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High-resolution ultraviolet spectra of sympathetically-laser-cooled Cd$^+$ ions

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Cadmium ions (Cd$^+$) have been cooled sympathetically below 1 K by means of Coulomb interactions with laser-cooled Be$^+$ ions that are trapped simultaneously in a Penning trap. The ultraviolet resonance lines ($5s\ ^2S_{1/2} \leftrightarrow 5p\ ^2P_{3/2}$, $\lambda = 214.5$ nm) of the cooled Cd$^+$ ions were observed using a frequency-doubled cw dye laser. This radiation is based on efficient frequency doubling in a nonlinear crystal [β-BaB$_2$O$_4$ (BBO)] with an external enhancement cavity. The spectra of the six isotopes ($A = 110$–114, and 116) were obtained using a natural isotope mixture, and the reliability of the measured isotope shifts is discussed.

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I. INTRODUCTION

The stored ion technique using ion traps has led to some unique experiments in the area of precision measurements. Its main advantage is that the ions in them can be stored for long periods of time without large perturbations such as those due to collisions with walls or buffer gases in a gas cell. This provides the basis for high-resolution spectroscopy, that is, narrow linewidths and large line Q's are permitted. In addition, the use of laser cooling on the stored ions suppresses Doppler effects. Because the ions are cooled below 1 K, the accuracy of these measurements is greatly improved. Laser cooling is therefore an important technique in achieving high-resolution spectroscopy. Nevertheless, the ion species that can be laser cooled directly in ion traps are limited and whether a particular species can be directly laser cooled depends on its energy level structure and on the availability of suitable lasers. Thus laser cooling can be used only on a limited number of ion species. In addition, the cooling laser itself causes ac Stark shifts. Sympathetic cooling [1] offers a solution to these problems. When an ion species that cannot be directly laser cooled is stored simultaneously with directly laser-cooled ions, it is sympathetically cooled by their Coulomb interactions. This suggests that in principle sympathetic cooling can cool any ion species regardless of its energy level and there is no need to be concerned about the perturbation of its energy level due to the cooling laser. Consequently, sympathetic cooling creates the possibility of cooling more ion species and this cooling also avoids light shifts. There are several types of ion traps in which laser cooling has been demonstrated, and the Penning trap [2] is one of these which uses static electric and magnetic fields. Its chief advantage is that a large collection of ions can be trapped and cooled without heating, such as rf heating in the Paul trap [2], since they are confined by means of static fields. This suggests that Penning traps are suitable for sympathetic cooling.

We report on sympathetic cooling of Cd$^+$ ions in a Penning trap, carried out by simultaneously trapping laser-cooled Be$^+$ ions. This is the first demonstration of trapping and cooling Cd$^+$ ions in ion traps and few experimental results on sympathetic cooling have so far been reported [1]. Isotope shifts in the ultraviolet resonance lines ($5s\ ^2S_{1/2} \leftrightarrow 5p\ ^2P_{3/2}$ $\lambda = 214.5$ nm) of Cd$^+$ ions have been observed. The spectra of the six isotopes ($A = 110$–114 and 116) which are dominant in a natural isotope mixture are obtained. The absolute isotope shifts are measured and the relative isotope shifts (the ratios of the isotope shifts) are compared with those of other transitions. Cd$^+$ ions are very attractive candidates for frequency standards, since their odd isotopes have large ground-state hyperfine splittings (approximately 15 GHz) [3]. It will be possible to use this ultraviolet transition as a cooling transition of Cd$^+$ ions. So the results described in this article will provide useful information for developing future frequency standards using laser-cooled Cd$^+$ ions. To our knowledge, only a few experimental results on the isotope shift between ^{116}Cd$^+$ and ^{110}Cd$^+$ in this resonance have been published [4,5] and the isotope shifts between the other isotopes have not yet been measured.

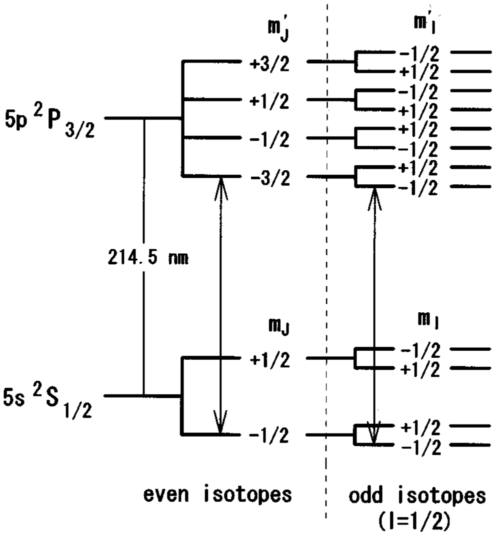

FIG. 1. Energy levels of the $5s\ ^2S_{1/2}$ and $5p\ ^2P_{3/2}$ states of Cd$^+$ for even and odd isotopes in a strong magnetic field. Two stable odd isotopes ^{111}Cd$^+$ and ^{113}Cd$^+$ have nuclear spin $I = \frac{1}{2}$. The figure is not drawn to scale. The observed transitions induced by a 214.5-nm laser are indicated by the arrows.

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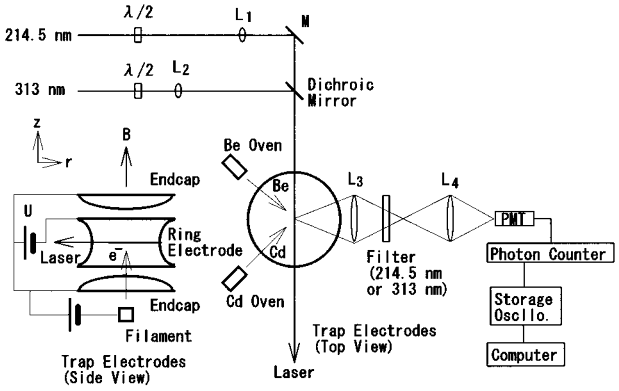

FIG. 2. Block diagram of the experimental setup. The 313-nm laser is obtained by intracavity doubling of a cw dye laser. The 214.5-nm laser is obtained from a frequency-doubled cw dye laser in a BBO crystal with an external enhancement cavity. These are not shown in the figure.

II. CADMIUM ION (CD^+)

The energy levels of the $5s\ ^2S_{1/2}$ and $5p\ ^2P_{3/2}$ states of Cd^+ ions are shown in Fig. 1, both for even and odd isotopes in a strong magnetic field. There are two stable odd isotopes ($A=111$ and 113) and both have nuclear spin $I=\frac{1}{2}$. Because Penning traps use a strong magnetic field (typically $B\approx 1$ T), the magnetic quantum numbers m_I and m_J become the good quantum numbers. The 214.5-nm laser drives the $5s\ ^2S_{1/2}\rightarrow 5p\ ^2P_{3/2}$ transition and according to the selection rules, the electric dipole transitions of $\Delta m_J=\pm 1$ and $\Delta m_I=0$ are driven by the light polarized perpendicular to the magnetic field. The frequency of the laser is scanned around the transitions indicated by the arrows in Fig. 1 to obtain the spectra. These transitions are cyclic. This laser also pumps the ions optically into the $5s\ ^2S_{1/2}(m_J=-\frac{1}{2})$ state for even isotopes and into the $5s\ ^2S_{1/2}(m_I=-\frac{1}{2}, m_J=-\frac{1}{2})$ state for odd isotopes [6]. The cadmium used in these experiments was a natural isotope mixture containing eight stable isotopes (1.25% ^{106}Cd, 0.89% ^{108}Cd, 12.49% ^{110}Cd, 12.89% ^{111}Cd, 24.13% ^{112}Cd, 12.22% ^{113}Cd, 28.73% ^{114}Cd, and 7.49% ^{116}Cd).

III. EXPERIMENTAL SETUP

A schematic diagram of the experimental setup is shown in Fig. 2. Trap electrodes for the Penning trap consist of two endcaps and a ring electrode, which are hyperboloids and are cylindrically symmetric. The static magnetic field B is applied along the symmetry axis (z axis) and the static voltage U is applied between the endcaps and the ring electrode. The motion of a single ion in a trap consists of a harmonic oscillation along the z axis and two circular motions in the x-y plane (cyclotron and magnetron motions) [2]. To create Be^+ and Cd^+ ions in the trap, two ovens are used, one for Be and the other for Cd, and the neutral Be and Cd atoms from the ovens are ionized by collisions with electrons emitted from a filament biased negatively relative to the endcap. This trap apparatus is mounted in a vacuum chamber and the pressure is maintained at about 10^{-8} Pa. The endcap separation is 16 mm and the inner diameter of the ring electrode is 22.6 mm. Typically, the voltage U is 5 V and the magnetic field B is about 1 T for the experiments reported here. The cooling laser for Be^+ ions, which has a wavelength of 313 nm, is obtained by intracavity doubling of a cw dye laser (CR-699-21 with Model No. 7500). The cooling procedure for Be^+ ions is described in detail elsewhere [7]. The 214.5-nm laser radiation for the detection of Cd^+ ions is obtained from a frequency-doubled cw stilbene three-dye laser (CR-699-21) in an angle phase-matched crystal of barium metaborate (BBO) with an external enhancement cavity. A Fabry-Pérot interferometer is used as a frequency marker to monitor the frequency of the fundamental laser ($\lambda=429$ nm). The maximum uv power is about 2 mW. The 313 and 214.5-nm laser beams are overlapped by a dichroic mirror and are directed through the trap. There are holes in the endcaps and ring electrode for introducing atomic, electron, and laser beams. The resonance fluorescence of ions passing through the hole in the ring electrode is collected and focused onto a photomultiplier tube (PMT). Two kinds of interference filters, one for Be^+ and the other for Cd^+ ions, are available and one of these is used as the need arises.

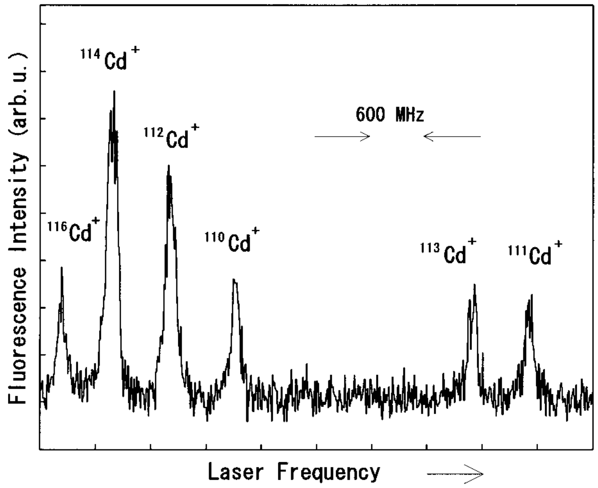

FIG. 3. Observed spectra of sympathetically cooled Cd^+ ions. The data were taken at $U=5.0$ V and $B=0.95$ T. The 214.5-nm laser frequency was scanned to obtain the spectra. The scan width was 7.6 GHz. Spectra are observed for the six isotopes. $m_I=+\frac{1}{2}$ transitions are not seen and the odd isotopes are therefore observed as single lines.

IV. RESULTS AND DISCUSSION

After loading Be^+ and Cd^+ ions into the trap, the Be^+ ions are cooled by the 313-nm laser. The laser frequency is fixed at approximately the maximum intensity point of the Be^+ fluorescence. The spectra of the sympathetically cooled Cd^+ ions, whose transitions are indicated by the arrows in Fig. 1, are obtained as shown in Fig. 3. The 214.5-nm laser frequency is scanned and its full scan width is about 7.6 GHz. To avoid the effects of cooling and heating on the Cd^+ ions as the 214.5-nm laser is scanned, its power is reduced as much as possible ($\approx 100\ \mu$W). Therefore, no asymmetry due to the cooling and heating is observed in the line shapes of the spectra [8]. The minimum Cd^+ ion temperature is found to be approximately 700 mK from the linewidth of the spectra (a Voigt profile is assumed and the value of the $5s\ ^2S_{1/2}$–$5p\ ^2P_{3/2}$ transition probability $A=294\times10^6$ sec^{-1} is used [9]), while the Be$^+$ ion temperature is approximately 400 mK. A similar temperature difference between different ion species has also been observed in other works [1]. The spot size is about 100 μm both for the 214.5 and 313-nm beams. Because the finite width of the laser-beam waist broadens the observed resonance line, it is necessary to determine the laser-beam spot size in order to estimate the error of the measured temperatures. In addition, we are preparing for probe lasers to measure temperatures of trapped ions precisely. We obtained the spectra of six stable Cd$^+$ isotopes ($A=110$–114 and 116). As described above, it has been reported that optical pumping of the nuclear hyperfine components for odd isotopes takes place. In our experiment, the following assumption is made to investigate the optical pumping. Unless the nuclear hyperfine components are optically pumped, half the number of each odd isotope in the ground state fluoresces, since the number of ions in the $m_I=+\frac{1}{2}$ and $-\frac{1}{2}$ ground states is the same and the ions in the $m_I=+\frac{1}{2}$ ground state do not fluoresce in this case. According to this assumption, the intensity of the ^{111}Cd$^+$ and ^{113}Cd$^+$ spectra becomes nearly half in comparison to that of the ^{110}Cd$^+$ spectrum, since these three isotopes have almost the same abundance ($\approx$12–13 %, as described above). Nevertheless, as shown in Fig. 3, the intensity of these three spectra is actually almost the same, showing that the assumption is wrong and the intensity ratio in the spectra roughly corresponds to the natural abundance regardless of even or odd isotopes. In addition, the $5s\ ^2S_{1/2}(m_I=+\frac{1}{2},\ m_J=-\frac{1}{2})\rightarrow 5p\ ^2P_{3/2}(+\frac{1}{2},-\frac{3}{2})$ transition of the odd isotopes is not observed in our experiment (the predicted positions are about 5 GHz lower than their observed transitions when the magnetic field $B=0.95$ T), also indicating that optical pumping into the $m_I=-\frac{1}{2}$ ground state due to the 214.5-nm laser takes place and odd isotopes are therefore observed as single lines which are the same as those in the case of even isotopes [10]. The natural isotope mixture has two other isotopes ($A=106$ and 108), but these spectra have not been obtained in this experiment. Their abundances are too small to detect the fluorescence. The improvement of the signal-to-noise ratio will make it possible to observe a single ion [11] and this will provide higher-resolution measurements of the spectra. The number of stored Cd$^+$ ions has not been determined in this experiment. If the total number of Cd$^+$ ions stored in the trap is less than 100, the ^{106}Cd$^+$ and ^{108}Cd$^+$ ions may not be loaded into the trap because of their small abundances. It is necessary to estimate fluorescence intensity from a single Cd$^+$ ion in this detection system.

To determine whether sympathetic cooling of the Cd$^+$ ions occurs, it is necessary to observe the intensity change in fluorescence from the Cd$^+$ ions when the 313-nm laser which cools the Be$^+$ ions is on and off, as shown in Fig. 4. The 214.5-nm laser frequency is fixed near the resonance of the ^{114}Cd$^+$ ions. When the 313-nm laser is turned off, the Be$^+$ ions begin to heat and spread. Consequently, the Cd$^+$ ions are sympathetically heated and the observed fluorescence decreases. After the 313-nm laser is blocked for 5 sec, it is turned on again. The Be$^+$ ions then begin to be cooled and compressed, causing sympathetic cooling of the Cd$^+$ ions and the return of their fluorescence to the previous value. This result clearly shows that sympathetic cooling takes place. As shown in Fig. 4 stored ions heat gradually when the cooling laser is blocked. The cause of ion heating in a Penning trap is imperfection in the trapping fields and collisions with background neutral molecules [12,13]. The former is caused by the trap asymmetry due to the imperfect hyperboloidal electrodes, contact potential due to deposition of Be and Cd atoms on the surface of the trap electrodes, and the tilted magnetic field with respect to the trap axis. Improvement of the trap axial symmetry and the vacuum will reduce the ion heating. According to the theory of the balance between the electric and magnetic forces on trapped ions [12], the highest static voltage $U_{\max}$ applied between electrodes is determined at a certain magnetic field. For Cd$^+$ ions at $B=0.95$ T, the highest voltage is $U_{\max}\approx25$ V. Nevertheless, in this experiment, fluorescence from Cd$^+$ ions is not observed when the static voltage U exceeds about 10 V. The space charge effect due to Be$^+$ and Cd$^+$ ion clouds probably influences the result.

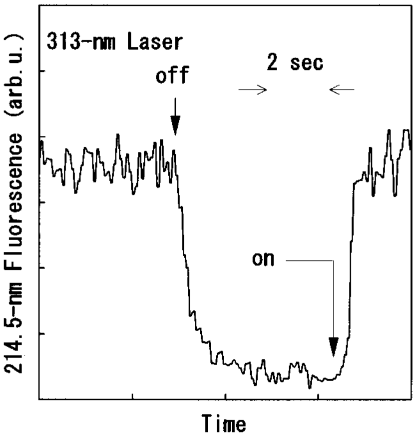

FIG. 4. Dependence of 214.5-nm fluorescence from Cd$^+$ ions on 313-nm laser light. The 214.5-nm laser frequency is tuned near the resonance of the ^{114}Cd$^+$ ions. When the 313-nm laser is turned off, the Be$^+$ ions heat gradually. As a result, the Cd$^+$ ions heat sympathetically and their fluorescence decreases. When the 313-nm laser is turned on, the Be$^+$ ions are cooled and the Cd$^+$ ions are cooled sympathetically. The 214.5-nm fluorescence therefore returns to the previous value.

In these experiments, the absolute isotope shifts of the $5s\ ^2S_{1/2}$–$5p\ ^2P_{3/2}$ line in Cd$^+$ are measured to be $\Delta\nu(110-112)=750\pm20$ MHz, $\Delta\nu(112-114)=680\pm30$ MHz, and $\Delta\nu(114-116)=520\pm20$ MHz. The observed frequency difference between $A=111$ and 113 is 610 ± 20 MHz and the isotope shift $\Delta\nu(111-113)$ is obtained with the correction of the observed value, since the hyperfine splittings in the $5s\ ^2S_{1/2}$ and $5p\ ^2P_{3/2}$ states are different between ^{111}Cd$^+$ and ^{113}Cd$^+$. The theoretical values of their hyperfine splittings are used [4,14], resulting in the correction of adding 159 MHz to the observed value and therefore $\Delta\nu(111-113)=770\pm20$ MHz is obtained. The experimental isotope shift values are the sum of two contributions, the field shift and the mass shift, and the mass shift is the sum of the normal mass shift and the specific mass shift. The normal mass shift is easily calculated by using $\nu m(M'-M)/M'M$, whose ν is the atomic transition frequency,

m is the electron mass, and M' and M are the masses of isotopes, but the specific mass shift is complicated to determine theoretically. The consistency of our experimental data is checked by using comparisons of relative isotope shifts which are ratios of the isotope shifts. The relative isotope shifts, according to Kuhn and Ramsden [14], are calculated using the values $\Delta\nu_R(A'-A)$ which are the corrected isotope shifts obtained by removing the normal mass shifts from the measured shifts. The relative isotope shifts obtained for the $5s\ ^2S_{1/2}$–$5p\ ^2P_{3/2}$ line ($\lambda = 214.5$ nm) in Cd^+ are $\Delta\nu_R(114-116)/\Delta\nu_R(110-112) = 0.73\pm0.04$, $\Delta\nu_R(112-114)/\Delta\nu_R(110-112) = 0.92\pm0.06$, and $\Delta\nu_R(111-113)/\Delta\nu_R(110-112) = 1.02\pm0.05$, and they agree within error margins with the values 0.65, 0.93, and 1.03, which are the relative isotope shifts of the $5p\ ^2P_{3/2}$–$5s^2\ ^2D_{5/2}$ line ($\lambda = 441.6$ nm) in Cd^+ obtained by Kuhn and Ramsden [14], except for the isotope pair 114-116. The cause of the discrepancy has not been fully clarified, but the absolute isotope shift between $A = 110$ and 116 is measured to be $\Delta\nu(110-116) = 1.95\pm0.07$ GHz, which is in agreement with the experimental result 70 ± 10 (10^{-3} cm^{-1}) obtained by Bishop and King [5]. This result supports the reliability of our experimental data. To discuss the isotope shifts in more detail, more precise measurements are required and the use of a King plot is necessary to separate the contributions of the field shift and the specific-mass shift.

V. CONCLUSION

In conclusion, we have performed laser spectroscopic measurements on the $5s\ ^2S_{1/2}$–$5p\ ^2P_{3/2}$ transition (214.5 nm) of Cd^+ ions. The Cd^+ ions were sympathetically cooled with the directly laser-cooled Be^+ ions in a Penning trap and the Cd^+ ion temperature reached approximately 700 mK. The spectra of six isotopes ($A = 110$–114, and 116) were observed, making use of a natural isotope mixture (no enriched isotopes) and the relative isotope shifts were compared with those of other transitions. The absolute isotope shift between $^{116}\mathrm{Cd}^+$ and $^{110}\mathrm{Cd}^+$ was found to be 1.95 ± 0.07 GHz, which was in agreement with other experimental results. These comparisons support the reliability of our experimental data. Using this method, high-resolution spectra can be obtained with a lower-power laser in comparison with other methods such as saturated absorption spectroscopy using a hollow-cathode lamp. The other two isotopes ($A = 106$ and 108) were not observed in this experiment. These results are useful for developing future frequency standards using laser-cooled Cd^+ ions and improvements in the detection system will allow more accurate measurements of the isotope shifts. Spatial separation of trapped ions will be observed to determine the characteristics of sympathetic cooling.

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