Storing quantum information in a solid using dark state polaritons

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The possibility of using a solid medium to store few-photon laser pulses as coupled excitations between light and matter is investigated. The role of inhomogeneous broadening and nonadiabaticity are considered, and conditions governing the feasibility of the scheme are derived. The merits of a number of classes of solid are examined.

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I. INTRODUCTION

In the last few years, many exciting effects in nonlinear optics have been made possible by using electromagnetically induced transparency (EIT). This allows a near-resonant probe field to experience extreme nonlinearities, while simultaneously using a second coupling field to cancel the associated absorption \[1, 2\]. Applications include nonlinear optics at low light levels \[3, 4, 5, 6\], full frequency conversion in distances so short that phase matching is not relevant \[7, 8, 9\], and quantum information storage \[10, 11\].

Another effect EIT allows is an extreme reduction of the group velocity of a light pulse \[2, 5, 12\]. This arises from the very large dispersion experienced by a probe field that is close to resonance. Slow light has been theoretically analyzed and experimentally demonstrated in gaseous media \[13\], BECs \[14\] and in solids \[15, 16\]. Group velocities of a few tens of meters per second have been achieved.

If the coupling field is allowed to become time-dependent, then the possibility of completely stopping and trapping the probe pulse arises \[11, 17, 18, 19\]. To do this one adiabatically reduces the coupling field to zero while the probe pulse is within the EIT medium. This results in the transfer of the probe pulse into a collective spin coherence between the atoms in the medium. As adiabatic passage techniques \[20\] are used, the collective atomic state storing the excitation is a "dark state", and contains no component of an upper level state which can decay. The lifetime of the dark state is thus governed by the ground state coherence dephasing rate, which can be as low as a few tens of Hertz. In the quantum picture these coupled electromagnetic and atomic excitations are best described as a single dressed state excitation which has been termed a dark state polariton by Fleischhauer and Lukin \[11\]. If the coupling field is subsequently adiabatically increased back to its original value, the spin coherence is transferred back into the electromagnetic field. The probe pulse is thus reformed and can propagate further.

As this scheme preserves the quantum state of the pulse, it allows the possibility of using such a method for quantum information storage and processing \[11\]. In addition, because the quantum state of the input pulse is mapped onto a many-atom collective excitation, it does not suffer from fundamental problems preventing the efficient coupling of a field to a single atom \[22\], and the scheme is robust and immune to many perturbing effects that can affect storage schemes utilizing single atoms in the context of cavity QED \[23, 24\].

Although a growing body of literature is beginning to consider storing classical light pulses in a solid \[15, 20\], a theoretical analysis of storing quantum information in a solid using the dark state polariton formalism has not been carried out. Such a scheme would be well worth considering, as solids have a number of advantages over gases. They are easy to prepare and store, stored information does not degrade due to atomic diffusion, and, above all much higher densities of interacting atoms can be achieved. For example, a common class of solids used within a quantum information context is rare-earth doped crystals, where the concentration of dopants can easily exceed the density of atoms in a gas by eight orders of magnitude. Outside this class of materials, nitrogen-vacancy centers in diamond have also been considered \[25, 26\]. These have the advantages of a strong oscillator strength and relatively long spin coherences. It is also conceivable that one could use doped glasses instead of crystals, although extreme inhomogeneous broadening must then be overcome.

The purpose of this paper is to extend the analysis of Fleischhauer and Lukin by considering the behavior of dark state polaritons in a solid, rather than gaseous, medium. We determine whether quantum information storage is still possible, given the large inhomogeneous broadening that is present in solids, derive conditions that must be met for successful storage, and finally discuss in which classes of systems the conditions can be met.

II. BASIC MODEL

We consider a standard three-level lambda system, as shown in Fig. \[11\]. \(E\) is a weak quantum field, while \(\Omega\) is the Rabi frequency associated with a strong classical coupling field. We assume that both fields propagate parallel to the \(z\) direction, reducing the system to a 1D problem.
Using a similar approach to \[22\], one can show that in the continuum limit the interaction Hamiltonian for this system is given by

\[
\hat{H}_{\text{int}} = -\frac{\hbar N}{L} \int dz \left[ g\sigma_{21}(z,t)\hat{E}(z,t) + \Omega(z,t)\sigma_{23}(z,t) + \text{H.c.} \right]
\]

(1)

where all quantities are taken to be slowly varying, both in time and space, i.e., we have transformed to a rotating frame. The coupling constant is given by \( g = d_{13}\sqrt{\nu/2\hbar\epsilon_0 V} \), where \( V \) is the interaction volume and \( d_{13} \) is the electronic dipole moment between states \( |1\rangle \) and \( |3\rangle \). \( N \) is the total number of dopant atoms in the interaction volume.

Within the slowly varying amplitude and phase approximation, the equation of motion for the quantum field \( \hat{E} \) is given by

\[
\left( \frac{\partial}{\partial t} + \frac{\partial}{\partial z} \right) \hat{E}(z,t) = igN\sigma_{13}(z,t)
\]

(2)

Using variables that are slowly varying in space and time one finds the atomic equations of motion for a single atom to be

\[
\dot{\sigma}_{11} = -\gamma_1\sigma_{11} + ig(\hat{E}^\dagger \sigma_{13} - \hat{E}\sigma_{31}) + F_1
\]

(3)

\[
\dot{\sigma}_{22} = -\gamma_2\sigma_{22} + i(\Omega^*\sigma_{23} - \Omega\sigma_{32}) + F_2
\]

(4)

\[
\dot{\sigma}_{33} = -\gamma_3\sigma_{33} + ig(\hat{E}\sigma_{31} - g\hat{E}^\dagger \sigma_{13}) + i\Omega\sigma_{32}^* - \Omega^*\sigma_{23} + F_3
\]

(5)

\[
\dot{\sigma}_{13} = \Gamma_1^\dagger \sigma_{13} + i(g\hat{E}(\sigma_{11} - \sigma_{33}) + \Omega\sigma_{12}) + F_{13}
\]

(6)

\[
\dot{\sigma}_{23} = \Gamma_2^\dagger \sigma_{23} + i(g\hat{E}\sigma_{31} + \Omega(\sigma_{22} - \sigma_{33})) + F_{23}
\]

(7)

\[
\dot{\sigma}_{12} = \Gamma_1^\dagger \sigma_{12} + i(\Omega^*\sigma_{13} - g\hat{E}\sigma_{32}) + F_{12}
\]

(8)

The atomic operators are defined by \( \sigma_{ij} = |i\rangle\langle j| \), \( \gamma_1 \) represents the population decay from state \( |1\rangle \), and the detunings are defined by

\[
\begin{align*}
\Gamma_{13}^\dagger &= -i\Delta_{13} - \gamma_{13} = -i(\Delta + \Delta\omega_{13}) - \gamma_{13} \\
\Gamma_{23}^\dagger &= -i\Delta_{23} - \gamma_{23} = -i(\Delta_0 + \Delta\omega_{23}) - \gamma_{23} \\
\Gamma_{12}^\dagger &= -i\Delta_{12} - \gamma_{12} = -i(\Delta - \Delta_0 + \Delta\omega_{12}) - \gamma_{12}
\end{align*}
\]

(9)

(10)

(11)

\( \gamma_{ij} \) represents the coherence decay between states \( |i\rangle \) and \( |j\rangle \); \( \Delta\omega_{ij} \) is the detuning of the inhomogeneously broadened line center from an isolated atom line center. \( \Delta \) and \( \Delta_0 \) represent the detuning of the laser from the inhomogeneous line center for the \( |1\rangle \) - \( |3\rangle \) transition and the \( |2\rangle \) - \( |3\rangle \) transition respectively.

The \( F_{ij} \) are \( \delta \)-correlated Langevin noise operators, and as such can be neglected in the adiabatic limit. We intend to remain close to this regime. It can also be noted that the magnitude of the noise correlations is related to the atomic decay via the fluctuation-dissipation theorem. As the essence of the transfer process involves utilizing dark states, ideally the upper state \( |3\rangle \) is never populated. As there is no dissipation, correlations involving the noise operators vanish. Although as one moves away from the purely adiabatic regime an admixture of the bright state with a \( |3\rangle \) component becomes excited, this component remains small, as will be seen in Section III B. Consequently we omit the writing of the noise operators in the analysis that follows.

We solve the atomic equations of motion perturbatively, using the expansion parameter \( \epsilon = g\hat{E}/\Omega \ll 1 \). Further, we assume that initially all the atoms are in state \( |1\rangle \), so that the zeroth order solutions for the atomic variables are \( \sigma_{ij}^0 = 1 \) if \( i = j = 1 \), and \( \sigma_{ij}^0 = 0 \) otherwise.

To first order in \( \epsilon \) we find

\[
\begin{align*}
\sigma_{12} &= -\frac{g\hat{E}}{\Omega} + \frac{1}{\Omega}(\partial_t - \Gamma_{13})\frac{1}{\Omega^2 + \Gamma_{12}\Gamma_{13}} g\hat{E}\Omega \\
\sigma_{13} &= \frac{ig}{\Omega}(\partial_t - \Gamma_{12}) \left[ \frac{\hat{E}\Omega}{\Omega^2 + \Gamma_{12}\Gamma_{13}} + \Gamma_{13} \frac{\partial_t}{\Omega^2 + \Gamma_{12}\Gamma_{13}} \right] \\
\sigma_{12} &= \frac{ig}{\Omega}(\partial_t - \Gamma_{12}) \left[ \frac{\hat{E}\Omega}{\Omega^2 + \Gamma_{12}\Gamma_{13}} + \Gamma_{13} \frac{\partial_t}{\Omega^2 + \Gamma_{12}\Gamma_{13}} \right]
\end{align*}
\]

(12)

(13)

These first order solutions are an excellent approximation to the true solutions, as the ratio between the probe field and the coupling field is extremely small. This can be seen by noting that within the context of quantum information storage the probe pulse will contain only a few photons, while, as we will see later, the coupling field must generally be of the order of kW/cm\(^2\) in order to overcome the inhomogeneous broadening.

The analysis above yields the relevant atomic equations of motion for a single ion with a specific detuning defined by its position within its host. As we are dealing with a collective effect, i.e., the incoming probe pulse excites many ions at different sites simultaneously, we need to average over the inhomogeneous broadening, ac-
counting for all possible detunings. Making the assumption that the inhomogeneous broadening is given by a Lorentzian, the averaged atomic quantities are given by
\[ \tilde{\sigma}_{ij} = \frac{W_{12}W_{13}}{\pi^2} \int \int \frac{d\Delta \omega_{12}}{(\Delta \omega_{12})^2 + W_{12}^2} \frac{d\Delta \omega_{13}}{(\Delta \omega_{13})^2 + W_{13}^2} \sigma_{ij} \]  
where the \( \Delta \omega_{ij} \) are given by \( |\omega| - |\Omega| \), and \( W_{12}, W_{13} \) are the inhomogeneous widths of the \(|1\rangle \rightarrow |2\rangle \) and the \(|1\rangle \rightarrow |3\rangle \) transitions respectively.

The integration results in rather involved expressions, and in order to make the analysis more tractable we make the following assumptions: Both the probe and coupling fields are on resonance with an isolated ion (that is, \( \Delta_{ij} = 0 \)), and \( \gamma_{ij} \ll W_{ij} \) (requiring that the inhomogeneous linewidth is far wider than the unBroadened linewidth). The first assumption can easily be met by choice of laser frequency, and the second is obviously met since the inhomogeneous linewidth is composed of many superimposed unbroadened linewidths. These assumptions result in the averaged atomic expressions
\[ \tilde{\sigma}_{12} = -\frac{g\tilde{E}\Omega}{\Omega^2 + W_{12}W_{13}} \left[ \frac{1}{\Omega^2} \frac{\partial}{\partial t} g\tilde{E}\Omega(-\gamma_{13}\Omega^2 + \gamma_{12}W_{13}) \right] + \frac{1}{\Omega^2} \frac{\partial}{\partial t} g\tilde{E}\Omega(-\gamma_{13}\Omega^2 + \gamma_{12}W_{13}) \]  
\[ \tilde{\sigma}_{13} = -\frac{g\tilde{E}\Omega(-\gamma_{12}\Omega^2 + \gamma_{13}W_{13})}{\Omega^2 + W_{12}W_{13}} \left[ \frac{1}{\Omega^2} \frac{\partial}{\partial t} g\tilde{E}\Omega W_{13} \right] + \frac{1}{\Omega^2} \frac{\partial}{\partial t} g\tilde{E}\Omega W_{13} \]  
\[ \tilde{\sigma}_{12} = -\frac{g\tilde{E}\Omega}{\Omega^2 + W_{12}W_{13}} \left[ \frac{1}{\Omega^2} \frac{\partial}{\partial t} g\tilde{E}\Omega(-\gamma_{13}\Omega^2 + \gamma_{12}W_{13}) \right] + \frac{1}{\Omega^2} \frac{\partial}{\partial t} g\tilde{E}\Omega(-\gamma_{13}\Omega^2 + \gamma_{12}W_{13}) \]  
\[ \tilde{\sigma}_{13} = -\frac{g\tilde{E}\Omega(-\gamma_{12}\Omega^2 + \gamma_{13}W_{13})}{\Omega^2 + W_{12}W_{13}} \left[ \frac{1}{\Omega^2} \frac{\partial}{\partial t} g\tilde{E}\Omega W_{13} \right] + \frac{1}{\Omega^2} \frac{\partial}{\partial t} g\tilde{E}\Omega W_{13} \]  
\[ \tilde{\sigma}_{12} = -\frac{g\tilde{E}\Omega}{\Omega^2 + W_{12}W_{13}} \left[ \frac{1}{\Omega^2} \frac{\partial}{\partial t} g\tilde{E}\Omega(-\gamma_{13}\Omega^2 + \gamma_{12}W_{13}) \right] + \frac{1}{\Omega^2} \frac{\partial}{\partial t} g\tilde{E}\Omega(-\gamma_{13}\Omega^2 + \gamma_{12}W_{13}) \]  
\[ \tilde{\sigma}_{13} = -\frac{g\tilde{E}\Omega(-\gamma_{12}\Omega^2 + \gamma_{13}W_{13})}{\Omega^2 + W_{12}W_{13}} \left[ \frac{1}{\Omega^2} \frac{\partial}{\partial t} g\tilde{E}\Omega W_{13} \right] + \frac{1}{\Omega^2} \frac{\partial}{\partial t} g\tilde{E}\Omega W_{13} \]  

Both \( \hat{\Psi} \) and \( \hat{\Phi} \) have bosonic commutation relations in the limit of few photons and many atoms. The action of \( \Psi^\dagger \) on the vacuum creates dark states \[11\], which contain no component of the excited state \[3\], and are therefore unaffected by spontaneous emission \[2\]. \( \hat{\Phi} \), on the other hand, creates states which couple to state \[3\] and which are therefore lossy. Consequently \( \hat{\Psi} \) and \( \hat{\Phi} \) are termed dark state and bright state polaritons respectively.

It is now clear that provided the system remains purely described by the excitation \( \hat{\Psi} \), by rotating the mixing angle from 0 to \( \pi/2 \) (equivalent to taking the control field from \( \Omega = \infty \) to \( \Omega = 0 \) one can losslessly transfer the quantum probe field into an atomic spin coherence, trapping the probe field in the medium. Ramping the control field back up rotates the spin coherence back into the probe field which is then released and able to propagate further.

Utilizing (2) along with (15)–(20), one can obtain
\[ (\partial_t + c \cos^2 \theta \partial_z) \hat{\Psi} = -\dot{\hat{\Phi}} - c \sin \theta \cos \theta \partial_z \hat{\phi} \]
\[ + g\sqrt{N} \sin \theta \left[ \frac{(-1)^2}{\Omega^2 + W_{12}W_{13}} \right] \]
\[ + \frac{W_{12}}{\Omega^2} \frac{\partial}{\partial t} \left[ \frac{\dot{\hat{E}}}{\Omega^2 + W_{12}W_{13}} \right] \]
\[ + \frac{W_{12}}{\Omega^2} \frac{\partial}{\partial t} \left( \frac{\dot{\hat{E}}}{\Omega^2 + W_{12}W_{13}} \right) \]

III. SOLUTIONS

We closely follow the analysis of Fleischhauer and Lukin \[11\].

As a starting point we introduce the two quantum fields \( \hat{\Psi} \) and \( \hat{\Phi} \). They are defined to be
\[ \hat{\Psi} = \cos \theta \hat{\Psi} - \sin \theta \sqrt{N} \hat{\sigma}_{12} \]  
\[ \hat{\Phi} = \sin \theta \hat{\Psi} + \cos \theta \sqrt{N} \hat{\sigma}_{12} \]
with the inverse relations
\[ \hat{\Psi} = \cos \theta \hat{\Psi} + \sin \theta \hat{\phi} \]  
\[ \sqrt{N} \hat{\sigma}_{12} = -\sin \theta \hat{\Psi} + \cos \theta \hat{\phi} \]

The time-dependent mixing angle \( \theta(t) \) is defined by
\[ \tan \theta = \frac{g\sqrt{N}}{\Omega(t)} \]

The first two terms on the right hand side are identical to those present in the gaseous medium considered in Ref. \[11\], where there is no inhomogeneous broadening and the ground state coherence lifetime \( \gamma_{12} \) is taken to be infinitely long. The remainder of the terms, however, are distinct to the case of a solid medium.
To obtain the final equation of motion for \( \hat{\Psi} \), we need to eliminate \( \hat{\Phi} \) from (24). This can be accomplished by using (15) and (18) to obtain

\[
\hat{\Phi} = \frac{g\sqrt{N}}{\Omega} \cos \theta \left[ \frac{\hat{E}W_{12}W_{13}}{\Omega^2 + W_{12}W_{13}} - \frac{1}{\Omega} \frac{\partial}{\partial t} \hat{E}(-\gamma_{13}\Omega^2 + \gamma_{12}W_{13}^2) + \frac{\partial}{\partial t} \hat{E}(-\gamma_{12}\Omega^2 + \gamma_{13}W_{12}^2) \right] \hat{\Psi}.
\]

Again making the replacement \( \hat{E} = \cos \theta \hat{\Psi} + \sin \theta \hat{\Phi} \) and performing the derivatives one finds the relation

\[
\hat{\Phi} = \left[ \frac{W_{12}W_{13} \sin \theta \cos \theta}{\Omega^2 + W_{12}W_{13}} - (\alpha + \beta) \right] \hat{\Psi} + \beta \cot \theta \hat{\Psi}
\]

where

\[
\alpha = \frac{\gamma_{13}\Omega^2(3W_{12}W_{13} - \Omega^2) + \gamma_{12}W_{13}^2(3\Omega^2 - W_{12}W_{13})}{(\Omega^2 + W_{12}W_{13})^3}
\]

\[
\beta = \frac{\sin^2 \theta ((\gamma_{12} + \gamma_{13})\Omega^2 - \gamma_{12}W_{13}^2 - \gamma_{13}W_{12}^2)}{(\Omega^2 + W_{12}W_{13})^2}
\]

From Eq. (25), it can be shown that to keep from populating the bright state polariton, that is to ensure that \( \hat{\Phi} \) is small relative to \( \hat{\Psi} \), we require that

\[
\Omega^2 \gtrsim 3W_{12}W_{13}.
\]

Thus to keep the bright state from being populated the strength of the control field \( \Omega \) must always dominate the inhomogeneous broadening.

If this criterion is met, we have

\[
\hat{\Phi} = \left[ \frac{W_{12}W_{13} \sin \theta \cos \theta}{\Omega^2 + W_{12}W_{13}} - (\alpha + \beta) \right] \hat{\Psi} + \beta \cot \theta \hat{\Psi}.
\]

The dynamics of the dark state polariton divide naturally into two subcases — one where the control field is altered so slowly that the evolution of the atomic states exactly follows the control field, and a second where some element of nonadiabaticity is considered.

### A. Adiabatic case

In totally adiabatic evolution, only the first term of (28) is relevant. Thus

\[
\hat{\Phi}_{\text{ad. lim.}} = \frac{g\sqrt{N}W_{12}W_{13}}{(\Omega^2 + g^2N)(\Omega^2 + W_{12}W_{13})} \hat{\Psi}
\]

Provided one remains in the regime given by (27) it is clear that \( \hat{\Phi} \) can be neglected relative to \( \hat{\Psi} \).

We note that Eq. (29) should contain a Langevin (vacuum) noise term so that the commutation relations are met. However since \( \langle \hat{\Phi}^\dagger \hat{\Phi} \rangle \ll \langle \hat{\Psi}^\dagger \hat{\Psi} \rangle \), the bright state polariton is never appreciably excited. It is thus possible to adiabatically transfer the electromagnetic probe pulse into an atomic dark state with no component projected onto the upper excited state, therefore avoiding destruction and noise due to spontaneous emission.

We are now able to derive the equation of motion for the dark state polariton in the adiabatic limit by using (28), ignoring the \( \hat{\Phi} \) terms and noting that as we are changing the control field adiabatically \( \dot{\theta} = 0 \). This yields

\[
(\partial_t + c\cos^2 \theta \partial_x) \hat{\Psi} = \frac{W_{12}W_{13} \sin^2 \theta}{\Omega^2 + W_{12}W_{13}} \hat{\Psi} - \sin^2 \theta \Gamma_\Psi \hat{\Psi}
\]

with

\[
\Gamma_\Psi = \frac{\Omega^2(\Omega^2\gamma_{12} - W_{12}^2\gamma_{13})}{(\Omega^2 + W_{12}W_{13})^2}.
\]

This result should be compared with that obtained using a gaseous medium, where no inhomogeneous broadening is present, and the ground state coherence time is taken to be infinitely long (11):

\[
(\partial_t + c\cos^2 \theta \partial_x) \hat{\Psi} = 0
\]

The first term on the right hand side of (30) is clearly a correction to the group velocity of the polariton pulse. Provided we remain in the regime given by (27) this results in a velocity correction factor close to unity.

It is equally clear that \( \Gamma_\Psi > 0 \) and so denotes a loss term. Furthermore, within the regime (27), \( \Gamma_\Psi \) is bounded by \( \gamma_{12} \), the dephasing rate of the ground state coherence. This is logical, and indicates that the maximum storage time is limited by the lifetime of the ground state coherence.

One difficulty remains: Because the power of the control field must dominate the inhomogeneous broadening, we cannot reduce it to zero in order to achieve a zero group velocity and stop the probe pulse. The minimum velocity occurs when \( \Omega^2 \approx W_{12}W_{13} \) and is given by

\[
v_g = c \cos^2 \theta \text{min} = \frac{cW_{12}W_{13}}{W_{12}W_{13} + g^2N}.
\]

Thus, in order to achieve a near-zero group velocity, one requires \( g^2 N \gg W_{12}W_{13} \).

In general, the solids of interest consist of rare-earth ion doped crystals. Consequently, \( W_{12} \) is of the order of tens of kilohertz, and \( W_{13} \) is of the order of gigahertz. \( g^2 N \), on the other hand, tends to be within a few orders of magnitude of \( \sim 10^2 \text{ Hz}^2 \). (These assumptions, along with solids other than rare-earth ions doped into crystal hosts, will be considered in greater detail in Section [14].)
on the medium chosen. Similarly, at the minimum control field strength $\sin \theta \approx 1 - W_{12} W_{13}/g^2 N$, indicating that practically all of the probe field has been transferred and stored.

This conclusion reproduces the gaseous medium result: a few-photon input pulse can have its quantum state stored as a spin coherence, provided the control field changes sufficiently slowly, and with a storage time bounded by the decay time of the ground state coherence. The group velocity of the polariton is $c \cos^2 \theta$, and approaches zero as the control field is reduced, ensuring that the pulse is slow enough to be considered stored.

To achieve this, we required two additional conditions that are a consequence of working in a solid:

$$g^2 N \gg W_{12} W_{13}$$  \hspace{1cm} (34)

$$\Omega^2 \gtrapprox 3W_{12} W_{13}.$$  \hspace{1cm} (35)

To what extent these conditions can be met in current materials will be considered in Section IV.

B. Nonadiabatic corrections

Although in principle one can modify the control field as slowly as one desires, and thus ensure that one remains in the adiabatic regime, this is not realistic for practical quantum information storage. As the storage time is bounded by the ground state coherence lifetime, one must at a minimum be able to complete a storage and retrieval operation within this period. This puts a lower bound on how slowly the control field can be turned off and on. Consequently one must determine the maximum speed at which the control field can be reduced to zero without nonadiabatic effects destroying the storage process. It is known that these nonadiabatic losses can be made negligible in a gaseous medium; we now consider whether the same can be made to hold in a solid medium.

As we wish to include first order nonadiabatic corrections, we cannot ignore all time derivatives as we did in the previous section. Making use of (28) and (23) we obtain the following equation of motion equation of motion for the dark state polariton:

$$\frac{\partial}{\partial t} + c \cos^2 \theta \frac{\partial}{\partial z} \Psi = -A(t) \Psi + B(t) c \frac{\partial}{\partial z} \Psi + C(t) c^2 \frac{\partial^2}{\partial z^2} \Psi$$  \hspace{1cm} (36)

where

$$A(t) = (1 + \gamma) \sin^2 \theta \Gamma \Psi + \dot{\theta} (\gamma \cot \theta + \gamma \tan \theta - (1 + \gamma) \delta \tan \theta - 2 \gamma^2 \cot \theta + \gamma^2 \tan \theta - 2g^2 N \gamma^2 \cot \theta \csc^2 \theta))$$

$$- \dot{\theta}^2 (1 - \gamma - \delta \tan^2 \theta)$$  \hspace{1cm} (37)

$$B(t) = -2 \gamma \cos^2 \theta - \beta \Gamma \Psi \sin^2 \theta \cos^2 \theta + \dot{\theta} \sin \theta \cos \theta (\alpha + \beta (1 + \cot^2 \theta - \delta - \gamma \cot^2 \theta))$$  \hspace{1cm} (38)

$$C(t) = \beta \cos^4 \theta$$  \hspace{1cm} (39)

and where

$$\gamma = \frac{\sin^2 \theta W_{12} W_{13}}{\Omega^2 + W_{12} W_{13}}$$  \hspace{1cm} (40)

$$\delta = \frac{W_{12} W_{13} (\Omega^2 - W_{12} W_{13})}{(\Omega^2 + W_{12} W_{13})^2}.$$  \hspace{1cm} (41)

$A(t)$ and $C(t)$ represent losses, and $B(t)$ represents a modification of the group velocity of the polariton.

To determine whether the transfer of the probe pulse into a trapped dark state can occur within an interval significantly shorter than the storage time, we must calculate how large $\theta$ can be without incurring significant nonadiabatic losses. We will follow the analysis of Ref. [11].

Eq. (36) can be solved by making the Fourier transform $\Psi(z, t) = \int dk \tilde{\Psi}(k, t) e^{-ikz}$. This gives

$$\tilde{\Psi}(k, t) = \tilde{\Psi}(k, 0) \exp \left( ik \int_0^t dt' (v_{gr}(t') - c B(t')) \right) \times \exp \left[ \int_0^t dt' (A(t') - k^2 c^2 C(t')) \right]$$  \hspace{1cm} (42)

where the first term is a group velocity correction and the last term contains the nonadiabatic losses and pulse-spreading effects we are interested in. To avoid losses, the integral in the exponent must be small relative to one. This results in the two conditions

$$\int_0^\infty dt' A(t') \ll 1$$  \hspace{1cm} (43)

$$k^2 c^2 \int_0^\infty dt' C(t') \ll 1.$$  \hspace{1cm} (44)

Since $\Omega^2 > 3W_{12} W_{13}$ we can construct an upper bound for $C(t)$ which gives

$$k^2 c^2 \gamma_{13} \int_0^\infty dt' \frac{\sin^4 \theta \cos^2 \theta}{g^2 N} \ll 1.$$  \hspace{1cm} (45)

This is identical to the condition derived for a gaseous medium, and can be shown to be equivalent to the condition that [11]

$$z \ll \frac{g^2 N}{\gamma_{13} L_p}$$  \hspace{1cm} (46)

where $L_p$ is the length of the probe pulse in the medium (i.e. after compression due to EIT effects) and $z$ is the distance the pulse travels in the medium before being completely stored, and can be seen as a lower bound on the medium length required. This condition in turn is equivalent to requiring only that the initial spectral width of the pulse before beginning deceleration fits within the initial EIT transparency window.

We turn now to the condition given by [12]. Looking at (47) the first term merely states that the polariton cannot be stored longer than the lifetime of the ground state coherence $1/\gamma_{12}$. 


Next we consider the term proportional to $\dot{\theta}^2$. We take the initial control field strength to be $\Omega_0$ and parameterize the final control field strength $\Omega(\tau)$ as $k = \Omega(\tau)/\sqrt{W_{12}W_{13}}$. Assuming a linear decrease in $\Omega$ over the time $\tau$, then integration of the term proportional to $\dot{\theta}^2$ with respect to time yields the condition

$$\tau \gg \frac{\gamma_{13}\Omega_0}{k^2(W_{12}W_{13})^{3/2}}$$

(47)

for $1 < k \lesssim 10$. This puts an upper bound on the speed one can reduce the control field field.

Finally we consider the term in $\dot{\theta}$ proportional to $\dot{\theta}$. As the integral is taken with respect to time, the presence of the $\dot{\theta}$ term ensures that there is no time dependence in the result. Thus effectively the integral is carried out with respect to $\theta$, and results in an overall loss factor. This loss factor is relatively insensitive to the precise values of all the parameters excepting the final control field strength $\Omega(\tau)$. Again if $k = \Omega(\tau)/\sqrt{W_{12}W_{13}}$ then the loss factor is approximately

$$\eta = \exp \left[ \frac{3 + 2k^2}{1 + k^2} + 2 \ln \frac{k^2}{1 + k^2} \right].$$

(48)

These are the conditions that must be met if we are to stop and store a probe pulse within a solid. Whether they can be met is strongly dependent on the solid that is used, and it is to this that we now turn.

**IV. PRACTICAL CONSIDERATIONS AND EXAMPLES**

The solids most often considered within the context of coherent optical behavior are rare-earth doped crystals. For a review of the general properties of these systems we refer to the recent paper by MacFarlane. The dopant rare-earth ions are characterized by low inhomogeneous broadening of their lower state hyperfine transitions, well-characterized energy levels, and existence of a zero-phonon line at low temperatures that coincides with the commonly used $f-d$ transitions with a reasonably large transition dipole moment but still a relatively narrow homogeneous optical width.

In addition, they are attractive for quantum information storage due to their high ratio of optical-transition inhomogeneous broadening to spin-transition inhomogeneous broadening, which allows the writing of many discrete channels via spectral hole burning and pulse compression by photon echo effects.

As has been made clear from the foregoing analysis, there are two primary quantities that govern the ability to transfer the quantum information from the probe pulse into a spin coherence. They are $W_{12}W_{13}$, the product of the inhomogeneous widths of the optical and spin transitions, and $g^2N$, the collective coupling strength of the medium.

The single-atom coupling is given by $g = d_{13}\sqrt{\nu/2\hbar c_0 V}$. The dipole moments for rare earths generally lie in the range $10^{-29} - 10^{-32}$ Cm. We choose $10^{-30}$ Cm as a representative value in the following. Assuming a wavelength of 1000 nm, we find the convenient relation that

$$g^2N [\text{Hz}^2] \sim \frac{N}{V} [\text{m}^{-3}],$$

(49)

that is, the collective coupling strength is simply given by the density of the dopant atoms in the medium. The density of rare-earths dopant ions in crystals can easily be as high as $10^{17} - 10^{19}$ cm$^{-3}$, depending on the dopant and matrix material. Thus $g^2N \sim 10^{23} - 10^{25}$ Hz$^2$, many orders of magnitude higher than what is possible in gases.

The magnitude of the optical and spin inhomogeneous broadening is strongly dependent on the rare-earth and on the electronic transition chosen. A typical range of values for $W_{13}$ is 1–10 GHz, while $W_{12}$ ranges from 100 Hz – 1 MHz. Consequently, one could expect $W_{12}W_{13} \sim 10^{15}$ Hz$^2$, and it is therefore clear that the condition $g^2N \gg W_{12}W_{13}$ is very easily met in these materials.

We now consider the power requirements of the coupling laser. If we wish to let the probe pulse enter the medium at speed $c$, and then reduce the coupling field strength to its minimum value, effectively stopping the pulse, it is necessary to rotate the mixing angle $\theta$ from 0 to $\pi/2$. A value of $\theta = 0$ corresponds to a coupling field of infinite intensity, or more realistically, $\Omega^2 \gg g^2N$. Optimistically assuming $W_{12}W_{13} = 10^{15}$ Hz$^2$, and $g^2N = 10^{17}$ Hz$^2$, we might require $\Omega^2(0) = 10^{19}$ Hz$^2$. Using

$$I = \frac{\Omega^2h^2c_0}{2d_{13}^2}$$

(50)

and assuming a dipole moment of $d_{13} = 10^{-30}$ Cm we see that this corresponds to a coupling laser intensity of 10 kW/cm$^2$. These numbers are only indicative, and it is possible to reduce the power requirements by choosing systems with smaller inhomogeneous broadening and reducing the dopant concentration.

One must also take into consideration the length of medium required to stop the pulse. Naively, if the coupling laser intensity is reduced from $\Omega(0) \gg g\sqrt{N}$ to its minimum value $\Omega(\tau) \sim \sqrt{W_{12}W_{13}}$ in time $\tau$, the distance the pulse travels is

$$z = \int_0^\tau c \cos^2 \theta(t) dt \approx c\tau.$$

(51)

Thus, bearing in mind the adiabaticity requirements, if $\tau \sim 10^{-6}$ s, the stopping distance is 300m. This may just be feasible for an experiment with a doped fiber, but certainly not for a crystal.

The correct approach, which obviates this difficulty as well as reducing the pump power required, is to ensure the coupling field has a strength such that the probe pulse is in the slow group velocity regime as soon as it enters the medium, namely $W_{12}W_{13} \ll \Omega(0)^2 \ll g^2N$. In this
As it is possible to make $g^2N$ extremely large in a solid, there is no difficulty in stopping the probe pulse within a few centimeters. The initial coupling laser Rabi frequency can now be orders of magnitude lower, provided it still dominates the inhomogeneous broadening, resulting in an initial coupling laser intensity of $\sim 100\ \text{W/cm}^2$.

The final conditions that must be met are the adiabaticity criteria, namely \cite{40, 47 and 48}, which exist only when one moves away from the adiabatic limit.

The first condition is that probe pulse bandwidth after entry to the medium must be within the EIT transparency window before the coupling laser intensity is reduced. In the case of a strong coupling field in a solid, the EIT window is given by $\Gamma_{EIT} = \Omega^2/W_{13}$, corresponding to a bandwidth of $\sim 10^8 - 10^9\ \text{Hz}$, depending on the initial coupling field strength. If a broader bandwidth is required, one merely needs to increase the coupling field strength.

Eq. (47) is a fairly weak condition. Using the numbers $W_{12}W_{13} \sim 10^{15}\ \text{Hz}^2$, $\Omega(0)^2 \sim 10^{17}\ \text{Hz}^2$ and $\gamma_{13} \sim 10^7\ \text{Hz}$ one obtains $\tau \gg 10^{-7}\ \text{s}$, which is certainly still orders of magnitude shorter than the limit of the storage time which is governed by $1/\gamma_{12}$.

The final condition \cite{45} is highly dependent on to what extent the final control field strength dominates the inhomogeneous broadening. Taking $\Omega(\tau) = 3\sqrt{W_{12}W_{13}}$ gives a damping factor of $\eta \approx \exp[-0.0007]$, which is negligible.

Thus, in broad, it appears that quantum information storage using this technique is feasible in rare-earth doped crystals. It is not clear, however, that there is one type of material that possesses all the properties that would make it an ideal candidate. The oscillator strength of the rare-earth itself is not too important, as it can generally be compensated for by altering the dopant density. The crucial quantities are the inhomogeneous broadening widths $W_{12}$ and $W_{13}$, the collective coupling $g^2N$ and the dephasing rate $\gamma_{12}$. The ideal material would have a low $W_{12}W_{13}$, a high $g^2N$, and a very low $\gamma_{12}$.

Some measures can be taken to reduce $W_{13}$. For example, Ham et al. introduce a repump laser to prevent spectral hole-burning by the coupling and probe lasers, and consequently limit the optical inhomogeneous broadening to the repump laser jitter ($\sim 1\ \text{MHz}$) \cite{31}. In the scheme described in this paper, we have assumed very weak probe fields for quantum information purposes, and so only a tiny fraction of the atoms make the transition to state $|2\rangle$, rendering such a repump laser unnecessary. Similar spectral hole-burning techniques, however, could be used prior to applying the probe pulse, selecting a subset of the ions within a particular spectral range \cite{25} and thus drastically reducing $W_{13}$. The drawback is a reduction in the interacting ion density, but as $g^2N \sim 10^{23}\ \text{Hz}^2$ is attainable, reducing the density by a factor of 1000 is certainly acceptable for a similar 1000-fold reduction in the inhomogeneous broadening.

The storage time, that is the time one may wait before the quantum field is released by the turning on of the strong pump field, is limited both by the homogeneous width $\gamma_{12}$ and the inhomogeneous width $W_{12}$ of the lower hyperfine transition in the ions. $\gamma_{12}$ serves as an absolute lower limit as discussed in the previous section. $W_{12}$ is a limit due to the fact that the phases of different ions evolve at different speeds due to inhomogeneity, meaning that after a time $1/W_{12}$ the stored information will no longer be coherent. In principle this can be overcome. Spin echo techniques have been exploited to compensate inhomogeneous frequency shifts and to observe features limited only by the homogeneous lower state width in ion doped crystals \cite{30}. In our case, however, it is not plausible that one can precisely invert the populations to a level of precision matching the almost insignificant number of photons stored in the medium. Thus in practice one is limited by the storage time $1/W_{12}$ rather than the longer storage time $1/\gamma_{12}$. It has been shown, however, that strong magnetic bias fields can reduce the inhomogeneous broadening significantly, and suggests that storage times of the order of 100 ms or more may be achievable \cite{32}.

For integration with current telecommunication technologies, it is natural to speculate about the possibilities to slow and store light in doped optical fibres and waveguides rather than crystals. In fibres and wave guides, where the ions are doped into a glass host, the inhomogeneous widths of both the optical and the hyperfine transitions are much larger than in crystals \cite{32}. Persistent hole burning has been demonstrated in glass fibres \cite{56}, and a natural strategy thus seems to be a preparation of the system by pumping all ions in a broad frequency range to passive spectator levels, leaving only ions which have their $1 \rightarrow 3$ and $1 \rightarrow 2$ transitions in desirable frequency windows in the middle of this range in their state $|1\rangle$. Considerable improvement of the hole burning must be achieved and further understanding of the homogeneous width of the transitions is clearly needed before serious attempts along this line can be carried out. As commented upon above, hole burning leads to a significant reduction of the number of ions available for the light storage. A crystal fibre may be doped only in the central rod which forms the central wave guide in the fibre \cite{57}. The light may thus be confined to a cross section about the size of the (resonant) absorption cross section of a single ion which, together with the achievable lengths of these fibres, may compensate for the low concentration, and make slowing and storage of light possible. It would also be possible to set up an optical cavity by writing a Bragg grating in the fibre \cite{58} (or by coating the faces of the crystals in our main proposal) and in this way enhance the interaction of the field with the atomic system, as it has been proposed for free atoms and for ions \cite{59}. An analysis of this proposal lies beyond the purpose of the present paper. Significant
non-linear dynamics, for example super-continuum generation, has been observed in crystal fibres at very high light intensities as a consequence of the non-linear susceptibility of the glass host [40]. For the above analysis to hold, one should avoid this parameter regime, but we do not exclude the possibility that useful effects may be derived from the optical nonlinearity of the host material in conjunction with the EIT dynamics due to the dopant ions.

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