Limits on Magnetically Induced Faraday Rotation from Polarized $^3$He Atoms

J. Abney, M. Broering, M. Roy, and W. Korsch
University of Kentucky
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Faraday rotation has become a powerful tool in a large variety of physics applications. Most prominently, Faraday rotation can be used in precision magnetometry. Here we report the first measurements of gyromagnetic Faraday rotation on a dense, hyperpolarized $^3$He gas target. Theoretical calculations predict the rotations of linearly polarized light due to the magnetization of spin-1/2 particles are on the scale of $10^{-7}$ radians. To maximize the signal, a $^3$He target designed to use with a multipass cavity is combined with a sensitive apparatus for polarimetry that can detect optical rotations on the order of $10^{-8}$ radians. Although the expected results are well above the sensitivity for the given experimental conditions, no nuclear-spin induced rotation was observed.

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I. INTRODUCTION

The Faraday effect is a well known dispersive phenomenon in which the plane of polarization of linearly polarized light is rotated while traversing a material placed in a magnetic field which is directed along the path of light propagation. Especially in recent years this circularly birefringent effect has evolved into a precision tool for a variety of applications. For example, it is used to measure permanent electric dipole moments in atoms [1], to determine magnetic field strengths and directions in the intergalactic medium [2], as part of precision magnetometers to test fundamental symmetries [3], to measure spin-noise [4], or to probe the process of light squeezing [5], just to name a few. However, all these applications rely on the interaction of light with the electric charge and thus the rotations are induced by electric polarizabilities of the studied systems. Such effects are strongly enhanced when the probing light is monochromatic with a frequency that is slightly detuned from the center of an atomic resonance [6]. In addition to this electrically induced rotation a similar effect should exist due to the magnetic interaction of light [7]. This so called gyromagnetic Faraday effect is significantly smaller and it exhibits a different frequency dependence than its electric counterpart. The experimental verification of this magnetic effect would open up new venues for studies of fundamental properties of Dark Matter, such as putting limits on possible anomalous magnetic moments of composite low-mass Dark Matter particles [8]. Furthermore, it could be considered for the development of new ways to monitor the polarization of dense spin-polarized targets. In this Letter we present the first measurements of the spin-induced Faraday rotation on a dense hyperpolarized $^3$He gas.

II. THE GYROMAGNETIC FARADAY EFFECT

The application of Faraday rotation appears to be exclusively devoted to the coupling of light to the electric polarizability of atoms and molecules. The theory of the interaction of light with electric free or bound charges is well established and the general behavior is described classically and quantum mechanically [9]. However, the magnetic contribution to this effect has attracted less attention in the literature. It has been pointed out in several papers [7, 10] that a rotation of the light can also be induced by magnetic moments. The only claimed observation has been in the infra-red on ferrite garnets [10, 11]. Such magnetic Faraday rotations are typically vastly smaller and they are experimentally more difficult to isolate. Ideally, in order to really investigate the magnetically induced rotation, a neutral particle with minimal intrinsic structure and an anomalous magnetic moment is the ideal candidate. Naturally free neutrons would be the perfect choice. However, the problem with neutrons is that they are unstable with a lifetime of about 880 s and therefore it is very difficult to produce dense targets. This is, for example, a major factor in the progress of improving the limit of the neutron permanent electric dipole moment [12, 13]. A system that has several of the advantages required here is helium-3. This helium isotope is predominantly in a singlet S ground state at room temperature, the single-electron excitation energy is relatively large ($\Delta E_{1S-1P} \approx 21$ eV), and it has a spin-1/2 nuclear ground state with a magnetic moment of $-2.12\mu_N$. Additionally, two well developed technologies exist to produce relatively dense and nuclear spin-polarized targets with high values of polarization. Typically such targets are either polarized by means of metastability exchange optical pumping (MEOP) or spin exchange optical pumping (SEOP). State of the art polarization values of up to about 80% have been reported at number densities reaching 7 amagats [14].

The theory for the gyromagnetic Faraday effect has been developed by Gardner and also Krinchik and Chetkin (see [8, 10]). We summarize the main ideas in the following. As shown in [8], a Faraday rotation induced by a sample of magnetic moments is given by:

$$\theta_{FR} = \mu_0 \frac{\hbar c \mu}{\rho P L},$$

where $\mu$ denotes the magnetic moment of the particle, $\rho$ is number density of the sample, $P$ is the spin-polarization, and $L$ is the sample length. The quadratic dependence on the magnetic moment is due to the replacement of the magnetization of the target by its polarization, $M = \mu_0 \rho P$. It should be emphasized that the induced rotation is, to leading order, independent of the frequency of the probing light. Using typical numbers for existing polarized $^4$He targets, for example a density of 4.4 amagats, a target length of 40 cm, and a spin-polarization of 55%, one would expect a spin induced rotation of about 120 nrad. In order to measure such small rotation angles, a dedicated experiment had to be conceived which combined such a polarized target with a highly sensitive Faraday rotation apparatus and a multipass cavity.
III. $^3$HE TARGET

For the experiment, the method chosen to polarize $^3$He is spin exchange optical pumping. The $^3$He gas is contained inside an aluminosilicate glass cell of similar style to the SEOP cells used at Jefferson Lab for electron scattering experiments [15]. A spherical pumping chamber on top is connected by a transfer tube to a 39.45 cm long cylindrical target chamber on the bottom. Rb vapor in the pumping chamber is polarized with a high power, narrowband (120 GHz) 795 nm laser whose light is converted into circular polarization with quarter-wave plates mounted rotation stages. The Rb then transfers this polarization to K vapor via spin exchange collisions. Both alkali metals then polarize $^3$He with spin exchange collisions and the $^3$He gas slowly gains nuclear spin polarization in this manner. The pumping chamber is contained inside a forced air oven heated to 235°C that is suspended on a platform which has vibration damping to reduce noise due to movements. Located below the oven is the target chamber which is in the path of the probe laser used to measure the Faraday rotation. Fig. (1) shows the basic geometry of the cell. The target chamber of the cell has flat windows with a diameter of 2.54 cm on the end to ensure minimal divergence of the probe laser. These windows are designed for use with a multipass optical cavity so the probe laser can be passed through the chamber multiple times to increase the optical rotation angle. A static magnetic field, stabilized with a feedback system and running parallel to the length of the target chamber and the probe laser provides an axis to align the nuclear spins. A separate magnetic field transverse to this is used to flip the spins with adiabatic fast passage (AFP) frequency sweep NMR. The $^3$He polarization can then be measured with a pair of pick-up coils on the sides of the target chamber. Absolute polarization is extracted by electron paramagnetic resonance frequency shift where the small Zeeman shift in the alkali metal resonance frequencies due to the effective magnetic field of the polarized $^3$He is detected. The specific cell used for this experiment has a $^3$He density of 4.4 amagats in the target chamber under experimental conditions and a maximum measured $^3$He polarization of 55%.

IV. MEASUREMENT METHOD

![Diagram of the experimental setup](image)

FIG. 1. Schematic of the experimental setup to measure the gyromagnetic Faraday rotation from polarized $^3$He.

The Faraday rotation is measured with a triple modulation technique in combination with lock-in amplifiers (LIA) [16]. A schematic of the setup is shown in Fig. (1). Laser light is polarized by a linear polarizer, traverses
the cavity mirrors and the cell target chamber, then passes into the polarization state analyzing optics encased in magnetic shielding. The optical rotation is converted into a change in intensity by a photoelastic modulator (PEM) and another linear polarizer with transmission axis at 45° to the first polarizer. After the second linear polarizer is an optical chopper and the detector. The PEM dynamically alters the polarization state of the light at its operating frequency of 50 kHz, detecting at the first harmonic gives a signal proportional to the optical rotation. DC drifts are controlled for by measuring the total light intensity at the optical chopper frequency of 1 kHz. To isolate the magnetic Faraday effect of the 3He nuclear spins from background static optical rotations, the final modulation is flipping the spins with AFP NMR. The spins switch from parallel to anti-parallel with respect to the static alignment field, each state having a different rotation associated with it. Every flip incurs some polarization loss due to magnetic field gradients, therefore constant spin flipping will result in an equilibrium polarization value smaller than the maximum and given by the balance between the pumping rate and the AFP losses. The optimum rate determined experimentally for the setup is one flip every 200 s yielding a $P_{eq} \sim 17\%$. Timing for the spins flips is handled by a computer that controls the function generator powering the transverse RF coils, the computer also sends a 2.5 mHz reference frequency to the final lock-in amplifier. In total there are three lock-in amplifiers that are referenced to the three modulation frequencies, one at 1 kHz for the chopper, one at 100 kHz for the PEM, and one at 2.5 mHz for the spin flipping. Both the DC LIA and the PEM LIA are connected to the detector and the de-modulated voltage output of the PEM LIA provides the input for the Spin Mod LIA. The ratio of the voltages from the LIAs with a prefactor gives the rotation angle

$$\theta = \frac{G_{PEM}}{12.22} \frac{V_{Mod}}{V_{DC}}. \quad (2)$$

The factor $G_{PEM}$ is the sensitivity setting of the PEM LIA in volts and $V_{Mod}, V_{DC}$ are the voltages measured by the Spin Mod LIA and DC chopper LIA, respectively. The 12.22 comes from a combination of various numerical factors needed to extract the true voltage amplitude from the LIA. To calibrate the system for the small values expected, nanoradian scale Faraday rotation from a glass sample with a known Verdet constant is measured with a square wave magnetic field oscillating at 2.5 mHz.

To boost the size of the signal expected from 3He a multipass cavity is implemented to increase the effective length of the target. Multipass cavities have been previously used to amplify optical rotations as part of sensitive magnetometers [17] and generate large rotation angles for spin noise measurements [18]. A pair of spherical concave mirrors with a 1.5 m focal length and coated with a high reflectivity coating form a Herriott style cavity [19] with the target chamber in between them. The laser beam reflects back and forth between the mirrors in an elliptical pattern. Each mirror has a hole offset from the center to allow the laser to enter and exit the cavity. While cylindrical mirrors can achieve a larger number of beam passes [20], spherical mirrors were chosen to better focus the beam and compensate for the divergence caused by the end windows on the target chamber. The mirrors sit inside a pair of coils to reduce the ambient longitudinal magnetic field to the 10 mG level. The maximum number of passes for the setup when taking data for the experiment is 13, determined by counting the number of beam spots on the surface of the mirrors. This number is limited by the large distance between the mirrors and intensity losses due to the glass windows on the cell.

Since the constant flipping of the nuclear spins reduces the equilibrium polarization to about 17% and the multipass cavity makes the effective length 13 times longer the calculated rotation value from the magnetic Faraday effect changes from 120 nrad to 455 nrad or 35 nrad per pass. The effect does not depend on the frequency of light so this value is true regardless of the probe laser used. Two different lasers were utilized to take measurements, a 633 nm HeNe with a maximum output power of 35 mW and a 405 nm external cavity diode laser with a maximum output power of 60 mW. As a result of larger intensity losses from the windows for blue light the maximum number of passes is reduced to 9, with the expected signal still in the measurable range.

The magnetic Faraday rotation is measured in the following way, the 3He cell is polarized to near maximum along the axis of the holding magnetic field with a magnitude of $B_0 = 21.5$ G. When the polarization is high an EPR calibration measurement is performed and the spin modulation is started via the computer, every 200 s the $B_1 = 200$ mG transverse oscillating magnetic field is turned on to flip the spins through frequency sweep NMR. The polarization is measured during every flip with the NMR pickup coils on the sides of the target chamber. At the same time, the quarter wave plates that convert the light from the pumping laser to circular polarization are rotated so the target continues to be pumped in the correct state. Once the
equilibrium polarization is reached the data collection is started for the three lock-in amplifiers. As a result of the low frequency integration time constants of 1000 s are applied to the Spin Mod and DC LIAs. The optical rotation is measured for several hours while the target is polarized to collect enough statistics before the pumping laser is turned off and the $^3$He polarization is allowed to decay to near zero. Without changing any other parameters, the optical rotation is measured while the target is not polarized. Fig. 2 shows the polarization and expected Faraday rotation during a typical measurement. The two data sets can then be compared to see if there is a magnetic Faraday effect from the $^3$He nuclear polarization. After enough statistics have been collected for the non-polarized target the pumping laser can be turned on again and the polarization will increase back to the equilibrium value. The measurement cycle can then be repeated.

V. RESULTS

Several data sets were taken for the 633 nm HeNe laser and the results are shown in Fig. 3. Each pair of polarized and non-polarized data points that were collected sequentially are plotted next to one another for comparison. A total of nine data sets are shown with an average $^3$He polarization 16.7±1.5%. Furthermore, theoretical Faraday rotation for this polarization is displayed along with the calculated error given by the shaded region. Error bars on the points are from statistics and show one standard deviation. The weighted averages for the polarized target data points and non-polarized target data points are the dashed lines. Not only do the measurements for the polarized target not match the expected value but they exhibit no difference from the non polarized measurements indicating that the effect was not observed. The process was repeated with the 405 nm external cavity diode laser, Fig. 4 shows a similar plot where the average $^3$He polarization was 17.0±1.5%. The larger error bars for the 405 nm wavelength are most likely due to more intensity noise in the laser. Again, there appears to be no effect due to the helium nuclear spins and the measured rotations are well below the theoretical prediction. Table I lists the averages for both wavelengths and the theory calculation.
FIG. 3. Measurements of the gyromagnetic Faraday rotation with a probe laser wavelength of 633 nm and 13 passes in the multipass cavity. Each pair of points corresponds to one measurement cycle of sequential polarized/unpolarized data sets with the averages of all the polarized markers and unpolarized markers given by the dashed lines.

TABLE I. Weighted averages of the results for both laser wavelengths.

| Wavelength | Polarized Average (nrad/pass) | Non-Polarized Average (nrad/pass) | Theory (nrad/pass) |
|------------|-------------------------------|----------------------------------|-------------------|
| 633 nm     | 6 ± 1                         | 7 ± 1                            | 36 ± 4            |
| 405 nm     | 11 ± 2                        | 14 ± 3                           | 37 ± 4            |

VI. SYSTEMATICS

Due to the fact that no signal from the nuclear spins was observed, a wide array of systematics were investigated to determine if any concealed or compensated for the gyromagnetic Faraday rotation from $^3$He. The full list is given in Table II. A few will be briefly mentioned in this section. It has been shown previously that lasers with a high intensity focused to a narrow beam width can have a self induced birefringence [21, 22]. Though the probe lasers seems far away from these extreme effects, the power was varied from several mW down to a few hundred µW for both lasers. No change was observed in the rotation measurements other than an increase in the statistical noise. A false signal at the spin modulation frequency due to a very small amount of pumping laser light leaking into the analyzing optics was discovered and corrected for by adding bandpass filters for the probe lasers along with additional shielding to block the light. There are several corrections to look for due to the glass cell windows and other gases inside the cell being inside the classical magnetic field from the polarized $^3$He atoms. This field is modulated at the same frequency as the spins and could also created false signals. The Verdet constants for the glass (Corning 1723) and N$_2$ are well known, the calculated Faraday rotation from these expected at this field strength is below the sensitivity of the apparatus. Likewise, resonant Faraday rotation at the D2 line of Rb has been measured in a 10 cm test cell at room temperature, when the same measurement is performed on the target chamber of the
helium cell no rotation angle is measured indicating that the density of Rb vapor is too low to be detected. Thus, at the far off resonance laser wavelengths used for the gyromagnetic Faraday rotation no contribution from Rb is expected. Finally, using the known Verdet constant for the electric part of the Faraday effect [23], the electrically induced rotation is too small to be detected at magnetic fields generated by the target magnetization.

VII. CONCLUSIONS

In summary, the gyromagnetic Faraday effect is a predicted optical rotation induced by magnetic moments. It is a purely magnetic effect that is separate from the standard Faraday effect which results from the coupling of light to electric polarizabilities or atomic or molecular resonances. Theoretically, the rotation angles expected are significantly smaller and exhibit no frequency dependence on the probe light which would allow the effect to be isolated from other optical rotations. We used a dense hyperpolarized $^3$He gas as a test system to attempt to discover the effect because it has many favorable qualities that lend to isolating the signal. A $^3$He target was polarized with spin exchange optical pumping and combined with a sensitive apparatus that can detect Faraday rotations several times smaller than the expected signal size to take several measurements at two different probe laser wavelengths. The results showed no nuclear spin induced rotation in the predicted range. Additionally, we observed no difference between a polarized and an unpolarized target indicating that the effect is even smaller than the limit that the setup can measure. One difference that should be noted is that the theory and estimates were for bare magnetic moments while the experiment was performed with entire atoms. The interaction of light with more complex systems might alter the coherent accumulation of the Faraday rotation angle [24].

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| Systematic Effect | Contribution to FR |
|-------------------|--------------------|
| Vary $|B_0|$ between 10 G and 21 G | No detectable effect |
| Vary probe laser power $(28 \text{ mW} \rightarrow 500 \mu \text{W})$ | No detectable effect |
| Change of DAQ (Use FFT instead of LIA) | No detectable effect |
| Change NMR flipping frequency $(2.5 \text{ mHz} \rightarrow 10 \text{ mHz})$ | No detectable effect |
| Vary target polarization $(P \approx 55\% \rightarrow P \approx 16\%)$ | No change |
| Vary number of passes in Herriott cavity $(15 \rightarrow 5)$ | No detectable effect |
| Higher order theory corrections (in frequency) | $<1\%$ of expected rotation |
| Correction due to spin misalignment with the probe laser | $<1\%$ of expected rotation |
| Contribution from electric FE from helium-3 | 0.019 nrad/pass (633 nm) 0.065 nrad/pass (405 nm) |
| Contribution from cell end windows | 1 nrad/pass (633 nm) 2 nrad/pass (405 nm) |
| Errors due to cavity mirrors | Signals scale as expected, no detectable effect observed |
| Depolarization due to cell window misalignment | $\ll 0.5\%$ |
| False signal from pumping laser light | Corrected with bandpass filters |
| FE from Rb in the target chamber | No detectable effect |
| FE from $\text{N}_2$ in the target chamber | 0.008 nrad/pass (633 nm) 0.015 nrad/pass (405 nm) |
| FE from air outside windows due to He magnetic field | 0.003 nrad/pass (633 nm) 0.007 nrad/pass (405 nm) |