Double photoionization of helium: Effect of the Coulomb repulsion on the angle-resolved cross section

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The effect of the Coulomb repulsion between the two ejected electrons on the angle-resolved cross section is examined; it is shown that in the general case the effective-charge approximation is not in agreement with solutions of the three-body Schrödinger equation with the appropriate asymptotic boundary conditions, and an approach is proposed in order to introduce the electron repulsion in the calculation of the sixfold differential cross section $d^6\sigma^{2+}/d^3\mathbf{k}d^3\mathbf{k}'$.

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Double photoionization (DPI) is a striking consequence of the electron correlation since it would be rigorously forbidden within the independent-particle model: It follows that DPI is much less probable than the single photoionization but it is a very powerful tool in the investigation of correlation effects. Recently, interest in DPI has greatly increased both from an experimental and from a theoretical point of view: The advent of intense synchroton radiation has allowed measurement of the total cross section for the DPI process [1] and analysis of the angular distribution for the fragment ions following DPI of H_2 [2]. From the theoretical point of view the literature offers a few approaches to the description of DPI: Many-body perturbation theory [3] has been used only for the calculation of the total DPI cross section, i.e., that integrated over the angular variables; on the other hand, the wave-function approach (WFA), pioneered by Byron and Joachain [4] for the DPI of helium atoms, has recently been applied by Le Rouzo and Dal Cappello [5] to the calculation of the angle-resolved cross sections. The case of DPI of helium is the most simple DPI process; nevertheless, the problem of the calculation of the angle-resolved cross section is very difficult because of the long range of the Coulomb interaction and because it displays the main effects of the electron repulsion in the final state, without the complications involved by structured cores. The purpose of this report is to show that the effective-charge approximation cannot be applied to the case under consideration and to describe an alternative approach to the problem of calculating the effects of the Coulomb repulsion on the angular distribution of the ejected electrons. The assumptions of the WFA (dipole approximation, nonrelativistic energies, neglect of spin-dependent interactions) are well satisfied in the case considered, so that the accuracy of the WFA depends essentially on the choice of the wave functions used to describe the initial and the final state of the system; the wave function for the initial state (bound) of the helium atom is known with very good accuracy so that the real problem consists in the choice of the final-state wave function, where both electrons occupy a continuum state. These double-continuum wave functions have been studied in recent years [6–9] but only their asymptotic expressions are known, so that the WFA usually neglects the Coulomb repulsion between the photoejected electrons, which are considered within the independent-particle model. Since the spin-dependent interactions are neglected, the spin part of the wave functions can be ignored while the spin selection rules allow only symmetric final wave functions; in the case of the DPI of the helium atom, the singlet final state is then represented by the orbital wave functions

$$ {}^1\Psi_f(Z,\mathbf{k},\mathbf{k}'|\mathbf{r}_1,\mathbf{r}_2)=\frac{1}{\sqrt{2}}[\psi(Z,\mathbf{k}|\mathbf{r}_1)\psi(Z,\mathbf{k}'|\mathbf{r}_2) + \psi(Z,\mathbf{k}|\mathbf{r}_2)\psi(Z,\mathbf{k}'|\mathbf{r}_1)] , \quad (1) $$

where $\psi(Z,\mathbf{k}|\mathbf{r})$ are pure Coulomb waves, i.e., eigensolutions of the Schrödinger equation corresponding to an electron with a positive energy moving in the Coulomb field created by a charge Z [10]; such functions have the form

$$ \psi(Z,\mathbf{k}|\mathbf{r})=(2\pi)^{-3/2}C(\eta(Z,k),\mathbf{k},\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}} , \quad (2) $$

where

$$ C(\eta,\mathbf{k},\mathbf{r})\equiv\Gamma(1-i\eta)e^{-\pi\eta/2}\,{}_1F_1(i\eta,1,-i(kr+\mathbf{k}\cdot\mathbf{r})) , \quad (3) $$

${}_1F_1$ is the confluent hypergeometric function [11], and

$$ \eta(Z,k)\equiv -mZe^2/\hbar^2k . \quad (4) $$

The wave functions (2) have been normalized according to the condition

$$ \int\psi^*(Z,\mathbf{k}'|\mathbf{r})\psi(Z,\mathbf{k}|\mathbf{r})d^3\mathbf{r}=\delta(\mathbf{k}'-\mathbf{k}) . \quad (5) $$

The Coulomb waves (2) have the asymptotic behavior of a plane wave plus an incoming spherical wave, both with a logarithmic distorted phase [12],

$$ \psi(Z,\mathbf{k}|\mathbf{r})\approx(2\pi)^{-3/2}\left[\exp[i\mathbf{k}\cdot\mathbf{r}-i\eta\ln(kr-\mathbf{k}\cdot\mathbf{r})]+f_c(\theta)\frac{\exp[-i(kr-\eta\ln(2kr))]}{r}\right] , \quad (6) $$

where the Coulomb scattering amplitude is given by

$$f_c(\theta)=\frac{\exp\{i\eta\ln[\sin^2(\theta/2)]\}}{2ik\sin^2(\theta/2)}\frac{\Gamma(1-i\eta)}{\Gamma(i\eta)}\ , \tag{7}$$

and θ is the angle between $\mathbf{k}$ and $\mathbf{r}$. From the calculations by Le Rouzo and Dal Cappello [5] it results clearly that the choice of the functions (1) as final-state wave functions, in the matrix element corresponding to the angle-resolved cross section, is not correct; in fact, the event where both electrons are ejected in the direction of the polarization vector of the incident photon is the most probable: in the case where the two electrons have the same energy, this event means that the two electrons are in the same spatial position simultaneously, which is physically impossible because of the Coulomb repulsion (the exclusion principle does not apply to this case because the two electrons have opposite spin). Le Rouzo and Dal Cappello, following Rudge and Seaton [13,14], examine the effect of variable charges $Z_1(\mathbf{k},\mathbf{k}'),Z_2(\mathbf{k},\mathbf{k}')$ on the angular distribution of the two ejected electrons; they suppose that $Z_{1,2}(\mathbf{k},\mathbf{k}')\to\infty$, so that $d\sigma^{2+}\to 0$ for $\mathbf{k}\to\mathbf{k}'$, but the ratio of the two differential cross sections, calculated with respect to the length or the velocity form of the dipole matrix element, is very scarcely improved by the introduction of the effective charges (3.7 versus 3.9); besides, the use of effective charges modifies greatly the angular distribution and introduces new peaks for which it is difficult to give a physical interpretation. The effective charges were used by Rudge and Seaton [13] in the calculation of the single ionization of the hydrogen atom by electron impact; however, their final-state wave function (Eq. 3.40 in Ref. [13]) cannot be applied to the case of DPI because it does not represent two electrons with asymptotic momentums $\mathbf{k}$ and $\mathbf{k}'$. The correct scattering wave function for the three-body Schrödinger equation has been derived recently by Brawner, Briggs, and Klar [6], and their results coincide with the asymptotic solutions previously obtained by Garribotti and Miraglia [7]; it has the following form:

$$\Psi(\mathbf{k},\mathbf{k}'|\mathbf{r}_1,\mathbf{r}_2)=\frac{1}{(2\pi)^3\sqrt{2}}[\exp(i\mathbf{k}\cdot\mathbf{r}_1+i\mathbf{k}'\cdot\mathbf{r}_2)C(\eta(Z,k),\mathbf{k},\mathbf{r}_1)\times C(\eta(Z,k'),\mathbf{k}',\mathbf{r}_2)$$
$$\times C(\eta(-1,k_{\rm rel}),\mathbf{k}_{\rm rel},\mathbf{r}_1-\mathbf{r}_2)+T_{1\leftrightarrow 2}]\ , \tag{8}$$

where $\mathbf{k}_{\rm rel}\equiv\mathbf{k}-\mathbf{k}'$ and $T_{1\leftrightarrow 2}$ stands for the term in square brackets with 1 and 2 interchanged. Note that if $e\to 0$, the solution (8) reduces to the ordinary double plane wave with the correct normalization factor. The solution (8) is completely symmetric in all pairwise Coulomb interactions and differs from the wave function (1) for the presence of a factor representing the relative motion of the electrons; for large r_1 r_2 Eq. (8) represents two plane waves with a logaritmic distorted phase

$$\Psi(\mathbf{k},\mathbf{k}'|\mathbf{r}_1,\mathbf{r}_2)\approx\frac{1}{(2\pi)^3\sqrt{2}}$$
$$\times\{\exp[i\mathbf{k}\cdot\mathbf{r}_1+i\mathbf{k}'\cdot\mathbf{r}_2-i\gamma(\mathbf{r}_1,\mathbf{r}_2)]$$
$$+T_{1\leftrightarrow 2}\}\ , \tag{9}$$

where the distorted phase γ is given by

$$\gamma(\mathbf{r}_1,\mathbf{r}_2)\equiv\eta(Z,k)\ln(kr_1+\mathbf{k}\cdot\mathbf{r}_1)$$
$$+\eta(Z,k')\ln(k'r_2+\mathbf{k}'\cdot\mathbf{r}_2)$$
$$+\eta(-1,k_{\rm rel})$$
$$\times\ln[k_{\rm rel}|\mathbf{r}_1-\mathbf{r}_2|+\mathbf{k}_{\rm rel}\cdot(\mathbf{r}_1-\mathbf{r}_2)]\ . \tag{10}$$

The solution (9) was derived by Redmond and discussed by Peterkop [8] in connection with the partial-wave expansion proposed by Altick [9]. If the asymptotic expression (6) is considered, it is easy to see that, in the general case, the wave functions (1) do not approximate for large values of r_1 and r_2, the solution (8) or (9), for any value of the effective charges $Z_1(\mathbf{k},\mathbf{k}')$, $Z_2(\mathbf{k},\mathbf{k}')$; this proves that the correlation between free electrons in a Coulomb field cannot be introduced by means of effective charges, as proposed by Le Rouzo and Dal Cappello. The introduction of effective charges is possible only in the case $k\gg k'$; in fact, since electron 2 is much slower than electron 1, it is physically meaningful to consider the asymptotic expression (9) for $r_1\gg r_2\gg 1$ [see also Ref. [9(a)]]; in this case one has

$$\mathbf{k}-\mathbf{k}'\approx\mathbf{k}\ ,$$

$$\mathbf{r}_1-\mathbf{r}_2\approx\mathbf{r}_1\ ,$$

so that the distorted phase in the solution (9) becomes

$$\gamma(\mathbf{r}_1,\mathbf{r}_2)=\eta(Z-1,k)\ln(kr_1+\mathbf{k}\cdot\mathbf{r}_1)$$
$$+\eta(Z,k')\ln(k'r_2+\mathbf{k}'\cdot\mathbf{r}_2)\ , \tag{11}$$

which coincides with the asymptotic expansion of Eq. (1) with effective charges $Z_1=Z-1$, $Z_2=Z$. This result is very reasonable: in fact, since electron 2 is much slower than electron 1, their interaction is stronger when both electrons are near the origin, so that the average effect of electron 2 over electron 1 is to screen the nuclear charge, while the inverse is not true because electron 1 passes near the origin too quickly to modify sensibly the motion of electron 2. When the two electrons have comparable energies, their interaction cannot be localized near the origin, so that a description of their repulsion cannot be done by means of an effective central field. However, the behavior of the wave function near the origin can be investigated by means of a classical analog; in fact, since the wave function (1) is obtained by the time reversal of the solution of the scattering problem [10], it is possible

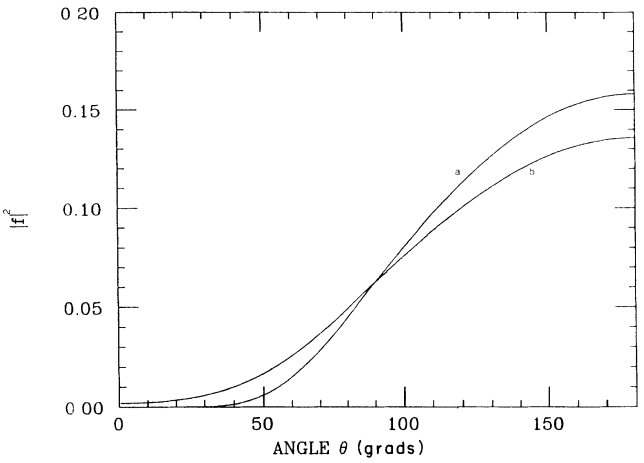

FIG. 1. The square modulus $|f(k_{\rm rel})|^2$ is represented as a function of the angle θ between $\mathbf{k}$ and $\mathbf{k}'$: Curve a refers to the case where both electrons have energy $\varepsilon=\varepsilon'=15$ eV, while curve b refers to the case where $\varepsilon=5$ eV and $\varepsilon'=25$ eV.

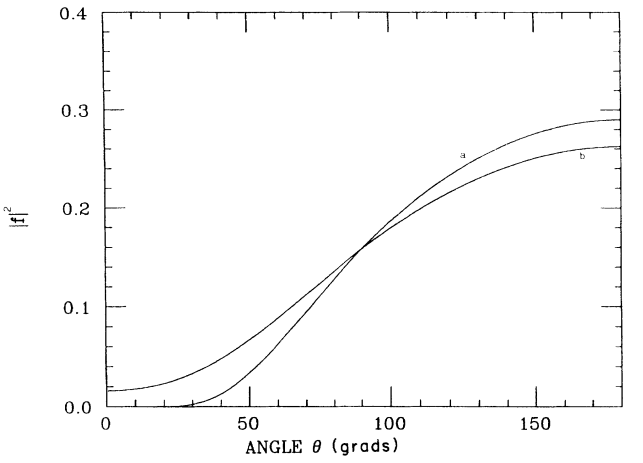

FIG. 2. The square modulus $|f(k_{\rm rel})|^2$ is represented as a function of the angle θ between $\mathbf{k}$ and $\mathbf{k}'$: Curve a refers to the case where both electrons have energy $\varepsilon=\varepsilon'=30$ eV, while curve b refers to the case where $\varepsilon=10$ eV and $\varepsilon'=50$ eV.

to describe the main features of the final state by considering the scattering by the nuclear charge of two electrons, incident with momentums $-\mathbf{k}$ and $\mathbf{k}'$: within this picture, it is clear that the main effect of the Coulomb repulsion between the two electrons is to remove each other from the neighborhood of the nuclear charge. This effect will be stronger when the two electrons have comparable energies and the angle between $\mathbf{k}$ and $\mathbf{k}'$ is small, that is, when $k_{\rm rel}$ is small: In fact, in this case the direction of the repulsion force is close to the normal to the electron momentums, so that the incident electron trajectories are strongly bent when the electrons are still far from the nuclear charge. The previous considerations show that near the origin, the wave function is lower in absolute value, with respect to the case where the Coulomb repulsion between the electron pair is neglected, say by a factor f; since the initial-state wave function is different from zero only near the origin, the dipole matrix element will approximately diminish by the same factor: by inspection of Eq. (8), such a factor is assumed to be

$$f(k_{\rm rel})=\Gamma(1-i\eta_{\rm rel})e^{-\pi\eta_{\rm rel}/2}\ , \tag{12}$$

where

$$\eta_{\rm rel}\equiv me^2/\hbar^2 k_{\rm rel}\ . \tag{13}$$

The square modulus of expression (12) is exactly the probability to find an electron with momentum $k_{\rm rel}$ near the origin, for a Coulomb field origined by a fixed charge $-e$ [consider that ${}_1F_1(a,b,x)\approx 1$ for $x\approx 0$] and it appears reasonable to use such an expression to weigh the effect of the Coulomb repulsion over the wave function near the origin, since the wave function (8) includes a multiplicative factor representing the relative motion of the two electrons (see Figs. 1 and 2). The approach proposed herein consists in choosing as a final-state wave function, the function (1) multiplied by the factor (12), but it is pointed out that such an approximation is to be considered valid only for small values of r_1, r_2; however, because of the exponential extinction of the initial-state wave function, the dipole matrix element will depend essentially on the behavior of the final-state wave function in such a spatial region. It is essential to investigate the behavior of the function (12) in the case $\mathbf{k}\to\mathbf{k}'$; it is easy to see that

$$f(k_{\rm rel})\to 0 \quad \text{for } \mathbf{k}\to\mathbf{k}'\ , \tag{14}$$

so that the double ejection of both electrons with the same $\mathbf{k}$ is forbidden without the introduction of an unphysical $-\infty$ effective charge in the origin. Besides, in the case $k\gg k'$, $\eta(Z,k)\ll 1$, since

$$|\Gamma(1+i\eta)|^2=\pi\eta/\sinh(\pi\eta)\approx 1 \quad \text{for } \eta\ll 1\ , \tag{15}$$

the main effect origined by the introduction of the factor (12) is the replacement of $\exp[-\pi\eta(Z,k)/2]$ by $\exp[-\pi\eta(Z-1,k)/2]$ into Eq. (1), which represents again the screening of the nuclear charge by the slower electron.

In conclusion, the introduction of the factor (12) in order to consider the effect of the Coulomb repulsion between the electron pair over the angle-resolved cross section presents the following two advantages, with respect to the effective-charge approximation:

(a) It gives directly the correct behavior in the two limit cases ($\mathbf{k}\to\mathbf{k}'$, $k\gg k'$) without the introduction of further hypothesis about the effective-charge functions $Z_1(\mathbf{k},\mathbf{k}')$, $Z_2(\mathbf{k},\mathbf{k}')$;

(b) It follows in a natural way from the asymptotic solution (8) of the three-body Schrödinger equation, while it has been shown that the effective-charge approximation is not in agreement with such a solution. Besides, the use of the factor (12) does not introduce new unphysical peaks in the angular distribution of the ejected electrons as does the effective-charge approximation [5].

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