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Cold Highly Ionized Ions: Comparison of Energies of Recoil Ions Produced by Heavy Ions and by Synchrotron Radiation X rays

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Abstract

The energies of highly excited, high-charge-state recoil ions produced by fast heavy-ion impact on target atoms ("hammer" method) have been compared with the energies of similar-charge-state recoil ions produced by vacancy cascades subsequent to inner-shell photoabsorption of tuned synchrotron radiation x rays ("scalpel" method). These comparisons show that the "hammer" method leads to recoil ion temperatures typically 4 orders of magnitude lower than those which occur in plasma sources in which ions of similar ionization and excitation states have comparable abundance, while the "scalpel" method leads to temperatures up to 6 orders of magnitude lower. Advantages and drawbacks of each method for potential precision spectroscopy of stored or trapped high charge state ions, and for production of extracted beams of low emittance for use in secondary ion-atom collision studies at eV to keV energies are discussed.

1. Introduction

In this paper we review recent progress in the quantitative determination of recoil ion energies produced using the "hammer" and "scalpel" methods, emphasizing those aspects which have principal applications to the central theme of this conference: access to the physics of stored and trapped particles. Generally, the colder the species of interest which are produced, be they neutral, singly, or multiply charged systems, the better, since the potential for

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refrigeration to still lower temperatures is a central concern of many scientists in the field. Production of highly ionized and excited ions at low temperatures poses a particularly severe challenge, since typical sources of such ions such as stellar, fusion, and laser plasmas typically involve temperatures in the 1 to 100 keV temperature region [1], and fast beam sources which achieve similar ionization-excitation states typically involve beam velocities $v/c \sim 0.1$ [2]. In both cases Doppler spreads and shifts tend to severely limit spectroscopic precision. Similarly, the emittance of plasma and fast beam sources is not attractive if one has the objective in mind of carrying out high-charge-state ion-atom collision experiments [3] at eV to keV energies under conditions where good energy and angular definition of high-charge-state projectile ions are important. As will be seen below, the "hammer" method permits achievement of 4 orders of magnitude lower temperature than plasma sources in which similar ionization-excitation states have comparable abundance, as well as 3 orders of magnitude advantage in v/c relative to fast beam sources. The "scalpel" method permits achievement of up to 6 orders of magnitude lower temperature, and 4 orders of magnitude advantage in v/c . These advantages are accompanied by the drawback of recoil sources being relatively weak sources, a disadvantage of less importance in trapping and storage devices than in many other applications.

The low recoil-ion energies intrinsic to the "hammer" method were first pointed out and verified by Sellin et al. [4], an observation soon thereafter followed up by Cocke [5] who exploited these recoils as a secondary source of keV-energy ions for the first in what is now a very long chain of active experiments on such topics as single and multiple electron capture by slow, highly charged ions [3,6].

Meanwhile, electron cyclotron resonance (ECR) ion sources have come on

line, and a new generation of cryogenic electron beam ion sources (such as CRYISIS at Atomfysik) are about to. These latter sources have many advantages for carrying out such collision experiments, especially in their intensity vs. charge state characteristics, but lack the attractively low potential emittance of the "scalpel" source.

In applications where production of high-charge-state ions of low emittance is an important consideration, and low production rates are sufficient or even desirable -- as they often are in trapping applications -- both the "hammer" and "scalpel" methods are likely to continue to be exploited to good advantage. Significant potential benefits of the "scalpel" technique include development of low emittance, sub-nanopulsed ion sources well suited to coincidence experiments in such areas as:

- angle-resolved high-charge-state ion-atom and ion-molecule collisions in the ~ 10 to few 100 eV range;
- new opportunities to study the interaction of "tunable" radiation simulating stellar radiation of virtually all temperatures with cold atoms and molecules (found for example in interstellar clouds);
- techniques for making precision spectroscopy on few-electron ions (e.g. intervals between high Rydberg states, Lamb shifts, etc.), which take advantage of the enhanced capabilities for trapping and further cooling that production of cold ions makes possible;
- new opportunities to study photoionization of stored or confined ions;
- development of new methods to study molecular interactions with surfaces, and molecular stereochemistry;
- study of site- and element-specific radiation damage at highly localized sites.

Further valuable development of the "hammer" method may also be

forthcoming in the foreseeable future. The possibility of producing a significantly more intense, parasitic source of bare and few-electron low-energy ions ($>10^{12}$ extracted ions/sec), "hammered" at MHz frequencies in a heavy-ion storage ring, has recently been conjectured by Ullrich et al. [7].

Despite the abundance of activity in the field of recoil ion production and use and the importance of the parasitic-ion-source possibility, few data concerning the energies and angular distributions of the primary recoil ions have been published [8]. Ullrich et al [8] have recently published a measurement of the entire recoil-energy distribution for Ne^{3+} ions produced by 1.4 Mev/u U projectiles, using a specially designed time-of-flight (TOF) spectrometer. Substantial new knowledge of intrinsic interest, as well as practical significance for design of parasitic sources, can be gained from measurements of recoil energies and angular distributions.

In this paper we review recent measurements of mean ion recoil energies E_r carried out using the scalpel method at the Stanford Synchrotron Radiation Laboratory, and using the hammer method at the ORNL EN tandem accelerator facility. As most of the data and details concerning the methods have been covered in recently published or soon to be published papers [8,9,10,11] we concentrate here on summarizing the results and interpreting them in the context of the principal concern of this meeting, the physics of stored and trapped particles. Further technical details may be found in the references [8,9,10,11].

2. Determination of mean recoil ion energies E_r .

2.1. "Scalpel" method determinations.

The "scalpel" technique involves use of a time-of-flight (TOF) analyzer similar in design to that used by our colleagues L. Liljeby and H. Danared at Atomfysik in Stockholm, to permit joint examination

of ion photoproduction rates and energies through study of the yields and widths of the corresponding TOF peaks. These peaks arise from the arrival of ions at a dual channelplate detector $\leq 1 \mu\text{s}$ subsequent to their creation by a synchrotron radiation x-ray burst in a dilute gas sample ($\sim 0.2 \text{ mT}$) maintained in the extraction region at the front end of the analyzer. The short length of the analyzer ($\sim 10 \text{ cm}$) 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.

An example of a TOF spectrum for Xe, irradiated by white x-rays from SSRL beam line IV-2 are shown in Fig. 1. The timing resolution achieved sufficed not only to make the desired temperature measurements but also to achieve adjacent mass isotopic resolution of the charge state (r) spectra of Xe. This fact alone qualitatively indicates low ion temperatures -- otherwise the TOF spectra would be hopelessly smeared.

In our initial study [9], beams of white (and later monochromatic) x-rays from an 8-pole wiggler operated at 15 kG were focussed by a toroidal mirror and collimated to a 1-mm diameter 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 up- and downstream of the uhv system housing the analyzer. Typical flux through the target was $\sim 10^{12} \text{ photons mm}^{-2} \text{ sec}^{-1}$, and total ion counting rates in the TOF detector were $\sim 1\text{--}5 \text{ kHz}$. Detector backgrounds from stray x-rays were found to be

negligible (< 3 Hz). The x-ray throughput was sufficient to permit the use of a Si(111) double crystal monochromator (bandpass 1×10^{-4}) to tune radiation above, below, and interleaving the L_1 , L_2 , L_3 edges of Xe to explore effects of tuning the x rays through an absorption edge.

Measurements of mean charge state $\bar{r}$ are presented in Table I. The mean charges listed can be compared with pioneering mass-spectroscopic measurements carried out long ago using x-ray guns and filters by Carlson et al. [12], and (for the case of Xe) very recent measurements by Tonuma et al. [13] using monochromatic x rays from the Photon Factory. In general, the agreement between our results and the measurements of Tonuma et al. is seen to be excellent, resolving a discrepancy we observed [9] in the mean charge state increment measured for Xe as the L_1 edge is crossed. Consideration of Coster-Kronig yields [14] for transfer of an L_1 vacancy to the $L_{2,3}$ subshells and of relative photoionization cross sections of the $L_{1,2,3}$ levels [15] leads to an estimated shift in mean charge state on the order of that seen by our group and by Tonuma et al.

Mean recoil energies were determined through study of the widths of the corresponding TOF peaks, using procedures similar to those described earlier [9]. For each charge state r the mean initial energy E_r was deduced from a fit of the peak widths at FWHM to the form

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

The constants α , β , γ represent contributions to measured flight time from, respectively, timing uncertainty; flight time variations due to field fringing; and recoil-ion energy [9,16]. The electric-field scaling parameter (κ) represents six different choices of proportionately scaled [9,16] electric field values in the TOF

analyzer. The essential correctness of the Ansatz for ΔE was well verified by the clustering of TOF data around fitted quadratic curves, as was illustrated in Fig. 2 of [9] for the rare gases ionized by synchrotron radiation, and in Fig. 2 below for Ar^{+n} recoil ions produced by 23 MeV Cl^{+5} incident projectile ions and detected in coincidence with emergent Cl^{+4} projectiles (simultaneous triple electron loss from the projectile). Comparative recoil energies determined in this manner are summarized in Table II, comment on which is deferred until these "hammer" measurements are summarized.

2.2 "Hammer" method determinations

A nearly identical TOF technique has been used to measure the energies of recoil ions produced using the "hammer" technique. Beams of $\sim .5 - 1$ MeV/u Cl^{+5} produced by the ORNL EN Tandem have been used to create argon and neon recoil ions which have been observed both in "singles" mode (averaged over all scattered ion charge states), and in coincidence with individual scattered ion charge states ranging from 6 to 8.

The nearly symmetric ion-atom collisions discussed here correspond to impact parameters $\sim .5$ au. [8,10]. Recoil energies for similar impact parameters estimated by simple elastic Coulomb scattering theory, in which $E_r \propto (qr)^2$, where q and r are the projectile and recoil charge states, respectively, are an order of magnitude smaller than those we observe. When present measurements of log recoil energy are plotted vs log qr ($q = 5$, $r = 6, 7, 8, 9$), a surprisingly linear dependence is observed, suggesting a power law of the form $E_r \propto (qr)^n$ where least-squares fit results give $n \sim 10$. This extraordinarily steep dependence is due, in part, to the low q ($q = 5$) used in the fits. Since projectile loss cross sections are large compared to capture cross sections for Cl^{+5} at these energies, most argon recoils are accompanied by scattered projectiles of charge state higher than

five. Similar plots for the data obtained in coincidence with scattered projectile charge state suggest an exponent $n \sim 5$, as illustrated in Fig. 3. This still strong dependence, far steeper than the $n = 2$ obtained for elastic Coulomb scattering is due primarily to the fact that effective charges seen by the target and projectile nuclei at the moment of closest approach are of much greater importance in determining recoil energy than are the asymptotic values (q,r) . In fact, good agreement with present measurements is obtained by using effective target and projectile charges obtained by Hartree-Fock calculations [8,10] to allow for the actual screening prevailing near the moment of closest approach.

These results illustrate a hazard in using a familiar and often useful rule-of-thumb estimate provided by Olson, Schlacter et al. [17] for estimating recoil energies:

$$E_r \text{ (in eV)} \approx 4 \times 10^{-4} q^2 r^2 / (M_T E_p b^2), \quad (2)$$

with M_T the target mass in u, E_p the projectile energy in MeV/u, and b in units of a_0 , and q and r the asymptotic final charges. This estimate coincides with the formula which is obtained for simple elastic Coulomb scattering for an approximately "half-Coulomb" collision (the target being neutral on the incoming half). It originally arose, however, from Olson's parameterization [17] of some classical trajectory Monte Carlo (CTMC) calculations he had made in calculating inelastic multiple ionization cross sections for 1 MeV/u collisions of bare ions ($q = 2-20, 44$) with the rare gases, assuming a projectile velocity appreciably greater than that of the target electrons (not the case here).

For the data discussed here, this estimate underestimates recoil

ion energies by nearly an order of magnitude at an impact parameter of the L shell radius of either collision partner. What is different is the extensive shell interpenetration accompanying the collisions discussed here, in contrast to large impact parameter ionization by bare nuclei passing well outside target shell radii, the case of principal interest in Ref. [17]. One anticipates that Eq. (2) provides a plausible and useful estimate for very highly charged, fast heavy ions achieving complete or nearly complete target electron stripping of an atom of significantly smaller nuclear charge. But for a more nearly symmetric collision, or in a situation where removing nearly all target electrons efficiently is a paramount consideration -- as is likely to be the case in trapping experiments on heavy 1-, 2-, or 3-electron ions -- then Eq. (2) may underestimate the recoil energy by an order of magnitude or more. The "hammer" data in Table II and Fig. 3 indicate this trend. Ions produced using the "scalpel" method are seen to be still colder from this relative standpoint.

3. Further remarks on the "scalpel" method; relevance to future trapping experiments.

While the "scalpel" method does indeed provide substantially colder ions than the "hammer" method, the charge state reached is limited by the element-specific ionization state reached when the Auger emission cascade, and associated electron shake-up and shake-off phenomena, have subsided. At currently available synchrotron radiation intensity levels, the time needed to achieve further ionization by sequential ionization of outer shells, or a second photoionization in an inner shell in the same atom, would seem to require trapping. Estimates of ionization states reached and the times required to achieve them have been discussed in earlier papers by Jones et al. [18] who considered the case of sequential photoionization only; and by

Church et al. [19], who considered a hybrid scheme based on a combination of inner-shell photoionization plus sequential stripping from outer shells.

That our initial efforts to achieve trapping of multiply charged argon ions in charge states 2^+ to 6^+ by photoionization of argon atoms in a Penning trap have been successful is illustrated in Fig. 4. Since these data were discussed earlier in this conference by Prof. Church, there is no need for further elaboration of them here.

Instead, we conclude by comparing and contrasting various aspects of the hammer, scalpel, and sequential ionization techniques as they pertain to present and potential future measurements on high charge state ions stored in Penning traps. The latter two methods are superior to the former insofar as they produce ions close to room temperature, which in turn permits trapping at low well depths, a circumstance which obviously eases the task of potential further cooling. Since the hammer excitation method involves pumping with beams of high charge state ions, and these are awkward to transport across the axial magnetic field lines characteristic of a Penning trap configuration, the hammer method involves creation of ions over the entire axial gap between the trap end caps, following passage through a magnet yoke and pole tips as well as the end caps themselves. Pumping with x-rays instead permits radial injection, which is not only easier to achieve geometrically, but also permits creation of ions over a sub-mm range of axial distances, limiting the axial energy spread of the subsequent stored ion motion.

A hybrid photoionization scheme, where the initial step consists of a vacancy cascade initiated by an inner shell photoionization event, permits production of an initially fairly high charge stage, e. g. r typically in the range 5 - 15. One thus avoids filling the trap with

space charge from low charge states, a risk run by both the hammer and sequential ionization methods. Since high r 's are easily confined by low magnetic fields, values of $B \sim 0.05$ T may be used. Use of iron free coils is thus a likely possibility, representing very considerable savings in cost and vacuum system complexity. The combination of low-well-depth trapping and small confining fields allows one to consider a confinement volume of a few cubic mm, and a typical cyclotron radius for the ion motion of $\rho \sim 1$ mm per charge.

While low confining well depth obviously eases the problem of further cooling, for x-rays the product $\hbar\gamma$, where γ is the natural width of a suitable excited level, is generally far too large to permit direct appropriation of the laser cooling concepts familiar from cooling studies at much softer laser photon energies, as discussed several times earlier in this meeting. At least two alternative choices exist. First, "sympathetic" cooling may be tried by simultaneously confining and laser cooling low-charge ions of similar charge-to-mass, relying on collisional energy transfer to cool the high charge state species of primary interest. A second possibility has been suggested recently by Church et al. [20], who point out that although most optical transitions in highly charged ions occur in the x-ray range, selected transitions occasionally fall in the range of cw or pulsed dye laser wavelengths. For example, several transitions between fine structure levels of the ground states of multiply charged argon ions occur at wavelengths ~ 500 nm. Since for room temperature ions Doppler widths correspond to $\lesssim 1$ GHz, laser cooling on such transitions may be possible (though at relatively slow rates).

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TABLE I. Average charge state $\bar{r}$ following inner-shell photoionization. Energy of monochromatized x rays (in eV) is indicated for each mean charge.

Deepest Vacancy	Present	$\bar{r}$ (expt.) ref. 12	ref. 13
Ne K	2.3 (white)	2.3	
Ar K	4.2 (white)	4.2	
Kr L ₁	5.4 (white)	6.7	
Xe L ₁	7.8 (5475)	9.1 (8400)	7.74 (5800)
Xe L ₂	7.7 (5400)		7.65 (5450)
Xe L ₃	7.5 (5050)	7.4 (4960)	7.45 (5100)
Xe M	6.2 (4750)	6.5 (1500)	6.01 (4500)

Table II. Comparison of argon recoil ion energies (in eV) produced using unmonochromatized x rays from SSRL with those produced using 23 and 27.6 MeV Cl^{+8} beams from the ORNL EN tandem accelerator detected in coincidence with Cl^{+9} where $q=7,8$.

Argon charge	SSRL x-rays	23 MeV Cl^{+8}		27.6 MeV Cl^{+8}	
		Cl^{+7}	Cl^{+8}	Cl^{+7}	Cl^{+8}
2	0.046(20) eV				
3	0.050(23)				
4	0.066(21)				
5	0.069(35)		.41 (.12)		.21 (.09)
6	0.086(36)	.27 (.15)	.63 (.12)	<.20	.35 (.12)
7	0.092(71)	.42 (.16)	1.14 (.25)	<.40	.71 (.21)
8	0.081(56)	.84 (.24)	2.26 (.35)	.47 (.28)	1.43 (.34)
9		2.08 (.50)	4.20 (.76)	1.66 (.54)	2.74 (.78)

Figure Captions

Fig. 1. Xe charge state distribution following L-shell vacancy production using synchrotron radiation. Plotted is number of ions detected as a function of flight time. Low ion temperature is indicated by the nearly complete isotopic resolution observed.

Fig. 2(a). Spectrum of Ar^{+7} recoil ions produced by 23 MeV Cl^{+5} and detected in coincidence with Cl^{+8} . Inset shows FWHM of Ar^{+8} , Ar^{+9} .

Fig. 2(b). Quadratic behavior of the square of Ar^{+7} TOF peak widths plotted as a function of $(qr)^{-1}$ (see text).

Fig. 3(a). Dependence of E_r on product (qr) of final projectile and recoil charge states for 1-, 2-, and 3-electron loss from 23 MeV Cl^{+5} projectiles ions.

Fig. 3(b). E_r vs qr for beams of 23, 27.6 and 33 MeV Cl^{+5} . Data have been fitted to straight lines (see text), both shifted for clarity. Both scales are logarithmic.

Fig. 4. Argon ion q/m signals obtained from ions stored in a 5 V deep Penning trap pumped by white x-rays from the Brookhaven National Light Source.