# 3-3 Precise Frequency Measurement using Trapped Zinc Ions

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The laser cooling of ions confined in the radio-frequency (RF) ion trap is capable of providing ideal samples for experiment in very high-resolution spectroscopy. We have laser-cooled all even isotopes of Zn<sup>+</sup> ions in a linear RF ion trap, and measured isotope shifts in the 4s  ${}^{2}S_{1/2}$ -4p  ${}^{2}P_{3/2}$  transition. Tunable continuous-wave coherent light near 202 nm was generated for this experiment by frequency conversion of light from solid-state lasers. The mass and the field shifts were estimated using a King plot. The results are useful for a future experiment to measure the frequency of the ground-state hyperfine splitting of Zn<sup>+</sup> ions precisely.

#### Keywords

Zinc ions, Deep ultraviolet, Ion trap, Isotope shift, Hyperfine splitting

### 1 Introduction

#### 1.1 Background and purpose

With the ion trap method, whereby the Coulomb force of an electric field generated between electrodes (i.e., in a microspace within the vacuum chamber) is used to confine ions (charged particles), anywhere from one to thousands of ions can be confined for a long period. In this state, if the ions are lasercooled through irradiation by coherent light of an appropriate wavelength they can be made to be nearly stationary without any perturbation resulting from the outer field[1]. Since the spectrum obtained by observing such ions does not featuring broadening of spectral line width or frequency shift originating from the Doppler effect, and since such observation is not limited in terms of time, the ion trap method has come to be accepted as an effective means of conducting precision frequency measurement. To date, Hg<sup>+</sup>, Yb<sup>+</sup>, Cd<sup>+</sup>, and similar ions have been studied for precision measurement of frequency in the microwave region[2]-[4], and in recent years, research employing Hg<sup>+</sup>, Sr<sup>+</sup>, Yb<sup>+</sup>, and In<sup>+</sup>, (and others) has expanded to include the optical region[5]-[7]. Among these measurements, experiments

that use Hg<sup>+</sup> and Cd<sup>+</sup> employ alkali metal electronic structures with ground states of  $ns^2S_{1/2}$ . With laser cooling, in order to force the ion into the ground state rapidly, it is often necessary to cause multiple light transitions to occur simultaneously. However, in the case of Hg<sup>+</sup> and Cd<sup>+</sup>, all that is required to cause laser cooling is to optically pump the ions from the ground state  $(ns^2S_{1/2})$  to the lowest excited state  $(np^{2}P_{J})$ ; the ions can then be laser-cooled by a comparatively simple light source. Zinc (Zn) is an atom of the same group as Hg and Cd in the periodic table, and Zn<sup>+</sup> has an alkali metal electronic structure. However, the transition suitable for laser cooling of Zn<sup>+</sup> is at a wavelength of 202 nm, deep in the ultraviolet region. Laser cooling of Zn<sup>+</sup> has not been performed to date because it is not easy to generate coherent light of this wavelength; hence Zn<sup>+</sup> has not been employed as an object of precision frequency measurement.

To generate coherent light of a wavelength in the vicinity of 200nm, a conventional largescale, complex light source that employs a single-mode  $Ar^+$  laser or the like has been required. However, in recent years, semiconductor and solid-state lasers have developed rapidly, meeting wide use in high-resolution

spectroscopy requiring frequency resolution of a few MHz or less. Although such lasers are more stable in terms of oscillation wavelength and optical power than gas and liquid lasers and excel in service life and operability, obtainable optical power and wavelength regions are restricted. Currently no laser is capable of directly oscillating light at the wavelength required for the laser cooling of Zn<sup>+</sup>. Therefore the current study was also aimed at the development of a wavelengthtunable coherent light source with a center wavelength of 202nm that uses wavelength conversion, in the hopes that such development would lead to the production of an ideal laser light source [8].

We performed laser cooling of zinc ions confined in an ion trap using the developed 202-nm light source, and observed the isotope shift thereof in a transition from  $3d^{10}4s^2S_{1/2}$  to  $3d^{10}4p^2P_{3/2}$ . The isotope shift is the difference in the transition frequency between two or more isotopes of an element in the same transition, and provides key information as to the atom's internal structure. This shift can be regarded as the sum of a factor based on nuclear mass (mass shift) and a factor based on nuclear volume (field shift)[9]. These factors affecting observed isotope shift were estimated by analysis in this study. Moreover, although odd isotopes of zinc ions could not be observed in this experiment, the isotope shifts of the odd isotopes were estimated from the findings for even isotopes. These estimates will be applied in the future in the course of our planned precision frequency measurement in the microwave region using zinc ions.

#### 1.2 Energy level of zinc ion

To date there have been a number of spectroscopic investigations of Zn<sup>+</sup>, and the Atomic Spectra Database of the NIST (National Institute of Standards and Technology)[10] provides detailed information on the energy levels of Zn<sup>+</sup>. However, no precision frequency measurements have made of the isotope shift corresponding to the optical transition from the ground  $(3d^{10}4s^2S_{1/2})$  state of Zn<sup>+</sup>. Some of the energy levels of the zinc ion are shown in Fig.1. Five isotopes of zinc occur naturally, with mass numbers and abundance ratios of 64 (48.6%), 66 (27.9%), 67 (4.1%), 68 (18.8%), and 70 (0.6%). The even isotopes among these (mass numbers of 64, 66, 68, and 70), when excited to the  $4p^2P_{3/2}$  state, feature only one ground state level to which they can be relaxed by spontaneous emission. They can then be laser-cooled only with an electromagnetic wave at a wavelength of 202.6-nm. On the other hand, the ground state of the odd isotope (mass number of 67) is composed of two hyperfine levels. The relaxation rate between these levels is very small. Therefore, two electromagnetic waves are required to perform laser cooling of the odd isotope. In our present study, the even isotopes of zinc ions were laser-cooled using deep ultraviolet coherent light having only one wavelength. Hyperfine splitting of the ground state of the odd isotope is predicted to be approximately 7.2 GHz[11]; the use of this frequency will enable precision frequency measurement in the microwave region.



# 2 Development of all-solid-state deep ultraviolet coherent light source

# 2.1 Outline of deep ultraviolet light generation

The  $\beta$ -BBO crystal is widely used as a nonlinear crystal to generate light of sufficient optical power up to wavelengths of approximately 200nm. Ultraviolet light up to 204.8 nm may be obtained by second harmonic generation using this crystal, but generation of 202-nm light requires a system of sum-frequency generation. Several combinations of fundamental wavelengths may be used in such sum-frequency generation. We focused on a combination of 266-nm light and 850-nm light. In recent years, the market has seen the introduction of a solid-state light source capable of continuously emitting high-power coherent light at a wavelength of 532 nm. Using this light source, 266-nm light of sufficient power can be obtained through second harmonic generation. At the same time, highpower 850-nm light can be generated using semiconductor lasers. Through sum-frequency generation of these two lights, an all-solidstate light source featuring a 202-nm wavelength is obtained. This sum-frequency generation requires a rather large phase matching angle of 61°. With increasing phase matching angle, the nonlinear effect of a crystal become smaller, but it is nevertheless possible to generate sufficiently large optical power with an angle of 61°. Fig.2 shows a light source developed in this study. The light source is composed of a master oscillator and power amplifier system for generating light at a wavelength of 850 nm, a semiconductor-laser-



pumped Nd:YVO<sub>4</sub> laser for generating 532nm light (Coherent Verdi), an external cavity for second harmonic generation (for 266-nm light), and an external cavity for sum-frequency generation (for 202-nm light)<sub>[8][12]</sub>.

### 2.2 Ultraviolet light (266 nm) generation by second harmonic generation using solid-state laser

The semiconductor-laser-pumped Nd:YVO<sub>4</sub> laser generates and outputs single-mode continuous-wave light at a wavelength of 532 nm by second harmonic generation inside the laser cavity. Since this laser outputs stable single-mode continuous-wave light featuring large optical power without complicated adjustment, it is excellent as a fundamental wave light source for wavelength conversion. The output 532-nm light is led to the external cavity for second harmonic generation. A  $\beta$ -BBO crystal (7-mm long, with both end facets cut at the Brewster angle) was used as a nonlinear crystal for wavelength conversion. As an external cavity for second harmonic generation, a commercial external cavity (Laser Analytical Systems) was employed, following our own modifications designed to enable the cavity to perform the generation of light of large optical power. These modifications included reduction of the temperature increase in the  $\beta$ -BBO crystal by gluing the crystal holder to a Peltier cooler and controlling crystal temperature during the experiments. We presented this improvement to the manufacturer of the external cavity, and the manufacturer provided us, free of charge, an improved new commercial model, which is currently in use. Fig.3 shows the optical power of the 266-nm light obtained as a function of the optical power of the 532-nm light. When 3.5-W 532nm light was made to enter the cavity, a 266nm optical output over 1 W was obtained. Although generally the output optical power increases in proportion to the square of the incident optical power in second harmonic generation, the observed power did not increase any more than this maximum power of about 1.2W. This phenomenon was ascribed

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to the fact that the high optical power of the 266-nm light resulted in non-uniform temperatures inside the crystal, despite the external temperature control, consequently limiting any increase in output.



### 2.3 Generation of deep ultraviolet light (202-nm) by sum-frequency mixing

The 202-nm light is obtained by the sumfrequency generation of light of 266 nm and light of 850 nm. Single-mode continuouswave light at 850 nm was obtained using a master-laser/power-amplifier system constructed based on semiconductor lasers. The light of the extended cavity diode laser (New Focus 6226) serving as a master laser was shaped with prism pairs, and was made to enter a power amplifier after passing through an optical isolator. A commercial high-output semiconductor laser was modified into the power amplifier used in this study. In this experiment, a taper-type semiconductor device with high gain was employed. Master laser light of optical power of 7.5 mW was input to the semiconductor amplifier, and the optical power was amplified to approximately 500 mW.

For efficient generation of light at the target wavelength of 202-nm, the optical power of the 850-nm light was increased within the external cavity using the sum-frequency generation. The external cavity was composed of two concave mirrors, each with a radius of curvature of 150 mm, and two planar mirrors. Spacing of the concave mirrors was set to 160 mm, and the optical path length for a round trip within the cavity measured 776 mm. The incident mirror featured a reflectivity of 97% and the other three mirrors each featured a reflectivity of 99.5% or more for a wavelength of 850 nm. When the light was resonated in the cavity, a somewhat elliptical focal spot with a diameter of approximately 0.1 mm was formed at the midpoint between the two concave mirrors. In this location, a  $\beta$ -BBO crystal (7-mm long, with end facets AR coated for 850-nm and 266-nm wavelengths) was placed to provide the phase matching angle. The round-trip length of the cavity was controlled by the Hansch-Couillaud method. The optical power of the 850-nm light inside the cavity was approximately 30 times as large as the incident power. Thus, when the incident power to the cavity is 420 mW, we can conclude that 850-nm light of approximately 13 W enters the crystal in the cavity.

The sectional intensity distribution of the output light beam from the cavity for second harmonic generation became elliptical due to walk-off in the  $\beta$ -BBO crystal. After correcting this ellipse to a near-circle with cylindrical lenses, the output light was focused into the BBO crystal in the cavity for sum-frequency generation. The focus of the 266-nm light was adjusted so that propagation of the 266-nm light and that of the 850-nm light overlapped sufficiently inside the crystal. The generated 202-nm light was taken out of the cavity using a mirror featuring reflectivity of 95% at a wavelength of 202 nm and transmissivity of 99% or more at a wavelength of 850 nm. Subsequently, the 202-nm light was passed through a prism, and its optical power was measured by a sensitivity-calibrated photodiode (Hamamatsu). Fig.4 shows the power of the 202-nm light as a function of the power of the 266-nm light. The incident power of the 850-nm light to the cavity was 420 mW. When the 0.8-W 266-nm light was made to enter the cavity, a maximum output of 0.32 mW was measured. Considering losses due to the mirrors and the like, this is equivalent to generation of 0.38 mW or more. When 266nm light of greater than 0.8 W was made to enter, the output power increased only minimally, while output power became unstable. At this time, the optical power of the 850-nm light in the cavity was about 13 times greater than the incident power, and the enhancement factor was reduced by half relative to the case in which the incident power of the 266-nm light was 100 mW. It was concluded that the incidence of the high-optical-power 266-nm light resulted in non-uniformity of temperature inside the crystal, which made the optical mode in the cavity unstable. With this light source, sum-frequency light ranging from 201.5 nm to 203.1 nm was successfully generated by changing the wavelength of the master laser. Moreover, by continuously sweeping the frequency of the master laser, a continuous frequency sweep of approximately 7 GHz was successfully observed at a wavelength of 202 nm.



# 3 Precision measurement of zinc ions by ion trap

# 3.1 RF ion trap and measurement equipment

Fig.5 shows the experimental arrangement of the ion trap and the laser cooling system

[13]. The linear-type ion trap used for the experiment can confine ions along the central axis of the trap, and hence was able to perform efficient laser cooling of the ions. The trap was composed of four rod electrodes and two ring electrodes, and was housed in an ultrahigh-vacuum chamber. The inside of the chamber was maintained at a vacuum pressure of  $2 \times 10^{-7}$  Pa or less using an ion pump. The rod electrodes each measure 5.0 mm in diameter and 56 mm in length, and are arranged in parallel to maintain a distance of 5.0 mm from the central axis of the overall trap to the central axes of the electrodes. RF voltage at a frequency of 4.2 MHz with an amplitude of  $\pm 250$  W was applied to the two rod electrodes disposed diagonally; the remaining two rod electrodes were grounded. The two ring electrodes, each having an inner diameter of 18 mm, were placed 16 mm apart from each other. A DC voltage of 100 V was applied on the ring electrodes to confine the ions in the axial direction of the trap.



Metal zinc featuring the natural isotopic abundance ratio was heated in an oven in the chamber to force the zinc atoms to flow into the trap through a nozzle. The electrons emitted from the filament collided with the atoms with an acceleration voltage of approximately 25 V, ionizing the atoms. The 202-nm light obtained by sum-frequency generation was

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made to enter the chamber and to travel along the central axis of the trap, causing the confined zinc ions to undergo photoexcitation. Fluorescence from the ions was collected with a lens and detected by a photomultiplier tube (Hamamatsu, R166P). Using the photon counting method, even weak fluorescence could be observed with excellent sensitivity. For laser cooling, 202-nm light of approximately 0.2 mW was generated, and the optical power reaching as far as the vicinity of the center of the trap was approximately 0.15 mW. Part of the 202-nm light was led to the temperature-stabilized Fabry-Perot etalon, and the power variations of the transmitted light, occurring at intervals of 150 MHz, were used as frequency markers. This etalon used a mirror featuring reflectivity of 98% at a wavelength of 202 nm. In order to verify optical transition of the zinc ion during the experiment, a hollow-cathode lamp was used to observe the opto-galvanic spectrum.

# 3.2 Laser cooling and measurement of isotope shift

Fig.6 shows the laser-induced fluorescence spectrum of laser-cooled zinc ions. The insert above the figure shows the frequency markers obtained using the etalon, and the numerals in the figure indicate the atomic mass numbers. The laser-induced fluorescence spectrum was observed by flowing a current to the oven and the filament for about two minutes to load the ions in the trap and sweeping the frequency of the 202-nm light at a speed of approximately 10 MHz/s. As shown in Fig.6, all of the even isotopes of zinc ions were observed. Since the abundance ratio of the isotope having a mass number of 70 was as small as 0.6%, the observation of this isotope demonstrated the high sensitivity of this measurement. From the photon counting rate that was detected with the photomultiplier tube, it was estimated that several <sup>70</sup>Zn<sup>+</sup> ions were observed, as indicated in the spectrum shown in Fig.6. In the optogalvanic spectrum of zinc ions observed as a reference, the isotopes were not separated because of large Doppler broadening. Nevertheless, using the peaks of the opto-galvanic spectrum and the frequency markers of the Fabry-Perot etalon as references for frequency, the ions were successfully laser cooled repeatedly in a single frequency range.



From the number of detected photons, we estimated that a few hundreds of ions were laser-cooled in the trap. When a large number of ions are laser-cooled, the spectral shape depends upon the efficiency of the laser cooling and absorption of an RF electric field induced by interactions between the ions; this is referred to as the efficiency of RF heating [14]. The sudden drop in fluorescence intensity seen on the high-frequency side of each spectrum peak of Fig.6 indicates that the frequency of the 202-nm light reaches the resonance frequency of transition, at which point the laser cooling changes to laser heating. The heated ions only minimally absorb light near the resonance frequency due to the Doppler effect. If the influence of the RF heating is large, the drop in intensity on the high-frequency side of the peak is slowed and the peak position of the fluorescence spectrum is shifted in the low-frequency direction. The acuteness of the drop in fluorescence intensity seen in Fig.6 indicates that the efficiency of laser cooling is significantly greater than the efficiency of RF heating. The full width at half maximum of the observed spectral line was approximately 50 MHz. We estimated that the zinc ions were laser-cooled to about 0.15 K by comparing the Voigt line shape corresponding to the reported transition intensity[10] with the peak line shape on the low-frequency side. Generally, with increasing amplitude of the RF electric field, the density of trapped ions becomes large, as does the extent of RF heating. On the other hand, with a low ion density, the signal-to-noise ratio of the spectrum decreases. From the results of experimentation at several different amplitudes, it was found that a minimum amplitude of ±250V was required to observe the low-abundance  $^{70}$ Zn<sup>+</sup> ion with sufficient intensity. In four out of ten experiments of laser cooling, the <sup>70</sup>Zn<sup>+</sup> ion was observed with a sufficient signal-tonoise ratio.

In order to examine the frequency shift of the peak due to RF heating, the reduction in fluorescence intensity observed on the highfrequency side of the peak was approximated by a straight line, and the frequency at the intersection of the line and the background level of the spectrum was recorded. In the spectral lines of the zinc ions having mass numbers of 64, 66, and 68, the frequency difference between these intersections and the vertexes of the peaks was approximately 10 MHz. On the other hand, in the zinc ion having a mass number of 70, the difference exceeded 10 MHz, and this difference featured variation. It is considered that <sup>70</sup>Zn<sup>+</sup> ions occurring in a small abundance ratio collided with other isotopes, to be heated even during laser cooling. The difference in the transition frequency between isotopes, i.e., the isotope shift, was determined not by the difference in the frequency between the peak vertexes in the spectrum but by that between the intersections obtained by the above-mentioned linear estimation operation. Although there may be slight differences between these intersections and actual resonance frequencies, we estimated that the difference is almost the same level among the spectral lines of the four isotopes when these isotopes were observed in a single

frequency sweep, and that the effect of this difference on the determination of the isotope shifts would therefore be negligible. The frequency shifts obtained by ten iterations of laser cooling were averaged, and the isotope shifts between the even isotopes were determined. The results are shown in Table 1. For the isotope shift between the isotopes having mass numbers of 68 and 70, there was a rather large standard deviation, as the reduction in fluorescence intensity on the high-frequency side of <sup>70</sup>Zn<sup>+</sup> was moderate. To perform more accurate measurement, two methods are effective: in one, a small number of ions are detected with high sensitivity using a small-sized trap; in another, the abundance ratio of <sup>70</sup>Zn<sup>+</sup> ions in the sample is increased artificially.

For the zinc ion, the isotope shift had been observed in the transition from  $3d^{10}4p^2P_{1/2}$  to  $3d^{9}4s^{22}D_{3/2}$  at a wavelength of 589 nm[15]. In this transition, since the number of s-orbital and *d*-orbital electrons changes simultaneously, a very large shift was observed even in a low-resolution spectroscopic experiment. In order to confirm that the isotope shift at a wavelength of 202 nm observed in the current study is consistent with the reported isotope shift at a wavelength of 589 nm, we performed a King plot as follows[9]. In a transition at a wavelength of X nm, an isotope shift  $IS_{PO}^{X}$ from the isotope <sup>Q</sup>Zn<sup>+</sup> to <sup>P</sup>Zn<sup>+</sup> can be regarded as a sum of the mass effect  $MS_{PQ}^{X}$  and the field effect  $FS_{PO}^{X}$ .

$$IS_{P,Q}{}^{X} = v_{P}{}^{X} - v_{Q}{}^{X} = MS_{P,Q}{}^{X} + FS_{P,Q}{}^{X}$$
(1)

where  $\nu_N^x$  is the transition frequency of the <sup>N</sup>Zn<sup>+</sup> ion. Denoting nuclear mass of the <sup>N</sup>Zn<sup>+</sup> ion by  $M_N$ , and electron mass by *m*, the mass effect  $MS_{RQ}^x$  is expressed by the equation

$$MS_{P,Q}{}^{X} = K^{X}(M_{P} - M_{Q}) / (M_{P} + m)(M_{Q} + m)$$
(2)

where  $K^{x}$  does not depend on  $M_{\text{N}}$  but rather depends on the transition. Using a combination of <sup>66</sup>Zn<sup>+</sup> and <sup>64</sup>Zn<sup>+</sup> for reference, the isotope shift  $IS_{\text{PQ}}^{x}$  is multiplied by the result  $\mu_{(\text{PQ})}$ of the following expression

#### $\mu_{(P,Q)} = (M_{66} - M_{64})(M_P + m)(M_Q + m) / (M_P - M_Q)(M_{66} + m)(M_{64} + m)$ (3)

to yield a modified isotope shift corresponding to the following expression:

$$\mu_{(P,Q)}IS_{P,Q}{}^{X} = MS_{66,64}{}^{X} + \mu_{(P,Q)}FS_{P,Q}{}^{X}$$
(4)

We calculated the modified isotope shift  $\mu_{(\rm RO)}IS_{\rm RO}^{202}$  for the observed isotope shift, and plotted these to  $\mu_{(PQ)}IS_{PQ}^{589}$  to obtain the King plot  $(\mu_{(PQ)}IS_{PQ}^{202}, \mu_{(PQ)}IS_{PQ}^{589})$  of Fig.7. In the figure, solid dots represent the modified isotope shifts obtained in the actual measurement and the solid lines represent standard deviations. Moreover, each pair of numerals indicates the mass numbers of the two isotopes for which the shift was measured. This King plot shows an almost linear relationship between the two sets of modified isotope shifts, verifying consistency of the isotope shifts determined in this experiment with the reported isotope shift at a wavelength of 589 nm. The relationship shown in Fig.7 is expressed as follows using the least square method.

$$\mu_{(P,Q)}IS_{P,Q}^{589} = (-2.07)\mu_{(P,Q)}IS_{P,Q}^{202} - 1.45$$
(5)

where  $IS_{PO}^{589}$  and  $IS_{PO}^{202}$  are given in GHz. Moreover, the slope -2.07 of the expression (5) gives an estimate of the ratio of the field shifts in the two transitions  $(FS_{PQ}^{589}/FS_{PQ}^{202})$ [9]. Following observations by Kloch et al., Foot et al. estimated contributions of the mass effect and the field effect in the isotope shift at a wavelength of 589 nm [16]. Using the  $MS_{66,64}^{589} = -3.58 \,\text{GHz}$  value reported there, expressions (4) and (5) can be used to calculate a value of  $MS_{66,64}^{202} = 1.03 \text{ GHz}$ . Furthermore, using this value of 1.03 GHz, the isotope shifts in Table 1, and the expression (4), we estimated the contributions of the mass effect and the field effect in the measured isotope shifts. The results are shown in Table 1.



Table 1	lsotope shift shifts	and mass a	nd volume
Isotope pair	Measured shift (GHz)	Mass shift (GHz)	Field shift (GHz)
66,64	0.676(6)	1.03	-0.35
68,66	0.670(4)	0.97	-0.30
70,68	0.568(10)	0.92	-0.35

#### 3.3 Measurement of hyperfine splitting

Since only one electromagnetic wave was used in this study, the <sup>67</sup>Zn<sup>+</sup> ion whose ground state was subjected to hyperfine splitting could not be laser cooled. Consequently, the transition frequency shift between <sup>64</sup>Zn<sup>+</sup> and  ${}^{67}\text{Zn}^+$  in a transition from  $4s^2\text{S}_{1/2}$  to  $4p^2\text{P}_{3/2}$  was estimated using expression (5). This shift can be regarded as a sum of the shift due to the isotope effect (isotope shift) and the shift due to hyperfine interaction (hyperfine splitting). Koch et al. derived -4.42 GHz for the isotope shift from <sup>64</sup>Zn<sup>+</sup> to <sup>67</sup>Zn<sup>+</sup> in a transition from  $4p^{2}P_{1/2}$  to  $4s^{22}D_{3/2}$ . Inserting this value in expression (5), we estimated the isotope shift from  ${}^{64}Zn^+$  to  ${}^{67}Zn^+$  in the transition from  $4s^2S_{1/2}$ to  $4p^2P_{3/2}$  as 1.14 GHz. Next, in order to estimate hyperfine splitting, it was necessary to consider the splitting of the ground state and of the  $4p^2P_{3/2}$  state. Theoretical predictions are

available for hyperfine splitting of the ground state of Zn<sup>+</sup>, Cd<sup>+</sup>, and Hg<sup>+</sup> (all within the same periodic group), and these predictions agree well with experiments with Cd<sup>+</sup> and Hg<sup>+</sup>[11]. On the other hand, no information is available on hyperfine splitting of the  $4p^2P_{3/2}$  state. However, generally, hyperfine splitting of the  $np^2P_{3/2}$  state is smaller than that of the  $ns^2S_{1/2}$ state, and hence the hyperfine splitting in question can be roughly estimated based on the hyperfine splitting of the  $4s^2S_{1/2}$  state. Assuming that the hyperfine splitting of the  $4p^{2}P_{3/2}$  state is sufficiently small, the spectrum of the transition from  $4s^2S_{1/2}$  to  $4p^2P_{3/2}$  of  ${}^{67}Zn^+$ can be regarded as composed of two transitions: a transition from  $4s^2 S_{1/2}$  (F = 2) to  $4p^2 P_{3/2}$ (F = 1, 2, 3), and a transition from  $4s^2S_{1/2}$  (F =3) to  $4p^2 P_{3/2}$  (*F* = 2, 3, 4). In this case, the frequency shift due to hyperfine splitting can be estimated based on the energy shift of the hyperfine levels of the ground state:

 $v_{\rm hfs} = (A/2)[F(F+1) \cdot J(J+1) \cdot I(F+1)]$ (6)

Here, the hyperfine constant A = 2.4 GHzis obtained from a predicted value of the hyperfine field strength, 451 tesla[11]. By adding  $IS_{67,64}^{202.6} = 1.14$  GHz to the value  $v_{hfs}$  of expression (6), we estimated that the transition from  $4s^2 S_{1/2}(F = 2)$  to  $4p^2 P_{3/2}(F = 1, 2, 3)$  and the transition from  $4s^2 S_{1/2}(F = 3)$  to  $4p^2 P_{3/2}(F = 3)$ 2, 3, 4) are frequency-shifted from the transition of <sup>64</sup>Zn<sup>+</sup> by approximately +4.1GHz and -3.1 GHz, respectively. Presently we are planning to perform precise measurement of hyperfine splitting in the ground state of <sup>67</sup>Zn<sup>+</sup>. In the projected experiment, the frequency of 202-nm light will first be fixed to the optical transition of <sup>67</sup>Zn<sup>+</sup>, and then the microwave will be irradiated to the ion. The microwave frequency will be swept and a frequency that resonates with the transition between the hyperfine levels will be detected as an opticalmicrowave double resonance signal. If collisional cooling<sup>[4]</sup>, whereby the ion is cooled by collision between itself and a He buffer gas, is used, the double resonance signal may be obtained even if the frequency of the 202-nm light is not exactly matched with the optical transition. Moreover, using laser cooling, the frequency of the hyperfine splitting of <sup>67</sup>Zn<sup>+</sup> in the microwave region can be precisely measured.

### 4 Conclusions

Zinc ions confined in the linear type ion trap were laser-cooled, and the isotope shifts in the transition from  $4s^2S_{1/2}$  to  $4p^2P_{3/2}$  at a wavelength of 202.6 nm were precisely measured. For this experiment, we developed a frequency-tunable coherent continuous-wave light source delivering optical power of 0.3 mW or more at a wavelength of 202 nm, as an all-solid-state light source employing a solidstate laser and a semiconductor laser. Using the laser cooling, the isotope shifts of all the even isotopes-including 70Zn+, at an abundance ratio of 0.6%-were determined. A King plot was established for the observed isotope shifts and the reported isotope shifts in the transition from  $4p^2P_{1/2}$  to  $4s^{22}D_{3/2}$ , and the mass shift and the field shift in the observed isotope shifts were estimated by analysis. The result obtained in this study can be used for future research, in which the frequency of the hyperfine splitting in the ground state of the odd isotope of zinc ions (67Zn+) will be precisely measured.

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