Raman scheme for adjustable bandwidth quantum memory

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Abstract

We propose a scenario of quantum memory for light based on Raman scattering. The storage medium is a vapor and the different spectral components of the incoming signal are stored in different atomic velocity classes. One uses appropriate pulses to reverse the resulting Doppler phase shift and to regenerate the signal, without distortion, in the backward direction. The different stages of the protocol are detailed and the recovery efficiency is calculated in the semi-classical picture. Since the memory bandwidth is determined by the Raman transition Doppler width, it can be adjusted by changing the angle of the signal and control beams. The optical depth also depends on the beam angle. As a consequence the available optical depth can be optimized, depending on the needed bandwidth. The predicted recovery efficiency is close to 100% for large optical depth.

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I. INTRODUCTION

The storage of quantum information in an atomic ensemble has received a great deal of attention over the past ten years or so. Protocols based on electromagnetic induced transparency (EIT) have been investigated by many groups both theoretically and experimentally, leading to storage and retrieval demonstration of both discrete and continuous quantum variables. Although successful, the EIT storage scheme suffers from time-bandwidth product limitations. In EIT-based protocols, the reduction of group velocity is used to spatially confine the input signal within the boundaries of the storage medium. Simultaneously, the signal spectrum must not exceed the bandwidth of the transparency window associated with EIT. Since the group velocity is inversely proportional to the width of the transparency window, it follows that, the larger the storage bandwidth, the shorter the temporal profile the memory can accommodate. This protocol has been demonstrated using level schemes in which inhomogeneous broadening does not play a significant role. Extension to inhomogeneously broadened systems does not improve the time-bandwidth product capabilities.

On the other hand, inhomogeneous broadening can be of critical importance in other protocols for storing quantum information. For example, Doppler broadening determines the storage bandwidth when a signal pulse is totally absorbed in an optically dense medium. While the temporal components of the input signal are distributed in atomic state coherence along the axial direction in EIT, the spectral components of the input signal are spread over the inhomogeneous frequency distribution of the atoms in the absorption protocol. Just as in EIT, information is stored in a long lifetime Raman coherence in the absorption protocol, but the maximum duration of the input signal is independent of the storage bandwidth, being ultimately limited only by the inverse homogeneous line width. The resulting time-bandwidth product capacity, given by the ratio of the inhomogeneous and homogeneous widths, is reminiscent of the photon-echo based storage techniques that were developed in the past. However, unlike some of those classical light storage schemes, the proposal in Ref. that we shall refer to as the MK protocol, is restricted to systems where the inhomogeneous broadening is provided by the Doppler effect.

A variant of the MK protocol has been proposed for solids in which the inhomogeneous broadening is linked to stochastic variations in atomic transition frequency that depend on
an absorbing center’s position in a host medium \[11, 12\]. Although there are several proof of principle experiments of this absorption-type protocol \[14, 15, 16\], all such experiments have involved classical input fields. It should be stressed that the large time-bandwidth product capacity is lost when the Doppler shift is replaced by a more stochastic source of inhomogeneous broadening. In order to keep control of the inhomogeneous phase shift, one selects a narrow spectral group of atoms at the beginning. An external field is used to spread this initial ensemble over the desired bandwidth. Hence the bandwidth is increased at the expense of the available optical density, i.e. at the expense of the capacity to trap the optically carried information within the material.

A key feature in the MK scenario is that the totally absorbed signal is restored without amplification. This contrasts with previous photon echo investigations where large retrieval efficiency results from strong amplification in an inverted medium \[17, 18, 19, 20, 21\]. Therefore, unlike these earlier works, the MK protocol is free from the noise associated with spontaneous and stimulated emission. This is closely related to the fact that the control fields do not interact with highly populated states. As a consequence few atoms are promoted to the upper electronic level. Another consequence is that, as the control fields interact with quasi-empty states, they are neither attenuated nor distorted as they travel through the active medium.

Quite surprisingly, the original MK scheme has not been demonstrated experimentally. A possible issue in atomic vapors is the short lifetime of the active optical transition upper level. Indeed, to combine a large optical density with the absence of collisions one has to work on strong lines with short upper level lifetime. As a consequence, some operations have to be carried out on a nanosecond time scale. Specifically, one needs nanosecond $\pi$-pulses to convert optical dipoles into Raman coherences and conversely. In addition, the input signal duration is limited to a few nanoseconds.

In this paper, we propose a Raman variant of the MK scheme that circumvents these limitations. Direct excitation of ground state coherence avoids the introduction of rapidly decaying quantities, yet retains the other advantages of the MK protocol. Quantum memories based on Raman scattering have been proposed in the past \[15, 22, 23\]. However, previous proposals did not fully examine the dynamics of the system, focusing on steady-state conditions, \[22\] or they considered a situation in which the temporal signal profile is mapped into a spatial distribution of the atomic ground state coherence \[15, 23\]. In our
scenario, the spectro-temporal features of the signal are stored in the spectral distribution of the ground state coherence. The experimental investigation presented in ref. [15] is actually very close to our situation, but the authors resort to a reversible magnetic field gradient to cover the signal spectrum and to reverse the atomic phase, which ultimately leads to a spatial mapping of the signal. In our case we use optical pulses to reverse the atomic phase.

Our objective in this paper is to describe the underlying physics of the Raman protocol, a goal that can be achieved within the confines of a theory in which all radiation fields are treated classically. The paper is arranged as follows: after presenting a picture of the overall process in section II, we develop a theory for each step of the protocol in sections III-V. In section VI we discuss the range of applicability of the storage method.

II. OUTLINE OF THE STORAGE AND RETRIEVAL PROCEDURE

The protocol consists of the four steps shown schematically in Fig. 1. We consider an ensemble of four-level atoms. The four levels can be magnetic state sublevels of the same or different ground state hyperfine levels. The atoms are prepared initially in state $|a\rangle$.

1. Stage 1

In the first stage, a quasi-monochromatic control field and the input pulse drive Raman transitions between levels $a$ and $c$. Each field is off-resonant for optical excitation of level $b$, but the difference of the field frequencies is close to that of the Raman transition. The control and input fields have a relative propagation vector $\mathbf{K}$ that leads to a Doppler shift $\mathbf{K} \cdot \mathbf{v}$ associated with the two-photon Raman transition. As a consequence the bandwidth that can be absorbed in this Raman process is on the order of $1/Ku$, where $u$ is a characteristic atomic speed. Of critical importance is that all frequency components of the signal field are depleted in an identical fashion (no pulse distortion) if the bandwidth is less than the inhomogeneous width. The medium is optically dense so the signal pulse is totally attenuated. In other words, as a result of stimulated Raman scattering, the signal pulse energy is totally transferred to the control field. Each atom has a negligibly small population in state $|c\rangle$ and the entire population stored in level $c$ is assumed to be small as well, assuming the signal pulse is weak. The control field is turned off following the depletion of the signal pulse. In
FIG. 1: (Color online) Level scheme and protocol steps. (a) the weak input signal, combined with a strong control field, drives the Raman transition $a \rightarrow c$. (b) the Doppler phase build-up is stopped by conversion of the coherence $\rho_{ac}$ into $\rho_{ad}$. This is accomplished by $\pi$-pulse excitation of the Raman transition $c \rightarrow d$. (c) the coherence $\rho_{ac}$ is recovered with the help of a second Raman $\pi$-pulse. (d) a backward propagating control field creates a coherence $\rho_{ab}$ which allows for the restoration of the signal pulse, propagating in the backward direction.

contrast to the MK protocol in which an optical coherence is created in the first stage, the signal field is transferred directly to a Raman coherence in our protocol that is immune to spontaneous emission decay.

2. **Stage 2**

The Raman coherence dephases following excitation as a result of the inhomogeneous broadening. As in a photon echo experiment, this dephasing can be reversed by the application of a second pair of pulses. However, if we were to use a two-pulse echo process, the Raman coherence excited by the signal would be affected by velocity changing collisions
over the _entire_ storage time between the two pulses. Instead, if we use a three-pulse echo configuration, the effect of collisions during most of the storage time can be suppressed. The second pulse pair, in effect, freezes the Doppler phase created by the first pulse pair. As in the MK protocol, this phase reversal must be carried out with a Raman pulse that leaves the population in level \( a \) unchanged. This is a crucial condition to ensure uniform illumination by the control fields; if any control field is resonant with a transition originating in level \( a \), the field will be strongly absorbed in the optically dense medium and unsuitable for this protocol [25]. All these requirements are satisfied if one applies a Raman \( \pi \) pulse between levels \( c \) and \( d \), following the excitation of the \( a - c \) Raman coherence. The relative \( \mathbf{K} \) vector of the two fields is the same as that in stage 1, so the net effect of the \( \pi \) pulse is to convert the \( a - c \) to \( a - d \) coherence, while freezing the Doppler phase evolution of this Raman coherence, just as in a stimulated photon echo. The net result is that the original pulse information is now stored in a Raman coherence that is, for the most part, "protected" from the effects of velocity-changing collisions.

3. **Stage 3**

To prepare the system for the retrieval stage, a \( \pi \) pulse is sent into the medium at some later time to restore the \( a - c \) coherence. This Raman pulse has its relative \( \mathbf{K} \) vector reversed and prepares the atoms with a spatial Raman coherence that allows for retrieval of the signal in stage 4.

4. **Stage 4**

A control pulse is sent into the sample that is identical to the initial control pulse, but with its propagation vector _reversed_. The first three stages produce a Raman coherence that allows a field to build up in a direction opposite to that of the input signal pulse. In other words, the depletion of the signal field is reversed and the original signal pulse is restored as it exits the sample propagating in a direction opposite to that of the original signal pulse. In principle, the pulse can be restored with close to 100% fidelity. In the context of creating a functional quantum memory device, splitting the phase reversal in two steps (stages 2 and 3) reduces the waiting time between the read-out pulse and the emission of the restored
signal, making the retrieved data available more rapidly after the read-out decision.

We now describe in detail how each of these steps can be achieved.

III. STORAGE STEP

Storage corresponds to the mapping of the optically carried information into atomic Raman coherence, accompanied by attenuation of the input signal field.

A. Buildup of a Raman atomic superposition state

The input signal is depleted by stimulated Raman scattering in a three-level $\Lambda$-system. The weak pulse to be stored, combined with the control field, resonantly excites the Raman transition $|a\rangle - |c\rangle$. Both fields are tuned off resonance from the optical transition to upper level $|b\rangle$. Storage is performed into the superposition of the ground substates $|a\rangle$ and $|c\rangle$. Since the atoms are prepared initially in state $|a\rangle$, the medium is transparent to the control field that uniformly illuminates all the active atoms. Assuming that the control field is constant during the signal pulse, we write the electric field of the control field as the plane wave

$$E_2(r, t) = A_2e^{i(k_2r - i\omega_2t)} + c.c. \quad (1)$$

where the envelope $A_2$ is a time- and space-independent parameter. The control field wave vector and frequency are denoted $k_2$ and $\omega_2$, respectively.

The signal pulse Rayleigh range is assumed to be much larger than the storage material length $L$ to insure that its diameter does not vary significantly as it propagates in the medium. The electric field of the signal field can be expressed as

$$E_1(r, t) = A_1(r, t)e^{ik_1r - i\omega_1t} + c.c., \quad (2)$$

where $A_1(r, t)$ is the envelope, $k_1$ the propagation vector, and $\omega_1$ the carrier frequency of this field. The spatial dependence of $A_1(r, t)$ reflects the radial distribution of the field and its attenuation along direction $k_1$. When $L/c$ is not much smaller than the pulse duration, retardation also contributes to the field envelope spatial dependence. The Rabi frequencies
associated with the signal and control fields are denoted by

\[ \Omega_1(r, t) = -\mu_{ba} A_1(r, t)/\hbar; \]
\[ \Omega_2 = -\mu_{bc} A_2/\hbar, \] (3b)

where \( \mu_{ba} \) and \( \mu_{bc} \) are optical dipole moment matrix elements. It is assumed that \( k_1 \approx k_2 \equiv k \).

In perturbation theory with the population of level \( a \) set equal to unity, the coupled equations for the optical and Raman coherences are:

\[ \dot{\rho}_{ab;m} = (i\Delta_1 - \gamma_{ab})\rho_{ab;m} + i\Omega_1^* [r_m(t), t] e^{-ik_1 \cdot r_m(t)} + i\rho_{ac;m} \Omega_2^* e^{-ik_2 \cdot r_m(t)} \] (4a)
\[ \dot{\rho}_{ac;m} = [i(\Delta_1 - \Delta_2) - \gamma_{ac}]\rho_{ac;m} + i\rho_{ab;m} \Omega_2 e^{ik_2 \cdot r_m(t)} \] (4b)

where \( \Delta_1 \) and \( \Delta_2 \) are the atom-field detunings for each optical transition, \( \gamma_{ab} \) is the decay rate for the \( a \rightarrow b \) coherence, \( \gamma_{ac} \) is the decay rate for the \( a \rightarrow c \) coherence, \( \rho_{ab;m} = \rho_{ab;m}^* e^{-i\omega_1 t} \) and \( \rho_{ac;m} = \rho_{ac;m} e^{-i(\omega_1 - \omega_2)t} \). These equations give the time evolution of the density matrix elements for atom \( m \), located at \( r_m(t) \) at time \( t \). The manner in which the spatial phases of the field are imprinted on the atoms is readily apparent in Eqs. (4). Under the assumption that \( \Omega_1, \Omega_1^{-1} d\Omega_1/dt, \gamma_{ab}, ku << \Delta_1 \), where \( u \) is the most probable atomic speed, the optical coherence adiabatically follows the field variations and can be written as:

\[ \rho_{ab;m} = -\Omega_1^* [r_m(t), t] e^{-ik_1 \cdot r_m(t)/\Delta_1} - \rho_{ac;m} \Omega_2^* e^{-ik_2 \cdot r_m(t)/\Delta_1}. \] (5)

Substituting this expression into Eq. (4b) one obtains

\[ \dot{\rho}_{ac;m} = \left[ i \left( \Delta_1 - \Delta_2 - \frac{\Omega_2^*}{\Delta_1} \right) - \gamma_{ac} \right] \rho_{ac;m} - i \frac{\Omega_1^* [r_m(t), t] \Omega_2 e^{iK \cdot r_m(t)}}{\Delta_1} \] (6)

where

\[ K = k_2 - k_1. \]

The detuning \( \Delta_1 - \Delta_2 \) can be adjusted to cancel the light shift \( |\Omega_2|^2/\Delta_1 \). Finally, the Raman coherence can be expressed as:

\[ \tilde{\rho}_{ac;m}(t) = -i \frac{\Omega_2}{\Delta_1} \int_{-\infty}^{t} dt' \Omega_1^* [r_m(t'), t] e^{iK \cdot r_m(t')} \] (7)

where it has been assumed that any decay of \( \tilde{\rho}_{ac;m} \) during the signal pulse can be neglected. The density matrix element can be expressed in terms of the atom position at time \( t \). Indeed, if collisions do not change the atomic velocity \( v_m \) during the signal pulse, the position at \( t' \) can be expressed as \( r_m(t') = r_m(t) - v_m(t - t') \), so that:

\[ \tilde{\rho}_{ac;m}(t) = -i \frac{\Omega_2}{\Delta_1} e^{iK \cdot r_m(t)} \int_{-\infty}^{t} dt' \Omega_1^* [r_m(t) - v_m(t - t'), t'] e^{-iK \cdot v_m(t - t')} \] (8)

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B. Stimulated Raman scattering of the input signal

It is assumed that, in the absence of the control field, the scattering of the probe field is negligible owing to the large detuning $\Delta_1$. As a consequence, the contribution to the polarization resulting from the first term in Eq. (5) can be neglected. On the other hand, the control field intensity is sufficiently large to allow for significant stimulated Raman scattering, resulting in a loss of signal field intensity as the signal field propagates in the medium. The Raman contribution to $\tilde{\rho}_{ab;m}$ is given by the second term on the right hand side of Eq. (5). To determine the modification of the signal field, we need to calculate the polarization associated with the $\tilde{\rho}_{ab;m}$ coherence. We write the macroscopic polarization as

$$P(r, t) = P_+(r, t)e^{i\mathbf{k}_1 \cdot \mathbf{r} - i\omega_1 t} + P_+(r, t) e^{-i(\mathbf{k}_1 \cdot \mathbf{r} - i\omega_1 t)}$$

In going over to a macroscopic polarization, we assume that, at any position $\mathbf{r}$ in the medium, one can define a slice of thickness $l \ll 2\pi/k$ in the $\mathbf{k}_1$ direction, containing many atoms. The macroscopic polarization at $(\mathbf{r}, t)$ is obtained by combining the contributions from all the atoms within the slice. Those atoms satisfy the condition $|\mathbf{k}_1 \cdot \mathbf{s}_m(t)| \leq l/2$, where $\hat{\mathbf{k}}_1$ is a unit vector in the $\mathbf{k}_1$ direction and $\mathbf{s}_m(t) = \mathbf{r} - \mathbf{r}_m(t)$. Therefore the positive frequency component $P_+(\mathbf{r}, t)$ is given by

$$P_+(\mathbf{r}, t) = \frac{\mu_{ab}}{\delta V} e^{-i\mathbf{k}_1 \cdot \mathbf{r}} \sum_{|\hat{\mathbf{k}}_1 \cdot \mathbf{s}_m(t)| \leq l/2} \tilde{\rho}_{ab;m}(\mathbf{r}, t),$$

where $\delta V$ represents the slice volume. Combining Eqs. (5), (8), (9), and (3a), we find

$$P_+(\mathbf{r}, t) = i \left| \frac{\mu_{ab}}{\delta V} \right|^2 \frac{|\Omega_2|^2}{\Delta_1^2} \sum_m \int_{-\infty}^{t} dt' A_1 [\mathbf{r} - \mathbf{v}_m(t - t'), t'] e^{i\mathbf{k}_1 \cdot \mathbf{v}_m(t - t')}.$$  

As was noted above, the contribution to $\tilde{\rho}_{ab;m}(\mathbf{r}, t)$ from the first term in Eq. (5) has been neglected since it adiabatically follows the field and vanishes for times greater than the pulse duration.

The atoms are uniformly distributed in space, with density $N$, and their normalized velocity distribution is represented by $W(\mathbf{v})$. Replacing the discrete sum by an integral, according to

$$\frac{1}{\delta V} \sum_m \rightarrow N \int d^3 \mathbf{v} W(\mathbf{v}),$$
enables us to transform Eq. (10) into
\[
P_+(\mathbf{r}, t) = i\frac{\mu_{ab}}{\hbar \Delta_1^2} N \int d^3 v W(v) \int_{-\infty}^{t} dt' A_1[\mathbf{r} - \mathbf{v}(t - t'), t'] e^{iK\cdot\mathbf{v}(t-t')}.
\] (11)

Provided the input signal spectral width \(\delta_s\) is smaller than \(Ku\), \(A_1[\mathbf{r} - \mathbf{v}(t - t'), t']\) can be taken out of the integral over \(t'\) and evaluated at \(t' = t\). In this limit the polarization \(P_+(\mathbf{r}, t)\) reduces to:
\[
P_+(\mathbf{r}, t) = i\frac{\mu_{ab}}{\hbar \Delta_1^2} N \int d^3 v W(v) \int_{-\infty}^{\infty} d\tau e^{iK v \tau}
\]
\[
= i \frac{N\pi}{k} \frac{\mu_{ab}^2 |\Omega_2|^2}{K\Delta_1^2} A_1(\mathbf{r}, t) (12a)
\]
\[
= \frac{i N\pi}{k} \frac{\mu_{ab}^2 |\Omega_2|^2}{K\Delta_1^2} W(0) \frac{k |\Omega_2|^2}{K\Delta_1^2} A_1(\mathbf{r}, t) (12b)
\]

where \(W(v)\) represents the one-dimensional velocity distribution.

This is the key result of this section. Owing to the large inhomogeneous width, the polarization is proportional to the field amplitude and depends locally on this amplitude. In other words, the polarization does not depend on the value of the field amplitude at earlier times as it would in the case of homogeneous broadening. The electric susceptibility \(\chi_R\) is defined by \(P_+(\mathbf{r}, t) = \epsilon_0 \chi_R A_1(\mathbf{r}, t)\) with the intensity absorption coefficient \(\alpha_R\) given by \(\alpha_R = k\text{Im}(\chi_R)\), which leads to
\[
\alpha_R = \frac{k |\Omega_2|^2}{K\Delta_1^2} \alpha_0
\] (13)

where \(\alpha_0\) represents the linear absorption coefficient on the inhomogeneously broadened transition \(a - b\). Provided \(\delta_s < Ku\), the signal propagates without distortion, which implies that all the signal spectral components are uniformly attenuated and stored in the atomic ensemble. The spectral components are mapped into Raman coherence in atoms whose velocities span an interval of order \(\delta_s/K\) along the direction \(\mathbf{K} = \mathbf{k}_2 - \mathbf{k}_1\). The parameter \(K\) should be adjusted in such a way that \(\delta_s/Ku\) is larger than unity to provide a sufficiently large bandwidth to store the signal pulse, but not too large, since the signal depletion and Raman storage varies inversely with \(K\).

IV. FREEZING THE DOPPLER PHASE

The input signal illuminates the storage medium during a time interval centered at \(t_1\). The control field is turned off following the signal pulse. At some later time, the Raman coherence, given by Eq. (8), can be expressed as:
\[
\tilde{\rho}_{ac,m}(t) = -i e^{iK\cdot\mathbf{r}_m(t) - iK\cdot\mathbf{v}_m + \gamma_{ac}(t-t_1)} \mathcal{R}_m
\] (14)
FIG. 2: (Color online) Schematic representation of the timing sequence for the entire protocol, showing the relative duration of the different pulses, and the various atomic quantities involved at the different stages.

where

\[
R_m = \frac{\Omega_2}{\Delta_1} \int_{-\infty}^{\infty} d\tau \Omega_1^* [r_m(t_1), t_1 + \tau] e^{iK \cdot v_m \tau}
\]

and it was assumed that \(\Omega_1 [r - \mathbf{v} \tau_p, t] \approx \Omega_1 [r, t]\), where \(\tau_p\) is the signal pulse duration. There is a build-up of Doppler phase associated with the Raman coherence that grows linearly as a function of time following the interaction with the signal pulse. Although this phase could be reversed at a later time, it is best to nip it in the bud to prevent any deterioration from velocity-changing collisions. To accomplish this task one can send in a Raman \(\pi\) pulse having the same \(K\) vector that transfers the amplitude from state \(|c⟩\) to an auxiliary level \(|d⟩\), or, equivalently, converts coherence \(\tilde{\rho}_{ac}\) into \(\tilde{\rho}_{ad}\).

Let \(\Omega_3(t)\) and \(\Omega_4(t)\) denote the Rabi frequencies on transitions \(|c⟩ - |b⟩\) and \(|b⟩ - |d⟩\), respectively. The radial extension of these fields is assumed to be larger than that of the signal pulse. Moreover, these fields are not attenuated as they propagate through the storage ma-
terial since interact with quasi-empty levels. Hence their Rabi frequency space dependence can be omitted. Both fields are detuned by the same amount $\Delta$ from resonance with their respective assigned transition. Therefore the $|c\rangle - |d\rangle$ two-photon transition is resonantly excited. The Rabi frequency of the equivalent two-level system is given by $\Omega(t)\Omega_\pi(t)/\Delta$. The two-level approximation holds provided $|\tilde{\Omega}_{3,4}(\Delta \pm \delta_s)|^2 << 1$, where $\Omega(\Delta)$ represents the time-to-frequency Fourier transform of $\Omega(t)$. Of course one must take care that none of those fields can excite state $|a\rangle$, where all the atomic population is concentrated. With these assumptions, one finds that $\rho_{ac}$ and $\rho_{ad}$ evolve according to

$$\dot{\rho}_{ac;m} = \frac{i}{\Delta} |\Omega_\pi|^2 e^{-i\phi_m(t)} \rho_{ac;m} - \gamma_{ad} \rho_{ad;m}$$  \hspace{1cm} (16a)$$

$$\dot{\rho}_{ad;m} = \frac{i}{\Delta} |\Omega_\pi|^2 e^{i\phi_m(t)} \rho_{ad;m} - \gamma_{ac} \rho_{ac;m},$$  \hspace{1cm} (16b)$$

where $\rho_{ad;m} = \rho_{ad;m} e^{-i(\omega_3 - \omega_4)t}$, $\phi_m(t) = (k_3 - k_4) \cdot r_m(t)$ and, for simplicity, we have set $\Omega_3(t) = \Omega_4(t) = \Omega_\pi(t)$ and neglected a light shift that is the same for levels $c$ and $d$. The pulses, having duration $\tau_\pi$, are applied at a time centered about $t = t_2$ and their duration is assumed to be sufficiently short, $\delta_s \tau_\pi << 1$, to resonantly excite all the atoms that were excited by the signal pulse (recall that atoms in the velocity range $|K \cdot v| \leq |\delta_s|$ are excited in stage 1). The time-diagram of the whole protocol is displayed in Fig. 2 showing the relative duration of the different pulses.

In terms of the coherence components at time $t_2^-$ just before the Raman $\pi$ pulse is applied, Eq. (16) can be solved, for $t > t_2$, as:

$$\tilde{\rho}_{ac;m}(t) = e^{-\gamma_{ac}(t-t_2)} \cos \left( \frac{\Theta}{2} \right) \tilde{\rho}_{ac;m}(t_2^-) + ie^{i\phi_m(t_2^-)-\gamma_{ad}(t-t_2)} \sin \left( \frac{\Theta}{2} \right) \tilde{\rho}_{ad;m}(t_2^-)$$  \hspace{1cm} (17a)$$

$$\tilde{\rho}_{ad;m}(t) = ie^{-i\phi_m(t_2^-)-\gamma_{ad}(t-t_2)} \sin \left( \frac{\Theta}{2} \right) \tilde{\rho}_{ac;m}(t_2^-) + e^{-\gamma_{ad}(t-t_2)} \cos \left( \frac{\Theta}{2} \right) \tilde{\rho}_{ad;m}(t_2^-),$$  \hspace{1cm} (17b)$$

where $\Theta = 2 \int dt |\Omega_\pi(t)|^2 / \Delta$, assuming that $\gamma_{ac}\tau_\pi, \gamma_{ad}\tau_\pi << 1$. Total conversion from $\rho_{ac}$ to $\rho_{ad}$ requires that $\Theta = \pi$. With initial conditions given by Eq. (14) and $\tilde{\rho}_{ad;m}(t_2^-) = 0$ one finds that

$$\tilde{\rho}_{ad;m}(t) = e^{i(k_4 - k_3 + K) \cdot r_m(t_2^-) - \gamma_{ad}(t-t_2)} e^{-i(K \cdot v_m + \gamma_{ac})t_2^-} R_m,$$  \hspace{1cm} (18)$$

where $t_{ij}$ is the time interval between the $j$th and $i$th pairs of pulses. With $k_3 - k_4 = K$, there is no build-up of Doppler phase following this second Raman pulse [26] - the Doppler phase has been frozen.
V. SIGNAL RECOVER Y

To prepare for signal retrieval at some later time, one sends in another Raman \( \pi \) pulse, centered at time \( t = t_3 \), to restore the \( \tilde{\rho}_{ac;m} \) coherence. This Raman pulse consists of two fields having propagation vectors \( k_4' \) and \( k_3' \) that drive the \( |d\rangle - |b\rangle \) and \( |b\rangle - |c\rangle \) transitions, respectively. The atom-field dynamics is again described by Eqs. (16), with \( \phi_m(t) \) replaced by \( \phi'_m(t) = (k_3' - k_4').r_m(t) \). To reverse the Doppler phase acquired in the time interval \( t_{12} \), we choose the propagation vectors such that \( k_3' - k_4' = -K \), which leads to a Raman coherence for \( t > t_3 \) given by

\[
\tilde{\rho}_{ac;m}(t) = i e^{i\phi'_m(t_3) - iK.v_m(t_{12} - \gamma_{ac}(t - t_3 + t_{12}) - \gamma_{ad}t_{23})} R_m
\]

(19)

To restore the signal one directs a quasi-monochromatic control field having frequency \( \omega_2 \) into the medium in the backward direction, with \( k_2'' = -k_2 \). This field should be switched on somewhat before time \( t = t_3 + t_{12} \), with the same Rabi frequency \( \Omega_2 \) as the initial coupling beam. The restored field can be written as

\[
E(r, t) = A(r, t)e^{ik_2''r - i\omega_1 t} + c.c.
\]

We now argue that, only if \( k_1'' = -k_1 \), can a phase matched signal be generated. To see this, we return to Eq. (19) and analyze the phase factor in this equation. Although \( R_m \) contains a Doppler phase factor [see Eq. (15)], it is of order \( Ku\tau \), where \( \tau \) is the signal pulse duration, and is small compared to the other Doppler phases appearing in Eq. (19), since we assume that \( t_{12} \gg \tau \). When the control field is sent into the medium, the Raman coherence (19) gives rise to an optical coherence on the \( a - b \) transition that is responsible for the generation of the restored field. From Eq. (15), one can deduce that the optical coherence \( \tilde{\rho}_{ab;m} \) varies as

\[
e^{i(k_2'.r_m(t) - iK.r_m(t) + iK.v_m(t_{12} - t_3 + t_{12}) - iK.v_m t_{12})} = e^{ik_1'.r_m(t) + iK.v_m(t_{12} - t_3)}
\]

This expression implies that a phase matched signal can propagate only in the \( k_1'' = -k_1 \) direction and that this signal can be nonvanishing (owing to the average over atomic velocities) only for times \( t \approx t_3 + t_{12} \). Thus, in what follows we assume that the restored signal field has propagation vector \( k_1'' = -k_1 \).
The existence of the restored field must be taken into account in the expression for the Raman coherence. Indeed, this field also combines with the control field to drive Raman transitions between levels $a$ and $c$. Adding this contribution to Eq. (19), one obtains the Raman coherence

$$
\rho_{ac;m}(t) = i e^{-i(K_r m(t) + i K_r v_m(t-t_3-t_{12}) - 2\gamma_{ac} t_{12} - \gamma_{ad} t_{23})} R_m
+ i \frac{\mu_{ab} \Omega_2}{\hbar \Delta_1} \int_{-\infty}^{t} dt' A[r_m(t'), t'] e^{iK' r_m(t')}.
$$

Substituting this equation into Eq. (5) one arrives at an expression for the optical coherence,

$$
\rho_{ab;m}(t) = i \frac{\mu_{ab} |\Omega_2|^2}{\hbar \Delta_1^2} e^{iK_r m(t)}
\times \left( e^{-\gamma_{ac}(t-t_3-t_{12}) - \gamma_{ad} t_{23}} \int_{-\infty}^{\infty} d\tau A^*_{1}[r_m(t_1), t_1 + \tau] e^{iK_r v_m(t-t_3-t_{12}+\tau)}
- \int_{-\infty}^{t} dt' A^*[r_m(t'), t'] e^{iK_r v_m(t'-t')} \right),
$$

that can be used to calculate the macroscopic polarization density, which is written as

$$
P_s(r, t) = P_{s+}(r, t) e^{-iK_1 r - i\omega_1 t} + P_{s+}^*(r, t) e^{-(iK_1 r - i\omega_1 t)},
$$

incorporating the fact that the signal is phase-matched in the $-k_1$ direction only. The polarization density is comprised of two components. One of them represents the source term that gives rise to the restored signal. This contribution originates from the first term on the right hand side of Eq. (21) and is given by

$$
P_{s+}^{(1)}(r, t) = -i \frac{\mu_{ab} |\Omega_2|^2 N}{\hbar \Delta_1^2} e^{-\gamma_{ac}(t-t_3-t_{12}) - \gamma_{ad} t_{23}}
\times \int d\mathbf{v} W(\mathbf{v}) \int_{-\infty}^{\infty} d\tau \mathcal{A}_1[r(t_1), t_1 + \tau] e^{-iK_r v(t-t_3-t_{12}+\tau)},
$$

where $\mathcal{A}_1[r(t_1), t'] \approx \mathcal{A}_1[r - vt_{13}, t']$. The polarization density depends on the field experienced by the participating atoms at their positions when they interacted with the signal field.

For the field to be restored, the condition $\mathcal{A}_1[r_m(t_1), t'] \approx \mathcal{A}_1[r, t']$ must hold. This condition is valid provided the distance travelled by the atoms in time $t_{13}$ is much smaller than the beam diameter, the spatial width of the input signal envelope, and the absorption
length $\alpha_R^{-1}$. Since $\delta_s << Ku$, one can take $A_1 [r, t]$ out of the integral over $t'$ and evaluate it at time $t' = t_1 - (t - t_3 - t_{12})$, which leads to

$$P_{+s}^{(1)} (r, t) = -i \frac{[\mu_{ab}]^2 [\Omega_2]^2 N}{h \Delta_1^2} e^{-\gamma_ac(t-t_3+t_{12})-\gamma_adt_{23}}$$

$$\times A_1 [r, t_1 - (t - t_3 - t_{12})] \int dv W(v) \int_{-\infty}^{\infty} d\tau e^{-iKv\tau}$$

(23)

Using Eqs. (12) and (13), one can re-express this in terms of $\alpha_R$ as

$$P_{+s}^{(1)} (r, t) = -2i\epsilon_0 \frac{\alpha_R}{k} A_1 [r, t_1 - (t - t_3 - t_{12})] e^{-2\gamma_ac(t-t_3-t_{12})}$$

(24)

The amplitude $A_1 [r, t_1 - (t - t_3 - t_{12})]$ is nonvanishing only for $t \approx t_3 + t_{12}$, reflecting the fact that Doppler rephasing occurs only for such times. We have used this fact to set $e^{-\gamma_ac(t-t_3-t_{12})} \approx e^{-2\gamma_ac(t-t_3-t_{12})}$.

The input signal was attenuated as it traveled through the storage medium. Taking into account the propagation and attenuation of the input field $A_1 [r, t]$, we can write $P_{+}^{(1)} (r, t)$ in terms of the input field at $z = 0$ as

$$P_{+s}^{(1)} (z, t) = -2i\epsilon_0 \frac{\alpha_R}{k} A_1 [0, t_1 - (t - t_3 - t_{12}) - z/c] e^{-\alpha_R z/2} e^{-2\gamma_ac(t-t_3-t_{12})}$$

(25)

the $z$ axis being directed along $k_1$.

The contribution to the polarization density from the second term on the right hand side of Eq. (21) corresponds simply to depletion of the restored field as a result of Raman transitions from level $a$ to $c$. This component can be written as

$$P_{+s}^{(2)} (z, t) = i\epsilon_0 \frac{\alpha_R}{k} A(z, t).$$

(26)

Finally, the restored field is a solution of the linearized Maxwell equation:

$$-\partial_z A(z, t) + \frac{1}{c} \partial_t A(z, t) = i \frac{k}{2\epsilon_0} \left[ P_{+s}^{(1)} (z, t) + P_{+s}^{(2)} (z, t) \right]$$

(27)

Making the change of variables $z' = z, t' = t + z/c$, and combining Eqs. (23) and (27), one obtains

$$\partial_{z'} A'(z', t') = -\alpha_R A_1 [0, t_1 - (t' - t_3 - t_{12})] e^{-\alpha_R z'/2} e^{-2\gamma_ac(t-t_3-t_{12})} + \frac{1}{2} \alpha_R A'(z', t'),$$

(28)

where $A'(z', t') = A(z', t' - z'/c)$. Solving this equation with the initial condition $A'(L, t') = 0$, we find

$$A(z, t) = A_1 [z, t_1 - (t - t_3 - t_{12})] e^{-2\gamma_ac(t-t_3-t_{12})} \left( 1 - e^{-\alpha_R (L-z)} \right).$$

(29)
For $\alpha_R L \gg 1$, the pulse is totally restored at time $t = t_3 + t_{12}$, neglecting decay of the Raman coherences. The latter equation represents the main result of the paper, showing the absence of distortion of the retrieved signal and its variation as a function of the optical depth $\alpha_R L$.

One can also verify that the atoms return to their initial state as the signal field is restored. Substituting Eq. (29) into Eq. (20) and using Eq. (15), one obtains

$$\tilde{\rho}_{ac;m}(t) = -i \frac{\mu_0 \Omega_2}{\hbar \Delta_1} e^{-i \mathbf{K} \cdot \mathbf{r}_m(t) + i \mathbf{K} \cdot \mathbf{v}_m(t - t_3 - t_{12}) - \gamma_{ac}(2t_{12} + t_{13})} \times \left( \int_{-\infty}^{\infty} dt' A_1^*(z, t') e^{i \mathbf{K} \cdot \mathbf{v}_m(t' - t_1)} - \left( 1 - e^{-\alpha_R(L-z)} \right) \int_{t_1 - (t_3 - t_{12})}^{\infty} dt' A_1^*(z, t') e^{i \mathbf{K} \cdot \mathbf{v}_m(t' - t_1)} \right).$$

For times $(t - t_3 - t_{12}) > 0$ the lower limit on the second integral can be replaced by $-\infty$ and this term cancels the first integral. In other words, the coherence vanishes once when the signal field is restored, provided $\alpha_R L \gg 1$.

Following the emission of the restored signal pulse, there remains in the medium a Raman coherence given by

$$\tilde{\rho}_{ac;m}(t) = -i \frac{\mu_0 \Omega_2}{\hbar \Delta_1} e^{-i \mathbf{K} \cdot \mathbf{r}_m(t) + i \mathbf{K} \cdot \mathbf{v}_m(t - t_3 - t_{12}) - \gamma_{ac}(2t_{12} + t_{13})} e^{-\alpha_R(L-z)} \int_{-\infty}^{\infty} dt' A_1^*(z, t') e^{i \mathbf{K} \cdot \mathbf{v}_m(t' - t_1)}.$$  

(31)

Comparing with expression with Eq. (3), and noting that the population in state $|a\rangle$ is approximately equal to unity, one can conclude that the population left in state $|c\rangle$ is

$$\rho_{cc;m}[(t_3 + t_{12})^+] = \left| \tilde{\rho}_{ac;m}(t_1^-) \right|^2 \left[ 1 - e^{-2\gamma_{ac}(t_{12} + 2t_{13})} \right] + \left| \tilde{\rho}_{ac;m}[(t_3 + t_{12})^+] \right|^2 \left[ 1 - e^{-\gamma_{ac}(2t_{12} + t_{13})} (1 - e^{-\alpha_R(L-z)}) \right].$$  

(32a)

Finally, summing over $z$, one finds the fraction $\eta$ of the initially excited atoms that is left in state $|c\rangle$ is given by

$$\eta = 1 - e^{-2\gamma_{ac}(t_{12} + 2t_{13})} \left( 1 - e^{-\alpha_R(L-z)} \right).$$  

(33)

If decay of the Raman coherence decay can be neglected, $\eta = e^{-\alpha_R L}$. During the storage process, a fraction $\eta_m = (1 - e^{-\alpha_R L})$ of the incoming signal radiation is used to excite the atoms to level $c$, the remaining signal passes through without being scattered. From the part that is stored, a fraction is lost at retrieval, even in the absence of relaxation. Indeed the restored field is $(1 - e^{-\alpha_R L})$ times smaller than the incoming one, according to Eq. (29).
Therefore one recovers a fraction $W_{\text{out}}/W_{\text{in}} = (1 - e^{-\alpha R L})^2$ of the incoming energy. The difference $\eta_{\text{in}} - W_{\text{out}}/W_{\text{in}} \approx e^{-\alpha R L}$ corresponds to the fraction $\eta$ of the excited atoms that is left in level $c$. To summarize, with a finite length material, information is lost in equal amounts at storage and retrieval, the storage loss associated with an incomplete depletion of the input field and the retrieval loss associated with excited state population $\rho_{cc}$ that is left in the medium.

VI. DISCUSSION

We have shown that nearly 100% recovery efficiency can be reached with the proposed quantum storage Raman scheme. The memory bandwidth, determined by the Raman transition Doppler broadening, can be tuned continuously from 0 to $2k u$. Since the atoms are not excited to the upper electronic level, the control pulse duration is not limited by the upper level lifetime, but only by the inverse signal bandwidth.

Our motivation in this work was to circumvent the time scale conditions imposed by the short upper level lifetime, since it is not easy to produce large area, large waist coherent pulses on the nanosecond time scale. The pulses to be considered here, whether input signal or control pulses, will most likely last for tens or hundreds of nanoseconds. This corresponds to a spectral width much smaller than the Doppler width $k u$, typically of order $10^9 s^{-1}$ or higher. The available bandwidth of the Raman scheme is determined by $K u$, where $K/k$ can be expressed in terms of the angle $\theta = (k_1, k_2)$ as $K/k = 2\theta \sin(\theta/2)$. According to Eq. (29), the Raman optical depth $\alpha R L$ should be as large as possible. Since $\alpha R L$ is inversely proportional to $K/k$, the value of $K/k$ should be matched to the bandwidth of the input pulse. For an input field spectral width of order 10 MHz, we are led to a nearly co-propagating configuration with $\theta$ of order 10 mrad.

The memory lifetime is limited by the atomic motion. As noticed in Section IV, the distance travelled by the atoms during the entire process should be much smaller than the beam diameter and the absorption length $\alpha_R^{-1}$. With a typical average speed of a few 100m/s, a memory lifetime of a few tens of microseconds limits the sample size to a few centimeters.

A critical issue in a multilevel system is the ability to selectively drive the target transitions. Field polarization can be used to provide the selectivity if the angle between the fields is small (or close to $\pi$); in this limit, one can use circularly polarized fields, as shown
FIG. 3: (Color online) Example of a possible excitation scheme. The Raman transitions connect the Zeeman sublevels of $F = 1$, $F = 2$ hyperfine levels. Each Raman transition involves a $\sigma_+$ and a $\sigma_-$ polarized beam.

Optical pumping by the strong control field determines the uncoupled initial state ($F = 1$, $m = -1$). In step (a), fields 1 and 2 create a Raman coherence $\rho_{1,-1;1,1}$, where the notation is $\rho_{F,m:F',m'}$. The Doppler dephasing is then stopped by a $\pi$-pulse, composed of two beams propagating along $k_3$ and $k_4$ in such a way that $k_4 - k_3 = k_1 - k_2$ [step (b)]. These fields convert the coherence $\rho_{1,-1;1,1}$ into $\rho_{1,-1;2,-1}$. The process in which fields 4 and 3 drive Raman transitions between states $|1,-1\rangle$ and $|1,1\rangle$ is suppressed, provided the hyperfine frequency separation is much larger the inverse pulse duration. In step (c) a Raman $\pi$ pulse having $k'_4 - k'_3 = -(k_4 - k_3)$ restores the $\rho_{1,-1;1,1}$ coherence and prepares the system in such
a fashion that the subsequent application of a control pulse having propagation vector \(-k_2\) restores the input signal field propagating in the \(-k_1\) direction at time \(t = t_3 + t_{12}\).

VII. CONCLUSION

We have proposed a Raman based quantum memory scenario that operates with close to 100\% efficiency. This protocol fits well to intermediate time scales, with signal duration of order 100ns, a time range that is well adapted to experiments based on high spectral purity continuous wave laser sources. We have shown that the spectral components are stored in different atomic velocity classes and have explained how to optimize the optical depth depending of the needed storage bandwidth. Since we have assumed that the signal field is weak and since spontaneous emission is negligible in this protocol, we expect the results to be unchanged if the classical input field is replaced by a quantized, pulsed radiation field.

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[25] In the MK protocol one prevents the control field from interacting with the populated level, not only to ensure uniform illumination of the medium, but also to avoid excitation to the
upper level that would entail amplification and spontaneous emission noise. This cannot occur in the present protocol.

[26] This connection between the inhomogeneous phase shift and spatial position is unique to the Doppler effect. The phase shift is connected simply to the distance travelled by the atoms. As a consequence, even a short laser pulse can result in an accumulated Doppler phase shift for the atoms. This rather paradoxical feature does not occur when the inhomogeneous broadening is caused by position dependent energy level shifts of stationary atoms, as in a solid.