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Akio Itoh University of Nebraska - Lincoln

M. Eugene Rudd University of Nebraska - Lincoln, erudd@unl.edu

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Recoil-ion production from zero-impact-parameter H⁺-Ar and H⁺-Kr collisions at 20-70 keV

Akio Itoh* and M. E. Rudd

Department of Physics and Astronomy, University of Nebraska-Lincoln, Lincoln, Nebraska 68588-0111

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Recoil ions emitted at 70° from H⁺-Ar and at 50° and 70° from H⁺-Kr collisions at 20–70 keV were analyzed by an electrostatic spectrometer. Ions selected in this way result from collisions at very small, essentially zero, impact parameters. Charge states up to 6 + were identified in the spectrum. The cross section for each charge state was measured. The data indicate that these close collisions produce a larger fraction of ions in high charge states than more distant collisions.

I. INTRODUCTION

The production of multiply charged recoil ions from ion-atom collisions is of considerable current interest since the mechanisms for making highly charged ions are not well understood even though such collisions have found application¹ as a source of highly charged ions for further experiments.

Pioneering work was done in this area by Afrosimov and Fedorenko² and by Morgan and Everhart.³ However, their data and much of the recent work⁴ has focused on recoils from heavy-ion impact on a variety of targets. Such studies are complicated by the fact that both of the collision partners can become charged in the collision. With proton impact the number of final charge states of the projectile is much more limited. Some cross-section data exist for low-energy protons on gases^{5,6} but these were measurements of the total production of ions as a function of charge state and projectile velocity. Since most of the recoil ions result from distant collisions, the charge distributions are characteristic of large-impactparameter collisions. Little is known about the mechanisms of production of multiply charged recoil ions in close ion impacts, even for the simplest case of proton collisions.

Hippler *et al.*⁷ have studied 300-keV proton-rare-gas collisions by detecting recoil ions of known charge states in coincidence with electrons ejected at three different angles. They have shown that the fraction of ions that are doubly and triply ionized generally increases as the energy of the coincident electron increases. Greater electron energies, of course, result from smaller-impact-parameter collisions. A statistical ionization model based on independent outer-shell ionization accounts for most of the cross section but Auger and Coster-Kronig transitions are also substantial contributers.

In this paper we report cross sections for ejection of recoil ions at 50° and 70°. Recoils at such angles result only from very close collisions, with distances of closest approach smaller than the radius of the K-shell in argon and in most cases also in krypton. These are therefore essentially zero-impact-parameter collisions. In this way, it is possible to study the rarer close collisions without their being masked by the more numerous ions from distant collisions. Argon and krypton were used as targets and the impact energies ranged from 20 to 70 keV.

II. EXPERIMENTAL METHOD

The apparatus used is similar to that used in previous measurements of electron production⁸ so only a brief description is given here. Protons from an rf ion source were accelerated and magnetically analyzed before entering the collision chamber and caught in a Faraday cup for charge integration. Ions from collisions in the static gas target ejected at 50° or 70° from the beam entered a parallel-plate electrostatic analyzer and those passing the analyzer were counted by a venetian-blind-type electron multiplier.

The proton beam had an angular full width at half maximum (FWHM) of 0.20°. Recoil ions from the collision were collimated to an angular range of 0.9° and accelerated by a 10-V potential difference before entering the parallel-plate electrostatic analyzer. Electrostatic deflection spectrometers, such as the one used, separate recoil ions according to the ratio of their energy to charge E_r/q . The resolution of the analyzer was 5.5%. The efficiency of the detector (EMI 9642/3B) was estimated from data provided by the manufacturer. Since the first dynode was at -3 kV, the impact energy on that dynode ranged from 3.2 to 12.8 keV for the various recoil energies and charge states up to 4 +. The efficiencies calculated varied between 0.60 and 0.75. At the higher charge states, the value 0.80 was estimated for the efficiency. From the measured pulse-height distribution a correction was also made for pulses lost because they were smaller than the setting of the discriminator. This efficiency was 75% at the setting used.

Data were taken at 2-6 different target-gas pressures between 0.12 and 0.70 mTorr. A Gaussian curve was fitted to each charge-state peak and the apparent cross section determined from the area under the curve. The final value for the cross section was obtained by extrapolation of the apparent cross sections to zero pressure. This procedure corrected for charge-changing collisions between the collision center and the analyzer. The small number of recoils detected at the lower proton energies and higher charge states have greater experimental fluctuations which limited the accuracy of the results in some cases.

The estimated contributions to the uncertainty of the cross sections were 8% from the measurement of pressure, 5% in the collection and integration of the beam current, and 12% from the uncertainty in the overall

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detection efficiency. Other sources of error were believed to be small except for the uncertainty in extrapolating to zero pressure mentioned above.

III. EXPERIMENTAL RESULTS

Figure 1 shows a complete spectrum of argon recoil ions from 1 to 1000 eV ejected at 70°. The cross sections have been multiplied by the recoil energy per unit charge to reduce the rapid falloff of the cross section with energy. The main goal of this investigation was the study of the "hard-collision" region which is characterized by small impact parameters and large projectile deflection angles. Such collisions produce recoil ions of high energies. These appear in the energy range above about 100 eV which includes the pronounced peaks. The "softcollision" recoils appear in the low-energy continuum up to about 100 eV. The terms "hard" and "soft" are relative only since even in the soft-collision region the inelastic energy loss Q calculated from conservation of momentum and energy is as much as 2600 eV at $E_r = 10$ eV and ranges up to a maximum of 5600 eV at $E_r = 200$ eV. A further investigation would be desirable to account for such large Q values in distant collisions.

Figure 2 shows the hard-collision region of the spectrum in more detail for three different impact energies. On the abscissas the energy per unit charge has been divided by the impact energy. This scales the spectra so that the peaks in the different spectra appear at the same place. The spectra at the three energies have been normalized so that the 2 + peaks are all of the same height. The widths of the peaks are very nearly the same with this scaling. The spectra at different energies are remarkably similar with only small variations in the relative sizes of the peaks.

The widths of the recoil peaks on the scale of $(E_r/q)/E_p$ depend on the angle and the target but are essentially independent of the projectile energy. The widths were 10% for krypton at 50°, 19% for krypton at 70°, and 16% for argon at 70°. Calculations were made of the expected contributions to the widths due to the thermal motion of the target, the acceptance angle of the analyzer, the angular spread of the beam, and the energy resolution of the analyzer. The calculated values account-



FIG. 1. Complete recoil-ion spectrum at 70° from H^+ -Ar collisions at 70 keV.



FIG. 2. Recoil-ion spectra at 70° for 20-, 45-, and 70-keV $\rm H^+$ collisions with argon. The three spectra have been normalized to the same height of the 2 + peak and have been shifted vertically for clarity. On the horizontal axis the energy per unit charge has been divided by the impact energy to bring the spectra in line. Gaussian functions have been used to fit the peaks.



FIG. 3. Recoil-ion spectra at 70° from 20- , 50- , and 70-keV $\rm H^+\text{-}Kr$ collisions. Other information as for Fig. 2.



FIG. 4. Recoil-ion spectra at 50° from 20- and 70-keV $H^+ + Kr$ collisions. Other remarks as for Fig. 2.



FIG. 5. Differential cross sections for production of ions of various charge states for H^+ impacts on argon and krypton. Lines marked "Th." are cross sections for the elastic cross section calculated by classical scattering theory.

ed for only 60-70% of the measured widths, the remainder possibly being due to the variation of Q values.

In principle, \hat{Q} values could be determined from the difference between the measured recoil energies and the recoil energies expected in elastic collisions. However, since this was not the objective of this experiment, neither the beam energy nor the recoil ion energies were measured with sufficient accuracy to allow this calculation. In fact, the measured recoil energies for krypton were somewhat higher than the expected values for elastic collisions, indicating a discrepancy of a few percent between the proton and recoil-ion energy calibrations.

The spectra from krypton are shown in Figs. 3 and 4. The 1 + peak at 50° and 70 keV is missing since it comes at an energy above the range of our analyzer. There is a noticeable shift in the position of the krypton peaks as the primary energy is varied. There is a similar but less pro-



FIG. 6. Ratios of cross-sections q + /1 + ion production vs energy for H⁺ collisions on argon and krypton. Triangles and dashed lines, present zero-impact-parameter data; circles and solid lines, total cross sections, multiplied by 4, for all impact parameters from DuBois, *et al.* (Ref. 5).

nounced shift in the argon spectrum. The cause of this shift is not known, but may be due to different distributions of Q values or to inaccuracies in our energy calibrations.

The cross sections for production of ions of various charge states are given in Table I and are plotted against impact energy in Fig. 5. The uncertainties in the measurements are shown by the error bars. Where not indicated, the uncertainty is about 15%. We also calculated differential cross sections for elastic recoils from a screened Coulomb potential using a method similar to that used by Bingham in compiling tables⁹ of classical scattering integrals. A transformation was made from the cross section per unit scattering solid angle to cross section per unit recoil solid angle. From these elastic cross sections and the sum over charge states of the measured cross sections, the ionization efficiencies were calculated

TABLE I. Differential cross sections for production of recoil ions. Units are m²/sr.

	Argon 70°			Krypton 50°			Krypton 70°		
	20 keV	50 keV	70 keV	20 keV	45 keV	70 keV	20 keV	50 keV	70 keV
1 +	1.94[-24] ^a	3.91[-25]	2.46[-25]	8.38[-25]	1.56[-25]		4.34[-24]	7.18[-25]	4.53[-25]
2 +	2.15[-24]	4.28[-25]	2.00[-25]	8.28[-25]	1.68[-25]	5.71[-26]	3.4[-24]	7.2[-25]	3.0[-25]
3 +	4.11[-25]	8.10[-26]	3.31[-26]	1.8[-26]	5.4[-26]	2.5[-26]	8.5[-25]	3.1[-25]	1.0[-25]
4 +		8.9[-27]		4.0[-26]	1.8[-26]	6.9[-27]	1.7[-25]	5.2[-26]	4.1[-26]
5 +				1.8[-26]	2.6[-27]	2.6[-27]	7.1[-26]	1.6[-26]	1.4[-26]
6+						1.2[-27]			
Sum	4.5[-24]	9.1[-25]	4.8[-25]	1.9[-24]	4.0[-25]	1.5[-25] ^b	8.8[-24]	1.8[-24]	9.1[-25]
Th.°	1.1[-23]	1.9[-24]	9.8[-25]	5.3[-24]	1.2[-24]	5.1[-25]	3.3[-23]	6.5[-24]	3.4[-24]
IE ^d	0.41	0.48	0.49	0.36	0.34	0.29	0.27	0.28	0.26

^aThe notation 1.94[-24] stands for 1.94×10^{-24} .

^bEstimated by setting $\sigma_{1+} = \sigma_{2+}$.

°Theoretical calculation of the elastic cross section.

^dIonization efficiency.

for each case and are given in Table I. While it might have been expected that collisions at such small impact parameters as these would have high ionization efficiencies, the results are between 25% and 50%. A possible explanation is that the detector efficiency for ions is lower than indicated in the manufacturer's literature.

Figure 6 shows the ratios σ_{q+}/σ_{1+} plotted versus projectile energy. The data for different angles did not vary appreciably and therefore were averaged for this comparison. These results for zero impact parameters are compared with the corresponding data of DuBois *et al.*⁶ for all impact parameters. The latter data have been multiplied by a factor of 4 to facilitate comparison. While the shapes of the curves are similar, the fraction of recoils in multiple charge states is higher by a factor of 4 or more for the close collisions than for the more distant ones which dominate DuBois's data.

Multiple ionization may be caused by the direct ejection of two or more electrons, by the capture of two or more electrons, or by a combination of capture and electron

*Present address: Rigaku Corp., Akaoji 14-8, Osaka 569, Japan.
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ejection. In all three cases the probability of multiple ionization should be greater for impacts at smaller impact parameters because the path length of the projectile in the electron charge cloud is greater.

Another mechanism for producing multiple ionization is the ejection of an inner-shell electron followed by one or more Auger transitions. An initial hole in the K shell of a multishell atom such as krypton can result in an Auger cascade with each transition resulting in the ejection of another electron. In this way a charge state of 4 can be reached in krypton. The production of initial K-shell vacancies is more probable in the close collisions in this experiment than in more distant ones. This offers a likely explanation of the relatively large production of 4 + ionsin krypton shown in Fig. 6.

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