Combined effect of light intensity and temperature on the magnetic resonance linewidth in alkali vapor cell with buffer gas*

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One of the peculiar phenomenons in non-zero magnetic resonance magnetometer is that, with the increase of the temperature, the magnetic resonance linewidth is narrowed at first instead of broadened due to the increasing collision rate. The magnetometer usually operates at the narrowest linewidth temperature to obtain the best sensitivity. Here, we explain this phenomenon quantitatively considering the nonlinear of the optical pumping in the cell and did experiments to verify this explanation. The magnetic resonance linewidth is measured using one amplitude-modulated pump laser and one continuous probe laser. The field is along the direction orthogonal to the plane of pump and probe beams. We change the temperature from 53 °C to 93 °C and the pumping light from 0.1 mW to 2 mW. The experimental results agree well with the theoretical calculations.

Keywords: atomic magnetometer, magnetic resonance, linewidth, temperature

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1. Introduction

The atomic magnetic resonance was first observed in nuclear by Rabi[1] and in electron by Zavoisky.[2] In the 1950s, Kaslter *et al.* developed optical methods to polarize and probe the spin of unpaired electron.[3,4] Since then, there appears a lot of researches on different magnetic resonance magnetometers, such as Bell–Bloom magnetometer,[5–7] Mx magnetomter,[8–10] Mz magnetomter,[11,12] CPT (coherent population trapping) magnetometer,[13,14] and SERF (spin exchange relaxation free) magnetometer,[15–18] etc. One of the most important issues of these magnetometers is the magnetic resonance linewidth which is related directly to the fundamental sensitivity of atomic magnetometer by $\delta B = (1/\gamma)\sqrt{\Delta\omega/nVt}$, where $\Delta\omega$ is the linewidth of the magnetometer, n is the density of the alkali atoms, V is the cell volume, t is the measurement time, and γ is the gyromagnetic ratio of the alkali atom.[15] The magnetic resonance linewidth is essentially decided by the sum of the relaxation rate and the pumping rate.[19–22] Relaxation due to collisions and wall are two main relaxation sources. The former is proportional to the density of the alkali atoms. William Happer *et al.* provide a comprehensive description of the relaxation mechanism and collision cross-section constants for different collision relaxation mechanisms and collision pairs in chapter ten of the book "Optically Pumped Atoms".[23] The later is still a research hotspot in vapor atomic spin field. When the alkali atoms encounter the bare glass surface of the cell wall, it can be captured for a little while (10^{-5} s–10^{-7} s) and be completely depolarized by the ions and molecules within the glass.[24] Paraffin,[25] OTS (octadecyltrichlorosilane),[26–28] and alkene[7,29] are all proved to be able to reduce the relaxation rate due to cell wall.

For the linewidth measurement in the relaxation experiments, one usually use a very low light intensity to isolate the effect of pumping rate to the magnetic resonance linewidth. In a real magnetometer, sometimes it is also expected to increase the output signal using a high light intensity. Optimization of the pumping rate thus becomes a subject in the atomic magnetometer research. If sensitivity is limited by technical noise, then one should optimize the quotient of the signal divided by the square of the linewidth. The optimized point happens when the collision relaxation is equal to all the other relaxation processes, including optical pumping.[30] In the case when the sensitivity is limited by the light Schottky noise, the sensitivity is optimal when the pumping rate is three fold of the collision relaxation rate.[31] If sensitivity is limited by the fundamental noise, then one should optimize the linewidth because the fundamental noise is proportional to the square of the magnetic resonance linewidth.

From the point of view of linewidth itself, if the other experiment parameters are kept constant, high temperature is expected to broaden the linewidth as elevating temperature increases the density of the alkali atoms, thus the collision contribution to the linewidth. However, experiment shows that

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the linewidth is narrowed at the beginning of the temperature increasing, i.e. from room temperature to around 70 °C.[5,32] We try to give a quantitative explanation of this phenomenon considering the nonlinear optical pumping rate along the cell and did experiment using Cs cell with buffer gas to verify the explanation. While we were preparing this paper, we noticed a new paper that analyzed the noise of SERF magnetometer considering this nonlinearity.[33] In this paper, we first discuss the linewidth broadening mechanism and give the theoretical explanation of the linewidth narrowing with the temperautre increase. Then magnetic resonance experiments are performed with the temperatures change from 53 °C to 93 °C and light intensity change from 0.1 mW to 2 mW. Finally, the experimental linewidth and the optimization temperature at different light intensity are shown which agree well with theoretical calculations.

2. Theoretical method

In order to explain the phenomenon of linewidth narrowing with temperature increasing quantitatively, we consider the main relaxation sources including the relaxation rate due to the pumping light $R_{\rm op}$ and the relaxation rate due to atom-atom collisions $R_{\rm col}$. As high pressure buffer gas is used in the experiment, we neglect $R_{\rm walls}$ in the theoretical analysis. The contributions of these relaxation sources to the linewidth can be expressed as follows:[31]

$$\Delta\omega = R_{\rm total} = R_{\rm op} + R_{\rm col}, \tag{1}$$

where $\Delta\omega$ is the magnetic resonance linewidth, $R_{\rm total}$ is the total relaxation rate and $R_{\rm op}$ is the pumping rate. $R_{\rm col} = R_{\rm se} + R_{\rm sd}$, where $R_{\rm se}$ is the spin-exchange relaxation rate and $R_{\rm sd} = R_{\rm sd}^{\rm self} + R_{\rm sd}^{\rm He} + R_{\rm sd}^{N_2}$, where $R_{\rm sd}^{\rm self}$, $R_{\rm sd}^{\rm He}$, and $R_{\rm sd}^{N_2}$ are due to collisions of alkali-alkali, alkali-^{4}He and alkali-N_2, respectively.

As the pumping light propagates through the cell, it is attenuated nonlinearly due to the absorption of the polarized alkali atoms. The attenuation depends on the optical depth and atom polarization,[34]

$$\mathrm{d}R_{\rm op}(x) = -n\sigma(\nu)\left(1 - \frac{R_{\rm op}(x)}{R_{\rm op}(x) + R_{\rm col}}\right)R_{\rm op}(x)\mathrm{d}x, \tag{2}$$

where n is the density of the alkali atoms, $\sigma(\nu)$ is the absorption cross section of the pumping light at frequency ν, and x is the position in the cell.

Equation (2) can be solved using Mathematica and the result is a Lambert W-function,

$$R_{\rm op}(x) = R_{\rm col}\mathrm{ProductLog}\left[\frac{R_{\rm op}(0)}{R_{\rm col}} \times \exp\left(\frac{R_{\rm op}(0)}{R_{\rm col}} - n\sigma(\nu)x\right)\right], \tag{3}$$

where $R_{\rm op}(0)$ is the pumping rate at the entrance window.

From Eq. (3) we can obtain numerical solution of $R_{\rm op}$ along the cell length. Figure 1 shows the pumping rate attenuation through the cell with different temperatures.

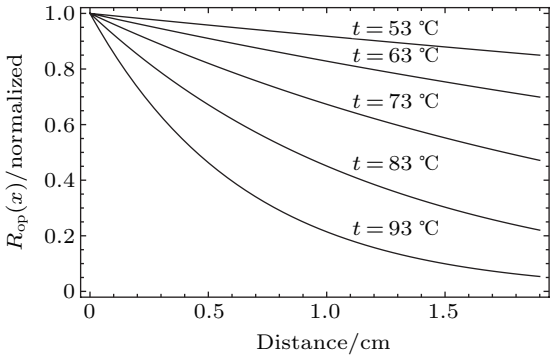


Fig. 1. Attenuation of the pumping rate as a function of distance from the entrance window of the cell at cell temperatures of 53 °C, 63 °C, 73 °C, 83 °C, and 93 °C, respectively.

The average pumping rate of the cell is given by,

$$R_{\rm op-ave} = \frac{1}{L}\int_0^L R_{\rm op}(x)\,\mathrm{d}x, \tag{4}$$

where L is the length of the cube cell, we set $L = 2$ cm to correspond to the later experiment.

The change of $R_{\rm op-ave}$ versus the cell temperature is shown in Fig. 2.

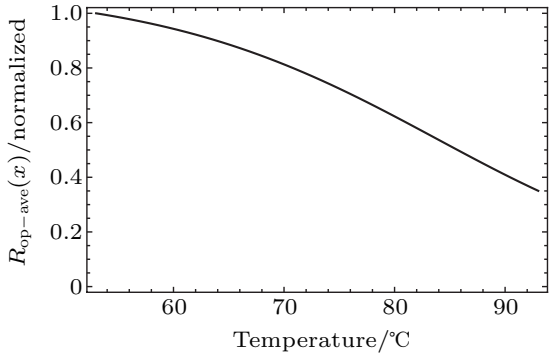


Fig. 2. $R_{\rm op-ave}$ as a function of the cell temperature. This curve is calculated by Eq. (4).

After getting the quantitative relationship between the pumping rate and the temperature, we analyze the collision relaxation rate $R_{\rm col}$ in the following. The general equation for collision relaxation is

$$R = n\sigma\bar{v}, \tag{5}$$

where n is the density of the alkali atoms, σ is the cross section of collisions. The density of Cs atoms is $n = (1/T)10^{21.866+A-B/T}$, here $A = 4.165$, $B = 3830$ according to Alcock,[35] and T is cell temperature in unit Kelvin. Here, the spin-destruction cross sections of Cs–Cs, Cs–He, and Cs–N_2 are 2×10^{-16} cm^2, 3×10^{-23} cm^2, and 6×10^{-22} cm^2, respectively.[36] The spin-exchange cross section of Cs–Cs collision is 2.1×10^{-14} cm^2.[37] The relative velocity of the collisional atoms is $\bar{v} = \sqrt{8k_{\rm B}T/\pi M}$ where $k_{\rm B}$ is Boltzmann

constant and M is given by $1/M = 1/m + 1/m'$ (m and m' are the masses of the alkali atom and the perturber, respectively).

As the electron spin precession is slowed down by the nuclear angular momentum, the relaxation rate of R_{sd} and R_{op} have a slowing-down factor $q = 2I + 1$. For Cs, $I = 7/2$ and $q = 8$. And the relaxation rate R_{se} has a slowing-down factor $q_{se} = 2I(2I-1)/3(2I+1)$ at high magnetic field.[38]

With the slowing-down factor, the collisional relaxation rate of R_{col} is given by

$$R_{col} = \frac{1}{q}R_{sd} + \frac{1}{q_{se}}R_{se}. \tag{6}$$

Figure 3 shows the relation between temperature and average pumping rate, the relation between temperature and collision relaxation rate and the relation between temperature and total relaxation rate according to Eq. (1), Eq. (4), and Eq. (6). From Fig. 3 we can find that R_{op-ave} and R_{col} have an opposite tendency with the temperature change, which results in the decrease of the magnetic resonance linewidth at the initial increase of the temperature.

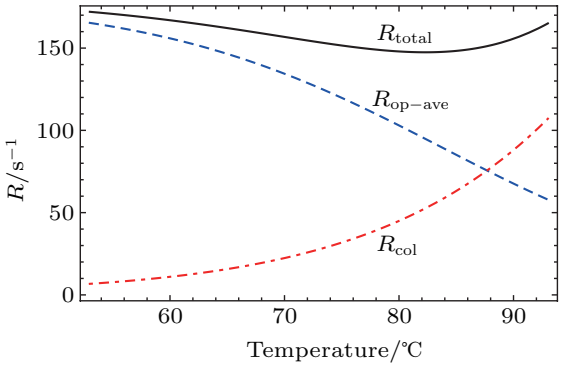


Fig. 3. (color online) Dependence of R_{col}, R_{op-ave}, and R_{total} on the cell temperature. R_{col} is the relaxation rate due to atom–atom collisions, R_{op-ave} is the the average pumping rate of the cell, and R_{total} is the the total relaxation rate. The three curves are calculated by Eq. (1), Eq. (4), and Eq. (6).

3. Experimental setup

The experimental setup is shown in Fig. 4. A diode laser generates a laser beam at the wavelength of 894.6 nm that matches with the D1 line of cesium. The amplitude of the light is modulated by an acoustic optical modulator (AOM) with a duty cycle of 50% at the Larmor frequency to achieve synchronous optical pumping. The atomic vapor cell is cube-shaped (2 cm×2 cm×2 cm) with flat windows, containing a drop of cesium metal, 50-Torr (1 Torr = 1.33322×10^2 Pa) N_2 and 700-Torr ^{4}He, here the N_2 of 50 Torr is used to avoid the radiation trapping and ^{4}He of 700 Torr is used to prevent wall collisions.[15,39] A boronnitride oven is used to heat the vapor cell to the temperature range of 53 °C to 93 °C. The heater is fabricated by using flexible substrate with pairs of heating wire so that the magnetic fields from the heating current can be counteracted. The cell and the oven are placed inside a three-layer μ-metal magnetic shields. The power of the pumping laser is changed by a series of attenuator of neutral density filter with the ODs from 0.1 to 1.5. The magnetic field component along the y axis is produced by a current generator (Thorlabs LDC201). The wavelength of the probe light is 894.6 nm and detuned from D1 resonance of Cs with $\Delta/2$ (Δ is the absorption linewidth of the alkali atoms) and the power of the probe light is 20 μW. Such a probe light is used to monitor the spin polarization and avoid the influence on the magnetic resonance linewidth. The probe light is detected by the polarimeter with a balanced photoreceiver (New Focus Models 2307) and demodulated by a lock-in amplifier (Stanford Research Systems SR830) whose reference signal is provided by a function generator (RIGOL4146) which also drives the AOM. The in-phase (I in Fig. 4) and the quadrature (Q in Fig. 4) signals of the optical rotation of the probe beam are used to measure the linewidth.

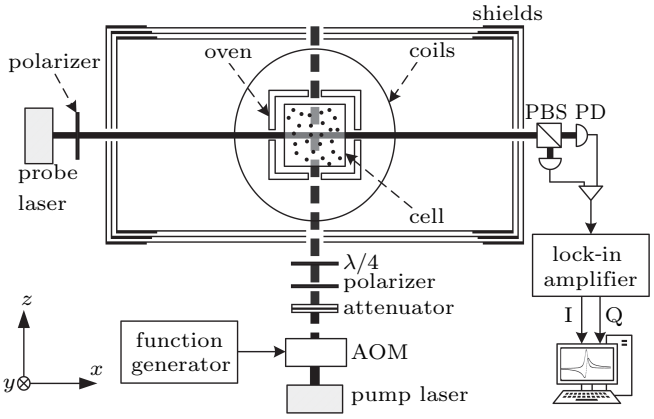


Fig. 4. Experiment setup for the magnetic resonance linewidth measurement.

4. Results and discussion

We measured experimentally the dependence of the magnetic resonance linewidth as a function of the cell temperature at different pump light intensity as shown in Fig. 5. The solid curves are based on Eq. (1), Eq. (4), and Eq. (6), where $L = 2$ cm, $R_{op}(0)$ is calculated by $R_{op}(0) = I\cdot\sigma/A\cdot E$, where I is the the light intensity, $\sigma = 1.83\times10^{-13}$ cm^2 is the absorption cross section of the pumping light, $A = 10$ mm^2 is the cross-sectional area of the laser and $E = h\nu$ is the single photon energy. In the range of the cell temperature from 53 °C to 93 °C, we use the attenuator to get six pump light intensities (0.114 mW, 0.466 mW, 0.891 mW, 1.414 mW, 1.826 mW, and 2.284 mW). At lower light power, the total relaxation rate is dominated by R_{col} since the average pumping rate of alkali atoms is very low. With the increase of light intensity, the average pumping rate come into play. When the light power is higher than 1.414 mW, R_{op-ave} cannot be ignored anymore. In this case, R_{op-ave} dominates the total relaxation rate at lower temperature and R_{col} dominates the total relaxation rate at higher temperature. And there is

a minimum magnetic resonance linewidth at certain temperature. In our experiment, the combination parameters corresponding to the minimum magnetic resonance linewidth are (0.114 mW, 53.3 °C), (0.466 mW, 66 °C), (0.891 mW, 74.7 °C), (1.414 mW, 78.2 °C), (1.826 mW, 83 °C), and (2.284 mW, 83.2 °C), respectively.

The coordination and optimization of the pump light intensity and the cell temperature depends on the magnitude of the signal. The signal is a function of the pumping rate and the density of the alkali atoms, that is, a function of the light intensity and cell temperature. The partial derivatives of the signal with respect to the pump light intensity and cell temperature are set to zero, respectively, and then we get nonzero solutions of the cell temperature and light intensity corresponding to the maximum signal magnitude. The nonzero solutions can be used to the coordination and optimization of the pump light intensity and the cell temperature. When considering the nonlinear of the optical pumping, the optimization of pumping rate means the average pumping rate in Eq. (4).

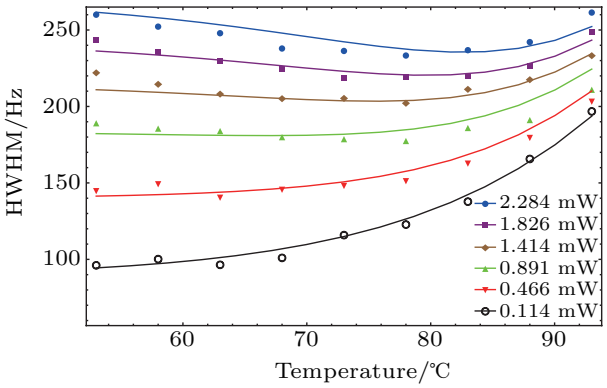


Fig. 5. (color online) Dependence of the magnetic resonance linewidth on the cell temperature for various values of light intensity. Plot markers correspond to experimental data, and the solid red curves are calculated by Eq. (1), Eq. (4), and Eq. (6).

Figure 6 is the temperature corresponding to the minimum magnetic resonance linewidth as a function of the pump light intensity. From Figs. 5 and 6, we can see that the model considering nonlinearity of pumping rate fits well with the experimental data.

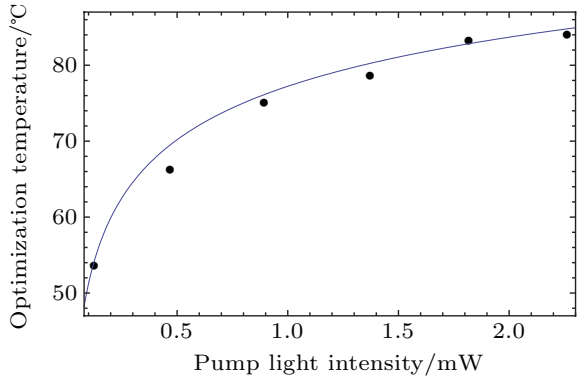


Fig. 6. (color online) Optimization temperature as a function of the pump light intensity. Plot markers correspond to fitted minimum value from the experiment data, and the solid curve is calculated theoretically.

5. Conclusion

In conclusion, we explained the phenomenon that the magnetic resonance linewidth is narrowed with the temperature increase by considering the nonlinear optical pumping rate. And we did specific experiments to verify the explanations. We show that the opposite tendency between $R_{\mathrm{op-ave}}$ and R_{col} with the temperature change results in this phenomenon. In the case of high light power, the pumping rate dominates the total relaxation rate at lower temperature, while the collision relaxation rate dominates the total relaxation rate at higher temperatures. When the light power is low, the magnetic resonance linewidth is mainly decided by the collision so that there is no linewidth narrow with the temperature increase. We also obtained the relation between the temperature corresponding to the minimum magnetic resonance linewidth and the pump light intensity from the measurement results and the theoretical analysis, respectively. The experimental results are in agreement with the theoretical calculations. This work is also helpful in seeking the optimized working point of the non-zero magnetic resonance magnetometer.

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