Enhancement of magneto-optic effects via large atomic coherence in optically dense media

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(Received 22 March 1999; revised manuscript received 1 September 1999; published 19 July 2000)

We utilize the generation of large atomic coherence in optically dense media to enhance the resonant nonlinear magneto-optic effect by several orders of magnitude, thereby eliminating power broadening and improving the fundamental signal-to-noise ratio. A proof-of-principle experiment is carried out in a dense vapor of Rb atoms. Applications such as optical magnetometry, the search for violations of parity and time-reversal symmetry, and nonlinear optics at low light levels are feasible.

PACS number(s): 42.50.Gy, 07.55.Ge

Resonant magneto-optic effects, such as the nonlinear Faraday and Voigt effects [1,2], are important tools in high-precision laser spectroscopy. Applications to both fundamental and applied physics include the search for parity violations [3–5] and optical magnetometry. In this paper, we demonstrate that the large atomic coherence associated with electromagnetically induced transparency (EIT) [6,7] in optically thick samples can be used to enhance nonlinear Faraday signals by two orders of magnitude while improving the fundamental signal-to-noise ratio.

In particular, our results show that by increasing atomic density and light power simultaneously the magneto-optic signal can be enhanced substantially, and the fundamental noise (shot noise) can be greatly reduced. A nearly maximal Zeeman coherence generated under these conditions preserves the transparency of the medium despite the fact that the system operates with a density-length product that is many times greater than that appropriate for $1/e$ absorption of a weak field. At the same time this medium is extraordinarily dispersive [8,9], such that even very weak magnetic fields lead to a large magneto-optic rotation. This effect is a direct manifestation of enhancement of linear and nonlinear optical properties of a dense, coherently prepared medium. In particular, the present result is of the same nature as those resulting in slow group velocities [10,11] and large nonlinear susceptibilities [6,12]. In this regime, the optical fields are coupled strongly to the atomic medium, and optical properties of the medium cannot be described by usual perturbation expansion of nonlinear optics.

Our experimental results demonstrate the possibility of improvement over the conventional thin-medium–low-intensity approach. This opens up interesting perspectives for applications such as optical magnetometry or the search for violations of parity and time-reversal symmetry. Furthermore, the present experiments demonstrate that sensitive phase measurements with a high signal-to-noise ratio can be carried out in dense coherent media using polarization states. This makes applications of this technique feasible for new regimes of nonlinear optics at a very low light levels [13].

This work also establishes the relationship between earlier studies of nonlinear magneto-optics and recent studies on EIT-related phenomena. The key physical effect (coherent trapping of atomic population) is essentially the same in both cases, with the difference lying in the parameter domain in which the work is performed. Typical nonlinear magneto-optics measurements are done in the regime of weak fields, low coherence, and low optical density. This regime corresponds to the smallest width of the magneto-optic resonances. Resonances are typically much broader in the EIT-like regime, when strong fields are involved. However, the value of long-lived coherence in this case is quite large, which allows operation in the optically dense regime, and makes some new optical phenomena possible.

There exists a substantial body of work on nonlinear magneto-optical techniques [1,2,5,14–18], which have been studied both in their own right and for applications. Such techniques can achieve high sensitivity in systems with ground-state Zeeman sublevels due to the narrow spectroscopic features associated with coherent population trapping [19]. The ultimate width of these resonances is determined by the lifetime of ground-state Zeeman coherences, which can be made very long by a number of methods (buffer gases and/or wall coating [5,18,20] in vapor cells, or atomic cooling or trapping techniques). These resonances are easily saturated, however, and power broadening deteriorates the resolution even for very low light intensities. For this reason, earlier observations of nonlinear magneto-optic features used small light intensities and optically thin samples, which corresponds to a weak excitation of Zeeman coherences. Recently, remarkable experiments of Budker *et al.* demonstrated an excellent performance of the magneto-optic techniques in this regime: very narrow magnetic resonances were observed in a cell with a special paraffin coating [5] (an effective Zeeman relaxation rate of $\gamma_0 \sim 2\pi \times 1$ Hz).

Typical measurements of the nonlinear Faraday effect involve an ensemble of atoms with ground-state Zeeman sublevels interacting with a linearly polarized laser beam. In the absence of a magnetic field, the two circularly polarized components generate a coherent superposition of the ground-state Zeeman sublevels corresponding to a dark state. A weak magnetic field B applied to such an atomic ensemble

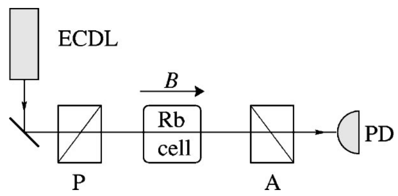

FIG. 1. Schematic of the experimental setup. The laser beam passes through polarizer (P), Rb cell, and analyzer (A), and is detected by the photodiode (PD).

causes a splitting of the sublevels and induces phase shifts $\phi_\pm$ which are different for right (RCP) and left (LCP) circularly polarized light.

In our experiment, shown schematically in Fig. 1, an external cavity diode laser (ECDL) was tuned to the 795-nm $F=2\rightarrow F'=1$ transition of the ^{87} RbD_1 absorption line. The laser beam was collimated with a diameter of 1 cm, and propagated through a 4-cm-long magnetically shielded vapor cell placed between two crossed polarizers. The cell was filled with natural Rb and a Ne buffer gas at a pressure of 3 Torr. The laser power was 2 mW at the cell entrance. The cell was heated to produce atomic densities of Rb near 10^{12} cm^{-3}. A longitudinal magnetic field was created by a solenoid placed inside the magnetic shields, and modulated at a rate of about 10 Hz. The ground-state relaxation rate was measured by decreasing sufficiently the laser power and the density until the absorption was low. The measured value of $\gamma_0 \approx 2\pi\times 400$ Hz (full width at half maximum) is attributed to time-of-flight broadening as well as to a residual inhomogeneous magnetic field. The magnetic field of the solenoid was calibrated with a commercial flux-gate magnetometer.

Figure 2 shows the result of direct measurement of the laser intensity at the photodetector after transmission through the system of two crossed polarizers ($\theta=45$ °), and a vapor cell as depicted by Fig. 1. We emphasize that no lock-in detection has been used for the data shown in Fig. 2, whereas

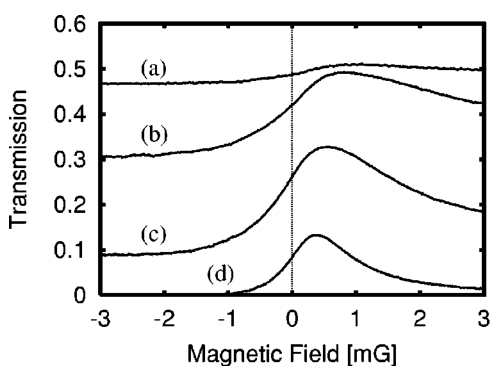

FIG. 2. Experimentally measured transmission through the system of Fig. 1, where the polarizer axes are 45° apart. The vertical scale is normalized such that unity corresponds to the transmission of the laser beam in the absence of the atomic cell and the polarizers. Thus, zero magnetic field at low density gives a transmission of 50%. The curves, from top to bottom, correspond to an increasing atomic density N of 1.1×10^{10}, 3.5×10^{10}, 1.1×10^{11}, and 2.5×10^{11} cm^{-3}.

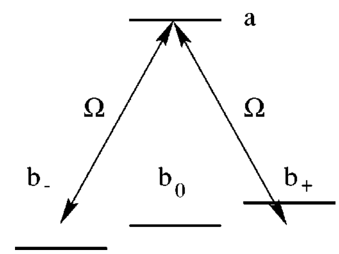

FIG. 3. A simplified, four-state model for observation of the nonlinear Faraday effect. Ω is the Rabi frequency of $\hat{\sigma}_\pm$ components of an $\hat{x}$-polarized laser field. The magnetic field B shifts $m=\pm1$ levels by $\pm\delta$.

in typical nonlinear Faraday measurements sophisticated detection techniques are usually required. We note that magneto-optical rotation angles increase with optical density as does the slope $\partial\phi/\partial B$ (Fig. 2). The latter increase is the essence of the method being described. Under the present conditions, rotation angles up to 0.7 rad have been observed (curves c and d) with a good signal-to-noise ratio. For very high densities the absorption becomes large even for high input laser intensity, and the amplitude of the magneto-optic signal does not grow with density any further.

From our measurements of the rotation angles, for the conditions outlined above we obtain

$$\partial\phi/\partial B\approx 10^3 \text{ rad/G}. \tag{1}$$

To put this result into perspective, we can estimate the shot-noise-limited sensitivity of this medium. The fundamental photon-counting error accumulated over a measurement time t_m scales inversely with the output intensity. That is, for a laser frequency ν [5],

$$\Delta\phi_{err}\simeq\frac{1}{2}\sqrt{\hbar\nu/[P(L)t_m]}, \tag{2}$$

where $P(L)$ is the power transmitted through the cell. Combining this with our measured rotation angles implies a shot-noise-limited sensitivity $B_{min}=\Delta\phi_{err}/(\partial\phi/\partial B)$ surpassing 10^{-11} G/$\sqrt{\text{Hz}}$. Although this is greater than the best observed value [4] and comparable to that in Ref. [5] it is important to note that this sensitivity is achieved in our present experiment despite nearly three orders of magnitude difference in the ''natural'' width of the Zeeman coherence γ_0. This demonstrates the very significant potential of the present technique, given that we can decrease γ_0 to the present state of the art.

We now turn to a theoretical consideration of this result. As a simple model, let us consider the interaction of a dense ensemble of atoms with ground-state angular momentum $F=1$ and an excited state $F=0$, as shown in Fig. 3. (Although the calculation presented in Fig. 4 represents a simulation of realistic rubidium hyperfine structure, this simple model, with well-chosen parameters, represents the qualitative physics quite well.) We consider a strong laser tuned to exact resonance with the atomic transition and disregard inhomogeneous broadening. The intensities and phases of the RCP and LCP components vary according to

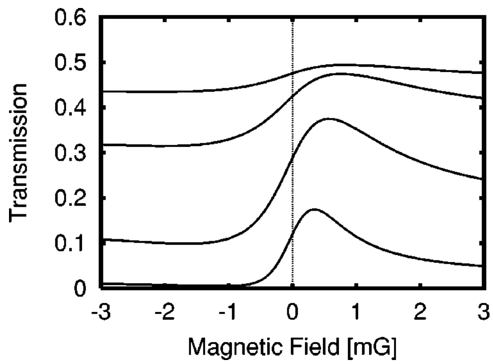

FIG. 4. Results of numerical simulations with parameters corresponding to Fig. 2.

$$\frac{1}{P}\frac{dP}{dz}=\kappa\gamma\frac{[2|\Omega|^2\gamma_0+\gamma(4\delta^2+\gamma_0^2)]\Delta\rho}{(2|\Omega|^2+\gamma\gamma_0-2\delta^2)^2+\delta^2(2\gamma+\gamma_0)^2}, \quad (3)$$

$$\frac{d\phi_\pm}{dz}=\pm\frac{\kappa\gamma\delta}{2}\frac{[4|\Omega|^2-4\delta^2-\gamma_0^2]\Delta\rho}{(2|\Omega|^2+\gamma\gamma_0-2\delta^2)^2+\delta^2(2\gamma+\gamma_0)^2}, \quad (4)$$

where $\Omega=\wp|E_\pm|/\hbar$ are the (equal) Rabi frequencies of the field components ($P\propto|\Omega|^2$), γ_0 and γ are the relaxation rate of Zeeman and optical coherences, respectively, $\delta=g\mu_B B/\hbar$ is the Zeeman level shift caused by a magnetic field B (g is a Landé factor), and $\kappa=3/(4\pi)N\lambda^2(\gamma_{a\to b}/\gamma)$ is the weak-field absorption coefficient (inverse absorption length), where $\gamma_{a\to b}$ is the natural width of the resonance. The population difference between the ground-state Zeeman sublevels and the upper state is $\Delta\rho$. This quantity is affected by optical pumping into the decoupled states (b_0 in Fig. 3), and depends upon cross-relaxation rates and applied magnetic field. For a weak magnetic field, $\Delta\rho\approx 1/3$.

One recognizes from Eq. (4) that in the case of optically thin media ($\kappa L\leqslant 1$ where L is the cell length) the phase shifts $\phi_\pm$ can be approximated by dispersive Lorentzian functions of δ, with amplitude $\phi_{max}=\kappa L\Omega^2/(2\Omega^2+\gamma\gamma_0)\Delta\rho$ and width $\delta_0=\gamma_0/2+\Omega^2/\gamma$. The former is typically rather small (on the order of mrad in the experiments of Refs. [1,2,5,15–18]), while the latter saturates when $|\Omega|^2$ exceeds the product $\gamma\gamma_0/2$, which corresponds to the usual power broadening of the magneto-optic resonance [5,16].

It is important to emphasize here that a principal difference between regimes involving low and high driving power lies in the choice of the density-length product and the degree of Zeeman coherence excited by the optical field:

$$\rho_{b\_b_+}=\frac{2|\Omega|^2\Delta\rho}{2|\Omega|^2+\gamma\gamma_0-2\delta^2+i\delta(2\gamma+\gamma_0)}. \quad (5)$$

Large coherence corresponds to a large population difference between symmetric (i.e., ''bright'') and antisymmetric (i.e., ''dark'') superpositions of Zeeman sublevels. In the low-power regime this difference is small corresponding to a small coherence. In a regime where the width of the resonance is determined by saturation, a very large (nearly maximal) Zeeman coherence is generated, as per Eq. (5). This is a signature of the so-called ''strong coupling'' regime of linear and nonlinear optics, in which susceptibilities cannot be derived from usual perturbation theory. At such large values of Zeeman coherence the magneto-optic signal is maximized if a large density-length product is chosen. In the case of a strong optical field ($|\Omega|^2\gg\gamma_0\gamma$) and weak magnetic fields $|\delta|<|\Omega|^2/\gamma,\sqrt{\gamma_0/\gamma}|\Omega|$, integration of Eqs. (3) and (4) yields, for the transmitted power and the rotation angle $\phi=(\phi_+-\phi_-)/2$,

$$P(L)=(1-\alpha_0 L)P(0), \quad (6)$$

$$\phi(L)=-(\delta/2\gamma_0)\ln[1-\alpha_0 L], \quad (7)$$

where $\alpha_0=\Delta\rho\kappa\gamma\gamma_0/2|\Omega_0|^2$, and Ω_0 corresponds to the input field.

Note that in the case of a strong input field and an optically thin medium we have $\alpha_0 L\ll 1$. However Eq. (7) shows that maximal rotation is achieved with a large density-length product $\alpha_0 L$. Clearly $\alpha_0 L$ cannot be too close to unity, since then no light would be transmitted. Using Eq. (2), one finds the optimum value $(\alpha_0 L)_{opt}=1-e^{-2}$, corresponding to a density-length product

$$\Delta\rho\kappa L|_{opt}=(\alpha_0 L)_{opt}|\Omega_0|^2/\gamma\gamma_0\gg 1. \quad (8)$$

In this case the total accumulated rotation angle is quite large, and the slope of its dependence upon B is maximal,

$$\partial\phi_{opt}/\partial B=g\mu_B/(\hbar\gamma_0), \quad (9)$$

which is in good agreement with our measured value. The significance of this result can be understood by noting that in a shot-noise-limited measurement, the minimum detectable rotation ϕ_{err} given in Eq. (2) is inversely proportional to the square root of the laser power.

To make a more realistic comparison of theory and experiment, we have carried out detailed calculations in which coupled density-matrix and Maxwell equations, including propagation through the medium and Doppler broadening, have been solved numerically for the two components of the optical field. The calculation takes into account a 16-state atomic system with energy levels and coupling coefficients corresponding to those of the Rb D_1 line. The results of these calculations are shown in Fig. 4, and are in excellent agreement with the experimental results. In particular, we note that our calculations predict the line shapes, the maximal rotation angle (which is apparently limited by the optical pumping into the $F=1$ $S_{1/2}$ hyperfine manifold), and the slope of the resonance curve.

It is important to comment at this point on possible limitations for the extension of the present technique into the domain of narrow resonances. For instance, in the case of a long-lived ground-state coherence, spin-exchange collisions can become a limiting factor for the Zeeman relaxation rate. In the case of Rb, these are a few tens of Hz at densities corresponding to the present operating conditions. We note, however, that it is possible to operate at lower densities by increasing the optical path length (e.g., by utilizing an optical cavity). Likewise, the role of the light shifts due to off-resonant coupling to, e.g., $F=1$ $S_{1/2}$ and $F=2$ $P_{1/2}$ hyperfine manifolds needs to be clarified.

For these reasons, we believe that the present approach is likely to improve the sensitivity of nonlinear magneto-optical measurements substantially. Therefore, we anticipate that this method will be of interest for sensitive optical magnetometry as well as for setting new, lower bounds to test for the violation of parity and time-reversal invariance [3,5]. Furthermore, as demonstrated here, the present technique can be used to carry out sensitive measurements of phase shifts in dense coherent media with a very high signal-to-noise ratio. (i.e., with substantially reduced technical noise contributions). For this reason interesting applications in nonlinear optics and nonlinear interferometry at low light levels [13] involving polarization states are very likely.

Note added. Recently, the relationship between electromagnetically induced transparency and nonlinear magneto-optics in *a thin medium* was extensively discussed by Budker *et al.* [11]

The authors warmly thank Leo Hollberg, Alexander Zibrov, and Michael Kash for useful discussions and Tamara Zibrova for valuable assistance. We gratefully acknowledge the support of the Office of Naval Research, the National Science Foundation, the Welch Foundation, and the Air Force Research Laboratory.

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