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Confined thermal multicharged ions produced by synchrotron radiation

D. A. Church and S. D. Kravis
Physics Department, Texas A&M University, College Station, Texas 77843

I. A. Sellin, C. S. O, J. C. Levin, and R. T. Short
Physics Department, University of Tennessee, Knoxville, Tennessee 37996
and Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831

M. Meron, B. M. Johnson, and K. W. Jones
Brookhaven National Laboratory, Upton, New York 11973
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Synchrotron x rays have been used to produce a confined multicharged ion gas near room temperature. Comparison of charge-state-number observations characteristic of ion formation and of ion storage, together with measurements of Ar-to-Ar^{q+} electron-transfer rate coefficients, provide information to estimate time constants for relaxation to thermal equilibrium and other stored-ion properties important to further development of the technique.

There is currently considerable interest in low-energy highly charged ions for the study of collision processes common in laboratory and astrophysical plasmas, for use in precision spectroscopic tests of quantum electrodynamics, and for the test of quantum-mechanical theories of electron transfer. K-electron photoionization of atoms followed by Auger electron ejection and electron shakeoff produces multicharged ions[1] with minimal energy transfer. We have used the "white" radiation on the X-26C beam line[2] of the National Synchrotron Light Source (NSLS) at Brookhaven National Laboratory to generate multicharged argon ions in a Penning ion trap, using proposed methods[3] designed to maintain and exploit their unique thermal production energies.[4] The result is a novel *stored thermal multicharged ion gas.* These efforts were originally motivated by a proposal for a low-energy high-brightness multicharged ion source called PHOBIS, in which sequential photoionization would generate high charge states.[5] In addition to potential studies of the photoionization of ions, our stored-thermal-ion technique opens a new energy frontier for highly-charged-ion collision studies.

We report here ion-storage and time-of-flight measurements which reveal considerable information about the cold charged-ion gas, and provide data for several calculations of stored-ion properties. The rate coefficients for the charge-changing collisions of near-room-temperature Ar^{q+} ions ($3 \leq q \leq 5$) with argon atoms were measured for the first time, and were used to estimate the relative equilibrium ion-charge-state numbers in the trap. With these results and with calculated photoionization cross sections, the temperature and characteristic relaxation time constants of the ion gas are estimated. From these, properties of the ion distribution in the trap are inferred, which are consistent with the measurements.

Figure 1 shows a detected spectrum of multicharged argon photoions confined in the Penning trap. This trap was of the standard type,[6] composed of a cylindrical ring elec-

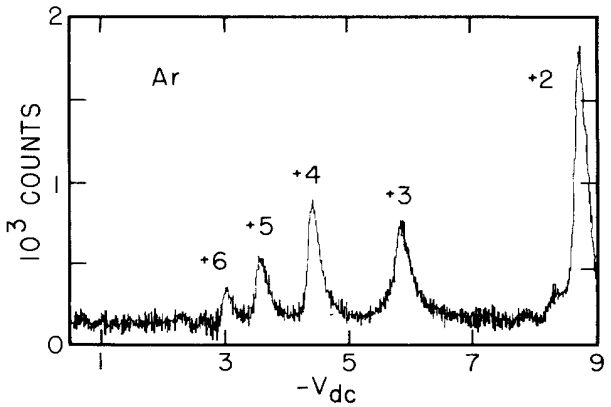

FIG. 1. Spectrum of argon charge states stored in a Penning ion trap with a 250 mV well depth at an argon pressure of 1.7×10^{-7} Torr, using unchopped, unmonochromatized synchrotron radiation. The small peaks to the left of Ar^{6+} are likely to be Ar^{7+} and Ar^{8+}.

trode and two end caps shaped to produce a dominant quadrupole potential when the end caps were at zero potential and a negative potential V was applied to the ring. The trap was mounted in a vertical uniform magnetic field $B \simeq 0.7$ T. A 2-mm-high, 0.75-mrad fan of bending-magnet synchrotron radiation from the X-26 beam line, sharply attenuated below 3 keV by a Be window, was channeled radially through a slot in the trap ring electrode. Since the total L- and M-shell photoionization cross sections are lower by at least an order of magnitude above 3 keV, basically only K-electron photoionization need be considered.[5] The radiation produced a "pancake" of ions near the minimum of the axial harmonic potential well of the trap, which had depth $D = V/2$, with D set at 250 mV. Together with B, the radial electric field produces a slow ion drift about the symmetry axis of the trap at angular frequency $\omega \geq \omega_- = 2.8$ kHz, rather than translational kinetic energy.

Recently, time-of-flight (TOF) measurements[4,7] were completed on various multicharged ions with charge states to $+9e$, which were created near room temperature by K-shell photoionization. Figure 2 shows the somewhat different TOF spectrum of argon ions produced on X-26C at the NSLS using this "scalpel"[4] technique. From the widths of the ion peaks in this and similar spectra, the average energies of the recoiling argon ions were calculated to be 0.04 eV, corresponding to a temperature near 480 K.

In the Penning trap with $D = 0.25V$, the mean additional axial energy derived from such ion production in the dc potential is $qDz^2/2z_0^2 = 2\times10^{-3}q$ eV. The cyclotron-motion degrees of freedom oscillating with angular frequency $\omega_+ \approx \omega_c$ should have a mean energy corresponding to the recoil temperature discussed above. Thus an ion gas is created with mean energy near 40 meV for all charge states. Decreasing the axial well depth to 125 mV begins to produce noticeably smaller ion signals, probably due to loss of the higher-energy ions of the equilibrium distribution which forms in the axial well. Note that these confining wells are ten times shallower than were possible with recoil ions having comparable charges, created using fast-heavy-ion impact.[8]

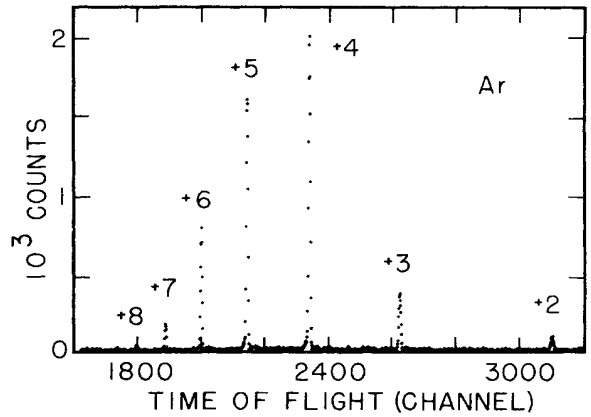

FIG. 2. Spectrum of argon charge states produced by unmonochromatized synchrotron radiation, as determined by time-of-flight spectroscopy.

The stored ions were detected using a resonantly excited tuned circuit made by connecting an inductor in parallel with the capacitance of the trap end caps. When the ring potential V was ramped to more negative values (larger magnitudes), the ion axial oscillation frequencies $\omega_z = (qV/mz_0^2)^{1/2}$ successively passed through the tuned circuit resonance, with the lowest charge state last, as Fig. 1 shows. The energy absorbed by the driven ions reduced the rf amplitude, producing modulations proportional to ion number which were amplified and detected.[9] Some ions other than argon ions, but with much higher mean energies, were detected when starting at deeper well depths. These hot ions are thought to arise from ionization of molecular residual gas followed by dissociation. Their signals were considerably broader than those of the cold Ar ions, making them difficult to identify. Ar^{2+} and Ar^{3+} signals of similar width were produced in separate measurements using electron-impact ionization, which generates ions throughout the axial potential. At the low well depths used to study the cold photoions, broad hot-ion signals essentially vanished, as Fig. 1 shows.

If the synchrotron beam is not blocked through a cycle of ion creation and detection lasting $\gtrsim 1$ s, competition between ion production through photoionization and ion loss by charge-changing electron-transfer collisions, with the target Ar gas, result in an equilibrium ion number in each charge state. The rate of ion change is $dN/dt = f_q G n l - N_q n k_q$ so that the equilibrium ion number is $N_q(\text{eq}) = f_q G l/k_q$, where $G = \int_{E_K}^{\infty} \sigma(E) F(E) dE$ is the integral above the K edge of the photoionization cross section per atom, $\sigma(E)$, times the number of photons/per sec, $F(E)dE$, incident through the trap in energy interval dE. The ion production length is $l \lesssim 2r_0 \approx 2.25$ cm, k_q is the rate coefficient for electron transfer with the target gas, and f_q is the fraction of ions produced in charge state q. G is 10^{-5} photon cm^2/s with 70 mA of electrons in the NSLS x-ray ring. From the equation for $N_q(\text{eq})$, the deviations of the equilibrium stored-ion numbers (Fig. 1) from the created numbers (Fig. 2) can be interpreted as due to different loss rates by electron transfer.

To calculate $N_q(\text{eq})$ we have obtained the rate coefficients for electron transfer from Ar to Ar^{q+} $(3 \leq q \leq 5)$ near room temperature, by measuring the storage time constant in the trap for these charge states at several pressures of Ar gas, with the photon beam blocked through the part of each cycle after ion information. Synchrotron x rays to form the ions were passed through the trap for about a second, and then pulsed off by mechanically rotating a shutter with a fast stepper motor. Tests showed that the closure times of the shutter were ≈ 5 msec, far shorter than the measured storage times of the ions. The losses of the stored-ion charges were found to be exponential. The time constants for these decays, τ_q, together with the target gas density n, enables a calculation of the rate coefficient $k_q = (n\tau_q)^{-1}$. The results of the rate-coefficient measurements are shown in Table I. For Ar^{2+}, k_2 was not measured. It is listed as both the maximum rate for ground and metastable states of the ion, obtained at 300 K from drift-tube measurements,[10] and as the rate from one ion-storage measurement[11] near 1.3 eV. Mea-

TABLE I. Measured rate coefficients for thermal Ar^{q+}-Ar charge-changing collisions. Estimated random uncertainties are in parentheses.

Charge q	Rate coefficient k (cm^3/s)
2	6.3×10^{-12} [a]
	8.9×10^{-10} [b]
3	$1.45(60)\times10^{-9}$
4	$1.7(4)\times10^{-9}$
5	$2.6(7)\times10^{-9}$

[a]Largest k value at 300 K, ground or metastable states (Ref. 10).
[b]Ion trap measurement (Ref. 11) near 1.3 eV.

surements of Ar^{q+}-Ar charge-changing cross sections have been previously made[12] at energies as low as $1.8q$ keV. These cross sections for $q=3$, 4, and 5 show the same trend with q as do our corresponding rate measurements, but these ion energies are so high relative to 40 meV that no meaningful comparison can be drawn. Our measured rate coefficients are slightly higher than Langevin orbiting rates,[9] which scale as q, if 50% capture probability is assumed.

Using the cold-ion data, relative equilibrium ion numbers $N_{q1}/N_{q2}=f_1k_2/f_2k_1$ were computed and tabulated with the data from Fig. 1 in Table II. The ratio N_5/N_4 is well reproduced, and the higher-charge-state ratios seem compatible with typical charge-transfer loss rates. The ratio N_3/N_4 is higher than expected and the ratio N_2/N_4 is much lower, or much higher than expected, depending on which rate coefficient is used. The 2+ charge-state ratio may be evidence that the rate is a strong function of ion energy between 300 K and 1.3 eV. However, charge-state equilibrium was probably not reached in our measurement, if the lower rate is appropriate. The 3+ ions must have an additional production rate, since the ion loss rate is experimentally determined. Production of Ar^{3+} by electron capture into higher charge states, or by photoionization of Ar^{2+}, are highly unlikely; a background contribution to the 3+ peak, which has a different shape, may be sufficient to explain the ratio.

There are expected to be appreciable rates for symmetric resonance charge transfer between atoms and thermal ions[13] with charges of 1+, 2+, and possibly 3+.

TABLE II. Ratios of measured numbers of stored ions in different charge states (from Fig. 1). Calculated values are based on the relative number of ions produced [from areas under the peaks in Fig. 2 and measured rate coefficients (Table I)]. The 4+ peak in each distribution was normalized to 1. Estimated uncertainties are in parentheses.

Charge states	Expt. ratio	Calc. ratio
(2+/4+)	2.5	66
		0.46(11)
(3+/4+)	0.9	0.44(18)
(4+/4+)	1.0	1
(5+/4+)	0.4	0.46(14)

These collisions will result in cooling, if $T_{ion} > T_{atom}$. The collision time constant, using the orbiting cross sections dictated by experiment,[13] will be near 0.1 s at the pressure at which the data in Fig. 1 were taken. Higher charge states are collisionally coupled to Ar^{2+} (see below), so they also will be cooled.

Using the magnitudes of parameters identified earlier, the estimated density of Ar^{4+} is $N_4(\text{eq})/v=\rho_4=6\times10^3$ cm^{-3}, where $v=2\pi r_0^2 z\simeq0.8$ cm^3. The self-collision time[14] of an Ar^{4+}-ion gas is given by

$$t_c=11.4A^{1/2}T^{3/2}/\rho_4 q^4\ln\Lambda\approx 50\text{ ms}\ ,$$

where A is the mass number and the shielding parameter $\ln\Lambda\approx15$. The self-collision time is the time constant for the ions to relax to temperature equilibrium via Coulomb collisions with like charges and masses, following a departure from equilibrium. Comparable times hold for the other stored charge states. This relaxation time constant equation has been tested for singly charged ions with measured temperature in a radio-frequency quadrupole trap.[15] The time constant for thermal equilibration between different mass and charge states A,q and A_1,q_1 is given by[14] $t_{eq}=5.87AA_1(T/A+T_1/A_1)^{3/2}/(\rho q^2q_1^2\ln\Lambda)\simeq0.1$ s for equilibration between the 3+ and 4+ ions, a typical value. For the customary ion storage times $\gtrsim1$ s, our working hypothesis of a stored multicharged ion gas in thermal equilibrium appears justified.

The ion-ion collisions leading to thermal equilibrium create torques which modify the (originally uniform) radial distribution of charge states, with higher-m/q-ratio ions migrating to larger radius, and small-m/q-ratio ions toward the symmetry axis.[16] The thermal equilibrium charge distribution should rotate at a well-defined frequency. The analysis of this centrifugal separation[16] presupposes a Debye length λ_D (cm) $=0.74[T\ (\text{eV})]^{1/2}\times[10^6/\rho\ (\text{cm}^{-3})]^{1/2}$ small compared to the charge distribution radius. From the parameters determined here for 4+ ions, λ_D is comparable to $r_0/2$, so centrifugal separation will be incomplete.[17] Nevertheless, it will probably lead to more rapid radial loss of the lowest charged ions, Ar^{2+} and Ar^{3+}, as the charge cloud slowly expands. However, since the Ar^{3+} ratio is higher than expected, rather than lower, such losses must proceed slowly compared to measurement times. No conclusion is drawn from the Ar^{2+} ratios.

Future employment of higher photon fluxes with no Be window, including radiation from wigglers or undulators, as well as from bending magnets, are planned. These improvements will lead to higher stored-ion densities, lower target pressure, more rapid relaxation, and an improved approach to the optimum parameters for centrifugal separation of the different charge states. These ions will make an ideal target for sequential photoionization to produce even higher charge states at comparable energies.[3]

We have produced and studied a novel confined ion gas of multicharged ions at thermal equilibrium near room temperature in a Penning ion trap. Measurements and calculations supporting this interpretation of the data have been presented. The first rate-coefficient measurements for charge-changing collisions of highly charged ions at such low energies have been obtained.

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