Compensation of ac Stark shifts in optical magnetometry

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The ac Stark shift of the resonance frequency of the nonlinear magneto-optic effect (NMOE) results in a fundamental broadening of this resonance which limits the precision of optical magnetometry based on NMOE. We have studied the dependence of the ac Stark shift versus frequency of the probing laser for the D_1 and D_2 lines of ⁸⁷Rb, and have shown that there exists a frequency where the shifts from different hyperfine components of the upper level cancel each other. This holds promise for an in-principle increase in the sensitivity of optical magnetometers. The influence of buffer gas on Faraday rotation and ac Stark shifts is also considered.

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The precision measurement of magnetic fields with optical methods is an exciting and rapidly developing branch of spectroscopy [1-5]. Recent studies in this area have focussed on the nonlinear magneto-optic effect (NMOE) [1,2,6] (also called the nonlinear Faraday effect) and have demonstrated the possibility of orders of magnitude improvement in sensitivity compared with currently existing devices [5,7].

The essence of NMOE magnetometry is as follows. Linearly polarized light, represented as a sum of two circular components of the same amplitude, travels through a medium with a Zeeman substructure on the relevant transitions as shown in Fig. 1(a). These two components create a coherent superposition of the magnetic sublevels, which is usually referred to as a "dark (or bright) state." A longitudinal magnetic field shifts the sublevels, changing the indices of refraction for the two circular polarizations by different (usually opposite) amounts. The phase difference accumulated by the two components results in a rotation of the initial polarization. Due to the high dispersion associated with a dark state, the phase difference between the two waves (or equivalently the rotation angle of the polarization) can be large, making this a particularly sensitive method for detecting small magnetic fields. The technique is called the nonlinear magnetooptic effect because the polarization rotation depends on the laser intensity.

The interaction of the radiation with nonresonant atomic transitions causes a shift of the levels called the ac Stark shift, or light shift. This effect is very important for precision



FIG. 1. (a) Simplified energy-level diagram showing the essence of NMOE. (b) The D_1 line $5s_{1/2}F=2 \rightarrow 5p_{1/2}$ and (c) $5s_{1/2}F=2 \rightarrow 5p_{3/2}$. In both *D* lines, the lower F=1 level is omitted because the ground-state splitting is much larger than the excited-state splitting.

magnetometry [7,8] as well as for atomic clocks and frequency standards [9]. For elliptically polarized light, this shift has been shown to be equivalent to an effective magnetic field [10] which results in a rotation of the polarization by an angle ϕ_{AC} even in the absence of a magnetic field. This effect is called self-rotation. (In general, a term equivalent to transverse electric field should also be included; however, in our case it is negligible due to low light power.) Although there is no self-rotation for linearly polarized light, this effect creates a nonlinear coupling between intensity and phase fluctuations of the incoming light leading to a jitter of the polarization axis. This must be taken into account when the sensitivity of a magnetometer based on NMOE is estimated [7].

Recent studies of the ac Stark shifts for the D_1 line of ⁸⁷Rb [8] showed experimentally that the ac Stark shift depends inversely on the detuning from nonresonant atomic hyperfine sublevels. The relevant energy levels are shown in Figs. 1(b) and 1(c). In this paper, we show that there is some laser detuning between the sublevels where the light shifts cancel. At this point the noncorrelated intensity fluctuations of each circular component have no effect on their relative phase, and the direction of the output polarization is not affected by the medium. Therefore, at this frequency the sensitivity of a NMOE magnetometer is limited only by the photon shot noise, allowing the use of higher laser power to achieve a better signal-to-noise ratio [5,11]. The absence of this rotation does not guarantee high magnetometer sensitivity, since the sensitivity is proportional to the slope of the polarization rotation with magnetic field $d\phi/dB$. This parameter should be large enough at the point of ac Stark suppression to maintain high sensitivity.

The experimental setup is shown in Fig. 2. An external cavity diode laser (ECDL) is tuned to the ground state F = 2 hyperfine component of either the D_1 or D_2 line of



FIG. 2. Diagram showing the experimental setup.



FIG. 3. (a) Measured rotation angle ϕ_{AC} for elliptical polarizations produced by rotations of the quarter wave plate in Fig. 2 by angles of 2°, 4°, and 6° degrees. (b) Measured Faraday rotation slope $d\phi/dB$ with linearly polarized light. Both (a) and (b) are for the ⁸⁷Rb D_1 line with no buffer gas and atomic density $N = 1.5 \times 10^{12}$ cm⁻³. (c) Calculated rotation angle ϕ_{AC} for the D_1 line. (d) Calculated Faraday rotation slope $d\phi/dB$.

⁸⁷Rb. The laser power at the entrance of the cell is 2 mW and its diameter is approximately 1 cm. A polarizer P_1 determines the initial linear polarization of the laser beam, and the ellipticity of the light is controlled by the rotation of a quarter-wave plate placed after the polarizer. An evacuated glass cylindrical cell (without buffer gas) of length 5.0 cm and diameter 2.5 cm containing saturated Rb vapor is placed inside a two-layer magnetic shield. A constant magnetic field along the direction of light propagation can be produced by a solenoid mounted inside the magnetic shield for measuring $d\phi/dB$. The atomic density of Rb is controlled by the temperature of the cell. A polarizing beam splitter is placed after the cell with its axis tilted 45° with respect to the first one. Photodiodes D_1 and D_2 collect the light from both channels of the polarizer, allowing simultaneous measurement of the polarization rotation and the transmission through the cell.

Experimental spectra for both the angle of self-rotation ϕ_{AC} and magnetic rotation slope $d\phi/dB$ are shown for the D_1 line in Figs. 3(a) and 3(b) and the D_2 line in Fig. 4. The rotation slope is measured for linearly polarized light by applying a small magnetic field (so the rotation angle is linearly proportional to the field) and then calculating the ratio between the rotation angle and the applied magnetic field. For each case the rotation angle at zero magnetic field (selfrotation) has been recorded for three different degrees of ellipticity of the laser beam [Figs. 3(a) and 4(a)]. In both cases there exists a value of the detuning where the ellipticity of the laser beam does not lead to any rotation. This compensation point is also independent on the laser intensity. For the D_1 line this is a point midway between the transitions to the two upper-state hyperfine levels. The rotation peaks are partially resolved, and in the middle point the value of rotation slope $(d\phi/dB)$ is about 0.4 of its maximum value. For the D_2 lines the point of compensation is near the center of the upper hyperfine manifold. It is important to note the compensation in the D_2 case, which occurs for an upper manifold consisting of three levels instead of two. In both of these pictures, there is another point on the high-frequency side where an extra compensation point appears. This is due to contamination of the cell by ⁸⁵Rb and demonstrates the interesting possibility to eliminate ac Stark shifts by tuning the laser between the transition for two different isotops.

To understand these results, we first analyze the simple case of motionless atoms by performing numerical simulations for the density-matrix propagation for the 13 levels of the D_1 line. The calculated rotation angle is shown in Fig. 5(a). We see that if the atoms are motionless there are two points where ac Stark shifts from different levels cancel each other very close to each resonance. Figure 5(b) shows the calculated rotation slope that predicts two sharp resonant peaks. This means that for motionless atoms a very small detuning can eliminate the effect of an ac Stark shift com-



FIG. 4. Same as Figs. 3(a) and 3(b) for the D_2 line and atomic density $N=8\times10^{10}$ cm⁻³. Zero detuning corresponds to the resonance with transition $5s_{1/2}F=2\rightarrow 5p_{1/2}F'=1$ for the D_1 line and to the center of the absorption on the transition $5s_{1/2}F=2\rightarrow 5p_{3/2}$ for the D_2 line.



FIG. 5. (a) Calculated rotation angle ϕ_{AC} and (b) calculated Faraday rotation slope $d\phi/dB$ for motionless atoms.

pletely without loss of sensitivity. However, the use of cold atoms is a complicated process that is not well suited to practical magnetometry.

For atoms in a vapor, Doppler averaging causes the cancellation points near the resonances to disappear. Thus, the shifts are compensated only in the point exactly between two transitions where the rotation slope is somewhat smaller than its maximum value. However, it has been demonstrated [11] that the proper increase of atomic density and laser power can increase the rotation rate significantly. This means that it is possible to compensate the self-rotation contribution by proper detuning of the laser without a significant loss of sensitivity.

The width of the NMOE resonance is determined by the relaxation time of the ground-state coherence, which in our case is the transient time of the atoms through the beam. There are a number of methods to increase this time: expansion of the laser beam, antirelaxation coatings on the walls of the cell [5,12], or use of buffer gas [13].

In the presence of buffer gas the transit time is determined by the diffusion of the Rb atoms through the buffer gas, and the lifetime of ground-state coherence can be increased by several orders of magnitude relative to that for the transit time of atoms across the laser beam in the absence of buffer gas. Because the coherence survives these collisions, the atoms will return to the light beam still in a coherent state, but with a different velocity. This process becomes extremely important if there is more than one unresolved or partially resolved excited state in the inhomogeneously (Doppler) broadened line, because the coherence may then be created on one transition and probed on another. (The same effect appears in coated atomic cells [5].) This implies that the steady-state solution used in the theoretical calculations for the evacuated cell does not give a correct physical picture for a cell with buffer gas.

To study this effect, we have used a cell with 0.12 torr of Kr buffer gas (length 4.0 cm and diameter 2.5 cm). The results are shown for the D_1 line in Figs. 6(a) and 6(b). We see that the ac Stark effect is eliminated by detuning between the resonances as before. However, in this case the rotation slope is also strongly suppressed in the same frequency region, being almost exactly zero at the point of compensation. In the case of the D_2 line (not shown) the rotation slope, while not zero, is much smaller at the point of compensation.

We can obtain a qualitative picture by considering the limit of high buffer-gas pressure where the atoms experience many collisions before the coherence decays, and by considering the simpler case of the D_1 line with only two upper hyperfine components. When the laser is tuned between the two upper levels there are equal probabilities for both cases of an atom being initially driven at one transition and probed on the other. However, because the matrix elements for transitions to the two upper levels have opposite signs, the corresponding atomic coherences produced by the external fields also have opposite signs. This results in the elimination of the average coherence between the transitions, leading to the disappearance of Faraday rotation. The experimental data show that in the presence of buffer gas the cancellation of



FIG. 6. (a) and (b) Same as Figs. 3(a) and 3(b) with 0.12 Torr of Kr buffer gas and atomic density $N=8\times10^{10}$ cm⁻³. (c) and (d) Same as Figs. 3(c) and 3(d) calculated under the assumption of velocity-changing collisions induced by a buffer gas.

Faraday rotation and ac Stark shifts occur at the same frequency. This implies that if buffer gas is used to increase the coherence decay time then the broadening because of the ac Stark effect cannot be compensated without strongly decreasing the signal. The result is critical when the quantum limit of sensitivity is estimated.

For a more quantitative approach, we have calculated the mean value of the atomic coherence by Doppler averaging the solution of the density-matrix equations in the approximation of given laser field. We then "plug-in" this coherence into the density-matrix equations and solve for the atomic polarization which gives the change in the polariza-

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tion of the laser field. This result is shown in Figs. 6(c) and 6(d) and agrees reasonably well with the data in Figs. 6(a) and 6(b). A detailed description of this model will be published elsewhere.

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