Polarization dependence of magnetic resonances in rubidium vapor using amplitude-modulated light: Theoretical and experimental investigations

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We report on experimental and theoretical investigations on the polarization dependence of magnetic resonance generated by synchronous optical pumping. Narrow linewidth magnetic resonances are obtained experimentally by using a rubidium vapor cell with antirelaxation coating. We studied the effect of light ellipticity on the amplitudes and widths of magnetic resonances when light modulation frequency matches with $2\Omega_i$ (alignment) and $\Omega_i$ (orientation) in a Bell-Bloom interaction geometry, with $\Omega_i$ corresponding to the Larmor frequency. Both $2\Omega_i$ and $\Omega_i$ resonance amplitudes showed a strong dependence on the light ellipticity. In addition, we also show that the duty cycle of light modulation can control the rate of variation of $2\Omega_i$ and $\Omega_i$ resonance amplitudes. This can find potential usage in applications that require in situ measurement of light ellipticity. We also studied the dependence of $2\Omega_i$ and $\Omega_i$ resonance amplitudes on the polarization angle by using a linearly polarized light. We observed that these amplitudes oscillate periodically with the polarization angle. This oscillatory behavior is sensitive to the tilt in magnetic field orientation from the polarization plane. Such a property could be used to realize a vector magnetometer using synchronous optical pumping. A density matrix-based theoretical model is developed to simulate the resonance spectrum for different light polarizations. The model accurately reproduces the above-mentioned experimental observations.

I. INTRODUCTION

Coherent excitation of atomic ensemble with resonant light produces atomic coherence (or atomic polarization) in the ground-state of the medium. These coherences cause interesting nonlinear magneto-optical phenomena such as Hanle effect [1], and nonlinear magneto-optical rotation (NMOR) [2,3] in the presence of magnetic field. Typically, these phenomena are observed by monitoring the changes in light transmission, scattered-light intensity or light polarization angle around the zero-magnetic field [4,5]. Synchronous optical pumping is a well-established technique in which atomic coherence is produced by optically pumping the atoms at the rate of their Larmor precession, thus creating a resonance at nonzero magnetic field. It is implemented by modulating the light amplitude [6,7], frequency [8,9] or polarization [10–12]. This method has been widely used in atomic magnetometry [13,14], and also, in precision measurement of permanent electric-dipole moment [15–17].

In synchronous optical pumping experiments, a spectrum containing multiple magnetic resonances is produced due to interaction of atoms with the frequency sidebands of modulated light [6,7]. Light polarization decides the type of magnetic resonance produced in this spectrum. Primarily, two types of magnetic resonances are produced due to (i) precession of atomic dipole moment (known as ‘orientation’) and (ii) precession of atomic quadrupole moment (known as ‘alignment’) [18,19]. In NMOR magnetometry, a linearly polarized light is used to produce ‘alignment’ resonance in a Faraday geometry where the magnetic field is applied along the light propagation direction [8]. Balanced polarimetric detection is used in NMOR magnetometry to measure polarization angle with high sensitivity [13,14,20]. Both frequency and amplitude modulation of light have been explored in NMOR magnetometry. Most synchronous optical pumping experiments to date have been performed using either circular or linearly polarized light; after it was first demonstrated by Bell and Bloom [21] using amplitude modulated light with a magnetic field perpendicular to the light propagation direction. Unlike NMOR, Bell-Bloom technique produces an ‘orientation’ resonance using a circularly polarized light. Recently, this technique has been extended for remote detection of geomagnetic field by exciting sodium atoms in the mesosphere [21–24].

It is known that the sensitivity of an optical magnetometer depends on the relative angle between light propagation direction and magnetic field direction. For example, a longitudinal magnetic field parallel to light propagation direction could not produce a magnetic resonance (considered as ‘dead-zone’) using Bell-Bloom technique. Studies have shown that periodic modulation of light polarization between two orthogonal states (right- and left-circular or linear) can eliminate the dead-zone problem as it produces both ‘alignment’ and ‘orientation’ resonances simultaneously [12,25]. Alternatively, excitation using

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elliptical light polarization could improve the directional response of the optical magnetometer. However, the effect of light ellipticity on magnetic resonances generated by synchronous optical pumping has not been very well studied. In this work, we present the outcome of our studies on the dependence of magnetic resonance on light ellipticity and polarization angle. A rubidium (Rb) vapor cell with octade-cytrichlorosilane (OTS) antirelaxation coating has been used in the experiment to produce narrow linewidth magnetic resonances. We show that elliptically polarized light can simultaneously produce both $2\Omega_L$ (alignment) and $\Omega_L$ (orientation) resonances in the Bell-Bloom interaction geometry. We have developed a theoretical model to simulate the resonance spectrum using light with a varying degree of ellipticity. Amplitudes of $2\Omega_L$ and $\Omega_L$ resonances show a strong dependence on the light ellipticity. Further, our study shows that the duty cycle of light modulation can effectively control the rate of variations in $2\Omega_L$ and $\Omega_L$ resonance amplitudes. Finally, we have presented a simple scheme for vector magnetometry to determine the magnetic field direction based on measurements of oscillations in the peak amplitudes of $2\Omega_L$ and $\Omega_L$ resonances with light polarization angle.

This paper is organized as follows. In Section II below, we discuss the theoretical model developed for studying the dependence of magnetic resonances on light ellipticity and polarization angle. A description of our experimental setup is provided in Sec. III. Results and discussions including comparisons between all experimental and theoretical results are presented in Sec. IV.

II. THEORETICAL MODEL

Our theoretical model is based on atomic density matrix equations. We consider a resonant excitation between the hyperfine ground state $F_g = 3$ and the hyperfine excited state $F_e = 2$ in $^{85}$Rb $D_1$ manifold, including all Zeeman sublevels in each hyperfine state. To study magnetic resonances, we considered a laser field propagating along the $z$-axis:

$$\vec{E}(t) = \hat{e}_z E_0 e^{-i\omega t} + c.c. \quad (1)$$

where $E_0$ is the amplitude of laser field with resonant frequency $\omega$. The polarization vector $\hat{e}_L$ of the field is described by

$$\hat{e}_L = \hat{e}_x \cos \epsilon + i \hat{e}_y \sin \epsilon \quad (2)$$

where $\hat{e}_x$ and $\hat{e}_y$ are the unit vectors along $x$- and $y$-axes, respectively and $\epsilon$ is the ellipticity angle of the laser field. All polarization states of the laser can be described by the value of $\epsilon$ ranging between $0^\circ$ to $45^\circ$. For our modeling purposes, we have considered (i) linear ($\epsilon = 0^\circ$) and (ii) elliptical polarizations ($\epsilon \neq 0^\circ$) of the optical field corresponding to a transition from ground state $F_g = 3$ to excited state $F_e = 2$. Fig. 1 depicts both of these scenarios. In Fig. 1(a), for the linearly polarized light, we consider the light propagation along the $\hat{z}$ direction, electric field vector is chosen along the $y$-axis and a magnetic field $B$ is considered parallel to the $x$-axis ($B = B_x$). To give a physical picture of light-atom interaction, we choose the axis of quantization to be along the magnetic field direction (i.e. $x$-axis). In the presence of magnetic field, the Zeeman sublevels shift by integer multiples of the Larmor frequency $\Omega_L$, i.e. $\pm m L \Omega_L (m \gamma B_x)$, where $\gamma$ is the Gyromagnetic ratio and $m_L$ is the magnetic quantum number of the sublevels. Since the linearly polarized field is chosen perpendicular to the quantization axis, it produces equal $\sigma^+$ and $\sigma^-$ transitions corresponding to $\Delta m_{eg} = \pm 1$ between the ground and excited state sublevels, as shown in Fig. 1a.

These two transitions create a three-level $A$-system involving one common excited state and two ground state Zeeman sublevels with $\Delta m_g = 2$, shown using a curved arrow in Fig. 1(a). A magnetic resonance is formed by coherent population trapping (CPT) due to a dark superposition of the participating ground state Zeeman sublevels. Magnetic resonance can also be produced by other $A$-systems (not shown in the figure) that could be formed in $F_g = 3 \rightarrow F_e = 2$ transition involving other Zeeman sublevels of $F_g$ and $F_e$ that satisfy the same condition $|\Delta m_g| = 2$. The transitions corresponding to elliptically polarized light are shown in Fig. 1(b). In this case, an additional $\pi$ transition corresponding to $\Delta m_{eg} = 0$ is introduced between the ground and excited state sublevels when light electric field vector becomes parallel to the quantization axis. This creates additional $A$-systems involving a superposition of
ground state sublevels satisfying $\Delta m_g = \pm 1$ [shown with curved arrows in Fig. 1(b)]. The strength of a $\pi$-transition increases with ellipticity and becomes equal to the $\sigma$ component at $\varepsilon = 45^\circ$.

In the synchronized optical pumping scheme, the amplitude of the laser field is modulated using a square-wave modulation function $\xi(t)$ with a duty cycle $\eta$. The laser field with amplitude modulation (AM) in the model is described as

$$\bar{E}(t) = \hat{E}_0 (1 + \Delta M \xi(t)) e^{-i\omega t} + c.c.$$

where $\Delta M$ is the modulation depth. The Fourier series expansion of the square-wave function $\xi(t)$ can be written as

$$\xi(t) = \sum_{m=1}^{\infty} g_m(\eta) \cos(m\Omega_{\text{mod}} t)$$

$$g_m(\eta) = \frac{2}{m\pi} \sin[m\pi\eta]$$

Here, the terms $g_m$ are Fourier coefficients of square-wave modulation function $\xi(t)$ and the integer index $m$ corresponds to different harmonics of $\xi(t)$. At a fixed modulation frequency $\Omega_{\text{mod}}$, the optical field consists of frequency sidebands at $\omega \pm m\Omega_{\text{mod}}$ along with a carrier at the laser frequency $\omega$. The amplitude $g_m$ of a particular sideband depends on duty cycle $\eta$ of $\xi(t)$ [26]. For example, at 50% duty cycle ($\eta = 0.5$), $g_m$ value for all even integer harmonics (even values of $m$) will be equal to zero and odd harmonics (odd values of $m$) will be nonzero. The modulated field has dominant first-order sidebands at frequencies $\omega \pm \Omega_{\text{mod}}$ for which $g_m$ value will be maximum. These sidebands in the modulated field cause synchronous pumping of the atoms to create multiple magnetic resonances at nonzero magnetic field satisfying the condition

$$\Omega_{\text{mod}} = k \Omega_L$$

where $k$ is the rank of the atomic spin polarization moment of the density matrix. These moments of the angular momentum state $F_g$ are related to coupling (or coherence) between Zeeman sublevels with $\Delta m_g \neq 0$ [19,27]. For a
given choice of the quantization axis, these coherences contribute to all atomic polarization moments with \( k \geq |\Delta m_g| \) and have a maximum possible rank \( k \) equal to \( 2F_g \). Generation and detection of multipole moments of rank \( k \leq 2 \) could be accomplished by using weak light, whereas higher rank moments \( (k > 2) \) require multiphoton interactions [27]. Our study is focused on magnetic resonances formed due to multipole moments of rank \( k \leq 2 \) at different light ellipticities. For linearly polarized light [Fig.1 (a)], the coherence condition \( \Delta m_g = k = 2 \) (quadrupole moment, also known as ‘alignment’) creates a resonance at \( \Omega_L = \Omega_{\text{mod}}/2 \). For elliptically polarized light [Fig.1 (b)], the coherence conditions \( \Delta m_g = k = 2 \) (quadrupole moment) and \( \Delta m_g = k = 1 \) (dipole moment, also known as ‘orientation’) create two resonances at \( \Omega_L = \Omega_{\text{mod}}/2 \) and \( \Omega_L = \Omega_{\text{mod}} \), respectively. For an arbitrary duty cycle \( \eta \) (\( \neq 0.5 \)), each frequency sideband corresponding to integer index \( m \) produces two resonances at \( \Omega_L = \pm m \Omega_{\text{mod}}/k \) corresponding to \( k = 2 \) and \( k = 1 \). As a general rule, when elliptically polarized light is used and magnetic field \( B_x \) is varied, resonances may occur at following frequencies:

\[
\begin{align*}
\Omega_L^{k=2} &= \pm \frac{\Omega_{\text{mod}}}{2}, \pm \Omega_{\text{mod}}, \pm \frac{3\Omega_{\text{mod}}}{2}, \pm 2\Omega_{\text{mod}} \ldots \quad (7)
\end{align*}
\]

\[
\begin{align*}
\Omega_L^{k=1} &= \pm \Omega_{\text{mod}}, \pm 2\Omega_{\text{mod}}, \pm 3\Omega_{\text{mod}}, \pm 4\Omega_{\text{mod}} \ldots
\end{align*}
\]

The resonance condition for a particular harmonic for \( k = 1 \) could also match with the one for a different harmonic for \( k = 2 \). For example, resonances for \( k = 2, m = 2 \) and \( k = 1, m = 1 \) occur simultaneously at \( \Omega_L = \pm \Omega_{\text{mod}} \). The combined resonances due to both \( k = 1 \) and \( 2 \) can be written as

\[
\begin{align*}
\Omega_L &= \pm \frac{\Omega_{\text{mod}}}{2}, \pm \Omega_{\text{mod}}, \pm \frac{3\Omega_{\text{mod}}}{2}, \pm 2\Omega_{\text{mod}}, \pm \frac{5\Omega_{\text{mod}}}{2}, \pm 3\Omega_{\text{mod}} \ldots \quad (8)
\end{align*}
\]

Here, the integer index \( n \) is used to label the position of all resonances formed by elliptically polarized light modulated with arbitrary duty cycle \( \eta \). The resonance at zero magnetic field (Hanle resonance) due to the carrier is represented by \( n = 0 \).

We calculate the magnetic resonances using the time evolution of atomic density matrix \( \rho \) given by the Liouville equation:

\[
\frac{\partial \rho}{\partial t} = \frac{i}{\hbar} [\hat{H}, \rho] - \frac{1}{2} \{ \hat{\Gamma}, \rho \} + R \quad (9)
\]

Here \( \hat{H} \) represents the total Hamiltonian of the atomic system in the rotating wave frame. It includes the internal atomic energy levels, light-atom interaction, and magnetic field-atom interaction. The relaxation matrix \( \hat{\Gamma} \) includes the spontaneous decay rate \( \Gamma \) of the excited state and the transient relaxation rate \( \gamma \) of each sublevel due to exit of atoms from the laser beam. Matrix \( R \) describes the repopulation of ground state sublevels due to decay rates \( \Gamma \) and \( \gamma \). The theoretical model is simplified by not considering the atomic motion (or velocity distribution), the effect of neighboring transitions, and the spatial distribution of laser intensity. Time-dependent density matrix equations obtained from the Eq. (9) are solved numerically to calculate the light absorption coefficient of the medium for ellipticity \( \varepsilon \) using the following expression:
\[ \alpha(t) \approx \sum_{e,g} \frac{\Gamma}{\Omega_R} \left( \beta_{e,g} \cos[\varepsilon] \Im[\rho_{e,g}(t)] + \beta'_{e,g} \sin[\varepsilon] \Re[\rho_{e,g}(t)] \right) \]  

(10)

where \( \Omega_R = \langle F_e | D | F_g \rangle E_o \) is the reduced Rabi frequency of the laser field and \( D \) is the dipole operator. The ground state and excited state sublevels involved in the optical transition decide the strength of coefficients \( \beta_{e,g} \) and \( \beta'_{e,g} \). To keep consistency with our experimental observation, we calculate the power \( P(t) \) in transmission using the following expression:

\[ P(t) = P_0 e^{-\alpha(t)L} = P_0 (1 - \alpha(t)L) \]  

(11)

Here, \( P_0 \) is the initial laser power which is set to unity to simplify the calculations. We have also assumed that \( \alpha L \ll 1 \) which is true for a thin medium. The in-phase and quadrature components of the magnetic resonances are calculated by demodulating \( \alpha(t) \) [eq. (10)] and \( P(t) \) [eq. (11)] at the first-harmonic of \( \pm \Omega_{\text{mod}} \) [28].

III. EXPERIMENTAL DESCRIPTION

A schematic diagram of the experimental setup is shown in Figure 2. A tunable external cavity diode laser with resonant wavelength 795 nm matching the \(^{85}\text{Rb} \ D_1 \) transition and linewidth less than 500 kHz is used in the experiment. Using a combination of half-wave plate \( \lambda/2 \) and a polarizing beam splitter (PBS), the laser beam is split into two paths. The reflected beam from the PBS is passed through an acoustic-optic modulator (AOM1) driven by 80 MHz radio-frequency signal of fixed amplitude. The first-order diffracted beam from the AOM1 is utilized in a saturation absorption spectroscopy (SAS) setup [29]. The laser is locked to \( F_g = 3 \rightarrow F_e = 2 \) transition of the \(^{85}\text{Rb} \ D_1 \) line using the Doppler-free absorption peak produced in the SAS setup.

Light transmitted through PBS is amplitude-modulated via AOM2 driven by 80 MHz radio-frequency signal using a rectangular pulse waveform generated through a function generator with an arbitrary duty cycle \( \eta \). The diameter of first-order diffracted beam is expanded from 2 mm to 8 mm using a couple of lenses in a telescopic configuration. The expanded beam increases the interaction time of atoms with the laser beam. Laser power to the vapor cell is controlled using a neutral density (ND) filter. Light ellipticity can be controlled from 0° to 45° using a
λ/4 plate placed in the beam path. A buffer gas free OTS-coated rubidium vapor cell is mounted at the center of a four-layer magnetic shield (MS) with a shielding factor of ~10^6. The MS contains a printed three-axis magnetic field coil installed inside its innermost layer. The coils are connected to three independent current sources to independently apply static and/or scanning magnetic field in any arbitrary direction as required in the experiment. The Rb vapor cell is kept at room temperature. Light transmitted through the cell is detected using a photodiode (PD). The OTS coating in the Rb vapor cell allows us to produce narrow linewidth magnetic resonances by reducing the wall collisions and thereby reducing the depolarization of Rb atoms [30,31]. Laser excitation of \( F_g = 3 \rightarrow F_e = 2 \) transition creates magnetic resonances with approximately 3% contrast, which are measured by demodulating the PD output using a lock-in amplifier operating at the first harmonic of the laser modulation frequency \( \Omega_{\text{mod}} \). The two channels of the lock-in amplifier allow us to measure simultaneously the in-phase (or amplitude) and quadrature (or phase) components of the magnetic resonances.

IV. RESULTS AND DISCUSSION

Figure 3 (a) shows the in-phase and quadrature components of experimentally observed magnetic resonances in the transmitted light for three different values (0°, 15° and 45°) of the light ellipticity. These measurements are performed by scanning the transverse magnetic field \( B_x \) around the zero-field. Modulation frequency of the laser is kept fixed at \( \Omega_{\text{mod}} = 3 \) kHz with duty cycle \( \eta = 0.5 \). Average intensity of the laser beam is set to 0.2 mW/cm^2. Average laser intensity in the experiment is set to 0.2 mW/cm^2. Parameters used in simulations: \( \Omega_R = 0.01 \Gamma, \gamma = 3 \times 10^5 \Gamma \). Plots are normalized with respect to the maximum amplitude in the respective signal. All the resonances are labelled using the integer index \( n \) described in section II.

![FIG. 3. Experimentally measured (a) and theoretically calculated (b) magnetic resonances using modulated light (\( \eta = 0.5 \) and \( \Omega_{\text{mod}} = 3 \) kHz) with different light ellipticity values in each row. Average laser intensity in the experiment is set to 0.2 mW/cm^2. Parameters used in simulations: \( \Omega_R = 0.01 \Gamma, \gamma = 3 \times 10^5 \Gamma \). Plots are normalized with respect to the maximum amplitude in the respective signal. All the resonances are labelled using the integer index \( n \) described in section II.](image)
\( n = \pm 1 \) resonances at non-zero magnetic field. Alternatively, \( n = \pm 1 \) ‘alignment’ resonances can be described as being produced by synchronous pumping of atoms with modulated light at Larmor frequency with atomic polarization moment \( k = 2 \). Since the magnetic field direction is transverse to the light propagation direction, resonances labeled as \( n = 0 \) and \( \pm 1 \) cannot be observed in polarization rotation measured using a balanced polarimeter setup, i.e. the amplitude of polarization rotation decreases as the cosine of angle between the light propagation direction and the magnetic field [32].

As light ellipticity is increased from zero, the \( \pi \) and \( \sigma^\pm \) components tend to form additional \( \Lambda \)-type systems satisfying the condition \( |\Delta m_s| = 1 \) between the ground state Zeeman sublevels. Therefore, for non-zero ellipticity, both \( |\Delta m_s| = 1 \) and \( |\Delta m_s| = 2 \) coherence conditions will produce resonances at zero magnetic field. Due to the presence of sidebands, coherence condition \( |\Delta m_s| = 1 \) creates ‘orientation’ resonances at \( \Omega_s = \pm \Omega_{\text{mod}} \) labeled as \( n = \pm 2 \) with polarization moment \( k = 1 \) [Fig. 3 (a) (middle row)]. At light ellipticity \( \varepsilon = 15^\circ \), the amplitudes of \( n = \pm 1 \) and \( n = \pm 2 \) resonances are approximately equal in both in-phase and quadrature signals. When ellipticity is further increased, the amplitudes of \( n = \pm 1 \) resonances decrease and nearly vanish at \( \varepsilon = 45^\circ \) corresponding to circularly polarized light [Fig. 3 (a) (bottom row)]. On the other hand, the amplitudes of \( n = \pm 2 \) resonances increase with increase in ellipticity and become maximum at \( \varepsilon = 45^\circ \). This behavior can be inferred from the increase in strength of the \( \pi \)-transition responsible for \( |\Delta m_s| = 1 \) coherence with increase in ellipticity. Theoretical results shown in Fig. 3(b) show a good agreement with our experimental observations shown in Fig. 3 (a). However, we observed a difference between theory and experiment in terms of the relative amplitudes of resonances for each ellipticity case. For example, unlike the experimental results shown in Fig. 3 (a) (middle row), the calculated resonances \( n = \pm 1 \) and \( n = \pm 2 \) in Fig. 3 (b) (middle row) do not have equal amplitudes for ellipticity \( \varepsilon = 15^\circ \). This discrepancy could have resulted from the simplification of our theoretical model discussed in section II.

Figure 4(a) shows experimentally measured peak amplitudes of \( n = 0, 1 \) and \( 2 \) resonances in the in-phase signal as a function of light ellipticity \( \varepsilon \) for duty cycle \( \eta = 0.5 \). Since zero-field \( n = 0 \) resonance has contributions from many degenerate ground state superpositions of sublevels satisfying conditions \( |\Delta m_s| = 1 \) and/or \( |\Delta m_s| = 2 \), it has much higher amplitude compared to \( n = 1 \) and \( n = 2 \) resonances. Increase in the amplitude of \( n = 0 \) resonance with ellipticity, is in agreement with previous reports where continuous laser excitation was utilized [33,34]. Since the strength of \( |\Delta m_s| = 2 \) coherence weakens with an increase in ellipticity from \( 0^\circ \) to \( 45^\circ \), the amplitude of \( n = 1 \)
resonance consequently diminishes, as shown in Fig. 4(a). Figure 4(b) shows the plots of full-width half maximum (FWHM) of $n = 0$, 1 and 2 resonances as a function of light ellipticity. The width of a resonance depends on the dephasing between the ground state sublevels, and on the Rabi frequencies (or matrix elements/coupling strengths) of the optical transitions involved in a particular $\Lambda$-system. Closer to zero ellipticity, $n = 0$ and $n = 1$ resonances have approximately equal FWHMs. The width of $n = 0$ resonance is increased by nearly 60% from 0.35 mG to 0.56 mG when $\varepsilon$ is changed from 5° to 25° and becomes approximately constant thereafter for higher ellipticities.

The other hand, the width of $n = 1$ resonance does not vary much with ellipticity. The $n = 2$ resonance shows a broader linewidth compared to $n = 0$ and $n = 1$ resonances for all ellipticities. Theoretical results shown in Fig. 4(c) and Fig. 4(d) are consistent with the corresponding experimental results shown in Fig. 4(a) and Fig. 4(b), respectively. The amplitudes and widths of $n = 1$ and $n = 2$ resonances were also measured from the quadrature signals, and found to exhibit similar dependencies on light ellipticity, as the ones measured from in-phase signals.

Next, we describe the effect of light ellipticity on the magnetic resonances for a lower duty cycle of light modulation $\eta = 0.3$ and by keeping the average laser intensity fixed at 0.2 mW/cm$^2$. In Fig. 5(a), higher-order magnetic resonances (e.g. $n = \pm 4$) are observed due to the presence of all sidebands in the modulated light. For $\varepsilon = 0°$, the in-phase signal shows resonances up to second-order (i.e. $n = \pm 2$) and the quadrature signal which is phase-sensitive, shows resonances up to the fourth-order (i.e. $n = \pm 4$) satisfying the resonance condition of $\Omega / \Omega_{mod} = m$ with $m = 4$ (i.e. fourth sideband of modulated light) and $k = 2$. The quadrature signal shows a dispersive line shape with same sign for the first three-orders and an opposite sign for the fourth-order (i.e. $n = \pm 4$) resonances [Fig. 5(a) (top row)] indicating a 180° phase change, possibly due to a sign reversal of the Fourier coefficient $g_k$.

As the light ellipticity is changed from zero, resonances up to eighth-order (i.e. $n = \pm 8$) are observed [Fig. 5(a)]. As discussed in Section II, the $k = 1$ resonance (for which $\Omega / \Omega_{mod}$ ) and the $k = 2$ resonance (for which $\Omega / \Omega_{mod}/2$ ) are produced simultaneously due to nonzero light ellipticity. The $\Omega / \Omega_{mod}$ values for these $k$'s are satisfied by two different $m$ values that correspond to two different sidebands of the modulated light. The

**FIG. 5.** Experimentally measured (a) and theoretical calculated (b) magnetic resonances for different light ellipticities (as labelled) using modulated light with $\eta = 0.3$ and $\Omega_{mod} = 3$ kHz. Other parameters used in simulations remain the same as in Fig. 3. Plots are normalized with respect to the maximum amplitude in respective signal. All the resonances are labelled using the integer index $n$ described in section II.
dominance of a particular side-band to form this type of higher-order resonance is decided by the strength of ground state coherence $|\Delta m_g|$. For example, $n = 4$ resonance at $\Omega_r = 2\Omega_{\text{mod}}$ is produced by the fourth sideband ($m = 4$) with polarization moment $k = |\Delta m_g| = 2$ and also, by the second sideband ($m = 2$) with polarization moment $k = |\Delta m_g| = 1$. For a non-zero light ellipticity $[\text{Fig. 5a (middle and bottom row)}]$, $n = 4$ dispersive resonance in the quadrature signal switches its sign with respect to $n = 4$ dispersive resonance for the zero light ellipticity case $[\text{Fig. 5a (top row)}]$. The switching of sign in $n = 4$ dispersive resonance with ellipticity is due to the dominance of the participating sideband from $m = 4$ to $m = 2$ (no sign reversal in $g_x$) satisfying the coherence condition $|\Delta m_g| = 1$ for $k = 1$. Similarly, the inter-sign reversal of $n = 8$ dispersive resonance with respect to $n = 4$ dispersive resonance at ellipticity $\varepsilon = 15^\circ$ or $45^\circ$ can be explained by the fact that it is predominantly formed by the strong fourth sideband (i.e. $m = 4$ with a sign reversal in $g_x$) of the modulated light. Fig. 5(b) shows corresponding theoretical results obtained for the same light ellipticities as in Fig. 5(a). The theoretical results reproduce most of the salient features observed experimentally in Fig. 5(a). The sign-changes of $n = 4$ and $8$ resonances in quadrature signals discussed above, are also clearly observed in the simulated results shown in Fig. 5(b).

![Image](attachment:figure6.png)

**FIG. 6.** Variation of experimentally measured (a) and theoretically calculated (b) difference $D$ in amplitudes of $n = 1$ and $n = 2$ resonances as a function of light ellipticity for different values of $\eta$. Other parameters used in the simulation are same as those described in Fig. 3.

Next, we measured the peak amplitudes of $n = 1$ and $n = 2$ resonances as a function of the light ellipticity for arbitrary duty cycle $\eta$ of the modulated light. The amplitudes of these resonances are found to vary at different rates with $\eta$. To study this dependence, we measured the amplitude difference ($D$) of $n = 1$ and $n = 2$ resonances as a function of the light ellipticity for different values of $\eta$. This is shown in Fig. 6(a). For $\eta = 0.5$, the $D$ value varies from positive to negative and passes through a zero-crossing around $\varepsilon = 15^\circ$ where $n = 1$ and $n = 2$ resonance amplitudes are equal. This is consistent with our results shown in Fig. 4(a). The zero-crossing is shifted towards a lower ellipticity $[\text{i.e. } \varepsilon = 11.7^\circ]$ with a decrease in $\eta$ from 0.5 to 0.3 $[\text{Fig. 6(a)}]$. The plots in Fig. 6(a) show three different duty cycles $\eta$ for which the difference $D$ varies quite linearly over ellipticity $\varepsilon$ ranging from $10^\circ$ to $30^\circ$. The red lines show linear fittings to the experimental data from which the slope $\partial D / \partial \varepsilon$ is calculated. The slope $|\partial D / \partial \varepsilon|$ increases from 4.1 mV/deg to 7.7 mV/deg by changing $\eta$ from 0.5 to 0.3. This slope can be used in applications that require in situ measurement of the light ellipticity. A higher slope will yield higher accuracy in $\varepsilon$ measurement, and the duty cycle $\eta$ can be used to adjust the slope. Figure 6(b) gives theoretical plots showing variations in $D$ with ellipticity for the same $\eta$ values. Theoretical results show a good agreement with the experiment, particularly, in reflecting the increase in slope $|\partial D / \partial \varepsilon|$ with lowering of duty cycle $\eta$. However, the zero-crossing points for theoretical plots in Fig. 6(b) do not match with those in Fig. 6(a) due to the simplification of our theoretical model.

So far, we have discussed the effect of light ellipticity on the higher-order magnetic resonances formed by a transverse magnetic field along the x-axis. Next, we discuss the dependence of these resonance amplitudes on the direction of the magnetic field. For this study, we kept the amplitude of total magnetic field fixed at $B = 5.1$ mG (i.e. $\Omega_L = 2.4$ kHz) and measured $n = 1$ and $n = 2$ resonances by changing the magnetic field direction. In this case, magnetic resonances are observed by scanning the modulation frequency $\Omega_{\text{mod}}$ of light from 1.5 kHz to 6 kHz.
Average laser intensity is kept fixed at 0.2 mW/cm² and the duty cycle \( \eta \) of light modulation is set at 50% (i.e. \( \eta = 0.5 \)). Linearly polarized light is used in this study. The orientation of the polarization vector \( \mathbf{E} \) with respect to the magnetic field \( \mathbf{B} \) is varied to induce coherence \( |\Delta m_\| \) for producing \( n = 1 \) and \( n = 2 \) resonances. Figure 7(a) shows the geometrical representation of the polarization vector \( \mathbf{E} \) and the magnetic field \( \mathbf{B} \) in a three-dimensional coordinate system. As shown, the polarization vector makes an angle \( \phi \) with the \( y \)-axis in the \( x-y \) plane. The direction of the magnetic field \( \mathbf{B} \) is defined by angles \( \theta \) and \( \psi \) in the spherical coordinate system, where \( \psi \) is the azimuthal angle and \( \theta \) is the polar angle. To illustrate the concept, we set the azimuthal angle of the magnetic field \( \psi = 30^\circ \) for all our measurements. For a given angle \( \theta \) of the \( \mathbf{B} \) field, the peak amplitudes of resonances at \( \Omega_{\text{mod}} = \Omega_L = 2.4 \) kHz (i.e. \( n = 2 \)) and \( \Omega_{\text{mod}} = 2\Omega_L = 4.8 \) kHz (i.e. \( n = 1 \)) are measured by changing the rotation angle \( \phi \) of the polarization vector \( \mathbf{E} \). The amplitudes of \( 2\Omega_L \) and \( \Omega_L \) resonances depend on the strength of \( \sigma \) and \( \pi \) transitions, which are determined by an angle between polarization vector and orientation of the magnetic field (i.e. axis of quantization), as discussed in section II.

Figure 7(b) shows the amplitudes of \( 2\Omega_L \) and \( \Omega_L \) resonances as a function of the angle \( \phi \) corresponding to three different values of \( \theta \). For \( \theta = 90^\circ \), the magnetic field \( \mathbf{B} \) is oriented in the \( x-y \) plane i.e. in the plane of light polarization [Fig. 7(a)]. When the polarization angle \( \phi = 30^\circ \), the electric field vector \( \mathbf{E} \) becomes parallel to \( \mathbf{B} \) resulting in a \( \pi \) only transition between the Zeeman sublevels of ground and excited states. In this case, due to the absence of \( \sigma \) transition, no coupling (or coherence) can be established between the ground state sublevels. Thus, amplitudes of both \( 2\Omega_L \) and \( \Omega_L \) resonances become zero at the polarization angle \( \phi = 30^\circ \). This can be seen in the plots shown in the top row of Fig. 7(b). The amplitude of \( 2\Omega_L \) resonance shows a plateau for polarization angle \( \phi \) between 10° and 30°.
60°. This is because it is only formed by σ transitions which remain weak over this range of angle. On the other hand, the amplitude of Ω₂ resonance (which is formed by both σ and π transitions) changes rapidly and reaches its maximum value at angle φ=75°, where the strengths of σ and π components become equal as the angle difference (φ – ψ) = 45°. When the polarization angle φ is changed further beyond 75°, the angle difference (φ – ψ) is increased beyond 45° resulting in a stronger σ transition. At φ =120° where (φ – ψ) = 90°, the light will only have σ components, which results in a maximum amplitude of 2Ω₂ resonance and minimum amplitude of Ω₃ resonance [Fig. 7(b) (top row)]. The amplitude of 2Ω₂ resonance shows the next plateau around φ =210° due to a resulting π only transition. Thus, the amplitude of 2Ω₂ resonance oscillates slowly with a periodicity of 180° in the polarization angle φ. On the other hand, the amplitude of Ω₃ resonance oscillates faster with a periodicity of 90° in angle φ for the case θ = 90°.

Next, we considered a tilt in the magnetic field direction (keeping its strength fixed) from the x-y plane by defining the angle θ to be less than 90° [Fig. 7(a)]. In this case, rotation of polarization angle φ cannot make the electric field vector E parallel to the magnetic field B to produce a π only transition. Therefore, unlike Fig. 7(b) (top row), polarization angles φ =30° and φ =210° will produce nonzero amplitudes in both 2Ω₂ and Ω₃ resonances for θ < 90° as shown in Fig. 7(b) (middle & bottom row). The oscillating amplitudes of 2Ω₂ and Ω₃ resonances with polarization angle φ get smaller by tilting the magnetic field B away from the x-y plane [Fig. 7(b) (middle and bottom rows)]. Also for θ < 90°, the oscillation frequency Ω₃ resonance become equal to the frequency of 2Ω₂ resonance (i.e. periodicity in φ = 180°). Experimental (dots) and theoretical (solid lines) results in Fig. 7(b) show very good agreement. When angle θ = 0°, the magnetic field B is along the z-axis (i.e. longitudinal), which is perpendicular to E for any choice of polarization angle φ. In this case, the amplitude of 2Ω₂ resonance does not depend on φ, hence, shows no oscillation. On the other hand, Ω₃ resonance at θ = 0° completely vanishes due to the absence of π transition. The oscillatory behavior showed here in Fig. 7 are different from the ones observed in Ref. [12] using a second-harmonic i.e. 2Ω₃ detection scheme.

Figure 7(c) shows oscillation amplitudes of 2Ω₂ and Ω₃ resonances as a function of the magnetic field tilt angle θ. The oscillations show a strong linear dependence over a range of tilt angle θ from 15° to 40°. Figure 7(c) can be used as a calibration curve for determining angle θ of the magnetic field, whereas locations of maxima and minima in 2Ω₂ and Ω₃ oscillations in Fig. 7(b) can used to find angle φ of the magnetic field. Measurements using both 2Ω₂ and Ω₃ resonances can improve accuracy of the synchronous optical pumping vector magnetometer. Similarly, the ratio of relative strengths between 2Ω₂ and Ω₃ resonances can be used as a response for avoiding the commonly encountered ‘dead-zone’ problem in the Bell-Bloom magnetometer [25].

V. CONCLUSIONS

We have investigated magnetic resonances at nonzero magnetic field created by synchronous optical pumping of the atoms using an OTS coated rubidium vapor cell. The effect of incident light ellipticity on the resonance spectrum is studied in the presence of magnetic field oriented perpendicular to the light propagation direction. Our study showed ground state coherences responsible for producing two types of magnetic resonances, strongly depend on the light ellipticity. Resonance (Ω₂ = ±Ω₃) satisfying the coherence condition |Δn| = 1 becomes stronger with increase in light ellipticity, whereas resonance (Ω₂ = ±Ω₃/2 ) satisfying the coherence condition |Δn| = 2 diminishes at higher ellipticity. We showed that the difference in amplitudes of Ω₂ =Ω₃ and Ω₂ = Ω₃/2 resonances varies linearly with ellipticity between 10° and 30°, which can be used for in situ measurement of light ellipticity. For non-zero light ellipticity, we also reported sign reversal in fourth-order dispersive resonance at 30% light duty cycle. We studied the dependence of 2Ω₂ and Ω₂ resonance amplitudes on the magnetic field direction using polarization rotation. The amplitudes of 2Ω₂ and Ω₂ resonances showed periodic oscillations with polarization rotation. These oscillations are found to be sensitive to the orientation of the magnetic field with respect to plane of polarization. This aspect can be utilized in developing a synchronous optical pumping vector magnetometer.
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