

## Inclusive and exclusive cross sections for multiple ionization by fast, highly charged ions in the independent-electron approximation

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Cross sections for the ionization of  $n$  of  $N$  electrons with equal single-electron ionization probability  $P$  are considered. When both  $N$  and the projectile charge  $q$  are large, the cross sections for single and double ionization are both found to be approximately linear in  $q$  at 1 MeV/amu. The ratio of double-to-single-ionization cross sections is independent of  $q$ . Moreover, first-order perturbation theory for the single-electron ionization probability  $P$ , which varies as  $q^2$ , is found to be applicable due to the damping of contributions with large  $P$  caused by factors of  $(1-P)^{N-n}$ . For large  $P$  there are differences between the inclusive probability  $\sum_n \binom{N}{n} P^n (1-P)^{N-n}$  and the probability  $NP$  commonly used for a target with  $N$  electrons. Both of these probabilities differ significantly from the exclusive probability  $NP(1-P)^{N-1}$  for the ionization of only one electron. For large  $N$  and large  $q$ , the exclusive ionization probabilities for removing exactly  $n$  of the  $N$  electrons tend to be concentrated in somewhat separate ranges of impact parameters  $b$ , defining impact-parameter "windows." The windows which we obtain using the quantum-mechanical semiclassical-Coulomb-approximation (SCA) probabilities are similar to those using classical Monte Carlo calculations. Model calculations, based on analytic fits to the SCA probabilities, are used to obtain approximate analytic expressions for single- and double-ionization cross sections and for the impact-parameter windows. Results of this simple model are in reasonable agreement with measured cross sections for single and multiple ionization of neon atoms by projectiles of charge  $q$  varying from 1 to 13 at a velocity corresponding to 1 MeV/amu.

### I. INTRODUCTION

For an understanding of an atomic or molecular collision it is in principle necessary to specify exactly what happens to each electron. However, since a detailed knowledge of each of possibly many electrons is difficult to obtain, it has in practice been useful to select a single, active electron, whose properties are specified, and to sum over the final states of the remaining, so-called passive electrons. Such procedures, which include sums over the final states of some electrons, are called inclusive. Studies<sup>1-11</sup> of the more difficult exclusive procedures, where the states of the electrons are specified, are becoming more common and are yielding more detailed information about atomic and molecular collisions. In this paper, using the independent-electron approximation (IEA), expressions for inclusive and exclusive cross sections are related, and some properties of exclusive cross sections for single and multiple ionization are examined.

A number of useful properties of inclusive and exclusive cross sections have been discussed by a variety of authors.<sup>1-14</sup> In this paper we shall work with the simple binomial distribution for the ionization probability function.<sup>15-17</sup> Although other methods are available, some of which incorporate a more complete quantum analysis,<sup>18,19</sup> the simple binomial distributions have been used to describe multiple-vacancy production in ion-atom collisions<sup>2-4,18-22</sup> and multiple ionization by strong laser pulses.<sup>23,24</sup> A substantial amount of experimental data has been analyzed using binomial distributions. For binomial distributions, if one sums over the final states of

all electrons in atomic shells other than the shell containing the active electron, the inclusive probabilities for the summed states is unity.<sup>12-14</sup> In this paper we form sums over states of electrons within the same shell. In this case there is a difference between the inclusive sum  $\sum_n \binom{N}{n} P^n (1-P)^{N-n}$  and the single-electron ionization probability  $P$  multiplied by the number of electrons  $N$  in the shell of the active electron.

Of particular interest in this paper are some properties of exclusive cross sections for multiple ionization by high-velocity projectiles with a large charge  $q$ . Using first-order perturbation theory, where the single-electron-ionization probability  $P$  varies as  $q^2$ , it is shown that both the single- and double-ionization cross sections are approximately linear in  $q$  and the ratio is independent of  $q$  for systems where  $N \gg 1$ . Furthermore, under these circumstances, it is shown that perturbation theory is applicable when  $n \ll N$ . For large  $q$  and large  $N$ , ionization of a particular number  $n$  of electrons tends to occur in distinct ranges of the impact parameter, i.e., distinct impact-parameter "windows," as was pointed out by Olson<sup>1</sup> using classical Monte Carlo calculations and by Cocks<sup>21</sup> using a statistical energy-deposition model. Calculations presented here using the semiclassical-Coulomb-approximation<sup>25</sup> (SCA) single-electron ionization probability  $P(b)$  are found to be in reasonable agreement with measured cross sections for single and multiple ionization of neon by projectiles of various charges at a velocity corresponding to 1 MeV/amu. Furthermore, using an analytic fit to the SCA single-electron ionization probability tail we show that the charge dependence of

few-electron ionization cross sections stems from the binomial distribution. The exact functional form of the single-electron ionization probability  $P(b)$  has only a small effect.

In Sec. II we briefly describe the independent-electron approximation using the binomial distribution of ionization probabilities and interrelate exclusive and inclusive cross sections. In Sec. III we develop calculations of exclusive cross sections for single and multiple ionization based on SCA calculations for  $P(b)$ . In Sec. IV we discuss the implications and limitations of our results.

Two new points are emphasized in this paper. First, when  $n \ll N$ , perturbation theory is applicable for calculations of total cross sections, even if the single-electron ionization probability  $P$  is sometimes close to unity. Second, the binomial distribution gives a constant ratio of double- to single-ionization cross sections when  $q$  is large.

## II. THEORY

In the independent-electron approximation, whatever happens to any one electron does not affect any of the other electrons, i.e., there is no correlation. In atomic scattering this approximation may be derived<sup>16,17,26</sup> assuming that (i) the projectile acts as a point charge, (ii) the internuclear motion may be treated classically, independent of the electronic activity, and (iii) the electron-electron Coulomb interactions are approximated by single-electron potentials. To obtain the usual binomial distributions it is additionally assumed that (iv) Pauli

exclusion effects may be ignored and (v) all electrons with a given shell have the same probability  $P$  for ionization. Studies concerning assumptions (ii), (iii), and (iv) may be found elsewhere.<sup>7-10,26</sup> Assumption (v) may be avoided by using multinomial distributions, where each electron has a different ionization probability.

In this paper we adopt all of the above simplifying assumptions, and in addition we ignore electron capture and excitation. If one wishes to extend our results to include capture and excitation, one may replace  $P = P_{\text{ion}}$  by  $P = P_{\text{ion}} + P_{\text{cap}} + P_{\text{exc}}$  and follow the development below, using a different charge scaling for  $P_{\text{cap}}$ . For  $N$  electrons with the same single-electron ionization probability  $P$ , the binomial distribution is formed by the expansion

$$1 = [P + (1-P)]^N = \sum_{n=0}^N \binom{N}{n} P^n (1-P)^{N-n}. \quad (1)$$

The exclusive probability of ionizing exactly  $n$  of  $N$  electrons is  $\binom{N}{n} P^n (1-P)^{N-n}$  and the inclusive probability is  $\sum_{n=0}^N \binom{N}{n} P^n (1-P)^{N-n}$ . The total cross section for ionization of exactly  $n$  of  $N$  electrons is found by integrating the exclusive probability over impact parameters, namely,

$$\sigma_n = 2\pi \binom{N}{n} \int_0^\infty P(b)^n [1-P(b)]^{N-n} b db. \quad (2)$$

For systems with  $s$  different shells, one may take

$$\begin{aligned} 1 &= [P_1 + (1-P_1)]^{N_1} [P_2 + (1-P_2)]^{N_2} \cdots [P_s + (1-P_s)]^{N_s} \\ &= \sum_{n_1=0}^{N_1} \binom{N_1}{n_1} P_1^{n_1} (1-P_1)^{N_1-n_1} \sum_{n_2=0}^{N_2} \binom{N_2}{n_2} P_2^{n_2} (1-P_2)^{N_2-n_2} \cdots \sum_{n_s=0}^{N_s} \binom{N_s}{n_s} P_s^{n_s} (1-P_s)^{N_s-n_s}. \end{aligned} \quad (3)$$

Since  $[P_j + (1-P_j)]^{N_j} = 1$ , summing over all the final states of electrons in a shell gives a factor of unity, and as a consequence that shell may be ignored. Such electrons are passive electrons. If  $P_k \ll 1/N_k$ , then the probability for ionizing a single electron in the  $k$ th shell is simply  $\binom{N_k}{1} P_k (1-P_k)^{N_k-1} \sim N_k P_k$ .

In this paper we consider only one shell with a total of  $N$  electrons. However, we shall consider the effects of large probabilities, corresponding to projectiles with a large charge  $q$ . Consequently, we may not ignore factors of  $(1-P)$ . In this case one may show that

$$NP \geq \sum_{n=1}^N \binom{N}{n} P^n (1-P)^{N-n} \geq NP(1-P)^{N-1}. \quad (4)$$

This is shown as follows. Consider

$$D = NP - \sum_{n=1}^N \binom{N}{n} P^n (1-P)^{N-n} = NP - [1 - (1-P)^N].$$

Now, since  $0 \leq P \leq 1$ , we have that

$$\frac{dD}{dP} = N[1 - (1-P)^{N-1}] > 0.$$

Since  $D=0$  at  $P=0$ , then the first inequality is proved for  $0 < P \leq 1$ . The second inequality is trivial since the right-hand side is the first in a sum of non-negative terms from the middle expression in Eq. (4).

This means that the probability  $NP$  commonly used for single ionization is greater than the correct inclusive sum for ionization of at least one electron. This sum in turn is greater than the exclusive single-ionization probability for electrons within a given shell. If  $P$  is small, then  $(1-P)^N \approx 1 - NP$ , and for  $P \ll 1/N$ , then all three quantities reduce to the same value, namely,  $NP$ .

As we later use perturbation theory to calculate single-electron-ionization probabilities, it is important to verify that small values of the probability make the largest contribution to the total exclusive ionization cross section. The probability function for  $n$ -electron ionization by a highly charged projectile peaks at a certain impact parameter  $b_e$ , defined by

$$\frac{d}{db} \left[ \binom{N}{n} [P(b_e)]^n [1-P(b_e)]^{N-n} \right] = 0.$$

The extremum condition

$$[n - NP(b_e)][P(b_e)]^{n-1} [1-P(b_e)]^{N-n-1} \frac{dP}{db}(b_e) = 0,$$

yields for a maximum,

$$P(b_e) = \frac{n}{N}, \quad (5)$$

for any monotonically decreasing  $P(b)$ , i.e., if  $dP/db \neq 0$ . Since the integrand in Eq. (2) is weighted with a factor of  $b$ , most contributions to the total cross section come from  $b > b_e$  for which  $P(b) < n/N$ . The independent-electron approximation limits the maximum value of the single-electron-ionization probability contributing significantly to few-electron ionization. Perturbation theory is applicable for any value of  $P(b)$  so long as  $n \ll N$ , because contributions for large  $P$  are damped by a factor of  $(1-P)^{N-n}$ .

### III. CALCULATIONS

In this section we consider exclusive probabilities and cross sections for the ionization of  $n$  of a total of  $N$  electrons in a single atomic shell with single-electron-ionization probability  $P(b)$  using the binomial distributions discussed in Sec. II. We give emphasis to cases where the number of electrons  $N$  and the projectile charge  $q$  are both large. We shall work at high velocities where the projectile velocity is large compared to the initial orbit velocity of the target electrons. The experimental single- and double-ionization cross sections at this velocity are approximately linear in  $q$ . The experimental ratio of double- to single-ionization cross sections is independent of  $q$ . Also for large  $N$  and  $q$ , the exclusive probabilities for ionizing  $n$  electrons tend to occur in distinct impact-parameter windows.

#### A. SCA calculations

Let us now consider the impact-parameter dependence of the exclusive probability of ionizing  $n$  of  $N$  electrons using the SCA single-electron-ionization probabilities tabulated by Hansteen, Johansen, and Kocbach.<sup>25</sup> The exclusive probability is given within the binomial distribution by the probability function

$$P_N^n(b) = \binom{N}{n} P(b)^n [1-P(b)]^{N-n}. \quad (6)$$

A typical curve for the SCA  $P(b)$  is shown in Fig. 1, corresponding to protons on hydrogen at  $v=6.3$  a.u. The SCA  $P(b)$  was calculated using the SCA code<sup>25</sup> to generate probability values for impact parameters larger than the tabulated range. Values of  $P(b)$  for other projectile charges  $q$  and target charges  $Z$  may be found from the well-known scaling law<sup>25</sup>

$$P(q, Z, v; b) = \frac{q^2}{Z^2} P(1, 1, v/Z, Zb). \quad (7)$$

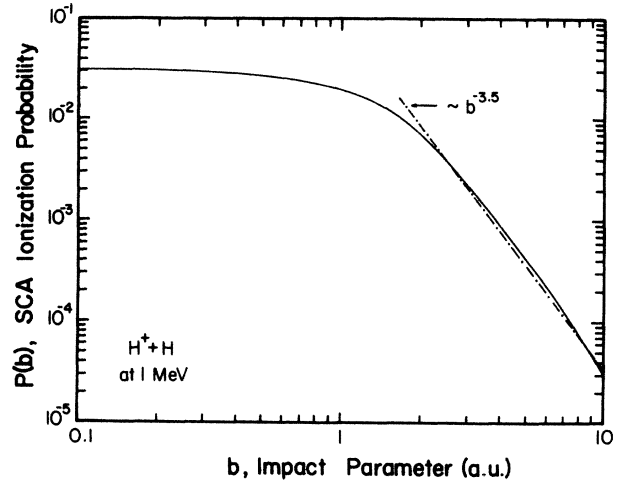


FIG. 1. SCA single-electron ionization probability  $P(b)$  for  $H^+ + H$  at 1 MeV (—). An analytical fit, Eq. (8), to the probability tail  $P(b) = Cb^{-3.5}$  (---).

For highly charged projectiles, this scaling law gives single-electron-ionization probabilities greater than 1. If  $P(q, b)$  actually were greater than 1, in violation of unitarity, then for some impact parameters the probability function in Eq. (6) would be negative, which is unphysical. Consequently, it is both necessary and reasonable to constrain  $P(q, b)$  to be no greater than 1 either by limiting  $P$  at  $P=1$  or by using a unitarized expression  $P_{\text{uni}} = 1 - e^{-P}$  for which some justification has been given.<sup>27</sup> Since in ionization of a multielectron target there is not much contribution except when  $P \ll 1$ , it is not important which method we use to limit  $P$ . We use the unitarized expression in the development of this subsection.

In Fig. 2 we show the impact-parameter dependence of the ionization probability function  $P_N^n(1, b)$  for  $n=1, 2$ , and 3 electrons in a shell with  $N=8$  electrons bombarded by 1 MeV protons. The probability functions become more sharply peaked about  $b=0$  as  $n$  increases. The factors of  $(1-P)$  have little effect since  $P_{\text{max}} \sim 0.03$ .

In Fig. 3 we show the exclusive probability functions for ionization of  $n=1, 2$ , and 3 electrons at  $v=6.3$  for  $q=10$ . Now there is a clear effect due to the  $(1-P)$  terms. In this case the single-ionization probability function is negligible except near  $b_e=3.6$ , where  $P(b_e)$  is about 0.125. This value  $P(b)$  indicates that application of the SCA method is not unreasonable, and error due to unitarization (or truncation at  $P=1$ ) is small. For double and triple ionization the probability function is peaked about  $b_e=2.8$  and 2.2, for which the values of  $P(b_e)$  are 0.25 and 0.375. The impact-parameter ranges contributing to single, double, and triple ionization are somewhat separate, forming windows in impact-parameter space for various degrees of multiple ionization. These windows form because at large impact parameters  $P(b)$  goes to zero while at small impact parameters  $[1-P(b)]$  goes to zero. These windows become more distinct as  $N$  increases.

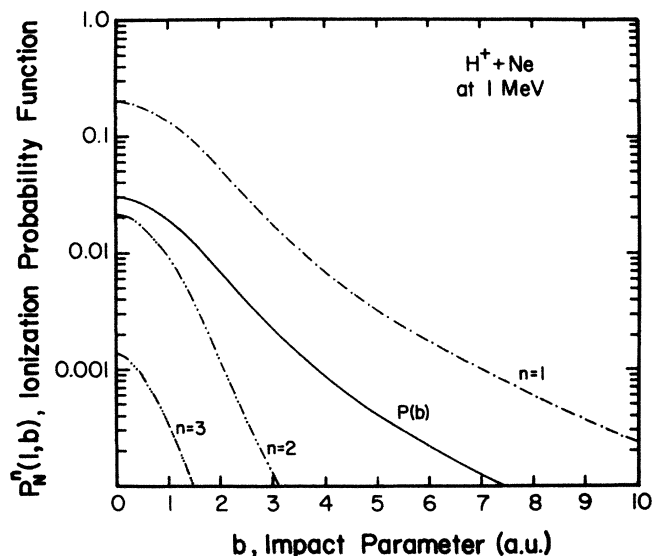


FIG. 2. Ionization probability function  $P_N^n(1,b)$  as a function of impact parameter for  $H^+ + Ne$  at 1 MeV/amu ( $v=6.3$  a.u.), for ionization of  $n=1, 2,$  and  $3$  from a total of  $N=8$  outer-shell electrons. The SCA single-electron-ionization probability  $P$  is represented by the solid curve. The exclusive ionization probability functions for  $n$ -electron ionization are  $P_8^1(b)$ , —·—;  $P_8^2(b)$ , —·—·—;  $P_8^3(b)$ , —·—·—·—.

In Fig. 4 we show total exclusive cross sections for  $n=1, 2,$  and  $3$  as a function of  $q$ , for 1 MeV/amu projectiles. They are compared with data of Cocke<sup>21</sup> and Gray *et al.*<sup>28</sup> for multiple ionization of neon by highly charged projectiles and with proton bombardment data of Wexler<sup>29</sup> and DuBois.<sup>30</sup> In our calculations we have very

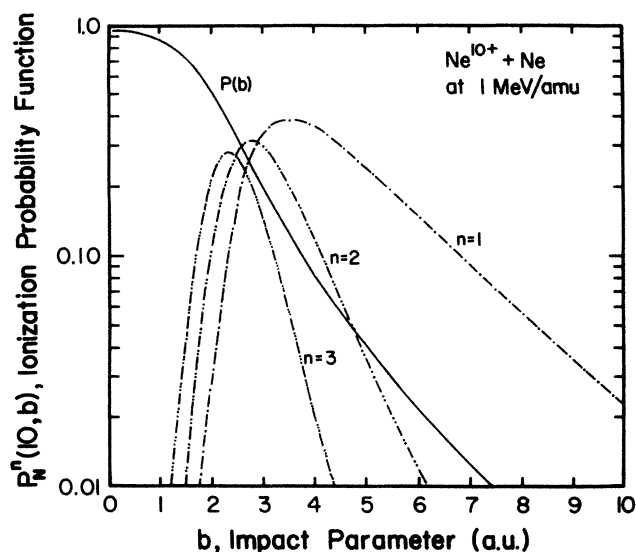


FIG. 3. Ionization probability function  $P_N^n(10,b)$  as a function of impact parameter for  $Ne^{10+} + Ne$  at 1 MeV/amu ( $v=6.3$  a.u.), for ionization of  $n=1, 2,$  and  $3$  from a total of  $N=8$  outer-shell electrons. The SCA single-electron-ionization probability  $P$  is represented by the solid curve. The exclusive ionization probability functions for  $n$ -electron ionization are  $P_8^1(b)$ , —·—;  $P_8^2(b)$ , —·—·—;  $P_8^3(b)$ , —·—·—·—.

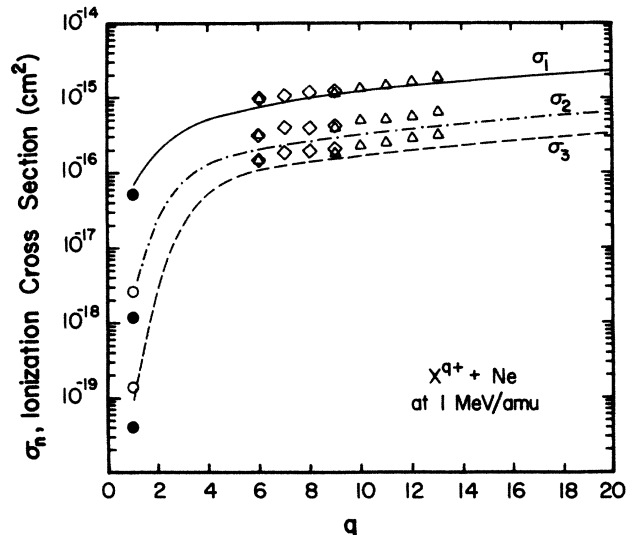


FIG. 4. Cross sections for ionization of  $n=1, 2,$  and  $3$  electrons out of  $N=8$  at 1 MeV/amu. SCA calculations:  $\sigma_1$ , —;  $\sigma_2$ , —·—;  $\sigma_3$ , —·—·—. Data:  $\Delta$ , Ref. 21;  $\diamond$ , Ref. 28;  $\circ$ , Ref. 29;  $\bullet$ , Ref. 30.

simply used  $K$ -shell SCA single-electron-ionization probabilities instead of  $L$ -shell SCA probabilities to illustrate that a rather simple model for  $P(b)$  can give good agreement with experimental results. Since hydrogenic wave functions are used, there is some uncertainty about the normalization of the SCA probabilities. Consequently, we normalize our probabilities to the total cross section for  $q=1$ , where the SCA results are at their best. Note that for large  $q$  even though the SCA  $P(q,b)$  and  $2\pi \int_0^\infty P(b)b db$  are both quadratic in  $q$ , the exclusive single- and double-ionization cross sections are approximately linear in  $q$ .

In Fig. 5 the ratios of double to single ionization and

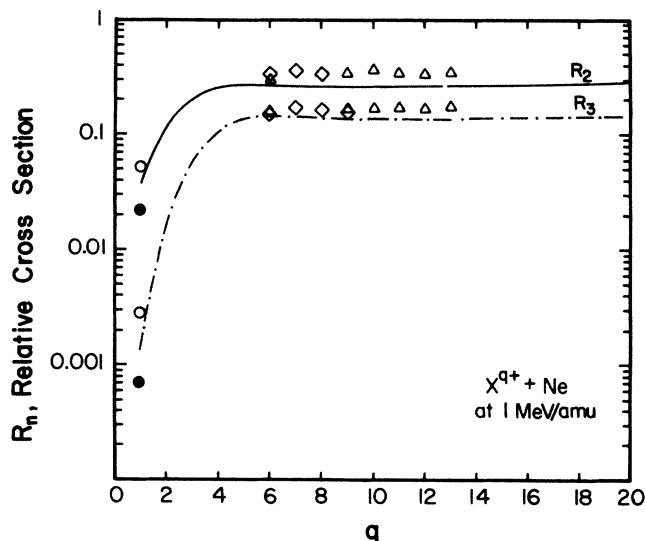


FIG. 5. Cross-section ratios  $R_2 = \sigma_2/\sigma_1$  and  $R_3 = \sigma_3/\sigma_1$  as a function of projectile charge at 1 MeV/amu. SCA calculations:  $R_2$ , —;  $R_3$ , —·—·—. Data:  $\Delta$ , Ref. 21;  $\diamond$ , Ref. 28;  $\circ$ , Ref. 29;  $\bullet$ , Ref. 30.

triple to single ionization,  $R_2$  and  $R_3$ , are plotted as a function of projectile charge  $q$  at 1 MeV/amu. These ratios increase approximately as  $q^2$  and  $q^4$ , respectively, for small- $q$  values. For  $q$  greater than 5 these ratios are practically constant.

In Fig. 6 the ratios of double to single ionization and triple to single ionization,  $R_2$  and  $R_3$ , are plotted as a function of projectile velocity  $v$  for  $q=12$ . The slow decrease in the calculated ratios is caused by a change in the power dependence [cf. Eq. (8)] of the tail of  $P(b)$  at increasing projectile velocity.

### B. Model calculations

In order to illustrate some properties of binomial distributions for large- $q$  projectiles, it is useful to develop an analytic fit to the SCA  $P(b)$  at large impact parameters. It was shown in Fig. 3 for  $q=10$  that the main contribution to few-electron ionization comes from collisions at large impact parameters where only the tail of  $P(q,b)$  contributes. The analytic form we have chosen for the single-electron-ionization probability is

$$P(b) = Cb^{-m} . \quad (8)$$

In Fig. 1,  $m=3.5$  gives a good fit to the large impact-parameter region as shown for  $q=1$ . As the projectile velocity increases,  $m$  decreases slowly, giving relatively more ionization at large impact parameters. Here we truncate the scaled probability  $P(q,b) = q^2 P(b)$  such that  $P(q,b) = 1$  for  $b < b_{\min}$ , where  $b_{\min}$  is the largest value of  $b$  such that  $P(q,b) = 1$  in the SCA approximation. The purpose of this simple analytical treatment is to show

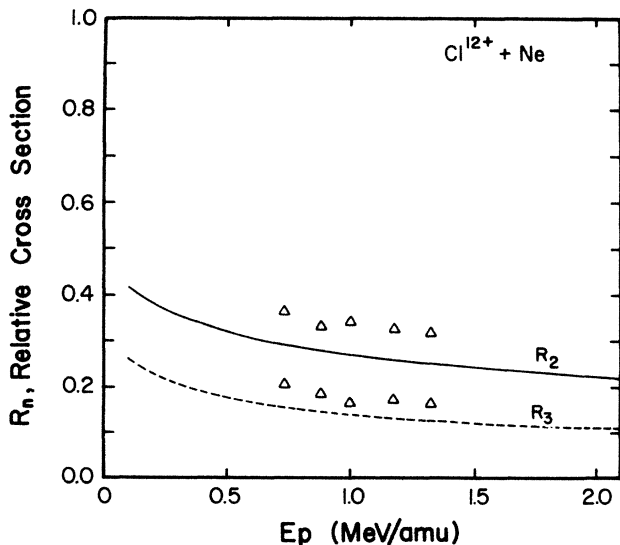


FIG. 6. Cross-section ratios  $R_2 = \sigma_2/\sigma_1$  and  $R_3 = \sigma_3/\sigma_1$  as a function of projectile energy for  $q=12$ . SCA calculations:  $R_2$ , —;  $R_3$ , - - -. Data:  $\Delta$ , Ref. 21.

that some properties of multiple ionization depend only slightly on the functional form of the single-electron-ionization probability. These properties are a general consequence of the binomial distribution resulting from the independent-electron approximation.

Let us first consider the ratio of double to single ionization at large  $q$  using our simple fit to the SCA  $P(b)$ , namely,

$$\begin{aligned} R_2 &= \frac{\sigma_2}{\sigma_1} = \frac{2\pi \binom{8}{2} \int_0^\infty P(q,b)^2 [1-P(q,b)]^6 b db}{2\pi \binom{8}{1} \int_0^\infty P(q,b) [1-P(q,b)]^7 b db} \\ &= \frac{1}{2} Cq^2 \frac{\int_{b_{\min}}^\infty (1-q^2 Cb^{-m})^6 b^{-2m+1} db}{\int_{b_{\min}}^\infty (1-q^2 Cb^{-m})^7 b^{-m+1} db} . \end{aligned}$$

Integrating the denominator by parts and using the fact that  $1 - q^2 P(q, b_{\min}) = 0$  yields,

$$R_2 = \frac{(m-2)}{2m} . \quad (9)$$

Note that this ratio is independent of  $q$  and  $N$ . But  $q$  must be large for Eq. (8) to hold in the impact-parameter region where single and double ionization are large. It may be similarly shown that

$$R_3 = \frac{\sigma_3}{\sigma_1} = \frac{(m-2)(m-1)}{3m^2} . \quad (10)$$

Again this is independent of  $q$  and  $N$ . Constant values for  $R_2$  and  $R_3$  may be similarly obtained by an exponential fit<sup>31,32</sup> to  $P(b)$ , i.e.,  $P(b) = P_0 e^{-ab}$ . For  $m=3.5$  these ratios are  $R_2=0.21$  and  $R_3=0.10$ , while the experimental ratios are 0.34 and 0.17, respectively. The SCA calculations of Sec. II A shown in Fig. 5 are in somewhat better agreement with experiment than these simpler calculations.

Next we evaluate the total cross section for single ionization using Eq. (8) for  $P(b)$ . The integration starts at  $b_{\min}$  because  $1-P=0$  for  $b < b_{\min}$ . Errors in  $P(q,b)$  near  $b_{\min}$  are not important, since both  $[1-P(q,b)]$  and the weighting factor of  $b$  in Eq. (2) tend to emphasize the regions of impact parameters where  $P$  is small and Eq. (8) is applicable. Now, using Eq. (8) in Eq. (2), we have for  $n=1$

$$\begin{aligned} \sigma_1 &= 2\pi \int_0^\infty P_N^1(q,b) b db \\ &= 2\pi \left[ \begin{matrix} 8 \\ 1 \end{matrix} \right] Cq^2 \int_{b_{\min}}^\infty (1-q^2 Cb^{-m})^7 b^{-m+1} db . \end{aligned}$$

Using integration by parts and the definition of  $b_{\min}$ , we obtain,

$$\sigma_1 = \sigma_0 q^{4/m}, \quad (11)$$

where

$$\sigma_0 = 2\pi N m^N \frac{N!}{\prod_{i=1}^{n+1} (im - 2)} C^{2/m}.$$

Since  $m$  is close to 4, the single-ionization cross section  $\sigma_1$  is nearly linear in  $q$  at large  $q$ , consistent with the behavior seen in Fig. 4. From the above analysis for  $R_2$  and  $R_3$ ,  $\sigma_2$  and  $\sigma_3$  have the same  $q$  dependence for large  $q$ .

Now we consider the impact-parameter dependence of the exclusive ionization cross sections for  $n < N$  and large  $q$ . Solving Eq. (8) for  $b_{\min}$  defined by  $P(b_{\min})=1$ , we have

$$b_{\min} = C^{1/m} q^{2/m}. \quad (12)$$

Next we find the impact parameter  $b_e$  where the exclusive probability function for ionizing  $n$  electrons peaks. Solving Eq. (5) for  $b_e$ ,

$$P(q, b_e) = Cq^2 b_e^{-m} = \frac{n}{N},$$

yields

$$b_e = \left( \frac{CN}{n} \right)^{1/m} q^{2/m}. \quad (13)$$

Note that both  $b_{\min}$  and  $b_e$  increase as  $q$  increases, having the same functional form  $q^{2/m}$ . The peak positions for various degrees of ionization tend to separate as  $n/N$  increases. These can be seen in Fig. 7 in which  $b_e$  is plotted as a function of projectile charge state.

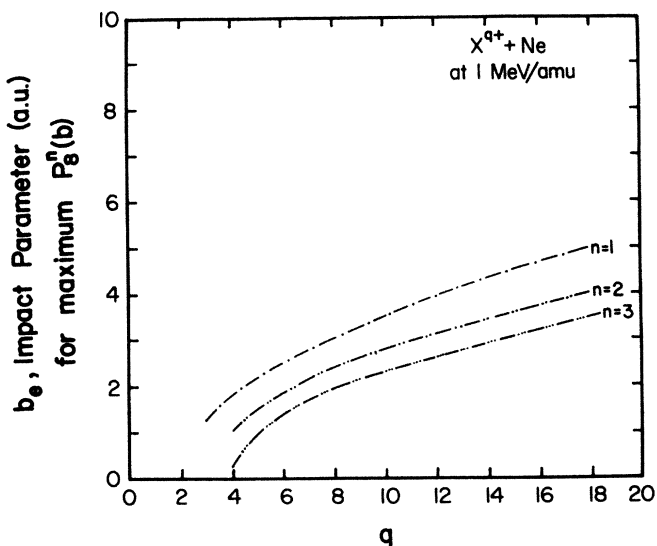


FIG. 7. Impact parameter  $b_e$  corresponding to the maximum of the probability function [cf. Eq. (5)], as a function of projectile charge for ionization of  $n=1, 2$ , and  $3$  from a total of  $N=8$  electrons.

#### IV. DISCUSSION

We have used perturbation theory within the independent-electron approximation to evaluate cross sections for multiple ionization of multielectron targets for high projectile charge  $q$ . This includes some contributions from large  $P(q, b)$ . The resulting error is small if  $n/N \ll 1$ . For example, for single ionization in a system with eight identical electrons, the maximum contribution comes from  $b_e$  where  $P(q, b_e) = n/N = 0.125$ . Since the integrand in Eq. (2) is weighted with a factor of  $b$ , most contributions to the total cross section come from  $b > b_e$ , where  $P(q, b) < 0.125$ . Numerical estimates using truncated and unitarized  $P(q, b)$  give an error of less than 10% due to the improper use of perturbation theory in evaluating total cross sections. It seems remarkable to us that perturbation theory indeed seems to work so well in the high- $q$  limit. Similarly we find that for double ionization with  $N=8$  that  $P(q, b_e)$  is 0.25 and the error introduced in the total cross section is less than 20%. The error in the cross sections increases with  $n$  and reaches about 40% for  $\sigma_3$ . The total cross sections monotonically decrease as  $n$  increases so that the large- $n$  cross sections never dominate the inclusive total cross section. This may not be the case if electron capture is included or if molecular orbital effects<sup>33</sup> are important. However, in the experimental systems we have used, capture contribution to few-electron ionization is negligible. This is evident from the work of Gray *et al.*,<sup>28</sup> on the collision system  $F^{9+} + Ne$  at 1 MeV/amu, in which the capture contributions for doubly and triply charged recoils was less than 1% and 10%, respectively. By using the independent-electron approximation we have ignored correlation and shakeoff effects.<sup>34</sup>

The functional dependence of  $P(q, b)$  selects the range of impact parameters which contribute to the exclusive ionization cross section, resulting in a window for ionization of  $n$  electrons. These windows<sup>1,19</sup> occur such that the deep inelastic collisions cause multiple ionization and the soft collisions contribute primarily to single ionization.

Using the independent-electron approximation we have shown that the ratio of double to single and triple to single ionization is independent of  $q$  for large  $q$ . Thus a ratio of double to single ionization independent of  $q$  (Refs. 35–37) does not always indicate a breakdown of the independent-electron approximation. For small  $q$  the ratio of double to single ionization does vary as  $q^2$ . In these cases where  $q$  is small, i.e.,  $P \ll n/N$ , a value of  $R_2$  which is independent of  $q$  is not consistent with the independent-electron approximation. In those cases<sup>35–37</sup> some other mechanism for double ionization, such as a shakeoff, may be dominating over independent interaction of each electron with the projectile. In the shakeoff mechanism  $R_2$  is independent of velocity  $v$  while the binomial distribution at large  $q$  the ratio  $R_2$  decreases slowly with increasing  $v$ .

Our present calculations are very simple and as noted above many improvements are possible. On the other hand, since the method is so simple it may be easily applied to more complicated systems. The wave functions

of outer shells of many systems decrease exponentially and may be regarded as isotropic, at least on the average. Applying the SCA  $P(b)$  may not be unreasonable for many atoms and molecules, whenever correlations may be ignored. In the region of intermediate  $q$  where the cross sections change from a  $q^2$  dependence to an approximately linear dependence on  $q$ , there are few data and further observations may be useful. For broad qualitative features of exclusive ionization cross sections our method may be useful and should be tested further.

## V. SUMMARY

Binomial distributions have been applied to collisions of atoms with fast projectiles of arbitrary charge  $q$ . The factor  $NP$ , commonly used for ionization of a single electron in a shell containing  $N$  electrons with the same single-electron ionization probability  $P$ , is an upper bound to the inclusive ionization probability. If  $P \ll 1/N$ , then both expressions are approximately equal.

We find that perturbation may be applied for exclusive ionization probabilities if  $n/N \ll 1$ , because factors of  $(1-P)^{N-n}$  suppress contributions where the single-electron ionization probability  $P(q,b)$  is large. Specifically, when  $P(q,b)$  is not smaller than  $n/N$ , the maximum contribution to the exclusive probability for ionizing  $n$  of  $N$  electrons occurs at  $P(q,b_e) = n/N$ . when  $N \gg 1$  and  $q$  is large, each of the exclusive ionization probabilities is contained within somewhat separate impact-parameter windows. For large  $q$  the ratio of double to single-total-ionization cross sections is independent of  $q$ . The single-ionization cross section is approximately linear in  $q$  in agreement with data for the ionization of neon by projectiles with  $q = 6-13$  at 1 MeV/amu.

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