Initial-state correlation in the electron-impact ionization of argon

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Ionization of the $3p$ orbital of argon by 113.5 and 200 eV incident electrons is studied by means of the post version of the CDW-EIS model. Results are presented for 2, 5, and 10 eV ejected electrons and compared to recent experimental data from Lohmann and collaborators. The present results are also compared to those obtained by other state-of-the-art theoretical methods. The present system is extremely sensitive to the way in which the post-collision interaction, the electronic exchange and the initial state correlation are considered. We conclude that none of the here discussed methods provides an ultimate description of the system and further theoretical work is needed.

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I. INTRODUCTION

Understanding atomic dynamics and interactions plays a key role in diverse areas such as astrophysics, radiation damage to solids and biological tissue, plasma processes, chemical reactions, etc. Thus, it is not surprising that since the pioneering work of Ehrhardt *et al.* [1] almost 40 years ago today [in which the first fully differential data for $(e,2e)$ reactions were presented], the ionization of atoms by light particle impact has been a topic subjected to wide interest from the atomic collisions physics community. As a result, light targets such as hydrogen and helium have been thoroughly explored experimentally as well as theoretically at different impact energies and geometric configurations [2–10].

As more data became available for the near-threshold region where perturbative methods are outside their validity range, nonperturbative theories were developed such as the convergent close coupling (CCC) [11] and, more recently, the exterior complex scaling [12] and the time-dependent close coupling [13] methods. These powerful numerical tools reached an unprecedented level of agreement with available experimental data. However, they have been mainly applied for the hydrogen target and only recently have been extended to other light targets and molecules [14,15].

On the other hand, in many fields such as astrophysics and irradiation of living matter there is a strong need of data for more complex reactants such as molecular and multielectronic atoms for which such a level of theoretical confidence has not been yet reached. Multielectronic targets such as the noble gases have been so far tackled by means of the distorted wave Born approximation (DWBA) and the distorted wave Born+R matrix (RM2) [16,17] models. Compared to the hydrogen target case, the multielectronic targets represent a completely different challenge since an additional problem is the choice of the effective potentials seen by the different particles in the one-active-electron representation and how the exchange terms are treated. An additional drawback related to these widely used approximations is that they neglect the post-collision interaction (PCI) between the receding projectile electron and the emitted electron. A standard procedure to include some of the PCI missing has been the implementation of a multiplying correlation factor to the DWBA differential cross sections. This factor has been modeled as the *e-e* Gamow factor, or the Ward and Macek factor [18].

During the last few years, Lohmann and co-workers have presented a set of measurements of fully differential cross sections (FDCSs) for electron impact single ionization of Ar at collision energies of 113.5 and 200 eV [19–24] which have turned out to be particularly challenging for theoreticians. They have focused on the coplanar configuration and have lately succeeded in measuring two coplanar FDCS over the whole angular range of the emitted electron for the ionization of the $3p$ shell of argon [24]. This set of data provides a unique benchmark to test state-of-the-art theoretical methods and to discuss the physical mechanisms involved in the collision process.

Initial theoretical studies on this system at these collision energies were performed by Prideaux *et al.* [17,25] and clearly revealed that none of the theories that proved successful at higher impact energies (DWBA, RM2) worked well at lower impact energies. Furthermore, the extension of the DWBA codes to the 3DWBA in order to fully account for PCI through the use of the 3DW approximation for the final state (the natural extension of the C3 wave function [8,26] to the case of arbitrary central potentials) did not lead to the expected accuracy. It was then argued that good agreement with experiment can probably be achieved if proper treatments of PCI and electronic exchange are combined.

In this paper, we focus on the role played by the initial state correlation for the ionization of the $3p$ shell of argon and present the first study of FDCS for single ionization of multielectronic targets in the low impact energy regime by means of the Continuum Distorted Wave (CDW) Eikonal Initial State (EIS) model in its post version.

In the next section we describe the theoretical method. In Sec. III, we use the hydrogen target as a test bench for our numerical implementation and we show our results for argon target at intermediate energies. We compare our results to those measured by Lohmann *et al.* and to those predicted by other state-of-the-art theoretical methods. Conclusions are drawn in Sec. IV. Atomic units are used throughout this work unless otherwise stated.

II. THEORETICAL MODEL

The CDW-EIS model was originally developed in the ion-atom context by Crothers and McCaan and has been extensively used during the last two decades to describe collisions involving different ions with either light targets such as hydrogen or helium [27–29] or multielectronic targets such as neon or argon [30–33]. For these targets (for which the contributions from the different shells had to be considered), the calculated total cross sections and doubly differential cross sections are in very good agreement with the available data.

In 1998, Jones and Madison successfully implemented the CDW-EIS model to study electron-hydrogen ionizing collisions at intermediate to low impact energies. In particular, they showed that the agreement with the experimental data was greatly improved at impact energies of 54.4 eV compared to a similar model which considered the C3 wave function for the final state but an uncorrelated Born initial state (C3Born). They later on published a detailed study of the electron-hydrogen system [34] and compared the results obtained by means of the CDW-EIS model to those of the C3Born model and the CCC method. Since the CDW-EIS model considers higher-order terms in the initial state correlation compared to the 3DWBA, its implementation for electron collisions on multielectronic targets is challenging since it provides a better asymptotic description of the initial channel.

The FDCS in the CDW-EIS model is given by

$$\frac{d^5\sigma}{dE_2 d\Omega_2 d\Omega_1} = N_{ee}(2\pi)^4 \frac{k_1 k_2}{k_0} \sum_{3p_0,3p_1,3p_{-1}} |T_{fi}|^2. \quad (1)$$

Here N_{ee} represents the number of identical electrons in the shell, $k_{1(2)}$ represents the momentum of the receding projectile and emitted electron respectively, and k_0 represents the impinging projectile momentum. The Gellman-Goldberger amplitude (T_{fi}) in Eq. (1) is represented by

$$T_{fi} = T_{fi}{}^{\mathrm{II}} + T_{fi}{}^{\mathrm{I}},$$

$$T_{fi}{}^{\mathrm{I}} = \langle \chi_{k_1}^- \chi_{k_2}^- D(r_{12}) | \boldsymbol{\nabla}_1 \cdot \boldsymbol{\nabla}_{12} - \boldsymbol{\nabla}_2 \cdot \boldsymbol{\nabla}_{12} | (\varepsilon_1 \varepsilon_{12} - 1) \Phi_i \varphi_i \rangle,$$

$$T_{fi}{}^{\mathrm{II}} = \langle \chi_{k_1}^- \chi_{k_2}^- D(r_{12}) | 1/r_{12} - V_{GS}(r_1) | \Phi_i \varphi_i \rangle. \quad (2)$$

The wave functions $\chi_{k_{1,2}}^-$ are distorted waves which are generated from central potentials for the ion core which mainly consist of the usual static screening potentials plus a local approximation for the electronic exchange. In this case, we have chosen the form provided by Gianturco and Schialla [35] which is particularly suited for particles in the low-energy regime. The infinite partial waves are considered in $\chi_{k_{1,2}}^-$, since the Kummer functions associated to the corresponding asymptotic charges, are corrected in the required number of partial waves until convergence to the Coulomb case is achieved. For Φ_{I} we have chosen a Clementi-Roetti wave function [36], and V_{GS} represents the core potential seen by the impinging electron. The distortion $D(\mathbf{r}_{12})$ is given by

$$D(\mathbf{r}_{12}) = N_3^- {}_1F_1[ia_3, 1, -ik_{12}r_{12} - i\mathbf{k}_{12} \cdot \mathbf{r}_{12}],$$

where the Coulomb normalization factor is given by $N_3^- = \exp(-\pi a_3/2)\Gamma(1-ia_3)$ and the Sommerfeld parameter $a_3 = 1/(2k_{12})$. In Eq. (2), φ_{I} represents the incident plane wave for the projectile while the eikonal distortions are $\varepsilon_j = \exp[ia_j \ln(k_j r_j - \mathbf{k}_j \cdot \mathbf{r}_j)]$.

The calculation of the transition amplitude has been developed by direct 6D numerical integration over the coordinates using the VEGAS adaptive Monte Carlo scheme. We estimate our numerical uncertainty to be less than 5%. In order to treat the continuum-continuum transition, we have used the wave-packet approach of Malcherek and Briggs [37]. Since we are concerned with very asymmetric collisions, we only include the direct amplitude neglecting the possibility of exchange between the impinging projectile and the active electron.

III. RESULTS AND DISCUSSION

In first place, we test our 6D numerical implementation on electron-hydrogen collisions at similar impact and emission energies than those afterwards considered for argon. In Fig. 1 we present FDCS obtained with the CDW-EIS model for impact energies of 54.4 and 150 eV at different emission geometries. The present results are in very good agreement with those published in Refs. [10,34], and as such, provide a good description of the data of Röder *et al.* [38,39].

We now turn to our results for argon and compare them to the relative experimental data by Lohmann and co-workers [21,22,24], which we normalize to our theory at the second structure of the binary peak. In addition, we also include the 3DWBA with electronic exchange included through the Furness and McCarthy potential (hereafter referred to as 3DWBA-FM model) and RM2 results from Ref. [17] that have also been renormalized in order to compare to our theoretical results. Due to the computational cost, we have not performed at this point any sort of convolution over the reported experimental uncertainties and the present FDCS have been obtained for discrete energy and angular configurations.

Based on physical grounds it reasonable to expect electronic exchange and PCI effects to be important when considering the emission of very low-energy electrons, so it was surprising to see that even the standard DWBA model without electronic exchange and PCI seemed to work well at the present impact energies for the binary peak structure [24], although a poor description of the recoil peak was observed. This clearly revealed how sensitive the FDCS profiles are to the way in which, at least, electronic exchange and PCI are balanced and indicates the difficulty in completely understanding this system.

In principle, the final state of the present CDW-EIS is similar to that employed by Pridaux *et al.* so the main discrepancies among these two theories can be confidently attached to the different representations of the initial state. On the other hand, 3DWBA-FM and RM2 have the same initial state correlation, so discrepancies among them should be mainly attached to the different modeling of the final state (exchange and PCI).

In Fig. 2 we show polar plots of the FDCS for a projectile impact energy of 113.5 eV and emitted electron energies of 2, 5, and 10 eV It is clearly seen from the $E_e = 2$ eV case, that the inclusion of higher order terms in the initial state translates into a more precise description of the shape of the FDCS compared to both the 3DWBA-FM and RM2 models.

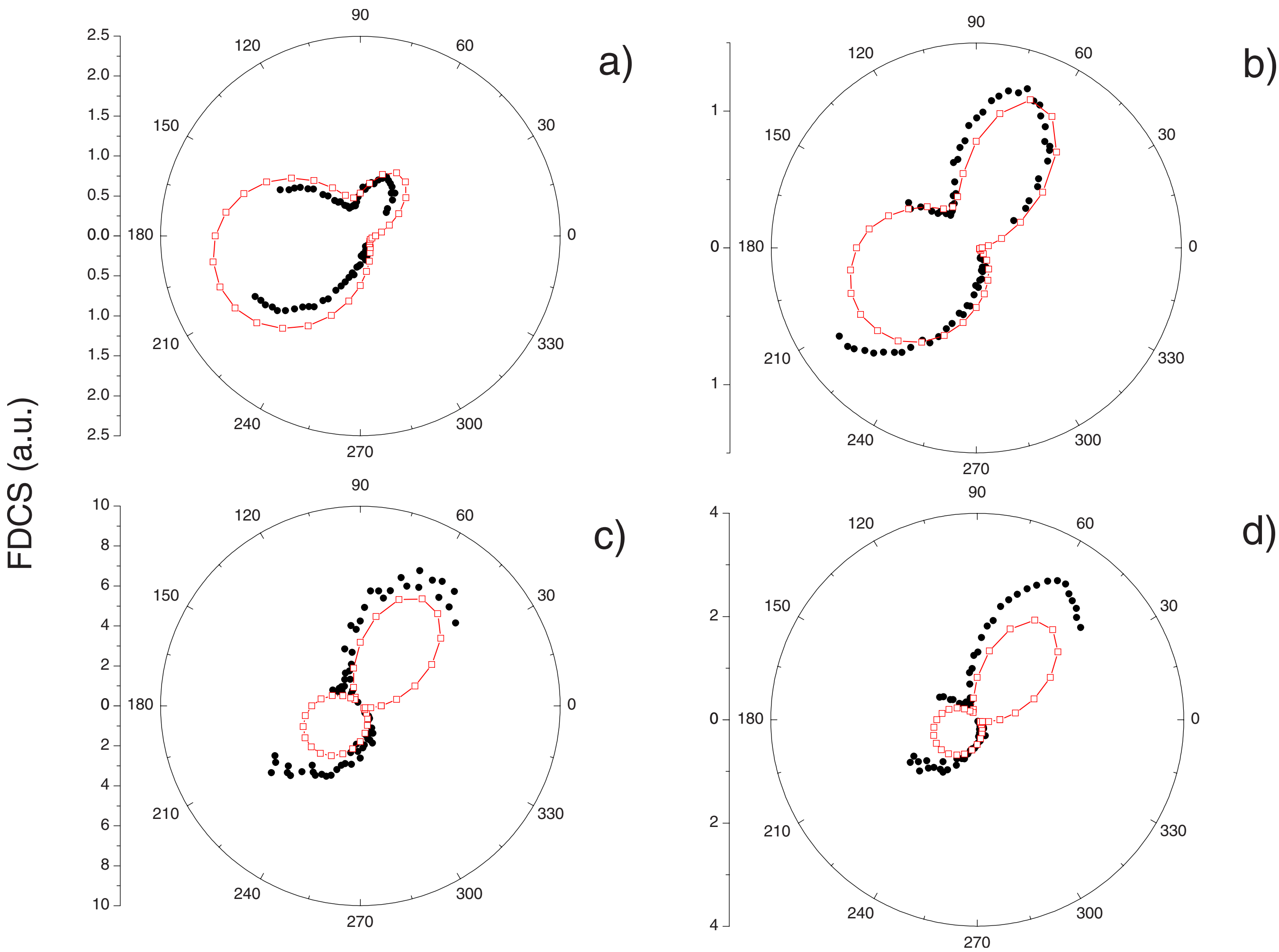

FIG. 1. (Color online) H(1*s*) ionization FDCS for 54.4 and 150 eV impact energy. The experimental data are those of Röder and co-workers [38,39]. The geometries considered are: (a) E_0=54.4 eV, E_2=5 eV, θ_1=4°, (b) E_0=54.4 eV, E_2=5 eV, θ_1=10°, (c) E_0=150 eV, E_2=5 eV, θ_1=4°, (d) E_0=150 eV, E_2=10 eV, θ_1=4°.

While 3DWBA-FM provides mainly a two-lobe structure, the CDW-EIS reproduces the four-lobe pattern exhibited by the data. Furthermore, RM2 clearly overestimates the forward emission possibly due to the lack of PCI. At E_e=5 eV, none of the theories provides a quantitative representation of the data. Both CDW-EIS and 3DWBA-FM predict a negligible emission in the direction of the scattered projectile probably due to the PCI interaction. RM2, on the other hand, again provides evidence of forward emission, because of the lack of PCI. Both models considering PCI provide a better representation of the backward emission. The CDW-EIS model predicts a lobe at approximately 200° which is also suggested by the data and more slightly by the RM2 model.

At E_e=10 eV the profile of the binary peak is accurately reproduced by the CDW-EIS and the 3DWBA-FM theories. The recoil peak at 255° is better reproduced in this case by the RM2 method what suggests that a more accurate incorporation of the electronic exchange between the emitted electron and the core should be important to properly reproduce the profile of that structure.

In order to understand the differences among the different theories and the data, we now analyze the momentum transfers involved. For the 2 eV case (k_e=0.38 a.u.) the momentum transferred by the projectile is Q=0.76 a.u. Hence, the electron emission is a fraction of the total momentum transferred to the target. In this case, we see that the inclusion of PCI as well as higher order terms in the initial state correlation compared to 3DWBA-FM produces better accord with the data. As the emission energy increases, 5 eV case (k_e=0.61 a.u.), the emitted electron momentum is closer to the total momentum transfer favoring the exchange of momentum between the electron with the target core. At 10 eV (k_e=0.86 a.u.) the electron is emitted with a momentum greater than that transferred by the projectile. Such can only be achieved at the expense of the recoiling core. Thus, this suggests that the very strong interaction between the incident electron and the core could be the reason why the RM2 provides a better agreement with the data in the recoil peak region compared with CDW-EIS and 3DWBA-FM which incorporate local approximations for the exchange terms. Moreover, since the data set is not as complete as for the 2 and 5 eV cases, it is not possible to test whether the lack of PCI by the RM2 model could lead to an overestimation of the nearly forward emission or not. Thus, we suggest that

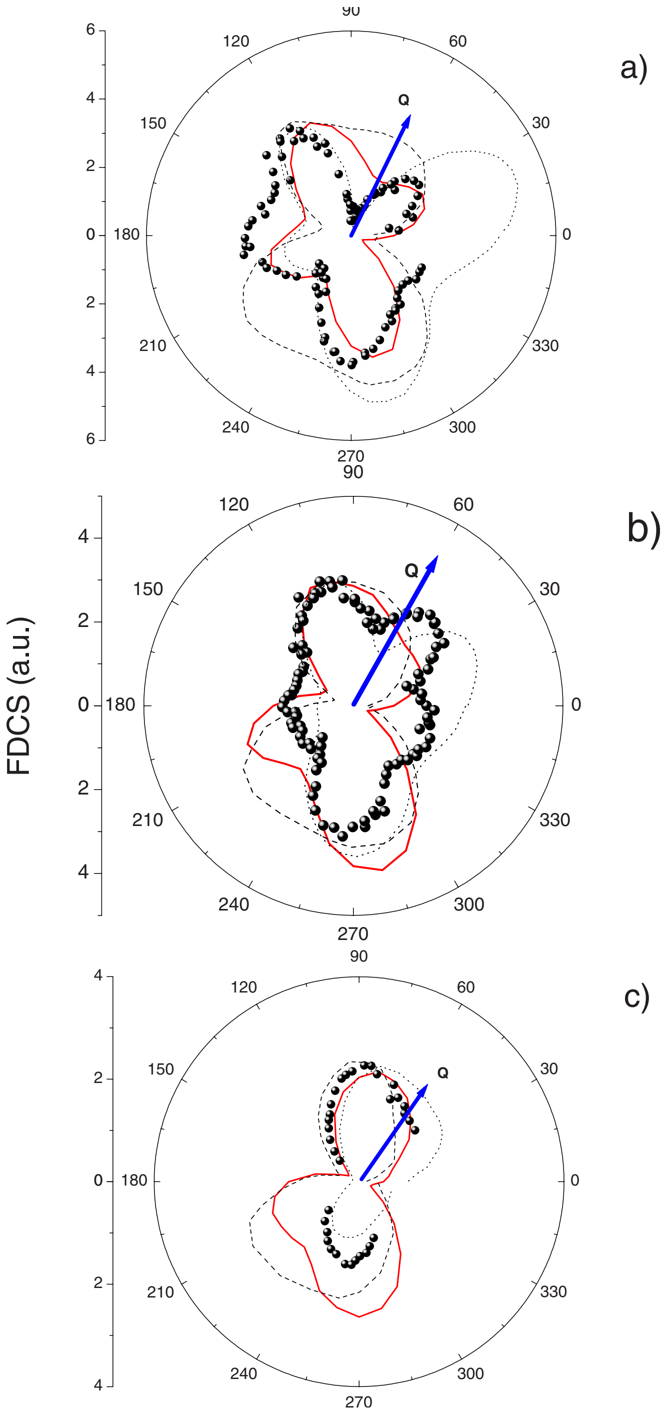

FIG. 2. (Color online) Ar(3p) single ionization FDCS for 113.5 eV incident electrons and (a) 2 eV, (b) 5 eV, and (c) 10 eV emitted electrons. Theories: CDW-EIS (solid line), 3DWBA-FM (dashed line), RM2 (dotted line). The experimental data is that of Lohmann and co-workers [21,24]. The blue arrow indicates the momentum transfer direction.

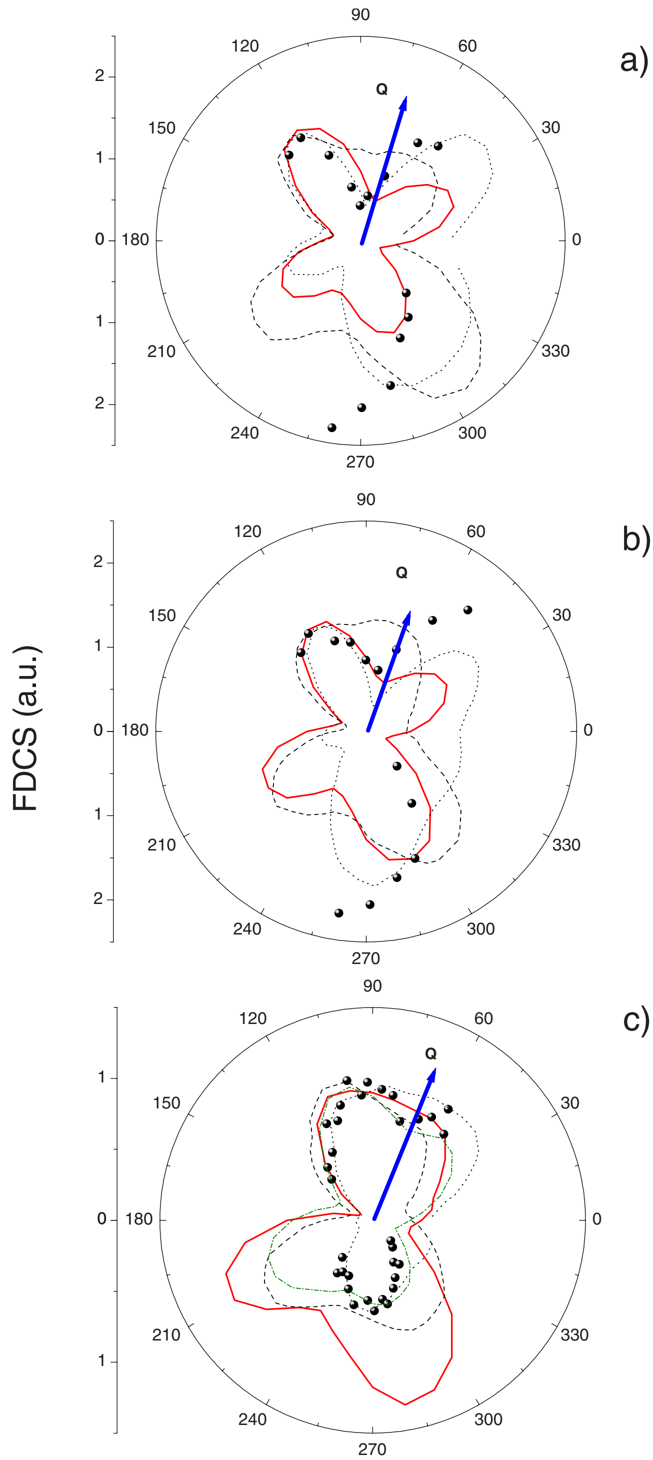

FIG. 3. (Color online) Ar(3p) single ionization FDCS for 200 eV incident electrons and (a) 2 eV, (b) 5 eV, and (c) 10 eV emitted electrons. Theories: CDW-EIS (solid line), 3DWBA-FM (dashed line), RM2 (dotted line), CDW-EIS (dot-dashed line) (static potential only). Experimental data: Stevenson and Lohmann [22].

future experiments check if a third lobe appears around 195° as suggested by the present CDW-EIS results.

We now consider the same emission geometries but for an impact energy of 200 eV (Fig. 3). In this case, the experimental data is that of Stevenson and Lohmann [22] and covers a partial fraction of the whole angular range.

For the 2 eV ionized electron case, we find that none of the theories is capable of providing an accurate description of the partial data set of Haynes and Lohmann. The CDW-EIS results again show a clear four-lobe structure and underestimate the recoil peak structure. It is interesting to notice that compared to the 113.5 eV impact energy case, the binary to recoil peak ratio predicted by the 3DW-EIS increases.

This ratio enhancement is in agreement with the observations of Ehrhardt and Röder for the hydrogen target [39] but it contradicts the trend suggested by the argon data which is just the opposite. More data covering the whole angular range would be very useful in order to shed light on this trend and would help elucidate whether or not this behavior could be a signature of the multielectronic character of the target. For 5 eV the recoil structure gains importance in the CDW-EIS results and similarly we observe an overestimation of this structure for 10 eV emitted electrons. Again, we attach this trend to the strong interaction between the recoiling core and the emitted electron which is not well described by the present local approximation for the electronic exchange. To reinforce this hypothesis, we also present the CDW-EIS results obtained by neglecting electronic exchange in the emitted electron-remaining core interaction and we have normalized them to the CDW-EIS results for the full potential. The fact that the static screening potential V_D is less attractive than the full potential V_D+V_{GS}, brings the recoil to binary ratio into much better accordance with that of the experimental data. Furthermore, we observe that compared to the 3DWBA results shown in Ref. [17] (where the electronic exchange was also neglected in the electron-core interaction) the present CDW-EIS improves the description at the recoil peak region, qualitatively reproducing the partial experimental data. Such a test highlights how sensitive the profiles are to the chosen representation for the electron-core interaction. Thus, for the emission energy of 10 eV we conclude that the RM2 method provides the closest agreement to the data although the forward scattering is overestimated due to the lack of PCI.

IV. SUMMARY AND CONCLUSIONS

In summary, we have performed a study of FDCS for single ionization of multielectronic targets by electron impact in the low impact energy regime by means of the CDW-EIS model. In contrast to the advantages observed for the hydrogen target, the extension of this model to a multielectronic target like argon did not lead to the same degree of success. Whether that is consequence of an inherent limitation of the model itself or the way in which multielectronic targets are usually modeled in the one-active electron scheme remains to be solved.

The present study suggests that none of the methods discussed provides an ultimate description of the present system. By analyzing the momentum transfers and emitted electron momenta, we proposed possible explanation of why the different theories work well under different energetic configurations. This explanation was based on the quality of PCI, electronic exchange, and initial state correlation of each representation. For those collisions in which the emitted electron has a momentum equal or higher than the momentum transferred by the projectile, the RM2 approach leads to a better description of the structures which result from a strong interaction between the emitted electron and the core (i.e., recoil peaks) due to its better representation of the electronic exchange. Possible remaining discrepancies could be attached to the lack of PCI and higher order terms in the initial state correlation.

In its present form, the CDW-EIS model still lacks an accurate treatment of electronic exchange affecting the agreement with the data when the emitted electron momentum is equal or greater than the total momentum transferred by the projectile. However, we observe that the initial state correlation plays a definite role at impact energies around 100 eV and the inclusion of higher order terms in the projectile-target interactions is desirable in any future theoretical model.

More sets of complete FDCS such as those recently published by Stevenson and Lohmann would be very valuable to help theoreticians improve the theoretical description of collision systems of this type. Furthermore, future experiments designed to obtain absolute data would also provide a more exhaustive test to the state-of-the-art theories.

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