R-matrix calculations for double differential cross sections in electron impact ionization of helium

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Short title: *R-matrix calculations for e-He DDCS*

PACS number(s): 34.80 D

Submitted to: *J. Phys. B: At. Mol. Opt. Phys.*

Date: 21 June 1995

Abstract. Double differential cross sections for electron impact ionization of helium are calculated within the R-matrix method. Angular dependences of the cross sections are presented for the primary (scattered) and secondary (ejected) electron for the case where the energy of the primary electron is large compared to that of the secondary electron at incident energies of 100, 200 and 300 eV. Comparison with experiment and other theories is made and the results are discussed based on the characteristics of the angle-integrated triple differential cross section.

1. Introduction

In a previous paper (Raeker *et al* 1994), we applied the R-matrix method of Bartschat and Burke (1987) to the calculation of single differential (with respect to energy loss of the projectile) and total cross sections for electron-impact ionization of helium from threshold to 400 eV. However, since the comparison of integrated cross sections is not a satisfying test of collision theories due to the angular integrations and, therefore, possible cancellation of errors, a more stringent test is desirable.

We have now extended the computer code of Bartschat (1993) to allow for the calculation of triple differential cross sections (TDCS) and, consequently, double differential cross sections (DDCS) by integration over the angles of one of the two outgoing electrons. It will be shown that the knowledge of the TDCS allows for an assessment of the reliability of our DDCS and provides explanations for possible deviations from experiment.

The DDCS is obtained from the TDCS by integrating over the angular variables of of the outgoing electron that is not experimentally detected. Its energy for a particular final ionic state is fixed by an energy measurement on the detected electron. Hence, the DDCS can be expressed as a function of the energy and the scattering angle of the detected electron. Since the energies of the two outgoing electrons are related by energy conservation, we only give the energy of the *slow* ejected electron throughout this paper.

The helium atom has provided a test ground for many collision theories. On the experimental side, measurements with high accuracy have been performed over a wide range of kinematic situations (cf Ehrhardt 1983, Teubner 1985). The existence of a large set of high-quality experimental data led to our choice of this system, and we take the opportunity to explain the particular features of our model in some detail. Since the general R-matrix code can be used directly for more complex systems as well, it is important to obtain some feeling for the accuracy of the present model.

Müller-Fiedler *et al* (1986) have published an extensive set of experimental data for the DDCS in e-He ionization for impact energies between 100 and 600 eV. They give results for detecting either the primary or the secondary electron. We also compare our DDCS with the distorted-wave Born calculations of McCarthy and Zhang (1989) and those obtained with the Glauber approximation by Ray *et al* (1991). In the latter paper, results were only given for the case where the primary electron is detected.

This paper is organized as follows. In section 2 we briefly outline the basic theory which has been fully described earlier (Bartschat and Burke 1987, Bartschat 1993), and we give details of the wavefunctions used in the calculation. In section 3 we present results for both kind of experiments (observation of the primary or the secondary electron) and interpret the data based on the characteristics of the binary and recoil lobes of the TDCS.

2. Theory

2.1. Scattering amplitudes and differential cross sections

In this paper, we describe the ionization process

$$e(\mathbf{k}_0) + \mathrm{He}\,(1s^2)\,{}^1\mathrm{S} \longrightarrow e(\mathbf{k}_1) + e(\mathbf{k}_2) + \mathrm{He}^+(1s). \qquad (1)$$

For collision processes with projectile energies $\geq 100\,\mathrm{eV}$, one can make the qualitative assumption that most of the incident electrons are scattered into small angles and remain *fast* while most of the ejected electrons have energies of only a few eV. (Quantum mechanically, this is supported by the fact that the "exchange" cross section is much smaller than the "direct" cross section.) We denote the linear momenta of the *fast* incident, *fast* scattered and *slow* ejected electrons by $\mathbf{k}_0$, $\mathbf{k}_1$ and $\mathbf{k}_2$, and the corresponding kinetic energies by E_0, E_1 and E_2, respectively. The two outgoing electrons can be "distinguished" by their energies which, to some extent, justifies their different treatment in our model:

(i) The *fast* ionizing electron denoted by $e(\mathbf{k}_0)$ and $e(\mathbf{k}_1)$ is described by distorted (Coulomb) waves.

(ii) The ejected electron $e(\mathbf{k}_2)$ is *slow*. Consequently, the initial and final (N+1) electron states Ψ_i and $\Psi_f\{\mathrm{He}^+(1s) + e(\mathbf{k}_2)\}$ are represented by an R-matrix (close-coupling) expansion.

(iii) The process is treated in LS coupling and only the Coulomb interaction between the ionizing electron and the target electrons is taken into account. Therefore, the spin components of the fast electron *alone* and of the total spin in the initial and final states of the *combined* [ejected electron + residual ion] system are conserved.

(iv) Exchange effects between the two final continuum electrons are neglected. Their contribution is rather small for incident energies above 200 eV, and so this approximation is fairly good for asymmetric collisions where the energies of the two outgoing electrons are very different.

(v) The long-range Coulomb interaction between the two outgoing electrons is neglected. This, of course, is the most problematic of our approximations.

For a particular final ionic state, the energy of the undetected electron is fixed through an energy measurement on the detected electron. In order to calculate the scattering amplitude explicitly, we choose the *collision system* as the coordinate frame. In this frame, the momentum of the incident beam direction defines the z–axis while the linear momentum of the detected electron is fixed in the xz–plane. After applying the multipole expansion of the Coulomb interaction and evaluating the angular integrals using standard techniques including the Wigner-Eckart theorem, the result for the scattering amplitude is given by

$$
\begin{aligned}
f(L_f M_{L_f}, & S_f M_{S_f}, \mathbf{k}_2 m_{s_2}; \mathbf{k}_1 m_{s_1}; L_0 M_{L_0}, S_0 M_{S_0}, \mathbf{k}_0 m_{s_0}) \\
&= -\delta_{m_{s_0} m_{s_1}} (S_f M_{S_f}, \tfrac{1}{2} m_{s_2} | S_0 M_{S_0}) \frac{4\sqrt{2\pi}}{\sqrt{k_0 k_1 k_2}} (-1)^{L_0 + M_{L_0}} \\
&\times \sum_{\substack{l_0 l_1 m_1 l_2 m_2 \\ L_1 M_{L_1} S_1 M_{S_1} \lambda}} \delta_{S_0 S_1} \delta_{M_{S_0} M_{S_1}} Y_{l_1 m_1}(\hat{\mathbf{k}}_1) Y_{l_2 m_2}(\hat{\mathbf{k}}_2)\, \mathrm{i}^{\lambda + l_0 + l_1} \\
&\times \exp[\mathrm{i}(\sigma_{l_2} + \sigma_{l_1} - \sigma_{l_0} + \delta_{l_1} - \delta_{l_0})] \\
&\times (l_1 \, {-m_1}, \lambda m_1 | l_0 0)(L_0 M_{L_0}, L_1 \, {-M_{L_1}} | \lambda m_1)(L_f M_{L_f}, l_2 m_2 | L_1 M_{L_1}) \\
&\times \langle \Psi_f^{L_1} \| \sum_{i=1}^{N+1} \sqrt{\frac{4\pi}{2\lambda + 1}} i^{\lambda} Y_{\lambda}(\hat{\mathbf{r}}_i) R^{\lambda}_{l_0 l_1}(k_0 k_1; r_i) \| \Psi_i^{L_0} \rangle. \qquad (2)
\end{aligned}
$$

The quantum numbers $L_0 S_0 M_{L_0} M_{S_0}$ define the initial atomic state and $L_f S_f M_{L_f} M_{S_f}$ are the quantum numbers for the final state of the ion. In partial wave expansion the notation of the three electrons e_i ($i = 0, 1, 2$) is given by $\mathbf{k}_i l_i m_i m_{s_i}$, and $\sigma_{l_i}, \delta_{l_i}$ are the Coulomb and potential phase shifts, respectively. The quantum numbers for the combined system of ejected electron plus residual ion are $L_1 S_1 M_{L_1} M_{S_1}$, and $(j_1 m_1, j_2 m_2 | j_3 m_3)$ denote Clebsch-Gordan coefficients. The spherical harmonics in (2) describe the directions of motion for the two outgoing electrons, with $\hat{\mathbf{k}}_1 \equiv (\theta_1, \phi_1)$ and $\hat{\mathbf{k}}_2 \equiv (\theta_2, \phi_2)$. We choose $\phi_j \equiv 0$ ($j = 1$ or 2) for the azimuthal angle of the detected electron. The reduced matrix element shows that, in our model, the initial and final states of the [ejected electron + residual ion] system are essentially being perturbed by the Coulomb interaction with the fast ionizing electron. Finally, the normalization of the continuum radial functions in (2) follows the convention of Bartschat and Burke (1987).

The double differential cross section (with respect to the direction of electron $e(\mathbf{k}_j)$, $j = 1$ or 2) is obtained after integrating over all possible directions of the undetected electron $e(\mathbf{k}_{j'})$ ($j' = 3 - j$):

$$\frac{d^2\sigma}{dE_2 d\hat{\mathbf{k}}_j} = \frac{k_1 k_2}{k_0} \underbrace{\frac{1}{2(2S_0+1)(2L_0+1)} \sum_{m_{s_0} M_{S_0} M_{L_0}}}_{\text{unpolarised initial atoms and electrons}} \underbrace{\sum_{m_{s_1} m_{s_2} M_{S_f} M_{L_f}}}_{\text{no observation of spins or magnetic sublevels in the final state}}$$
$$\times \underbrace{\int d\hat{\mathbf{k}}_{j'}}_{\text{undetected electron}} |f(L_f M_{L_f}, S_f M_{S_f}, \mathbf{k}_2 m_{s_2}; \mathbf{k}_1 m_{s_1}; L_0 M_{L_0}, S_0 M_{S_0}, \mathbf{k}_0 m_{s_0})|^2 \quad (3)$$

The sums over the magnetic quantum numbers arise from the experimental preparation of the electron and target beams (unpolarized electrons in collision with unpolarized helium atoms) and the fact that no spin selection is performed in the electronic channel. The kinetic energy E_2 of the secondary electron is given by

$$E_2 = E_0 - E_1 - I_f \quad (4)$$

where I_f is the ionization potential for production of the residual ion in state f.

2.2. Numerical details

The use of an R-matrix basis to represent the initial and final (N+1) electron states was described in detail by Bartschat and Burke (1987). One important requirement of the Belfast R-matrix codes is the need to expand both the initial and final (N+1) electron wavefunctions in terms of *one* set of bound orbitals and R-matrix basis functions. While this automatically guarantees orthogonality between the initial and final state inside the R-matrix box, it is sometimes difficult to represent all physically important orbitals to a sufficient accuracy with a limited basis. This is particularly true for ionization of helium where the 1s (Hartree-Fock) orbital in the ground state is very different from the 1s orbital in any of the excited states; the latter is almost a hydrogenic orbital with effective charge $Z = 2$ which is also the appropriate 1s orbital for the residual ion. As another example, the 2s orbital in the ion is again significantly different from a suitable 2s orbital for the description of the $(2s^2)^1S$ autoionizing resonance.

For the calculations reported in this paper, we have taken the Hartee-Fock 1s orbital for the $He(1s^2)$ ground state from the tables of Clementi and Roetti (1974) to ensure a good representation of the initial state, together with hydrogenic (Z=2) 2p,3p,3d orbitals and $\overline{2s}$ and $\overline{3s}$ pseudo-orbitals optimized on the $He^+(1s)$ and $He^+(2s)$ ionic states. Our calculated energy of the He bound state is -2.8891 (in atomic units), and the energy obtained for the ionic ground state is -1.9999.

Distorted waves for angular momenta up to $l = 90$ were calculated for the *fast* electron to ensure converged results. The incoming electron wave was calculated in the static potential of the neutral He ground state whereas the scattered electron was treated as a distorted Coulomb wave moving in the $He^+(1s)$ static potential. We accounted for multipole ranks $\lambda \leq 4$ of the radial operator describing the Coulomb interaction between projectile and target. This led to five coupled continuum states and partial waves for the secondary ejected electron with $l_2 \leq 6$. For each l_2, twenty-five inner-region continuum orbitals were constructed. The size of the inner region of 19.032 atomic units is large enough to contain the initial bound state and the final ionic state. With this set of values, we obtained converged DDCS for the He ionization process of interest with projectile energies up to 400 eV. Along with the DWBA we performed a second calculation in which the primary electron was treated as a plane wave. This effect will be discussed in section 3.

In a previous paper (Raeker *et al* 1994) we chose the hydrogenic ($Z = 2$) He^+ orbitals (1s,2s,2p,3s,3p,3d) to represent the residual ion in the R-matrix expansion. A detailed analysis of the TDCS, however, revealed that we needed further improvement. While a plane-wave + R-matrix calculation for 256 eV incident energy and a scattering angle of 4° for the fast projectile was expected to give the same results as the first Born calculations by Byron *et al* (1986), the actual results showed a significant overestimation of the binary peak and a simultaneous underestimation of the recoil peak. In fact, the six He^+ orbitals listed above, even in conjunction with the R-matrix continuum orbitals, could not represent the tail of the He($1s^2$) ground state wavefunction to sufficient accuracy. Further studies revealed that numerical results for the ionization process depend in a very sensitive way on the representation of the initial bound state. Consequently, a low ground state energy alone does not indicate an appropriate representation of the initial state wavefunction for the ionization process.

The new R-matrix basis set described in this section, especially the inclusion of the Hartree-Fock 1s orbital, avoids these problems, and the TDCS calculation with these orbitals reproduced the first Born results of Byron *et al* (1986) very well. This fact gives us confidence that the other parts of our newly developed computer codes are working properly.

3. Results and discussion

We present results for the DDCS for e-He ionization at incident electron energies (E_0) of 100, 200 and 300 eV. The relevant parameters of our calculation, *i.e.* the energies of the incident, scattered and ejected electrons, were chosen in accordance with the experiments performed by Müller-Fiedler *et al* (1986). Note that incident energies below 100 eV are too low considering the requirement for an asymmetric energy sharing between the two outgoing electrons in our approach.

The primary-electron DDCS is plotted as a function of the scattering angle θ_1 in figure 1. The characteristic features of the experimental results are well reproduced by the present model. Good agreement is found at $E_0 = 200$ and 300 eV for small secondary energies E_2. A comparison is made with the distorted wave Born approximation (DWBA) of McCarthy and Zhang (1989) and the Glauber approximation (GA) of Ray *et al* (1991). At larger values of E_2, the present results as well as the DWBA tend to overestimate the experimental data. As expected, the largest deviation from experiment occurs for $E_0 = 100$ eV and a relatively fast ejected electron. This is the case where the kinematic setup does not meet the asymmetry requirement in our model. Hence, additional effects, such as exchange between the fast projectile and the target electrons as well as long-range electron–electron correlations, should be taken into account. The predicted cross sections are too large at small incident energies, but the results are consistent with our previously published total cross sections (Raeker *et al* 1994, figure 1) at $E_0 = 100\,\mathrm{eV}$.

The above results can be explained by looking at the characteristics of the TDCS shown in figure 2 for $E_0 = 256\,\mathrm{eV}$, $E_2 = 3\,\mathrm{eV}$ and $\theta_1 = 4°$. Our plane-wave + R-matrix calculation coincides with the standard first Born approximation (FBA) and exhibits the following well-known characteristics: (i) the binary peak is too high, (ii) the recoil peak is too low, and (iii) the FBA does not reproduce the shift of the lobes towards the direction of larger ejection angles. When one uses distorted Coulomb waves instead of plane waves for the fast electron, the binary peak moves to larger angles and, therefore, closer to the experimental peak, but the recoil peak moves away from the experimental data. Nevertheless, we judge it as a minor improvement over the first Born result.

Note that the primary-electron DDCS is proportional to the area below the TDCS curve. Consequently, the deviations of our theory from the experimental lobes cancel each other, to some extent, when one integrates over the ejection angle of the secondary electron. For small ejection energies, the TDCS exhibits a recoil peak that is comparable in magnitude to the binary peak, so that substantial cancellation of errors is possible. However, with increasing energy of the secondary electron, the recoil peak becomes relatively smaller, so that the binary contribution becomes dominant. Hence the compensation effect decreases, and our theory overestimates the cross sections as shown by the lines C, D and E in figure 1.

The secondary-electron DDCS, depending on θ_2, are shown in figures 3 and 4. The agreement with experiment is not as good in this case, with the largest deviations occurring again for relatively low incident and relatively high ejected electron energies. In addition to the distorted wave Coulomb calculation, a plane wave calculation was performed, and the results are also shown in figure 4. Again, the curves can be understood with the help of figure 2. Since integration is performed over the scattering angle of the primary electron (θ_1), we now have to consider the difference between theory and experiment for each ejection angle in figure 2. As can be seen from figure 1, the dominating contribution comes from scattering into small angles; hence, figure 2 with $\theta_1 = 4°$ is representative for the entire process. (The different projectile energy does not change the validity of this qualitative argument.) At small ejection angles, the left side of the binary peak ($0° \geq \theta_2 \geq -50°$) dominates over the small contributions from the right side of the recoil peak ($0° \leq \theta_2 \leq 50°$) (the latter being equivalent to scattering with $0° \geq \theta_2 \geq -50°$ when $\theta_1 = -4°$). On the other hand, the recoil peak dominates for large ejection angles, and hence we can see that the small theoretical secondary-electron DDCS at large ejection angles ($\theta_2 \geq 120°$) are due to the underestimation by the theoretical TDCS in the region of the recoil peak. While our arguments rely on a somewhat simplified picture, the overall effects due to the integration over the angles of one of the outgoing electrons are explained correctly because of the cylindrical symmetry of the TDCS around the axis of momentum transfer. Finally, on the left side of the binary peak, the plane wave treatment yields even larger cross sections than the distorted Coulomb wave approach. This is clearly reflected in figures 3 and 4.

4. Conclusions

The TDCS and DDCS calculations reported in this paper provide more detailed information about previous work on total ionization cross sections. The experimental data for the primary-electron DDCS for electron impact ionization of helium are well reproduced by the DWBA+R-matrix approach at 200 and 300 eV. Furthermore, it was found that the calculation of secondary-electron DDCS is very sensitive to the long-range Coulomb interaction between the two outgoing electrons, and this effect should be taken into account to improve the results. A second Born approach and the inclusion of "post collision effects" is also desirable. Work in this direction is currently in progress.

Acknowledgments

The authors would like to thank Professor P.G. Burke for many stimulating discussions and Professor K. Blum for his continuous support and interest in this project. This work was supported, in part, by the Deutsche Forschungsgemeinschaft in Sonderforschungsbereich 216, "Polarization and Correlation in Atomic Collision Complexes", the European Community under contract ERB CHRX CT920013, NATO under grant CRG-930056 (AR and KB), the UK EPSRC under grant GR/H59862 (RHGR), and the United States National Science Foundation under grant PHY-9318377 (KB). One of us (AR) wishes to thank the Queen's University of Belfast for their support and hospitality.

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Figure captions

Figure 1. Double differential cross section for electron impact ionization of He($1s^2$) and detection of the scattered electron as a function of the scattering angle. The energies of the incident electron are a) 100 eV, b) 200 eV, c) 300 eV. The ejected electron energies are A: 2 eV, B: 4 eV, C: 10 eV, D: 20 eV, E: 40 eV, respectively. The DDCS for 2 eV (curve A) and 4 eV (curve B) are multiplied by 100 and 10, respectively. Present work (DWBA+R-matrix) ——, McCarthy and Zhang (1989) – – –, Ray *et al* (1991) · · · ·. The experimental data of Müller-Fiedler *et al* (1986) are denoted by □ .

Figure 2. Triple differential cross section for electron impact ionization of He($1s^2$) at an incident energy/ejected energy/scattering angle combination of 256 eV/3 eV/4° as a function of the ejection angle. DWBA+R-matrix ——, PW+R-matrix – – –, FBA of Byron *et al*(1986) · · · ·, experiment of Müller-Fiedler *et al* (1985) •.

Figure 3. Same as figure 1 for observation of the ejected electron. The experimental results of Goruganthu *et al* (1986) are represented by △. The DDCS in part a) A is multiplied by 2. Error bars have been left out when smaller than the symbol.

Figure 4. Double differential cross section for electron impact ionization of He($1s^2$) and detection of the ejected electron as a function of the ejection angle for incident/ejected electron energy of 300/4 eV. DWBA+R-matrix ——, plane wave + R-matrix — · — ·, McCarthy and Zhang (1989) – – –, Müller-Fiedler *et al* (1986) □.