R matrix with pseudostates calculation for single and double ionization of helium by photon impact

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We have applied the *R* matrix with pseudostates (RMPS) method to calculate single and double ionization of helium by photon impact. Using the velocity gauge of the dipole operator, excellent agreement with high-precision experimental data is obtained for photon energies between threshold and 200 eV. While some discrepancies remain between results from different gauges, we demonstrate that the RMPS method can produce satisfactory results if either the length, velocity, or acceleration form of the dipole operator is used. [S1050-2947(97)50709-8]

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Double photoionization of helium has attracted much attention, both from experimentalists and theorists, for many years. This fundamental problem, involving the correlated motion of two free electrons in the final state, has been approached theoretically by several methods, usually characterized as perturbative (Born-type) or nonperturbative (close-coupling based). A list of recent references can be found in the work of Forrey *et al.* [1].

The *R* matrix with pseudostates (RMPS) method, as formulated by Bartschat *et al.* [2], has recently been applied successfully to electron scattering from various light atoms and ions [3–7]. The approach is essentially a combination of the standard *R*-matrix method, as reviewed by Burke and Robb [8], with the ideas of the ''convergent close-coupling'' (CCC) approach of Bray and Stelbovics [9,10]. As such, it should be able to treat ionization, ionization plus excitation, and double ionization by photon impact in a similar manner to the work presented by Meyer and Greene [11] and by Kheifets and Bray [12]. Interestingly, however, both of the latter papers presented only results obtained in the velocity and acceleration forms of the dipole operator, due to apparent problems with the length gauge.

In order to check the ability of the RMPS approach to accurately predict single- and double-photoionization cross sections, we have modified the most recent *R*-matrix code of Berrington, Eissner, and Norrington [13] and implemented all the changes outlined by Bartschat *et al.* [2]. These include a much more stable orthogonalization procedure to deal with the overcomplete basis of pseudo and continuum orbitals, improved orthogonalization of all bound orbitals, modification of the bound orbitals near the *R*-matrix boundary to ensure a consistent logarithmic derivative of the new continuum basis after Schmidt orthogonalization, and the omission of high-energy poles in the calculation of the *R* matrix from the eigenvalues and eigenvectors of the Hamiltonian matrix. The latter change is essential for the proper evaluation of the Buttle correction [14], since it avoids double counting of the poles whose eigenvalues are pushed up due to the orthogonalization procedure.

We then performed an RMPS calculation with the following 23 orbitals (and thus the corresponding states of He^+) included in the close-coupling plus correlation expansion: the physical orbitals $1s$, $2s$, $2p$, $3s$, $3p$, and $3d$ as well as a set of pseudo-orbitals $\overline{n\ell}$ up to $8s$, $8p$, $7d$, and $6f$. The latter pseudo-orbitals represent the higher discrete as well as the target continuum states. They were constructed by taking the minimum linear combination of Sturmian-type orbitals $r^i e^{-\alpha r}$ orthogonal to the above-mentioned orbitals. The pseudostates were then obtained by diagonalizing the target Hamiltonian in the above basis.

In the present calculation, we chose range parameters $\alpha=(1.10,1.20,1.27,1.30,1.33,1.40,1.50)$ and averaged the results (see below). This choice of α's ensured that the pseudostates with $\overline{n}=4$ were always bound, while all states with $\overline{n}\geqslant 5$ lay in the ionization continuum of He^+. Excitation to the latter states represents double ionization in our model. As described earlier [5,15], however, the small number of discrete pseudostates requires a correction to be made to estimate the contribution to double ionization from excitation of discrete pseudostates. The *R*-matrix radius was chosen as $27a_0$ to ensure that all physical and pseudo-orbitals fit properly into the box. The values for the ground-state energy of He were always below -2.902 a.u., with very minor variations depending on the actual α value. Finally, 40 continuum orbitals per angular momentum of the photoelectron were sufficient to obtain converged results up to incident photon energies of 200 eV.

Figure 1 shows our results for photoionization of helium in the vicinity of the $(3s3p)^1P^o$ resonance. Very good agreement is obtained between the length and velocity gauge results of the cross section for leaving the He^+ ion in its ground state, and the form of the background cross section agrees much better with that measured by Lindle *et al.* [16] than the form obtained in a previous ten-state (discrete states only) *R*-matrix calculation [17]. Nevertheless, our predictions lie approximately 3% above the experimental data. To investigate this discrepancy, we performed several calculations with different numbers of states and α values. We believe that the present theoretical results are converged to better than 1%.

Moving on to the region beyond the double-photoionization threshold at 79 eV, we present in Fig. 2 results for photoionization of $He(1s^2)^1S$ with the residual He^+ ion in its ground state $(1s)^2S$ or in excited states with prin-

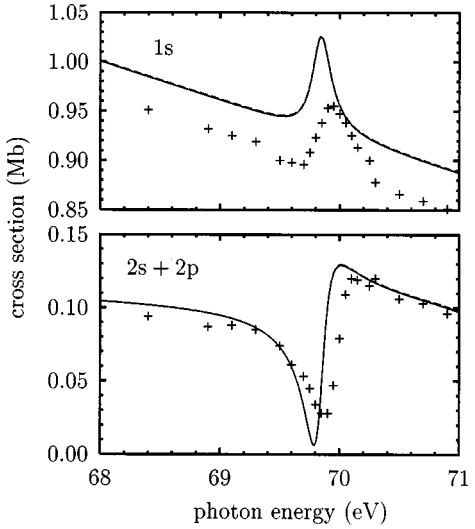

FIG. 1. Cross sections for photoionization of $He(1s^2)^1S$ with the He^+ ion remaining in its ground state $(1s)^2S$ (top) or being excited to either the $(2s)^2S$ or $(2s)^2P^o$ state (bottom) as a function of the incident photon energy in the vicinity of the $(3s3p)^1P^o$ resonance. The results obtained with the length and velocity forms of the dipole operator (indistinguishable on the graph) are compared with the experimental data of Lindle *et al.* [16].

cipal quantum numbers $n=2$ or $n=3$. Note that the structures in the $n=2,3$ results between 80 and 100 eV are unphysical. They originate from Rydberg resonance series converging to pseudothresholds and could, for example, be smoothed out by performing more calculations with different α values or by convoluting the results with a finite energy resolution (see below). Also shown in Fig. 2 are results for the total photoionization cross section, including single and double ionization. For single ionization with the He^+ ion in states with $n\leqslant 3$, we obtain very good agreement between the results from the length and velocity gauges of the dipole operator, while those obtained in the acceleration gauge appear too high for ionization to $He^+(1s)$ and too low for ionization plus excitation to $n=2,3$. Since single ionization leaving He^+ in its ground state is the dominating process, the predictions for the total ionization cross section obtained in the acceleration gauge are also larger than the high-precision experimental results of Samson *et al.* [18,19]. For energies below 100 eV, our length and velocity gauge results (indistinguishable on the graph) lie slightly above the experimental data, while essentially perfect agreement with experiment is obtained for photon energies between 100 and 200 eV. The acceleration gauge results converge towards the other curves and the experimental values for energies above 150 eV.

Figure 3 demonstrates the effects of averaging the results of different α values as well as including the above-mentioned correction to the double-ionization cross sections from excitation of discrete pseudostates. The results for a single α value (1.27) are strongly affected by pseudoresonances. Since the R-matrix method is very efficient in calculating the results for many energies, we have chosen a very narrow mesh of incident photon energies (approximately 1000 individual points) to illustrate this effect. We found that the length gauge results were most sensitive to pseudoresonances, which may, in part, explain the problems reported in other calculations for this gauge [11,12]. Note that the correction is most important in the near-threshold regime, due to the very small number of open channels to represent double ionization. Finally, we show the curves obtained after convoluting the corrected results with a Gaussian of energy resolution $\Delta E=\sqrt{e}$, where e is the energy (in Rydbergs) above the double-photoionization threshold. Such a procedure was also used by Meyer *et al.* [20] to smooth out remaining unphysical structures in numerical results.

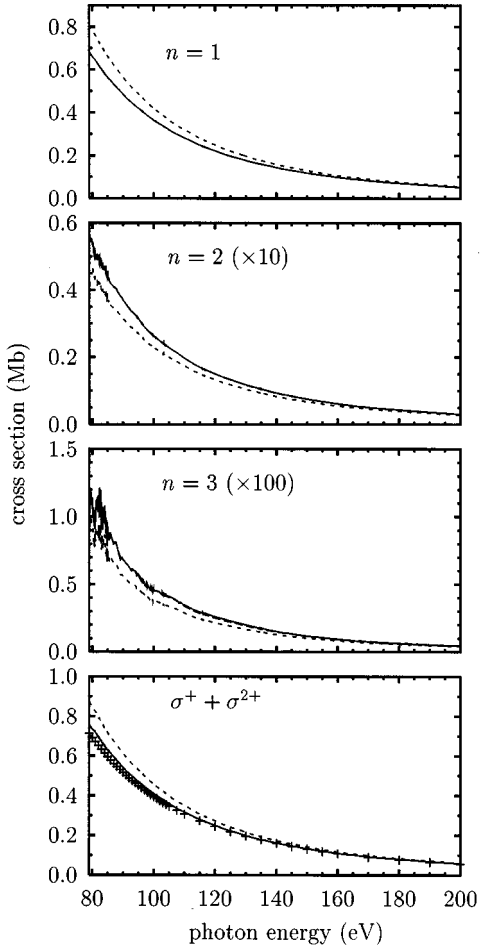

FIG. 2. Cross sections for photoionization of $He(1s^2)^1S$ with the He^+ ion in levels with various principal quantum number n, as well as the total (single plus double) photoionization cross section as a function of the incident photon energy. Solid line, length, and velocity gauge results; dashed line, acceleration gauge. The experimental data for the total cross section are taken from Samson *et al.* [18,19].

Finally, we show in Fig. 4 the ratio of double- to single-photoionization cross sections. Not surprisingly, the acceleration gauge results are significantly lower than the experimental data. This discrepancy is not only due to problems with this gauge in predicting double photoionization alone (i.e., the numerator is too small), but also due to problems in the calculation of single photoionization to He^+ in its ground state (i.e., the denominator is too large). Our length gauge results, while agreeing well with the experimental data of Levine *et al.* [21], lie significantly above the experimental results of Dörner *et al.* [22] and the most recent results of Samson and collaborators [19]. Furthermore, problems appear in the length gauge results for energies above 170 eV, where the predicted increase in the ratio is not seen in any of

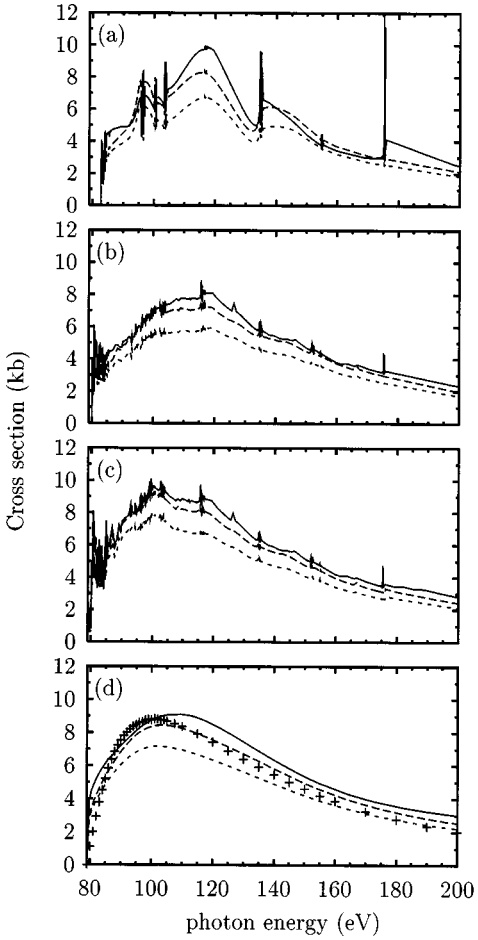

FIG. 3. Cross section for double photoionization of $He(1s^2)^1S$ as a function of the incident photon energy. (a) Results for $\alpha = 1.27$, (b) results averaged over seven α values, (c) averaged results corrected for excitation from discrete pseudostates, (d) convoluted results (see text). Solid line, length gauge; long dashes, velocity gauge; short dashes, acceleration gauge. The experimental data are from Samson *et al.* [18,19].

the experimental data sets. However, our velocity gauge results, which we (like other authors [11,12]) would trust the most for the present case of interest, agree very well with the experimental results of Refs. [19,22].

In conclusion, we have demonstrated that the RMPS method can, indeed, be used to calculate single- and double-photoionization cross sections with accuracies similar to those obtained from related close-coupling-type approaches.

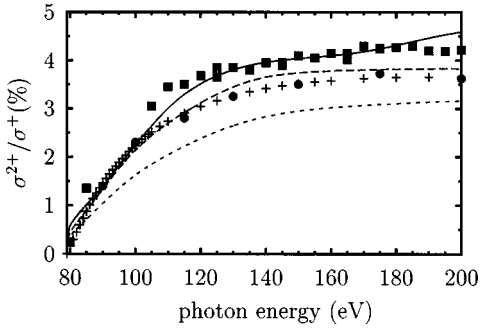

FIG. 4. Ratio of cross sections for double to single photoionization of $He(1s^2)^1S$ as a function of the incident photon energy. The results, obtained from the smoothed curves presented in Fig. 3, are compared with the experimental data of Levine *et al.* [21], Dörner *et al.* [22], and Samson *et al.* [19].

This is an important result in itself, since the generality of the Berrington, Eissner, and Norrington codes, in connection with the present modifications, will immediately allow for calculations of this kind to be performed for more complex targets than atomic helium. Although we did not achieve complete agreement between the results obtained from different gauges in the present calculation, we believe that the problem can be solved, in principle, by systematically increasing the basis set. A first check can be performed by looking at the results for single photoionization leaving the residual ion in its ground state.

Finally, we note that Meyer, Greene, and Esry [23] have recently used a finite element basis set in connection with the eigenchannel *R*-matrix method [24]. They obtained very good agreement between their results in all three gauges. While these findings are very promising, in particular as a benchmark for future work on this problem, the computational effort was very large. In contrast, the present calculation was performed on a desktop DEC-Alpha workstation within a few hours of CPU time. Very little additional computer resources would be required to obtain results for a large number of photon energies, for example, to map out the Rydberg resonance structure associated with the discrete levels of He^+.

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