

# Observation of sympathetically cooled $^{43}\text{Ca}^+$ ions in a linear ion trap

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**Abstract** We report the observation of sympathetically cooled  $^{43}\text{Ca}^+$  (natural abundance 0.135%) in a linear ion trap utilizing simultaneously trapped isotope ions as coolant. We investigated different possibilities of realizing efficient sympathetic cooling and observed the peaks of the hyperfine transitions of  $^{43}\text{Ca}^+$  under various experimental conditions.

## 1 Introduction

Calcium has six stable isotopes,  $^{40}\text{Ca}$  (abundance 96.9%),  $^{42}\text{Ca}$  (0.647%),  $^{43}\text{Ca}$  (0.135%),  $^{44}\text{Ca}$  (2.09%),  $^{46}\text{Ca}$  (0.004%),  $^{48}\text{Ca}$  (0.187%), and a long-lived ultra trace isotope  $^{41}\text{Ca}$  ( $^{41}\text{Ca}/\text{Ca} = 10^{-15}$ – $10^{-14}$ ,  $t_{1/2} = 1.04 \times 10^5$  yr). The isotope analysis of calcium finds applications in many fields, such as biomedical tracers or dating [1–6]. The measurement of  $^{41}\text{Ca}$  by means of laser based techniques such as Resonance Ionization Mass Spectrometry (RIMS) or Atom Trap Trace Analysis (ATTA) [7] is a more flexible alternative to the current routine analysis involving Accelerator Mass Spectrometry, which requires a large facility.

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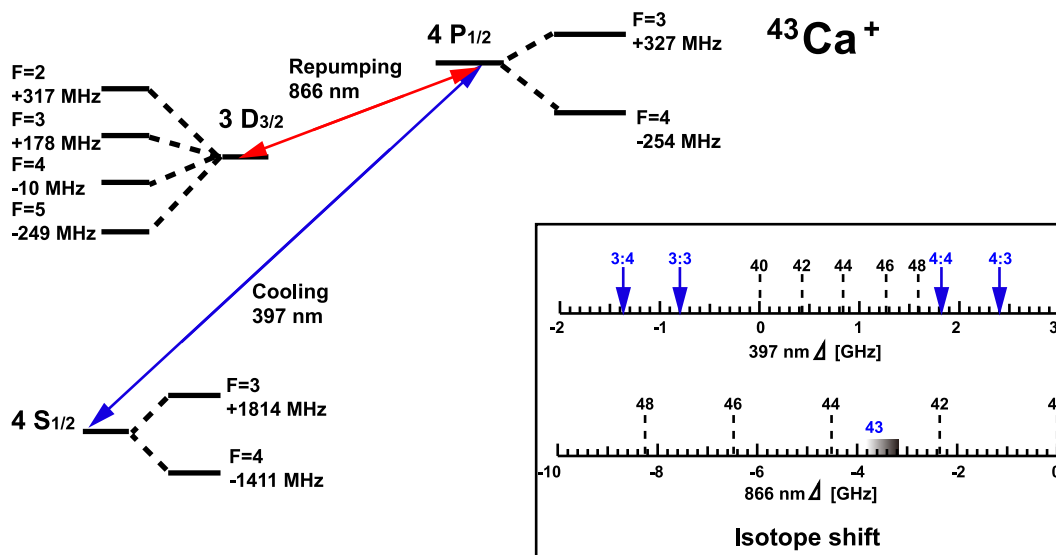
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We developed a novel method of measuring Ca isotopes utilizing ion trapping—laser cooling techniques [8–10]. This method enables us to observe even  $\text{Ca}^+$  isotopes,  $^{40,42,44,48}\text{Ca}^+$ , by means of the isotope shifts of the transition wavelengths and the mass selective function of the linear ion trap [10]. Cooling trapped ions enhances the Laser Induced Fluorescence signal and makes it possible to observe single isotope ions without destructive measurements. This method has two isotope-selective schemes, which are mass selectivity and laser spectroscopy. These features make our scheme flexible and open the possibility of applying it to the measurement of ultra trace isotopes such as  $^{41}\text{Ca}$  [8].

The nuclear spin of  $^{43}\text{Ca}^+$  ( $I = 7/2$ ) is the same as that of  $^{41}\text{Ca}^+$ , and its abundance is 0.135% so we can more easily demonstrate and evaluate our method using  $^{43}\text{Ca}^+$  before treating  $^{41}\text{Ca}^+$ . There are several points that need to be taken into consideration.

The optical transitions of  $^{43}\text{Ca}^+$  are complicated because of the hyperfine structure due to the nuclear spin (Fig. 1). In order to create a closed cycle to directly laser cool  $^{43}\text{Ca}^+$ , hyperfine repumping with multiple lasers or microwave radiation is required. Since the polarization of the lasers may cause population trapping in the degenerate Zeeman sub-levels of the hyperfine states [11] it becomes necessary to apply a magnetic field and carefully consider the polarization effects [12–16].

Experiments with  $^{43}\text{Ca}^+$  utilizing an isotope-selective loading method with enriched samples [13, 17] or resonant photo ionization [16, 18–20] have been reported. We have already observed  $^{48}\text{Ca}^+$ , the abundance of which is comparable with that of  $^{43}\text{Ca}^+$ , so we should be able to directly manipulate  $^{43}\text{Ca}^+$  among other isotopes in our system. However, the neighboring isotopes of  $^{43}\text{Ca}^+$  could affect the cooling of the ions [20]. The effect of the motion of trapped ions needs to be investigated because the inter-



**Fig. 1** Energy levels of  $^{43}\text{Ca}^+$  ion [12, 13, 28]. *Inset:* The isotope shifts of  $\text{Ca}^+$  isotopes [32, 33]

actions of the ions in an ion trap is stronger than that in ATTA. The motion of different isotopes can affect the cooling efficiency of the ions of interest via sympathetic cooling/heating.

To avoid the technical difficulties of directly cooling  $^{43}\text{Ca}^+$  we use sympathetic cooling [21–23] for the pre-cooling of the ions of interest. In this method directly laser cooled ions cool the target ions via Coulomb interaction and thermal exchange. In this way atoms [21] and molecules [22] which have complicated energy levels, or even polymer molecules [23] with mass numbers exceeding 10,000 amu, can be cooled to low temperatures. Sympathetic cooling of odd isotopes has been previously reported, e.g.,  $^{25}\text{Mg}^+$  by  $^{24}\text{Mg}^+$  [24],  $^{135}\text{Ba}^+$  by  $^{138}\text{Ba}^+$  [25],  $^9\text{Be}^+$  by  $^{24}\text{Mg}^+$  [26],  $^{27}\text{Al}^+$  by  $^9\text{Be}^+$  [27] and  $^{43}\text{Ca}^+$  by  $^{40}\text{Ca}^+$  [28]. We simultaneously trap the coolant isotopes and  $^{43}\text{Ca}^+$  using laser ablation [10] and investigate the sympathetic cooling of  $^{43}\text{Ca}^+$ . With the laser ablation method one cannot selectively load an individual isotope of interest so the mass selectivity function of the linear quadrupole trap is required.

In this article, we report the observation of sympathetically cooled  $^{43}\text{Ca}^+$  generated by laser ablation. First, we describe our experimental setup for the  $^{43}\text{Ca}^+$  hyperfine transitions. Then, we investigate the neighboring isotopes as coolants and observe the hyperfine transitions of  $^{43}\text{Ca}^+$  under various experimental conditions.

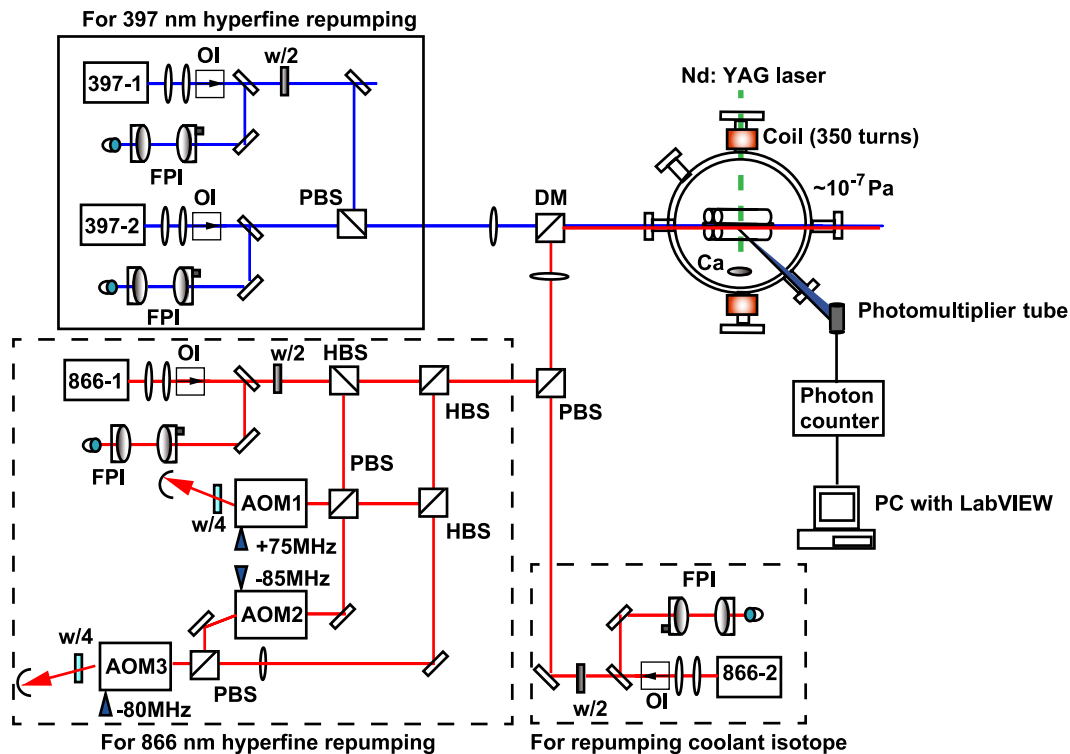
## 2 Experimental system

Figure 2 shows the schematic diagram of our experimental system. Our trap consists of four linear electrodes (10 mm diameter, closest distance from the trap center to the electrodes  $r_0 = 4.3$  mm) and two end cap electrodes (5 mm outer

diameter and 3 mm inner diameter). In order to generate the trapping potential we apply an RF voltage ( $V_{\text{rf}} = 35\text{--}300$  V,  $\Omega/2\pi = 1.5$  MHz) and a DC voltage ( $V_{\text{cap}} = 1\text{--}30$  V) to the linear and end cap electrodes, respectively. These electrodes are placed in a vacuum chamber, whose pressure is maintained at  $1.7 \times 10^{-7}$  Pa by a turbo molecular pump and a rotary pump. A Ca target with a purity of 99.5%, is placed on a pedestal below the trap electrodes. To easily generate calcium ions from the solid Ca target, we utilize laser ablation with a frequency doubled Nd:YAG pulsed laser [29].

In order to laser cool the  $\text{Ca}^+$  ions we utilize external cavity laser diodes (ECLD). To excite  $^{43}\text{Ca}^+$  an additional 397 nm ECLD is required because of the wide splitting of the 397 nm transition (3.2 GHz). We can excite the  $4S_{1/2}$ ,  $F = 3 \rightarrow 4P_{1/2}$ ,  $F' = 4$  ( $F : F' = 3 : 4$ ) for the repumping transition (397-2) and  $4S_{1/2}$ ,  $F = 4 \rightarrow 4P_{1/2}$ ,  $F' = 4$  or  $3$  ( $F : F' = 4 : 4, 4 : 3$ ) for the cooling transition (397-1).

For the 866 nm transition (866-1), we use three acousto optic modulators (ISOMET 1205C-2) as the splitting of the 866 nm transition is 600 MHz. The modulation frequencies are +75 MHz (AOM1),  $-85$  MHz (AOM2), and  $-80$  MHz (AOM3), respectively. We apply the double pass method to the AOM1 and AOM2. Utilizing the first order diffractions of the AOMs, we can excite the four hyperfine transitions,  $4P_{1/2}$ ,  $F = 3 \rightarrow 3D_{3/2}$ ,  $F' = 3$  ( $F : F' = 3 : 3$ ),  $4P_{1/2}$ ,  $F = 3 \rightarrow 3D_{3/2}$ ,  $F' = 2$  ( $F : F' = 3 : 2$ ) and  $4P_{1/2}$ ,  $F = 4 \rightarrow 3D_{3/2}$ ,  $F' = 5$  ( $F : F' = 4 : 5$ ),  $4P_{1/2}$ ,  $F = 4 \rightarrow 3D_{3/2}$ ,  $F' = 4$  ( $F : F' = 4 : 4$ ). Furthermore, we also make use of an additional 866 nm ECLD (866-2) for efficiently repumping the coolant isotopes. The 397 nm laser powers are 2 mW for repumping (397-2) and 1.7 mW for cooling (397-1). The 866 nm laser powers are 0.7 mW



**Fig. 2** Experimental setup with additional 397 and 866 nm lasers for hyperfine repumping and sympathetic cooling. Abbreviations, OI: Optical Isolator, FPI: Fabry–Pérot Interferometer, PBS: Polarizing

Beam Splitter, HBS: Half Beam Splitter, DM: Dichroic Mirror, AOM: Acoustic Optic Modulator, w/2: 1/2 wave plate, w/4: 1/4 wave plate

(0 MHz), 0.8 mW (+150 MHz), 0.4 mW (−245 MHz), 5.3 mW (for the coolant isotope).

To avoid population trapping in the dark states we utilize a pair of coils to generate a magnetic field of 0.5 mT. They are placed on the vertical viewports along the vertical direction.

### 3 Influence of neighboring isotopes

In order to directly cool  $^{43}\text{Ca}^+$ , the frequencies of the two 397 nm and three 866 nm repumping lasers are set to resonance. The frequency of the cooling laser is set to −200 MHz from the resonance of  $F : F' = 4 : 4$ . To suppress effects of the dominant isotope,  $^{40}\text{Ca}^+$ , we load the ions at  $V_{\text{rf}} > 235$  V, where  $^{40}\text{Ca}^+$  cannot be trapped [10]. However, using the technique of the rf field perturbation [30] we find that  $^{42}\text{Ca}^+$ ,  $^{43}\text{Ca}^+$ ,  $^{44}\text{Ca}^+$  still remain in the trap. We found that these other isotopes prevent  $^{43}\text{Ca}^+$  from cooling down to low temperature in our experimental setup. The experiments using combinations of lasers show that the  $^{42}\text{Ca}^+$  ions non-resonantly absorb the laser photons of 397 nm ( $F : F' = 3 : 4$ ) and 866 nm ( $F : F' = 3 : 3$ ),  $^{44}\text{Ca}^+$  ions, 397 nm ( $F : F' = 3 : 4$ ) and 866 nm ( $F : F' = 4 : 4$ ), respectively.

## 4 Observation of sympathetically cooled $^{43}\text{Ca}^+$

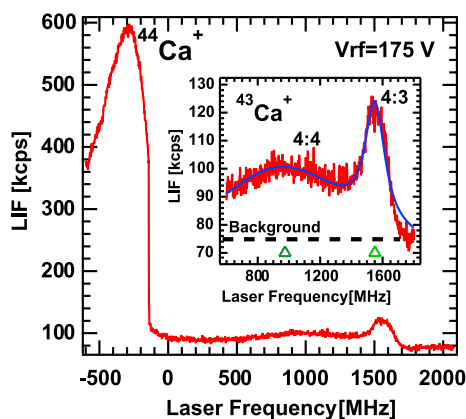
The natural abundance of  $^{43}\text{Ca}^+$  is lower than those of the neighboring isotopes, so eliminating them by exciting the nonlinear resonances of the trapping potential will also affect the behavior of the trapped  $^{43}\text{Ca}^+$ . We discuss two possible scenarios for the sympathetic cooling of  $^{43}\text{Ca}^+$  by neighboring isotopes. (a) Sympathetic cooling by  $^{42}\text{Ca}^+$ : the laser cooling of  $^{42}\text{Ca}^+$  will also cause the laser cooling of  $^{44}\text{Ca}^+$ . Furthermore, the number of trapped coolant ions ( $^{42}\text{Ca}^+$ ) is smaller than that of hot isotope ions ( $^{43}\text{Ca}^+$ ) because  $V_{\text{rf}} > 235$  works as a low mass filter to eliminate lighter ions ( $^{42}\text{Ca}^+$ ). (b) Sympathetic cooling by  $^{44}\text{Ca}^+$ : in the cooling process we can eliminate  $^{42}\text{Ca}^+$  due to laser heating, so we can sympathetically cool  $^{43}\text{Ca}^+$  by utilizing the laser cooled  $^{44}\text{Ca}^+$  as a coolant. Furthermore, the number of the coolant ions is by one order of magnitude larger than that of  $^{43}\text{Ca}^+$ , so the cooling efficiency would be higher than that for  $^{42}\text{Ca}^+$ . Therefore we employ the laser cooled  $^{44}\text{Ca}^+$  as a coolant for  $^{43}\text{Ca}^+$ .

### 4.1 Effect of coolant $^{44}\text{Ca}^+$ ions

We directly laser cool  $^{44}\text{Ca}^+$  first and observe the spectra of  $^{44}\text{Ca}^+$  and  $^{43}\text{Ca}^+$  as a function of the cooling laser frequency (397-1). The 397 nm for repumping  $F : F' = 3 : 4$

(397-2) and 866 nm (866-1) laser frequencies are set to the hyperfine components of  $^{43}\text{Ca}^+$ , which enables us to cool directly  $^{43}\text{Ca}^+$  as the cooling laser frequency (397-1) is close to the resonance of  $^{43}\text{Ca}^+$ . In order to efficiently cool the coolant isotope  $^{44}\text{Ca}^+$ , we irradiated an additional 866 nm laser for repumping  $^{44}\text{Ca}^+$  (866-2) and then observed the spectrum (Fig. 3). The 866 nm laser for  $^{44}\text{Ca}^+$  (866-2) is shut off when the frequency of the cooling laser (397-1) exceeds the resonance of  $^{44}\text{Ca}^+$  to avoid laser heating of  $^{44}\text{Ca}^+$ . Three peaks, for  $^{44}\text{Ca}^+$  and for the hyperfine transitions of  $F : F' = 4 : 4$  and  $4 : 3$  of  $^{43}\text{Ca}^+$ , can be observed and they correspond to their isotope shifts [16, 20]. Compared with the conditions without the additional 866 nm laser for  $^{44}\text{Ca}^+$  (866-2), the cooling of  $^{44}\text{Ca}^+$  is more efficient and due to sympathetic cooling the intensity of  $^{43}\text{Ca}^+$  is improved. The achieved temperatures derived from the fitting of each peak of the isotopes to the Voigt function are 14.8 K for  $^{44}\text{Ca}^+$  and 11.8 K for  $^{43}\text{Ca}^+$ , respectively. We used symmetric shapes of the function for the fitting so that the temperatures may be used as reference, because the experimentally obtained spectra actually drop just above resonances and do not fit the function well. These temperatures, however, show sympathetic cooling of  $^{44}\text{Ca}^+$  works as pre-cooling of  $^{43}\text{Ca}^+$  because the temperature of  $^{43}\text{Ca}^+$  is lower than that of  $^{44}\text{Ca}^+$ .

We estimated the LIF intensities of single  $^{43}\text{Ca}^+$  and  $^{44}\text{Ca}^+$  both to 1.3 kcps based on the detection efficiency derived from our previous experimental results [9]. From the observed peak intensities, we can roughly estimate that the numbers of trapped  $^{43}\text{Ca}^+$  and  $^{44}\text{Ca}^+$  are 30 ions and 390 ions, respectively. That ratio corresponds to the order of the natural abundance of Ca isotopes. The line width of

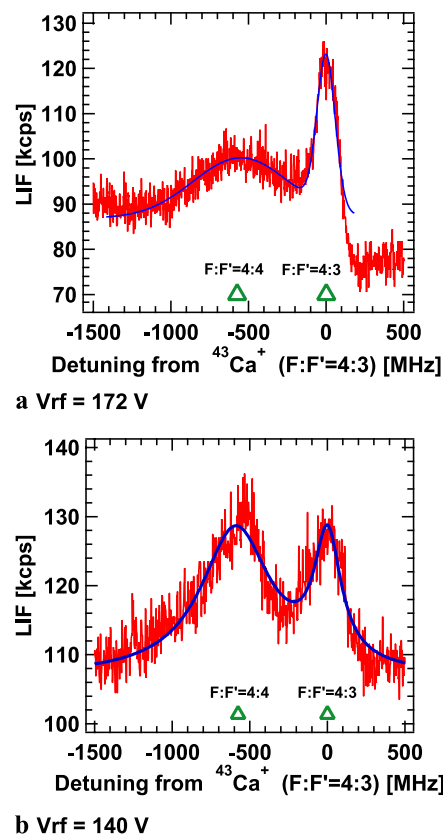


**Fig. 3** Excitation spectra of trapped  $\text{Ca}^+$  isotopes. *Inset:* Spectra of sympathetically cooled  $^{43}\text{Ca}^+$ . The markers ( $\Delta$ ) show the hyperfine transitions of  $F : F' = 4 : 4$  and  $F : F' = 4 : 3$ . The blue lines are fitted to the experimental data. The two peaks correspond to the hyperfine transitions. From the peak intensities, we estimate the numbers of trapped isotopes to be 390 ions for  $^{44}\text{Ca}^+$  and 30 ions for  $^{43}\text{Ca}^+$ , respectively.  $V_{\text{rf}} = 175$  V corresponds to  $q^{43}\text{Ca}^+ = 0.457$ ,  $q^{44}\text{Ca}^+ = 0.454$

$F : F' = 4 : 3$  is narrower than that of  $F : F' = 4 : 4$ . This corresponds to the direct detection of  $^{43}\text{Ca}^+$  in an ion beam experiment [31]. The red side wing of the  $F : F' = 4 : 4$  peak is slightly higher than that of background level. We start the laser frequency scanning from below the peak of  $^{44}\text{Ca}^+$  and turn off the 866 nm laser for  $^{44}\text{Ca}^+$  when the laser frequency exceeds the resonance of  $^{44}\text{Ca}^+$  to suppress the laser heating of  $^{44}\text{Ca}^+$ . Since non-resonant absorption of the laser photons for  $^{43}\text{Ca}^+$  occurs, the laser heating of  $^{44}\text{Ca}^+$  may occur with lower efficiency, which may also affect the peak shape of  $F : F' = 4 : 4$ .

#### 4.2 Effect of the RF trapping voltage

The RF voltage is set to the condition where a nonlinear resonance of  $^{40}\text{Ca}^+$  occurs and only  $^{43}\text{Ca}^+$  and  $^{44}\text{Ca}^+$  can be trapped. Nonlinear resonances occur in the stable region



**Fig. 4** The effect of the RF voltage on the cooling spectra of  $^{43}\text{Ca}^+$ . After sympathetic cooling, we observed the spectra of  $^{43}\text{Ca}^+$  by scanning laser frequency from the resonance  $^{44}\text{Ca}^+$ . The markers ( $\Delta$ ) show the hyperfine transitions  $F : F' = 4 : 4$  and  $F : F' = 4 : 3$ . The blue lines are fitted to the observed spectra. (a)  $V_{\text{rf}} = 172$  V ( $q_{43} = 0.457$ ), 390  $^{44}\text{Ca}^+$  ions and 30  $^{43}\text{Ca}^+$  ions are trapped. (b)  $V_{\text{rf}} = 140$  V ( $q_{43} = 0.372$ ), 100  $^{44}\text{Ca}^+$  ions and 10  $^{43}\text{Ca}^+$  ions are trapped. The LIF intensity and S/N of (a) are higher than those of (b) because of the number of trapped isotopes. However, the line widths of peaks of (b) are narrower than those of (a). The achieved temperature are (a) 12 K and (b) 6.5 K. The intensity of LIF per single ion increases with the temperature

of the Mathieu diagram because due to imperfection of the electrodes the trap potential has higher order components which are not included in the Mathieu equations [30]. After sympathetic cooling, we observed the cooling spectra of  $^{43}\text{Ca}^+$  scanning the laser frequency (397-1) from the resonance of  $^{44}\text{Ca}^+$ . The result of the scanning is shown in Fig. 4. Decreasing of the RF voltage makes the trapping potential shallower so the number of the trapped ions decreases. The Mathieu  $q$  values of Figs. 4(a) and (b) are 0.457 and 0.372, respectively. On the one hand the LIF intensity and S/N are decreased, and on the other hand, the line widths of the both peaks are narrower than those for high RF voltage because of the improvement of the cooling efficiency. The achieved temperature for the  $F : F' = 4 : 3$  transition is reduced from 12 K to 6.5 K. Furthermore, the temperature of the coolant  $^{44}\text{Ca}^+$  also decreases so the effect of the wing of the  $^{44}\text{Ca}^+$  peak may be ignored and the LIF level is almost the same as the background level. However, in our experimental setup we cannot decrease it below  $V_{\text{rf}} = 140$  V because nonlinear resonances are generated and the number of trapped  $^{43}\text{Ca}^+$  may decrease.

## 5 The elimination of the coolant isotope $^{44}\text{Ca}^+$

In principle, by utilizing the nonlinear resonances we should be able to eliminate only  $^{44}\text{Ca}^+$  while leaving  $^{43}\text{Ca}^+$ . However, we could not observe any LIF of  $^{43}\text{Ca}^+$  after the elimination of  $^{44}\text{Ca}^+$ . Our previous study shows the nonlinear resonance of an isotope affects the behavior of simultaneously trapped different isotopes [10]. Due to the distortion of the trapping potential using DC voltage, the elimination of the heavy isotopes can be difficult. Furthermore, the abundance of  $^{44}\text{Ca}^+$  is about ten times higher than that of  $^{43}\text{Ca}^+$ . Therefore,  $^{43}\text{Ca}^+$  can be also eliminated from the trap in the elimination process of  $^{44}\text{Ca}^+$ .

References [16, 20] reported that the 397 nm repumping laser for  $^{43}\text{Ca}^+$  (397-2) is turned off during the isotope-selective loading in order to generate laser heating for the other isotopes. We also tried to eliminate other isotopes with this method, but we could not observe any LIF. Because in our ion loading method a lot of ions enter the trap region simultaneously, the small number of trapped  $^{43}\text{Ca}^+$  also may be heated by other laser heated isotopes and eliminated from the trap. Compared to [20], our trap rf frequency is small so that the amplitude of the rf voltage cannot be decreased so much.

## 6 Conclusion

We developed a hyperfine repumping laser system and magnetic field coils for laser cooling of  $^{43}\text{Ca}^+$ . We found that the

neighboring isotopes,  $^{42}\text{Ca}^+$  and  $^{44}\text{Ca}^+$ , which are simultaneously trapped isotopes, affect the laser cooling of  $^{43}\text{Ca}^+$ . Utilizing the neighboring isotope  $^{44}\text{Ca}^+$  as a coolant ion, we realized sympathetic and direct cooling of  $^{43}\text{Ca}^+$  and observed the peaks corresponding to the hyperfine structure,  $F : F' = 4 : 4$  and  $F : F' = 4 : 3$  even though  $^{44}\text{Ca}^+$  remains in the trap. In order to apply these techniques to the isotope analysis of calcium we should directly observe low abundance isotopes, such as  $^{43}\text{Ca}^+$  and  $^{46}\text{Ca}^+$ . To realize the direct observation, we plan to use the resonant ionization loading method.

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