Production and storage of low-energy highly charged ions by laser ablation and an ion trap

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Low-energy multiply charged ions of the refractory elements tungsten and molybdenum have been produced and stored using a novel technique that couples laser ablation with a radio-frequency ion trap. The charge states of the ions stored range from $q=+1$ to $+4$ for tungsten and from $q=+1$ to $+6$ for molybdenum. Approximately 10^5 ions with energy less than 5×10^{-3} eV/amu are stored for a period in excess of 2 s. The technique can be used to store low-energy multiply charged ions of almost any element.

In the last decade several techniques have been developed for the production of near-thermal-energy highly charged ions. Cocke[1] produced highly charged ions by the impact of swift heavy ions; this technique was later combined with ion storage to carry out experiments on low-energy charge transfer.[2,3] More recently, Short *et al.*[4] have developed a technique that uses synchrotron radiation to produce near-room-temperature highly stripped ions of the noble gases after inner-shell excitation. In these experiments, however, choices of parent gases for the ions are limited, and only a few varieties of singly and multiply charged ions can be produced. Thermal energy multiply charged ions of refractory elements such as tungsten and molybdenum, so important to our current fusion energy program,[5] have not been studied. Reported here is a novel yet simple approach that overcomes some of the limitations of earlier techniques. Useful quantities of low-energy multiply charged ions can be produced and stored for almost any element.

The method combines the production of ions by laser ablation with ion trap techniques. Laser ablation has been used previously to produce both neutral atoms and ions from solids. Measures and Kwong[6] have reported the production of neutral chromium atoms. Ehler,[7] Irons,[8] and Phaneuf[9] reported the generation of multiply charged ions from a variety of target materials. The number and charge states of ions produced by laser ablation depends on the laser energy and the laser power density. The number produced per laser shot is large, typically ranging from 10^{15} to 10^{19}. To date only singly charged ions from laser ablation have been trapped. Johnson and Kwong[10] trapped Al^+ in a cylindrical rf trap for the purpose of measurements of the lifetime of metastables. Knight[11] was able to capture Be^+, C^+, Al^+, Fe^+, and Pb^+ ions in an electrostatic trap.

The inability to trap multiply charged ions in the above-mentioned experiments most likely arises from the ions high kinetic energy (tens to hundreds of eV) relative to the traps low potential wells (a few eV). In order to trap multiply charged ions from ablation plasmas, it is therefore essential to reduce the kinetic energy of the ions. We have achieved the "cooling" of laser-ablated ions by using two separate beams that cross at right angles inside the ion trap. A few ions in one beam undergo collisions with ions from the other with the result that some of the scattered ions lose almost all their kinetic energy (and remain inside the trap) while others approximately double theirs (and leave the trap). A simple classical analogue of this cooling effect is the elastic collision of two billiard balls of identical mass M and speed v which approach one another at zero-impact parameter along exactly orthogonal trajectories, say from $-\alpha$ along the x and y axes. Using conservation of energy and momentum, it is trivial to show that after a perfectly elastic collision, one ball remains at the origin with zero velocity, and the other moves away along the line $y=x$ with a speed $\sqrt{2}$ times larger. In the case of collisions of multiply charged ions from the crossed ablation beams, only a small fraction of the number of collisions will result in ions which remain in the trap. Since there are 10^{15} or more ions in each ablation pulse, a "small fraction" is still a very large number. In practice, the number of ions that can be stored is limited to about 10^6 because of space charge effects.

The schematic of the facility is shown in Fig. 1. The timing diagram for the production, trapping, and detection of the ions is shown in Fig. 2. Two pulsed beams of ions are created by the ablation of either tungsten or molybdenum targets using the focused light from a frequency-doubled Nd:YAG laser. The ablation beams intersect each other at right angles at the center of a radio-frequency ion trap. Ions enter the trap through holes cut in the trap's ring electrode. The ablation beams are collimated by carefully positioned, grounded skimmers which minimize back scattering of ions from the expanding cloud by preventing them from striking the ring electrode or its mounting structure. A Langmuir probe, used for diagnostic purposes, is positioned outside but near the ring electrode. The contents of the trap are sampled at adjustable times after the ablation event by applying a negative-bias voltage [1-μs full width at half maximum (FWHM)] to one of the trap's end caps. Ions extracted from the trap are detected by a channel electron multiplier (CEM) which is gated on after the ablation event. Gating of the CEM is necessary to avoid damage to the detector caused by the large quantities of ions pro-

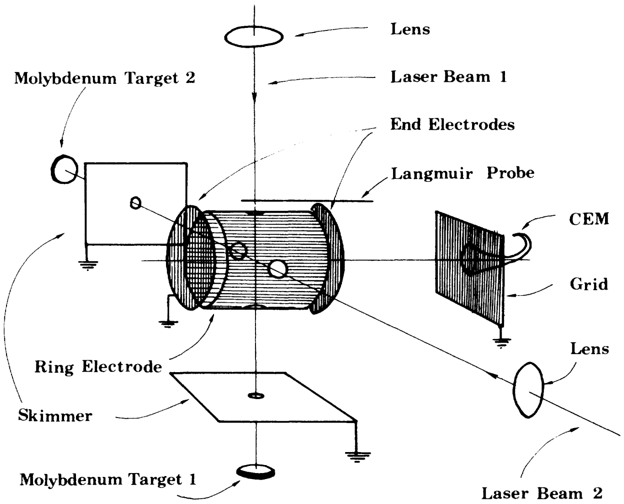

FIG. 1. Laser ablation and ion-trap apparatus.

duced during laser ablation. As shown in Fig. 3, a large signal, with very good signal to noise, is observed after both ablation beams are allowed to intersect. The size of the ion signal is comparable to the maximum ion signal obtained by electron-impact ionization of a known gas such nitrogen or argon suggesting that the trap is nearly filled. If either of the ablation-producing laser beams is blocked, no signal is observed. Measurements using the Langmuir probe indicate that the ions in each beam have a mean energy of 250 eV, a value much greater than the highest trapping potential of 42 eV used in this study. Collisions which lower the energy of a reasonable fraction of the ions are clearly taking place.

The charge-to-mass ratio of the ions captured was initially identified by operating the trap near the edges of its stability for specific ions. Figure 4 shows the theoretical stability diagram for molybdenum ions obtained from the solution of the Mathieu equation for the trajectories of

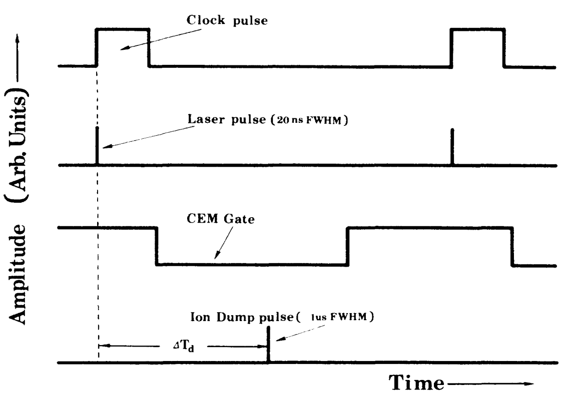

FIG. 2. Ion-trap and laser timing diagram. The channel electron multiplier (CEM) is gated off during laser ablation to avoid damage to it from the large number of laser-induced ions. The ion dump pulse is used to allow stored ions to exit the trap and move toward the CEM. The ion dump pulse is delayed relative to the ablation event ΔT_d ranging from 20 ms to 2 s.

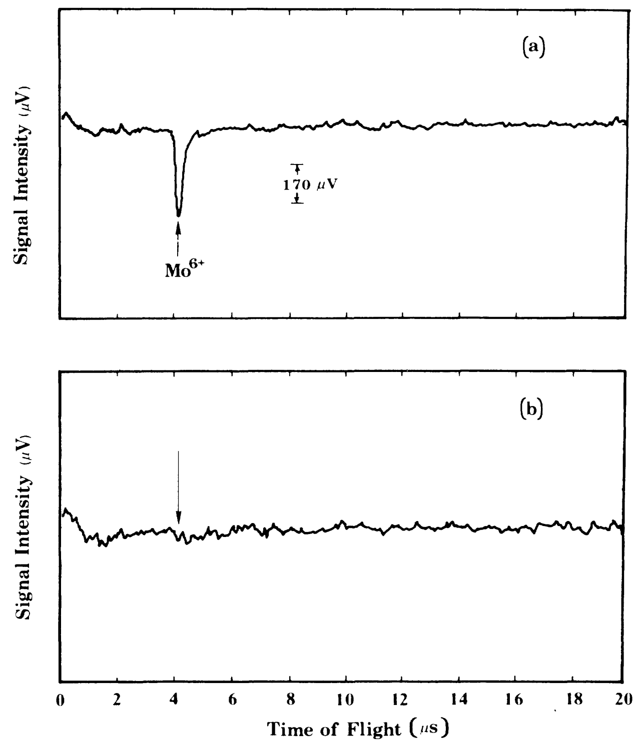

FIG. 3. Typical ion signals for Mo^{6+}. The trap was set to store only Mo^{6+}. The trap contents were sampled by applying a -70-V pulse to the end cap facing the CEM. The data in (a) were obtained using both ablation beams and the data in (b) with only one beam. An ion signal is clearly present in (a) but not in (b). Based on the ion optics collection efficiency of 0.2% and the channel electron multiplier gain of 10^5 driving a load of 200 Ohms, the observed signal indicates 10^5 ions are stored.

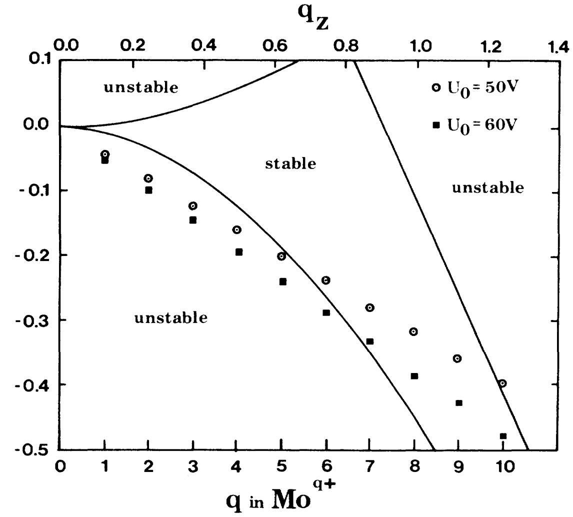

FIG. 4. Ion stability diagram for molybdenum. The ion trap with $r_0=1.63$ cm, $z_0=1.68$ cm is operated at $V_0=315$ V,$f=0.8$ MHz. Ions in the region labeled "stable" can be stored in the trap. The dc voltage U_0 on the ring electrode selects which ions are stored. Increasing U_0 from 50 to 60 V shifts Mo^{6+} out of the stable region. An ion signal is observed at $U_0=50$ V but not at $U_0=60$ V.

ions in a periodic hyperbolic potential.[12,13] The trap used in this study has a cylindrical electrode, and so the potential surfaces near its center only approximate those of an ideal hyperbolic electrode. However, the trap has been tested using N_2^+, N^+, Ar^+, and Ar^{2+} produced by electron impact. When the operating point of the trap is set at the edge or beyond the stable regions for each of the aforementioned ions, no storage is observed. It can be concluded, therefore, that the stability diagram for an ideal trap can be used to characterize the stable region for our rf trap with its cylindrical electrode. To determine the highest charge state of molybdenum ions, the trap was biased so that only Mo^{q+} with $6 \le q \le 9$ were inside the stable region. An ion signal with excellent signal to noise was observed. However, no signal was observed when the trap was biased so that Mo^{q+} with $1 \le q \le 6$ was outside the stable region. Since Mo^{6+} was the only ion that is shifted out of the stable region, the highest charge state of molybdenum observed must have been 6.

The charge-state identification has been confirmed by pulsing the ions into a time-of-flight spectrometer using a fixed acceleration voltage. Shown in Fig. 5 is a plot of the ions time of flight normalized to the time of flight for $q=1$ plotted against $q^{-1/2}$. The values of the times of flight and the distance traveled indicate that the ions are accelerated by an effective potential of 32 V for all ions. The ion dump pulse on the end cap is phase locked to the rf waveform. We believe that the effective potential is due to the sum of dc and rf bias on the ring electrode and the leading edge of the dump pulse bias on the end cap. An uncertainty weighted least-squares fit to the points gives a slope of 0.97±0.09. This clearly indicates that we have trapped and correctly identified multiply charged ions of Mo^{q+} with $1 \le q \le 6$. We have carried out similar experiments for tungsten and observed W^{q+} with $1 \le q \le 4$. The highest charge state obtained in our current experiments is believed to be limited by the power density of the ablation laser, which is operated at 2×10^8 W/cm^2.

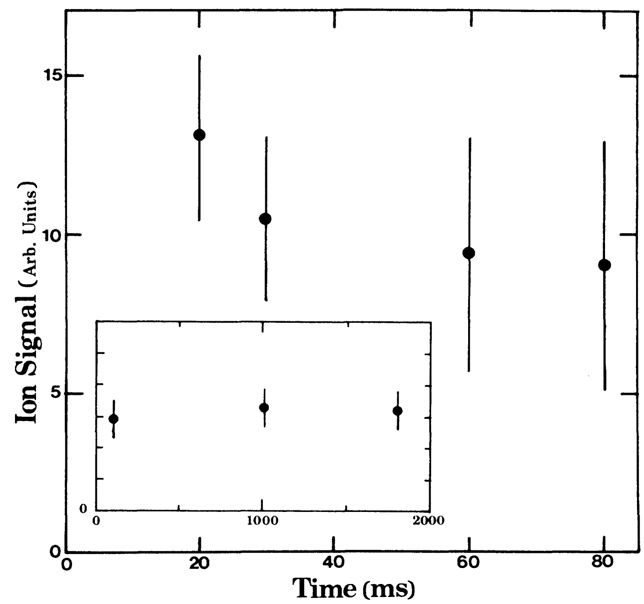

FIG. 6. W^{4+} signal amplitude as a function of storage time. The storage time is in excess of 2 s. The main figure shows the ion signal over a period of 80 ms, while the inset shows ion signal essentially constant to 2 s. The initial decay of the ion signal may be due to enhanced background gas pressure following laser ablation.

The storage time of the ions in the ion trap was determined by monitoring the decrease of the ion signal magnitude when the stored ions are pulsed out of the trap at progressively longer delay times. A plot of the data obtained is shown in Fig. 6. The storage time is in excess of 2 s for all ions stored. Depending on the charge state, the ultimate storage time is determined by the charge-

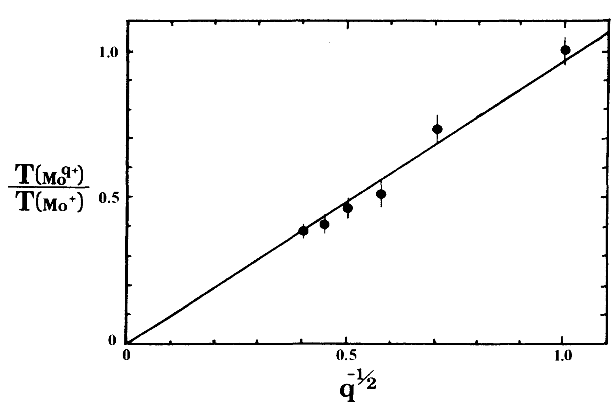

FIG. 5. Ions time of flight and charge-state identification. Shown plotted are measured times of flight normalized to the time of flight for Mo^+ vs the charge state $q^{-1/2}$. The data fit a straight line with a slope of 0.97±0.09, clearly indicating a correct charge-state identification. q_z and a_z are the parameters used in the Mathieu equation (Refs. 12 and 13).

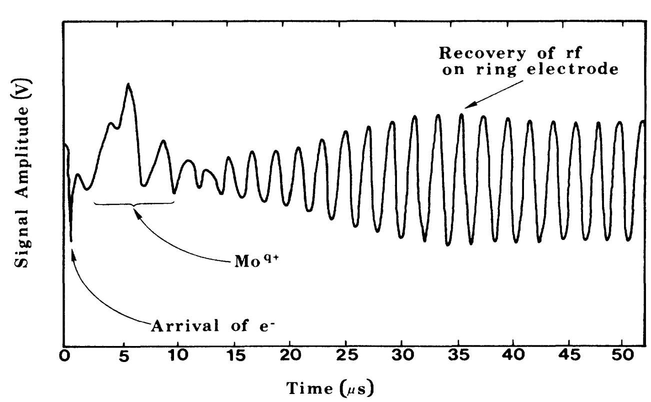

FIG. 7. Langmuir probe signal. The arrival of electrons and ions at the trap is detected almost immediately after ablation indicating that the ions' energies must be near 250 eV. Note that the probe also picks up a rf signal from the ring electrode. The presence of the electrons and ions from the ablation pulse influence greatly the rf amplitude at the ring electrode, effectively shorting it out momentarily.

transfer rate or the elastic collisions rate between the stored ions and the background gas which is about 3×10^{-9} Torr.

The pseudopotential well for trapping tungsten and molybdenum ions is estimated to be between 1.1 and 42 eV in this investigation depending on the operating parameters for the charge states observed. The maximum well depth gives the upper limit on the energy of the stored ion. An absolute upper limit of the energy of the stored W^{4+} and Mo^{6+} is 0.7 and 1.2 eV/amu, respectively. The upper bound for the energy of the trapped ion is lower for a lower charge state. However, the actual energies of these trapped ions must be substantially lower. Figure 7 shows a temporal profile of the laser-induced plasma monitored by the Langmuir probe. Electrons from the ablation plasma first stream into the ion trap followed by the slower-moving heavy ions. The high density of the fast-moving electrons temporarily shorts the ring electrode to ground as indicated by the sharp reduction of rf pickup by the Langmuir probe. The ions moving behind the electrons therefore enter the ion trap without seeing the rf field. The rf recovers its full amplitude only 30 μs after the arrival of the plasma electrons. During this short duration, the trap is unable to store any ion. Ions with energies above 4×10^{-3} eV/amu traverse the full diameter of the trap while the rf recovers. Ions which are still inside the trap after the rf has fully recovered must therefore have less than 4×10^{-3} eV/amu.

In summary, low-energy multiply charged ions have been produced and trapped for periods in excess of 2 s for all ions in this investigation. The relatively long storage time, the substantial number density, and the near universality of the technique will enable the study of various processes involving slow ions in a wide variety of low-energy multiply charged ions.

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