Time-dependent close-coupling studies of the electron-impact ionization of excited-state helium

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The time-dependent close-coupling theory is applied to the study of the electron-impact ionization of helium from the excited $(1s2s)$ configuration. Calculations are made in an effort to resolve the discrepancy between theoretical calculations and existing experimental measurements for this cross section. We find good agreement with the existing convergent close-coupling calculations of Bray and Fursa [J. Phys. B **28**, L197 (1995)], but are in substantial disagreement with the experimental measurements of this quantity by Dixon *et al.* [J. Phys. B **9**, 2617 (1976)].

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I. INTRODUCTION

The electron-impact ionization of helium is the second most simple electron-atom scattering problem, after the electron-hydrogen problem. Due to its practical experimental advantages it has been studied extensively by experiment over many years. As well as its importance in the understanding of fundamental atomic physics, helium is also an important element in fusion plasma devices, where it has been used extensively as a diagnostic in recent years [1]. This use in fusion physics has demonstrated the need for accurate electron-impact ionization cross sections from excited states of atoms. The cross sections from all excited states must be known to accurately estimate the ionization balance. Although, of course, the ground state will usually be the most populated, sometimes extremely large cross sections arising from ionization from excited states counteracts this in making these cross sections just as critical. Recently, there has been a joint theoretical study of the electron-impact ionization of lithium from its ground and first excited state, where excellent agreement was obtained between three leading nonperturbative techniques [2]. The extremely large cross section calculated from the excited state, as well as the very large differences between these calculations and perturbative distorted-wave theoretical calculations, has highlighted the need for further study of electron-impact ionization from excited states.

With this in mind, we now turn our attention to the electron-impact ionization of helium from its first excited state. In recent times, theory has attempted to match the level of experimental study of helium by application of many leading nonperturbative theoretical techniques. The most comprehensive application of theory to the electron-helium system to date has been by the convergent close-coupling (CCC) method. This method has been used to investigate electron-helium scattering over a wide range of incident electron energies [3,4] and has generally given excellent agreement with experiment for the total ionization cross section from the ground state. A review of earlier calculations using the CCC method in electron-helium scattering is given by Fursa and Bray [4], which has been updated recently [5]. In recent years, the CCC method has also been employed to study the more difficult problem of energy and angular differential cross sections for electron scattering of helium [6–8], and in general, theory is found to be in good agreement with experiment. The time-dependent close-coupling method has also been used to calculate both the total integral and single differential cross sections for helium [9]; again very good agreement was found with experiment for both cases. Also, the semiempirical binary-encounter dipole method has been applied to helium [10]. At very high incident electron energies (up to 10 keV) good agreement was found with experiment.

In this paper, we examine electron-impact ionization of helium from the excited $(1s2s)$ configuration, using the time-dependent close-coupling method. Our calculations are made in an effort to resolve discrepancies between theory and experiment for electron scattering from the metastable states of helium. The CCC method, while giving good agreement for the total ionization cross section from the ground state of helium, gives cross sections [4,11] that are almost a factor of two lower than experimental measurements [12] for ionization from the $1s2s\ ^{1,3}S$ states. This anomaly with experiment also extends to studies of electron-impact excitation of helium. Whereas for electron-impact excitation from the ground state there is good agreement between CCC calculations, *R* matrix with pseudostates (RMPS) [13] calculations, and experimental measurements; for electron-impact excitation from the metastable $(1s2s)\ ^3S$ state of helium, there are large differences between theory and experiment. For excitation of the $n=2$ states from the $(1s2s)\ ^3S$ state, the CCC and RMPS theories are in very good agreement with each other, but are up to a factor of two lower than experimental measurements [14,15]. This problem remains unresolved.

In our calculations, we use a configuration-average approximation for the $1s2s$ configuration and resolve our ionization cross sections from the ^1S or ^3S states using the corresponding branching ratios, as used in previous configuration-average time-dependent calculations [16]. In this case, the branching ratios to both states are the same and equal to 1, so that the configuration-average approximation employed here predicts the same cross section for both ionization from the ^1S and ^3S states. We present total ionization cross sections over a range of incident electron energies using a Fourier-transform method which has been recently applied to electron-impact ionization of Li^{2+} [17]. This allows

us to calculate ionization cross sections over a wide range of incident electron energies.

In the following paragraphs, we briefly describe the time-dependent close-coupling theory used to calculate electron scattering from helium. We then present our results for the total ionization cross section and compare to other theoretical and experimental results. We conclude with a brief summary.

II. THEORY

In considering electron scattering from helium, we first make the ''frozen-core'' approximation employed in previous time-dependent calculations [9] and in the CCC calculations discussed earlier, where we freeze the $1s$ electron. This is a good approximation in the energy range considered here.

We first calculate the $1s$ hydrogenic ground state of He^+. A complete set of radial orbitals $P_{nl}(r)$ for helium are then obtained by diagonalization of the single-particle Hamiltonian

$$h(r) = -\frac{1}{2}\frac{\partial^2}{\partial r^2} + \frac{l(l+1)}{2r^2} - \frac{Z}{r} + V_D(r) + V_X(r), \qquad (1)$$

where the ''direct'' potential terms are given by

$$V_D(r) = \int_0^\infty \frac{\overline{P}_{1s}^2(r_1)}{\max(r_1, r)} dr_1 \qquad (2)$$

and act to shield the outer active electrons from the full Coulomb potential of the nucleus. The $\overline{P}_{1s}(r_1)$ radial orbital is the exact $1s$ orbital of He^+. The local ''exchange'' potential is given by

$$V_X(r) = -\alpha\left(\frac{24\overline{\rho}_{1s}(r)}{\pi}\right)^{1/3}, \qquad (3)$$

where $\overline{\rho}_{1s}(r)$ is the radial probability density for the $1s$ orbital of He^+. In this equation, α is a parameter which may be adjusted so that the single-particle energies for each angular momentum are in good agreement with a configuration-average experimental spectrum. For example, the ionization potential of the ground state of helium, made up of the exact $1s$ He^+ orbital and a $1s$ orbital obtained from diagonalization of the Hamiltonian in Eq. (1), is found to be 24.58 eV, using a suitable value for α, in excellent agreement with the exact experimental value. The ionization potential of the $1s2s$ configuration (again, made up from the exact $1s$ He^+ orbital and a $2s$ orbital obtained from diagonalization) is found to be 4.34 eV, in good agreement with the configuration-average experimental value.

The two-electron wave functions are constructed following previous time-dependent calculations [9] as antisymmetrized products of a radial wave packet and the $P_{2s}(r)$ radial orbital. From projection onto the time-dependent Schrödinger equation a set of time-dependent close-coupled partial differential equations is obtained

$$i\frac{\partial P_{l_1 l_2}^{LS}(r_1, r_2, t)}{\partial t} = T_{l_1 l_2}(r_1, r_2) P_{l_1 l_2}^{LS}(r_1, r_2, t) + \sum_{l_1', l_2'} U_{l_1 l_2, l_1' l_2'}^{L}(r_1, r_2) P_{l_1' l_2'}^{LS}(r_1, r_2, t), \qquad (4)$$

where $T_{l_1 l_2}(r_1, r_2)$ contains kinetic energy, centrifugal barrier, nuclear, direct Hartree, and local exchange operators; and $U_{l_1 l_2, l_1' l_2'}^{L}(r_1, r_2)$ couples the various $(l_1 l_2)$ scattering channels. These are then propagated in time for each LS symmetry as previously. Following our previous work on Li^{2+} [17], we propagate two time-dependent wave functions at energies of 40 and 140 eV, and after propagation, extract wave functions at many energies using the Fourier-transform approach as shown in detail in Ref. [17].

As in our previous time-dependent calculations, the close-coupled equations are solved on a lattice of uniform mesh spacing, in this case $\Delta r = 0.2$ a.u., with 512 points. After the time propagation, the probabilities and cross sections for ionization may be extracted by the usual projections onto products of bound and continuum radial orbitals for helium, obtained in the initial diagonalization.

III. RESULTS

The time-dependent close-coupling method was used to generate ionization cross sections over a range of incident electron energies from 10 to 190 eV. For each angular momentum from $L = 0-6$, two time-dependent wave functions were propagated at energies of 40 and 140 eV, and subsequent wave functions were extracted, as described, at energy increments of 10 eV. As in previous time-dependent calculations of total ionization cross sections, our results were ''topped up'' with distorted-wave calculations for high angular momentum above $L = 6$.

Electron-impact ionization cross sections for the He $1s2s$ configuration are presented in Fig. 1. In Fig. 1(a), we focus on the peak region of the ionization cross section and in (b) we present the ionization cross section at higher energies. We show the time-dependent close-coupling calculations compared with the experimental measurements of Dixon *et al.* [12], as well as distorted-wave calculations. The experimental measurements were made with metastable helium atoms predominantly in the $1s2s\ ^3S$ state. Our time-dependent calculations are substantially lower than the experimental measurements over the entire energy range by almost a factor of 2. The distorted-wave calculations, which were made using a previous configuration-average set of programs [18], are substantially higher than the time-dependent calculations near the peak of the cross section (though closer to experiment in this range), are still lower than the experimental measurements in the higher-energy range by a factor of 2. The distorted-wave calculations are also in good agreement with early Born calculations [19] which show the same trends.

In an effort to resolve this discrepancy, we also calculated the distorted-wave ionization cross section from the $1s$ shell

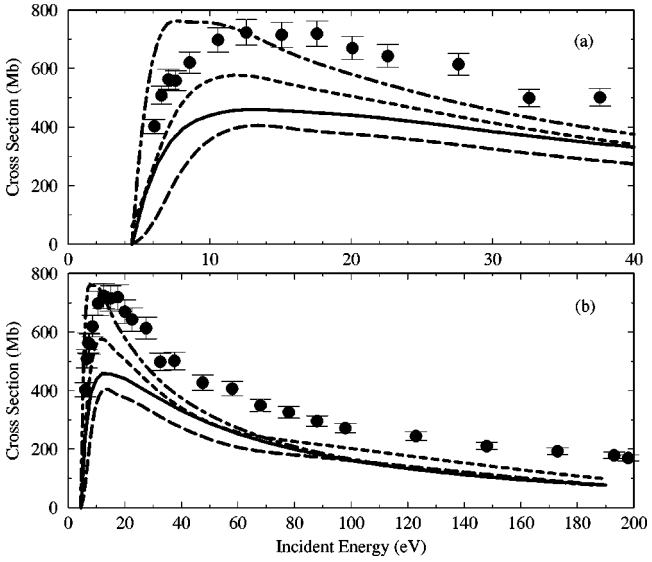

FIG. 1. Electron-impact ionization cross section for He from the $1s2s$ configuration. The filled circles are the experimental measurements of Dixon *et al.* [12], the short- and long-dashed lines are CCC calculations from the ^1S and ^3S states of He $1s2s$, respectively [5], the solid line is the time-dependent close-coupling calculation, and the dot-dashed line is the distorted-wave, calculation ($1.0\ \text{Mb} = 1.0\times10^{-18}\ \text{cm}^2$).

of He ($1s2s$). This was found to be almost negligible on the scale of the $2s$ ionization cross section. Contributions from excitation-autoionization from the $1s$ were also calculated and these, too, were found to be a small fraction of the $2s$ ionization. In any case, since both these contributions had thresholds above 40 eV incident electron energy, they could not affect the region around the peak of the ionization cross section.

We also show CCC calculations [5,11] made for both the $1s2s\ ^1S$ and $1s2s\ ^3S$ states. Near the peak of the cross section the time-dependent calculations fall between this set of calculations, which is to be expected as we present the configuration-average cross section of these two states. As expected, in the higher energy region the time-dependent, distorted-wave, and CCC ^3S calculations are all in very good agreement with each other, and all consistently lower than the experimental measurements. As discussed, the branching ratios for both of these states (^1S and ^3S) is in fact 1, so the configuration-average method predicts the same cross section from both the $1s2s\ ^1S$ and ^3S states. The differences between the CCC results for the ^1S and ^3S states, and between these and our time-dependent calculations are probably due to the different orbitals used in the respective calculations. As shown by Fischer [20], there is a degree of term dependence in the $2s$ orbital which gives rise to differences in this orbital when optimized on the ^1S or ^3S state.

IV. SUMMARY

In this Brief Report, we have presented time-dependent calculations of the electron-impact ionization of excited helium ($1s2s$). We have compared our calculations with experimental measurements and other nonperturbative calculations made using the convergent close-coupling method. Our results are also in good agreement with unpublished RMPS calculations made by Bartschat [21].

It is clear that there is a serious discrepancy between theory and experiment. The experimental measurements used metastable helium atoms predominantly in the ^3S state. However, there exists no agreement between the time-dependent and CCC theories and experiment for any energy range. New experimental measurements in this area are required to further investigate this discrepancy. We hope that this work will highlight the need for continuing experimental work in ionization from excited states of light atoms. We note that our configuration-average approximation for electron-impact ionization of atoms does not take into account term dependence in bound and continuum orbitals. We are currently working on this nontrivial extension of the method.

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