Multiple electron emission from noble gases colliding with proton beams, including postcollisional effects

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(Received 30 November 2006; revised manuscript received 28 February 2007; published 11 May 2007)

The process of multiple electron ionization of Ne and Ar noble gases produced by impact of proton beams is studied in the framework of the independent-electron model. The role played by different mechanisms is analyzed, including intershell Auger and intrashell Coster-Kronig electron emission, which follow the production of vacancies due to direct interaction of the projectile with the target electrons. The present results, obtained with the continuum distorted wave–eikonal initial state (CDW-EIS) approximation, confirm previous predictions given by a different theoretical model. Semiempirical approximations are introduced by using analytical single-ionization probabilities with adjustable parameters determined from CDW-EIS total cross sections. The small computational time required to calculate multiple ionization cross sections with these semiempirical approximations and the good agreement found with existing experimental data and with results obtained with more elaborated theoretical models make them good candidates to study electron emission from complex targets.

DOI: 10.1103/PhysRevA.75.052708 PACS number(s): 34.50.Fa

I. INTRODUCTION

Different physical mechanisms contribute to the reaction of multiple electron ionization of atomic targets by impact of ion beams. They can be produced during the collision as the result of the direct interaction of the projectile with the target electrons or as the result of a postcollisional relaxation of the residual target when the projectile is far away. In the first case, processes of direct target ionization with or without electron capture can contribute. In the second case, shell vacancies produced in the first step of the reaction can be followed by intershell Auger and/or intrashell Coster-Kronig electron emission. Moreover, these postcollisional mechanisms can provoke additional Auger-type cascades [1].

In recent years, active research has been devoted to understanding the physics involved in the reaction of multiple ionization of noble gases. This activity was partially motivated by discrepancies found between existing experimental data and theoretical predictions. A theoretical time-dependent independent-particle model was employed to analyze the cases of Ne and Ar targets [2,3]. Single-particle time-dependent Schrödinger equations were solved using the basis generator method (BGM), where the target ground state was described in terms of the optimized potential method. A very good description of experiments was obtained for single-electron loss of the target for the cases of protons impacting on Ne and Ar atoms [1,4–6]. However, for higher degrees of target electron loss, the theory predicted larger cross sections than the corresponding ones given by measurements at intermediate impact velocities. On the contrary, at large enough collision velocities the theoretical results gave a large underestimation of experimental cross sections. Discrepancies at these high energies were attributed to postcollisional Auger processes following previous K-hole production in the Ne target case [2]. For Ar targets it was also suggested that they could be due to L-shell vacancy production followed by Auger and Coster-Kronig transitions combined with independent M-shell vacancy formation [3].

Recent measurements of absolute cross sections for single and multiple ionization of Ne by fast protons were reported in the collision energy range 0.75–3.5 MeV, showing through the use of simple theoretical estimations that contributions from time-delayed postcollisional mechanisms play a principal role in the description of multiple electron emission reactions [7]. Further theoretical work was reported using the above-mentioned nonperturbative BGM to solve the independent-particle Schrödinger equations [8]. A multinomial statistics was applied to calculate total cross sections for q degrees of target electron loss, confirming the relevance of Auger-type postcollisional effects for $q>1$, at high impact energies. It was proven that the importance of these effects increases as q increases. However, a theoretical overestimation of experimental data is still observed at intermediate collision velocities for q values larger than 1.

Very recently, a many-electron description was introduced to study multiple ionization of noble gases [9]. This model, which is based on the transport equation for an impacting ion in an inhomogeneous electron density and where ionization probabilities were obtained using the shell-to-shell local plasma approximation with the Levine and Louie dielectric function [10] to take into account the shell binding energy, gives a good representation of experimental data for Kr and Xe targets. However, as could be expected for a many-electron approximation, a large overestimation of BGM results and so of experimental data is found for multiple ion-

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ization at intermediate collision energies when lighter targets are considered.

The interest of the present work is focused on the possibility of generating simple models to allow multiple ionization cross sections to be obtained, consuming short computational time. We will base the generation of these simple models on the use of the continuum distorted wave–eikonal initial state (CDW-EIS) approximation, which has been employed with success to study single ionization of multielectronic targets interacting with ion beams [11–14]. We will treat collisions in a high enough energy domain where electron capture processes can be neglected. As in previous works, the independent-particle approximation will be used. It is an almost obligatory limitation of theoretical models due to the difficulty of considering in a proper way the dynamical electron correlation for multielectronic targets. The present work is motivated by the possible application of simple models in the determination of the response to ion irradiation of complex targets like, for example, polymer chains, DNA, liquid water, or biological tissue. In the case of irradiation of biological matter, it is necessary to know how the projectile beam deposits energy on the target. It is a very complicated process, which is usually reduced to the study of water radiolysis. Thus, in the first stage, which is known as the physical one, the target molecules are ionized, so that simple and multiple ionization cross sections must be used as input to Monte Carlo codes employed to describe this stage, and consequently the simulation of the following physicochemical and chemical stages [15–18]. Moreover, for radiotherapy an efficient and rapid determination of radiation dose is required. Thus, it is important in this case to avoid the use of multiple ionization models involving large computational times, like the BGM and CDW-EIS approximation.

Atomic units will be used except where otherwise stated.

II. THEORY

The reaction of multiple emission of target electrons provoked by the impact of protons is here described using an independent-particle model. Thus, the interaction between the electrons is considered in the mean-field approximation, so that the dynamical evolution of each one of them is assumed to be completely independent of the evolution of the other ones. In order to calculate exclusive probabilities and cross sections for q degrees of target ionization, a binomial statistical distribution is employed [19,20]. Following Refs. [7,8], the probability for ionization degree q, as a function of the impact parameter ρ, is given by the following expression:

$$P_q(b) = \sum_{q_1+q_2+\cdots+q_N+\alpha=q} \prod_{i=1}^{N} \binom{N_i}{q_i} p_i^{q_i}(\rho) \times [1-p_i(\rho)]^{N_i-q_i} \mathcal{P}(q_1,q_2,\ldots,q_N,\alpha), \quad (1)$$

where q_i and N_i indicate the ionization degree and the occupation number of the i subshell, respectively, α is the number of postcollisional electrons emitted from the target, which is composed of N subshells, and q is the total number of ejected electrons. Also, $p_i(\rho)$ is the probability for single ionization per electron for each atomic subshell and $\mathcal{P}(q_1,q_2,\ldots,q_N,\alpha)$ is the probability for postcollisional emission of α electrons after the creation of q_i vacancies in each one of the i subshells. In the same expression, the binomial coefficient is given by $\binom{N_i}{q_i}=N_i!/q_i!(N_i-q_i)!$. According to Ref. [8], the probability $\mathcal{P}(q_1,q_2,\ldots,q_N,\alpha)$ can be obtained using the expression

$$\mathcal{P}(q_1,q_2,\ldots,q_N,\alpha) = \sum_{\alpha_j=0}^{\alpha} \prod_{j=1}^{q_1} \mathcal{M}_1(q_2,q_3,\ldots,q_N,\alpha_j) \prod_{j=q_1+1}^{q_1+q_2} \mathcal{M}_2(q_3,\ldots,q_N,\alpha_j) \times \cdots \times \prod_{j=q_1+q_2+\ldots+q_{N-2}+1}^{q_1+q_2+\ldots+q_{N-1}} \mathcal{M}_{N-1}(q_N,\alpha_j) \quad (2)$$

considering that

$$\sum_{j=1}^{q_1+q_2+\ldots q_{N-1}} \alpha_j = \alpha. \quad (3)$$

In Eq. (2), $\mathcal{M}_x(\ldots,\alpha_j)$ is the probability for the postcollisional production of α_j electrons provoked by only one vacancy in the inner subshell x, accompanied by direct outershell ionization. These probabilities must take into account the fact than more than one electron can be directly ionized, reducing thus the number of electrons that can be ejected after the collision. In order to estimate them, we use the experimental photoionization probabilities $m_x(\alpha_j)$ for the postcollisional production of α electrons if only one electron is directly removed from the subshell x, while all other electrons remain bound to the target [21]. Thus, it is easy to obtain that

$$\mathcal{M}_1(q_2,q_3,\alpha) = \begin{cases} 1 - \dfrac{8-q_2-q_3}{8}[1-m_1(0)] & \text{for } \alpha = 0, \\ \dfrac{8-q_2-q_3}{8} m_1(\alpha) & \text{for } \alpha > 0, \end{cases}$$

$$\mathcal{M}_2(q_3,\alpha)=\begin{cases}1-\dfrac{6-q_3}{6}[1-m_2(0)] & \text{for } \alpha=0,\\ \dfrac{6-q_3}{6}m_2(\alpha) & \text{for } \alpha>0,\end{cases} \tag{4}$$

for the case of a Ne target, and

$$\mathcal{M}_1(q_2,q_3,q_4,q_5,\alpha)=\begin{cases}1-\dfrac{16-q_2-q_3-q_4-q_5}{16}[1-m_1(0)] & \text{for } \alpha=0,\\ \dfrac{16-q_2-q_3-q_4-q_5}{16}m_1(\alpha) & \text{for } \alpha>0,\end{cases}$$

$$\mathcal{M}_2(q_3,q_4,q_5,\alpha)=\begin{cases}1-\dfrac{14-q_3-q_4-q_5}{14}[1-m_2(0)] & \text{for } \alpha=0,\\ \dfrac{14-q_3-q_4-q_5}{14}m_2(\alpha) & \text{for } \alpha>0,\end{cases}$$

$$\mathcal{M}_3(q_4,q_5,\alpha)=\begin{cases}1-\dfrac{8-q_4-q_5}{8}[1-m_3(0)] & \text{for } \alpha=0,\\ \dfrac{8-q_4-q_5}{8}m_3(\alpha) & \text{for } \alpha>0,\end{cases}$$

$$\mathcal{M}_4(q_5,\alpha)=\begin{cases}1-\dfrac{6-q_5}{6}[1-m_4(0)] & \text{for } \alpha=0,\\ \dfrac{6-q_5}{6}m_4(\alpha) & \text{for } \alpha>0,\end{cases} \tag{5}$$

for the case of an Ar target. The subscripts 1, 2, 3, and 4 in Eqs. (4) and (5) correspond to the 1s, 2s, 2p, and 3s subshells, respectively. The probability $\mathcal{P}(q_1,q_2,\ldots,q_N,\alpha)$ accounts for the production of multiple inner-shell vacancies during the collision.

Total cross sections for q-order ionization of atoms can be obtained as

$$\sigma_q=2\pi\int \rho P_q(\rho)d\rho. \tag{6}$$

To calculate the single-electron probabilities $p_i(\rho)$ we employed the CDW-EIS and the semiempirical exponential (EM) models [1,22]. In the CDW-EIS model, the initial electron orbital wave function is distorted by a multiplicative eikonal phase associated with the projectile-electron Coulomb interaction. In the exit channel, the emitted electron is described by a double product of a plane wave and two continuum factors, associated with its interactions with the residual target and with the projectile. Thus, the electron is considered to move in the combined field created by the residual target and the projectile [11,13]. CDW-EIS single-electron probabilities $p_i(\rho)$ are calculated following the procedure indicated in Ref. [23]. For further details, the reader is referred to that work.

In the exponential model, it is assumed that the single-ionization probability per electron for a given subshell presents an exponential dependence on the impact parameter,

$$p_i(\rho)=p_i(0)\exp(-\rho/r_i), \tag{7}$$

where r_i is a characteristic interaction distance for each electronic orbital and $p_i(0)$ is the single-ionization probability for zero impact parameter.

CDW-EIS reduced probabilities $\rho p_i(\rho)$ for impact of protons on Ne atoms are presented in Fig. 1, at collision energies of 0.5 and 9 MeV. Roothaan-Hartree-Fock orbitals [24] and effective Coulomb continuum factors are employed in the initial and exit channels, respectively [11]. It can be observed that the maxima of reduced probabilities are produced for impact parameters that coincide approximately with the Hartree-Fock mean radius of the different subshells [25]. Also, the positions of these maxima present a weak dependence on the impact energy. As reduced probabilities calculated with expression (7) present maxima at $\rho=r_i$, we choose r_i according to two criteria: (i) as the corresponding Hartree-Fock mean radius for each different subshell, and (ii) as the radii given by the Bohr atomic model, $r_i=n_i/(-2\varepsilon_i)^{1/2}$, with ε_i the orbital energies (which in our case are chosen as the Rothaan-Hartree-Fock values). It must be noted that for im-

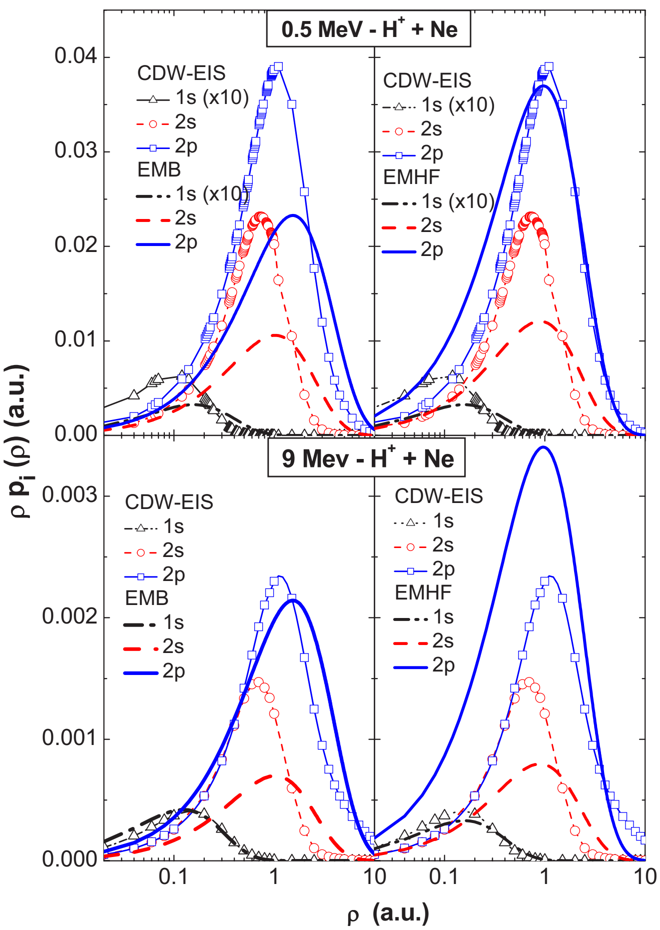

FIG. 1. (Color online) Reduced single-electron probabilities calculated applying the CDW-EIS, EMB, and EMHF models, for impact of protons on Ne atoms at collision energies of 0.5 and 9 MeV.

pact of protons on Ne and Ar targets it has been obtained using experimental data that, at high enough collision energies, r_i is approximately equal to the Hartree-Fock mean shell radius when outer-shell ionization is considered, presenting a weak dependence on the collision energy [1].

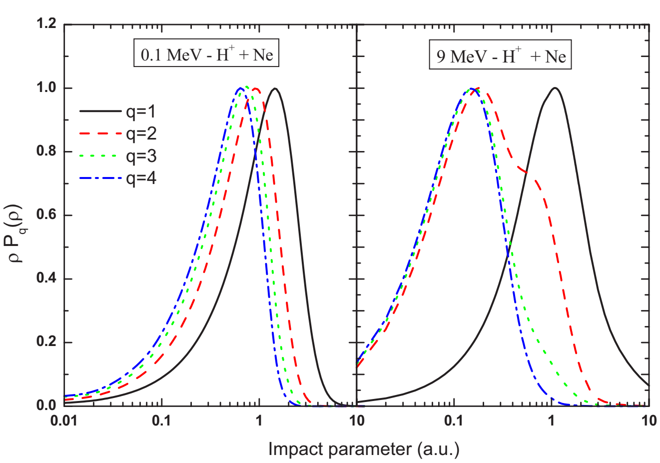

FIG. 2. (Color online) CDW-EIS reduced probabilities for multiple ionization of Ne for impact of protons at 0.1 and 9 MeV. The maxima of the curves are normalized to 1.

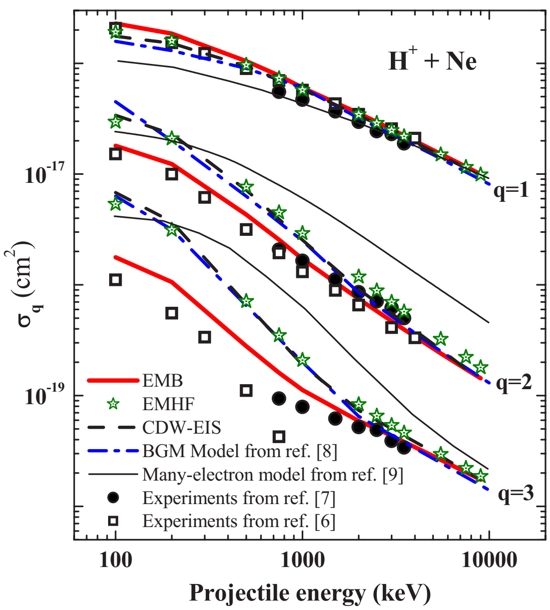

FIG. 3. (Color online) Total cross sections for ionization degree q of Ne by proton impact.

The parameter $p_i(0)$ is determined to reproduce CDW-EIS total cross sections for single-electron emission per electron for each subshell, but using expression (7). It is clear that with this simple expression only a general description of the qualitative behaviors of CDW-EIS impact parameter probabilities can be expected. The corresponding reduced probabilities obtained using the exponential model with Hartree-Fock (EMHF) or Bohr (EMB) subshell radii are also included in Fig. 1. The EMHF and EMB calculations are in reasonable agreement for the $1s$ and $2s$ subshells, but they differ for the $2p$ subshell. For the $2p$ case, which dominates the direct single-emission reaction, EMHF reduced probabilities are in good comparison with CDW-EIS ones for the impact energy of 0.5 MeV, but overestimate them for the 9 MeV case. On the contrary, EMB $2p$ calculations underestimate CDW-EIS results at 0.5 MeV but they compare fairly well for the 9 MeV case.

III. RESULTS AND DISCUSSION

CDW-EIS reduced probabilities for multiple ionization from Ne (normalizing their maxima to 1) are presented in Fig. 2. This figure shows that as q and the impact energy increase (for $q>1$), the collision is dominated by closer encounters. Similar behaviors also appear in the EMHF and EMB descriptions (not shown in the figure).

In Fig. 3, total cross sections σ_q (with $q=1$, 2, and 3) for impact of protons on Ne targets are shown. The present calculations obtained using the CDW-EIS, EMHF, and EMB models are compared with previous theoretical predictions corresponding to the BGM [8] and the many-electron model [9], and with experimental data [6,7]. The close agreement obtained between the CDW-EIS and BGM models for all q values confirms the theoretical predictions previously given by the latter. The results corresponding to the many-electron

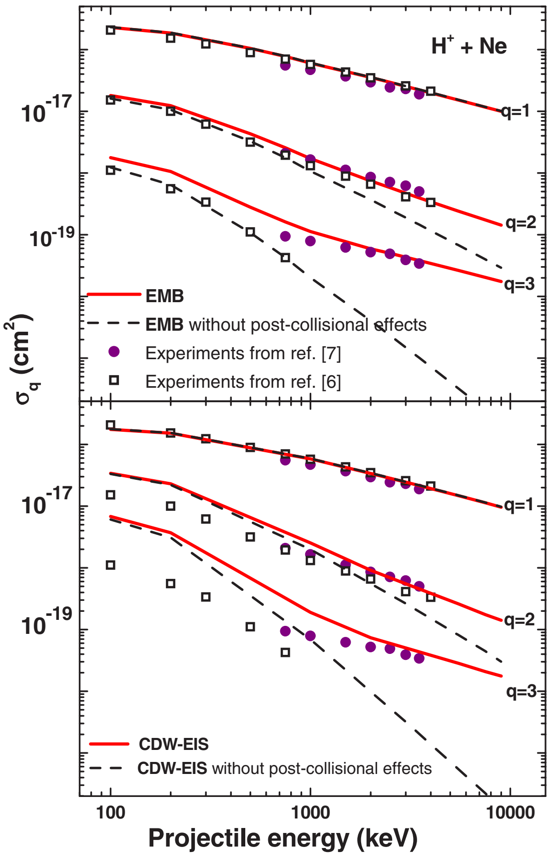

FIG. 4. (Color online) Comparison between total cross sections for ionization degree q of Ne, by proton impact, calculated with and without inclusion of postcollisional effects.

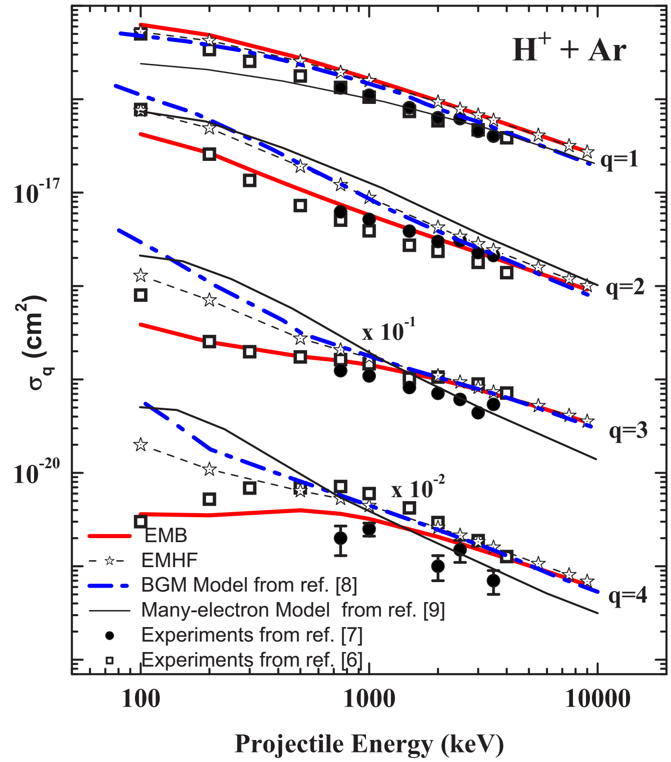

FIG. 5. (Color online) Total cross sections for ionization degree q of Ar by proton impact.

approximation overestimate experimental data for $q=2$ and 3 in the whole energetic range considered, for this target with a relative small number of electrons [9].

CDW-EIS and BGM cross sections are also in agreement with EMHF ones for all q values at intermediate energies, where *direct* multiple ionization dominates [7,8]. This behavior can be explained in terms of the concordance of CDW-EIS and EMHF reduced probabilities shown in Fig. 1 for the $2p$ subshell. At higher energies, EMHF results are slightly larger than those of the CDW-EIS model and BGM. For $q > 1$, the CDW-EIS, BGM, and EMHF results overestimate experimental data at collision energies smaller than approximately 2 MeV. The source of this disagreement with experiments could be the use of an independent-particle model to describe multiple electron reactions. Also, it should be noted that EMB cross sections are in better agreement with experiments for all q values over the whole energy domain considered. However, this agreement should be taken with caution considering the fact that we are dealing with a semiempirical approximation.

In Fig. 4, total cross sections σ_q obtained by employing the CDW-EIS and EMB models, with or without inclusion of postcollisional effects, are shown. As in previous calculations [7,8], the dominant role played by postcollisional electron emission for $q>1$, at high enough impact energies, is confirmed by the present results. According to Fig. 2, at high collision velocities the projectile preferably penetrates the K-shell atomic region, ionizing K-shell electrons and thus favoring postcollisional emission. In this velocity regime, EMHF and EMB reduced probabilities $\rho p_i(\rho)$ are in good agreement with CDW-EIS ones for emission from the K shell (see Fig. 1). It could explain the accordance obtained when the simple exponential models and the more elaborated CDW-EIS and BGM models are employed to calculate σ_q cross sections (see also Fig. 3).

The case of impact of protons on Ar targets can be analyzed from Figs. 5 and 6. EMHF and EMB calculations are done with the parameters $p_i(0)$ and r_i chosen following the criteria used for the Ne case. So r_i are taken as the Hartree-Fock and Bohr radii corresponding to the different subshells and $p_i(0)$ is determined using the numerical CDW-EIS model from Ref. [13]. From Fig. 5, EMHF and EMB cross sections σ_q are compared with previous BGM predictions [8] and with experimental data [6,7]. For $q=1$, a reasonably good agreement is obtained between the three models shown, even when both exponential approximations present values slightly larger than the BGM ones. Also, the three models overestimate the experimental results at intermediate energies. However, it can be considered that, in general, the different theories adequately describe the measurements. The many-electron model results coincide with those given by the three other approximations at high impact velocities, but underestimate them at lower energies. For larger q values, EMHF and BGM results are in agreement with experiment for high enough collision energies, given a large overestimation of them at lower ones. Also, for this target case, the EMB model presents results that are closer to measurements for the whole energetic domain here considered, except be-

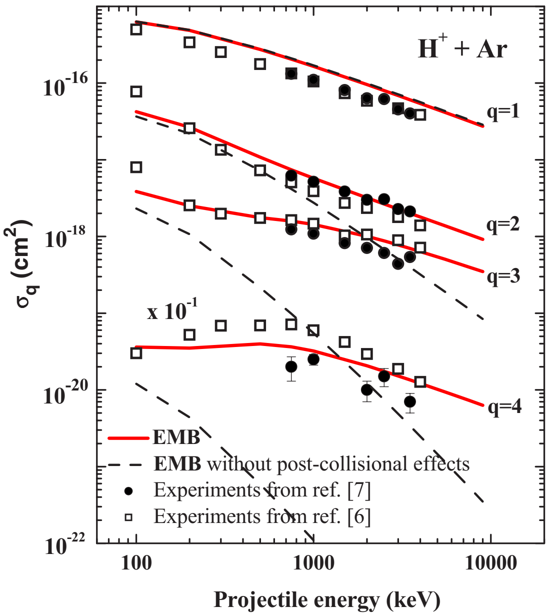

FIG. 6. (Color online) Comparison between total cross sections for ionization degree q of Ar, by proton impact, calculated with and without inclusion of postcollisional effects.

low 0.2 MeV. As could be expected, for $q>1$, the many-electron model gives a better representation of experiments than for the Ne case.

In Fig. 6, the importance of postcollisional effects for $q>1$ is emphasized using EMB calculations (with and without Auger-type contributions), confirming previous predictions given for Ar atoms [8]. It can be observed that postcollisional emission dominates the multielectron production at high enough energy for $q=2$, and dominates over the whole energetic domain for $q>2$. The importance of this effect increases as q increases.

IV. CONCLUSIONS

The multiple electron ionization of atomic targets has been considered, paying particular attention to the contribution of postcollisional mechanisms (Auger and Coster-Kronig emission). The cases of impact of protons on Ne and Ar noble gases, for which experimental data are available, are analyzed.

CDW-EIS calculations of cross sections for q ionization degree confirm previous theoretical predictions given by the BGM. It is also shown that for single-electron emission the maxima of reduced probabilities present a weak dependence on the collision velocity. However, the situation is different for multiple ionization, where for high velocities the maxima move to a small parameter region where K-shell ionization dominates, favoring postcollisional effects.

Theoretical exponential models, EMHF and EMB, with adjustable parameters obtained from the CDW-EIS model, are also introduced. Their main advantage is the small computational times required for the calculation of impact-parameter-dependent probabilities. Multiple ionization cross sections are then easily computed using a binomial statistical distribution. The EMHF model gives cross sections in general agreement with the corresponding ones resulting from the use of models that involve large-scale computers and long computational times. Also, the EMB calculations are closer to experimental values at intermediate impact energy for all ionization degrees. The use of these semiempirical models is supported by comparisons with CDW-EIS direct ionization cross sections. Thus, they offer the possibility of their application to the study of the interaction of proton beams with atomic targets and complex molecules, especially when a rapid computation of cross sections is required, as happens in radiotherapy.

ACKNOWLEDGMENTS

The authors acknowledge support from CONICET, Argentina.

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