Modulation induced splitting of the magnetic resonance

Rita Behera\textsuperscript{1,2} and Swarupananda Pradhan\textsuperscript{1,2,3} \email{spradhan@barc.gov.in and pradhans75@gmail.com}

\textsuperscript{1} Laser and Plasma Technology division, Bhabha Atomic Research Centre, Trombay, Mumbai-400085, India
\textsuperscript{2} Department of Atomic Energy, Homi Bhabha National Institute, Mumbai-400094, India

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Abstract

The splitting of magnetic resonance induced by a frequency modulated linearly polarized light field is presented for Hanle experimental configuration. The experiment is carried out with thermal Rubidium atoms in an anti-relaxation coated glass cell. The observed splitting corresponds to steady-state behaviour of the system over an integrated time scale that is longer than the modulation frequency. The splitting is only observed for dark resonances, with left and right circularly polarized light simultaneously coupled to the atomic system. The split components are separated by half of the modulation frequency with the central component lying at zero magnetic field. The absence of odd split components at single photon resonance is consistent with the theoretical calculation. The different components of the split profile have distinct response to the direction of the transverse magnetic field. The response is captured by the theoretical calculation and the underlying physical mechanism is discussed. These studies will be useful in metrology and advancement of vector atomic magnetometer.

Keywords: magnetic resonance, quantum interference, frequency modulation, transverse magnetic field, frequency comb, single photon resonance

(Some figures may appear in colour only in the online journal)

1. Introduction

The ubiquitous magnetic field carries vital information on the associated physical processes and thus, its measurement is an important part of the contemporary science [1–5]. Apart from fundamental interest, the prospect of highly sensitive magnetometers in a variety of applications has led to exceptional growth in the associated research area [6–8]. The intriguing aspect of atomic interaction with photon gets augmented near zero magnetic field due to interplay of quantum interference, optical pumping and level crossing assisted population redistribution. The semi-classical density-matrix based calculation is a prominent tool for extracting the underlying physical mechanism behind the observations [1, 9–19]. In general, a comprehensive knowledge of the laser atom interaction process is instrumental for the advancement of modern science and atomic devices. The zero-field magnetic resonance, realized through Hanle configuration is central to many of the leading magnetometry technique. The Hanle kind of magnetic resonance in an atomic system is realized by scanning the magnetic field along the laser propagation direction using resonant as well as off resonant (few GHz away from the atomic resonance) light field.

One variant of Hanle method uses modulated light field for study of magnetic resonance [9–14]. It incorporates non-linear magneto-optic rotation for study of atomic spin dynamics near zero magnetic field. The modulation applied to either laser amplitude, frequency or polarization leads to synchronous oscillation of the atomic polarization at the modulated frequency. The oscillating atomic polarization is phase sensitively detected, mostly using a polarimetric set-up. The experimental procedure extracts the oscillation at different harmonics of the modulating frequency by using lock-in amplifier. The amplitude of these oscillations shows resonances as the
Larmor’s frequency matches with the harmonics of the applied modulation. Resultantly, there is an apparent splitting in the Hanle kind of resonance. However, it may be noted that the process of signal acquisition is different from the conventional Hanle technique. The above configuration is labelled as FM method in the subsequent discussion.

We have observed a series of magnetic resonances resembling to a split profile due to the frequency modulated light field, while utilizing a different experimental approach. This method closely resembles with the Hanle configuration and excerpts a different physical attribute [from the frequency modulation (FM) method] of the phenomenon. Similar to the FM method, a frequency modulation ($f_m$) is applied to the light field. However, an additional low frequency modulation ($m_m$) is applied to the magnetic field along the laser propagation direction (longitudinal magnetic field). The transmitted light by the atomic sample is detected in reference with the $m_m$ by using a lock-in amplifier. Since the frequency of $m_m$ is much slower than $f_m$, the oscillations at the frequency of $f_m$ do not contribute to the acquired signal. The signal represents a steady state attribute of the system and is studied as a function of the longitudinal magnetic field. This experimental configuration is termed as magnetometric (MM) method.

In summary, the FM method addresses the oscillation in the transmitted light at different harmonics of $f_m$, where as the integrated transmitted light intensity is studied in the MM method. These two methods represent two different physical scenarios albeit both originates due to the modulation in the light field and studied as a function of the magnetic field.

The parametric dependence of the signal by MM method is studied with respect to the laser polarization, laser detuning, and transverse magnetic field. To the best of our knowledge, these dependences are not addressed in the FM method also. The split profiles in MM method have shown strong dependence to the coherent population trapping (CPT) signal, generated between the two lower hyperfine levels [20, 23, 24]. A solenoidal coil and two set of rectangular coils are used for controlling the magnetic field along and transverse to the laser propagation direction respectively. The utilized electromagnets are spectrscopically calibrated with respect to the coherent population trapping (CPT) signal, generated between the two lower hyperfine levels [20, 23, 24]. All the quoted magnetic field ($B_x$, $B_y$, $B_z$) in this article are with reference to the zero-magnetic field derived from the splitting of the CPT resonance. The ambient magnetic field is shielded by enclosing the vapor cell along with the solenoidal coil in four layers of Mu-metal sheets. The innermost shield has a diameter of 25 mm and length of 50 mm. It is coated with octadecyltrichlorosilane for anti-relaxation (AR) that is expected to provide a coherence relaxation time close to ~50 ms [19, 21, 22]. A solenoidal coil and two set of rectangular coils are used for controlling the magnetic field along and transverse to the laser propagation direction respectively. The utilized electromagnets are spectrscopically calibrated with respect to the coherent population trapping (CPT) signal, generated between the two lower hyperfine levels [20, 23, 24]. All the quoted magnetic field ($B_x$, $B_y$, $B_z$) in this article are with reference to the zero-magnetic field derived from the splitting of the CPT resonance.

The transition identification is difficult owing to the associated Doppler broadening and the quoted values of $\Delta$ is only indicative.

The linearly polarized transmitted beam by the PBS, with polarization axis in the $x$-direction is used for the experiment. A quarter wave plate (QWP) is used after the PBS to change the polarization state of the light field for study related to the laser polarization dependence of the signal. The beam is passed through an atomic cell containing Rb atoms of natural isotopic composition at room temperature (Rb2). The atomic cell has a diameter of 25 mm and length of 50 mm. It is coated with octadecyltrichlorosilane for anti-relaxation (AR) that is expected to provide a coherence relaxation time close to ~50 ms [19, 21, 22]. A solenoidal coil and two set of rectangular coils are used for controlling the magnetic field along and transverse to the laser propagation direction respectively. The utilized electromagnets are spectrscopically calibrated with respect to the coherent population trapping (CPT) signal, generated between the two lower hyperfine levels [20, 23, 24]. All the quoted magnetic field ($B_x$, $B_y$, $B_z$) in this article are with reference to the zero-magnetic field derived from the splitting of the CPT resonance. The ambient magnetic field is shielded by enclosing the vapor cell along with the solenoidal coil in four layers of Mu-metal sheets. The innermost shield has a diameter of 90 mm to accommodate a notch structure of the atomic cell. The magnetic shields are made from a sheet of 0.35 mm thickness except 1 mm thickness sheet for the outermost shield.

The scanning of the longitudinal ($B_z$) field is done by a ramp generator (scanner). An oscillating field generated by an oscillator (oscillator2) is used for modulating the $B_z$ field as shown in Figure 1. A part of the frequency modulated laser beam after passing through a Rb vapour cell (Rb1), is phase sensitively detected with respect to the modulation applied to the laser field and is used for laser frequency stabilization. The other part of the beam after interaction with Rb atoms in an AR coated vapour cell (Rb2), is phase sensitively detected with respect to modulation applied to the $B_z$ field for study of magnetic resonances. The laser polarization axis is along $x$-direction.

2. Experimental set-up

The experiment is carried out with a vertical cavity surface emitting diode laser (VCSEL) tuned to the rubidium (Rb) D1 line at 795 nm. The laser beam has $\sim$100 $\mu$W power with a knife edge width of $\sim$4.5 mm, and line width <100 MHz. The laser frequency is modulated at a frequency $\omega_m$ with an amplitude $A_m$ (12 kHz and $\sim$1.63 GHz respectively, unless specified). The primary objective of this modulation is to generate a frequency comb for study of its effect on the Hanle resonance. It is also used for generating an error signal for laser frequency stabilization.

The schematic diagram of the experimental set-up is shown in figure 1. A part of the laser beam ($\sim$10%) is split by the use of a half-wave plate and a polarization beam splitter cube (PBS). This beam (in the reflected port of the PBS) is passed through a Rb vapour cell and detected by a photodiode (PD1). An oscillator (oscillator1) with a frequency $\omega_m$ is used to modulate the injection current of the VCSEL. The PD1 signal is phase sensitively detected by a lock-in amplifier (lock-in amp1) in reference with $\omega_m$ to generate an error signal for laser frequency stabilization [8, 14, 20]. A servo lock loop modifies the laser frequency to the zero crossing of the error signal at Rb-85 $F = 3 \rightarrow F' = 2, 3$ transition (unless specified) with a stability of $\sim<$50 MHz. The lock position corresponds to a frequency detuning ($\Delta$) $\sim$+25 × $\Gamma$ from the Rb85 $F = 3 \rightarrow F' = 2$ transition, where $\Gamma$ ($\sim$5.6 MHz) is the natural line width. The transition identification is difficult owing to the associated Doppler broadening and the quoted values of $\Delta$ is only indicative.

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in figure 1. The transmitted light through rubidium cell (Rb2) is phase sensitively detected using a lock-in amplifier (lock-in amp2) in reference with the oscillator2 and is termed as MM signal. The oscillating magnetic field has a frequency of 55 Hz with an amplitude $\sim 200$ nT. The frequency of modulation applied to the magnetic field is kept slower than $\omega_m$ to isolate the MM signal from the oscillation at $\omega_m$ and its harmonics present in the amplitude of the light field. This is in contrast to the signal in the FM method that exclusively represents the oscillation at $\omega_m$ and its harmonics [9–14]. Thus, the MM signals presented in this work are derived using a different technical approach as compared to all prior investigations. The MM method has a technical advantage compared to the conventional FM method as the contribution of the residual amplitude modulation to the signal profile is inherently circumvented.

3. Theoretical outline

The objective of the theoretical calculation is to capture the observed splitting of the magnetic resonance and their response to the transverse magnetic field. The calculation is carried out by using some of the basic blocks available in the ‘atomic density matrix’ package [1, 17, 18, 25]. The density matrix formalism for calculating the response of an atomic ensemble near zero magnetic field, while interacting with a light field has been described by several group [1, 9–13, 15–18]. The model along with associated interaction, relaxation and procedure for calculation (in consistency with the MM Method) are briefly discussed for completeness. The level diagram of an atomic transition ($J = 1 \rightarrow J' = 0$) coupled with a linearly polarized light field for different experimental condition is shown in figures 2(A)–(C). As can be seen, the simultaneous couplings of different side modes varies with the experimental condition and are used for interpreting the experimental observation in the later part of this article. This is an ideal model for study of the physical processes associated with the atomic system coupled with a frequency modulated light field. The $m_f = 0$ ground state is not coupled to any light field and mimics the uncoupled ground hyperfine level in the realistic alkali atoms. A representative diagram of the frequency comb with the spectral profile of the atomic transition is shown in figure 2(D). The total Hamiltonian corresponding to the interaction of a light field ($\mathcal{E}$) with the atom in presence of a magnetic field ($\mathbf{B}$) is given by $\hat{H} = \hat{H}_0 - \hat{d} \cdot \mathcal{E} + \hat{\mu} \cdot \mathbf{B}$.

The input light field propagating along z axis $(E_0 (\cos \epsilon x + i \sin \epsilon y) \cos \omega t)$ is frequency modulated by a sinusoidal wave $A_m \cos (\omega_m t)$, where $\epsilon$, $A_m$, and $\omega_m$ are ellipticity, depth of modulation, and modulation frequency respectively. The resultant frequency modulated field (taking care of the accumulated phase) can be represented as $E_0 (\cos \epsilon x + i \sin \epsilon y) \cos \left(\omega t + \frac{\Delta \omega_m t}{\omega_m} \right)$, where $\Delta \omega_m t/\omega_m$ represents $\Gamma/3$ and $\Delta\omega_m t/(\omega_m\gamma\rho)$ is the quantum jump operator. The collisional relaxation among all the ground Zeeman states is considered with equal probability. The relaxation matrix (R) and repopulation matrix ($\Lambda$) associated with the ground state coherence relaxation rate ($\gamma_c$) are given by $\hat{L}_\rho = -\frac{1}{2} [R, \rho] + \Lambda$. The evolution of the density matrix for this system is obtained by solving

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the Lindblad master equation $\frac{d\rho}{dt} = -\frac{i}{\hbar}[H, \rho] + L_d\rho + L_t\rho$. The utilized notations in the above description have their customary meaning.

The influence of transverse magnetic field to the atomic dynamics can be incorporated by transforming the electric field vector (of the light field) along the direction of magnetic field [26, 27]. However, coupling between the Zeeman ground state by the transverse field can be equally used. This approach preserves the quantization axis along the laser propagation direction [18, 25]. We have utilized the later approach for calculation of the signal profile. The selection rule $\Delta m_f = \pm 1$, for magnetic coupling between ground level Zeeman states is used. The coupling strength is decided by the amplitude of corresponding magnetic field.

The amplitude of the temporal oscillation of density matrix element, at different harmonics of corresponding magnetic field. The laser frequency is locked at $\Delta \sim +25 \Gamma$ with respect to Rb85 $F = 3 \rightarrow F' = 2$ transition.

The various experimental parameters like laser polarization, laser intensity, magnetic field direction and amplitude, atomic properties, and inhabitant composition in the cell dictate the relative domination of these effects [8–29]. The signal acquired through the MM method with linearly polarized light shows splitting of the Hanle kind of resonance ($\Delta = +25 \times \Gamma$ from Rb85 $F = 3 \rightarrow F' = 2$ transition), as shown in figure 3. The dispersive signal profile is due to the use of phase sensitive detection technique. The positive or negative slope of the signal at the line centre corresponds to enhanced transmission or absorption respectively. The role of $B_z$ field modulation on the signal profile is examined by changing the frequency and amplitude of the magnetic modulation form 13 Hz to 79 Hz and 10 nT to 400 nT respectively. The features of the MM$_c$ signal profile remained intact except for the changes in the amplitude and associated noise level. It is verified that magnetic field modulation neither introduces any extra broadening nor alters the MM$_c$ signal profile. The $B_z$ field separation between the neighbouring split components is found to be $\sim 1108 nT$ ($\omega_{\mu 01}$) for $\omega_{\mu} = 12 kHz$, as shown in figure 2. The gyromagnetic ratio for Rb-85 ground level is $\sim 4.7 Hz nT^{-1}$ that gives the theoretical value of $\omega_{\mu}/2$ to be $\sim 1276 nT$ [30]. The close value of the $\omega_{\mu 01}$ with the theoretical value for $\omega_{\mu}/2$ indicates that the resonance occurs as the Larmor’s frequency approaches a harmonic of $\omega_{\mu}/2$. Similar splitting is also observed for laser locked at a detuned position from the Rb87 $F = 2 \rightarrow F' = 1$ transition. The gyromagnetic ratio for Rb-87 ground level is 7 Hz nT$^{-1}$ that gives the theoretical value of $\omega_{\mu}/2$ (for $\omega_{\mu} = 12 kHz$) to be $\sim 857 nT$. The measured separation between nearby split component is $\sim 748 nT$ ($\omega_{\mu 02}$) that is also close to the corresponding theoretical value. The magnetic field values presented in this article are derived from the calibrated coils with respect to the CPT signal between the two lower ground levels. The reference CPT signals are associated with systematic shift for circularly polarized light field [20, 23, 24].
Further, the accuracy in the amplitude of the bias voltage (used for calibrating the coils) contributes to the error in the calibration. The separation between the split components in figure 3 provides a better way for calibration of the longitudinal magnetic field. It will circumvent the above sources of error as linearly polarized light is used and the separation between the split components depends on the frequency of modulation (in the light field). The frequency of a voltage source is a better standard compared to its amplitude. However, it is not used in the current article.

The observed splitting in the MM method is due to enhanced macroscopic atomic polarization (steady state) as Larmor’s frequency became resonant with any harmonics of \( \omega_m/2 \). There is a resemblance with the FM method, where the oscillations (at the harmonics of \( \omega_m/2 \)) of the atomic polarization are resonantly enhanced under identical condition. The splitting is predominantly observed for linearly polarized light and its polarization dependence provides information on the underlying mechanism. A single enhanced absorption profile (without splitting) is observed for either left or right circularly polarized light as shown in figure 3. In the current experimental conditions, it is impossible to realize a common level excitation for a pure circularly polarized light field. The Zeeman redistribution (subsequent to the optical pumping) has been envisaged as a mechanism for the observed enhanced absorption of circularly polarized light in absence of orthogonal magnetic field in prior-art [24]. However, role of Zeeman redistribution due to residual orthogonal magnetic field cannot be ruled out in the current experimental set-up. It reveals that the signal originating due to optical pumping followed by Zeeman redistribution does not show any split profile in MM method. This is in contrast to FM method, where splitting is observed for circularly polarized light.

For the linearly polarized light field, quantum interference plays role in the atomic dynamics near zero magnetic field as shown in figure 2. The dark and bright resonances (enhanced transmission and absorption) are generally associated with \( F_g \rightarrow F_e \leq F_g \) and \( F_g \rightarrow F_e > F_g \) class of transition respectively [16, 18, 20, 26, 27]. In the MM method with linearly polarized light, enhanced transmission is observed for all Rb85 and Rb87 D1 transition with \( F_e \leq F_g \) except for Rb85 \( F = 2 \rightarrow F' = 2 \). The possible error in the resonance position of Rb85 \( F = 2 \rightarrow F' = 2 \) transition is overruled by acquiring the signal in several detuned positions, where all the profile showed enhanced absorption. The contradiction is due its spectral overlap with the Rb85 \( F = 2 \rightarrow F' = 3 \) transition that exhibits bright resonance. The dark resonances are found to be transformed to bright resonance as the laser polarization is changed from linear to circular (figure 3). The relative strength (with respect to signal for linearly polarized light shown in figure 3) of the signal for various transitions is shown in table 1. The positive and negative polarity of the amplitudes indicates enhanced transmission and absorption respectively. 

None of the bright resonances \( (F_g \rightarrow F_e > F_g) \) of Rb85 and Rb87 D1 transition showed splitting of the magnetic resonance irrespective of the laser polarization. Similarly, splitting is not observed for purely circularly polarized light as all the transitions shows enhanced absorption. Thus, the observation of splitting in the MM method is facilitated by simultaneous coupling of both left \((\sigma-\)\) and right \((\sigma+)\) circularly polarized light to a dark resonance. We could not observe similar splitting of the Hanle resonance in buffer gas (@25 Torr nitrogen gas) environment. This is due to very large homogeneous (collisional) broadening (~few GHz) that leads to spectral overlap among the transitions. It is consistent with the above discussed anomaly for the Rb-85 \( F = 2 \rightarrow F' = 2 \) transition where splitting is not observed. The increase in the homogeneous width also broadens the split components as discussed in the theoretical section and obscures the observation of the splitting. Thus, AR coated cell is not only required for improving the Zeeman coherence time but also an ideal system for observation of the split profile in the MM Method.

The modulation of the VCSEL injection current by \( A_m \cos(\omega_m t) \) leads to the generation of a frequency comb with side bands spaced at \( \omega_m \). The corresponding modulation index is \( h_m = A_m/\omega_m \). The intensity of the \( n \)th side-band \( (I_n) \propto |J_{n}(h_m)|^2 \), where \( J_n(h_m) \) is \( n \)th-order ordinary Bessel function. The \( \sigma-\) and \( \sigma+\) components of these side bands constitute CPT states as the Larmor’s frequency becomes resonant with the harmonics of \( \omega_m/2 \). The factor 1/2 arises due to the frequency shifting between \( m_j = -1 \) and \( m_j = +1 \) (participating in CPT, figure 2) at twice rate of the associated Larmor’s frequency. Thus, the separation and relative amplitude of the split components of the magnetic resonance corresponds to \( \omega_m/2 \) and \( \propto |J_{n}(h_m)|^2 \), respectively, and is consistently observed in figure 4(A). As the value of \( A_m \) is increased, CPT states at higher Larmor’s frequency are observed due to coupling of more power to the higher side modes. The derivative of the calculated signal profile is shown in figure 4(B) that can be compared with the experimental profile. The calculated split profile consistently explains the experimental observation under different conditions and is discussed in the following part of this article. The success of this simplified model (shown in figure 2) proves the ground state Zeeman coherence as the mechanism behind the observation.
calculation in absence of transverse magnetic field. The position of consistent with the characteristic of Bessel function. (B): theoretical parameters, except for calculations are carried out using experimental values of the curve) is consistent with the experimental observation. The Figure 4.

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\[ \rho + \text{and} \sigma - \text{component of each mode constitutes a lambda system. The higher order modes also form similar lambda system at different detuned position and are not shown in figure 2(A). These lambda systems have different contribution to the signal depending on the value of } \Delta \text{ and } A_m. \text{ In contrast, the } \rho + \text{ and } \sigma - \text{ component from two different laser modes constitutes the CPT state for the non-central components of the split profile. The } m_f = 0 \text{ ground state in figure 2 resembles to the non-coupled ground hyperfine level in the realistic atomic system. The population of the } m_f = 0 \text{ state is reduced due to the creation of CPT state (leading to diminishing optical pumping) at each of the split components.}

The calculated split components at } n \times \omega_m/2 \text{ are not observed for laser frequency resonant } (\Delta = 0) \text{ with the atomic transition. The phase of } \pm n \text{th side modes around the carrier frequency follows the relationship } J_{-n}(h_m) = (-1)^n J_n(h_m). \text{ Thus, the } +1 \text{ and } -1 \text{ side modes have opposite phase with each other. For this specific case } (B_z = \omega_m/2 \text{ and } \Delta = 0), \text{ two symmetrically placed (with respect to the excited level) lambda systems forms a close loop excitation as shown in figure 2(B). Since one of the field is in opposite phase with respect to the other fields (due to the above relationship), the quantum interference is destroyed in this symmetric close loop excitation. Similar physical mechanism leads to the disappearance of the split components at all } n \times \omega_m/2 \text{ for } \Delta = 0. \text{ The actual process is little complicated than this simplified description. The calculation is done with } A_m \gg \Gamma \gg \omega_m \text{ that is analogous to the experimental condition } A_m \gg \text{ Doppler width } \gg \omega_m. \text{ Thus the atomic sample is resonant with only a part of the frequency comb (} A_m \gg \text{ transition width) that contains a large number of frequency components (transition width } \gg \omega_m) \text{ as shown in figure 2(D). The central part of the frequency comb is in resonance with the atomic system for } \Delta = 0. \text{ Since very high value of } h_m \text{ is used, the immediate (first) neighbours of any component of the frequency comb have similar amplitude but are in opposite phase in the central part of the frequency comb. Similar relationship exists between the third (all odd) neighbours of any frequency component in this regime. So as discussed above, the split components at } n \times \omega_m/2 \text{ for } \Delta = 0 \text{ are not observed. This property of Bessel function is progressively broken away from the centre of the frequency comb. Finally at both tail end of the frequency comb, immediate neighbour of any frequency component are in same phase (figure 2(D)). For } \Delta \neq 0, \text{ the frequency components away from the centre of the frequency comb are in resonance with the atomic system. Thus, one of the lambda systems becomes dominant over the other or both acts constructively in the close loop excitation as the above symmetry is broken. Consequently, the split components at } n \times \omega_m/2 \text{ are observed for } \Delta \neq 0, \text{ irrespective of sign of the detuning. The coupling scheme for } B_z = \omega_m \text{ and } \Delta = 0 \text{ is shown in figure 2(C), where the } \rho + \text{ component of the } +1 \text{ side mode forms a lambda system with the } \sigma - \text{ component of the } -1 \text{ side mode. It is evident from the coupling diagram that the split component at multiple of } \omega_m \text{ is observed for both on and off resonant light field.}

The experimentally observed detuning dependence of the split components is shown in figure 5. The primary feature of

![Figure 4](image-url)

(A) experimentally observed dependence of the split profile on modulation parameters (\(\omega_m, A_m\)). The behaviour is consistent with the characteristic of Bessel function. (B): theoretical calculation in absence of transverse magnetic field. The position of the split components for \(\Delta = 0\) (blue curve) and \(+25\ \Gamma\) (black curve) is consistent with the experimental observation. The calculations are carried out using experimental values of the parameters, except for \(A_m\). The difference in the amplitude of the signal for \(\Delta = 0\) and \(+25\ \Gamma\) is due to negligence of the thermal velocity distribution of the atoms and laser line width in the calculation.
the MMz profiles are consistent with the calculated signal profile for $\Delta = +25 \Gamma$. The experimental signal profile at $\Delta = -15 \Gamma$ and $+110 \Gamma$ exhibit similar behaviour to the calculated profile for $\Delta = 0$, where the components at $\omega_m/2$ are suppressed. However, the resonance position for Rb85 $F = 3 \rightarrow F' = 2$ and $3$ transitions are at $\Delta = 0$ and $+65 \Gamma$ respectively. It may be noted that the signal profile will have contribution from both of the transitions due to thermal velocity of the atoms. A detail calculation with the realistic atomic system (incorporating thermal averaging, laser line-width and actual modulation parameters) is required to address some of the discrepancies. Nevertheless, the transition identification is difficult for Rb85 $F = 3 \rightarrow F' = 2$ and $3$ transitions owing to Doppler broadening and the specified values of $\Delta$ are indicative only.

The detuning dependence of the MMz signal profile is further studied for Rb87 $F = 2 \rightarrow F' = 1$ transition coupled with a linearly polarized light. It is an ideal system to study the detuning dependence as there is no overlapping transition. The experiment is carried out with $\omega_m = 12$ kHz and $A_m \sim 1.63$ GHz, same as in figure 5. The split component at $\omega_m/2$ are not observed for laser tuned to the Rb-87 $F = 2 \rightarrow F' = 1$ transition as shown in figure 6. These components appear for off resonant light field and the observation is consistent with the calculated profile shown in figure 4(B). The disapperance of the split components at $n \times \omega_m/2$ for $\Delta = 0$ can be utilized as a new sub-Doppler spectroscopic technique for precise measurement of atomic energy levels while using a broad frequency comb.

The relative amplitude of the split components in figure 6 is different from the corresponding signal for Rb85 atoms shown in figure 3, despite using same value of $\omega_m$ and $A_m$. The split components for Rb87 atoms become more prominent at higher value of $A_m$ and signal profile appears similar to figure 3. Thus, the relative amplitude of the split components depends on the details of the transition apart from $A_m$. The issue (difference in $A_m$) can be addressed by using all experimental parameters and realistic atomic system for the calculation. However, the utilized simple model demonstrates the generality of the phenomena that will have application in other system like ultra-cold atoms. It also provides the vital physical processes without going through the involved calculation.

The MMz signal profile shows additional features in AR coated cell for higher amplitude of $A_m$ (~0.5 GHz). The higher $A_m$ also broadens the single photon absorption spectrum, similar to the buffer gas filled cell. It is due to generation of a wider frequency comb whereas a large homogeneous broadening was responsible for the buffer filled gas filled cell as has been discussed earlier. The laser frequency is locked to the zero crossing of this broad absorption signal, where the central mode of the frequency comb is expected to be at $\Delta \sim +25 \Gamma$ from the Rb87 $F = 3 \rightarrow F' = 2$ transition. The split components at higher $B_z$ field are prominently observed as shown in figure 7, despite having a broad single photon resonance width. This is consistent with the calculation, where a larger $A_m$ leads to observation of the higher order split components whereas larger homogeneous broadening has detrimental role for observation of the split profile. It may be recalled that split profile was not observed for buffer gas filled cell.

The higher side-mode of the light field are close to the Rb87 $F = 2 \rightarrow F' = 2, 1$ transitions at large value of $A_m$. The spacing between split components ($\omega_m/2$) for Rb85 and Rb87 atoms are $\omega_{1,01}$ (~1108 nT) and $\omega_{1,02}$ (~748 nT) respectively.
Thus, the split components of the Hanle resonance for Rb85 and Rb87 atoms appear at different $B_z$ magnetic field in the MM$_z$ signal profile (figure 7). The split components from both the species are merged together at several $B_z$ field leading to apparent amplified signal, whereas some components have diminished amplitude due to partial overlap with each other. It will be interesting to further explore the experiment to find out the single photon resonance (of the central mode of the frequency comb) for different transition by observing the depleted split components at $\omega_m/2$. This cannot be achieved by conventional sub-Doppler spectroscopic technique (like saturation absorption spectroscopy) due to wider spectrum of the frequency comb. A detailed investigation in this regard is beyond the scope of this article.

The direction of the ambient magnetic field with respect to the polarization axis of the light field plays a critical role in the dynamics of the laser-atom interaction. The $B_x$ field is parallel to the light polarization axis, whereas the $B_x$ and $B_z$ field are perpendicular to it. The distinct response of the atomic dynamics to the $B_x$ and $B_z$ field are reflected in the MM$_z$ signal profile as shown in figure 8. The quantum interference and optical pumping are the driving mechanism behind the MM$_z$ signal profile. The interplay and their dominance depend on the relative amplitude between the magnetic field along and transverse to the polarization axis of the light field. The calculated change in the population of the $m_f = 0$ ground state provides vital information on the associated physical process. The depleted population of $m_f = 0$ ground state signifies the population trapped in the dark state (prohibited optical pumping), whereas increase in its population is associated with the bright state.

The transverse $B_z$ field being orthogonal to the laser polarization axis, gets added with the scanning $B_z$ field. The resultant scanning field becomes $\sqrt{B_x^2 + B_z^2}$ and is perpendicular to the laser polarization axis. The position of the split components gets shifted with change in the $B_z$ field as shown in figures 8(A1) and (B1). The position of the split component will remain unaltered if plotted against $\sqrt{B_x^2 + B_z^2}$ instead of $B_z$ field. This may be realized from the larger shift in the position of the $\pm 1$ split components (at $\pm \omega_m/2$) towards the centre as compared to $\pm 2$ split components (at $\pm \omega_m$) for the same value of $B_z$ field. Consequently, the central component gets attenuated with increase in the amplitude of $B_z$ field and eventually vanishes (skipped during the scanning) for $B_z \sim 900$ nT. On further increase in the $B_z$ field, the $\pm 1$ split components merge together and appears as the central resonance (not shown here). In contrary to the central component in absence of the orthogonal field, this apparent central structure at higher $B_z$ field is originated due to quantum interference involving the $\sigma^+$ and $\sigma^-$ components from two different modes of the frequency comb. The dark state is established at each of the split component (for different $B_z$ field value) as has been seen by the depletion (calculated) in the population of the $m_f = 0$ ground state.

The transverse $B_z$ magnetic field along the laser polarization axis, influences the MM$_z$ signal profile in a different manner. The relative amplitude of the $B_z$ field as compared to the scanning $B_z$ field, dictates the onset of different physical mechanism. For the $B_z$ scan in the regime $B_z < B_{z1}$, the $B_z$ field gets added to the scanning $B_z$ field ($\sqrt{B_x^2 + B_z^2}$), in a similar way to the additional $B_z$ field. Thus, the split components shift towards the centre (figures 8(A2) and (B2)) as the amplitude of $B_z$ field is increased. The split components in this regime behave similar to the additional $B_z$ field. For the $B_z$ scan regime with $B_z < B_{z1}$ (near the central split component), the dark state is disturbed due to the orthogonal magnetic field. It is consistently observed by enhanced optical pumping of the atomic population to the $m_f = 0$ ground state in the calculated data. Similar mechanism has been reported in different experimental configuration where the dark resonance is transformed to a bright resonance in presence of transverse magnetic field [16, 18, 26, 27]. Since there is a depletion in the population of the $m_f = \pm 1$ ground state, enhanced transmission of the optical field is observed despite the dark state is disturbed by the $B_z$ field. This mechanism dominates at the central part of the MM$_z$ profile till $B_z \sim B_{z1}$, leading to broadening of the central resonance with increase the value of $B_z$ field. The calculated population of the $m_f = 0$ ground state shows steady increase with amplitude of the $B_z$ field in this regime. The amplitude of the $\pm 1$split components also get reduced apart from shift towards centre, as the amplitude of the $B_z$ field approaches $\omega_m/2$. It is shown in the plot for $B_z \sim \pm 900$ nT and proves the detrimental role of the $B_z$ field for the dark state. On further increase in the $B_z$ field, a single board profile is observed without any split components (not shown here). As has been stated, this broad profile is due to optical pumping resulting from the collapse of the dark state. This further establishes the requirement of dark state for observation of the split components in the MM$_z$ method. In summary, the central component
Figure 8. Experimental MM signal (A1) and (A2) and calculated signal (B1) and (B2) profiles for different amplitude of $B_x$ and $B_y$ field as a function of $B_z$ field. The calculated signal profile remarkably captures the details of experimental observation. The signal profiles are identical for change in the polarity of $B_x$ and $B_y$ field (not shown here). The experiment is carried out at $\omega = 12$ kHz and $A_m \sim 1.63$ GHz, whereas the calculated profiles corresponds to $\omega = 12$ kHz and $A_m \sim 100$ MHz.

gets skipped during scanning of the $B_z$ field as the amplitude of $B_y$ field is increased, whereas enhanced optical pumping (to the uncoupled $m_f = 0$ state) facilitated by the $B_x$ field leads to broadening of the central component. Nevertheless, the side components move towards the centre of the MM profile as either of $B_x$ or $B_y$ field is increased. The distinct response of the MM profile to the direction of the transverse magnetic field will be useful for advancement of three axis atomic magnetometer.

5. Conclusions

The splitting of the Hanle resonance by a frequency modulated light field is investigated using MM method and its distinctions with the FM method are discussed. The integrated atomic polarization gets resonantly enhanced as the Larmor frequency becomes resonant with the harmonics of the applied modulation. This is an additional feature to the oscillating atomic polarization studied in the conventional FM method. The splitting of the magnetic resonance for different experimental parameters is studied using a linearly polarized light field. The essential conditions required for observation of splitting in the MM method are pointed out. The sharp dependence of the split components (at the odd integral multiple of $\omega_m/2$) on the single photon detuning can have application in metrology using frequency comb. The contradiestinctive nature of the split components to the direction of magnetic field, along with the underlying physical processes is presented.

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Disclosures

The authors declare no conflicts of interest.
ORCID iDs

Swarupananda Pradhan https://orcid.org/0000-0003-2646-1836

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