Interactions between non-resonant rf fields and atoms with strong spin-exchange collisions

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We study the interactions between oscillating non-resonant rf fields and atoms with strong spin-exchange collisions in the presence of a weak dc magnetic field. We find that the atomic Larmor precession frequency shows a new functional form to the rf field parameters when the spin-exchange collision rate is tuned. A strong modification of atomic Larmor frequency appears when the spin-exchange rate is comparable to the rf field frequency. This new effect has been neglected before due to its narrow observation window. We compare the experimental results with density matrix calculations, and explain the data by an underdamped oscillator model.

Interactions between non-resonant rf fields and atoms can modify the atomic Zeeman spectroscopy. The pioneering work of Haroche et al. [11] showed that in a system with negligible collisions and rf fields oscillating perpendicular to the bias dc field, atomic gyromagnetic ratio is modified through “rf photon dressing”. This result has been applied to resonant transfer of coherence [2] and precision measurements searching for new physics beyond the standard model [3–5]. There are further investigations extended to cases with more complex orientations or components of the rf fields [6–9]. Moreover, the Hamiltonian describing such interactions is directly related to a class of dynamical control of the trap potentials in cold atom experiments [10, 11].

Eq. [1] shows the modification of the gyromagnetic ratio through a functional dependence on rf field parameters when collisions are negligible.

\[ \gamma(B_{rf}, \omega_{rf}) = \gamma(0) J_0(\gamma(0) B_{rf}/\omega_{rf}), \] (1)

where \( \gamma(0) \) is atomic gyromagnetic ratio without external fields, \( J_0 \) is the zero order Bessel function, \( B_{rf} \) and \( \omega_{rf} \) are the amplitude and frequency of the rf field.

However, spin-exchange interactions are normally the dominant collisions between atoms in alkali atom vapor cell experiments. This interaction conserves total angular momentum of the internal states. Recently it has been used for quantum manipulation to generate deterministic entanglement in a sample of ultracold atoms [12]. When the spin-exchange collision rate \( A_{se} \) is much larger than the atomic Larmor frequency, there exists a spin-exchange-relaxation-free (SERF) regime [13, 14]. In this regime, the observed atomic gyromagnetic ratio is also modified due to the weighted average time of atoms staying in different hyperfine states [15], which in turn makes \( \gamma(0) \) in Eq. [1] smaller than the free atomic gyromagnetic ratio \( \gamma_0 \).

There are connections between spin-exchange collisions and rf fields through their interactions with the atoms. Ref. [16] uses the resonant rf fields to effectively cancel the phase evolution difference between ground hyperfine states, and extend the range of the SERF regime. In this paper, we perform atomic spectroscopy in the presence of non-resonant rf fields and spin-exchange interactions. We focus on the low bias field regime, where the atomic Larmor frequency is much smaller than the rf field frequency \( \omega_{rf} \). By tuning the atomic density, we scan the spin-exchange collision rate over a large range. A region of particular interest appears when \( A_{se} \) is comparable with \( \omega_{rf} \). There we find a surprisingly large modification of the atomic Larmor frequency. We also find the amplitude of this modification decreases rapidly as the bias field increases, which makes this effect only observable in a narrow window of experimental parameters.

We perform the experiments on cubic cells with an external size of 10 mm made from Pyrex glasses. The cells sit inside 5-layer cylindrical \( \mu \)-metal shields with the innermost layer length of 41 cm and diameter of 18 cm. An ac current heated oven is used to control the cell temperature. Fig. [1] shows the experimental setup. We have tested two atomic systems with different nuclear spin numbers. For \( I = 5/2 \), we use a cell filled with \(^{85}\)Rb atoms and 600 torr \( \text{Ne} \) gas. For \( I = 3/2 \), we use a cell filled with \(^{87}\)Rb atoms, \( \text{K} \) atoms with natural abundance, 50 torr \( \text{Ne} \) and 150 torr \( \text{Ne} \) gas. Each experiment cycle is 1 sec in length. In the first half cycle, we optically pump the alkali atoms at zero field using a circularly polarized beam on resonance with the corresponding atomic \( D_1 \) transition. In the second half cycle, we turn off the pumping beam, apply a bias field \( B_0 \) less than 1 mG in the \( y \) direction, turn on a kHz rf field along the \( z \) direction, and probe the spin precession signal with a far off-resonance linearly polarized light using the Faraday rotation effect.

Figure [2(a)] shows a typical experimental data. When the effects of rf fields could be treated as perturbations, we keep the leading terms and fit data using the equation:

\[ y = a e^{-t/T} \sin(\omega_L t + \phi_0)[1 + c_1 \sin(\omega_{rf} t + \phi_1) + c_2 \sin(2\omega_{rf} t + \phi_2)] + b, \] (2)

where \( \omega_L \) is the Larmor precession frequency due to the bias field \( B_0 \), the terms with \( \omega_{rf} \) and \( 2\omega_{rf} \) comes from modulation of the atomic polarization orientation due to the rf field. We define the parameter \( R = \omega_L(\gamma_0 B_{rf}/\omega_{rf})/\omega_L(0) \) \( (\omega_L(0) = \gamma(0) B_0) \) to character-
ize the interaction between atoms and rf fields, and probe the effects of spin-exchange collisions on this interaction from the changes in $R$ as we scan the spin-exchange collision rate $A_{se}$. In the experiment, we tune $A_{se}$ from $10^3$ to $10^5$ s$^{-1}$ by changing the cell temperature from 60 °C to higher than 160 °C. We calibrate $A_{se}$ by measuring $T_2$ at a large bias field in the low polarization limit, and using the nuclear slow-down factors [17].

Figure 2(b) shows $R$ as a function of rf field parameters for atoms with nuclear spin $I = 5/2$, $\omega_{rf} = 2\pi \times 4$ kHz, and $B_0 = 0.3$ mG. As we scan $A_{se}$, $R$ remains a monotonic function of $\gamma_0 B_{rf}/\omega_{rf}$, but shows a complex relation with $A_{se}$, which deviates from the zero order Bessel function in Eq. [1]. To better understand their relation, we fix $\gamma_0 B_{rf}/\omega_{rf} = 0.391$, and plot $R$ as a function of $A_{se}$ in Fig. 2(c). There are two turning points in this plot. When $A_{se} < 6 \times 10^3$ s$^{-1}$, $R$ increases together with $A_{se}$. This is due to the fact that the system starts to enter the SERF regime, the gyromagnetic ratio without external rf fields $\gamma(0)$ decreases when $A_{se}$ increases as shown in the same plot, and $R$ in turn decreases slower as a function of $B_{rf}$ according to Eq. [1]. For $A_{se} > 6 \times 10^3$ s$^{-1}$, $\gamma(0)$ is stable but $R$ shows a non-monotonic relation with $A_{se}$, and a minimum value appears at $A_{se} \sim 3.6 \times 10^4$ s$^{-1}$. This new spectroscopy result shows the strong influence of spin-exchange interactions on rf spectroscopy when $A_{se}$ and $\omega_{rf}$, the spin-exchange collision rate and the “dressing” rf field frequency, are comparable.

We simulate the spin dynamics in this experiment using the standard density matrix formalism [15]

$$\frac{d\rho}{dt} = \frac{\alpha h_{rf} I \cdot S, \rho}{i\hbar} + \frac{\mu_B g_s [B \cdot S, \rho]}{i\hbar} + A_{se} \varphi (1 + 4 \langle S \cdot S \rangle - \rho) + A_{sd} \varphi - \rho) + R_{op} \varphi (1 + 2\dot{s} \cdot S) - \rho,$$

where $\varphi = \rho/4 + S \cdot \rho S$ is the nuclear spin part which is unaffected by spin collisions [19], $B = B_0 \hat{y} + B_{rf} \sin \omega_{rf} t \hat{z}$, $A_{sd}$ is the spin-destructive collision rate, $\dot{s}$ is the spin of the optical pumping beam, and $\langle S \rangle = \text{Tr}(S \rho)$. Using the experimental parameters, we extract $\omega_L$ from the evolution of $\langle S_x \rangle$ in the simulation. To improve the quantitative agreement between the simulation and the experimental data, we have also tried to include an attenuation of 4% of the rf field amplitude in the simulation without changing other independently calibrated parameters. This is probably due to the attenuation of rf fields by materials around the cell which makes us overestimate the rf amplitude using only the coil parameters calibrated in dc fields. The results with and without the extra rf attenuation are shown in Fig. 2(c). We adopt the simulation results using this modified rf amplitude in the rest of this paper.

**FIG. 1:** (Color online) A schematic plot of the experiment setup.

**FIG. 2:** (Color online) (a) A typical experimental data with a fitting curve. The data is taken using the $I = 3/2$ system with $B_0 = 0.3$ mG, $A_{se} = 4.7 \times 10^4$ s$^{-1}$, $\omega_{rf} = 2\pi \times 4$ kHz, and $B_{rf} = 2.6$ mG. (b) Experimental data of $R$ as a function of $\gamma_0 B_{rf}/\omega_{rf}$ at different $A_{se}$, where $I = 5/2$, $B_0 = 0.3$ mG, and $\omega_{rf} = 2\pi \times 4$ kHz. (c) Experimental data (solid circles) and simulation results (lines) of $R$ as a function of $A_{se}$ at $\gamma_0 B_{rf}/\omega_{rf} = 0.391$ in plot (a), where the dash line shows modified simulation result with a $B_{rf}$ 4% smaller than the experimental parameter. This plot also includes the experimental data (triangle points) of $\gamma(0)/\gamma_0$ as a function of $A_{se}$. 


Several experimental parameters contribute to the modified rf spectroscopy in the strong spin-exchange collision regime. Fig. 3(a) shows results at different rf field frequencies. For clear comparisons, we have chosen $B_{rf}$ so that the minimum $R$ is around 0.6 for different $\omega_{rf}$. The results show that the curve shifts with $\omega_{rf}$. To better characterize this shift, we define $A_{se,m}$ as the value of $A_{se}$ at minimum $R$, and plot $A_{se,m}$ as a function of $\omega_{rf}$ in Fig. 3(b). The data shows a linear relation between $A_{se,m}$ and $\omega_{rf}$ with a slope of 1.52, which agrees with the simulation result $A_{se,m} = 1.42\omega_{rf}$ within 10%.

![FIG. 3: (Color online) (a) Experimental (solid points) and simulation results (dash lines) of $A_{se}$ as a function of $A_{se}$ for different rf field frequencies. The data is taken for atoms with nuclear spin $I = 5/2$ at $B_0 = 0.3$ mG. (b) Experimental (solid points) and simulation results (open points) of $A_{se,m}$ at different rf field frequencies $\omega_{rf}$ for atoms with nuclear spin $I = 3/2$ (triangle points) and $I = 5/2$ (square points). (c) Experimental data of the depolarization rate due to the rf fields for $\omega_{rf} = 2\pi \times 4$ kHz (nabla points) and 8 kHz (diamond points), and other parameters are the same as in plot (a).](image)

We further check the same behavior for atoms with nuclear spin $I = 3/2$, which requires larger $A_{se}$ to reach the strong spin-exchange regime due to the larger atomic gyromagnetic ratio. Fig. 3(b) shows the results of $A_{se,m} > 3 \times 10^4$ s$^{-1}$ for atoms with $I = 3/2$, and both the experimental data an the simulation results show a linear slope of 1.30. This value agrees with the $I = 5/2$ system within 15%. Compared with the factor of 2 difference in the gyromagnetic ratio in the SERF regime and a 50% difference in nuclear spin slowing down factor of the transversal spin relaxation for such two systems, we conclude that the dependence of $A_{se,m}$ on the nuclear spin is weak.

We gain physics insight on this observation by using a damped oscillator model. Without the rf fields, atoms follow a spin-temperature distribution due to the dominant spin-exchange collisions \[19\]. The rf field drives atomic transitions between Zeeman levels which disturb the spin-temperature distribution, and the spin-exchange collisions try to drive atoms back to the original distribution, entering as a damping factor for the oscillations. This damping effect increases linearly with $A_{se}$ when $A_{se} \ll \omega_{rf}$, and decreases inversely with $A_{se}$ when $A_{se} \gg \omega_{rf}$ \[20\]. Therefore, we expect the damping effect to be strongest when $A_{se}$ is comparable with $\omega_{rf}$. Because the Larmor precession and rf transitions are coupled together in the spin precessions, the damping effect described above affects the spin precessions in the same way. To characterize the damping effects due to the rf fields, we define the depolarization rate $1/T_{rf}$ by comparing the changes of $T$ in Eq. 2 with and without rf fields. Fig. 3(c) shows the results for $1/T_{rf}$, which confirms the conclusion above. Due to the similarity with the damped oscillator model, we adopt such a model with the damping coefficient equal to $1/T_{rf}$, and the damping ratio is proportional to the ratio of $1/T_{rf}$ to the oscillation frequency. For the oscillating components in Eq. 2 we only need to consider the $\omega_L$ term, because the frequencies in other terms are much larger than $\omega_L$ and damping ratios are negligible. In the underdamped oscillator model, the oscillation frequency decreases as the damping ratio increases. Minimum values of $\omega_L$ and $R$ appear in the region with the largest damping rate, where $A_{se}$ is comparable with $\omega_{rf}$.

According to the model above, an increase in Larmor frequency $\omega_L$ reduces the damping ratio, and results in a decreased modification on $R$. Fig. 4(a) shows the experimental and simulation results for $R$ as a function of $A_{se}$ at different $B_0$, with other parameters the same as those in Fig. 2(b). We find that the contrast of the curve decreases by a factor of 6 as $B_0$ increases from 0.3 mG to 0.6 mG, such a rapid decrease in the contrast would lead to no experimentally observable relation between $R$ and $A_{se}$ when $B_0$ is larger than 1 mG. However, $A_{se,m}$ is weakly dependent on $B_0$ as shown in Fig. 4(b), where $A_{se,m}$ changes less than 20% when $B_0$ increases from 0.3 mG to 0.6 mG.

In conclusion, we have studied the interactions between non-resonant oscillating rf fields and atoms with strong spin-exchange collisions. When the collision rate and the rf field frequency are comparable ($A_{se} \sim 1.4\omega_{rf}$), we
FIG. 4: (Color online) (a) Experimental (solid points) and simulation (dash lines) results of $R$ as a function of $A_{se}$ at different bias fields $B_0$, where $I = 5/2$, $\omega_{rf} = 2\pi \times 4$ kHz, and $\gamma_0 B_{rf}/\omega_{rf} = 0.391$. (b) Experimental (solid points) and simulation (empty boxes) of $A_{se,m}$ for the plots in plot (a).

We observe a strong modification of the atomic Larmor frequency, which is confirmed by density matrix simulations. The amplitude of this modification decreases rapidly when the bias field increases. This effect has been neglected before probably due to its narrow observation window. This new phenomena is explained by an underdamped oscillator model, and the damping effect comes from the disturbance of the spin-temperature distribution determined by the strong spin-exchange collisions. Atomic magnetometers based on the same system studied in this paper has emerged to be a promising and complementary tool in biomagnetic applications [21].

Identification of these effects is helpful in calibrations of atomic gyromagnetic ratio and spin-exchange collision rates in such systems. The presence of strong spin-exchange interactions also changes the position of the minimum gyromagnetic ratio point, where atoms are least sensitive to both magnetic fields and rotations. A system working on such a point could be used for spin control related applications.

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