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DIFFERENTIAL CROSS SECTIONS FOR ELECTRON EMISSION IN HEAVY ION-ATOM COLLISIONS

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INTRODUCTION

When a fast charged particle passes through matter, the primary manner in which energy is lost is through ionizing collisions. Ionized electrons are ejected from the target atom or molecule and if the projectile possesses bound electrons, they too may be released as free electrons by the interaction. Investigation of the energy and angle of emission of ionization electrons provides detailed information on the interaction processes. Analysis of cross sections for electron emission enables evaluation of the significance of screening, projectile stripping, and continuum-charge-transfer as well as the applicability of Z^2 scaling in ionization by heavy charged particles. Electron spectra also provide evidence of atomic structure effects through autoionization and Auger transitions.

For collisions of heavy ions, it is common to refer to three main classes of excitation and ionization phenomena: coulomb ionization, which is most important at relatively high incident velocities; charge transfer processes, which can, when exothermic, be important even at very low collision energies; and shell interpenetration effects, which are especially important at intermediate energies. The high energy coulomb ionization process is perhaps the most well developed in terms of theoretical understanding. Theoretical calculations based on semiclassical and plane wave Born approximations have been relatively successful for inner shell ionization and for outer shell ionization by fast structureless ions.[1-3] There remains the question as to the projectile energy necessary to qualify as high energy and the relevance of these approximations to ions posessing electronic structure. The definition of high energy is further complicated by competition between coulomb processes and ionization resulting from interpenetration of electronic shells.

For low energy collisions, where the velocity of the collision is small compared to the orbital velocity of the participating bound electrons, the collision may be studied as a diatomic molecule using the Born-Oppenheimer method.[4] In this method, the system is treated using the full Hamiltonian with the internuclear kinetic energy removed and then treating the latter as a perturbation. This procedure is well suited for the study of very low energy colliding systems; however, as the kinetic energy is increased, it is necessary to consider translational factors to account for the translational kinetic energy of the electrons bound to the projectile. In addition, this method of describing the collision involves the interaction of potential energy curves for specific electronic states which has limited the application to inner shell excitation where the number of interacting levels is small. This report will not attempt to review the extensive literature regarding inner shell excitation in heavy ion collisions since several excellent reviews have been published.[1,5-10]

In this report, we will explore the systematics of differential ionization cross sections for incident charged particles which possess atomic structure. Although little has been published regarding outer shell ionization by structured projectiles, sufficient data are becoming available to provide some insight into the collision process. Cross sections differential in electron emission energy and angle were reported by Cacak and Jorgensen[12] for 50 to 300 keV Ne^+-Ne and Ar^+-Ar collisions, for 30 MeV O^{+n} collisions by Stolterfoht, et al.,[13] for 0.6 to 1.5 MeV H_2^+-H_2 collisions by Wilson and Toburen,[14] and we have recently measured cross sections for incident helium and carbon ions in the energy range 0.3 to 4 MeV. This discussion will concentrate on systematics of emission cross sections for the intermediate and high energy projectiles where we can test the applicability of high energy approximations and illustrate the dramatic changes in emission spectra observed as the projectile energies decrease. Since heavy ion interactions are important in many research fields, it is particularly useful to determine where simple methods of cross section scaling may be appropriate.

HIGH ENERGY IONS

For sufficiently fast incident ions, the principle features in ejected electron spectra can be readily identified in terms of relatively simple mechanisms of ionization. These features are illustrated in Fig. 1 for collision systems which vary from ionization of a simple molecular target by fast protons to ionization of molecules containing inner electronic shells by a multi-pli-charged projectile carrying bound electrons. The spectra shown in Fig. 1 were obtained by multiplying the emission cross section by the respective ejected electron energy and plotting vs the electron energy. One can interpret these cross sections as the probability per unit energy (eV) for the transfer of energy from the projectile into kinetic energy of ionized electrons. This presentation is convenient for display of emission cross sections since the dynamic range is much smaller than the corresponding ionization cross sections. The spectra shown in Fig. 1 illustrate how increasing the complexity of the collision system adds new features to the emission spectra. The simplest spectrum shown in Fig. 1 is for ionization of molecular hydrogen by fast protons. This data, from work at our laboratory,[15] can be described as containing essentially two basic features: a low energy electron peak resulting from soft glancing collisions (large impact parameters), and a high energy electron peak resulting from direct binary encounter collisions between the incident proton and a bound electron (small impact parameters).

Relatively simple collision systems such as those involving incident fast protons can be treated with a good deal of success both qualitatively and quantitatively by high energy approximations such as binary encounter theory and the Born approximation,[2,6,7] The results of ionization of molecular oxygen by protons[15] shown in Fig. 1B illustrate the effect of adding complexity in the target molecule. The peak which appears at about 500 eV is the contribution from Auger electrons ejected as a consequence of inner shell vacancies produced in the target atom. Another effect of additional electronic structure in the target atom or molecule is broadening of the binary encounter and low energy peaks in the electron spectrum relative to the corresponding spectrum for ionization of molecular hydrogen. The designation (T) following the identification of spectral features in Fig. 1 refers to electrons originating from the target, whereas the designation (P) refers to electrons arising from the moving projectile.

One of the simplest examples of spectra which include electrons originating from the projectile is shown in Fig. 1C for ionization of molecular hydrogen by H_2^+.[14] The electron bound to the projectile enters into the

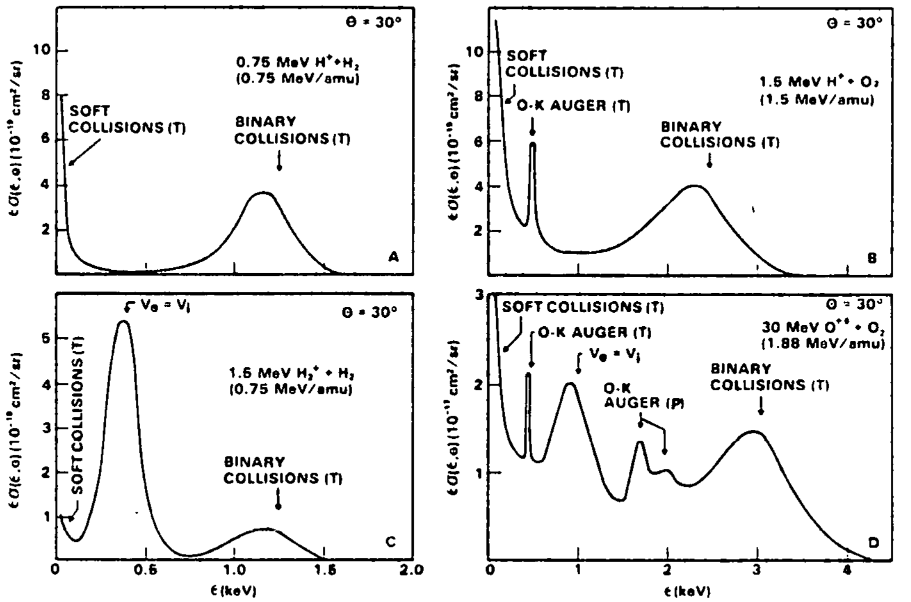

Fig. 1. Cross sections times electron energy for ejection of electrons with an emission angle of 30° in collisions of (A) 0.75 MeV $H^+ + H_2$; (B) 1.5 MeV $H^+ + O_2$; (C) 1.5 MeV $H_2^+ + H_2$; and (D) 30 MeV $O^{+5} + O_2$. The H^+ and H_2^+ data are from our laboratory; $H^+ + H_2$, Ref. 15; $H^+ + O_2$, Ref. 16; and H_2^+, Ref. 14; and the oxygen ion data are from Stolterfoht, et al., Ref. 13.

collision dynamics in several ways. For example, it screens the projectile charge in collisions involving large impact parameters reducing the relative yield of low-energy electrons and, most notably in Fig. 1C, the stripping of this electron from the H_2^+ ion contributes a very strong peak to the spectrum of ionized electrons. The peak contributed by electron loss from the projectile is observed at an ejected electron energy corresponding to $v_e = v_i$, where v_e is the ejected electron velocity and v_i is the incident ion velocity. This electron loss peak is also a prominent feature of the electron spectrum resulting from ionization of molecular oxygen by fast O^{+5} ions.[13] The ionized electron spectrum resulting from O^{+5} impact shown in Fig. 10 exhibits all of the features of the simpler collision systems plus contributions from Auger transitions which originate from inner shell ionization of the projectile. The energy of these Auger electrons exhibits a kinematic (Doppler) shift to nearly 2 keV when observed at 30° in the laboratory reference frame.

The purpose of presenting spectra for various high energy collision systems is to illustrate the features common to each. From these spectra, one has confidence that ionization mechanisms can be identified, at least in a qualitative sense. The next question to consider is to what extent one can quantitatively understand these features which have been identified. For incident protons, a great deal of experimental and theoretical work has been performed and a high degree of success has been demonstrated in quantitative understanding of the collision cross sections.[2,3] Our knowledge of cross sections for structured projectiles is far less advanced. For a very simple system such as ionization of molecular hydrogen by H_2^+ ions, we found that we could separate components of the electron spectra into a contribution of electrons which were stripped from the projectile and an underlying continuum similar to that observed in ionization of molecular hydrogen by protons. The data in Fig. 2 illustrate the similarity of proton and H_2^+ spectra above and below the H_2^+ electron loss peak. Here we simply multiply the cross sections for equal velocity protons by a factor of 2 to obtain the contribution to the H_2^+ spectra for target ionization. The two nuclei of the H_2^+ ion are assumed to act independently in ionization of the target molecule. The effect of screening these nuclei by the bound electron is apparent for ejection of low energy electrons but excellent agreement occurs between H_2^+ and scaled proton spectra for high energy ejected electrons. Using proton spectra as an estimate for target ionization, electron stripping contributions can be determined with a reasonably high degree of accuracy. One may expect because the translation kinetic energy of the H_2^+ electron is large compared to its binding energy that the electron stripping cross sections will be similar to those obtained from scattering free electrons. The insert in Fig. 2 shows data from our measurements[16] where indeed the electron yields were found in excellent agreement with the elastic electron scattering data of Williams.[17] It should be noted that similar analysis for electron loss by H^- and He^+ ions with comparable ion energies and electron emission angles less than 10° has not resulted in agreement with the elastic scattering model.[18,19]

As we consider slightly lower energy projectiles, the difficulty of determining the target continuum background becomes an increasing problem in separating projectile and target ionization. In Fig. 3, spectra are shown for ionization of argon by equal velocity He^+, He^{++}, and H^+ ions. The equivalent velocity proton data have been multiplied by 4 in accord with Z^2 scaling for comparison to the helium ion results. In these spectra for ejection of electrons with an emission angle of 15°, it is apparent that the spectra for bare charged particles do not provide an adequate guide for determination of the ejected electron continuum underlying the electron loss peak. The relative yield of low-energy electrons ejected from the target is considerably reduced for He^+ over the bare charged particles because of the screening effect of the projectile electron. Since the ejection of low-energy electrons occurs predominately through large impact parameters, one can expect screening to become increasingly important as the ejected electron energy decreases. The relative cross

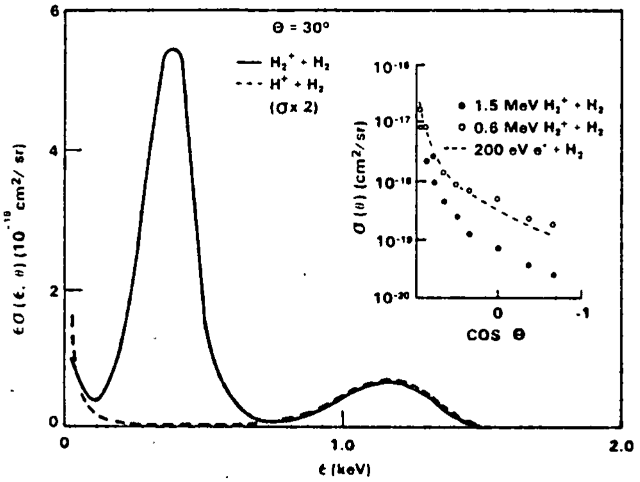

Fig. 2. Electron emission in H_2^+ and H^+ collisions with H_2. The insert compares the electron loss cross section obtained by integrating the loss peak with respect to ejected electron energy with measurements of elastic scattering of electrons. The electron scattering data are from Ref. 17.

sections for bare charged particles are also seen to be different whereas high energy approximations would predict similar spectral shapes differing quantitatively by the factor Z^2. Since spectra for bare charged particles do not provide adequate estimates of the shape of the emission spectra for target ionization, the best one can do is draw a reasonable guess of the He^+ target ionization continuum. Using such an estimate, the projectile stripping contribution obtained differs by an order of magnitude from what would be expected from scattering free electrons. From this, we can conclude that either our separation of target and projectile ionization was inaccurate or, more likely, the incident ion is moving too slow for this approximate high energy estimate to be valid.

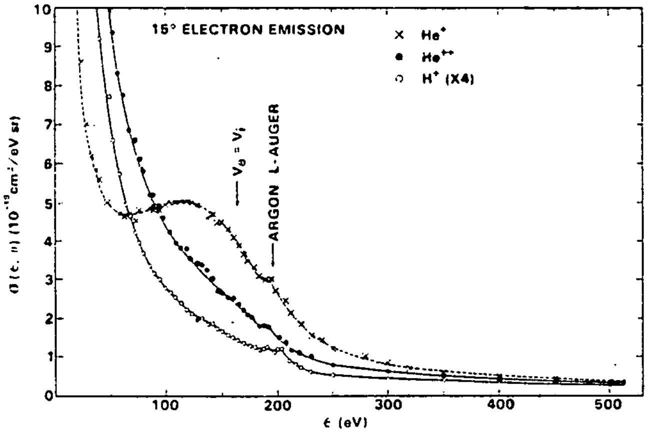

Fig. 3. Cross sections for ejection of electrons from argon by 0.3 MeV/amu He^+, He^{++}, and H^+ ions. The proton data have been multiplied by 4 in accord with Z^2 scaling for comparison to the helium ion results.

INTERMEDIATE ENERGY IONS

To this point, we have discussed what are normally considered high energy collisions; where ion velocities are several times the velocity of the bound electrons participating in the collision. We have seen that although a quantitative description may be dificult to obtain, basic spectral features follow as expected from extrapolation of results for simple collision systems using estimates based on high energy atomic collision theory. As we consider lower energy projectiles, the spectral features change dramatically. The results shown in Fig. 4 compare the ejected electron spectra and yields for ionization of molecular hydrogen by 1.3 MeV singly charged carbon ions and approximately equal velocity protons. Note that the proton data of Rudd, et al.[21] have been multiplied by 5 in order to compare spectral shapes on this linear scale indicative of the large difference in the cross sections of ionization by these two incident ions. Although the ion velocity is only about a factor of 5 lower than the ions shown in Fig. 1 and is still about twice the velocity of the target electrons, there is essentially no similarity between the spectra for H^+ and C^+ impact. Arrow mark the positions where one might expect to see features in the spectra associated with ionization mechanisms of the type discussed above for fast charged particles.

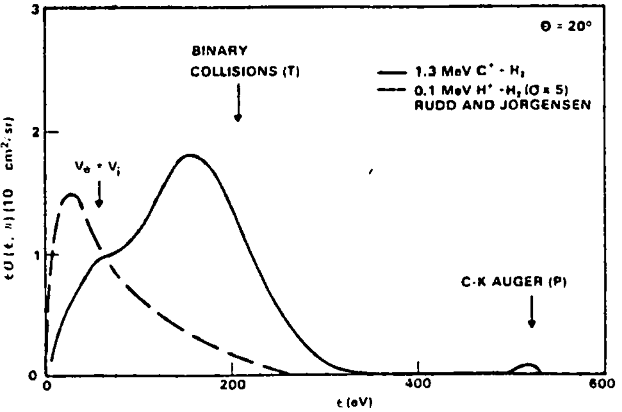

Fig. 4. Cross section times ejected electron energy for electron ejection from argon by 1.3 MeV C^+ and approximately equal velocity protons. The proton data are from the work of Crooks and Rudd, Ref. 24.

A clearly identifiable feature in the electron spectrum for collisions of C^+ with molecular hydrogen shown in Fig. 4 is the Auger electron peak associated with K-shell ionization of the projectile. Because of kinematics, the Auger peak in this spectrum occurs at sufficiently high energies to be clearly separated from the continuum electrons. The intensity of this peak, when corrected for kinematic effects,[7] provides a measure of the cross section for ionization of the carbon K-shell. If we assume that the fluorescence yield for K-shell ionization is similar to that for neutral carbon and that the collision partners can be interchanged with regard to which is moving, then this Auger intensity should yield a K-shell ionization cross section comparable to ionization of carbon by approximately 100 keV protons. The carbon K-shell ionization cross section obtained from our Doppler shifted Auger intensity is $1.8 \times 10^{-19} cm^2$ which is in excellent agreement with the value derived from the proton impact x-ray data of Langenberg and van Eck[20] which yields $1.73 \times 10^{-19} cm^2$. An implication of this close agreement is that the fluorescence yield varies little between singly ionized carbon and a neutral carbon atom.

Other features in the spectrum of electrons resulting from 1.3 MeV carbon ionization of molecular hydrogen are less clearly identified. Evidence of a small amount of contribution from electrons stripped from the projectile is observed as a shoulder on the carbon induced spectrum at the position where the ejected electron velocity is comparable to the ion velocity. The major contribution to the energy loss spectrum for incident carbon ions is for ejection of electron energies near 150 eV in contrast to the proton induced spectrum which shows no indication of peaking in this region. Although it is at a lower energy than expected, we attribute this peak to electrons ejected from the target molecule in binary collisions with the incident carbon ion. The actual peak energy and intensity results from a combination of the influence of the effective charge of the carbon ion as determined by screening, the binding energy of the target electrons, and the collision kinematics. Results of the work of Stolterfoht[7] for high-energy oxygen ions as well as our measurements for helium ions[22] show that screening of the nuclear charge varies strongly with the amount of energy transferred during the collision. For ejection of low energy electrons produced in soft collisions (large impact parameters), the bound electrons provide an efficient screen of the projectile charge. For ejection of higher energy electrons, smaller impact parameters are required to transfer sufficient momentum and screening becomes less efficient. This was illustrated to some extent in Fig. 3 where cross sections for emission of high energy electrons by He^+ and He^{++} are equal. This variable screening effect will contribute to the spectrum for energy transfer for C^+ impact by increasing the probability of electron ejection for increasing electron energies. In Fig. 4, the cross section for an ejected electron energy of 150 eV by carbon ion impact is approximately 25 times larger than the corresponding proton impact value. If we assume Z^2 scaling, this implies that the carbon ion had an effective charge of +5 in the interaction which yielded 150 eV electrons. Such a high charge indicates a small impact parameter; one near the radius of the K-shell electrons of the carbon ions. For ejected electron energies above 150 eV, the spectrum in Fig.4 decreases due to kinematics; the probability of energy transfer decreases dramatically for energies beyond the classical kinematic limit. The fact that the target electrons are bound influences the ejected electron spectrum in two ways. The bound electrons have an initial velocity distribution which contributes to the width of the "binary encounter peak" and the fact that the electrons are bound rather than free contributes to the shift of the peak to lower electron energies than calculated for a free electron; the free electron energy calculation is shown by an arrow in Fig. 4

Some interesting characteristics of the electron emission spectra for incident carbon ions can be observed by comparing cross sections for different charge states of the incident ion. Data for ejection of electrons at 50° and 130° are shown in Fig. 5 for ionization of argon by 3 MeV C^+, C^{+++}, and equal velocity protons (0.25 MeV/amu). The carbon ion data have been divided by 36 (nuclear charge squared) in light of previous data for helium ions and oxygen ions which indicate that the nuclear charge is effectively unscreened for ejection of high energy electrons.[7,22] This scaling procedure again is successful for high-energy electrons as the scaled cross sections for C^+, C^{+++}, and H^+ all converge. This scaling procedure is not expected to be relevant to lower energy ejected electrons but it does give some measure of the degree to which the projectile charge is screened as the energy transfer decreases, i.e., average impact parameter increases. Also, note that the magnitude of screening appears to be dependent on the electron emission angle. The cross section for emission of 20 eV electrons is a factor of 2.3 larger for C^{+++} than for C^+ for an ejection angle of 50° and only a few percent larger for an ejection angle of 130°. These data illustrate the complicated nature of projectile screening and the difficulty of scaling cross sections for incident heavy ions.

The contribution to the ejected electron spectra of electrons stripped from the projectile is most evident in Fig. 5 in the spectrum for emission at 130°. The contribution is largest for C^+, as expected, since it carries a larger number of bound electrons. It is interesting to consider the collision in the rest frame of the carbon ion where the interaction mav be viewed

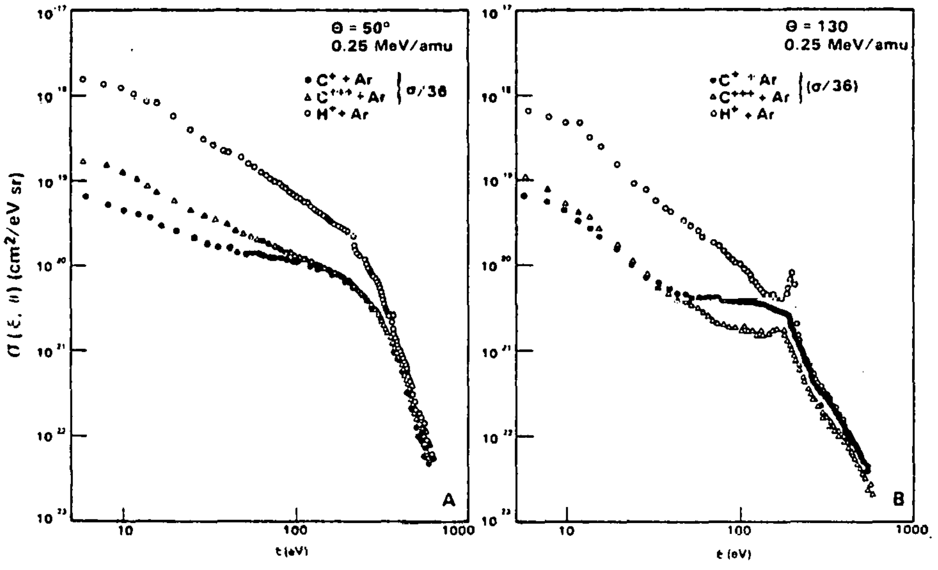

Fig. 5. Electron emission cross sections for electron emission from argon by 3 MeV C^+, C^{+++}, and equal velocity H^+ impact. The proton data are from Ref. 3.

as ionization of a carbon ion by an incident proton; the incident proton may be screened somewhat since it is actually a constituent of a hydrogen molecule. The ionized electron spectrum in the carbon ion rest frame should look very similar to spectra such as those shown in Fig. 1 for ionization by protons. For example, one would expect a peak for ejection of low-energy electrons (soft collisions) and a binary encounter peak for ejection of fast electrons. Since binary collisions deal with large energy transfer, the bound electrons interact as though nearly free and one would expect the cross section to depend on the number of bound electrons. According to the description of Drepper and Briggs,[23] the binary encounter electrons associated with ionization of the projectile will be observed predominantly at large angles as observed in the laboratory. Analysis of the relative intensity of the electron loss peaks observed at 130° in Fig. 5B indicates intensities proportional to the number of projectile electrons. After making an approximate correction to account for target ionization, the relative cross sections of stripping electrons from C^+, C^{++}, and C^{+++} ions vary approximately as 10, 7, and 3, respectively, for laboratory ejection energy of 135 eV and emission angle of 130°. The relative cross sections decrease somewhat faster than the relative number of projectile electrons due to the increased binding energy of the higher charge states. The relative contribution of stripped electrons to the emission spectra for small laboratory emission angles represented by the 50° spectrum in Fig. 5A cannot be reliably determined because the cross sections for stripping plus target ionization appear similar for both C^+ and C^{+++} impact. One cannot tell whether the cross sections for stripping have become less dependent on projectile charge state for emission into small angles or whether the target ionization cross section has become sufficiently large relative to the stripping cross section to dominate the spectrum.

SINGLE DIFFERENTIAL ELECTRON EMISSION CROSS SECTIONS

The contribution to ejected electron spectra from electrons stripped from the projectile is difficult to determine in low velocity collisions because the stripping cross section may be small and masked by the variation in projectile screening with ejected electron energy. An indication of the significance of projectile stripping and screening as a function of projectile velocity can be obtained by comparing ejected electron spectra plotted as the ratio of measured cross sections to the corresponding Rutherford values. This ratio, Y(E,T) is given by:

$$Y(E,T) = \frac{T}{4\pi a_0^2 R^2} E^2 \sigma(\varepsilon) \qquad (1)$$

where R is the Rydberg unit of energy (13.605 eV), T is the kinetic energy of the incident ion in units of an equivalent velocity electron ($T = \frac{1}{2} m_e v_i$, where m is the electron mass and v_i is the ion velocity), a_0 is the Bohr radius (0.529A), and E is the energy transfer in ejection of an electron of energy ε ($E = \varepsilon + I$, where I equals the electron binding energy). The single differential cross section $\sigma(\varepsilon)$ is obtained from measured double differential cross section by integration with respect to electron emission angle. An advantage of converting the emission cross sections to the form given by Y(E,T) is that cross sections which vary by many orders of magnitude can be displayed for convenient analysis of spectral features on a linear scale. In addition, when Y(E,T) is plotted vs R/E, as in Fig. 6, the area under the curve is proportional to the total ionization cross section. This provides a graphical illustration of the significance of spectral features as contributions to the ionization process. The comparison of

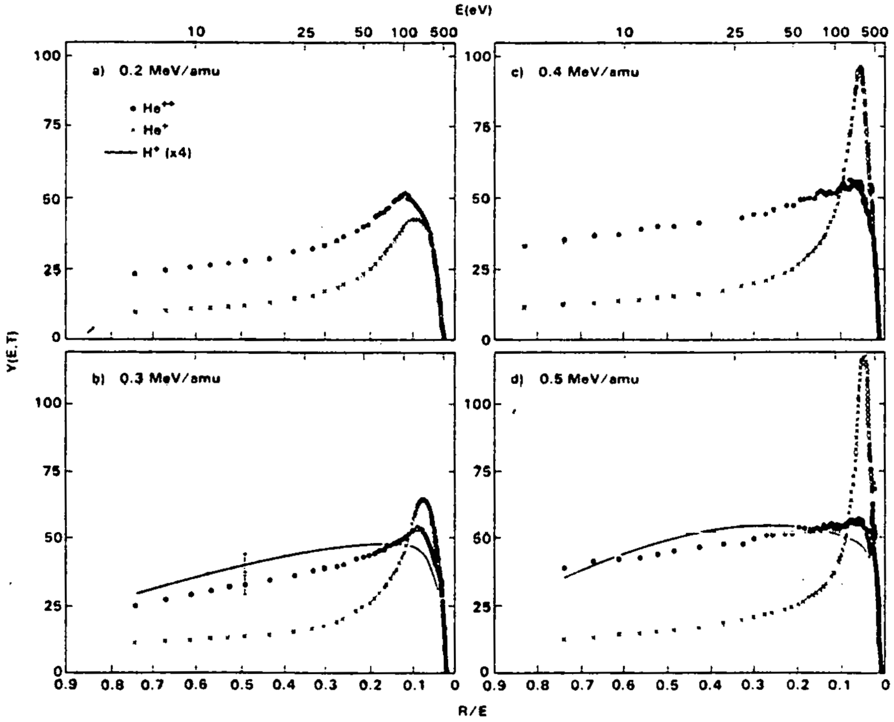

Fig. 6. Electron emission from water vapor by H^+, He^+, and He^{++} impact. See the text for definition of the quantity Y(E,T). The proton data are from Ref. 16.

electron spectra for ionization of water vapor by He^+, He^{++}, and H^+ ions of several energies shown in Fig. 6 clearly indicates the importance of screening of the projectile charge in interactions involving small energy transfer. For larger energy transfer, screening becomes inefficient and results for He^+ and He^{++} impact are similar. It is also evident that screening becomes somewhat less significant as the energy of the incident ion decreases. The most dramatic change in the spectra with increasing ion energy is the increase in intensity of the electron loss contribution to the ionization process. At the highest ion energy shown (0.5 MeV/amu), the stripped electron peak dominates the spectrum for He^+ impact and at the lowest ion energy, there is no evidence of this contribution. The proton data shown in Fig. 6 have been multiplied by a factor of 4 for comparison to the helium ion data to determine the reliability of Z^2 scaling for this energy range. The deviations from Z^2 scaling, although small, are significant and are discussed in detail elsewhere.[22] The primary purpose of Fig. 6 is to illustrate the effect of projectile structure on electron energy spectra. Since equal areas under these curves contribute equally to the total ionization cross section, it is apparent that a large fraction of the ionization cross section for fast He^+ impact is contributed by fast electrons. The mean energy of electrons will, therefore, be much larger for ionization by He^+ than for equal velocity protons or alpha particles. A similar plot for ionization of argon by carbon ions is shown in Fig. 7 along with data for equal velocity protons from the work of Crooks and Rudd.[24] The dashed lines are estimates of the cross sections for low-energy electrons; the carbon ion measurements are unreliable for electron energies less than about 10 eV. The ion velocities here are about 30% lower than the lowest energy helium ion data shown in Fig. 6, so we would not expect a very large contribution from projectile stripping. Still, the carbon ion data show a large contribution of fast electrons relative to the proton induced spectra. Although there is some evidence of a contribution to the spectra from electrons stripped from the carbon ion (see the shoulder on the 1.3 MeV C^+ spectrum at about 60 eV), the principle reason for the rise in the spectrum at higher electron energies is attributed to a decrease in the efficiency of projectile screening for large energy transfer. Again, the significance of this effect is that the mean energy of electrons ejected in ionizing collisions will be much greater for the structured ions than for bare projectiles of the same velocity. Further measurements are required to obtain systematics of interaction processes such as these where screening is particularly important. It is hoped that such measurements will not only provide data needed for application in several research fields but provide incentive for theoretical calculations which, when compared to experimental data, will provide insight into the collision process.

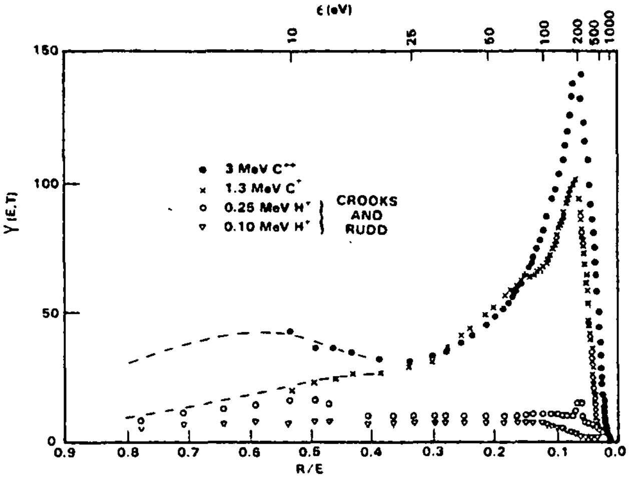

Fig. 7. Electron emission from argon by 1.3 MeV C^+ and 3 MeV C^{++} impact. $Y(E,T)$ is described in the text and the proton data are from Ref. 3. The dashed lines are estimates of the cross sections for low-energy electrons; the measurements are unreliable for energies less than about 10 eV.

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