PHYSICAL REVIEW A **67**, 011801(R) (2003)

Light propagation in an atomic medium with steep and sign-reversible dispersion

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(Received 11 October 2002; published 17 January 2003)

We show that ground-state Zeeman coherence prepared by two-photon Raman transitions in alkali atoms results in steep controllable and sign-reversible dispersion. Pulse propagation with small negative as well as positive group velocity of light ($-c/5100$ and $c/41\,000$) in a Cs vapor cell is reported. Energy exchange between copropagating light components through long-lived Zeeman coherence with enhanced absorption or transmission has been observed.

DOI: 10.1103/PhysRevA.67.011801 PACS number(s): 42.50.Gy, 32.80.Qk, 42.25.Bs

The coherent superposition of ground-state atomic sublevels forms the basis for a number of intriguing and sometimes counterintuitive phenomena. One of the most striking manifestations of a long-lived Zeeman coherence prepared by resonant light on a degenerate two-level transition is a dramatic modification of the absorption. Depending on the parameters of the optical transitions, the absorption may be almost cancelled or essentially enhanced. Such absorption variations are described as electromagnetically induced *transparency* (EIT) [1,2] or *absorption* (EIA) [3–5]. In other words, a coherent superposition of Zeeman sublevels can be ''dark'' or ''bright.''

According to the classical theory of dispersion the real and imaginary parts of the refractive index of an atomic medium at low pressure may be written in the form

$$(n-i\kappa)=1+(Ne^2/4\pi\nu_0 m)(\nu_0-\nu+i\gamma/2\pi)^{-1},$$

where N is the atomic density; e and m are the electron charge and mass; ν, ν_0, and γ are the frequency of the optical field, the resonant frequency, and the linewidth, respectively. At resonance ($\nu=\nu_0$), the dispersion $dn/d\nu$ and the absorption coefficient $\alpha=2k_0\kappa$ (k_0 is the vacuum wave number) depend on γ. Thus, the dispersion may be expressed as $dn/d\nu=\alpha(\nu_0)/k_0\gamma$.

A large dispersion is usually associated with a strongly absorbing medium. However, even moderate absorption variations within a very narrow spectral region result in an extremely steep dispersion. The ultimate width of the absorption EIT or EIA resonance is determined by the ground-state relaxation time, which may be rather long, up to 1 s. As a result, very steep refractive index variation accompanies a coherent absorption resonance with subnatural width. However, the slopes of $n(\nu)$ in the two cases are opposite: the EIT medium possesses a normal or positive dispersion ($dn/d\nu>0$), while in the EIA medium the dispersion is anomalous or negative ($dn/d\nu<0$) [6]. Possible ways to obtain steep positive and negative dispersion were discussed theoretically [7].

The propagation of light through a highly dispersive medium has attracted special attention following several experiments on ''*slow light*.'' A pulse of light travels through a medium with a group velocity $V_g=2\pi(d\nu/dk)=c/[n+\nu(dn/d\nu)]$, where c is the speed of light in vacuum. A steep normal dispersion in the medium leads to a dramatic decrease of V_g. Indeed, extremely small values of the group velocity as a result of EIT have been demonstrated both in a cold atomic cloud [8] and in a hot vapor cell [9]. The effect of ''*slow light*'' has played a key role in the so-called ''*light storage*'' [10].

The group velocity V_g may also be negative if the dispersion is anomalous and $|dn/d\nu|>1/\nu$. $V_g<0$ implies that the peak of a pulse exits the medium before it enters in. This counterintuitive situation has been observed. Light pulse propagation with $V_g=-c/23$ has been obtained in a GaP:N crystal [11]. Anomalous dispersion in the spectral region between two closely spaced Raman gain lines in Cs vapor has led to $V_g=-c/315$ [12]. However, the value of the anomalous dispersion of an atomic medium with a long-lived ''bright'' Zeeman coherence is much higher [6]. Recently, pulse propagation in such a medium with a negative group velocity $V_g\simeq-c/3600$ has been reported [13].

The aim of this paper is to study some peculiarities of light propagation in an atomic medium in which the dispersion is highly enhanced by long-lived Zeeman coherence. The experiment was performed with cesium vapor as the resonant medium. The long-lived coherent states in Cs were prepared by the interaction with resonant light within the D_2 line. A simplified scheme of our experimental setup is shown in Fig. 1.

An extended-cavity diode laser was used as the source of

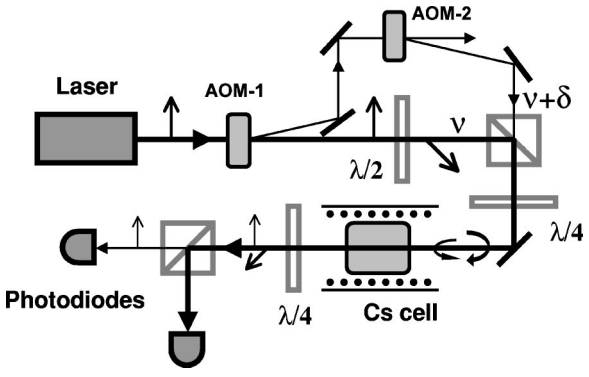

FIG. 1. Experimental setup.

resonant light. Two acousto-optic modulators produced mutually coherent drive and probe beams with tunable frequency offset $\delta=|\nu_1-\nu_2|\approx\Gamma/2\pi$, where $\Gamma/2\pi$ is the natural width of the optical transitions. The two beams were carefully combined into a copropagating bichromatic beam and passed through the Cs cell. They were then separated by a Glan-Thomson prism and detected with fast photodiodes. The intensity of the off-resonant drive component at the probe photodiode was reduced by a factor of 5×10^3. The cross section of the bichromatic beam in the cell was about 1 cm, while the maximum intensities of the drive and probe components were 4 and 0.8 mW/cm^2, respectively. The 2-cm-long Cs cell without buffer gas was placed in a μ-metal shield to reduce the ambient magnetic field to milligauss level. A controllable longitudinal magnetic field was produced by a solenoid mounted inside the magnetic shield. The density of Cs was varied within the range $N=(0.7-8)\times10^{11}$ cm^{-3} by changing the cell temperature. Gaussian-shaped as well as sharp-edge square probe pulses were applied to interrogate the light propagation.

Opposite circular polarizations of the probe and drive components were set with polarizers and quarter wave plates. At $\delta=0$ the bichromatic beam may produce a coherence between ground-state magnetic sublevels belonging to the same hyperfine level as a result of Raman transitions involving one photon from the drive field and a second photon from the probe. Each component alone is not able to produce coherence between the magnetic sublevels in the ground state.

Since the Doppler broadening exceeds the hyperfine splitting of the $6P_{3/2}$ state, the bichromatic light is resonant with atoms within three velocity groups. Nonabsorbing ''dark'' superpositions of Zeeman sublevels in the ground state may be prepared, according to Ref. [14], through the $6S_{1/2}(F_g=3)$-$6P_{3/2}(F_e=2,3)$ and $6S_{1/2}(F_g=4)$-$6P_{3/2}(F_e=3,4)$ transitions, while a more absorbing ''bright'' superposition can be induced through the $6S_{1/2}(F_g=4)$-$6P_{3/2}(F_e=5)$ transition [15]. We assumed that the dispersion is proportional to absorption variations and inversely proportional to the width of the coherent resonance: $dn/d\nu\sim\Delta\alpha/\Delta\nu$. To find a spectral region where coherent effects are more pronounced and to optimize experimental parameters, the double frequency sweeping technique was used (Fig. 2).

The maximum contrast of EIA resonances on the high-frequency slope was close to 60%. The typical width of the high-contrast coherent resonance $\Delta\nu$ measured at fixed optical frequency was about 250 kHz.

A medium with a ground-state Zeeman coherence induced by a constant drive and a pulse probe radiation affects the shape of a pulse during propagation, as shown in Fig. 3. In the spectral region, where EIT takes place, the transmitted intensity grows with time due to pumping into the nonabsorbing state (curve c). Thus, the centroid of the pulse is delayed with respect to the off-resonance reference. In the EIA spectral region the situation is reversed (curve b). At the beginning of the pulse the transmitted intensity is higher. This causes a shift of the centroid toward the leading edge of the pulse.

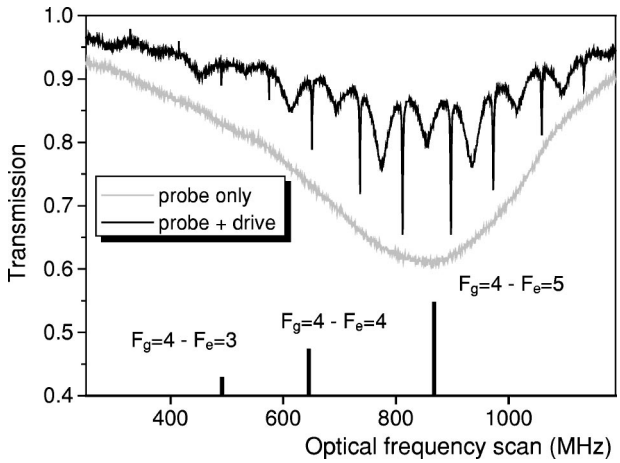

FIG. 2. Transmitted probe intensity with and without the drive component versus optical frequency on the $6S_{1/2}(F_g=4)$-$6P_{3/2}$ transitions. The offset δ is swept 15 times faster (at 750 Hz) in a few megahertz range. The probe absorption is saturated in the presence of the drive. However, during the slow scan when the condition for Raman transitions is fulfilled ($\delta=0$), the absorption of the probe is enhanced due to a ''bright'' state, producing sharp spikes in the transmission.

Some delay or advance is expected because the pulses move in a medium with steep dispersion. Indeed, for a pulse traveling with V_g the time delay with respect to the off-resonant pulse is $\Delta T=(L/c-L/V_g)$, where L is the length of the medium. The values of ΔT can be easily measured even for $L\simeq1$ cm if $|V_g|\ll c$.

To avoid strong distortion of the pulse, the spectral width should be within the narrow region of constant dispersion. In our case this means that the smooth probe pulse should be longer than $\Delta t>1/2\pi\Delta\nu\approx0.7$ μs.

The propagation of a Gaussian-shaped probe pulse with low V_g through Cs vapor in a ''dark'' Zeeman state is shown in Fig. 4. The pulses of resonant light are delayed relative to the reference. The position of the pulse maximum is found by a Gaussian fit to the experimental data. The time delay is

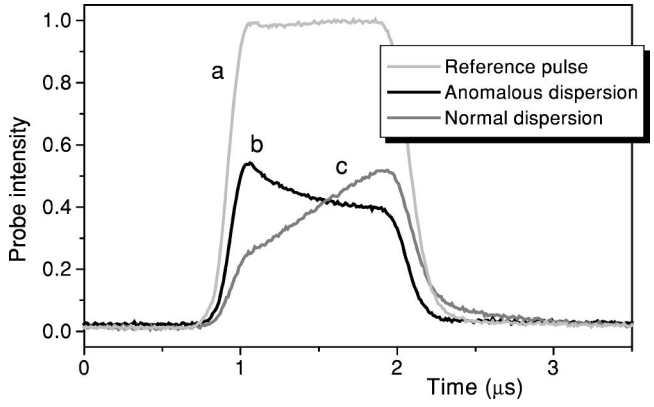

FIG. 3. Distortion of sharp-edge probe pulses in Cs vapor. Curve a represents the off-resonance probe pulse propagating with $V_g\approx c$. Curves b and c correspond to the optical frequency tuned to the $6S_{1/2}(F_g=4)$-$6P_{3/2}(F_e=5)$ and $6S_{1/2}(F_g=3)$-$6P_{3/2}(F_e=2)$ transitions, respectively.

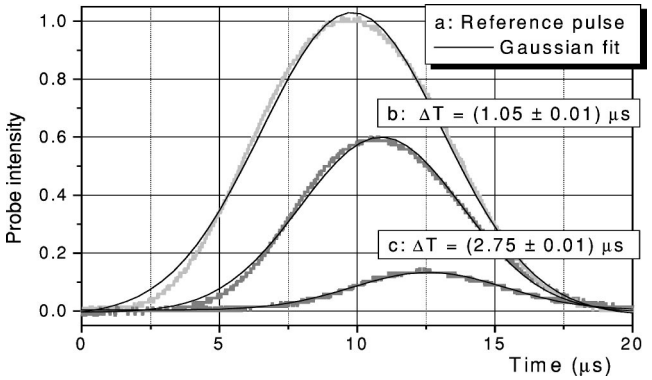

FIG. 4. Pulse delay measurements in Cs vapor with steep normal dispersion. Off-resonance pulse (*a*) is used as a reference. Curves *b* and *c* correspond to the optical frequency tuned to the $6S_{1/2}(F_g=3)$-$6P_{3/2}(F_e=2)$ transition at atomic densities of 4×10^{11} cm^{-3} and 7×10^{11} cm^{-3}, respectively.

larger for higher Cs density because $dn/d\nu\sim N$. The delay $\Delta T\approx$ 2.75 μs for the 2-cm-long cell corresponds to $V_g\approx c/41\,000\approx7.3$ km/s.

We emphasize an interesting feature of a Zeeman coherence. The sign of the steep dispersion may be easily changed by tuning the laser frequency to another optical transition. In the EIA spectral region Cs vapor possesses steep anomalous dispersion due to a ''bright'' coherence. As a result the top of the probe pulse is advanced in time compared to the reference (Fig. 5). The advance is $\Delta T=(0.10\pm0.01)$ μs for the 35% absorption, while for the higher absorption (73%) the shift in time is clearly visible and equals $\Delta T=(0.34\pm0.01)$ μs. The ratio of the time advance to the pulse duration is about 9% and is three times larger than in Ref. [12]. At the same time the distortion of the pulse shape is not strong. The maximum anomalous dispersion of the medium is estimated to be $dn/d\nu\approx(c/V_g\nu)\approx-1.02\times10^{-11}$ Hz^{-1}. Reduction of the width of the coherent resonance by increasing the lifetime of ground-state Zeeman coherence is a natural way for obtaining higher anomalous dispersion.

Thus, the probe pulse propagates through the coherently

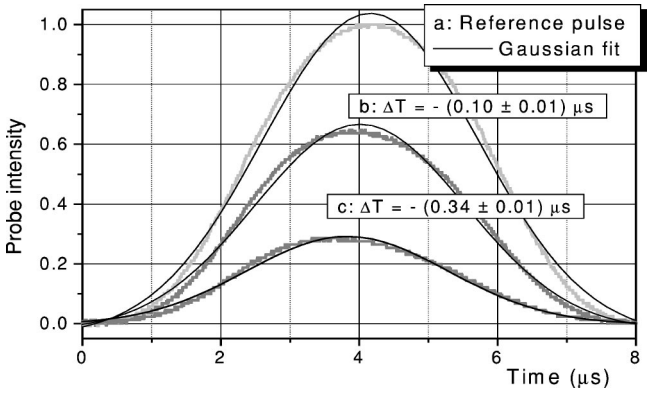

FIG. 5. Negative pulse delay (advancement) in Cs vapor with steep anomalous dispersion. Off-resonance pulse (*a*) is a reference. Curves *b* and *c* correspond to the optical frequency tuned to the $6S_{1/2}(F_g=4)$-$6P_{3/2}(F_e=5)$ transition at atomic densities of 1.5×10^{11} cm^{-3} and 7×10^{11} cm^{-3}, respectively.

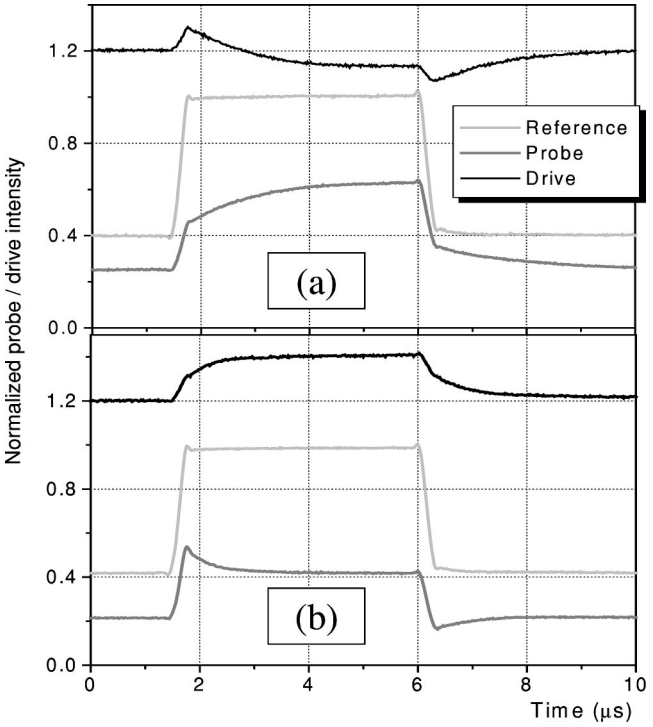

FIG. 6. Transient effects in the transmitted probe and drive components. The intensities are normalized to the off-resonant probe intensity. (a) and (b) correspond to the optical frequency tuned to the $6S_{1/2}(F_g=3)$-$6P_{3/2}(F_e=2)$ and $6S_{1/2}(F_g=4)$-$6P_{3/2}(F_e=5)$ transitions, respectively.

prepared Cs vapor faster than the reference pulse. In spite of the counterintuitive nature of light propagation with a negative group velocity, there is no violation of the principle of causality. This phenomenon can be described within classical electrodynamics as pointed out in Ref. [16]. Light propagation with $V_g<0$ is a result of interference within a wave packet between spectral components moving with phase velocity $V_{ph}=c/n$.

An interesting energy exchange between the probe and drive components was discovered in a coherently prepared Cs vapor, as shown in Fig. 6. It is worth noting that the coherently prepared medium possesses enhanced Kerr nonlinear susceptibility both in ''dark'' [17] and in ''bright'' [18] states at low light intensity. To investigate this effect we consider the sharp-edge probe pulse propagation when it is added to some constant level in the presence of a constant drive component. First we consider the EIT case [Fig. 6(a)]. The constant part of the bichromatic radiation partially pumps atoms into a coherent ''dark'' superposition mainly through the cycling transitions $F_g=3$-$F_e=2$. The incoherent part of the population mostly accumulates in the extreme magnetic sublevels ($m=+2,+3$) due to σ^+ transitions driven by the more intense drive component. The sharp-edge probe pulse modifies this steady-state distribution. The atoms in $m=+2,+3$ are excited to the $6P_{3/2}$ state by the leading edge of the pulse. They return to the ground state after emitting stimulated photons into the drive, thus the transmitted intensity of the drive increases. The absorption at the leading edge of the pulse is rather strong, but decreases due to pump-

ing into the modified dark state, corresponding to a new ratio between the drive and probe components. This transient process leads to a slow increase of the probe intensity. Steady state can be reached within 0.1 to 1 μs, depending upon the light intensity, in agreement with the expected value of the ground-state relaxation time.

A rather long tail appears in the transmitted probe intensity after switching off the pulse. A fraction of the drive intensity leaves the cell as the probe. This can be explained in terms of imprinting of information about the pulse into the Zeeman coherence. The ground-state Zeeman coherence can be considered as a phase grating and this intensity exchange is evidence of transient behavior in the stimulated scattering from the modified phase grating. Thus, more efficient nonlinear mixing (or stimulated scattering) is responsible for some reduction of the drive intensity. Almost perfect energy transfer from the probe to the drive and then back to the probe takes place.

We note that this process occurs not only for the ''dark'' coherent superposition, but also for the ''bright'' state. However, in the latter case the situation is in some sense reversed. The probe pulse transmission through the EIA medium is shown in Fig. 6(b). The front edge of the transmitted pulse is enhanced due to relatively low absorption during the pulse rise time. After switching off the pulse, one can see a positive tail in the drive intensity. The process of stimulated scattering for the ''bright'' superposition has the opposite effect, that is, the probe intensity is transferred into the drive. The relaxation time (about 1 μs) for this process also depends on the light intensity.

Here we present only the initial observations of the energy exchange between light components, which accompany a long-lived Zeeman coherence. We are currently investigating further aspects of this process, focusing our attention on the low intensity limit.

In conclusion, we have shown that the dispersive properties of an atomic medium can be dramatically modified as a result of a long-lived Zeeman coherence. Light pulse propagation with small negative as well as positive group velocity ($V_g \approx -c/5100$ and $V_g \approx c/41\,000$) in Cs vapor has been achieved. The unprecedentedly high anomalous dispersion is due to EIA. Even higher dispersion can be obtained in an atomic medium with a longer ground-state lifetime, e.g., using a sample of ultracold atoms.

A reversible exchange between the drive and probe components through Zeeman coherence in the atomic vapor has been observed not only for a ''dark'' but also for a ''bright'' coherence. This exchange has the same origin as the optical storage effect [10].

Atomic media with unique dispersive and nonlinear properties may be useful, for example, for acceleration of data processing and further investigations of ground-state Zeeman coherence are required.

The authors gratefully acknowledge helpful discussions with A.S. Zibrov, V.I. Yudin, A.V. Taichenachev, A.B. Matsko, and R.J. McLean.

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