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Yangying Fu (傅杨颖), Xiaohu Liu (刘小虎), and Jie Yuan (袁杰)

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Light narrowing of cesium magnetic-resonance lines in a radio-frequency atomic magnetometer

Yangying Fu (傅杨颖), Xiaohu Liu (刘小虎) and Jie Yuan (袁杰)

AFFILIATIONS
1 College of Advanced Interdisciplinary Studies, National University of Defense Technology, Changsha, Hunan 410073, China
2 Luoyang Electronic Equipment Test Center of China, Luoyang, Henan 471003, China

Electronic mail: jieyuan@nudt.edu.cn

ABSTRACT

The magnetic-resonance lines of alkali atoms are broadened considerably by the spin-exchange collisions for a radio-frequency magnetometer operating at high temperature. The resonance linewidths of cesium atoms are derived by solving the relaxation equations. When spin-exchange relaxation dominates, a light narrowing effect is predicted. For the cesium atomic radio-frequency magnetometer operates at high temperature, a remarkable narrowing of the cesium magnetic-resonance lines are observed by increasing the pump power. The Cs-Cs spin-exchange relaxation is partially suppressed by light narrowing. This study helps to expand the applications of radio-frequency magnetometers in the high-sensitivity radio-frequency magnetic-field detection.

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INTRODUCTION

Biomedical applications, fundamental investigations, and radio communication need the devices which can detect radio-frequency (rf) magnetic-field with high-sensitivity. However, the existing optical magnetometers, such as scalar magnetometers, SERF (Self-Exchange Relaxation-Free) magnetometers, and NMOR (Nonlinear Magnetic-Optical Rotation) magnetometers are generally used for the detection of static or quasi-static magnetic fields, with their sensitivity reduces at high frequency. The recently developed tunable rf atomic magnetometers enables the high-sensitivity detection of rf signals. In order to enhance the optical rotation signal, the rf magnetometer typically operates at a high temperature. Nevertheless, the magnetic-resonance lines are broadened considerably by the fast spin-exchange collisions between alkali atoms in this regime. In order to improve the sensitivity of rf atomic magnetometer, the spin-exchange relaxation should be suppressed. To achieve the resonance detection of rf magnetic-field, a large bias field is needed. Therefore, the completely elimination of the spin-exchange relaxation is unrealizable. However, it can be partially suppressed by light narrowing, which provides an alternative for improving the sensitivity of radio-frequency atomic magnetometer.

Some researches have been conducted on the light narrowing effects in optically pumped alkali vapor. N. D. Bhaskar et al. studied the influence of cesium number density on the light narrowing effect. S. Appelt et al. reported the light narrowing effects of optically pumped Rb vapor in high-pressure gas cells, and made a quantitative analysis of the light narrowing effect. T. Scholtes et al. demonstrated an optically pumped Mx magnetometer with miniaturized cesium cell using the light narrowing effect. In this letter, the cesium magnetic-resonance linewidths are derived by solving the relaxation equations in Liouville space. When the Cs-Cs spin-exchange relaxation dominates, a narrowing of the cesium magnetic-resonance linewidth is predicted as the pump power is increased. A cesium atomic radio-frequency magnetometer is built to study the light narrowing effect. The spin-exchange relaxation of cesium atoms is partially suppressed by light narrowing, and the cesium magnetic-resonance line is narrowed considerably.
THEORY AND SIMULATION

For an optical-pumping cell with high-pressure buffer gas, the spin relaxation causes by the spatial diffusion of the polarized atoms is negligible. In the Liouville space, the relaxation equations of alkali atoms can be expressed as

$$\frac{d}{dt} \rho = -\Lambda \rho.$$

(1)

Where $\Lambda$ is the relaxation operator, $\rho$ is the density matrix in the Liouville space. Spin-exchange collisions, spin-destruction collisions and optical pumping are three main mechanisms which destroy the spin-polarization of alkali atoms. For the optical-pumping cell with high-pressure buffer gas, the magnetic-field gradient causes additional relaxation, however, it can be removed by compensation method.\textsuperscript{14–18}

The eigenvalues of the relaxation operator $\Lambda$ are complex numbers for the transverse coherence,\textsuperscript{13} and the real parts of that represent the damping of the free coherence for the Zeeman transitions, and the imaginary parts describe the precession frequencies of the coherence. The real and imaginary parts of the eigenvalues for the relaxation operator $\Lambda$ were derived in Ref. 13. The real part is given by

$$R(F, m|\Lambda|F, \bar{m}) = \delta_{FP} \sum \delta_{m,m^*} \left( R^{2}[1] + 1 - 4 R^2 \right) - R_S \left( \frac{1}{2} \left( 1 - \frac{|F|^2}{2 - 4 R^2} \right) \right),$$

(2)

and the imaginary part is

$$\text{Im}(F, m|\Lambda|F, \bar{m}) = \omega_{m,m^*} \delta_{FP} \delta_{mm^*}.$$  

(3)

Thereinto

$$\left( F, \bar{m} | S_s \right) = \left( \frac{1}{2} \right)^{1/2 - F} \sqrt{1 - \frac{1}{4 |F|^2}}$$

(4)

$$R = R_S + R_D + R_{OP},$$

(5)

$$R_S = PR_S + R_{OP} P_S,$$

(6)

and

$$P = \frac{R_{OP}}{R_{OP} + R_D}.$$  

(7)

Where $R_S$ is the spin-exchange rate, $R_D$ is the spin-destruction rate. $R_{OP}$ represents the optical-pumping rate, which is proportional to the pump power $I_{pump}$ with $R_{OP} = \alpha I_{pump}$. $\alpha$ is the optical-pumping rate constant, which is independent of the pump power.\textsuperscript{7} $\omega_{m,m^*}$ represents the coherence resonance frequency of the Zeeman transition. $P$ is the electron-spin polarization, and $s_z$ describes the mean longitudinal photo spin of the pump light. The mean azimuthal quantum number $m = m - 1/2$ defines a $\Delta m = 1$ ground-state Zeeman transition inside one hyperfine multiplet with quantum number $F$. $I$ is the nuclear spin quantum number of the alkali atoms, and $[I] = 2I + 1$. The factor $Q_m$, describes the probability that the nucleus will have the mean azimuthal number $\bar{m}$, given by

$$Q_m = \frac{2P(1 + P)\bar{m} \left( 1 - P \right)^{\bar{m} + 1} (1 - P)^{1 - \bar{m}}}{(1 + P)^{1 + 1} (1 - P)^{1 + 1}}.$$  

(8)

The coherence strength of the Zeeman transition is proportional to the population difference $\Delta P$ between the two coupled sublevels, and $\Delta P = Q_{m_1} - Q_{m_2}$.

For the $F = 4$, $\bar{m} = 7/2$ ground-state transition of cesium atoms pumped by a left circularly polarized light, the line broadening $\Delta v_l$ due to spin-destruction relaxation, optical pumping, and spin-exchange relaxation is

$$\Delta v_l = \frac{9}{16} R_{SD} + \frac{1}{8} R_{OP} + R_S \left[ \frac{9 - 7P}{16} - \frac{1}{4} \frac{P(1 + P)^{7/2}}{P(1 + P)^{7/2} - (1 - P)^{7/2}} \right].$$  

(9)

The first term on the right-side of Equation (9) is the spin destruction rate. The second term is the optical pumping rate, which is proportional to the pump power. The last term characterizes the Cs-Cs spin-exchange rate. The term in the square brackets is called “the reduction factor” in this letter, which depends only on the electron-spin polarization $P$ of the cesium atoms. The reduction factor decreases as the electron-spin polarization is increased.

The cesium magnetic–resonance line will be broadened as the pump power is increased, since the second term in Equation (9) being proportional to the pump rate, which is called “light broadening”. On the other side, the reduction factor decreases with increasing pump power, thus the linewidth decreases by the spin-exchange relaxation will be compressed, this effect is named as “light narrowing”. For a high density optical-pumping cell, where the spin-exchange relaxation dominates, the light narrowing will exceed the light broadening. Then, the magnetic-resonance line will be narrowed by increasing the pump power.

Light narrowing occurs when most atoms are pumped into the stretched state, i.e. the state with maximum or minimum magnetic quantum number. Conservation of angular momentum requires these atoms remain in the stretched state after collisions, so spin-exchange collisions of atoms in the stretched state do not introduce spin-relaxation. As shown in Equation (8), when all the atoms are pumped into the stretched state, the coherence strength is zero. Hence, in order to generate coherence between two Zeeman sublevels, there must be a fraction of atoms in the non-stretched state. Therefore, the spin-exchange relaxation is partially suppressed rather than completely eliminated.

For the optical-pumping cell with high-pressure buffer gas, the resonance linewidth and the reduction factor are calculated and profiled in Figure 1 for the case of $^{133}$Cs with $R_{SD} = 2000$s\textsuperscript{-1} and $R_{OP} = 20$s\textsuperscript{-1}. Under this condition, the spin-exchange relaxation is the dominant relaxation mechanism of cesium atoms. We see that there is an optimal pumping rate at which the linewidth is minimized. Below that value, the polarization of cesium atoms is small, thus the light narrowing cannot be completely realized. At the optimal pumping rate, the
FIG. 1. The cesium magnetic-resonance linewidth (red) and the reduction factor (blue) as a function of the pumping rate.

complete light narrowing is achieved. While above that value, the line broadening due to optical pumping builds up, and the linewidth begins to increase slowly with the pumping rate.

Figure 2 describes the resonance linewidths of cesium atoms as a function of the pumping rate at four different temperatures, assuming that only spin-destruction collisions, spin-exchange collisions and optical pumping contribute to the spin-relaxation of cesium atoms. The temperatures are T=85°C (magenta), T=80°C (blue), T=75°C (red), and T=70°C (black), respectively. The light narrowing effects are observed as the pumping rates are increased for all the four cases. The theoretical result predicted a 61% reduction in linewidth as the pumping rate is increased at T=85°C. At T=80°C, T=75°C, and T=70°C, the reductions in linewidths are 54%, 46%, 42%, and 37%, respectively. The most pronounced light narrowing effect occurs at T=85°C. Since the cesium atoms number density increases with the temperature, the Cs-Cs spin-exchange rate magnifies with the increasing temperature. Then, the linewidth has a strong dependence on the spin polarization and the pumping rate at a higher temperature. Therefore, the most significant light narrowing effect is observed at T=85°C.

EXPERIMENTAL SETUP

Figure 3 is the sketch of the cesium atomic radio-frequency magnetometer. The spherical Cs cell contains 450 Torr helium and 50 Torr nitrogen, and is heated by the heaters in the oven. Helmholtz coils generate the bias field along the pump beam direction, and the RF coils generate the rf field along the y-axis. The balanced detector is used to detect the optical rotation angle of the polarization plane of the probe beam. By adjusting the current value in the Helmholtz coils, the longitudinal bias field can be varied from a few nT to a few hundred of μT, and the magnetometer can be tuned to detect rf magnetic-field anywhere within kilohertz to gigahertz with high sensitivity.

RESULT AND CONCLUSION

Through fitting the cesium magnetic-resonance line with a Lorentzian curve, the linewidth (FWHM, Full Width at Half Maximum) is extracted from the fitting solution. In our experiments, the measured linewidths are the total ones, which contain the line broadenings arise from radio-frequency irradiation, magnetic-field gradient, and the probe beam. According to the Bloch formula, the measured linewidth is given by

$$\Delta v = \frac{1}{\pi} \left( \Delta v_1^2 + \eta B_1^2 + \Delta v_{\text{gr}}^2 + \Delta v_{\text{probe}}^2 \right)^{1/2}. \quad (10)$$

Where $\Delta v_1$ is the total linewidth causes by spin-destruction collisions, Cs-Cs spin-exchange collisions, and the optical pumping of pump laser, as shown in Equation (9). The second term on the right-side of Equation (10) describes the line broadening due to radio-frequency irradiation. $B_1$ is the amplitude of the rf magnetic-field, and $\eta$ is a coefficient of order $\mu_B/\hbar$. $\Delta v_{\text{gr}}$ characterizes the line broadening causes by the magnetic-field gradient, and $\Delta v_{\text{probe}}$ denotes the line broadening arises from the probe beam. For our experimental conditions, $\Delta v_1 \gg \eta B_1$, $\Delta v_{\text{gr}}$, $\Delta v_{\text{probe}}$, the line broadenings cause by radio-frequency irradiation, magnetic-field gradient and the probe light are small contributions. These elements can be treated as perturbations in our light narrowing experiments.

As shown in Figure 3, the pump power can be adjusted by the attenuator, so the pump power is selected as the variable in our light narrowing experiments. Because the pumping rate is proportional to the pump power, the change in variables is reasonable. The cesium magnetic-resonance signals for two different pump powers are described in Figure 4, the cell temperature is 85°C. The pump powers of the upper trace and the lower one are 1.12 mW and 0.09 mW, respectively. Through fitting the resonance signal with a Lorentzian curve, the FWHM is extracted from the fitting solution.
The pump power of the upper trace is almost 12.5 times larger than the lower one, while the linewidth is 0.85 times narrower. The results show that the cesium magnetic-resonance line is narrowed by improving the pump power.

Figure 5 shows the measured linewidths and magnetometer signal responses as a function of the pump power at $T=\text{85} \, ^\circ\text{C}$. As the results indicate, the measured linewidth decreases dramatically with the pump power when the pump power is lower than 2.10 mW. Above that power, the measured linewidth begins to increase slightly. The amplitude of the magnetometer signal increases with the pump power, and reaches a maximum at the ideal pump power. The cesium magnetic-resonance linewidth drops from 1127.2 Hz at 0.04 mW to 894.5 Hz at 2.10 mW, while the amplitude of the magnetometer response increases from $6.27 \times 10^{-4}$ V at 0.04 mW to $4.94 \times 10^{-3}$ V at 2.10 mW. From a purely phenomenological point of view, the figure-of-merit of the magnetometer can be described by the amplitude-tolinewidth ratio. The results show that the sensitivity of the cesium atomic radio-frequency magnetometer can be improved by light narrowing.

Figure 6 shows the results of the measured linewidths of cesium atoms at three different temperatures $T=\text{70} \, ^\circ\text{C}$, $T=\text{75} \, ^\circ\text{C}$, and $T=\text{80} \, ^\circ\text{C}$. Under our experimental conditions, the line broadenings arise from magnetic-field gradient, radio-frequency irradiation, and probe light are just small.
contributions, which can be treated as perturbations for the study of light narrowing effect. The Cs-Cs spin-exchange relaxation is the dominant relaxation contribution for the three cases, and light narrowing effects are observed as predicted. In addition, the most pronounced light narrowing effect is observed at T=80 °C. A 20% reduction in linewidth is observed as the pump power is increased. The experimental result is lower than the theoretically predicted value. Because of the absorption of cesium atoms by the glass wall, the actual cesium atoms number density is always less than the theoretical one. Figure 7 shows the measured cesium atoms number density at various temperatures. The experimental results indicate that the actual cesium atoms number density is always less than the theoretical one. Figure 8 describes the influence of cesium atoms number density on the light narrowing effect. The results in Figure 8 indicate that the light narrowing effect is more pronounced for a larger cesium atoms number density. Therefore, a smaller linewidth reduction than the theory predicted value is reasonable, considering the absorption of cesium atoms by the glass wall. For a higher temperature, the cesium magnetic-resonance linewidth can be further narrowed. The results proved that light narrowing can be an effective method to suppress the magnetic-resonance lines of radio-frequency magnetometers which operates at high density.

In conclusion, the light narrowing effects are studied both in theory and experiment. When Cs-Cs spin-exchange relaxation dominates over all the other relaxation mechanisms, a light narrowing effect is predicted by improving the pump power. A cesium atomic radio-frequency magnetometer is designed to verify the light narrowing effect. For the magnetometer operates at high temperature, a linewidth reduction is observed as the pump power is increased. The study in this letter provides an attractive alternative for lengthening the spin-relaxation time of alkali atoms, and improving the sensitivity of the radio-frequency magnetometer operates at high density. However, the measurements in this letter were performed at a frequency where the individual lines in the resonance are unresolved by nonlinear Zeeman shifts. The unresolved lines will appear to broaden the resonance. As the pump power is increased, the stretched state will become preferentially populated and the contributions of the other states to the resonance will become reduced. This could lead to an effect similar to that expected from reduced broadening due to Cs-Cs spin-exchange. Therefore, the analysis could be clouded by this effect, along with the uncertainty in the measurement of the cesium atoms number density. This issue will be studied in our future work.
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