Nonlinear magneto-optical rotation with modulated light in tilted magnetic fields

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Larmor precession of laser-polarized atoms contained in anti-relaxation-coated cells, detected via nonlinear magneto-optical rotation (NMOR) is a promising technique for a new generation of ultra-sensitive atomic magnetometers. For magnetic fields directed along the light propagation direction, resonances in NMOR appear when linearly polarized light is frequency- or amplitude-modulated at twice the Larmor frequency. Because the frequency of these resonances depends on the magnitude but not the direction of the field, they are useful for scalar magnetometry. New NMOR resonances at the Larmor frequency appear when the magnetic field is tilted away from the light propagation direction in the plane defined by the light propagation and polarization vectors. These new resonances, studied both experimentally and with a density matrix calculation in the present work, offer a convenient method for NMOR-based vector magnetometry.

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When linearly polarized light, frequency- or amplitude-modulated at \( \Omega_m \), resonantly interacts with an atomic vapor in the presence of a magnetic field, the polarization of the light can be observed to rotate synchronously with the modulation. This effect is known as nonlinear magneto-optical rotation with frequency-modulated light (FM NMOR) \( \frac{\hbar}{2} \) or amplitude-modulated light (AMOR) \( \frac{\hbar}{2} \). It occurs when \( \Omega_m \) is a subharmonic of the quantum beat frequency; the quantum beat frequency is at the first or second harmonic of the Larmor frequency \( \Omega_L \) for the lowest-order effect discussed here. Higher-order effects involve quantum beat frequencies at other multiples of \( \Omega_L \). \( \frac{\hbar}{2} \). The width of the resonance between \( \Omega_m \) and \( \Omega_L \) is given by the relaxation rate of the atomic ground-state coherences. When the atomic vapor is contained in a paraffin-coated cell, in which ground-state atomic coherences can survive on the order a second, the widths of the resonances can be as small as 0.6 Hz. \( \frac{\hbar}{2} \). These resonances allow extremely precise measurements of the magnetic field over a wide field range, with magnetometric sensitivities exceeding \( 10^{-11} \text{ G/}\sqrt{\text{Hz}} \) for low fields (competitive with the best atomic magnetometers, see for example Ref. \( \frac{\hbar}{2} \), as well as SQUID magnetometers \( \frac{\hbar}{2} \)) and reaching \( 4 \times 10^{-10} \text{ G/}\sqrt{\text{Hz}} \) for higher fields (up to 1 G). \( \frac{\hbar}{2} \). The FM NMOR method used in the present work, as well as AMOR, can be applied to studies of nuclear magnetic resonance and magnetic-resonance imaging \( \frac{\hbar}{2} \), measurements of geophysical fields \( \frac{\hbar}{2} \), and tests of fundamental symmetries \( \frac{\hbar}{2} \). The same approach can also be used in construction of chip-scale atomic magnetometers \( \frac{\hbar}{2} \).

Although the magnetometric method based on FM NMOR enables sensitive measurements of the magnetic field, the measurements are scalar, i.e., the position of a given resonance depends only on the magnitude, and not the direction, of the magnetic field. However, the relative magnitudes of the FM NMOR resonances can depend on the magnetic field direction. Thus, a detailed analysis of the FM NMOR signal could give some information about the direction of the magnetic field. Towards this end, we study here the dependence of the FM NMOR signal on the magnetic field direction.

In the Faraday geometry, in which the magnetic field is along the light propagation direction, the main resonance occurs at \( \Omega_m = 2\Omega_L \), because of the symmetry of the optically pumped state, as discussed below. We find that when the magnetic field direction is tilted in the plane perpendicular to the light polarization axis, only this resonance at \( 2\Omega_L \) is observed, with its amplitude depending on the tilt angle. However, when the magnetic
field is tilted toward the light polarization axis, a new resonance appears at \( \Omega_L \); the relative magnitudes of the resonances at \( \Omega_L \) and \( 2\Omega_L \) depend on the tilt angle.

The scheme of the experiment is shown in Fig. 1. An anti-relaxation-coated buffer-gas-free vapor cell, containing isotopically enriched \(^{85}\text{Rb}\), was placed within a four-layer magnetic shield. The magnetic shield provided passive attenuation of dc magnetic fields by a factor of \( 10^6 \) \[15\]. A set of three mutually orthogonal magnetic-field coils placed inside the innermost layer enabled compensation of the residual average magnetic field and first-order magnetic field gradients inside the shield. The coils were also used for generation of an arbitrarily oriented magnetic field inside the shield. The rubidium atoms interacted with a 2-mm diameter laser light beam, linearly polarized along the \( y \)-axis. An external cavity diode laser was tuned to the rubidium D1 line (795 nm) and its central frequency was stabilized with a dichroic atomic vapor laser lock \[16, 17\] at the low-frequency wing of the \( F = 3 \rightarrow F' \) transition. The laser-modulation frequency ranged from 100 Hz to 400 Hz with 300 MHz (peak to peak) modulation depth and light power was \( \sim 3 \) \( \mu \)W. Upon passing through the cell, the light was analyzed using a balanced polarimeter. The polarimetric signal was detected with a lock-in amplifier at the first harmonic of \( \Omega_m \) and stored on a computer.

The FM NMOR signals were studied as the magnetic field direction was tilted in both the \( xz \)-plane (perpendicular to the light-polarization axis) and the \( xy \)-plane (containing the light-propagation vector and light-polarization axis). Figure 2 shows the signals with the magnetic field \( \mathbf{B} \) in the \( xz \)-plane at various angles to the light propagation direction. The strength of the magnetic field was equal for all the measurements (\( \Omega_L = 98.5 \) Hz). The main resonance occurs at \( \Omega_m = 2\Omega_L \). The small resonance at \( \Omega_L \) is a result of using the modulated pump beam as a probe. For the frequency-modulated probe the time dependence of the light-polarization plane is observed, even if the atomic polarization in the medium is static. This effect has been analyzed in detail in Ref. \[18\] Sec. IV. The amplitudes of the resonances decrease with increasing angle in the \( xz \)-plane (Fig. 3). The experimental signals are in qualitative agreement with the predictions of a density-matrix calculation \[22\] of FM NMOR for a model \( F = 1 \rightarrow F' = 0 \) transition (Fig. 1).

The observed dependence on the magnetic-field direction can be understood by considering the time evolution of the atomic polarization. In the absence of a magnetic field, the linearly polarized light creates atomic alignment (polarization with a preferred axis, but no preferred direction) along the \( y \)-axis. This alignment can be visualized with a method described in, for example, Ref. \[19\]: a surface is plotted whose radius in a given direction is equal to the probability of finding the maximum projection of angular momentum along that direc-

![Fig. 1: Experimental setup. The magnetic field coils mounted inside the innermost shielding layer and used for creation of arbitrarily oriented magnetic field are not shown.](image1)

![Fig. 2: The FM NMOR in-phase and quadrature signals vs. modulation frequency recorded for various angles between the magnetic field and the light propagation direction in the \( xz \)-plane. A small residual signal observed for 90° is a result of a small \( y \)-component of the magnetic field.](image2)

![Fig. 3: The amplitude of the FM NMOR signals recorded at 2\( \Omega_L \) vs. the tilt angle of the magnetic field in the \( xz \)-plane. The solid line is a cosine fit to the experimental points.](image3)
tion. When a magnetic field is applied, the alignment precesses around the magnetic-field direction at the Larmor frequency \([22]\). Since in this case \( \mathbf{B} \) is perpendicular to the alignment axis, the polarization returns to its original state after a 180° rotation, i.e., in half a Larmor period \([23]\). Thus optical rotation, induced by the rotating linear dichroism, is periodic at twice the Larmor frequency \([24] \). Because only linear dichroism transverse to the light propagation direction (i.e., in the \(yz\) plane) can produce rotation, the amplitude of the rotation decreases as the cosine of the angle between \( \mathbf{B} \) and the light propagation direction, as seen experimentally in Fig. 3.

Figure 4 shows the FM NMOR signals for the magnetic field tilted in the \(xy\) plane at various angles to the light propagation direction; corresponding theoretical results for a \( F = 1 \rightarrow F' = 0 \) transition are shown in Fig. 7. When the magnetic field is parallel to the light propagation direction a strong FM NMOR resonance is observed only at \( \Omega_m = 2\Omega_L \). However, when the magnetic field is tilted toward \( y \)-axis an additional resonance appears at \( \Omega_m = \Omega_L \). The amplitude of this resonance becomes more pronounced with increasing the angle, while the amplitude of the resonance at \( 2\Omega_L \) decreases (Fig. 5). However, when the magnetic field is tilted by more than 60° the amplitude of the resonance recorded at \( \Omega_L \) also starts to decrease and reaches zero when the magnetic field is directed along the \( y \)-axis.

Once again, these signals can be explained by considering the time-evolution of atomic polarization \([25] \). Because the magnetic field is no longer perpendicular to the alignment axis, the polarization takes a full Larmor period to return to its original state. The anisotropy of the medium in the \( yz\)-plane is modulated at two frequencies: \( \Omega_L \) and \( 2\Omega_L \), as reflected in the plot of time-dependent optical rotation \([26] \). The larger the tilt angle between \( \mathbf{B} \) and the light propagation direction, the bigger the difference between the polarization states at the beginning and middle of the Larmor periods, increasing the signal at \( \Omega_L \). However, for large angles, the magnetic field direction is nearly along the alignment axis, reducing the effect of the field on the polarization and thus reducing the optical rotation.

The data presented in Figs. 2 and 3 can be also understood in a language of atomic coherences. When the magnetic field is in the \(xz\)-plane, the light polarization axis is perpendicular to the magnetic field. Thus, with the quantization axis along the magnetic field, the light can only create coherence between the \( m = 1 \) and \( m = -1 \).
FIG. 6: The FM NMOR signal vs. modulation frequency $\Omega_m$ recorded for different tilt angles of the magnetic field in the $xy$-plane. We proved with theoretical simulations that the small signal observed at $\Omega L$ for the magnetic field oriented along $y$-axis is due to the misalignment between the magnetic field and the light polarization direction.

FIG. 7: Theoretical calculation of FM NMOR signal vs. modulation frequency for a $F = 1 \rightarrow F = 0$ transition for various tilt angles of the magnetic field in the $xy$-plane.

Zeeman sublevels. The frequency splitting between these sublevels is $2\Omega_L$, so the resonance is observed at this frequency. However, when the magnetic field is tilted in the $xy$-plane, the light is a linear superposition of polarizations parallel and perpendicular to the magnetic field. In this case, the light can create coherences between sublevels with $\Delta m = 1$ and $\Delta m = 2$, so resonances are observed at both $\Omega L$ and $2\Omega L$.

FIG. 8: The amplitude of the FM NMOR signals recorded at $\Omega L$ and $2\Omega L$ vs. the tilt angle of the magnetic field in $xy$-plane.

In conclusion, we have studied resonances in nonlinear magneto-optical rotation with frequency-modulated light (FM NMOR) when the magnetic field is tilted at angles away from the direction of light propagation. When the magnetic field is tilted in the plane defined by the light polarization and light propagation vectors, a new FM NMOR resonance appears at the Larmor frequency. The amplitude of the resonance depends on the tilt angle and thus can be useful for vector magnetometry or for aligning a scalar magnetometer to reduce heading errors. When the magnetic field is tilted in the orthogonal plane, no new resonance appears at the Larmor frequency. For tilt angles in both planes, the amplitude of the FM NMOR resonance at twice the Larmor frequency decreases with increasing tilt angle. The effects have been modeled with density matrix calculations, yielding good agreement with the experimental observations, and can be understood through visualization of the time-dependence of the atomic polarization moments.

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FIG. 9: As Fig. 8 but with B in the xy plane at 17.5° to the light propagation direction \( \hat{x} \). (a) The alignment axis in this case is not perpendicular to the magnetic field, so it takes a full Larmor period \( \tau \) to return to its original state. (b) The optical rotation. The optical anisotropy in the yz-plane has components at \( \Omega_L \) and \( 2\Omega_L \).


[22] The optical Bloch equations are solved using the “matrix-continued-fraction method” in the manner of Ref. 20; the density matrix is expanded in a Fourier series in harmonics of the modulation frequency, and the resulting matrix recursion relation for the Fourier coefficients is inverted as a matrix continued fraction, which is evaluated by assuming a high-harmonic cutoff.

[23] In this work the light power was sufficiently low to exclude effects such as alignment-to-orientation conversion that cause more complicated polarization evolution.