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LETTER TO THE EDITOR

Projectile- and target-charge dependent effects in ionizing collisions of H^+ and He^{2+} with He, Ne and Ar atoms

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Abstract. The spectra of electrons emitted in collisions between H^+ and He^{2+} projectiles and He, Ne and Ar targets at energies of 50 and 100 keV amu^{-1} have been studied. The data are in qualitative agreement with results of Irby *et al*, but are in disagreement with recent measurements of Bernardi *et al*. It is shown that the observed electron spectra have a dependence on both target-ion and projectile effective charge that can be understood qualitatively in terms of 'saddle-point' ionization. Several issues relevant to saddle-point ionization are discussed.

When protons of intermediate speed ($1\ \text{au} \leqslant v_P \leqslant 3\ \text{au}$) ionize atoms, a large fraction of the ejected electrons are closer to the midpoint between the two nuclei than to either the proton or the target nucleus (Olson *et al* 1987). The Coulomb forces of each of the nuclei on these electrons are comparable. Thus, 'two-centre' effects (Meckbach *et al* 1986, Stolterfoht *et al* 1987) cannot be viewed as a perturbation of the collision process; they dominate it at intermediate energies. Classically one might expect that some fraction of these electrons, and especially of those ejected in the forward direction, will be ionized by a 'saddle-point' mechanism, in which a target electron with a velocity of about $\frac{1}{2}v_P$ finds itself near the saddle region of electrostatic potential between the two nuclei. Feeling no strong force in this region, its velocity changes little, and it 'drifts' out of the target-atom region without being captured by the receding proton (see, e.g., Bandarage and Parson 1990 and references therein). By changing the charge of the incident projectile in collisions of this type, the position and velocity of the saddle point is changed, and one would expect to see corresponding changes in the forward-ejected electron spectra to the extent that the saddle-point mechanism is important.

Evidence for such a projectile-charge dependent effect has been reported by Irby *et al* (1988). In their experiment, the velocity spectra of electrons ejected at 17° with respect to the projectile-beam direction were measured when H^+ and He^{2+} nuclei bombarded He. They found that the peak in the velocity spectrum shifted to lower values when, for a given projectile velocity, He^{2+} replaced H^+. This change was attributed to a corresponding shift in the saddle-point speed, given by

$$v_{sp} = \frac{v_P}{1+(q_P/q_T)^{1/2}} \tag{1}$$

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where v_P is the projectile speed, and q_P and q_T are the effective nuclear charges of the projectile and target, respectively. An increase in q_P thus reduces the saddle-point velocity. Irby *et al* (1988) found that for projectile velocities corresponding to 100 keV amu^{-1}, the electron-velocity spectra peaked at about 0.7 v_P and 0.6 v_P for incident H^+ and He^{2+} respectively, whereas at 60 keV amu^{-1}, the peak shifted from 0.8 to 0.6 v_P. Since q_T has a value of +1 (assuming single ionization) only for large separations between the ejected electron and the residual ion, one might expect the electronic spectra to exhibit similar target-dependent saddle-point shifts as well.

To further study the projectile-charge dependence of ejected-electron spectra, and to investigate possible target effects, we have made new measurements of forward-angle electron velocity distributions with H^+ and He^{2+} projectiles and He, Ne and Ar targets. Our data confirm the 'saddle-point' shift reported by Irby *et al*, and are in qualitative disagreement with the recently reported results of Bernardi *et al* (1989, 1990).

The present measurements were made using an apparatus substantially different from that of Irby *et al*. It has been described extensively in the literature (Rudd *et al* 1966, Cheng *et al* 1989 and references therein). Beams of H^+ and $^3He^{2+}$ were extracted from an RF ion source, accelerated, and mass analysed. Upon entering the target chamber, they were collimated by apertures biased for secondary electron suppression, and traversed a 12 cm diameter cylinder filled with the target gas before being stopped in a deep, guarded Faraday cup. Electrons emitted from the observed interaction region at angles of 10° or 20° from the beam direction passed through solid-angle defining slits, were energy analysed in an electrostatic parallel-plate analyser, and detected by an electron multiplier. The data were taken automatically by a computer. Beam current and target pressures varied insignificantly during individual runs.

In order to compute absolute differential cross sections from the raw data, a number of corrections were required. Background gas and slit-scattering count rates were typically 3% of the signal rate at 10°. At 20°, these rates amounted to 4% for H^+ projectiles and 10% for He^{2+}. The target-pressure dependence of the spectra was studied carefully. The pressure was measured using a temperature-corrected and stabilized capacitance manometer. Electrons emitted from the interaction region can be scattered from their original trajectories by target gas and not be detected. Thus, a total scattering 'absorption' correction was made using known cross sections (Buckman and Lohmann 1985 and references therein). This correction, which depends on electron energy, was less than 3% for He targets on all runs, but was occasionally as high as 7% for Ar targets. Target pressures of all three gases were varied between 0.6 and 0.1 mTorr. For He and Ne targets, the uncorrected raw data exhibited no pressure dependence. The Ar raw spectra exhibited ~10% systematic differences over this pressure range. These differences were completely eliminated, however, upon application of the absorption corrections. This gives us confidence that the corrections were applied properly.

Incident projectiles can capture electrons from gas in the beam line and target chamber. Thus, the Faraday cup reading will differ from the actual beam current responsible for target ionization. Moreover, some fraction of the detected electrons will have been produced by projectiles other than H^+ or He^{2+}. Using known pressures and electron-capture cross sections, and by measuring the change in Faraday cup current when target gas was admitted to the chamber, we have estimated the fractions of H°, He^+ and He° in the beam at the centre of the target volume to be at most 4%, 5% and 1%, respectively, for the worst case of Ar targets and 50 keV amu^{-1} projectiles. In all other cases, charge-transfer contaminants were less than 2% of the beam. While

these effects must be accounted for in determining absolute cross sections, we note again that the shape of the velocity spectra exhibited no pressure dependence that was not removed by the absorption correction. Thus, the effect of projectile contaminants in altering spectral shapes is negligible.

The uncertainty in the absolute cross sections reported here, including uncertainties in detector efficiency, solid angle acceptance, analyser resolution, target pressure and beam current, as well as the statistical counting uncertainty, is about 30%. For electron energies below about 10 eV, the uncertainty is about 50%. The relative values of cross sections for H^+ and He^{2+} projectiles, however, should be accurate to about 10% at all electron energies.

Several velocity spectra from our measurements are shown in figure 1. They are compared with the data of Bernardi *et al* (1989, 1990). A few comments should be made at this point with regard to the way we have chosen to present these results. Differential electron-ejection cross sections have been reported in the literature in three forms: $d\sigma/d\Omega\, dE$, $d\sigma/d\Omega\, dv$ and $d\sigma/d\boldsymbol{v}$. These are simply related by

$$\frac{d\sigma}{d\Omega\, dv} = mv\frac{d\sigma}{d\Omega\, dE} = v^2\frac{d\sigma}{d\boldsymbol{v}} \tag{2}$$

where m is the electron mass. While the information content of any of these is essentially the same, we feel that plots of $d\sigma/dv\, d\Omega$ best elucidate the physics of the electron-ejection process by showing directly the distribution of electron speeds for a given direction and solid angle of emission. Values of $d\sigma/d\boldsymbol{v}$, on the other hand, are kinematically enhanced at small speeds due to the increasingly large number of emission directions encompassed in a unit-volume element of velocity space. In this regard, we emphasize that the mid-velocity maxima seen in figure 1 are a dynamical signature of the ionization physics, and not kinematic artefacts of plotting the data as $d\sigma/dv\, d\Omega$. In a very real sense, plots of $d\sigma/dv\, d\Omega$ give a truer picture of the global ionized-electron distribution than do those of $d\sigma/d\boldsymbol{v}$. One might be tempted to conclude, e.g. from the $d\sigma/d\boldsymbol{v}$ plot in figure 2 of Meckbach *et al* (1986), that most ionized electrons are to be found in close proximity to either the receding projectile or the residual ion. In fact, practically the opposite is true; most of the electrons emerge in a large, broad maximum roughly midway between the two positive charge centres (Olson *et al* 1987).

Finally, we note that the presence of 'saddle-point' maxima at forward angles of emission and the projectile-charge dependent shifts of these maxima can be seen clearly in graphs of $d\sigma/dE\, d\Omega$ as well as those of $d\sigma/dv\, d\Omega$ (see, e.g., Gay *et al* 1988, figure 4). Moreover, plots of $d\sigma/dv\, d\Omega$ do not always yield mid-velocity maxima; if $d\sigma/dE\, d\Omega$ falls faster than $E^{-1/2}$ near the origin, $d\sigma/dv\, d\Omega$ will monotonically decrease in this region as well. An example is shown in figure 2, where the velocity spectrum of electrons emitted at 10° following electron impact ionization of He is displayed. The lack of a saddle-point maximum is obvious and unsurprising, given the charge of the projectile.

Figure 1 illustrates the significant discrepancies between our data (which are in qualitative agreement with that of Irby *et al* (1988)) and those of the Bariloche group. We have considered possible causes of these discrepancies and, lacking detailed knowledge of the Bariloche apparatus, can suggest only one possibility: the large projectile-charge-dependent shifts which we and Irby *et al* have observed could be explained if, in both the Rolla and Nebraska experiments, a significant contaminant of lower-velocity ions was present in the projectile beam. The only mechanism for such a contaminant to be introduced would be collisional dissociation of H_3^+ after

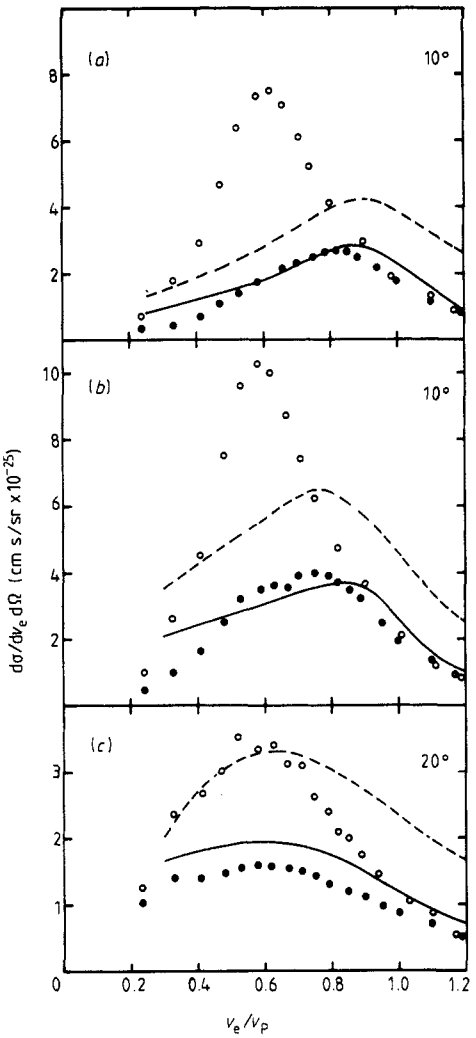

Figure 1. Doubly differential velocity cross sections for projectile energies of 100 keV amu^{-1} for (*a*) Ne and ((*b*) and (*c*)) He. The velocity scales are normalized to the projectile velocity (2 au) in all cases. Full curves represent the H^+ projectile data of Bernardi *et al* (1989, 1990); broken curves represent their $^3He^{2+}$ results. The present H^+ and $^3He^{2+}$ data are indicated by full and open circles, respectively. In the (*b*) and (*c*) the H^+ data have been multiplied by two. In (*a*) our $^3He^{2+}$ results have been multiplied by 0.5. The relative experimental results of Bernardi *et al* have been placed on an absolute scale by normalization to their theoretical calculations.

acceleration but prior to magnetic selection. Resulting H_2^+ ions would be bent into the target beamline at a magnetic field only 6% lower than that needed for $^3He^{2+}$, but their velocity would be 0.71 times that of the $^3He^{2+}$ ions.

We do not believe that such contamination is a serious problem in our work for several reasons. In both the Rolla and Nebraska experiments, beamline pressures and lengths were such that at most 0.4% of the H_3^+ produced in the source would be converted to H_2^+ upstream of the magnetic bend (Berkner *et al* 1973, Williams and

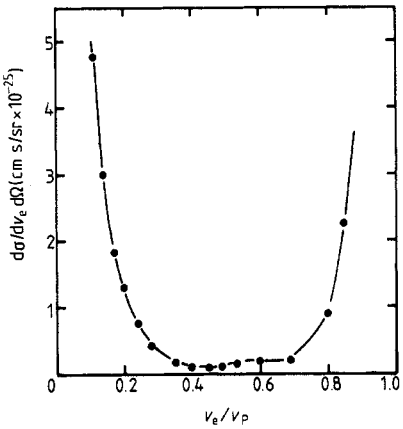

Figure 2. Doubly differential 10° velocity cross sections for ionization of Ar targets by 250 eV electrons (Rudd and DuBois 1977).

Dunbar 1966). The Rolla experiments were performed using a microwave ion source with a mixture of ^{3}He and Ar gas. Direct H_3^+ currents were always well below 1 nA. This corresponds to at most 4 pA of contaminant H_2^+, while typical $^3He^{2+}$ beams were greater than 1 nA. The Nebraska runs were made with mixtures of ^{3}He and H_2 in an RF source, and maximum H_3^+ currents of at most 10 nA (corresponding to 0.04 nA of H_2^+), compared with $^3He^{2+}$ currents greater than 1.7 nA. In one test, pure H_2 was used in the source and an 80 nA current of H_3^+ was observed. No contaminant H_2^+ was seen at the level of 20 pA. Given the above observations, the fact that both magnetic analysers had sufficient resolution to separate the two peaks, and the good agreement between the Nebraska and Rolla data, taken with different sources and gas mixtures, we must conclude that the velocity spectral shifts we observe are not due to slow H_2^+ contamination. This leaves unanswered the question of the disagreement with Bernardi *et al.* We note that it is unlikely that the Argentina experiments suffer from significant H_2^+ contamination. They observe an electron capture to the continuum cusp due to He^{2+} at 0° which has, to within 1.5%, the correct energy value.

Since the saddle-point position depends on q_T as well as q_P, one might also expect target-dependent shifts in the velocity-spectral maxima for a given projectile. To illustrate this idea, we have computed, using Slater's rules (Eyring *et al* 1964), crude estimates of q_T of the ionic cores, as seen by a valence electron. These values are 1.7, 2.93 and 2.25 for He, Ne and Ar, respectively. While this procedure ignores the fact that q_T is actually a function of distance between the ionized electron and the ion (with an asymptotic value of 1), a more elaborate calculation is probably not justified, given the qualitative nature of our saddle point arguments. Figure 3 summarizes the 10° data at both 50 and 100 keV amu^{-1}. The error bars result from uncertainty in the peak positions of the velocity spectra. Shown also is the geometric result (equation 1) for the saddle-point velocity. We note that the monotonic decrease, within experimental uncertainty, of the normalized peak positions with increasing ratios of q_P/q_T, in qualitative accord with (1). Interestingly, the zero-range potential calculations of Burgdörfer *et al* (1988, 1990) predict a qualitatively similar dependence of the velocity-spectral maximum on the charge ratio q_P/q_T. The decrease occurs either for a given projectile or a given target. Neon target spectra, for example, tend to exhibit peak positions at the highest velocities for a given projectile charge because they have the

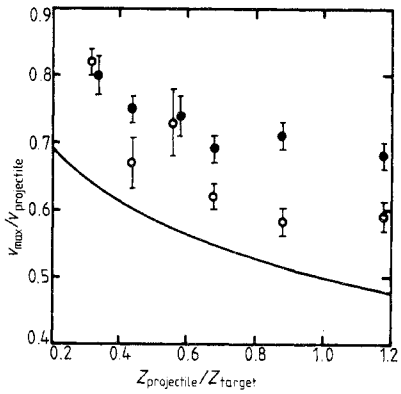

Figure 3. Ratios of the velocity corresponding to the peak in $d\sigma/dv_e\, d\Omega$ to the projectile velocity, v_P, at 10°, as a function of q_P/q_T (see text). Open circles are for 100 keV amu^{-1} projectiles; full circles are for 50 keV amu^{-1} projectiles. The full curve is the function given by equation (1). For a given projectile velocity, the data from left to right correspond to target/projectile combinations of H^+/Ne, H^+/Ar, H^+/He, He^{2+}/Ne, He^{2+}/Ar and He^{2+}/He, respectively.

largest q_T. The He^{2+} peak values are systematically below those for H^+ projectiles. Thus, our data are described comprehensively in a qualitative way by the simple saddle-point picture. More detailed calculations are obviously required to give quantitative information about individual velocity peak positions.

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