Polarization-ellipse rotation by induced gyrotropy in atomic vapors

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We study theoretically and experimentally a new mechanism for the rotation of the polarization ellipse of a single laser beam propagating through an atomic vapor with a frequency tuned near an atomic resonance. The results of a theoretical treatment for the case of a $J = 1/2$ to $J = 1/2$ atomic transition show that a rotation of the polarization ellipse of the laser beam will occur as a result of ground-state optical pumping and that the angle of rotation is independent of the laser intensity over a broad range of laser intensities. The predictions of this theoretical model are tested experimentally through the use of potassium vapor and are found to agree with the experimental data.

One of the simplest nonlinear optical interactions is the rotation of the polarization ellipse of a laser beam as it propagates through a nonlinear medium.[1] The rotation arises from the tensor nature of the third-order nonlinear susceptibility, and the magnitude of the rotation increases with increasing laser intensity. In this Letter we describe a novel nonlinear-optical mechanism that also leads to the rotation of the polarization ellipse of a laser beam propagating through an atomic vapor. The ellipse rotation described here results from the nonlinearity induced by ground-state optical pumping, and the magnitude of the rotation is found to be independent of the laser intensity over a broad range of intensities.

The optical nonlinearity associated with ground-state optical pumping has been shown to lead to effects such as phase conjugation by four-wave mixing[2-5] and optical multistability.[6,7] Saikan[8] has calculated the tensor components of the third-order nonlinear susceptibility for transitions between atomic resonance lines of arbitrary total angular momentum J, and, for certain cases, the polarization ellipse rotation described by Maker and Terhune[1] is expected to occur. Baer and Abella[9,10] have observed an intensity-independent rotation of the direction of polarization of photon echoes.

Here we study the propagation of an elliptically polarized laser beam through an atomic vapor for the specific case in which the laser frequency is tuned near a $J = 1/2$ to $J = 1/2$ atomic transition. For laser intensities much greater than the saturation intensity associated with the optical pumping of the electronic ground state, the nonlinear response of the atomic vapor cannot be described by a $\chi^{(3)}$ nonlinearity. In fact, we find that the response of the atomic vapor leads to a rotation of the polarization ellipse that is independent of the laser intensity for a broad range of intensities. The rotation angle depends only on the ellipticity of the laser beam, the absorption path length, and the detuning of the laser beam from resonance. The physical origin of the rotation of the polarization ellipse is described as follows: An elliptically polarized laser beam contains unequal amounts of the two circular components, and this difference produces an imbalance in the ground-state populations. Consequently, each circular component of the laser beam experiences a different refractive index, and this effect causes the polarization ellipse of the laser beam to rotate. We performed experiments using potassium vapor and observed rotations as large as 40°. All our results are in good agreement with the theoretical predictions.

We assume that the complex electric-field amplitude $\mathbf{E}$ inside the atomic vapor can be decomposed in a circular polarization basis such that $\mathbf{E} = (E_+\hat{\epsilon}_+ + E_-\hat{\epsilon}_-)$, where $\hat{\epsilon}_\pm = (\hat{x} \pm i\hat{y})/\sqrt{2}$ are the unit vectors for left-hand and right-hand circular polarizations, respectively. The density-matrix equations that describe the time evolution of the atomic polarizations $\sigma_\pm$ and the atomic populations $n_{1\pm}$ and $n_{2\pm}$ of the $J = 1/2$ to $J = 1/2$ atomic transition are then given by[11]

$$\frac{d\sigma_\pm}{dt} = \left(-\frac{1}{T_2} + i\Delta\right)\sigma_\pm \mp ig(n_{2\pm} - n_{1\mp})E_\pm, \quad \text{(1a)}$$

$$\frac{dn_{1\pm}}{dt} = \frac{1}{3T_1}n_{2\pm} + \frac{2}{3T_1}n_{2\mp} \mp \frac{1}{2T_g}(n_{1+} - n_{1-}) \mp i(g^*E_\mp{}^*\sigma_\mp - \text{c.c.}), \quad \text{(1b)}$$

$$\frac{dn_{2\pm}}{dt} = -\frac{1}{T_1}n_{2\pm} \mp i(g^*E_\pm{}^*\sigma_\pm - \text{c.c.}), \quad \text{(1c)}$$

where T_1 is the population lifetime of the electronic transition, T_2 is the dipole-dephasing time, T_g is the ground-state recovery time, $\Delta = (\omega - \omega_0)$ is the detuning of the laser frequency from that of the electronic transition, and the atomic coupling constant g is proportional to the dipole moment μ by $g = \sqrt{2/3}\,\mu/\hbar$.

We solve Eqs. (1) for σ_+ and σ_- under steady-state conditions and under the assumption that the inten-

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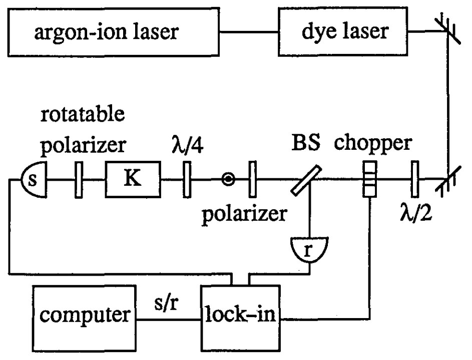

Fig. 1. Experimental setup used to measure polarization-ellipse rotation in potassium vapor. BS, beam splitter; s, signal detector; r, reference detector.

sities $I_\pm = |E_\pm|^2$ of the circularly polarized components are much less than the electronic saturation intensity $I_s^e = (1 + \delta^2)/4|g|^2T_1T_2$, where $\delta = \Delta T_2$, but much greater than the optical-pumping saturation intensity $I_s^g = I_s^e T_1/T_g$. In practice, this inequality can be satisfied for a broad range of intensities because T_g is typically several orders of magnitude greater than T_1 for atoms having an S ground state.[12] The complex amplitude $\mathbf{P}$ of the macroscopic polarization is related to the atomic polarizations through $\mathbf{P} = \sqrt{2/3}N\mu(\sigma_+\hat{\epsilon}_+ - \sigma_-\hat{\epsilon}_-)$, where N is the atomic number density. The complex polarization amplitude, which drives the complex electric field in Maxwell's equations, can be written in the form $\mathbf{P} = \chi_L(\mathbf{E}\cdot\mathbf{E})\mathbf{E}^*/(\mathbf{E}\cdot\mathbf{E}^*)$, where $\chi_L = -N|\mu|^2T_2/3\hbar(i + \delta)$ is the linear susceptibility.

For the case in which a single monochromatic laser beam propagates through the atomic vapor and the detuning is much greater than the atomic linewidth ($\delta \gg 1$), the equations for the electric-field amplitudes $E_\pm$ show that propagation through the vapor introduces only a change in the phase of each of the two electric-field components. Rotation of the polarization ellipse occurs when the two amplitude components E_+ and E_- undergo different phase changes. The electric-field amplitudes after propagating though a distance L of atomic vapor can be expressed as $E_\pm(L) = E_\pm(0)\exp[i(\phi \pm \theta)]$, where

$$\theta = \frac{\alpha_0 L}{2\delta}\frac{I_+(0) - I_-(0)}{I_+(0) + I_-(0)} \tag{2}$$

gives the rotation angle of the polarization ellipse and where $\phi = -\alpha_0 L/2\delta$ represents an inconsequential overall phase shift, and $\alpha_0 = 4\pi k N|\mu|^2T_2/\hbar$ is the line-center absorption coefficient. Note that the rotation angle θ is independent of the laser intensity, and the direction of rotation depends on both the sign of the detuning and on the ellipticity of the incident beam under our assumed conditions.

Experimental measurements of the rotation of the polarization ellipse were performed by using the $4^2S_{1/2} \leftrightarrow 4^2P_{1/2}$ transition of atomic potassium vapor and a heat pipe with an interaction length L of ~5 cm. A helium buffer gas at a pressure of 75 Torr was used to collisionally broaden the transition to a width of 2 GHz, which is greater than that (440 MHz) of the ground-state hyperfine splitting. The temperature of the oil used to heat the outer walls of the interaction region of the heat pipe was fixed at a value of 100°C. From the known values of the dipole moment and the population lifetime,[13] the saturation intensity of the electronic transition is found to be 3.5 W/cm^2. By using modulation spectroscopy, we measured the ground-state recovery time T_g and found it to be equal to 1.4×10^{-5} s. From this measurement, the optical-pumping saturation intensity is found to be 6.3 mW/cm^2.

Figure 1 shows the experimental setup. A quarter-wave plate is used to create a known ellipticity on the input beam. The polarization-ellipse rotation angle θ is then related to the angle β between the c axis and the direction of the (linear) polarization of the field incident upon the quarter-wave plate by the expression $\theta = (-\alpha_0 L/2\delta)\sin(2\beta)$.

We assume in our theoretical analysis that the normalized detuning δ is much greater than unity. If this were not so, the two circular components of the electric field would experience different amounts of absorption, causing the ellipticity of the laser beam to change as it propagates through the atomic vapor. Therefore, in our experiments, we monitor the ratio of the major to minor axis of the polarization ellipse to ensure that the ellipticity of the laser beam does not change. We also ensure that the intensity of each of the circular components of the laser beam is much greater than the optical-pumping saturation intensity. At fixed laser intensity, this condition places restrictions on the ellipticity of the laser beam that can be used. We

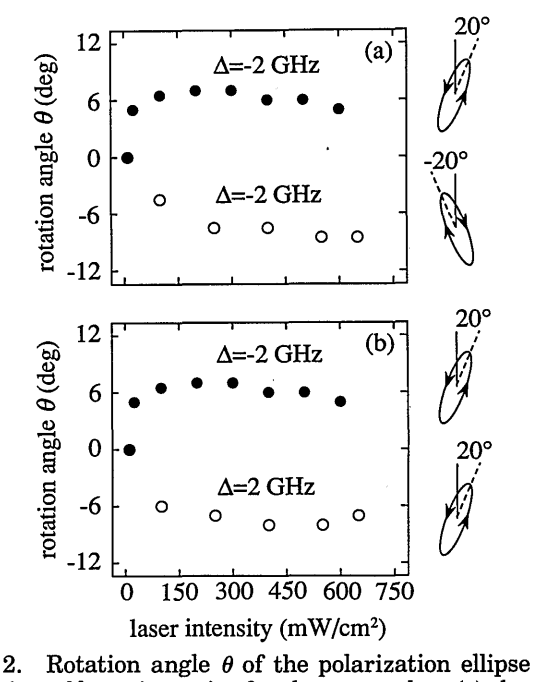

Fig. 2. Rotation angle θ of the polarization ellipse as a function of laser intensity for the cases when (a) the laser detuning is held fixed and the laser ellipticity takes on opposite values and (b) the laser ellipticity is held fixed and the laser detuning takes on opposite values. The direction of rotation changes when an opposite ellipticity is used or when the sign of the detuning changes. Notice that the magnitude of the rotation angle is insensitive to the laser intensity for laser intensities between 50 and 600 mW/cm^2.

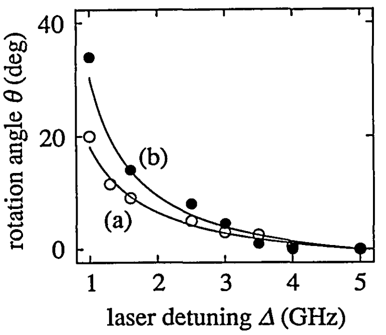

Fig. 3. Rotation angle θ of the polarization ellipse as a function of laser detuning for an input laser intensity of 400 mW/cm^2 and an input laser ellipticity characterized by $\beta = -20°$ [curve (a)] and $\beta = -30°$ [curve (b)]. In both cases, the solid curve shows a best-fit hyperbola through the data.

estimate that our experimental uncertainty in measuring the polarization-ellipse rotation angle θ is $\sim 1°$.

Our experimental results are shown in Figs. 2 and 3. Figure 2 shows the rotation angle θ as a function of laser intensity for two cases: fixed laser detuning but two ellipticities of opposite sign [Fig. 2(a)] and a fixed ellipticity but two detunings of opposite sign [Fig. 2(b)]. In all cases the rotation angle remains essentially constant for laser intensities between 50 and 600 mW/cm^2, more than a factor of 10 change in laser intensity. Note that the direction (but not the magnitude) of the angular rotation changes when an opposite ellipticity is used or when the sign of the detuning changes, consistent with Eq. (2).

Figure 3 is a plot of angular rotation as a function of laser detuning for two different input laser ellipticities ($\beta = -20°$ and $-30°$). In both cases the solid curve shows a best-fit hyperbola through the data, which is consistent with the Δ^{-1} dependence predicted in our theory.

A calculation of the magnitude of the angular rotation by using our theory requires an accurate value for the absorption path length $\alpha_0 L$. The value of the absorption path length is difficult to calculate, however, because the potassium number density in a heat pipe is not constant throughout the interaction region. From our experimental data displayed in Fig. 3, we estimate that the absorption path length $\alpha_0 L$ has a value of ~ 0.67. This corresponds to an average number density N of 5×10^{10} cm^{-3}. We believe that this value is within the experimental error for estimation of the number density in the interaction region. The value for the absorption path length is consistent with the magnitude of the angular rotations shown in Fig. 2.

We conducted similar experiments using the $4^2S_{1/2} \leftrightarrow 4^2P_{3/2}$ transition of potassium. Polarization-ellipse rotation, however, was not observed experimentally for this transition. The absence of ellipse rotation can be understood by including the effects of relaxation between magnetic sublevels of the excited state that result from collisions with the buffer gas.[14,15] These collisions decrease the efficiency of optical pumping of the ground state for the case of the $J = 1/2$ to $J = 3/2$ transition, whereas for the case of the $J = 1/2$ to $J = 1/2$ transition the collisions can increase the efficiency of ground-state optical pumping.

In conclusion, we have predicted theoretically and verified experimentally the existence of a new mechanism for polarization-ellipse rotation caused by ground-state optical pumping in atomic vapors. The rotation depends only on the ellipticity of the laser beam, the absorption path length, and the detuning of the laser beam from resonance.

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