Laser ablation and two-step photo-ionization for the generation of $^{40}$Ca$^+$

H Shao$^{1,2,3}$, M Wang$^{1,2,3}$, M Zeng$^{1,2,3}$, H Guan$^{1,2,4}$ and K Gao$^{1,2}$

1. Introduction

Trapped and laser-cooled ions are proven to be ideal candidates for optical frequency standards [1, 2] and quantum information processing [3, 4], due to their environmental isolation and long interrogation time. There are similar applications for the measurement of lifetimes and lifetime ratio [5–9], branching fractions [10, 11], and transition matrix elements [12, 13]. As detrimental destruction to vacuum conditions is inappreciable through accurate control by laser ablation, an ideal vacuum condition is beneficial to the experimental accuracy for limiting the effects of collisions and mixing [14, 15]. In the method of electron impact ionization with a resistively heated oven, dark ions are often produced, which affecting vacuum pressure, causing collisions between the ultra-cold ions and background gases, results in trapped ion depopulation [8]; reactions that generate molecule-ions [16], leading to the loss of individual ions. The situation is similar to the pulse laser ablation (PLA) at high intensity, without requiring other lasers for photo-ionization, ablation pulses can directly create ions. Thus some multiple dark ions can be loaded into the trap simultaneously, and undergo sympathetic cooling by calcium ions. The heating effect of dark ions not only affects the cooling efficiency during ion loading. The efficiency of the target ions, leading to the loss of individual ions. The situation is similar to the pulse laser ablation (PLA) at high intensity, without requiring other lasers for photo-ionization, ablation pulses can directly create ions. Thus some multiple dark ions can be loaded into the trap simultaneously, and undergo sympathetic cooling by calcium ions. The heating effect of dark ions not only affects the cooling efficiency during ion loading, but also further affects measurements taken with atomic precision spectroscopy. The laser ablation method, combined with use of ionization lasers, gives rise to accurate control and high efficiency during ion loading. The similar method of $^{40}$Ca$^+$ ion loading will benefit the study of the characteristics of many kinds of trapped ions such as $^{37}$Al$^+$, $^{86}$Sr$^+$, $^{138}$Ba$^+$, $^{159}$Hg$^+$, etc.

To our knowledge, up until now the methods of electron impact ionization [17–19] and photo-ionization [20–31] have been the universal methods for producing ions in an ion trap. The traditional method of electron impact ionization is an effective tool that was first used for the generation of ions decades ago. The disadvantages of this method are the lack of isotope selectivity, the lack of control over the number of loading ions, and that the
residual gas in the vacuum can also be ionized. The fluxed sample also deposits onto the trap electrodes during the loading process, which causes patch potentials and increases the heating rate of trapped ions from the motional ground state when compared with the case of clean electrode surfaces [17–19]. Later, in order to overcome the shortcomings of electron impact ionization, the method of using a resistively heated oven with photo-ionization was universally adopted. Using photo-ionization to load a single calcium ion into a Paul trap was first demonstrated by Kjærgaard et al [21], who adopted the \( 4s^2 \, ^1S_0 \rightarrow 4s5p \, ^1P_1 \) transition at a wavelength of 272 nm for excitation far above the ionization limit. Later, Gulde et al [22] and Schuck et al [23] demonstrated another scheme with the \( 4s^2 \, ^1S_0 \rightarrow 4s4p \, ^1P_1 \) transition at 423 nm followed by excitation close to the continuum by ultraviolet radiation (from an LED) near 390 nm. Following the success of this transition, Lucas et al [24] and Tanaka et al [25, 26] performed \(^{40}\text{Ca}^+\) loading as well as the rare isotopes of \(^{44}\text{Ca}^+, \, ^{46}\text{Ca}^+\) and \(^{48}\text{Ca}^+\). Recently, a different scheme demonstrated by Zhang et al [27] used a 423 nm-wavelength laser combined with another of 732 nm for the realization of \(^{40}\text{Ca}^+\) loading. By adjusting the temperature of the atomic oven and using the appropriate-wavelength ionizing lasers, this method allowed isotopic selectivity and avoided ionization of the residual gases, but the problems of electrode contamination and uncontrollable loading rate persisted. Recently, with the development of laser techniques, PLA as a simple and versatile experimental method with efficient, clean and controllable advantages has been adopted. Due to experimental requirements, Zimmermann et al [28] adopted this scheme for the direct generation of \( \text{Th}^+ \) ions, and Leibrandt et al [29] for \(^{88}\text{Sr}^+\) ions, during the ablation process. Hendricks et al [30] adopted this scheme for generating calcium ions with an ablation beam at a wavelength of 1064 nm, and a photo-ionization laser at a wavelength of 272 nm, in a linear Paul trap. In that paper, the ablation depth was studied as a function of pulse energy, and the influence of vacuum pressure was also monitored. The relationship between properties of Rydberg atoms and laser power was also investigated. Later, Sheridan et al [31] performed a detailed investigation of the difference between this all-optical ion generation technique and the resistively heated oven method.

In this paper, in order to accurately load a single \(^{40}\text{Ca}^+\) ion with maximum probability and avoid the dark ion problem, we use 532 nm-wavelength laser pulses to generate calcium atoms and a two-photon resonance-enhanced photo-ionization scheme with ultraviolet laser sources of 423 nm and 375 nm wavelengths to acquire a single ion. This method can avoid heat load during loading process, which is especially important for cryogenic systems [32–34], and increase the rate at which ions can be reloaded after they are lost. The laser ablation technique can also govern the shape of the heated area, and the ability to target small areas makes it possible to avoid large pressure rises during loading and obtain a faster recovery time for returning to the initial level of pressure [30]. Ion-trapping experiments typically aim to load either a single ion or small numbers of ions, so a large continuous flux of atoms passing through the trap structure is undesirable. Therefore, the aforementioned advantages will provide important benefits for optical frequency standards. The effects of intensity and frequency of the three lasers are studied carefully, with results showing that the short loading time makes it easier to produce a single ion within a few seconds. With optimal parameters, only one ion is generated in the trap in each loading process in a controllable and clean manner, which can successfully reduce electrode contamination and avoids the issue of dark ions affecting atomic precision spectroscopy. The method could potentially be used to generate ions in miniature ion traps, especially chip-scale traps [35, 36], with only tens of microns between the trapped ions and the electrode surface.

The ablation and photo-ionization schematic is shown in figure 1. First, calcium atoms are generated by the laser pulses when the energy density exposure on the surface of calcium target exceeds the threshold value of relaxation processes such as thermal conduction, which requires the laser pulse duration to be smaller than the relaxation time (figure 1(a)). The dipole transition \( 4s4s \, ^1S_0 \leftrightarrow 4s4p \, ^1P_1 \) of neutral calcium is then excited by a laser operating at a wavelength of 423 nm, and followed by excitation to the continuum by a laser at a wavelength of 375 nm (figure 1(b)). Lastly, the newly-ionized ion is trapped in a linear trap and Doppler cooled by 397 nm-wavelength and 866 nm-wavelength lasers (figure 1(c)).

2. Experimental apparatus

In our experiment, three lasers are used for loading the ion into the trap. The experimental apparatus is shown in figure 2. First, a 532 nm-wavelength ablation laser is used for producing the atoms, this wavelength was chosen due to the requirements of nanosecond pulse duration and high pulse energy for ablation experiments. The laser has a pulse width of approximately 20 ns at full-width half-maximum (FWHM), and an effective output power of 128 mW under a 1 kHz repetition rate. Short duration pulses allow the energy density in the region to exceed the ablation limit and facilitate the removal of material from the target. For conveniently aligning the laser beams, 532 nm was selected as the visible wavelength, also this wavelength will be more strongly absorbed by metals than infrared radiation. The three lenses in figure 2 are used to keep the beam waist on the calcium target below 30 \( \mu \text{m} \). A signal generator combined with an acousto-optical-modulator (AOM) controls the ablation
laser power and exposure time of the calcium surface while randomly changing the ablation region by changing its frequency in the range of 180–220 MHz. The 423 nm-wavelength photo-ionization laser is used for driving the dipole transition from the ground 4s4s1P1 state to the intermediate 4s4p1P1 state, while another laser with a wavelength of 375 nm is used for driving the transition from the 4s4p1P1 state to the continuum. The two photo-ionization lasers are coupled to one fiber and controlled by a shutter, with a beam waist of ∼100 μm in the trap center. By this process, the ion is generated. The ion trap is mounted in a vacuum chamber with a vacuum pressure of approximately 2.0 × 10⁻⁹ Pa to maintain a stable environment for long-lifetime trapping. An ideal vacuum environment is not only beneficial for reducing the effect of collisions between trapped ions and background gases, but also for preventing the loss of ions through reactions with those gases that generate new molecule-ions. The chambers with relevant attachments are constituted of titanium metal, a material that possesses nonmagnetic characteristics, which is beneficial for magnetic control. Once the ion is successfully loaded, the 397 nm-wavelength laser with a power of 35 μW and beam waist of 40 μm and 866 nm-wavelength laser with a power of 47 μW and beam waist of 86 μm are used to cool the newly loaded ions to the Lamb-Dicke regime. The powers of 397 and 866 nm under their beam waist are found to be the optimum values for a single ion in our trap. As the lifetime of the 4p1P₁/2 state in the ⁴⁰Ca⁺ ion is approximately 7 ns [13], it is very suitable for Doppler cooling at 397 nm. The laser beam is red-detuned by 100 MHz from the resonant frequency of the dipole transition 4 s²S₁/₂ ↔ 4 p²P₁/₂. As there is a probability that the ion could decay from the 4p²P₁/₂ state to...
the 3d\(^2\)D\(_{3/2}\) state\(^{[10]}\) resulting in the cooling being interrupted, an 866 nm-wavelength laser is applied to depopulate the 3d\(^2\)D\(_{3/2}\) state. The 866 nm-wavelength laser frequency is locked in the position of the 4p\(^2\)P\(_{1/2}\) - 3d\(^2\)D\(_{3/2}\) resonance. The cooling and detection lasers at 397 nm and 866 nm are frequency-locked to a high-finesse ULE (ultra-low-expansion) cavity using the Pound-Drever-Hall scheme, with a typical linewidth (FWHM) of ~100 kHz. The power of the 423 nm and 375 nm lasers can be adjusted from 0 to 3.5 mW and 1 mW by using separate attenuators, while the 532 nm-wavelength laser can be adjusted from 0 to 128 mW. The fluorescence of the 4p\(^2\)P\(_{1/2}\) → 4s\(^2\)S\(_{1/2}\) dipole transition is recorded at 397 nm. The signal detection is carried out by combining an Electron-Multiplying Charge-coupled Device (EMCCD) and a photomultiplier tube (PMT) with the 397 nm and 866 nm lasers switched on. A narrow-band-pass filter and a pinhole with a diameter of 50 μm are placed in front of the PMT and EMCCD to block the background scattering light. All vacuum windows are coated with antireflection film to ensure high transmittance while reducing background light which may affect signal detection. A computer recorded all experimental data and simultaneously controlled the photon counter, AOMs, and shutters via a PIC-6733 DAQ card with microsecond accuracy.

### 3. Ion loading scheme

A simplified sequence for the measurement of loading a single \(^{40}\)Ca\(^{+}\) ion is shown in figure 3. This sequence consists of three major steps for a complete round of measurement. In the first step \((t_1)\), the 397 nm and 866 nm lasers are employed to measure the background level as the state with no ions. After confirming the background level, the ion loading process is performed in the second step in a process where all five lasers are switched on simultaneously for an interval. The atoms are generated by 532 nm-wavelength ablation pulses to the calcium metal surface while photo-ionization is performed with 423 nm and 375 nm ultraviolet lasers in the center of the trap. If the ion is successfully produced, the detected signal will appear greater than background level. In this instance, the computer sends a command to the AOM and shutter in the 523 nm, 423 nm, and 375 nm light routes simultaneously to switch those lasers off. The loading time is denoted as \(t\). The delay time between ion production and ionization lasers switch off is about 200 ms. In order to distinguish the ion level from the background level, an optimal threshold value is needed. In the last step \((t_2)\), the 397 nm and 866 nm lasers are combined with the photon counter for the recording of the ion’s fluorescence. When the 866 nm-wavelength laser is blocked, the fluorescence drops to the background level instantly, demonstrating that the ions have been generated. The ion is then released and the cycle is repeated until six of the same processes are completed. In figure 3, the high steps represent the time when the lasers or counter are switched on, and the low steps represent the off-state.

Before carrying out the study, the PLA is first performed in an auxiliary system where a mass spectrometer is utilized. The beam waist and power are consistent with the system previously described, except that the vacuum
Pressure is inferior, in the order of $10^{-6}$ Pa. The red and blue lines in figure 4 show mass scans with the atomic beam production by PLA turned off and on, respectively. From figure 4 we can see that a peak appears at a mass of 40 atomic units relative to the background level when the ablation laser of 532 nm is incident on the calcium target surface, which demonstrates that the calcium atoms can be generated through this form of PLA.

Figure 5 shows the single trapped $^{40}$Ca$^+$ ions generated by 532 nm-wavelength laser ablation and two-step photo-ionization of combined 375 nm and 423 nm lasers. The photon counts and EMCCD signals of the loaded ions are shown in the cases from one to four loaded ions. The photon counts represent the numbers recorded in 200 ms recording intervals. The photon counts of a single ion varied with a 397 nm detuning frequency is shown in the top left corner, in which the photon counts represent the numbers recorded in 0.15 ms recording intervals.

Figure 4. Pulse laser ablation (PLA) for the generation of calcium atoms. The measurement was performed in an auxiliary system with a vacuum pressure of $1.0 \times 10^{-6}$ Pa. Gas compositions were monitored by a mass spectrometer. The red line represents the compositions of background gases while the blue line is the atomic signal. The result shows that an obvious peak signal at a mass of 40 atomic units appeared when the PLA was applied to the calcium target compared to the background level.

Figure 5. Single trapped $^{40}$Ca$^+$ ions generated by 532 nm-wavelength laser ablation and two-step photo-ionization of combined 375 nm and 423 nm lasers. The photon counts and EMCCD signals of the loaded ions are shown in the cases from one to four loaded ions. The photon counts represent the numbers recorded in 200 ms recording intervals. The photon counts of a single ion varied with a 397 nm detuning frequency is shown in the top left corner, in which the photon counts represent the numbers recorded in 0.15 ms recording intervals.
4. Loading time measurements

A single calcium ion can be successfully loaded by PLA and the two-step photoionization scheme, but which is affected by many factors such as laser frequency and power. Maintaining a short loading time and avoiding dark ion generation are the key reasons to be considered in optical frequency standard. In this section, the loading time is measured as a function of the power and detuning frequency of the 532 nm, 423 nm and 375 nm lasers.

In the first experiment, the loading time as a function of the 532 nm-wavelength pulse laser power is studied. We used a model of power function to display the tendency of loading time with 532 nm laser power, the results are shown in figure 6. Each point represents the average loading time of six independent experiments, while the error bars in the figure indicate the standard deviation of the mean. From the red data points of figure 6 we can see that the loading time is improved from 10 s to 2 s when the power is varied from 50 mW to 88 mW. However, when the laser power is set below 50 mW, we cannot directly detect ion generation by the 532 nm-wavelength laser alone. These results demonstrate that an ion can be directly produced by the ablation laser at high intensity.

The loading time is verified to be further improved by experimentally introducing the 423 nm and 375 nm lasers. Here, the 423 nm and 375 nm lasers are operated at a power of 3 mW and 1 mW, respectively, with a waist of 100 μm, and the 423 nm-wavelength laser frequency is 709.077 80 THz. As shown in the blue data points of figure 6, there is a significant decrease in loading time for each successive point with the inclusion of the photoionization laser beams, especially when the 532 nm-wavelength laser power is low. It is important to note that the enhancement in this case is not obvious when the 532 nm-wavelength laser power is greater than 30 mW. Therefore, short loading time can be reached with high ablation and photoionization laser power.

In the second experiment, the loading time as a function of the 423 nm-wavelength laser detuning frequency is studied. The results are shown in figure 7. Again, each point represents the average loading time of six independent experiments, and the error bars in the figure indicate the standard deviation of the mean. From figure 7, we can see that the loading time is decreased from 10 s to 2 s with the detuning frequency set close to the resonant frequency of the 4s4s → 4s4p dipole transition. Fitting a Gaussian to the loading times measured as a function of the laser frequency yields a value of 709.077 74 THz for the centroid of this transition.

Aside from the frequency of the 423 nm-wavelength laser, the loading time may also be affected by the laser power. In the third experiment, the loading time as a function of the 423 nm and 375 nm laser powers are investigated independently. The ablation laser with a wavelength of 532 nm is set to a power of 30 mW in 1 kHz pulses as only atoms are created at this power without the direct generation of ions, allowing the change in loading time with the power of the 423 nm and 375 nm lasers to be observed. Figure 8 shows the results of plotting the loading time as a function of the power of the photo-ionization lasers. We used a model of power function to display the tendency of loading time with 423 nm and 375 nm lasers’ power. The blue points represent the loading time with varying 423 nm-wavelength laser power, performed with a power of 1.0 mW for the 375 nm-wavelength laser, while the red points represent the loading time with varying 375 nm-wavelength laser power, with a power of 3.0 mW for the 423 nm-wavelength laser. The results show that at lower PLA intensities the loading times are improved with increasing powers of both the 423 nm and 375 nm lasers, but as the powers increase further the effect saturates. When the power of the 375 nm and 423 nm lasers reach 1.0 mW...
and 2.0 mW, respectively, the mean loading time can stably reach 4 s, but does not appear to improve further with increasing laser power. From experimental results, we conclude that powers of 30 mW at 523 nm, 1 mW at 375 nm, and 2 mW at 423 nm are the optimal parameters for producing a single calcium ion when combined with the 423 nm-wavelength laser frequency of 709.0774 THz. With these parameters selected, we find that the single ion can be loaded with a short loading time of approximately 4 s in each loading process. The single ion loading probability was briefly investigated by 30 independent experiments with the appropriate parameters and detected the single ion loading of 24. Moreover, with these parameters, we can successfully avoid generated dark ions affecting atomic precision spectroscopy measurements.

5. Conclusion

In this paper, we present a scheme that utilizes pulse ablation and two ultraviolet lasers for the photo-ionization of calcium in a miniature linear Paul trap, and report the appropriate and critical parameters for accurately and effectively loading a single $^{40}$Ca$^+$ ion in optical frequency standard. The atoms are generated by the method of
PLA, which is characteristically efficient and clean, thereby greatly reducing the pollution produced. Compared to electron impact ionization with a resistively heated oven, the main advantage of laser ablation is that it can be precisely controlled in order to restrict the amount of fluxed calcium depositing onto the trap electrodes. The decreased loading time is also desirable. Typically, a single ion is loaded in less than 10 s. More importantly, by this method, a single ion is generated in the trap with high probability, so we can successfully avoid the dark ions which can affect the atomic precision spectroscopy measurements. Furthermore, the laser ablation technique has the potential to substantially reduce the heat load associated with loading ions, which is especially important for cryogenic systems, and increase the rate at which ions can be reloaded after they are lost. The laser ablation technique can also govern the shape of the heated area, and the potential for targeting small-sized areas makes it possible to avoid large pressure rises during loading and obtain much faster recovery times for returning to initial pressure levels. Laser ablation technique can be applied for the generation of a number of atoms and ions conducted in ultra-high vacuum condition, which brings extensive benefits to optical frequency standards, quantum simulation and quantum information processing.

Acknowledgments

This work is financially supported by The National Key Research and Development Program of China (Grants No. 2017YFA0304401), the National Science Foundation of China (Grants No. 91336211, No. 11034009, No. 11474318, and No. 11622434), the Strategic Priority Research Program of the Chinese Academy of Sciences (Grant No. XDB21030100) and Youth Innovation Promotion Association CAS (2015274).

ORCID iDs

H Shao https://orcid.org/0000-0002-8739-4457

References

[1] Huang Y, Guan H, Liu P, Bian W, Ma L, Liang K, Li T and Gao K 2016 Phys. Rev. Lett. 116 013001
[2] Huang Y, Guan H, Bian W, Ma L, Liang K, Li T and Gao K 2017 Appl. Phys. B 123 166
[3] Roos C F, Chwalla M, Kim K, Riebe M and Blatt R 2006 Nature 443 316
[4] Benhelm J, Kirkmair G, Roos C F and Blatt R 2008 Nat. Phys. 4 463
[5] Guan H, Shao H, Qian Y, Huang Y, Liu P-L, Bian W, Li C-B, Sahoo B K and Gao K-L 2015 Phys. Rev. A 91 022511
[6] Shao H, Huang Y, Guan H, Qian Y and Gao K 2016 Phys. Rev. A 94 042507
[7] Kreuter A et al 2005 Phys. Rev. A 71 032504
[8] Barton P A, Donald C J S, Lucas D M, Stevens D A, Steane A M and Stacey D N 2000 Phys. Rev. A 62 032503
[9] Shao H, Huang Y, Guan Hand Gao K 2018 J. Phys. B: At. Mol. Opt. Phys. 51 045002
[10] Ramm M, Pruettavarinar T, Kokish M, Talakdar I and Haffner H 2013 Phys. Rev. Lett. 111 023004
[11] Gerringa R, Kirkmair G, Zajc G, Benhelm J, Blatt R and Roos C F 2008 Eur. Phys. J. D 50 13
[12] Shao H, Huang Y, Guan H, Li C, C, T and Gao K 2017 Phys. Rev. A 95 053415
[13] Hettrich M, Ruster T, Kaufmann H, Roos C F, Schmiegelow C T, Schmidt-Kaler F and Poschinger U G 2015 Phys. Rev. Lett. 115 143003
[14] Knoop M, Vedel M and Vedel F 1995 Phys. Rev. A 52 3763
[15] Knoop M, Vedel M and Vedel F 1998 Phys. Rev. A 58 264
[16] Khanyile N 2015 Vibrational spectroscopy of sympathetically laser-cooled CaH+ PhD Thesis Georgia Institute of Technology
[17] Turchette Q A et al 2000 Phys. Rev. A 61 062518
[18] DeVoe R G and Kurtsiefer C 2002 Phys. Rev. A 65 063407
[19] Deslauriers L, Haljan P C, Lee P J, Brickman K A, Blinov B B, Madsen M J and Monroe C 2004 Phys. Rev. A 70 042508
[20] Dygdala R S, Karasek K, Giammanco F, Kobus J, Paj-janek-Zawadzka A, Raczywnski A, Zaremba J and Zielinski M 1998 J Phys. B: At. Mol. Opt. Phys. 31 2259
[21] Kjeriaard N, Hornekar L, Hommesen M A, Videsen Z and Drewsen M 2000 Appl. Phys. B 71 207
[22] Guile S, Ritter D, Barton P, Kaler F S, Blatt R and Hogervorst W 2001 Appl. Phys. B 73 861
[23] Schuck C, Almendros M, Rohde F, Henrich M and Eschner J 2010 Appl. Phys. B 100 765
[24] Lucas D M, Ramos A, Home J P, McDonnell M J, Nakayama S, Stacey J P, Webster S C, Stacey D N and Steane A M 2004 Phys. Rev. A 69 012711
[25] Tanaka U, Matsuushi H, Morita I and Urabe S 2005 Appl. Phys. B 81 795
[26] Tanaka U, Morita I and Urabe S 2007 Appl. Phys. B 89 195
[27] Zhang J, Xie Y, Liu P, Ou B, Wu W and Chen P 2017 Appl. Phys. B 123 145
[28] Zimmermann K, Okhapkin M V, Herrera-Sancho O A and Peik E 2012 Appl. Phys. B 107 883
[29] Leibbrandt D R, Clark R J, Labaziewicz J, Antoiti P, Balk W, Brown K R and Chuang I L 2007 Phys. Rev. A 76 035403
[30] Hendricks R J, Grant D M, Herskind P F, Dantan A and Drewsen M 2007 Appl. Phys. B 88 507
[31] Sheridan K, Lange W and Keller M 2011 Appl. Phys. B 104 775
[32] Poitzsch M E, Bergquist J C, Itano W M and Wineland D J 1996 Rev. Sci. Instrum. 67 129
[33] Schwarz M et al 2012 Rev. Sci. Instrum. 83 083115
[34] Brandl M F et al 2016 Rev. Sci. Instrum. 87 113103
[35] Wright K et al 2013 New J. Phys. 15 033004
[36] Craig D A, Linke N M, Harty T P, Ballance C J, Lucas D M, Steane A M and Alcock D T C 2013 Appl. Phys. B 114 5