Nonlinear Magneto-optics and Reduced Group Velocity of Light in Atomic Vapor with Slow Ground State Relaxation

D. Budker,^{1,2,*} D. F. Kimball,¹ S. M. Rochester,¹ and V. V. Yashchuk^{1,3}

¹Department of Physics, University of California, Berkeley, California 94720-7300

²Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720

³B. P. Konstantinov Petersburg Nuclear Physics Institute, Gatchina, Russia 188350

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The dynamics of resonant light propagation in rubidium vapor in a cell with antirelaxation wall coating are investigated. We change the polarization of the input light and measure the time dependence of the polarization after the cell. The observed dynamics are shown to be analogous to those in electromagnetically induced transparency. Spectral dependence of light pulse delays is found to be similar to that of nonlinear magneto-optic rotation. Delays up to ≈ 13 ms are observed, corresponding to a 8 m/s group velocity.

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Long-lived atomic coherences lead to narrow resonances that can be observed either in the spectral domain using pump-probe light beam arrangements, or in level-crossingtype experiments involving a single light beam. Longlived ground state coherences can be obtained in alkali metal vapor using cells with antirelaxation paraffin coating [1,2] in which the coherences survive tens of thousands of collisions with the walls of the cell. Previously, we used such a cell with ⁸⁵Rb vapor at room temperature to obtain resonances in the nonlinear magneto-optic effect (NMOE) corresponding to a relaxation rate $\gamma_{rel} \approx 2\pi \cdot 1$ Hz [3]. The long-lived coherences are also important for electromagnetically induced transparency (EIT) [4]. Recently, attention has been drawn to slow group velocities of light obtained due to enhanced dispersion in the conditions of EIT [5–9]. For example, in Ref. [8], light propagation through an optically dense sample of sodium atoms above and below the Bose-Einstein condensation threshold was investigated and group velocities as low as $v_g = 17 \text{ m/s}$ and group delays $\tau_d \approx 7 \ \mu s$ were observed.

Here, we investigate resonant light propagation through rubidium vapor contained in a cell with antirelaxation wall coating and elucidate the relation between slow group velocities and NMOE. The maximum obtainable light pulse delay scales as the inverse of the relaxation rate [see, e.g., Ref. [5], and Eq. (4), below]; thus large delays $\tau_d \approx$ 13 ms are achieved in our case. The optical thickness of the atomic vapor that we use is $\approx 1-2$ absorption lengths, which allows investigation of pulse propagation and reshaping phenomena both in conditions of EIT and electromagnetically induced opacity (EIO). In addition, it is demonstrated that EIT and EIO, and consequently the group velocity of light, can be controlled using weak magnetic fields ($B \ge 1 \ \mu G$) corresponding to the Larmor frequency $\Omega_L = g_F \mu B \ge \gamma_{rel}$. Here g_F is the ground state Lande factor, and μ is the Bohr magneton.

The apparatus used for the present measurements (Fig. 1) is largely the same as for our earlier work [3,10].

The vapor cell (diameter 10 cm), contained in a multilayer magnetic shield, and a glass Faraday rotator are positioned between crossed polarizer and analyzer. Diode lasers are used to produce cw light at 795 nm (D1 transition) or 780 nm (D2 transition). The linearly polarized light induces alignment of the atomic vapor. If a current pulse is applied to the Faraday rotator coil, causing light polarization before the cell to rotate, the polarization state after the cell exhibits a transient response. The transmitted probe light intensity is measured by a photodetector after the analyzer and its time dependence is investigated.

For $F \to F'$ transitions with F' = F or F - 1, measuring the time dependence of the output polarization is equivalent to performing an EIT experiment (see Fig. 2). (For the $F \to F + 1$ transitions, optical pumping leads to increased absorption [11,12], corresponding to EIO.) In traditional EIT, state 1 is populated and probe light (with frequency ω_n) is tuned to a resonance with the $1 \to 2$



FIG. 1. Experimental setup. For pulse delay measurements, a current pulse is applied to the Faraday rotator coil, causing light polarization before the cell to rotate. The transmitted probe light intensity is measured by a photodetector (PD). In order to measure the spectral dependence of the delay, we apply sinusoidal current to the Faraday rotator, and detect the phase shift of the transmitted intensity using a lock-in amplifier. Faraday rotation and the dark and bright transmission resonances are measured as described in Refs. [3,10].



FIG. 2. (a) Schematic diagram of energy levels illustrating the relation between the present experimental arrangement and EIT. (b) Conceptual illustration of the imaginary part of the refractive index for the probe light. Dot-dashed line: in the absence of the coupling light— n_p ; dashed line: in the presence of coupling light for probe polarization parallel to the coupling light polarization— n_p^{\parallel} ; solid line: in the presence of coupling light for probe polarization orthogonal to the coupling light polarization— n_p^{\perp} . Here the width of the nonlinear feature is the ground state relaxation rate γ_{rel} , and for simplicity we ignore the hole burning effects (see, e.g., Ref. [3]).

transition. Coupling light (ω_c) is applied resonant with the $3 \rightarrow 2$ transition; states 2 and 3 are both unpopulated in the absence of light.

In our case, states 1 and 3 are superpositions, known as dark and bright states, of the (degenerate) ground state Zeeman sublevels M_F [13]. They are equally populated in the absence of light. When the coupling light is applied, the populations of the ground state sublevels redistribute due to optical pumping: the bright state depopulates, and the vapor becomes dichroic. In general, for $\omega_p \neq \omega_c$, the index of refraction n_p^{\perp} for probe light of orthogonal polarization to that of the coupling light is different from the index of refraction n_p^{\parallel} for probe light of the same polarization (Fig. 2b). However, for $|\omega_p - \omega_c| \ll$ γ_{rel} , we have $n_p^{\perp} \approx n_p^{\parallel} \approx n_c$, where n_c is the refractive index for the coupling light. This is because when the pump and coupling frequencies coincide, the situation is equivalent to illuminating the medium only with the coupling light with slightly rotated polarization direction. In this experiment, the probe beam is of the same central optical frequency as the coupling light and is produced with the Faraday rotator by slightly rotating the input polarization. As long as the polarization rotation induced by the rotator is small (as is always the case here), the coupling light can be considered constant, while the probe intensity goes as $I_p(t) \propto i^2(t)$, where i(t) is the timedependent current applied to the Faraday rotator.

To analyze the probe light propagation dynamics in our experimental conditions, we consider a probe field Emodulated at the frequency $\Omega: E = E_0 \exp[i\omega_c(n_p z/c - t)]\sin(\Omega t)$, where z is the propagation direction. A field of this form corresponds to light at the two frequencies $\omega_p^{\pm} = \omega_c \pm \Omega$. Under certain simplifying assumptions (sufficiently low light power [3], few absorption lengths, zero transverse magnetic fields), the contribution of the narrow feature due to the coupling light (Fig. 2b) to the effective complex refractive index is

$$n_p = A\kappa \left(\frac{\gamma_{\rm rel}c}{\omega_c l_0}\right) \frac{1}{\omega_p - \omega_c - i\gamma_{\rm rel}},\qquad(1)$$

where A is a dimensionless coefficient incorporating the details of the level structure of a particular atomic system, $\kappa = d^2 E_c^2 / (\gamma_0 \gamma_{rel}) < 1$ is the optical pumping saturation parameter (d is the transition dipole moment, E_c is the coupling light electric field, γ_0 is the natural width of the transition), l_0 is the unsaturated absorption length of the vapor, and we neglect terms which vary slowly with respect to the probe frequency.

Substituting (1) into the expression for the electric field, one finds that the light intensity modulation acquires a phase shift

$$\phi = A\kappa \frac{2\gamma_{\rm rel}\Omega l/l_0}{(\Omega^2 + \gamma_{\rm rel}^2)},\tag{2}$$

where l is the length of the cell. Using a similar approach to analyze nonlinear Faraday rotation, one finds that the rotation at a given magnetic field is [14,15]

$$\Phi = A\kappa \frac{\gamma_{\rm rel} \Omega_L l/l_0}{[(2\Omega_L)^2 + \gamma_{\rm rel}^2]}.$$
(3)

From Eqs. (2) and (3), one sees that within the approximations used in this derivation, the ratio of phase shift and rotation does not depend on laser frequency, and thus the two effects are expected to have the same spectral shape (determined by the spectral dependence of $A\kappa/l_0$).

Probe light pulses long enough that all Fourier components satisfy $|\omega_p - \omega_c| = \Omega \ll \gamma_{rel}$ propagate without change of shape. Thus the delay for each frequency component of the light intensity modulation is given by $\phi/(2\Omega)$, so that the pulse delay is

$$\tau_d = A \kappa \frac{l/l_0}{\gamma_{\rm rel}} \,. \tag{4}$$

The group velocity is given by $v_g = l/\tau_d$.

The magnetic field dependence of slow group velocities (measured as described below), nonlinear Faraday rotation, and the dark resonance is illustrated in Fig. 3. Application of a magnetic field causes mixing between the dark and bright states via Larmor precession. Thus a field corresponding to a Larmor frequency comparable to the relaxation rate (i.e., $\geq 1 \ \mu G$ in our case; see Fig. 3) controls all these effects.



FIG. 3. Comparison of transmission (the dark resonance) (a), nonlinear optical rotation (b), and phase shift (c). Maximum phase shift shown in (c) corresponds to a group delay ≈ 13 ms and group velocity ≈ 8 m/s. The laser is tuned to the center of the lower-frequency peak of the $F = 3 \rightarrow F'$ transition group of the D1 line (see Figs. 5b and 5c). Input light intensity: 180 μ W/cm²; beam diameter: 3.5 mm. The width of the resonant features corresponds to a relaxation rate $\gamma_{rel} = 2\pi \times 1.3$ Hz. The line shape irregularities in (c) are believed to be due to average residual magnetic fields (~0.1 μ G).

An example of a pulse delay measurement is shown in Fig. 4. We apply Gaussian current pulses to the Faraday rotator (width: 200 ms; maximum polarization rotation: 25 mrad) and observe delayed pulses on the photodetector with $B_z \approx 0$. No delay is observed when a magnetic field of 10 μ G is applied or when the laser is tuned off-resonance. The value of the constant A, which is determined by the complex multilevel structure of the Rb system, turns out to be $\ll 1$, so that $\tau_d \ll 1/\gamma_{rel}$. To minimize pulse reshaping, the pulse duration has to be long enough so that its spectral width is less than $\gamma_{rel} =$ $2\pi \times 1.3$ Hz; thus the pulse delay constitutes a small fraction of the pulse width. Note that if the vapor density were increased by heating the cell, both the number of absorption lengths and the relaxation rate, dominated by spin exchange [3], would increase proportionally to atomic density. As long as the assumptions employed in the derivation of Eqs. (1)–(4) are satisfied, the overall delay would not be expected to change significantly. However, increasing the relaxation rate would increase the "EIT bandwidth" [5], allowing the use of shorter pulses, so that



FIG. 4. An example of pulse delay measurement on the D1 line. The laser is tuned to the same frequency as in Fig. 3. Dotted line: time-dependent signal recorded by the photodetector with $B_z \approx 0$; solid line: same with $B_z = 10 \ \mu G$ (corrected for the time-independent Faraday rotation produced by the magnetic field). The timing of the latter curve is within experimental uncertainty identical with that recorded for off-resonant laser light. The measured delay in this particular case is $\tau_d = 2.5(1)$ ms. Similar observations were also made on the D2 line. The inset shows an enlarged view of the highlighted area of the plot.

delayed and nondelayed pulses could be better resolved at the output.

In order to measure the spectral dependence of the delay, we apply sinusoidal current $[i(t) = i_0 \sin \Omega t; \Omega =$ $2\pi \times 0.5$ Hz] to the Faraday rotator (Fig. 1), and detect the phase shift of the transmitted intensity. The result of a phase shift spectrum measurement for the D1 line is shown in Fig. 5 which also shows for comparison the nonlinear Faraday rotation spectrum and the transmission spectrum measured under similar experimental conditions. The Faraday rotation spectrum (Fig. 5b) is considerably different from the transmission spectrum (Fig. 5a) [16]. For example, the rotation spectrum is split into two peaks with sub-Doppler widths for the D1 $F = 3 \rightarrow F'$ transition group. The phase shift spectrum (Fig. 5c) qualitatively reproduces this behavior. Moreover, the ratios of the heights of corresponding peaks of optical rotation and delay agree with the prediction of Eqs. (2) and (3) within \sim 30%. We believe that the residual differences between the delay and the nonlinear Faraday rotation spectra may be attributable to effects neglected in the above analysis, including those due to residual magnetic fields and light broadening. It is interesting to note that we observe negative group delays (most pronounced near the center of the $D1F = 3 \rightarrow F'$ transition group). In linear optics, negative group velocities and group delays are well known to occur due to frequency dependent absorption (pulse reshaping) in regions of anomalous dispersion (see, e.g., Ref. [17]).

In conclusion, we have demonstrated the relation between reduced light group velocity and nonlinear magneto-optics. The ultranarrow resonances obtained in atomic vapor with slow ground state alignment relaxation lead to the observed group delays $\tau_d \approx 13$ ms and



FIG. 5. Comparison of the transmission spectrum (a), nonlinear optical rotation with $B_z = 1.3 \ \mu$ G (b), and phase shift (c) on the D1 line. The sign of the optical rotation for the $F = 3 \rightarrow F'$ and $F = 2 \rightarrow F'$ transitions is opposite due to the difference in the g factors. Negative phase shifts, most pronounced near the center of the $F = 3 \rightarrow F'$ transition group, correspond to negative group velocity.

velocities $v_g \approx 8$ m/s. Their magnitude and spectral dependence are in agreement with predictions based on the observed NMOE.

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*e-mail: budker@socrates.Berkeley.edu

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