Ultraslow Propagation of Matched Pulses by Four-Wave Mixing in an Atomic Vapor

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We have observed the ultraslow propagation of matched pulses in nondegenerate four-wave mixing in a hot atomic vapor. Probe pulses as short as 70 ns can be delayed by a tunable time of up to 40 ns with little broadening or distortion. During the propagation, a probe pulse is amplified and generates a conjugate pulse which is faster and separates from the probe pulse before getting locked to it at a fixed delay. The precise timing of this process allows us to determine the key coefficients of the susceptibility tensor. The fact that the same configuration has been shown to generate quantum correlations makes this system very promising in the context of quantum information processing.

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Slow group velocities, valuable for all-optical signal processing, are obtained at a resonance peak of the transmission spectrum of a medium, and a number of different implementations of this principle have been demonstrated. They rely either on a reduction of the absorption, such as electromagnetically induced transparency (EIT) [1], coherent population oscillations [2], and dual absorption lines [3], or on a gain resonance, like stimulated Brillouin scattering [4] and stimulated Raman scattering [5]. To be useful in the context of all-optical signal processing, an optical delay line should be able to produce a fractional delay (defined as the ratio of the delay to the duration of the pulse) larger than unity with only modest absorption and pulse broadening. Recent developments [4,6,7] have shown the benefits of using an amplifying medium to alleviate the absorption and distortion issues usually associated with slow light [8].

We have examined the group velocity reduction effects due to nondegenerate four-wave mixing (4WM) in hot rubidium vapor and have obtained large and tunable fractional delays with very little distortion. The presence of gain in this system makes it in principle possible to stack such delay lines and achieve fractional delays only limited by pulse broadening. Another notable feature of the amplification in the 4WM process is the generation of a conjugate pulse which is coupled to the probe and which propagates alongside it, similar to matched pulses in EIT systems [9]. We have studied the interplay between the 4WM coupling of the probe and conjugate and the Raman coupling of the probe and pump and showed that the system embodies the paradigm of the ideal double-lambda system [10]. This interplay leads to the ultraslow propagation of matched probe and conjugate pulses and to the enhancement of the 4WM gain. Such an enhancement, when applied to cross-phase modulation, is the key element of recent optical quantum information processing proposals [11–13]. The system that we study has already been shown to generate intensity-difference squeezing between the probe and conjugate beams in a cw regime under similar conditions [14], and quantum-correlated matched pulses generated in this way could also be a useful resource for quantum information processing.

Our apparatus is described in Ref. [14], and consists of a linearly polarized, continuous, strong (up to 280 mW) pump and a cross-polarized, pulsed, weak (0.5 mW) probe propagating at a small angle (0.5°) through a 2.5 cm-long ^{85}Rb cell heated to 90–140 °C [Fig. 1(a)]. The pump and the probe are near-resonant with a Raman transition between the two hyperfine electronic ground states of ^{85}Rb, with a controllable detuning δ [Fig. 1(b)] and have $1/e^2$ radii of 600 and 350 μm, respectively. The residual 2-photon Doppler broadening due to the small angle is a few MHz. The detuning from the $5P_{1/2}$ excited state is $\Delta_1/2\pi \approx 850$ MHz, and the peak pump intensity (up to 45 W/cm^2) is high enough to excite off-resonant Raman transitions with a detuning $\Delta/2\pi \approx 4$ GHz from the excited state. The double-lambda [10] is closed by the con-

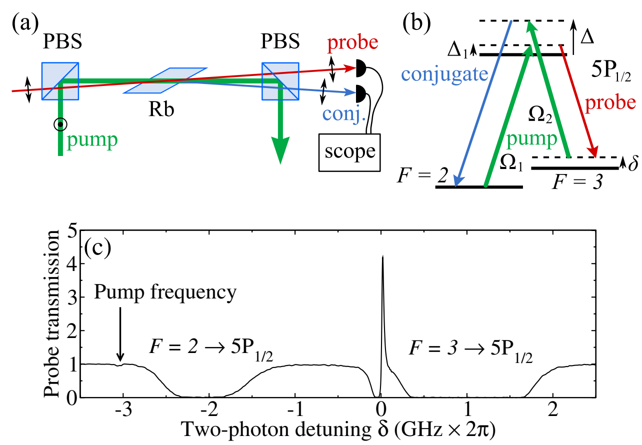

FIG. 1 (color online). (a) Experimental setup. PBS: polarizing beam splitter. (b) Energy-level diagram of the D1 line of ^{85}Rb, showing the double-lambda scheme. Note that the pumps Ω_1 and Ω_2 are in fact the same laser beam. (c) Probe transmission profile versus two-photon detuning δ.

jugate beam which emerges on the other side of the pump from the probe, with the same polarization as the probe. The combination of the beam polarizations and the Zeeman substructure makes the system a four-level system (the two virtual excited states are orthogonal). The probe amplification is sharply resonant in δ, as shown in Fig. 1(c), with a gain that can reach 30. The gain feature leads to a strong dispersion of the index of refraction and thus a low group velocity for the probe.

We measure the group velocity delay by recording the arrival time of a 70 ns-long (full width half maximum [FWHM]) Gaussian probe pulse with and without the atomic medium (reference pulse). Figure 2(a) shows an example in which the parameters are set to provide a large probe gain ($G = 13$). By varying the two-photon detuning δ and the pump intensity, one can tune the probe delay and achieve a fractional delay larger than 0.5 such that the pulse remains Gaussian and is broadened by less than 10% of its original width. If one accepts a variable gain, the tunability extends from zero to the maximum attainable delay. This low level of distortion is remarkable in comparison with that seen in some EIT experiments [8]. The maximum delay corresponds to a group velocity of $c/500$, where c is the speed of light in vacuum. It is in general possible to tune the pump intensity, the pump and probe detunings, and the temperature to achieve an overall gain of unity. Figure 2(a) also shows the record of the conjugate intensity. A striking feature is the emergence of the conjugate pulse *before* the probe pulse. This relative delay is a fundamental property of the dynamics of the system and was predicted and observed in Refs. [15–17] in the case of resonance on the "lower" lambda transition ($\Delta_1 = 0$).

Larger delays, shown in Fig. 2(b), can be achieved by setting δ between the gain peak and the Raman absorption dip present at $\delta \lesssim 0$ [see Fig. 1(c)]. The competition between large amplification and large absorption leads to complex dynamics which can result in pulse breakup, in a similar fashion to the dual-field solitons predicted to exist in three-level systems [18]. Here, we restrict ourselves to the regions of low absorption where our system can be consistently described by the simple concepts developed in the theory of Refs. [16,19].

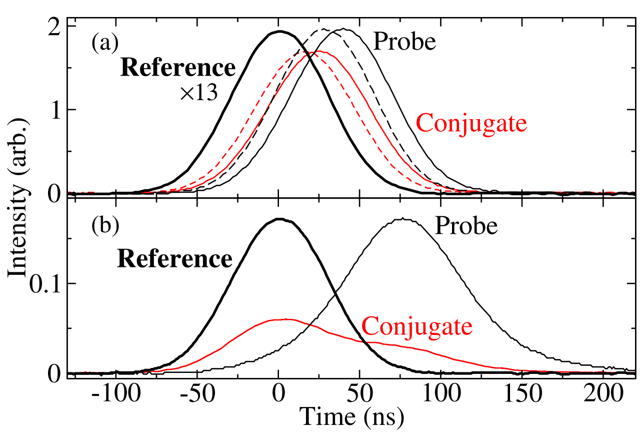

FIG. 2 (color online). (a) Slow-light effect near the peak of the resonance. The reference pulse is magnified 13 times. Thin solid and dashed lines: probe pulses for the parameters ($\delta/2\pi$, pump power) equal to (10 MHz, 280 mW) and (22 MHz, 200 mW), respectively. The smaller amplitude solid and dashed lines are the corresponding conjugate pulses. The probe pulse is broadened by 5% in the less retarded case and 10% in the more retarded case. (b) Slow-light effect closer to the Raman absorption dip.

We neglect the hyperfine splitting of the excited state. Averaged over the Zeeman substructure, the dipole moments of all four transitions are equal, which gives Ω_1 and Ω_2, the peak resonant Rabi frequencies of the pump for the lower and the "upper" lambda, respectively, the same value, denoted Ω. The double-lambda system in its ideal incarnation operates in the limit $\Delta_1 \ll \Delta$ and has the following crucial features. First, a ground state coherence is established by the "lower," more resonant lambda. The coherence has a lifetime $1/\gamma_c$, limited by magnetic fields, collisions, and the transit time in the laser beams, and corresponds to a dark state in which the absorption of the probe is reduced (EIT). Second, the "upper," less resonant lambda slightly perturbs this coherence and creates a resonant atomic polarization at the probe and conjugate frequencies via 4WM, while keeping the population in the excited state near zero. The dynamics of the system can thus be broken down into two intertwined processes: EIT and 4WM.

For a strong pump, most of the atomic population is in the ground state $F = 3$, and the Fourier components $\mathcal{E}_p(\omega)$ and $\mathcal{E}_c(-\omega)$ of the slowly-varying envelopes of the probe and conjugate fields (of wave vectors $\mathbf{k}_p$ and $\mathbf{k}_c$) obey the equations, in the adiabatic approximation [16,19]:

$$(i\omega + c\partial_z)\mathcal{E}_p = i\eta\Delta_R\mathcal{E}_c^* - \eta\left[i\left(\delta + \omega - \frac{\Omega^2}{4\Delta}\right) + \gamma_c\right]\mathcal{E}_p \tag{1}$$

$$(i\omega + c\partial_z)\mathcal{E}_c^* = -i\eta\Delta_R\mathcal{E}_p. \tag{2}$$

We assumed perfect phase matching, $\Delta \gg \Delta_1$, and low pump depletion. In the limit of $\Omega^2/4\Delta_1 \gg \delta$, γ_c, and $\Delta \gg \gamma$ (where $\gamma/2\pi = 6$ MHz is the linewidth of the atomic transition), the coefficients in Eqs. (1) and (2) are [19] $\eta = g^2N/[\Omega^2/4 + \Delta_1(\delta + \omega + i\gamma_c)] = c/v_g \gg 1$ and $\Delta_R = \Omega^2/4\Delta$. Here, $g^2 = ck\wp^2/(2\varepsilon_0\hbar)$, $k = k_p \approx k_c$, N is the atomic density, and $\wp$ is the average dipole moment acting on the probe and the conjugate.

The interpretation of Eqs. (1) and (2) is straightforward. The probe field $\mathcal{E}_p$ is slowed down by a factor η due to the EIT interaction with the pump [second drive term on the right-hand side of Eq. (1)]. The EIT resonance is light-shifted by the pump on the upper lambda and occurs at $\tilde{\delta} \equiv \delta - \frac{\Omega^2}{4\Delta} = 0$. In addition, $\mathcal{E}_p$ and $\mathcal{E}_c$ are cross-coupled with a coupling constant $\alpha = \eta\Delta_R$, responsible for the 4WM amplification. The presence of η highlights the role of the longer interaction time due to the slow-down effect in obtaining a sizeable nonlinear coupling [12]. The other factor in α, the so-called Raman bandwidth Δ_R, is the Rabi frequency of a fictitious resonant Raman transition driven on both legs by the pump field and with an intermediate Raman detuning Δ. As shown by the absence of any dependence on Δ_1, the 4WM dynamics is dominated by the upper lambda, which acts as a bottleneck. The propagation equations are asymmetrical, as the direct term for the conjugate is missing [Eq. (2)]. Its imaginary part would correspond to a slow-light effect and is negligible compared to the same term for the probe because $\Delta \gg \Delta_1$. Its real part would correspond to a Raman amplification, scaling as $1/\Delta^2$, and is negligible compared to the cross term α which scales as $1/\Delta$. As a result, in the absence of the 4WM coupling ("bare" fields), the probe and the conjugate propagate at velocities v_g and c, respectively. The finite decoherence γ_c translates into a small absorption of the probe.

Our system departs from the ideal case described by the expressions of η and α given above in many respects. Unlike the experiments described in Refs. [15,16,19], which were performed with a resonant probe and a weak pump, our probe is tuned to the side of the Doppler profile, in an already almost transparent region. As a result, the position of the gain peak is not tied as closely to a narrow EIT window. It depends on the balance between the losses, which include the Raman absorption dip and the absorption from the Doppler broadened 1-photon transition and the 4WM gain. Factors influencing the peak position are the spread of values for Δ_1 due to the Doppler broadening, the spread of values for Ω_1 and Ω_2 due to the Zeeman degeneracy, the contribution of the usual dispersion of the Doppler broadened vapor to the slow down of the probe, and the only approximate phase matching. In practice, this means that the position of the gain peak varies by up to 20 MHz depending on parameters like the temperature Δ_1 and the probe intensity.

In spite of the added complexity and the difficulty of directly calculating η and Δ_R, we assume that the propagation Eqs. (1) and (2) are still valid over most of the resonance peak, provided that the peak is at $\tilde{\delta} \approx 0$ and that $\gamma_c \ll 2\Delta_R$. Solving them in the limit $\eta \gg 1$ and starting from a probe field $\mathcal{E}_0(\omega)$ and no conjugate field, one finds

$$\mathcal{E}_p(\omega, z) = \mathcal{E}_0(\omega) \exp\left(i\sigma(\omega)\frac{z}{c}\right) \times \left[\cosh\left(\xi(\omega)\frac{z}{c}\right) + i\frac{\sigma(\omega)}{\xi(\omega)} \sinh\left(\xi(\omega)\frac{z}{c}\right)\right] \quad (3)$$

$$\mathcal{E}_c^*(\omega, z) = \mathcal{E}_0(\omega) \exp\left(i\sigma(\omega)\frac{z}{c}\right)\frac{\alpha(\omega)}{i\xi(\omega)} \sinh\left(\xi(\omega)\frac{z}{c}\right) \quad (4)$$

where $\xi(\omega) = \sqrt{\alpha(\omega)^2 - \sigma(\omega)^2}$ and $\sigma(\omega) = \frac{\eta(\omega)}{2} \times (\tilde{\delta} + \omega + i\gamma_c)$. Since $\gamma_c \ll 2\Delta_R$, $\xi(\omega)$ is real. As expected, past an initial linear growth of the conjugate, both fields grow exponentially in distance with $(\xi - \frac{\eta}{2}\gamma_c)/c$ as the linear gain coefficient. In the limit $\tilde{\delta} = 0$, one has $\xi \approx \alpha$.

Equations (3) and (4) show that the fields accumulate a phase across the medium, denoted $\theta(\omega)$. The main contribution to the group delay $\frac{d}{d\omega}\theta(\omega)$ for both fields comes from the first exponential and gives a common delay $\tau = \frac{d}{d\omega}\mathrm{Re}[\sigma(\omega)]z/c|_{\tilde{\delta}=0,\omega=0} = \eta z/2c$. In other words, the probe and the conjugate are slowed down by half the bare slow-down factor η. The probe experiences an extra delay due to the second term in Eq. (3). At large gain, the cosh and sinh functions are equal, and the additional delay is $\Delta\tau = \frac{d}{d\omega}\mathrm{Re}[\sigma(\omega)/\xi(\omega)]/(1 - \mathrm{Im}[\sigma(\omega)/\xi(\omega)])|_{\tilde{\delta}=0,\omega=0} = \eta/[2\xi - \eta\gamma_c] \approx \eta/2\xi$. At low gain, a first order expansion in z gives $\Delta\tau = \eta z/2c$. Thus, the conjugate pulse is created without delay by the probe and travels at a velocity $2v_g$ ($\ll c$). The probe pulse travels initially at a velocity v_g and then locks onto the conjugate by accelerating to $2v_g$ when the delay reaches $\eta/2\xi$ (at a gain close to 2).

We test this interpretation by first scanning the two-photon detuning δ, using 120 ns-long (FWHM) probe pulses and a pump power of 280 mW, which corresponds to a spread of ω of 10 MHz around zero and to $\Omega/2\pi = 420$ MHz. Figure 3 shows the measured gain, the probe delay $\tau + \Delta\tau$, which includes the contribution of all the slowing effects, and the differential delay $\Delta\tau$. The contribution to the probe delay of the usual dispersion effect, which is measured with the pump intensity strongly reduced, is found to be 8 ± 2 ns. From the experimental data and the theoretical expressions of τ and $\Delta\tau$, one can deduce a value of η and ξ for each δ, in the limit of large gain and small γ_c. Inserting these values into Eq. (3) (with $\tilde{\delta} \approx 0$) and adjusting γ_c to 0.5γ (making the linear losses equal to 14% of the peak linear gain), one can reproduce the gain curve with reasonable accuracy. For our parame-

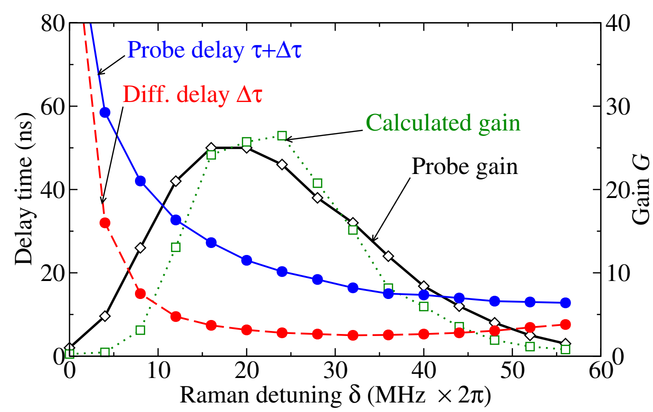

FIG. 3 (color online). Two-photon detuning scan at a temperature of 140 °C, using 120 ns-long pulses. The bare state 2-photon resonance corresponds to $\delta = 0$. The calculated gain, inferred from the delays, assumes linear losses equal to 14% of the peak linear gain.

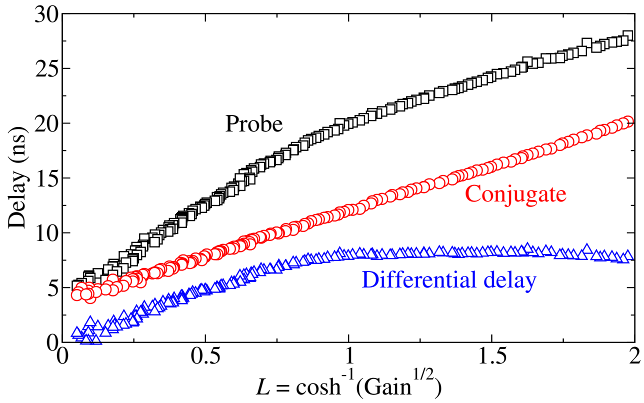

FIG. 4 (color online). Probe delay $\tau + \Delta\tau$, conjugate delay τ, and differential delay $\Delta\tau$ as a function of a pseudo propagation distance.

ters, the EIT resonance is light-shifted to $\delta = 11$ MHz $\times 2\pi \approx 2\gamma$, close to the observed gain maximum ($\tilde{\delta} = 6$ MHz $\times 2\pi \approx \gamma$). It can be shown that in the above calculations, the approximation $\tilde{\delta} \approx 0$ is valid as long as $\tilde{\delta} \ll 2\Delta_R$ (4γ for our beam parameters). For $|\tilde{\delta}|$ larger than a few γ, that is to say in the wings of the gain peak, the approximation is expected to break down.

Next, we directly observe the locking between the probe and the conjugate during the propagation. It is impractical to continuously vary the distance of propagation, and we instead vary the atomic density N via the temperature, which is equivalent. Indeed, σ and ξ are proportional to N through their dependence on η, and changing N is like renormalizing z in the solutions (3) and (4). According to (3), the renormalized propagation length L is related to the probe intensity gain G by $L = \cosh^{-1}(\sqrt{G})$. The detuning δ is set to 15 MHz $\times 2\pi$, near the gain maximum, the pump power is still set to 280 mW, and the measured delays as a function of the renormalized distance for a temperature scan of 50 °C around 120 °C are shown in Fig. 4. The two regimes of propagation are very clear. First, the pulses separate in time, and second, they lock to each other at a fixed delay. A direct evaluation of $2\Delta/\Omega^2 \approx \Delta\tau$ in the ideal case using our beam parameters gives a value of 7 ns, comparable to the one measured near the gain maximum (see Fig. 3). We checked qualitatively that $\Delta\tau$ increases when the pump intensity decreases. It is worth noting that the detail of the low-gain transient regime depends on the initial conditions. For instance, swapping the frequencies of the probe and the conjugate would lead to an initial propagation in which the probe travels at a velocity c while the conjugate travels at a velocity $2v_g$.

Finally, an important feature of the model is that the gain saturates with the pump intensity. For a gain peak location δ and a decoherence γ_c both of the order of γ, ξ saturates when $\Omega \gg 2\sqrt{\Delta_1\gamma} = 140$ MHz $\times 2\pi$. In agreement with this prediction, we observe that G starts saturating at our operating intensity.

To conclude, we have observed the ultraslow propagation of probe and conjugate pulses with matched shapes and group velocities through a rubidium vapor. The study of the coupled propagation gives access to the atomic dynamics through a simple model that reflects a few key concepts. Although the hypothesis of the model does not match precisely the conditions of the experiment, our findings on slow propagation and delay locking of the probe and conjugate pulses are generic to the double-lambda system. The absence of attenuation and distortion suggests the possible existence of a dual-field soliton, such as the one predicted in Ref. [20].

This work is also relevant to recent advances in the generation of nonclassical light. The fact that the same double-lambda scheme successfully generated intensity-difference squeezed twin beams [14] raises two comments. First, as pointed out in Ref. [16], the propagation effects studied in this Letter, which are fundamentally different from those observed in parametric down-conversion [21], have an impact on the squeezing spectrum. Second, the system could be used with gain close to unity to slow light in the quantum regime in order to manipulate photonic qubits, possibly more efficiently than with EIT alone [22].

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