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Ionization and charge exchange in multiply-charged-ion—helium collisions at intermediate energies

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A Bohr classical model for He is applied to multiply charged ions colliding with He at intermediate energies. Reactions studied are single-electron capture, single and double ionization, and electron-capture ionization for projectile charge states $q=1+$ to $100+$, and the energy range $E=1-5$ MeV/amu. The dominant electron-removal collision process is single ionization. For the higher-charge-state ions, single-charge exchange is found to be primarily due to transfer ionization, a two-electron process where one electron is ionized and the other is captured by the projectile. For low-charge states, the single- and double-ionization cross sections are close to the expected q^2 and q^4 scaling. The calculations are in reasonable agreement with available experimental data.

I. INTRODUCTION

When Hirschfelder *et al.*[1] began their early work using the classical trajectory Monte Carlo method on the $H+H_2$ system, they completed only one trajectory due to the time taken to complete calculations "by hand." Work using classical mechanics in atomic scattering problems was laid aside due to lack of computing machines. However, since Gryzinski's early work[2] and the availability of ever faster computers, the role of classical mechanics in atomic scattering problems has enjoyed a renaissance. More specifically, the classical trajectory Monte Carlo method (CTMC) has become an important technique in calculating cross sections for chemical reactions. Olson *et al.*[3] have proved that the CTMC method is a very successful tool for treating collisions between multiply charged ions and atomic hydrogen at energies of approximately 500 keV/amu ($v\approx 10^9$ cm/sec).

In this paper we address the problem of calculating the electron-loss cross sections in the velocity range from 1.40×10^9 cm/sec (1 MeV/amu) to 3.08×10^9 cm/sec (5 MeV/amu) for a four-body system. The reactions studied were

$$A^{q+}+\mathrm{He}\rightarrow\begin{cases} A^{(q-1)+}+\mathrm{He}^{+}, & \text{1-cex} \quad (1a)\\ A^{q+}+\mathrm{He}^{+}+e, & \text{1-ion} \quad (1b)\\ A^{(q-2)+}+\mathrm{He}^{2+}, & \text{2-cex} \quad (1c)\\ A^{q+}+\mathrm{He}^{2+}+2e, & \text{2-ion} \quad (1d)\\ A^{(q-1)+}+\mathrm{He}^{2+}+e, & \text{cex-ion} \quad (1e)\end{cases}$$

where 1-cex is single-charge exchange; 1-ion is single ionization; 2-cex is double-charge exchange; 2-ion is double ionization; and cex-ion is combined single-charge exchange and single ionization (transfer ionization).

Fully stripped ions were investigated, with charge states of $q=1+$, $2+$, $3+$, $5+$, $10+$, $20+$, $50+$, and $100+$. Experimental data are available for some of the ions and reactions studied here and will be compared to our results.

The CTMC method is quite useful because it is easily applied to a wide range of incident ions and can concurrently compute cross sections for electron capture, impact ionization, and capture ionization.[4] The method, also, preserves the unitarity of the S matrix and is nonperturbative. When there is a strong interaction involving, as we have in this study, a highly charged ion colliding with a low-Z target, perturbation theories such as the low-order Born approximation are questionable. Also, this is a three-dimensional calculation that includes all particles and all forces between them; these forces include such results as the recoil of the target nucleus.

It is well known that the classical two-electron atom is unstable and will autoionize unless properly constrained in the initial orbits of the electrons. For this reason, we have incorporated the method of Kirschbaum and Wilets,[5] thus preventing the collapse of the atom by use of the Heisenberg uncertainty principle. Kirschbaum and Wilets also make use of the Pauli exclusion principle, however, with He, this is not necessary, since the two electrons are in different spin states.

Our work directly addresses a difficult many-body ($n=4$) problem and provides the first systematic set of calculations of single and double ionization in a range of intermediate collision energies. The discussion of the theoretical method and a presentation of the results follow.

II. THEORY

The method we have used is based on solving Hamilton's equations of motion for the four-body system, which includes the incident ion, target nucleus, and two electrons initially bound to the target nucleus. The stages of the computer scattering experiment are: preparation of initial conditions and system setup, collision, and analysis of final states. The equations are solved for a variety of sets of initial conditions, including impact parameter, relative velocity of projectile to target, and positions and momenta of electrons. For each set of initial conditions, the classical trajectories of the nuclei are calculated from a

large internuclear separation to the distance of closest approach, and out again to a large internuclear separation.

The electrons were not kept independent of each other. They interacted with one another directly by their $1/r_{12}$ repulsive potential. The orbital eccentricity was zero, thus defining a circular orbit. We use the "actual" charge of 2+ rather than an effective charge for the target nucleus.

At the end of the trajectories, if both electrons remain bound to the target nucleus, then a ground or an excited state of the He atom results. If one electron remains bound to the target and one becomes bound to the projectile then single capture results. If one electron remains bound to the target and one electron is free (not bound to either target or projectile) then single ionization results. If both electrons are bound to the projectile then double capture results. If both electrons are free then double ionization results. If one electron is bound to the projectile and one is free then capture ionization results. For a good statistical distribution defining the cross sections, the number of trajectories used was 5000 to 10 000.

The work of Abrines and Percival[6] on the Bohr hydrogen atom was extended to encompass four particles rather than three. We made the approximation that the four bodies obey Newtonian laws during the collision. The units used were atomic units; in these units the initial binding energy of each electron to the He target atom is 0.903 a.u. (which includes the repulsive electron-electron term).

The computer experiment used step-by-step integration of Hamilton's equations of motion by means of the Runge-Kutta-Gill method. The integration began at a distance of about $5q$ a.u. from the target nucleus (q is the charge state of the incident ion), which was at rest and at the origin at time $t=0$.

A. Hamilton's equations of motion

The Hamiltonian for the four-body system is

$$\mathscr{H}=\frac{p_a^2}{2m_a}+\frac{p_b^2}{2m_b}+\frac{p_c^2}{2m_c}+\frac{p_d^2}{2m_d}+\frac{Z_aZ_b}{r_{\rm ab}}+\frac{Z_aZ_c}{r_{\rm ac}}+\frac{Z_aZ_d}{r_{\rm ad}}+\frac{Z_bZ_c}{r_{bc}}+\frac{Z_bZ_d}{r_{bd}}+\frac{Z_cZ_d}{r_{cd}}\ , \tag{2}$$

where the subscripts a, b, c, and d refer to the incident ion, target nucleus, and the two electrons, respectively,

$$\dot{Q}_i=\frac{\partial\mathscr{H}}{\partial p_i}\quad\text{and}\quad\dot{P}_i=-\frac{\partial\mathscr{H}}{\partial q_i}\ . \tag{3}$$

Using the indices 1,2,3 for the x,y,z coordinates of the projectile, 4,5,6 for the target nucleus, 7,8,9 for the first electron, and 10,11,12 for the second electron, we can specify the velocities of the particles by the following:

$$\dot{Q}(j=1,3)=\frac{c(j+12)}{m_a},\quad \dot{Q}(j=4,6)=\frac{c(j+12)}{m_b}\ ,$$
$$\dot{Q}(j=7,9)=\frac{c(j+12)}{m_c},\quad \dot{Q}(j=10,12)=\frac{c(j+12)}{m_d}\ , \tag{4}$$

where m_a is the mass of the projectile, m_b is the mass of the target nucleus, m_c is the mass of the first electron, and m_d is the mass of the second electron.

The indices $j=13,14,15$ are used to specify the x,y,z components of the momentum for the projectile; $j=16,17,18$ for the target nucleus, $j=19,20,21$ for the first electron and $j=22,23,24$ for the second electron.

By taking the appropriate partial derivatives, the momenta are described by

$$\begin{aligned}
\dot{P}(j=13,15)=&-\frac{Z_aZ_b}{r_{ab}^3}[c(j-9)-c(j-12)]\\
&-\frac{Z_aZ_c}{r_{ac}^3}[c(j-6)-c(j-12)]\\
&-\frac{Z_aZ_d}{r_{ad}^3}[c(j-3)-c(j-12)]\ ,\\
\dot{P}(j=16,18)=&\frac{Z_aZ_b}{r_{ab}^3}[c(j-12)-c(j-15)]\\
&-\frac{Z_bZ_c}{r_{bc}^3}[c(j-9)-c(j-12)]\\
&-\frac{Z_bZ_d}{r_{bd}^3}[c(j-6)-c(j-12)]\ ,\\
\dot{P}(j=19,21)=&\frac{Z_bZ_c}{r_{bc}^3}[c(j-12)-c(j-15)]\\
&+\frac{Z_aZ_c}{r_{ac}^3}[c(j-12)-c(j-18)]\\
&-\frac{Z_cZ_d}{r_{cd}^3}[c(j-9)-c(j-12)]\ ,\\
\dot{P}(j=22,24)=&\frac{Z_aZ_d}{r_{ad}^3}[c(j-12)-c(j-21)]\\
&+\frac{Z_bZ_d}{r_{bd}^3}[c(j-12)-c(j-18)]\\
&+\frac{Z_cZ_d}{r_{cd}^3}[c(j-12)-c(j-15)]\ ,
\end{aligned} \tag{5}$$

where Z_a, Z_b, Z_c, and Z_d are the charges of the projectile, the target nucleus, and electron 1 and electron 2, respectively.

When the collision caused no further significant perturbation, the integration of Hamilton's equations was stopped. Then "exit tests" were performed based on the calculation of energies. The energies were calculated as follows:

$$E_{ij}=27.211\left\{\frac{1}{2}\left[\frac{m_im_j}{(m_i+m_j)}\right]\times\left[(v_{j_x}-v_{i_x})^2+(v_{j_y}-v_{i_y})^2+(v_{j_z}-v_{i_z})^2\right]+\frac{Z_iZ_j}{r_{ij}}\right\}. \tag{6}$$

(The factor 27.211 converts a.u. to eV.)

The tests were based on four criteria, not two as in previous papers,[7,8] With the addition of the fourth body, the second electron, and the electron-electron term, two addi-

tional criteria became necessary. The criteria followed for the determination of reactions were then as follows:

if $E_{\rm tot}<0$ and $E_{ac}\geq 0$ and $E_{ad}\geq 0$, the excited state is implied;

if $E_{ac}<0$ and $E_{ad}<0$ and $E_{bc}\geq 0$ and $E_{bd}\geq 0$, 2-cex is implied;

if $E_{ac}\geq 0$ and $E_{ad}\geq 0$ and $E_{bc}\geq 0$ and $E_{bd}\geq 0$, 2-ion is implied;

if $E_{ac}\geq 0$ and $E_{ad}<0$ and $E_{bc}\geq 0$ and $E_{bd}\geq 0$, capture of electron d by projectile and ionization of the electron c is implied;

if $E_{ac}<0$ and $E_{ad}\geq 0$ and $E_{bc}\geq 0$ and $E_{bd}\geq 0$, capture of electron c by projectile and ionization of the electron d is implied;

if $E_{\rm tot}\geq 0$ and $E_{ac}\leq 0$ or $E_{ad}\leq 0$ capture of electron c or d by projectile while the uncaptured electron remains bound to the target nucleus is implied;

if $E_{tot}\geq 0$ and $E_{ac}\geq 0$ and $E_{ad}\geq 0$, ionization of electron c or d while the other electron remains bound to the target nucleus is implied;

E_{ij} are the various interparticle energies and E_{tot} is the total electronic energy, $E_{\rm tot}=E_{bc}+E_{bd}+(27.211/r_{cd})$.

B. Determination of electronic initial conditions

To prevent the collapse of the classical He atom, we imposed the Heisenberg uncertainty principle using the method of Kirschbaum and Wilets[5] as a constraint on our He atom model in the form of

$$r_i p_i \geq \xi\hbar \ . \tag{7}$$

We can use rp rather than $\Delta r\Delta p$ since we are dealing with a so highly localized space. These constraints are approximated by potentials of the general form

$$V_c(p,r)=\frac{f(r,p)}{r^2} \ , \tag{8}$$

and with the particular form of

$$V_c=\frac{(\xi\hbar)^2}{4\alpha r^2 m_e}\exp\left\{\alpha\left[1-\left[\frac{rp}{\xi\hbar}\right]^4\right]\right\} . \tag{9}$$

Recall that in utilizing atomic units, $\hbar=m_e=e=1$. ξ is a dimensionless constant which must be determined; α is an adjustable hardness parameter. We used a large value, $\alpha=50$, which gives more nearly the Heisenberg constraint $rp\geq\hbar$.[5,9] The classical Hamiltonian for the two electrons combined with the ξ constant give a complete description of the model and may be used to determine the ground-state configuration of atoms.

To determine the ξ constant, we have applied the restraining potential used by Kirschbaum and Wilets, Eq. (9), on the hydrogen model in the ground-state configuration. This means that

$$rp=\xi \ \text{ or } \ p=\frac{\xi}{r} \ . \tag{10}$$

Applying this potential, Eq. (9), to the two-electron target Hamiltonian yields a new target Hamiltonian,

$$\mathscr{H}=\sum_{i=1}^{2}\left[\frac{\xi^2}{2r_i^2}-\frac{Z}{r_i}\right]+\frac{1}{2}\sum_{i\neq j}\frac{Z}{r_{ij}} + \sum_{i=1}^{2}\frac{\xi^2}{4\alpha r_i^2}\exp\left\{\alpha\left[1-\left[\frac{r_i p_i}{\xi}\right]^4\right]\right\} . \tag{11}$$

where i represents either the first or the second electron, and $Z=2$ for He.

We require $\partial\mathscr{H}/\partial r_i=0$ for the stationary state. With this requirement we can find expressions for r, p, and E,

$$\frac{\partial\mathscr{H}}{\partial r_1}=\frac{Z}{r_1^2}-\frac{1}{(r_1+r_2)^2}-\frac{\xi^2}{r_1^3}\exp\left\{\alpha\left[1-\left[\frac{r_1p_1}{\xi}\right]^4\right]\right\} - \frac{\xi^2}{2\alpha r_1^3}\exp\left\{\alpha\left[1-\left[\frac{r_1p_1}{\xi}\right]^4\right]\right\} . \tag{12}$$

The exponential terms are unity for $r_i p_i=\xi\hbar$, and since $r_1=r_2$ we have $r_1+r_2=2r_1$, then

$$\frac{\partial\mathscr{H}}{\partial r_1}=\frac{Z}{r_1^2}-\frac{1}{(r_1+r_2)^2}-\frac{\xi^2}{r_1^3}-\frac{\xi^2}{2\alpha r_1^3}=0 \ ,$$

$$r=\left[\frac{4}{4Z-1}\right]\xi^2\left[\frac{2\alpha+1}{2\alpha}\right] . \tag{13}$$

To find p, we impose the condition of the stationary state $rp=\xi$ or $p=\xi/r$,

$$p=\frac{4Z-1}{4\xi}\left[\frac{2\alpha}{2\alpha+1}\right] . \tag{14}$$

To derive an expression for E, we return to the two-electron Hamiltonian, Eq. (11), and substitute for r and for p the expressions above in Eqs. (13) and (14).

Recall that $r_1=r_2$, also, $p_1=p_2$, and the exponential terms are equal to unity. E then reduces to

$$E=-\frac{(4Z-1)^2}{16\xi^2}\left[\frac{2\alpha}{2\alpha+1}\right] . \tag{15}$$

We can also determine expressions for the first and second ionization potentials. For the first ionization potential

$$V_{\rm IP_1}=-\frac{(16Z^2-16Z+3)}{32\xi^2}\left[\frac{2\alpha}{2\alpha+1}\right] . \tag{16}$$

For the second ionization potential, $V_{\rm IP_2}$

$$V_{\rm IP_2}=-\frac{(16Z^2-1)}{32\xi^2}\left[\frac{2\alpha}{2\alpha+1}\right] . \tag{17}$$

To determine the value of ξ, with $\alpha=50$, we algebraically manipulate Eq. (16) for $V_{\rm IP_1}$ and solve for ξ, with $V_{\rm IP_1}=-24.587$ eV $=-0.904$ a.u. (the spectroscopic value). This gives a value of $\xi=1.094\,49$. Using this calculated value of ξ yields a value of $V_{\rm IP_2}=-44.2777$ eV $=-1.6272$ a.u. rather than the true value of $V_{\rm IP_2}=-54.416$ eV $=-2.0000$ a.u., making the sum of the two ionization potentials $V_{\rm IP_1}+V_{\rm IP_2}=-68.8647$ eV $=-2.5312$ a.u. rather than the experi-

mental value of -79.003 eV $=-2.904$ a.u., a difference of 12.8%. The dominant process is the first (single) removal by ionization. Thus, it is more important to accurately model the first ionization potential with error in the second ionization potential than vice versa. With these values for ξ and for α, the values for r and p are $r=0.6914$ a.u. and $p=1.58$ a.u.

III. RESULTS

The results of the previous sections were incorporated into a computer code to obtain cross sections that can be compared to experiments. Specifically, we modeled the target atom using the Kepler equation to define the electrons in orbits; also, we utilized the Euler angle transformations to place the orbits in three-dimensional space. Further, we modeled an incoming beam of projectiles to simulate a uniform density beam based on a grid defined by the square of the impact parameter divided by the number of grids. The incoming beam was divided into ten equal annular areas with an equal number of scattering attempts in each area.

Cross sections were calculated and evaluated for reactions (1a) through (1e) with fully stripped ions in charge states from $q=1+$ to $q=100+$. The projectile energy range was varied from 1 to 5 MeV/amu. Our calculations utilized the CTMC four-body method, which included the electron-electron potential energy term.

The cross sections Q_R (R may represent ionization, charge exchange, double ionization,...) was calculated by

$$Q_R=\left[\frac{N_R}{N}\right]\pi b_{\max}^2 , \tag{18}$$

where $b_{\max}$ is the maximum impact parameter, N_R is the number of collisions satisfying the criteria for reaction, and N is the total number of trajectories calculated. The probable error from counting events is determined by

$$\Delta Q_R=Q_R\left[\frac{(N-N_R)}{NN_R}\right]^{1/2} . \tag{19}$$

The statistical uncertainty, Eq. (19), was $<10\%$ and most often $<5\%$.

The results for single and double ionization, electron capture, and charge-exchange ionization are shown in Figs. 1 through 3 for the collision energies 1, 2, and 5 MeV/amu. (Double-charge exchange was too small for our statistics.) These results are compared to experimental data of coincidence measurements by several researchers, most of which are only available for the 1-MeV/amu projectile energy and are displayed in Fig. 1.

In general, our theoretical values were in good agreement with the experimental data. Obvious exceptions are in the double ionization, $q=6,7,8$, where our values are approximately 30% below those of Hvelplund *et al.*[10] Regarding single ionization, for $q=1,2$, our values lie below those of Shah and Gilbody by approximately 20%.[11] At $q=1+$, double ionization, our value is approximately 25% below that of Shah and Gilbody.[11] For combined single-charge-exchange and ionization at charge states $q\lesssim 10+$, our values lie approximately 20% below the

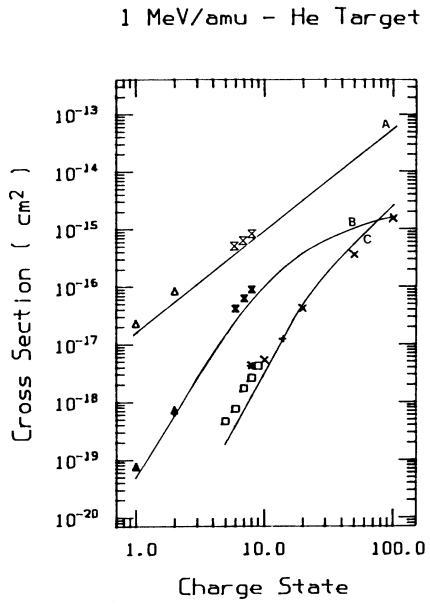

FIG. 1. Cross sections vs charge state for He atom (1 MeV/amu). Curves are theoretical results: *A*, 1-ion; *B*, 2-ion; *C*, cex-ion. ×, Schlachter scaling rules (Ref. 12). Experimental data are △, Shah and Gilbody (Ref. 11) 1-ion; ▲, Shah and Gilbody (Ref. 11) 2-ion; ⧖, Hvelplund *et al.* (Ref. 10) 1-ion; ⧗, Hvelplund *et al.* (Ref. 10) 2-ion; +, Scheibel *et al.* (Ref. 15) 1-cex; ✱, Knudsen *et al.* (Ref. 13) 1-cex; □, Guffey *et al.* (Ref. 14) 1-cex.

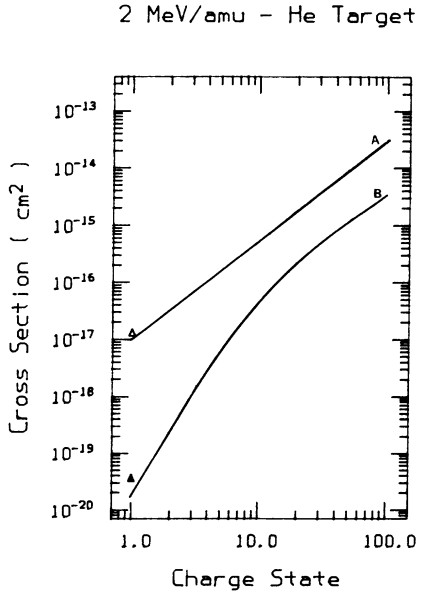

FIG. 2. Cross sections vs charge state for He atom (2 MeV/amu). Curves are theoretical results: *A*, 1-ion *B*, 2-ion. Experimental data are △, Shah and Gilbody (Ref. 11) 1-ion; ▲, Shah and Gilbody (Ref. 11) 2-ion.

empirical scaling formula of Schlachter *et al.*[12] as well as the experimental data of Knudsen *et al.*[13] and Guffey *et al.*[14] For $q > 10+$ our calculated values lie approximately 10% above the scaling formula and also above the datum of Schiebel *et al.*[15]

For Fig. 2, 2 MeV/amu, our calculations are in reasonable agreement with the proton data of Shah and Gilbody[11] for both single and double ionization. At all collision energies, the dominant collision process is single ionization, the most "effortless" process to accomplish, followed by double ionization, and then by single-charge-exchange ionization; excepting $q \gtrsim 60+$ at 1 MeV/amu where combined single-charge-exchange and ionization becomes dominant over double ionization.

The single-ionization cross section calculated at the three energies are found to scale as approximately $q^{1.78\pm0.05}$. This charge-state dependence is less than predicted by the Born approximation q^2, but is in agreement with recent measurements by Be *et al.* who find $q^{1.84\pm0.06}$ for fully stripped ions in charge states $q \leq 10+$ at 1.05 MeV/amu.[16] The possible reason for the difference between the Born result and our calculations is that all the electron-removal channels are explicitly included in the CTMC calculations, thus reducing the amount of flux that can be associated with single ionization. Also, as one proceeds to higher charge states at a given energy, or lower values of E/q, experimental measurements indicate a reduced dependence of the ionization cross section on the charge state.[17]

From Figs. 1—3, the double-ionization cross sections are found to approximately follow the Born prediction of approximately q^4 for low charge states (values of $E/q \gtrsim 0.5$ MeV/amu). For lower values of E/q the double-electron-removal reactions parallel those of single ionization and follow an approximate q^2 dependence. Analysis of the calculated transition probabilities show the double-electron removal values saturate the small-impact-parameter region at the 50% to 90% range for values of $E/q \lesssim 0.2$ MeV/amu. Thus, it is unlikely that a scaling based on a perturbation treatment, i.e., a q^4 dependence, would be valid for such "strong" collisions.[3,17]

The single-capture events are composed of "true" single capture, reaction (1a), and the capture-ionization (or transfer-ionization) channel, reaction (1e). Our calculations indicate that above $q = 10+$ at 1 MeV/amu, the double-electron event, reaction (1e), dominates over true single capture, reaction (1a).[18] Analysis of the transition probabilities indicates the electron-capture processes dominate at small impact parameters, while ionization proceeds to large impact parameters that greatly exceed the orbital radius of the He atom. For collisions with high-charge-state ions at the energies studied here, it is difficult not to have electron capture without subsequent ionization of the He atom. The scaling calculation of the single capture for $q \lesssim 20+$ is found to proceed similar to that for double ionization, approximately q^4, which is consistent with that observed by Schlachter *et al.*,[12] $q^{3.9}$. For higher charge states for which there are no data, we find the single capture (primarily capture ionization) competing with the double-ionization channel as shown in Fig. 1.

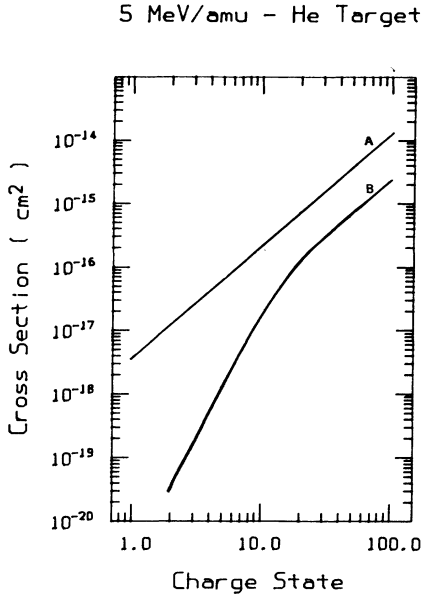

FIG. 3. Cross sections vs charge state for He atom (5 MeV/amu). Theoretical results are shown: *A*, 1-ion; *B*, 2-ion.

Figure 4 shows a reduced plot of single-electron removal by impact ionization. The plot is reduced to a common curve by dividing both the energy and the cross section by the charge state.[3,17] The curve varies approximately as q^2/E.

The line curve fit through the points can be parametrized by

$$\sigma_{1\text{-ion}} = [(1.46\times10^{-17})q^{1.78}/(E^{0.78})]\ \text{cm}^2\ , \tag{20}$$

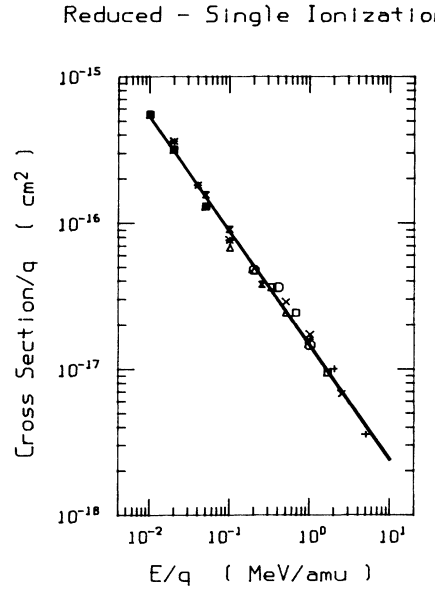

FIG. 4. Reduction of single-ionization cross sections. $+$ =H; $\times$,He; □, Li; ○,B; △, Ne; ⧖, Ca; ✱, Sn; ■, Fm.

where $\sigma_{1\text{-ion}}$ is the single-ionization cross section, q is the charge state of the projectile, and E is in MeV/amu. Note that the behavior of σ as approximately q^2/E is the same as the classical prediction for single-electron removal.[19]

A principal result of this study is the observance of the two-electron nature of the single-charge capture. This behavior has previously been noted, at lower energies, by Andersen *et al.*[20] and by Datz *et al.*[21] We have compared out calculations to available experimental data and also to the scaling rules which were derived by Schlachter *et al.*[12] for the purpose of testing theoretical computations for charge-transfer predictions. Our values are in reasonable agreement with both the Schlachter points and experimental data (Fig. 1). The double-electron-removal process necessary for single-electron capture by high charge ions helps explain why the charge-state scaling given by Schlachter *et al.*,[12] $q^{3.9}$, deviates from the q^3 dependence predicted by theoretical models based on a single-electron transition.[22,23]

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