We report a new technique to observe magneto-optical resonance with synchronous modulation of two resonant light fields. The resonance is created through coherent population trapping (CPT) by virtue of Raman excitation due to resonant light fields. Due to higher resonance amplitude at longer duty cycle close to 50%, the magnetic field sensitivity is improved in comparison to the conventional single resonant beam case. We achieve an average magnetic field sensitivity of 41 pT Hz$^{1/2}$ for duty cycle ranging between 35% to 10%. We have developed a theoretical model based on density matrix equations to verify our experimental observations. This technique could be used in remote magnetometry with mesospheric sodium for improving the sensitivity in geomagnetic field measurement.

Optically pumped atomic magnetometers are useful in sensitive detection of magnetic fields in biomagnetics [1,2], fundamental science [3] and geophysics [4]. Different techniques have been devised to improve the sensitivity and the dynamic range of atomic magnetometers [5-8]. For example, the spin-exchange relaxation free (SERF) magnetometer can measure very weak fields with sub-femtotesla sensitivity [6] and the synchronous pumping of the atoms with modulated light allows one to measure the geophysical range magnetic fields with high precision [8]. Magnetic field measurements are typically performed by monitoring either the intensity [7] or the rotation in polarization angle [5] of the light transmitted through an atomic sample used in the magnetometer. Recently, magnetic field measurement based on fluorescence light emitted from sodium (Na) atomic ensemble is demonstrated for potential application in remote magnetometry [9,10]. These laboratory experiments opened up the possibility of conducting geomagnetic field mapping over a large area using mesospheric sodium fluorescence [11–13]. Trial sky experiments have been performed using back-scattered fluorescence in sodium D$_2$ line by using a single amplitude-modulated resonant beam in the Bell-Bloom (BB) geometry [14]. Currently, measurement sensitivity in the sky experiments is very low compared to the laboratory based atomic magnetometers [9]. Thus, new techniques need to be explored for improving the sensitivity further in a remote magnetometer.

The method involving two continuous resonant light fields has been widely studied in coherent population trapping (CPT), particularly for designing miniaturized vapor cell atomic clocks [15,16]. In this case, two light fields couple the $m_F=0$ Zeeman sublevels in two hyperfine ground-states of alkali atoms to create a magneto-insensitive CPT resonance, which is utilized as a reference for correcting the phase drift of a local oscillator in the atomic clock. In present work, we use two amplitude-modulated resonant fields to study magnetic-resonances in Na D$_1$ line and demonstrate magnetometer operation based on the magnetic resonance observed in fluorescence. Resonance signal at Larmor frequency $\Omega_\phi$ is measured by detecting fluorescence with an applied magnetic field perpendicular to the light propagation direction, which is consistent with the sky experiment [11–13]. Measurements are performed at different modulation duty cycles with constant peak intensity of the light fields. The amplitude and width of the magnetic field are chosen with strong dependence on the light duty cycle. Unlike the $\Omega_\phi$ resonance obtained using a single modulated field in the BB geometry [9], higher resonance amplitude is observed at longer duty cycles close to 50% in the two beam case. An average magnetic field sensitivity of 41 pT Hz$^{1/2}$ is achieved for duty cycles ranging from 35% to 10%. The experimental observations are supported by theoretical results obtained using the atomic density matrix analysis.

Figure 1 illustrates the schematic of the experimental arrangement. A frequency-doubled Raman fiber amplifier with a narrow bandwidth (< 1 MHz), and a maximum output power of 2 W, tuned to Na D$_1$ resonance (589.756 nm) is used in the experiment. The laser wavelength is monitored and controlled by utilizing a saturation absorption spectroscopy (SAS) setup using buffer-gas-free Na reference cell. As the amplitude-modulated beam in our experiments is frequency shifted, the input laser beam to the SAS setup is also frequency shifted by AOM1 by the same frequency as AOM2. Using the SAS peaks, the laser is tuned at $\Delta\nu=2→F_2→2$ transition of Na D$_1$ line. Laser beam used in our
experiments is amplitude-modulated at 80MHz with AOM2. The first-order diffracted beam is used as a carrier beam for a resonant electro-optic phase modulator (EOM) from QUBIG (PM-Na23_17K3). The EOM is driven by a signal generator (Stanford Research Systems SG384) to create optical sidebands at frequencies ±1.771 GHz with respect to the carrier frequency. The output of the signal generator is amplified using a radio frequency (RF) amplifier before sending it to the EOM. The carrier to sideband power ratio is set to 1:2, measured using a Fabry-Perot Interferometer (Thorlab SA210-5B). Using a two-lens telescopic configuration (lenses L2 and L3), the laser beam diameter is expanded from 2 mm to 8 mm. Experiments are conducted using a Na vapor cell, containing 10 Torr of neon buffer gas. The laser beam intensity at the cell is controlled using a neutral density (ND) filter, and the laser beam polarization is adjusted to circular using a λ/4 plate. The cell is wrapped with a twisted bilayer Nichrome wire (to cancel the magnetic field produced by applied DC current) and heated to 92° Celsius which yields a vapor density of about 1.6×10^9 atoms/cm^3. The cell is kept inside a non-magnetic chamber for thermal insulation, which is then installed at the center of a two-layered magnetic shield (μ-metal) ensuring a shielding factor of ~10^5. Any residual ambient magnetic field is further cancelled using the three pairs (three-axes) of Helmholtz coils mounted inside the shield. These coils are also utilized to apply a constant field in all three directions using a voltage-controlled current source (VCCS). We have performed all experimental measurements using the fluorescence light collected in the perpendicular direction to that of light propagation k0. A photomultiplier tube (PMT) placed at the focus of the lens L4 is used to collect the fluorescence light. An ultranarrow band-pass filter (Alluxa 589.45-10DG) is kept before the PMT to reduce the background light in the fluorescence. Resonance signals are detected using a low-pass filter (LPF) at a cut-off frequency f_c=1 kHz [10].

Figure 2 shows some of the possible A-excitation schemes using resonant light fields in the presence of constant magnetic fields along different directions. In Fig. 2 (a), the carrier signal (strong) is tuned to F_s=2→F_p=2 transition, and one of the side band (weak) is resonant to F_s=1→F_p=2 transition. Both carrier and side band beams are σ+ polarized, which gives rise to two-photon excitations by coupling the Zeeman sublevels of hyperfine ground states F_s=2 and F_p=1 via a common sublevel of excited state F_s=2 in the A-system in the presence of a longitudinal magnetic field B_i. In the absence of magnetic field, all the Zeeman sublevels (or A-systems) are degenerate and a single CPT resonance occurs (not shown) when the frequency difference (Δ) between carrier and side band matches with the ground state hyperfine splitting (ΔG) of Na i.e. Δ = Δ_s - Δ_e = Δ_0 [16,17], where Δ_s and Δ_e are the single photon detunings of the carrier and the side band from their resonant frequencies ω_c and ω_e, respectively. Figure 3 shows the observed CPT spectrum as a function of difference detuning Δ in the presence of fixed (constant) magnetic fields. The detuning Δ is varied by scanning the side-band detuning Δ using the signal generator (through EOM) with fixed Δ_c=0. To improve signal-to-noise ratio (SNR), the resonances are measured using a low-pass filter (LPF) at a cut-off frequency f_c=1 kHz [10].

Using a digital oscilloscope, the signal is averaged over 50 samples. A total intensity in the carrier and one of side band is set to 5.2 W/m^2. The axis of quantization is considered along the direction of total effective magnetic field (B). First, in the presence of longitudinal field (B=B_i), the resonant excitations with (σ', σ") polarized fields create A_1-A_2 systems [as shown in Fig. 2(a)]. A central CPT resonance at Δ=0, created by the A_2 system, and two additional resonances at Δ±2Ω_e formed by A_1 and A_3 systems [16,17], are observed [Fig. 3 (solid red curve)], where Ω_e=γB is the Larmor frequency, γ is the Gyromagnetic ratio. In
addition to field $B_0$, when a transverse magnetic field ($B_x$ or $B_y$) is applied, the axis of quantization tilts in the plane of light polarization (i.e. $x$-$y$ plane). This creates additional $\pi$- and $\sigma$-transitions along with $\sigma^*$-transitions and forms new $\Lambda$-systems as shown in Fig. 2(b & c). The new $\Lambda$-$\Lambda_{10}$ systems created by $(\sigma^*, \pi)$ excitations contribute to form the additional CPT resonances at $\Delta=\pm \Omega$, and $\pm 3\Omega$, increasing total number of CPT dips to seven in the spectrum [Fig. 3 (dashed blue curve)]. The contrast of a CPT dip depends on the population distribution among ground-state Zeeman sublevels and coupling strengths of the $\sigma^*$, $\sigma$ and $\pi$-transitions involved in corresponding $\Lambda$-system [17].

Next, we measure the magnetic resonances, produced by the resonant two-photon excitation, by changing the $\Omega_m$ at fixed

![Amplitude vs $\Omega_m$ (kHz)](image)

Fig. 4. Experimentally (a) and theoretically (b) obtained amplitudes of the magnetic resonances as the function of $\Omega_m$ at different $\Delta$. Parameters used in the simulations: Rabi frequencies $\Omega_m=2\Omega=0.031\Gamma$ and transit decay rate $\tau=10^{-6}\Gamma$, where $\Gamma$ is the spontaneous decay rate.

magnetic field $B_x=B_y=85.7$ mG ($\Omega_m=60$ kHz) [Fig. 4a]. Since the low-pass filter is insensitive to signal phase, it allows us to scan the $\Omega_m$ over a wide range from 40 kHz to 200 kHz [10]. At a duty cycle of 50%, the modulated light will have dominant first-harmonic frequencies $\omega_c+\Omega_m$ and $\omega_c-\Omega_m$ of the carrier and side band beams, respectively. These new frequency components in the modulated light fields will create additional $\Lambda$-systems independently as well as with unmodulated frequencies $\omega_c$ and $\omega_c$. For example, the frequency difference of each pair $(\omega_c+\Omega_m, \omega_c-\Omega_m)$ and $(\omega_c, \omega_c+\Omega_m)$ matches with the hyperfine splitting $\Delta_{th}$ of $\Delta=0$ and $\Delta=\pm \Omega_m$ respectively. Therefore, at fixed frequency detuning $\Delta$, when the magnetic field is applied in an arbitrary direction, the magnetic resonances can occur at $\Omega_m=\Omega, 2\Omega, 3\Omega, \pm \delta B$, and $3\Omega, \pm \Delta$ under two-photon resonant conditions. Experimentally, for $\Delta=0$ (Fig. 4a: blue curve), we observe the magnetic resonances at $\Omega_m=60$ kHz ($\Omega_m$ and $\Omega_m=180$ kHz ($3\Omega_m$) generated by strong $(\sigma^*, \pi)$ transitions [refer to Fig. 2(c)] in the presence of field $B_0$. On the other hand, when only a transverse magnetic field $B_x$ is applied, resonance at $\Omega_m=120$ kHz ($2\Omega_m$) is formed by weak $(\sigma^*, \sigma^*)$ or $(\sigma, \sigma)$ transitions [refer to Figs. 2a, 2b]. Hence, it may not be possible to detect $2\Omega$ resonance as observed in Fig. 4(a) due to low sodium vapor density in the experimental cell at the set temperature of 92°C. When the difference detuning $\Delta=10$ kHz is introduced, the resonances $\Omega_m=\Omega, \pm \delta B$ and $\Omega_m=3\Omega, \pm \delta B$ are observed around $\Omega_m=\Omega$ and $\Omega_m=3\Omega$, respectively (Fig. 4a: red curve). For $\Delta=10$ kHz, there is no two-photon resonance condition at $\Omega_m=3\Omega$, and $\Omega_m=\Omega$, in the $\Lambda$-transitions resulting from the two CPT fields. Hence, no $\Omega_m=3\Omega$

resonance is observed in Fig. 4(a) at $\Delta=10$ kHz. However, the $\Omega_m=\Omega$ resonance can be seen for both $\Delta=0$ and $\Delta=10$ kHz. This observation can be explained by considering that for $\Delta=10$ kHz, the $\Omega_m=\Omega$ resonance could also be formed by a single field in the Bell-Bloom (BB) geometry [10] through $\Lambda_{12}$-system, as shown in Fig. 2(d). To further validate our experimental observations, we developed a theoretical model based on the density matrix equations by considering three hyperfine sub-levels of Na D$_1$ line with two amplitude-modulated light fields [18]. We have simplified our model by neglecting the effect of neighboring transition in the excited state, the atomic motion, and the spatial distribution of light intensity. The theoretical calculations produce similar results (Fig. 4b) as observed experimentally (Fig. 4a). In addition, the theoretical plots also manifest weak resonances at $\Omega_m=120$ kHz ($2\Omega_m$) formed by $(\sigma^*, \sigma^*)$ and $(\sigma, \sigma)$ CPT fields at $\Delta=0$, and by a single field at $\Delta=10$ kHz with $\Lambda_{12}$-transitions [refer to Fig. 2d].

Finally, we demonstrate an atomic magnetometer based on $\Omega_m=\Omega$. Resonance produced using two resonant amplitude-modulated CPT fields ($\Delta=0$). The sensitivity of this magnetometer depends on the amplitude and linewidth of the resonance. Figure 5(a) shows experimentally measured peak amplitude (red dots) and width (blue dots) of the $\Omega$ resonance at different duty cycles of light modulation. The peak intensity of light fields is fixed to 5.2 W/m$^2$ for all measurements. The light modulation frequency $\Omega_m$ is varied around the Larmor frequency $\Omega_m=180$ kHz, i.e. magnetic field $B_m=257$ mG. The cut-off frequency $f_c$ of LPF is set to 1 kHz and the signal is averaged over 50 samples on the oscilloscope. As the duty cycle is lowered from 50%, the amplitude of the resonance increases initially, and maximizes around 35% duty cycle (Fig. 5a: red curve). The amplitude decreases almost linearly over duty cycle ranging from 30% to 10%. This CPT technique with two resonant fields delivers the $\Omega_c$ resonance with higher peak amplitudes near 50% duty cycle [Fig. 5(a)] unlike the single field $\Omega_m$ resonance obtained in the Bell-Bloom (BB) geometry [9]. To keep the resonance peak intensity constant, the average light field intensity needs to be adjusted to lower values as the duty cycle is lowered. Therefore, the linewidth of resonance reduces linearly with duty cycle [Fig. 5a: black curve]. The theoretical results shown in Figure 5(b) qualitatively agree with experimental results shown in Figure 5(a).

Particularly, calculated $\Omega_m$ resonance also has higher peak amplitude at a 50% duty cycle.

Next, to quantify the performance of our magnetometer, we measured the noise in the low-pass filter output signal using a fast-Fourier-transform (FFT) spectrum. The shot-noise (SN) limited sensitivity $\delta B$ of a magnetometer is given by
\[ \delta B = \Delta f / (\gamma \cdot \text{SNR}) \]  

(1)

where \( \Delta f \) is the full-width at half-maximum (FWHM) of the resonance in units of Hz, \( \gamma = 6.989812 \text{ Hz/nT} \) is the Gyromagnetic ratio for sodium atom, and \( \text{SNR} \) is the signal to noise ratio with units of Hz\(^1/2\). Figure 6(a) shows the magnetic noise spectrum at resonance (\( \Omega_m = \Omega_L = 180 \text{ kHz} \)). At a duty cycle 35%, the root-mean-square of magnetic-noise is measured around 45 pT/Hz\(^1/2\) for frequencies ranging from 1 Hz to 1 kHz (Fig. 6a: red line). Since the LPF has \( f_c = 1 \text{ kHz} \), the noise decreases rapidly for frequencies above 1 kHz. The SN-limited sensitivity \( \delta B \) at different duty cycles is shown in Fig. 6(b). As the duty cycle is decreased from 50% to 35%, the sensitivity \( \delta B \) improves by a factor of two. For lower duty cycles, the sensitivity remains constant without much variation. Our results show an average sensitivity of 41 pT/Hz\(^1/2\) (Fig. 6b: dashed line) for duty cycles between 35% to 10%. In the reported literature, using single beam amplitude-modulated light in the BB geometry, the highest sensitivity is recorded at ≤ 25% [12,13] in the sky experiments. That limits the average laser power used in the measurements. A better sensitivity at longer duty cycles opens the possibility of using higher average laser power in the measurement, which is particularly important to increase the return fluorescence from mesospheric sodium.

The magnetic field sensitivity can be further improved by modifying the current experimental setup. For example, in the current setup, the EOM generates two side bands and only one of the side band beams participate to form the resonance along with carrier beam. The remaining side band beam adds a background noise, thus reducing the overall contrast of the resonance. The amplitude of the \( \Omega_m \) resonance also depends on the power ratio between the two CPT fields. Currently, a maximum carrier to sideband power ratio of 0.5 is possible due to our experimental constraints. We expect a further increase in resonance amplitude with equal light powers in both fields. In summary, we have developed a new technique to improve the sensitivity of an atomic magnetometer based on \( \Omega_m \) resonance. This technique uses two resonant amplitude-modulated fields instead of a single modulated-field in the Bell-Bloom geometry. We also showed that our technique significantly enhances the SN sensitivity of resonance at longer duty cycles than previously reported. With two resonant fields, atoms in both ground states contribute to the optical pumping process, leading to strong \( \Omega_m \) resonance. An additional \( 3\Omega_m \) resonance is also observed using two resonant CPT fields, which can be used for the magnetic field measurement as well. The \( 3\Omega_m \) resonance is not possible to obtain with a single field in BB configuration. Magnetic field sensitivity in our technique is comparable for both longer and shorter duty-cycle pulses. An average magnetic field sensitivity of 41 pT/Hz\(^1/2\) is measured with pulse duty cycle ranging from 35% to 10%. At same peak power, the longer duty-cycle pulses have higher average power, which can be utilized to enhance return fluorescence from mesospheric sodium in the sky experiments. In addition, the dark \( \Omega_m \) resonance of sodium \( D_1 \) line with reduced photon shot noise is expected to show better magnetic field detection sensitivity in the sky experiments [19]. We expect to conduct remote magnetometry experiments using this technique in the sodium \( D_1 \) line in the near future.

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