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Mechanisms of double ionization of atoms by electron impact

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Recent high-energy $(e,3e)$ experiments indicate that the one-step mechanism (shake-off process) is insufficient. We have performed an *ab initio* calculation that takes into account all the two-step mechanisms of double ionization. This calculation uses the second Born approximation and correlated wave functions for the initial bound state. We show that the two-step processes are not negligible and must be added to the one-step process with the production of interference terms. [S1050-2947(98)51002-5]

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I. INTRODUCTION

Double ionization of neutral atoms by electron impact is due to different processes, namely, (i) a direct process consisting of the ejection of two electrons without any internal rearrangement of bound electrons and (ii) an indirect process resulting in an ejection of a single inner-shell electron followed by Auger decay. We consider here the direct process that involves the so-called shake-off (SO), two-step 1 (TS1) and two-step 2 (TS2) mechanisms [1]. We restrict this study to the case of high incident energy, which allows us to describe the incoming and scattered electrons by plane waves. This restriction corresponds to the kinematic conditions used in the latest experiments [2,3]. The SO process is a single interaction between the incident electron and one target electron [1,4]. It leads to a first ejected electron. This first ionization is followed by a relaxation process due to the sudden change of potential that is responsible for a second ejection. This one-step mechanism (because there is only one interaction between the incoming electron and the target) can be described by the first Born approximation [5]. The so-called TS1 process [1] consists of a first interaction between the incoming electron and one target electron. It leads to a first ejected electron that interacts with another target electron. This second interaction leads to an ejection of another bound electron. This TS1 mechanism is described by the second Born approximation because two interactions are involved. The so-called TS2 process [1] takes into account two interactions between the incoming electron and the target. The first is concerned with the collision of the incoming electron with one target electron. Then the intermediate scattered electron interacts with another target electron. These two interactions can be incorporated in the second Born approximation too.

Up to now there has been no calculation that includes these three mechanisms. Popov *et al.* [6] have studied the TS1 mechanism when both ejected electrons have high energy. They have used some approximations and a free parameter in their formalism. They have included interference effects by adding the SO and TS1 amplitudes. Very recently, El Mkhanter and Dal Cappello [7,8] have performed an *ab initio* TS2 calculation without any free parameter. They have also considered the interference effects by adding the SO and the TS2 amplitudes. In this work we consider all the three mechanisms without any free parameter.

II. THEORY

We investigate the case of the double ionization of helium where the incoming and scattered electrons are fast (their energies vary between 1 keV [9] and 5 keV [2,3]) and can be described by plane waves. The fivefold differential cross section (FDCS) in the standard Born approximation [4] is written as (atomic units are used throughout)

$$\frac{d^5\sigma}{dE_1 dE_2 d\Omega_s d\Omega_1 d\Omega_2} = \frac{k_s k_1 k_2}{k_i} |M_{\mathrm{SO}} + M_{\mathrm{TS1}} + M_{\mathrm{TS2}}|^2, \tag{1}$$

where $d\Omega_s$, $d\Omega_1$, and $d\Omega_2$ denote, respectively, the elements of solid angles for the scattered and the two ejected electrons. The energy intervals are represented by dE_1 and dE_2. The amplitudes M_{SO}, M_{TS1}, and M_{TS2} correspond, respectively, to the SO, TS1, and TS2 processes. The FDCS was measured in 1989 in a difficult experiment [10], where the three electrons (the scattered and the two ejected) in the final state were detected in coincidence. These experiments constitute the best way to investigate finer details of the mechanisms of the double ionization. The SO process leads to

$$M_{\mathrm{SO}} = -\frac{1}{2\pi}\left\langle \psi_f^{(-)}(\vec{k}_1,\vec{k}_2;\vec{r}_1,\vec{r}_2) e^{i\vec{k}_s\cdot\vec{r}_0}\left| -\frac{2}{r_0} + \frac{1}{|\vec{r}_{01}|} + \frac{1}{|\vec{r}_{02}|}\right| \psi_i(\vec{r}_1,\vec{r}_2) e^{i\vec{k}_i\cdot\vec{r}_0}\right\rangle, \tag{2}$$

where $\vec{k}_i$, $\vec{k}_s$, $\vec{k}_1$, and $\vec{k}_2$ denote, respectively, the momenta of incident, scattered, the first, and second ejected electrons. The initial-state wave function of helium $\psi_i(\vec{r}_1,\vec{r}_2)$ is that of Silverman *et al.* [11], which includes both radial and angular correlations. The final-state wave function is the approximate Brauner-Briggs-Klar wave function (Dal Cappello

et al. [12]), which describes each ejected electron by a Coulomb wave function and includes the repulsive Gamow factor. This Gamow factor takes into account the Coulomb interaction between each ejected electron. Equation (2) has been studied by several authors (see, for instance, Popov *et al.* [6] and El Mkhanter *et al.* [13]). The TS1 process makes use of the second Born approximation (Tweed [4], Popov *et al.* [6])

$$M_{\mathrm{TS1}}=-\left(\frac{1}{\pi}\right)\sum_n \int \frac{d\vec{k}_a}{(2\pi)^3[k_i^2-k_a^2-k_s^2-2I_n]}\left\langle \psi_f^{(-)}(\vec{k}_1,\vec{k}_2;\vec{r}_1,\vec{r}_2)\left|\frac{1}{|\vec{r}_{12}|}\right|\psi_n^{(+)}(\vec{k}_a;\vec{r}_1,\vec{r}_2)\right\rangle\left\langle \psi_n^{(-)}(\vec{k}_a;\vec{r}_1,\vec{r}_2)e^{i\vec{k}_s\cdot\vec{r}_0}\left|-\frac{2}{r_0}+\frac{1}{|\vec{r}_{01}|}+\frac{1}{|\vec{r}_{02}|}\right|\psi_i(\vec{r}_1,\vec{r}_2)e^{i\vec{k}_i\cdot\vec{r}_0}\right\rangle, \tag{3}$$

where I_n represents the energy necessary to eject one target electron leaving the residual ion He^+ in its ground state and ψ_n is a product of an ingoing or outgoing Coulomb wave function (which represents the first intermediate ejected electron) multiplied by the wave function of the He^+ atom. This term is difficult to calculate since it involves the product of three Coulomb waves. It needs a long time, even on a fast computer (it is a six-dimensional numerical integration), and so only the case $n=1$ case is investigated. The TS2 process leads to [8]

$$\begin{aligned}M_{\mathrm{TS2}}=&-\left(\frac{1}{\pi}\right)\sum_n \int \frac{d\vec{k}_b}{(2\pi)^3[k_i^2-k_b^2-k_1^2-2I_n]}\left\langle \psi_f^{(-)}(\vec{k}_2,\vec{r}_2)e^{i\vec{k}_s\cdot\vec{r}_0}\left|\frac{1}{|\vec{r}_{02}|}\right|e^{i\vec{k}_b\cdot\vec{r}_0}\psi_n(\vec{r}_2)\right\rangle\\ &\times\varphi(|\vec{k}_1-\vec{k}_2|)\left\langle \psi_n^{(-)}(\vec{k}_1;\vec{r}_1,\vec{r}_2)e^{i\vec{k}_b\cdot\vec{r}_0}\left|-\frac{2}{r_0}+\frac{1}{|\vec{r}_{01}|}+\frac{1}{|\vec{r}_{02}|}\right|\psi_i(\vec{r}_1,\vec{r}_2)e^{i\vec{k}_i\cdot\vec{r}_0}\right\rangle\\ &-\left(\frac{1}{\pi}\right)\sum_n \int \frac{d\vec{k}_b}{(2\pi)^3[k_i^2-k_b^2-k_2^2-2I_n]}\left\langle \psi_f^{(-)}(\vec{k}_1,\vec{r}_1)e^{i\vec{k}_s\cdot\vec{r}_0}\left|\frac{1}{|\vec{r}_{01}|}\right|e^{i\vec{k}_b\cdot\vec{r}_0}\psi_n(\vec{r}_1)\right\rangle\\ &\times\varphi(|\vec{k}_1-\vec{k}_2|)\left\langle \psi_n^{(-)}(\vec{k}_2;\vec{r}_1,\vec{r}_2)e^{i\vec{k}_b\cdot\vec{r}_0}\left|-\frac{2}{r_0}+\frac{1}{|\vec{r}_{01}|}+\frac{1}{|\vec{r}_{02}|}\right|\psi_i(\vec{r}_1,\vec{r}_2)e^{i\vec{k}_i\cdot\vec{r}_0}\right\rangle,\end{aligned} \tag{4}$$

with

$$\varphi(|\vec{k}_1-\vec{k}_2|)=e^{\pi\xi_{12}}\Gamma(1-i\xi_{12}) \tag{5}$$

and

$$\xi_{12}=\frac{1}{|\vec{k}_1-\vec{k}_2|}. \tag{6}$$

Equation (4) represents well the successive interactions of the incoming electron with two target electrons. The intermediate scattered electron, described here by a plane wave $e^{i\vec{k}_b\cdot\vec{r}_0}$, collides with the last bound electron (1 or 2) after a first ionization without $(n=1)$ or with simultaneous excitation. Because of these two successive interactions in the TS2 mechanism, the symmetry around the momentum transfer $\vec{k}_i-\vec{k}_s$ is broken. We note that the same result is obtained by using the second Born approximation to describe $(e,2e)$ collisions at low incident energy [14]. The evaluation of the integral over $d\vec{k}_b$ or $d\vec{k}_a$ (for the TS1 process) needs a great amount of care [15] since the integrand is singular when $\vec{k}_a^2=\vec{k}_i^2-\vec{k}_s^2-2I_n$, $\vec{k}_b^2=\vec{k}_i^2-\vec{k}_1^2-2I_n$, $\vec{k}_b^2=\vec{k}_i^2-\vec{k}_2^2-2I_n$, $\vec{k}_a=\vec{k}_1$, $\vec{k}_a=\vec{k}_2$, $\vec{k}_b=\vec{k}_s$ in Eqs. (3) and (4).

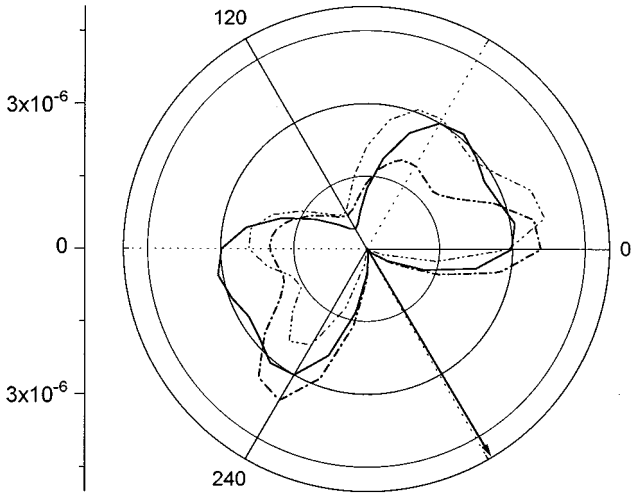

FIG. 1. The fivefold differential cross section (FDCS) in atomic units for the $(e,3e)$ double ionization of helium in coplanar geometry. The incident, scattered, and ejection energies are 5000, 4901, and 10 eV, respectively. The scattering angle is 1°. One ejected electron is detected along the momentum transfer direction, while the other is detected at variable angles. The FDCS is plotted in polar coordinates as a function of the direction $\vec{k}_2$ of the second ejected electron. The incident electron is moving along the x axis. —, SO mechanism; -·-·-, SO and TS1 mechanisms; -..-..- SO, TS1, and TS2 mechanisms.

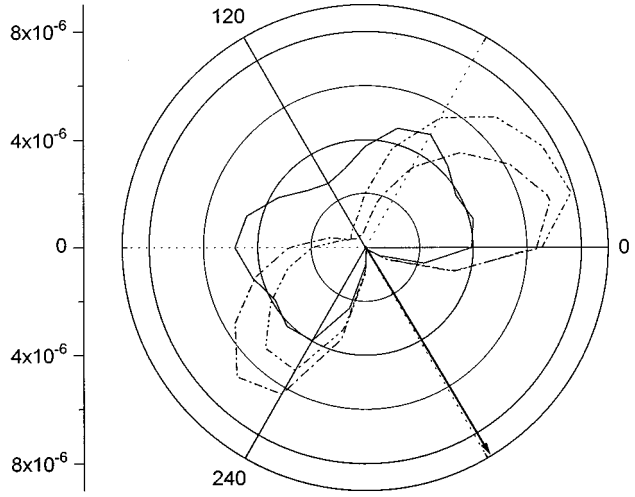

FIG. 2. Same as Fig. 1 except for the ejected energies $E_1 = 15$ eV and $E_2 = 5$ eV. The faster ejected electron is detected along the momentum transfer direction.

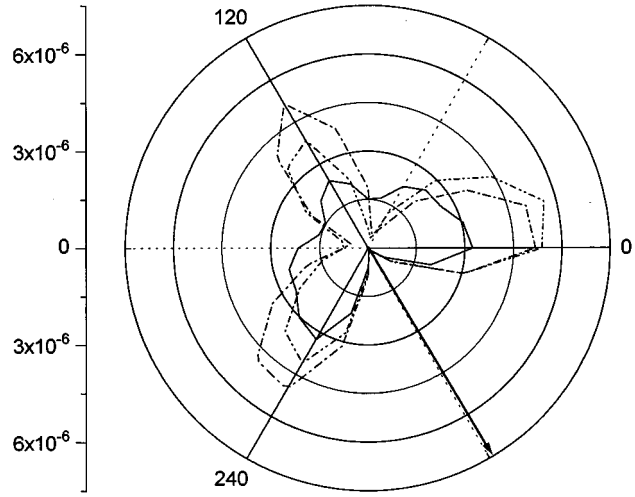

FIG. 3. Same as Fig. 1 except for the ejected energies $E_1 = 5$ eV and $E_2 = 15$ eV. The slower ejected electron is detected along the momentum transfer direction.

III. RESULTS

To our knowledge, up to now no $(e,3e)$ experiments on helium have been performed. We use here the same kinematic conditions of Lahmam-Bennani *et al.* [2], who recently measured the FDCS of the double ionization of noble gases (argon and neon). The scattered electron is fast and is detected at a low angle. Each ejected electron has low energy $(E_1 = E_2 = 10$ eV). All of these $(e,3e)$ measurements are coplanar: the scattering plane that contains the vectors $\vec{k}_i$ and $\vec{k}_s$ is characterized by the azimuthal angle $\phi_s = 0$. We investigate the case where one ejected electron is detected along the momentum transfer, while the second ejected electron is detected at variable angles. This situation is very sensitive to the mechanisms used to describe the double ionization because the contributions of SO and TS1 are symmetrical with regard to the direction of the momentum transfer, whereas the contribution of TS2 is not symmetrical. It has been shown [2,3] that for noble gases this symmetry around the momentum transfer is broken for equal energy sharing. We now find that in the present case of the double ionization of helium this symmetry is broken too, even for unequal energy sharing.

Figure 1 shows the case of equal energy sharing $(E_1 = E_2 = 10$ eV). The symmetry around the momentum transfer is broken and it is not possible here to say that one mechanism is more important than another. All the mechanisms contribute significantly. This seems to contradict the observation of McGuire [16], who estimated that the total cross section in the TS1 and the TS2 mechanisms is negligible at high incident energy (~ 5 KeV).

Figure 2 exhibits a case of unequal energy sharing $(E_1 = 15$ eV, $E_2 = 5$ eV). The faster ejected electron is detected along the momentum transfer direction. We observe that the symmetry is broken too. We notice that interference effects destroy the amplitude where the slower ejected electron is detected opposite the momentum transfer direction. This theoretical finding is important and could be checked in experiments in the near future.

Figure 3 shows another case of unequal energy sharing $(E_1 = 5$ eV, $E_2 = 15$ eV). Here the slower ejected electron is detected along the momentum transfer. The symmetry around the momentum transfer is broken again but the shift is small. It seems that the SO and TS1 contributions are the most important. We also notice that the TS1 mechanism is important when the direction of the faster ejected electron is antiparallel to that of momentum transfer direction.

IV. CONCLUSION

We have calculated the fivefold differential cross section of electron-impact double ionization of helium for high incident and low ejection energies. In this calculation we have taken account of all the contributions of one-step and two-step mechanisms. We have also included the accurate correlated wave function for the initial state and pairwise Coulomb interaction between the two ejected electrons and the ion in the final state. We have shown that the TS2 mechanism is important in the case of equal sharing as has been observed in recent experiments. We have also found that the TS1 mechanism is the most important process for unequal energy sharing when the direction of the faster ejected electron is opposite that of momentum transfer.

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