

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 [3–6], full frequency conversion in distances so short that phase matching is not relevant [7–9], 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 [11,17–20]. 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 [21] 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 uti-

lizing 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-metal-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 Λ system, as shown in Fig. 1. \hat{E} is a weak quantum field, while Ω 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 one-dimensional (1D) problem.

Using a similar approach to [27], one can show that in the continuum limit the interaction Hamiltonian for this system is given by

$$\hat{H}_{\text{int}} = -\frac{\hbar N}{L} \int dz [g\sigma_{21}(z,t)\hat{E}(z,t) + \Omega(z,t)\sigma_{23}(z,t) + \text{H.c.}], \quad (1)$$

where all quantities are taken to be slowly varying, both in time and space, i.e. we have transformed to a rotating frame.

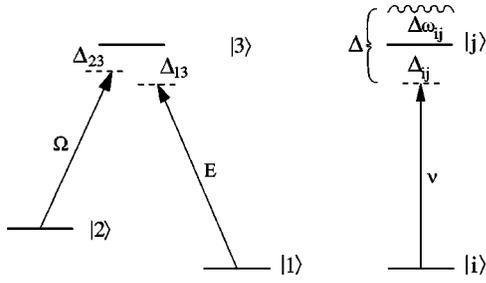


FIG. 1. Left: three-level scheme. \hat{E} is a weak quantum field while Ω is a classical field. Right: detuning scheme for a particular atom. The wavy line represents the inhomogeneous line center for the atomic ensemble, thus $\Delta\omega_{ij}$ represents the detuning between the upper level of a particular atom and the inhomogeneous line center.

The coupling constant is given by $g = d_{13}\sqrt{v/2\hbar}\epsilon_0V$, 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

$$\left(\frac{\partial}{\partial t} + c\frac{\partial}{\partial z}\right)\hat{E}(z,t) = igN\sigma_{13}(z,t). \quad (2)$$

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

$$\dot{\sigma}_{11} = -\gamma_1\sigma_{11} + ig(\hat{E}^\dagger\sigma_{13} - \hat{E}\sigma_{31}) + F_1, \quad (3)$$

$$\dot{\sigma}_{22} = -\gamma_2\sigma_{22} + i(\Omega^*\sigma_{23} - \Omega\sigma_{32}) + F_2, \quad (4)$$

$$\dot{\sigma}_{33} = -\gamma_3\sigma_{33} + i(g\hat{E}\sigma_{31} - g\hat{E}^\dagger\sigma_{13} + \Omega\sigma_{32} - \Omega^*\sigma_{23}) + F_3, \quad (5)$$

$$\dot{\sigma}_{13} = \Gamma'_{13}\sigma_{13} + i[g\hat{E}(\sigma_{11} - \sigma_{33}) + \Omega\sigma_{12}] + F_{13}, \quad (6)$$

$$\dot{\sigma}_{23} = \Gamma'_{23}\sigma_{23} + i[g\sigma_{21}\hat{E} + \Omega(\sigma_{22} - \sigma_{33})] + F_{23}, \quad (7)$$

$$\dot{\sigma}_{12} = \Gamma'_{12}\sigma_{12} + i(\Omega^*\sigma_{13} - g\hat{E}\sigma_{32}) + F_{12}. \quad (8)$$

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

$$\Gamma'_{13} = -i\Delta_{13} - \gamma_{13} = -i(\Delta + \Delta\omega_{13}) - \gamma_{13}, \quad (9)$$

$$\Gamma'_{23} = -i\Delta_{23} - \gamma_{23} = -i(\Delta_0 + \Delta\omega_{23}) - \gamma_{23}, \quad (10)$$

$$\Gamma'_{12} = -i\Delta_{12} - \gamma_{12} = -i(\Delta - \Delta_0 + \Delta\omega_{12}) - \gamma_{12}. \quad (11)$$

γ_{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. Δ and Δ_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 δ -function-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 Sec. 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 ϵ we find

$$\sigma_{12} = -\frac{g\hat{E}}{\Omega} + \frac{1}{\Omega}(\partial_t - \Gamma_{13})\frac{1}{\Omega}(\partial_t - \Gamma_{12})\frac{g\hat{E}\Omega}{\Omega^2 + \Gamma_{12}\Gamma_{13}}, \quad (12)$$

$$\sigma_{13} = \frac{ig}{\Omega}(\partial_t - \Gamma_{12})\frac{\hat{E}\Omega}{\Omega^2 + \Gamma_{12}\Gamma_{13}} + \frac{ig}{\Omega}(\partial_t - \Gamma_{12}) \times \left[\frac{\Gamma_{13}}{\Omega^2}\partial_t\frac{\hat{E}\Omega}{\Omega^2 + \Gamma_{12}\Gamma_{13}} + \frac{\Gamma_{12}}{\Omega}\partial_t\frac{\hat{E}}{\Omega^2 + \Gamma_{12}\Gamma_{13}} \right]. \quad (13)$$

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/cm² 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, accounting for all possible detunings. Making the assumption that the inhomogeneous broadening is given by a Lorentzian, the averaged atomic quantities are given by

$$\bar{\sigma}_{ij} = \frac{W_{12}W_{13}}{\pi^2} \int \int \frac{d\Delta\omega_{12}}{(\Delta\omega_{12})^2 + W_{12}^2} \frac{d\Delta\omega_{13}}{(\Delta\omega_{13})^2 + W_{13}^2} \sigma_{ij}, \quad (14)$$

where the $\Delta\omega_{ij}$ are given by Eqs. (9)–(11), 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

$$\bar{\sigma} = -\frac{g\hat{E}\Omega}{\Omega^2 + W_{12}W_{13}} - \frac{1}{\Omega^2} \frac{\partial}{\partial t} \frac{g\hat{E}\Omega(-\gamma_{13}\Omega^2 + \gamma_{12}W_{13}^2)}{(\Omega^2 + W_{12}W_{13})^2} - \frac{1}{\Omega} \frac{\partial}{\partial t} \frac{g\hat{E}(-\gamma_{12}\Omega^2 + \gamma_{13}W_{12}^2)}{(\Omega^2 + W_{12}W_{13})^2}, \quad (15)$$

$$\bar{\sigma}_{13} = \frac{-ig\hat{E}(-\gamma_{12}\Omega^2 + \gamma_{13}W_{12}^2)}{(\Omega^2 + W_{12}W_{13})^2} - \frac{ig}{\Omega^3} \frac{\partial}{\partial t} \frac{\hat{E}\Omega W_{12}W_{13}}{\Omega^2 + W_{12}W_{13}} - \frac{ig}{\Omega^2} \frac{\partial}{\partial t} \frac{\hat{E}W_{12}^2}{\Omega^2 + W_{12}W_{13}} + \frac{ig}{\Omega} \frac{\partial}{\partial t} \frac{\hat{E}\Omega}{\Omega^2 + W_{12}W_{13}} + \frac{ig}{\Omega} \frac{\partial}{\partial t} \frac{1}{\Omega} \frac{\partial}{\partial t} \frac{\hat{E}(-\gamma_{12}\Omega^2 + \gamma_{13}W_{12}^2)}{(\Omega^2 + W_{12}W_{13})^2} + \frac{ig}{\Omega} \frac{\partial}{\partial t} \frac{1}{\Omega^2} \frac{\partial}{\partial t} \frac{\hat{E}\Omega(-\gamma_{13}\Omega^2 + \gamma_{12}W_{13}^2)}{(\Omega^2 + W_{12}W_{13})^2}. \quad (16)$$

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

$$\hat{\Psi} = (\cos \theta)\hat{E} - (\sin \theta)\sqrt{N}\bar{\sigma}_{12}, \quad (17)$$

$$\hat{\Phi} = (\sin \theta)\hat{E} + (\cos \theta)\sqrt{N}\bar{\sigma}_{12}, \quad (18)$$

with the inverse relations

$$\hat{E} = (\cos \theta)\hat{\Psi} + (\sin \theta)\hat{\Phi}, \quad (19)$$

$$\sqrt{N}\bar{\sigma}_{12} = -(\sin \theta)\hat{\Psi} + (\cos \theta)\hat{\Phi}. \quad (20)$$

The time-dependent mixing angle $\theta(t)$ is defined by

$$\tan \theta = \frac{g\sqrt{N}}{\Omega(t)}. \quad (21)$$

Both $\hat{\Psi}$ and $\hat{\Phi}$ have bosonic commutation relations in the limit of few photons and many atoms. The action of $\hat{\Psi}^\dagger$ on the vacuum creates dark states [11], which contain no component of the excited state $|3\rangle$, and are therefore unaffected by spontaneous emission [2]. $\hat{\Phi}$, on the other hand, creates states which couple to state $|3\rangle$ 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 Eqs. (15)–(20), one can obtain

$$[\partial_t + c(\cos^2 \theta)\partial_z]\hat{\Psi} = -\dot{\theta}\hat{\Phi} - c(\sin \theta)(\cos \theta)\partial_z\hat{\Phi} + g\sqrt{N}(\sin \theta) \left[\frac{(-\Omega^2\gamma_{12} + W_{12}^2\gamma_{13})\hat{E}\Omega}{(\Omega^2 + W_{12}W_{13})^2} + \frac{W_{12}W_{13}}{\Omega^2} \frac{\partial}{\partial t} \frac{\hat{E}\Omega}{\Omega^2 + W_{12}W_{13}} + \frac{W_{12}^2}{\Omega} \frac{\partial}{\partial t} \frac{\hat{E}}{\Omega^2 + W_{12}W_{13}} \right]. \quad (22)$$

We make the assumption that $W_{12} \ll W_{13}$, that is, that the inhomogeneous width of the ground state is much less than the width of the upper, excited, state. In solids the upper state broadening is normally several orders of magnitude larger than that of the ground state, making this an excellent approximation. We can thus neglect the the last term in Eq. (22) relative to the second to last term.

After using Eq. (19) to eliminate \hat{E} and carrying out the differentiation, Eq. (22) can be written

$$[\partial_t + c(\cos^2 \theta)\partial_z]\hat{\Psi} = -\dot{\theta}\hat{\Phi} - c(\sin \theta)(\cos \theta)\partial_z\hat{\Phi} + (\tan \theta) \times [(\cos \theta)\hat{\Psi} + (\sin \theta)\hat{\Phi}] \times \left[\frac{\Omega^2(\sin \theta)(W_{12}^2\gamma_{13} - \Omega^2\gamma_{12})}{(\Omega^2 + W_{12}W_{13})^2} + \frac{\dot{\theta}}{\cos \theta} \frac{W_{12}W_{13}(\Omega^2 - W_{12}W_{13})}{(\Omega^2 + W_{12}W_{13})^2} \right] + \frac{(\sin^2 \theta)W_{12}W_{13}}{\Omega^2 + W_{12}W_{13}} [\dot{\Psi} + (\tan \theta)\dot{\Phi}] - (\tan \theta)\dot{\theta}\hat{\Psi} + \dot{\theta}\hat{\Phi}, \quad (23)$$

where we have used both Ω and θ even though they are related, as this makes the expression more compact. 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 γ_{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 Eq. (23). This can be accomplished by using Eqs. (15) and (18) to obtain

$$\hat{\Phi} = \frac{g\sqrt{N}}{\Omega}(\cos \theta) \left[\frac{\hat{E}W_{12}W_{13}}{\Omega^2 + W_{12}W_{13}} - \frac{1}{\Omega} \frac{\partial \hat{E}}{\partial t} \frac{\Omega(-\gamma_{13}\Omega^2 + \gamma_{12}W_{13}^2)}{(\Omega^2 + W_{12}W_{13})^2} - \frac{\partial \hat{E}(-\gamma_{12}\Omega^2 + \gamma_{13}W_{12}^2)}{\partial t (\Omega^2 + W_{12}W_{13})^2} \right]. \quad (24)$$

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

$$\hat{\Phi} = \left[\frac{W_{12}W_{13}(\sin \theta)(\cos \theta)}{\Omega^2 + W_{12}W_{13}} - (\alpha + \beta)\dot{\theta} \right] \hat{\Psi} + \beta(\cot \theta)\dot{\hat{\Psi}} + \left[\frac{W_{12}W_{13} \sin^2 \theta}{\Omega^2 + W_{12}W_{13}} - \alpha\dot{\theta}(\tan \theta) \right] \hat{\Phi}, \quad (25)$$

where

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

$$\beta = \frac{\sin^2 \theta [(\gamma_{12} + \gamma_{13})\Omega^2 - \gamma_{12}W_{13}^2 - \gamma_{13}W_{12}^2]}{(\Omega^2 + W_{12}W_{13})^2}. \quad (26)$$

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

$$\Omega^2 \gtrsim 3W_{12}W_{13}. \quad (27)$$

Thus to keep the bright state from being populated the strength of the control field Ω must always dominate the inhomogeneous broadening.

If this criterion is met, we have

$$\hat{\Phi} = \left[\frac{W_{12}W_{13}(\sin \theta)(\cos \theta)}{\Omega^2 + W_{12}W_{13}} - (\alpha + \beta)\dot{\theta} \right] \hat{\Psi} + \beta(\cot \theta)\dot{\hat{\Psi}}. \quad (28)$$

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 non-adiabaticity is considered.

A. Adiabatic case

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

$$\hat{\Phi}_{\text{ad.lim.}} = \frac{g\sqrt{N}\Omega W_{12}W_{13}}{(\Omega^2 + g^2N)(\Omega^2 + W_{12}W_{13})} \hat{\Psi}. \quad (29)$$

Provided one remains in the regime given by Eq. (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 polar-

iton 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 Eq. (23), ignoring the $\hat{\Phi}$ terms, and noting that as we are changing the control field adiabatically $\dot{\theta}=0$. This yields

$$[\partial_t + c(\cos^2 \theta)\partial_z] \hat{\Psi} = \frac{W_{12}W_{13} \sin^2 \theta}{\Omega^2 + W_{12}W_{13}} \dot{\hat{\Psi}} - (\sin^2 \theta)\Gamma_\Psi \hat{\Psi}, \quad (30)$$

with

$$\Gamma_\Psi = \frac{\Omega^2(\Omega^2\gamma_{12} - W_{12}^2\gamma_{13})}{(\Omega^2 + W_{12}W_{13})^2}. \quad (31)$$

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]:

$$[\partial_t + c(\cos^2 \theta)\partial_z] \hat{\Psi} = 0. \quad (32)$$

The first term on the right-hand side of Eq. (30) is clearly a correction to the group velocity of the polariton pulse. Provided we remain in the regime given by Eq. (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), Γ_Ψ is bounded by γ_{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

$$v_g = c(\cos^2 \theta_{\min}) = \frac{cW_{12}W_{13}}{W_{12}W_{13} + g^2N}. \quad (33)$$

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

In general, the solids of interest consist of rare-earth-ion-doped crystals [15]. Consequently, W_{12} is of the order of tens of kilohertz, and W_{13} is of the order of gigahertz. g^2N , on the other hand, tends to be within a few orders of magnitude of $\sim 10^{21}\text{Hz}^2$. (These assumptions, along with solids other than rare-earth ions doped into crystal hosts, will be considered in greater detail in Sec. IV.) The minimum polariton velocity is thus perhaps few tens of meters per second, although this is highly dependent on the medium chosen. Similarly, at the minimum control field strength $\sin \theta \approx 1 - W_{12}W_{13}/g^2N$, 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 suffi-

ciently 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,

$$g^2 N \gg W_{12} W_{13}, \quad (34)$$

$$\Omega^2 \gtrsim 3 W_{12} W_{13}. \quad (35)$$

To what extent these conditions can be met in current materials will be considered in Sec. 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 preceding section. Making use of Eqs. (28) and (23) we obtain the following equation of motion for the dark-state polariton:

$$\begin{aligned} [\partial_t + c(\cos^2 \theta) \partial_z] \Psi = & -A(t) \Psi + B(t) c \frac{\partial}{\partial z} \Psi \\ & + C(t) c^2 \frac{\partial^2}{\partial z^2} \Psi, \end{aligned} \quad (36)$$

where

$$\begin{aligned} A(t) = & (1 + \gamma)(\sin^2 \theta) \Gamma_\Psi + \dot{\theta} [\gamma(\cot \theta) + \gamma(\tan \theta) - (\alpha + \beta) \\ & \times (\tan \theta)(\sin^2 \theta) \Gamma_\Psi - (1 + \gamma) \delta(\tan \theta) - 2\gamma^2(\cot \theta) \\ & + \gamma^2(\tan \theta) - 2g^2 N \gamma^2(\cot \theta)(\csc^2 \theta)] - \dot{\theta}^2(\alpha + \beta)(1 - \gamma \\ & - \delta \tan^2 \theta), \end{aligned} \quad (37)$$

$$\begin{aligned} B(t) = & -2\gamma(\cos^2 \theta) - \beta \Gamma_\Psi(\sin^2 \theta)(\cos^2 \theta) \\ & + \dot{\theta}(\sin \theta)(\cos \theta) [\alpha + \beta(1 + \cot^2 \theta - \delta - \gamma \cot^2 \theta)], \end{aligned} \quad (38)$$

$$C(t) = \beta \cos^4 \theta, \quad (39)$$

and where

$$\gamma = \frac{(\sin^2 \theta) W_{12} W_{13}}{\Omega^2 + W_{12} W_{13}}, \quad (40)$$

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

$A(t)$ 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 $\dot{\theta}$ can be without incurring significant nonadiabatic losses. We will follow the analysis of Ref. [11].

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

$$\begin{aligned} \tilde{\Psi}(k, t) = & \tilde{\Psi}(k, 0) \exp \left[ik \int_0^t dt' [v_{gr}(t') - cB(t')] \right] \\ & \times \exp \left[\int_0^t dt' [A(t') - k^2 c^2 C(t')] \right], \end{aligned} \quad (42)$$

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

$$\int_0^\infty dt' A(t') \ll 1, \quad (43)$$

$$k^2 c^2 \int_0^\infty dt' C(t') \ll 1. \quad (44)$$

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

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

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

$$z \ll \frac{g^2 N}{\gamma_{13} L_p^2}, \quad (46)$$

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 Eq. (43). Looking at Eq. (37) 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 . We take the initial control field strength to be Ω_0 and parametrize the final control field strength $\Omega(\tau)$ as $k = \Omega(\tau)/\sqrt{W_{12}W_{13}}$. Assuming a linear decrease in Ω over the time τ , then integra-

tion of the term proportional to $\dot{\theta}^2$ with respect to time yields the condition

$$\tau \gg \frac{\gamma_{13}\Omega_0}{k^7(W_{12}W_{13})^{3/2}} \quad (47)$$

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

Finally we consider the term in (43) 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 θ , 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

$$\eta = \exp\left[\frac{3+2k^2}{(1+k^2)^2} + 2 \ln \frac{k^2}{1+k^2}\right]. \quad (48)$$

Equations (46), (47), and (48) 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. In general, the rare-earth-dopant ions are characterized by low inhomogeneous broadening of their lower state hyperfine transitions and well-characterized energy levels.

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 [29] and pulse compression by photon echo effects [30].

In these materials the transitions of interest are usually $f-f$ or $f-d$. The $f-f$ transitions are characterized by having a low inhomogeneous linewidth for the optical transitions, and have relatively low oscillator strengths. $f-d$ transitions, on the other hand, have larger optical inhomogeneous broadening, and also have relatively large oscillator strengths. An additional advantage of the $f-d$ transitions is that many of them coincide with the existence of a zero-phonon line at low temperatures [15].

Due to these properties, our focus will be on materials with suitable $f-d$ transitions. A wide class of rare-earth-doped crystals satisfying these criteria can be found in [15].

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 $g = d_{13}\sqrt{v/2\hbar\epsilon_0V}$. The dipole moments for $f-d$ transitions in rare-earths generally lie in the range 10^{-29} – 10^{-31} C m. We choose

10^{-30} C m as a representative value in the following. As an order-of-magnitude estimate we assume a wavelength of 1000 nm, and find the convenient relation that

$$g^2N(\text{Hz}^2) \sim \frac{N}{V}(\text{m}^{-3}), \quad (49)$$

that is, the collective coupling strength is simply given by the density of the dopant atoms in the medium. The density of rare-earth 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} for $f-d$ transitions is 40–300 GHz [15], while W_{12} ranges from 100 Hz – 10 Mhz [28,32]. Consequently, as a representative value 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 θ 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

$$I = \frac{\Omega^2 \hbar^2 c \epsilon_0}{2d_{13}^2} \quad (50)$$

and assuming a dipole moment of $d_{13} = 10^{-30}$ C m we see that this corresponds to a coupling laser intensity of 10 kW/cm². 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 τ , the distance the pulse travels is

$$z = \int_0^\tau c \cos^2 \theta(t) dt \approx c\tau. \quad (51)$$

Thus, bearing in mind the adiabaticity requirements, if $\tau \sim 10^{-6}$ s, the stopping distance is 300 m. 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 case

$$z = \frac{\Omega(0)^2}{3g^2N} c\tau. \quad (52)$$

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 ~ 100 W/cm².

The final conditions that must be met are the adiabaticity criteria, namely Eqs. (46), (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 $\Gamma_{EIT} = \Omega^2/W_{13}$ [15], corresponding to a bandwidth of $\sim 10^6 - 10^8$ Hz, depending on the initial coupling field strength. If a broader bandwidth is required, one merely needs to increase the coupling field strength.

Equation (47) is a fairly weak condition. Using the numbers $W_{12}W_{13} \sim 10^{15}$ Hz², $\Omega(0)^2 \sim 10^{17}$ Hz², and $\gamma_{13} \sim 10^7$ Hz, one obtains $\tau \gg 10^{-7}$ 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 (48) is highly dependent as 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 general, it appears that quantum information storage using this technique is feasible in rare-earth-doped crystals. It is not clear, however, whether there is one type of material that possesses all the properties which 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 γ_{12} . The ideal material would have a low $W_{12}W_{13}$, a high g^2N , and a very low γ_{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 (~ 1 MHz) [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 [28] and thus drastically reducing W_{13} . The drawback is a reduction in the interacting ion density, but as $g^2N \sim 10^{23}$ Hz² 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, which 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 γ_{12}

and the inhomogeneous width W_{12} of the lower hyperfine transition in the ions. γ_{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 [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 this suggests that storage times of the order of 100 ms or more may be achievable [32].

For integration with current telecommunication technologies, it is natural to speculate about the possibilities of slowing and storing light in doped optical fibers and waveguides rather than crystals. In fibers and waveguides, 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 [33–35]. Persistent hole burning has been demonstrated in glass fibers [36], 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 1-3 and 1-2 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. There are, however, a number of techniques that could be employed to compensate for this. For example, a crystal fiber may be doped only in the central rod which forms the central waveguide in the fiber [37]. The light is thus 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 fibers, may compensate for the low concentration, and make slowing and storage of light possible. Another possibility is to set up an optical cavity by writing a Bragg grating in the fiber [38] (or by coating the faces of a crystal if a fiber is not used) 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 [39].

A detailed analysis of light slowing and stopping in fibers as opposed to crystals is beyond the scope of this paper. However, we would like to note that significant nonlinear dynamics, for example supercontinuum generation, has been observed in fibers at very high light intensities as a consequence of the nonlinear susceptibility of the glass host [40]. If our rare earth-doped crystal analysis is to be directly applicable to fibers, these intensities should be avoided. On the

other hand, it is quite possible that in the high-intensity regime the nonlinear susceptibility coupled with the EIT dynamics could lead to new and interesting effects.

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