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Properties of Cold Ions Produced by Synchrotron Radiation and by Charged Particle Impact

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Argon recoil ions produced by beams of 0.8 MeV/u Cl^{5+} have been detected by time-of-flight(TOF) techniques in coincidence with the loss of from one to five projectile electrons. Recoil-ion energies have been determined to be more than an order of magnitude higher than those of highly-charged ions produced by unmonochromatized synchrotron radiation. Charge-state distributions, however, show similarities, suggesting that loss of projectile electrons corresponds, in some cases, to inner-shell target ionization producing vacancy cascades. In an essential improvement to the usual multinomial description of ionization in the independent-electron-ejection model, we find the inclusion of Auger vacancy cascades significantly alters the description of the recoil ion spectra corresponding to projectile-electron loss. These conclusions are consistent with impact parameters inferred from determinations of mean recoil energy.

Beams of low-energy, high-charge-state ions, typically formed from recoils produced by swift heavy-ion impact, or in recent years, by ECR sources, have long been used to study charge-changing reactions with gas targets. As beam energies are pushed lower and lower [1], the inherent recoil energy resulting from the primary ionization may begin to limit the resolution of the low-energy secondary beam. Our measurements of energies of recoil ions indicate that highly-charged ions produced by synchrotron radiation have kinetic energies up to two orders of magnitude less than do similar charge states of recoil ions produced by charged-particle impact. The results may be applicable to development of a very cold ion source featuring low energy spread and good angular definition.

The independent-electron-ejection model has long been employed, with substantial success, to describe multiple-vacancy production in target gases by beams of energetic charged projectiles [2]. In this model, whose validity requires that the collisional velocity be much larger than the orbital velocity of the electrons being ionized, electron ejection is described by a binomial distribution of outer-shell vacancies. Extension of the model to include inner -shell ionization is accomplished by introducing an independent binomial distribution for each inner shell. In most treatments, the effect of vacancy cascades following inner-shell ionization on the final recoil-ion charge distribution is thus neglected. We have made measurements of Ar recoil-ion charge-state distributions in coincidence with nearly symmetric charge-changed projectiles at 0.8 MeV/u whose interpretation is simplified by the assumption of Auger vacancy cascades filling target L-shell vacancies, beyond the customary binomial distribution of M-shell ionization, in order to obtain good agreement with recoil-ion charge distributions[3]. Inclusion of vacancy cascades also permits improved interpretation of Ar and Kr recoil-ion charge- state distributions published recently by Müller et al [2], who obtained recoil-charge fractions for a large number of projectile-target systems at 1.4

MeV/u. Earlier non-coincidence work by Meron et al [4] discussed the contribution of Auger decay of inner-shell vacancies in low-Z projectile charge -state distributions following small-impact-parameter, large-scattering-angle collisions with light gas targets at lower energy. We note here that vacancy cascades are also very important for impact parameters ≈ the shell radii of the active electrons and that coincidence measurements can aid in separating the comparative quantitative importance of such vacancy cascades from pure Coulomb ionization.

Rare-gas TOF spectra were obtained using synchrotron x radiation produced by an eight-pole wiggler at Stanford Synchrotron Radiation Laboratory to photoionize atomic inner shells in a dilute gas sample. The experimental method and spectra for krypton and xenon have been previously reported [5]. An example of an Argon TOF spectrum is displayed in Fig. 1. The high charge states which result are due to shakeoff accompanying the Auger cascade of a single K-shell vacancy. This spectrum is in good agreement with previous measurements [6] and theoretical treatments [7]. The narrow peak widths are near the experimental time resolution and indicate qualitively the low recoil energy of the multiply- charged ions.

Using similar techniques at the Oak Ridge National Laboratory EN Tandem Facility, we have measured argon recoil-ion distributions and mean recoil- ion energies produced by beams of Cl^{5+} at 0.8 MeV/u, corresponding to a beam velocity matching target L-electron velocities, as a function of the degree of coincident projectile ionization (Fig. 2). The familiar shift to higher recoil charges with increasing projectile-electron capture, often noted in the past [1], is clearly visible here for the case of increasing projectile- electron loss. Peak widths are observed to increase as a function of final projectile charge state indicating greater recoil-ion kinetic energy associated with such processes [8].

The determination of mean recoil energy can be accomplished through study of the widths of individual charge state TOF peaks as a function of spectrometer electric field strength. In short, we assume three contributions to peak width: electronic timing uncertainty, field fringing, and recoil energy. Only recoil energy varies as the inverse square of the spectrometer electric field. A second-order-polynomial least-squares fitting procedure permits extraction of the recoil energy from the quadratic coefficient. Our method for determining recoil energies, and corresponding impact parameters, is described in more detail elsewhere [5,8], and is illustrated in Fig. 3 for 3-electron loss from Cl^{5+} on argon at 0.7 MeV/u. Note how broad the peaks are as a result of recoil energy imparted to the target by the charged projectile. Recoil energies of highly charged ions are much higher when produced by particle impact than by synchrotron radiation, as can be seen from Fig 4.

We have previously [3] compared recoil-ion charge distributions corresponding to single- and double-projectile-electron capture and loss for 0.7 MeV/u Cl^{8+} on Ar. For that system, consideration of impact parameters and velocity-matching conditions suggests that projectile- electron capture occurs primarily from the target L shell. The resultant charge distributions for single (double) capture are described well by the assumption of target Auger vacancy decays following projectile capture of one (two) target L electrons. The single and double projectile-electron loss spectra strongly resemble the single and double projectile-electron capture spectra, suggesting that loss of one or two projectile electrons is closely associated with loss of one or two target L electrons, respectively. We have made estimates of recoil-ion distributions following Auger decay of one or two L-shell vacancies in Ar assuming a binomial distribution of M- shell ionization. Charge distributions following a single L-shell photoionization have been established both experimentally [6] and theoretically [7] to peak at q=2 (P(2)=.65) with shakeoff and Coster-Kronig decays of L_1 vacancies producing a substantial component of q=3 (P(3)=.32) and q=4 (P(4)=.03). In the independent electron model, the probability of removing m of the 8 M- shell electrons in Ar, where P_M is the ionization probability of removing each electron, is the product

$$P(m) = \binom{M}{m}(P_M)^m(1-P_M)^{M-m}.$$

The time for the projectile to traverse the target ($\approx 10^{-17}$ sec), during which time all M-shell ionization is assumed to occur, is small compared to the L-vacancy lifetime ($\approx 10^{-15}$ sec). We thus suppose that the fraction of each final charge state r=q+m is just the product P(q)xP(m) while imposing the constraint that there are only (8-m) M-shell electrons remaining to be autoionized in the subsequent vacancy cascade. We make the approximation that each inner-shell vacancy decays as it would in a neutral atom but is cut off by the supply of available M electrons. We assume direct Coulomb ionization of L electrons produces at most a small additional contribution to the ionization. Measured recoil-ion charge-state fractions for loss of three to five electrons (which necessitates dipping into the L-shell) are depicted, together with estimates based on this simple model(P_M=0.5, a value consistent with results of Tonuma et al [9]), in Fig. 5b. In fact, P_M can be expected to depend on the projectile charge, higher charges producing greated M-shell ionization [9]. For the three-to-five electron-loss processes, the Auger decay of three to five inner-shell vacancies superimposed upon some M-shell ionization effectively empties the M-shell, producing charge distributions with similar shapes regardless of variations in P_M. Recoil-ion distributions resulting from the loss of one or two projectile electrons, however, exhibit differences both in peak shape and mean(Fig. 5a). The loss of a single projectile electron from Cl^{5+}, presumably from the M shell, produces a recoil-ion distribution which can be reproduced well by a binomial distribution of 8 M-shell electrons (P_M=.37).

The quasimolecular model can be invoked to help explain why the loss of L- shell electrons from the target is accompanied by ionization of the same number of L-electrons from the target. In the quasimolecular model, L-shell ionization in slow Ar-Ar collisions proceeds via excitation (with virtually unit probability) of 4fσ molecular orbitals(MO) into continuum orbitals whenever impact parameters are $\leq$ 0.5 a.u.[10] Although the quasimolecular model is of limited applicability to the higher-energy non-adiabatic Cl-Ar collisions discussed here, perhaps some MO character persists. A contribution to the loss cross section may also arise from the direct interaction between a projectile and a target electron in which both are ionized.

The present work suggests that in fast, symmetric collisions between energetic, charged ions and neutral gas targets the projectile selectively ionizes velocity-matched target electrons while producing less stripping of the outer electrons than is often assumed. The final target charge distribution follows from the subsequent Auger cascade(s). This result is in good agreement with earlier data [3,11]. The mechanism by which loss of one or more projectile L electrons could result in ionization of the same number of target L-electrons for the nearly symmetric but non-adiabatic collision discussed here is unclear, but may be related to the promotion of 4fσ orbitals into the continuum producing L-shell vacancies, as predicted by the MO model for low-energy, symmetric collisions. This work was supported in part by the National Science Foundation, and by the U. S. Department of Energy under Contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc. SSRL is supported by the U.S. Department of Energy, Office of Basic Energy Sciences, and the National Institutes of Health, Biotechnology Resource Program, Division of Research Resources.

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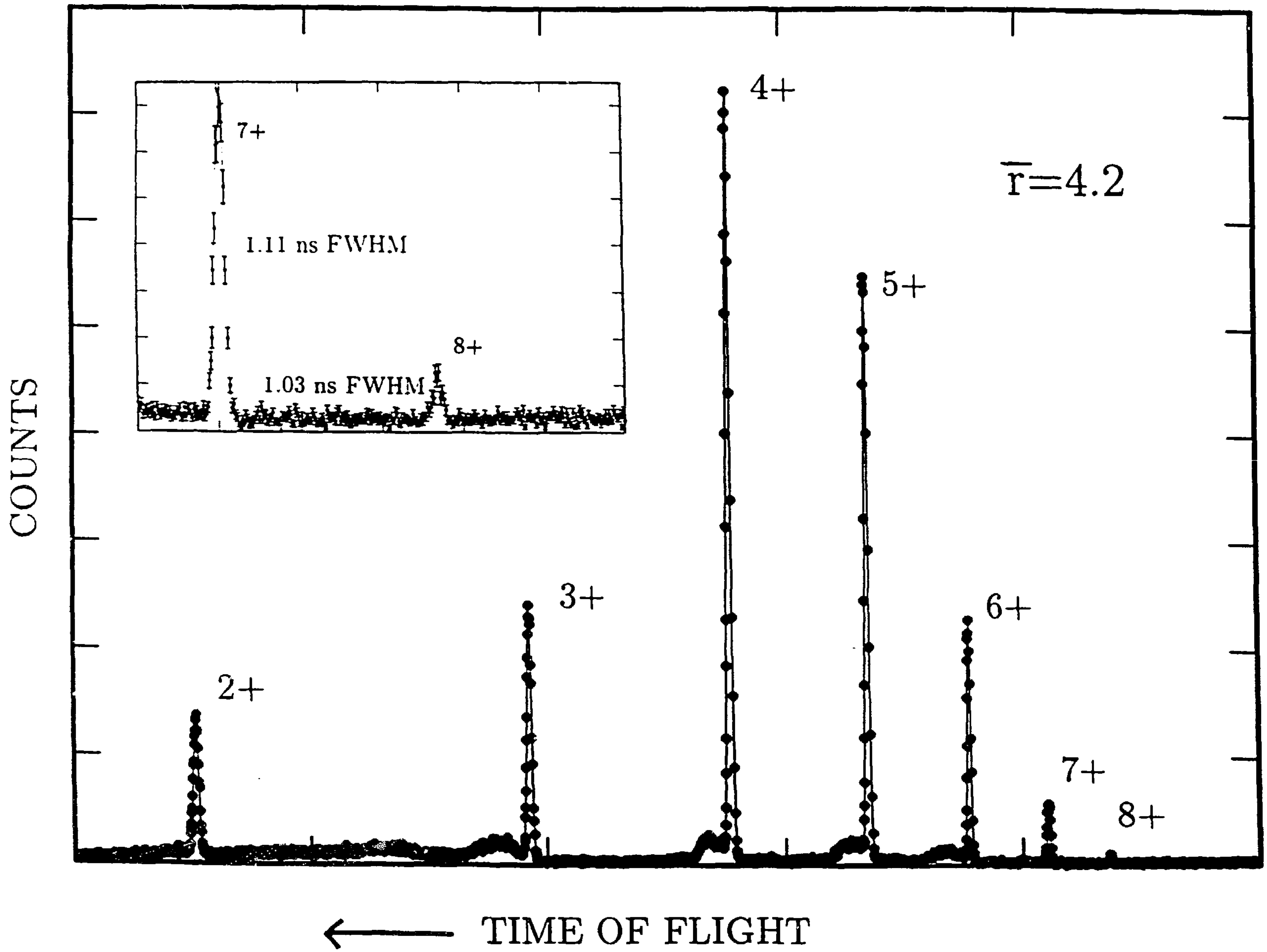
Fig. 1. Time-of-flight spectra for argon produced by vacancy cascades following K-shell photoionization by synchrotron radiation. Inset shows FWHM of Ar^{7+}, Ar^{8+}.

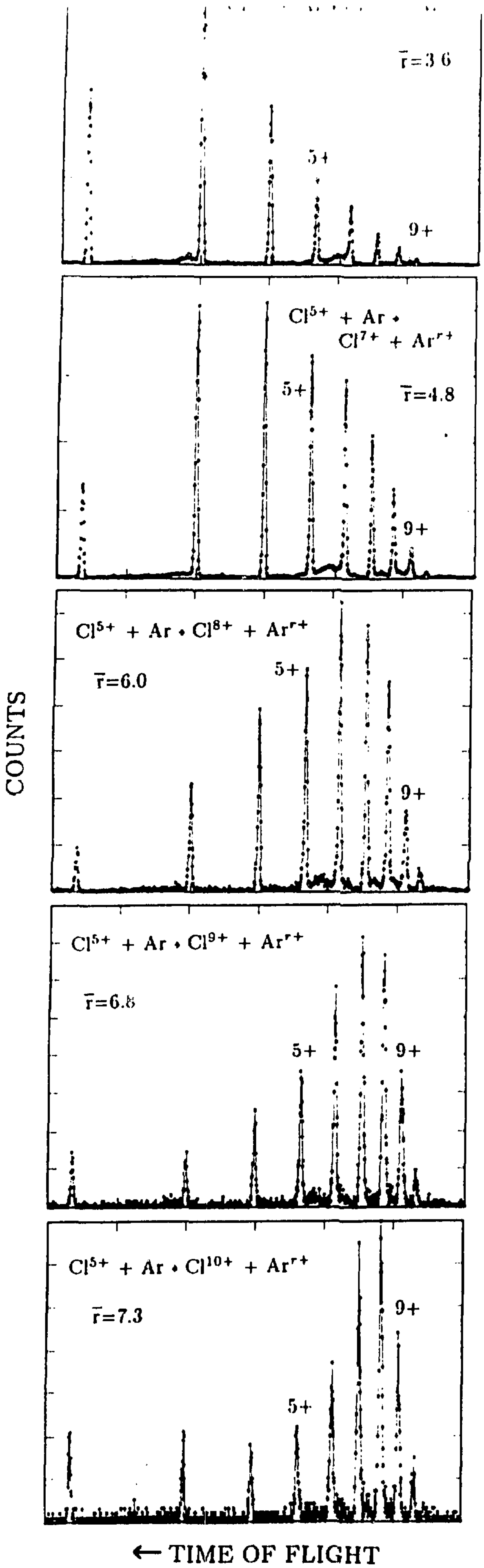
Fig. 2. Ar recoil-ion spectra detected in coincidence with (from top to bottom) loss of one, two, three, four, and five electrons from 0.8 MeV/u Cl^{5+}. Mean recoil-ion charge is indicated. Lower charge states for four- and five-electron loss are produced primarily by higher-cross-section processes involving small amounts of $Cl^{8+,9+}$ contamination in the beam.

Fig. 3. (a) Spectrum or Ar^{r+} recoil ions produced by 0.8 MeV Cl^{5+} and detected in coincidence with Cl^{8+}. Inset shows FWHM of Ar^{7+}, Ar^{8+}. (b) Quadratic behavior of the square of Ar^{r+} TOF peak widths plotted as a function of (charge x electric field)$^{-1}$.

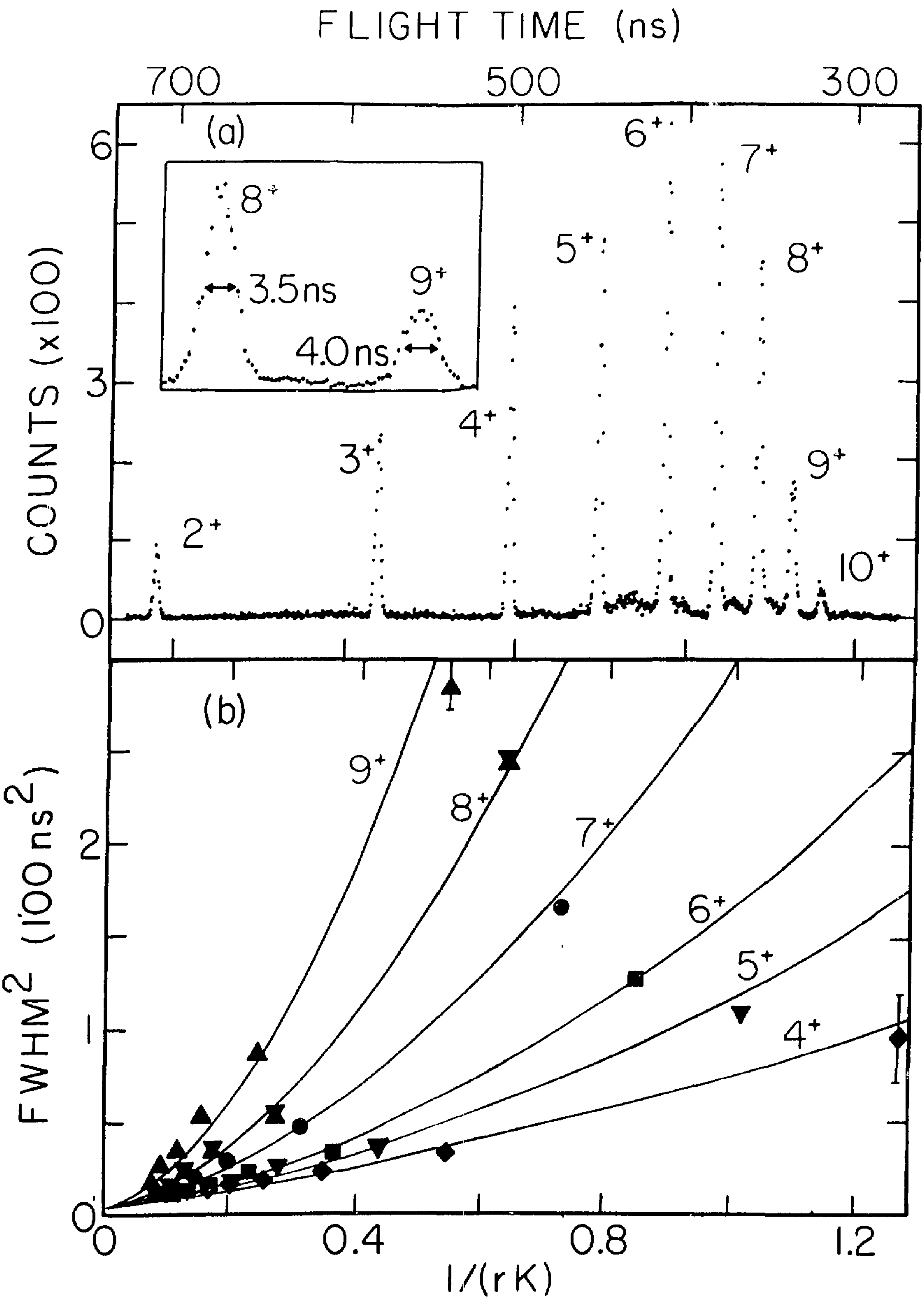
Fig. 4. Comparison of mean recoil energies of ions produced by charged particle impact and by synchrotron radiation.

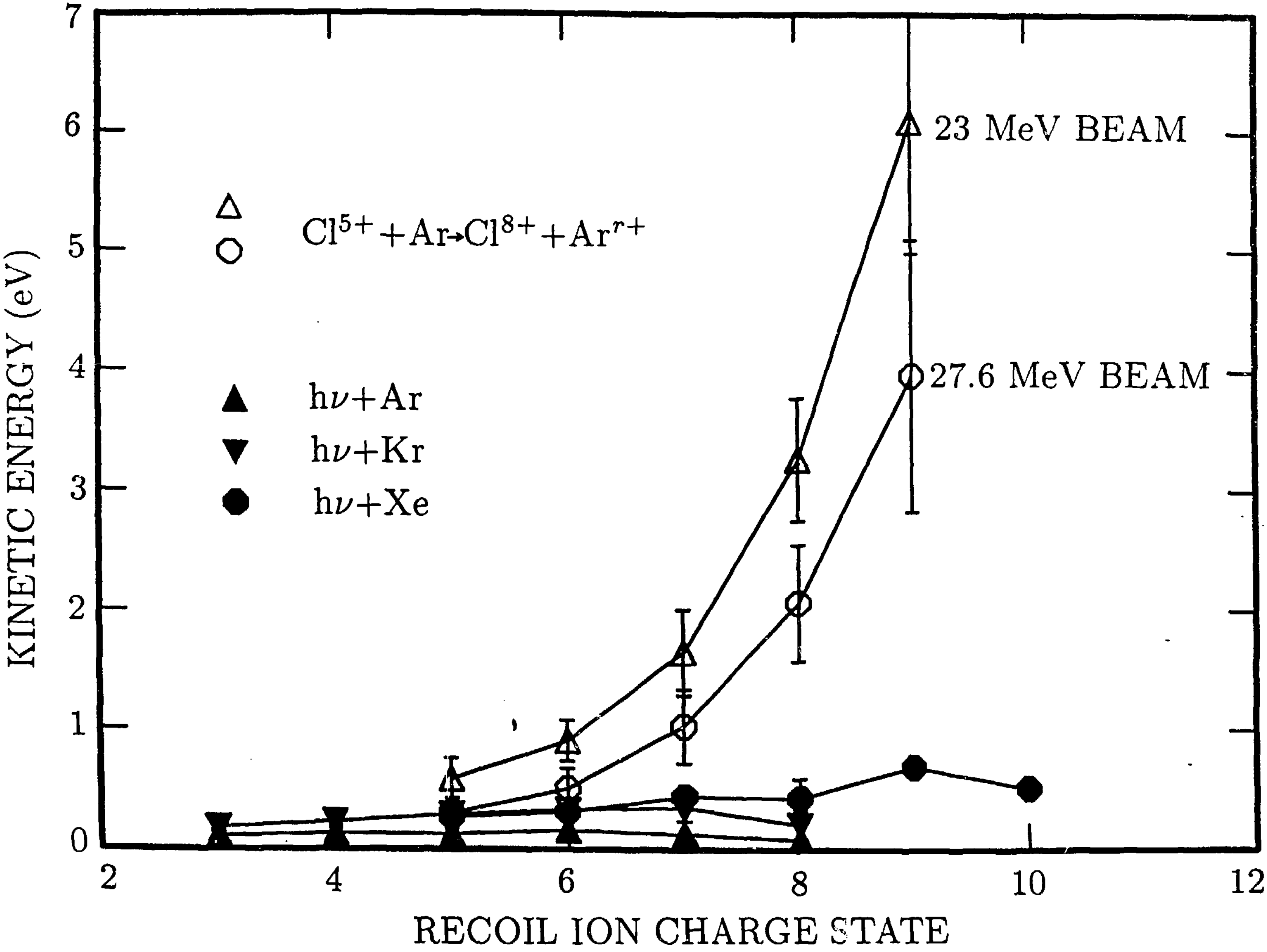
Fig. 5. Plotted as a function of charge-mean charge are argon recoil ions detected in coincidence with (a) one- and two-electron loss and (b) three-, four-, and five-electron loss from 0.8 MeV/u Cl^{5+}. The data points in (a) are connected to guide the eye. The solid line in (b) is prediction based on decay of one L vacancy accompanied by a binomial distribution of M-shell ionization by the projectile.


COUNTS
TIME OF FLIGHT
r̄=4.2
2+
3+
4+
5+
6+
7+
8+
1.11 ns FWHM
1.03 ns FWHM

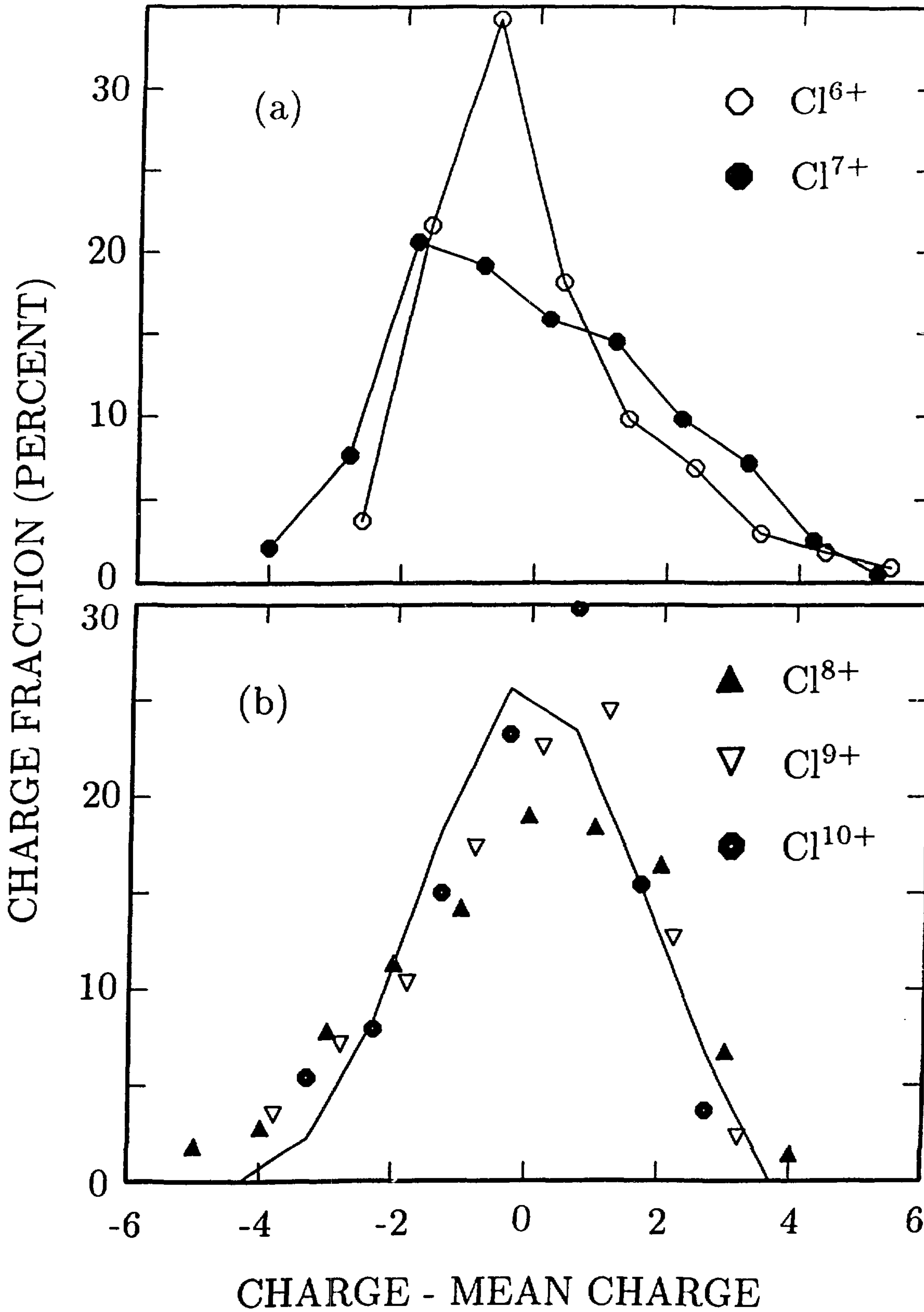


r̄=3.6
5+
9+
Cl5+ + Ar → Cl7+ + Arr+
r̄=4.8
5+
9+
Cl5+ + Ar → Cl8+ + Arr+
r̄=6.0
5+
9+
Cl5+ + Ar → Cl9+ + Arr+
r̄=6.8
5+
9+
Cl5+ + Ar → Cl10+ + Arr+
r̄=7.3
5+
9+
COUNTS
← TIME OF FLIGHT


FLIGHT TIME (ns)
700
500
300
(a)
6
3
0
COUNTS (x100)
8+
3.5ns
9+
4.0ns
2+
3+
4+
5+
6+
7+
8+
9+
10+
(b)
FWHM^2 (100 ns^2)
2
1
0
9+
8+
7+
6+
5+
4+
0.4
0.8
1.2
1/(rK)



KINETIC ENERGY (eV)
RECOIL ION CHARGE STATE
0
1
2
3
4
5
6
7
2
4
6
8
10
12
23 MeV BEAM
27.6 MeV BEAM
$Cl^{5+} + Ar \rightarrow Cl^{8+} + Ar^{r+}$
$h\nu + Ar$
$h\nu + Kr$
$h\nu + Xe$


(a)
Cl^{6+}
Cl^{7+}
(b)
Cl^{8+}
Cl^{9+}
Cl^{10+}
CHARGE FRACTION (PERCENT)
CHARGE - MEAN CHARGE
0
10
20
30
-6
-4
-2
0
2
4
6