Calculations for (e,3e) and (γ ,2e)

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Abstract : an ab initio calculation using correlated wavefunctions for the target as well as for the two ejected electrons is reported for the case of the double ionization of rare gases by electron impact (e, 3e) and photon impact (γ ,2e).

Good agreement is observed with available (e,3e) and (γ ,2e) experimental results .

1 INTRODUCTION

The double ionization of atoms has recently received a considerable amount of interest . Determination of fully differential cross sections have been performed on krypton and xenon(1,2,3), on argon(4) and on helium(5) for the case of photon impact (γ ,2e) . The same kind of experiments are done for the double ionization by electron impact (e,3e) of argon(6) and krypton(7) where we have an additional difficulty at detecting the three electrons in coincidence . Few theoretical models are now available . A model based on the Wannier theory(8) and the use of a Gaussian function to represent the electron-electron interaction gives results which do not agree with the first (γ ,2e) experiments on helium . Ab initio calculations made by Le Rouzo and Dal Cappello(9) and Dal Cappello and Le Rouzo (10) using an effective charge model also give results different from the experiments(11) . An important improvement has been observed when Joulakian et al(12) and Maulbetsch et al(13) applied the correlated BBK wavefunction(14) respectively to the calculation of the (e,3e) and (γ ,2e) cross sections for helium . This model provides the best agreement with the recent experiments of Schwartzkopf et al(5) .

The application of the BBK wavefunction needs tedious calculations and time consuming procedures when applied to the double ionization of many electron atoms .For this reason we have developped an approximate BBK model(15)

constructed by the product of Coulomb waves multiplied by a Coulomb correlation factor arising from the normalisation of the electron-electron interaction . This factor apparently contains the major part of the final state correlation as it gives results which are in good agreement⁽¹⁵⁾ with those obtained by the complete BBK function at least for the kinematics we have considered.

The purpose of this paper is to apply the approximate BBK to the double ionization of noble gases in order to show the influence of the initial and final state wavefunctions in the calculation of the differential cross sections .

2. THEORY

The theory used in this paper has been previously detailed in the paper of Dal Cappello et al⁽¹⁵⁾ and Hda et al⁽¹⁶⁾ . We first investigate the double ionization of an helium atom by an incident electron . We use plane waves for the initial and final states of the fast colliding electron and we obtain the plane wave Born approximation for the fivefold differential cross section :

$$\sigma^{(5)} = \frac{d^5 \sigma}{d\Omega_s d\Omega_1 d\Omega_2 d(k_1^2/2) d(k_2^2/2)} = \frac{4 k_s k_1 k_2}{k_i K^2} (|M|^2) \quad (1)$$

where $\sigma^{(5)}$ represents the fivefold differential cross section with respect to the three solid angles (respectively of the scattered and the two ejected electrons) . The momentum transfer is $\vec{K} = \vec{k}_i - \vec{k}_s$, where $\vec{k}_i$ and $\vec{k}_s$ are the momentum of the incident and scattered electrons respectively ,while $\vec{k}_1$ and $\vec{k}_2$ are the momenta of the two ejected electrons .

The matrix element M is given by :

$$M = \frac{1}{K} \langle \Psi_f | -2 + \sum_{n=1}^2 e^{i\vec{K} \cdot \vec{r}_n} | \Psi_i \rangle \quad (2).$$

where ψ_i is the wavefunction for the initial state and ψ_f that of the final state.

The dipole approximation (dipole-length formulation⁽⁹⁾) used in double photoionization (DPI) is obtained when $\vec{K}$ tends to zero :

$$M \rightarrow M_{DPI} = i \langle \Psi_f | \vec{u} \cdot \vec{r}_1 + \vec{u} \cdot \vec{r}_2 | \Psi_i \rangle \quad (3).$$

here the unit vector $\vec{u} = \frac{\vec{K}}{K}$ represents the direction of the electric field for DPI processes .

A simplification is obtained by expanding ψ_i in terms of products of one electron orbitals :

$$\phi = Y_l^m(\hat{r}) R(n, \varepsilon, r) \quad (4).$$

and ψ_f as the product of two Coulomb wavefunctions multiplied by the Coulomb correlation factor :

$$\begin{aligned} \psi_f = & \frac{e^{i\vec{k}_1 \cdot \vec{r}_1}}{(2\pi)^{3/2}} e^{iZ/2k_1} \Gamma(1+iZ/k_1) {}_1F_1\left(\frac{-iZ}{k_1}, 1; -i(k_1 r_1 + \vec{k}_1 \cdot \vec{r}_1)\right) \\ & \frac{e^{i\vec{k}_2 \cdot \vec{r}_2}}{(2\pi)^{3/2}} e^{iZ/2k_2} \Gamma(1+iZ/k_2) {}_1F_1\left(\frac{-iZ}{k_2}, 1; -i(k_2 r_2 + \vec{k}_2 \cdot \vec{r}_2)\right) \\ & e^{-\pi/4k_{12}} {}_1F_2\left(1-i/2k_{12}\right) \end{aligned} \quad (5).$$

$$\text{and } \vec{k}_{12} = \vec{k}_1 - \vec{k}_2.$$

This model can be applied to the double ionization of other atoms under simple approximations⁽¹⁶⁾ if we consider only the two active electrons⁽¹¹⁾. In the double ionization of the krypton atom whose outermost electrons occupy the orbital 4p the residual ion is left in one of the 15 possible states of the 4p⁴ subshell. Neglecting spin-orbit coupling there are three possible terms, namely 1S, 1D and 3P. When the energy resolution is too poor to select a particular energy level of the ion (7) we must add up the cross sections for all these states before comparing the theoretical calculations to the experimental results.

The initial state for the two active electrons of the helium atom can be described by many accurate wavefunctions which are available in the literature. In order to show the importance of radial and angular correlations, three wavefunctions will be considered.

a) A first wavefunction of Silvermann et al⁽¹⁷⁾ that includes (1s) radial correlations (33% of the total correlation energy).

b) A second wavefunction of Silvermann et al⁽¹⁷⁾ that includes (1s) radial and (2p) angular correlations (80% of the total correlation energy).

c) A wavefunction of Tweed and Langlois⁽¹⁸⁾ which includes (1s,2s) radial and (2p,3p,3d) angular correlations (94% of the total correlation energy).

For the case of the ground state of other noble gases, the choice of accurate wavefunctions is limited to the wavefunctions calculated by superposition of configurations. Here we will use the following eight components⁽¹⁹⁾ (for instance, in the case of krypton 4p⁶)

$$\begin{aligned} & 4p^6, \quad 4p^4(1S)4d^2(1S), \quad 4p^4(1S)5s^2(1S), \quad 4p^4(1S)5p^2(1S), \\ & 4p^4(3P)5p^2(3P), \quad 4p^4(3P)4d^2(3P), \quad 4p^4(1D)5p^2(1D), \\ & 4p^4(1D)4d^2(1D) \end{aligned} \quad (6).$$

Our model may be applied to the double ionization of any atom if we restrict ourselves to the first-order process (the colliding particle, electron or photon, interacts with one electron of the target, the second electron of the target leaves it because it sees a change of the potential). It is usual to neglect the second-order processes for high incident energy. These second-order processes⁽²⁰⁾ require the inclusion of the second Born terms approximation which seems to be very difficult to apply for the double ionization with the same wavefunction for the final state.

3. RESULTS AND DISCUSSION :

The recent experimental results of Schwarzkopf et al⁽⁵⁾ for the DPI of helium allows us to check our model. By applying the dipole length formulation (3) we have calculated the differential cross section for the case of linear polarisation. Fig. 1 shows that our calculated cross sections are in reasonable agreement with the experimental results provided that the initial state wavefunction is sufficiently correlated. More precisely it shows clearly that angular correlation must be accounted for, since when we use a wavefunction with only radial correlation we observe important discrepancies between theory and experiment.

If we remember that Le Rouzo et al⁽⁹⁾ and Maulbetsch et al⁽¹³⁾ have shown that the correlation in the final state is necessary to describe the DPI of helium, we can conclude that a good model must include both the angular correlation in the initial state and the correlation in the final state. Nevertheless it seems actually difficult to use these kind of experiments to distinguish between a highly correlated wavefunction⁽²⁰⁾ and a less correlated wavefunction⁽¹⁷⁾. It is interesting now to consider the same kinematical conditions for a future (e,3e) experiment on helium. Fig. 2 shows the result of the calculations using relation (2). The shapes of the curves of fig.1 and fig.2 are similar⁽¹⁵⁾ for these energies (10 eV for each ejected electron). A small difference appears in the direction opposite to that of the momentum transfer: the differential cross section for DPI is always zero but it is non-zero for (e,3e) in the case of helium. Fig. 3 illustrates a comparison between our model and the "true" BBK model⁽¹²⁾ for an (e,3e) collision with a first electron leaving the target in a direction perpendicular to the momentum transfer. A good agreement is found between the "true" BBK model calculated with an Hylleraas initial state wavefunction and our model when we use a highly correlated wavefunction for the initial state. It is interesting to note that the two lobes are not equal in an (e,3e) collision but that they are equal for a (γ ,2e) collision (fig.4). The importance of each lobe is sensitive to the

amount of correlation included in the initial state wavefunction.

As a second example we now investigate the case of many-electron atoms. The recent experiments of Lahmam-Bennani et al on krypton⁽⁷⁾ lead to interesting verifications of the theoretical results. Fig.5 shows a comparison between experiment and the results computed in our model using the correlated wavefunction (5) and Hartree-Fock wavefunction⁽²¹⁾ for the initial state. The agreement with experiment is good and we can see small differences between the shapes of the two curves and large differences between their amplitudes. In other typical experimental situations the cross section exhibits⁽¹⁶⁾ the same behaviour.

The experiment of Lahmam-Bennani et al on argon⁽⁶⁾ has also been performed in the same kinematical conditions as that on krypton but with a different scattering angle. As shown in fig. 6 the cross sections calculated with the Hartree-Fock wavefunction and with a superposition of configurations for the initial state are very different. We obtain a fairly good agreement with the available experimental data only if we account for the electronic correlation in the initial state. The situation is quite different for neon as shown in fig. 7. Contrary to the cases of argon and krypton the correlation in the initial state has only a minor influence on the calculated cross section. This result is surprising at first sight and it is probably related to the fact that for argon and krypton the most important added configuration ($3d^2$ and $4d^2$ respectively) belongs to the same complexe as the dominant configuration ($3p^6$ and $4p^6$ respectively) while this is not the case for the configurations $2p^4 3d^2$ and $2p^6$ for neon.

3 CONCLUSION

We have shown that our model including the correlation in the final state and in the initial state is able to explain the major part of $(e,3e)$ and $(\gamma,2e)$ experiments. These kind of experiments constitute the best way to understand the mechanisms of the double ionization and to "measure" the correlation in an atom.

Fig.1.

The triple differential cross sections for the $(\gamma, 2e)$ double ionization of helium, plotted as a function of the direction $\hat{k}_2$ of one electron, for a fixed direction $\hat{k}_1$ along $\hat{u}$. Ejected electron energies are 10 eV.

Final-state wavefunction : approximate BBK model.

Initial-state wavefunction :

— wavefunction of Silvermann et al(17) (radial correlation)

....wavefunction of Silvermann et al(17) (radial and angular correlation)

-.-.-wavefunction of Tweed et Langlois(18) (radial and angular correlation)

| experiments of Schwartzkopf et al(5)

↑ direction of the polarisation vector $\hat{u}$ and $\hat{k}_1$

Fig.2.

The differential cross sections for the $(e, 3e)$ double ionization of helium. Incident, scattered and ejected energies are respectively 5099 eV, 5000 eV and 10 eV.

The scattering angle is -0.4° . The differential cross sections are plotted as a function of the direction $\hat{k}_2$ of one electron, for a fixed direction $\hat{k}_1$ along $\hat{K}$.

Final-state wavefunction : approximate BBK model.

Initial-state wavefunction :

— wavefunction of Silvermann et al(17) (radial correlation)

....wavefunction of Silvermann et al(17) (radial and angular correlation)

-.-.-wavefunction of Tweed et Langlois(18) (radial and angular correlation)

↑ direction of the momentum vectors $\hat{K}$ and $\hat{k}_1$

Fig.3

The differential cross sections for the (e,3e) double ionization of helium. Incident, scattered and ejected energies are respectively 5099 eV, 5000 eV and 10 eV. The scattering angle is -0.4° . The differential cross sections are plotted as a function of the direction $\vec{k}_2$ of one electron, for a fixed direction $\vec{k}_1$ perpendicular to the momentum transfer $\vec{K}$.

Final-state wavefunction : "true" BBK model.

Initial-state wavefunction :

..... wavefunction of Hylleraas

↑ direction of the momentum vector $\vec{K}$

↑ direction of $\vec{k}_1$

Fig.4

Same as Fig.1 except :

↑ direction of the polarisation vector $\vec{U}$

↑ direction of $\vec{k}_1$

Fig. 5.

The fivefold differential cross section (FDCS) atomic units for the $(e,3e)$ double ionization of krypton ($4p^6$). Electrons energies are 5568.4 eV, 5500 eV, 20 eV and 10 eV, respectively for incident, scattered, first and second ejected electrons. The scattering and the first ejected electron angles are -1° and 255° . The FDCS is plotted as a function of the direction E_2 of the second ejected electron.

— superposition of configuration of Langlois(19)

..... Hartree - Fock wavefunction of Clementi(21)

⊥ experiments of Lahmam-Bennani(7)

↑ direction of the momentum transfer R

↑ direction of the first ejected electron E_1

Fig. 6

Same as Fig.5 except : for the $(e,3e)$ double ionization of argon ($3p^6$).

Electron energies are 5553 eV, 5480 eV, 20eV and 10eV, respectively for incident, scattered and ejected electrons. The scattering and the first ejected angles are -0.45° and 255° .

Fig.7.

Same as Fig.5 except : for the (e,3e) double ionization of neon ($2p^6$).

Electron energies are 5596 eV , 5500 eV , 20eV and 10eV , respectively for incident , scattered and ejected electrons . The scattering and the first ejected angles are $-0^\circ 45'$ and 255° .

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