Dynamics of single or double ionization of small systems from coincidence electron impact experiments

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Abstract. In this brief review, we illustrate the potentialities and the power of electron-electron coincidence studies to investigate the dynamics of single ionization SI [(e,2e) case] or double ionization DI [(e,3e) case] processes. An example for (e,2e) SI of rare gas atoms is presented with a new insight into the behaviour of the recoil versus the binary intensity. A second example for (e,2e) SI of molecules is used to illustrate the observation of a purely molecular effect, namely the signature of interference effects due to the two-center nature of the H₂ molecule. The third example discusses an unprecedented triple coincidence study involving the scattered – ejected – Auger electrons emitted from an argon target. It is shown that the method allows to disentangle the contribution of various DI mechanisms.

1. Introduction
In an electron impact single ionization (SI) or double ionization (DI) experiment, an incident electron (indexed 0) scatters off an atomic or molecular target provoking the emission of one or 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 coincidence to insure that they are issued from the same ionizing event. These are the so-called (e,2e) [1] or (e,3e) [2] experiments, relative to the SI or DI cases, respectively. An impressive amount of work has been published to-date concerning the (e,2e) experiments, see for instance the reviews [3-5] and to a lesser extent concerning the (e,3e) experiments, see for instance the reviews [3,6]. In these studies one might grossly distinguish three kinematical regimes: (i) The dipolar regime, reached at high impact energy (several keV) and very small momentum transfer to the target (few tenths of an au); here the incident projectile acts as a pseudo photon. (ii) The Bethe ridge regime (also called electron momentum spectroscopy (EMS) regime), reached when the momentum transferred to the target is fully absorbed by the ejected electron, the nucleus acting as a spectator. Here the experiments are used to gather information about the target structure. And (iii) the more general case where neither of the above two conditions is fulfilled. The experiments are then used to investigate the dynamics of the SI or the DI process. In this paper, we will briefly discuss a few examples which all belong to this third category, i.e. the kinematics is far away from both the dipolar limit and from the Bethe ridge. These examples are meant to illustrate the very wide possibilities offered by both (e,2e) and (e,3e) techniques to investigate in the finest details some dynamical aspects of electron impact ionization studies.

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The experimental set up used for these investigations has been fully described elsewhere [7]. 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. (e,2e) results on simple atoms

We first discuss results for SI of the rare gas atoms He, Ne, and Ar. These results have been described in all details in [8]. Here, we will only concentrate on one particular aspect, which is the relative evolution of the recoil scattering vs the binary scattering.

Figure 1 shows (e,2e) triple differential cross sections (TDCS) for the three targets, obtained for a fixed scattered electron observed with an energy $E_a$ of 500 eV at a scattering angle of -6° with respect to the incident direction, and a sequence of ejected electron energies ranging from 12 to above 200 eV. The vertical thin line indicates the momentum transfer direction and it’s opposite. Many various theoretical models were applied (with some success) to describe these experimental data. We only mention the two of them which are needed for the present discussion.

First, the Convergent Close Coupling (CCC) model [9] which is known to produce very accurate TDCS results, at least for electron impact SI of He when the residual ion is left in its ground state. Indeed, we find very good agreement between CCC results and our data. Second, the Distorted Wave Born Approximation [8] corrected for the post collision interaction (PCI) by a simple Gamow factor (DWBA-G). At all ejection energies, the CCC (where they exist) as well as the DW calculations are in fairly good agreement with experimental data, except at the largest $E_b$ value of 205 eV where the ejected and the scattered electrons acquire comparable energies and their PCI becomes strong. Hence, it may not be fully accounted for by the simplified Gamow factor. Also, we observe that the general agreement with experiments deteriorates at all energies from the lighter to the heavier target. For
instance, at 37 eV, the theory predicts a prominent central peak positioned in between the binary and recoil peaks, which is not confirmed experimentally. But, besides from these general remarks, what we would like to emphasize from figure 1 is the steady decrease in the case of He of the recoil peak maximum intensity, relative to the magnitude of the binary peak, as the energy of the ejected electron increases. Indeed, the origin of the recoil peak is qualitatively understood as being due to reflection of the ejected electron wave from the atomic potential well. The relative recoil-to-binary intensity ratio, RB, is proportional to the reflection coefficient, which is decreasing as the energy of the ejected electron is getting larger. However, this is not the case for all three targets. For instance, the RB intensity ratio is, in fact, increasing with the ejected electron energy in the case of Neon 2s. Whereas in Argon 3s, we observe that, as the ejected energy increases, the experimental RB ratio is first decreasing, then it becomes minimal at 74 eV and increases again towards larger ejected energies. This tendency is confirmed by the DWBA-G model, except for the lowest energy, 17 eV, where the theory predicts a relatively large recoil lobe in a blunt contradiction to the experiment.

These findings are summarized in Figure 2, where the RB ratios are plotted vs the ejected electron energy for all three targets. The experimental RB ratio for He is decreasing with increasing ejected energy. Conversely, this experimental ratio in Ne is steadily growing across the whole energy range studied here. Whereas the corresponding ratio in Ar is non-monotonic: there is a hint of a minimum in between 37 and 74 eV, and a considerable growth towards larger energies, while the ratio seems to be flattening at the smallest energies of 17 and 12 eV.

![Figure 2. Recoil-to-binary intensity ratio plotted as a function of ejected electron energy for He (left panel), Ne (middle panel) and Ar (right panel).](image)

A theoretical analysis of the different behaviors of the RB ratio was given by Kheifets *et al* [8]. Very briefly, the argument is based on the idea that the RB ratio is determined by the slow electron movement in the ionic potential, which is a superposition of the Coulomb potential of the nucleus and the Hartree-Fock HF potential of the ionized electron core. The smooth, slowly varying Coulomb potential alone is a poor reflector, and it is the short-range HF potential that is chiefly responsible for the formation of the recoil peak. The qualitative change in the RB ratio dependence on the ejected electron-energy from He to Ar can be explained by variation of reflectivity of the short-range HF potential. The reflectivity increases profoundly from He to Ne and to Ar due to specific scattering phase behavior.

### 3. (e,2e) results on simple molecules

We now turn to (e,2e) results obtained on molecules, and we choose to illustrate this point by the observation of a specific molecular effect, that is the signature of interference effects in the TDCS for the H\textsubscript{2} molecule [10], similar to the Cohen-Fano oscillations predicted for DDCS’s [11]. The same idea of a molecular double slit behavior as viewed on electron impact TDCS’s was theoretically suggested by Stia *et al* [12]. Very briefly, the idea is the following: we measured (e,2e) angular
distributions for H₂ at a number of ejected electron energies, similar to those displayed in figure 1. These H₂ distributions are then compared to those for He. Figure 3 is an example of such comparison.

**Figure 3.** TDCS measured for He (full circles) and H₂ (open circles). $E_a = 500$ eV, $\theta_a = -6^\circ$ and $E_b = 205$ eV (upper panel) and 37 eV (lower panel). The results from the two targets are normalized to each other at the maximum of the binary lobe.

We observe that in one case, at the ejection energy of 37 eV, the recoil intensity in H₂ is diminished with respect to He, whereas it is enhanced at the energy of 205 eV. Such recoil intensity destruction or enhancement in the molecular case was theoretically shown by Stia *et al* [12] to result from Young type interference in which the two sources of coherent emission are the two atomic centers in the molecular target. In fact, Stia *et al* introduced the well known interference factor, I, which reads

$$I = 1 + \left[ \frac{\sin(q\rho)}{q\rho} \right]$$

where $\rho$ is the equilibrium internuclear distance in the molecule and $q$ is the momentum of the recoiling ion. Of course, $q$ depends on the ejected electron energy and hence on its wavelength. Figure 4 shows, for three ejected electron energies, the interference factor, I, derived from our experiments compared to the one predicted by the above formula.

**Figure 4.** Theoretical interference factor, I, as compared to the experimental ratio of TDCS measured for He and H₂. $E_a = 500$ eV, $\theta_a = -6^\circ$ and $E_b = 37$, 74 and 205 eV from top to bottom, respectively.

Qualitative good agreement is seen in the three cases between theory and experiments, in spite of the large error bars due to the fact that, in certain angular ranges, we are taking the ratio of two small quantities. These data clearly show a suppression of the recoil peak intensity with respect to the binary one at 37 and 74 eV, whereas the data at $E_b = 205$ eV display its prominent enhancement. This reasonably good agreement of the experimental results with the predictions of Stia *et al* suggests that the present observations can be ascribed, in the 37 and 74 eV cases, to destructive interference effects arising from the two-centre nature of H₂, or to constructive interferences in the 205 eV case. We note
that, to our best knowledge, this is the first time that both the destructive and constructive characters of the interference process are simultaneously observed in the same (e,2e) experiments.

4. (e,3e) results
We now come to discuss some DI results and we choose to illustrate these by (e,3e) experiments on Ar with Auger emission [13]. We investigate here electron impact indirect DI events involving first the removal of one fast electron ejected from an inner shell, namely here the Ar-2p shell, followed by an electronic rearrangement which leads to the ejection of one fast Auger electron with an energy 205 eV in this Ar case. We deliberately choose the energies of both continuum electrons to be equal, hence the ejected and the Auger electrons are fully indistinguishable (we do not know which is which!!). This indirect process is in competition with the direct DI process involving the removal of 2 electrons from the outer 3p-shell, with identical energies, 205 eV each. Both direct and indirect DI processes yield the same final state, hence they might interfere.

The kinematics of these (e,3e) experiments is as follows. A coplanar geometry is used. The incident electron with energy $E_0 = 953$ eV is observed after scattering with an energy $E_a = 500$ eV and under a scattering angle of $6^\circ$. The two indistinguishable electrons labeled here b and c for convenience are observed at the same energy in the angular ranges $20^\circ – 160^\circ$ and $200^\circ – 340^\circ$, and all three electrons are detected in triple coincidence.

The data are displayed in figure 5 as a 2-D representation of the measured (e,3e) intensity vs. $\theta_b$ and $\theta_c$ in these angular ranges.

![Figure 5.](image)

The picture is very rich but also very difficult to interpret. Very briefly, we can distinguish several islands (or peaks) of maximum intensity, some of them are labeled B, F, L and R, for which a tentative interpretation is given, associated with the emission diagrams shown at the bottom of the figure, and some others are not labeled.

In peak B, both atomic electrons are emitted backwards (hence B) with respect to the incident direction, at respective angles of $120^\circ$ and $240^\circ$ (or $-120^\circ$). We remind that the fast scattered electron is observed at $6^\circ$, i.e. very close to $0^\circ$. Hence, the 3 electrons emerge at about $120^\circ$ from each other, which is the configuration which minimizes the Coulomb repulsion between these final state electrons. In peak F, the two electrons are emitted in the forward direction, at angles of $+60^\circ$ and $-60^\circ$. This observation, coupled with other observations from complimentary (e,3-1e) experiments which are not
discussed in the present paper, see [13] for more details, leads us to attributing this peak F to a shake-off (SO) - 2step - indirect DI where the first step is an (e,2e) SI of the 2p-shell followed in a second step by a quasi-isotropic Auger emission.

In peaks L and R, (standing for Left and Right with respect to the incident beam) one electron is emitted close to +90° in L and close to -90° (or +270°) in R, while the 2nd electron appears at -50° or +50°. These 90° angles are reminiscent of a classical binary collision between two particles of equal mass, hence the interpretation of these peaks as being due to a direct DI in the so-called “Two Step 2” (TS2) process, that is, a binary collision first occurs between the incident electron and one target electron, followed by a 2nd step where the projectile hits a 2nd target electron in an (e,2e) collision. The other peaks, unlabelled, may speculatively be attributed to the interference between the competing direct and indirect DI processes, leading to the same final state.

5. Summary and conclusion
We have discussed in this paper a number of new (e,2e) experiments for ionization of a series of atoms and molecules. For the rare gases, the particular kinematics (characterized by large q-momentum imparted to the recoiling ion) enhances the recoil scattering intensity. Investigating this recoil intensity shows that for the sequence He, Ne, Ar, a different behavior of the RB ratio is observed and it is attributed to the difference in reflectivity of the HF ionic potential. For molecules, we have seen the signature of Young type (destructive or constructive) interference effects, due to the two-centre nature of H₂. We also briefly described (e,3e) experiments involving for the first time coincidence detection of scattered – ejected – Auger electrons, and we have shown that it is possible to disentangle from the data the contribution of different DI mechanisms.

As a final remark we want to add that we badly need further development of theoretical models to help understanding all these very complex SI as well as DI processes.

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References
[1] Ehrhardt H, Schulz M, Tekaat T and Willmann K 1969 Phys. Rev. Lett. 22 89
[2] Lahmam-Bennani A, Dupré C and Duguet 1989 Phys. Rev. Lett. 63 1582
[3] Lahmam-Bennani A J. Phys. B: At. Mol. Opt. Phys. 24 2401
[4] Weigold E and McCarthy I E 1999 Electron Momentum Spectroscopy (Kluwer Academic, Dordrecht/Plenum Publishers, New York)
[5] Lahmam-Bennani A 2002 J. Electron Spectrosc. Relat. Phenom. 123 365
[6] Berakdar J, Lahmam-Bennani A and Dal Cappello C 2003 Phys. Reports 374 91
[7] Catoire F, Staicu Casagrande E M, Lahmam-Bennani A, Duguet A, Naja A, Ren X G, Lohmann B and Avaldi L, 2007 Rev. Sci. Instrum. 78 013108
[8] Kheifets A S, Naja A, Staicu Casagrande E M and Lahmam-Bennani A 2009 J. Phys. B: At. Mol. Opt. Phys. 42 165204
[9] Bray I, Fursa D V, Kherifets A and Stelbovics A 2002 J. Phys. B: At. Mol. Opt. Phys. 35 R117
[10] Staicu Casagrande E M, Naja A, Mezdari F, Lahmam-Bennani A, Bolognesi P, Joulakian B, Chuluunbaatar O, Al-Hagan O, Madison D H, Fursa D V and Bray I 2008 J. Phys. B: At. Mol. Opt. Phys. 41 025204
[11] Cohen H D and Fano U 1966 Phys. Rev. 150 30
[12] Sia C R, Fojón O A, Weck P F, Hanssen J and Rivarola R D 2003 J. Phys. B: At. Mol. Opt. Phys. 36 L257
[13] Naja A, Staicu-Casagrande E M, Ren X G, Catoire F, Lahmam-Bennani A and Dal Cappello C 2007 J. Phys. B: At. Mol. Opt. Phys. 40 2871