasing and rephasing is superposed with the natural evolution due to the inhomogeneous dephasing. Hence, in the case of FID, the intensity of the light after the rephasing should reach the intensity of the unperturbed FID signal.

3. The experiment

In the experiment we used a \( \text{Y}_2\text{SiO}_5 \) crystal doped with erbium ions Er\(^{3+}\) (with 10 ppm concentration). The atoms were excited on the transition \( ^4\text{I}_{15/2} \rightarrow ^4\text{I}_{13/2} \) at the telecom wavelength of 1536 nm [30]. The \( \text{Y}_2\text{SiO}_5 \) crystal has three mutually perpendicular optical-extinction axes labelled \( D_1 \), \( D_2 \) and \( b \). The direction of light propagation \( \vec{k} \) is along the \( b \)-axis. The dimensions of the Er\(^{3+}\) : \( \text{Y}_2\text{SiO}_5 \) crystal are 6 mm \( \times \) 3.5 mm \( \times \) 4 mm along the \( b \)-, \( D_1 \)- and \( D_2 \)-axis, respectively. The crystal was cooled to 2.6 K in a pulse tube cooler (Oxford Instruments). The inhomogeneous width is approximately 250 MHz and the optical peak absorption depth of the crystal is 2. In order to create the electric field gradient, we implemented a quadrupole scheme [22] using four electrodes attached directly to the crystal, perpendicular to the \( D_2 \)-axis (see inset of figure 1). Such a configuration creates an electric field in the \( D_2 \) direction which changes linearly along the axis parallel to the light propagation. The electrodes were thin aluminium stripes, each of 1 mm width and spaced by 1.5 mm. To switch the electric field we used a fast electrical switch with a switching time of 10 ns and minimal/maximal voltage \(-100/\pm 100 \text{ V}\).

The experimental setup is shown in figure 1. The light source was a free running external cavity diode laser (Toptica) at 1536 nm with laser bandwidth of approximately 2 MHz. The light was amplitude modulated by an AOM in order to create the excitation pulses at a repetition rate of 10 kHz, with duration 3 \( \mu \text{s} \). The light was then coupled to a single mode optical fibre and passed through a variable fibre attenuator, before being focused in free space through the crystal.

Figure 1. Experimental scheme used to demonstrate the electric control of collective atomic coherences. Light pulses created with an acousto-optic modulator (AOM1) are attenuated with a fibre variable attenuator (VAR ATT) and focused through an Er$^{3+}$: Y$_2$SiO$_5$ crystal cooled at 2.6 K in a pulse tube cooler. The excitation pulses are then blocked by an optical gate implemented with a fibre AOM2 and the weak FID signal at the single photon level is detected with a superconducting single photon detector (SSPD). The inset shows the crystal with the quadrupole configuration of electrodes that produce the electric field gradient along the light propagation direction.

in the cryostat. The beam waist (diameter) in the crystal was 70 $\mu$m. After the crystal, the light was again coupled into a single mode fibre and sent through a fibre coupled AOM that served as optical gate in front of the detector to block the excitation pulses. With respect to the applications in quantum memories, where usually only few photons are absorbed, the measurements were made in the low excitation regime (with an excitation pulse area $\ll \pi$, typically with about $10^6$ photons in the excitation pulse). As the amplitude of the FID signal is strongly nonlinear with respect to the excitation intensity, the FID signal was extremely weak (about 50 photons) and was detected with a superconducting single photon detector [41].

4. Experimental collapses and revivals of collective spontaneous emission

We now present experimental results of the observation of collapse and revival of collective emission. In a first experiment, we apply a positive Stark pulse after the end of the optical pulse and we observe the decay rate of the FID for different values of electric fields. The results are shown in figure 2(a). We see that the decay becomes faster when the electric field increases, due to the applied broadening. For high value of electric fields, we also observe a small revival after the FID goes to zero. This is due to the spectral distribution of the induced broadening, which approaches a square shape when the field increases. In figure 2(b), we plot the
Figure 2. (a) Decay of the FID for different values of electric fields. The electric field is switched on after the pulse and kept constant afterwards. (b) Broadening of the excited atoms as a function of the voltage applied on the electrodes. The value plotted is the FWHM of the resonance frequency distribution given by the Fourier transform of the decay curves in (a).

Figure 3. Collapse and revival of collective emission for different electric field sequences. The unperturbed FID signal is represented by the dashed line. The dashed–dotted curve represents the damped FID signal when the electric field is not switched. The voltage applied on the electrodes is ±95 V. (a) Temporary revival obtained when the polarity of the electric field is reversed at time $\tau$ and the field remains constant afterwards. (b) Revival obtained when the polarity of the electric field is reversed at time $\tau$ and the field is turned off at time $2\tau$. The revived signal then follows the unperturbed FID signal.
Figure 4. Multiple collapse and revival obtained when the polarity of the electric field is reversed repeatedly. The voltage applied on the electrodes is ±95 V.

The full-width at half-maximum (FWHM) of spectral distribution, given by the Fourier transform of the FID decay. The linear dependence confirms that the spectral distribution of the excited atoms is proportional to the applied electric field. We then show that the controlled dephasing is reversible, by switching the polarity of the Stark pulse after a time $\tau$ and observing the revival of collective emission due to the rephasing of the atoms. Different sequences of electrical pulses can be used in order to manipulate the atomic coherence in the desired manner. In figure 3, we present two examples of such sequences. If the electric field is kept constant after the switching, the atoms are in phase again after a time $2\tau$ and we observe a temporary revival of the collective emission at this time (figure 3(a)). However, if the field is switched off to zero at the time $2\tau$, the controlled phase evolution is frozen and the natural inhomogeneous dephasing governs the evolution. In that case, the revived signal follows the unperturbed FID signal (figure 3(b)). The dashed curve represents the original unperturbed FID signal. If the electric field is switched repeatedly, it is also possible to induce multiple revivals, as shown in figure 4.

In all cases it is clearly observed that the quality of rephasing is excellent, i.e. the revived FID signal reaches almost the unperturbed signal. This suggests that the process of manipulating atomic coherence using electric fields does not introduce any substantial noise which would cause additional decoherence. In order to have a more quantitative estimation of the quality
of the rephasing, we induced temporary revivals at different times. We varied the duration of the first Stark pulse, and hence the moment of the switching $\tau$. We measured the visibility $V$ of the revival as a function of $\tau$. $V$ is defined as $V = I_{\text{revival}} / I_{\text{FID}}$, where $I_{\text{revival}}$ is the maximal intensity of the revival and $I_{\text{FID}}$ is the intensity of the unperturbed FID at the corresponding time. In order to have an accurate estimation of the visibility, it is important that the revival and the reference measurement (unperturbed) FID are taken in the same experimental conditions. Since the FID intensity is very dependent on the laser intensity and to a lesser extent on the laser frequency, and that the typical measurement times are a few hundred seconds, it is difficult to ensure the same experimental conditions for the two measurements. Particularly, the drift of the laser frequency inside the inhomogeneous line can yield different results for two consecutive measurements. In order to overcome this problem, we implemented a measurement sequence with two subsequent optical excitation pulses within 10 $\mu$s. The electric field was applied for one pulse, while the other pulse served as reference. The result is shown in figure 5. We observe that the visibility stays constant above 0.95 within the error bars for all delays. In figure 5, we also plot the time of the revivals as a function of $\tau$. We measure a slope of $1.99 \pm 0.02$, which confirms that the revivals happen after a time $2\tau$.

These results show that the dephasing and rephasing of the collective atomic coherence can be controlled to a very high degree. This is a crucial capability for applications in photonic quantum storage based on CRIB. More generally, this ability to switch on and off at will the collective emission of light from the sample is an interesting resource for quantum state engineering and quantum state manipulation.
5. Conclusion

In conclusion, we have shown that the collective atomic coherence of an ensemble of erbium ions embedded in a solid state matrix can be controlled to a high degree on the timescale of tens of nanoseconds using external electric fields. Controlled inhomogeneous dephasing and rephasing was implemented using a reversible linear gradient of electric field on the crystal.

We used optical FID to test our capacity to manipulate the atomic coherence. In particular, we showed that the controlled dephasing and rephasing of the atomic dipoles results in collapses and revivals of the collective emission of light from the sample. The experimental results show that the use of the electric field does not introduce any substantial decoherence and enables us to manipulate the atomic coherence efficiently on the timescale of tens of nanoseconds. It thus provides a useful resource in quantum information science, in particular for quantum storage applications.

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Phase-noise measurements in long-fiber interferometers for quantum-repeater applications

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Many protocols for long distance quantum communication require interferometric phase stability over long distances of optical fibers. In this paper we investigate the phase noise in long optical fibers both in laboratory environment and in installed commercial fibers in an urban environment over short time scales (up to hundreds of μs). We show that the phase fluctuations during the travel time of optical pulses in long-fiber loops are small enough to obtain high visibility first-order interference fringes in a Sagnac interferometer configuration for fiber lengths up to 75 km. We also measure phase fluctuations in a Mach-Zehnder interferometer in installed fibers with arm length 36.5 km. We verify that the phase noise respects Gaussian distribution and measure the mean phase change as a function of time difference. The typical time needed for a mean phase change of 0.1 rad is of order of 100 μs, which provides information about the time scale available for active phase stabilization. Our results are relevant for future implementations of quantum repeaters in installed optical fiber networks.

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I. INTRODUCTION

Distributing quantum resources over large distances is an important experimental challenge in quantum information science [1]. One possibility to overcome losses in optical fibers is to implement a quantum-repeater architecture [2] where the total distance is split into several segments. Entanglement between neighboring nodes in each segment is created independently and stored in quantum memories. The entanglement is then extended to longer distances by entanglement swapping.

One way to create entanglement between remote material systems is by quantum interference in the detection of a single photon emitted by the atomic systems [3–9] [see Fig. 1(a)]. The remote atoms are excited in a coherent fashion and the emitted fields are combined at a beam splitter placed at a central location. If the two fields are indistinguishable, the information about the origin of the photon is erased and the detection of a single photon projects the two atomic systems in an entangled state. This type of entanglement creation is probabilistic, but heralded. It has been proposed both for single systems (e.g., single atoms [3,4] or nitrogen-vacancy centers in diamond [6,7]) and for atomic ensembles [5,8]. In the case of ensembles, the creation of entanglement by single photon detection results in entangled number states, with one joint collective atomic excitation delocalized between the remote ensembles, as has been recently demonstrated experimentally [8,9]. The advantage of this type of entanglement is that only one photon must be created, transmitted, and detected, which enables a greater probability per trial to obtain the desired entangled state, as compared to the creation of entanglement by detection of two photons [10–14]. However, the drawback of this method is that it requires interferometric stability over large distances, which is generally considered to be a challenging experimental task. Phase noise in the quantum channels acts as a decoherence in the entanglement generation, in the same way as atomic dephasing acts as a decoherence in the entanglement storage process.

Several solutions have been proposed to alleviate the phase stability problem. (i) First, the two atomic systems can be excited in a Sagnac interferometer configuration [6] [see Fig. 1(b)]. In this way, the excitation lasers for the two memories and the emitted photons travel the same path in a counterpropagating fashion. Hence as long as the phase fluctuations are slower than the travel time, the phase difference is automatically zero. (ii) The second possibility is to stabilize actively the phases, with respect to a reference laser. This requires that the phase fluctuations are not too fast in order to be able to implement a feedback loop [8]. (iii) Finally, a third possible solution has been proposed in [5] for the case of atomic ensembles, by implementing two chains of entangled memories in parallel, with the same quantum channel linking them. In that case, entanglement is generated independently in the two chains, and stored in the memories. By selecting the case where at least one excitation is present in each node, one can obtain an effective two-excitations maximally entangled state. Effective entanglement between remote quantum nodes can thus be created by asynchronous two photon quantum interference [15]. In that case, the phase of the quantum channels must be constant only during the time Δt between the detection heralding the entanglement in the two chains.

While there is an active theoretical activity in developing new quantum-repeaters architectures based on entanglement generation by single photon quantum interference [5–7,16–19], no study has addressed so far the feasibility of the implementation of such protocols in installed telecom networks with respect to phase stability. In this context, we report in this paper measurements of the phase noise in long-fiber interferometers over short time scale. Our results show that the phase in long optical fibers (several tens of km) remains stable at an acceptable level for times of order of 100 μs, in realistic environments.

The measurement of phase noise in optical fibers is also relevant for other applications. There is currently an active area of research aiming at the transmission of frequency references over large distances in optical fibers, in order to synchronize or compare remote optical-frequency atomic clocks [20–24]. In this case, the phase noise in the fiber link directly translates into a spectral broadening of the frequency refer-
The two quantum memories (QMs) are placed in the arms of a balanced Mach-Zehnder interferometer and excited with a common laser. The emitted fields are combined at a beam splitter, which erases information about the origin of the emitted photon. The detection of a single photon with detector after the beam-splitter projects the remote QM into an entangled state. In order to generate entanglement, interferometric stability must be preserved for the duration of the experiment. (b) Sagnac configuration. The excitation pulses for each QM are first reflected at the other QM, using, e.g., optical switches. In this way, the excitation lasers for the two systems and the emitted photons travel the same path in a counter-propagating way. Straight lines indicate excitation lasers and wavy lines indicate emitted fields [6].

In order to perform the phase-noise measurements, we used two interferometric techniques, namely Sagnac and Mach-Zehnder (MZ) interferometry. The Sagnac configuration allows us to study the feasibility of solving the phase stability problem by cancelling the phase fluctuations with the geometry of the interferometer. Moreover, it permits us to visualize the effects of phase fluctuations directly on the visibility of first-order interference, which can then be used as a measure of the fidelity of the quantum communication. However, with this technique we can infer the phase noise only on a time scale shorter than the travel time of the pulses in the interferometer. On the other hand, the MZ interferometer provides some information about the structure of the phase fluctuations over larger time scales. This information is important for the two other proposed techniques to alleviate the phase-noise problem, namely active stabilization and asynchronous two-photon interference. The measurements were performed both in spooled optical fibers in the laboratory, and in installed commercial fibers in an urban environment. The details of the measurements are presented in Secs. II and III and discussed in Sec. IV.

II. SAGNAC INTERFEROMETRY

Information about the phase noise of a long optical fiber can be inferred by measuring first order interference fringes in a Sagnac interferometer. In such an interferometer the two paths correspond to pulses counterpropagating in the same fiber loop. For small distances, the phase in the loop does not have the time to fluctuate during the travel time of the pulses, which leads to a zero phase difference between the two paths of the interferometer. By changing the phase difference between the two pulses (for instance using time resolved phase modulation) it is possible to obtain an interference fringe. If there is no phase noise in the interferometer during the travel time of the pulses, the visibility will be perfect. As mentioned before, this phase fluctuation cancellation technique can be exploited in order to generate remote entanglement of quantum memories by single photon interference. However, for long fibers, the phase might have time to fluctuate during the travel time of the pulses, because the pulses travel through a given segment of the fiber at different times. In that case, the visibility of the interference fringe will be reduced. Hence one can use the visibility as a measure of the phase noise over a time scale shorter than the time of propagation of the light in the interferometer. Let us now investigate in more detail the relation between visibility and phase noise.

Consider that the intensity \( I \) at the output of the interferometer is of the form

\[
I(\delta \phi) = \frac{I_0}{2} \left[ 1 + \cos(\phi + \delta \phi) \right],
\]

where \( I_0 \) is the average intensity and \( \phi \) is a constant phase given by the difference between the two arms of the interferometer. In this model the instantaneous value of intensity \( I \) is given by a particular phase fluctuation \( \delta \phi \). Next we assume that the distribution of phase fluctuations reads

\[
p(\delta \phi) = \frac{1}{\sigma \sqrt{2\pi}} e^{-\delta \phi^2/2\sigma^2},
\]

where \( \sigma \) is the width of the Gaussian distribution. The assumption of a Gaussian distribution of the phase noise will be justified by our results presented in the next section. In order to obtain some characteristic phase-noise independent value \( \langle I \rangle \) of intensity, one has to average the intensity described by Eq. (1), such that

\[
\langle I \rangle = \int_{-\infty}^{\infty} d\delta \phi p(\delta \phi) I(\delta \phi) = \frac{I_0}{2} \left[ 1 + \cos \phi e^{-\sigma^2/2} \right].
\]

We can thus find a direct relation between the visibility \( V \) and the distribution of the phase noise described by \( \sigma \)

\[
V = e^{-\sigma^2/2}.
\]
Let us now describe our experimental setup, shown in Fig. 2. The light source is a single mode distributed feedback laser diode at 1556 nm (linewidth 1 MHz). The light emitted by the laser is modulated by an integrated LiNbO$_3$ optical amplitude modulator (AM) which creates 1–10 $\mu$s rectangular light pulses with repetition rate 1 kHz. These pulses are split at the fiber beam splitter and sent to the Sagnac interferometer. We apply the phase to only one of the pulses by gating a phase modulator (PM) with a pulsed variable voltage. Finally, the interfering light pulses are combined at the fiber beam splitter and the resulting interference signal is detected using a detector and oscilloscope. The area of the pulse is then plotted as a function of the phase shift. The phase is scanned linearly, which results in a sinusoidal interference fringe. The visibility of the fringe is determined by a sinusoidal fit. All the visibilities were obtained after the subtraction of the detector noise from the signal. The phase in the modulator is varied slowly in order to sample the entire Gaussian distribution defined in Eq. (2). The typical time for scanning one fringe is chosen to be about 10 min.

We used several polarization controllers (PC) in order to ensure the optimal polarization alignment. At the same time we made sure that this alignment was quite stable over tens of minutes so that the quick intensity changes are not due to polarization fluctuations. For the experiment in the laboratory conditions, we used spools with different fiber lengths. The spools were inserted in a polystyrene box in order to prevent fast thermal fluctuations. On the other hand, for the measurement in installed fibers, our building is directly connected to the commercial Swisscom network. We had at our disposal two links connecting our building to two different telecom stations in the Geneva area, about 17.5 and 18.25 km away from our building, respectively. Each link was composed of two fibers, connected together at the telecom stations. In this way, we could use two fiber loops of 35 and 36.5 km, respectively, going to different locations. By connecting the two loops together at our building, one can form a 71.5 km long Sagnac interferometer. To connect our laboratory to the telecom network, we used about 150 m of optical fibers.

A typical example of interference fringe is shown in Fig. 3, for the 36.5 km long Sagnac interferometer in installed fibers. One can see that the signal is very stable, even for duration of order of minutes. This is important especially in the case when one needs to accumulate sufficiently large photon statistics. We investigated the dependence of visibility on the fiber length both in laboratory and real world conditions. The results are shown in Fig. 4. For short distances, the visibility is almost perfect. As expected, a slight decrease of the visibility can be observed when increasing the fiber length, due to phase fluctuations during the travel time of optical pulses. However, the visibilities remain high even for fiber lengths $\sim 75$ km. Note that in the 71.5 km interferometer in installed fibers, there is a significant change in visibility for the measurements performed during the day or the night. From the measured fringe visibilities, and attributing all the loss of visibility to phase noise, we are able to express an upper bound for the mean phase change for a given distance using Eq. (4). For instance, for the maximal distance in installed fibers $L=71.5$ km, the measured visibility during the day $V=93.6\%$ corresponds to $\sigma=0.36$ rad and the measured visibility during the night $V=98\%$ corresponds to $\sigma=0.26$ rad.
The typical distance between two adjacent datapoints was a few cm, i.e., much less than the coherence length of the laser. We used the polarization controller PC in order to optimize the signal at the output. The resulting interference signal is detected by detector D and oscilloscope OSC.

\[ \sigma = 0.2 \text{ rad for a travel time } 360 \mu s. \] One can see that the measurements performed in installed fibers in the urban environment give similar visibilities as those in the laboratory (within the error). The cases where the visibilities in the installed fibers are better than the visibilities expected in the laboratory (25 km interferometer in the laboratory and the 35 and 36.5 km interferometer in the telecom network) might be explained by global vs local disturbance: the long fibers used in laboratory were on spools and when some disturbance such as a mechanical vibration appeared, it was applied simultaneously to the whole length of the fiber whereas in the telecom network such disturbance applied only to the local part of fiber.

### III. MACH-ZEHNDER INTERFEROMETRY

As mentioned above, Sagnac interferometry allows us only to obtain information about the integrated phase noise over a time scale shorter than the travel time of a light pulse in the interferometer. In order to obtain a more quantitative analysis over longer time scales, we used Mach-Zehnder interferometry. The experimental setup is shown in Fig. 5. The two arms of the interferometer are composed by the two fiber loops connecting our laboratory to the two telecom stations. In order to balance the lengths of the two arms, we had to add a spool of 1.5 km of fiber in our laboratory. We introduced a polarization controller into one arm of the interferometer in order to align the polarization at the output coupler. Similarly to the experiment with the Sagnac interferometer, we verified that there are no quick intensity changes due to polarization fluctuations. The resulting interference signal is detected by a detector and an oscilloscope with 1 and 6 GHz bandwidth, respectively. We verified that there are no significant changes of light intensity at the detector in the time scale of few \( \mu s. \) It is thus sufficient to use a few datapoints to describe the intensity changes for this time difference without loss of information. In our case the typical distance between two adjacent datapoints was 2–4 \( \mu s, \) which is then the final temporal resolution of the measurement. We also assured that during measurement in this setup the lengths of both arms of the interferometer were equilibrated in such a way that their difference was of order of centimeters. This is much less than the coherence length of the laser (66 m in the optical fiber) corresponding to the measured bandwidth 1 MHz, and leads to negligible phase noise induced by the frequency drift of the laser. In this section we will focus only on measurements performed in the installed fibers.

If we consider two interfering light fields, we can find the dependence of the measured intensity \( I \) on time \( t \) in the form

\[
I(t) = \frac{I_{\text{max}} - I_{\text{min}}}{2} [1 + \cos \varphi(t)] + I_{\text{min}},
\]

where \( I_{\text{max}} \) and \( I_{\text{min}} \) are maximal and minimal measured intensity and \( \varphi \) is the actual phase difference between the two fields. For our purposes we call from now on this phase difference simply the phase. An example of such a measurement is shown in Fig. 6. From the measurement of \( I(t) \) and using Eq. (5) we can directly find the time dependence of the phase. However, around maxima and minima of the signal, a relatively large change in the phase causes only a small change in the intensity and thus introduces a bigger error to the data analysis. On the other hand, on the slope of a measured signal, a relatively small change of phase causes significant change in the intensity. For this reason we restricted our analysis only to this region, omitting maxima and minima; this provides us with less data but should not qualitatively change the obtained statistics. We also verified that the interference is not caused by retroreflections in the interferometer.

Once we calculated the temporal evolution of the phase, we can investigate the phase changes as a function of time. For a time difference \( \tau \) we can find a set of corresponding phase differences \( \{ \delta \varphi_j \} \), where \( \delta \varphi_j = |\varphi(t_j + \tau) - \varphi(t_j)| \), and the final phase difference as the average over \( j \), schematically:

\[
\tau: \{ \delta \varphi_1, \delta \varphi_2, \ldots \} \rightarrow \Delta \varphi_\tau.
\]

An example of dependence \( \Delta \varphi \) vs \( \tau \) is shown in Fig. 7. Actually every measurement will give a slightly different (but still monotonously rising) curve, i.e., the times needed for, e.g., a phase change of 0.1 rad will be different. The different measurements are then averaged and statistical er-
correlations can be calculated as shown later in Fig. 9. Another interesting information one can obtain from such a set of phase differences is their distribution. The distribution for a time difference corresponding to the travel time in the interferometer (τ=182 μs) is shown in Fig. 8. In the data processing to obtain this curve, we assume that long terms phase drifts are negligible on this time scale and that the phase fluctuations are random. Thus a positive (negative) intensity change is attributed to a positive (negative) phase change. Hence the Δφ are considered here without absolute value. The Gaussian fit in Fig. 8 shows that the phase-noise distribution corresponds well to a Gaussian distribution. This can be explained if we describe the phase fluctuations in terms of random walk theory [25].

Note that the Gaussian distribution [Eq. (2)] describes fully the phase noise at a given time τ. It is possible to show that there is a direct relationship between this distribution and Δφ calculated in Eq. (6), such that

One of the possibilities to quantify the quick phase changes is to fix some value of the phase difference, which can still be tolerated in the mentioned quantum communication protocols and look what is the corresponding time needed for such phase change. We took for example the value of Δφ=0.1 rad, which according to Eq. (4) corresponds to a visibility of 99.5% and we found the corresponding time intervals τ_{0.1}. This value will be further justified in the following section. We investigated how these time intervals changed in installed fibers as a function of the time of day. The results are plotted in Fig. 9. It is obvious that the phase is more disturbed during the day, than during the night, with τ_{0.1} moving from ~100 μs during the day to ~350 μs during the night. This suggests that a big part of the induced phase noise is due to vibration caused by external disturbances, e.g., traffic.

IV. DISCUSSION

In this section, we discuss in more detail the results obtained with the two interferometric methods. A first interesting point to consider is the dependence of the measured phase noise on fiber length. A common assumption is that the variance of the phase noise var(Δφ) is proportional to the length of fiber L, where the constant of proportionality is the diffusion coefficient [19],

$$\text{var}(\Delta \phi) = DL.$$  

For the measurements in the Sagnac configuration, we can express the diffusion coefficient as a function of the width σ and using Eq. (4) as a function of the visibility V.

$$D = \frac{\text{var}(\Delta \phi)}{L} = \sigma^2 \frac{1}{L} = -2 \ln(V) \frac{1}{L}.$$  

Figure 10 shows the calculated diffusion coefficient as a function of fiber length both for spooled fibers in laboratory
and for installed fibers. We observe that different fiber lengths yield the similar diffusion coefficient (of order of $D = 8 \times 10^{-4}$ rad$^2$ km$^{-1}$). This confirms the assumption that the variance of phase noise is proportional to the fiber length. As expected from Fig. 4, there is, however, a difference for the value of $D$ obtained for the 71.5 km Sagnac interferometer in installed fibers during the day and during the night. This difference might be due to two factors. (i) The phase noise is larger during the day than during the night, as shown in Fig. 9. (ii) In this analysis, all the loss of visibility is attributed to the phase noise, such that the coefficient $D$ is an upper bound. For the 71.5 km interferometer, however, the alignment of polarization was difficult due to a noisy signal. In this case, it is thus likely that a part of the visibility loss is also caused by a polarization mismatch. Note that we do not observe a significant difference between day and night for the Sagnacs interferometers of lengths 35 km and 36.5 km. This might be due to the fact that in this case the whole fiber is in the same cable.

Let us now discuss the results obtained with the Mach-Zehnder interferometer. If we consider the theory of random walk to explain the distribution of phase noise, we should obtain a histogram, i.e., a distribution of phase noise in the form of a Gaussian, see Fig. 8. At this point the theory corresponds well to the observed results. The other conclusion arising from random walk theory is that the function plotted in Fig. 7 should depend on $\tau$ as $\tau^x$ where $x = \frac{1}{2}$. In our case, we find value of $x$ ranging from 0.7 to 0.9. At this point, we do not have a model for the phase fluctuations that explains the observed dependence.

We also considered the effect of a potential slow thermal drift on the observed phase noise. A phase shift of 0.1 rad corresponds to an optical path length difference between the two arms of the interferometer of about 25 nm for a wavelength of 1550 nm. Assuming a drift constant in time (for instance due to a slow temperature variation), an optical path difference of 25 nm in 100 $\mu$s would lead to an optical path difference of 90 cm per hour. We measured the optical lengths of the two fibers with a high resolution photon counting optical time domain reflectometer [26], and found that the difference remained constant within a few cm over the course of 12 h (from 1 p.m. to 1 a.m.). This shows that the observed phase noise is most likely due to other causes than a slow thermal drift.

Finally, let us compare the results obtained with the two methods. From the phase noise $\Delta \varphi(\tau = L_{MZ}/c)$ obtained with a Mach-Zehnder interferometer with arm length $L_{MZ}$, one can predict the visibility one should obtain with a Sagnac interferometer of length $2L_{MZ}$, with the help of Eqs. (4) and (7). In our case the visibility calculated from the Mach-Zehnder interferometer of $L_{MZ} = 36.5$ km is $V = (99.0 \pm 0.7)^\%$ during the night (between 11 p.m. and 1 a.m.) and $V = (97.1 \pm 2.4)^\%$ during the day. This is compatible (within the error) with the results for the Sagnac interferometer of 71.5 km shown in Fig. 4.

V. APPLICATIONS TO QUANTUM REPEATERS

Let us now analyze our results with respect to applications in quantum repeaters. As mentioned in the Introduction, there are several ways to alleviate the phase stability problems. The Sagnac configuration presented in Sec. II is very similar to the self-aligned configuration proposed in [6] [See Fig. 1(b)]. In this setup, the two ensembles are excited by a laser pulse traveling in opposite direction in the same fiber loop. In this way, the optical path lengths for the two excitation or emission paths are identical, as long as the phase of the loop is stable for a duration equal to the travel time. Our measurement in urban environment shows that first order interferences with high visibilities can be obtained in such a configuration, for fiber lengths up to 71.5 km (corresponding to a maximal physical distance between the two nodes of 35 and 36.5 km). This shows that from the point of view of phase stability, it is feasible to generate entanglement by single photon quantum interference between two quantum memories separated by tens of km, without active stabilization. Moreover, by using the measurements of the diffusion coefficients (during the night) presented in Fig. 10 and Eq. (9), visibilities higher than 90% can be inferred for fiber lengths of order 250 km.

Alternatively, it is also possible to generate entanglement by single photon detection with a Mach-Zehnder configuration. In that case, however, the phase must be stable for the duration of the experiment, i.e., for a duration several orders of magnitude longer than the typical time of phase fluctuations. Hence in this configuration it is necessary to stabilize the phase actively. Our measurement shows that the phase remains constant at an acceptable level for quantum communication purposes (see below) for duration of order of 100 $\mu$s so that the phase noise at frequencies higher than a few tens of kHz can be neglected. Active stabilization at this frequency range seems within reach of current technology (although it remains experimentally challenging). There is currently an active experimental effort to transmit phase references over long distances in optical fibers. In this context, the distribution of a phase reference with subfemtosecond jitter over an actively stabilized fiber link of 32 km length has been recently demonstrated [22].
As mentioned in the Introduction, in order to perform quantum protocols with entangled number states involving one delocalized excitation, one possibility is to implement two chains of entangled memories in parallel and to obtain an effectively entangled state (with two excitations) by post-selection in the final stage [15]. In that case the relevant time scale for phase stability is the time between successful entanglement generations in the two chains. For standard quantum-repeater protocols based on asynchronous two photon interference, $\Delta t$ can be quite long (orders of seconds), mainly because of the small probability of success per trial and of the limited repetition rate due to the communication time [17]. This means that the phase of the quantum channels should remain stable during this time which is four orders of magnitude longer than the typical phase fluctuation time measured in our experiment ($100 \mu s$). Hence in that case, either a self-aligned configuration or an active stabilization are required. However, it is in principle possible to increase by several orders of magnitude the entanglement generation rate with spatial, spectral or temporal multiplexing. For example, in [17], it was suggested that the use of quantum memories allowing the storage of multiple temporal modes (multimode memories) could decrease the required time $\Delta t$ in order to generate successful entanglement between remote quantum nodes from seconds to tens of microseconds. Our measurement shows that the phase fluctuations on this time scale are acceptable.

Let us finally evaluate how the phase noise propagates in a quantum-repeater architecture and estimate the fidelity that might be obtained with the measured phase noise. In the following analysis for the sake of simplicity we consider only the errors caused by the phase noise. For an elementary link of quantum repeater the ideal state of two entangled memories can be written in the form [17]

$$|\psi_{\Phi}\rangle = \frac{1}{\sqrt{2}}(|10\rangle + e^{i\Phi}|10\rangle), \quad (10)$$

where $\Phi$ is the phase difference in between the two arms of the interferometer. If we consider the phase noise in optical fibers, the state for a particular phase shift $\delta \varphi$ is

$$|\psi(\delta \varphi)\rangle = \frac{1}{\sqrt{2}}(|01\rangle + e^{i(\Phi+\delta \varphi)}|10\rangle). \quad (11)$$

For phase shifts $\delta \varphi$ distributed with Gaussian distribution given by Eq. (2), the state of the entangled memories is now given by the density matrix $\hat{\rho}_{\text{real}}$

$$\hat{\rho}_{\text{real}} = \int d\delta \varphi p(\delta \varphi)|\psi(\delta \varphi)\rangle\langle \psi(\delta \varphi)|. \quad (12)$$

Fidelity can then be calculated from its definition

$$F = \langle \psi_{\text{id}}|\hat{\rho}_{\text{real}}|\psi_{\text{id}}\rangle = \frac{1}{2}(1 + e^{-\sigma^2/2}). \quad (13)$$

The important thing to point out is that if we consider not one but $N$ elementary links and the corresponding entanglement connections [5], the phase shift in Eq. (11) becomes $\delta \varphi = \delta \varphi_1 + \delta \varphi_2 + \cdots + \delta \varphi_N$ and factor $\sigma$ in Eq. (13) for fidelity becomes $\sigma^2 = \sigma_1^2 + \sigma_2^2 + \cdots + \sigma_N^2$. This is the way the errors add in the considered protocol.

As an example we consider two distant locations separated by $1000 km$, connected with eight elementary links [5,17]. We fixed a desired fidelity $F=0.9$ for the entangled state between the two distant places. The fidelity can be also expressed in terms of observed visibility as $F=(1+V)/2$. Using Eq. (4) and the fact that $\sigma^2 \sim L$, we can calculate the allowed mean phase change $\Delta \varphi_{\text{lim}}$ for the 36.5 km segment of fiber. We find $\Delta \varphi_{\text{lim}}=0.1 rad$, which motivated our choice of this value in Sec. III. Note that for experimental realizations, other factors will also contribute to the decrease of fidelity, such as dark counts, probability of generating multiple photons, distinguishability between photons, and memory errors.

**VI. CONCLUSION**

The creation of entanglement between remote quantum memories by single photon interference is attractive for quantum-repeater applications because it is much less sensitive to loss than two-photon schemes, and thus it increases the probability per trial to generate the desired entangled state. This in turns results in shorter times to distribute entanglement over long distances. However, the need for interferometric phase stability over long distances is generally considered problematic from an experimental point of view. In this context, we presented an experimental investigation of phase fluctuations over short time scales in long-fiber interferometers. Our results show that the phase remains stable at an acceptable level for quantum communication protocols for durations of order $100 \mu s$ for a 36.5 km long Mach-Zehnder interferometer in installed telecom fibers. This typical time for phase fluctuation should allow an active stabilization of the phase for long interferometers. We also showed experimentally that the phase fluctuations in installed fibers are slow enough to guarantee high visibility first-order interference for Sagnac interferometers over a length of several tens of km, without any active stabilization. This demonstrates the feasibility of observing single photon interference over these lengths, and of its use to generate entanglement between remote quantum memories without active phase stabilization. Our measurements have been performed in installed fibers in a noisy urban environment. An increase in stability is thus expected when using underwater fibers.

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Spectral hole-burning spectroscopy in Nd$^{3+}$:YVO$_4$


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We present spectral hole-burning measurements on the 879 nm, $^4I_{9/2} \rightarrow ^4F_{3/2}$ transition in Nd$^{3+}$:YVO$_4$. We observe antiholes in the spectrum along with long lived spectral holes, which demonstrates optical pumping between the ground state levels. The spectral holes are narrow (homogeneous linewidth of 63 kHz) at 2.1 K with a 300 mT applied magnetic field. We also perform preliminary spectral tailoring in this material by creating a 40 MHz wide transmission window in the inhomogeneous absorption. These results show the potential of the Zeeman levels in Nd doped materials to be used for spectral tailoring for quantum and classical information processing.

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I. INTRODUCTION

Rare-earth ion doped solids have been the subject of much investigation in the last decades because of their excellent optical coherence properties. Many applications have been proposed for these materials such as classical data storage by photon echo or spectral hole burning, radio frequency spectrum analysis, as well as laser frequency locking. These materials are now also considered for the storage of quantum states carried by single photons, in the context of quantum information science. Many protocols for storing single photons in ensembles of atoms have been proposed, e.g., electromagnetically induced transparency (EIT) and photon echo techniques based on controlled reversible inhomogeneous broadening (CRIB). Most of the schemes have in common, the need for a level structure with available optical transitions, a so-called lambda system. To initialize the memory, it is necessary to be able to efficiently transfer and durably store the population between the ground state levels, for example, using optical pumping via the excited state. For some protocols such as CRIB, it is also necessary to be able to tailor the absorption profile, for example, to isolate a narrow absorption peak within a wide transparency window. In this context, several materials have been proposed and investigated as potential materials. Three-level lambda systems with efficient optical pumping have been so far demonstrated in praseodymium, europium and thulium doped crystals. Population storage in the Zeeman sublevels of Nd$^{3+}$:LaF$_3$ has also been observed in a different context.

One of the experimental difficulties of implementing a quantum memory with rare-earth ion doped solids is that the optical transitions between the ground state and the excited states typically have small oscillator strengths. Hence, it is difficult to obtain the high optical depths required to achieve efficient storage and retrieval of photonic quantum states. Similarly, experiments based on the spontaneous Raman scattering aiming at creating entanglement between atomic ensembles in the solid state require high optical powers, making filtering of the classical beam more challenging. For these reasons, it is interesting to consider rare-earth ions with the highest possible oscillator strength for the optical transition. The most studied systems in this context are the praseodymium doped crystals which have excellent optical and hyperfine coherence times. However, these systems have a small separation between hyperfine ground states (a few megahertz). This is a strong limitation for applications that require a large spectral bandwidth. Moreover, the transition used in Pr doped crystals is at a wavelength of 600 nm, where only dye lasers which are difficult to frequency stabilize are available.

In this paper, we investigate experimentally a candidate ion, Nd$^{3+}$, for quantum storage applications. In general this ion is attractive for quantum and classical information processing because of its $^4I_{9/2} \rightarrow ^4F_{3/2}$ transition around 880 nm where diode lasers and efficient single photon detectors are available. In addition, Nd$^{3+}$ is a Kramers ion and thus has a strong first-order Zeeman effect which gives a large ground state splitting under the application of a magnetic field. This large splitting, in turn, translates to a large storage bandwidth for the quantum memory. Recent proposals for quantum repeaters would take advantage of a large storage bandwidth to speed up entanglement distribution. In particular, YVO$_4$ as a host material for Nd$^{3+}$ ions is of interest because of the high oscillator strength of the ~880 nm transition. Moreover, it has been shown via photon echo measurements that the optical coherence time of
this transition is 20 μs at magnetic fields higher than 1.5 T. We investigate the 879 nm transition in Nd3+:YVO4 using spectral hole-burning (SHB) techniques and show population storage in the Zeeman levels of the ground state leading to spectral holes that are long lived as compared to the excited-state lifetime. In addition, we demonstrate spectral holes of 125 ± 4 kHz at a temperature of 2.1 K and applied magnetic field of 300 mT. Moreover, we show that it is possible to create a wide (40 MHz) transmission pit in the inhomogeneous absorption profile by optically pumping ions between the Zeeman levels.

II. THEORETICAL BACKGROUND

In SHB spectroscopy of ions with only one ground state and one excited state, an intense laser is used to excite ions from the ground state to the excited state. The population difference can then be probed by attenuating the laser and scanning the frequency around the burning frequency. The ions in the excited state lead to a hole in the inhomogeneous absorption profile manifested as an increase in transmitted light at the corresponding transition wavelength. The width of the hole is limited by the homogeneous linewidth of the transition, spectral diffusion, and power broadening. In the case of an ion with two ground states and two excited states, for instance, corresponding to the Zeeman levels, the spectrum is more complex as the burning frequency can be in resonance with transitions from either of the two ground states to either of the two excited states. These four alternatives correspond to four different classes of ions within the inhomogeneous profile. Ions can be optically pumped via the excited state into the other ground state, leading to an increase in absorption from this other ground state (an antihole). Thus, in addition to the central hole, one expects additional holes as well as antiholes at spacings given by the Zeeman level spacings. Figure 1(a) shows the hole-burning spectrum for one class of ions, the complete spectrum for the four classes is shown in Fig. 1(b).

The YVO4 crystal is uniaxial, and Nd3+ ions substitute for Y3+ ions in sites of D3d point symmetry. In a crystal field, the ground state 4I_{9/2} splits into five Kramers doublets, only the lowest being populated at liquid helium temperatures, and the 4F_{3/2} excited state splits into two Kramers doublets. Under an applied magnetic field, the ground and excited-state doublets each split into two Zeeman levels. The optical lifetime of the 4F_{3/2} state has been measured by stimulated photon echo to be 40 μs.

In order to have efficient optical pumping and trapping in the ground state level, it is necessary to have a relaxation time between the ground state levels, which is much longer than the optical T1 lifetime of the excited state. The signature of storage in the ground state Zeeman levels is the appearance of antiholes at the ground state Zeeman splitting and a central hole with a decay time longer than the excited-state T1 lifetime.

In SHB experiments, one measures the transmitted intensity after the crystal of length L. The transmitted intensity at position L, I(L), is given by

$$I(L) = I(0)e^{-\alpha L},$$

where I(0) is the incident intensity and α is the absorption coefficient. The absorption coefficient at a frequency ω is linearly proportional to the population difference, N1 − N2, where N1 and N2 is the number of ions in the ground (excited) state at ω. Therefore, SHB measurements can be used to extract information about the dynamics of the population, in particular, if there is no population in the excited state, then the absorption coefficient is linearly proportional to the ground state population. This is the case for the measurements presented in this paper as the excited-state lifetime is much shorter than waiting time between the spectral hole-burning pulse and the probe pulse. All SHB spectra presented in this paper are the natural logarithm of the measured transmitted intensity.

III. EXPERIMENTAL SETUP AND/OR METHODS

The YVO4 single crystals doped with Nd3+ were grown by spontaneous nucleation from a Pb2V2O7 flux. Reagent grade PbO and V2O5, Nd2O3 (99.99%) and Y2O3 (99.99%) were used as starting materials in suitable amounts. The Dop−

cing concentration was 0.001% (Nd/Y nominal molar ratio). The batch was set in a 50 cm³ covered platinum crucible and heated to 1300 °C inside a horizontal furnace. After a soaking time of 12 h, the temperature was lowered to 850 °C at a rate of 3–4 °C/h, then the crucible was drawn out from the furnace and quickly inverted to separate the flux from the
crystals grown at the bottom of the crucible. The flux was dissolved by using hot diluted nitric acid. YVO$_4$ crystallizes in the $I41/amd$ space group, with cell parameters $a=b=7.118$ Å and $c=6.289$ Å and $Z=4$.\textsuperscript{30}

The experiments were performed on the 879 nm, $^4I_9/2 \rightarrow ^4F_{3/2}$ transition in a Nd$^{3+}$:YVO$_4$ crystal with a thickness of 0.9 mm along the direction of light propagation, perpendicular to the crystal $c$ axis. Light at 879 nm from a micro-lase Ti:sapphire laser (MBR-110) in continuous mode with an output power of $\sim$100 mW is modulated by an acousto-optic modulator (AOM) in a double pass setup. By modulating both the amplitude and frequency of the rf wave produced by an arbitrary wave form generator which is used to drive the AOM, we can create pulses and scan the frequency of the light. A $\lambda/2$ plate is used to control the polarization of the light on the sample.

We made measurements in two different cryostats: a He bath cryostat (Cryovac, model 100) with external Helmholtz coils which can produce a magnetic field of up to 15 mT at the sample and a He bath cryostat (Oxford instruments, Spectromag) with a superconducting magnetic that can produce a field of up to 8 T at the sample. The light was focused directly onto the sample in the cryostat and the transmitted light was focused onto a detector (Thorlabs, model PDB150A). A pickoff mirror before the cryostat directed a small percentage of the incident light onto an identical detector to normalize the signal to laser and AOM intensity fluctuations.

IV. RESULTS AND DISCUSSION

A. Spectral hole-burning mechanism

The inhomogeneous absorption spectrum was measured in transmission by slowly scanning the frequency of the Ti:sapphire laser source (see Fig. 2). The inhomogeneous broadening is roughly 2.1 GHz and the absorption is strongly polarized with $\alpha L$ of $\sim$3.7 for light polarized parallel to the crystal $c$ axis (filled circles) and $\sim$0.6 for light polarized perpendicular to the crystal $c$ axis (open circles).

B. Homogeneous linewidth

The homogeneous linewidth $\Gamma_h$, of a transition can be measured directly via spectral hole burning and the spectral hole linewidth $\Gamma_{\text{hole}}$ as $\Gamma_h = \Gamma_{\text{hole}}/2$ in the limit where there is no significant spectral diffusion and negligible laser linewidth and power broadening. The homogeneous linewidth has been previously measured to be 15 kHz at a magnetic field higher than 1.5 T using two pulse photon echoes.\textsuperscript{30}
However, spectral diffusion effects can broaden the hole considerably compared with linewidth values measured via two pulse photon echoes, where the width is measured at a much shorter time scale.\textsuperscript{32} The relevant width for a CRIB-based memory is the width of the spectral hole that can be prepared, as the storage time in the excited state is limited to the inverse of the width of the prepared spectral hole.\textsuperscript{23}

Spectral holes measured at $T$ = 2.1 K, for fields $B$ = 15 and 300 mT parallel to the crystal $c$ axis, are shown in Fig. 4. For an applied field of 15 mT, the spectral hole linewidth is 3.8 ± 0.2 MHz, whereas for an applied field of 300 mT, the spectral hole narrowed significantly to 125 ± 4 kHz, respectively. This is an upper limit on the homogeneous linewidth as we do not take into account the effect of power broadening or the laser linewidth.

The narrow holes at higher field confirm that the width of the spectral hole is reduced with the application of a field which freezes out magnetic interactions as was observed in two pulse photon echo measurements\textsuperscript{26} where a $\Gamma_h$ of 40 kHz was measured for the same applied magnetic field. The fact that via SHB we measure a value that is only slightly larger indicates that spectral diffusion does not play an important role in this sample at 300 mT for the given magnetic field dependence, 1.8 MHz/mT [see Figs. 5(a) and 5(b)]. In addition, there are weak hole and antihole with larger magnetic field dependences, 14.8 and 12.9 MHz/mT, respectively. An excited-state Zeeman $g$ factor, $g_E$ = 1.05, can be extracted from the magnetic field dependence of the weak hole (see Fig. 1), which corresponds well to published values.\textsuperscript{33} The $g$ factor of the weak antihole, 0.92, agrees with that of the Zeeman ground state splitting.\textsuperscript{33} The magnetic field dependence of the strong antihole corresponds to a $g$ factor of 0.13, which is the difference between our measured $g$ factors for the excited and ground states, as one expects from Fig. 1.

In the case of $B$ perpendicular to $c$, we observe a strong hole with a magnetic field dependence of 3.92 MHz/mT [see Figs. 5(c) and 5(d)]. There is also a broad and weak antihole structure with a larger magnetic field dependence from which it was not possible to extract a Zeeman $g$ factor. From the strong hole, one can extract an excited-state $g$ factor of 0.28. This value differs from that measured by Mehta et al.,\textsuperscript{33} 0.18 ± 0.04; however, it is closer to the value of 0.34 which they obtain by crystal field calculations including the Zeeman effect. Note that SHB spectroscopy offers a higher spectral resolution than the fluorescence spectroscopy used in those measurements. The pattern of antiholes at the Zeeman level spacings in Fig. 5, along with the appearance of the central hole at times longer than the excited-state lifetime (Fig. 3) with the application of a magnetic field, confirms that we have population trapping in the Zeeman levels.

\section*{D. Zeeman level lifetime}

In order to characterize the lifetime of the population trapping, we investigated the dynamics of the central spectral hole. After a burning pulse of 2 ms, we probed the hole with a series of readout pulses of up to 20 ms. In this way, we could measure the decay of the hole as a function of time. We performed this measurement for $B$ parallel to $c$ for temperatures from 1.7 to 4.2 K and magnetic fields from 5 to 15 mT. Over this range, we found no clear dependence of the spectral hole lifetime on the temperature or magnetic field. A typical measurement is shown in Fig. 6 for $T$ = 2.1 K and $B$ = 5 mT with a lifetime of 2.1 ± 0.4 ms. We then made a measurement at the higher magnetic field $B$ = 300 mT (at 2.1 K), as shown in Fig. 6. At this higher field, the lifetime increased to 6 ± 1 ms. Note that we observe an offset in the decay which could be due in part to a long lived component of the spectral hole. This could be due to trapping in hyperfine and superhyperfine levels, as discussed in Sec. IV A.

In the case of Zeeman levels of a paramagnetic ion such as Nd$^{3+}$, we expect spin lattice relaxation (SLR) to be an important relaxation process. The SLR rate, which is the inverse of the Zeeman state lifetime, has $T^4$ and $B^4$ dependences on the temperature and applied magnetic field, respectively.\textsuperscript{34–36} Instead, we find no temperature or magnetic field dependence of the lifetime at low fields. A larger applied $B$ field leads to an increase in the lifetime. This indicates that the limiting factor in the lifetime at low fields is not the SLR but rather that the relaxation is driven by interactions which are frozen out by the application of the large magnetic field. This observation is consistent with the behavior of the width of the spectral hole (Fig. 4), which strongly

FIG. 4. Spectral hole for applied magnetic fields $B$ = 15 mT (open circles) and 300 mT (filled circles) parallel to the crystal $c$ axis. Temperature of the sample, $T$ = 2.1 K. Points are data and lines are a fit to a Gaussian with full widths at half maximum of 3.8 ± 0.2 MHz and 125 ± 4 kHz, respectively.

C. Hole-burning spectrum

For larger frequency scans, one can see the pattern of holes and antiholes expected for a four level system with population trapping in the ground state. In order to characterize the transitions, we measured the spectrum for magnetic fields from 0 to 15 mT for the two inequivalent directions of the magnetic field ($B$ parallel to the $c$ axis and $B$ perpendicular to the $c$ axis). The spectra are shown in Fig. 5.

In the case of $B$ parallel to $c$, we observe a strong antihole close to the central hole with a relatively small magnetic field dependence, 1.8 MHz/mT [see Figs. 5(a) and 5(b)]. In addition, there are weak hole and antihole with larger magnetic field dependences, 14.8 and 12.9 MHz/mT, respectively. This is an upper limit on the homogeneous linewidth as we do not take into account the effect of power broadening or the laser linewidth.
narrowed with the larger applied magnetic field. Magnetic spin interactions between neighboring neodymium ions as well as those between neodymium and vanadium ions in the host material are possible sources for the observed Zeeman relaxation. Similar discrepancies between the expected Zeeman lifetime due to SLR and the actual measured lifetime have been reported. In general, it appears that the interactions that drive spin relaxations at low magnetic field have not been thoroughly investigated.

E. Spectral tailoring

To measure the percentage of ions trapped and to demonstrate that the Zeeman levels can be used in spectral tailoring of the absorption, we burned a wide transmission pit in the inhomogeneous absorption profile. During a burning time of 10 ms, we made 200 frequency scans of the laser over 40 MHz. Then, after a delay of 10 μs, we reduced the intensity and scanned the laser over 60 MHz in 500 μs to probe the spectrum. The pit burning was done for the case where the burning frequency is in the center of the inhomogeneous

FIG. 5. Transmission spectra for hole burning with (a) $B \parallel c$ and (c) $B \perp c$. The separation of the holes and antiholes from the central hole as a function of applied field is plotted in (b) for $B \parallel c$ and (d) for $B \perp c$. The lines are fits to the data and the $g$ values extracted are for $B \parallel c$, $g_G=0.92$, $g_E=1.05$, and $g_E-g_G=0.13$, and for $B \perp c$, $g_E=0.28$. 

FIG. 6. Area of spectral hole as a function of waiting time at $T=2.1$ K with applied magnetic fields of 5 mT (open circles) and 300 mT (filled circles). The decay is fit to an exponential with lifetimes of $2.1 \pm 0.4$ and $6 \pm 1$ ms, respectively.
energy level, for instance, a crystal field level in the ground state multiplet, using a second laser. It should also be possible to use rf waves to transfer population between excited-state Zeeman levels to improve the branching ratio.

V. CONCLUSION

We have performed spectral hole-burning spectroscopy on the 879 nm, \(^4I_{9/2} \rightarrow ^4F_{3/2}\) transition in Nd\(^3+\):YVO\(_4\) where we observe strong spectral holes under the application of a small magnetic field. The spectral holes are long lived compared with the excited-state lifetime, indicating the population trapping in the Zeeman sublevels. At low magnetic fields (5–15 mT) and temperatures from 1.7 to 4.2 K, we measure spectral holes of several megahertz and a Zeeman level lifetime of \(\sim 2\) ms which does not depend on magnetic field or temperatures in this range. In contrast, with a larger applied magnetic field (300 mT), the holes narrow considerably to 125 kHz and the Zeeman level lifetime increases to 6 ms. These results indicate that the dominant relaxation process between the Zeeman levels at low magnetic fields is not spin lattice relaxation as one would expect but a magnetic spin interaction which can be partially frozen out by the application of a moderate magnetic field.

The signatures of the Zeeman level population trapping, antiholes at the ground state splitting difference, and a long lived central hole are clearly observed. We can identify the holes and antiholes due to transitions between the four Zeeman levels of the excited state and the ground state and determine \(g\) factors for these levels. In addition, we presented preliminary spectral tailoring results by creating a 40 MHz wide transmission hole in the inhomogeneous absorption.

These results show the potential for the Zeeman levels in Nd\(^3+\) doped materials to be used to implement a three-level lambda system. The large ratio between the Zeeman level lifetime and the optical \(T_1\) lifetime is promising for efficient pumping and storage. The high absorption \((a \sim 40/\text{cm} \text{ for Nd}^{3+}:\text{YVO}_4)\) will allow for a high optical depth, which is necessary for efficient memories, with a relatively short crystal. Moreover, there are other host materials such as YAlO\(_3\) (Ref. 38) and \(\text{Y}_2\text{SiO}_5\) (YSO),\(^{39}\) where the oscillator strength of the \(^4I_{9/2} \rightarrow ^4F_{3/2}\) transition, while smaller than in YVO\(_4\), is also relatively large compared with other rare-earth ion transitions. In addition, rare-earth ions doped into YSO have been shown to have good optical coherence properties due to the low nuclear magnetic moments of the host ions.\(^{26}\) Therefore, we believe that Nd\(^3+\) doped crystals are interesting for future applications in classical and quantum information processing.

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SPECTRAL HOLE-BURNING SPECTROSCOPY

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Long-distance entanglement distribution with single-photon sources

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We present an efficient architecture for quantum repeaters based on single-photon sources in combination with quantum memories for photons. Errors inherent to previous repeater protocols using photon-pair sources are eliminated, leading to a significant gain in efficiency. The requirements on the single-photon sources and on the photon detectors are within reach of current technology.

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The entangled-state distribution over long distances is a challenging task due to the limited transmission efficiencies of optical fibers. To overcome this problem, quantum repeaters are likely to be required [1]. The basic principle of quantum repeaters consists in decomposing the full distance into shorter elementary links. Quantum memories allow the creation of entanglement independently for each link. This entanglement can then be extended to the full distance using entanglement swapping.

The protocol proposed here is similar to the well-known Duan-Lukin-Cirac-Zoller (DLCZ) scheme [2] and to its recent modification based on photon pairs and multimode memories (P²M³) [3], in that entanglement for an elementary link is created by the detection of a single photon. However, both protocols rely on sources that create correlated pairs of excitations, namely, one atomic excitation and one photon in the case of the DLCZ scheme and two photons in the case of P²M³. These correlations allow one to establish entanglement between distant memories based on the detection of a photon which could have come from either of two remote sources.

Our protocol uses single-photon sources, making it possible to eliminate errors due to two-pair emission events, which are unavoidable for Refs. [2,3]. This leads to a significant improvement in the achievable entanglement distribution rate. Moreover, our scheme is compatible with the use of multimode memories [3] and spatial and frequency multiplexing [4] which promise additional speedups.

We begin by recalling the basic principles of the P²M³ protocol. The DLCZ protocol is equivalent for the purposes of the present discussion. The architecture of an elementary link is represented in Fig. 1(a). The procedure to entangle two remote locations A and B requires one photon-pair source and one memory at each location. The pair sources are coherently excited such that each of them can emit a pair with a small probability \( p/2 \), corresponding to the state

\[
[1 + \sqrt{p/2}(a^\dagger a'^\dagger + b^\dagger b'^\dagger) + O(p)]|0\rangle;
\]  

(1)

\( a, a' \) and \( b, b' \) are the pairs of modes emitted by the sources located at A and B, respectively, and \( |0\rangle \) is the vacuum state. The modes \( a \) and \( b \) are stored in memories close to the respective sources while the modes \( a' \) and \( b' \) are sent through optical fibers to a station located halfway between A and B, where they are combined on a beam splitter. Omitting for simplicity the phase acquired by the photons during their transmission, the modes after the beam splitter are \( \bar{a} = (a' + b')/\sqrt{2} \) and \( \bar{b} = (a' - b')/\sqrt{2} \) (see Ref. [3] for a more complete discussion). The detection of a single photon in mode \( \bar{a} \), for example, creates the state \( 1/2(a^\dagger + b^\dagger)|0\rangle \), which corresponds to a single delocalized excitation.

The modes \( a \) and \( b \) are stored in memories, and the stored single excitation can be written as an entangled state \( 1/\sqrt{2}(|1_a0_b\rangle + |0_a1_b\rangle) \), where \( |0_a1_b\rangle \) and \( |1_a0_b\rangle \) denote zero and one photon, respectively, stored in the memory at A (B). This entanglement can further be extended to long distances using entanglement swapping [2,3].

The performance of the described protocol, which has many attractive features, is limited by a fundamental error mechanism. Even if the pair sources are ideal—i.e., even if they emit at most one pair each—there is a probability \( p^2/4 \) that two pairs will be emitted in total. If this is the case and if one photon is lost during its transmission through the fiber or by detector failure, the detection of a single photon in the mode \( \bar{a} \) or \( \bar{b} \) generates the state \( |1_a1_b\rangle \), corresponding to two full memories. This state, which is not the desired entangled state, introduces errors and limits the fidelity of the created entanglement. To preserve high fidelity, one has to use sources with low emission probability \( p \ll 1 \), such that the probability to get simultaneous emissions at A and B is sufficiently small. This limits the achievable distribution rate of

FIG. 1. (Color online) Schematic architecture of an elementary link connecting two locations A and B for (a) the P²M³ protocol that uses photon-pair sources [3] and (b) the new single-photon source protocol. Sources, memories, and detectors are represented by circles, squares, and half-circles, respectively. Vertical bars labeled BS denote beam splitters. For both (a) and (b), the detection of a single photon behind the central beam splitter projects the two memories into an entangled state.
entangled states. The same problem occurs for the DLCZ protocol, which, for the purpose of the present discussion, differs from the P^2M^3 protocol only by the fact that the modes a and b are created directly in the memories [2,3].

Our proposed scheme using single-photon sources is free of these fundamental errors. The architecture of our scheme is represented in Fig. 1(b). The two remote locations each contain one single-photon source and one memory. When they are excited, each of the two sources ideally creates one photon. The photons created at A and B are sent through identical beam splitters with reflection and transmission coefficients α and β satisfying |α|^2 + |β|^2 = 1, such that after the beam splitters, the state of the two photons is (|αa1 + βa'(1)|)(|ab'1 + βb'(1)|)0, which can be rewritten as

\[ |α^2a'b' + αβ(a''1b'1 + a'b''1) + β^2a'1b''1|0⟩. \]  (2)

The modes a and b are stored in memories. The modes a' and b' are coupled into optical fibers and combined on a beam splitter at a central station, with the modes after the beam splitter denoted by \( \bar{a} \) and \( \bar{b} \) as before. We are interested in the detection of one photon—for example, in the mode \( \bar{a} \). Let us consider the contributions from the three terms in Eq. (2). The term \( α^2b'1|0⟩ \), which corresponds to two full memories, cannot generate the expected detection and thus does not contribute to the entanglement creation. The term \( (a''1b'1 + a'b''1)|0⟩ \) may induce the detection of a single photon in mode \( \bar{a} \) with probability \( α^2β^2|⟨\bar{a}|b'1⟩|^2 \) \( β^2|⟨\bar{a}|b''1⟩|^2 \), \( |⟨\bar{a}|b'1⟩|^2 + |⟨\bar{a}|b''1⟩|^2 \), and \( |⟨\bar{a}|b'1⟩|^2 |⟨\bar{a}|b''1⟩|^2 \) associated with entangled memories. Note that in contrast with the P^2M^3 protocol, the entanglement creation uses correlations between modes a'-b' and a-b' rather than correlations between a-a' and b-b'. The state created by the detection of a single photon in mode \( \bar{a} \) is thus given by

\[ β^2|⟨\bar{a}|b'1⟩|^2|0⟩ + |α^2|ψ⟩⟨ψ|. \]  (3)

where \( |ψ⟩ = \frac{1}{\sqrt{2}}(|a'1b'1⟩ + |a'b''1⟩) \). The state \( |ψ⟩ \) corresponds to an entangled state of the two memories located at A and B, \( \frac{1}{\sqrt{2}}(|0_10_2⟩ + |0_11_2⟩) \), while the vacuum state \( |0⟩ \) corresponds to \( |0_10_2⟩ \); i.e., both memories are empty. We emphasize that none of the three terms in Eq. (2) leads to a component of the form \( |1_10_2⟩ \). This is a crucial difference compared to the pair-source protocols (DLCZ and P^2M^3).

The further steps are as for the DLCZ protocol [2,3]. Neighboring links are connected via entanglement swapping, creating an entangled state \( \frac{1}{\sqrt{2}}(|0_10_2⟩ + |0_11_2⟩) \) between two distant locations A and Z. Moreover, each location contains two memories, denoted \( A_1 \) and \( A_2 \) for location A, etc. Entangled states of the given type are established between \( A_1 \) and \( Z_1 \) and between \( A_2 \) and \( Z_2 \). By post-selecting the case where there is one excitation in each location, one generates an effective state of the form

\[ \frac{1}{\sqrt{2}}(|A_11_Z⟩ + |A_21_Z⟩). \]  (4)

The vacuum component in Eq. (3) does not contribute to this final state, since if one of the two pairs of memories contains no excitation, it is impossible to detect one excitation in each location. The vacuum components thus have no impact on the fidelity of the final state. This is not the case for components involving two full memories as in Refs. [2,3], which may induce one excitation in each location and thus decrease the fidelity. Note that vacuum components, which exist for the single-photon source protocol already at the level of the elementary links, occur for the pair-source protocols as well, starting after the first entanglement swapping procedure [2].

The pair-source protocols require a fixed phase relationship between the two pair sources—i.e., between the \( a''1b'1 \) and \( a'b''1 \) terms in Eq. (1) [3]. There is no equivalent requirement for the single-photon source protocol, since the phase between the \( a''1b'1 \) and \( a'b''1 \) terms in Eq. (2) depends only on the beam-splitter transformation, and not on the phase of the pump laser. It is important for all considered protocols that the photons from the two sources are indistinguishable and that the fiber lengths are stable on the time scale of the entanglement creation for an elementary link [3].

As we have indicated before, the absence of fundamental errors proportional to the entanglement creation rate leads to very significantly improved entanglement distribution rates for the single-photon source protocols. We now discuss this improvement quantitatively. The time required for a successful creation of an entangled state of the form (4) is given by [3]

\[ T_{\text{tot}} = \frac{3}{2} n + 1 \frac{1}{L_0} \frac{1}{P_0P_1 \cdots P_nP_{\text{pr}}}, \]  (5)

where \( L_0 = L/2^n \) is the length of an elementary link, L is total distance, and \( n \) is the nesting level of the repeater; \( P_0 \) is the success probability for entanglement creation in an elementary link; \( P_i \) (with \( i \geq 1 \)) is the success probability for entanglement swapping at the \( i \)th level, and \( P_{\text{pr}} \) is the probability for a successful projection onto the state Eq. (4).

For the DLCZ protocol, the success probability for entanglement creation in an elementary link is \( P_0 = πη_1η_2 \), while for the new single-photon source protocol \( P_0 = 2p_1β^2η_1η_2 \). Here \( p_1 \) is the probability that the source emits one photon \( (p_1 = 1 \) in the ideal case). The weight of the vacuum component at each nesting level is larger in the single-photon source protocol, and thus the success probabilities \( P_i \) (with \( i \geq 1 \)) for entanglement swapping are somewhat lower. However, the probability \( P_0 \) can be made much larger than in the photon-pair source protocols. Overall, this leads to higher entanglement distribution rates, as we detail now.

One can show from Eq. (5) that the total time required for the entanglement distribution with single-photon protocol is
In our examples, we have chosen $p_1=0.95$. The single-photon source protocol achieves an advantage over the DLCZ protocol as soon as $p_1>0.67$. Efficient sources are thus required for profiting from the proposed protocol; cf. below.

Our architecture is compatible with the use of multimode memories. The indicated average times for entanglement creation can thus be reduced by several orders of magnitude depending on the number of modes the memory can store [3]. Spatial and frequency multiplexing [4] could further increase the distribution rates.

We have shown that the single-photon protocol has no fundamental error mechanism; i.e., the fidelity of the created entangled states will be equal to 1 as long as all components of the architecture work perfectly. However, imperfections do affect the fidelity. A first significant imperfection comes from dark counts—i.e., detector clicks in the absence of photons. A dark count of one of the detectors located at the central station can be associated with two full memories, if the photons emitted by the two sources located at A and B are in the modes $a$ and $b$. The corresponding state $|1_A 1_B\rangle$ does not coincide with the expected entangled state and thus decreases the fidelity. With the above parameter values (for 1000 km), one finds that the probability for a detector to give a dark count has to be smaller than $4.6 \times 10^{-6}$ in order to achieve a final fidelity of $F=0.9$. This is realistic. Transition-edge sensor detectors can already resolve telecommunication photons of 4 ns duration at a repetition rate of 50 kHz, with an efficiency of 0.88 and negligible noise [6]. In the long run, NbN detectors promise to resolve even shorter pulses at higher rates [7].

Two-photon emissions cause errors that are similar in nature to those for dark counts. Such emissions might be due, e.g., to pump laser scattering. For the same values as above one finds that the probability for each source to emit two photons has to be smaller than $3.7 \times 10^{-4}$ in order to achieve a final fidelity of $F=0.9$. Single-photon sources as required for the presented protocol—i.e., with high probability of single-photon emission and low probability of two-photon emission—can be realized with a variety of approaches. For first demonstration experiments, the most promising approach may be the use of asynchronous heralded single-photon sources based on parametric down-conversion, where $p_1>0.6$ and $p_2$ of order $10^{-4}$ have already been achieved at 1.5 μm [8]. Even higher $p_1>0.8$ has been reported in Ref. [9] at 780 nm. In the long run, sources based on quantum dots [10] embedded in microcavities [11] are likely to offer higher repetition rates, which is important in order to fully profit from multimode memories. Quantum dot sources [12] and single atoms inside high-finesse cavities [13] are also potential candidates. Efficient quantum memories are essential for the implementation of the present protocol. Several approaches have been proposed and realized experimentally based on off-resonant Raman interactions [14], electromagnetically induced transparency (EIT) [15,16], or photon echo [17,18]. For EIT, efficiencies up to 45% [16] as well as single-photon storage [19] have been demonstrated with atomic gases. Rare-earth-ion-doped crystals are very attractive systems for the realization of quantum memories because of their excep-

### Table I. Average times for entanglement distribution over various distances for the DLCZ and single-photon source (SPS) protocols. The optimal nesting level $n$ and beam-splitter transmission coefficient $\beta^2$ are given. We assume high-efficiency memories and photon detectors ($\eta_m=\eta_d=0.9$). For the DLCZ protocol, the fidelity of the final state constrains the probability $p$ of photon-pair emission to be small; e.g., for $F=0.9$, one has to choose $p=0.003$ for both 1000 and 1500 km and $p=7 \times 10^{-4}$ for both 2000 and 2500 km. In the case of the single-photon source protocol, the fidelity is not fundamentally limited by the success probability of single-photon emission $p_1$, chosen to be equal to 0.95. As shown in the last column, the gain for the single-photon source protocol compared to the DLCZ protocol increases from a factor of 18 for 1000 km to a factor of 42 for 2500 km. Note that the indicated average times could further be decreased by several orders of magnitude using, e.g., multimode memories [3].

<table>
<thead>
<tr>
<th>Distance (km)</th>
<th>DLCZ (s)</th>
<th>SPS (s)</th>
<th>$n$</th>
<th>$\beta^2$</th>
<th>Gain</th>
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<td>1000 km</td>
<td>4600</td>
<td>3</td>
<td>250</td>
<td>0.11</td>
<td>18</td>
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<tr>
<td>1500 km</td>
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<td>3</td>
<td>1560</td>
<td>0.11</td>
<td>18</td>
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<tr>
<td>2000 km</td>
<td>156700</td>
<td>3</td>
<td>6000</td>
<td>0.08</td>
<td>26</td>
</tr>
<tr>
<td>2500 km</td>
<td>650000</td>
<td>4</td>
<td>15300</td>
<td>0.08</td>
<td>42</td>
</tr>
</tbody>
</table>

$$T_{\text{tot}} = \frac{3^{n+1} \prod_{k=1}^n \left[ \frac{\sqrt{2} - (\sqrt{2} - 1)p_1 \alpha^2 \eta}{\eta_d \eta_{\eta_d}^{n+3} \beta^2 \alpha^{2n+4} \eta^{n+2}} \right]}{2c \eta_{\eta_d}}.$$

Here $\eta = \eta_m \eta_d$, where $\eta_m$ is the memory efficiency, and we assume photon-number-resolving detectors with efficiency $\eta_d$; $\eta = \exp[-L_0/(2L_{\text{sat}})]$ is the fiber transmission efficiency, and $c=2 \times 10^8$ m/s is the photon velocity in the fiber. To evaluate the potential performance of our scheme, we calculate the average total time for an entangled state distribution for a distance $L=1000$ km with $L_{\text{sat}}=22$ km, corresponding to photons at the telecommunications wavelength of 1.5 μm. We assume $\eta_m = \eta_d = 0.9$. From Eq. (6) one can show that the optimal nesting level for our protocol for these parameter values is $n=3$, corresponding to eight elementary links. Assuming $p_1=0.95$ and optimizing the formula (6) over $\beta$ gives $T_{\text{tot}} = 250$ s with $\beta^2=0.11$. For the DLCZ protocol, $T_{\text{tot}}$ is also minimal for eight links. The calculation of the errors due to double-pair emission shows that in order to achieve a final fidelity $F=0.9$ one has to choose $p=0.003$, leading to $T_{\text{tot}} = 4600$ s. The new single-photon source protocol thus reduces the average time for a successful distribution of an entangled state over 1000 km by a factor of 18. This gain can be understood by comparing the values of $P_0$ for the two protocols. For the new protocol, $P_0=0.01$, whereas for the DLCZ protocol, $P_0=0.0001$. The difference between the calculated factor of 18 and the ratio between the $P_0$ values is due to the lower success probabilities for entanglement swapping.

Different distances from 1000 to 2500 km are considered, and the corresponding gain with respect to the DLCZ protocol is shown in Table I. This gain increases with distance. For example, it reaches a factor larger than 40 for 2500 km. The proposed protocol thus improves the entanglement distribution over long distances very significantly.
tionally long optical and spin coherence times (up to 6 ms [20] and 30 s [21], respectively). Storage times greater than 1 s have already been demonstrated in this system using EIT [22]. Furthermore, a memory efficiency of 13% was recently reported [18] using controlled reversible inhomogeneous broadening (CRIB). The CRIB technique is very attractive for realizing high-efficiency multimode memories [3]. The main experimental challenge consists in increasing the optical depth in order to achieve higher efficiencies—e.g., using multipass configurations.

We have proposed a quantum repeater protocol based on single-photon sources that eliminates the fundamental errors due to double-pair emission which limit the performance of previous protocols [2,3]. It is interesting to note that an important initial motivation for the development of single-photon sources was their application for the point-to-point quantum key distribution (QKD), while quantum repeaters were thought to require photon-pair sources. In fact, a high-bit-rate point-to-point QKD is achieved more conveniently using weak laser pulses with decoy-state protocols [23]. On the other hand, we have shown that single-photon sources are very promising for the implementation of efficient quantum repeaters.

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[5] The two photons bunch at the beam splitter, leading to a two-photon state in the mode $\tilde{a}$ with probability 1/2. For an overall efficiency $\eta_1\eta_2 \leq 1$, this state leads to a single detection with probability $2\eta_1\eta_2$.