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Ultraslow propagation of an optical pulse in a three-state active Raman gain medium

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We investigate an active Raman gain scheme for significant group velocity reduction. We show that this scheme, which is fundamentally different from the electromagnetically induced transparency scheme, is capable of achieving ultraslow and distortion-free propagation of a pulsed probe field. We demonstrate the group velocity behavior that is drastically different from the conventional electromagnetically induced transparency scheme, and we show that the new scheme can be used to accurately determine the decoherence rate of a long-lived state. In addition, the Raman gain scheme has the advantage of being broadly tunable, an important feature that may have potential applications.

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The propagation of optical fields in dispersive media has been extensively studied [1–6]. In general, the index of refraction of an optical medium changes rapidly when the center frequency of an optical field is tuned close to one of the strong resonances of the medium. This rapid change of the index leads to significant modification of the propagation velocity of the optical field, which is inversely proportional to the slope of the dispersion function in frequency domain. The rapidly changing region of the refractive index usually, however, overlaps with the peak absorption of the field, preventing the region from being used for active wave propagation control. An index manipulation technique such as electromagnetically induced transparency (EIT) [7] can circumvent this difficulty. Using this technique, several groups [8–11] have demonstrated many order-of-magnitude reductions of the group velocity of optical pulses in highly resonant media. In addition, a passive Raman scheme [12,13] (i.e., no gain) has also been demonstrated to be capable of achieving ultraslow group velocity.

In this Rapid Communication we report an ultraslow wave propagation scheme that is very different from the EIT and passive Raman schemes [8–13]. Contrary to the conventional EIT scheme, which relies on a transparency window created in the frequency spectrum of a strongly absorbing medium, we use a far-detuned, active Raman gain scheme. As we will show, such a system (1) is capable of significant group velocity reduction, (2) has a very different group velocity dependence on the driving field, (3) has excellent signal to noise (S/N) ratio due to the Raman gain and probe field being distortion free, (4) allows accurate determination of the decoherence rate of a long-lived coherent population trapping state, and (5) has the advantage of being widely tunable. This last feature may have applications in optoelectronics where tunable and distortion-free ultraslow optical wave propagation are desirable.

Our experiment is based on the active Raman gain scheme studied by Payne and Deng [14]. It was predicated that, for a far-detuned three-state Λ scheme with active Raman gain, the group velocity of an optical pulse can be reduced significantly when the two-photon resonance is maintained. Furthermore, the driving field power dependence of the group velocity is opposite to that encountered in the EIT scheme. More importantly, under the same driving conditions the active Raman gain scheme often performs better than the EIT scheme [15]. Because of the large detuning and the active gain, the probe field maintains or gains its amplitude, resulting in excellent S/N ratio and distortion-free propagation. This is to be contrasted to the EIT-based schemes where attenuation and pulse shape distortion can be severe under ultraslow propagation conditions [16].

We use ^{85}Rb to demonstrate the active Raman gain, far-detuned three-state Λ scheme for significant group velocity reduction. An external cavity diode laser system produces nearly 400 mW power and is locked to a rubidium reference cell. The output of the laser passes through a 3 GHz acousto-optical modulator (AOM) to generate two laser beams with frequency difference of the ground state hyperfine levels $|1\rangle=|5S_{1/2},F=2\rangle$ and $|3\rangle=|5S_{1/2},F=3\rangle$ of ^{85}Rb atoms. The AOM is driven by a rf synthesizer which allows accurate adjustment so that the two-photon resonance can be precisely achieved and maintained. The rubidium cell is 100 mm in length and 25 mm in diameter with antireflection coating on both ends, and is placed in a temperature controlled enclosure. Experiments were carried out in the temperature region of $T=60$ °C to $T=90$ °C.

Figure 1 shows the energy level diagram and relevant laser couplings for the present study. The probe and the driving fields are jointly tuned to and kept on $|1\rangle=|5S_{1/2},F=2\rangle\to|3\rangle=|5S_{1/2},F=3\rangle$ two-photon resonance. Both lasers are detuned, however, on the high energy side of the $5P_{1/2}$ hyperfine manifold with a large (3 GHz) one-photon detuning. Consequently, the driving field is also on $|1\rangle=|5S_{1/2},F=2\rangle\to|2\rangle=|5P_{1/2},F=3\rangle$ resonance, acting as an optical pumping field that converts and maintains all population in the state $|3\rangle=|5S_{1/2},F=3\rangle$ (see discussions later). The off-resonance driving field couples the $|3\rangle=|5S_{1/2},F=3\rangle\to|2\rangle=|5P_{1/2}\rangle$ transition, creating a medium with active Raman gain. It is on this Raman gain curve where we study the propagation characteristics of a weak pulsed probe field.

The system described above can be understood using a simple lifetime broadened three-state model where nearly all population remains in the initial state $|3\rangle$ (Fig. 1). This is because the one-photon detuning is sufficiently large and the ac Stark shift produced in the $5P_{1/2}$ hyperfine states by the

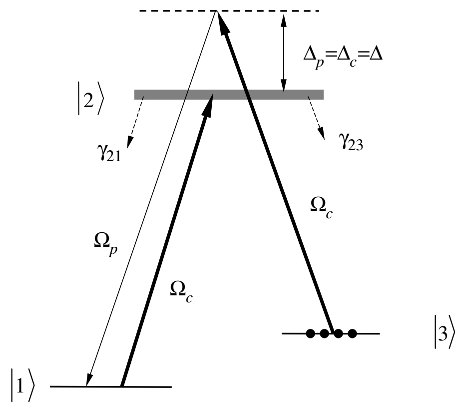

FIG. 1. Energy level diagram and relevant laser couplings. Level assignment: $|1\rangle=|5S_{1/2},F=2\rangle$, $|2\rangle=|5P_{1/2},F=3\rangle$, $|4\rangle=|5P_{1/2},F=2\rangle$, and $|3\rangle=|5S_{1/2},F=3\rangle$. Because of the large one-photon detuning, the hyperfine splitting will be neglected and $|2\rangle$ and $|4\rangle$ will be treated as a single level. Possible four-wave mixing from $|2\rangle$ to $|3\rangle$ is not shown.

optical pumping field under our driving conditions is small. Consequently, Doppler broadenings of the hyperfine states are unimportant and the contributions by the two hyperfine states can be easily included. For simplicity we neglect the optical pumping field, the Doppler broadening, and the hyperfine splitting. We further assume that nearly all population is maintained in the state $|3\rangle$. Since the one-photon detuning is very large and the probe field is weak [12,13], there is never appreciable population that can be transferred to the state $|1\rangle$. The equations of motion for the relevant density matrix elements are given as (assuming $\rho_{33}\approx 1$)

$$\frac{\partial \rho_{12}}{\partial t} \approx -i\Omega_c^* e^{i\Delta t}\rho_{13} - \gamma_{12}\rho_{12}, \tag{1a}$$

$$\frac{\partial \rho_{13}}{\partial t} \approx i\Omega_p^* e^{i\Delta t}\rho_{23} - i\Omega_c e^{-i\Delta t}\rho_{12} - \gamma_{13}\rho_{13}, \tag{1b}$$

$$\frac{\partial \rho_{23}}{\partial t} \approx i\Omega_p e^{-i\Delta t}\rho_{13} + i\Omega_c e^{-i\Delta t} - \gamma_{23}\rho_{23}, \tag{1c}$$

where $\Omega_p=D_{21}E_{p0}/(2\hbar)$, $\Omega_c=D_{23}E_{c0}/(2\hbar)$, and γ_{ij} are the half Rabi frequencies of the probe and pumping fields, and the decoherence rate of the respective transitions. In addition, $\Delta_p=\Delta_c=\Delta$ is the one-photon detuning to the state $|2\rangle$ by the probe and pump fields. In our experiment these two one-photon detunings are identical so that the two-photon resonance is maintained.

Payne and Deng have shown that the positive frequency part of the probe field amplitude, in the case of unfocused plane wave and within the slowly varying amplitude and adiabatic approximations, must satisfy the wave equation which, in the Fourier transform space, is given by

$$\frac{\partial \Lambda_p}{\partial z} - i\frac{\omega}{c}\Lambda_p = \frac{i\kappa_{12}|\Omega_c|^2 W(\omega)}{(\Delta - i\gamma_{23})(\Delta + i\gamma_{21})}\Lambda_p, \tag{2a}$$

$$W(\omega) = \frac{1}{(\omega + i\gamma_{31}) - \dfrac{|\Omega_c|^2}{(\Delta + i\gamma_{21})}}, \tag{2b}$$

where Λ_p is the Fourier transform of the probe field Rabi frequency Ω_p and ω is the Fourier transform variable. In addition, $\kappa_{12}=2\pi N_0\omega_p|D_{21}|^2/c$ where N_0, ω_p, and D_{12} are the concentration, probe transition frequency, and the corresponding dipole moment for the probe transition, respectively. Equations (1) and (2) must be solved self-consistently in order to correctly predict the propagation characteristics of the probe field. Payne and Deng have carried out a non-steady-state calculation and shown the group velocity of a probe pulse as

$$V_g \approx \frac{1}{\kappa_{12}}\left(\frac{\gamma_{31}\Delta}{2\Omega_c}\right)^2, \tag{3}$$

where $|\Delta| \gg \gamma_{21}, \gamma_{23}$ and $|\Delta| \gg |\Omega_c|$ have been used. Equation (3), which shows an *inverse quadratic dependence* on the driving field Rabi frequency, is very different from the EIT scheme where the slow group velocity shows a *quadratic dependence* on the driving field Rabi frequency. Our experiment is aimed at demonstrating three predictions given in Eq. (3): (1) the existence of the nondistorted (i.e., probe field attenuation and spreading) ultraslow propagation in an active Raman gain medium; (2) the characteristic inverse parabolic dependence of the group velocity on the driving field Rabi frequency; and (3) quadratic dependence on the large one-photon detuning Δ.

In Fig. 2 we show a delayed probe propagation (left panel) and a Raman gain spectrum (right panel). For a $\tau=25$ μs probe pulse [17], the delay is $\delta t=2.1$ μs. This corresponds to a group velocity of $v_g\approx 1.6\times 10^{-4}c$, where c is the speed of light in vacuum. The delay fraction is about 8.4%. If the identical driving condition is used in an EIT scheme one would have a group velocity that is about 20 times faster [15]. Thus, even though an EIT scheme may be able to reach much slower group velocity [8–11] at a great expense of significant probe pulse distortion and attenuation (typically worse than 80% [16]), in regions where an EIT scheme shows small loss and distortion, the Raman gain medium is superior to the EIT scheme [15]. It should be noted that the 8.4% delay fraction of the probe pulse is remarkable for a complex system such as room-temperature ^{85}Rb. At the room-temperature ^{85}Rb is far more complex than ^{87}Rb because the hyperfine splitting is much smaller than the mean Doppler width of the line. This implies that both $F=2$ and $F=3$ levels of the $5P_{1/2}$ manifold must be treated on an equal footing for the overall system response. Theoretical investigation indicates that the absorption is far more severe with ^{85}Rb than with ^{87}Rb. This is one of the primary reasons why room-temperature ^{85}Rb is not suitable for an EIT type of group velocity reduction experiment and therefore studied much less for this purpose. This complexity, however, gives rise to very rich dynamic behavior of the ^{85}Rb [18].

In Fig. 3 we show the group velocity of a probe pulse as a function of the driving field Rabi frequency. Contrary to the EIT scheme where the group velocity of a probe pulse

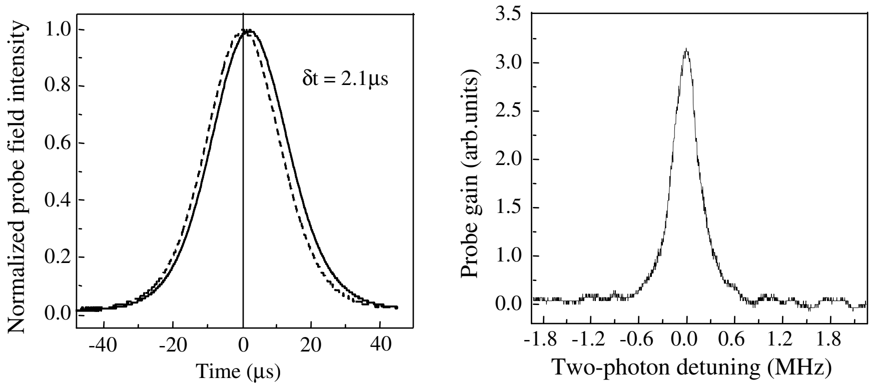

FIG. 2. Left panel: Typical data showing the delayed propagation of a $\tau=25$ μs probe pulse. Right panel: two-photon Raman gain as a function of two-photon detuning. In both data $|\Omega_c| \approx 2\pi\times 25$ MHz, and temperature $T=60$ °C. The two traces in the left panel have been normalized to show the preservation of the pulse shape. Here, the dashed line represents the reference pulse which travels through the air whereas the solid line depicts the probe pulse that has propagated through the active Raman medium.

under the weak driving condition is a *quadratic* function of the driving field Rabi frequency, the group velocity of the active Raman gain medium is an *inverse quadratic* function of the driving field Rabi frequency. This observation agrees well with Eq. (3) which was first predicted in Ref. [14]. In the slowest group velocity region the Raman gain is about a factor of 2.

In Fig. 4 we show the group velocity of a probe pulse as a function of the one-photon detuning Δ of the driving field with two-photon resonance being maintained. As expected from Eq. (3), the group velocity increases as a quadratic function of the one-photon detuning. This quadratic behavior is also in accord with the passive Raman scheme for ultraslow propagation velocity reported before [12,13].

We note that, in our experiment, we have observed a relative flat group velocity region of about 500 MHz (one-photon detuning near the smallest group achievable under the conditions used). This is an important feature of the active Raman gain scheme. Indeed, this relative insensitive group velocity on a wide range of one-photon detuning is one of the advantages of the present scheme over the EIT scheme which requires that both fields be on exact one-photon resonance in order to avoid unmanageable probe field loss and distortion. In the case of the usual three-state Λ type EIT scheme, the probe field spectrum must be sufficiently narrower than the induced transparency window which becomes increasingly small as the driving field is reduced for ultraslow propagation. In the case of Raman gain scheme with large one-photon detunings the different frequency components of a broad bandwidth optical field experience the same velocity reduction effect, making it potentially useful for possible broad bandwidth optoelectronic devices. We further note that the probe pulse exhibits excellent Gaussian shape. No detectable pulse shape distortion has been found in the χ-square fitting routine.

It is important to point out that Eq. (3) can be used to obtain accurate experimental value of the decoherence rate γ_{31} at room temperature. This is another key feature of this scheme. The Raman gain nature of the scheme gives excellent S/N ratio and sufficient time delay so that group velocity

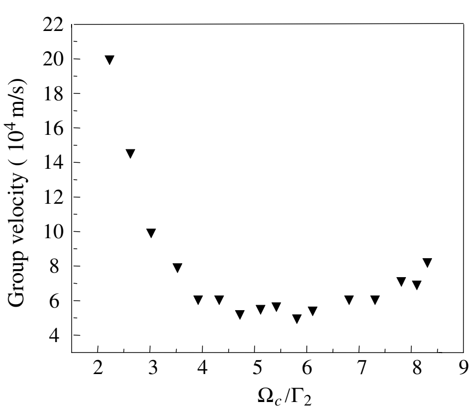

FIG. 3. Plot of the probe pulse group velocity as a function of driving field Rabi frequency. The data shows the inverse quadratic dependence as predicted in Eq. (3). The horizontal axis is in the unit of $\Gamma_2=2\pi\times 5.6$ MHz. At higher driving field deviation from non-depleted ground state approximation occurs and the group velocity increases, as indicated at large Ω_c. $T=60$ °C.

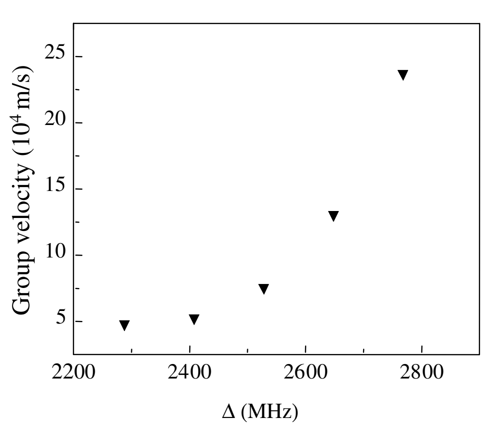

FIG. 4. Plot of the probe pulse group velocity as a function of driving field detuning Δ. The data show the expected quadratic dependence as predicted in Eq. (3). Near the bottom of the curve there is a region where the group velocity does not change significantly as the detuning changes for about several hundred MHz. For this plot $|\Omega_c|=2\pi\times 25$ MHz and $T=60$ °C.

can be very accurately measured. Since the accuracy of the detuning and Rabi frequency measurements can be well maintained, this yields an accurate value of the decoherence rate. Such information is not obtainable from the EIT related group velocity measurement since the decoherence rate γ_{31}, to the first order, does contribute to the group velocity. On the other hand, this decoherence rate is the leading contribution to the probe pulse attenuation in the EIT scheme [16]. Currently, the only direct way [19] to measure the decoherence rate γ_{31} at room temperature is to measure the decay of the light initiated from the state $|3\rangle$ at different "storage" time in the light storage and retrieval method (or variations of that scheme) [20]. One then fits peaks of the signal to an exponential function to deduce the decoherence rate. Such a procedure involves many uncertainties such as the propagation dependent absorption and pulse distortion. Both effects can significantly alter the amplitude of the signal which is the basis for fitting to an exponential function.

It is also worth pointing out some other differences between the present scheme and the usual EIT scheme. In the usual three-state Λ type EIT scheme the transparency is a result of interference between two one-photon channels and is enabled only with an intense driving field. In the present scheme, the absorption is reduced by the large one-photon detuning. The active Raman gain modifies the dispersion properties of the atomic system, leading to steep slope and sharp dispersion structures and resulting in nondistorted ultraslow propagation of the probe pulse tuned on two-photon resonance.

We finally comment on the possible four-wave mixing (FWM) process. Since the driving field E_c is also on resonance with $|1\rangle-|2\rangle$ transition it is possible that a FWM process $|2\rangle-|3\rangle$ can occur (both optical pumping and FWM fields are neglected in our simple model). Under this circumstance we have the combination of a Λ scheme with active gain ($|3\rangle \xrightarrow{\Omega_c} |2\rangle \xrightarrow{\Omega_p} |1\rangle$) and a Λ-EIT scheme with absorption ($|1\rangle \xrightarrow{\Omega_c} |2\rangle \xrightarrow{\Omega_m} |3\rangle$). Since the FWM field, together with Ω_c, forms the EIT scheme it also propagates with a significantly reduced group velocity. This interesting double Λ scheme with gain-loss combination is very different from the double Λ scheme widely studied in literature, where both single Λ branches are EIT in nature (i.e., attenuation). Further studies of this gain-loss combination double Λ scheme are ongoing.

In summary, we have demonstrated experimentally significant group velocity reduction of an optical pulse using a three-state *active Raman gain scheme*. We have shown that the new scheme exhibits very different driving field dependence characteristics in comparison with the three-state absorptive EIT scheme, and it also has the advantage of being broad tunable. In addition, we have shown that the new scheme allows a direct, nondecay type of measurement of the decoherence rate of the members of the ground state manifold.

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[15] For example, with $|\Omega_c|=2\pi\times 25$ MHz, $\gamma_{13}=2\pi\times 100$ kHz, $\Delta_c=\Delta_p=\Delta=2\pi\times 3$ GHz, and $\kappa_{12}=10^{10}/(\mathrm{cm\,s})$, a lifetime broadened three-state system yields $V_g^{(Raman)}\approx 0.06V_g^{(EIT)}$, where $V_g^{(EIT)}=|\Omega_c|^2/\kappa_{12}$ is the group velocity obtainable with the usual EIT scheme under the same driving conditions.
[16] Note that the leading contribution to the probe field attenuation in a three-state EIT scheme is $e^{-\kappa_{12}z\gamma_{13}/|\Omega_c|^2}$. For $\kappa_{12}=10^{10}/(\mathrm{cm\,s})$ and $|\Omega_c|=2\pi\times 10$ MHz, even with $\gamma_{13}=2\pi\times 10$ kHz, the probe attenuation for a 10 cm propagation is still about $e^{-1.6}\approx 0.2$, i.e., 80%. See L. Deng *et al.*, Opt. Commun. **212**, 101 (2002).
[17] With $\tau=25$ μs, $\gamma_{13}\tau\approx 12\gg 1$ so the conditions used in deriving Eq. (3) are well satisfied; see Ref. [14].
[18] A detailed theoretical study that compares room-temperature ^{85}Rb and ^{87}Rb for both EIT and active Raman gain schemes will be published separately.
[19] The decoherence rate γ_{13} can also be deduced or inferred using other techniques. For instance, it can be deduced from the EIT bandwidth measurements at low intensities.
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