Electron impact double ionization of rare gases: A kinematical analysis of the non-first order effects at intermediate impact energy

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Abstract. The (e,3-1e) four-fold differential cross sections (4DCS) are measured for the electron impact double ionization of rare gas atoms (He, Ne, Ar) in coplanar asymmetric geometry for a wide range of ejected electron energies (5 to 144 eV) and at an incident energy of about 600 - 700 eV. Though we only show here a sample of the results, all experimental angular distributions of the 4DCS are characterized by large angular shifts of the forward and backward lobes with respect to the momentum transfer direction or its opposite, respectively. This validates our previously published results [Lahmam-Bennani et al 2002 J. Phys. B: At. Mol. Opt. Phys. 35 L59] which were questioned by Götz et al [2003 J. Phys. B: At. Mol. Opt. Phys. 36 L77]. A qualitative, kinematical analysis is given which allows relating these shifts and the observed structures (or sub-lobes) in the cross section distributions to the second order, ‘two-step 2’ double ionization mechanism, which is shown to predominate over the first-order ‘shake-off’ and ‘two-step 1’ mechanisms under the present kinematics.

1. Introduction

In an electron impact double ionization (DI) experiment, an incident electron (indexed 0) scatters off an atomic or molecular target provoking the emission of two electrons from the target. Though indistinguishable, these electrons are indexed for convenience “a” for the fast-scattered one and “b” and “c” for the slower-ejected ones. A kinematically complete experiment is realized when these electrons are energy and angle selected and are detected in triple coincidence to insure that they are issued from the same ionizing event. However, these so-called (e,3e) experiments [1,2] are very difficult to perform, because the corresponding cross sections are inherently very low due to their multiply (five-fold) differential character. Therefore, it is also of interest to consider (e,3-1e) experiments [3,4] where, with respect to the (e,3e) case, one electron (say “c”) is not detected, hence only necessitating a double coincidence experiment similar to an (e,2e) single ionization (SI) but with energetics corresponding to DI. Such experiments were clearly demonstrated [4] to provide a very sensitive mean to identify the various mechanisms responsible for electron impact DI and to gauge their relative importance.

A few years ago, there was an article published by our group reporting (e,3-1e) experiments for DI of He at about 600 eV impact energy [5]. In this paper, we claimed that evidence is given of the great importance of second- or higher-order effects in the projectile-target interaction. These effects mostly

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showed up in the measured angular distributions as large shifts of the forward and backward lobes with respect to the momentum transfer direction +K, or its opposite, -K. It is well known [6] that these shifts cannot be reproduced by first-order models, for example the ‘first Born – 3 Coulomb waves’ (FB-3C) model described in [5] which is peaking, as expected, in the ± K directions.

A year later, in 2003, Götz et al [7] published a tentative theoretical description of our results, where the four-particle continuum was described in different approximations leading up to a 6C wave function which takes all two-body interactions into account as well as the interactions between each outgoing electron and the nucleus, and hence goes beyond the second Born approximation. However, the authors could not reproduce the strong asymmetries seen in the measured data, so that they came to implicitly question the experiments by writing: “we suggest that it is very important to establish the validity or otherwise of the asymmetry in the experimental data.”

Given this questioning, we decided to re-examine the problem by taking advantage of the extended capabilities and sensitivity of our spectrometer gained by including to it new developments [8]. Our aim is to produce more extensive experimental data which would validate or invalidate the observations we made in our earlier work, and against which the theoretical predictions can be tested. The experimental set-up used for these investigations has been fully described elsewhere [8]. It essentially features multi-angle analysis and double/triple coincidence detection of the two/three final state electrons, based on three toroidal analysers coupled with three position sensitive detectors (PSD). The key point is that the angular distribution of the scattered or ejected electrons in the collision plane is preserved upon arrival on the PSD’s.

2. Results and discussion

Since we are aiming to discuss the results in terms of DI mechanisms, it is convenient to start this section with a brief description of the different DI processes usually invoked in the literature. Direct electron impact DI may proceed mostly via three routes [9]. (i) In the shake-off mechanism (SO), the incident electron interacts only once with a single target electron which is ejected without further interaction with other target electrons. Subsequent ejection of a second electron occurs, due to the sudden change in the distribution of potential seen by this electron. (ii) In the two-step mechanism termed TS1, the projectile electron interacts with one target electron which subsequently collides with a second electron leading to ejection of the pair. (iii) In the two-step mechanism termed TS2, the projectile electron successively collides with two target electrons, resulting in their double ejection. Both SO and TS1 involve only one single interaction of the projectile with the target, hence their denomination as first order or first Born mechanisms (in the Born series). In contrast, TS2 involves two successive projectile - target interactions and hence is a second order or second Born mechanism. The most obvious signature of the presence of these mechanisms is that first order mechanisms yield ejected electron(s) angular distributions which are symmetrical with respect to the momentum transfer vector direction; whereas this symmetry is broken in the case of second order, TS2 mechanism, see [6]. Another high-order mechanism was recently invoked by M Schulz et al [10] in the context of p + He collisions. The authors labelled it Two-Step 1–Elastic mechanism (TS1-El). It can be viewed as follows: first, a TS1 process leads to the ejection of both target electrons, and then the projectile is elastically scattered from one of the two continuum electrons (hence the alternative label ‘3steps – 2 interactions’ 3S2). As in TS2, the symmetry about K direction is broken because of this double interaction of the projectile with the target. In the following, we will concentrate only on the contribution of TS2.

Let us now discuss the results of (e,3-1e) experiments. Several targets were investigated (He, Ne, Ar, H2, N2), under various kinematics. We used a fixed scattered electron energy and angle, 500 eV and -6 degrees respectively. (Throughout this work, positive angles are counted clockwise starting from the incident direction). In contrast, the energies of the ejected ‘b’-electron (Eb) which is detected and that of the ‘c’-electron (Ec) which is undetected were varied over a large range, from 5 to 144 eV. Only a selection of these results will be shown here (the full results will be published elsewhere), as the observations and conclusions are more or less the same for all cases. But before discussing the DI
data, it is important to mention that the experimental procedure and the good response of the spectrometer for measuring coincidence angular distributions were validated as follows. Very briefly, the \((e,2e)\) triple differential cross section (TDCS) for SI of the He 1s orbital is measured at the energies \(E_a\) and \(E_b\) of the current DI experiments. The so-obtained experimental TDCS-distributions are compared with the calculated convergent close coupling (CCC) results. The CCC model is commonly known to produce very accurate TDCS results for electron impact SI of He when the residual ion is left in its ground state. Consequently, the very good agreement obtained between experiments and CCC calculations clearly validates our measurement procedure.

With this, we can now look with confidence to our DI results. Some of the He data are displayed in figure 1 for two energy sharings \((E_b:E_c) = (37:5)\) and \((74:5)\) eV. They are compared with the predictions of the FB-3C model where the final state is described by the product of three Coulomb waves. All the experimental as well as theoretical distributions of the 4DCS display the well-known two-lobes structure: a forward lobe pointing roughly in the momentum transfer direction, \(+K\), and a backward lobe pointing in the opposite direction, \(-K\), (these two directions are indicated by the vertical dashed lines). Also, the calculated backward-to-forward maximum intensity ratio is more or less in agreement with the measured one at \((37:5)\) eV, but not so at \((74:5)\) eV where the theory predicts quite a small backward lobe. However, the most striking observations from figure 1 are the breaking of symmetry about \(\pm K\) directions, the large angular shifts (~30° to 60°) of the measured lobes from these directions, and the existence of structures or sub-lobes in these lobes. For all energy sharings considered in this work, the symmetry breaking and the large angular shifts are very much alike those we reported earlier in Ref. [5] which were obtained at \((E_b : E_c) = (51:10)\) eV using an older version of our spectrometer. This close resemblance between the ‘old’ results and the extended new data definitely confirms the validity of our previous results, and should put a final end to the question left open by Götz et al [7] about this validity.

In terms of DI mechanisms, the features observed in figure 1 (i.e. breaking of symmetry, large angular shifts and existence of structures in the measured lobes) cannot be present in any first-order mechanism, namely the SO and TS1 which involve only one single interaction of the projectile with the target. Hence, they are clear signatures of non-first order processes such as TS2 or TS1-El which involve two or more successive target - projectile interactions. In the absence of detailed second- or higher-order calculations which could be compared with our data (such calculations are very difficult to perform), it was proposed in [11] a qualitative interpretation of the origin of the observed structures,
as well as an estimate of the angular positions of the sub-lobes, based on kinematical arguments for TS2 built on our knowledge of SI (e,2e) processes. For the sake of brevity, the discussion is briefly summarized in the following and is illustrated using the data of Fig. 1(a) at \( (E_b : E_c) = (37 : 5) \) eV and \( E_0 = 621 \) eV. However, the ideas and the resulting conclusions hold for all the data and targets considered in this work.

TS2 is a two-step process: in the first step, the slowest c-electron (with energy 5 eV in figure 1) is ejected in an (e,2e)-like process where the corresponding scattered electron (call it ‘a*’) has an energy \( E_{a*} \) imposed by the energy balance, \( E_{a*} = E_0 - E_c \). IP = 591 eV (IP is the He SI potential) and appears with the highest probability at the Compton scattering angle, \( \pm \theta_{a*} \), corresponding to the Bethe ridge condition \[ E_0 - E_{a*} = E_0 \sin^2(\theta_{a*}) \]. This corresponds to \( \theta_{a*} \approx \pm 12^\circ \). The \( \pm \) sign stands for the fact that the intermediate, scattered a*-electron may appear on both sides of the incident beam direction. The associated c-electron in this (e,2e) process is most likely ejected in the corresponding momentum transfer direction, that is \( \theta_{c*} \approx \pm 62^\circ \) with respect to the incident direction. Note that neither the a*-nor the c-electrons are detected in the (e,3-1e) experiments.

In the second step of TS2, the a*-electron plays the role of the incident projectile in a new (e,2e) ionization of the target, resulting in the pair of electrons effectively detected, that is the a-scattered one with \( (E_a = 500 \text{ eV}, \theta_a = -6^\circ) \) and the fast b-ejected one with \( E_b = 37 \) eV. Here, two scenarios may occur, depending whether the intermediate a*-electron is scattered under \( \theta_{a*} \approx \pm 12^\circ \) or under \( +12^\circ \). We show in the following that these 2 scenarios might be responsible for the forward and backward sub-lobes observed in the experimental data.

Scenario 1 corresponds to \( \theta_{a*} \approx -12^\circ \). In the corresponding (e,2e) event, the a-electron is detected at \( \theta_a = -6^\circ \), and the b-electron is essentially ejected in the corresponding \( \theta_b \)-direction, that is \( \theta_b \approx -48^\circ \) from the a*-incident direction (at the high energies involved here, the recoil intensity appearing about \(-K\) direction is quite small). In other words, the b-electron is mostly ejected at \( \theta_b \approx -60^\circ \), or alternatively at \( \theta_b \approx (360^\circ - 60^\circ) = 300^\circ \) from the primary incident beam direction. Scenario 2 corresponds to \( \theta_{a*} \approx +12^\circ \). In a similar way, we find that the b-electron is mostly ejected at \( \theta_b \approx +78^\circ \) from the primary incident beam direction.

In the experimental data (figure 1(a)), the backward and the forward intensity distributions are peaking at about \( 305^\circ \) and \( 85^\circ \). On the one hand, these values are quite far from the \( \pm K \) directions, \( (41^\circ \text{ and } 221^\circ, \text{ respectively}) \) where the first-order SO and TS1 contributions should be at their maximum. But on the other hand, these values are in excellent agreement with the values \( 300^\circ \) and \( 78^\circ \) derived from scenarios 1 and 2 above. We thus conclude that the positions of the backward and the forward lobes in the measured data are essentially determined by the TS2 contribution, according to these two scenarios. In contrast, SO and TS1 contributions are of course also present, but their intensity appears to be appreciably smaller than that of the TS2.

This discussion and the resulting conclusions may be likewise repeated for the other data we obtained either for He at different energies \( (E_b : E_c) \), or for other targets. Two more examples are given respectively in figure 1(b) for DI of He and in figure 2 for Ne: the splitting in two sub-lobes of both the forward and backward intensity distributions is also clearly seen, and the position of the large angle sub-lobes nicely agrees with the prediction of our qualitative two-step-2 interpretation indicated by the vertical dotted lines.

In summary, all our data show that for all targets and energies investigated, and under the present kinematics, the “non-first order mechanisms such as TS2” dominates over the first-order ones, SO and TS1, and are mostly responsible for the structures and angular positions of the measured lobes. The argument is very nicely supported by very recent second-Born calculations by Dal Cappello et al [13] displayed in figure 3 for the He case at \( (37:5) \) eV. As noticed above, the first Born-3C prediction does not reproduce the observed shifts and structures of the cross section distribution, whereas the 2nd Born treatment (dashed curve) very clearly constitutes a considerable improvement, both for the position of the forward lobe which is correctly reproduced and for the structure of the backward lobe which is qualitatively well predicted.
Similarly, TS2 results by M Schulz and co-workers (not shown here, and to be published elsewhere) are also in quite good agreement with experiments.

3. Conclusion
In conclusion, we have presented in this contribution a sample of our new (e,3-1e) experiments for DI of a series of atoms and molecules, at an incident energy of about 600-700 eV and different energy sharings (E_b:E_c) among the two ejected electrons. All our data display large shifts of the forward and backward lobes in the ejected electron angular distributions, thus confirming and validating our earlier observations [5].

We have discussed a qualitative analysis of the data based on kinematical arguments which shows that under the present kinematics the non-first order mechanisms such as TS2 dominate over the first order ones (SO and TS1), and are mostly responsible for the structures or sub-lobes observed in the measured distributions. Other mechanisms such as TS1-El may also contribute, but the present kinematical analysis cannot be straightforwardly applied to these mechanisms.

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