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Double photoionization of helium using R-matrix methods

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The eigenchannel R-matrix method for single photoionization is extended to describe a class of two-electron escape processes. We apply this approach to calculate the double photoionization of helium by single-photon absorption for photon energies in the range 80–280 eV. Pseudoresonances in the double continuum are eliminated by performing a Gailitis average and by averaging the final spectrum over the size of the R-matrix box. Calculations are presented in both the velocity and acceleration gauges. The ratio of double- and single-photoionization cross sections, a key parameter for characterizing electron correlations, is compared to existing theoretical and experimental values.

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Difficulties in the description of the two-electron continuum have plagued most theoretical techniques for decades. In the standard close-coupling approach, the double continuum is represented by an integral over an uncountable set of continuum states of the single-electron ion, multiplied by corresponding unknown wave functions for the second electron. Numerical determination of this uncountable set of wave functions would be exceedingly difficult. Most approximate treatments of double continuum processes have relied instead upon perturbative approaches, such as many-body perturbation theory, the Born approximation, or a distorted-wave Born-type approximation. Such methods have been shown to give realistic approximations to many double-continuum cross sections at sufficiently high energies. At the same time, methods that use a discrete representation of all possible excitation channels have enjoyed tremendous success at low energies where only single escape is energetically allowed. This includes for instance the R-matrix method, variants of the close-coupling method that use pseudostates, and the convergent close-coupling (CCC) approach. In the present paper we show that the eigenchannel R-matrix method is capable of describing a class of processes in the two-electron continuum, excluding energies very close to the threshold energy for double escape. Other coupled-channel methods have been similarly extended to treat double continua in recent years, notably the intermediate energy R-matrix method [1] and the CCC approach [2]. It is important to explore this difficult energy region, because of its intrinsic theoretical interest, and also because even processes involving single-electron escape could be adversely affected if the double-continuum regime is poorly represented. The contributions to the escaping electron flux from the single- and double-continuum regions are linked in some sense, e.g., through constraints such as the oscillator strength sum rule.

One sensitive probe of electron correlations in helium is the ratio R of double- and single-photoionization cross sections ($R \equiv \sigma^{2+}/\sigma^{+}$). Double photoionization could not occur in the absence of electron-electron correlations, at least at the order of first-order perturbation theory for the photon-atom interaction, as it is a sum of individual electron operators. Perturbative treatments of electron correlations rely on the assumption that one of the ionized electrons leaves the target area relatively rapidly. For this situation, in which the electron kinetic energies overwhelm the electron-electron interaction potential, the final-state correlation effects are minimal. The assumption that the escaping electrons have very different speeds is approximately valid for photon energies much larger than the ionization potential of the target, which is the energy range considered here.

Considerable effort has been expended to obtain values of the ratio R for helium in the low photon energy regime, both experimentally [3–9] and theoretically [10–14]. Many-body perturbation theory (MBPT) is applied in most of the theoretical calculations. The differences between various many-body calculations are due to differences in the choice of basis sets and in the methods used to estimate higher-order corrections. In this study, theoretical calculations of double-photoionization cross sections are performed by extending the eigenchannel R-matrix method. While this approach also faces difficulties when applied to the escape of two electrons, it should be informative to see results from a completely different formulation.

In R-matrix theory, configuration space is partitioned into two (or more) regions by a sphere of radius $r = r_0$, as shown in Fig. 1. The most complicated physics is included in the finite internal region (region I in Fig. 1), called the reaction volume. In the external region, electron-electron correlation effects are neglected, which is a good approximation for large enough values of r_0. Once the wave function is determined both inside and outside the reaction volume, the full solution is obtained by matching the logarithmic derivative of the wave function across the surface of the reaction volume and solving the Schrödinger equation in the exterior region.

For this study we adopt the eigenchannel R-matrix method, which calculates variationally a particular set of Schrödinger solutions; each solution has a constant normal logarithmic derivative across the surface of the reaction volume. The implementation of this approach used here starts from numerical solutions of the one-electron radial Schrödinger equation for He$^+$, calculated inside the reaction volume subject to the boundary condition that the radial wave function vanishes at the boundary $r = r_0$. This generates a set of (closed-type) solutions having a discrete energy spectrum. The lowest-energy states (with principal quantum numbers

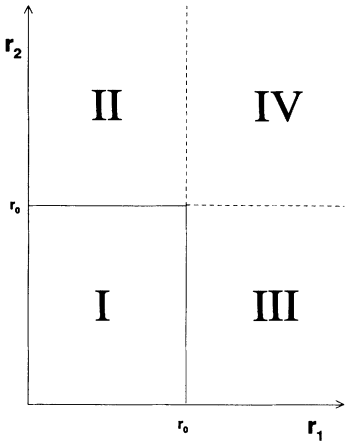

FIG. 1. Radial configuration space for two electrons used in R-matrix theory. Region I represents the reaction volume, regions II and III represent single electron escape, and region IV represents the double continuum.

$n \leq 3$) are very similar to the ordinary physical bound states of He$^+$, since they vanish at large distances anyway. These solutions are next used to construct a two-electron basis set. The two-electron trial wave function at each final-state continuum energy E is written as a linear combination of these basis functions, which now describes a continuum energy state. A variational method described elsewhere [15] is used to solve for the optimal coefficients in the basis set expansion of the trial wave function. We neglect correlation effects outside the reaction volume.

If only closed-type one-electron orbitals were used to construct two-electron trial functions, no electrons would be able to escape from the reaction volume, because the wave function would be exactly zero everywhere on its surface. Accordingly we introduce "open-type" orbitals as in Ref. [15], which are also eigenfunctions of the He$^+$ Hamiltonian but which are nonvanishing at $r = r_0$. Each "channel" in our problem is represented by one closed-type state of He$^+$ for the "inner" electron, multiplied by each member of the complete set of closed-type basis functions for a given outer electron partial wave, and by two open-type basis functions for that partial wave as well. In the present study, 22 closed-type basis functions and two open basis functions are included for each channel. Note, however, that there are no two-electron basis functions included that involve open-type orbitals for each electron, which one might think would be needed to represent double escape from the reaction volume. Wave functions in the outer region are approximated by a linear combination of Coulomb functions with unit charge for the outer electron, multiplied by a He$^+$ eigenstate for the inner electron, followed by antisymmetrization. The channels included in the present photoionization calculations are $ns\epsilon p$, $np\epsilon s$, $np\epsilon d$, and $nd\epsilon p$. A numerical test showed that f waves have a relatively minor effect on both the single- and double-photoionization cross sections. Approximately one thousand total two-electron configurations are used to describe the final states reached in single and double photoionization.

Previous applications of eigenchannel R-matrix methods have treated the escape of a single electron from the reaction volume (regions II and III in Fig. 1). The boundary conditions imposed by these calculations do not allow for the possibility of direct electron escape into the double-continuum region of configuration space (region IV in Fig. 1). The eigenchannel R-matrix method is extended in this study to obtain double-photoionization cross sections for helium, using a point of view similar to that applied by Robicheaux *et al.* [16] to the double continuum of $H + e + e$. We accomplish this by reinterpreting the meaning of the remaining He$^+$ bound-state electron in our single photoionization treatment, despite the fact that we actually impose the same (single-escape) boundary conditions. The central idea is to recognize that if the complete set of He$^+$ closed-type eigenfunctions in the R-matrix box is used to represent the "inner" electron in a close-coupling expansion, then those eigenfunctions that lie at positive energies (relative to the double escape threshold, 78.98 eV for the case of helium) must physically represent a (discretized) continuum state of He$^+$. Consequently we can simply interpret all flux escaping in such channels with positive-energy thresholds as flux that contributes to double photoionization. All flux escaping in channels at negative energies is similarly interpreted as being associated with single photoionization, even if the negative-energy He$^+$ box eigenstate does not coincide with a physical He$^+$ eigenstate. Physically, this corresponds to the assumption that most of the photon's energy is absorbed by one of the electrons, leaving the He$^+$ ion in either a negative-energy bound state or a discretized positive-energy continuum state. Samson *et al.* [3] have achieved some success using a model based on this qualitative picture.

One well-known consequence of using a discrete spectrum of positive-energy He$^+$ "ionization thresholds" is the appearance of pseudoresonances in the double-ionization cross section. These pseudoresonances arise simply as an artifact of the artificial boundary conditions imposed on the finite reaction volume; they have no real physical meaning. It is desired to eliminate these pseudoresonances in the double continuum to obtain a smooth cross section. This can be accomplished by implementing the Gailitis [17,18] and box averaging techniques (the "box" is another term for the reaction volume). The Gailitis technique is useful for averaging over resonances near channel thresholds. The technique is easily implemented in the multichannel quantum defect theory (MQDT) part of the calculation, simply by treating a closed ionization channel (which has $E_i > E$) as though it were open when solving the MQDT equations for the photoionization cross section. In this analysis, Gailitis averaging is performed from 2 eV below each He$^+$ threshold all the way up to threshold. This automatically removes all or most of the Rydberg resonances converging to that particular ionization threshold. To implement box averaging in this study, the cross sections are calculated for five different box sizes (reaction volume radii) in the range 10–12 atomic units, after which they are averaged to obtain the final cross sections reported here.

Calculations of double-photoionization cross sections have been attempted in the length, velocity, and acceleration gauges. The length gauge calculations give unreasonably high values of double-photoionization cross sections for our extended R-matrix method, while the results obtained using

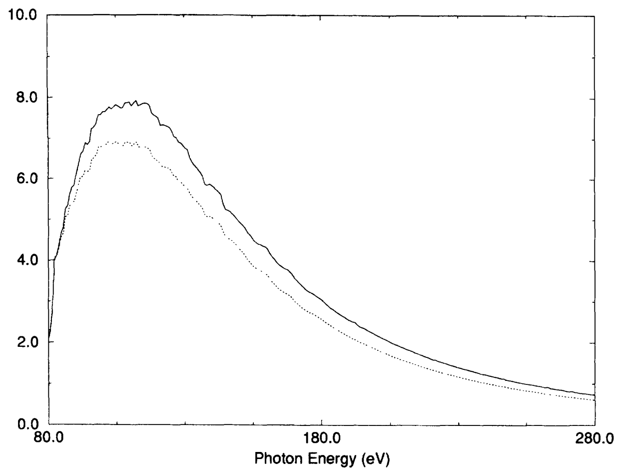

FIG. 2. The averaged double-photoionization cross section calculated for helium is shown as a function of photon energy. The solid curve represents calculations performed in the velocity gauge, while the dotted curve represents calculations performed in the acceleration gauge.

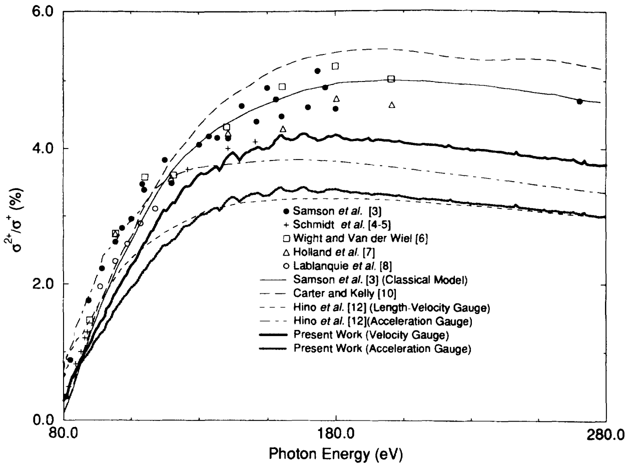

FIG. 3. The ratio of double- and single-photoionization cross sections of helium is shown versus the photon energy. The individual symbols represent experimental measurements, while the lines represent theoretical calculations.

the velocity and acceleration gauges are stable, consistent with each other, and in reasonable agreement with previous calculations. In view of the fact that the length gauge weights the large-r part of the wave function the largest, which is precisely where our unphysical boundary conditions probably cause our variational solutions to be the least accurate, the large errors in the length gauge calculation might have been anticipated in advance. Consequently we do not report the length gauge results here. The averaged double-photoionization cross sections obtained in the velocity and acceleration gauges are shown in Fig. 2. There are no signs of artificial pseudoresonances, although the averaging schemes still leave some irregularities in the energy dependence of the cross sections which could presumably be eliminated by averaging over more values of the box size r_0.

Our calculated values for the ratio R of the single- and double-photoionization cross sections are compared in Fig. 3 to several existing experimental and theoretical values. The curve generated by Samson's simple classical model agrees well with the available experimental data, despite the fact that the model fails to account for the symmetry of the final state reached in the photoionization process. The MBPT calculations of Carter and Kelly [10] and of Hino *et al.* [12] represent upper and lower limits on the theoretical values shown in Fig. 3. Our eigenchannel R-matrix results agree more closely with the results of Hino *et al.*, although the ordering of the velocity and acceleration gauge curves is reversed in these two sets of calculations. Pan and Kelly [19] have calculated a maximum R value of almost 5%, which is somewhat larger than the maximum value obtained in both of our calculations. Proulx and Shakeshaft's [13] cross section calculations, which cover an energy range from the double-ionization threshold to roughly 60 eV above threshold, agree well with our velocity gauge calculations. Our results are slightly smaller than the experimental values. This discrepancy is comparable to our theoretical "error bar," which we take to be the difference between our velocity and acceleration gauge calculations.

Our study has applied the eigenchannel R-matrix theory to calculate double-photoionization cross sections of helium. Other variants of R-matrix theory have been used to describe processes in which electron-electron correlations are important. Scholz *et al.* [20] have presented a modified R-matrix theory to address electron-atom and electron-molecule scattering at intermediate energies, including the two-electron continuum. In their approach, the internal region of configuration space is further divided into a number of subregions to treat inelastic effects. A T-matrix energy averaging technique, which involves continuation to complex energies, is implemented to eliminate pseudoresonances and obtain smooth cross sections. Callaway and Oza [21] have applied a variational pseudostate method to address electron-electron correlations. In addition to the basis states of the close-coupling method, they introduce a set of discrete states to represent the atomic continuum. Cross sections calculated for electron-hydrogen scattering using both of these methods are somewhat in discrepancy with experimental results, illustrating the difficulty involved in treating correlation effects. More recently, Bray and Stelbovics [22] have calculated cross sections using the convergent close-coupling formalism; their results agree more closely with existing experimental data.

It may not be obvious why the present calculations using artificial boundary conditions at $r=r_0$ should properly represent the flux leading to double ionization. In particular, we essentially neglect the wave function in region IV of Fig. 1 initially, although our reinterpretation of positive-energy partial cross sections amounts to allowing the inner electron to eventually escape to infinity through region IV. Our view is that by describing nonperturbatively the electron correlation and exchange effects within the reaction volume (region I), we properly describe the initial outward propagation of the electron pair that is induced by the photon absorption. The electrons are mutually deflected as they continue outward to infinity beyond the reaction zone, and this deflection is not accurately described by our approach, but we nevertheless expect that the amount of flux leading to double escape will not differ greatly from our prediction. However, because we do not accurately describe the physics of the two-electron escape beyond the reaction volume, we cannot expect our approach to give a realistic description of other details of the double-photoionization process such as the photoelectron distributions in energy and angle.

In conclusion, we have applied an alternative approach to calculate double-photoionization cross sections of helium. The R-matrix method is extended by reinterpreting the meaning of discrete He^+ states having a positive-energy eigenvalue. We eliminate pseudoresonances in the double continuum by implementing Gailitis and box averaging to obtain smooth ionization cross sections. Although a discrepancy in the ratio of double and single photoionization exists between our calculations and experimental results, this discrepancy is comparable to the uncertainty in our results. At much higher photon energies, electrons are ejected primarily by Compton scattering processes [23,24], which can in principle be included in this theoretical method.

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