Postcollisional effects in multiple ionization of diatomic molecules by ion impact

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Multiple electron ionization of diatomic molecular targets is studied for impact of proton beams. It is shown that for these molecular targets, as it was previously predicted for atomic ones, postcollision Auger type emission following inner-shell vacancy production dominates the reaction at high enough impact energies. Moreover, it is proven that this dominance increases as the ionization degree increases.

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I. INTRODUCTION

The present work deals with multiple electron ionization of diatomic molecular targets by impact of fast ion beams. During the collision process, when the projectile interacts with the target electrons, the emission to the continuum of one or more of them can be produced, generating vacancies in the different molecular orbitals. Thus in the time interval corresponding to a first step, the residual molecule is promoted to a highly excited state as a consequence of its *direct* interaction with the ion beam. In a postcollisional step, once the projectile is far away, the vacancies in the inner orbitals are filled by residual target electrons, such that the excess of energy is simultaneously employed to induce photon emission or to produce ionization of other bound electrons, which are thus autoionized through Auger-type emission [1–3]. In fact, this Auger-type emission corresponds to intershell Auger and/or intrashell Coster-Kronig electron ionization. These ejected electrons generate new vacancies, so that photon emission and/or electron autoionization are again produced. These mechanisms can be subsequently repeated so that additional Auger-type cascades can be observed in the target [4].

Special attention was devoted to the Auger autoionization reaction since its discovery in the 1920s, due to the important role that it plays in different scientific areas (see, for example, Refs. [5,6]). In more recent years, active research was developed in order to understand the physical mechanisms involved in multiple ionization of noble gases. In a pioneering work by DuBois and Manson [7], a detailed analysis for the case of proton impact on Ar and Kr showed that Auger transitions following the production of direct inner-shell vacancies dominate the multiple ionization cross sections at high enough impact energies. It was also suggested that the same behavior could be expected for Ne. Later, Heber *et al.* [8] measured single and multiple ionization cross sections for impact of 1 MeV/amu-O^{x+} and F^{x+} ions on Ar atoms. They found that to describe the experimental results it was necessary to take into account a charge multiplication effect of the recoil target via Auger decay of *L*-shell vacancies. However, they claimed that this charge multiplication effect should not appear for Ne targets.

An independent-particle model was employed to study multiple electron loss of the target in the cases of Ne and Ar atoms, where single-particle time-dependent Schrödinger equations were solved using the basis generator model (BGM), describing the target bound state in terms of the optimized potential model [9–11]. For proton impact a good representation of experimental data [7,12–15] was obtained. However, for both targets and high ionization degree the theoretical description overestimates (underestimates) the existing measurements at intermediate (high) collision energies. The discrepancy at high impact energies was corrected including Auger type postcollisional emission in the theoretical model [9–11,14]. Instead of using a multinomial statistics as it was done in previous works, Archubi *et al.* [16] solved the transport equation for an ion traveling through an inhomogeneous electron density corresponding to the atom, which leads to a Poisson distribution. Ionization probabilities were obtained using the shell-to-shell local plasma approximation with the Levine and Louie dielectric function [17] to take into account the energy ionization threshold of each shell. This model gives an adequate description of experimental total cross sections, for different *q*-ionization degrees, for proton impact on Kr and Xe targets. Also, a statistical energy-deposition model (SED) was used by Kabachnik *et al.* [18,19] to study multiple ionization of atoms and diatomic molecules. They include a free parameter in the calculation of *q*-fold ionization probabilities to consider the contribution of nondirect ionization processes. Recently, we have studied the cases of the impact of protons on Ne and Ar atoms, employing a binomial statistics in a similar way as it has been done before in Refs. [9–11] but where single ionization probabilities were calculated using the continuum distorted wave eikonal initial state (CDW-EIS) and exponential models [20]. The total cross section results for different *q*-ionization degrees confirmed the behavior obtained by using the BGM model. At high enough energies, the multiple ionization reaction is dominated by Auger type emission following previous process of direct ionization produced by the projectile. Moreover, this dominance becomes more important as *q* increases.

The main scope of the present work is to analyze the influence of postcollisional Auger type emission on the process of multiple electron ionization of diatomic molecules after interaction with a fast ion beam. We select the case of impact of protons on CO molecules for which experimental data exist at impact energies high enough to neglect electron

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capture reactions. As in our above mentioned work on atomic targets, we employ the independent electron approximation and binomial distributions to calculate cross sections for q-ionization degree. The diatomic molecule is described as an ensemble of two independent atoms with their nuclei separated by the corresponding molecular equilibrium distance. In a previous work [21], we have proposed this simple molecular representation to study the dependence of multiple ionization cross sections with the molecular orientation. Using the CDW-EIS model we found good agreement with experimental cross sections, differential in the molecular orientation, for impact of 5.9 MeV/amu-Xe^{18+} on N_2 and O_2 targets [22,23]. Theoretical total cross sections for different target ionization degrees will be also presented for impact of protons on N_2 and O_2 in order to provide an incentive for the development of new experiments. Atomics units will be used in the following except where otherwise stated.

II. THEORY

Let us consider that a bare ion of nuclear charge Z_P interacts with a diatomic molecular target at intermediate and high impact energies. Using the straight line version of the impact parameter approximation, we will consider that the target internuclear vector remains as *frozen* during the collision. This approximation will be valid if the collision time is much smaller than the target vibrational ones; it is, at high enough projectile velocities. Considering that the target is randomly oriented and averaging over all target orientations, the total cross section for q-ionization degree can be expressed as

$$\sigma_q = \frac{1}{2}\int_0^{\pi}\int_0^{2\pi}\int_0^{\infty} P_q(\mathbf{b}) b \sin\theta \, db \, d\varphi \, d\theta, \qquad (1)$$

where $\mathbf{b}=(b,\varphi,\pi/2)$ is the impact parameter vector considering a reference frame with the origin in the middle point of the internuclear distance of the molecule, the z axis is given by the direction and sense of the impact velocity $\mathbf{v}$ and the y axis is contained in the plane defined by the z axis and the internuclear vector (see Fig. 1). Also, in Eq. (1), $P_q(\mathbf{b})$ is the q-degree ionization probability, b is the modulus of the impact parameter, φ is the angle formed by the vector $\mathbf{b}$ and the x axis, and θ is the angle formed by the internuclear vector $\mathbf{R}$ and the z axis.

A. Multiple direct ionization

By simplicity we assume that the molecule is composed by its two constituent atoms separated by the ground state–molecular equilibrium distance, each one of these j-atomic centers $(j=1,2)$ containing N_j electrons. This image of the molecular target was previously used with some success by different authors [21,24–26] to study multiple ionization of diatomic molecules. During the ionization reaction each one of these centers loses q_j electrons, in such a way that the total number of electrons lost by the target is $q=q_1+q_2$.

So, the q-ionization degree probability can be expressed as

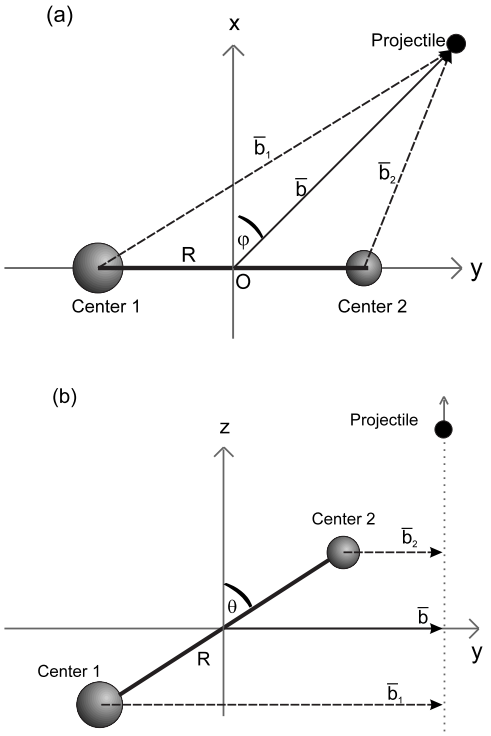

FIG. 1. Schematic representation of the spatial orientation of a diatomic molecule: (a) side view; (b) top view.

$$P_q(\mathbf{b}) = \sum_{\substack{q_1,q_2=0 \\ q_1+q_2=q}}^{N_1,N_2} P_{q_1}(b_1) P_{q_2}(b_2), \qquad (2)$$

with $b_j=[b^2+(R/2\sin\theta)^2+(-1)^j R\ b\ \sin\theta\cos\varphi]^{1/2}$ the modulus of the impact parameter vector $\mathbf{b}_j$ defined with respect to each one of the j-molecular centers (see Fig. 1). Into an independent electron approximation and using a binomial distribution, the probability $P_{q_j}(b_j)$ for the q_j-ionization degree of the j center given in Eq. (2) is given by

$$P_{q_j}(b_j) = \sum_{q_{k_j}=0}^{N_{k_j}} \prod_{k_j=1s,2s,2p,\ldots} \binom{N_{k_j}}{q_{k_j}} p_{k_j}^{q_{k_j}}(b_j) \times [1-p_{k_j}(b_j)]^{N_{k_j}-q_{k_j}}, \qquad (3)$$

$$\sum_{k_j=1s,2s,2p,\ldots} q_{k_j}=q_j$$

In this expression, k_j indicated the orbital k of the atomic center j, N_{k_j} and q_{k_j} are the total number of electrons of the k_j orbital and the number of electrons ionized from the k_j-orbital, respectively, with p_{k_j} the single ionization probability per electron of k_j.

Following the previous work by Galassi *et al.* [20], we employ different theoretical models to calculate p_{k_j}: the CDW-EIS [27–29] and two versions of the exponential model. In the CDW-EIS model, the atomic bound states described in the Roothaan-Hartree-Fock approximation [30] are distorted by eikonal phases accounting for the *active* electron (the one to be ionized) moving in a continuum state

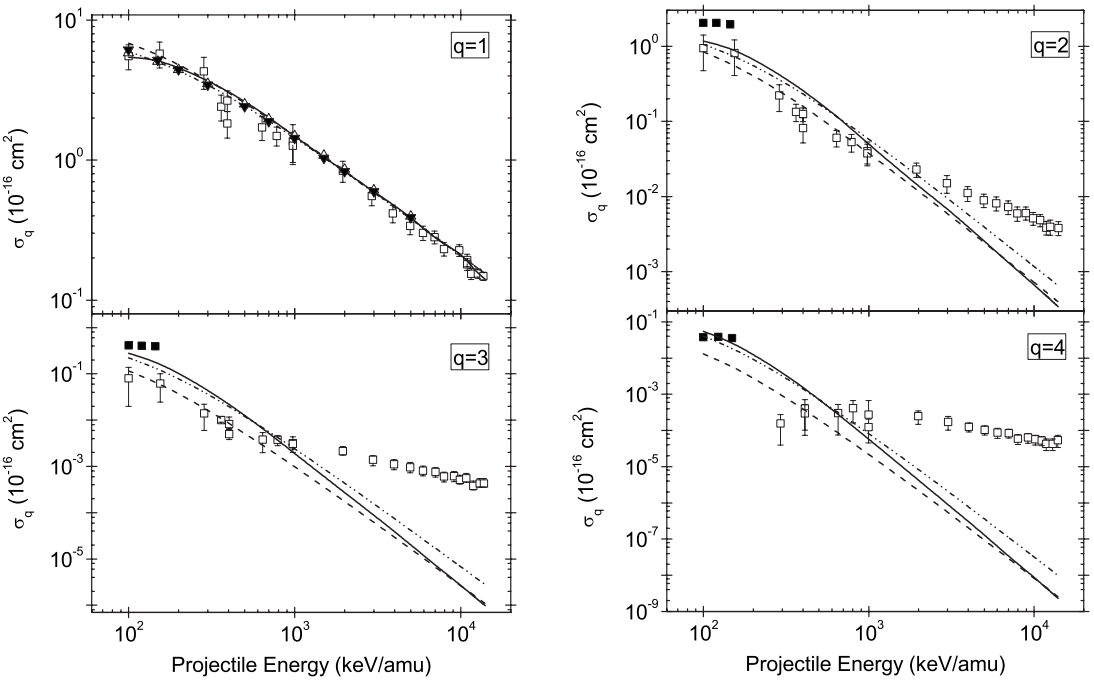

FIG. 2. Total cross sections for q-ionization degree of CO molecules by proton impact. Theory without Auger-type contributions: — CDW-EIS model; —··— EMRHF model, — — — EMB model. Experiments: Ref. [34] (□), Ref. [37] (■), Ref. [35] (△), and Ref. [36] (▼).

of the projectile. In the exit channel, the continuum state of this *active* electron is described as a triple product of a plane wave, a continuum factor of the projectile, and an effective coulombic continuum factor of the residual target. In the exponential model, the probabilities p_{k_j} corresponding to the atomic orbital of each j-molecular center are supposed to present a decreasing exponential behavior as a function of the impact parameter b_j:

$$p_{k_j}(b_j) = P_0^{k_j} \exp(-b_j/r_{k_j}), \tag{4}$$

where the parameters r_{k_j} are chosen according to two criteria: (i) as the corresponding Hartree-Fock mean radius of the different subshells k_j [31], and (ii) as the radius given by a Bohr atomic model, $r_{k_j}=n_{k_j}/(-2\varepsilon_{k_j})^{1/2}$, with ε_{k_j} the Roothaan-Hartree-Fock orbital energy of each k_j [30] and n_{k_j} the principal quantum number. These two series of r_{k_j} values give origin to the two different exponential models: the Roothaan-Hartree-Fock exponential model (EMRHF) in the case of using the prescription (i) and the Bohr exponential model (EMB) in the case of using the prescription (ii). The parameters $P_o^{k_j}=p_{k_j}(b_j=0)$ are obtained by fitting the single ionization total cross sections per electron obtained employing Eq. (4) with the CDW-EIS ones corresponding to the different k_j orbitals.

B. Multiple ionization including postcollisional effects

Let us consider now that inclusion of postcollisional electron emission. Let α be the total number of electrons postcollisionally emitted. Thus the probability of emission of q electrons (including the processes of direct and postcollisional ionization) can now be expressed as

$$P_q(\mathbf{b}) = \sum_{\substack{q_1,q_2,\alpha=0 \\ q_1+q_2+\alpha=q}}^{N_1,N_2,\alpha^{\max}} \sum_{\substack{\alpha_1,\alpha_2=0 \\ \alpha_1+\alpha_2=\alpha}}^{\alpha_1^{\max},\alpha_2^{\max}} P_{q_1,\alpha_1}(b_1) P_{q_2,\alpha_2}(b_2), \tag{5}$$

where N_j (with $j=1,2$) is the total number of electrons in the j center and $P_{q_j,\alpha_j}(b_j)$ is the probability of emission of q_j electrons by direct interaction with the projectile and of α_j postcollisional electrons. This probability takes the form

$$P_{q_j,\alpha_j}(b_j) = \sum_{\substack{q_{k_j}=0 \\ \sum_{k_j=1s,2s,2p,\ldots} q_{k_j}=q_j}}^{N_{k_j}} \mathcal{P}(q_{1s_j}, q_{2s_j}, \ldots; \alpha_j) \times \prod_{k_j=1s,2s,2p,\ldots} \binom{N_{k_j}}{q_{k_j}} p_{k_j}^{q_{k_j}}(b_j)[1-p_{k_j}(b_j)]^{N_{k_j}-q_{k_j}} \tag{6}$$

with $\mathcal{P}(q_{1s_j}, q_{2s_j}, \ldots, \alpha_j)$ the probability of emission of α_j postcollisional electrons from the jth center, after emission of q_{k_j} electrons from each k_j orbital due to direct ionization. According to Ref. [11], $\mathcal{P}(q_{1sj}, q_{2sj}, q_{2pj}, \alpha_j)$ for each jth center can be obtained using the expression

$$\mathcal{P}(q_{1s_j},q_{2s_j},q_{2p_j};\alpha_j) = \sum_{\substack{\alpha_{h_j}=0 \\ \Sigma\alpha_{h_j}=\alpha_j}}^{\alpha_{h_j}^{\max}} \prod_{h_j=1}^{q_{1s_j}} \mathfrak{M}_{1s_j}(q_{2s_j},q_{2p_j};\alpha_{h_j}) \prod_{k=1+q_{1s_j}}^{q_{1s_j}+q_{2s_j}} \mathfrak{M}_{2s_j}(q_{2p_j};\alpha_{h_j}), \tag{7}$$

where $\mathfrak{M}_{k_j}(\ldots,\alpha_{h_j})$ is the probability for postcollisional production of α_{h_j} electrons after production of only one vacancy in the inner subshell k_j, accompanied by direct outer-shell ionization. The information that more than one outer-shell electron can be emitted by direct ionization must be included in these probabilities. To calculate them it is assumed that the postcollisional ionization reaction is independent of the nature of the primary vacancy production [32]. Thus we use the experimental photoionization probabilities $\mathfrak{B}_{k_j}(\alpha)$ for the production of α electrons if only one electron is removed from the k_j subshell by interaction with the photon beam, while all the other electrons remain bound to the target [33]. For the different shells of the atomic compounds of the molecule, we have

$$\mathfrak{M}_{2s_j}(q_{2p_j};\alpha_{h_j}) = \begin{cases} 1 - \dfrac{N_{2p_j} - q_{2p_j}}{N_{2p_j}}[1 - \mathfrak{B}_{2s_j}(0)] & \text{for } \alpha_{h_j} = 0 \\ \dfrac{N_{2p_j} - q_{2p_j}}{N_{2p_j}}\mathfrak{B}_{2s_j}(\alpha_{h_j}) & \text{for } \alpha_{h_j} > 0 \end{cases},$$

$$\mathfrak{M}_{1s_j}(q_{2s_j},q_{2p_j};\alpha_{h_j}) = \begin{cases} 1 - \dfrac{N_{2s_j} + N_{2p_j} - q_{2s_j} - q_{2p_j}}{N_{2s_j} + N_{2p_j}}[1 - \mathfrak{B}_{1s_j}(0)] & \text{for } \alpha_{h_j} = 0 \\ \dfrac{N_{2s_j} + N_{2p_j} - q_{2s_j} - q_{2p_j}}{N_{2s_j} + N_{2p_j}}\mathfrak{B}_{1s_j}(\alpha_{h_j}) & \text{for } \alpha_{h_j} > 0 \end{cases} \tag{8}$$

Using Eqs. (1) and (5)–(8), it is possible to calculate multiple ionization cross sections.

III. RESULTS AND DISCUSSION

Let us first consider the case of multiple ionization of a CO target by its direct interaction with the projectile. In Fig. 2, total cross sections for impact of protons are presented for different ionization degrees as a function of the collision energy. For single electron ionization, results obtained using the three theoretical models described above are in close agreement between them and in general good agreement with existing experimental data from Wells *et al.* [34] over all the energetic range considered. The coincidence is extended to the measurements of total ionization cross sections given by Rudd *et al.* [35,38]. For higher q values: CDW-EIS cross sections are in accordance with EMHF (EMB) ones at intermediate (high) impact energies. For $q=2,3$ and collision energies lower than 1 MeV/amu, EMB results and experiments show good agreement but CDW-EIS and EMHF models overestimate the measurements at intermediate energies. For $q=4$, CDW-EIS and EMHF results tend to the data obtained by Siegmann *et al.* [36] at the lower energies considered. Moreover, for $q \geq 2$ and energies larger than approximately 1 MeV/amu a large discrepancy is found between all theoretical results and experimental data [34]. It should be noted that this discrepancy increases as q increases and cross sections can differ by four orders of magnitude for $q=4$! The same behavior was previously obtained for the case of atomic targets and it was attributed to contributions coming from postcollisional Auger type effects [7,9–11,20].

We consider, in the following, the inclusion of Auger type emission after creation of inner-orbital vacancies by the projectile beam. To our knowledge, experimental $\mathfrak{B}_{k_j}(\alpha)$ photoionization probabilities were not measured either for C or for O atoms. So, we estimate them from measurements existing for Ne [33], considering the C and O cases as Ne ions with absence of four and two outer electrons, respectively. We take thus $\mathfrak{B}_{1s}^{C}(\alpha)=\mathfrak{W}_{1s}^{Ne}(0,4;\alpha)$, $\mathfrak{B}_{2s}^{C}(\alpha)=\mathfrak{W}_{2s}^{Ne}(4;\alpha)$ and $\mathfrak{B}_{1s}^{O}(\alpha)=\mathfrak{W}_{1s}^{Ne}(0,2;\alpha)$, $\mathfrak{B}_{2s}^{O}(\alpha)=\mathfrak{W}_{2s}^{Ne}(2;\alpha)$ for the C and O targets, respectively.

In Fig. 3, CDW-EIS, EMHF, and EMB total cross sections for $q=1,2,3,4$ including postcollisional Auger type emission are presented together with experimental data previously shown in Fig. 2. From this comparison it is evidenced that the postcollisional emission dominates the reaction of multiple electron emission from the diatomic CO molecule at high enough impact energies. Moreover, as it was observed for atomic targets, the effect becomes more pronounced as the q-ionization degree increases. In order to test the adequacy of the prescription described above to estimate $\mathfrak{B}_{k_j}(\alpha)$ we have also extrapolated the tendency of experimental photoionization probabilities as the atomic nuclear charges decrease, for a vacancy in the K and L shells. The modifications obtained in the corresponding σ_q cross sections are much smaller than contributions coming from Au-

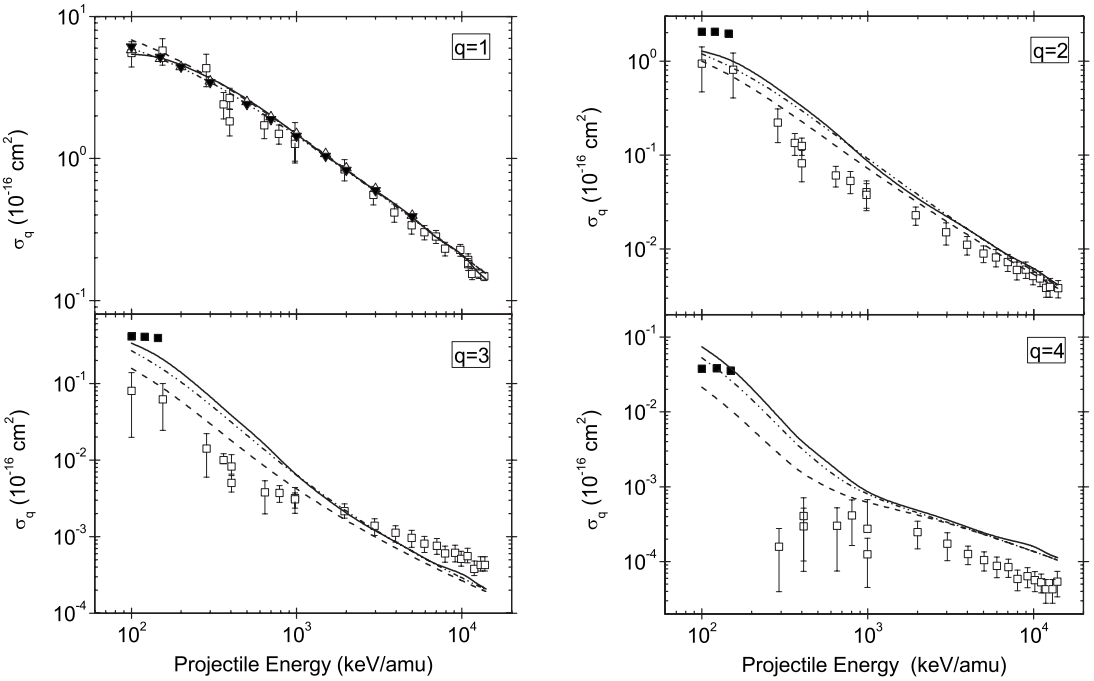

FIG. 3. Total cross sections for q-ionization degree of CO molecules by proton impact. Theory including Auger-type contributions: — CDW-EIS model; —··— EMRHF model, — — — EMB model. Experiments: Ref. [34] (□), Ref. [37] (■), Ref. [35] (△), and Ref. [36] (▼).

ger type autoionization, confirming thus the validity of our predictions.

Two possible sources of the overestimation of experimental data from reference [34] given by theoretical calculations at lower impact energies for $q=2,3$ could be the use of an independent-particle model to describe multiple electron reactions and/or the use of a simple representation of the molecule. For these ionization degrees the EMB model gives the better representation of the measurements. However, for $q=3,4$, CDW-EIS and EMHF results tend to the experimental data from Ref. [36] at energies lower than 200 keV/amu.

In Figs. 4 and 5, CDW-EIS σ_q cross sections are presented for impact of protons on two other diatomic targets of biological and astrophysical interest, N_2 and O_2, for which experimental data for total ionization exists [35–38]. We employ a similar procedure to the one used for CO to evaluate direct ionization and Auger type contributions. Again, σ_1 cross sections give a good representation of measurements. Unfortunately, no experimental σ_q cross sections are given in the literature for $q \geq 2$. For these multiple ionization reactions, the behavior obtained theoretically is analogous to the case of CO targets. Auger type emission dominates at high enough collision energies and this comportment becomes more important as the q-ionization degree increases.

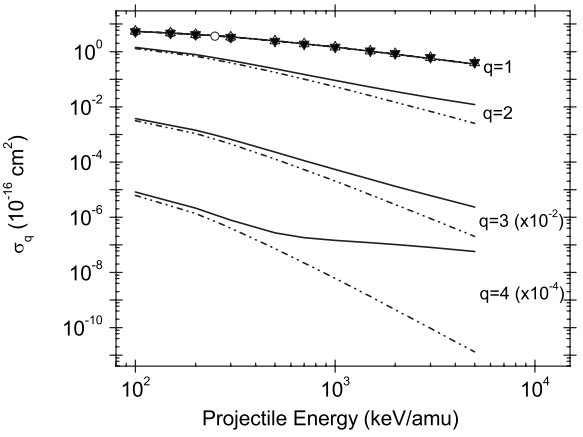

FIG. 4. Total cross sections for q-ionization degree of N_2 molecules by proton impact. CDW-EIS Model calculations: — *with* and —··— *without* Auger-type contributions. Experiments: Ref. [35] (△), Ref. [36] (▼), and Ref. [38] (○).

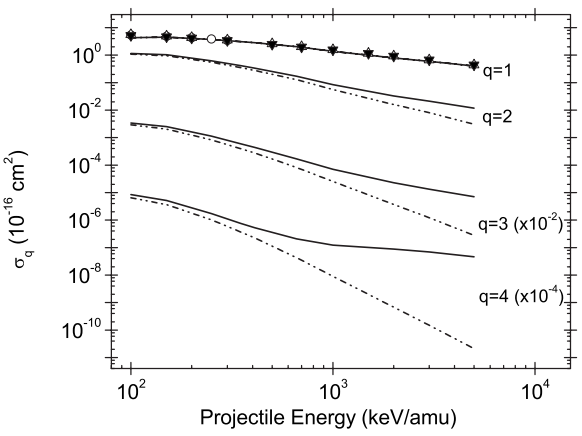

FIG. 5. Total cross sections for q-ionization degree of O_2 molecules by proton impact. CDW-EIS Model calculations: — *with* and —··— *without* Auger-type contributions. Experiments: Ref. [35] (△), Ref. [36] (▼), and Ref. [38] (○).

IV. CONCLUSIONS

Multiple ionization of diatomic molecular targets was studied for impact of fast proton beams. A simple description of the molecule as two atomic compounds separated by the equilibrium internuclear distance is employed. Contributions from direct ionization by interaction of the target with the projectile and from postcollisional Auger type autoionization are disentangled. Direct ionization is evaluated using the CDW-EIS and exponential models and photoionization experimental data are employed to estimate the postcollisional reactions. By comparison with experiments it is shown that, for CO molecules, Auger type emission dominates the multiple electron ionization reaction at high enough collision energies, as it was previously shown to happen with atomic targets. The effect is proven to increase as the ionization degree increases. A similar behavior is also theoretically found for other diatomic molecules like N_2 and O_2 but for which multiple ionization experimental cross sections do not exist. New experimental results related to these systems would be welcome.

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