Calculation of double photoionization of helium using the convergent close-coupling method

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We use the convergent close-coupling formalism to calculate the double-to-single-photoionization cross-section ratio in helium for photon energies from threshold up to 400 eV. Our results for the velocity and acceleration forms of the electromagnetic interaction operator are very close, and lie in between the measurements of Dörner *et al.* [Phys. Rev. Lett. **76**, 2654 (1996)] and Levin *et al.* [Phys. Rev. Lett. **76**, 1220 (1996)]. [S1050-2947(96)50508-1]

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Helium double photoionization is a fundamental problem and, as such, has been at the forefront of theoretical and experimental atomic physics for the past 30 years. Much of the attention has been focused on the ratio of the double-to-single-photoionization cross sections $R=\sigma^{2+}/\sigma^{+}$ studied as a function of the photon energy ω. Although this ratio is now well established in the asymptotic region of large photon energies of several keV, there still exists uncertainty in this ratio at the low-to-intermediate photon energies from the double-photoionization threshold up to several hundred eV. It is generally agreed that $R(\omega)$ has a broad maximum at a photon energy of $\simeq$200 eV. However, the value of R_{max} varies substantially in both available theory and experiment.

The latest experimental undertakings by Dörner *et al.* [1] and Levin *et al.* [2] were aimed at eliminating any uncertainty in the value of $R(\omega)$. However, these most recent, and presumably most accurate, measurements produced conflicting results. Dörner *et al.* [1] reported systematically lower $R(\omega)$ with $R_{max}=3.5\%$ as opposed to the value of 4.0% by Levin *et al.* [2]. Latest theoretical data [3–8] also vary greatly depending on the method used by the authors and the gauge (length, velocity, or acceleration) chosen for the electromagnetic interaction. This uncertainty in the experimental and theoretical results gives us an incentive to use an alternative approach to the helium double ionization by applying the convergent close-coupling (CCC) method introduced for the e-H system by Bray and Stelbovics [9], and extended to hydrogenic atoms and ions by Bray [10].

In our formalism we consider the double photoionization as a two-stage process. Stage 1 is single ionization that is followed by electron-impact ionization of the resultant He^+ ion. The one-electron states of the He^+ ion are described by a Laguerre square-integrable basis with excitation of the positive-energy pseudostates corresponding to ionization of the He^+ ion and therefore the double ionized channels for photon-impact ionization of helium. The CCC method has been extensively tested for the e-He^+ scattering system, and has yielded quantitative agreement with the measurements of electron-impact total ionization [11], suggesting that we should be able to obtain accurate double-photoionization cross sections for helium. The primary limitation on the accuracy of our approach is a fair description of the helium-atom single photoionization leading to the He^+-ion ground and various excited states. We believe this can be achieved by employing a highly correlated helium ground-state wave function together with an accurate solution of the e-He^+ excitation and ionization problem using the CCC method.

In the present study, as in our previous work on the helium ionization with excitation [12], we use a multiconfiguration Hartree-Fock wave function for the helium ground state. As a variational wave function, it is mostly accurate near the origin and at intermediate distances that contribute most significantly to the ground-state energy. The accuracy of this wave function is marginal at large distances that are negligible in terms of the energy. This puts a certain limitation on the use of the length form of the electromagnetic interaction operator, which enhances the area far from the origin. So, we restrict ourselves to the velocity and acceleration gauges only.

The idea of using a discretized continuum for the description of the helium double photoionization is not a new one. It has been used previously by Meyer and Greene [7] in the R-matrix formalism, and by Tang and Shimamura [5] within the hyperspherical coordinate space close-coupling method. What makes the present work different is the implementation of the discretization procedure that is realized within the momentum space close-coupling formalism.

We use the multichannel expansion for the final-state wave function of the system He^+ ion plus ejected electron:

$$|\Psi_j^{(-)}(\boldsymbol{k}_b)\rangle = |j\boldsymbol{k}_b^{(-)}\rangle + \sum_i \int d^3k \frac{\langle \boldsymbol{k}^{(+)} i|T|j\boldsymbol{k}_b^{(-)}\rangle}{E-k^2/2-\epsilon_i+i0}|i\boldsymbol{k}^{(+)}\rangle, \tag{1}$$

with boundary conditions corresponding to an outgoing wave in a given channel $|j\boldsymbol{k}_b^{(-)}\rangle$ and incoming waves in all other channels $|i\boldsymbol{k}^{(+)}\rangle$. Here $E=k_b^2/2+\epsilon_j$ is the final-state energy. The channel wave function $|j\boldsymbol{k}_b^{(-)}\rangle$ is the product of a one-electron orbital ϕ_j, obtained by diagonalizing the He^+ Hamiltonian in a Laguerre basis, and a Coulomb $Z=1$ outgoing wave $\chi^{(-)}(\boldsymbol{k}_b)$. The half off-shell T matrix in Eq. (1) is the solution of the corresponding Lippmann-Schwinger integral equation [10].

The photoionization cross section, as a function of the photon energy ω, corresponding to a particular bound electron state j, is given by [13]

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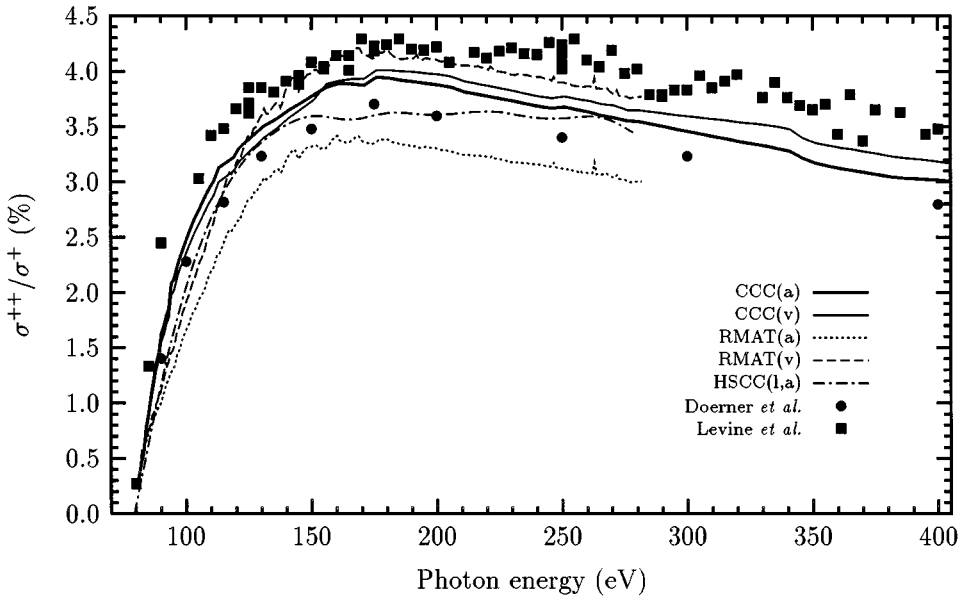

FIG. 1. Ratio of double-to-single photoionization cross sections of the ground state of helium. The present calculations are denoted by CCC. Those denoted by RMAT and HSCC are the R-matrix calculations of Meyer and Green [7] and the hyperspherical close-coupling calculations of Tang and Shimamura [5], respectively. The parentheses indicate the gauge. The measurements are by Dörner *et al.* [1] and Levin *et al.* [2].

$$\sigma_j(\omega)=\frac{4\pi^2}{\omega c}\sum_{m_j}\int d^3k_b|\langle\Psi_j^{(-)}(\boldsymbol{k}_b)|\mathcal{D}|\Psi_0\rangle|^2 \times\delta(\omega-E_f+E_0), \tag{2}$$

where $c\simeq 137$ is the speed of light in atomic units.

For the ground-state wave function Ψ_0 in Eq. (2) we used a seven-term multiconfiguration Hartree-Fock expansion [12]. This gave us a reasonably good account of the ground-state correlations judging by the ground-state energy $E_0=-2.90181$ a.u., recovering 95.4% of the correlational energy. Another indication of the quality of our ground-state wave function is the limit $R_{\omega\to\infty}$ at large photon energies. According to Åberg [14], this limit depends only on the ground-state wave function. Our ratio $R_{\omega\to\infty}=1.67\%$ is in a good agreement with the measurement of Spielberger *et al.* [15], who carefully separated the Compton scattering from the photoionization and reported the value of 1.72%.

The dipole electromagnetic operator $\mathcal{D}$ can be written in one of the following forms commonly known as length, velocity, and acceleration [13]:

$$\mathcal{D}^r=\omega(z_1+z_2),\quad \mathcal{D}^\nabla=\nabla_{z_1}+\nabla_{z_2},\quad \mathcal{D}^{\dot\nabla}=\frac{2}{\omega}\left(\frac{z_1}{r_1^3}+\frac{z_2}{r_2^3}\right). \tag{3}$$

Here we assume that the photon is polarized along the z axis. The length form enhances the large distance contribution to the radial integral of Eq. (2). As we mentioned above, the variational ground-state wave function is inaccurate in this region. So we deem the length form unreliable and do not present it. Incidentally, Meyer and Greene [7] discarded their length form calculation as well, though they argued that their final-state wave function is unreliable at large distances because of the coordinate space discretization procedure.

We separate the contribution from the final channels $|j\boldsymbol{k}_b^{(-)}\rangle$ into single and double ionization according to the energy of the ϵ_j, which is positive for the double ionized channels and negative for the singly ionized channels. We also ensure that the negative-energy state cross sections, contributions to the ionization plus excitation cross sections, are multiplied by the projection of the state onto the true target discrete subspace [16]. This way we have a relatively clear separation between the discrete and continuous spectrum of the e-He^+ excitation. As a first check of our calculations we find that our single-photoionization cross sections are identical in the three gauges and are in absolute agreement with the most recent data of Samson *et al.* [17].

The results of our calculations for $R(\omega)$ are presented in the figure along with the latest experimental data of Dörner *et al.* [1] and Levin *et al.* [2]. For clarity of presentation we only compare with other theories that use a similar foundation for these calculations. These are the eigenchannel R-matrix method (RMAT) of Meyer and Greene [7] and the hyperspherical close-coupling approach of Tang and Shimamura [5]. Comparison with other experiments and theories may be found in these references. Convergence in our calculations was achieved by performing 52-state calculations consisting of 13 states for each He^+ target-space orbital angular momentum $\ell=0,\ldots,3$. Some unphysical oscillation is visible in our results, which is due to pseudoresonances associated with the pseudothresholds. The calculations have been performed at nearly 100 energy points suitably distributed over the presented energy range. Note that no averaging of the CCC results is undertaken; we simply rely on taking sufficiently large Laguerre basis sizes to ensure that pseudoresonances are of sufficiently small magnitude [18].

Our results, both in the velocity and acceleration forms, are in between the two sets of most recent experimental data, with a difference of around 10% from either set. The RMAT calculation of Meyer and Greene [7] in the velocity gauge is close to our velocity gauge result. However, their acceleration form result is substantially lower than ours. This R-matrix method has also demonstrated an ability to obtain accurate ionization cross sections (a few eV above threshold) in the e-He^+ problem [19], and so we suspect that the source of the discrepancy may be the choice of the ground He state. This supposition could be readily tested by using identical

ground He states in both calculations. The hyperspherical close-coupling results of Tang and Shimamura [5] give a different shape to those obtained from the other calculations, but also generally lie in between the two sets of measurements. Given that both of their length and acceleration forms yielded much the same results, and that we are in agreement at the higher energies, we suspect that the difference may be due to the solution of the e-He^+ part of the problem.

In summary, it appears that the ratio of double-to-single photoionization of helium is still unresolved either experimentally or theoretically to a satisfactory accuracy. On the theoretical front, we need to improve the ground-state description to ensure that all gauges give similar results. Furthermore, it would be helpful for theorists to establish a definitive set of cross sections for e-He^+ excitation and ionization in the ^1P wave. We believe the CCC method is able to solve this problem very accurately, and these results may be used to test the accuracy of other theories before application to photoionization. Apart from attempting to improve the description of our He ground state, we will apply the CCC theory to the calculation of the $(\gamma,2e)$ differential cross sections.

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