Electron loss of fast projectiles in collisions with molecules

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(Received 2 May 2011; published 19 July 2011)

The single- and multiple-electron loss of fast highly charged projectiles in collisions with neutral molecules is studied within the framework of a nonperturbative approach. The cross sections for single-, double-, and triple-electron losses are calculated for the collision system $Fe^{q+} \rightarrow N_2$ ($q = 24, 25, 26$) at the collision energies 10, 100, and 1000 MeV/nucleon. The effects caused by the collision multiplicity and the orientation of the axis of the target molecule are treated. It is shown that the collision multiplicity effect leads to considerable differences for the cases of perpendicular and parallel orientations of the molecular axes with respect to the direction of the projectile motion, while for chaotic orientation such an effect is negligible.

DOI: 10.1103/PhysRevA.84.012704 PACS number(s): 34.50.−s, 34.10.+x

I. INTRODUCTION

Stripping, or electron loss by heavy projectiles in the collisions with atoms, has been the subject of extensive studies during the last decade. Various approaches for the theoretical study of projectile electron loss cross sections in collisions with neutral targets are presented in Ref. [1] and in the monograph by Voitkiv and Ullrich [2], where a detailed discussion of the experimental results can be found. In the theoretical study of such processes, both the treatment of the effects caused by the strong field of the projectile and accounting for the transitions in the target state are important. Such a treatment requires using nonperturbative methods for the calculation of stripping cross sections [3,4]. Recently such an approach was developed on the basis of sudden perturbation approximation and was used for the study of electron losses by fast highly charged projectiles with neutral gas atoms [5,6].

Despite the fact that considerable progress has been made in the theoretical study of electron loss by fast projectiles, most of the treatments are restricted because they consider atomic targets. However, the study of electron loss by fast highly charged projectiles in collisions with molecular targets is of fundamental and practical importance because of the number of new effects that cannot be observed in the case of atomic targets.

One such effect is the strong dependence of the ionization (both for the projectile and the target) cross section on the orientation of the molecular axis with respect to the direction of the collision velocity. Previously the existence of such an effect was mentioned in Refs. [7–9], where the ionization of molecular targets in their collisions with fast highly charged ions was studied.

Another important effect that appears in the collision of fast highly charged projectiles with molecules is the so-called collision multiplicity effect [10]. It implies that after the collision with the first atom of the target, before the relaxation, the projectile collides with the second atom being in the excited state. Considerable contributions from multiplicity effects to the ionization cross section and energy losses of fast highly charged ions with diatomic molecules [10] and nanoparticles [11] were found recently.

In particular, it was shown in Refs. [10,11] that the orientation and multiplicity effects occur when the time interval between two subsequent collisions is less than the relaxation time. Furthermore, it is clear that the multiple collisions of the projectile with target molecule atoms occur when the direction of the projectile motion is close to the orientation of the target molecule axis. Qualitatively, the role of the orientation effects can be understood for a diatomic molecule as follows. In the study of fast collisions of highly charged projectiles with diatomic molecules, one should take into account the two-step processes which include collisions of the projectile with the first atom of the molecule which cannot relax into the ground state before the collision with the second atom of the target. We note that in the present paper we consider the case when the time between two subsequent collisions is much shorter than the characteristic period of the projectile electrons. Therefore, the processes we deal with can be interpreted as the variation of the "atomic-double-slit" processes in which projectile electrons interact with both molecular centers in a coherent way (see, e.g., Ref. [12]). It is important to note that the collision multiplicity effects contribute only to the projectile ionization or excitation cross section without causing any changes in the transitions of target states. Indeed, in subsequent collisions with each atom of the target molecule the ionization or excitation occurs in different atoms. It is clear that the above arguments are true for polyatomic molecules, too.

In this paper we develop a nonperturbative approach which is based on the use of sudden perturbation approximation and we use it for the calculations of the electron loss cross sections of fast highly charged projectiles in collisions with polyatomic molecules. The method allows to take into account all the transitions, in both target and projectile electronic states. Moreover, it is possible to achieve considerable simplification of the expressions for the electron-loss cross sections in the cases of high enough projectile charges and multielectron targets.

The calculations of the electron-loss cross sections and their dependence on the orientation of the molecular axis are presented for single, double, and triple stripping of fast highly

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charged iron ions. It is also shown that the collision multiplicity effect leads to considerable difference between the stripping cross sections for the cases of parallel and perpendicular orientation of the molecular axis with respect to the direction of projectile motion. However, for a chaotic orientation of the axis of the target molecule, such an effect is negligible. Finally, it should be noted that for the calculation of the multiple stripping cross section of the fast highly charged projectiles in their collisions with multielectron targets one should use nonperturbative methods, since the Born approximation is not applicable for these processes. Our approach also can be considered a nonperturbative one. However, our results concerning the dependence of the cross sections on the molecule's axis orientation can be obtained within the perturbative approach, too—at least for a single-electron ionization.

This paper is organized as follows. In the next section we present a detailed description of the approach used, including a derivation of the nonperturbative expressions for the stripping cross section and its applications for the calculations of the multiple stripping cross sections of fast highly charged projectiles in collisions with the nitrogen molecule, N_2. Section III presents discussions of the results, and in Sec. IV we give some concluding remarks.

II. THEORETICAL BACKGROUND

The system we are going to treat includes a fast highly charged projectile colliding with a molecule that consists of multielectron atoms (number of electrons, $N_A \gg 1$). For such a collision system, the cross section for excitation or ionization of the projectile from states $|0\rangle$ to $|k\rangle$ (provided arbitrary transition can occur in the target state) with high accuracy (error is proportional to $1/N_A$) in the Glauber approximation can be written as [6,10]

$$\sigma = \int \left| \langle k| \exp\left\{ -\frac{i}{v} \int_{-\infty}^{+\infty} U dX \right\} |0\rangle \right|^2 d^2\mathbf{b}, \tag{1}$$

where $\mathbf{v}$ is the projectile velocity and the x axis is directed along the vector $\mathbf{v}$. If $\mathbf{b}$ is the impact parameter, and the target position is fixed with one of the nuclei being at the origin of the coordinate system, then the position of the projectile nucleus can be given by the vector $\mathbf{R} = (X,\mathbf{b})$. Atomic units are used here and in the following. In Eq. (1) the potential U describes the interaction of the projectile electrons with the target, which is considered a lengthy object. In other words, the cross section in Eq. (1) is expressed in terms of the charge density of target electrons [10,11]. Formally, Eq. (1) has the same form as that for the ionization cross section in the "frozen" approximation for the target electrons. However, as shown in Ref. [4], for the multielectron targets ($N_A \gg 1$) this formula describes (with the error order of $1/N_A$) the transition cross section for the projectile electrons summed over the complete set of final states of the target electrons. For a molecule consisting of multielectron atoms, the electronic density is almost the same as the sum of that for isolated atoms. Therefore, for such a case we can consider the target as consisting of isolated and noninteracting atoms [10,11]. We describe the charge density of each atom in the target within the Hartree-Fock-Slater model [13], in which the spatial charge density can be written as

$$\rho_m(r) = -\frac{Z_m}{4\pi r} \sum_{i=1}^{3} A_{m,i}\alpha_{m,i}^2\, e^{-\alpha_{m,i} r}, \qquad \sum_{i=1}^{3} A_{m,i} = 1, \tag{2}$$

where Z_m is the charge of the mth atomic nucleus, and $A_{m,i}$ and $\alpha_{m,i}$ are the tabulated constants that can be found in Ref. [13]. The potential created by a molecule at the point $\mathbf{r}$ can be written in terms of potentials created by each atom of the molecule:

$$\varphi(\mathbf{r}) = \sum_{m=1}^{N} \frac{Z_m}{d_m} \Phi_m(d_m), \tag{3}$$

where N is the number of atoms in the target, d_m is the distance from the mth nucleus in the target to the observation point $\mathbf{r}$, and $\Phi_m(r)$ is the screening function for the mth atom. In the Hartree-Fock-Slater model, the screening function can be written as

$$\Phi_m(r) = \sum_{i=1}^{3} A_{m,i} \exp(-\alpha_{m,i} r).$$

Furthermore, let us introduce the following notation: $\mathbf{r}_p$ is the coordinate of the projectile electrons with respect to its nucleus ($p = 1,2,\ldots,N_P$), N_P is the total number of projectile electrons, and $\mathbf{R}_m = (X_m,\mathbf{b}_m)$ are the distances between the projectile nucleus and the nucleus of the mth atom of the target. If $\mathbf{b}_m$ is the impact parameter with respect to the mth atom, then $d_m = |\mathbf{R}_m + \mathbf{r}_p|$.

Therefore, the interaction potential between the target and projectile electrons can be written as

$$U = -\sum_{p=1}^{N_P} \varphi(\mathbf{r}_p) = -\sum_{m=1}^{N}\sum_{p=1}^{N_P} \frac{Z_m}{|\mathbf{R}_m + \mathbf{r}_p|} \Phi_m(|\mathbf{R}_m + \mathbf{r}_p|). \tag{4}$$

Taking into account Eq. (4) for the "eikonal phase" involved in Eq. (1), we have

$$\begin{aligned} \chi &= -\frac{1}{v} \int_{-\infty}^{+\infty} U dX \\ &= \sum_{m=1}^{N} \frac{2Z_m}{v} \sum_{p=1}^{N_P} \sum_{i=1}^{3} A_{m,i} K_0(\alpha_{m,i}|\mathbf{b}_m + \mathbf{s}_p|), \end{aligned} \tag{5}$$

where $K_0(z)$ is the lowest-order McDonald function and $\mathbf{s}_p$ is the projection of the vector $\mathbf{r}_p$ onto the impact parameter plane. As shown in Ref. [6], Eq. (1) with the eikonal phase given in Eq. (5) is applicable for relativistic collisions, too. Thus Eq. (1) describes the cross section for the transition of the projectile electrons from the initial state $|0\rangle$ to a state $|k\rangle$ under the assumption that arbitrary transitions can occur in the states of target electrons. The accuracy of the formula depends on the number of target electrons, N_A; i.e., the error is of order of the quantity $\sim 1/N_A$ for $N_A \gg 1$.

In the following we consider the highly charged projectiles whose "visible" (effective) charge, Z_P, is much larger than unity (for instance, for the Fe^{10+} projectile, $Z_P = 10$, while

the nucleus charge is $Z = 26$). Then the characteristic size of the projectile is much less than that of an atom of the target molecule. Therefore, we can assume that the mean field created by an atom acts uniformly on the projectile electrons, corresponding to expansion of the eikonal phase in Eq. (5) in terms of a small parameter, s_p/b. This allows us to rewrite Eq. (1) for orthogonal $|0\rangle$ and $|k\rangle$ in the following form:

$$\sigma = \int \left| \langle k| \exp\left(-i \sum_{m=1}^{N} \mathbf{q}_m \sum_{p=1}^{N_P} \mathbf{r}_p \right) |0\rangle \right|^2 d^2\mathbf{b}, \qquad (6)$$

where

$$\mathbf{q}_m = \frac{2Z_m}{v} \sum_{i=1}^{3} \alpha_{m,i} A_{m,i} K_1(\alpha_{m,i} b_m) \frac{\mathbf{b}_m}{b_m} \qquad (7)$$

has the meaning of the momentum transfer to each projectile electron due to the collisions with the mth atom of the target, and $K_1(z)$ is the first-order McDonald function.

Furthermore, we note that Eq. (6) can be applied for the collisions of highly charged ($Z_P \gg 1$) projectiles with the neutral molecules consisting of multielectron atoms. This requires fulfilling by Z_t the condition $Z_t \gg 1$, with $t = 1,2,\ldots,N$ being the number of atoms in the target; i.e., Z_1 is the nucleus charge of the first atom of the target, Z_N is the nucleus charge of the Nth atom, and so on.

Since the expression for the cross section is derived within the Glauber approximation, the energy of the projectile, E, should be much larger than that of the projectile-target interaction U (i.e., $E \gg U$ and $kL \gg 1$, where k is the projectile momentum and L is the interaction radius for the potential U). In the case of a neutral molecular target whose size is much larger than that of the projectile, as the quantity L one can take the characteristic size of the target. It is clear that this condition is fulfilled for a fast highly charged projectile. In addition, to make use of our approach we have to assume that the sudden perturbation approximation is valid; i.e., for each target atom (interaction) collision time, $\tau_c \sim L/v$ between the projectile and target is much shorter than the period of the fastest (inner) electron, τ_e, to be ionized:

$$\tau_c \ll \tau_e.$$

Fulfillment of this condition means that target electrons cannot change their positions during the collision time. In this case target electrons can be considered as being in fixed positions during the collision [14]. For relativistic projectiles the above condition can be written as $\sqrt{1 - v^2/c^2}\ L/v \ll \tau_e$. The quantity τ_e can be estimated for each fixed collision system. For a multielectron collision system, with most of the electrons to be ionized being in outer shells, we have $\tau_e \sim 1$.

As mentioned above, in order to make use of Eq. (6) for the calculation of the projectile stripping cross sections in the collision with neutral molecular targets, the following condition should be fulfilled:

$$Z_P = Z - N_P \gg 1,$$

where N_P is the number of projectile electrons before the collision and Z is the charge of the projectile nucleus. This means that the (unscreened) charge of the projectile should be large enough that the projectile is considered a highly charged particle. An important point in the calculations of the projectile's multiple stripping cross section in the collisions with polyatomic molecules is the effects of collision multiplicity and effects of molecular axis orientations. To include these effects in the consideration, we need to assume that two or more atoms in the target are on the same line which is parallel to the vector $\mathbf{v}$. To demonstrate collision multiplicity and molecular axis orientation effects, we consider the simplest target, a diatomic molecule. We use $\mathbf{L}$ to denote the orientation vector of the molecular axis and assume that it is directed along the line connecting nuclei of two atoms in the molecule. Then the cross section presented by Eq. (6) is the function of vector $\mathbf{L}$; i.e., $\sigma = \sigma(\mathbf{L}) = \sigma(\theta,\phi)$. Furthermore, we represent vector $\mathbf{L}$ in terms of spherical coordinates with the angles ϕ, θ and assume the axis of the spherical coordinate system is directed along the vector $\mathbf{v}$.

In the following we are interested in the stripping cross section averaged over the angle ϕ:

$$\sigma(\theta) = \frac{1}{2\pi} \int \sigma(\theta,\phi)\, d\phi. \qquad (8)$$

Angle θ is called the orientation angle of the molecular axis. Then the multiplicity effect can be characterized in terms of θ-dependent relative correction; δ is defined as $\delta = (\sigma(\theta) - \sigma_\perp)/\sigma_\perp$, where $\sigma_\perp$ is the stripping cross section at $\theta = \pi/2$. It is easy to see that $\sigma_\perp$ described the collision when the target molecular axis is perpendicular to the projectile direction.

III. RESULTS AND DISCUSSION

Let us now apply Eq. (6) to the collision system consisting of a fast Fe^{q+} projectile and N_2 molecule. For simplicity we consider the cases when $q = 24,25,26$. Let us start from the simplest case: stripping of a hydrogenlike projectile in the collision with a diatomic molecule for which the cross section can be written as

$$\sigma(\theta,\phi) = \int d^2b \int d\mathbf{k} \mid \langle \mathbf{k} \mid \exp\{-i(\mathbf{q}_1 + \mathbf{q}_2)\mathbf{r}\} \mid 0\rangle \mid^2 . \qquad (9)$$

Here $\mathbf{r}$ is the coordinate of the projectile electron with respect to the projectile nucleus, $\mathbf{k}$ is the momentum of the projectile electron lost, and $\mathbf{q}_j$ is the momentum transfer that can be written as $\mathbf{q}_j = \frac{2Z_A}{v} \sum_{i=1}^{3} \alpha_i A_i K_1(\alpha_i b_j) \frac{\mathbf{b}_j}{b_j}$, where $\mathbf{b}_j$ is the impact parameter with respect to the jth nucleus of the target ($j = 1,2$).

Using Eqs. (8) and (9), we have calculated the stripping cross section $\sigma(\theta)$ of the hydrogenlike projectile Fe^{25+} in the collision with nitrogen molecule N_2 for different orientation angles and collision energies. A quantity we are interested to analyze is the correction to the projectile stripping cross section due to the collision multiplicity, which is given as $\delta(\theta) = [\sigma(\theta) - \sigma_\perp]/\sigma_\perp$. In Fig. 1, the results of the calculation of such a quantity for collision system $Fe^{25+} \rightarrow N_2$ are presented. The continuous line in this figure represents $\delta(\theta)$ for the collision energy 10 MeV/nucleon, while long-dashed and short-dashed lines are the results for the energies 100 and 1000 MeV/nucleon, respectively. The angle θ is given in radians.

Now let us consider stripping of a heliumlike projectile, Fe^{25+}-N_2 in the collision with a N_2 molecule. In this case,

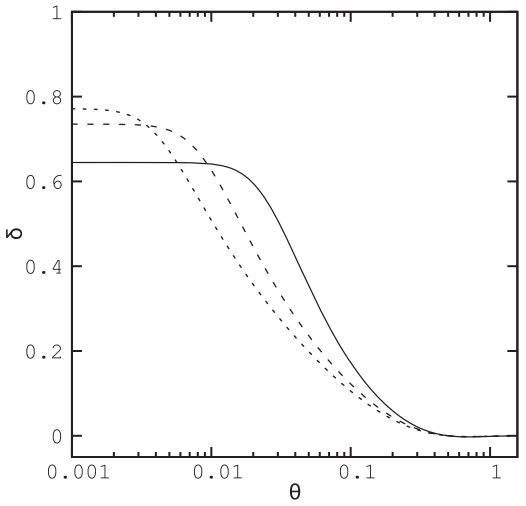

FIG. 1. The dependence of the quantity $\delta(\theta) = (\sigma(\theta) - \sigma_\perp)/\sigma_\perp$ on the orientation of the target molecule axis θ for the collision system $Fe^{25+} \rightarrow N_2$ with $\sigma_\perp = \sigma\ (\theta = \pi/2)$. The continuous line is the result for the collision energy 10 MeV/nucleon; the long-dashed line is the result for the collision energy 100 MeV/nucleon and the short-dashed line is $\delta(\theta)$ for the energy 1000 MeV/nucleon. The values of θ are given in radians.

according to Eq. (6) the cross section for the transition of a projectile from state $|0,0\rangle$ to $|n_1,n_2\rangle$ can be written as

$$\sigma(\theta,\phi) = \int d^2b \mid \langle n_1,n_2 \mid e^{-i(\mathbf{q}_1+\mathbf{q}_2)(\mathbf{r}_1+\mathbf{r}_2)} \mid 0,0\rangle \mid^2 , \qquad (10)$$

with $\mathbf{r}_1$ and $\mathbf{r}_2$ being the coordinates of the projectile electrons with respect to the projectile nucleus.

Following Refs. [15,16], in our calculations (final and initial), two-electron states of the projectile are described as symmetric products of one-electron hydrogenlike wave functions with effective charges equal to the degree of ionization. Single $\sigma^{1+}(\theta)$ and double $\sigma^{2+}(\theta)$ electron loss cross sections can be calculated using Eq. (10). Here the cross section for double stripping, $\sigma^{2+}(\theta)$, corresponds to a transition of both electrons into the continuum state, while the single projectile ionization cross section $\sigma^{1+}(\theta)$ describes the transition of one of the electrons into the continuum by exciting another one into a state of the discrete spectrum. Correspondingly, the quantity δ can be estimated for both cases. In Figs. 2(a), 2(b), and 2(c) we plotted δ as a function of orientation, θ for the collision energies 10, 100, and 1000 MeV/nucleon, respectively. The continuous line in this figure is the result for single electron loss, $\delta = [\sigma^{1+}(\theta) - \sigma_\perp^{1+}]/\sigma_\perp^{1+}$, while the dashed line describes the calculation of δ for double ionization, i.e., $\delta = [\sigma^{2+}(\theta) - \sigma_\perp^{2+}]/\sigma_\perp^{2+}$.

Finally, we use Eq. (6) for the calculation of multiple stripping of a three-electron (lithiumlike) projectile, Fe^{23+}, in the collision with a N_2 molecule. In this case the electron loss cross section describing the transition from state $|0,0,0\rangle$ to state $|n_1,n_2,n_3\rangle$ can be written as

$$\sigma = \int d^2b \mid \langle n_1,n_2,n_3 \mid e^{-i(\mathbf{q}_1+\mathbf{q}_2)(\mathbf{r}_1+\mathbf{r}_2+\mathbf{r}_3)} \mid 0,0,0\rangle \mid^2 , \qquad (11)$$

where $\mathbf{r}_1$, $\mathbf{r}_2$, and $\mathbf{r}_3$ are coordinates of projectile electrons.

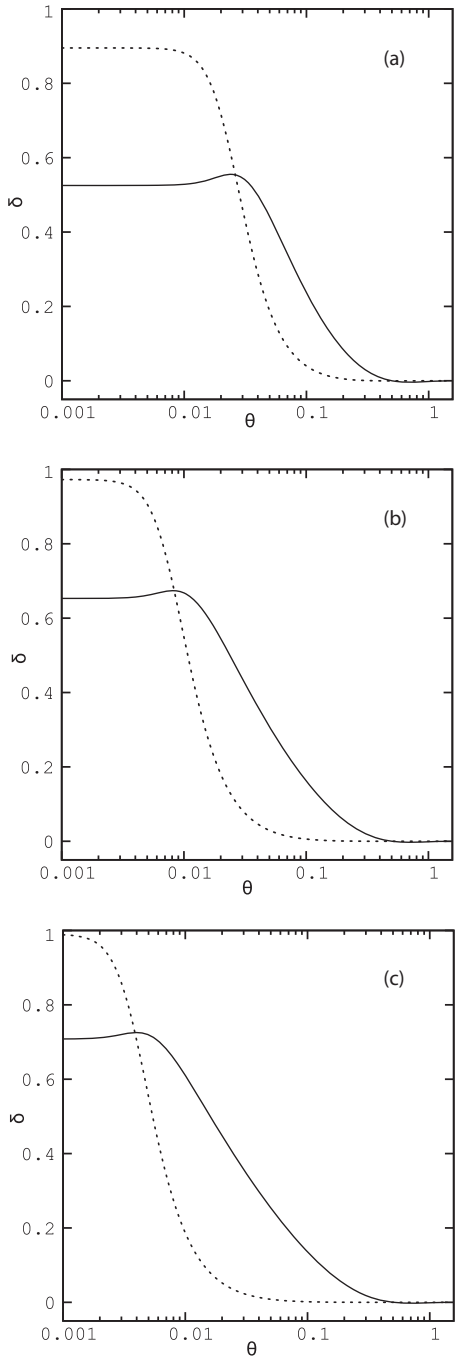

FIG. 2. The dependence of the quantity $\delta(\theta) = (\sigma(\theta) - \sigma_\perp)/\sigma_\perp$ on the orientation of the target molecule axis θ for the collision system $Fe^{24+} \rightarrow N_2$ for the energies (a) 10, (b) 100, and (c) 1000 MeV/nucleon. The continuous line is the result for single electron loss, and the dashed line is the result for double electron loss by projectile.

As in the case of a heliumlike projectile, the wave functions of the three electron states are taken as the symmetric product of one-electron (hydrogenlike) wave functions with effective charges equal to the degree of ionization discussed in Refs. [15,16]. Single, double, and triple electron loss cross sections of the Fe^{23+} projectile can be calculated using Eq. (11). Single electron loss implies ionization of one

electron, while the other two electrons can be excited into any state of the discrete spectrum. Similarly, in the case of double electron loss, two electrons are lost, while the other electron is excited into any bound state. As in the cases of single and double electron losses, we have calculated the quantity δ for the cases of single-, double-, and triple-electron losses. Figures 3(a), 3(b), and 3(c) present the results of calculation of $\delta(\theta)$ for the collision energies 10, 100, and 1000 MeV/nucleon, respectively. The continuous line in this figure is the result for single-electron loss, $\delta = (\sigma^{1+}(\theta) - \sigma_\perp^{1+})/\sigma_\perp^{1+}$; the long-dashed line describes calculation of δ for double ionization, $\delta = (\sigma^{2+}(\theta) - \sigma_\perp^{2+})/\sigma_\perp^{2+}$; and the short-dashed line is the plot of $\delta = (\sigma^{3+}(\theta) - \sigma_\perp^{3+})/\sigma_\perp^{3+}$.

As is seen from Figs. 1–3, for all collision energies, stripping cross sections considerably depend on the orientation angle θ; the difference between perpendicular and parallel orientations is about 50%–100%. It is easy to estimate from the geometrical analysis the value of the angle θ at which the multiplicity effect becomes essential. Since in all cases the projectile collides with the neutral atoms of the target molecule whose sizes are ~ 1, for the internuclear distance of target atoms denoted by L, the orientation can be estimated as $\theta \leqslant 1/L$. Therefore, for the nitrogen molecule, for instance, we have $L = 2.07$, which gives the estimate $\theta \leqslant 0.5$. This can be seen also from Figs. 1–3.

Also, it should be noticed that the behavior of the quantity δ is almost the same for all collision partners. To check this, we have performed calculations (similar to the above) for one-, two-, and three-electron Ni and Xe projectiles colliding with N_2, O_2, and Au_2 targets. The difference from the above treated collision systems we observed for these systems was too small to present them in this paper.

Finally, since in the experiment the cross sections for chaotic orientation are usually measured, we have performed calculations of the stripping cross section averaged over the angle θ assuming uniform distribution on the orientation angle:

$$\overline{\sigma} = \int \sigma(\mathbf{L}) \frac{d\Omega}{4\pi} = \int_0^\pi \sigma(\theta) \frac{1}{2} \sin\theta \, d\theta.$$

As shown in the results of such calculations, the corrections due to the collision multiplicity for the case of chaotic orientation is too small: on the order of 0.1%. In this case the difference between the cross sections $\overline{\sigma}$ and $\sigma_\perp$ is very small. Thus the collision multiplicity effect is considerable only for the case of regular (nonchaotic) orientation of the molecular axis, while for the chaotic orientation it becomes negligible.

IV. CONCLUSIONS

The electron losses of fast highly charged projectiles in collisions with neutral molecules has been studied. Based on the Glauber approximation, a nonperturbative approach is developed to estimate single and multiple stripping cross sections. Using the method, single-, double-, and triple-electron-loss cross sections of the fast Fe^{25+}, Fe^{24+}, and Fe^{23+} ions in the collisions with a N_2 molecule are calculated. The effect of collision multiplicity, caused by the collisions of the projectile with separate atoms of the target, is analyzed.

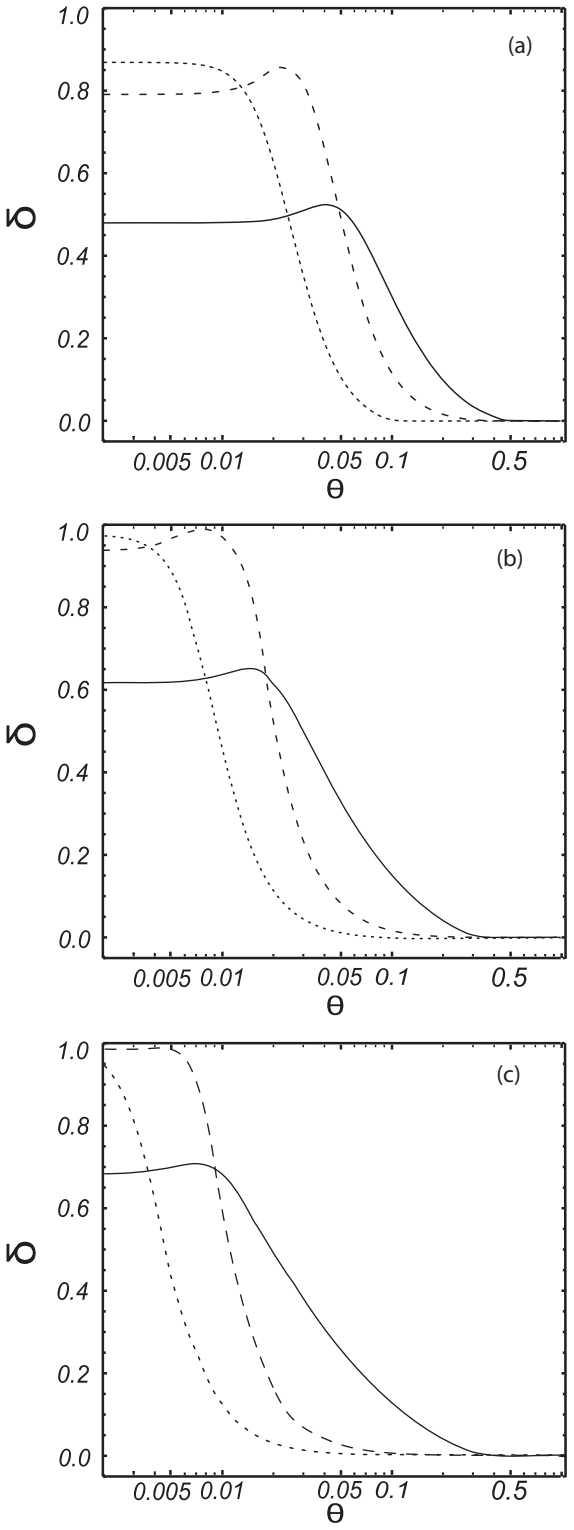

FIG. 3. The dependence of the quantity $\delta(\theta) = (\sigma(\theta) - \sigma_\perp)/\sigma_\perp$ on the orientation of the target molecule axis θ for the collision system $Fe^{23+} \rightarrow N_2$ for the energies (a) 10, (b) 100, and (c) 1000 MeV/nucleon. The continuous line is the result for single-electron loss, the long-dashed line is the result for double-electron loss, and the short-dashed line is the result for triple-electron loss by projectile.

It is shown that the multiplicity effect is essential for the case when the orientation of the target molecule axis is

parallel or perpendicular to the projectile velocity direction, whereas for chaotic orientation such an effect is negligible. We note that in all the cases we calculate the total electron loss cross sections; i.e., we perform summation (integration) over all the continuum states of the electron after the ionization. Therefore, in the present paper we did not discuss the energies and directions of the emitted electrons. The above developed method is rather simple for application and can be used for any (polyatomic) target molecules and for the projectiles of arbitrarily high (including relativistic) velocities.

ACKNOWLEDGMENTS

We are grateful to Davron Matrasulov for valuable discussions and useful comments. This work is supported by the Russian Foundation of Basic Studies (Grant No. 08-02-00711-a) and by the Russian Federation President (Grant No. MK-3592.2011.2).

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