Optical Magnetometer: Quantum Resonances at pumping repetition rate of 1/n of the Larmor frequency

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Abstract

Quantum sub-resonances at 1/n are observed experimentally in a SERF type pulsed optical magnetometer. This is a type of synchronization phenomenon. Sequential short pump laser pulses of 70-200 μsec, circularly polarized are employed for optical pumping of the alkali vapor in 5 layers shielded magnetometer. The repetition rate of these pulses is 1/n of the Larmor frequency at specified magnetic field produced within the shield. Signal resonances are obtained at these 1/n frequencies. Mixed alkali atoms of K and Rb cells are studied, where one specie is pumped while the probe is on the other species that is polarized by spin exchange. The effect of spin destruction, spin exchange and collisions are studied, in order to account for the width of the resonances. Quantum calculations of three levels Lambda model for this phenomenon exhibit a dip at the resonance frequency in the absorption spectrum for both cases of pulsed and CW pump modes and an evidence for EIT. Such a decrease in the absorption intensity which is the result of quantum interferences may cause false sensitivity in the optical magnetometer and in the determination of tiny magnetic fields. Methods in which the magnetic detection employ only frequency changes do not suffer from this effect.

Introduction

Atomic magnetometers are studied vigorously in the last decades since a record sensitivity below 1 fT was achieved in a SERF configuration. Various configurations of optical magnetometers as well as their applications have been published along and in comparison, with non-optical highly sensitive magnetometers. A dual use, scalar and vectorial optical magnetometer and an unshielded pulsed magnetometer increase the applications span. In this article, we present a particular effect of quantum resonances achieved in a SERF configuration Atomic magnetometer by repetitive short pulse optical pumping, in which the repetition rate is 1/n the atom internal Larmor frequency.

A quantum theoretical three levels Lambda scheme was studied, which presents a simplification of the active alkali atoms levels. It shows that the presence of a sequential pulsed pump at a repetition rate equal to 1/n of any internal atomic frequency exhibits resonances. Expanded details of this model are given below. In optical magnetometer, magnetic interaction (μ·H) generates Zeeman splitting at the Larmor frequency and characteristic internal Zeeman
frequency. Thus, one expects to see the above resonances when the pump is a sequential at repetition rate of $f = 1/n$ the Larmor frequency. The present study exhibits this effect in the particular case of the Atomic magnetometer. We utilize the scalar mode of the optical magnetometer in a pulse mode excitation and measure the temporal response of the decaying magnetic moment or the population in the related Zeeman level.\textsuperscript{5} Intuitively, this effect is a result of the synchronization of the pump rate with the internal eigenfrequency of the quantum system, the alkali atom in the present case of the Optical magnetometer. Although the repetition rate has no quantal characteristics, Quantum resonances occur due to the synchronization. Quantum calculations of three levels system model indicate possible false absorption measurement at resonance frequency employed in various magnetometry sensitivity level on the order of $10^{-8}$, due to possible EIT effect utilizing pump-probe in CW or pulsed modes. Methods in which the magnetic detection employs only frequency changes do not suffer from this effect.\textsuperscript{6}

**Experimental**

In an atomic magnetometer alkali atoms in a glass cell are optically pumped by a circularly polarized laser beam. A magnetic field rotates the polarized atoms at Larmor frequency depicted by a linearly polarized probe beam. In a SERF configuration of an atomic magnetometer the cell is warmed to increase alkali density and is magnetically shielded to reduce to minimum any external fields. In this experiment, we used a pulsed mode of optical pumping. Pulses with duration of less than 1 msec at a variable repetition rate are used. The schematic of the pulsed operation of the magnetometer is shown in Fig. 1. A glass cell containing a mixture of Rb and K alkali atoms, 52 Torr of $\text{N}_2$ and 2.5 amagats He buffer gas is placed inside a five-layer $\mu$-metal magnetic shields. The cell is heated to 180°C by hot air flow. Magnetic fields inside the shields were nulled by a system of 3 pairs of compensation coils, producing calibrated, uniform magnetic fields along $x$, $y$, and $z$ directions.
In the present work, a mixture of two active alkali atoms, Rb and K, was used in which we separate the pump from the probe/detection processes. In this method, there is optical pumping on one of atomic species resulting in population of the extended state of the Zeeman level. This process involves many cycles of excitations, collisions, decays and thermal noise. This is followed by spin exchange to atoms of the other species, that encounters only direct exchange of the spin, avoiding many noise related effects. Probing is done on the second alkali. Thus, reduction of the "noise" on the detection signal is obtained, as evidenced by our experimental results and mentioned elsewhere.\textsuperscript{9,10}

Optical pumping was accomplished by circularly polarized laser light propagating in the $z$-direction tuned to the center of the D1 line of the pumped alkali atoms. The pump has been performed with “\textit{Intense}” CW Laser Diode. However, the diode is adapted to pulse mode and driven with DEI PCO-6141 high power pulsed current source. The laser wavelength was tuned to the atomic transition by an external cavity Littrow configuration. Fig. 2 presents the laser pulse shape, measured with DET 10 A/M Thorlabs Si Detector. (1 ns Rise Time, 0.8 mm$^2$ sensor area). Laser pulse shape of 200µs and 70 µs pulse durations are shown.

![Fig. 1. Experimental set-up.](image)
A linearly polarized probe beam generated by New Focus Velocity tunable diode laser, propagates in the $x$-direction. It is detuned by 0.8 nm from the D1 or D2 line of the probed alkali atom to reduce absorption and depolarization. The polarization plane of the probe beam is modulated by a Faraday modulator at a frequency $\omega_m \approx 3.1$ kHz. The probe beam passes through the cell and an analyzer, and is detected by a ThorLabs DET36A photodiode. The photo diode is connected to a SRS SR850 digital Lock-in amplifier. The filtered signals are connected to an SRS SR760 spectrum analyzer and a LeCroy Wave-Runner 44XI 400MHz Oscilloscope for analysis.

The Bloch equation describing the magnetometer is

$$\frac{d}{dt} S = \gamma S \times B - IS$$  \hspace{1cm} (1)

Where $\gamma$ is the gyromagnetic ratio, $I$ is the spin relaxation rate due all causes. The analytic solution of this equation is:

$$P_x = e^{-It} \sin(\gamma B_z t)$$  \hspace{1cm} (2)

Fig. 3 presents a typical magnetometer response to a pump laser pulse, a decaying sine function oscillating at the Larmor frequency.$^5$

Fig. 3. A typical magnetometer response to a pumping laser pulse when a magnetic field is applied to the atoms, after the end of the pumping pulse. The decaying sine function oscillates at the Larmor frequency.$^5$ Note the amplitude of peak to peak (pk-pk) of the first sine in the oscillogram.
We investigated the magnetometer signal versus the pulsed pump repetition rate. A DC magnetic field was applied in the y-direction, corresponding to $\omega_L$ Larmor frequency of the atoms in the vapor cell. We found a synchronization between the Larmor frequency $\omega_L$ and the pump pulse repetition rate, $f$, leading to resonance features at $2\pi f = \omega_L / n$, where $n$ is integer (inverse harmonics).

We present in Fig. 4 the first "peak-to-peak" amplitude response of the magnetometer as function of pulse repetition rate by dots. The Larmor frequency is 220 Hz. Resonances at 110Hz, 73.3Hz, 55Hz, (1/2, 1/3, 1/4… of the Larmor) are clearly observed corresponding to $f = \omega_L / n$. A fitting of the obtained resonance curve with analytical solution for Bloch equation of single shot pumping is presented by the solid line. There is a very good agreement between the experiment and theory of both the Bloch equation and a good qualitative agreement with the quantum model presented in Reference [7] and in the theory chapter of this paper. As expected, the width of the resonances vs repetition rate decreases as the repetition rate decreases.

![Graph](image)

**Fig. 4.** First oscillation peak-to-peak amplitude vs pulse repetition rate. Analytical solution curve is shown by the solid line, dots are experimental values.

**Mixed atoms experiments:**

Our vapor cell contains Rb and K metals allowing several pump-probe configurations. One specie is pumped while the probe was on the other species that is polarized by spin exchange. We investigate several options looking for better signal to noise measurements and narrower resonances. The configurations are:

1. Rb D1 atoms pumping and K D1 atoms probing,
2. K D1 atoms pumping and Rb D1 atoms probing,
3. Rb D1 atoms pumping and Rb D2 atoms probing,
4. K D1 atoms pumping and K D2 atoms probing.

For each configuration, the magnetometer lasers system was rebuilt at the required wavelengths. A constant magnetic field corresponding to 180Hz and 220Hz Larmor frequencies was applied to the atoms in the vapor cell. Amplitude of the first oscillation $A_{pk-pk}$ (Fig.3) of the magnetometer signal was measured as a function of pumping repetition rate. The obtained resonance curves for different magnetometer configurations are summarized in Fig. 5 and Fig. 6. with a detailed description in figure captions.

![Graph](image-url)

Fig. 5. Amplitude of the first oscillation of the magnetometer signal vs. pump pulses repetition rate for different magnetometer configurations Rb(pump)-K(probe), K(pump)-Rb(probe), Rb(pump)-Rb(probe), K(pump)-K(probe). Magnetic field corresponding to 220Hz Larmor frequency is applied to the atoms.

Signals from an optically pumped atomic magnetometer are proportional to the x component of the spin polarization. Therefore, the Bloch equations describing the behavior of the spin polarization are used to simulate the behavior of these magnetometers. The Bloch equations for such a magnetometer using a hybrid cell containing high pressure (2.5 amagats) of He as buffer gas, K vapor and Rb vapor at a time when no optical pumping is employed is given by Equations 3 and 4.: 

$$\frac{d}{dt} S^K = \gamma^K S^K \times B + R_{SE} S^{Rb} - (R_{SD} + R_{SE}) S^K$$  \hspace{1cm} (3)
\[
\frac{d}{dt} S^{Rb} = \gamma^{Rb} S^{Rb} \times B + R_{SE} S^K - (R_{SD}^{Rb} + R_{SE}^{Rb}) S^{Rb} \tag{4}
\]

Where \( \gamma \) is the gyromagnetic ratio, \( R_{SD} \) is the spin relaxation rate due to spin destruction collisions, and \( R_{SE} \) is the spin relaxation rate due to spin-exchange collisions. The diffusion term is neglected because of the high pressure of the buffer gas. There exists an analytical solution to eqs. 3 and 4. Using the simple geometry depicted in Fig.1, and in the simplest case, assuming initial conditions such that at the end of the pump pulse, the polarization of the 2\(^{nd}\) species is zero and the polarization of the 1\(^{st}\) species is in the x axis direction, and further assuming that the spin exchange rate and the spin destruction rate are the same for the two species, the polarizations of the two species are given by Eq. 5 and Eq. 6:

\[
P_{1x} = e^{-\gamma B_z t} \sinh(R_{SE+SD} t) \sin(\gamma B_z t) \tag{5}
\]

\[
P_{2x} = e^{-\gamma B_z t} \cosh(R_{SE+SD} t) \sin(\gamma B_z t) \tag{6}
\]

The time evolution of the polarizations of the two species is depicted in Fig.7.

Fig. 6. Normalized resonance curves for 180Hz Larmor frequency for the different pump-probe magnetometer configurations. Magnetometer signal for Rb pumping is stronger, but the FWHM is wider for Rb pumped atoms at all the measured magnetic fields and narrower for potassium pumping.
Bloch equations formalism for the resonance curves

The Bloch equations describing the atomic system mentioned above are given in [10]. In the simple geometry of Fig.1. The Bloch equation can be solved analytically. Using the pulsed pump CW probe scheme, one can reconstruct the waveforms depicted in Fig. 3. and calculate the amplitude of the first oscillation vs. the pulse rate of the pump laser. The analysis results are presented for two hybrid magnetometers: Potassium pumped and Rubidium probed and vice-versa. The results are given in Fig. 8 and Fig. 9.
Fig. 8. Normalized resonance curves for 120Hz Larmor frequency for the K pumped Rb probed configuration. The dots represent the experimental points, the solid line shows the calculation results. The rate of spin destruction is calculated to be $\Gamma_{SD} = 6.6 \text{ sec}^{-1}$. 
Fig. 9. Normalized resonance curves for 120Hz Larmor frequency for the Rb pumped K probed configuration. The dots represent the experimental points, the solid line shows the calculation results. The rate of spin destruction is calculated to be $\Gamma_{SD}=13 \text{ sec}^{-1}$.

**Spin destruction rates of Potassium and Rubidium:**

It is obvious from the experimental data and the analysis presented in Figs. 5-9 that the spin destruction (SD) rate for Rb pumped spin is much larger than that of K pumped spins. The alkali metals cell used in the experiment consisted of 2.5 amagats of $^4\text{He}$, 60 torr of $\text{N}_2$, and a mixture of K and Rb delivering a ratio of 1:20 of vapor density of Rb and K respectively at 180°C. Using the data given in ref. [10] for the spin-destruction (SD) cross sections of K and Rb by the various atoms in the cell, and the slowing down factor q mentioned in ref. [10] which is 6 for K and 10.8 for natural Rb$^1$, one can predict that the SD rate of Rubidium in our experiment should be higher by a factor of 4 than that of Potassium. The main source of this factor is the higher cross section for SD by $^4\text{He}$ gas. Our measurements yield a factor of 2, in reasonable agreement with the phenomenological model prediction.
Extended Theory

In the sake of completeness, we briefly sketch here our previous work, in which an atomic SERF magnetometer was modelled as a three-level system driven by two lasers. The following system Hamiltonian has been used

\[ H = \hat{d}_{ac} \cdot \vec{E}_{ac} J_1(t) + \hat{d}_{bc} \cdot \vec{E}_{bc} J_2(t) + \hat{d}_{ac} \cdot \vec{E}_{bc} J_2(t) e^{-i\omega t} + \hat{d}_{bc} \cdot \vec{E}_{ac} J_1(t) e^{-i\omega t} + H.c., \]  

(7)

where \( \hat{d}_{ac} = d_{ac}\sigma_{ac} \) and \( \hat{d}_{bc} = d_{bc}\sigma_{bc} \) are dipole operators of the corresponding transitions, and \( \sigma_{ac} = |c\rangle\langle a| \) and \( \sigma_{bc} = |b\rangle\langle a| \) are atomic raising and lowering operators. The first two terms in Eq. (7) represent resonant interaction between the driving fields \( E_{ac} \) and \( E_{bc} \) and the corresponding transitions. The last two terms describe cross interaction between the driving field \( E_{ac} \) (\( E_{bc} \)) and the transition \( |b\rangle \rightarrow |c\rangle \) (\( |a\rangle \rightarrow |c\rangle \)), in which case the fields are automatically detuned by the atomic frequency \( \pm \omega_{ab} \) from the corresponding transitions. Functions \( f_1(t) \) and \( f_2(t) \), of the type

\[ f(t) = \sum_n \{ \delta(t - n\tau) \} \]  

(8)

which determine the time dependencies of the driving field amplitudes \( E_{ac} \) and \( E_{bc} \), respectively, are chosen either as \( 1 \), in the case of cw field, or as \( g(t - n\tau) \) in the case of a sequence of short pulses, or any combination thereof. The function \( g \) here is taken to be Gaussian, but any appropriate pulse function can be used as well. Solving numerically the master equation with the Hamiltonian (7) the coherences and level populations were calculated for various values of the system parameters. The dependence of the oscillation amplitude of \( \text{Im}[\rho_{bc}] \), which is responsible for absorption on the probe transition \( |b\rangle \rightarrow |c\rangle \), on the repetition rate \( \omega_{\text{Rep}} \), exhibits, apart of intuitively expected main resonance at \( \omega_{\text{Rep}} = \omega_{ab} \), other resonances at fractional frequencies \( \omega_{\text{Rep}} = \omega_{ab}/n \) with \( n \) being an integer number, which agrees well with experimental result shown in Fig. 10.
Fig. 10. Oscillation amplitude of the absorption on the probe transition as a function of the pulse repetition rate $\omega_{\text{Rep}}$ (solid line). Apart from expected resonance at $\omega_{\text{Rep}} = \omega_{ab}$ there are additional resonances at $\omega_{\text{Rep}} = \omega_{ab}/n$, where $n$ is an integer number.

Further calculations show electromagnetically induced transparency (EIT) and Coherent Population Trapping (CPT) shown in Fig. 11, as expected in a three levels system.

Fig. 11. Populations of atomic levels as functions of time at quantum $1/n$ resonances. There is clear CPT at fractional resonances $\omega_{\text{Rep}} = \omega_{ab}/n$, at $n > 3$. 
It is important note for atomic magnetometer measurements that rely on measuring absorption intensity of the probe, particularly in the case where single laser is used for both pump and probe CW lasers. There is coherence among the pump and probe that may result experimentally in EIT. This is detrimental for the measure of the absorption amplitude, and in turn the detection sensitivity of a magnetometer. We note that EIT is readily observed in Alkali atoms. This is observed in our simplified theoretical calculation, Fig. 12. This may jeopardize the accuracy in such measurements in atomic magnetometers, as the probe intensity measurement may not reflect truly the change due to the magnetic field. This problem is not present in the case where magnetic field is measured solely from changes in the Larmor frequency rather than absorption intensity.

Fig. 12. Comparison of the probe absorption spectra for CW (bottom line) vs pulse (upper line) pump. EIT is clearly manifested in both cases. This may cause false positive sensitivity in optical magnetometer measurements of minute magnetic field, since absorption amplitude may adversely show lesser magnitude than it is in reality.
Another interesting result is obtained at a specific resonant condition when the Rabi frequency $\Omega_{ac} = \vec{a}_{ac} \cdot \vec{E}_{ac}/\hbar$ of the pump laser equals the Larmor frequency $\omega_{ab}$. In this case in addition to $1/n$ resonances there appear more resonances at $\omega_{Rep} = 2\omega_{ab}/(2n+1)$, as can be seen in Fig. 13. These resonances are quite prominent due to resonance condition of $\Omega_{ac} = \omega_{ab}$.

Fig. 13. Fractional resonances at $\omega_{Rep} = 2\omega_{ab}/(2n+1)$ under resonant condition $\Omega_{ac} = \omega_{ab}$.

Intuitively $2/(2n+1)$ peaks result from synchronization of populating $\rho_{cc}$. Population $\rho_{bb}$ of the intermediate level oscillates at Larmor frequency, which in this case is resonant to Rabi frequency $\Omega_{ac}$ of the pumping laser, the population $\rho_{cc}$ of the upper level oscillates at double frequency $2\Omega_{ac}$. This can be seen in Fig. 14.
While population $\rho_{bb}$ of the intermediate level oscillates at Larmor frequency, which in this case is resonant to Rabi frequency $\Omega_{ac}$ of the pumping laser, the population $\rho_{cc}$ of the upper level oscillates at double frequency $2\Omega_{ac}$.

In another condition the $2/(2n+1)$ resonances are extremely weak and not observed experimentally [6].

**Summary**

Operation of the optical magnetometer in a pulsed sequential mode at repetition rate at $1/n$ value of Larmor frequency of the active atom exhibits resonances as function of the pulse pump laser repetition rate. These resonances are in qualitative agreement with quantum theory of three levels Lambda system, pumped in a similar manner.

Interesting narrowing of these resonances is obtained when two alkali atoms are used in which one species acts as the pumping agent and transfers the spin excitation population to the other specie atom. By this method, isolation of the probe from the pump is achieved along with narrower resonances and higher "Q value". In particular, it is found that for this narrowing, the right choice is to use Potassium atom as the pump and the Rubidium atom as the probing one.

An important caution note is presented in this work for measurements of the magnetic field in methods that rely on absorption intensity to determine minute magnetic field changes. These are only reliable when there is no EIT (in CW pump) or absorption dip (in pulsed) effects. The method employing solely measuring changes in the Larmor frequency does not suffer from such problem.
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