Frequency-Dependent-Light-Shift Suppression in Light Narrowing Regimes

Yue Chang, Yu-Hao Guo, and Jie Qin

1. Beijing Automation Control Equipment Institute, Beijing 100074, China
2. Quantum Technology R&D Center of China Aerospace Science and Industry Corporation, Beijing 100074, China

Alkali-metal-vapor magnetometers, using coherent precession of polarized atomic spins for magnetic field measurement, have become one of the most sensitive magnetic field detectors. Their application ranges from practical uses such as detections of NMR signals, to fundamental physics research such as searches for the permanent electric dipole moments. One of the main noise sources of the atomic magnetometers comes from the light shift that depends on the frequency of the pump laser. In this work, we theoretically study the light shift, based on the master equation, taking into account the relaxation due to the optical pumping and the collision between the alkali atoms and between the alkali and the buffer gas. To speed up the numerical simulation, and more importantly, to acquire the intuitive picture, we adiabatically eliminate the excited states and obtain an effective master equation in the ground state subspace. This effective master equation shows that in the light narrowing regime, where the line width is reduced while the coherent precession signal is enhanced, the frequency-dependency of the light shift is largely suppressed. Our theoretical predictions agree with the experimental results in cesium magnetometers.

I. INTRODUCTION

Alkali-metal-vapor atomic magnetometers, which have become one of the most sensitive devices for magnetic field detection, find applications in various areas ranging from practical uses such as NMR signal detections, to fundamental physics research such as searches for the permanent electric dipole moments. The physics behind the atomic magnetometers is as follows: polarized alkali-metal spins precess along the magnetic field that needs to be measured, with the precession angle being proportional to the magnitude of the magnetic field. The precession is monitored by a probe beam, while the spins are polarized by optical pumping. However, in the measurement of the Larmor frequency, the AC stark shift is resulted from the (the pump beam here)-matter interaction, behaves as an effective magnetic field to the spins, and subsequently shifts its precession frequency. This frequency-dependent light shift will decrease the measurement accuracy, if the pump beam’s frequency has fluctuations. One way to reduce this frequency-dependency of the light shift is to decrease the pump beam’s intensity, or to increase the line broadenings of the alkali atoms’ excited states, but either of them will lower the atomic polarization, which reduces the precession signal.

Recently, we have found that in the cesium vapor magnetometer, with buffer gas N₂, without tuning the pump beam’s intensity or the excited states’ life time, the light shift’s dependency of the laser frequency can be highly suppressed in the light narrowing regime, where the line width of the spin precession signal is decreased and the fundamental sensitivity, which is inversely proportional to the square root of the spin’s transverse relaxation time, is improved. In the experiment, the alkali atoms’ hyperfine structure levels in the ground state with $F = I - 1/2$, where $I$ is the nuclear spin of the alkali atom, are coupled to the first excited states (D1 transition) via a nearly resonant circularly polarized pump beam propagating along or antiparallel to the static field’s direction. The intuitive picture of this frequency-dependent-light-shift suppression mechanism is as follows: in the light narrowing regime, the $F = I - 1/2$ ground states are pumped (the pump beam’s frequency is resonant with the transition between the ground states with $F = I - 1/2$ and the excited states) and the alkali atoms are highly polarized, i.e., they are mostly pumped to the $F = I + 1/2$ ground state with the quantum magnetic number $m = F$ (or $-F$). Then the dependency of the light shift on the pump beam’s frequency is suppressed since the light frequency is far off resonance with the frequency of the transitions between the $F = I + 1/2$ ground state and the first excited states, i.e., the hyperfine splitting for the ground states is much larger than the excited states’ energy broadening.

To analyze this frequency-dependent-light-shift suppression in details, in this paper, we theoretically study this phenomenon using the master equation, taking into account the light-matter interaction, the relaxation due to the collisions between the alkali atoms and between the alkali atoms and buffer gas. Here, the interaction between the pump light and the alkali atoms is modeled using the dipole approximation and rotating-wave approximation. We show that in the weak-driven limit, where the Rabi frequency—the coupling strength between the ground and excited states—is much smaller than the excited states’ decay rates, the excited states can be adiabatically eliminate and we acquire an effec-
tive master equation in the subspace consisting only the ground states. This can not only dramatically reduce the calculation power and time required to solve the nonlinear master equation (Note that when treating the the spin-exchange interaction, the mean field approximation is applied. Thus, the master equation is nonlinear), but also show explicitly the intuitive picture of the suppression of the light shift dependency on the frequency, as well as the light narrowing effect [21].

The paper is organized as follows: In section II we model the system by the master equation for the density matrix evolution of the alkali atoms. The Hilbert space of this master equation involves all the ground states and the first excited states. In section III we adiabatically eliminate the excited states in the weak-driving limit and acquire an effective master equation in only the ground state subspace. It is shown that this effective master equation can give the rate equations [27] used in many contexts. And when the line broadening of the excited states is much larger than the hyperfine splittings, it is reduced to the master equation that only consists of the electronic spin operators, as shown in Ref. [17], leading to the spin temperature distribution. Then in section IV we study the linear response of the alkali atoms to the small transverse oscillating magnetic field, both analytically and numerically, explaining the frequency-dependent-light-shift suppression in the light narrowing regime. We also compare our theoretical results to the experimental data in the cesium magnetometer and find good agreement. At last, in section V we summarize the work.

II. FULL MASTER EQUATION DESCRIPTION

In this section, we will give the master equation depicting the time evolution of the density matrix $\rho(t)$ of the alkali atoms. This master equation involves all the energy levels in the ground state and the first excited states. The energy levels of the alkali atoms are shown in Fig. 1 where the fine structure levels are denoted by $^2S_{1/2}$ for the ground states and $^2P_{1/2}$ for the excited states. And the hyperfine interaction further splits the energy levels, with the splitting between $F = I + 1/2$ and $F = I - 1/2$ states in the ground (first excited) states as $\Delta_S (\Delta_P)$. The pump beam (the solid red arrow in Fig. 1) propagating along the $z$-direction is left-handed circularly polarized, and its frequency is tuned to induce the D1 transition. This light-matter interaction contributes to the master equation as

\[
\mathcal{L}^{(1)} \rho = -i \left[ H_{\text{int}}, \rho \right] + \Gamma_{sd} \sum_{l = 0, 1} \left( \langle s | \rho | p_l \rangle \langle p_l | s \rangle - \frac{1}{2} \{ \rho, | p_l \rangle \langle p_l | \} \right),
\]

where $\Gamma_{sd}$ is the spontaneous decay rate, $| s \rangle$ and $| p_l \rangle$ are the electron’s orbital states $1s$ and $2p_l$, respectively, and in $| p_l \rangle$, $l$ is its quantum magnetic number. The Hamiltonian

\[
H_{\text{int}} = \Omega (| s \rangle \langle p_1 | + | p_1 \rangle \langle s |),
\]

where we have used the dipole approximation and rotating-wave approximation [25] [26], and $\Omega$ is the Rabi frequency. Here, the radiation trapping [28] is not included, since the quenching [18] gas can largely remove it. Note that without loss of generality, we have assume the pump laser propagated parallel to the magnetic field to be measure, which defines the magnetic number of the hyperfine state. But the model can be easily generalized to the antiparallel case, or equivalently, the parallel laser with right-handed circular polarization. This will not change the conclusion of this paper.

The second kind of interactions include the hyperfine interaction between the electrons and nuclei, and the Zeeman splitting due to the static magnetic field $B_z \hat{z}$ to be measured:

\[
\mathcal{L}^{(2)} \rho = -i \left[ H_{\text{hf}} + H_{\text{Zee}}, \rho \right]
\]

where the hyperfine interaction $H_{\text{hf}}$ is

\[
H_{\text{hf}} = \sum_{M} \Delta_s | s_a, M \rangle \langle s_a, M | - \Delta_p | p_b, M \rangle \langle p_b, M | + \Delta \sum_{F} | p_F, M \rangle \langle p_F, M |,
\]

FIG. 1. D1 transition of the alkali atom. Here, $^2S_{1/2}$ and $^2P_{1/2}$ represent ground and first excited states respectively in the fine structure, and through the hyperfine interaction between the electrons and nuclei, these levels are further split, with the splitting $\Delta_S (\Delta_P)$ for the $^2S_{1/2}$ ($^2P_{1/2}$) states. A pump beam (the solid red arrow) with Rabi frequency $\Omega$ and detuning $\Delta$ (with respect to the frequency difference between the $a = I - 1$ ground and excited states) induces transitions between the ground and first excited states. Another much weaker laser, the probe laser (the dotted green arrow), with linear polarization, is far detuned from the $^2S_{1/2}$ - $^2P_{1/2}$ transition. Thus, it is not included in the theoretical treatment.

$H_{\text{int}}$ depicts the coupling between the pump beam and the alkali atoms is written in the rotating frame with respect to the laser’s frequency as

\[
H_{\text{int}} = \Omega (| s \rangle \langle p_1 | + | p_1 \rangle \langle s |),
\]

$\Omega$ is the Rabi frequency. Here, the radiation trapping [28] is not included, since the quenching [18] gas can largely remove it. Note that without loss of generality, we have assume the pump laser propagated parallel to the magnetic field to be measure, which defines the magnetic number of the hyperfine state. But the model can be easily generalized to the antiparallel case, or equivalently, the parallel laser with right-handed circular polarization. This will not change the conclusion of this paper.

The second kind of interactions include the hyperfine interaction between the electrons and nuclei, and the Zeeman splitting due to the static magnetic field $B_z \hat{z}$ to be measured:

\[
\mathcal{L}^{(2)} \rho = -i \left[ H_{\text{hf}} + H_{\text{Zee}}, \rho \right]
\]

where the hyperfine interaction $H_{\text{hf}}$ is

\[
H_{\text{hf}} = \sum_{M} \Delta_s | s_a, M \rangle \langle s_a, M | - \Delta_p | p_b, M \rangle \langle p_b, M | + \Delta \sum_{F} | p_F, M \rangle \langle p_F, M |,
\]
and the Zeeman splitting Hamiltonian $H_{Zee}$ is
\begin{equation}
H_{Zee} = \sum_{F=a,b} \sum_{M} M \omega_p \langle s_F M \rangle \langle s_F M |.
\end{equation}

Here, $a = I + 1/2$, $b = I - 1/2$, $|s_F M\rangle$ is the hyperfine structure state in the ground ($|s_F M\rangle$) or excited states ($|p_F M\rangle$), with the total angular momentum $F$ and its projection in the $z$-direction $M$, $\omega_p \equiv \omega_s = -\omega_p = \gamma_e B_x/(2I+1)$ is the Larmor frequency of the atom, where $\gamma_e$ is the gyromagnetic ratio of the electron. Note that only the linear Zeeman splitting in the ground states has been included, since other interactions with the magnetic filed, such as the nonlinear Zeeman interaction and the Zeeman splitting in the excited states, are too small to influence the result. And the energies of the excited states have beam shifted with respect to the pump beam’s frequency.

Besides the light-matter interaction, hyperfine interaction and Zeeman splittings, there exist relaxation of the electronic spins mainly resulted from the collision between alkali atoms (spin exchange), and collision between the alkali atom and the buffer gas (spin destruction), which is $N_2$ in the experiment. The two kinds of collisions lead to dissipations in the master equation as
\begin{equation}
\mathcal{L}^{(3)}(\rho) = \gamma \left( S \cdot \rho S - \frac{1}{2} \{ \rho, S \cdot S \} \right)
+ \frac{i}{2} \gamma_{se} \langle S_+ \rangle \left( S_+ \rho S_- - S_- \rho S_+ + \{ \rho, S_+ \} \right)
+ \frac{i}{2} \gamma_{se} \langle S_- \rangle \left( S_- \rho S_+ - S_+ \rho S_- + \{ \rho, S_- \} \right) + \text{H.c.}
+ \Gamma_{pb} \sum_{m=0,\pm 1} A_m \rho A_m^\dagger - \frac{1}{2} \{ \rho, A_m^\dagger A_m \}.
\end{equation}

where $S$ is the electronic spin operator in the ground states, $\gamma_{se}$ is the spin exchange rate, and $\gamma = \gamma_{se} + \gamma_{sd}$ is the total relaxation rate with the spin destruction rate $\gamma_{sd}$. Besides the spin relaxation, the collision also causes excited state broadening, with $\Gamma_{pb}$ the pressure broadening of the $^2P_{1/2}$ states due to the collision of the alkali atoms with the $N_2$ molecule, inducing the alkali atom decaying to the ground state without emitting photons, with the jump operators $A_m$, defined as
\begin{equation}
A_0 = \sum_{m=\pm 1/2} |^2S_{1/2}, m\rangle \langle ^2P_{1/2}, m |,
\end{equation}
\begin{equation}
A_{\pm 1} = \left| ^2S_{1/2}, \pm \frac{1}{2}\right\rangle \left\langle ^2P_{1/2}, \pm \frac{1}{2} \right|.
\end{equation}

It can be shown straightforwardly that in the spin relaxation described by $\mathcal{L}^{(3)}(\rho)$, the spin exchange interaction does not change the mean values of the spin, i.e., $\partial_t \langle S \rangle = 0$ if we set $\gamma_{sd} = 0$ and $\Gamma_{pb} = 0$, while the spin destruction interaction exponentially decrease the spin’s mean values, i.e., $\partial_t \langle S \rangle = -\gamma_{sd} \langle S \rangle$ if we set $\gamma_{se} = 0$ and $\Gamma_{pb} = 0$.

To measure the precession frequency, a small oscillating magnetic field $B_x e_x \cos \omega t$ along the $x$-direction is applied, leading to a time-dependent term in the master equation as
\begin{equation}
\mathcal{L}^{(4)}(\rho) = -i \gamma_e B_x \cos \omega t \ [S_x, \rho].
\end{equation}

As a result, the full master equation is
\begin{equation}
\partial_t \rho = \sum_{n=1}^4 \mathcal{L}^{(n)}(\rho).
\end{equation}

In the superspace, the density matrix can be written as a column vector with dimension $4 \times (4I + 2)^2$. Note that the master equation is nonlinear due to the spin exchange interaction, which makes numerical simulation time and power consuming. On the other hand, more importantly, the physics can not be revealed in such a big set of nonlinear equations. Therefore, in the next section, we will simplify this master equation by adiabatically eliminating the excited states.

III. EFFECTIVE MASTER EQUATION IN THE GROUND STATE SUBSPACE

To gain the physics insights and speed up the numerical solution to the master equation, we will adiabatically eliminate the excited states in the weak-driving limit, where the coupling strength between the ground and excited states is much smaller than the energy level broadening of the corresponding excited state, i.e., $\Omega \ll \sqrt{2/3\Omega} \ll \Gamma_{sd}/2 + \Gamma_{pb}$, which is met in the experiment. Furthermore, when $\gamma_e B_x \ll \gamma$, we can apply the linear response theory [29] and consider the effect of the transverse field at the very end. Therefore, in this section, we will drop the term $\mathcal{L}^{(4)}(\rho)$ in the master equation.

We start the adiabatic elimination by rewriting the master equation [10] as
\begin{equation}
\partial_t \rho = (\mathcal{L}_0 + \mathcal{L}_1) \rho,
\end{equation}
where the perturbation
\begin{equation}
\mathcal{L}_1 \rho \equiv -i [H_{\text{in}}, \rho]
\end{equation}
and the zero-th order term $\mathcal{L}_0 \rho = (\sum_{n=1}^3 \mathcal{L}^{(n)} - \mathcal{L}_1) \rho$.

Then we define two projection operators [25] $\mathcal{P}$ and $\mathcal{Q} = 1 - \mathcal{P}$, where $\mathcal{P}$ projects any operators to the ground state subspace. For instance, when performing in the density matrix, $\mathcal{P} \rho$ gives
\begin{equation}
\mathcal{P} \rho = \sum_{FMF'M'} \langle s_F M | \rho | s_{F'} M' \rangle \langle s_{F'} M' | s_F M \rangle.
\end{equation}

Then the master equation can be written in the $\mathcal{P}$ and $\mathcal{Q}$ spaces respectively as
\begin{equation}
\partial_t \mathcal{P} \rho = \mathcal{P} \mathcal{L}_0 \mathcal{P} \rho + \mathcal{P} \mathcal{L}_0 \mathcal{Q} \rho + \mathcal{P} \mathcal{L}_1 \mathcal{Q} \rho,
\end{equation}
\begin{equation}
\partial_t \mathcal{Q} \rho = \mathcal{Q} \mathcal{L}_0 \mathcal{Q} \rho + \mathcal{Q} \mathcal{L}_0 \mathcal{P} \rho - \mathcal{Q} \mathcal{L}_1 \mathcal{P} \rho.
\end{equation}
\[
\frac{\partial}{\partial t} \rho = \mathcal{L}_0 \rho + \mathcal{L}_1 \rho + \mathcal{L} \rho. \tag{15}
\]

The solution of \( \rho \) to Eq. (15) is

\[
\rho(t) = \int_0^t e^{\mathcal{L}(t-t')} \mathcal{L}_1 \rho(t') \, dt', \tag{16}
\]

where we have assumed \( \rho(0) = 0 \). Substituting this solution of \( \rho(t) \) into Eq. (14), to the second-order of \( \rho \), we acquire the density matrix in the ground state subspace evolving as

\[
\frac{\partial}{\partial t} \rho_g(t) \approx \mathcal{P} \mathcal{L}_0 \rho(t) + \mathcal{P} \mathcal{L}_1 \int_0^t e^{\mathcal{L}_0 (t-t')} \mathcal{Q} \mathcal{P} \rho(t') \, dt'
+ \mathcal{P} \mathcal{L}_0 \int_0^t e^{\mathcal{L}_0 (t-t')} (1 + \int_{t'}^t e^{-\mathcal{L}_0 (t''-t')} \mathcal{Q} \mathcal{L}_1 e^{\mathcal{L}_0 (t''-t')} \mathcal{Q} \mathcal{L}_1 \rho(t) \, d(t') \tag{17}
\]

As a result, the effective master equation for the density matrix in the ground state subspace is

\[
\frac{\partial}{\partial t} \rho_g = -i [H_{hf} + H_{zee}, \rho_g] + \gamma \left( \mathbf{S} \cdot \rho_g \mathbf{S} - \frac{1}{2} \{\rho_g, \mathbf{S} \cdot \mathbf{S}\} \right)
+ \frac{1}{2} \gamma_{se} \langle S_z \rangle (S_z \rho_g S_z - S_z \rho_g S_z + \{\rho_g, S_z\}) + \frac{1}{2} \gamma_{se} \langle S_z \rangle (S_z \rho_g S_z - S_z \rho_g S_z + \frac{1}{2} \{\rho_g, S_z\}) + \text{H.c.}
+ 3 \sum_{n=1}^3 \sum_{FM'FM} \left[ \Gamma^{(n)}_{FM'FM'} J^{(n)}_{FM'FM} \rho_{g} J^{(n)\dagger}_{FM'FM} \right] - \sum_{FM'FM} (\Gamma_{FM} + \Gamma^*_{FM'}) J_{FM} \rho_{g} J_{FM'}, \tag{18}
\]

where the density matrix in the ground state subspace \( \rho_g = \mathcal{P} \rho(t) \), the jump operators \( J^{(n)}_{FM'FM} \) are

\[
J^{(1)}_{FM'FM} = |F', M + 1 \rangle \langle FM|, \tag{19}
\]

\[
J^{(2)}_{FM'FM} = |F' \rangle \langle FM|, \tag{20}
\]

and the effective decay rates are

\[
\Gamma^{(1)}_{FM'FM'} = \frac{\Gamma_{pb} \Omega^2}{\Delta_{FF'}} g_{2;FMFM'g_{1;F'M+1,F'M'+1}}, \tag{23}
\]

\[
\Gamma^{(2)}_{FM'FM'} = g_{2;FMFM'g_{2;F'M'} \sum_{F_1,F_2} \Delta_{FF1}^* \Delta_{FF2}^* g_{1;F_1M+1,F_2M'+1}}, \tag{24}
\]

\[
\Gamma^{(3)}_{FM'FM'} = g_{2;FMFM'g_{1;F'M+2,F'M'+2} \sum_{F_1,F_2} \Delta_{FF1}^* \Delta_{FF2}^* g_{1;F_1M+1,F_2M'+1} g_{2;F_1M+1,F_2M'+1}}, \tag{25}
\]

\[
\Gamma^{(4)}_{FM} = \sum_{F'} \frac{\Omega^2}{\Delta_{FF'}} g_{2;FMFM'g_{1;F'M+1,F'M+1}}, \tag{26}
\]

where we have applied the Markov approximation \([25, 26]\) to replace \( \rho(t') \) by \( \rho(t) \) in the integral, in the assumption that the exponent \( e^{\mathcal{L} t} \) decays in a time scale much smaller than the time scale of \( \rho(t) \). Under the same consideration, and if only the long term behavior is in concern, the upper limit \( t \) in the integration can be extended to \( +\infty \) \([25, 26]\).
with the coefficients
\[ g_{n,F,M|F',M'} = F(n, M) F'(n, M') \]  
and the effective energy difference
\[ \Delta_{FF'} = \Gamma_{pb} + i \Delta_{FF'}. \]
Here, \( F(n, M) \) with \( n = 1, 2 \), is the Clebsch-Gordan coefficient defined as
\[ F(n, M) = \left\langle I, M - \frac{(-)^{n-1}}{2}, rac{1}{2}; \frac{(-)^{n-1}}{2} | FM \right\rangle, \]
and the energy differences \( \Delta_{aa} = \Delta - \Delta_s, \Delta_{ab} = \Delta - \Delta_s - \Delta_p, \Delta_{ba} = \Delta_s \) and \( \Delta_{bb} = \Delta - \Delta_p \). Note that in the derivation of Eq. (18), we have assumed the hyperfine splitting \( \Delta_s \) is the largest energy such that the density matrix element with different \( F \) can be neglected. Moreover, we have assumed that \( \Gamma_{sl}/2 \ll \Gamma \approx \Gamma_{pb} \) and \( \gamma \approx \omega_p \ll \Delta_s \). Thus, the spontaneous decay term in the master equation, and in the effective energy difference \( \Delta_{FF'} \), the contribution from the electronic spin relaxation and from the Zeeman splitting can be neglected.

It is shown from the jump operators \( J_{FM,F'M'}^{(n)} \) that the right-circularly polarized light can increase the angular momentum of the ground state atom by 0, 1, or 2. Especially, when \( \Delta_p \ll \Gamma \), namely, the splitting of the hyperfine structure in the excited state is much smaller than its energy broadening, the energy difference \( \Delta_p \) can be ignored. In this case, \( \Gamma_{FM,F'M'}^{(3)} \approx 0 \). Moreover, if only the excited state splitting \( \Delta_p \) but also the ground state splitting \( \Delta_s \) are much smaller than \( \Gamma \), the master equation can be written in a compact form as in Ref. [17]:
\[ \partial_t \rho_g = \left( \mathcal{L}^{(2)} + \mathcal{L}^{(3)} \right) \rho_g + \Gamma_{OP} \left[ S_+ \rho_g S_- + S_- \rho_g S_+ + \frac{1}{2} \left\{ S_z, \rho_g \right\} - \frac{3}{4} \rho_g \right] - i \Delta_{LS} \left\{ S_z, \rho_g \right\}, \]
where the optical pumping pumping rate
\[ \Gamma_{OP} = \frac{\eta^2 \Gamma}{\Gamma^2 + \Delta^2} \]
and the light shift
\[ \Delta_{LS} = -\frac{\eta^2 \Delta}{\Gamma^2 + \Delta^2}. \]
This master equation (30) leads to the spin temperature distribution [18] [17], where the population is the same in the states with the same \( M \).

When the condition \( \Delta_s \ll \Gamma_{pb} \) is not fulfilled, the spin temperature distribution is not valid. Thus, we consider the generic master equation (18). If further ignoring the coherence between the hyperfine states with different \( F \) in the spin relaxation term in Eq. (18), one can obtain the rate equations [27], which gives the evolution of the diagonal elements of the density matrix, decoupled to the off-diagonal terms. In this case, the steady-state solution to the master equation has only the diagonal terms non-vanishing, namely, the polarization is along the \( z \)-direction and the mean value \( \langle S_z \rangle \) in \( \mathcal{L}^{(3)} \) vanishes.

For the cesium atom, whose nuclear spin is \( I = \frac{1}{2} \), and the energy splittings \( \Delta_S = 9.193 \text{GHz}, \Delta_P = 1.686 \text{GHz} \), with the parameters \( \gamma = 1.31 \text{KHz}, \gamma_s = 1.31 \text{KHz}, \Omega = 0.49 \text{MHz} \), we plot the electron’s polarization \( \langle S_z \rangle \) in the steady-state, as a function of the detuning \( \Delta \) in Fig. 2 for different excited state broadening \( \Gamma \). When the excited state’s decay rate \( \Gamma \) is much smaller than both \( \Delta_S \) and \( \Delta_P \), as shown by the blue solid line in Fig. 2, there are four peaks in \( \langle S_z \rangle \), each of which corresponds to a resonant transition between the ground and excited states. On the other hand, when \( \Gamma \approx \Delta_P \) and \( \Gamma \ll \Delta_S \), as shown by the red dashed line in Fig. 2, the excited states with different \( F \) can not be distinguished, thus two of the closest peaks merge and only two peaks are left in \( \langle S_z \rangle \).

Moreover, Fig. 2 shows that the polarization is larger when pumping the \( F = b \) ground states ((a) and (b) in Fig. 2) other than pumping the \( F = a \) ground states ((c) and (d) in Fig. 2). This is because pumping the \( F = b \) states will remove more populations from these states, leaving the atom populated mostly in the \( F = a \) ground states, which contribute more to the electron’s polarization. To confirm this, we plot the diagonal terms of the density matrix, i.e., the population distribution in

\[ \Gamma = 200 \text{MHz} \]
\[ \Gamma = 1 \text{GHz} \]

FIG. 2. Electron’s polarization in the steady-state solutions to the effective master equation in the ground state subspace, as functions of the pump beam’s detuning \( \Delta \). For small decay rate \( \Gamma = 200 \text{MHz} \) of the excited state that \( \Gamma \ll \Delta_S \) and \( \Delta_P \) (blue solid line), there are four peaks ((a)-(d) in the electron’s polarization, corresponding to four resonant frequencies \( \Delta_{FF'} = 0 \). Moreover, the populations in different hyperfine states with the detuning at these four peaks ((a)-(d) are shown in Fig. 3(a)-(d), respectively. When \( \Gamma \) is increased so that \( \Gamma \approx \Delta_P \) but \( \Gamma \ll \Delta_S \) (red dashed line), the ground states with \( F = a \) and \( F = b \) can still be distinguished, but this is not the case for the excited states. As a result, there are two peaks, corresponding to \( \Delta_{ab} = 0 \) and \( \Delta_{aa} = 0 \).
different hyperfine states in Fig. 3 at the pump beam's frequencies corresponding to the four resonant peaks (a)-(d) in the solid blue line in Fig. 2. Here, integers from 1 to 16 in the horizontal axis represent the state |4, −4⟩, |4, −3⟩,..., |4, 4⟩, |3, −3⟩,..., |3, 2⟩, |3, 3⟩. Taking two resonant detunings Δ = 0 and ΔS, the population in different hyperfine state shows that the spin temperature distribution, where states with the same m have the same population, is not valid.

In the next section, we will use the effective master equation (18) to study the linear response [29] of the atoms to an oscillating transverse magnetic field.

IV. FREQUENCY-DEPENDENT-LIGHT-SHIFT SUPPRESSION AND LIGHT NARROWING

In the presence of the oscillating magnets field in the x-direction, the master equation reads

$$\partial_t \rho_g = \mathcal{L}_0 \rho_g + \mathcal{L}_1 \rho_g,$$

where

$$\mathcal{L}_0 \rho_g = -i [\hat{H}_b + H_{zz}, \rho_g] + \gamma \left( \langle S \cdot \rho_g S - \frac{1}{2} \{\rho_g, S \cdot S\}\right) + \frac{1}{2} \gamma_{se} \langle S_z \rangle (S_+ \rho_g S_\downarrow - S_- \rho_g S_\uparrow + \{\rho_g, S\})$$

and

$$\mathcal{L}_1 \rho_g = \frac{1}{2} \gamma_{se} \langle S_z \rangle \left( S_\downarrow \rho_g S_\uparrow - S_\uparrow \rho_g S_\downarrow + \frac{1}{2} \{\rho_g, S\} \right) + \text{H.c.} + \mathcal{L}_I \rho_g.$$  (35)

When $\gamma_{se} B_x$ is much smaller than the decay rate $\gamma$ or $\Gamma_{FM,FM'}$, the interaction $\mathcal{L}_I$ can be treated as a perturbation. To the first-order of $\mathcal{L}_I$, $\rho_g$ in the long term limit is composed of three parts

$$\rho_g = \rho_g^{(0)} + \rho_g^{(+)} e^{i \omega t} + \rho_g^{(-)} e^{-i \omega t},$$

where $\mathcal{L}_0 \rho_g^{(0)} = 0,$

$$\langle \mathcal{L}_0 - i \omega \rangle \rho_g^{(+)} + \mathcal{L}_1^{(+)} \rho_g^{(0)} = 0,$$  (37)

with

$$\mathcal{L}_1^{(+)} \rho_g^{(0)} = \frac{i}{2} \gamma_{se} B_x \left[ S_x \rho_g^{(0)} \right] + \frac{1}{2} \gamma_{se} \text{Tr} \left( S_x^+ \rho_g^{(0)} \right) \times \left( S_- \rho_g^{(0)} S_\uparrow - S_\uparrow \rho_g^{(0)} S_- + \frac{1}{2} \{\rho_g^{(0)}, S\} \right) + \text{H.c.}$$  (38)

and $\rho_g^{(-)} = \rho_g^{(+)}$. Note that $\mathcal{L}_1^{(+)}$ is dependent on $\rho_g^{(+)}$ through the mean value $\langle S_x \rangle$. Therefore, $\mathcal{L}_1^{(+)} \rho_g^{(0)}$ consists of two parts: $\mathcal{L}_1^{(+)} \rho_g^{(0)} = \mathcal{L}_0^{(+)} \rho_g^{(0)} + \mathcal{L}_1^{(+)} \rho_g^{(0)}$, where $\mathcal{L}_0^{(+)}$ contains $\rho_g^{(0)}$ and $\mathcal{L}_1^{(+)} \rho_g^{(0)} = \mathcal{L}(4) \rho_g^{(0)}$. As a result, the solution of $\rho_g^{(+)}$ is

$$\rho_g^{(+)} = -\left( \mathcal{L}_0 + \mathcal{L}_1^{(+)} - i \omega \right)^{-1} \mathcal{L}_1^{(+)} \rho_g^{(0)},$$

and the electron's polarization in the x-direction can be written as

$$\langle S_x (t) \rangle = \text{Re} \langle S_x^+ \rangle \cos \omega t - \text{Im} \langle S_x^+ \rangle \sin \omega t.$$  (40)
where \( \langle S^z \rangle = 2 \text{Tr} \left[ S_x (\rho_q^{(0)}) \right] \). Note that \( \langle S^z \rangle \) is a function of \( \omega \), and in the experiment, the Larmor frequency \( \omega_L \) is determined by the zero-crossing \( \omega_0 \) of \( \text{Re}(S^z) \), and the line width \( w \) is defined as half of the difference between frequencies corresponding to the maximum and minimum of \( \text{Re}(S^z) \).

As shown in Sec. [11], there are only diagonal terms in the steady-state \( \rho_g^{(0)} \). Thus, in the superspace [20], \( \tilde{L}_1^{(+)} \rho_g^{(0)} \) is a column vector in the subspace \( \{ |FM \rangle \langle FM \pm 1 | \} \), and \( \tilde{L}_0 + \tilde{L}_1^{(+)} \) is a matrix that does not couple this subspace \( \{ |FM \rangle \langle FM \pm 1 | \} \) to the others. Here, we have ignored the coherence between states with different \( F \), for the same reason as in Sec. [11]. In general, the zero-crossing \( \omega_0 \) and the line width \( w \) are obtained by diagonalizing the matrix \( \tilde{L}_0 + \tilde{L}_1^{(+)} \), which can only be done numerically. But we can analyze the diagonal terms of \( \tilde{L}_0 + \tilde{L}_1^{(+)} \) to acquire some physical insight.

When the Larmor frequency \( \omega_L \) is much larger than the dissipation rates that contribute to the real parts of the eigenvalues of \( \tilde{L}_0 + \tilde{L}_1^{(+)} \), the zeros-crossing \( \omega_0 \) will be around \( \pm \omega_L \), the eigenvalues of \( \tilde{L}^{(2)} \) in the subspace \( \{ |FM \rangle \langle FM \pm 1 | \} \). Here we focus on the the frequency range of \( \omega \) around \( \omega_L \), corresponding to the subspace \( \{ |aM \rangle \langle a, M + 1 |, |bM \rangle \langle b, M - 1 | \} \). Especially, in the highly polarized case, the diagonal element of \( \tilde{L}_0 + \tilde{L}_1^{(+)} \) in concern is in the basis \( |a, a - 1 \rangle \{ aa \} \), with the element \( \omega - \gamma \), where the frequency

\[
\tilde{\omega} = \omega_L + \frac{1}{2I + 1} \frac{\Omega^2 \Delta_{aa}}{I^2 + \Delta_{aa}^2} \tag{41}
\]

and the line broadening

\[
\tilde{\gamma} = \frac{\Omega^2}{2I + 1} \frac{\Gamma}{I^2 + \Delta_{aa}^2} \frac{I + 1}{2I + 1} \gamma + \frac{1}{2I + 1} \gamma_{ex} - \frac{1}{2I + 1} \gamma_{ex} \langle S_z \rangle. \tag{42}
\]

For fully polarized atoms, i.e., \( \langle S_z \rangle = 1/2 \),

\[
\tilde{\gamma} = \frac{\Omega^2}{2I + 1} \frac{\Gamma}{I^2 + \Delta_{aa}^2} \frac{I + 1}{2I + 1} \gamma_{ex} \tag{43}
\]

and the relaxation from the spin-exchange does not contribute to the line width, which makes perfect line narrowing [21][22] possible. It has been shown that pumping the \( F = b \) ground states, i.e., turning the detuning such that \( \Delta \approx 0 \), is more efficient for the polarization. Thus, in the vicinity of this resonant frequency, \( \tilde{\omega} \) can be approximated as \( \tilde{\omega} = \omega_L + \delta \omega \), where

\[
\tilde{\omega}_L = \omega_L - \frac{1}{2I + 1} \frac{\Omega^2 \Delta_a}{I^2 + \Delta_a^2}, \tag{44}
\]

is independent of the pump beam’s frequency, and

\[
\delta \omega = \frac{1}{2I + 1} \frac{\Omega^2 \Delta}{I^2 + \Delta^2} \tag{45}
\]

is the frequency-dependent light shift that causes measurement inaccuracy, if the pump laser’s frequency has fluctuations. Because of the large hyperfine splitting \( \Delta \), the frequency-dependent light shift \( \delta \omega \) can be strongly suppressed.

The diagonal element of the matrix \( \tilde{L}_0 + \tilde{L}_1^{(+)} \) has given an intuitive picture how the dependency of the light shift on the frequency is suppressed in the light narrowing regime. To acquire the quantitative result, we numerically solve Eq. (39) and search for the zero-crossing \( \omega_0 \) and the line width \( w \). The results are shown in Figs. [4] with the same parameters as in Fig. [2] with the magnetic field \( B_z = 0.1 \text{G} \). For \( \Gamma = 200 \text{MHz} \), which is much smaller than \( \Delta_F \), both the excited and ground states with different \( F \) can be distinguished. Thus, in the light shift shown in Figs. [4](a), in the vicinity of the frequency \( \Delta_S \), where the \( F = a \) ground states are pumped, the solid blue line has two zero-crossings corresponding to the two resonant frequencies, and the light shift changes a lot while the frequency varies. However, when \( \Delta \approx 0 \), i.e., when the \( F = b \) ground states are pumped, the frequency-dependent light shift is highly suppressed, leaving the zero-crossing much less visible. This is resulted from the large detuning \( \Delta \), as shown in Eq. (45). Meanwhile, the line width (solid blue line in Figs. [4](b)) has two dips at the resonant frequencies when pumping the \( F = b \) ground states, corresponding to the two polarization peaks in Fig. [2], which is the light narrowing effect. But when pumping the \( F = a \) ground states, two peaks at the resonant frequencies in the line width show up, since the light induced line broadening surpassed the polarization induced light narrowing. When increasing \( \Gamma \) to 1\text{GHz}, the excited states can not be distinguished, thus there are less peaks and dips in the light shift and line width, as shown in the dashed red lines in Figs. [4]. Note that at large detuning limit (-5 and 15\text{GHz} for instance), the light’s effect tends to vanish. As a result, the light shift goes to zero and the line width tends to be a constant, independent of the pump beam’s Rabi frequency \( \Omega \), its detuning \( \Delta \), and the excited states’ decay rate \( \Gamma \) [20].

In the experiment of Cesium magnetometers, the magnetic field being measured is also 0.1\text{G} and is illuminated by a right-handed circularly polarized pump laser propagating parallel to the magnetic field (note that the atoms will be optically pumped to the states \( |a, -a \rangle \) in the absence of the spin relaxation, which is different from Fig. [3] since the polarization of the pump laser is different), the power of the pump beam is 700\text{mW}, shining into the cubic cesium vapor, with its inner size 4 \times 4 \times 4 \text{mm}^3 and the temperature 90 Celsius. Nitrogen—the buffer gas—is of 100 and 700\text{Torr}, respectively. Correspondingly, for the numerical simulation, the Rabi frequency is \( \Omega = 4.1 \text{MHz} \), the spin exchange rate is \( \gamma_{ex} = 1.31 \text{KHz} \), the same as in Fig. [2] and \( \gamma = 1.53 \text{KHz} \), \( \Gamma = 0.66 \text{GHz} \) for the 100\text{Torr Nitrogen case, while for the 700\text{Torr Nitrogen case,} \( \gamma = 1.65 \text{KHz} \), \( \Gamma = 4.2 \text{GHz} \). Moreover, the transverse magnetic field \( B_x = 3 \text{nT} \), which is small enough to assure the linear response theory. With these parame-
FIG. 4. Light shift (a) and line width w (b), as functions of the pump beam’s detuning ∆. The parameters are the same as in Fig. 2, and the light shift has been normalized to the Larmor frequency.

ters, the light shift and line width are plotted in Fig. 5 for the 100Torr Nitrogen case, and Fig. 6 for the 700Torr Nitrogen case. In both cases, the theoretical predictions agree with the experimental data very well, even for some details. For instance, there is point in the vicinity of ∆ = 0, where the first-order derivative of the light shift has a large variance, forming a structure like a “dip”, as shown in the inset in Fig. 5(a). But this “dip” is suppressed by the large energy difference ∆s. In both the 100 and 700Torr cases, it shows that the frequency-dependent light shift is strongly suppressed in the light narrowing regime. We note that the temperature and pressure measured in the atomic vapor might not be very accurate, and the cross section for the collision may vary in different regimes. Thus, the relaxation rates we used for numerical simulations are tuned a little bit around the ones calculated from the density of the gas and the cross section, in order to fit the experimental data better.

V. CONCLUSIONS

We have studied in details the mechanism of the light shift and light narrowing effects in alkali-metal-vapor magnetometers. Starting from the full master equation for the alkali atom’s density matrix, we acquire the effective master equation in the ground state subspace, by adiabatically eliminating the excited states in the weak driving limit. This effective master equation can not only help saving power and time for the numerical simulation, but also reveal the intuitive picture of the frequency-dependent light shift suppression: in the light narrowing regime, the pump beam’s frequency is near the frequency of the transition between the F = a ground states and the excited states, and the atom is strongly pumped to the state |a,a⟩. In this case, the light shift is suppressed since the pump beam’s frequency if largely detuned from the transition frequency between the F = a ground states and the excited states. We compare the theoretical results to the experimental data, and find they agree with each other very well.

We note that the effective master equation we have obtained is generic, which can lead to the spin temperature distribution in the limit that the hyperfine splittings can be ignored when the broadening of the excited states is much larger than it. Therefore, the effective master equation is valid in a wide parameter regime, and is applicable in many other topics to be explored, for instance, in the...
ACKNOWLEDGMENTS

This work was supported by National Natural Science Foundation of China grants 61473268, 61503353 and 61627806.

[1] I. K. Kominis, T. W. Kornack, J. C. Allred, and M. V. Romalis, Nature 422, 596 (2003).
[2] D. Budker, Nature 422, 574 (2003).
[3] D. Budker and M. Romalis, Nat. Phys. 3, 227 (2007).
[4] V. V. Yashchuk, J. Granwehr, D. F. Kimball, S. M. Rochester, A. H. Trabesinger, J. T. Urban, D. Budker, and A. Pines, Phys. Rev. Lett. 93, 160801 (2004).
[5] I. M. Savukov and M. V. Romalis, Phys. Rev. Lett. 94, 123001 (2005).
[6] T. G. Walker and W. Happer, Rev. Mod. Phys. 69, 529 (1997).
[7] N. Fortson, P. Sandars, and S. Barr, Phys. Today 56, 33 (2003).
[8] J. M. Amini, C. T. Munger, and H. Gould, Phys. Rev. A 75, 063416 (2007).
[9] B. M. Roberts, V. A. Dzuba, and V. V. Flambaum, Annu. Rev. Nucl. Part. Sci. 65, 63 (2015).
[10] W. Happer, Rev. Mod. Phys. 44, 169 (1972).
[11] W. Happer and W. A. Van Wijngaarden, Hyperfine Interact. 38, 435 (1987).
[12] W. Happer, Y.-Y. Jau, and T. Walker, Optically pumped atoms (John Wiley & Sons, 2010).
[13] M. Auzinsh, D. Budker, and S. M. Rochester, Optically polarized atoms (Physics of Atoms and Molecules (Oxford University Press, New York, 2010), 2010).
[14] S. H. Autler and C. H. Townes, Phys. Rev. 100, 703 (1955).
[15] W. Happer and B. S. Mathur, Phys. Rev. 163, 12 (1967).
[16] B. S. Mathur, H. Tang, and W. Happer, Phys. Rev. 171, 11 (1968).
[17] S. Appelt, A. B.-A. Baranga, C. J. Erickson, M. V. Romalis, A. R. Young, and W. Happer, Phys. Rev. A 58, 1412 (1998).
[18] S. J. Seltzer, Developments in alkali-metal atomic magnetometry (Princeton University, 2008).
[19] V. Schultze, B. Schillig, R. IJsselsteijn, T. Scholtes, S. Woetzel, and R. Stolz, Sensors 17, 561 (2017).
[20] Y. Guo, S. Wan, X. Sun, and J. Qin, Appl. Opt. 58, 734 (2019).
[21] S. Appelt, A. Ben-Amar Baranga, A. R. Young, and W. Happer, Phys. Rev. A 59, 2078 (1999).
[22] T. Scholtes, V. Schultze, R. IJsselsteijn, S. Woetzel, and H.-G. Meyer, Phys. Rev. A 84, 043416 (2011).
[23] T. Scholtes, S. Pustelny, S. Fritzsche, V. Schultze, R. Stolz, and H.-G. Meyer, Phys. Rev. A 94, 013403 (2016).
[24] D. A. Steck, “Cesium d line data,” (2003).
[25] D. F. Walls and G. J. Milburn, Quantum Optics, SpringerLink: Springer e-Books (Springer Berlin Heidelberg, 2008).
[26] C. Gardiner and P. Zoller, Quantum Noise: A Handbook of Markovian and Non-Markovian Quantum Stochastic Methods with Applications to Quantum Optics, Springer Series in Synergetics (Springer, 2004).
[27] S. Lang, S. Kanorsky, T. Eichler, R. Müller-Siebert, T. W. Hänsch, and A. Weis, Phys. Rev. A 60, 3867 (1999).
[28] A. F. Molisch and B. P. Oehry, Radiation trapping in atomic vapours (Oxford University Press, 1998).
[29] A. L. Fetter and J. D. Walecka, *Quantum theory of many-particle systems* (Courier Corporation, 2012).

[30] W. Happer and A. C. Tam, *Phys. Rev. A* **16**, 1877 (1977).

[31] K. Jensen, V. M. Acosta, J. M. Higbie, M. P. Ledbetter, S. M. Rochester, and D. Budker, *Phys. Rev. A* **79**, 023406 (2009).

[32] G. Bao, A. Wickenbrock, S. Rochester, W. Zhang, and D. Budker, *Phys. Rev. Lett.* **120**, 033202 (2018).