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Isotope selective loading of an ion trap using resonance-enhanced two-photon ionization

N. Kjærgaard*, L. Hornekær, A.M. Thommesen, Z. Videsen, M. Drewsen

Institute of Physics and Astronomy, Aarhus University, 8000 Aarhus C, Denmark
(E-mail: nielskj@ifa.au.dk)

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Abstract. We have demonstrated that resonance-enhanced two-photon ionization of atomic beams provides an effective tool for isotope selective loading of ions into a linear Paul trap. Using a tunable, narrow-bandwidth, continuous wave (cw) laser system for the ionization process, we have succeeded in producing Mg^+ and Ca^+ ions at rates controlled by the atomic beam flux, the laser intensity, and the laser frequency detuning from resonance. We have observed that with a proper choice of control parameters, it is rather easy to load a specific number of ions into a string. This observation has direct applications in quantum optics and quantum computation experiments. Furthermore, resonant photo-ionization loading facilitates the formation of large isotope-pure Coulomb crystals.

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The field of laser cooling of ions in electromagnetic traps ranges from the study of large Coulomb crystals [1–4] to strings containing only a few ions [5–8]. A common method to create the ions of interest has so far been to bombard a neutral atomic beam with electrons. If isotopic pure samples are needed, this method can cause problems, however, since the energetic electrons can ionize atoms from the residual gas in the vacuum chamber, as well as unwanted isotopes in the atomic beam. Moreover, several charge states can often be formed. Altogether, this leads to impure Coulomb crystals which have to be subjected to purification by altering the trap parameters. Since purification typically leads to a substantial loss of the desired ions, a direct and controlled technique of producing large and isotope-pure crystals is of considerable interest. In quantum optics experiments (for example, quantum computation experiments), the creation of a string containing a specific number of ions is often of special interest [5–10]. For such experiments a loading technique based on controllable parameters would be superior to the purification method with its trial and error principle.

In this paper, we propose to use resonance-enhanced two-photon ionization instead of electron bombardment in order to overcome the problems stated above. Resonant multi-step photo-ionization has already been exploited with success in ion trap mass spectrometry [11–14]. Here, pulsed lasers are applied to obtain a high selectivity of analyte ions, but due to the large laser bandwidths and power broadening of the atomic energy levels isotopic selectivity is usually not achieved. Detection of hydrogen isotope atoms was, however, recently reported [14]. In the experiments presented below, we use a narrow bandwidth low power cw-laser system to resonantly ionize Mg atoms via the $3s^2\,{}^1S_0 \leftrightarrow 3s\,3p\,{}^1P_1$ transition at 285 nm and Ca atoms via the $4s^2\,{}^1S_0 \leftrightarrow 4s\,5p\,{}^1P_1$ transition at 272 nm. During trapping in a linear Paul trap [15], the ions are laser cooled and form Coulomb crystals. Using the fluorescence from the ions induced by the cooling laser for imaging the crystals, we demonstrate that this method of loading the trap can facilitate isotopic selectivity, as well as providing an easy way of loading a specific number of ions into a string.

1 Laser systems and experimental setup

For the ionization of both Ca and Mg atoms a laser system consisting of a single-mode, tunable, ring cw dye laser optically pumped by an Ar^+ laser is applied. The dye used was Pyrromethene 556 with a peak power operating wavelength of 553 nm; the laser output was frequency doubled in a 5-mm-long BBO crystal placed in an external bow-tie cavity. This frequency-doubling unit was designed to operate at 560 nm, but is sufficiently versatile to function at the wavelengths needed for the ionization of both Mg and Ca atoms, i.e., 570 nm and 544 nm, respectively.

Figure 1 shows the schematic level schemes of ^{24}Mg, $^{24}Mg^+$, ^{40}Ca, and $^{40}Ca^+$ together with the transitions used in the photo-ionization and laser cooling processes in the experiments. In the case of Mg, the two-photon ionization proceeds from the ground state via the $3s\,3p\,{}^1P_1$ state. From the $3s\,3p\,{}^1P_1$ state the atoms can be ionized either by a second photon at 285 nm or by one of the cooling laser photons at 280 nm (see Fig. 1a,b). The cooling transition $3s\,{}^2S_{1/2} \leftrightarrow 3p\,{}^2P_{3/2}$ of $^{24}Mg^+$ is driven by a laser system almost identical to the one used for the ionization. Similarly, ^{40}Ca can be resonantly ionized through excita-

*Corresponding author.

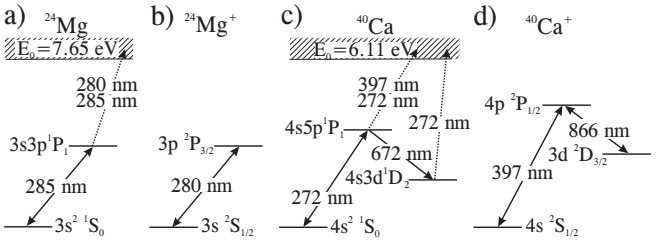

Fig. 1a–d. Schematic level schemes for **a** ^{24}Mg, **b** ^{24}Mg$^+$, **c** ^{40}Ca, and **d** ^{40}Ca$^+$ identifying the transitions involved for laser ionization and cooling

tion to the $4s\,5p\,^1P_1$ state. The atoms can be ionized by another photon at 272 nm either directly from the $4s\,5p\,^1P_1$ state, or from the metastable $4s\,3d\,^1D_2$ state which is populated through spontaneous decay of the $4s5p\,^1P_1$ state (see Fig. 1c,d). As in the case of Mg, the cooling laser light driving the $4s\,^2S_{1/2} \leftrightarrow 4p\,^2P_{1/2}$ transition at 397 nm for cooling the ^{40}Ca$^+$ ions, can also lead to enhanced photo-ionization, but only from the $4s\,5p\,^1P_1$ state in ^{40}Ca. To cool the ^{40}Ca$^+$ ions, the $4s\,^2S_{1/2} \leftrightarrow 4p\,^2P_{1/2}$ transition at 397 nm is driven by frequency-doubled cw light from a Ti:sapphire laser pumped by an Ar$^+$ laser. The doubling process is achieved in an external bow-tie cavity by passage of the fundamental wavelength laser beam through a 12-mm-long LBO crystal. To avoid optical pumping into the $3d\,^2D_{3/2}$ state of ^{40}Ca$^+$ during laser cooling, a diode laser resonant with the $4p\,^2P_{1/2} \leftrightarrow 3d\,^2D_{3/2}$ transition at 866 nm is applied.

The experimental arrangements used in the case of Mg and Ca are shown in Figs. 2 and 3, respectively. In both cases, the ionizing laser beam is directed along the axis of a linear Paul trap to interact with the atomic beams at the trap center. The atomic beams effuse from ovens and are collimated to a full divergence angle of 30 mrad. The beams

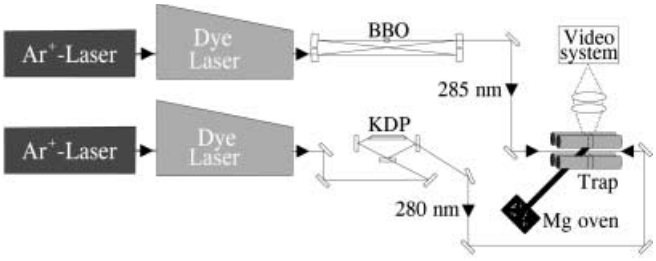

Fig. 2. Schematic experimental setup for photo-ionization of Mg and laser cooling of ^{24}Mg$^+$

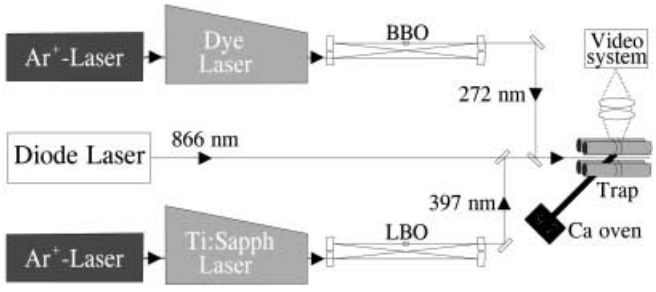

Fig. 3. Schematic experimental setup for photo-ionization of Ca and laser cooling of ^{40}Ca$^+$

emerge at an angle of 45° to the trap axis. The cooling laser beams enter co-linearly with the ionization laser (counter-propagating for Mg and co-propagating for Ca). The linear Paul trap used consists of four parallel cylindrical electrodes with an applied RF field as in the quadrupole mass filter. To obtain axial confinement, each electrode is longitudinally sectioned into three pieces, such that a positive dc voltage can be added to the eight end-pieces [16]. The middle section is 4 mm long and the outer parts 25 mm each. The diameter of the electrodes is 4 mm, and the minimum distance to the trap center axis is chosen to be 1.75 mm to obtain the best approximation to a perfect quadrupole field [17]. A RF frequency $\Omega = 2\pi \times 5.1$ MHz and an amplitude $U_{\mathrm{RF}} = 30$–80 V are typically used. Laser-induced fluorescence of the trapped, cooled ions can be recorded by an image intensified digital video system monitoring the trapped ions in a direction perpendicular to the trap axis.

2 Results and discussion

Figure 4a shows pictures of ^{24}Mg$^+$ Coulomb crystals loaded into the trap at different relative detunings of the ionization laser. The ionization laser having a total power of about 2 mW was focused to a diameter of about 0.3 mm and was present for 20 s. During loading, the cooling laser was present and detuned a few atomic line widths below resonance of the cooling transition, ensuring good capturing of the ^{24}Mg$^+$ isotope. Moreover, the UV cooling laser light serves to enhance the rate of the final step in the ionization process (see Fig. 1a). After loading, the laser-induced fluorescence from the cooling laser was used to image the trapped ^{24}Mg$^+$ ions. We notice a maximum in production of ^{24}Mg$^+$ ions (≈ 3500 ions detected) at the ionization laser detuning denoted by 0 GHz in Fig. 4a. The number of ^{24}Mg$^+$ ions decreases when the ionization laser is detuned away from resonance. For positive detunings, there is, however, an onset in the trapping production of the isotopes ^{25}Mg$^+$ and ^{26}Mg$^+$. This is seen to introduce a sharp, linear boundary at the top and bottom of the image projections of the crystals originating from heavier elements lying in shells outside the ^{24}Mg$^+$ crystals. The isotopes ^{25}Mg and ^{26}Mg have natural isotopic abundances of 10% and 11%, respectively, and the isotope shift for the transition $3s^2\,^1S_0 \leftrightarrow 3s\,3p\,^1P_1$ used for ionization of these two isotopes is 0.73 GHz and 1.41 GHz [18], respectively. The velocity spread of the atoms in the direction of the ionizing laser beam gives rise to a Doppler broadening of the resonances of about 800 MHz FWHM. This is much larger than the 80-MHz natural line width of the $3s\,3p\,^1P_1$ state as well as the hyperfine splitting in the case of ^{25}Mg$^+$ [19]. Figure 4b shows the result of a loading with a relative ionization laser detuning of 1.4 GHz and a loading time of 50 s. The shape of the crystal indicates that many heavier ions are trapped along with ^{24}Mg$^+$. By applying the cooling laser at the wavelength of the cooling transition of ^{26}Mg$^+$ during loading while the ionization laser detuning is kept at 1.4 GHz, we see (Fig. 4c) that ^{26}Mg$^+$ ions primarily account for these heavier ions, and that an almost pure crystal of this isotope is formed. We note a minor fraction of lighter isotopes at the left end. The reason for this asymmetry is the laser cooling force pushing the ^{26}Mg$^+$ ions towards the right in the trap potential. The asymmetry in

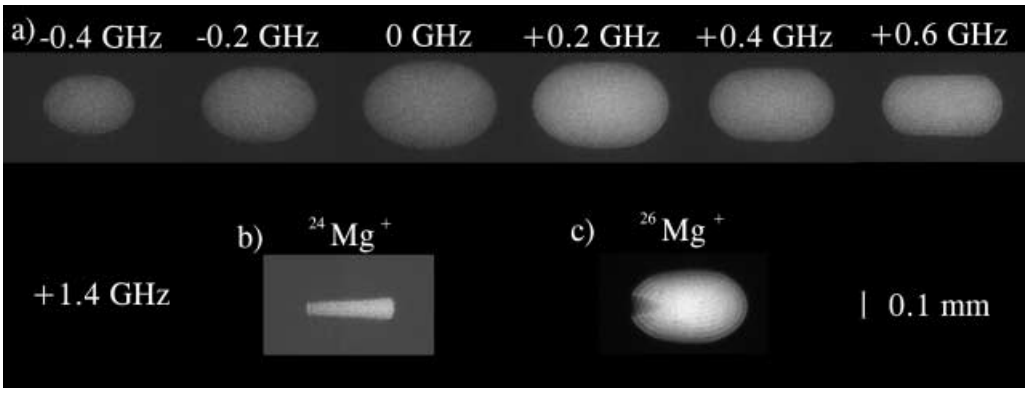

Fig. 4. a Production of $^{24}Mg^+$ for different relative ionization laser detunings with a trap loading time of 20 s. **b** Production of $^{24}Mg^+$ for a relative ionization laser detuning of 1.4 GHz with a trap loading time of 50 s. **c** Loading with the cooling laser close to resonance of $^{26}Mg^+$ for a relative ionization laser detuning of +1.4 GHz resulting in an almost pure $^{26}Mg^+$ crystal

Fig. 4b is due to the same effect, but this time acting on the $^{24}Mg^+$ ions.

Using the setup for Ca depicted in Fig. 3, we prove that the photo-ionization method outlined here is not restricted to Mg. In Fig. 5, a large, pure $^{40}Ca^+$ Coulomb crystal has been produced using the scheme described in Sect. 1. The crystal consists of about 5000 ions. Due to its atomic level structure, $^{40}Ca^+$ is of particular interest in quantum optics and quantum computation experiments (see, for example, [8]). Here, a specific number of a few ions has to be loaded into the trap and hence control of this regime is of considerable importance. This type of loading is readily accomplished with photo-ionization by using the laser detuning, the laser intensity and atomic beam flux as control parameters to keep the production rate low: When a specified number of trapped ions is observed by the camera, the ionization laser is simply blocked. For cavity quantum electrodynamics (QED) experiments involving ions, the photo-ionization technique will also facilitate loading of ions between the closely spaced mirrors without charging up the nearby surroundings as it is likely to be the case for electron bombardment. Figure 6a shows a string of 21 $^{40}Ca^+$ ions produced in this way. As a further proof of principle, we supply pictures of strings consisting of 1–12 $^{24}Mg^+$ ions in Fig. 6b.

The ultimate situation for isotope selective loading of an ion trap by two-photon resonant ionization is achieved when the Doppler broadening is less than the natural line width of the resonant level. In this case, which can be obtained in a setup with a well collimated, atomic beam crossed at a 90° angle with the resonant ionizing laser beam, the impurity levels can often be $\lesssim 1$ %, for example, in the case of Mg. Hence, it should be feasible to preferably load the isotope $^{25}Mg^+$, which is of special interest in quantum optics experiments due to its hyperfine-splitted ground state. Atomic elements such as Sr, Ba, and In, which are often used in ion trap experiments, can similarly to Mg and Ca be ionized by a resonant two-photon process with near UV photons. For these three elements, very pure loading of at least some of the naturally occurring isotopes should be possible. In the case of Sr, this can be achieved by applying light resonant with, for example, the $5s^2\,{}^1S_0 \leftrightarrow 5s\,6p\,{}^1P_1$ transition at 293 nm. For Ba, the $6s^2\,{}^1S_0 \leftrightarrow 5d\,6p\,{}^1P_1$, $6s\,7p\,{}^1P_1$ transitions at 350 nm and 307 nm, respectively, can be used. Finally, for In, resonant ionization can be obtained through the $5p\,{}^2P_{1/2} \leftrightarrow 6s\,{}^2S_{1/2}$ transition at 410 nm. With an isotope shift of this transition of -258 MHz [20] for $^{113}In^+$ with respect to $^{115}In^+$ and a line width of 22 MHz [21], it should be possible to reduce the relative occurrence of $^{113}In^+$ to below 10^{-3}. In connection with recently proposed ion quantum computers based on $^{25}Mg^+$ ions sympathetically cooled by $^{115}In^+$ ions [22], resonance-enhanced two-photon ionization seems to be a very attractive tool.

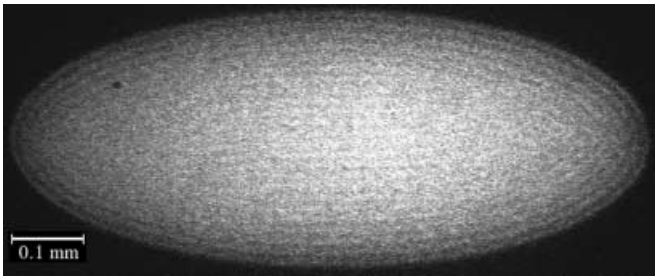

Fig. 5. Large $^{40}Ca^+$ crystal produced by resonant photo-ionization

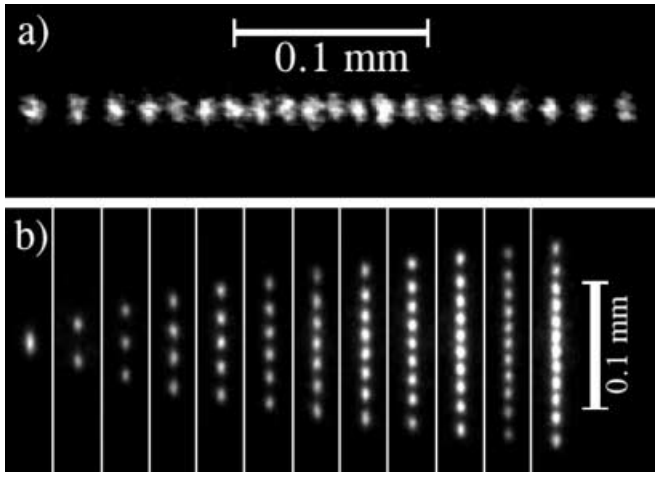

Fig. 6. a Laser-cooled $^{40}Ca^+$ string consisting of 21 ions produced by resonant photo-ionization. **b** Laser-cooled $^{24}Mg^+$ ions produced by resonant photo-ionization; strings with 1–12 ions are shown

3 Conclusion

We have demonstrated that resonant photo-ionization of an atomic beam provides a controlled method of loading an ion trap. The advantages as compared to the electron bombardment method are several-fold. Using the laser fre-

quency detuning as a control parameter, specific isotopes can be selectively loaded. Besides being a method for obtaining very large, isotope-pure Coulomb crystals, loading of isotopes with low natural abundance should also be possible. By using the laser frequency detuning, the laser intensity, and the atomic beam flux as control parameters, resonant photo-ionization can facilitate loading of a specific number of ions into a string, which can have direct applications in quantum computation and cavity QED experiments. The present experiments have shown the feasibility of resonant photo-ionization loading in the cases of Mg and Ca, but the method should easily be extended to other elements.

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