Determination of the ground-state hyperfine splitting of trapped $^{113}Cd^+$ ions

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The ground-state hyperfine splitting of trapped $^{113}Cd^+$ ions has been measured to be 15 199 862 858(2) Hz. The transition was detected by a laser microwave double-resonance method in the presence of He buffer gas. The result was corrected for the second-order magnetic-field dependence. This paper also discusses the possibility of using Cd^+ ions as a microwave-frequency standard. [S1050-2947(96)05906-9]

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I. INTRODUCTION

The ground-state hyperfine splittings of trapped ions such as $^{199}Hg^+$, $^{137}Ba^+$, $^{171}Yb^+$, and $^{43}Ca^+$ have been investigated using the laser microwave double-resonance technique [1–9]. Using the results of these measurements as a possible microwave-frequency standard is a subject of substantial interest. Among the above-mentioned ions, $^{199}Hg^+$ and $^{171}Yb^+$ ions are the preferred candidates because of their simple ground-state hyperfine structure and large hyperfine splitting frequencies. The advantage of $^{199}Hg^+$ ions is that it has the largest hyperfine splitting among those ions studied; moreover, excellent frequency stability has been reported [5]. But it is difficult to generate the optical excitation wavelength of 194 nm because it requires a complicated laser system. Particularly when we consider the reduction of the second-order Doppler shift, a laser with that wavelength is indispensable for laser cooling [6]. As for $^{171}Yb^+$ ions, many investigations [7–9], including even the performance of a prototypical frequency standard [9], have been published recently. However, the loss of population into the $^2F_{1/2}$ level whose lifetime is longer than a week is not a negligible problem. In addition, one has to use two light sources with wavelengths of 369.5 and 935.2 nm.

In this paper we report on the measurement of the ground-state hyperfine splitting of Cd^+ ions using the optical microwave double-resonance method. To the authors' knowledge, Cd^+ ions have not yet been investigated using the ion-trapping technique. However, in a related experiment our group has recently confined Cd^+ ions and employed sympathetic cooling with Be^+ ions in a Penning trap. In that work we determined the isotope shifts precisely [10]. Hamel measured the hyperfine structure in an optical pumping experiment using the spin polarization transfer from metastable He atoms to Cd^+ ions [11,12] and obtained 15 240±200 MHz for $^{113}Cd^+$ ions. Brimicombe *et al.* obtained 14.89(0.15) GHz for $^{111}Cd^+$ using a hollow-cathode discharge tube [13]. Our rf trap experiment on $^{113}Cd^+$ ions improved the precision of this measurement by seven orders of magnitude. We also show that Cd^+ ions can be regarded as a promising candidate for a microwave-frequency standard.

II. CADMIUM ION

There are eight stable isotopes of the Cd atom. Of interest in this paper are the natural abundances of the odd isotopes ^{111}Cd and ^{113}Cd, which are 12.8% and 12.3%, respectively, both of which have a nuclear spin of $I=\frac{1}{2}$. Figure 1 shows the energy-level diagram of the odd isotopes. The ground-state hyperfine splitting of the energy levels is about 15 GHz. The wavelength of the optical transition between the $S_{1/2}$ and $P_{3/2}$ levels is 214.5 nm. The laser microwave double-resonance method is performed with these transitions. The wavelength of 214.5 nm is accessible by several methods. In particular, it can be produced by quadrupling the frequency of the output of an 858-nm semiconductor laser when using two successive frequency-doubling stages [14]. Cd^+ ions have a simple energy-level structure, a large ground-state hyperfine splitting, and need only a relatively simple diode laser based light source. Those are the reasons why we investigated the feasibility of using Cd^+ ions as a frequency standard.

III. EXPERIMENT

Figure 2 shows the experimental setup. The ion trap has hyperboloidic electrodes: the inner ring diameter is $r_0=14$ mm and the end-cap separation is $2z_0=20$ mm. An alternating trap voltage of 240 V amplitude was applied at a frequency of 570 kHz together with a superimposed constant voltage of 5 V. Helium buffer gas of about 5.3×10^{-4} Pa was used to slow down the ions and to damp their oscillation amplitude inside the trap. For the optical excitation of the

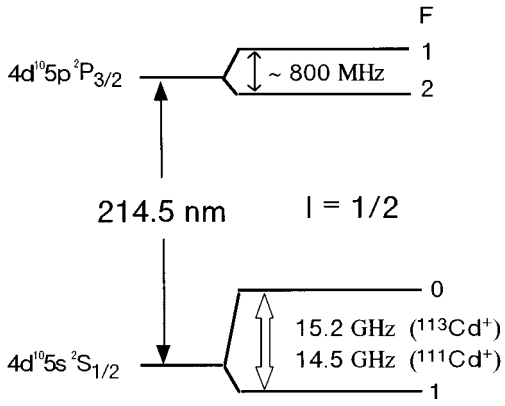

FIG. 1. Part of the energy diagram of the odd isotopes of Cd^+. Both $^{111}Cd^+$ and $^{113}Cd^+$ have a nuclear spin of $I=\frac{1}{2}$. The hyperfine-structure splitting of excited states is unresolved due to a Doppler broadening of 2 GHz in this experiment.

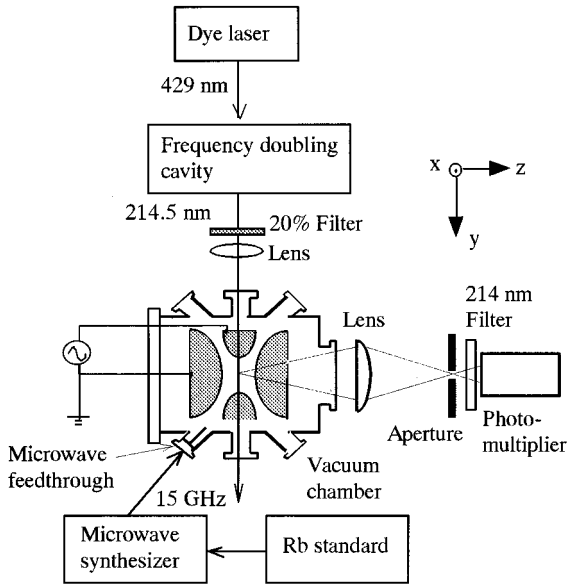

FIG. 2. Experimental setup. Helmholtz coils for compensation of the magnetic field are not shown here for simplicity.

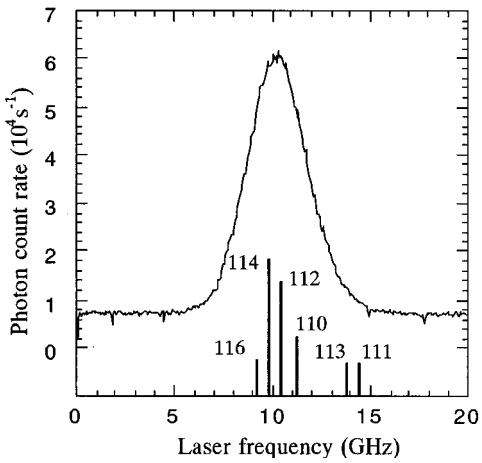

FIG. 3. Observed fluorescence spectrum from the cloud of the natural Cd^+ ions. Vertical lines indicate the resonance lines of the even isotopes and the hyperfine components $[S_{1/2}(F=1)$-$P_{3/2}(F=2)]$ of the two odd isotopes, which are expected from the results of [10]. The numbers beside the lines and the length of the lines show the mass numbers and the natural abundance, respectively.

$S_{1/2}$-$P_{3/2}$ resonance transition at 214.5 nm, we used a frequency-doubled cw dye laser that emits at a wavelength of 429 nm. The output is frequency doubled in a bow-tie ring cavity [15] through a β-barium-borate crystal. The maximum harmonic power was 3 mW. When optimized for the experiment, only 40 μW were needed at the output of the frequency-doubling cavity. This value was further reduced to 7 μW at the location of the ion cloud through a filter and the window of the chamber. The laser beam was focused to a diameter of about 0.7 mm at the ion cloud. The microwaves were created by a synthesizer (Wiltron 68359B) phase locked to a Rb frequency standard (Hewlett-Packard 5065 A) whose accuracy and short-term stability are $\pm 1\times 10^{-11}$ and 5×10^{-12} s^{-1}, respectively. The microwaves are emitted from a typical SMA coaxial feedthrough oriented toward the gap between the ring electrode and the lower end cap. In this arrangement the polarization of the microwave was almost parallel to the x axis, as could be confirmed by observing the double-resonance signals [16]. Three orthogonal pairs of Helmholtz coils compensated stray magnetic fields and also allowed us to generate a magnetic field of up to 900 mG in any direction. Resonance fluorescence from the ion cloud was collected by a lens spatially filtered by an aperture and directed to a photomultiplier connected to a photon counter.

IV. RESULTS AND DISCUSSION

First we observed the fluorescence signal from trapped natural Cd^+ ions by scanning the laser through the resonance with a 214.5-nm optical transition (Fig. 3). In the related experiment of sympathetic cooling of Cd^+ and Be^+ ions using a Penning trap [10], we measured the isotope shifts of the six isotopes. From these results, the overlap of the resonance lines of the four even isotopes, one of the hyperfine components $[S_{1/2}(F=1)-P_{3/2}(F=2)]$ of $^{113}Cd^+$, and that of $^{111}Cd^+$ could be inferred as shown in Fig. 3. Comparing the spectrum obtained with the expected lines, we estimated an optical Doppler width of 2 GHz. Under this condition, the resonance lines of even isotopes overlapped and the fluorescence from the odd isotopes cannot be observed due to optical pumping, which takes place quickly. Because the optical Doppler width is greater than the excited-state hyperfine splitting of about 800 MHz, the excited states are completely unresolved. Easy hyperfine pumping, for example, from the $S_{1/2}(F=1)$ to the $S_{1/2}(F=0)$ state is, in this case, obtained when the laser frequency is tuned to one of the hyperfine components of the resonance line such as $S_{1/2}(F=1)$ to $P_{3/2}$. In our experiment, we tuned the laser frequency by about 4 GHz higher than the peak of the total spectrum in order to dominantly excite the ions in the $S_{1/2}(F=1)$ state.

In addition, in the final laser microwave double-resonance experiment, we used a 95% enriched sample of ^{113}Cd in order to improve the signal-to-noise radio. The corresponding measurement on $^{111}Cd^+$ has not been carried out yet. Using the weak fluorescence signal from the residual even isotopes as an optical frequency marker, we tuned the 214.5-nm radiation at the resonance between the $S_{1/2}(F=1)$ and $P_{3/2}$ states of $^{113}Cd^+$.

An induced microwave transition at 15 GHz connecting the $S_{1/2}(F=0)$ state and the $S_{1/2}(F=1)$ state repopulated the empty level, leading to a corresponding increase in the observed fluorescence. In a weak magnetic field the hyperfine energy level of $F=1$ is split into three Zeeman sublevels. We observed all possible $\Delta F=1$, $\Delta m_F=0,\pm 1$ transitions as a function of magnetic field B and used the $(F=0, m_F=0)$-$(F=1, m_F=0)$ resonance (0-0 transition) for determining the hyperfine splitting, because it is, to first order, independent of magnetic fields and inhomogeneities. The various magnetic-field, values were determined by measuring the splitting of the first-order field-dependent $\Delta m_F=\pm 1$ lines.

Figure 4(a) shows a double-resonance signal of the 0-0 transition. It consists of a narrow central component and sidebands that result from phase modulation by the secular

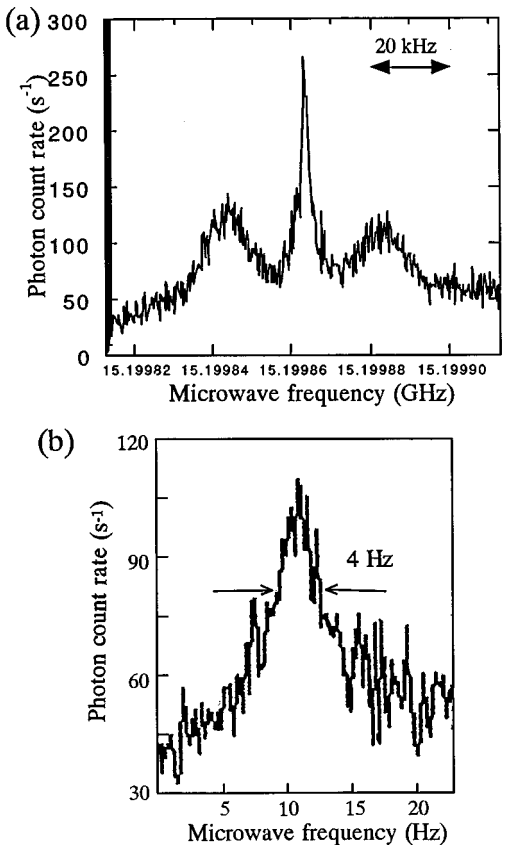

FIG. 4. (a) Laser microwave double-resonance spectrum of the ground-state 0-0 transition of $^{113}Cd^+$. (b) 0-0 transition spectrum with greater resolution at lower microwave power than in (a).

ion motion. The amplitude of these components depends on the orientation of an externally applied magnetic field with respect to the polarization axis of the applied microwave field [16]. We adjusted the magnitude and direction of the applied magnetic field with the Helmholtz coils to make the central component clear. At a microwave power lower than that in Fig. 4(a), we obtained a 0-0 transition signal with a linewidth of 4 Hz in a magnetic field of 260 mG, as shown in Fig. 4(b). The signal was obtained by scanning the microwave frequency in steps of 0.1 Hz for 505 ms, and by recording with a gate time of 800 ms. By repeated scanning around the center of the spectrum the center frequency of the 0-0 transition was obtained with an uncertainty of ± 1 Hz.

We also measured the second-order magnetic-field dependence of the 0-0 transition by varying the field from 140 to 918 mG and used the Breit-Rabi formula, which yielded

$$\nu_{0,0}(B) = \nu_{0,0}(0) + 257.6B^2$$

(B in gauss), for extrapolations to $B=0$ (Fig. 5). The main error in the horizontal axis arises from the uncertainty of the applied magnetic-field value because the linewidth of the microwave resonance signals of $\Delta m_F = \pm 1$ transitions is broadened by the inhomogeneity of the stray field. As shown in [16], the behavior of the 0-0 transition and that of $\Delta m_F = \pm 1$ transitions are quite different. If we adjust the direction of the applied magnetic field to make the resonance of the 0-0 transition clear, the signals of the $\Delta m_F = \pm 1$ transitions become too small to be observed. Therefore, when we

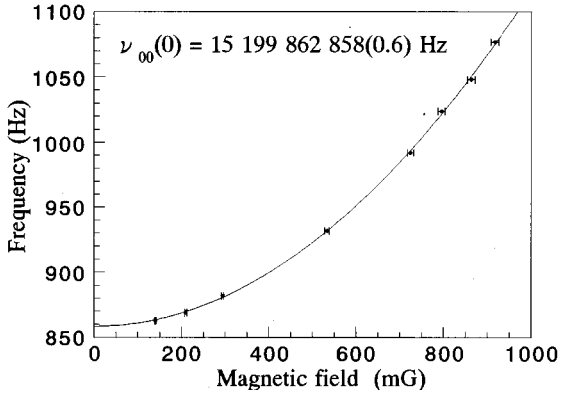

FIG. 5. Magnetic-field dependence of the 0-0 transition. The magnetic field was determined by observing the $\Delta m_F = \pm 1$ transitions. We fitted a parabola through the experimental points corresponding to the Breit-Rabi formula.

observed the $\Delta m_F = \pm 1$ transitions, we applied a higher microwave power than that in Fig. 4(b). It followed that the linewidth of these resonances became broader. In addition, the linewidth of $\Delta m_F = \pm 1$ transitions is generally broader than that of the 0-0 transition. For example, the linewidth of the $\Delta m_F = \pm 1$ transitions was about 15 kHz in the magnetic field of 140 mG. We determined the frequency separation of these resonances with the uncertainty of ± 6 kHz, which corresponded to the uncertainty of the magnetic field about ± 2 mG. In the larger magnetic field, the linewidth of these transitions became broader, leading to larger uncertainty of the applied magnetic field, as shown in Fig. 5. The error in the extrapolation to zero magnetic field is 0.6 Hz.

The light shift must be considered when the double-resonance experiment is carried out with continuous irradiation of both the laser and the microwave. To avoid the effect of the light shift, we reduced the laser intensity to 18 $\mu W/mm^2$ at the ion cloud. Under this condition, the expected frequency shift is negligible and smaller than the current resolution of the double-resonance signals.

The final result of our measurements yields a ground-state hyperfine splitting of $^{113}Cd^+$ of

$$\nu_{0,0}(0) = 15\ 199\ 862\ 858(2)\ \text{Hz}.$$

The uncertainty is limited by the linewidth of the double-resonance signal. It can be further reduced by shielding the magnetic field and by reducing the microwave power while improving the signal-to-noise ratio.

We also observed the double-resonance signal of $^{111}Cd^+$ using the natural isotopes. The obtained value is not was precise as that of $^{113}Cd^+$ because of the difference in the signal-to-noise ratio. This value confirmed, however, that our result is lower than Hamel's result of $^{111}Cd^+$ ions by 40 MHz. This difference is also equal to the difference found for $^{113}Cd^+$. Using an enriched $^{111}Cd^+$ sample will make it possible to determine the hyperfine splitting as precisely as it was determined when using $^{113}Cd^+$.

V. CONCLUSION

We have reported on laser microwave double-resonance spectroscopy investigations of a collision-cooled $^{113}Cd^+$ ion

cloud in a rf trap. The ground-state hyperfine structure splitting has been determined by seven orders of magnitude more precisely than by previous methods. Several characteristics of the Cd^+ ions make them attractive for use as a microwave-frequency standard. Employing an all-solid uv laser whose fundamental oscillator is a semiconductor laser as a light source will enable us to construct a compact frequency standard in the future.

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