Production of Very-Low-Energy Highly Charged Ions by Synchrotron Radiation

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Very-low-energy highly charged ions have been produced by use of white and monochromatic x rays from a wiggler line at the Stanford Synchrotron Radiation Laboratory to generate vacancy cascades following inner-shell photoionization. Recoil-ion energies have been determined and are shown to correspond essentially to room temperatures, even for high charge states. Promising applications to study of the interaction of stellar radiation with cold matter, to high-brightness ion-source development, to precision spectroscopy, and to angle-resolved chemical-physics reactive-scattering studies are discussed.

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We report here for the first time on experiments which determine the average energy of high-charge-state ions created in vacancy cascades subsequent to the production of single K vacancies in Ne and Ar, and L vacancies in Kr and Xe, by x rays from a Stanford Synchrotron Radiation Laboratory (SSRL) wiggler line. These "recoil" energies are found to be so small that they lie well within the range of characteristic energies of the Maxwell-Boltzmann distribution of the target gas at room temperature. We therefore may speak of a noninteracting ensemble of highly charged ions at (approximately) *room* temperature, even though details of the modification of the original equilibrium distribution due to multiple ionization have not yet been fully determined. The ion temperatures are $\gtrsim 6$ orders of magnitude lower than those characteristic of laboratory and stellar plasmas in which ions of corresponding ionization states have comparable abundances. These temperatures are also ~ 2 orders of magnitude smaller than those characteristic of recoil ions produced with charged-particle impact as the direct ionizing agent[1] (the so-called "hammer" beam method initiated by L. Cocke and co-workers after $\lesssim 1$ eV energies had been pointed out and verified by Sellin *et al.*). We discuss below possible novel applications of our x-ray "scalpel" method to atomic spectroscopy and collisions physics, to astrophysics, and to chemical-physics scattering experiments.

Use of a unique time-of-flight (TOF) analyzer[1] has permitted joint examination of ion photoproduction rates and temperatures through study of the yields and widths of the corresponding TOF peaks. These peaks arise from the arrival of ions at a dual channel-plate detector ~ 1 μs subsequent to their creation by a synchrotron x-ray burst in a dilute gas sample (~ 0.2 mT) maintained in the extraction region at the front end of the analyzer. The timing resolution of $\lesssim 1$ ns was achieved by use of a time compression feature of the analyzer, of stable, fast-rise-time TOF start pulses derived from the storage-ring rf electronics, and of careful low-noise time-pickoff techniques applied to pulses from the detector. The synchrotron-radiation burst itself has a measured FWHM of ~ 300 ps.[2]

Beams of white (and later monochromatic) x rays from an eight-pole wiggler operated at 15 kG were focused by a toroidal mirror and collimated to a 2-mm-diam x-ray beam at the position of the gas target. The critical energy of the radiation was 4 keV (corresponding to the 2-GeV electron-beam energy) and was attenuated below 3 keV by Be windows. This target was viewed by the vertically mounted TOF analyzer through a 3-mm-long slit located just above the x-ray beam. X-ray intensity was monitored by ion chambers positioned both upstream and downstream of the UHV system housing the analyzer. Typical flux through the target was $\sim 10^{12}$ photons mm^{-2} sec^{-1}, and total ion counting rates in the TOF detector were ~ 1–5 kHz. Detector backgrounds from stray x rays were found to be negligible ($\lesssim 3$ Hz).

The x-ray throughput was sufficient to permit the

use of a Si(111) double-crystal monochromator to tune the radiation (bandpass 1×10^{-4}) above, below, and interleaving the L_1, L_2, and L_3 edges of Xe to explore the effects of tuning the x rays through an absorption edge. The timing resolution achieved sufficed not only to make the desired temperature measurements but also to achieve adjacent-mass-isotopic resolution (Fig. 1) of the charge-state (q) spectra of Ne, Ar, Kr, and Xe. This fact alone qualitatively indicates low ion temperatures—otherwise the TOF spectra would be hopelessly smeared. The results achieved largely agree with and make substantial improvements on the pioneering efforts of Carlson and co-workers,[3] who used x-ray guns and filters to study q spectra of recoil ions arising from similar vacancy cascades in the same gases; and of Hastings and Kostroun,[4] who used monochromatic x rays from the Cornell High Energy Synchrotron Source storage ring to examine the dependence of Kr q's on tuning through the Kr K edge.

Our results improve upon this fine earlier work in two ways. First, our ion counting rates are about 2 to 3 orders of magnitude higher than those of Carlson *et al.*, who moreover had insufficient x-ray intensity to permit the use of a monochromator. Second, the timing resolution achieved, the key to making the temperature measurements of central concern here, is significantly better than that available to Hastings and Kostroun. In our analyzer (as in theirs) the finite thickness of the source region—a source of spread[5] in time of arrival at the detector—is compensated to first order. An added advantage is the short length of our analyzer, $\lesssim 25\%$ of that used by Hastings and Kostroun. Short length is important to keep flight times of interest in the range between successive x-ray bursts. Also, event statistics benefit from a short timing cycle, since more cycles are accommodated in the beam time available.

Examples of TOF spectra are shown in Fig. 1. With few exceptions the match of these and many other q spectra with those of Carlson *et al.*[3] is excellent when the energy range of the x rays employed is similar, showing that except near shell edges final q distributions are not sensitive to the x-ray spectral profile. The close match in mean charge $\bar{q}$ (cf. Table I) also demonstrates flat detection sensitivity versus q and absence of other serious systematic errors in both experiments, whose methods vastly differ. Two exceptions are worth noting. First, some of the low-q data of Carlson *et al.*[3] for all four gases exhibit lower abundances than ours. Carlson *et al.*[3] note, however, that properties of their mass-spectrometric equipment made data acquisition less reliable for low q. Second, while the shifts in $\bar{q}$ that we observe as the L_3 and L_2 edges of Xe are crossed are similar to those seen by Carlson *et al.*, the shift of ~ 0.1 charge state that we observe as the L_1 edge is crossed is much smaller than 1.7, as seen by them. Consideration of Coster-Kronig transition probabilities[6] for transfer of an L_1 vacancy to the $L_{2,3}$ subshells

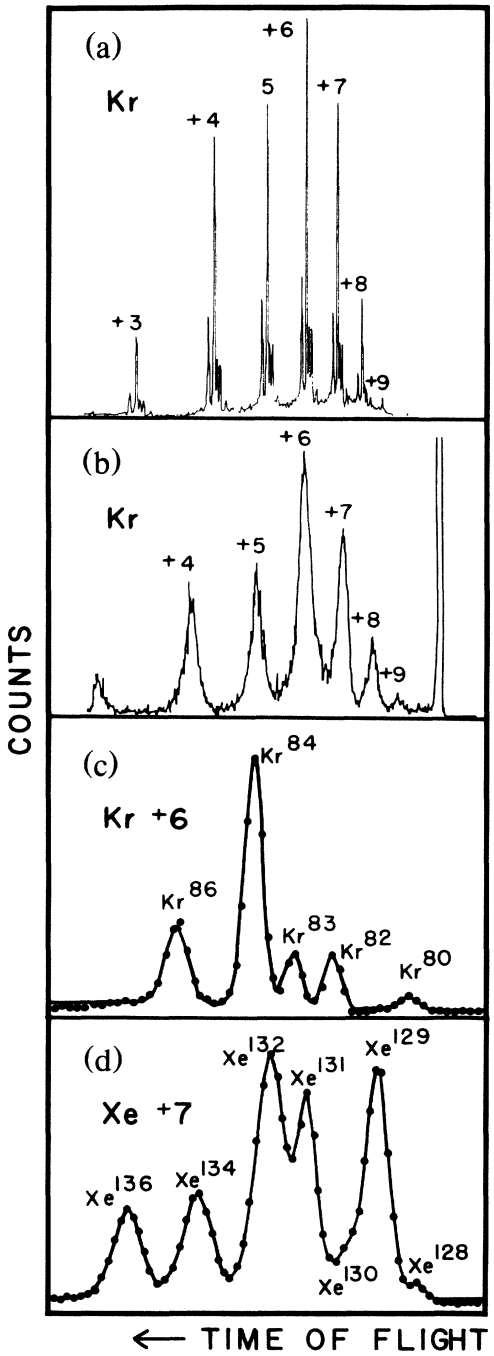

FIG. 1. (a) Kr charge-state distribution following L-shell vacancy production. Plotted is number of ions detected as a function of flight time. (b) Earlier work by Hastings and Kostroun (Ref. 4) displaying similar results. (c) Expanded view of a Kr peak from (a), demonstrating isotopic resolution. (d) Expanded view of a Xe peak from a typical Xe spectrum.

TABLE I. Average recoil energy U_0 and charge state $\bar{q}$ following inner-shell vacancy (as noted for each gas) produced by photoionization.

Gas	U_0 (expt.) (eV)	U_0 (est.) (eV)	$\bar{q}$ (expt.)	$\bar{q}$ (Ref. 3)
Ne (K)	0.16	0.11	2.3	2.3
Ar (K)	0.041	0.047	4.2	4.2
Kr (L)	0.038	0.032	5.4	6.7
Xe (L)	0.040	0.030	7.8	9.1

leads to an estimated shift of ~ 0.5 unit. Resolution of the discrepancy will be sought at the earliest available opportunity.

Figure 2 displays plots of suitably scaled data from which approximate ion temperatures were deduced (see Table I). For each gas the temperature was deduced from a fit of the widths at FWHM of the symmetrical TOF peaks by the functional form

$$\Delta t^2 = \alpha + \beta/q\kappa + \gamma/(q\kappa)^2. \quad (1)$$

Here α, β, and γ are assumed constant for each gas, q is the charge state, and κ is an electric field scaling parameter representing four different values ($\frac{1}{8}$, $\frac{1}{4}$, $\frac{1}{2}$, 1) of proportionately scaled electric fields in the TOF analyzer. The first term represents overall electronic timing resolution; the second term reflects a width associated with variation of flight time with trajectory separation from the TOF axis (due to field fringing)[5]; and the third arises from finite recoil energy U_0 at the moment of ion birth.[5]

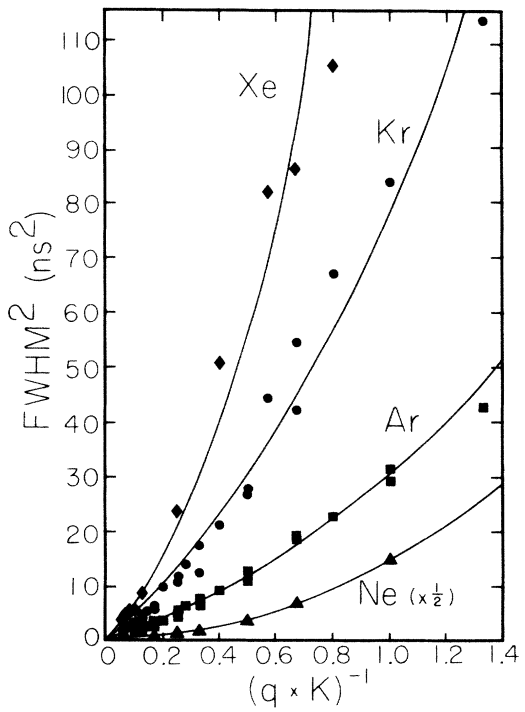

FIG. 2. Dependence of TOF peak width Δt on electric field scaling parameter κ and charge state q. Plotted is $(\Delta t)^2$ as a function of $1/q\kappa$ for each gas.

Figure 2 shows data clustering around fitted curves whose universality for all q verifies the correctness of our *Ansatz* for Δt^2. The near-zero fitted α value corroborates the good timing resolution claimed. The influence of quadratic terms is evident. When fitted values of γ are translated into U_0 values, only a weak dependence of temperature on q is observed, consistent with *no* dependence at the factor-of-2 error level. Strictly, the axial contribution to U_0 is what is observed; it is likely that ions directed along the horizontal electric vectors of the x-ray photons are slightly hotter (cf. the discussion of Ne data below). Error bars on U_0, difficult to estimate since the addition-in-quadrature assumption in Eq. (1) has not yet been tested, can be narrowed. Improved fitting programs for the multi-isotope TOF peaks will also aid in reduction of error bars, likely permitting extraction of a weak charge-state dependence as well. Comparisons of U_0 with estimates based on an extremely simple, physically well-founded model are displayed in Table I. The assumption is that the first Auger event provides one of three appreciable contributions to U_0; the second is a contribution due to photoelectron recoil; the third is initial thermal spread. Here we have neglected competing radiative decay channels (well known to have low fluorescence yields). While in most cases the added momentum imparted to the target particle in the primary photoionization event is small, high photoelectron energy leads to substantial ion recoil for Ne, whose K edge lies far below the $\simeq 3$-keV-window cutoff. Although the horizontal x-ray polarization gives rise to photoelectron emission velocities peaked perpendicular to the axial direction which therefore contribute little to the axial energy, components along the TOF axis are appreciable for Ne. Consideration of the axial velocity components of the dipole distribution leads to an increase in anticipated U_0 for Ne recoils, which is consistent with the hotter Ne temperatures observed (cf. Table I).

Having established the ability to create highly charged ions in a room-temperature environment, as well as the ability to extract and manipulate ions born in it, we conclude by citing examples of the abundance

of interesting experimental possibilities that this work opens, within atomic physics and more broadly. Intriguing though speculative (no data) related papers by Jones, Johnson, and Meron and by Church *et al.* have been published.[7] The former authors proposed to create a trapped-ion source utilizing successive photoionization from the outer shells inward to create highly charged ions in realistic periods of time (e.g., Ar^{18+} in ~ 7 s). Church *et al.* discussed the feasibility of storage of highly charged ions (e.g., Ar^{17+}) in a Penning trap, and concluded that storage at near thermal energies should be possible. The present experiment not only provides proof of near thermal energies, but also presents hard data concerning ion yields per transmitted x ray. It also unequivocally establishes the feasibility of the x-ray pumping portion of the proposed ion-trapping scheme.

Development of a *very cold (or x-ray vacancy cascade) ion source* promises even greater benefits to angle-resolved atomic collision studies. Such a source has extremely high potential brightness, since synchrotron x rays can be focused to spot sizes ~ 100 μm in diameter, and the measured energy spreads are very low. Extraction of ions to form very-low-energy beams ($\lesssim 10$ eV) of low energy spread and good angular definition appears within reach. Low-brightness beams having otherwise attractive properties can be achieved by monochromatization of ions (at great expense in intensity) extracted from a source region using the "hammer" method.[1] Our x-ray "scalpel" method shares nearly all of the advantages of the "hammer" method, but features 2 orders of magnitude improvement in reducing ion-temperature limits. An additional significant advantage is the subnanosecond pulsed nature of the excitation. Such high-quality pulsing lends itself naturally to development of coincidence and TOF experiments with excellent possibilities for noise rejection.

Certain atomic-physics experiments may benefit materially from exploitation of the techniques discussed here. Chief among these possibilities are precision-spectroscopy experiments—for example, high-precision measurements of Lamb shifts and intervals between high Rydberg levels of one-, two-, and three-electron ions. As pointed out some time ago by Sellin *et al.*,[1] the use of target-ion recoil spectroscopy may permit the reduction of Doppler shifts and spreads (which typically limit the precision of fast-beam approaches to this problem) by 3–4 orders of magnitude in v/c. The "scalpel" method promises to reduce these spreads by an *additional 1–2 orders of magnitude in* v/c.

Beyond the fundamental atomic-physics problems which such a room-temperature source may help address, it appears to have high potential for applications to the study of the interaction of radiation from stars of all temperatures with cold matter (e.g., interstellar clouds), for development of a very-cold ion source for multiply charged ions of high brightness and ion-optic quality, and for chemical physics, where use of such ion-source technology may result in high-resolution ion beams of kinetic energy $\lesssim 10$ eV suitable for use in novel angle-resolved chemical charge-transfer and reactive-scattering studies.

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