

## Multiple ionization of atoms and molecules in collisions with fast ions. II. Ion-molecule collisions

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(Received 6 August 1997)

An extended version of the statistical energy-deposition model is used for a description of multiple ionization of molecules by energetic ion impact. The energy transferred to the molecule during the collision is calculated as a function of the impact parameter within the local-electron-density approximation for a fixed molecular orientation. The fluctuating character of the deposited energy is taken into account by introducing a straggling of the energy transfer. For a given deposited energy the probability of multiple ionization of the molecule is assumed to be proportional to the volume of phase space available at the considered ionization state. The total ionization cross section is obtained by integrating over impact parameters and averaging over all orientations of the molecule. Sample calculations for collisions of He and F ions with N<sub>2</sub> and CO molecules are presented and compared with experimental data. A dependence of the multiple ionization cross section on the molecular orientation is considered. A strong molecule alignment effect has been obtained in agreement with recent experimental findings. [S1050-2947(98)00702-1]

PACS number(s): 34.50.Gb

### I. INTRODUCTION

In recent years detailed experimental investigations of the dissociative ionization of small molecules by energetic ion impact have been carried out by several groups [1]. This process is of fundamental importance in many areas of science and technology, for example, in studies of astrophysical plasmas and upper planetary atmospheres [2] or in studies of radiation damage to biological tissues [3]. Moreover, it was found that in ionizing collisions with heavy multiply charged ions, highly charged fragment ions are copiously produced [4]. Therefore, such experiments provide unique information upon the dissociation of multiply ionized molecular ions that is not accessible in experiments with electron or photon beams.

Recent progress in the investigation of the dissociative ionization is due to the use of the coincidence time-of-flight technique [5–9] that gives the possibility to study various dissociation channels, their relative abundances, kinetic-energy distribution in a particular channel, etc. It was shown that coincident measurements with a position- and time-sensitive multiparticle detector provide a complete three-dimensional image of the breakup process for each individual event [9]; this means that the dissociation of highly ionized molecules can be even studied for a definite orientation of the molecular axis.

Quick progress in the experimental studies demands a development of reliable theoretical methods for treating the ion-molecule ionizing collisions. However, here the situation is far from satisfactory. To the best of our knowledge, the multiple ionization of multielectron molecules by fast ion impact was treated only within the independent-electron

model [10,11]. However, in applications of this method, which proves to be very successful for the description of ion-atom collisions, the molecular properties of the target are practically ignored [10,11].

In our previous paper [12], hereafter referred as I, an extended version of the statistical energy-deposition (SED) model was developed to describe the multiple ionization in fast ion-atom collisions. The basic ideas of the SED model were formulated by Russek and his collaborators [13–15] and further developed by Cocke [16]. The model implies that the multiple ionization is viewed to proceed in two stages. In the first one, part of the kinetic energy of the projectile is transferred to electronic excitations of the target system. Then, in the second stage, after the partners depart from one another, the deposited energy is distributed among all target electrons and the system subsequently autoionizes to reach its final ionization state.

In I we extended the Russek-Meli-Cocke model in two respects. First, the deposited energy for each projectile trajectory is considered as a fluctuating quantity with a certain distribution, and the ionization probability is calculated as a weighted average over this distribution. Second, the mean value and the straggling of the deposited energy are calculated within the Lindhard-Scharff [17] local-electron-density approximation. In this approximation the energy transferred to the target is determined by the target ground state electron density. Therefore, this approach can be easily extended to the case of ion-molecule collisions provided the electron density of the target molecule is known.

In the present paper we describe an application of the extended SED model to ion-molecule collisions. As examples, collisions of He and F ions with diatomic N<sub>2</sub> and CO molecules are considered.

In the next section we discuss in more detail the SED model and in particular the calculations of the energy transferred to a molecule in a fast ion-molecule collision. In Sec. III sample calculations of multiple ionization cross sections are presented and discussed. Finally in Sec. IV we discuss a

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theoretical description of the effect of molecular alignment on the multiple ionization, which was recently revealed experimentally [18–20]. We give conclusions in Sec. V. Atomic units are used throughout unless otherwise indicated.

## II. STATISTICAL ENERGY-DEPOSITION MODEL OF MOLECULAR IONIZATION

We consider ionizing collisions of light ions with simple molecules in the energy range 50 keV/amu to 5 MeV/amu. In this energy region the collision time is much shorter than the characteristic time of molecular rotations or vibrations. Therefore, we may consider an ion collision with a molecule of fixed orientation and interatomic distance, which we assume to be equal to the equilibrium distance. Moreover, we suppose that the electrons are removed slowly in comparison with the collision time and rapidly in comparison with the nuclear motion in the molecule, a condition that is commonly expected to be fulfilled in the processes considered. Accordingly, the dissociative ionization of molecules may be considered as proceeding in several stages. The first stage is a collision of the projectile with the molecule in which part of the kinetic energy of the projectile is deposited into the target. The second stage consists of autoionization (evaporation) of several electrons from the highly excited molecule and formation of the transient molecular ion state. Finally, this transient molecular ion dissociates into the final system of ion fragments.

The first two stages resulting in multiple ionization of the molecule are treated here within the framework of the statistical energy-deposition model. The energy transferred in the collision (or rather the distribution of the transferred energies) is considered as a reasonably well defined function of the collision parameters and will be treated in the next subsection. Given that the deposited energy is known, the probability for each final ionization level is calculated as suggested by Russek and Meli [15]. It is proportional to the volume of phase space available in a particular ionization state, and it is directly related to the deposited energy and the ionization potentials of the various levels.

### A. Energy transfer in ion-molecule collisions

In the considered energy range the recoil energy is much less than the energy transferred to the electronic shell of the target [21]. In other words, the energy loss of the projectile is approximately equal (with negative sign) to the deposited electronic energy. In order to calculate this deposited energy we use the well-known approximation used in the stopping power theory, the so-called local-plasma or local- (electronic) density approximation (LDA). It is based on the idea of Lindhard and Scharff [17] who suggested to consider each volume element of the target atom as an independent electron plasma of uniform density that is equal to the electron density of the atom. Using the known stopping power of an electron gas and integrating over the atomic volume with the known electron density a good description of the energy loss is achieved [22,23]. The LDA has also been successfully applied for calculating the energy loss in molecular targets [24]. We use this approximation in order to calculate the energy deposition in ion-molecule collision.

We assume (see I) that the projectile ion is a point charge  $Z_1$  moving along a straight-line trajectory with a constant velocity  $v$  and an impact parameter  $\mathbf{b}$ , which is taken from the center of mass of the target molecule. The energy loss in a collision is a statistical process and the transferred energy may be characterized by the mean energy and energy straggling.

Within the LDA the mean deposited energy for a certain ion trajectory can be calculated as a line integral along the trajectory

$$\bar{E}(\mathbf{b}) = \frac{4\pi Z_1^2}{v^2} \int_{-\infty}^{\infty} dz \rho(\mathbf{r}) L(\rho(\mathbf{r}), v), \quad (1)$$

where the  $z$  axis is chosen along the ion beam direction,  $\mathbf{r} = \{\mathbf{b}, z\}$ ,  $\rho(\mathbf{r})$  is the electron density of the molecule [we normalize the density to the total number of electrons in the molecule:  $\int d\mathbf{r} \rho(\mathbf{r}) = Z_2$ ], and  $L(\rho(\mathbf{r}), v)$  is the usual stopping number. Convenient approximate expressions for calculating  $L(\rho(\mathbf{r}), v)$  for a free-electron gas have been suggested by Lindhard and Winther [25] within the framework of the linear response dielectric formalism [see expressions (6)–(8) in I].

The stopping cross section may be obtained by integrating the energy loss  $\bar{E}(\mathbf{b})$  over all impact parameters:

$$S = \int d^2 \mathbf{b} \bar{E}(\mathbf{b}). \quad (2)$$

Similarly, the straggling of the deposited energy can be calculated within the Lindhard-Scharff model as a line integral along the trajectory:

$$W_{LS}^2(\mathbf{b}) = 4\pi Z_1^2 \int_{-\infty}^{\infty} dz \rho(\mathbf{r}) \frac{\Omega^2(\rho(\mathbf{r}), v)}{\Omega_B^2}, \quad (3)$$

where

$$\Omega_B^2 = 4\pi Z_1^2 Z_2, \quad (4)$$

and  $\Omega^2(\rho(\mathbf{r}), v)$  is the straggling in a free-electron gas, which we have calculated using analytical approximations suggested by Bonderup and Hvelplund [26] [see expressions (11) and (12) in I].

The straggling has been corrected allowing for the spatial correlations of the collisions with electrons inside the molecule (so-called bunching effect [27], see I for details). Note that in our approach we do not have to apply an additional correction due to the spatial correlations of atoms in a molecule which was discussed by Sigmund [28], because we calculate the electronic density of the molecule *ab initio* instead of considering the molecule as a system of individual atoms.

The total straggling may be obtained as an integral over all impact parameters

$$\Omega^2 = \int d^2 \mathbf{b} W^2(\mathbf{b}). \quad (5)$$

The electronic density of the molecules that enters in expressions (1) and (3) was calculated in the Hartree-Fock ap-

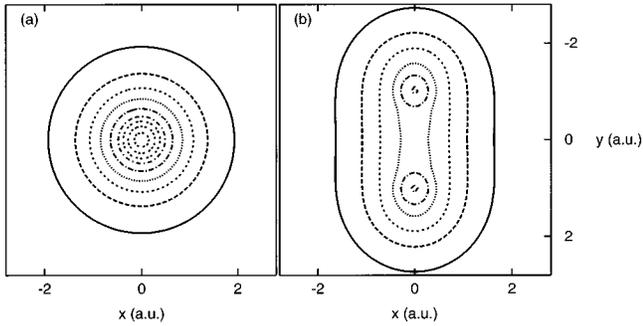


FIG. 1. Contour plot of the deposited energy (in eV) calculated for collisions of  $\text{He}^{2+}$  ions with a  $\text{N}_2$  molecule at the energy of 2 MeV. Projectile velocity is along the  $z$  axis. The molecular axis is oriented (a) along the ion beam, or (b) perpendicular to the beam. The outermost solid line corresponds to an energy of 10 eV. Other lines are drawn in steps of 20 eV (a) or 15 eV (b). The innermost line corresponds to an energy of 170 eV (a) and 85 eV (b).

proximation using the MOLPRO code [29–31]. The ground-state electronic wave function was obtained using the double-zeta plus polarization Gaussian basis set for the optimized geometry of the molecule. The impact parameter distribution of the energy transfer and of the straggling depends, naturally, on the molecular orientation with respect to the ionic beam. In Fig. 1 we show, as an example, the contour plot of the deposited energy distribution for the case of 2-MeV  $\text{He}^{2+}$  scattering from a  $\text{N}_2$  molecule. The results for two orientations of the molecule are shown: (a) when the molecule is aligned with the beam direction, (b) when it is perpendicular to the beam. Note that in the orientation (a) the transferred energy is much larger in the central region than in the case (b). This is easy to understand since when the projectile moves along the molecular axis it encounters more electrons in the central region than when it moves perpendicular to the axis. The energy loss, which is proportional to the electron density sampled by the projectile, is larger in the aligned case. Similar results are obtained for the energy straggling (see Fig. 2).

The total energy loss, calculated for the considered case, is  $32.4 \times 10^{-15}$  eV  $\text{cm}^2/\text{atom}$ , which agrees well with the experimental value  $32.7 \times 10^{-15}$  eV  $\text{cm}^2/\text{atom}$  [32]. The calculated straggling of the energy loss is 8.0

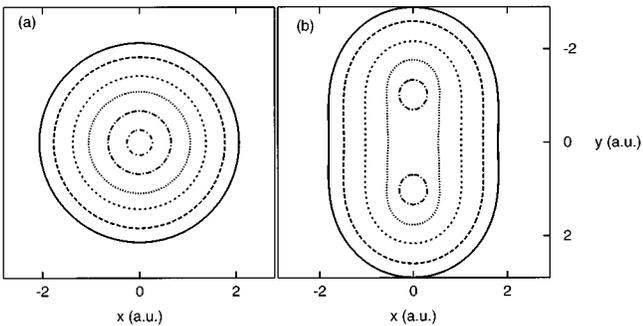


FIG. 2. The same as in Fig. 1 but for the energy straggling (in  $\text{eV}^2$ ). The outermost solid line corresponds to a straggling of  $10^3$   $\text{eV}^2$ . Other lines correspond to a straggling of 2.5, 10, 20, 50, respectively, in units  $10^3$   $\text{eV}^2$ . The innermost line corresponds to a straggling of  $5 \times 10^4$   $\text{eV}^2$  (a) and  $2 \times 10^4$   $\text{eV}^2$  (b).

$\times 10^{-12}$   $\text{eV}^2$   $\text{cm}^2/\text{atom}$ . It is less than the experimental value  $9.3 \times 10^{-12}$   $\text{eV}^2$   $\text{cm}^2/\text{atom}$  [33], but greater than the value of  $6.6 \times 10^{-12}$   $\text{eV}^2$   $\text{cm}^2/\text{atom}$  given by Chu in Ref. [34]. Thus we may conclude that the LDA calculations give a rather accurate account of the deposited energy and energy straggling.

Note that in the LDA the total energy loss averaged over all impact parameters is independent of the molecular orientation. This is so because each elementary volume element contributes to the energy loss independently, and the total energy loss is a simple sum of these contributions. For the same reason the energy transfer to a heteronuclear diatomic molecule depends only on its alignment but not on its orientation.<sup>1</sup> This is a property of our approximation. In principle, the energy transfer can depend on the orientation, for instance, if we take into account a deceleration of the projectile along the trajectory or the Coulomb deflection of the projectile in the field of the target nuclei. Such effects, however, should be small for the systems considered here.

The deposited energy distribution for each trajectory is assumed to be Gaussian

$$w(E_T, \mathbf{b}) = \frac{1}{\sqrt{2\pi}W(\mathbf{b})} \exp\left(-\frac{[E_T - \bar{E}(\mathbf{b})]^2}{2W^2(\mathbf{b})}\right) \quad (6)$$

with parameters calculated in the LDA according to the above expressions.

## B. Multiple ionization cross sections: sample calculations

Knowing the deposited energy distribution one can calculate the probability of multiple ionization as a convolution of this distribution with the probability of ionization calculated for each deposited energy [see expressions (1) and (16) in I].

At this stage of the calculation the knowledge of the ionization potentials is necessary for each state of molecule ionization. Using the MOLPRO program [29–31] we have calculated the total ground-state energies of the multiply ionized molecular ions. The difference of energies of two sequential molecular ions was taken as the corresponding ionization potential,  $\mathcal{E}_i = E_{i-1} - E_i$ . This procedure means that we consider multiple ionization as a sequential emission of electrons, which is slow enough to provide sufficient time for relaxation; it may be considered as an extreme case (the other extreme would be a frozen molecular orbitals approximation). The calculated ionization potentials for  $\text{N}_2$  and  $\text{CO}$  molecules are presented in Table I.

As is clear from the discussion in the Introduction and the previous section, our calculations are performed for a fixed molecular orientation, and the calculated multiple ionization cross sections may depend on the alignment of the molecule with respect to the ionic beam. This alignment effect is considered in the next section. For randomly oriented molecules the cross section has to be averaged over all molecular orientations.

Before considering the results of sample calculations we would like to discuss a contribution of the electron capture

<sup>1</sup>Here we use the term *orientation* in its narrow sense as a vector characteristic of the system.

TABLE I. Calculated ionization potentials (in eV) for  $N_2$  and CO molecules and molecular ions for different states of ionization.

	1	2	3	4	5	6	7	8	9	10
$N_2$	15.92	29.79	41.63	54.37	68.13	88.31	97.61	114.6	162.6	191.0
CO	13.31	28.64	39.68	57.84	80.24	71.29	102.1	128.5	168.8	190.0

process, to multiple ionization. It is well known that electron capture can strongly affect the ionization process, especially at not very high collision energies. Direct experiments in which the projectile and the recoil ion were detected in coincidence [35] showed that the transfer-ionization channel can contribute considerably to the total ionization cross section. The question arises if capture channels are included in the SED model. In his paper of 1979 Cocke [16] argued that in principle, the SED model takes into account all phase space available to the outgoing electrons. Part of this space may be that which is occupied by electrons bound to the projectile. However, he concluded that the answer to the above question is not clear. We are inclined to believe that the capture channels are excluded from the SED model. The basic assumption of the model is that the electron emission occurs *after* the collision when the fast projectile is already far from the interaction region. Therefore a captured electron as well as any other fast “directly” knocked out electrons are not considered by the model. Practically, this means that if the capture channels are important the corresponding cross sections should be added to the one calculated within the SED model.

As a first example we now consider the multiple ionization of  $N_2$  molecules by  $He^{2+}$  ions at 0.5-MeV/amu energy. At this energy the contribution of electron capture is negligible. Thus we calculated the ionization cross section in the SED model using the energy deposition given in the preced-

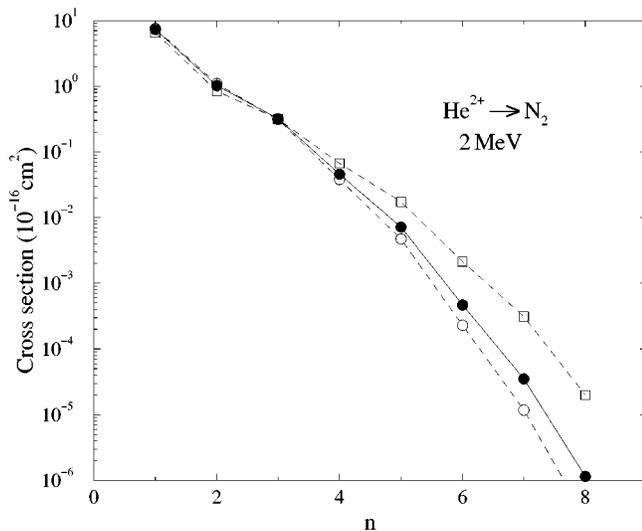


FIG. 3. Cross sections for removing  $n$  electrons from  $N_2$  by 2-MeV  $He^{2+}$  impact. Calculated points are connected by lines to guide the eyes. Black dots connected by solid lines represent the cross sections averaged over all orientations of the molecule. Points connected by dashed lines represent the cross sections for the molecular axis aligned with the beam (squares) and perpendicular to it (open circles).

ing section. The results of the calculations are presented in Fig. 3 for two different orientations of the molecule as well as averaged over all orientations. We recall that in the SED model there is a free parameter  $g$  (see I) characterizing the mean square matrix element of autoionization. We have chosen it to be 0.01. This is close to the values we used in our previous calculations (I) of ion-Ne-atom collisions at slightly higher energy. Besides, with this value we obtain good agreement between calculated and experimental cross sections for a  $He^+$ -Ar collision at the same collision energy 2 MeV [36], Ar having ionization potentials very similar to those of  $N_2$ . One can see from Fig. 3 that the multiple ionization cross sections steeply decrease with increasing degree of ionization as is known from ion-atom collisions. Furthermore, the cross sections for multiple ionization clearly show the alignment dependence, which will be discussed in more detail in the following section. In order to demonstrate the influence of the energy straggling we calculated the same cross sections disregarding straggling ( $W=0$ ). The results are shown in Fig. 4. One can see that the slope of the curves has increased. Moreover, the difference between the cross sections for the two orientations is drastically increased too. The straggling of deposited energies smooths the alignment dependence.

Similar calculations have been performed for  $F^{4+}$  scattering from CO molecules at an energy of 1 MeV/amu. In this calculation we assumed that the  $F^{4+}$  ion can be considered as a point charge with  $Z_1=4$ . This is justified, at least for not very high degrees of molecular ionization, because the radius of the ion is small (0.2 a.u.) in comparison with the dimensions of the molecular orbitals. The results of the calculations are compared with the experimental data [6] in Fig. 5

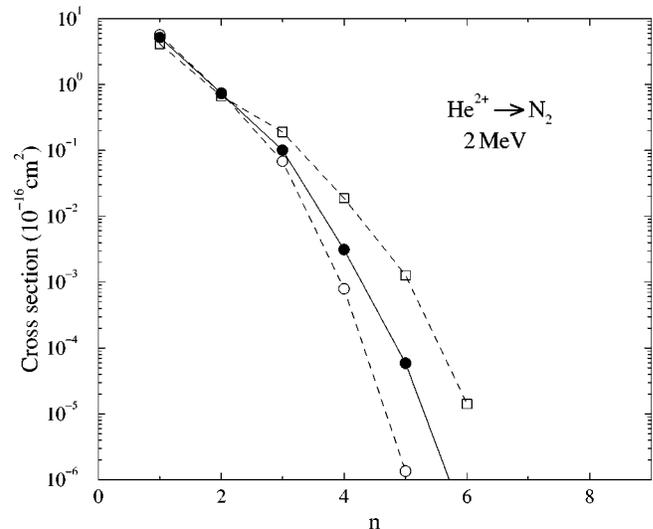


FIG. 4. The same as in Fig. 3 but disregarding the straggling of the deposited energy ( $W=0$ ).

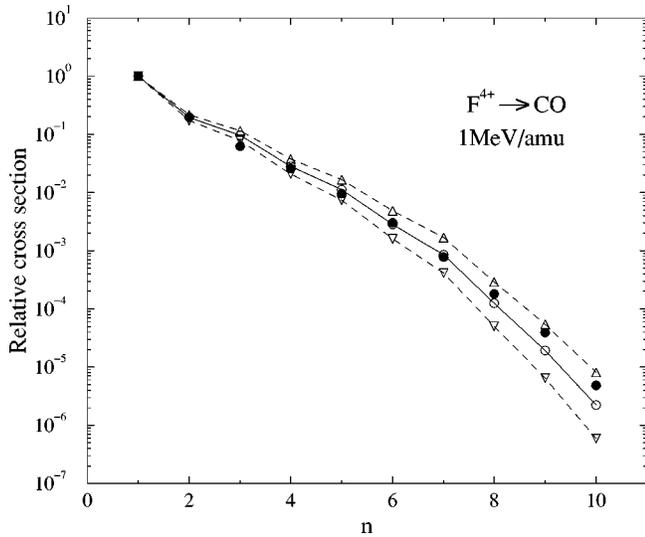


FIG. 5. Cross sections for removing  $n$  electrons from CO by 1-MeV/amu  $F^{4+}$  impact relative to the single ionization cross section. Calculated points are connected by lines to guide the eyes. Solid lines with open circles:  $g=0.007$ ; dashed lines, triangles pointed down:  $g=0.005$ ; triangles pointed up:  $g=0.01$ . Black dots: experimental data from Ref. [6].

where the relative multiple to single ionization cross section ratios are displayed. The parameter  $g$  was fitted to obtain good agreement with the experimental data. The fitted value  $g=0.007$  is close to the value 0.01, which was chosen in I to describe  $F^{4+}$ -Ne collisions at the same energy. With this choice of  $g$  the SED model yields very good agreement with experiment. In order to show the sensitivity of the results to the parameter  $g$ , we present in Fig. 5 the results of the calculations for a smaller (0.005) and a larger (0.01) value of  $g$ . With increasing  $g$  the cross-section ratios increase while the single ionization cross section decreases.

The calculated value of the single ionization cross section is  $8.29 \times 10^{-16} \text{ cm}^2$ , which is less than the experimental value  $(17 \pm 8) \times 10^{-16} \text{ cm}^2$ . We note that in our previous SED-LDA calculation of ion-atom collisions the single ionization cross section was also underestimated (see I). This can probably be explained by a shortcoming of the LDA. Due to the long-range character of the Coulomb force, the energy transfer decreases less steeply at large impact parameters than predicted by the LDA, where it is approximately proportional to the electron density of the system. This has been demonstrated in I (see Fig. 1 in I) where we compared the impact parameter dependence of the energy transfer calculated within the LDA and the more accurate semiclassical approximation [37,38]. The LDA underestimates the energy loss at large impact parameters that contribute considerably to the single ionization cross section. Note that the ratio of the single ionization cross sections for  $F^{4+} + \text{CO}$  and  $F^{4+} + \text{Ne}$ , which, in fact, was measured in [6], is given by the SED-LDA model more accurately than the individual cross sections. The experimental value of the ratio is  $4.0 \pm 0.7$  while the theory gives 3.6.

### III. MOLECULE ALIGNMENT EFFECT IN MULTIPLE IONIZATION

We have already noticed in the preceding section that the multiple ionization cross section is influenced by the align-

ment of the molecule with respect to the beam direction. In this section we discuss the alignment effect in more detail.

Qualitatively, the influence of molecule alignment on the multiple ionization cross section can be easily explained by the anisotropy of the electron-density distribution. As was first suggested by Wohrer and Watson [39] on the basis of a very simple two-atom picture and an independent-electron model, a significant suppression of the multiple ionization cross section may be expected in the perpendicular orientation of the molecular axis with respect to the beam as compared to the parallel one (see also [40]). Within the SED model this effect is related to the anisotropy of the energy deposition along the trajectory (see Fig. 1). In order to remove several electrons a large energy deposition is needed. When a diatomic molecule is oriented along the beam the probability of a large energy deposition is greater than for an orientation perpendicular to the beam because the projected electron density sampled by the projectile is larger for trajectories close to the molecular axis. At perpendicular orientation the probability of large energy transfer is small and the multiple ionization cross section is suppressed. It is clear that more pronounced orientation effects are expected with increasing degree of ionization since for those a larger energy transfer is required.

Experiments [18–20] confirm the qualitative picture described above. Recent measurements [20] show a strong alignment effect in multiple ionization for  $\text{He}^+$  collisions with  $\text{N}_2$  molecules at collision energies of 100–300 keV. Below we compare the predictions of the SED model with the experimental data.

In contrast to the examples considered above, the  $\text{He}^+$  ion cannot be treated as a point charge with  $Z_1=1$ . The dimension of the electron cloud of  $\text{He}^+$  is comparable with the dimensions of the target electron shells; thus, the screening of the projectile nucleus charge by the electron is not complete. We take the screening into account by introducing the ion effective charge  $Z_{\text{eff}}$  according to the definition

$$\left. \left( \frac{dE}{dx} \right) \right|_{\text{He}^+} = Z_{\text{eff}}^2 \left. \left( \frac{dE}{dx} \right) \right|_{Z_1=1}, \quad (7)$$

where  $(dE/dx)|_{Z_1=1}$  is the calculated energy loss for a point charge  $Z_1=1$  moving with the same velocity as the ion, and  $(dE/dx)|_{\text{He}^+}$  is the experimental  $\text{He}^+$  ion energy loss. This effective charge not only includes screening effects but also the influence of charge changing processes on the energy transfer. Of course, the effective charge depends on the collision energy. In the following we present the results of calculations for a collision energy of 200 keV. Similar results have been also obtained for other ion energies. The energy loss calculated within the LDA for collision of a point charge  $Z_1=1$  with an  $\text{N}_2$  molecule at this velocity is  $12.8 \times 10^{-15} \text{ eV cm}^2/\text{atom}$ . The corresponding experimental energy loss for a  $\text{He}^+$  ion is  $34.6 \times 10^{-15} \text{ eV cm}^2/\text{atom}$  [32]. Therefore, the effective charge is  $Z_{\text{eff}}=1.65$ , which is in agreement with conventional expectations.

Using this effective charge we calculated the impact parameter dependence of the energy loss and straggling for various orientations of the  $\text{N}_2$  molecule. For calculating the multiple ionization cross sections we have chosen the param-

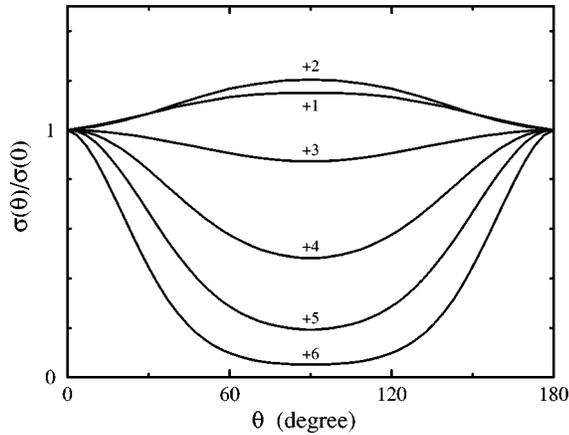


FIG. 6. The calculated cross sections for multiple ionization of  $N_2$  molecules in collisions with 200-keV  $He^+$  ions as functions of the angle  $\theta$  between the molecular axis and the beam direction. Curves are normalized at  $\theta=0^\circ$ .

eter  $g=0.1$ , which gives good agreement for the multiple to single ionization cross section ratios in  $He^+-Ne$  and  $He^+-Ar$  collisions at the same collision energy [36]. The results are shown in Fig. 6 as a function of the alignment angle  $\theta$  ( $\theta=0$  corresponds to the molecular axis being parallel to the beam direction). For convenience, the calculated cross sections are divided by the cross-section values at  $0^\circ$ . The corresponding absolute cross sections are presented in Table II where we also give the cross sections averaged over all orientation angles as well as the multiple-to-single ionization cross-section ratios. The results presented in Fig. 6 agree qualitatively with those obtained from the simplified model [39,40]. At small degree of ionization the variation of the cross section with the alignment angle is weak with a slight increase at perpendicular orientation of the molecule relative to the beam direction. When several electrons are removed the cross sections have a pronounced minimum at the perpendicular orientation that becomes deeper for higher degrees of ionization.

The calculated cross sections (multiplied by  $\sin\theta$ ) are compared with the experimental data (histogram) in Fig. 7. For higher degrees of ionization ( $n=3-5$ ) the theory describes the experimental alignment dependence well. It is interesting that for double ionization of  $N_2$  the experimental distribution is peaked at  $90^\circ$  much more strongly than predicted by theory. One can speculate that this enhancement may be explained by the contribution of transfer ionization, which is not included explicitly in our model. In fact, the capture cross section for 200-keV  $He^+$  is  $2.6 \times 10^{-16} \text{ cm}^2$  [41], i.e., about 20% of the total ionization cross section.

TABLE II. Calculated cross sections for the  $n$ -fold ionization of  $N_2$  molecules with the molecular axis aligned with the  $He^+$  beam [ $\sigma_n(0)$ ] and averaged over all orientations ( $\sigma_n^{av}$ ) in  $10^{-16} \text{ cm}^2$ ;  $R$  is the ratio of the averaged cross sections  $\sigma_n^{av}/\sigma_1^{av}$ .

	$\sigma_1$	$\sigma_2$	$\sigma_3$	$\sigma_4$	$\sigma_5$
$\sigma_n(0)$	5.81	1.99	0.808	0.117	0.0139
$\sigma_n^{av}$	6.47	2.28	0.736	0.0733	0.005
$R(\%)$	100.	35.3	11.4	1.13	0.080

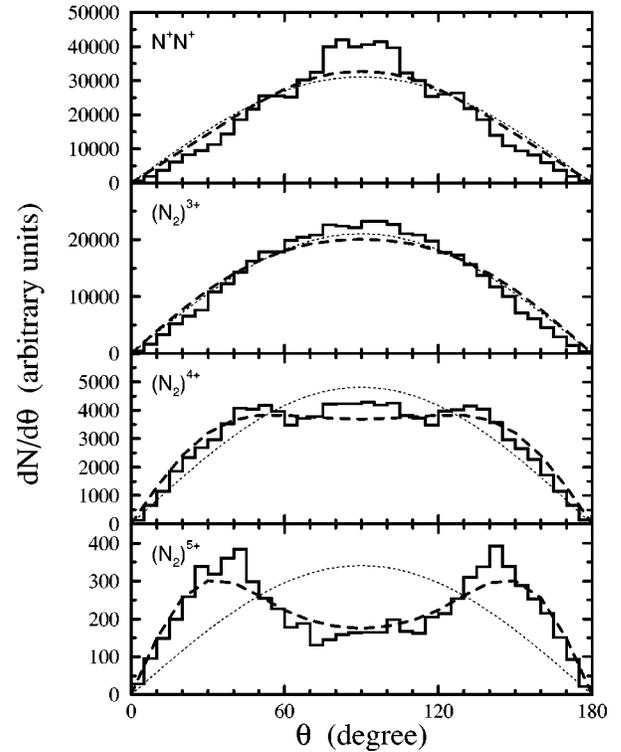


FIG. 7. Alignment dependence for the multiple ionization of  $N_2$  molecules in collisions with 200-keV  $He^+$  ions. Histograms show the experimental results [20], dashed curves are the theoretical results of the SED-LDA model, dotted curves show the sine distribution. All curves are normalized to have the same area as the experimental histograms, and  $\theta$  is the angle between the ion beam direction and the molecular axis.

Thus one can expect a significant contribution of transfer ionization to the double ionization. For the transfer ionization of diatomic molecules another orientation effect is known (see [42] and references therein), which is interpreted as an interference effect in electron capture from the two centers of the molecule. This interference effect leads to a predominant ionization of molecules having their axis perpendicular to the beam. Our estimates show that the interference effect [42] can be very large at least for the dominant electron capture to the  $2s, 2p$  levels of the  $He$  ion. Therefore, the observed maximum at  $90^\circ$  in the angular distribution for double ionization (and possibly some excess cross section for triple and fourfold ionization) can probably be explained by the interference effect in transfer ionization. Since competing ionization mechanisms give different orientation effects, one can expect that such measurements and a quantitative analysis of the angular distributions could shed more light on the dominant ionization mechanism.

#### IV. CONCLUSION

We have described an application of the SED model, together with an LDA calculation of the deposited energy, to a description of multiple ionization of simple molecules by fast ion impact. Using only one adjustable parameter, the statistically averaged square of the autoionization matrix element, we have been able to describe the removal of up to 10 electrons from diatomic molecules with the cross sections

differing by as much as 5 orders of magnitude. For the considered systems this free parameter is practically independent of the projectile and the target and varies slightly with the projectile energy. The model describes well not only the total cross sections averaged over molecular orientations, but also the strong variation of the multiple ionization cross section with the angle between the molecular axis and the ionic beam.

The simplicity of the model allows one to extend it to the cases of more complicated polyatomic molecules where more elaborate calculations are hardly feasible. The application of the model so far is limited to an energy range where capture processes are not significant. Electron capture and related processes of transfer ionization can be considered as competing processes, and in a first approximation their con-

tribution can be added to the cross sections calculated within the SED model.

#### ACKNOWLEDGMENTS

This work has been supported by the Center for Interdisciplinary Research of Bielefeld University in the research group "Interaction of Oriented Molecules" and in part by the Deutsche Forschungsgemeinschaft (DFG). V.N.K. gratefully acknowledges the financial support from the Alexander von Humboldt Stiftung. N.M.K. is grateful to the Center for Interdisciplinary Research of Bielefeld University for its hospitality. We are indebted to Professor J.H. McGuire for illuminating discussions and to Dr. K. Stark for his help with the MOLPRO calculations.

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