Magneto-optical resonances in fluorescence from sodium D₂ manifold

RAGHWINDER S. GREWAL¹, GOUR S. PATI¹, RENU TRIPATHI¹, *ANTHONY W. YU², MICHAEL KRAINAK² AND MICHAEL PURUCKER²

¹Division of Physical and Computational Sciences, Delaware State University, Dover, DE 19901, USA
²NASA GSFC, Greenbelt, MD 20771, USA
*Corresponding author: rtripathi@desu.edu

We report on magneto-optical resonances observed in sodium fluorescence from D₂ manifold with an intensity modulated light illuminating a sodium (Na) cell containing Ne buffer gas. Resonances are measured in fluorescence emitted perpendicular and backward to the light propagation direction in the cell. Properties of these resonances are studied by varying the magnetic field at fixed light modulation frequency, and vice-versa. A dark resonance having maximum amplitude for laser wavelength close to the crossover peak, is observed. The origin of dark resonance observed in Na D₂ line is discussed. Pulse modulation with low-duty cycle shows higher-harmonic resonances of the modulation frequency and sub-harmonic resonances of the Larmor frequency. Present study is aimed towards improving the understanding of magneto-optical resonances for remote magnetometry applications with mesospheric sodium.

Return fluorescence obtained from laser excitation of mesospheric sodium is conventionally used as laser guide star to correct wavefront distortion in astronomical observations [1]. Recently, laser excitation of mesospheric sodium has received considerable attention in remote sensing of geomagnetic field [2-4]. The method, first proposed by Higbie et al [2], was based on the measurement of spin precession of mesospheric sodium atoms by polarizing them through synchronous optical pumping with a modulated laser beam. In 2016, Kane et al [3] made the first experimental observation and measured the geomagnetic field with sensitivity 162 nT Hz⁻¹/². The measurement sensitivity was further improved to 28 nT Hz⁻¹/² by Bustos et al [4]. Polarization-modulated light was used to measure geomagnetic field with an uncertainty of 30 nT [5]. Sensitive measurement and large-scale mapping of geomagnetic field can provide better insight into various processes impacting the mesosphere region, thus, enabling several important scientific studies, for example, magnetic fluctuations at mesospheric altitude can be used to determine large-scale ocean currents [6], mapping local current structures in the dynamo region [7], and magnetic structures in the earth’s upper mantle [8]. Unlike other alkali atoms (K, Rb, Cs), magneto-optical resonance is not studied extensively in sodium atom. Understanding light-atom interaction and the origin of magneto-optical resonances in sodium atom via laboratory experiments is necessary for perfecting remote magnetometry and further improving its sensitivity. Recently, Fan et al [9] performed a detailed study of magnetic resonance in sodium D₁ line using a sodium cell with high buffer gas pressure. Effects of different parameters like laser intensity, duty cycle of modulation and magnetic field on sensitivity were analyzed. The measurements were done using sodium fluorescence in a direction perpendicular to the laser beam axis.

In this letter, we focus on studying magneto-optical resonances in fluorescence from Na D₂ line, which has been used in all sky experiments [3-5]. We study the properties of magnetic resonances by detecting fluorescence at directions perpendicular and backward to laser beam propagation in a sodium cell. Resonances are acquired using two independent methods: phase-sensitive lock-in detection is performed using a high bandwidth lock-in amplifier, and direct amplitude measurement is performed using a low-pass filter (or integrator). We measured the resonances as functions of magnetic field and laser modulation frequency (or pulse repetition rate) with different pulse duty cycles. We also explain the origin of dark resonance observed in Na D₂ line using the action of a repump beam, and a theoretical model including buffer gas induced collisional dephasing in the excited state.

The schematic of the experimental setup is shown in Fig. 1. A narrow-band frequency-doubled Raman fiber amplifier laser tuned to Na D₂ line is used in the experiment. A combination of half-wave plate and PBS is used to control the optical power in the laser lock setup. Two different methods are employed to monitor the laser wavelength using buffer gas free Na reference cells. First, the Doppler-broadened fluorescence spectrum at 90° angle to light propagation direction is collected using an avalanche photo diode (APD) and second, the Doppler-free absorption peaks/dips are observed in transmitted light from a saturation absorption spectroscopy (SAS) setup. Using offset lock with the help of a proportional integral (PI) controller (Newport LB1005 Servo...
Controller), the laser is locked on different positions in Na D₂ line by utilizing the fluorescence spectrum from the APD. The SAS setup is used only to find the exact position of the laser with respect to $F_{2}$ → $F_{0}$ (1, 2, 3) [referred as D₂a transition], $F_{2}$ → $F_{0}$ (0, 1, 2) [referred as D₂b transition] and the crossover peak present in the Doppler-free absorption spectrum [10]. The laser intensity is modulated using an acoustic-optic modulator (AOM) driven by the rectangular pulse waveform with different duty cycles, and the first-order diffracted beam is selected as probe beam for the experiment [11]. A resonant electro-optic phase modulator (BDM) is used to create optical sidebands as repump at frequencies ±1.710 MHz with respect to the carrier (or probe beam) frequency. Before passing through the experimental Na cell containing 10 Torr Ne buffer gas, the beam diameter is expanded to approximately 8 mm (using two lenses L2 & L3) to increase the number of interacting Na atoms in the beam volume and prevent transit-time broadening.

A neutral density (ND) filter is used to control the probe beam power to the cell. Using a $\lambda$/4 plate, the light polarization is set to circular. A non-polarizing pellicle beam splitter of diameter 2" transmits ~50% laser power onto the experimental Na buffer cell. The experimental cell is kept inside a two-layered magnetic shield (µ-metal) enclosure, which reduces the ambient magnetic field by a factor of ~10². Three pairs of Helmholtz coils are installed inside the shield to further reduce the ambient field by canceling the residual field. Coils along y-axis perpendicular to the light propagation vector (k₀) are utilized to apply fixed or scanning magnetic field, as per experimental needs. Temperature of the Na buffer cell is kept at 86°C to keep sodium atomic density low (~1.0×10⁹ atoms/cm³ [12]), thus making it comparable to the number of interacting Na atoms in the mesosphere column (length ≈ 10 km). Magnetic resonances are obtained by collecting fluorescence light from the Na buffer cell at two different angles. First, fluorescence at 90° angle to the light propagation direction is detected using a PMT at the focal plane of a lens L5. Second, fluorescence at 180° angle (i.e. backward) to the light propagation direction is detected using another PMT at the focus of lens L4. The backscattered fluorescence is reflected by the pellicle beam splitter. To eliminate background light in fluorescence, an ultra-narrow band-pass filter (Alluxa 589.45-1 OD6) is used before the PMT. The resonance signals are acquired by two independent methods: (a) demodulating the signal with a lock-in amplifier (Stanford Research System SR865), operating at the first harmonic of the light modulation frequency $\Omega_{m}$ and (b) measuring the signal directly through a low-pass filter (Stanford Research Systems SR560).

Fluorescence measurements are first performed in the direction perpendicular to light propagation and by scanning the magnetic field (B₀) along y-axis while keeping $\Omega_{m}$ fixed. Figure 2 shows magneto-optical resonances observed corresponding to different duty cycles of light modulation. The probe beam is modulated at fixed $\Omega_{m} = 20$ kHz frequency with an average intensity of 0.25 mW/cm². The laser is locked closer to the crossover peak (observed midway between D₂a and D₂b transitions) where maximum amplitude of the magneto-optical resonance is observed. Fluorescence signal is demodulated using the lock-in amplifier; the in-phase and the quadrature-phase components are shown respectively, in Fig. 2(a) and 2(b).

When the laser is modulated with 50% duty cycle [Fig. 2 (top row)], synchronous optical pumping of atoms leads to optical resonances at Larmor frequency $\pm 2 \Omega_{0}$ (B₀ = $\Omega_{0}$) [13], as seen in both in-phase and quadrature-phase signals, where $\gamma$ is the Gyromagnetic ratio. The in-phase signal shows the amplitude, and the quadrature signal shows the phase of the resonance. The width (i.e. full width at half maximum, FWHM) of the resonance near zero magnetic field is measured to be 3.2 mG. As the duty cycle of modulation is lowered to 35% [Fig. 2 (middle row)], the second harmonic signals corresponding to $\Omega_{x} = 2 \Omega_{0}$ are also seen along with the first harmonic. The presence of higher order resonances at lower duty cycles is due to the Fourier components present at integer multiples of $\Omega_{m}$ in the modulated waveform [14]. For duty cycle of 20% [Fig. 2 (bottom row)], the in-phase signal clearly shows up to third harmonic and the quadrature signal shows resonances up to fourth harmonic. Since the quadrature signal is sensitive to the signal phase, it allows to detect weak signal.

Next, we measured the magnetic resonances directly using a low-pass filter for different duty cycles of modulation [Fig. 3]. The cut-off frequency ($f_{c}$) for low-pass filter is set to 1 kHz and the signal is averaged over 100 samples using a digital oscilloscope. The low-pass filter measures only the signal amplitude i.e. insensitive to the
phase. Signals measured with low-pass filter [Fig. 3] resemble the in-phase signals of lock-in amplifier [Fig. 2 (first column)]. The advantage of using low-pass filter is that the scanning rate of magnetic field (or modulation frequency at fixed field) can be faster compared to the lock-in detection where the scanning rate is limited by its long time constant, hence it reduces the data processing time. Figure 4 shows the plots of magneto-optical resonances when the magnetic field $B_m$ is fixed at 258.6 mG and the modulation frequency $\Omega_m$ of rectangular pulses of the laser beam is varied from 50 kHz to 200 kHz. Unlike lock-in detection, the low-pass filter is phase-insensitive and therefore, the resonance signal is measured over a wide range of $\Omega_m$. Laser beam modulation with 50% duty cycle shows magnetic resonance at Larmor frequency [Fig. 4(a)]. When the duty cycle is lowered to 20%, resonances also occur at the second and third sub-harmonics of Larmor frequency [Fig. 4(b)]. This is in agreement with the previously reported results for Na D$_1$ line [9].

Fig. 3. Magneto-optical resonances: direct measurement using a low-pass filter for different modulation duty cycles of modulation(a) 50% and (b) 20% at $\Omega_m$=20 kHz.

Fig. 4. Magneto-optical resonances observed by changing the laser beam modulation frequency $\Omega_m$: Direct measurement using low-pass filter at different duty cycles (a) 50% and (b) 20%.

The presence of cycling transition $F_g=2\rightarrow F_e=3$ in Na D$_2$ line in a Doppler-broadened medium, is expected to increase the fluorescence (higher absorption) at the center of magnetic resonance [2], which is the characteristic of bright resonance. However, we observed a dark resonance in the experiment i.e. decrease in fluorescence at the center of resonance. To understand the observed behavior, we locked the laser at different positions in Na D$_1$ line with and without the repump beam by switching the EOM on and off, respectively [Fig 5]. The resonance, obtained when the laser beam is tuned to D$_{2a}$ transition (without the repump beam), is shown in Fig. 5 (a). Average laser intensity in this case, is set to 0.25 mW/cm$^2$ with 20% duty cycle and the magnetic field is kept fixed at 258.6 mG. Compared to the amplitude observed near the crossover, we observed a reduction in the amplitude of resonance by a factor of $\sim$1.2. However, the sign of resonance did not change. The EOM is turned on, one of the side bands produced by EOM gets resonant with the Na D$_{2a}$ transition and acts as the repump beam. Due to 50% loss of power in the two sidebands, the total laser beam power is doubled to keep the probe beam power same. This results in an increase of resonance width from 2.5 kHz to 3.7 kHz as measured from the fittings of experimental data [Fig 5 (top row red line)], and signal amplitude is also increased by a factor of $\sim$1.8 without observing a change in its sign. The laser is then tuned to Na D$_{2b}$ transition [Fig. 5 (c)] keeping all the other experimental parameters and conditions same as in Fig. 5(a). Due to the presence of cycling transition $F_g=1\rightarrow F_e=0$ in the Na D$_{2b}$ line, the resonance is expected to be a dark resonance. Experimentally, no resonance is observed in the signal [Fig 5 (c)]. A dark resonance is observed only when the repump beam, resonant to D$_{2a}$ transition, is switched on [Fig. 5 (d)]. The role of probe and repump beams is reversed in this case, thus, indicates that the magnetic resonances happen only due to light absorption in the D$_{2a}$ transition. The D$_{2b}$ transition has less absorption compared to D$_{2a}$ transition [15], which suggests that magnetic resonance in D$_{2b}$ transition is weak and may not be possible to observe due to low sodium density in the cell under the present experimental conditions. The observed dark resonance in D$_{2a}$ transition can be explained by considering excited state dephasing of $F_g=2\rightarrow F_e=3$ transition due to collision of Na atoms with buffer gas atoms. The collisional decay typically suppresses the coherence transfer due to spontaneous emission from the excited state, which results in the formation of bright resonance for $F_g=3\rightarrow F_e=4$ transition [16,17]. Therefore, the sign of resonance in Na D$_2$ line is reversed in the presence of buffer gas atoms. Remote magnetometry with mesoscopic sodium atoms can only be performed with backscattered fluorescence [3,4]. Therefore, measuring magnetic resonances in backscattered fluorescence is consistent with remote magnetometry. Typically, in laboratory experiments, reflection from the cell window prevents one to perform fluorescence measurements in the backward direction [9]. In our experiment, Na cell with wedge window is used to eliminate backward reflection. Only half of the backscattered fluorescence is reflected by the p-BS (shown in Fig. 1). Figure 6 shows magneto-optical resonances obtained using backscattered fluorescence light. Light modulation frequency is set to 20 kHz at duty cycle 20% and the average intensity is set to 0.25 mW/cm$^2$. Figure 6(a) shows quadrature phase signal obtained by demodulation using the lock-in amplifier as $B_m$ becomes. The quadrature component shows resonances up to fourth harmonic, which matches our earlier observation in Fig. 2 (bottom row). However, the in-phase component did not show any resonance due to weak backscattered fluorescence. Instead, we used a low-pass filter at $f_c=1$ kHz to measure the resonance amplitude [Fig 6 (b)]. A faster field scan at 4 Hz allowed us to perform long sample averaging on the oscillosc-
Experimental results shown in Fig. 5(a). Next, we measured the resonances by scanning the modulation frequency $\Omega_{\text{m}}$ in Fig. 6(c) and(d). First, the measurements are performed at low magnetic field ($B_y = 29$ mG) to observe resonances at subharmonics of the Larmor frequency [Fig. 6(c)]. Although the resonances are weak, but second and third subharmonics are visible along with the resonance at Larmor frequency. Magnetic field is then increased to 258.6 mG [Fig. 6(d)]. A resonance of width 2.6 kHz is observed at Larmor frequency corresponding to the applied magnetic field.

In Na D$_2$ line, Fan et al. [9] showed that fluorescence from sodium atoms when excited by circular polarized light, is likely to be directed backward, thus resulting in a dip at the center of resonance for fluorescence emitted in the direction perpendicular to the laser beam. However, in our experiments the resonance measured in backward direction also showed the dip at the center of resonance [Fig. 6(d)]. In contrast to the explanation in reference [9], this observation suggests that the observed dip in Na D$_2$ line is not dependent on the detector position.

In conclusion, we have investigated magneto-optical resonances in sodium D$_2$ line using fluorescence from sodium atoms in a vapor cell containing Ne buffer gas. We performed measurements using fluorescence emitted both in perpendicular and backward direction to the laser beam propagation axis. Maximum amplitude of magnetic resonances was measured near the cross-over peak. Due to collisional dephasing in the excited state of $F=2\rightarrow F'=3$ transition, a dark resonance is observed in sodium D$_2$ line. This is validated using a theoretical model. Resonance in $F=1\rightarrow F'=0$ transition was also investigated in the presence of a repump beam. Higher-harmonic resonances and subharmonic resonances were observed with modulated light by employing respective magnetic field and modulation frequency scans. Resonances acquired using lock-in amplifier and low-pass filter were compared. Resonances measured in backscattered fluorescence were found considerably weaker but matches with the observed resonance in perpendicular direction. The present work has improved the understanding of magneto-optical resonances in sodium D$_2$ transitions, which will be useful in conducting remote magnetometry experiments in the near future.

References
1. W. Happer, G. J. MacDonald, C. E. Max, and F. J. Dyson, J. Opt. Soc. Am. A 11, 263 (1994).
2. J. M. Higbie, S. M. Rochester, B. Patton, R. Holzlöhner, D.B. Calia, and D. Budker, Proc. Natl. Acad. Sc. 108, 3522 (2011).
3. T. J. Kane, P. D. Hillman, C. A. Denman, M. Hart, R. P. Scott, M. E. Purucker, and S. J. Potashnik, Geophys. Res. Space Phys. 123, 6171 (2018).
4. F. P. Bustos, D. B. Calia, D. Budker, M. Centrone, J. Hellemeier, P. Hickson, R. Holzlöhner, and S. Rochester, Nat. Commun. 9, 3981 (2018).
5. F. P. Bustos, D.B. Calia, D. Budker, M. Centrone, J. Hellemeier, P. Hickson, R. Holzlöhner, and S. Rochester, Opt. Lett. 44, 138 (2019).
6. R. H. Tyler, S. Maus, and H. Lühr, Science 299, 239 (2003).
7. S. Maus, F. Yin, H. Lühr, C. Manoj, M. Rother, J. Rauberg, I. Michaelis, C. Stolle, and R. D. Müller, Geochem. Geophys. Geosyst. 9, Q07021 (2008).
8. M. Blanc, and A. D. Richardson, J. Geophys. Res. 85, 1669 (1980).
9. T. Fan, L. Zhang, X. Yang, S. Cui, T. Zhou, and Y. Feng, Opt. Lett. 43, 1 (2018), and supplementary material on arXiv:1710.04342 (2018).
10. C. Y. She, and J. R. Yu, Appl. Opt. 34, 1063 (1995).
11. Laser locking using the fluorescence spectrum was adjusted such that the frequency shift (+80 MHz) introduced by AOM brings the probe beam closer to the Doppler-free absorption peaks in D$_{2a}$ and D$_{2b}$ transitions observed using the SAS setup.
12. G. D. Domenico, and A. Weis, Vapor Pressure and Density of Alkali Metals, available online at http://demonstrations.wolfram.com/VaporPressureAndDensityOfAlkalimets/.
13. W. E. Bell, and A. L. Bloom, Phys. Rev. Lett. 6, 280 (1961).
14. W. Gawlik, L. Krzemień, S. Pustelny, D. Sangla, J. Zachorowski, M. Graf, A. O. Sushkov, and D. Budker, Appl. Phys. Lett. 88, 131108 (2006).
15. F. P. Bustos, R. Holzlöhner, D. Budker, S. Lewis, and S. Rochester, Proc. SPIE 9909, 210 (2016).
16. A. V. Taichenachev, A. M. Tumaikin, and V. I. Yudin, Phys. Rev. A 61, 013802 (1999).
17. H. Faílache, P. Valente, G. Ban, V. Lorent, and A. Lezama, Phy. Rev. A 67, 043810 (2003).
18. S. M. Rochester, Atomic Density Matrix package, available online at http://rochesterscientific.com.

![Fig. 6. Magneto-optical resonances obtained from backscattered fluorescence light using lock-in amplifier (a) and low-pass filter (b)-(d).](image)

![Fig. 7. Calculated magneto-optical resonances for $F=2\rightarrow F'=3$ transition:](image)