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Direct-Transition Features in Stripping Collisions of Heavy Neutral Atoms and Ions*

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Previously published experimental data on single-electron stripping for over 100 different neutral-neutral and almost 100 ion-neutral collision pairs are compared with Firsov's statistical ionization model. Reasonable agreement is found for a number of reactions, in particular for collisions between some rare-gas atoms. However, marked disagreement exists for many other collisions. From a model assuming a single-step transition of the ionized electron from the bound state into the continuum, a scaling law is developed semiempirically which is found able to reduce a large majority of these published data to within a factor of two of a general cross-section curve. Major disagreements with this model occur only in cases which are particularly well described by the Firsov model.

I. INTRODUCTION

Over the last 15 years, extensive experimental data have been accumulated on ionization and stripping in collisions of heavy atoms, molecules, and/or ions. However, possibly with the exception of inner-shell ionization, comparatively little understanding of these processes has developed. This is particularly true for single-electron stripping processes in collisions between heavy neutral atoms. In spite of a long list of experimental results,[1–46] only very few theoretical calculations of the corresponding cross sections have been published so far. Good *a priori* calculations only exist for the lightest collision partners H and/or He. If heavier collision partners are involved, it is generally assumed that the ionization process proceeds by means of a large number of crossings and pseudocrossings of the interatomic-potential curves through which the electronic wave function "diffuses" during the encounter. Then, ionization supposedly occurs by auto-ionization from states above the ionization threshold. Correspondingly, any theoretical treatment appears to be necessarily statistical in nature. So far, three independent approaches in this direction have been published by Mittleman and Wilets,[47] by Russek and co-workers[48] and by Firsov.[49] Of these, only Firsov's paper arrives at actual predications for total cross sections independent of unknown phenomenological parameters. Since its publication, Firsov's formula has been compared and found to agree reasonably well with a limited set of experimental data, mostly involving collisions of rare-gas particles, and therefore is relatively widely quoted and accepted.

In this paper, a much wider range of experimental results on the stripping of neutral-atom and ion projectiles in collisions with neutral target atoms is collected, involving many projectiles from hydrogen to uranium and target gases ranging from rare gases to some alkalies and some diatomic gases. Comparing this set of data with Firsov's formula, a reasonable agreement of experimental and theoretical cross-section values is found around the maxima of the cross-section curves. However, considerable disagreement is observed in many cases where cross sections have been measured at smaller velocities, with the experimental values dropping much faster than predicted at the smaller energies. The latter behavior seems to indicate that, in these cases, the ionization process is dominated by a single large-step transition of the electron, from the ground state either directly into the continuum or into a highly excited state, rather than by a gradual statistical excitation.

To investigate this point, a scaling law, more adequate for direct ground-state-to-continuum transitions, is developed semiempirically with the aim to reduce these cross sections to a general functional dependence. The corresponding derivations are based on the assumption that the electron transition occurs simply on account of the time-dependent perturbation of the projectile by the target atom or molecule. Because of a general lack of detailed knowledge, in addition a number of somewhat arbitrary assumptions are introduced

concerning the time dependence and strength of the interaction, the corresponding effective interaction distance, and the relative probabilities for ionization from subshells having various angular momenta. The cross-section scaling law resulting from this rather coarse approach is able to reduce, to within a factor of about two, over 80% of the collected experimental data to a general cross-section curve which approximately coincides with the cross-section curve for the stripping of hydrogen atoms. Major discrepancies between this formula and the existing data are limited to collisions of rare-gas atoms which seem to be better described by Firsov's formula. The plausibility of the above assumptions and possible modifications expected from future detailed studies are discussed.

II. EXPERIMENTAL DATA

To provide as wide a base as possible for any comparison, a literature search for experimental results on stripping cross sections published over the last 20 years was conducted. In this search, single-electron-loss cross sections $\sigma_{i,i+1}$ for more than 200 different neutral-neutral and ion-neutral collision pairs were discovered and were included in this analysis.[50]

All these data were obtained by one of two experimental methods: (a) by creation of a neutral beam from an ion beam by passage through a separate charge-transfer cell, removal of the remaining ions from the beam, passage of the neutrals through the collision chamber and subsequent magnetic separation of the resulting neutrals and ions; or (b) by passage of the ion beam itself through the target gas and measurement of the various charge-state percentages as a function of the target thickness.

In most cases, intensities of the various charge states of the projectiles were determined assuming identical secondary electron emission coefficients—an assumption which, in general, appears reasonable. In addition to the usual error possibilities in the determination of various experimental parameters, such as pressures, currents, etc., mainly two specific error sources are possibly important: the scattering of the projectile in the stripping reaction and contributions from metastable projectiles. Because of scattering, some of the reaction products may not be able to reach the detector if too narrow a detection geometry is used; the resulting cross section would appear too low, particularly at low beam energies. A sizable number of authors seem to have devoted some thoughts and actual rough checks on this point. Only a comparatively few papers (e.g., Refs. 2, 22, 39, 51, 52) state explicit results on the scattering in stripping collisions either in the form of actual angular distributions or in the form of percentages of reaction products found in certain angular intervals. From these data, it appears that the scattering angles generally increase with the ionization energy of the projectile and with the atomic numbers of target and projectile. In addition, the cone angle into which a certain percentage of the stripped particles is scattered seems roughly proportional to between $1/E$ to $1/E^2$. As an example, the product of beam energy and cone angle for 50% of the reaction products varies from about 1–2 keV deg for stripping of 1-keV hydrogen neutrals in various gases to over 50 keV deg for single-electron stripping of 50-keV Ar^+ ions. Because of a lack of more detailed knowledge in this area, and because of the insufficient description of beam geometries in most papers, errors which might have arisen are hard to estimate, though it appears that significant errors may have resulted in some cases for beam energies below, say, 20–50 keV. Above these energies, however, these errors probably were negligible in most cases.

Admixtures of metastable atoms in the beam would result in seemingly higher cross sections. In most cases, beyond hydrogen or helium, little is known about the production cross sections of metastable atoms or about their stripping cross sections. However, known cross sections for production of metastables are sufficiently small with respect to the corresponding ground-state charge-transfer cross sections that it appears reasonable to assume that the over-all contributions of excited atoms to the measured cross sections in general are small.

For a number of the most frequently used target gases, the top parts of Figs. 1–9 show a representative sample of published cross-section curves for single-electron loss of a wide range of neutral and ionic projectiles. Only for H and He projectiles did various measurements significantly overlap in energy range. In these cases, discrepancies between the various data generally were small and averaged curves were plotted for these reactions. Errors of most of the other data are quoted to be less than or about 10–20%. However, on account of the above error possibilities and from a comparison with cross sections for total electron production, we think that considerably larger errors may have occurred in some cases. This is particularly obvious in some of the cases where overlapping cross-section curves for O and N projectiles were reported by different authors[22,32] and where the more recent measurements, which also include losses of more electrons, were higher by a factor of up to three. In general, it appears that errors of the order of at least 15–30% for the energy dependence of most published cross sections and sometimes considerably larger errors of the absolute cross-section values may be present. How-

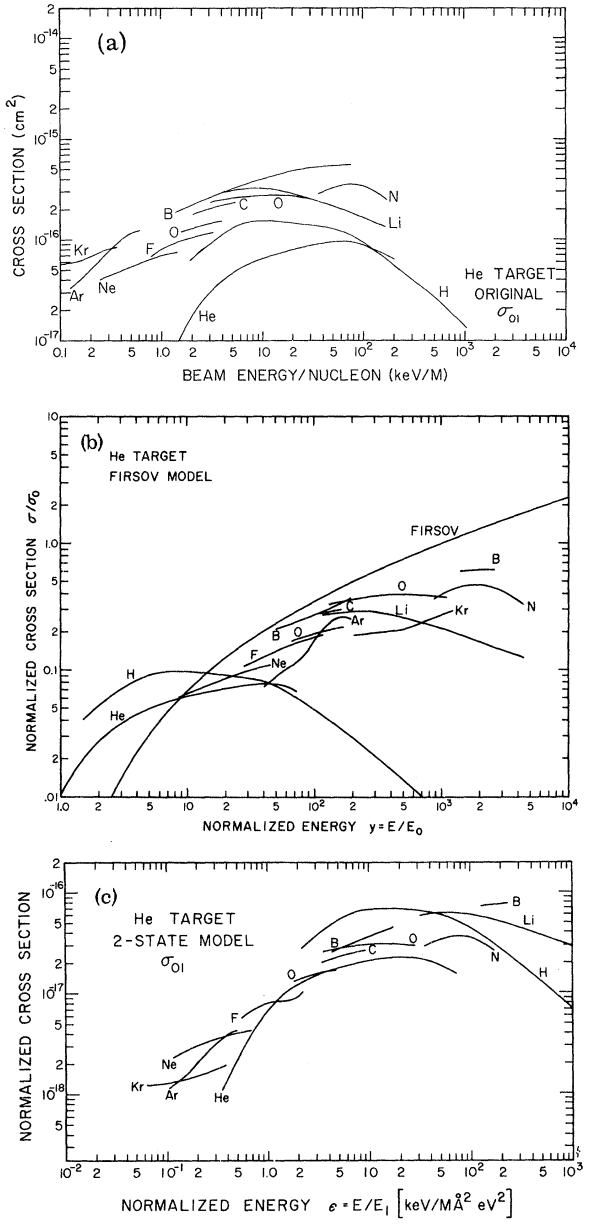

FIG. 1. Stripping of neutral atoms in helium.

ever, because of insufficient knowledge of all details, all cross sections are plotted as given in the original publications.

Since multielectron-loss cross sections generally are small compared with single-electron-loss cross sections, total ionization cross sections $q_0 = \sigma_{01} + \sigma_{02} \cdots$ are quoted in this paper as σ_{01}.

III. COMPARISON WITH FIRSOV THEORY

As mentioned, three independent statistical treatments of the ionization process have been published. Mittleman and Wilets[47] try to calculate directly the probability of diffusion of the electron wave function through statistically distributed excited states via curve crossings and then the ionization from the probability flux across the ionization threshold. Starting from Zener's[53] theory of transitions via pseudocrossings of molecular potential curves, they derive a general dependence of the normalized cross sections on the normalized beam energy. The corresponding scale factors are related to two parameters in the Zener theory, but no estimate of these parameters for actual cases is given. The shape of the general curve can be fitted reasonably to most experimental results; but, since only the average slope of the experimental curves can be used for fitting of the parameters, a comparison

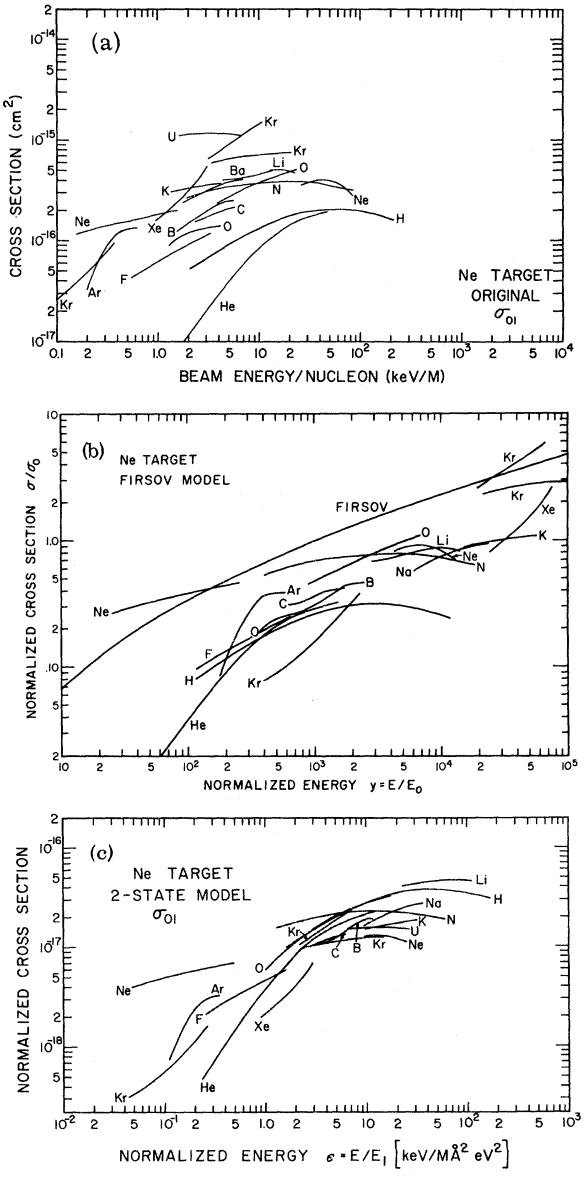

FIG. 2. Stripping of neutral atoms in neon.

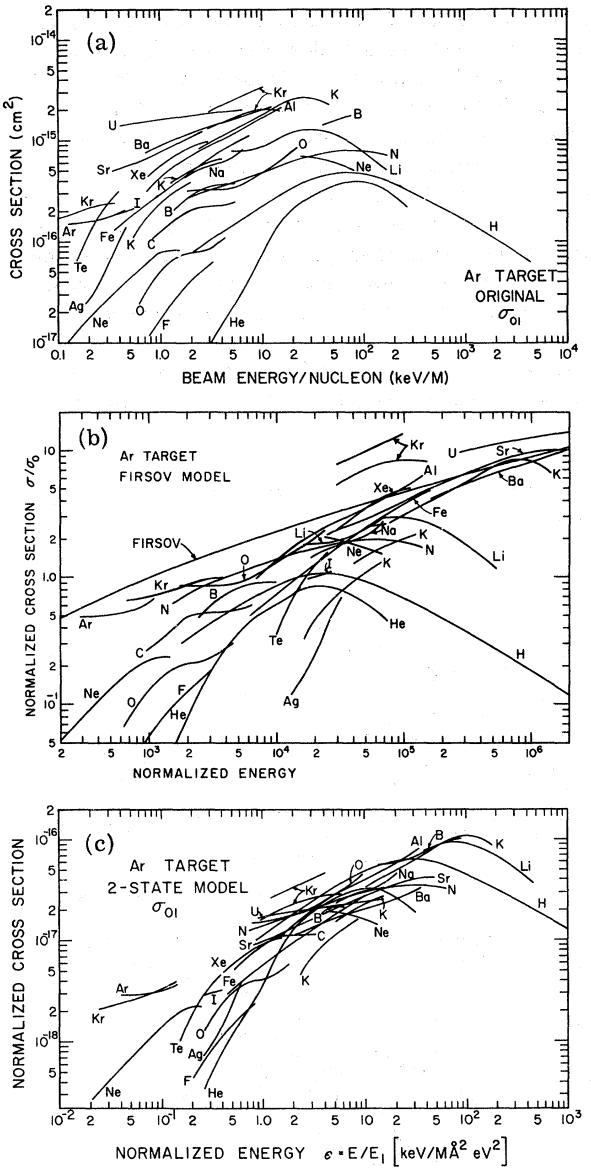

FIG. 3. Stripping of neutral atoms in argon.

between experiments and this theory becomes very ambiguous, particularly in view of the mentioned experimental uncertainties.

An entirely different statistical approach to this problem was undertaken independently by Russek and his co-workers[48] and by Firsov.[49] They both start out from a Thomas-Fermi model of the colliding atoms, calculate the "frictional heating" of this electron gas during the encounter, and then consider the ionization process as an evaporation of electrons from the heated electron gas. By a proper choice of parameters, Russek and co-workers are able to reproduce quite well some experimentally determined angular distributions of charge-state percentages resulting from a few high-energy small-impact parameter collisions of rare-gas particles. But no total cross sections have been calculated for more general cases.

Only Firsov's treatment leads to results which can be compared with the collected experimental data. His final result, determining the total electron production in heavy-particle collisions, is given in the form

$$\sigma = \sigma_0 \left[(v/v_0)^{1/5} - 1 \right]^2, \tag{1}$$

with

$$v_0 = \left[(23 \times 10^6)\, E_i/(Z_p + Z_t)^{5/3} \right] \text{ cm/sec}$$

and

$$\sigma_0 = \left[(33 \times 10^{-16})/(Z_p + Z_t)^{2/3} \right] \text{ cm}^2 ,$$

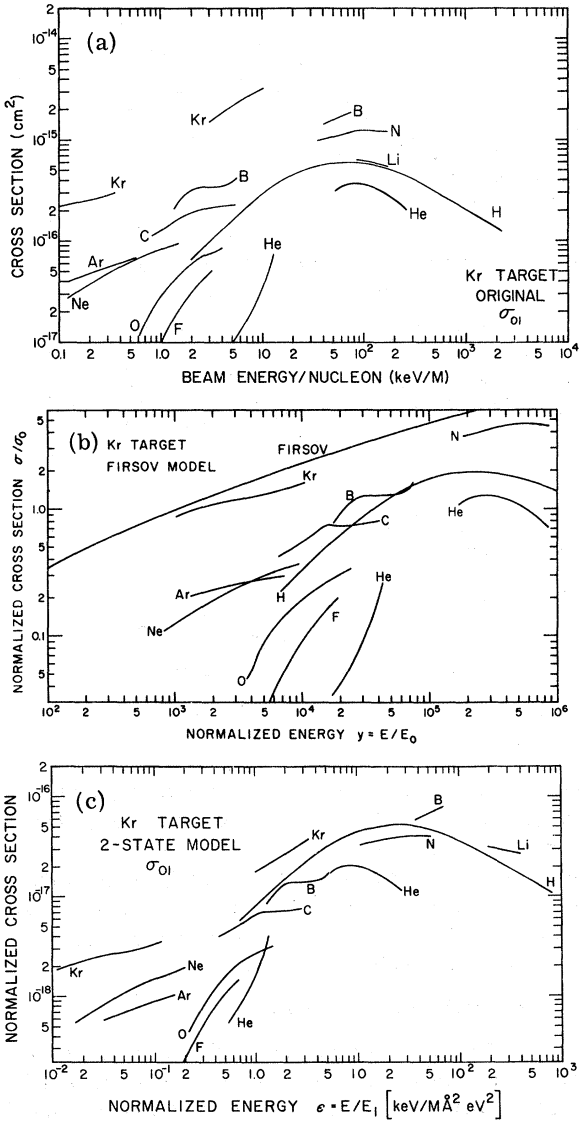

FIG. 4. Stripping of neutral atoms in krypton.

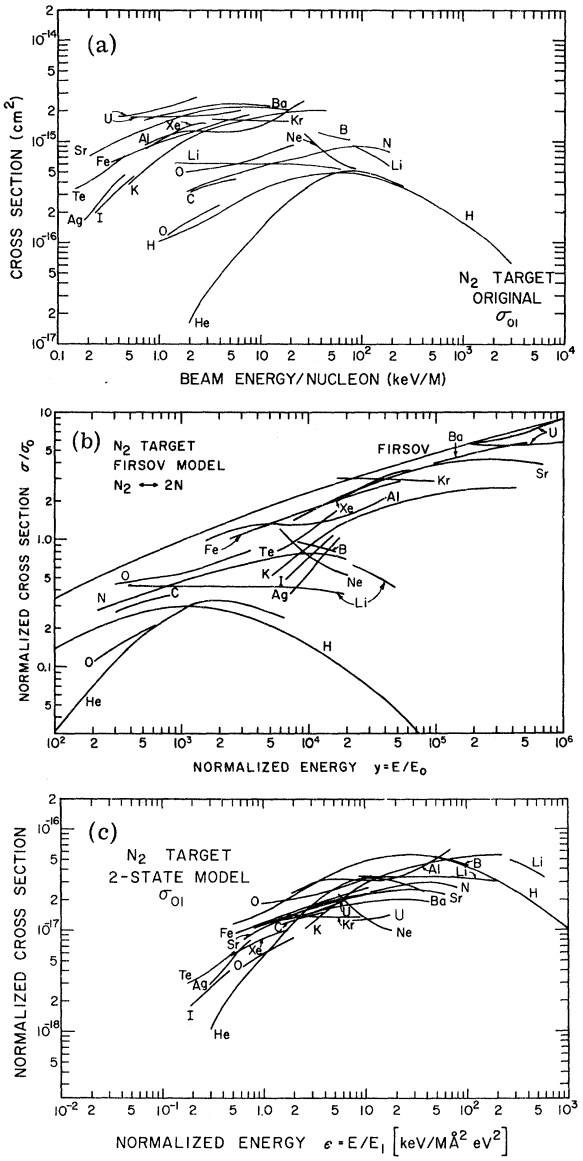

FIG. 5. Stripping of neutral atoms in nitrogen.

where v is the projectile velocity, E_i is the ionization energy in eV, and Z_p, Z_t are the atomic numbers of projectile and target atom, respectively. By introducing the mass M of the projectile and the beam energy E, one obtains instead

$$\sigma/\sigma_0 = [(E/E_0)^{1/10} - 1]^2 , \qquad (1a)$$

where

$$E_0 = \tfrac{1}{2}\, m_p M v_0^2 = 2.7\times10^2\, ME_i^2/(Z_p+Z_t)^{10/3}\ \text{eV},$$

M and m_p denoting atomic-mass number of the projectile and proton mass in g, respectively. This formula basically was derived for total electron production in neutral-neutral collisions, i.e., for stripping of the projectile particle plus ionization of the target particle. But it should also give a reasonable estimate for stripping cross sections alone, particularly when the ionization energies of both collision partners are comparable. Correspondingly, in the middle parts of Figs. 1–9, Firsov's result is plotted in this normalized form and compared with the experimental results for various targets and projectiles. For this purpose, in all cases, the ionization energy of the projectile was used in calculating E_0. Figures 10 and 11 show a composite of the original data and the correspond-

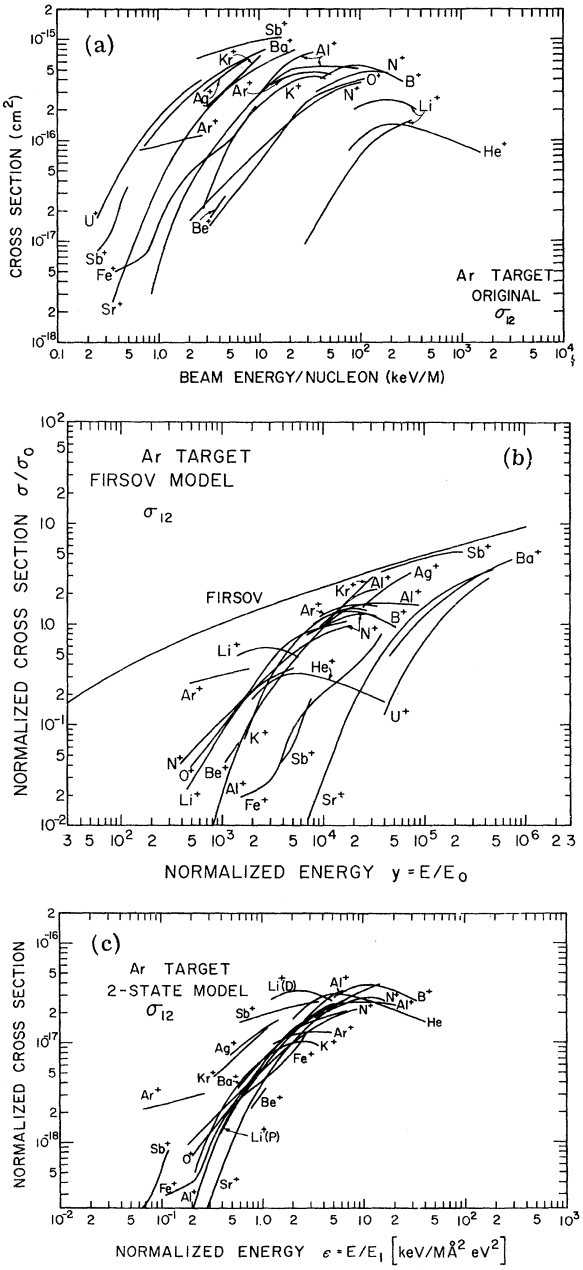

FIG. 6. Stripping of singly charged ions in argon.

ing Firsov plots for neutral-neutral collisions. They include also additional experimental data for other targets. Recognizing that on the average, a factor of about two should be subtracted from the theoretical Firsov predictions to account for target ionization, it seems that formula (1) gives a good estimate in a number of cases, in particular for collisions between rare-gas atoms.

However, two deficiencies are apparent. Formula (1) does not reproduce the falling high-energy parts of the experimental curves for neutral hydrogen. Also, in many cases, the decrease of the

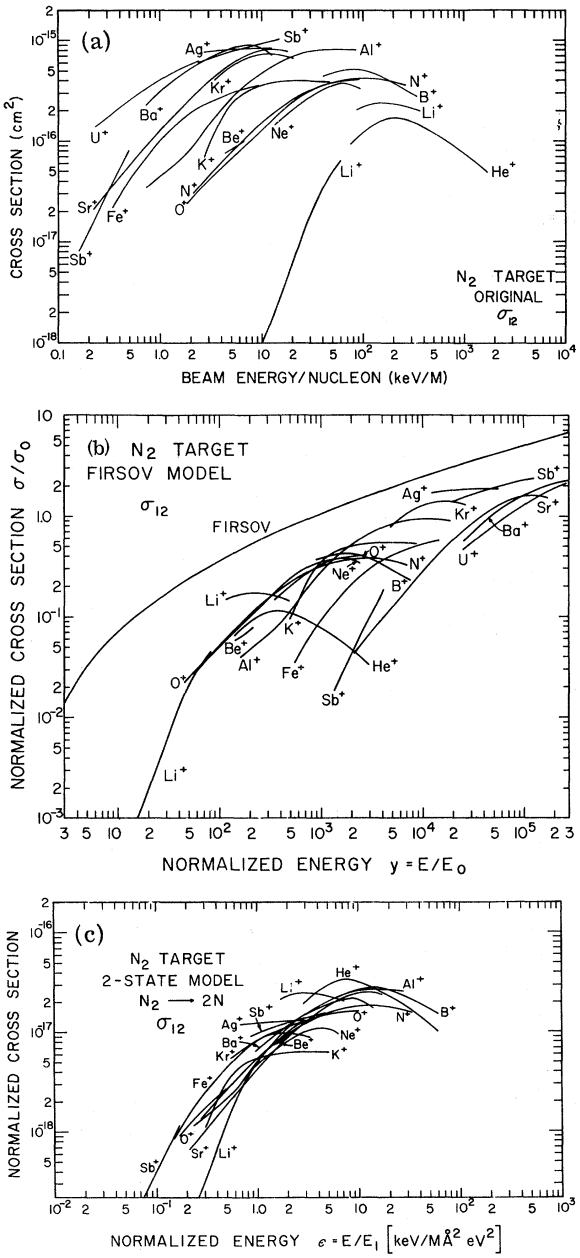

FIG. 7. Stripping of singly charged ions in nitrogen.

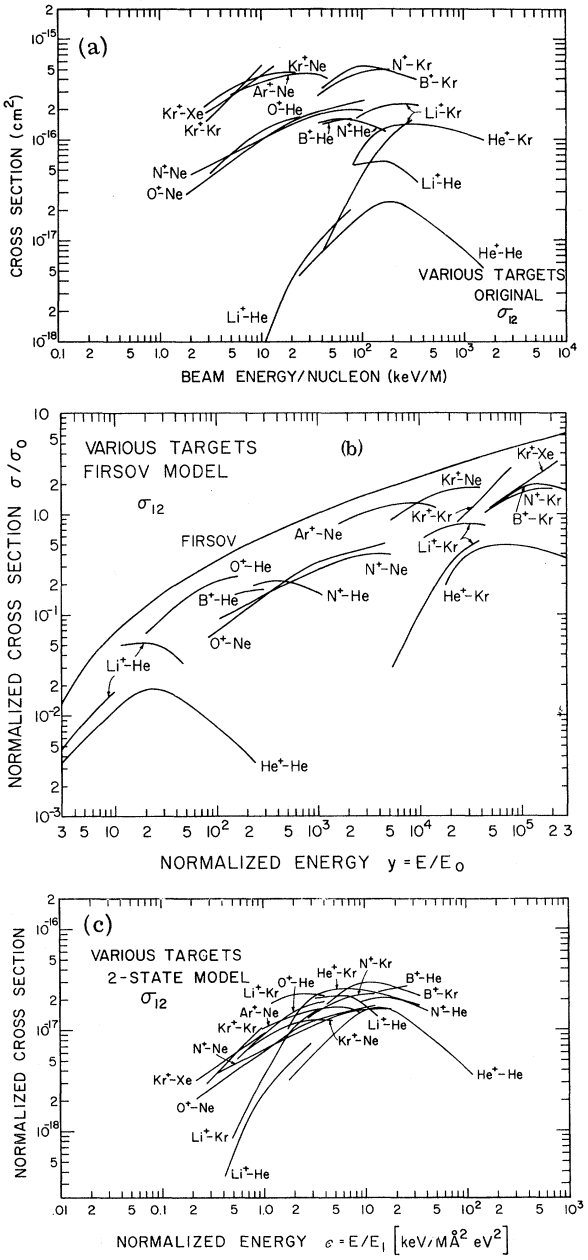

FIG. 8. Stripping of singly charged ions in various gases.

cross section toward smaller energies is clearly steeper than predicted by Firsov and leads to considerable discrepancies. Of these deficiencies, the first is not very serious, since the applicability of this formula to neutral hydrogen collisions is very doubtful because of the single electron and the correspondingly large ratio between the electron numbers of both collision partners. For more electron projectiles, a similar decrease in the single-electron-loss channel could be explained

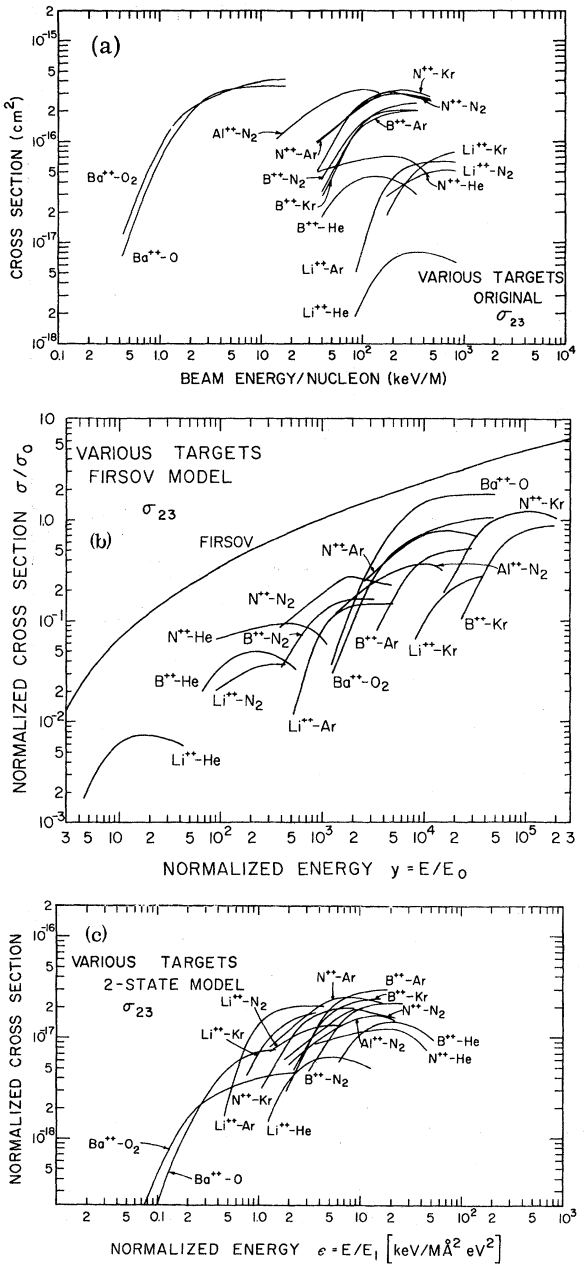

FIG. 9. Stripping of doubly charged ions in various gases.

by depletion of this reaction channel due to increases in the multielectron-loss channels.

However, more serious is the existence of the second mentioned deficiency. Its occurrence is not limited to collisions involving very light projectiles where the above arguments again could be used as an explanation, but this effect also appears in a number of reactions involving higher-mass projectiles and targets where the atomic number of the collision partners becomes comparable. In principle, this effect could be produced by the mentioned experimental errors due to scattering; but the scattering effects certainly are small at most of the beam energies in question.

A possible explanation is suggested by the physical model which this theory is supposed to describe, namely, the ionization of the electron via a large number of curve crossings. If only a direct transition between the ground state and the ionized state were involved, a comparatively faster decrease of the cross sections toward small energies is expected in the "near-adiabatic" energy range, i.e., for beam velocities below the cross-section maximum predicted by the Massey criterion.[54] The comparatively slow decrease of the cross section in Firsov's formula, even for velocities far below the Massey velocity, is a direct result of the assumed curve crossings which strongly reduce the effective energy gap between the two states and thus permit transitions to occur even at very small velocities. The fact that a number of cross sections decrease much faster than predicted by Firsov and are comparable with the steepness of the cross sections for hydrogen projectiles seems to indicate that the curve-crossing mechanism is not as effective in these cases as Firsov predicts.

IV. TWO-STATE MODEL

To investigate this point, we sought to develop a semiempirical scaling law (including a scaling of absolute cross-section values and of beam energies) which is more adequate for direct transition between the two states and which should be able to reduce to a general function as many cross-section curves as possible. Since the stripping reactions of neutral H and He atoms are the most likely to occur by direct transition, the corresponding cross sections were to obey this scaling law particularly well.

A. Scaling of Beam Energies

As a starting point, the stripping is considered as resulting from the time-dependent perturbation of the projectile by the target atom or molecule. Assuming that this perturbation is caused mostly by the electrostatic potential $V(|\vec{r}-\vec{R}_n(t,\rho)|)$ of the target particle [where $\vec{R}_n(t,\rho)$ and ρ denote the classically time-dependent position of the target nucleus in the frame of the projectile and the impact parameter, respectively], the probability of an electron transition from the bound state Ψ_i to a continuum state Ψ_f during a collision with impact parameter ρ is given in first order by

$$P_{if}(\rho) = (4\pi^2/\hbar^2)\,|\,W_{if}(\omega_{if},\rho)\,|^2\,, \tag{2}$$

where

$$\omega_{if} = (E_f - E_g)/\hbar \approx E_i/\hbar\,, \tag{3}$$

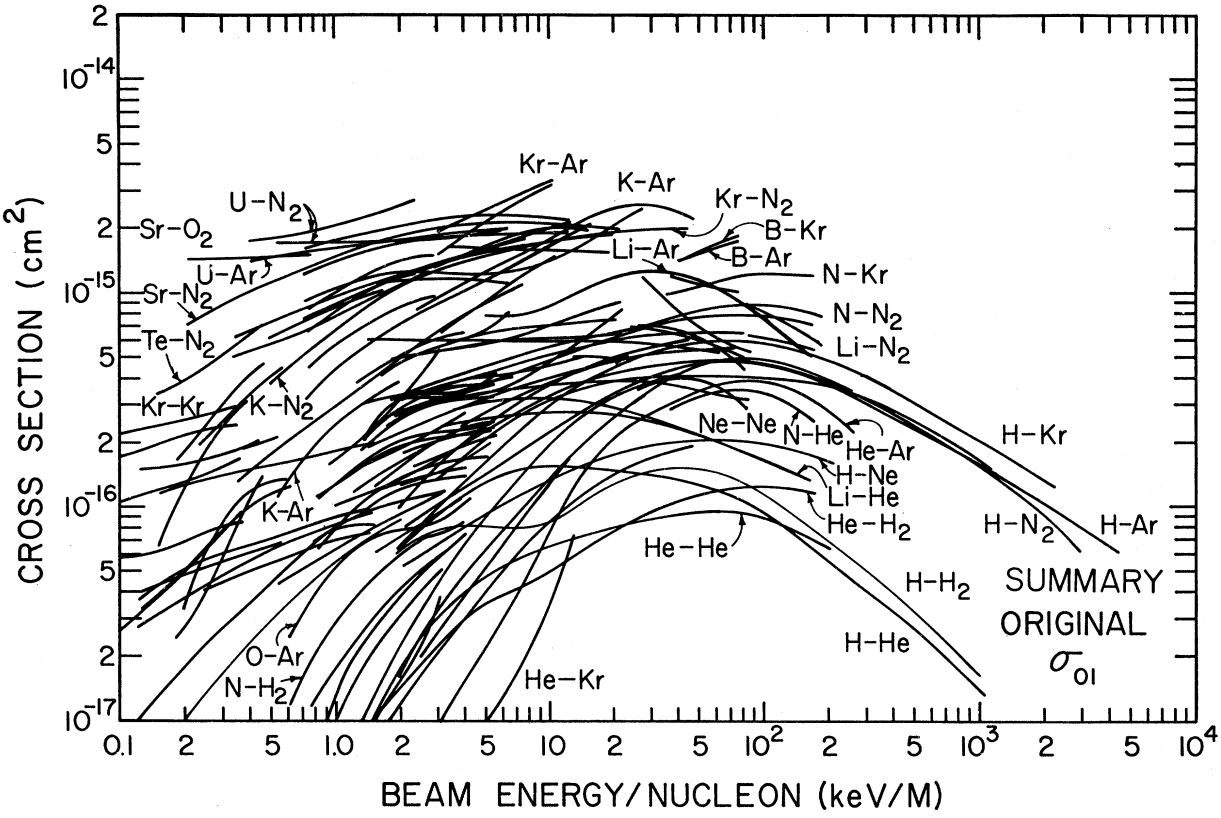

FIG. 10. Summary of original data on stripping of neutral projectiles in gases including all data used in analysis.

and

$$W_{if}(\omega,\rho)=(1/2\pi)\int_{-\infty}^{\infty} V_{if}(t,\rho)\,e^{i\omega t}\,dt \tag{4}$$

is the Fourier transform of the time-dependent matrix element

$$V_{if}(t,\rho)=\int \Psi_i^*(\vec{r})\,V(|\vec{r}-\vec{R}_n(t,\rho)|)\,\Psi_f(\vec{r})\,d^3r\,. \tag{5}$$

Integration of P_{if} over all impact parameters and all continuum states Ψ_f then yields the total cross section.

To obtain a scaling law for the beam energy, straight particle trajectories and constant particle velocity during the collision are assumed, as is reasonable for most impact parameters and particle energies in question. In this case, the energy dependence of $\vec{R}_n(t,\rho)$, and therefore also of $V_{if}(t,\rho)$, appears simply through the product (vt), and thus we obtain

$$W_{if}(\omega,\rho)=\frac{1}{2\pi}\,\frac{1}{\omega}\left(\frac{\omega}{v}\int_{-\infty}^{\infty} V_{if}(\xi,\rho)\,e^{i(\omega/v)\xi}\,d\xi\right), \tag{6}$$

i.e., the energy dependence of the final cross section should scale according to $v/\omega_{if}\sim v/E_i$.

Beyond that, we assume[55] that $V_{if}(t,\rho)$ can effectively be normalized for all collision pairs to an "interaction strength" V_0 and an "interaction time" τ which is related to an "interaction distance" R by $\tau=R/v$, i.e., we assume that we can write

$$V_{if}(t,\rho)=V_0(Z_p,Z_t,E_i,\ldots)\times F(vt/R(Z_p,Z_t,E_i,\ldots),\rho)\,,$$

where V_0 and R may depend on the various parameters of the collision partners, whereas the function F does not otherwise depend on them.

In this case, the total cross section for ionization of an electron from an original state having the ionization energy E_i^k becomes

$$\sigma_{\rm ion}(\Psi_k)=\sum_f 2\pi\int_0^{\infty} P_{kf}(\rho)\,\rho\,d\rho$$
$$=\sum_f \frac{2\pi V_0^2(Z_p,Z_t,E_i^k,\ldots)}{(E_i^k)^2}\left(\frac{E_i^k R}{\hbar v}\right)^2\int_0^{\infty}\rho\,d\rho$$
$$\times\left|\int_{-\infty}^{\infty} F(\eta,\rho)\,e^{i(E_i^k R/\hbar v)\eta}\,d\eta\right|^2, \tag{7}$$

where $\sum_f$ denotes a summation over all continuum states Ψ_f. In this formula, the beam energy enters only through the ratio $v/E_i^k R$ and a normalization of the beam energies E according to

$$\epsilon=E/E_1\,, \tag{8}$$

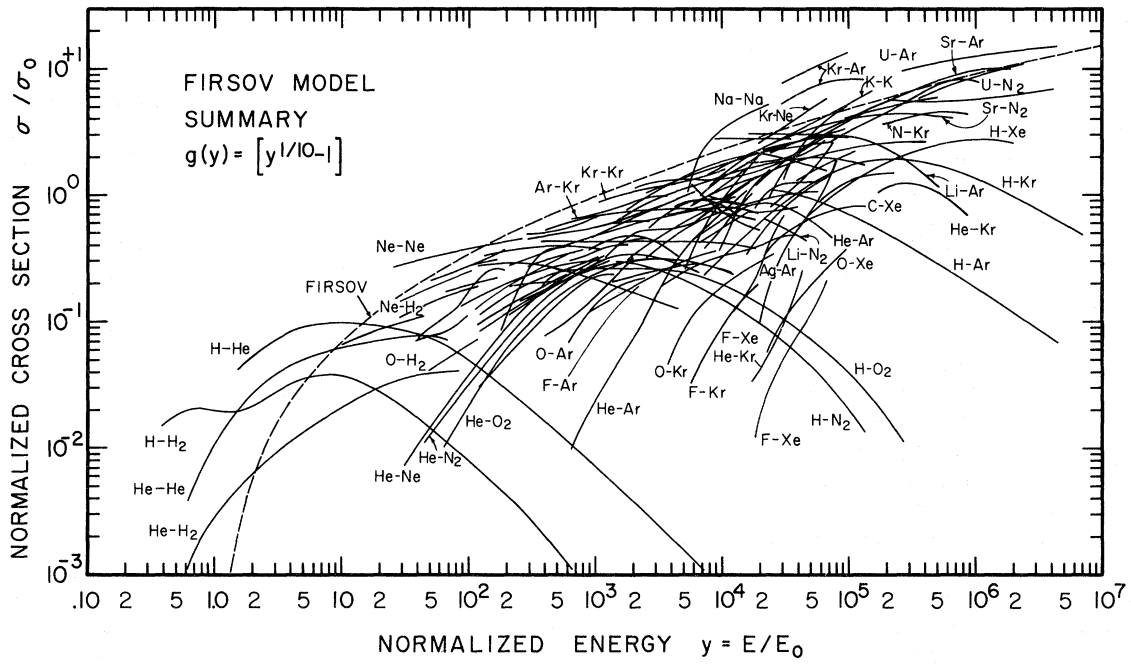

FIG. 11. Firsov graph of data from Fig. 10.

where

$$E_1 = MR^2\,(E_i/13.6)^2$$

(E_i denoting the official ionization energy of the projectile which is normalized to that of atomic hydrogen and M is the atomic-mass number of the projectile) appears indicated.

When applied to the cross-section maximum, this scaling of the beam energy, of course, corresponds to the well-known "Massey criterion"[54] determining the scaling of the beam energies at which inelastic cross sections have their maximum.

Using this scaling, the cross section for the ionization of an electron from the state Ψ_k may be written in the form

$$\sigma_{\rm ion}(\Psi_k) = S(Z_p, Z_t, E_i^k, \ldots)\, G((E_i^k/E_i)\epsilon), \qquad (9)$$

where the two unknown functions S and G may also depend on Ψ_i, though it is heuristically assumed that only S does.

Summing over all valence electrons of the projectile, its total single-electron-loss cross section becomes

$$\sigma_{\rm ion}^{\rm tot} = \sum_k \sigma_{\rm ion}(\Psi_k)$$

$$= \sum_k S(Z_p, Z_t, E_i^k, \ldots)\, G((E_i^k/E_i)\epsilon)\,, \qquad (10)$$

which can be written

$$\sigma_{\rm ion}^{\rm tot} = n_{\rm eff}\, S(Z_p, Z_t, E_i)\, G(\epsilon)\,, \qquad (11)$$

where $S(Z_p, Z_t, E_i, \ldots)\, G(\epsilon)$ is the ionization cross section for the outermost electron and $n_{\rm eff}$ is defined so as to include the contribution of the other valence electrons.

In this formula, the selection of the "interaction radius" R is an open question and needs further study. However, to a first approximation, we expect that this radius is mainly determined by the size of the target atom. This size determines the Fourier power spectrum of the perturbations of the electrostatic potential at any point of the projectile (after averaging over all impact parameters). Due to the involvement of continuum wave functions with varying energies and, thus, with varying phases relative to the bound state, we expect this local Fourier spectrum to be a reasonable first-order approximation for the over-all spectral distribution of the interaction and thus for the energy dependence of the cross section. In this case, R should depend only on the target atom and thus be roughly identical for all collisions involving the same target. For this reason, the data shown in Figs. 1–9 are separated according to targets. The selection of actual target "sizes" can be obtained from a comparison of collisions with projectiles and different targets and is discussed briefly later.

B. Scaling of Absolute Cross Sections

At present, no *a priori* arguments can be given for the functional dependence of S on the parameters of the collision partners. Therefore, a purely empirical analysis of the existing experimental data was performed.

From these data, it appeared that the cross sec-

tions plotted versus normalized energies increased with the atomic number of both target and projectile, roughly according to a $Z^{2/3}$ dependence. After some trials, a proportionality with $(Z_p^{2/3}+Z_t^{2/3})$ seemed to fit the data best (the indices p and t referring to projectile and target, respectively).

From the classical Thompson cross section,[56] a proportionality of the absolute cross section with E_i^{-2} is predicted. Similarly, the factor $1/\omega$ in front of the large parenthesis in Eq. (6) would lead to a factor $1/E_i^2$ in the final cross section. On the other hand, both the transition matrix elements V_{if} and the number of continuum states reached by the interaction may depend on E_i and thus lead to some other dependence. Also, from a first look at the data set, a less pronounced dependence on E_i seemed more realistic. Therefore, in the analysis, a factor $E_i^{-\alpha}$ with $\alpha < 2$ was included in S.

In Sec. IV A, it was mentioned that the function S may depend on the initial wave function Ψ_i. In particular, a dependence on the orbital angular momentum number l of the bound state may be expected, possibly similar to the dependence of the dipole oscillator strength from various angular momentum states to the continuum.[57] Supporting this, a comparison of the cross sections for projectiles of silver, iron, and uranium, which contain a relatively large number of d electrons in the outer shell, with those cross sections for other projectiles indicates that these d electrons have a clearly smaller ionization probability than p, and particularly, s electrons. Therefore, the inclusion of a weight factor $p(l)$ in S seemed appropriate. Thus, in total, Eq. (10) was assumed to take the form

$$\sigma_{i,i+1}=\sum_k p(l_k)\,(E_i^k/13.6)^{-\alpha}\,(Z_p^{2/3}+Z_t^{2/3})\,\sigma_{TM}(\epsilon_k)\ ,$$

which can be written

$$\sigma_{i,i+1}=n_{\mathrm{eff}}\,(E_i/13.6)^{-\alpha}\,(Z_p^{2/3}+Z_t^{2/3})\,\sigma_{TM}(\epsilon)\ , \quad (12)$$

where σ_{TM} was to represent the general normalized cross-section curve of this model and $\epsilon = E/M\,E_i^2 R_t^2$ (where R_t = "target radius").

In this formula, for convenience, we have normalized all ionization energies to that of hydrogen and we can put $p(l=1)=1$. Thus, n_{eff} describes the sum of the contributions from all electrons in the outer shell (possibly having different ionization energies E_i^k and angular momenta l_k) relative to the ionization of a possibly nonexistent s electron with the official ionization energy E_i of the projectile. In principle, n_{eff} is dependent on the beam energy, because of the energy dependence of the logarithmic slope of $\sigma_{TM}(\epsilon)$. However, for most reactions, this energy dependence was not very significant over the investigated energy ranges. Therefore, in the analysis, n_{eff} was computed for each reaction from the average slope and assumed constant.

C. Analysis of Experimental Data

In the actual analysis, published experimental single-electron stripping cross sections σ_{01}, σ_{12}, and σ_{23} for about 200 different neutral-neutral and ion-neutral collision pairs were included. From an initial analysis of cross-section curves that remained roughly constant over a wider range of beam energies, i.e., that were measured close to their maxima, it appeared that these cross sections increased with the atomic number of the projectile and target, with the valence of the projectile, and with decreasing ionization energy of the projectile. First, a more detailed analysis was performed for the 120-odd different neutral-neutral collisions. As mentioned, the dependence on the atomic numbers of the collision partners seemed best approximated by the factor $(Z_p^{2/3}+Z_t^{2/3})$ in Eq. (12). After exclusion of this factor, the cross sections for all such reactions using the same target were grouped together and plotted according to the energy normalization mentioned in Sec. IV B and using a fixed "target radius" for each plot. From these plots, the dependence of the cross sections on the ionization energy of the projectile and the number of electrons in its outer shell appeared even more pronounced. To investigate these points more closely, the ionization energies and angular momenta of all the outer-shell electrons for all the projectiles were obtained from Ref. 58. All of the ionization energies were sufficiently small so that no loss of a second electron by Auger autoionization subsequent to the first ionization could occur. These ionization processes would contribute mainly to a double-stripping cross section and would not be seen in the single-loss data. It became evident that a relatively large number of d electrons in Ag, U, and Fe have ionization energies similar to the outer-s and -p electrons. On the other hand, the cross sections for these projectiles were comparable with cross sections for projectiles having a small number of s and p electrons. Thus, the ionization probability $p(l)$ for d electrons appeared significantly smaller than that for electrons with smaller angular momentum. Similarly, it seemed that $p(1) < p(0)$.

For a more quantitative test, a sample of reactions, mainly including those with Ar and He targets, were investigated using various sets of $p(l)$'s. In these calculations, it was assumed that the dependence on E_i^k was proportional to $(E_i^k)^{-\alpha}$ (with $\alpha = 0,\ \frac{1}{2},\ 1$), as seemed compatible with a comparison of reactions with chemically similar projectiles. Best results were obtained assuming $\alpha \approx \frac{1}{2}$, $p(2) \approx 0.15-0.3$, and $p(1) \approx 0.5-0.7$ [by definition $p(0)=1$].

Thereafter, all experimental data for neutral-neutral and ion-neutral collisions normalized ac-

TABLE I. Effective number n_{eff} of outer-shell electrons and ionization energies E_i for various projectiles.

Projectile	H	He	Li	B	C	N	O	F	Ne	Na	Al		
E_i(eV)	13.6	24.5	5.4	10.8	11.2	14.5	13.5	17.3	21.5	5.1	6		
n_{eff}	1.0	2.0	1.0	1.4	2.0	2.3	1.9	2.7	4.1	1.0	1.5		
Projectile	Ar	K	Fe	Kr	Sr	Ag	Te	I	Xe	Cs	Ba		
E_i(eV)	15.7	4.3	7.8	14	5.7	7.5	9	10.6	12	3.9	5.2		
n_{eff}	4.2	1.0	2.2	4.1	2.0	1.5	2.8	2.9	4.1	1.0	2.0		
Projectile	U	He^+	Li^+	Be^+	B^+	N^+	O^+	Ne^+	Al^+	Ar^+	K^+		
E_i(eV)	6.0	54.1	75	18.1	25	29.5	35	41	18.7	27.8	31.7		
n_{eff}	2.0	1.0	2.0	1.0	2.0	2.4	2.8	3.7	2.0	3.8	4.6		
Projectile	Fe^+	Kr^+	Sr^+	Ag^+	Sb^+	Xe^+	Cs^+	Ba^+	Li^{++}	B^{++}	N^{++}	Al^{++}	Ba^{++}
E_i(eV)	16.2	26	11	21.4	18	21	23.4	10	122	37.8	47.4	28.3	36
n_{eff}	1.2	3.7	1.0	2.5	2.3	3.7	4.1	1.0	1.0	1.1	2.0	1.5	3.7

cording to Eq. (12) using $\alpha = \frac{1}{2}$, $p(2) = 0.2$, and $p(1) = 0.6$. The resulting plots are shown for the most often used target gases in the lower parts of Figs. 1–9. Table I gives the n_{eff} values which were used for various projectiles. The target radii used for energy scaling are discussed later. The plots for the other target gases used, in particular, Xe, H_2, and O_2 are similar to the ones shown.

Figures 1–4 show the results for the atomic target gases He, Ne, Ar, and Kr. In all cases, most of the cross-section curves fall into quite a narrow range of the normalized two-state plots. This is true both for the flat and for the steep sections of the curves, and the steep sections coincide considerably better than in the Firsov plots.

Major deviations occur only for low-energy stripping of some of the rare-gas atoms in rare gases, in particular, for the combinations Ne-Ne, Ar-Ar, Kr-Ar, Kr-Kr, Ar-Kr, and possibly Ne-He and Kr-He. In these cases, the experimental results lie considerably above the bulk of the other curves and also exhibit a clearly different slope. It is noteworthy that most of these collisions are much better described by the Firsov model; otherwise, no serious deviations from the general range of cross-section values are apparent. However, in a number of cases, the slopes of the measured curves differ from the slopes generally prevalent in the respective range of the normalized energy. In particular, this occurs for some collisions involving B, C, and O projectiles (see also Fig. 5 for N_2 target). For oxygen projectiles, this anomaly may be due to experimental errors since considerable discrepancies exist between the older low-energy stripping measurements of Fogel *et al.*[18,22] and the more recent high-energy measurements of Fite *et al.*[32] which fall well into the general range. Even though Fite and co-workers measure attenuation cross sections $q_0 = \sum_k \sigma_{0k}$, i.e., include also multi-electron-loss collisions, at the energies in question, the cross sections for multi-electron-losses should be clearly smaller than the single-loss cross section σ_{01}, and thus most of this discrepancy is probably experimental. The measurements with boron and carbon projectiles were done by the same authors who did oxygen and thus may have been subject to the same errors. In addition, slopes of relatively short curve sections may be changed considerably due to statistical errors. On the other hand, the slopes of other curves, in particular of some curves belonging to projectiles of atomic nitrogen, and uranium,[32] point out of the normal range at low energies. Again, in many of these cases, the respective cross sections are quite well described by the Firsov model, but the difference between both models is not very distinct in this energy range and good low-energy measurements are needed to decide which model applies in these cases. In addition, an over-all tendency appears to exist for a shift of the cross-section maxima with increasing ionization energy towards smaller energies and lower cross-section values than would be required by our model.

The two-state plot for the Xe target, in which only eight different projectiles were included, is similar to the one for the Kr target. Again, only two low-energy curves, relating to Kr-Xe and Ne-Xe collisions, fall out of the normal range.

Concerning the effective "radii" R applicable to these reactions, we can only make comparisons with other known radii like the "gas-kinetic" or "atomic" radii[59,60] for the various targets. As previously mentioned, the same radius R was used in the normalization of all collisions involving the same target atom, i.e., for each of the figures. Since the general function of $\sigma_{TM}(\epsilon)$ is not known at present, no absolute values for the effective radii of the various targets can be derived from this scaling. However, relative values can be obtained from a comparison between the normalized

graphs for various targets. For this purpose, the effective radius of Ar was arbitrarily chosen equal to its gas kinetic radius 1.50 Å[59] in Fig. 3. The corresponding radii for the other rare gases, He, Ne, and Kr, are 0.95, 1.12, and 1.6 Å, respectively. These numbers were used in the two-state graphs of Figs. 1, 2, and 4, and the general agreement between various figures is reasonable. A somewhat better fit could be achieved by using $R = 0.85$, 1.20, and 1.7 Å, respectively. Correspondingly, the best fit for Xe would require $R \approx 1.9$ Å compared with a gas-kinetic radius of 1.7 Å. Except for He, very similar ratios between these rare gases apply for the "atomic radii."

Figure 5 shows the corresponding graph for N_2, assuming $R = 1.4$ Å. This corresponds to an "atomic radius" for N of 0.92 Å compared with 0.98 Å for argon. On the other hand, an optimal fit between the data for Ar and those for N_2 would require a radius of approximately 1.25 Å, which compares with a gas-kinetic radius for the entire molecule of 1.6 Å. For the other target gases, Xe, H_2, and O_2, good fitting would require the radii 1.9, 1.0, and 1.2 Å, respectively, compared with the gas-kinetic radii 1.8, 1.1, and 1.5 Å, respectively. For the atomic oxygen target $R = 1.1$ Å would be about optimal. Thus, it appears that the basic use of "target radii" is able to give a reasonable first approximation, though it is too early to determine exactly which of the various radius determinations in use produces the best scaling result. The few data available for alkali gas targets indicate that relatively large radii around 1.9 and 2.4 Å are necessary to produce reasonable scaling.

Unfortunately, very little is known at present on radii of molecular curve crossings through which a single-step transition of the electron into the continuum might occur, and, therefore, no similar comparison can be made. On the other hand, a collisional transition of this type does not appear very likely for most cases for which our model applies. In collisional electronic transitions of this type, generally a much slower energy dependence of the cross sections is expected and also observed, even at quite small energies. (Compare for instance the charge transfer reaction[61] $He^+ + Ar \rightarrow He + Ar^+$ which was shown[62] to proceed via molecular curve crossings.) Also, our derivations concerning the energy scaling implicitly assume the effective absence of such crossings.

In Fig. 5, both the Firsov graph and the two-state graph for collisions involving a molecular nitrogen target were plotted assuming that the two nitrogen atoms act independently, i.e., the published molecular cross sections were divided by two and $Z_t = 7$ was used. Direct experimental evidence for such procedure is limited, at best,[63] to collisions with energies above the Massey maximum. In the near-adiabatic range, recent measurements by Lo and Fite[46] of σ_{12} on atomic oxygen indicate that the respective cross sections are not very different from those using a molecular oxygen target; on the average, they may be smaller by at most about 30%. On the other hand, considering the statistical orientation of the target molecule, this assumption still appears reasonable as a general first approximation. In Fig. 5, the difference between both models is not very distinct. The two-state model gives a somewhat better over-all fit; but again, some curves, in particular those for N, Fe, and possibly O projectiles, tend to fall out of the general range at the low-energy end.

The plots for molecular oxygen targets containing 12 different reactions are very similar to the ones for molecular nitrogen; those for molecular hydrogen, containing five different reactions, show an agreement with the two-state model which is similar to the agreement found in the rare-gas targets.

For an over-all comparison of the two-state scaling for neutral-neutral collisions, Fig. 12 presents a composite of all two-state plots of Figs. 1–9, including some additional data also shown in Figs. 10 and 11. With the exception of the curves for rare-gas–rare-gas collisions, most experimental data are confined to a relatively narrow range around a central curve which, for practical applications, can be roughly approximated by

$$\delta_{TM}(\epsilon) \approx 3.2 \times 10^{-14} \left[\frac{\epsilon}{(\epsilon^{2/3} + 30^{2/3})^3} \right]^{1.2} .$$

For $\epsilon \lesssim 0.2$ deviations from this curve generally are smaller than a factor of 2.5.

Figures 6–8 show the corresponding graphs for a number of σ_{12} reactions. In this case, a larger number of reactions were measured over a wide energy range. The steep rise of the cross section at small beam energies is even more frequent than in the σ_{01} case and leads to very pronounced deviations from the Firsov model. As in the earlier Firsov graphs, the ionization energy of the projectile was used in the normalization. The use of any lower ionization energy would increase the disagreement further. In contrast, the two-state graphs using the same radii as the corresponding earlier figures show a good grouping of most curves; and of particular interest is the fact that this grouping occurs for all targets exactly in the same area of the normalized coordinates as for the neutral-neutral collisions. At low normalized energies, the curves appear, on the average, to be somewhat steeper than in the neutral-neutral collisions. This may, however, be a result of experimental errors: The ionization energies in these collisions generally are higher than in neutral-atom stripping and thus

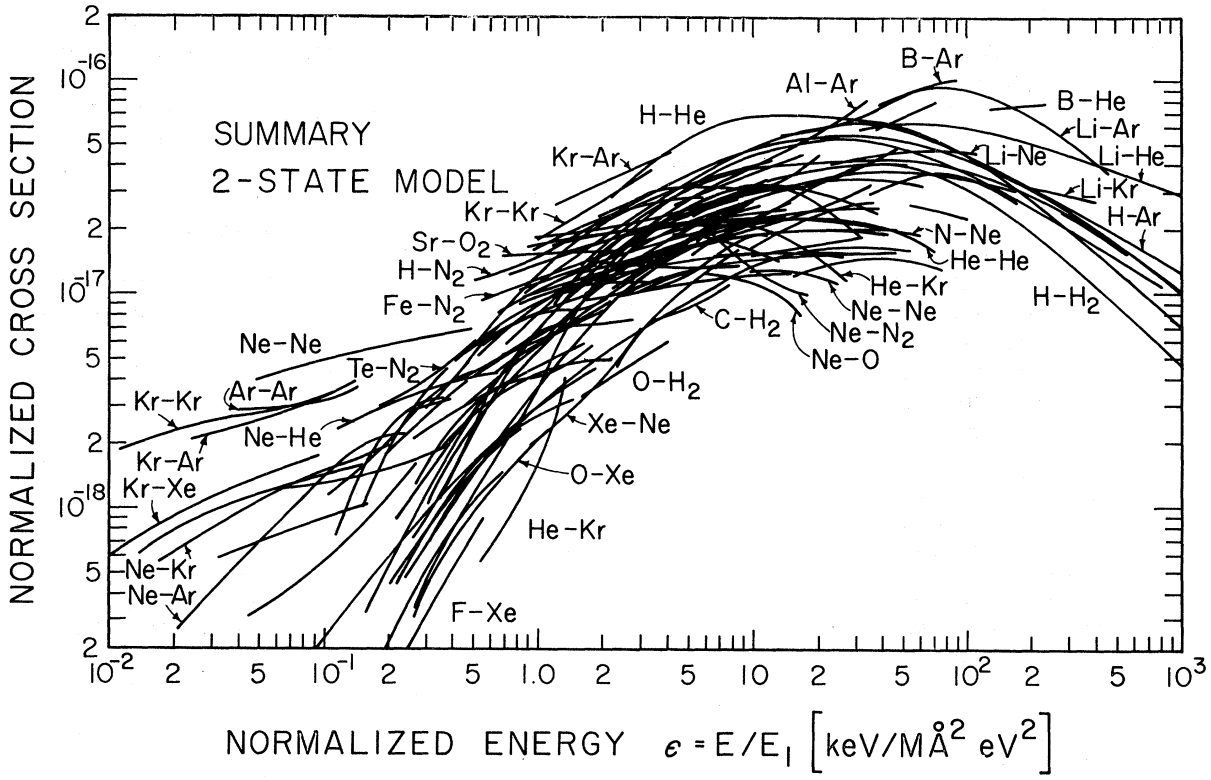

FIG. 12. Two-state model graph of data from Fig. 10.

may have led to enhanced scattering losses. In this sample of experimental results, only the case of $Ar^+ + Ar$ deviates seriously from the two-state scaling. The data of Dmitriev *et al.*[37] on Li^+-Ar and Li^+-Kr, indicated by (D), fall somewhat out of the general range, but also disagree strongly with the data of Allison *et al.*[42] Beyond that, only a few slopes are at variance with that of the general curve. But, again, curves for projectiles with higher ionization energy tend to peak at somewhat lower beam energies and lower cross-section values.

Similar remarks apply to collisions involving targets of Xe, H_2, and O_2 which are not reproduced in these figures. As mentioned, the few curves measured for atomic oxygen targets are very similar to the corresponding curves involving molecular oxygen targets.

Finally, Fig. 9 shows graphs for some collisions involving doubly charged ions. Again, the normalized plots were obtained using the same procedures and parameters as in the earlier figures. In this case, the two-state plot becomes somewhat more diffuse though it still is clearly better than the corresponding Firsov graph. Some more single-electron-loss cross sections $\sigma_{i,i+1}$ for higher-charged low-Z ions were investigated. It appears, that they fall roughly into the same general range of the normalized plot, though the deviations become larger and the maxima again seem to occur at lower values of the normalized energy.

V. DISCUSSION

In total, it appears that this rather coarse model is able to describe quite well a large portion of the measured single-electron-loss cross sections, in particular those exhibiting steep slopes at low energies. Barring major errors in the experimental results, in these latter cases, serious deviations from this model, so far, are limited to collisions between rare-gas particles which seem better described by the statistic model of Firsov. In many other cases, no clear distinction between both models can be made. Therefore, no valid claim as to the range of applicability of both models for various collision pairs can be made at present. This has to await further experimental studies at lower normalized energies, which have to include careful checks on the angular scattering in these reactions.

Major questions certainly arise as to the validity and accuracy of some of the assumptions made concerning the differences between various angular momenta of the bound state, the dependence of the cross sections on the ionization energies, and the atomic numbers of the collision partners and also

concerning the use of the target radii for the energy scaling. In all these areas improvements may be possible by more detailed empirical studies. In particular, it appears that slight changes of the scaling of energy and absolute values recognizing the mentioned shifts in the position and cross-section values of the curve maxima may give a better fit, with best results probably obtainable with an energy scaling proportional to $1/E_i^{3/2}$ (instead of $1/E_i^2$) and a scaling of the absolute values proportional to $1/E$ (instead of $1/E_i^{1/2}$). Also, detailed theoretical single-electron model calculations would be useful in regard to these questions. Such calculations also could give indications as to the influence of chemical effects between the collision partners and of the coherence between various electrons in the outer shells. As additional possibility,[64] combining some aspects of the Firsov model and the two-state model, one could consider that the determining single-step transition of the stripped electron only proceeds to a highly excited state (possibly the lowest available) and then further excitation and ionization occurs in a statistical fashion.

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