Optical Ramsey spectroscopy of a single trapped $^{88}Sr^{+}$ ion

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Coherent optical spectroscopy has been performed on the narrow $5s\ {}^2S_{1/2}-4d\ {}^2D_{5/2}$ quadrupole transition in a single Doppler-cooled and trapped $^{88}Sr^{+}$ ion. High-contrast optical Ramsey spectra have been observed with fringe visibilities up to ~90%. The visibility decreased as the free precession period was increased, and was limited by the interrogation laser's coherence. The effect of varying the relative phase of the second Ramsey pulse was investigated. By measuring the difference in excitation probability on reversing a 90° relative phase shift between the two Ramsey pulses, we have demonstrated Ramsey's anti-symmetric line shape in the optical domain. A significant advantage of this line shape is the zero crossing at line center and the independence of this center frequency on drifts in signal amplitude. This optical Ramsey line shape is suitable for stabilizing a local oscillator to an atomic reference transition in an optical frequency standard. All observed optical Ramsey signals are well described by a model using the optical Bloch equations.

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I. INTRODUCTION

Ramsey's method of separated oscillatory fields [1], originally developed to improve the accuracy of molecular beam resonance experiments, has found widespread use in precision atomic and molecular spectroscopy [2]. When compared with conventional single-pulse interrogation techniques, Ramsey spectroscopy provides a reduction in the observed linewidth and is insensitive to inhomogeneities in the fields experienced during the free precession period between fields. These advantages led to the technique's use in interrogating the hyperfine clock transition in cesium beam clocks [3]. Laser-cooled atoms enabled rf spectroscopy to be performed in an atomic fountain [4], allowing significantly increased interaction times together with corresponding improvements in resolution.

Early demonstrations of Ramsey spectroscopy in the optical domain were performed using a neon atomic beam [5] and a sodium vapor cell [6]. The prospect of high-precision optical experiments using Ramsey's method was illustrated by the observation of the Sagnac effect in a calcium beam interferometer [7]. The intercombination transition in laser-cooled ^{40}Ca atoms, interrogated using Ramsey's method, now represents the forefront of research into optical frequency standards based on high-Q optical transitions in neutral atoms [8–12]. Optical Ramsey spectra have been reported for similarly narrow transitions in single trapped ions of $^{199}Hg^{+}$ [13] and $^{88}Sr^{+}$ [14], which are also being developed as optical frequency standards. The work of Rafac *et al.* [13] presents results confirming that a Ramsey experiment results in a narrower linewidth when compared to a single-pulse experiment of the same interrogation cycle duration. In addition, high-contrast, high signal-to-noise Ramsey spectra have been used to study decoherence of quantum superpositions of the narrow $4s\ {}^2S_{1/2}-3d\ {}^2D_{5/2}$ quadrupole transition in trapped $^{40}Ca^{+}$ ions [15,16]. Furthermore, Ramsey's technique has been used to characterize ac-Stark shifts induced by an intense off-resonant laser on the same transition [17].

Reported absolute measurements of optical clock transitions in single ions of $^{199}Hg^{+}$ [11], $^{88}Sr^{+}$ [18,19], $^{115}In^{+}$ [20], and $^{171}Yb^{+}$ [21,22] have interrogated the clock transition using a single-pulse excitation scheme, rather than using a Ramsey scheme. The majority of these experiments [11,18,19,21] stabilized the laser to the line center by comparing alternate measurements of a relatively low excitation rate on either side of the Lorentzian line shape.

The characteristic signal observed in Ramsey spectroscopy is a result of quantum interference between the atomic excitation amplitudes of the two temporally separate interactions. Hence, any relative phase shifts between the two oscillatory fields, as experienced by the atomic sample, result in a shift of the observed Ramsey resonance frequency [2]. Unintended phase shifts, due to atomic motion in the free precession period, are significant in the optical domain. In two-photon optical Ramsey spectroscopy, this limitation can be circumvented using a standing wave laser field [23,24]; an atom absorbing a photon from each direction sees a position-independent optical phase. Alternatively, the geometry of Bordé [25], containing four traveling waves, is used in existing ^{40}Ca neutral experiments [8–12]. Systematic uncertainties in such systems can be reduced by sub-Doppler cooling [26–28], and should be reduced further by confining the atomic sample in an optical dipole trap [29]. In contrast to atoms, ions cooled and trapped in the Lamb-Dicke regime are confined to a region small compared to the wavelength of the interrogating fields. Therefore, uncontrolled phase shifts due to the particle's motion are much less of a problem, and single-photon optical Ramsey spectroscopy using two temporally separated oscillatory fields is possible [13–17].

An atomic sample experiencing unintended and *uncontrolled* relative phase shifts between the two oscillatory fields is obviously undesirable. However, Ramsey pointed out significant additional advantages to be gained from a *controlled* variation of the relative phase shift $\delta\Phi$ between the two $\pi/2$

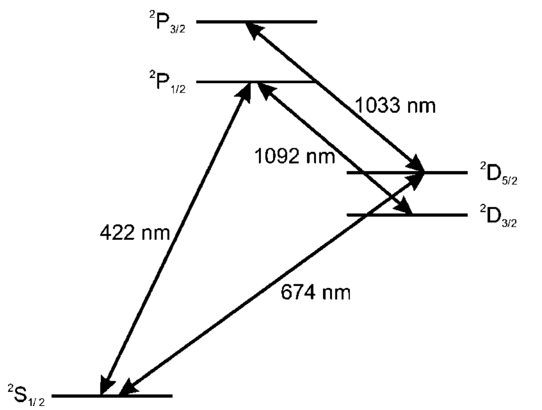

FIG. 1. Relevant energy levels of $^{88}Sr^+$. The ion is Doppler-cooled on the $5s\ ^2S_{1/2}$–$5p\ ^2P_{1/2}$ transition at 422 nm. Repumper light at 1092 nm prevents optical pumping into the metastable $4d\ ^2D_{3/2}$ state. Ramsey's method of separated oscillatory fields is used to interrogate the narrow $5s\ ^2S_{1/2}$–$4d\ ^2D_{5/2}$ quadrupole transition at 674 nm. If excited to the $4d\ ^2D_{5/2}$ state, the ion is returned quickly to the electronic ground state by driving the $4d\ ^2D_{5/2}$–$5p\ ^2P_{3/2}$ transition at 1033 nm.

pulses [30], which shifts the fringes of the Ramsey spectrum in a predictable fashion. A substantial benefit is gained simply by measuring the difference in transition probability when $\delta\Phi$ is switched from +90° to −90° (hereafter referred to as phase-modulation technique no. 1). The advantage of this measurement is that it results in an antisymmetric resonance signal, containing a zero crossing at line-center. A further advantage is that the observed line center frequency is insensitive to drifts in signal amplitude. A second phase-modulation technique (no. 2) records the change in transition probability when $\delta\Phi$ is switched from 0° to 180°, resulting in a symmetric line shape with a maximum signal at line center. Both Ramsey phase-modulation techniques are relatively insensitive to drifts in background signal. These are well-known techniques for rf and microwave experiments with beams, however, to the best of our knowledge, there have been no reported demonstrations in the optical domain.

This paper reports a series of coherent optical spectroscopy experiments on the 674 nm $5s\ ^2S_{1/2}$–$4d\ ^2D_{5/2}$ quadrupole transition (see Fig. 1) in a single $^{88}Sr^+$ ion, which is trapped and cooled in the Lamb-Dicke regime. This transition is being developed as a future optical frequency standard [18,19], and is a recommended radiation for realization of the meter [31]. The same transition is also of use for experimental investigations in quantum information processing, since the analogous transition in $^{40}Ca^+$ has been used in this respect with outstanding success [32,33]. In the work reported here, single-pulse excitation of one $5s\ ^2S_{1/2}$–$4d\ ^2D_{5/2}$ Zeeman component has demonstrated the coherent nature of the excitation with the observation of Rabi flopping in a single $^{88}Sr^+$ ion. Conventional Ramsey spectroscopy (with no relative phase shift $\delta\Phi$ between the two $\pi/2$ pulses) has been demonstrated and used to investigate the coherence of the experimental system. A controlled variation of the relative phase shift $\delta\Phi$ between the two $\pi/2$ pulses was observed to vary the Ramsey excitation probability in the expected oscillatory manner. By measuring the difference in Ramsey transition probability when $\delta\Phi$ is switched from +90° to −90°, an optical Ramsey line shape, with the significant advantage of a zero-crossing signal at line center, has been demonstrated. Ramsey's second phase-modulation technique has also been demonstrated. All optical Ramsey line shapes are well described by a numerical integration of the optical Bloch equations.

This paper is organized as follows. First, Sec. II presents a theoretical description of Ramsey's method of separated oscillatory fields, applied to an optical transition in a single Doppler-cooled, trapped ion. The main features of the experimental apparatus are outlined in Sec. III. Results on single-pulse excitation of the $^{88}Sr^+$ quadrupole transition are presented in Sec. IV. Optical Ramsey spectra, including demonstrations of Ramsey's phase-modulation techniques, are described in Sec. V. In conclusion, Sec. VI discusses the timely relevance of controlled variation of the relative phase shift $\delta\Phi$, and Ramsey's phase-modulation technique no. 1, with regard to quantum information studies and optical clocks, respectively.

II. THEORY OF A SINGLE-ION RAMSEY EXPERIMENT

This section presents a realistic model to describe the coherent optical spectroscopy experiments reported in this paper. A two-level atomic system (ground state $|S\rangle$, excited state $|D\rangle$, resonant frequency ω_{atom} and excited-state spontaneous decay rate Γ_{DS}) interacts with a laser of frequency ω_{laser}, phase Φ, and bandwidth Γ_{laser}. The Rabi frequency Ω_{R} characterizes the atom-laser coupling strength. Finite laser bandwidth (i.e., limited laser coherence) and spontaneous decay are accounted for in a manner similar to that of Boyd [34]. The system is complicated further by the fact that the motion of a Doppler-cooled ion in a trap affects the interaction of the laser with the atomic system, and so this also needs to be accounted for. It can be shown that the optical Bloch equations describing the interaction of a single laser pulse with the two-level system are

$$\dot{\tilde{\rho}}_{DS} = \left[i(\omega_{\text{laser}} - \omega_{\text{atom}}) - \left(\Gamma_{\text{laser}} + \frac{1}{2}\Gamma_{DS} \right) \right] \tilde{\rho}_{DS} - i\frac{\Omega_R}{2} e^{-i\Phi}(\rho_{DD} - \rho_{SS}), \tag{1a}$$

$$(\dot{\rho}_{DD} - \dot{\rho}_{SS}) = -\Gamma_{DS}[1 + (\rho_{DD} - \rho_{SS})] + i\Omega_R(e^{-i\Phi}\tilde{\rho}_{DS}^* - e^{i\Phi}\tilde{\rho}_{DS}), \tag{1b}$$

where ρ_{SS} and ρ_{DD} are the ground– and excited-state populations. In the case of a single interaction we can, without loss of generality, assume $\Phi = 0$. Numerical integration of Eqs. (1), over the period of the pulse duration τ (subject to the boundary conditions $\rho_{SS} = 1$, $\rho_{DD} = 0$, and $\tilde{\rho}_{DS} = 0$ at $t = 0$), describes Rabi flopping of an atomic system under single-pulse excitation. In the case of a Ramsey experiment, $\Phi = 0$ for the first pulse and $\Phi = \delta\Phi$ for the second pulse, where $\delta\Phi$ represents the relative phase shift between the two interaction zones.

In a Ramsey experiment, an atomic transition is interrogated using two coherent fields of equal duration τ and Rabi frequency Ω_R, separated by a period T of free precession. The first $\pi/2$ pulse prepares the ion in a coherent superposition $(1/\sqrt{2})[|S\rangle+|D\rangle]$. If ω_{atom} differs from ω_{laser}, a relative phase shift between the atom and the laser will be accumulated during the free precession period, and fringes, approximately separated by a frequency $1/(\tau+T)$, are observed in the spectral line shape. A relative phase shift $\delta\Phi$ of the second $\pi/2$ pulse with respect to the first [30] leads to a shift in the fringe pattern. Measuring the difference in transition probability when $\delta\Phi$ is reversed from +90° to −90° yields an antisymmetric Ramsey signal. For the Ramsey case, the Bloch equations (1) are integrated numerically for a system initially prepared in the ground state (i.e., $\rho_{SS}=1$, $\rho_{DD}=0$, and $\tilde{\rho}_{\text{DS}}=0$ at $t=0$). The system experiences a first $\pi/2$ pulse with $\Phi=0$, the free precession period with $\Omega_R=0$, and a second $\pi/2$ pulse with $\Phi=\delta\Phi$, resulting in the final excitation probability ρ_{DD}.

Conventional Ramsey experiments are performed with zero relative phase shift between the two $\pi/2$ pulses, i.e., $\delta\Phi=0$. Introducing relative phase shifts of $\delta\Phi=+90°$, 180°, and −90° between the two Ramsey pulses modifies the spectrum. Phase-modulated Ramsey line shapes are recorded by measuring a difference in excitation probability on switching $\delta\Phi$ from +90° to −90° (technique no. 1), and from 0° to 180° (technique no. 2). Use of the optical Bloch equations can be extended to simulate all of these line shapes.

A laser driving a narrow quadrupole transition of a single Doppler-cooled trapped ion is a close experimental approximation to the above model. However, consideration must also be given to the fact that the cold ion is confined to a harmonic potential, and can therefore occupy any one of the quantized vibrational energy levels $|n\rangle$ in the trap. In practice, over many measurements, the Doppler-cooled ion would display a distribution over $|n\rangle$ with a probability governed by its mean vibrational quantum number and the resulting thermal distribution. It is known that the laser driving the optical carrier transition between the two electronic states $|S\rangle$ and $|D\rangle$ couples to each of the vibrational states $|n\rangle$ with a Rabi frequency Ω_n that is dependent on $|n\rangle$ [35,36]. In one dimension,

$$\Omega_n = \Omega e^{-\eta^2/2} L_n(\eta^2), \tag{2}$$

where η is the Lamb-Dicke parameter of the trapped ion, Ω is the total coupling strength, and $L_n(\eta^2)$ is the nth-order Laguerre polynomial. The Rabi oscillations and Ramsey line shapes for a Doppler-cooled ion can be calculated by summing the numerically integrated solutions for each vibrational state $|n\rangle$, in proportion to the thermal occupation probability of that state. This weighted summation over all vibrational states for a single Doppler-cooled ion is a similar requirement to integrating over the atomic/molecular velocity distribution in a beam experiment [1]. Undoubtedly, the cleanest method of circumventing this complication is to prepare the ion in the motional ground state of the trap $(n=0)$ prior to making the Ramsey excitation [16].

III. EXPERIMENTAL SETUP

The basic apparatus used in this series of experiments has been described in extensive detail by an earlier publication [37]. A short description is presented here, highlighting recent improvements to the apparatus. A single $^{88}Sr^+$ ion, confined in a rf trap, is laser-cooled on the $5s\ ^2S_{1/2}-5p\ ^2P_{1/2}$ transition at 422 nm (see Fig. 1). Since the $5p\ ^2P_{1/2}$ state also decays to the metastable $4d\ ^2D_{3/2}$ state, a repumper laser at 1092 nm is required to maintain efficient Doppler cooling. A probe laser at 674 nm is used to conduct spectroscopic investigations of the narrow $5s\ ^2S_{1/2}-4d\ ^2D_{5/2}$ electric quadrupole transition. An ion excited to the $4d\ ^2D_{5/2}$ state is returned to the $5s\ ^2S_{1/2}$ state by driving the $4d\ ^2D_{5/2}-5p\ ^2P_{3/2}$ transition with a clear-out laser at 1033 nm.

The ion trap is of the "endcap" design [37,38] and held at a pressure of 10^{-11} mbar. A 15.9 MHz trap drive frequency results in motional frequencies of $(\omega_r,\omega_z)/2\pi = (2.15, 3.80)$ MHz. The ion may be laser-cooled along any of three noncoplanar axes, a necessity for reducing the ion's micromotion in three dimensions [39] and realizing confinement in the Lamb-Dicke regime. Fluorescence at 422 nm is imaged onto a photomultiplier tube, and around 4×10^4 counts per second are detected when the cooling laser is tuned to line center. The count rate due to background scatter was less than $1\times10^{-3}\ \text{s}^{-1}$.

The 422 nm cooling laser is a frequency-doubled diode laser system. The fundamental laser is stabilized to a low-finesse, tunable reference cavity to reduce the short-term linewidth at 422 nm to $\leqslant 1$ MHz. To ensure long-term frequency stability, the cooling laser is offset-locked to the $5s\ ^2S_{1/2}(F''=2)-6p\ ^2P_{1/2}(F'=3)$ transition in ^{85}Rb (at 422 nm) using a saturated absorption spectrometer. A neodymium-doped fiber laser provides the repumper radiation at 1092 nm, and a 1033 nm extended-cavity diode laser is used to return the ion from the $^2D_{5/2}$ back to the ground state.

The 674 nm probe laser system for spectroscopy of the quadrupole transition is a master and slave arrangement based on extended-cavity diode lasers [37]. The master laser is stabilized to an ultralow-drift reference cavity via a Pound-Drever-Hall lock [40], and the slave is offset-locked to the master via a tunable rf offset. Previously, the drive current of the master laser was modulated to produce the sidebands for the Pound-Drever-Hall lock. The drawback of this approach is that current modulation results in a significant residual amplitude modulation of the laser light, and is undesirable for minimizing the frequency drift of the laser [41]. Drifts in residual amplitude modulation cause drifts in the lock point of the error signal, and consequently drifts in the laser frequency. To reduce this limitation, a 36 MHz resonant electro-optic modulator is now used to generate the sidebands on the master for the stabilization scheme. There are two independent slave lasers, both locked to the master. The first provides a weak probe beam, whereas the second provides an intense probe beam with up to 1.5 mW into a $2w_0=30\ \mu$m spot at the ion. The probe laser linewidth is $\leqslant 2.4$ kHz.

Absorption of the 674 nm probe laser by the $5s\ ^2S_{1/2}-4d\ ^2D_{5/2}$ electric quadrupole transition in the single

$^{88}Sr^{+}$ ion is measured using pulsed-probe spectroscopy [42]. A magnetic field of 360 μT is applied to split the quadrupole transition into its various Zeeman components. The k vectors of both probe beams are at an angle 30° to the applied magnetic-field vector, and to the radial plane. In all the investigations reported here, the carrier transition of the $5s\ ^2S_{1/2}\ (m_j=-\frac{1}{2})-4d\ ^2D_{5/2}\ (m_j=-\frac{1}{2})$ Zeeman component was used. In a single interrogation cycle the ion is Doppler-cooled, optically pumped into the $m_j=-\frac{1}{2}$ ground-state sublevel, and interrogated by the probe laser with a specific set of parameters (i.e., detuning, intensity and pulse duration). The cooling laser is switched on to determine the ion's state via Dehmelt's electron shelving method [43]. If a transition to the $^2D_{5/2}\ (m_j=-\frac{1}{2})$ state is detected, the 1033 nm laser drives the ion back to the electronic ground state and cooling cycle via the $^2P_{3/2}$ state. The interrogation cycle is repeated a set number of times at each value of the parameter being varied to measure the excitation probability. Varying the probe laser detuning results in an absorption spectrum of the transition. Varying the pulse duration records the temporal evolution of the excitation. An acousto-optic modulator (AOM) is used to vary the phase of the optical beam probing the ion. It has been shown [44] that a phase shift $\delta\Phi_A$ of the AOM rf drive signal causes a change in the optical phase $\delta\Phi o$ of the diffracted beam, where $\delta\Phi_O=\delta\Phi_A$. An arbitrary relative phase shift in the range $-180° \leqslant \delta\Phi \leqslant +180°$ can be realized easily by means of a direct digital synthesis frequency source.

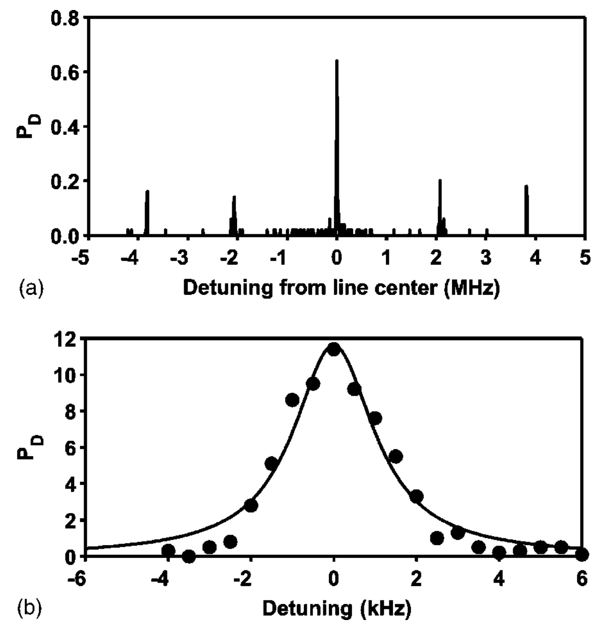

FIG. 2. Excitation spectra of the quadrupole transition with a weak probe laser beam. (a) Quadrupole transition in zero magnetic field, with carrier and radial and axial sidebands at ±2.15 MHz and ±3.80 MHz (50 interrogations per point). (b) $5s\ ^2S_{1/2}\ (m_j=-\frac{1}{2})-4d\ ^2D_{5/2}\ (m_j=-\frac{1}{2})$ transition resolved by a 360 μT magnetic field (1000 interrogations per point, 1 ms pulse duration). The 2.4 kHz observed linewidth is attributed to frequency fluctuations of the interrogation laser and any ac magnetic fields.

IV. SINGLE-PULSE EXCITATION OF THE $5s\ ^2S_{1/2}-4d\ ^2D_{5/2}$ TRANSITION

Prior to experiments in the magnetic bias field, Fig. 2(a) shows the spectrum of the quadrupole transition in zero magnetic field recorded using a weak probe laser beam. The k vector of the probe laser beam is oblique to the radial plane, and therefore is sensitive to the radial and axial modes of the ion's motion. The spectrum in Fig. 2(a) exhibits motional sidebands at 2.15 MHz and 3.80 MHz, due to the ion's radial and axial motions, respectively. The Lamb-Dicke parameter, given by $\eta=k\cos\theta\sqrt{\hbar/2m\omega}$, is therefore $\eta_r=0.042$ for the radial motion and $\eta_z=0.018$ for the axial motion (where θ is the angle between the k vector and the motional directions). The data of Fig. 2(a) indicate that the ion is confined in the Lamb-Dicke regime.

A 360 μT bias field is applied to resolve individual Zeeman components of the quadrupole transition. Figure 2(b) shows the excitation spectrum of the $5s\ ^2S_{1/2}\ (m_j=-\frac{1}{2})-4d\ ^2D_{5/2}\ (m_j=-\frac{1}{2})$ carrier transition with the probe laser beam well below saturation to avoid power broadening. Each point in Fig. 2(b) is the result of 1000 interrogations and such a spectrum takes several minutes to acquire. The 2.4 kHz observed linewidth is attributed to ac-magnetic fields and the probe laser linewidth over this time scale.

The ion was excited coherently on the $5s\ ^2S_{1/2}\ (m_j=-\frac{1}{2})-4d\ ^2D_{5/2}\ (m_j=-\frac{1}{2})$ carrier transition using the intense probe laser beam (1.5 mW in $2w_0=30\ \mu$m). With the laser detuning fixed at line center, the duration of the interrogation pulse was varied. High-contrast Rabi oscillations in the excited-state population were observed as shown in Fig. 3. The apparent damping observed is due to the differing Rabi frequencies Ω_{n_r,n_z} of the laser for each motional state $|n_r,n_z\rangle$ of the ion. The Rabi frequency for a specific motional state $|n_r,n_z\rangle$ is given by [36]

$$\Omega_{n_r,n_z}=\Omega e^{-\eta_r^2/2}L_{n_r}(\eta_r^2)e^{-\eta_z^2/2}L_{n_z}(\eta_z^2), \tag{3}$$

where η_r and η_z are the radial and axial Lamb-Dicke parameters of the trapped ion, and Ω is the total coupling strength.

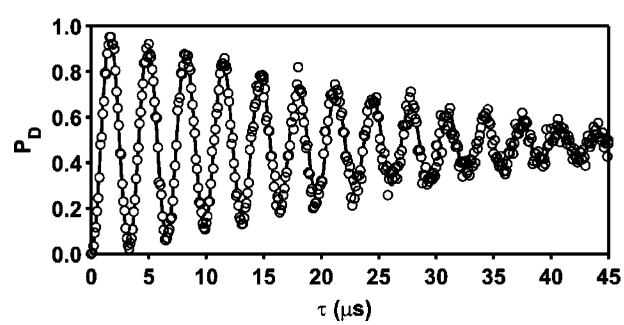

FIG. 3. Single-pulse coherent excitation on the $5s\ ^2S_{1/2}\ (m_j=-\frac{1}{2})-4d\ ^2D_{5/2}\ (m_j=-\frac{1}{2})$ transition. The apparent damping of the Rabi oscillations, due to the ion's thermal distribution over vibrational energy levels of the trap, suggests mean vibrational quantum numbers $\overline{n_z}=8$ and $\overline{n_r}=14$. The solid curve is a fit to the experimental data points and is described in the text.

As discussed in Sec. II, the observed Rabi flopping can be simulated by numerical integration of the optical Bloch equations, summed over many vibrational levels with the appropriate probabilistic weight. Alternatively, the excitation probability as a function of pulse duration is well approximated by superposing the Rabi oscillations for each state as follows:

$$P_D(t) = \frac{A}{2}\left(1 - e^{-\Gamma t}\sum_{n_r,n_z} P_{n_r}P_{n_z}\cos(\Omega_{n_r,n_z}\tau)\right), \qquad (4)$$

where the state $|n_r,n_z\rangle$ has occupation probability $P_{n_r}P_{n_z}$ and A is the occupation probability of the $m_j=-\frac{1}{2}$ Zeeman sublevel of the ground state. The term Γ accounts for decoherence mechanisms such as the finite coherence time of the driving field. Equation (4) was fitted to the Rabi oscillation data of Fig. 3 using a maximum likelihood method. To simplify the calculations, it was assumed that the ion's radial and axial motions are equally well cooled. Consequently, the relationship between the mean radial and axial vibrational quantum numbers is constrained. The fit yields mean vibrational quantum numbers of $\overline{n_r}=14$ and $\overline{n_z}=8$, and decoherence rate $\Gamma/2\pi=0.46$ kHz. The decoherence term Γ is attributed to the linewidth of the interrogation laser over the short time scale of the measurements in Fig. 3. This time scale is significantly shorter than that of Fig. 2(b). The vibrational quantum numbers should be compared with the Doppler limit ($E_{\min}=\hbar\Gamma/2$) of $\overline{n_r}=4.3$ and $\overline{n_z}=2.1$. As an alternative to the method described here, $\overline{n_r}$ and $\overline{n_z}$ can be inferred independently by recording Rabi oscillations on the upper motional sidebands, as has been reported for $^{40}Ca^{+}$ [45].

Figure 3, therefore, shows that the ion is well cooled and that high-contrast coherent oscillations are possible. Recall also that Fig. 2(a) demonstrates confinement of the ion in the Lamb-Dicke regime. Both of these features are important prerequisites for realizing high-contrast optical Ramsey fringes using a single trapped ion.

V. RAMSEY SPECTROSCOPY OF THE 5s $^2S_{1/2}$–4d $^2D_{5/2}$ TRANSITION

Optical Ramsey spectroscopy of the $5s\ ^2S_{1/2}\ (m_j=-\frac{1}{2})$–$4d\ ^2D_{5/2}\ (m_j=-\frac{1}{2})$ transition was performed in a manner almost identical to that of the single-pulse spectroscopy detailed in Sec. IV. In the Ramsey case, however, the single probe pulse was replaced with two distinct pulses of duration τ separated by a period T of free precession. For all Ramsey experiments presented in this paper, the probe beam power was reduced to $\sim 75\ \mu$W, in a spot size of $2w_0=30\ \mu$m centered on the ion. The Rabi frequency and the duration of $\pi/2$ pulse were determined from the observation of the first complete Rabi oscillation with the probe laser beam at this reduced intensity.

To calculate the line shapes, the optical Bloch equations (1) were integrated numerically (as described in Sec. II) for each harmonic-oscillator level, and summed with a statistical weight determined by the thermal occupation probability of each level. In all instances, the best fit to the data was determined using a maximum likelihood estimator, with $\tau_c=1/\Gamma_{\text{laser}}$ as the only free parameter.

The noise of a single ion's Ramsey line shape displays binomial statistics. For a specific set of experimental parameters, P_S and $P_D=(1-P_S)$ are the probabilities of the atom being in the ground and excited states, respectively. N independent measurements result in a signal of NP_D observed excitations, with standard deviation $\sqrt{NP_SP_D}$. So for a single-ion experiment, the statistical noise of the central Ramsey fringe is largest halfway up the fringe and approaches zero at the maximum and minima. Itano *et al.* [46] have analyzed these Ramsey signal fluctuations in detail using a quantum treatment, calling the fluctuations *quantum projection noise*. In contrast to a single ion, Ramsey experiments in atomic beams and fountains are subject to fluctuations in the atom number. By measuring the populations of the two states of the clock transition, the quantum projection noise limit can be reached [47].

A. Conventional Ramsey spectroscopy

Using the probe laser beam of reduced intensity, a single Rabi oscillation period was recorded, and the Rabi frequency $\Omega/2\pi$ was determined to be 15.7 kHz. Conventional Ramsey experiments (i.e., $\delta\Phi=0°$) were performed with $\pi/2$-pulse duration $\tau=15.5\ \mu$s, chosen to maximize the observed Ramsey fringe contrast. The experimentally determined parameters (Ω,τ) were a good approximation to a $\pi/2$-pulse, since $\Omega\tau=0.49\pi$. Conventional Ramsey experiments were conducted using periods of free precession in the range $T=20\ \mu$s through $T=150\ \mu$s. In all instances, the ion was interrogated 100 times at each detuning. As T increased, the resolution of the scans was increased accordingly. The experimental results are presented in Fig. 4, together with fitted theoretical predictions of the Ramsey line shapes (solid curves) for the experimental parameters Ω,τ, and T. The predicted signals were determined using a maximum likelihood estimator, with the laser coherence decay rate Γ_{laser} being the sole free parameter. Note that the model assumes only random laser phase fluctuations and that Γ_{laser} is the resulting laser linewidth.

The contrast of the Ramsey fringes falls as T increases, however the apparent value of Γ_{laser} (as determined from the fitted line shapes) increases throughout the data. Moreover, the data are at first sight inconsistent with the 2.4 kHz linewidth of Fig. 2(b) (i.e., $1/\Gamma_{\text{laser}}=55\ \mu$s), which suggests that the contrast would decay faster than is observed in Fig. 4. However, at slower scan rates the experiment becomes more sensitive to probe laser frequency drifts and low Fourier frequency noise components. Scan rates are slower for the higher-resolution Ramsey scans, and slower still for the single-pulse experiment of Fig. 2(b). Hence the short-term linewidth of the probe laser is considerably less than that suggested by Fig. 2(b).

Any significant heating of the ion during the Ramsey sequence would also reduce the fringe contrast. However, this is unlikely to be the case since the time scale of the experiments performed here is less than 1 ms, whereas heating rates as low as one phonon in 190 ms have been reported for a trap of similar dimensions [48].

B. The effect of oscillator phase

The effect of varying the relative phase difference $\delta\Phi$ between the Ramsey pulses is illustrated as follows. The

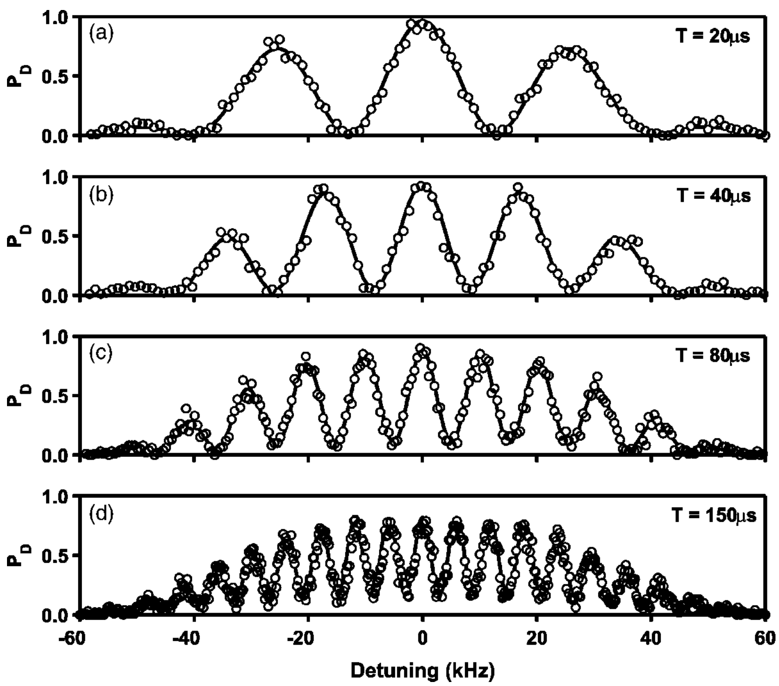

FIG. 4. Conventional Ramsey spectroscopy of the $5s\ ^2S_{1/2}\ (m_j=-\frac{1}{2})-4d\ ^2D_{1/2}\ (m_j=-\frac{1}{2})$ transition. The data shown used consecutive pulses of 15.5 μs duration, separated by periods T of (a) 20 μs, (b) 40 μs, (c) 80 μs, and (d) 150 μs. Each data point is the result of 100 interrogations. The solid lines are fitted simulations with $\Omega/2\pi$ $=15.7$ kHz, and laser coherence decay rate $\Gamma_{\text{laser}}/2\pi$ of (a) 0.19 kHz (b) 0.29 kHz, (c) 0.40 kHz, and (d) 0.53 kHz.

probe laser was tuned to line center and the ion was interrogated with successive $\pi/2$ pulses using the Ramsey method. The pulse separation T was fixed and the excitation probability was measured as a function of the relative phase shift $\delta\Phi$ between the two pulses. The results, presented in Fig. 5, exhibit a maximum at line center when $\delta\Phi=0°$ and 360°, and a minimum when $\delta\Phi=180°$. The solid curve shows the expected form of the variation, and the data demonstrate precise control of the optical phase of the interrogation field relative to the ion. Since any phase shift (between the atomic system and the laser field) may be introduced in this manner, the technique is therefore ideal for canceling any unwanted, but measurable, accumulated phase difference.

C. Ramsey's phase-modulation techniques

These techniques measure the change in transition probability on switching the relative phase shift $\delta\Phi$ of the second Ramsey pulse between two values $\delta\Phi_x$ and $\delta\Phi_y$ [30]. The parameters for phase-modulation technique no. 1 are

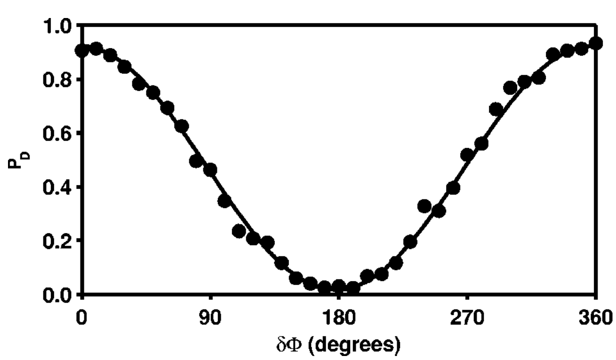

FIG. 5. Excitation probability at line center on the $5s\ ^2S_{1/2}\ (m_j=-\frac{1}{2})-4d\ ^2D_{5/2}\ (m_j=-\frac{1}{2})$ transition as the relative phase shift $\delta\Phi$ between the Ramsey pulses is varied.

$\delta\Phi_x=+90°$ and $\delta\Phi_y=-90°$, and for technique no. 2 are $\delta\Phi_x=0°$ and $\delta\Phi_y=180°$. At each laser detuning, the ion was interrogated by $\pi/2$ pulses (duration $\tau=15$ μs, separation $T=40$ μs , and relative phase shift $\delta\Phi_i$), with 25 sets of the sequence $i=\{x,y,y,x,y,x,x,y\}$. This interrogation sequence was chosen to reject systematic errors in the spectra due to time-dependent linear and quadratic drifts of any background signal that may exist [49]. The change in transition probability is simply the mean difference between the probability for each relative phase shift. The line shape for phase-modulation technique no. 1 is shown in Fig. 6(a). The individual Ramsey line shapes for $\delta\Phi_x=+90°$ and $\delta\Phi_y=-90°$ are shown in Figs. 6(b) and 6(c), respectively, and are constituent subsets of the data of Fig. 6(a). The solid curves in Fig. 6 are numerical integrations of the optical Bloch equations (1), fitted to the data with the laser coherence decay rate as the sole free parameter. Recording the spectrum using phase-modulation technique no. 2 results in the data appearing in Fig. 7(a). Again, the line shapes for the individual phases are constituent subsets of the data of Fig. 7(a), and are shown in Fig. 7(b) ($\delta\Phi_x=0°$) and Fig. 7(c) ($\delta\Phi_y=180°$).

VI. DISCUSSION

The results presented in this paper are of direct and timely relevance to quantum information studies with trapped ions and also to optical atomic frequency standards. The importance and relevance of (i) controlled variation of the relative phase shift $\delta\Phi$ between the two Ramsey pulses, and (ii) measuring the difference in transition probability on switching $\delta\Phi$ from +90° to −90°, is now discussed.

In optical Ramsey experiments with a single $^{40}Ca^+$ ion, Häffner *et al.* [17] studied phase shifts induced by an intense off-resonant laser beam incident on the ion during the period

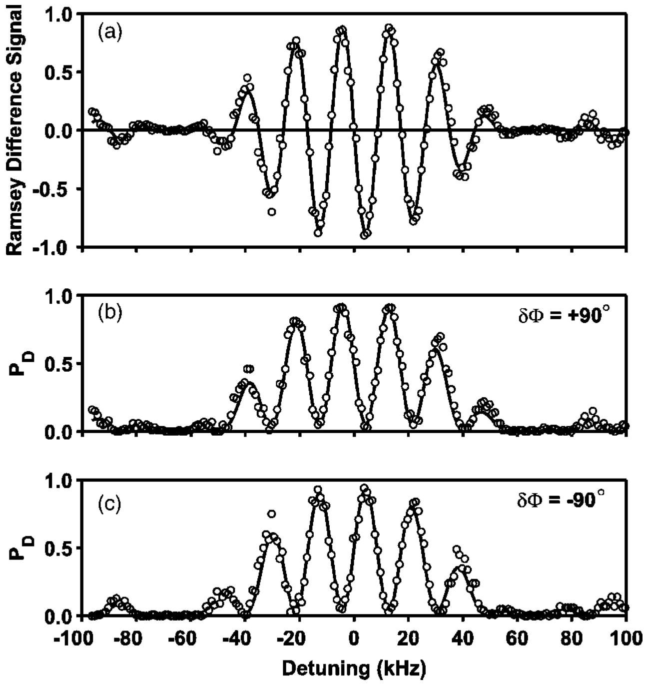

FIG. 6. (a) Ramsey fringe pattern observed when measuring the change in excitation probability on switching the Ramsey pulse relative phase shift $\delta\Phi$ between +90° and −90° (phase-modulation technique no. 1). Each point results from 100 interrogations of each phase. (b) Data for Ramsey measurement with $\delta\Phi=+90°$ phase only. (c) Data for Ramsey measurement with $\delta\Phi=-90°$ phase only. The data in (b) and (c) are subsets of the data shown in (a). The solid curves are calculated using the optical Bloch equations and fitted to the data. The antisymmetric line shape in (a) crosses zero at line center.

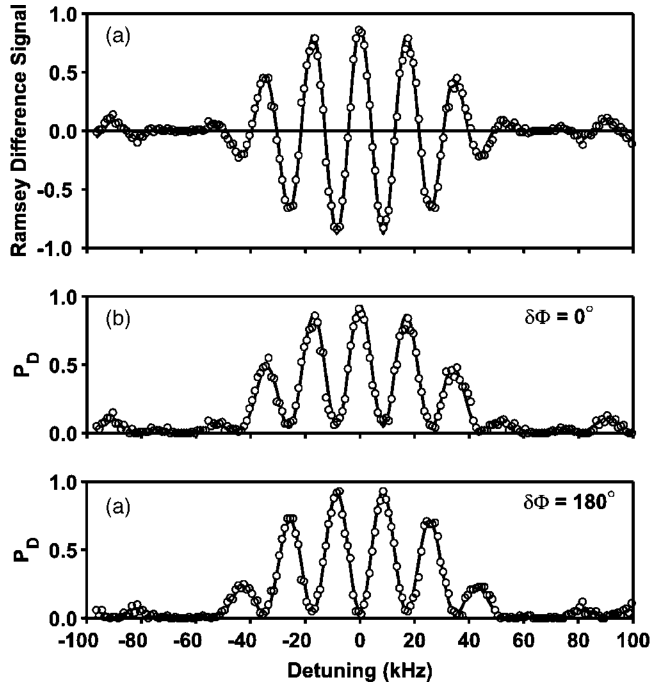

FIG. 7. (a) Ramsey fringe pattern observed when measuring the change in excitation probability on switching the Ramsey pulse relative phase shift $\delta\Phi$ between 0° and 180° (phase-modulation technique no. 2). Each point results from a total of 200 interrogations, 100 of each phase. (b) Data for Ramsey measurement with $\delta\Phi=0°$ phase only. (c) Data for Ramsey measurement with $\delta\Phi=180°$ phase only. The data in (b) and (c) are subsets of the data shown in (a). The solid curves are calculated using the optical Bloch equations and fitted to the data.

of free precession. This study was motivated by the occurrence of such phase shifts when manipulating quantum states in trapped ion quantum information experiments. These phase shifts, induced by the intense off-resonant laser, were simply due to ac-Stark shifts of the transition's energy levels. In order to compensate the ac-Stark shift, Häffner *et al.* used a second off-resonant laser beam to induce an equal but opposite ac-Stark shift to that caused by the first laser beam. The present work suggests that the same effect could be achieved by an appropriate adjustment of the phase of the second Ramsey interaction. The results of Fig. 5, where the laser phase has been varied, demonstrate a relatively simple means of achieving this via the rf signal to an acousto-optic modulator.

Optical frequency standards require a local oscillator to be stabilized to a high-Q atomic resonance transition. With a trapped ion as the atomic reference, local oscillator (i.e., laser) stabilization has traditionally used a frequency-modulation technique [11,18,19,21]; a weak pulse probes the high- and low-frequency sides of the Lorentzian profile, and any imbalance in these absorption signals provides a discriminant. As demonstrated in Fig. 6, Ramsey's phase-modulation technique no. 1 yields an antisymmetric discriminant with a zero crossing at line center. This technique is therefore highly relevant for laser stabilization to a narrow optical transition. A further desirable feature of this Ramsey signal is that the frequency of the line center is independent of drifts in signal amplitude. Such drifts, due to deviations from the $\pi/2$-pulse condition, will only affect the gradient of the signal at line center. It is useful to note that as long as the relative phase shifts $\delta\Phi$ remain identical in magnitude, but of opposite sign, the zero crossing of Fig. 6 will remain at line center. Therefore, small offsets θ in the phase, such that $\delta\Phi=\pm(90°+\theta)$, result only in a small change of the fringe pattern's gradient at line center.

This laser stabilization technique, together with a detailed analysis of associated systematic shifts for optical frequency standards, will be presented in a forthcoming publication [50]. This same phase-modulated Ramsey technique will also be of benefit to neutral atom optical frequency standards when the atomic sample is appropriately confined, allowing a two-pulse Ramsey experiment [29].

VII. SUMMARY

This paper has presented results on coherent optical excitation of the narrow $5s\ ^2S_{1/2}$–$4d\ ^2D_{5/2}$ quadrupole transition in a solitary $^{88}Sr^+$ ion, which is Doppler-cooled and trapped in the Lamb-Dicke regime. High-contrast Rabi flopping has been observed under single-pulse excitation of one Zeeman component carrier transition. The apparent damping of the Rabi oscillation is due to the thermal probability distribution of the ion over the vibrational states of the trap, and the variation of laser Rabi frequency with the ion's vibrational state. Fitting the predicted function to the Rabi flopping signal yields mean radial and axial vibrational quantum numbers of $\overline{n_r}=14$ and $\overline{n_z}=8$.

Optical Ramsey spectroscopy has also been demonstrated on one Zeeman component carrier transition. High-contrast Ramsey fringes have been observed with visibilities up to ~90%. A controlled variation of the phase of the second Ramsey pulse, relative to the first, has been observed to alter the Ramsey excitation probability in a predictable manner. Varying the optical phase by an arbitrary amount is of use in compensating unwanted atomic phase shifts during a sequence of coherent pulses to manipulate an atomic state, as in quantum information processing studies.

In addition to conventional Ramsey spectroscopy, Ramsey's two phase-modulation techniques have been demonstrated. On measuring the change in excitation probability on introducing a 180° relative phase shift, a Ramsey line shape symmetric about line center is observed as predicted. In contrast, by measuring the change in excitation probability on reversing a 90° relative phase shift, the predicted antisymmetric optical Ramsey line shape was observed. The significance of the antisymmetric optical Ramsey signal is that it contains a zero crossing at line center, and is an ideal discriminant signal for stabilizing a local oscillator laser to a narrow atomic reference transition in an optical frequency standard. All observed Ramsey signals are well described by a model based on a numerical integration of the optical Bloch equations.

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