RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 52, NUMBER 5 NOVEMBER 1995

Double photoionization of helium at low photon energies

Jian-Zhi Tang* and Isao Shimamura†

The Institute of Physical and Chemical Research (RIKEN), Hirosawa 2-1, Wako-shi, Saitama 351-01, Japan

(Received 18 April 1995)

The cross sections for single and double photoionization of helium at energies from the threshold of double ionization to 280 eV are calculated by combining the previously developed hyperspherical close-coupling method with a discretization procedure for the continuum spectrum of He^+. No pseudoresonances due to the discretization are found. Both the length and acceleration forms of the dipole operator are used, and the results hardly depend on these different gauges. The results are compared with previous theories and experiments.

PACS number(s): 32.80.Fb, 31.15.Ja, 31.25.−v

The theoretical calculation of atomic processes involving two electrons in the continuum, or double continuum, is one of the most difficult problems in atomic physics. An example is the double photoionization of helium, in which two electrons are emitted. Double ionization by a single photon could not occur if the electron-electron correlation were absent, since the photon-atom interaction is the sum of one-electron operators. Especially at low photon energies, the two electrons in the continuum are strongly correlated. Therefore, this correlation effect must be taken into account accurately, and must be difficult to treat by perturbation theory. On the other hand, the cross sections σ^{++} for double ionization are much smaller than those for single ionization, σ^{+}, and the determination of σ^{++} by subtracting σ^{+} from the total photoionization cross section σ_t is bound to be plagued by the loss of accuracy.

The many-body perturbation theory (MBPT) treats the electron-electron correlation perturbatively, and has been successful in the calculations of σ^{++} for He at high photon energies ω [1–3]. The cross sections at very high ω may be calculated by using an asymptotic-ω formulation [4,5]. A recently proposed method for calculating σ^{++} for low ω [6] uses a trial function that is simple at short electronic distances from the nucleus but that satisfies the asymptotic boundary condition that is correct in the stationary-phase limit. Another method circumvents the correct asymptotic boundary condition for two outgoing electrons, invoking projection operators to separate double ionization from single ionization [7].

There is a class of methods for single photoionization and electron-impact excitation that describes the asymptotic wave function in a close-coupling-type expansion form. If one uses bound-state-like pseudostates to represent the target continuum states by discretization, one may extract σ^{++} from calculations similar to those for single photoionization. A method combining this technique with the eigenchannel R-matrix method was applied recently to low-ω σ^{++} for He with some success [8]. However, the spurious oscillations of σ^{++} found as a function of ω had to be smoothed out artificially.

In this paper, we combine the hyperspherical close-coupling (HSCC) method [9] with the technique of pseudostates for discretizing the continuum He^+ states, and calculate σ^{++} for He from the threshold to 280 eV. This method is nonperturbative and takes full account of the electron-electron correlation effect. The calculated σ^{++} seems to be free from spurious oscillations, and is found to be almost gauge independent, which is a feature absent in previous theoretical σ^{++}.

For the two-electron wave function of He expressed as $\Psi(\mathbf{r}_1,\mathbf{r}_2)=(R^{5/2}\sin\alpha\cos\alpha)^{-1}\Phi(R,\alpha,\Omega)$ in terms of the hyperspherical coordinates, the Schrödinger equation may be written as

$$\left(-\frac{1}{2}\frac{\partial^2}{\partial R^2}+\frac{H_{\rm ad}}{R^2}-E\right)\Phi(R,\alpha,\Omega)=0, \tag{1}$$

where $R=(r_1^2+r_2^2)^{1/2}$ is the hyperradius, $\alpha=\arctan(r_2/r_1)$ is the hyperangle, and Ω denotes collectively the four angles $(\hat{\mathbf{r}}_1,\hat{\mathbf{r}}_2)$. The adiabatic Hamiltonian $H_{\rm ad}$ for a fixed R, written in the hyperspherical coordinates, may be found in Ref. [9]. The eigenfunctions of $H_{\rm ad}$ define hyperspherical adiabatic channel functions [10].

The configuration space is divided into two regions, the inner region ($R<R_M$) and the outer or asymptotic region ($R>R_M$). In the inner region, the wave function $\Phi^{(\rm in)}$ is expanded as

$$\Phi^{(\rm in)}(R,\alpha,\Omega)=\sum_{\mu}F_{\mu}^{(E)}(R)\phi_{\mu}(\alpha,\Omega;R) \tag{2}$$

in terms of diabatic basis functions ϕ_μ defined in Refs. [9,11]. Substituting Eq. (2) into the Schrödinger equation (1), one obtains close-coupling equations for $F_\mu^{(E)}(R)$. These equations may be solved by well-established computational methods, and independent sets of solutions at the boundary $R=R_M$ may be obtained; further details may be found elsewhere [9]. Extensive studies of two-electron systems [9,11–15] have shown that the strong electron-electron correlations in the inner region for processes with one electron in continuum are well described by the expansion in terms of the rapidly convergent basis functions $\{\phi_\mu\}$, which are set up numerically and are independent of the total energy E. This suggests that the same HSCC method should also be power-

*Electronic address: tang@rikvax.riken.go.jp

†Electronic address: shimamura@rikvax.riken.go.jp

ful for describing the electron-electron correlations in the inner region for double photoionization in the low-energy region.

To extract the information on double photoionization, we use a discretization method [8,16] in the outer region $R > R_M$. In other words, the double continuum or the two-electron continuum is represented by an electron interacting with a He^+ ion in the continuum that is represented by a discretized pseudostate. More specifically, the wave function in the outer region is expressed as

$$\Psi_j^{(\mathrm{out})}(\mathbf{r}_1,\mathbf{r}_2)=r_>^{-1}\sum_i \varphi_i(\mathbf{r}_<,\hat{\mathbf{r}}_>)[f_i(r_>)\delta_{ij}-g_i(r_>)K_{ij}], \tag{3}$$

where $r_< = \min(r_1,r_2)$ and $r_> = \max(r_1,r_2)$. The functions f_i and g_i are regular and irregular Coulomb functions for open channels, linear combinations of exponentially decaying and growing functions for weakly closed channels, and an exponentially decaying function and a null function ($\equiv 0$) for strongly closed channels; see Ref. [9] for details. The function φ_i is written as

$$\varphi_i = R_{\tilde{n}l}(r_<)\mathscr{Y}_{LM}(\hat{\mathbf{r}}_1,\hat{\mathbf{r}}_2) \tag{4}$$

in terms of an eigenfunction $\mathscr{Y}_{LM}$ of the square of the total orbital angular momentum and its projection onto a quantization axis. The function $R_{\tilde{n}l}(r_<)$ is an eigenfunction of the radial Schrödinger equation of the hydrogenic He^+ ion subject to the boundary condition that it vanish at a boundary $r_< = r_M = R_M/\sqrt{2}$, i.e.,

$$R_{\tilde{n}l}(r_M)=0. \tag{5}$$

Under this condition, the He^+ levels are completely discretized. One may assume that the eigenstates with eigenvalues $\epsilon_{\tilde{n}l} < 0$ represent a spectrum of true bound states; some lowest eigenvalues are expected to be good approximations to true bound-state energies, and other, higher eigenvalues represent collectively a range of the bound-state spectrum. Positive eigenvalues may be assumed to represent the continuum. In other words, one may interpret the channels with $\epsilon_{\tilde{n}l} < 0$ in the asymptotic solutions (3) as representing single ionization of He and the channels with $\epsilon_{\tilde{n}l} > 0$ as representing double ionization [8,16].

We calculate the K matrix by smoothly matching the solutions obtained in the inner region with the outer-region solutions at $R = R_M$, using a two-dimensional matching procedure [11]. The wave functions of the final states of single and double photoionization are thus set up. The cross sections may be obtained from the matrix elements of the dipole operator between those final states and the initial state, the latter being calculated by the conventional HSCC method [11]. One may use any of the three forms, or gauges, of the dipole operator, namely, the length form, the velocity form, and the acceleration form. These forms weigh different spatial regions of the wave functions, but should give an identical result if the wave functions are exact. Actually the exact helium wave functions of the initial and final states are unknown, and the cross sections depend necessarily on the choice of the gauge.

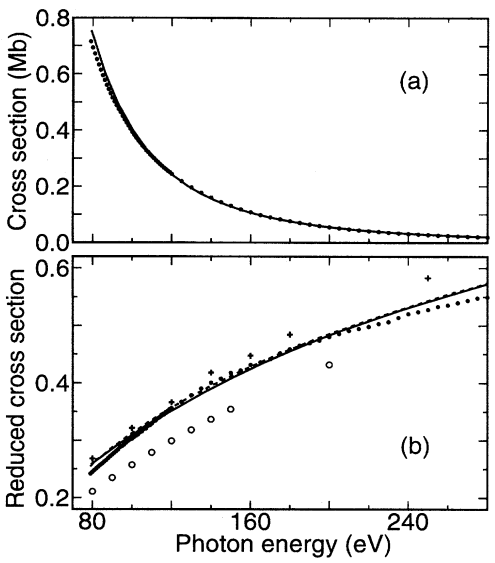

FIG. 1. Single-photoionization cross sections for He. (a) Solid curve: present results. Solid circles: experiment [18]. (b) Reduced cross sections σ_r^+ of Eq. (7). Solid curve: present results. Broken curve: eigenchannel R-matrix method [8]. ●: experiment [18]. +: B-spline method [19]. ○: many-body perturbation theory [2].

In the present calculation, 175 diabatic basis functions ϕ_μ were used in the expansion (2) to construct the final continuum states, and the matching procedure was performed at $R_M = 30$ a.u. Both the matching radius and the number of basis functions were varied in test calculations (several R_M between 20 and 70 a.u. being tested), and the convergence in σ^{++} within several percent and in σ^+ within three digits was confirmed. The good convergence of σ^+ even with R_M as small as 30 a.u. is due to the dominance of σ^+ by the partial cross section for the production of $He^+(1s)$, whose wave function is well localized and is almost exactly reproduced under the boundary condition (5). Both the length and acceleration forms of the dipole operator were used and the results were confirmed to be almost the same, as seen below; the velocity form was not used because of the numerical difficulty in the hyperspherical-coordinate calculations in this form [17].

The single-ionization cross section calculated using the acceleration form is shown in Fig. 1(a) and is compared with the results of recent experiment [18]. We find a good agreement, except near the double ionization threshold of 79.0 eV, where the present results are slightly higher than the experimental data.

To demonstrate the difference between various theoretical results, we define a reduced cross section σ_r^+ as follows. The single-ionization cross section σ^+ has an asymptotic form

$$\sigma^+ \sim \sigma_h \equiv \sigma_0 \omega^{-3.5} \tag{6}$$

at high photon energies ω (higher than a few keV), where σ_0 is a constant independent of ω. The cross section deviates from the $\omega^{-3.5}$ scaling law at low ω. The reduced cross section is defined by

$$\sigma_r^+ = \sigma^+/\sigma_h \tag{7}$$

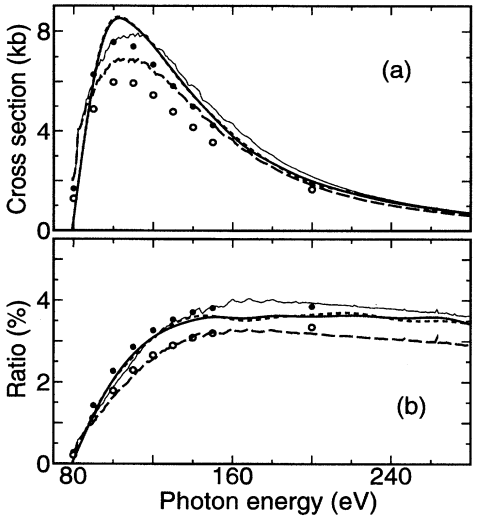

FIG. 2. (a) Double-photoionization cross sections for He. (b) The ratio of the cross sections for double to single photoionization. Solid and short-broken curves: present results from the acceleration and length forms of the dipole operator. Long-broken and fine solid curves with wiggles: eigenchannel R-matrix method, acceleration and velocity forms [8]. ● and ○: many-body perturbation theory, acceleration and length forms [2].

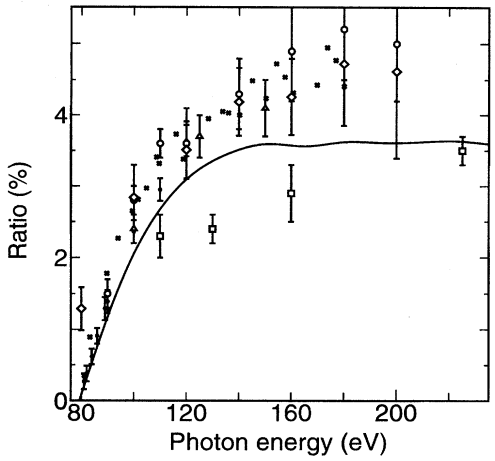

FIG. 3. Comparison of the calculated ratio (solid curve) of the cross sections for double to single photoionization with experimental data of Ref. [20] (●), Ref. [21] (□), Ref. [22] (△), Ref. [23] (○), Ref. [24] (◇), and Ref. [25] (×).

at all values of ω, and tends to 1 in the high-ω limit. The reduced cross sections from the present calculation and some recent calculations [2,8,19] are compared in Fig. 1(b); a detailed comparison of the cross sections from other theories and experiments is found in Ref. [18]. The present calculation reproduces almost exactly the results of the eigenchannel R-matrix calculation of Ref. [8]. The cross section calculated from multichannel continuum states by a least-squares approach in a B-spline basis set [19] is systematically higher than the experiment and the present results, while the result of the MBPT calculation [2] is systematically lower than the experiment and other theories.

Figure 2(a) compares the calculated σ^{++} with the results of the MBPT calculation [2] and the eigenchannel R-matrix calculation [8]. The present theory is seen to produce an length-form result (weighing the large-R part of the wave functions the largest) and an acceleration-form result (weighing the small-R part the largest) that are in good agreement with each other and that show no oscillations as functions of the photon energy ω. The results of the eigenchannel R-matrix method were obtained by smoothing out the spurious oscillations that resulted from the use of pseudostate basis functions; small oscillations are found even after the smoothing. These functions were defined by an artificial boundary condition similar to Eq. (5). The two-electron excitation configurations involving these pseudostate basis functions in the expansion of the inner-region wave function led to pseudoresonances in σ^{++}. The present method avoids the use of pseudostates in the inner region, and this appears to be the reason for the absence of spurious oscillations in σ^{++} in the present work.

The velocity-form and acceleration-form results of the eigenchannel R-matrix calculations differ from each other, and sandwich the present results between them at the higher energies in the figure, though both of them are smaller than the present results near the maximum. In fact, the length-form eigenchannel R-matrix calculations gave "unreasonably high values of double-photoionization cross sections" [8], which are therefore not reported in Ref. [8]. The good agreement between the present length-form and acceleration-form results is considered to stem from the adequate account of the strong electron-electron correlations in the inner region by the use of the hyperspherical diabatic-basis expansion. The length-form and acceleration-form results of MBPT calculations are significantly different from each other and both are smaller than the present results. This may imply that the perturbative treatment of the electron-electron correlations by the MBPT has not converged. It has been discussed [4] that the acceleration form, which weighs the near-nuclear part of the wave function the largest, is more reliable than the length form at high energies. Figure 2(a) appears to support this even at low energies, if we assume the present results to be accurate.

We note that an earlier MBPT calculation in Ref. [1] gave much larger σ^{++} than any of the results shown in Fig. 2(a). Recent calculations by using a simple trial function satisfying reasonable asymptotic boundary conditions [6] and by using projection operators to circumvent the boundary conditions [7] agree well with the present results.

The ratio σ^{++}/σ^{+} is usually measured in place of σ^{++}, since the absolute value of the latter is difficult to determine. This ratio is also considered to serve as a probe of electron correlations in the study of the physics of double photoionization. This ratio is shown in Fig. 2(b) and is compared with the results of other theories, corresponding to Fig. 2(a).

The discrepancy among different theories at the higher energies in Fig. 2 is emphasized more in Fig. 2(b) for the ratio σ^{++}/σ^{+} than in Fig. 2(a) for σ^{++} itself. On the other hand, the discrepancy in σ^{++}/σ^{+} becomes smaller at energies close to the maximum of σ^{++}. Thus, for example, the ratio obtained from the acceleration-form MBPT calculation

is close to and slightly larger than the present result near the maximum, though σ^{++} itself is considerably smaller than the present result near the maximum because of the smaller σ^{+} by MBPT, as shown in Fig. 1(b). This illustrates the importance of comparison between different theories not only in terms of σ^{++}/σ^{+} (as in most literature) but also in terms of σ^{++} directly.

Comparison of the present acceleration-form ratio with available experimental data [20–25] is made in Fig. 3. Most data lie above the present result, with the exception of the oldest data. We note that a recent measurement between 85 and 150 eV, still unpublished [26], has led to cross-section ratios that are in excellent agreement with the present calculations.

In conclusion, we calculated the cross sections for single and double photoionization of helium from 80 to 280 eV by extending the hyperspherical close-coupling method. The length and acceleration forms of the dipole operator led to cross sections for double photoionization that were in good agreement with each other. The present method uses a discretization procedure for the He^{+} continuum to represent the double continuum states of He in the asymptotic region. This circumvents the use of the correct asymptotic boundary condition for two outgoing electrons. This necessarily introduces drawbacks. First, the region of the configuration space in which the two electrons escape far from the nucleus is neglected, and hence the current flux flowing into an asymptotic region with $r_1 \simeq r_2$ is ignored, as discussed in Ref. [8]. Second, calculations close to the eigenenergies of the pseudostates may sometimes be deteriorated by the effects of pseudothresholds. With these drawbacks, however, the present method produced accurate low-energy cross sections for helium.

One of the authors (J.-Z.T.) was supported by the Special Researchers' Basic Science Program of RIKEN. We would like to thank Professor C. H. Greene for sending us calculated cross sections prior to publication. We would also like to thank Professor H. Schmidt-Böcking, D. Kato, K. Hino, and S. Watanabe for useful discussion.

[1] S. L. Carter and H. P. Kelly, Phys. Rev. A **24**, 170 (1981).
[2] K. Hino, T. Ishihara, F. Shimizu, N. Toshima, and J. H. McGuire, Phys. Rev. A **48**, 1271 (1993).
[3] K. Hino, P. M. Bergstrom, and J. H. Macek, Phys. Rev. Lett. **72**, 1620 (1994).
[4] A. Dalgarno and H. Sadeghpour, Phys. Rev. A **46**, 3591 (1992).
[5] R. C. Forrey, H. R. Sadeghpour, J. D. Barker, J. D. Morgan III, and A. Dalgarno, Phys. Rev. A **51**, 2112 (1995), and references therein.
[6] D. Proulx and R. Shakeshaft, Phys. Rev. A **48**, R875 (1993).
[7] M. Pont and R. Shakeshaft, Phys. Rev. A **51**, 494 (1995).
[8] K. W. Meyer and C. H. Greene, Phys. Rev. A **50**, R3573 (1994).
[9] J.-Z. Tang, S. Watanabe, and M. Matsuzawa, Phys. Rev. A **46**, 2437 (1992).
[10] J. H. Macek, J. Phys. B **1**, 831 (1968); C. D. Lin, Adv. At. Mol. Phys. **22**, 77 (1986).
[11] J.-Z. Tang, S. Watanabe, and M. Matsuzawa, Phys. Rev. A **46**, 3758 (1992).
[12] J.-Z. Tang, S. Watanabe, M. Matsuzawa, and C. D. Lin, Phys. Rev. Lett. **69**, 1633 (1992).
[13] J.-Z. Tang and I. Shimamura, Phys. Rev. A **50**, 1321 (1994).
[14] J.-Z. Tang and I. Shimamura, Phys. Rev. A **51**, R1738 (1995).
[15] J.-Z. Tang, C. D. Lin, B. Zhou, and I. Shimamura, Phys. Rev. A **51**, 4694 (1995).
[16] D. Kato and S. Watanabe, Phys. Rev. Lett. **74**, 2443 (1995).
[17] C.-H. Park, A. F. Starace, J. Tan, and C.-D. Lin, Phys. Rev. A **33**, 1000 (1986).
[18] J. A. R. Samson, Z. X. He, L. Yin, and G. N. Haddad, J. Phys. B **27**, 887 (1994).
[19] P. Decleva, A. Lisini, and M. Venuti, J. Phys. B **27**, 4867 (1994).
[20] R. Wehlitz, F. Heiser, O. Hemmers, B. Langer, A. Menzel, and U. Becker, Phys. Rev. Lett. **67**, 3764 (1991).
[21] T. Carlson, Phys. Rev. **156**, 142 (1967).
[22] V. Schmidt, N. Sandner, H. Kuntzemüller, P. Dhez, F. Wuilleumier, and E. Källne, Phys. Rev. A **13**, 1748 (1976).
[23] G. R. Wight and M. J. van der Wiel, J. Phys. B **9**, 1319 (1976).
[24] D. M. P. Holland, K. Codling, J. B. West, and G. V. Marr, J. Phys. B **12**, 2465 (1979).
[25] R. J. Barlett, P. J. Walsh, Z. X. He, Y. Chung, E. M. Lee, and J. A. R. Samson, Phys. Rev. A **46**, 5574 (1992).
[26] V. Mergel *et al.* (private communication).