Multi-state interferometric measurement of nonlinear AC Stark shift

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We demonstrate measurement of quadratic AC Stark shifts between Zeeman sublevels in an $^87$Rb Bose–Einstein condensate using a multi-state atomic interferometer. The interferometer can detect a quadratic shift without being affected by relatively large state-independent shifts, thereby improving the measurement precision. We measure quadratic shifts in the total spin $F = 2$ state due to the light being near-resonant to the D$_1$ line. The agreement between the measured and theoretical detuning dependences of the quadratic shifts confirms the validity of the measurement. We also present results on the suppression of nonlinear spin evolution using near-resonant dual-color light pulses with opposite quadratic shifts.

I. INTRODUCTION

Atom–field interactions often cause shifts in atomic energy levels, referred to as AC Stark shifts in the semiclassical treatment. These shifts often depend on the atomic spin state. Linear and quadratic energy shifts (light shifts) of Zeeman sublevels can arise from spherical tensor operators of, respectively, rank-1 (vector) and 2 (tensor) in the irreducible decomposition of the interaction Hamiltonian [1, 2]. Quadratic shifts enable advanced quantum state manipulation such as dynamical spin control [3] and nuclear–electronic spin entanglement [4]. Quadratic shifts have recently been used to generate the Schrödinger’s kitten state in cold Dy atoms of large spin $J = 8$ [5]. On the other hand, quadratic shifts are often detrimental for precise measurements, such as in atomic clocks. Even a small energy shift can be a dominant uncertainty in state-of-the-art precise measurements. Quadratic shifts are also harmful in spin detection via Faraday rotation [6, 7]. In a Faraday rotation measurement, near-resonant light gives a large signal but may also change the atomic spin state through nonlinear spin evolution due to quadratic shifts [2, 7, 8, 9].

Accurate measurement of quadratic or tensor shifts is important for building a sound basis for quantum control as well as for precise measurements. The tensor shifts in clock transitions in alkali atoms have been measured using the Ramsey method with a hot vapor [5] and a cold-atom fountain [10]. Tensor shifts in cold lanthanide atoms in the ground state have been determined using Kapitza–Dirac diffraction from a pulsed standing wave [11], trap frequency measurement [12, 13] and modulation spectroscopy in an optical lattice [14]. In these measurements [9, 14], the tensor shift is distinguished from other shifts based on the difference in the frequency dependence of the scalar and tensor shifts and/or the polarization dependence of the tensor shift. Tensor shift measurements distinguished in this way tend to be uncertain due to technical issues, such as imperfect polarization control at the atomic position. It is difficult to precisely determine a tensor shift much smaller than a state-independent scalar shift. This is the case for most atom experiments, although lanthanide atoms can have large tensor polarizability at specific light frequencies [11, 14].

In the present study, we demonstrate the detection of quadratic light shifts using a multi-state atomic interferometer [13, 14] in a Bose–Einstein condensate (BEC) of $^87$Rb atoms. This scheme is insensitive to state-independent light shifts and directly measures the quadratic light shift, thereby realizing a sensitive measurement by avoiding the uncertainty in distinguishing the tensor shift from other shifts. Direct tensor shift detection is also advantageous in that we can measure the shift in a particular experimental configuration without needing to change light frequencies or polarization. Furthermore, as we can measure the tensor shift without relying on a priori theoretical knowledge including light frequency and polarization dependence of the shift, the measurement may be used for checking the validity of a theory. Conversely, by verifying that the measured frequency dependence of the quadratic shift is consistent with the theory, the validity of the measurement scheme is confirmed. We confirm the validity of our measurement in this manner. We also demonstrate suppression of nonlinear spin evolution using dual-color light pulses near the D$_1$ transition, with the light frequencies and powers chosen on the basis of the light shift measurement to null the net quadratic shift.

The paper is organized as follows. In Sec. II we present our experimental method and setup. The experimental results are described in Sec. III. We conclude the paper in Sec. IV.

II. EXPERIMENTAL METHOD AND SETUP

We produce a BEC of typically $3 \times 10^5$ $^87$Rb atoms in a vacuum glass cell [15]. The BEC is trapped in a crossed optical trap. The trap is composed of an axial beam at a wavelength of 852 nm and a radial beam at 976 nm.
The axial and radial beam waists are approximately 30 μm and 70 μm, respectively. The axial and radial trap frequencies are measured to be 2π × (123, 16) Hz. A bias magnetic field $B$ of 15 μT is applied along the axial beam along the $z$ axis. We initially prepare the atoms in the $|F, m_z⟩ = |2, 2⟩$ state, where $F$ is the quantum number for the total angular momentum of the atoms in the ground state and $m_z$ denotes the magnetic sublevel.

The experimental configuration for the light shift measurement is shown in Fig. 1(a). We measure the quadratic light shift due to a light pulse near-resonant to the $D_1$ line ($\lambda = 795$ nm), which propagates along the $x$ direction. The $D_1$ light is generated by a distributed feedback (DFB) laser and is frequency offset–locked to a master external cavity diode laser (ECDL). The frequency of the ECDL is stabilized to the $F = 2 \rightarrow F' = 1$ resonance line, where $F'$ represents the total angular momentum for the excited $^2P_{1/2}$ state. The beam power is controlled by an acousto-optic modulator (AOM). The beam is directed to the atoms through an optical fiber after the AOM. The beam is almost collimated before the AOM cell and has a Gaussian profile with a 1/e²-radius of $\omega_0 = 0.75$ mm. We control the polarization state at the atomic position using a half-wave plate (HWP) and a quarter wave plate (QWP) before the atom cell. Just after the cell we adjust the polarization of the light to be linearly polarized. The angle between the polarization plane and the direction of the magnetic field, $\theta$, is adjusted to 54.7°, at which the nonlinear spin evolution of the precessing atoms is averaged over a Larmor period if the light is continuous [7, 17].

We perform atomic interferometry with the time sequence depicted in Fig. 1(b). In this sequence, we use the spin echo method to suppress the influence from low-frequency fluctuations of the bias magnetic field. We apply light pulses between the middle $\pi$ pulse and last $\pi/2$ rf pulse. Each magnetic sublevel experiences an AC Stark shift and acquires a phase shift during the light pulse. Different phase shifts between the magnetic sublevels result in a change in the population of each sublevel after the last $\pi/2$ rf pulse. We measure the sublevel populations by absorption imaging along the $x$ axis after a time-of-flight of 20.6 ms with Stern–Gerlach spin separation (see Fig. 1(c)).

The AC Stark shift due to light near-resonant to the $D_1$ line is derived from the light-shift Hamiltonian [2]:

$$\hat{\mathcal{H}}_{\text{shift}} = \sum_{F'} \frac{\hbar \Omega_{0F}^2}{4\Delta_{F,F'}} \{C_{F,F'}^{(0)}|\vec{\varepsilon}|^2 + iC_{F,F'}^{(1)}(\vec{\varepsilon}^* \times \vec{\varepsilon}) \cdot \vec{F} + C_{F,F'}^{(2)}(|\vec{\varepsilon}|^2 - \frac{1}{3} \vec{F}^2|\vec{\varepsilon}|^2)\},$$

(1)

where $\Delta_{F,F'}$ is the amount of light detuning from the transition frequency between the $F$ and $F'$ states, $C_{F,F'}^{(k)}$ is a rank-$k$ tensor coefficient representing the angular momentum dependence [2] and $\vec{\varepsilon}$ is the polarization vector for the light. $\Omega_0$ is defined by

$$\Omega_0 = \frac{\langle P_{1/2}|d||S_{1/2}\rangle E}{\hbar},$$

(2)

where $\langle P_{1/2}|d||S_{1/2}\rangle = 2.537 \times 10^{-29} \text{ C} \cdot \text{m}$ [18] is the reduced matrix element for the $D_1$ dipole transition and $E$ is the field amplitude. $\Omega_0$ can be expressed using the beam power, $P$, as

$$\Omega_0 = \frac{\langle P_{1/2}|d||S_{1/2}\rangle}{\hbar} \sqrt{\frac{4P}{\pi c^2 \omega_0^2 \eta}} \equiv \sqrt{\eta P},$$

(3)

where $c$ is the speed of light and $\varepsilon_0$ is the electric constant. Hereafter we consider linearly polarized light, which introduces no vector shift and produces a state-dependent shift solely through the tensor component. The state-dependent Hamiltonian can be written as

$$\hat{\mathcal{H}}_{\text{depend}} = \frac{\hbar \Omega_0^2}{4\Delta_{\text{HFS}}} \chi (F_x \cos\theta + F_y \sin\theta)^2,$$

(4)
where $\Delta_{\text{HFS}} = 2\pi \times 814.5$ MHz is the hyperfine splitting between the $F' = 1$ and $F'' = 2$ states and

$$\chi = \sum_{F'} \frac{C^{(2)}_{F,F'}}{\Delta_{F,F'}} \Delta_{\text{HFS}}$$

represents the dependence of the coupling strength on the light frequency. We refer to $\chi$ as the coupling coefficient. In an experiment using the ground $F = 2$ state, $\chi$ depends on the laser frequency as

$$\chi = \frac{\Delta_{\text{HFS}}}{12} \left( \frac{1}{\Delta} - \frac{1}{\Delta - \Delta_{\text{HFS}}} \right),$$

where $\Delta \equiv \Delta_{21}$ (see Fig. 3(b)). Hereafter, we consider detuning with respect to the transition frequency between the $F = 2$ and $F'' = 1$ states.

The spin dynamics for $H_{\text{depend}}$ can be easily described if the quantization axis is selected to be along the direction of the polarization axis, $z'$. The state before sending the light pulse, $|\psi\rangle = \sum m_{z'} \beta_{m_{z'}} |m_{z'}\rangle$, with $\beta_{m_{z'}}$ being the probability amplitude for each sublevel $m_{z'}$ in this frame, evolves under the influence of a rectangular pulse of width $\tau$ and power $P$ into

$$|\psi\rangle = \sum m_{z'} \beta_{m_{z'}} e^{i \chi m_{z'}^2 \xi P \tau} |m_{z'}\rangle,$$

where

$$\xi = \frac{\eta}{4 \Delta_{\text{HFS}}}.$$

Here we consider evolution due to a single pulse for simplicity. The extension to the multi-pulse case is straightforward. We assume that the pulse width, $\tau$, is sufficiently shorter than the period of Larmor precession and neglect evolution due to the magnetic field during the pulses. In Eq. (7) we omit the global (state-independent) phase shift, which has no relevance to the population change in the magnetic sublevels. If the spin evolution during the pulse is purely caused by the light shift, the state after the last $\pi/2$ pulse in the $m_{z}$ basis is written using the Wigner D-matrix $[19]$, $D^j(\alpha, \beta, \gamma)$, as

$$|\psi_{\text{end}}\rangle = D^2(0, -\frac{\pi}{2}, 0) D^2(0, 0, -\phi) D^2(-\theta, 0, 0)^\dagger\ A D^2(-\theta, 0, 0) D^2(0, 0, \phi) |\psi_0\rangle,$$

where $A = \text{diag}(e^{i \xi P \tau}, e^{i \xi P \tau}, 1, e^{i \xi P \tau}, e^{i \xi P \tau})$ represents the time development by light pulses, $\phi$ is the spin angle in the $x-y$ plane with respect to the $y$ axis at the starting time of the light pulse and $|\psi_0\rangle = (-1/\sqrt{4}, i/\sqrt{4}, \sqrt{6}/4, -i/\sqrt{4}, -1/4)^T$ represents the coherent spin state (CSS) along the $y$ axis ($\phi = 0$) as the basis of the $z$ axis. Here $D^j(\alpha, \beta, \gamma)$ is defined in terms of the Euler angles $(\alpha, \beta, \gamma)$ around the $z$–$y$–$z$ axes as

$$D^j_{\alpha,\beta,\gamma}(\alpha, \beta, \gamma) = \langle jq | \hat{R}(\alpha, \beta, \gamma) | jq \rangle,$$

where $\hat{R}(\alpha, \beta, \gamma) = e^{-i \alpha \hat{j}_z} e^{-i \beta \hat{j}_y} e^{-i \gamma \hat{j}_z}$ is the rotational operator [19]. We calculate the magnetization, $m = \langle \psi_{\text{end}} | \hat{F}_z | \psi_{\text{end}} \rangle$, using Eq. (9). The calculated $m$ is a function of $\chi P \tau$, but its explicit expression is too long to show here. We note $m = 2 \cos^3(\chi P \tau)$ if $\theta = 0$ [20].

![Figure 2](image-url) Power dependence of magnetization, $m$, for $\Delta/2\pi$ of (a) $-840$ MHz, (b) $240$ MHz, (c) $340$ MHz, (d) $440$ MHz, (e) $540$ MHz and (f) $640$ MHz. The red symbols (circles and triangles) represent the measured magnetization. The circle data points are used for fitting (see text for details) and the blue solid lines are the fitting curves. The green dashed line is the simulated magnetization including the spontaneous emission effect. The calculation and simulation are performed for a single pulse of $\tau = 667$ ns.
III. RESULTS

A. Measurement of quadratic light shifts

We first detect quadratic light shifts produced by a single pulse of $\Delta/(2\pi) = -840$ MHz, which induces less light-assisted collisional atom losses. The pulse has an almost rectangular shape and its length is fixed to $\tau = 667$ ns. The pulse is applied 0.125 ms after the $\pi$ pulse, when the spin orientation is along the $x$ axis ($\phi = \pi/2$). We confirm the spin direction by spin-sensitive phase contrast imaging.

We observe changes in the sublevel population using a Stern–Gerlach measurement, as shown in Fig. 1(c). We experimentally obtain the magnetization using

$$m = \sum_i i N_i,$$

where $N_i$ is the number of atoms in the $|F, m_z = i\rangle$ state ($i = -2, -1, 0, 1, 2$) after the read-out pulse and $N_{tot} = \sum_i N_i$ is the total number of atoms. The magnetization is plotted as a function of the pulse power in Fig. 2(a).

We fit the data using $(1 - \delta p)\langle \psi_{end}\rangle F_r|\psi_{end}\rangle$, where $\delta p$ is introduced to account for experimental imperfect state preparation and control. In the fitting (blue line in Fig. 2(a)), we use Eq. 9 with $\theta = 54.7^\circ$ and $\phi = 90^\circ$, in correspondence with the experiment. The fitting gives $\chi = -0.0404(8)$. The value in parentheses denotes the standard deviation of $\chi$ calculated from three sets of data.

Next, we measure the quadratic light shifts for other light frequencies with positive detunings $\Delta/(2\pi) = +\{240, 340, 440, 540, 640\}$ MHz. The results for each detuning are depicted in Figs. 2(b)–2(f). We observe large changes in $m$. For these positive detunings, $\chi$ is much larger than that for $\Delta/(2\pi) = -840$ MHz and the observed large change in $m$ is reasonable. The spontaneous emission rates also become large for these detunings and optical pumping due to spontaneous emission may also result in spin change. To evaluate the contribution of spontaneous emissions, we numerically calculate the dynamics of the magnetization using an atomic master equation including spontaneous emission. A theoretically predicted value of the coupling coefficient, $\chi_{tho}$, is used in the simulation. We plot the simulation results in Fig. 2 (green dotted lines). The numerical simulation indicates that the spontaneous emissions are not negligible for beam powers larger than approximately 5 mW, which we were not able to investigate in detail due to the limited available beam power. In the fitting shown by blue lines in Figs. 2(b)–2(f), to obtain experimental values of the coupling coefficient, $\chi_{exp}$, we use data points at beam powers for which the magnetization obtained by the simulation differs from the matrix calculation by less than 0.06 to lessen the effect of spontaneous emissions on the estimation of $\chi$. The fitted values of $\chi_{exp}$ are shown in Table I.

The frequency dependence of $\chi_{exp}$ is consistent with the theoretical curve given by Eq. 10, as shown in Fig. 3(a). Note that interferometric detection does not reveal the sign of $\chi$. We determine the sign of $\chi$ at each $\Delta$ in Fig. 3(a) to coincide with the theoretically determined sign. We also note the exact determination of $\chi$ requires precise calibration of the beam intensity at the atom position. It is more appropriate to analyze the ratio between $\chi_{exp}$ and $\chi_{tho}$ to estimate the validity of the measurement. This ratio is shown in Table I. The sample standard deviation of the ratios, which represents the precision of the measurement, is 0.02 (2%).

B. Suppression of nonlinear spin evolution

We also demonstrate suppression of spin evolution due to a quadratic shift by using near-resonant dual-color light pulses. From Eq. 6, we can see that $\chi$ is positive if $0 < \Delta < \Delta_{HFS}$ and negative otherwise. If we combine negative ($\Delta < 0$) and positive ($0 < \Delta < \Delta_{HFS}$) de-
tuned light with a power ratio of $p_{neg}/p_{pos} = \chi_{pos}/\chi_{neg}$, the quadratic light shift should vanish and nonlinear spin evolution should be suppressed. We use the DFB laser used in the above experiment at a fixed detuning of $\Delta_{+}/(2\pi) = -840$ MHz. We prepare another ECDL for quadratic shift compensation. The ECDL is frequency offset-locked to the master ECDL, and its detuning is set to $\Delta_{+}/(2\pi) = \{240, 340, 440, 540, 640\}$ MHz. The two laser beams are mixed at a non-polarizing beam splitter (NPBS) before the AOM for power control. We adjust the ratio of the DFB laser power to the ECDL power using a HWP and PBS before the NPBS. We set the interval between pulses to 9 pulses to observe the changes in magnetization more clearly. We observe that the change in magnetization for $\Delta = 0$ in Fig. 6. The coupling strength to the atoms via the circular component, as shown in Fig. 4(b), is set to 3 times stronger than that to the linearly polarized beam coupling.

The magnetization change in the experiment is larger than that in the simulation including the spontaneous emissions. In the analysis, we count the number of kicked atoms, $N_{\text{sup}}$, to the total number of atoms, $N_{\text{tot}}$, as a measure of superradiant scattering. In the analysis, we count the number of kicked atoms in the regions adjacent to the zero moment component (see the lower panel of Fig. 4) as $N_{\text{sup}}$. We plot $N_{\text{sup}}/N_{\text{tot}}$ for the magnetic sublevels of $m_z = +1$ and +2.
We consider that superradiance enhances the spin relaxation rate and may increase the magnetization change. We estimate from Fig. 5(a) that the scattering rate is enhanced by up to approximately 30%. The Raman superradiance \[25\] to the \( F = 1 \) state should also contribute to the scattering rate enhancement. We also measure the atom number loss, which changes the population among the magnetic sublevels and causes spin relaxation. However, we do not observe a significant dependence of the loss rates on \( \Delta_+ \). The larger than expected magnetization change might be partly due to the enhancement of spontaneous emission near the critical temperature for Bose–Einstein condensation \[20\].

**IV. CONCLUSIONS AND OUTLOOK**

We have demonstrated the successful measurement of quadratic light shifts using an atom interferometer. The measured dependence of the shift due to detuning is consistent with the theoretical prediction. In addition, we suppressed the influence of nonlinear light shifts by using dual-color light pulses. Dual-color light is applicable to high-precision measurement in a probe, especially for BEC magnetometers \[17\]. A dual-color probe is useful not only for improving the signal-to-noise ratio in spin measurements by increasing the probe strength, but also for preventing disturbances when creating a spin squeezed state via quantum nondemolition measurements \[27–31\].

Further improvements of this measurement scheme are feasible. Reducing technical noises during absorption imaging and spin manipulation by rf pulses will lead to higher sensitivity. The bias magnetic field may produce a slight difference between the experimental data and the theoretical prediction calculated by the Wigner D-matrix and interaction Hamiltonian, in which we have neglected the spin evolution due to the magnetic field. Therefore, to improve the estimation accuracy for the coupling coefficients, it may be effective to use a low magnetic field. These improvements may make it possible to estimate physical quantities such as transition matrix elements and hyperfine splittings.

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