High-resolution laser microwave double-resonance spectroscopy of hyperfine splitting of trapped $^{113}Cd^+$ and $^{111}Cd^+$ ions

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The ground-state hyperfine splittings of laser-cooled $^{113}Cd^+$ and $^{111}Cd^+$ ions confined in a linear Paul trap have been measured to be 15 199 862 854.96(0.12) Hz and 14 530 507 349.9(1.1) Hz, respectively, using a microwave-optical double-resonance technique. The results for $^{113}Cd^+$ agrees with previous measurements very well, and the precision of the $^{111}Cd^+$ is improved by 7 orders of magnitude compared to the best results reported in the literature. The possibility of a microwave frequency standard based on laser-cooled cadmium ions is also discussed.

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I. INTRODUCTION

Trapped ions are suitable for precision frequency metrology due to the long interaction time between the ions and the applied radiation field. Recently, a statistical measurement uncertainty of 7.0×10^{-18} between two optical clocks based on Al^+ ions has been achieved by Chou *et al.* [1]. In the microwave spectral region, frequency standards based on ions are also of great interest. For example, $^{199}Hg^+$, $^{137}Ba^+$, $^{9}Be^+$, $^{171}Yb^+$, and $^{113}Cd^+$ have been investigated by different groups in the past decades [2–8]. Because of its simple energy levels, $^{113}Cd^+$ ion is a good candidate for a microwave frequency standard with laser cooling and detection using only a single laser. Besides that, trapped ions are also widely applied in quantum information processing [9,10]. Based on a trapped $^{111}Cd^+$ ion, direct entanglement between an ion and a photon has been demonstrated [11]. However, to the best of our knowledge, there are very few precisely measured results for the ground-state hyperfine splitting of cadmium ions. For $^{113}Cd^+$, Hamel and Vienne obtained 15 240(200) MHz by polarization transfer from metastable He atoms to Cd^+ ions [12,13], and Tanaka *et al.* obtained 15 199 862 858(2) Hz using the laser excitation, microwave double-resonance method [7]. Recently, Jelenkovic *et al.* obtained 15 199 862 855(0.2) Hz by the microwave-optical double-resonance technique [8]. For $^{111}Cd^+$, Brimicombe *et al.* obtained 14.89(0.15) GHz using a hollow-cathode discharge tube [14].

In this paper we report the measurements of the ground-state hyperfine splittings of $^{113}Cd^+$ and $^{111}Cd^+$ ions using the microwave-optical double-resonance technique. The ions trapped in a linear Paul trap are cooled by a frequency-quadrupled, 214.5-nm tunable diode laser. Meanwhile, the same laser is used to optically pump and detect the ions, which offers us good signal-to-noise ratio (SNR). With Ramsey's separated fields interrogation technique, we precisely measure the ground-state hyperfine splittings of $^{113}Cd^+$ and $^{111}Cd^+$. Our result for $^{113}Cd^+$ agrees with those of Jelenkovic *et al.* very well [8], and the measurement precision of $^{111}Cd^+$ is improved by 7 orders of magnitude compared to the results of Brimicombe *et al.* [14]. These results are also important steps towards the development of a laser-cooled cadmium ion microwave frequency standard [15].

II. EXPERIMENT

The simple energy-level structure of the odd isotope ions of cadmium, as shown in Fig. 1, is very suitable for frequency standards. First of all, the ground-state hyperfine splitting ν_{00} (about 15.2 GHz for $^{113}Cd^+$ and 14.5 GHz for $^{111}Cd^+$) is larger than that of all the ions mentioned above except mercury. Secondly, the ions can be laser cooled and detected by a laser with high efficiency due to the cycling transition between the states of $S_{1/2}(F = 1)$ and $P_{3/2}(F = 2)$. During the laser-cooling process, ions can be trapped in the dark state of $S_{1/2}(F = 0)$ because of the relatively small hyperfine splitting of the $P_{3/2}(F = 2)$ state. In order to prevent this complication, a cooling laser with circular polarization connecting two Zeeman sublevels of the cycling transition is applied. Meanwhile, microwave radiation is applied to repump the ions from the dark state to $S_{1/2}(F = 1)$ [16]. Finally, the same laser for the cooling process can be used to pump the ions by using an acousto-optic modulator (AOM) to blueshift the laser frequency to reach the $P_{3/2}(F = 1)$ hyperfine level.

The experimental setup is shown in Fig. 2. The 214.5-nm laser system is a frequency-quadrupled, tunable diode laser system (TA-FHG Pro, Toptica, Inc). This laser is compact and easy to use with about 5 mW uv laser output. During the measurement, the frequency of the uv laser is stabilized to the megahertz level. The uv laser beam is split into two by a polarizing beamsplitter (PBS). One beam is circularly polarized via a quarter-wave plate and directed to pass through the ion trap for ion cooling and detection. The other beam is frequency shifted by an AOM and passes through the trap in the reverse direction for optically pumping the ions. Two optical shutters are inserted into the paths of the two beams, respectively, and are controlled by a computer to

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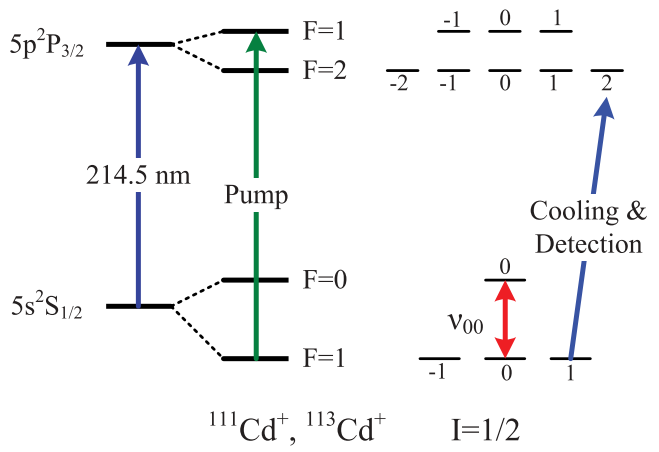

FIG. 1. (Color online) Schematic energy levels of the odd isotopes of cadmium ions. A laser connecting the transition between $S_{1/2}(F=1)$ and $P_{3/2}(F=1)$ is used to pump ions into the state of $S_{1/2}(F=0)$. Another laser is applied to cool the ions and detect the state of ions via the cycling transition between $S_{1/2}(F=1,m_F=1) \leftrightarrow P_{3/2}(F=2,m_F=2)$.

switch the beams on and off during the measurement sequence. The fluorescence signal from the ions is collected by a lens and detected by a photomultiplier tube (PMT) in the photon-counting mode. The microwave is generated by a signal generator referenced to a cesium clock and directed to the ions via a horn antenna.

The ions are trapped in a linear, quadrupole Paul trap consisting of four rods. The radius of the rod electrodes is 3 mm; the minimum distance from the center of the trap to the electrode surface is 8.5 mm; and the separation between the two ring endcaps is 49.5 mm. The trap is driven by a high-voltage radio frequency (rf) driver. The rf field has an amplitude of 300 V at a frequency of 971 kHz. The dc potential applied on the endcaps is approximately 30 V. The cadmium oven points perpendicularly to the center of the trap and is made from a stainless steel tube filled with a small piece of natural cadmium metal. The oven can be heated to the desired temperature by applying a controllable current through it. On the opposite side of the oven, an electron gun is installed to ionize the atomic cadmium by electron bombardment. The trap is enclosed in a stainless-steel vacuum chamber pumped to a background pressure of 2×10^{-10} mbar. A static magnetic field parallel to the electrodes is applied via a Helmholtz coil pair, and the ambient magnetic fields in other directions are compensated by another two pairs of Helmholtz coils.

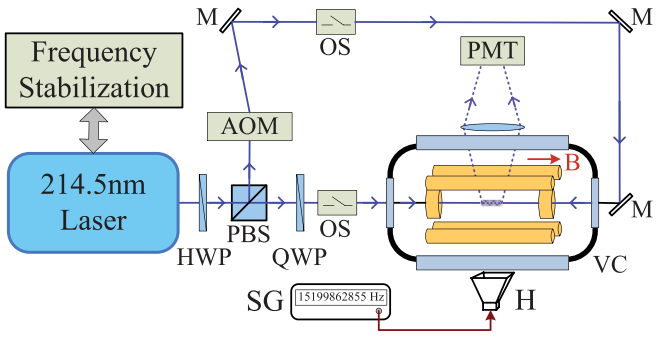

FIG. 2. (Color online) Schematic diagram of experimental setup. AOM: acousto-optic modulator; H: horn antenna; HWP: half-wave plate; M: mirror; OS: optical shutter; PBS: polarizing beamsplitter; PMT: photomultiplier tube; QWP: quarter-wave plate; SG: signal generator; VC: vacuum chamber. A static magnetic field, B, is applied and its direction is parallel to the trap electrodes.

III. RESULTS AND DISCUSSION

The microwave-optical double-resonance spectra of the 0-0 ground-state hyperfine transition of $^{113}Cd^+$ and $^{111}Cd^+$ are measured. Figure 3 shows two examples of the experimental results.

For the Rabi resonance spectra, every data point shown in Fig. 3(a) is obtained after the following sequence of operations: (i) Cooling: a uv laser is tuned to the red side of the $S_{1/2}(F=1,m_F=1) \leftrightarrow P_{3/2}(F=2,m_F=2)$ transition, and a resonant microwave radiation is switched on to repump half of the ions from the dark state back to the $S_{1/2}(F=1,m_F=0)$ state. (ii) Optical pumping: the cooling laser is blocked by

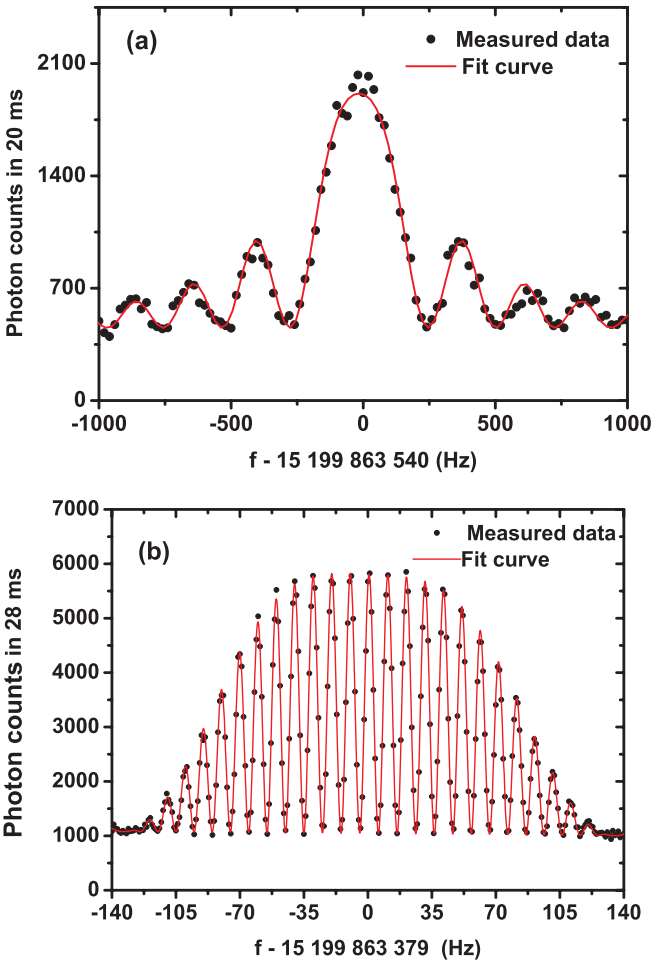

FIG. 3. (Color online) Microwave-optical double-resonance spectra of the 0-0 ground-state hyperfine transition of $^{113}Cd^+$ ions. (a) Rabi resonance fringes for ion interrogation time of 5 ms. (b) Ramsey fringes obtained using 0.1-s separation of free procession between two microwave pulses of 5 ms. The solid lines in (a) and (b) are fits through the experimental data.

an optical shutter, and the repumping microwave radiation is switched off. Then the frequency-shifted laser beam resonant with the $S_{1/2}(F=1) \leftrightarrow P_{3/2}(F=1)$ transition is applied to pump all the ions into the $S_{1/2}(F=0, m_F=0)$ state. (iii) Microwave interrogation: a microwave pulse of $\tau = 5$ ms at frequency f is applied to the ions while all lasers are blocked. (iv) Detection: a circularly polarized laser resonant with the $S_{1/2}(F=1, m_F=1) \leftrightarrow P_{3/2}(F=2, m_F=2)$ transition is applied, and the fluorescence signal is detected by the PMT.

In the experiment, we scan the microwave frequency f over ± 1 kHz about the center frequency f_0. The solid line in Fig. 3(a) is a nonlinear curve fit of the measured data according to the equation of magnetic dipole transition probability [17]

$$P_{\text{Rabi}}(2\pi f) = \frac{b^2}{\Omega^2} \sin^2 \frac{\Omega}{2}\tau \tag{1}$$

with

$$\Omega = \sqrt{(2\pi f - 2\pi f_0)^2 + b^2}, \tag{2}$$

where b is the Rabi angular frequency acting on the ions, f is the frequency of the microwave, f_0 is the center frequency of the transition spectrum, and τ is the duration of the microwave pulse.

For the Ramsey fringes shown in Fig. 3(b), the data is obtained by the similar measurement sequence to that of Rabi except the third step. In this microwave interrogation step, Ramsey's method of separated oscillating fields is applied: two successive, phase-coherent microwave pulses of duration $\tau = 5$ ms interact with the ions, separated by a free precession period of $T = 0.1$ s. By scanning the microwave frequency over ± 140 Hz about f_0, we obtain the Ramsey fringes shown in Fig. 3(b). Then the experimental results are fitted according to the following equation [18]:

$$P_{\text{Ramsey}}(2\pi f) = \frac{4b^2}{\Omega^2} \sin^2 \frac{\Omega}{2}\tau \left(\cos \frac{\Omega}{2}\tau \cos \frac{\Omega_0}{2} T - \frac{\Omega_0}{\Omega} \sin \frac{\Omega}{2}\tau \sin \frac{\Omega_0}{2} T \right)^2, \tag{3}$$

where $\Omega_0 = 2\pi(f - f_0)$, τ is the duration of each microwave pulse, and T is the separation time between two pulses. With this method, the center frequency of the spectra can be obtained with a statistical error of less than 0.02 Hz for $^{113}Cd^+$ and less than 0.1 Hz for $^{111}Cd^+$.

According to the Breit-Rabi formula

$$\nu_{00}(B) = \nu_{00}(B=0) + K_0 B^2, \tag{4}$$

where B is the static magnetic field, $\nu_{00}(B)$ is the 0-0 ground transition frequency in the static magnetic field of B, and K_0 is 257.5 Hz/G^2 for $^{113}Cd^+$ and 269.3 Hz/G^2 for $^{111}Cd^+$ [19]. $\nu_{00}(B=0)$ can be obtained from measurements of $\nu_{00}(B)$ at different magnetic field intensities. In our experiment, the magnetic field is generated by a Helmholtz coil pair whose magnetic flux density at the midpoint is proportional to the current in the coil; hence $\nu_{00}(B=0)$ can be obtained after measuring different resonance frequencies at different coil currents. First, the Ramsey fringes as shown in Fig. 3(b) are measured for a preset current I through the coil. Then,

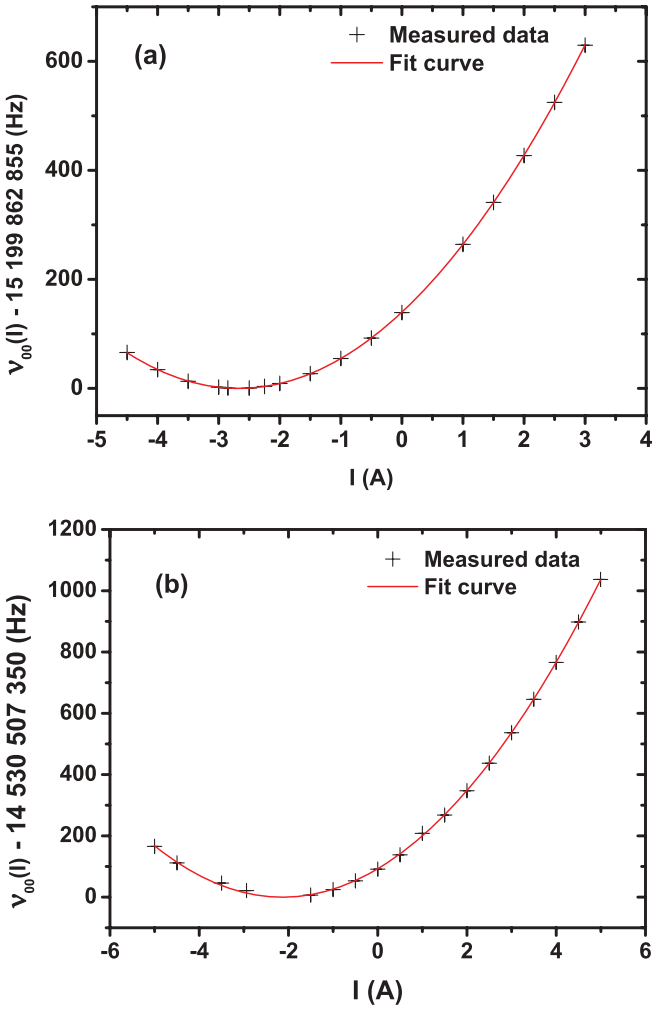

FIG. 4. (Color online) The 0-0 ground transition frequency versus the current in the magnetic coils: (a) results for $^{113}Cd^+$ ions and (b) for $^{111}Cd^+$ ions.

$\nu_{00}(I)$ is obtained by a fit according to Eq. (3), and the above two steps are repeated for other current values. The same measurements are applied for both $^{113}Cd^+$ and $^{111}Cd^+$ ions and the results are shown in Fig. 4. By fitting the experimental data, we obtain the ground-state hyperfine splittings of $\nu_{00,113} = 15\,199\,862\,854.96(0.11)$ Hz and $\nu_{00,111} = 14\,530\,507\,349.9(1.1)$ Hz for $^{113}Cd^+$ and $^{111}Cd^+$, respectively, where the errors are the statistical errors during extrapolating the 0-0 ground transition frequency to zero field.

In estimating the measurement errors, we neglect the light shift because all of the lasers are blocked by optical shutters during microwave interrogations. Our experiment is very similar to the work of a microwave frequency standard based on laser-cooled $^{171}Yb^+$ ions carried out by Warrington *et al.* [20]. We also measured the temperature of the ions to be about 1 K, estimated that the number of ions is about 10^4 and the density is about 2×10^7 cm^{-3}, and the second-order Doppler shift is at the level of 10^{-14} [20,21]. The frequency reference of the measurements is derived from a commercial cesium clock with a frequency accuracy of 5×10^{-13} calibrated by the National Institute of Metrology of China (NIM). The main limit of our measurements is the uncertainty of the magnetic field. In order

to obtain the optimum currents for the two pairs of compensation Helmholtz coils to compensate the magnetic field in the orthogonal directions, we scan the current in the compensation coils to maximize the fluorescence signal while the cooling laser is on. By this method, the currents can be determined with an uncertainty of ±5 mA, and the estimated residual magnetic field in the orthogonal directions is less than 10 mG. According to Eq. (4), the error of the second-order Zeeman shift due to this residual magnetic field is less than 0.03 Hz.

Therefore, the results of our measurements of the ground-state hyperfine splittings of $^{113}Cd^+$ and $^{111}Cd^+$ are

$$\nu_{00,113}(B=0) = 15\,199\,862\,854.96(0.12)\ \text{Hz} \tag{5}$$

and

$$\nu_{00,111}(B=0) = 14\,530\,507\,349.9(1.1)\ \text{Hz}, \tag{6}$$

respectively, where the error estimate is the combination in quadrature of the statistical errors during the spectra fitting and the extrapolation of the transition frequency, and the systematic uncertainty due to the residual magnetic field. The result of $^{113}Cd^+$ agrees with the measurement by Jelenkovic *et al*. [8] very well, and validates the precision of the technique applied in our measurement. The precision of $^{111}Cd^+$ has been improved by 7 orders of magnitude compared to the result of Brimicombe *et al*. [14]. The measurement of $^{111}Cd^+$ is not as precise as that of $^{113}Cd^+$ because of the SNR difference.

The measurement of the 0-0 ground-state hyperfine splittings of both $^{113}Cd^+$ and $^{111}Cd^+$ ions is a key step toward building a laser-cooled, microwave frequency standard based on cadmium ions. Park *et al*. predicted a frequency stability of better than $5 \times 10^{-14}\tau^{-1/2}$ and a frequency uncertainty of 4×10^{-15} for the frequency standard based on laser-cooled $^{171}Yb^+$ ions [6]. Considering the fact that laser-cooled $^{113}Cd^+$ is similar to the laser-cooled $^{171}Yb^+$ in many ways, a similar performance is also expected for cadmium ion clocks.

IV. CONCLUSION

We have reported high-resolution measurements of the ground-state hyperfine splittings of both $^{113}Cd^+$ and $^{111}Cd^+$ ions confined in a linear Paul trap by a laser microwave double-resonance technique, and obtained the values of 15 199 862 854.96(0.12) Hz for $^{113}Cd^+$ and 14 530 507 349.9(1.1) Hz for $^{111}Cd^+$, respectively. The value of $^{113}Cd^+$ agrees with previous results very well, and the value for $^{111}Cd^+$ is improved by 7 orders of magnitude compared to the best results reported in the literature. These measurements are not only significant for the development of microwave frequency standards, but also for experiments on quantum information processing based on cadmium ions.

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