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# Isotope-selective trapping of rare calcium ions using high-power incoherent light sources for the second step of photo-ionization

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ABSTRACT Rare calcium isotope <sup>48</sup>Ca<sup>+</sup> (0.187%) has been selectively loaded in a linear Paul trap using two ultraviolet light emitting diodes with the output power of 85 mW for the second excitation in a two-step photo-ionization process. Isotope selectivity has been achieved by utilizing the isotope shifts for the  $4s^{2}$   ${}^{1}S_{0}$ -4s4p  ${}^{1}P_{1}$  transition of neutral calcium atom. Sympathetic cooling of  ${}^{48}Ca^{+}$  ions has been demonstrated using  ${}^{40}Ca^{+}$  ions as refrigerant ions. Purification of rare isotope  ${}^{42}Ca^{+}$  ions (0.647%) from a mixture of  ${}^{40}Ca^{+}$  (96.9%) and  ${}^{42}Ca^{+}$  ions has been performed by adjusting the detuning of the cooling laser frequency, which overcomes the imperfect selectivity for some rare isotopes having close resonance frequencies to that of  ${}^{40}Ca$  in the  $4s^{2}$   ${}^{1}S_{0}$ -4s4p  ${}^{1}P_{1}$  transition. The methods can be applied to  ${}^{43}Ca^{+}$  ion (0.135%) that has been considered as one of the attractive candidates for quantum information processing as well as for an optical frequency standard.

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### 1 Introduction

A string of laser-cooled ions confined in a linear Paul trap has been considered as a promising system for the development of quantum information processing (QIP) since the Cirac–Zollar proposal [1]. One of the attractive candidates for QIP is calcium ion because it has a number of advantages as a qubit [2, 3]. Among the naturally occurring calcium isotopes (Table 1), experiments for the development of QIP using calcium ions have been succeeded with the <sup>40</sup>Ca<sup>+</sup> so far [4, 5]. On the other hand, the odd isotope <sup>43</sup>Ca<sup>+</sup> is another candidate since its hyperfine structure is suitable for qubit operation [2]. Other isotopes are also of interest in the experiments of sympathetic cooling [6], which is considered to be one of the required techniques in the construction of a large scale QIP [7–9]. The difficulty in using isotopes other than <sup>40</sup>Ca<sup>+</sup> is their low natural abundances ranging from 2% to 0.004%.

Recently, photo-ionization for the loading of calcium ions into a Paul trap was performed by several groups [10–14]. Besides isotope-selectivity, photo-ionization has a number of advantages: no charging of insulating parts of the trap structure, reduction of unwanted material sputtered onto the trap electrodes from the oven due to its high efficiency [13, 14]. The latter is of importance since investigation of the heating of trapped ions [15] indicates that clean electrode surfaces reduce the heating rate of trapped ions from the motional ground state. Kjærgaard et al. first demonstrated loading of calcium ions [10] using the  $4s^2 {}^1S_0 - 4s5 p {}^1P_1$  transition at 272 nm followed by excitation far above the ionization limit. Gulde et al. demonstrated other scheme with the  $4s^2 {}^1S_0 - 4s4p {}^1P_1$  transition at 423 nm followed by the excitation close to the continuum by the ultraviolet radiation near 390 nm, and showed that photo-ionization is over five orders of magnitude more efficient than the electron bombardment ionization [13]. Using the same transitions, Lucas et al. [14] first performed laser cooling of rare isotopes <sup>43</sup>Ca<sup>+</sup> and <sup>46</sup>Ca<sup>+</sup>. They also showed that the diode laser used for the second excitation in the photoionization of <sup>40</sup>Ca could be replaced by an incoherent light source and pointed out the possibility of loading of the rarer isotopes with a high-power incoherent light.

In this paper, we report selective loading of rare calcium isotopes using high-power light emitting diodes (LEDs) for the second excitation in photo-ionization, which greatly simplifies a setup of light sources. We employ the photoionization scheme demonstrated by Gulde et al. [13], so isotope selectivity is achieved by utilizing the isotope shifts for the  $4s^2 \, {}^{1}S_{0}-4s4p \, {}^{1}P_{1}$  transition. First we directly laser-cool the rare isotope  ${}^{48}Ca^{+}$  ions that are selectively loaded. We also demonstrate sympathetic cooling of the selectively loaded  ${}^{48}Ca^{+}$  ions by use of the laser-cooled  ${}^{40}Ca^{+}$  ions. We perform purification of  ${}^{42}Ca^{+}$  ions from a mixture of  ${}^{40}Ca^{+}$  and  ${}^{42}Ca^{+}$ ions, which overcomes the imperfect selectivity of rare iso-

Mass number	Natural abundance	Isotope shifts [MHz] Ca $4s^2 {}^1S_0 - 4s4p {}^1P_1$
40	96.9 %	0
42	0.647 %	394
43	0.135 %	612
44	2.09 %	774
46	0.004 %	1160
48	0.187 %	1513

**TABLE 1** Abundances of naturally occurring calcium isotopes and isotope shifts for the  $4s^2 {}^1S_0$ - $4s4p {}^1P_1$  transition at 423 nm

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topes having close resonance frequencies to that of <sup>40</sup>Ca in the  $4s^{2} {}^{1}S_{0}$ – $4s4p {}^{1}P_{1}$  transition. Since we have only one cooling laser at present, we can laser-cool and observe only even isotopes. However, the methods demonstrated here could be applied for the <sup>43</sup>Ca<sup>+</sup> ion that is an attractive candidate not only for QIP but also for an optical frequency standard based on trapped ion.

# 2 Photo-ionization scheme and experimental apparatus

The schematic diagram of energy levels of neutral calcium is shown in Fig. 1a as well as that of singly ionized calcium (Fig. 1b). Isotope-selectivity is achieved by use of the isotope shifts for the  $4s^2 {}^1S_0 - 4s4p {}^1P_1$  transition. A 423-nm ultraviolet diode laser is available for excitation of the transition. Since the natural linewidth of the transition (35 MHz) is sufficiently small compared with the isotope shifts [16] (Table 1), it is expected that perfect selectivity of isotopes is achievable. In practice, however, purification is necessary for some isotopes due to their very low abundances compared with <sup>40</sup>Ca and the residual Doppler broadening. The second step of the photo-ionization is achieved by a photon near 390 nm [13]. Not only a coherent laser source but also an incoherent light source is available for the second excitation [14]. After photo-ionization and trapping, laser cooling is performed with the  $4s^2S_{1/2}-4p^2P_{1/2}$  transition at 397 nm and the  $4p^2 P_{1/2} - 3d^2 D_{3/2}$  transition at 866 nm as a repumping process. Isotope shifts for the  $4s^2S_{1/2}-4p^2P_{1/2}$  transition can be utilized for cooling specific isotopes while heating other isotopes by adjusting the detuning of the 397-nm laser frequency. Details will be described in Sect. 3.

All of coherent light sources in our setup, i.e. the light source for the first step of the photo-ionization and those for laser cooling of the singly ionized calcium, are diode lasers as shown in Fig. 2a. Except one 866-nm laser for repumping, each diode laser forms an external-cavity diode laser (ECDL) with a diffraction grating in the Littrow configuration. Each ECDL is frequency-stabilized by use of a temperature-controlled Fabry–Perot cavity that has a thin Brewster glass plate inside. A dispersion-shape error signal is obtained by means of the Hänsch–Couillaud method [17] then fed back to the current and the PZT voltage of the ECDL through a PI controller. Frequency drift of the ultraviolet ECDL system is typically less than 5 MHz for 200 seconds and that of the near infrared ECDL system is less than 1 MHz for the same period. The output of the 397-nm ECDL is re-



FIGURE 1 Schematic diagrams of energy levels of neutral (a) and singly ionized (b) even calcium isotopes. Relevant wavelengths for photo-ionization and laser cooling are also shown



**FIGURE 2** (a) Experimental setup for the photo-ionization and laser cooling. ECDL, external-cavity diode laser; LD, laser diode; LED, light emitting diode; L, lens; BS, beam splitter; DM, dichroic mirror; CM, concave mirror; PMT, photomultiplier. The ECDL system consists of an external-cavity diode laser, a temperature-controlled Fabry–Perot cavity, and optical components for locking the laser frequency to the resonance of the cavity. (b) Linear trap assembly. Optical paths of lasers and LEDs irradiated into the trapping region are also shown. After passing through the trap, the LEDs are reflected back to the trapping region by the concave mirrors. Calcium beam is set to be perpendicular to the 423-nm laser beam that propagates along the trap axis (*z* axis). The trap assembly is placed in the vacuum chamber

flected with a grating then passed through a spatial filter with the diameter of  $100 \,\mu\text{m}$  to eliminate background incoherent radiation [18, 19], then introduced to a single-mode fiber to obtain a proper spatial mode.

The second excitation in the photo-ionization is driven with two high power ultraviolet LEDs (Nichia NCCU001E) with the output power of 85 mW, the peak wavelength of 380 nm, and the spectral width of 15 nm. Using LED is preferable because it is much cheaper than diode laser and no stabilization of current and temperature is required for its operation. However, it is difficult to collimate the output of the LED due to its very poor directivity. Optical components for collimating the LED are composed of one condenser lens with the focal length of 10 mm and three plano-concave lenses. In the current setup, the distance between the LED and the trap center is not same for the LED1 and for the LED2, so combinations of the focal lengths of the plano-concave lenses are different. After passing through the vacuum chamber, both beams are reflected back to the trapping region with concave mirrors. Since the image of the LED does not have a Gaussian profile but a complicated structure, it is difficult to estimate the intensity at the ion. The averaged intensities at the trapping region of the LED1 and LED2 propagating forward were about  $20 \text{ mW/cm}^2$  and  $5 \text{ mW/cm}^2$ , respectively.

Linear trap assembly placed in a vacuum chamber is formed of four rods and two dc electrodes at the edge of the rods (Fig. 2b). The diameter and length of the rods are 5 mm and 50 mm, respectively. The perpendicular distance from the trap axis to the trap electrodes is 2.5 mm. The trap is operated with an alternating voltage of 5.25 MHz and 240 V<sub>amp</sub>. To confine the ions axially, a dc voltage of 50 V is applied to the end electrodes. Calcium oven is set under the trap so that the calcium beam intersects the trapping region in perpendicular to the 423-nm laser beam irradiated along the trap axis. Two apertures having 1-mm diameter hole are used for collimation of the calcium beam to avoid the first-order Doppler shifts in the  $4s^2 {}^1S_0 - 4s4p {}^1P_1$  transition as possible. The power and intensity of the 423-nm laser at the trapping region were approximately  $18 \,\mu\text{W}$  and  $85 \,\text{mW/cm^2}$ , respectively. This intensity is comparable with the saturation intensity of the  ${}^{1}S_{0}$ - ${}^{1}P_{1}$  transition  $(I_{\text{sat}} = \pi hc/3\lambda^{3}\tau = 60 \text{ mW/cm}^{2})$ , where h is Planck's constant, c is the light velocity,  $\tau$  is the lifetime, and  $\lambda$  is the wavelength). At the lower intensity, it is difficult to tune the 423-nm laser frequency accurately to the resonance of the rare isotopes at the typical oven current in our setup. Conventional loading with an electron gun is also available in our system. The 397-nm and 866-nm diode laser beams are overlapped with a dichroic mirror then irradiated to the trapping region from the opposite side to the 423-nm beam. Fluorescence intensity from the calcium beam or trapped ions is observed with a photomultiplier followed by a photon counter and a recorder. An interference filter for blocking the near infrared radiation and a spatial filter for reducing the stray scattered light are placed in front of the photomultiplier.

### 3 Results

PMT counts [10<sup>4</sup>counts/sec]

-1

Fluorescence spectra from the neutral calcium beam obtained by scanning the frequency of the 423-nm laser about 3 GHz are shown in Fig. 3. Since the signal from the <sup>40</sup>Ca is much larger than those of other isotopes, only fluorescence from the <sup>44</sup>Ca can be observed in the same scale as shown in the inset. As the vertical axis is represented in linear scale and magnified to observe the rare isotopes, the peak of the <sup>40</sup>Ca signal is truncated. All of isotopes other than <sup>46</sup>Ca (0.004%) were observed. The linewidth of the spectra



Frequency detuning at 423 nm [GHz]

0

**FIGURE 3** Fluorescence spectra from neutral calcium beam for the  $4s^2 {}^1S_0 - 4s4 p {}^1P_1$  transition. The frequency of the 423 nm diode laser is scanned about 3 GHz. The vertical axis is represented in linear scale, which is the reason why the spectrum from  ${}^{40}Ca$  is truncated. The whole profile of the spectrum from  ${}^{40}Ca$  is shown in the *inset* 

was measured to be about 70 MHz, which is larger than the natural linewidth. This is considered to be due to the power broadening and the residual Doppler broadening, although we collimate the calcium beam with two apertures. As shown in the Fig. 3, spectra of the <sup>42</sup>Ca, <sup>43</sup>Ca, and <sup>44</sup>Ca isotopes and the tail of the <sup>40</sup>Ca spectrum overlap, which indicates a purification process is required for these isotopes.

Selective loading and laser cooling of <sup>48</sup>Ca<sup>+</sup> were performed as follows. First we tune the 423-nm laser frequency to the <sup>48</sup>Ca resonance with monitoring the fluorescence from the atomic beam. After the photomultiplier is turned off, the LEDs and the ECDLs are irradiated to the trapping region for 2 minutes at the oven current of 2.1 A. To cool the trapped ions, the 397-nm laser is detuned about 400 MHz to the long wavelength side of the resonance. The 397 nm radiation does not function as a light source for the second excitation of photo-ionization. After loading, the current of the oven, 423-nm laser and the LEDs are turned off, then the fluorescence is observed with the photomultiplier. The most probable impurity is <sup>40</sup>Ca<sup>+</sup>, whose percentage is calculated to be 10% based on the Voigt profile with the linewidth of 70 MHz. However, since the isotope shift in the 866-nm transition is more than 8 GHz, we can confirm which isotope, <sup>40</sup>Ca<sup>+</sup> or <sup>48</sup>Ca<sup>+</sup>, contributes the observed fluorescence signal. Figure 4 shows a fluorescence profile of the laser-cooled <sup>48</sup>Ca<sup>+</sup> as the frequency of the 397-nm laser is scanned over the  $4s^2S_{1/2}-4p^2P_{1/2}$  transition. The discontinuity of the spectrum represents that crystallization occurred. This spectrum was obtained using two LEDs, however, we found that one of them is enough for photo-ionization of <sup>48</sup>Ca. When using only LED2, the fluorescence intensity at the peak of the spectrum was about one-third of that observed in Fig. 4.

Sympathetic cooling was performed with  ${}^{48}Ca^+$  ions using  ${}^{40}Ca^+$  ions as cooling ions. Photo-ionization of the  ${}^{48}Ca$ isotope was carried out with the 423-nm laser and the LEDs as mentioned above. The  ${}^{40}Ca^+$  ions were loaded in the conventional electron bombardment method because we have only one 423-nm laser system and the possibility of loading  ${}^{48}Ca^+$ with the electron bombardment method can be negligible in our experimental condition. During loading, the 397-nm laser was detuned by 400 MHz to the long wavelength side from the resonance of  ${}^{40}Ca^+$ . The detuning is about 2.1 GHz lower from the  ${}^{48}Ca^+$  resonance considering the isotope shift for



**FIGURE 4** Fluorescence spectrum from selectively loaded  ${}^{48}Ca^+$  ions obtained by scanning the 397-nm laser frequency

the 397-nm transition [20] of 1696 MHz. Therefore, we consider that the 397-nm laser effectively cools only the <sup>40</sup>Ca<sup>+</sup> ions. As for the 866-nm transition, two lasers whose frequencies were different by the isotope shift [21] of about 8.3 GHz were irradiated. The oven current and loading time were 2.1 A and 2 minutes, respectively. Figure 5 shows the fluorescence spectra obtained by scanning the 397-nm laser from lower frequency to the resonance. The frequency difference between higher and lower peaks is in coarse agreement with the isotope shift between the <sup>40</sup>Ca<sup>+</sup> and <sup>48</sup>Ca<sup>+</sup> ions. Note that the ratio of the photon-counting rates of these peaks is much different from the ratio of the natural abundances. It can be considered that <sup>48</sup>Ca<sup>+</sup> ions are selectively loaded by photo-ionization then sympathetically cooled by the <sup>40</sup>Ca<sup>+</sup> ions.

In the case of the rare isotopes  ${}^{42}Ca$ ,  ${}^{43}Ca$ , and  ${}^{44}Ca$ , we cannot achieve good selectivity by the photo-ionization scheme. As a benchmark of the future experiment with  ${}^{43}Ca^+$ , we performed the purification of  ${}^{42}Ca^+$  whose natural abundance is similar to that of  ${}^{43}Ca^+$  from the mixture of  ${}^{40}Ca^+$  and  ${}^{42}Ca^+$ . The elimination of the unwanted isotope can be realized by adjusting the detuning of the 397-nm laser so that the laser cools the required isotope and simultaneously heats the unwanted isotope [14, 22, 23]. The isotope shift between  ${}^{40}Ca^+$  and  ${}^{42}Ca^+$  for the 397-nm transition is 425 MHz [20] and the  ${}^{40}Ca^+$  resonance lies in the long wavelength side. Detuning of the 397-nm laser by 200 MHz to the long wavelength side from the  ${}^{42}Ca^+$  resonance allows us to eliminate the  ${}^{40}Ca^+$  while cooling  ${}^{42}Ca^+$ .

For a reference, first we photo-ionized the <sup>42</sup>Ca without eliminating <sup>40</sup>Ca<sup>+</sup>. While loading, we detuned the 397-nm laser by 200 MHz to the long wavelength side from the  ${}^{40}Ca^+$ , so both of the isotopes were laser-cooled by the 397-nm laser. Two 866-nm lasers were irradiated simultaneously. The loading time was 2 minutes at the oven current of 2.0 A. The result shown in Fig. 6a indicates that large number of <sup>40</sup>Ca<sup>+</sup> were trapped and laser-cooled. Next we loaded the ions in the same manner as above, but detuned the 397-nm laser by 200 MHz to the long wavelength side from the  ${}^{42}Ca^+$ , that is, the laser cools the  ${}^{42}Ca^+$  but heats the  ${}^{40}Ca^+$  (Fig. 6b). The ratio of the peaks changed but a mixture of the two isotopes is still observed. We considered that it might be difficult to heat the <sup>40</sup>Ca<sup>+</sup> and cool the <sup>42</sup>Ca<sup>+</sup> as long as the ions are in the cloud state, that is, the resolution of the spectra is not good enough. Finally, we shortened the loading time (50 seconds) to load



FIGURE 5 Fluorescence spectra from laser-cooled  ${}^{40}Ca^+$  ions and sympathetically cooled  ${}^{48}Ca^+$  ions

smaller number of ions. In this case, we could observe only spectrum from  ${}^{42}Ca^+$  (Fig. 6c).

### 4 Discussion

The effect that should be taken into account as a process of trapping impurities is charge exchange [12, 14]. In the calcium experiments, if n is a mass number of a rare calcium isotope, the following process can occur during loading:

$${}^{40}\text{Ca} + {}^{n}\text{Ca}^{+} \rightarrow {}^{40}\text{Ca}^{+} + {}^{n}\text{Ca}^{+}$$

Due to this process, the rare isotope that is selectively loaded could be replaced by  ${}^{40}Ca^+$  because most of the isotope in the neutral calcium beam is  ${}^{40}Ca$ . We observed the effect of the charge exchange in our setup as follows. We loaded and laser-cooled  ${}^{44}Ca^+$  ions then heated the calcium oven with monitoring the spectra by repeatedly scanning the 397-nm



**FIGURE 6** Fluorescence spectra from  ${}^{40}Ca^+$  and  ${}^{42}Ca^+$  ions. (a) The loading time was 2 minutes. The 397-nm laser was detuned so that both isotopes are laser-cooled. (b) Same as (a) except for the detuning of the 397-nm laser. The detuning was 200 MHz to the long wavelength side of the  ${}^{42}Ca^+$  resonance so that  ${}^{42}Ca^+$  are laser-cooled while  ${}^{40}Ca^+$  are heated. (c) The loading time was 50 seconds. The detuning was 200 MHz to the long wavelength side of the  ${}^{42}Ca^+$  resonance

laser frequency about 1200 MHz. The scanning range was set to cover the isotope shift between the  ${}^{40}Ca^+$  and  ${}^{44}Ca^+$  for the 397-nm transition (842 MHz [20]). Two 866-nm lasers were irradiated so that we could detect both  ${}^{40}Ca^+$  and  ${}^{44}Ca^+$  ions if they were simultaneously confined. Four minutes after the oven was turned on, we observed a small peak of fluorescence appeared at the resonance frequency of the  ${}^{40}Ca^+$  ions. We consider that the charge exchange would not be dominant during the typical loading time in our current setup and that this process would not be a cause of trapping impurity ions.

The other effect to be considered is the possibility of loading ions only by the 390 nm radiation as reported in [14]. We checked it by irradiating only LEDs during the loading time but no ion was observed. This may be due to relatively large dimension of our trap.

# 5 Conclusion and outlook

We have performed selective loading of a rare calcium isotope whose natural abundance is 0.187% using two high-power LEDs as the light sources in the second excitation of the photo-ionization process. Though the 390-nm radiation can be generated with a UV diode laser, utilizing LED is preferable because neither a stable current source nor a temperature controller is required for its operation.

Sympathetic cooling of  ${}^{48}Ca^+$  by laser-cooled  ${}^{40}Ca^+$  has been demonstrated with the same setup. Sympathetic cooling is one of the key technologies for the large scale QIP [7–9] and several groups investigated with isotopes [24] or different species [25] for this purpose. Using isotopes is advantageous since the heating rates of modes are expected to be low in the case of ion species of nearly equal mass if we choose appropriate modes [26]. It also requires a relatively simple experimental setup because many common optical components can be used for cooling or detecting of different isotopes. Although the different species of ions would be preferable when the effect of the spontaneous emission is taken into account [25], sympathetic cooling with calcium isotopes will be useful for preliminary experiments such as mode analysis or formation of an ion string with controlled number of isotopes.

We have also observed that purification of the rare isotopes is possible even though the isotope shifts are not large enough to resolve different isotopes. New lasers for laser cooling of the odd isotope  ${}^{43}Ca^+$  are now in preparation. We will apply the method demonstrated here for  ${}^{43}Ca^+$  ion that is considered to be a target of the QIP as well as the optical frequency standard based on calcium ion.

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