Removal of irrelevant isotope ions in the presence of laser cooling in a rf trap

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Abstract. An additionally applied rf perturbation excites the secular motion of isotope ions selectively in a rf trap. Simultaneous application of laser cooling has achieved complete removal of irrelevant isotope ions and kept a large number of relevant ones.

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A rf trap is expected to be a prospective device for frequency standards [1], high-resolution spectroscopy [2], and quantum-mechanical experiments related to quantum jump [3] and quantum electrodynamics [4], because it can offer an ideally isolated ion system. For these applications, the technique of laser cooling [5, 6] is one of the essential elements because the trapped ions should be at extremely low temperature. If the trapped ion species has some isotopes, they are usually confined simultaneously in the rf trap. In the presence of other isotopes the ion temperature may be high because of the rf heating [7]. In this study a novel in situ method of the selective removal of irrelevant isotopes is proposed and successfully demonstrated.

In the present experiment, isotopes of magnesium ions of natural abundance in the rf trap with two cap electrodes and one ring electrode are selectively removed. Magnesium ions can be laser-cooled by a single laser of 280 nm at high efficiency because of the high spontaneous emission rate (43 MHz) and are advantageous as coolant ions of sympathetic cooling [8, 9]. The natural abundance ratio of stable magnesium isotopes is $^{24}Mg : ^{25}Mg : ^{26}Mg = 8 : 1 : 1$. $^{25}Mg^+$ and $^{26}Mg^+$ ions are removed as a demonstration of the isotope removal in the experiment. In the method of isotope removal, the additionally applied rf field selectively excites isotope ion motion in the trap potential and drives them out of the trap. The principle of mass selection is based on the difference of the charge-to-mass ratio of each isotope, as commonly used in mass separation techniques. The dependence of the resonance (secular) frequency of ion motion in the trap potential on the charge-to-mass ratio is used. In [10] the isotope separation of europium ions is performed with the numerous resonances of ion motion caused by the deviations from the ideal harmonic potential. In the reference [11] isotope separation of cadmium ions in a linear rf trap is carried

out only by the laser cooling, but a few irrelevant ions remain because of the effect of sympathetic cooling. The merits of using the additional rf field in this study are not only that the separation efficiency depends less on the structure of the apparatus, but also that the ion motion can be controlled easily by adjusting the additional rf amplitude. To keep many relevant ions in the trap, the laser cooling is simultaneously applied, and as a result achievements in purity and trapped number of relevant ions are substantially improved.

1 Principle

The experimental apparatus is an ordinary type [12]. The trap electrodes of hyperboloid of revolution are arranged in the conventional quadrupole configuration. The ac voltage of the frequency of $\Omega/2\pi$ and the amplitude of V_{ac} is applied between a ring and two cap electrodes, and no dc voltage is applied. The ion motion can be assumed to be harmonic in the time-averaged pseudopotential. The frequency of the secular oscillation along the symmetry axis of revolution is approximately given by

$$\omega_s/2\pi = \frac{\sqrt{2}eV_{ac}}{\pi m \Omega \left(r_0^2 + 2z_0^2\right)} . \tag{1}$$

Here, e and m are the charge and the mass of the trapped ion, respectively. r_0 and $2z_0$ are the radius of the ring electrode and the spacing between the cap electrodes, respectively.

For the excitation of the secular motion along the symmetry axis of the trap, an additional rf perturbation voltage (V_p) is applied between the top and bottom cap electrodes. When the frequency of the perturbation coincides with the secular frequency, the ion motion is resonantly excited and the ions are expelled spatially from the trap.
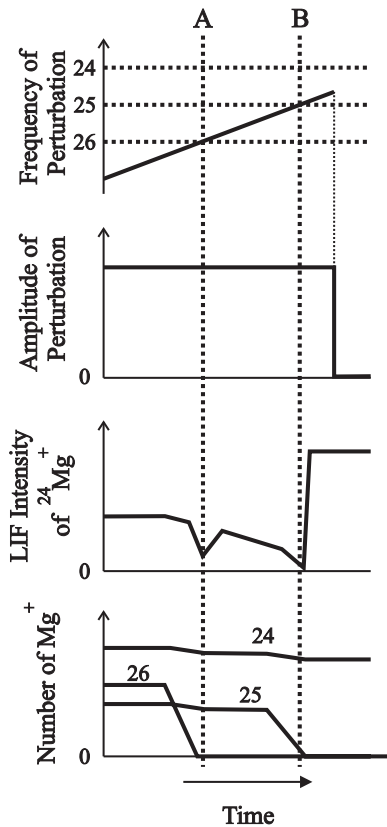
In the cloud of two or more ion species, there are two kinds of resonant motion [13], namely, the individual oscillation, in which each ion moves in a harmonic pseudopotential independently and the secular frequency depends on the charge-to-mass ratio of each ion e_i/m_i, and the collective oscillation, in which the whole ions move collectively in a potential and the secular frequency depends on the charge-to-mass ratio of the whole ions $\sum e_i / \sum m_i$. In the case of

the simultaneous trapping of the isotope ions, the ions tend to move collectively because the mass difference, and therefore the difference in the secular frequencies of the individual oscillations, are small [13]. However, the phase correlation among the individual ion motions may be broken at the larger trajectory of the ions caused by applying the larger perturbation amplitude. Then it is expected that the selectively excited ion species is removed from the trap.

The relevant ions, however, are also excited by the perturbation through the collision with the irrelevant ions. In order to prevent the loss of the relevant ions by the indirect excitation, the selective laser cooling is employed simultaneously. The laser-induced fluorescence (LIF) caused by the cooling laser is used to monitor the behaviors of ions [14].

The parametric instability in the quadrupolar perturbation field applied between the ring and the cap electrodes is reported [15, 16]. In the present experiment the ions are subjected to forced oscillation in the dipolar perturbation field applied between two cap electrodes. The dipolar field is not zero at the center of the trap and can excite the motion of ions located at the center.

The timing chart of the removal procedure of $^{25}Mg^+$ and $^{26}Mg^+$ is schematically shown in Fig. 1. The laser frequency is fixed at which $^{24}Mg^+$ is cooled during the procedure. The frequency of the rf perturbation for the excitation of ion motion is swept from the lower to the higher (the top chart in Fig. 1). The perturbation is turned off at the end of the frequency sweep (the second chart). The LIF intensity of $^{24}Mg^+$ is monitored throughout the procedure (the third chart). The fourth chart shows the number of each isotope. The perturbation frequency is increased and coincides with the secular frequency of $^{26}Mg^+$ (at the vertical line A in Fig. 1), where the motion of $^{26}Mg^+$ is resonantly excited and they are expelled from the trap. At this point, the motion of $^{24}Mg^+$ is also excited through the collision with $^{26}Mg^+$. The density of $^{24}Mg^+$ becomes low because of this heating effect and the LIF intensity of $^{24}Mg^+$ at the center of the trap decreases. As the perturbation frequency is further increased, the LIF intensity recovers. When the perturbation frequency coincides with the secular frequency of $^{25}Mg^+$ (at B in Fig. 1), $^{25}Mg^+$ ions are similarly removed. Now the ion cloud contains only $^{24}Mg^+$ and then the lower ion temperature is achieved in the absence of the rf heating caused by the collisions with other isotopes. Consequently the ion density increases and the LIF intensity increases drastically, which is used as a trigger to stop the frequency sweep.

When pure ions composed of only $^{25}Mg^+$ are required, for example, the perturbation frequency is swept from lower for the removal of $^{26}Mg^+$ and from higher for the removal of $^{24}Mg^+$ and the laser frequency is fixed to cool $^{25}Mg^+$, in cooperation with a microwave radiation which repumps $^{25}Mg^+$ back to the cooling cycle.
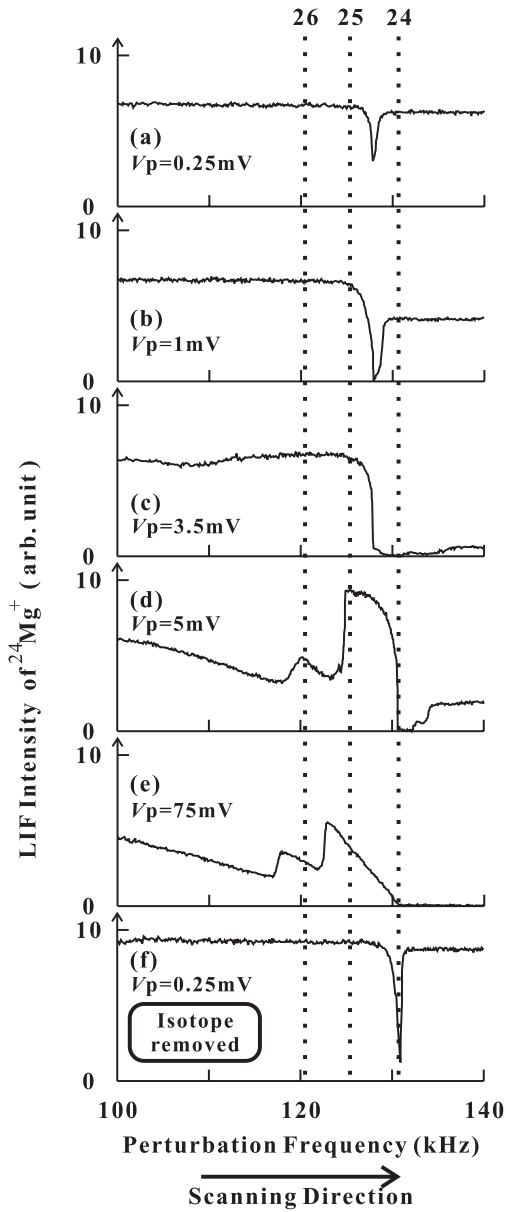
2 Experimental setup

In this experiment the removal of $^{25}Mg^+$ and $^{26}Mg^+$ is tried as an example. The experimental setup is as follows. r_0 is 7.5 mm and $2z_0$ is 10.8 mm. The pressure in the vacuum chamber is 10^{-7} Pa. For the cooling and the detection of $^{24}Mg^+$, the ultraviolet (UV) laser light at 280 nm is used, which excites the ions from the $3s\ ^2S_{1/2}$ state to the $3p\ ^2P_{3/2}$ state. The UV light is obtained by the second-harmonic generation of the tunable dye laser at wavelength $\lambda = 560$ nm. The absolute frequency of the laser is measured with the etalon fringes and the calibrated fluorescence lines of iodine molecule, and tuned at 200 MHz below the frequency of the $^{24}Mg^+$ fluorescence line. The laser power is about 1 mW. The LIF of $^{24}Mg^+$ is detected by a photomultiplier tube. The amplitude and the frequency of the trapping rf voltage (V_{ac} and $\Omega/2\pi$) are 107 V and 2.06 MHz, respectively. The secular frequency of $^{24}Mg^+$ is calculated to be 130.7 kHz from (1). Magnesium atoms from the heated oven are ionized at the center of the trap by electron bombardment. The initial ratio of isotope abundance of trapped ions is natural one.

3 Results and discussions

In Fig. 2, the results of the observed LIF signal of $^{24}Mg^+$ are shown as a function of the perturbation frequency for several perturbation amplitudes V_p. The perturbation frequency $\omega_p/2\pi$ is swept from 100 kHz to 140 kHz in 80 s.

In the case of $V_p = 0.25$ mV (Fig. 2a), the individual motion is not excited but the collective motion is. Then the dip of LIF intensity appears at the frequency which is determined

Fig. 1. Schematic timing chart for the isotope removal. The *horizontal lines* in the top chart imply the secular frequencies of $^{26}Mg^+$, $^{25}Mg^+$, and $^{24}Mg^+$, respectively. The *vertical lines* A and B correspond to the moments at which the perturbation frequency coincides with the secular frequencies of $^{26}Mg^+$ and $^{25}Mg^+$, respectively

Fig. 2. The observation of LIF intensity of ^{24}Mg$^+$ as a function of the perturbation frequency for several perturbation amplitudes before (a–e) and after (f) the isotope removal. Perturbation frequency is scanned from 100 kHz to 140 kHz in 80 s. The perturbation amplitude is shown in the figure. The secular frequency of each isotope is shown by the *vertical line*

by the averaged mass and does not appear at three frequencies corresponding to those of individual isotopes. In the case of $V_p = 1$ mV (Fig. 2b), the LIF intensity of ^{24}Mg$^+$ does not recover completely because of the loss of the ions. In the case of $V_p = 3.5$ mV (Fig. 2c), most of the ions are lost when the perturbation frequency coincides with the resonance frequency of the collective ion motion. The isotope removal is impossible with these perturbation amplitudes.

On the other hand, when the larger perturbation is applied (5 mV in Fig. 2d and 75 mV in Fig. 2e), the individual motion is excited and the resonances occur at three frequencies. The drastic increases of LIF of ^{24}Mg$^+$ mentioned above are observed at 125 kHz for (d) and at 122 kHz for (e), respectively. When the perturbation frequency coincides with the

resonance frequency of ^{24}Mg$^+$, the LIF intensity decreases and does not recover because of the loss of ^{24}Mg$^+$.

In Fig. 2d,e the increases of the LIF intensity of ^{24}Mg$^+$ by the loss of ^{25}Mg$^+$ start to occur at a lower frequency than the resonance frequency of ^{25}Mg$^+$. The reason for the removal at a lower frequency is that the amplitude of the motion of ^{25}Mg$^+$ exceeds a critical trapping size, which is determined by the dimensions of the trapping electrodes, and ^{25}Mg$^+$ ions are removed before the perturbation frequency reaches the exact resonance. This interpretation may be accepted as follows. Assuming the linear damping force of the ion motion as $4\pi m \kappa v$, where v is the velocity of the ion and $2\pi\kappa$ is a damping coefficient, the amplitude of the ion motion Z is represented by

$$Z = \frac{eV_p}{8\pi m z_0 \omega_s} \frac{1}{\sqrt{\kappa^2 + \delta^2}}, \tag{2}$$

where δ is a detuning of the perturbation frequency from the resonance. This equation is derived by solving the equation of motion of the forced oscillation in the harmonic potential and assuming $\kappa, \delta \ll \omega_s/2\pi$. If Z exceeds a constant Z_{cr} at the perturbation frequency detuning of δ_{cr}, the ions of ^{25}Mg$^+$ are removed and the LIF intensity increases. The square of δ_{cr} is a linear function of the square of V_p and the observed result is shown in Fig. 3. From the gradient and the ordinate intercept of the fitted line, it is found that $\kappa = 0.3$ kHz and $Z_{cr} = 1$ mm. The value of the damping coefficient κ agrees with the line width of the spectral line in Fig. 2a.

It should be mentioned that the continuous decrease of the LIF intensity around 125 kHz in Fig. 2e is not due to the loss of ^{24}Mg$^+$ but due to the increase of ion temperature. This is confirmed by the result that no hysteresis is observed when the perturbation frequency is scanned from the higher (but lower than the resonance of ^{24}Mg$^+$) to the lower. By the interruption of frequency sweep after the increase of LIF and before the loss of ^{24}Mg$^+$, the ion cloud composed of only ^{24}Mg$^+$ can be obtained. The result of Fig. 2a–e implies that the perturbation amplitude should be larger than 5 mV to remove irrelevant isotopes in this experimental setup.

Figure 4 shows the isotope removal procedure as a function of time in the case of $V_p = 10$ mV. The result is reproducing the third chart of Fig. 1. Figure 2f is the chart of repeated
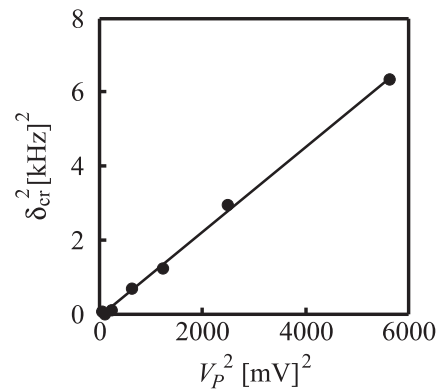
Fig. 3. The dependence of the square of δ_{cr} on the square of V_p. The *circles* of the experimental results and the *line* fitted by the method of least squares are shown
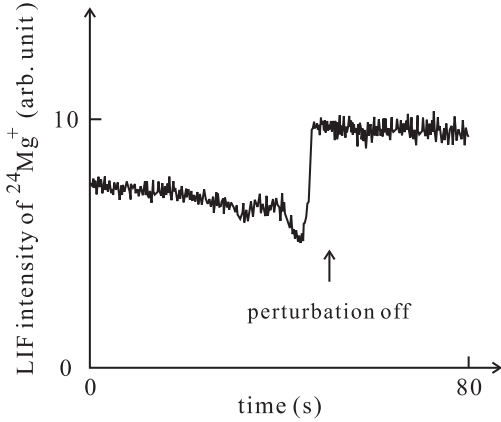
Fig. 4. The LIF intensity of $^{24}Mg^+$ during the isotope removal procedure. The perturbation frequency sweep is 0.4 kHz/s from 110 kHz. The perturbation amplitude is 10 mV. At the moment denoted by an arrow in the figure, the perturbation amplitude is set to be zero

frequency scan after the isotope removal of Fig. 4 is carried out. The resonance appears only at the secular frequency of $^{24}Mg^+$ and hence it is found that the other isotopes are almost removed.

It should be noted that no increase of LIF of $^{24}Mg^+$ is observed when the perturbation frequency is fixed a little lower than δ_{cr} of $^{25}Mg^+$ for a while. When the perturbation frequency is increased a little from there, the LIF intensity of $^{24}Mg^+$ starts to increase immediately. This result implies that each ion of $^{25}Mg^+$ is expelled not gradually but immediately when the expelling condition of the perturbation is realized. Therefore, the removal of $^{26}Mg^+$ and $^{25}Mg^+$ is independent of the sweep rate of the perturbation frequency.

The isotope removal is confirmed by the observation of LIF spectral lines of Mg^+ as shown in Fig. 5. Before the isotope removal (Fig. 5a), the spectral line of $^{26}Mg^+$ is also observed. The LIF line of $^{25}Mg^+$ is absent because of poor laser-cooling efficiency as a result of the presence of hyperfine levels. The area of each LIF spectral line in Fig. 5a corresponds to the isotope abundance and it is found that $^{24}Mg^+ : ^{26}Mg^+ = 8.4 : 1$. This result is consistent with the natural abundance of magnesium. After the isotope removal
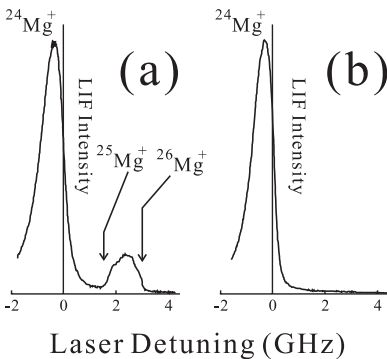
Fig. 5a,b. LIF spectral lines of trapped Mg^+ before (**a**) and after (**b**) the isotope removal. The abscissa implies the detuning of the laser frequency from the line center of $^{24}Mg^+$. The LIF intensities are normalized to equal the peak intensities in both cases

(Fig. 5b), LIF lines of $^{25}Mg^+$ and $^{26}Mg^+$ are not observed. The present detection system can catch a single ion of $^{26}Mg^+$. Consequently there are no $^{26}Mg^+$ ions after the isotope removal. It is concluded that 100% pure isotope ions composed of only $^{24}Mg^+$ are obtained in the trap.

The ion temperatures in both cases in Fig. 5 can be roughly estimated from the deviation of the spectral line peak from the atomic center frequency and the line width [17]. The ion temperature before the isotope removal is about 10 K, whereas it is about 100 mK after the removal. In this experiment the laser power broadening mainly contributes to the line width.

It is interesting to know how many $^{24}Mg^+$ ions are left in the trap after the isotope removal. It depends on the laser-cooling condition, namely, the laser detuning and power. For example, when the laser power is 250 μW, only a few $^{24}Mg^+$ ions remain, and no increase of LIF of $^{24}Mg^+$ is observed. On the other hand, when the laser power is 1 mW, more than 5000 $^{24}Mg^+$ ions remain (the ion number is estimated by the LIF intensity and the detection efficiency). The obtained ion cloud consists of sufficiently many ions to be applied to the experiments such as that of sympathetic cooling.

Next, we roughly estimate the cooling rate and the necessary time for the LIF increase caused by the loss of $^{25}Mg^+$. The kinetic energies of the trapped ions before and after the LIF increase are $Nk_B T_i$ and $Nk_B T_f$, respectively. Here, N is the number of the trapped ions, k_B the Boltzmann constant, and T_i and T_f the ion temperatures before and after the LIF increase, respectively. At the moment of increase of LIF intensity, the ions are cooled from 10 K to 100 mK. The laser-cooling rate may be estimated from $n\gamma h\Delta$, where n is the number of the ions irradiated by the cooling laser, γ the cycle of the laser-cooling process, h the Planck constant, and Δ the laser detuning. Therefore, the necessary time of the cooling process t is represented by

$$t = \frac{Nk_B (T_i - T_f)}{n\gamma h\Delta} . \qquad (3)$$

The ratio of N/n is estimated to be 10^3 in this experiment from the dimensions of ion cloud and the laser beam radius. If the laser is intense enough to saturate the transition, γ is the spontaneous emission rate. Then t is calculated to be about 2 s. The agreement of this calculation with the experimental result shown in Fig. 4 implies that the LIF increase is due to the condensation of $^{24}Mg^+$ caused by the effective laser cooling in the absence of rf heating by the isotopes.

The critical (lowest) amplitude of the perturbation for the isotope removal V_{cr} depends on the trapping voltage V_{ac}. The results are $V_{cr} = 15$ mV, 5 mV, and 25 mV for $V_{ac} = 86$ V, 107 V, and 129 V, respectively. It is found that the critical amplitude is minimum when $V_{ac} = 107$ V in the present experiment condition. This result is qualitatively explained as following. When the trapping potential is deep ($V_{ac} = 129$ V), the motion of the ions is small and the irrelevant ions tend to remain in the trap even if the perturbation resonates with their motion. Then the higher perturbation voltage is required to remove them. On the other hand, when the trapping potential is shallow ($V_{ac} = 86$ V), the ion temperature is low [18] and the phase of the ion motion is more correlative than in the

case of higher temperature. Then the ions tend to move collectively and a large amplitude is required to break the phase correlation.

Lastly, we mention about the applicability of this method to the heavier ions such as Hg^+ (200.6 atomic mass units (amu)) and Yb^+ (173.0 amu). In the present setup, the resolution of the secular frequency (spectral line width) in Fig. 2a is $\kappa = 0.3$ kHz. It is expected that the resonant motion of each isotope can be excited separately if the difference between the secular frequencies of the isotopes of the heavier ion species is over $\kappa = 0.3$ kHz. For example, the difference between the secular frequencies of $^{200}Hg^+$ and $^{201}Hg^+$ is over 0.3 kHz in the case of $V_{ac} = 300$ V, which satisfies the condition for stable trapping [12].

4 Conclusion

We have demonstrated the isotope removal of Mg^+ which is based on the additional rf excitation of the secular motion of trapped ions. Laser cooling carried out simultaneously suppresses the loss of relevant ions. As a result, complete removal of irrelevant ions and retention of a large number of the relevant ion are achieved. The obtained cloud of pure isotope ions becomes dense and of low temperature because the rf heating is reduced. The principle of this method is applicable not only to the isotope removal of Mg^+, but also to the removal of the heavier ions, such as Yb^+ and Hg^+, though the values of the trapping parameters may be different from those in the case of Mg^+. This method is also applicable to the removal of impurities, such as CO^+ and MgC^+, which

are likely generated from the residual gas by the electron bombardment.

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