Core-Excited Resonance Enhancement in the Two-Photon Complete Fragmentation of Helium

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A time-dependent close-coupling method is used to calculate, for the first time, fully differential cross sections for the complete fragmentation of helium by two photons. Surprising differences in the magnitude of the total-integral cross sections are found in comparisons with other calculations. These differences are found to be due to a core-excited resonance enhancement of the two-photon process for both single and double ionization. These calculations provide theoretical support for ground-breaking measurements expected to be obtained from free-electron laser experiments in the near future.

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The future development of high-powered free electron lasers at x-ray wavelengths promises to open up new avenues for the study of strongly correlated quantal dynamics [1]. At present, synchrotron light sources at x-ray wavelengths are being used to study the one-photon double ionization of atoms [2]. Absolute measurements of the energies and angles of emission of the two strongly correlated photoelectrons [3] provide a stringent test of recently developed nonperturbative theoretical methods [4–6]. In anticipation of future x-ray laser experiments, several theoretical predictions have been made of total probabilities and cross sections for the two-photon double ionization of helium [7–10]. In these studies the nonperturbative methods needed to treat the quantal three body problem involving two free photoelectrons in the Coulomb field of the nucleus have been further extended to include a nonperturbative interaction with a strong electromagnetic field.

In this Letter, we use a time-dependent close-coupling method [7] to calculate the first energy and angle differential cross sections for the two-photon double ionization of helium. As has been found in one-photon studies [11], the triple-differential cross section in ejected energy and the emission angles of the two photoelectrons provides the most stringent test for a comparison of theory and experiment. At equal ejected energies and for nearly equal emission angles the two outgoing photoelectrons remain highly correlated to quite large distances, well beyond the validity of any perturbative approach. For two-photon absorption, the total angular momentum of the two photoelectrons is a combination of $L = 0$ and $L = 2$ waves, resulting in dramatic differences with one-photon absorption differential cross sections. In an initial attempt to avoid complications from resonance processes, we limit our two-photon energies to those just above the total fragmentation threshold energy of helium. Thus, a generalized differential cross section may be defined over a wide range of laser field intensities, making easier comparisons with future experimental results. In addition, the time-dependent close-coupling approach for the calculation of triple-differential cross sections may be easily extended to the multiphoton double ionization studies of energy [12–14] and angular [15–17] distributions that have been made with high-power lasers at ultraviolet wavelengths.

In the following paragraphs we first give a brief description of the time-dependent close-coupling method and set out the equations defining the total integral, energy, and angle differential cross sections for two-photon double ionization, where we use atomic units unless otherwise stated. We then present results for these processes and compare, where possible, with other theoretical methods. We conclude with a brief summary.

The time-dependent Schrödinger equation for a two-electron atom in a strong, time-varying laser field can be written as

$$i\,\frac{\partial\Psi(\boldsymbol{r}_1,\boldsymbol{r}_2,t)}{\partial t} = (H_{\rm atom} + H_{\rm rad})\Psi(\boldsymbol{r}_1,\boldsymbol{r}_2,t)\,, \quad (1)$$

where expressions for $H_{\rm atom}$ and $H_{\rm rad}$ can be found in [6,7]. We choose the laser frequency $\omega = 45$ eV such that none of the doubly excited states of He, or excited states of He^+, are accessed by the direct two-photon process, as shown by the first process in Fig. 1. The intensity is sufficiently low that the Stark shift does not bring any of these states into this region, and that double-electron above threshold ionization processes are not probable. Following standard procedure, we expand the total wave function in coupled spherical harmonics and substitute it into Eq. (1) to derive a set of time-dependent partial differential equations for the radial wave functions $P^{LS}_{l_1 l_2}(r_1, r_2, t)$. These can be used to obtain, via a Coulomb transform, the wave functions in momentum space $P^{LS}_{l_1 l_2}(k_1, k_2, t)$ as discussed in [6].

The ground state of helium is found by relaxation of the time-dependent Schrödinger equation in imaginary time. The ground state wave functions are then time evolved by solving the time-dependent Schrödinger equation in real time subject to a 10-cycle pulse of linearly polarized light which has constant amplitude and is ramped on and off smoothly over one laser field period. We note that this is quite different from a previous time-dependent calculation [7] where the electric field amplitude was described

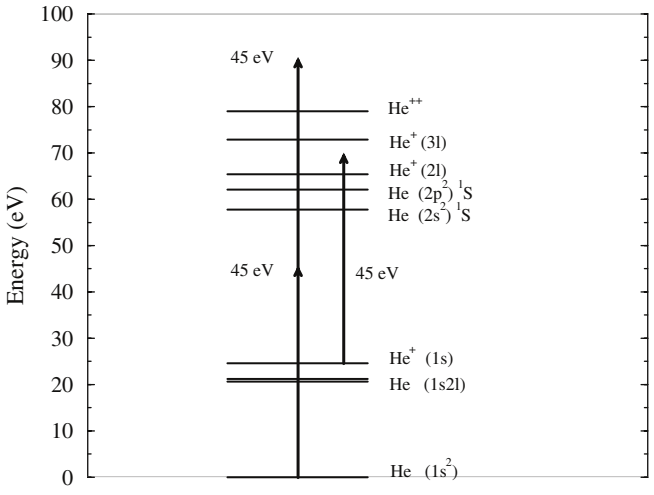

FIG. 1. Energy level diagram for helium, showing the two-photon double ionization process. It is clear that, for a photon energy of 45 eV, the direct path is nonresonant. However, a 45 eV photon from the ground state of He^+ lies between the $2l$ and $3l$ levels of He^+.

by a sine-squared pulse. The Schrödinger equation is solved on a 600×600 point lattice with a mesh spacing of $\Delta r = 0.1$. It was found that increasing the lattice to 1000×1000 points made a difference of no more than 3% in the results presented here. The results presented here are insensitive to the laser field ramped on/off over a longer number of periods. After propagation for several field periods after the laser pulse has been ramped off, standard projection techniques are used to extract the single or double ionization probability as required.

This description of the electric field amplitude allows us to define a total-integral cross section for single and double ionization by one or two photons in the following manner:

$$\sigma(\gamma, e) = \left(\frac{\omega}{I}\right)\frac{\mathcal{P}_{nk}}{\tau}, \qquad \sigma(2\gamma, e) = \left(\frac{\omega}{I}\right)^2\frac{\mathcal{P}_{nk}}{\tau},$$
$$\sigma(\gamma, 2e) = \left(\frac{\omega}{I}\right)\frac{\mathcal{P}_{kk}}{\tau}, \qquad \sigma(2\gamma, 2e) = \left(\frac{\omega}{I}\right)^2\frac{\mathcal{P}_{kk}}{\tau}, \tag{2}$$

where $\mathcal{P}_{nk}$ and $\mathcal{P}_{kk}$ are the probabilities for single and double ionization, respectively. Detailed expressions for these quantities in terms of the radial wave functions can be found in previous publications [6,7]. Also in these equations I is the intensity of the pulse and τ is the time for which the laser field is at full intensity.

By suitable projection onto the appropriate states all of these cross sections may be extracted from the time-dependent calculation. For a photon frequency of 45 eV, the single-photon single ionization cross section $\sigma(\gamma, e)$ is found to be 2.6 Mb, which is in excellent agreement with the experimental measurements of Samson *et al.* [18]. Next we calculated the $\sigma(\gamma, 2e)$ cross section at a photon energy of 99 eV using our new constant amplitude laser pulse method. This is an important check as the method described in this Letter is quite different from the Green's function approach used, with great success, to calculate total and differential cross sections in previous time-dependent calculations [6], which are in excellent agreement with experiment and other nonperturbative calculations [3–5]. Our value of 8.3 kb is in good agreement with the value calculated in [6], which gives us some confidence in our new constant amplitude laser pulse method of calculating cross sections as defined in Eq. (3).

The two-photon single ionization cross section $\sigma(2\gamma, e)$ at 45 eV is found to be 1.5×10^{-51} cm^4 s. Our two-photon double ionization cross section $\sigma(2\gamma, 2e)$ at 45 eV is found to be 1.2×10^{-52} cm^4 s. The cross section was calculated at an intensity of 10^{14} W cm^{-2} and is found to be only weakly dependent on intensity, with a slight decrease (increase) in the cross section at higher (lower) intensities. The ratio of these two quantities (0.08) is found to be in good agreement with calculations of this ratio in a similar energy range by Pindzola and Robicheaux [7], Parker *et al.* [8], and estimated from Nikolopoulos and Lambropoulos [9], where in all cases this ratio was found to be around 10%. However, the magnitude of our two-photon cross sections is considerably higher than those quoted in [9], where it was found that the $\sigma(2\gamma, e)$ cross section at 45 eV was 1.2×10^{-53} cm^4 s and the $\sigma(2\gamma, 2e)$ at 45 eV is found to be 1.5×10^{-54} cm^4 s. We also note that the value quoted in [10] of 1.3×10^{-54} cm^4 s for the $(2\gamma, 2e)$ cross section is again much lower than our value. This large difference was puzzling.

In an effort to resolve this discrepancy, we carried out a series of perturbation theory calculations [19,20], which were second order in the dipole operator with Hartree-Fock bound and continuum states, based on the expression

$$\sigma(2\gamma, e) = \frac{16\pi^2\omega^2}{c^2 k_f}\left|\sum_n \frac{\langle\psi_f|D|\psi_n\rangle\langle\psi_n|D|\psi_i\rangle}{E_i - E_n + \omega}\right|^2, \tag{3}$$

where c is the speed of light, $D = \hat{\boldsymbol{\epsilon}} \cdot (\mathbf{r}_1 + \mathbf{r}_2)$ is the dipole operator in the length gauge, and the sum is over a complete set of virtual intermediate states, $|\psi_n\rangle$. We note that this sum includes a principal value and pole contributions for photon energies ω greater than the ionization potential of helium. Our results for $|\psi_f\rangle = 1sk_f s\,{}^1S$ and $1sk_f d\,{}^1D$ are presented in Table I, under the column labeled perturbation theory, for photon energies of 15, 27.2, and 45 eV. The perturbation theory results are compared with the multichannel bound and continuum state calculations of Nikolopoulos and Lambropoulos [9], Proulx and Shakeshaft [21], and the present time-dependent close-coupling method. The time-dependent results are further subdivided into a $\sigma(2\gamma, e)$ total cross section leaving the ion in the $1s$ state, and a total cross section summed over the ground and all excited states of the ion. It is clear that at 15 eV all methods are in excellent agreement. However, as the photon energy is increased, the total time-dependent results increase significantly, whereas the contribution to the time-dependent

TABLE I. The $(2\gamma, e)$ cross sections for helium at various photon energies ω. We present the results of Proulx and Shakeshaft [21], Nikolopoulos and Lambropoulos [9], and our perturbation theory calculations. We also show two time-dependent close-coupling calculations, where TDCC($1s$) denotes the results of our calculations where the helium ion is left in the $1s$ state, and TDCC(tot) denotes the results of our calculations which are summed over the ground and all excited states of the helium ion. All results are in cm^4 s.

ω (eV)	Proulx and Shakeshaft	Nikolopoulos and Lambropoulos	Perturbation Theory	TDCC(1s)	TDCC(tot)
15	⋯	1.1×10^{-51}	1.2×10^{-51}	1.2×10^{-51}	1.2×10^{-51}
27.2	2.7×10^{-52}	1.0×10^{-52}	2.5×10^{-52}	4.5×10^{-52}	5.5×10^{-52}
45	⋯	1.2×10^{-53}	3.3×10^{-53}	1.0×10^{-52}	1.5×10^{-51}

results leaving the ion in the $1s$ state remains relatively close to the values obtained by the other multichannel state calculations [9,21] and our own perturbation theory calculations.

At a photon energy of 45 eV the largest contribution to the total time-dependent result leaves the ion in the $2p$ and $3p$ states. As shown in Fig. 1, a 45 eV photon from the ground state of He^+ lies between the He^+ $(2l)$ and He^+ $(3l)$ channels. We thus carried out an additional perturbation theory calculation, which was second order in the dipole operator using a mixed basis state set, at 45 eV in which $|\psi_f\rangle = 2pk_f p\,{}^1S$ and $|\psi_f\rangle = 2pk_f p\,{}^1D$, for which we found $\sigma(2\gamma, e) = 2.3 \times 10^{-51}$ cm^4 s, a value almost 70 times larger than the perturbative value for the $1skL\,{}^1L$ final states. This core-excited resonant process is included in the time-dependent calculations, but appears to be missing in the multichannel bound and continuum state calculations of [9]. As a further check, we repeated our calculations at photon energies which were closer to the $2p$ and $3p$ channels, respectively (see Fig. 1), and found increases in the ionization cross section from these channels. This core-excited resonance process is also responsible for our large $\sigma(2\gamma, 2e)$ cross section as previously quoted.

We turn now to a discussion of differential cross sections for two-photon double ionization. The single-differential cross section for two-photon double ionization may be written as

$$\sigma(2\gamma, 2e) = \int_0^{\pi/2} \frac{d\sigma}{d\alpha}\, d\alpha = \int_0^{E} \frac{d\sigma}{dE_1}\, dE_1\,, \quad (4)$$

where $E_1 = \frac{k_1^2}{2}$, E is the excess photon energy given by $E = \frac{k_1^2}{2} + \frac{k_2^2}{2}$, and we write the differential cross section in hyperspherical angle α as

$$\frac{d\sigma}{d\alpha} = \left(\frac{\omega}{I}\right)^2 \frac{1}{\tau}\frac{2}{\pi}\int_0^{\infty} dk_1 \frac{2}{\pi}\int_0^{\infty} dk_2 \times \delta\left[\alpha - \tan^{-1}\left(\frac{k_2}{k_1}\right)\right] \times \sum_{L=0,2}\sum_{l_1,l_2} |P^{{}^1L}_{l_1 l_2}(k_1, k_2, t)|^2. \quad (5)$$

Our single-differential cross section for two-photon double ionization of helium at a photon energy of 45 eV is shown in Fig. 2 at intensities of 10^{14} and 10^{15} $\mathrm{W\,cm^{-2}}$. As expected, the area enclosed by the single-differential cross section gives the total integral cross section. The lower single-differential cross section at the higher intensity reflects the lower total-integral cross section at higher intensities. We note that the form of the single-differential cross section is similar in most respects to that obtained from $(\gamma, 2e)$ calculations for helium [6]. The shape is broadly concave, but in this case, we see a slight peak in the single-differential cross section at equal energy sharing $E_1 = E_2$. Although this peak persists in the cross section even when more $l_1 l_2$ pairs are added to the partial waves, we do not believe that it is physical, as the single differential cross section is seen to oscillate slightly as a function of the pulse length of the laser.

In a similar manner, we can define the triple-differential cross section as

$$\frac{d^3\sigma}{d\alpha\, d\Omega_1 d\Omega_2} = \left(\frac{\omega}{I}\right)^2 \frac{1}{\tau}\frac{2}{\pi}\int_0^{\infty} dk_1 \frac{2}{\pi}\int_0^{\infty} dk_2\, \delta\left[\alpha - \tan^{-1}\left(\frac{k_2}{k_1}\right)\right] \times \left|\sum_{L=0,2}\sum_{l_1,l_2} (-i)^{l_1+l_2} e^{i(\sigma_{l_1}+\sigma_{l_2})} P^{{}^1L}_{l_1 l_2}(k_1, k_2, t) Y^{{}^1L}_{l_1 l_2}(\hat{k}_1, \hat{k}_2)\right|^2, \quad (6)$$

where the momentum space amplitudes are weighted by the appropriate Coulomb phases σ_l since we project onto products of Coulomb radial waves, and $Y^{{}^1L}_{l_1 l_2}(\hat{k}_1, \hat{k}_2)$ denotes the standard expression for coupled spherical harmonics. Triple differential cross sections for the two-photon double ionization of helium with a photon energy of 45 eV are shown in Fig. 3, for coplanar geometry, for equal energy sharing between the electrons, for four values of θ_1 as shown, and over a range of θ_2. Because of the different angular momenta values of the final-state partial waves (^1S and ^1D), the triple-differential cross sections are quite different in shape from the corresponding $(\gamma, 2e)$ case,

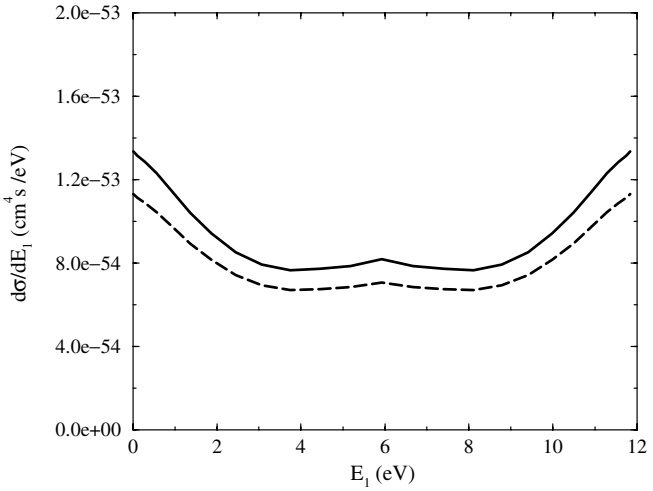

FIG. 2. Single differential cross section for helium for a photon energy of 45 eV, over a range of E_1, the energy of the first ejected electron. The solid line denotes the cross section at 10^{14} W cm^{-2}, and the dashed line denotes the cross section at 10^{15} W cm^{-2}.

where the final state is a ^1P partial wave. As expected, the shape and magnitude of the triple differential cross section is dominated by the $Y_{pp}^{^1L}$ $(L = 0, 2)$ channel, since this path is the largest contribution to the total integral cross section, as previously discussed. We plot the cross sections for θ_2 from 0° to 360° in order to show the peak in the cross section near $\theta_2 = 180°$ evident at all values of θ_1.

The large peak at $\theta_2 = 180°$ in the cross section for $\theta_1 = 0°$ is to be expected as it represents the "back-to-back" ejection of the two electrons, which is the most probable process due to electron-electron correlation. This peak is evident at the other values of θ_1, and its position

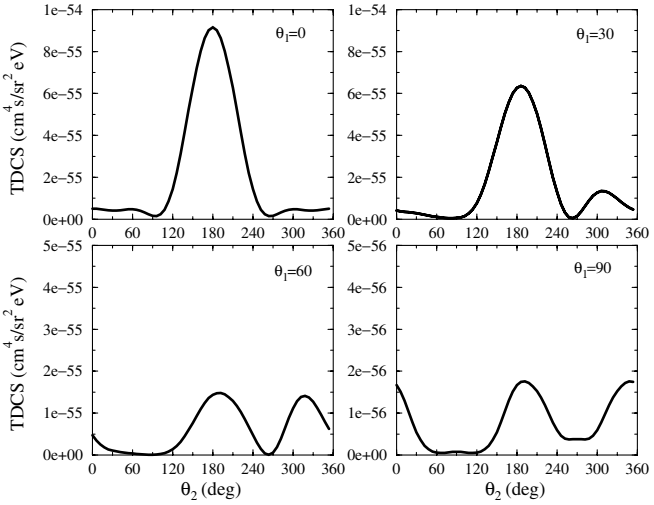

FIG. 3. Triple differential cross section for helium, for a photon energy of 45 eV, for equal energy sharing between the electrons. The angle of the first ejected electron is as shown, and we present cross sections over a range of θ_2, the angle of the second ejected electron from 0° to 360°.

increases from 180° with larger values of θ_1. As θ_1 is increased, a second peak in the cross section develops at higher values of θ_2 above 300°. There is very little cross section in the forward scattering range $(0° < \theta_2 < 90°)$. It is clear also that the magnitude of the cross section drops quickly as θ_1 is increased. Integration over all energies and angles of the electrons produces the total integral cross section as required.

In summary, we have presented time-dependent calculations of the total-integral, single-differential, and triple-differential cross sections for the two-photon double ionization of helium at a photon frequency of 45 eV. Calculations of the total-integral cross section for both the $(2\gamma, e)$ and $(2\gamma, 2e)$ processes are much higher than the only other calculations [9,10]. This is due to a core-excited resonance enhancement, in an energy range previously thought to access only the direct two-photon ionization process. Single- and triple-differential cross sections are also presented at a photon energy of 45 eV, although for these there are no calculations or measurements with which to compare. It is hoped that these calculations will provide a benchmark for future experiments.

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