Solid state quantum memory using complete absorption and re-emission of photons by tailored and externally controlled inhomogeneous absorption profiles

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Solid state quantum memory using complete absorption and re-emission of photons by tailored and externally controlled inhomogeneous absorption profiles

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Abstract

We present a scheme for storage and retrieval of the quantum state of a single-photon or a weak classical or non-classical optical wave-packet, in a solid-state medium. We describe how the spectral absorption profile of a material such as a rare-earth-ion-doped crystal can be manipulated to efficiently absorb and subsequently reemit a wave-packet, in a photon echo-like process. We discuss how the scheme can be realised experimentally and suggest candidate materials for the storage medium.

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1. Introduction

In the fields of quantum information processing and quantum optics there is a need for ways to implement storage as well as transmission of quantum states. Single photons or multi-photon wave-packets are usually considered good carriers of quantum state information, including non-classical states such as squeezed states, but they are in general not suitable for quantum state storage. Therefore, it is interesting to look for ways of mapping the states of photonic wave-packets to stationary quantum systems, such as atoms, and vice versa. Here, we present a scheme for storing the quantum state of a photon wave-packet in the long-lived ground state sublevels of an ensemble of absorbers in a solid-state medium, using a technique that is similar to techniques for pulse shape storage using photon echoes. The technique is suitable for weak or single-photon wave-packets.
because it can, in principle, offer complete reconstruction of the wave-packet and because no additional fields are present at the time the stored state is re-emitted.

The promise of high storage densities and high processing bandwidths have made all optical storage and processing of information using photon echoes a well developed field [1–3]. Reliable data storage and processing has been demonstrated, using a variety of related schemes, in gases [4] and in solids [5–8]. In these schemes, information about the phases of the incoming field is stored, but for faithful quantum state storage and recall one has to overcome the fact that the photon echo process is generally not efficient. For instance, in the case where all fields are propagating in the same direction there will be fundamental limits on the efficiency because the medium has to have a certain optical depth to ensure that the field to be stored is absorbed, but this also means that the recalled field will be partly absorbed or distorted while propagating out of the medium.

A recently proposed scheme overcomes this and uses the photon echo process for efficient quantum state storage in a gaseous medium [9]. In this scheme, the absorption process in an ensemble of absorbers is effectively reversed. This is a consequence of the fact that the Doppler broadening of a transition is effectively reversed for waves propagating in opposite directions. This enables efficient storage with high fidelity, but the storage time will be limited by the atomic movement in the gas. A scheme for obtaining an equivalent effect in solids has been proposed in [10], using spin–lattice interaction and RF-pulses to enable storage of sub-millimetre wavelength photons. In this paper, we suggest the use of an externally applied inhomogeneous field to create and control the inhomogeneous broadening of an ensemble of absorbers used to store a wave-packet.

We propose using rare-earth-ion doped inorganic crystals (RE materials) for implementing the scheme in [9] in solids. The optical transitions of the rare-earth dopants in these materials have useful properties that make them interesting for a range of applications in optical information processing and quantum optics [11–15], as well as for use as laser crystals, and many spectroscopic studies of RE materials have been performed [16]. The properties of RE materials include long optical coherence times, in some cases up to several milliseconds [17,18] and long-lived ground state hyperfine levels. The lifetime of these states is hours or days in some cases and the coherence time of the spin-/hyperfine transitions can be tens of milliseconds or more [19]. Significantly, experiments with electromagnetically induced transparency (EIT) and slow or stopped light, which are closely related to the ideas presented here, have been performed in RE materials [20,21].

Storage of light using EIT and the associated reduced group velocities was first demonstrated in atomic vapour [22] and can be analysed in terms of dark-state polaritons (form-stable coupled excitations of light and matter) [23]. There are also several other quantum state schemes based on, e.g., atomic ensembles [24–26] or atoms in cavities [27]. The scheme presented here shares some of the characteristics of EIT-based storage, such as the need for high optical densities and large reference field Rabi frequencies, but has a significant advantage, when few-photon fields are to be stored, in that the fields are separated in time.

This paper is organised as follows: First we give a brief description of the scheme for quantum state storage in a medium with a Doppler broadened transition, using a simplified version of the full quantum mechanical treatment presented in [9]. Here, we place the emphasis on the requirements and necessary properties of the medium. We then show how the same conditions can be reached in solids, specifically rare-earth doped crystals, through optical pumping and the application of a spatially varying external field. As an explicit example, we then analyse the steps for the particular case of praseodymium doped Y$_2$SiO$_5$ (Pr:YSO), a material which has been used in various quantum optical experiments [13,21]. The paper ends with a step-by-step summary of the scheme and some concluding comments.

2. General principles

The technique proposed in [9], exploits the fact that frequency shifts due to the Doppler effect are
opposite for counter-propagating fields. This makes it possible to create a rephasing process, which is a time-reverse of the process where an incoming wave-packet is absorbed. The quantum state of the wave packet is mapped onto a superposition of two close laying and slowly dephasing states of the absorbers in the medium between the absorption and reemission processes. An energy level diagram and a basic geometry for the technique are shown in Fig. 1. Transition frequencies $\omega_{12}$ and $\omega_{23}$ can typically be in the optical or infrared ranges, whereas $\omega_{13}$ should be smaller, e.g., of the order of a few GHz or less.

Initially all absorbers in the medium should be in the state $|1\rangle$. The incoming wave-packet that is to be stored, containing frequencies around $\omega_{12}$, is completely absorbed on transition $|1\rangle \rightarrow |2\rangle$.

We can write the state of a specific absorber after this first pulse as

$$\psi = \alpha |1\rangle + \beta |2\rangle,$$

where $|\beta|^2$ is the probability that this absorber has absorbed a photon from the wave-packet. The inhomogeneous (Doppler) width, $\Delta$, of the ensemble should be large enough for all spectral components of the wave-packet to be absorbed, i.e., $\Delta > \delta \omega_{wp}$, where $\delta \omega_{wp}$ is the spectral width of the wave-packet. The absorption depth, $xL$, should be large enough for the wave-packet to be completely absorbed. Also, both transitions $|1\rangle \rightarrow |2\rangle$ and $|3\rangle \rightarrow |2\rangle$ should have low decoherence and relaxation rates, compared to the total duration of the wave-packet, in order not to lose any phase or amplitude information during the optical pulses.

The absorbers will now rapidly dephase relative to each other, due to the inhomogeneous broadening of the transition. Omitting factors describing the spatial evolution, the state a time $\tau$ after the first pulse can be expressed as

$$\psi = \alpha |1\rangle + \beta |2\rangle \exp \left(-i \omega_{12} \tau \right),$$

where $\omega_{12}$ is the Doppler shifted absorption frequency of absorber $j$. Next, a short time, $\tau$, after the absorption event, a $\pi$-pulse propagating in the same direction as the wave-packet is applied to transition $|2\rangle \rightarrow |3\rangle$. This pulse can be efficient and maintain its shape throughout the medium provided that the number of absorbers in states $|2\rangle$ and $|3\rangle$ is low. The effect of the pulse is to turn the coherence on the $|1\rangle \rightarrow |2\rangle$ transition, containing the information about the quantum state of the absorbed wave-packet, into a coherence between states $|2\rangle$ and $|3\rangle$ and thus the state after the pulse can be written

$$\psi = \alpha |1\rangle + \beta |3\rangle \exp \left(-i \omega_{12} \tau \right).$$

The properties of the wave-packet have now been mapped onto the collective properties of the $|1\rangle \rightarrow |3\rangle$ transition of the ensemble of absorbers. If states $|1\rangle$ and $|3\rangle$ are Zeeman or hyperfine levels, this may be described in terms of spin coherence. The storage time, $T$, will be limited by the relaxation and dephasing rates of the transition, but in a gaseous medium the main limitation may be

![Fig. 1. (a) Energy level diagram and temporal diagram for the photon echo-like quantum state storage technique. (b) Geometry of the scheme. The figure shows the incoming and re-emitted wave-packet and the laser pulses used to move probability amplitude between the optically excited state $|2\rangle$ and the storage state $|3\rangle.](image-url)
movement of the absorbers out of the interaction region. For 1 cm diameter laser beams and typical thermal velocities, this limits the storage time to be of the order of 10 μs. The quantum state of absorber \( j \) after time \( T \) will be

\[
| \psi \rangle = \alpha | 1 \rangle + \beta' | 3 \rangle \exp (-i \omega_{12} T) \exp (-i \omega_{13} T).
\]

If the inhomogeneous broadening of transition \(| 1 \rangle \rightarrow | 3 \rangle \) is small, the last exponent will be the same for all absorbers and can be disregarded in the following.

The information is recalled by applying a second \( \pi \)-pulse (the recall pulse), also with frequency \( \omega_{23} \) but counter-propagating compared with the first \( \pi \)-pulse (right to left in Fig. 1(b)), which again moves the coherence to transition \(| 1 \rangle \rightarrow | 2 \rangle \). Because the Doppler shifts of transitions \(| 1 \rangle \rightarrow | 2 \rangle \) and \(| 3 \rangle \rightarrow | 2 \rangle \) are effectively inverted for fields propagating in opposite directions, the phases of the absorbers will now evolve such that the original dephasing of absorbers with different Doppler shifts is undone, for a field propagating in the reverse direction. The state after the recall-pulse is

\[
| \psi \rangle = \alpha | 1 \rangle + \beta'' | 2 \rangle \exp (-i \omega_{12}^{(+)} T) \times \exp (-i \omega_{12}^{(-)} (t - T - T)).
\]

Here, \( \omega_{12}^{(+)} = \omega_{12}^{0} + (1 + v_j c) \) and \( \omega_{12}^{(-)} = \omega_{12}^{0} - (1 - v_j c), \) with \( v_j \) being the velocity of the \( j \)-th atom. If relaxation can be neglected and if the \( \pi \)-pulse transfers are efficient, the probability amplitude \( \beta'' \) will be equal to the original amplitude \( \beta \), apart from a constant phase factor determined by the phases of the two \( \pi \)-pulses. Thus, a time \( T \) after the second \( \pi \)-pulse (cf. Fig. 1), absorbers with all values of the inhomogeneously broadened transition frequency \( \omega_{12}^{0} \) will re-phase throughout the medium. The rephasing of coherences in an ensemble of atoms gives rise to a macroscopic dipole moment, which causes the emission of coherent transients such a photon echoes and free induction decay. At the time of the rephasing, the quantum state of the atomic medium will be the same as immediately after the absorption of the initial wave-packet (up to a constant phase factor), except that the spectrum is now reversed. The evolution of the quantum state will be a time-reverse of the absorption process, leaving all atoms in the ground state and creating an output field that is radiated in the back direction. The amplitude of the output wave-packet exactly copies that of the absorbed wave-packet, but its temporal profile will be reversed and the spectrum will be reversed around the centre frequency [9]. In this way, the reversal of a controlled inhomogeneous broadening leads to a time-reversal of coherent interactions between an ensemble of absorbers and the electro-magnetic field, which gives a general mechanism for implementing absorption and subsequent re-emission of photonic wave-packets. In the description above, the spatial dependence of the absolute phase has been omitted. This part essentially describes the phase-matching of the wave propagation of the four fields involved in the process, taking into account the centre frequency, \( \omega_{12}^{0} \), and the time it takes for the laser pulses to propagate through the medium [9].

3. Implementing the scheme in a solid-state medium

By using carefully selected solid-state materials cooled to cryogenic temperatures it is possible to avoid some of the limitations of a gas, such as the movement of atoms out of the interaction region and changes of phase or velocity due to collisions, while still retaining useful properties such as long optical coherence times and ground state sublevels with low relaxation and dephasing rates. However, since the scheme outlined above relies on the ability to “invert” the inhomogeneous broadening of a transition, using the properties of the Doppler shift, it needs to be modified in order to be implemented in a solid medium.

Rare-earth-ion doped crystals (RE materials) can have optical transitions with homogeneous line-widths of the order of kHz or less, e.g., 50 Hz in Er\(^{3+}\):Y\(_2\)SiO\(_3\) [18]. For most materials, the ground state is also split into long-lived hyperfine levels that are suitable as storage states. Typical level splittings range from a few MHz (e.g., in Pr: YSO) through tens of MHz (e.g., in Eu\(^{3+}\):Y\(_2\)SiO\(_3\)) to several GHz (e.g., in Pr\(^{3+}\):CaF\(_2\)). Usually the narrow absorption lines and different hyperfine transitions are initially buried within a broad
(~1–100 GHz) inhomogeneous absorption line, where the broadening is caused by the fact that different ions are located in slightly different surroundings in the host. Thus, a first step is to use optical pumping to ensure that an ensemble of ions can be addressed using frequencies $\omega_{12}$ and $\omega_{23}$ to drive transitions from two ground state sub-levels to the optically excited state. By using optical pumping to transfer ions between hyperfine levels, two spectral regions around $\omega_{12}$ and $\omega_{23}$ can be completely emptied of absorbing ions. A narrow peak of ions absorbing at $\omega_{12}$ from a specific hyperfine level can then be created by pumping back ions from another hyperfine level [28,29].

An example of such an isolated group of ions in a specific hyperfine level, with zero background absorption, is shown in Fig. 2. The minimum spectral width of this absorbing peak of ions, $\Gamma_n$, will be set by the laser line-width, by the optical homogeneous line-width and possibly by inhomogeneous broadening of the hyperfine splittings. If the optically excited state is also split into hyperfine levels and the transition strengths to different levels are significantly different it may be useful to select only ions which absorb on a specific hyperfine transition, which can be done using slightly more elaborate pumping schemes [29].

By applying a static magnetic field, degenerate levels may be split into Zeeman components, which gives additional freedom in choosing the separations between the energy states that are to be used. In addition, in many materials the application of a magnetic field enhances the coherence times, by inhibiting spin flips in the host material.

The absorbing peak, created using the procedure above, will fill the role of the homogeneously broadened absorption lines of atoms in a gaseous medium and the next step will be to induce a reversible externally controlled inhomogeneous broadening of this absorption line, as will be described below. An important figure of merit, when considering losses, will be the ratio of the spectral width of the prepared peak to the width it will be broadened to. The original width of the peak will correspond to relaxation or dephasing processes that will not be compensated for, and the bandwidth that it can be broadened to will determine the maximum bandwidth of the stored wave-packet.

A controlled inhomogeneous broadening of the absorption line can be accomplished by applying an inhomogeneous external field that shifts the resonance frequencies of ions in different positions in the medium by different amounts. If the field is reversed, the resonance frequency shifts and thus the inhomogeneous absorption line, will be inverted. Many rare-earth-ions have permanent electric dipole moments when doped into a solid, and this dipole moment is different for the ground and the excited state. This causes the resonance frequency to be Stark shifted when an electric field is applied. This shift is typically of the order of $100 \text{ kHz} / (\text{V cm}^{-1})$ [30], which means that the ensemble of ions can be broadened to tens of MHz by the application of modest voltages across the crystal. An example of frequency shifting of a transition in these materials, due to the application of an electric field across the sample, is shown in Fig. 3 [31]. The field should vary longitudinally along the propagation direction of the light pulses, to ensure that all frequency components are completely absorbed within the full transverse extent of the wave-packet.
It will be desirable to induce a large spectral width, $\Gamma_{\text{ih}}^*$, of the broadened absorption line, since this will determine what bandwidth can be used for storage. However, the maximum broadening may be limited by how large absorption can be achieved. A requirement for efficient storage of an incoming wave-packet is that the absorption depth is large, $\alpha L \gg 1$, throughout the storage bandwidth, so that all of the wave-packet is absorbed. Since the absorption is due to an ensemble of ions originally absorbing in a narrow frequency interval around $\omega_{12}$ and subsequently spread out spectrally, the original absorption must be high. In order to minimise losses due to relaxation and dephasing it is desirable to have a large value of the ratio between the original width of the absorbing feature to the width of the broadened line, $\Gamma_{\text{ih}}^*/\Gamma_h^*$, but this ratio will also determine how much higher the initial absorption must be than the desired absorption when the absorbers are spread out over the storage bandwidth, $\Gamma_{\text{ih}}^*$. With a ratio $\Gamma_{\text{ih}}^*/\Gamma_h^* \approx 100$ the initial absorption should be at least $\alpha L > 300$, if we require that at least 95% of the incoming light is absorbed. RE materials with low doping concentrations, e.g., 0.05% Pr:YSO, can have an absorption exceeding 10 cm$^{-1}$, which means that the required absorption might be achieved by using a higher doping and a long sample. Alternatively, the necessary absorption depth could be achieved by using a shorter sample in a multiple-pass configuration or by using a wave-guide or (crystalline) fibre doped with rare-earth ions.

After the incoming wave-packet has been absorbed, the coherent optical excitation distributed throughout the ensemble of absorbers should be moved to the storage states by transferring all probability amplitude from state $|2\rangle$ to state $|3\rangle$, and vice versa when the wave-packet is to be recalled. This is conventionally done by applying a laser pulse with pulse area $\pi$ to the transition $|2\rangle \rightarrow |3\rangle$. The pulse should be spectrally flat over the storage bandwidth, which for a fixed frequency pulse means that it should have a duration shorter than $1/\Gamma_{\text{ih}}^*$ and at the same time it preferably should not affect ions on nearby transitions. This makes it attractive to use frequency chirped pulses that are equivalent to $\pi$-pulses \cite{32}, since these
pulses can more easily cover a specific spectral region and can be made to have less off-resonant excitation. Experiments with rapid adiabatic passage and complex hyperbolic secant pulses have also shown efficient and selective transfer of population between quantum states in RE materials [33–35]. The use of chirped pulses may also help in achieving the necessary pulse area in materials where transitions have low oscillator strengths. Many of the transitions in RE materials that may be considered are only weakly allowed and achieving a pulse area of $p$ using a short pulse may require intensities that are experimentally difficult to realise using CW lasers. The $\pi$-pulses should also have small intensity variations over the spatial extent of the region where the wave-packet has been absorbed, implying that these optical fields should have a larger area than the wave-packet in pulse 1.

If the stored wave-packet contains few or only one photon, it will be important not to excite many other ions, in addition to the ones storing the state of the absorbed wave-packet, during read-out. This is because the fluorescence from these other ions may drown the signal. In this case it may be advantageous to remove the static electric field before applying the first storage $\pi$-pulse and only apply it again (with reverse polarity) after the recalling $\pi$-pulse has been applied. In this way it will only be necessary to move population amplitude between states $|2\rangle$ and $|3\rangle$ within a narrow frequency interval of width $f_0$ around the frequency $\omega_{23}$, which makes it easier to avoid off-resonant excitation. This requires that the absorption of the weak wave-packet does not excite a significant fraction of the ions to state $|2\rangle$, because then the $\pi$-pulses can propagate essentially undisturbed through the sample as there are very few ions absorbing on transition $|2\rangle \rightarrow |3\rangle$.

If the electric field is removed before the first $\pi$-pulse and only reapplied after the recall pulse, as suggested above, the time of reemission of the stored wave-packet will be determined by the timing of the electric field. The ions will dephase during the time, $T_e$, between the absorption of the wave-packet and the removal of the field creating the inhomogeneous broadening, and will rephase again, during the recall procedure, a time $T_e$ after the reapplication of the field with opposite polarity.

4. Implementing the scheme in Pr$^{3+}$:Y$_2$SiO$_5$

We now turn to a specific example of how to realise quantum state storage, using the $^3\text{H}_4 \rightarrow ^1\text{D}_2$ transition of Pr$^{3+}$ ions in YSO. Although it is likely that there are other RE materials more suited for an implementation, Pr:YSO is a useful example because its optical properties are well studied and because many of the steps necessary for the scheme have already been demonstrated in related experiments. Some relevant material data for Pr:YSO can be found in Table 1. The homogeneous line-width is 2.4 kHz and the ground state is split into three doubly degenerate states, conventionally labeled $| \pm 1/2\rangle$, $| \pm 3/2\rangle$, and $| \pm 5/2\rangle$, separated by 10.2 and 17.3 MHz [36,37]. The lifetime of the hyperfine levels is around 100 s and the coherence time, $T_2$, is at least 0.5 ms, with a possible increase up to 80 ms if a magnetic field of appropriate orientation and strength is applied [19]. The first steps are to burn a 10–15 MHz wide pit in the absorption profile, leaving the ions to be used in state $|5/2\rangle$, and then move back a peak of ions, with a spectral distribution width less than 50 kHz, into state $|1/2\rangle$. Advanced optical pumping schemes achieving this have been successfully demonstrated, using a frequency stabilised dye laser [29]. Fig. 2 shows an absorbing peak of ions in Pr:YSO, with a linewidth of less than 100 kHz.

The next step is to broaden the absorbing feature spectrally by applying an electrical field. The Stark shift of the transition in Pr:YSO is 110 kHz/V cm$^{-1}$, which means that the application of a spatially varying field between $-25$ V and $+25$ V over a 1 cm thick crystal will cause the absorption of an ensemble of ions to broaden by 5 MHz. Due to the close laying transitions to the different hyperfine levels of the excited state (separated by 4.6 and 4.8 MHz) the largest bandwidth that can be used without complications in Pr:YSO is approximately 4 MHz.

The incoming wave-packet should now be completely absorbed by the inhomogeneously
broadened ensemble on the transition from level $|±1/2\rangle$ in the ground state to $|±3/2\rangle$ in the excited state. Assuming a ratio $\Gamma_\text{inh}/\Gamma_\text{h} = 4$ MHz/50 kHz $\approx 80$, the absorption in the prepared absorbing peak needs to be at least $2L > 240$, yielding an absorption of $\alpha L > 3$ of the broadened line. If a group of ions, absorbing on the $|±1/2\rangle$ – $|±3/2\rangle$ transition, is prepared in a Pr:YSO crystal with a Pr doping concentration of 0.05%, it can have an absorption of about $\alpha \approx 5$ cm$^{-1}$, so the total absorption length would need to be 40–50 cm. The electric field can then be reduced to zero, which means only a narrow (∼50 kHz) spectral interval needs to be transferred to the storage states, by the application of π-pulse. The pulses should be resonant with the transition between the $|±3/2\rangle$ hyperfine levels in the optically excited state and the ground state, thus storing the quantum state of the absorbed wave-packet as a superposition between levels $|±1/2\rangle$ and $|±3/2\rangle$ in the ground state. For the transition $^3\text{H}_4 \rightarrow ^3\text{D}_2$ in Pr:YSO a Rabi-frequency of ∼1 MHz can be achieved using a laser beam with 30 mW power and a diameter of 100 μm. With this, a spectral region of around 2 MHz can be inverted using a fixed frequency pulse. Applying a Gaussian pulse with a FWHM of 1 μs gives intensity variations of 1% over 100 kHz and a spectral intensity $<10^{-6}$ of the maximum intensity 4 MHz off resonance. A longer, smoothly chirped pulse can presumably give the same transfer with less off-resonant excitation. Since the hyperfine transition in Pr:YSO has an inhomogeneous broadening of up to 50 kHz it will be necessary to apply refocusing RF or Raman pulses during the storage time, if the storage time is greater than a few microseconds, cf. [38].

The stored wave-packet is recalled by again applying a π-pulse, or equivalent, to the transition between the $|±3/2\rangle$ hyperfine levels in the optically excited state and the ground state, and then reapplying the electrical field with inverted polarity.

The efficiency of an implementation using Pr:YSO will be limited by the ratio $\Gamma_\text{inh}/\Gamma_\text{h}$ that we have chosen, which puts severe constraints on the duration of the optical pulses. Optical dephasing occurs on a time-scale of $T_\text{2} = 1/\pi \Gamma_\text{h}$ and the efficiency of the rephasing process decreases according to $\exp(-4\tau/T_\text{2})$. Using pulses with a duration of ∼1 μs, the losses due to optical dephasing will be of the order of 50%. In principle, $\Gamma_\text{h}$ could be as low as a few kHz in Pr:YSO, which would lead to losses due to dephasing of only a few percent. However, in Pr:YSO this would lead to significant off-resonant excitation (see below) due to the small ground state hyperfine splittings and a proof-of-principle experiment would be easier choosing a $\Gamma_\text{h}$ of 50 kHz. Homogeneous dephasing on the storage transition contributes another factor $\exp(-2T/\hbar \delta h_2)$, where $T$ is the storage time and $\hbar \delta h_2$ is the rate of dephasing on the storage transition. In Pr:YSO this occurs on a time-scale of a ms, which will introduce a few percent losses, for a storage time of the order of a few tens of μs. In most rare-earth-ion doped materials the dephasing rates increase rapidly above a critical temperature somewhere in the range 5–10 K, but is essentially independent of temperature below this. Losses in the optical equipment, e.g., due to reflexes in cryostat windows or in-coupling losses, will further decrease the efficiency. Thus, the overall efficiency of this implementation is likely to be lower than 40%. This could still be sufficient for proof-of-principle experiments, as the efficiency of conventional optical pulse storage using photon echo techniques is limited to a few percent.

Another factor that can affect the fidelity of the storage is off-resonant excitation. The storage process occurs at frequencies inside a spectral region of width $\delta$, which has been emptied of absorbers. Outside this region will be the rest of the broad inhomogeneous absorption line, with a large

<table>
<thead>
<tr>
<th>$\lambda$</th>
<th>605.977 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Gamma_\text{inh}$</td>
<td>4.4 GHz</td>
</tr>
<tr>
<td>$\Gamma_\text{h}$</td>
<td>2.1 kHz</td>
</tr>
<tr>
<td>$T_1$</td>
<td>164 μs</td>
</tr>
</tbody>
</table>

Ground state hyper-fine level separations 10.2 MHz

Excited state hyper-fine level separations 4.6 MHz

Hyperfine level $T_1$ $\sim 100$ s
absorption. The absorption at frequencies inside the emptied region, caused by off-resonant excitation, will be

\[ a = a_i \left( 1 - \frac{2}{\pi} \tanh \frac{\delta}{\Gamma_h} \right), \]

where \( a_i \) is the absorption in the unmodified absorption line and \( \Gamma_h \) is the usual optical homogeneous linewidth. For Pr:YSO, with \( \Gamma_h = 2.4 \text{ kHz} \) and a distance of \( \sim 4 \text{ MHz} \) to the edge of the spectral pit, we obtain \( a \approx 2 \times 10^{-4} a_i \), and \( aL \approx 0.15 \). This will lead to attenuation of the storage and recall-pulses and, perhaps more importantly, the ions excited can contribute spontaneously emitted photons, of which some will be emitted at the time of and in the direction of the output field.

For achieving efficiencies above 90% other materials would need to be used. There are several materials that should allow the dephasing to be reduced to a few percent instead of several tens of percents. Since the optical homogeneous linewidths in Er:YSO or Eu:YSO can be more than 20 times narrower than in Pr:YSO, it is likely that the losses due to optical dephasing could be proportionally decreased by implementing the scheme in these materials. Further, the proposed scheme could be implemented in a holeburning material where wider spectral intervals can be emptied of absorbers, e.g., in rare-earth-ion doped materials with hyperfine splittings in the range of GHz instead of MHz. Due to the large range of available materials, we believe that it is possible to find materials where the storage efficiency is no longer limited by the properties of the storage medium.

5. Step-by-step summary of the scheme

The different steps of the scheme are depicted in Fig. 4.

1. Prepare the absorption profile using optical pumping, so that an ensemble of absorbers can be addressed on transitions \( |1 \rangle \rightarrow |2 \rangle \) and \( |3 \rangle \rightarrow |2 \rangle \), at frequencies \( \omega_{12} \) and \( \omega_{32} \), without exciting other absorbers. (Fig. 4(a)).
2. Prepare the absorbing ensemble in state \( |1 \rangle \), creating a narrow, highly absorbing peak at frequency \( \omega_{12} \). (Fig. 4(b)).
3. Apply an electric field that varies along the propagation direction, causing a Stark shift of the resonance frequencies of the absorbers and broadening the absorbing peak to the desired bandwidth. (Fig. 4(c)). The medium is now ready to store the quantum state of an incoming wave-packet.

4. An incoming photon/wave-packet, resonant with $\omega_{12}$ propagates through the medium and is completely absorbed, mapping the state of the wave-packet a coherence between states $|1\rangle \rightarrow |2\rangle$ of the absorbers.

5. The applied electric field is reduced to zero a time $T_e$ after the wave-packet entered the medium.

6. A $\pi$-pulse or an equivalent chirped pulse is applied to transition $|2\rangle \rightarrow |3\rangle$, turning the superposition between states $|1\rangle$ and $|2\rangle$ into a superposition between states $|1\rangle$ and $|3\rangle$. (Fig. 4(d)) The quantum state of the absorbed wave-packet is now stored as a coherence between the sub-levels of the optical ground state. The possible storage time is limited by spin decoherence and dephasing due to inhomogeneous broadening of the spin transition.

7. If necessary, apply $\pi$-pulses on transitions $|1\rangle \rightarrow |2\rangle$ and $|2\rangle \rightarrow |3\rangle$ (refocusing Raman pulses [38]), or RF pulses on the $|1\rangle \rightarrow |3\rangle$ transition, to flip the spin coherence and counteract the effects of dephasing due to inhomogeneous broadening of transition $|1\rangle \rightarrow |3\rangle$.

8. Apply a $\pi$-pulse or an equivalent chirped pulse, propagating in the opposite direction compared to the pulse in step 6, to the transition $|2\rangle \rightarrow |3\rangle$, thus turning the superposition between states $|1\rangle$ and $|3\rangle$ into a superposition between states $|1\rangle$ and $|2\rangle$.

9. Apply the same electric field as earlier, but with opposite polarity.

10. When the field is turned on, the absorbed wave-packet will be re-emitted, propagating in the backward direction, after a time $T_e$, which is equal to the time between the absorption of the wave-packet and the removal of the static electric field after the absorption process (steps 4 and 5).

6. Summary

We have described a technique for storing the quantum state of a single photon or the properties of a weak optical pulse, in a solid, for photons in the optical or near infrared regime. Implementation of the scheme is possible with existing technology and there is a choice of material to best fit the intended application. The possibility to tailor and manipulate the spectral absorption properties of RE materials with long-lived hyperfine levels, as described here, gives rich possibilities for applications in quantum optics or quantum information processing. In particular we believe that the presented technique, or extensions of it, may be of use in the fields of quantum cryptography [39], quantum teleportation [40] or for the storage or generation of entangled macroscopic ensembles [41].

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References