

Quasiclassical double photoionization from the $2^{1,3}\text{S}$ excited states of helium including shake-off

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Abstract

We account for the different symmetries of the $2^{1,3}\text{S}$ helium excited states in a quasiclassical description of the knockout mechanism augmented by a quantum shake-off contribution. We are thus able to formulate the separate contributions of the knockout and shake-off mechanisms for double photoionization for *any* excess energy from the $2^{1,3}\text{S}$ states. Photoionization ratios and single-differential cross sections calculated for the $2^{1,3}\text{S}$ excited states of helium are found to be in very good agreement with recent theoretical results.

1. Introduction

A two-electron transition in an atom after the absorption of a single photon is only possible due to electron–electron correlations. The study of two-electron transitions in helium, the simplest atomic target with two electrons, probes the role of electron–electron correlations in their purest form. As a result, there has been extensive experimental and theoretical study of double photoionization from the ground state of the helium atom.

In addition to the role of electron–electron correlations, the study of two-electron escape by a single-photon absorption from the singlet, 2^1S , and triplet, 2^3S , excited states of helium probes the role of symmetry in the two-electron dynamics. This is particularly important for quasiclassical approximations where symmetries and quantum interferences are difficult to include. With the exception of a few earlier studies in the high-energy limit [1, 2], it is only recently that the study of double photoionization from the $2^{1,3}\text{S}$ excited states of helium has attracted considerable theoretical interest. Experimental measurements of photon absorption cross sections from the helium excited states still remain a challenge. In these recent studies sophisticated fully numerical *ab initio* methods, namely the *R*-matrix method [3] and the convergent close-coupling method [4], are used to compute integral double-ionization cross sections, double-to-single ratios, and, only very recently, the time-dependent close-coupling method is used to compute single- and triple-differential cross sections as well [5]. However, due to the numerical nature of these sophisticated *ab initio* calculations, approximate methods are also essential in uncovering the underlying mechanism of the two-electron dynamics in the

double-escape process. Such approximate methods have been successfully developed for the double photoionization from the helium ground state [6, 7]. The current work focuses on an approximate method to describe double photoionization from the helium excited states.

In the double-photoionization process the redistribution of the energy, following the photon absorption, is often discussed in terms of two mechanisms, that is, knockout and shake-off dominant in the low- and high-energy limits, respectively. In the knockout mechanism, often referred to as the TS1 (two-step-one) process, one electron, the primary, absorbs the photon and undergoes a hard collision with the secondary electron, thus knocking it out of the atom. The knockout mechanism dominates at low energies where the interaction time of the two electrons is large and can be described classically [6, 8]. For knockout, electron correlations are essential in the final continuum state. On the other hand, in the shake-off mechanism the primary electron absorbs the photon and leaves the atom very fast without undergoing a direct interaction with the secondary electron. The secondary electron feels the sudden change in the atomic field and relaxes in one of the bound or continuum eigenstates of the remaining ion. The shake-off mechanism is quantum mechanical in nature and prevails at high energies where the interaction time of the two electrons is short. For shake-off, electron correlations are important in the initial state before the photon is absorbed.

In the following, we formulate the explicit but separate contributions of the two mechanisms for double photoionization from the $2^{1,3}\text{S}$ helium excited states for *any* excess energy by extending the model developed to separate the contribution of the two mechanisms for the helium ground state in [6]. The separation is achieved by a *quasiclassical* formulation of the knockout process. By construction it is free from any shake-off contribution which is purely *quantum mechanical*. This separation not only facilitates the calculation of double photoionization but offers considerable insight into the process concerning for example the similarity with electron impact ionization of He^+ . Compared to the ground state, formulating the separate contributions of the two mechanisms from the helium excited states is a much harder problem, since the different symmetry of the $2^{1,3}\text{S}$ states has to be accounted for in the model.

We express the two steps of the double-photoionization process from the helium excited states, absorption of the photon and subsequent redistribution of the energy, as

$$\sigma_X^{++} = \sigma_{\text{abs}} P_X^{++}, \quad (1)$$

where X stands for either knockout or shake-off and P_X^{++} is the double-electron-ionization probability. In what follows we discuss how to obtain P_{KO}^{++} and then P_{SO}^{++} for the $2^{1,3}\text{S}$ helium states.

2. Knockout ionization

After the photon absorption by the primary electron, we describe the subsequent evolution of the two electrons using the classical-trajectory Monte Carlo (CTMC) phase space method. CTMC has been successfully used to describe charged particle impact processes [9–13]. We model the initial phase space of the trajectories quantum mechanically using a Wigner distribution [14]. To do so, we first find the initial two-electron wavefunction. Since electron correlations are not important in the initial state of the knockout mechanism, we choose our initial state as the independent-electron wavefunction

$$\Psi_0(\mathbf{r}_1, \mathbf{r}_2) = \phi_{1s}^{Z_{1s}}(\mathbf{r}_1)\phi_{2s}^{Z_{2s}}(\mathbf{r}_2), \quad (2)$$

where $\phi_{1s}^{Z_{1s}}(\mathbf{r}_1) = (Z_{1s}^3/\pi)^{1/2}e^{-Z_{1s}r_1}$ and $\phi_{2s}^{Z_{2s}}(\mathbf{r}_2) = (Z_{2s}^3/(8\pi))^{1/2}(1 - Z_{2s}r_2/2)e^{-Z_{2s}r_2/2}$ are hydrogenic 1s and 2s orbitals with effective charges Z_{1s} and Z_{2s} , respectively. $\Psi_0(\mathbf{r}_1, \mathbf{r}_2)$ given

by equation (2) is not a symmetrized wavefunction; thus, we do not yet account for the different symmetries of the triplet 2^3S and the singlet 2^1S helium excited states. To find the effective charges Z_{1s} and Z_{2s} we use the two-electron ionization energies $I_{2^1\text{S}/2^3\text{S}}^{++} \approx 2.146(2.175)$ for the $2^1\text{S}(2^3\text{S})$ states given in [2], and the ionization energies of the 1s and 2s orbitals as follows. The energy needed to remove the 1s electron after the 2s electron has been ionized is 2 au and thus from $I_{1s} = Z_{1s}^2/2 = 2$ we find $Z_{1s} = 2$ for both the triplet and the singlet states. From $I_{2^1\text{S}/2^3\text{S}}^{++} - I_{1s} = I_{2s} = Z_{2s}^2/8$ we find $Z_{2s} \approx 1.081$ (1.184) for the 2^1S (2^3S) states, respectively. Atomic units are used throughout this paper unless otherwise indicated.

While for the $1s^2$ helium ground state the photon is necessarily absorbed by a 1s electron, for the $1s2s$ configuration of the $2^{1,3}\text{S}$ helium states the photon can be absorbed by a 1s or a 2s electron. For the values of the effective charges given above, one can show that the cross section is much larger for photon absorption from a 1s electron than from a 2s electron using the independent-electron picture [15]. Therefore, we take the primary electron to be that on the 1s orbital. With the PEAK approximation, exact in the high-photon-energy limit [16], we assume that the photon absorption happens directly at the nucleus. This significantly reduces the initial phase space to be sampled by the CTMC method.

The initial phase space distribution, $\rho(\Gamma)$, is the two-electron density immediately after the photon absorption given by

$$\rho(\Gamma) = N\delta(\mathbf{r}_1)\rho_2(\mathbf{r}_2, \mathbf{p}_2), \quad (3)$$

where N is a normalization constant. The initial distribution of the primary electron is $\delta(\mathbf{r}_1)$ (PEAK approximation) while that of the secondary electron is given by

$$\rho_2(\mathbf{r}_2, \mathbf{p}_2) = W_\psi(\mathbf{r}_2, \mathbf{p}_2)\delta(\epsilon_2^{\text{in}} - \epsilon_B), \quad (4)$$

where $W_\psi(\mathbf{r}_2, \mathbf{p}_2)$ is the Wigner distribution function of the two-electron wavefunction with the primary electron at the nucleus, $\mathbf{r}_1 = 0$,

$$\psi(\mathbf{r}_2) = \Psi_0(\mathbf{r}_1 = 0, \mathbf{r}_2)(\langle\Psi_0(\mathbf{r}_1 = 0, \mathbf{r}_2)|\Psi_0(\mathbf{r}_1 = 0, \mathbf{r}_2)\rangle)^{-1/2}. \quad (5)$$

In equation (4), we take the energy of the secondary electron immediately after photon absorption, ϵ_2^{in} , to be fixed on the 2s energy shell $\epsilon_B = -Z_{2s}^2/8$. From equation (2), it follows that $\epsilon_2^{\text{in}} = p_2^2/2 - Z_{2s}/r_2$. The excess energy available to the two-electron system after photon absorption is determined by the photon energy to be

$$E = \omega - I_{2^1\text{S}}^{++}, \quad E = \omega - I_{2^3\text{S}}^{++} \quad (6)$$

for the 2^1S and 2^3S states, respectively. Due to the PEAK approximation the primary electron can have any energy necessary so that, together with the initial energy ϵ_2^{in} of the secondary electron, it adds up to the excess energy E in equation (6). After modelling the initial phase space distribution, we propagate the electron trajectories using the classical equations of motion (CTMC). Regularized coordinates [17] are used for the propagation of the electron trajectories to avoid problems with trajectories starting at the nucleus ($\mathbf{r}_1 = 0$). The doubly ionized trajectories are those that end with the asymptotic energies of both electrons being positive. To evaluate the double-electron escape probability each trajectory is weighted by the initial phase space Wigner distribution.

So far we have treated the two electrons as distinguishable particles; that is, we distinguish between the primary and the secondary electron. Singlet and triplet probabilities require a proper symmetrization of the probability amplitude upon exchange of the two electrons. The probability amplitude, however, is not available from our purely classical propagation. Close to the threshold $E \rightarrow 0$, previous analysis has shown [18] that the symmetrized singlet/triplet

probabilities can be formulated using only classical information in the form

$$\frac{dP_{\text{KO}}^{++}}{d\epsilon} = \frac{1}{2} \left(\sqrt{\frac{dP_{\text{KO}}^{++}(\epsilon, E)}{d\epsilon}} \pm \sqrt{\frac{dP_{\text{KO}}^{++}(E - \epsilon, E)}{d\epsilon}} \right)^2, \quad (7)$$

where $dP_{\text{KO}}^{++}(\epsilon, E)/d\epsilon$ is the probability for both electrons escaping when the primary electron is ejected with energy $0 \leq \epsilon \leq E$. Since the KO mechanism is dominant for low energies we adopt equation (7) also for finite excess energies as an approximation. There it provides an upper bound for the difference between singlet and triplet contributions of the quasiclassically formulated KO mechanism. To evaluate $dP_{\text{KO}}^{++}(\epsilon, E)/d\epsilon$ we divide the energy interval $[0, \epsilon]$ into N equally sized bins and find the doubly ionized trajectories which fall into the bins. In our calculations we take $N = 21$ for excess energies up to 80 eV and $N = 27$ for higher excess energies. The double-ionization probability P_{KO}^{++} is obtained by integrating over all possible energies that an electron can be ejected with; that is,

$$P_{\text{KO}}^{++} = \int_0^E \frac{dP_{\text{KO}}^{++}}{d\epsilon} d\epsilon. \quad (8)$$

Note that for the case of the helium ground state the double-ionization probability P_{KO}^{++} is worked out without using the differential probabilities [6].

3. Shake-off ionization

Assuming that the primary electron is suddenly removed from the atom, Aberg [19] found that the probability for the shaken (secondary) electron to relax on a hydrogenic eigenstate of the remaining ion for *any* excess energy is

$$P_{\alpha}^{\nu} = |\langle \phi_{\alpha} | \psi^{\nu} \rangle|^2 / \langle \psi^{\nu} | \phi^{\nu} \rangle, \quad (9)$$

where $\psi^{\nu}(\mathbf{r}_2) = \int d^3\mathbf{r}_1 v^*(\mathbf{r}_1) \Psi_0(\mathbf{r}_1, \mathbf{r}_2)$ and $\nu(\mathbf{r}_1)$ is the primary electron wavefunction after it has left the atom. The primary electron is in an s state before the photon absorption and in a p state afterwards. $\Psi_0(\mathbf{r}_1, \mathbf{r}_2)$ is the initial state wavefunction of the $2^{1,3}\text{S}$ helium excited states and ϕ_{α} is a hydrogenic eigenstate of the bare nucleus ($Z = 2$) that is either a bound ($\alpha = n$) or a continuum state ($\alpha = \epsilon$). Aberg [19] has further shown that when the primary electron leaves the atom with very high energy ($\nu(\mathbf{r}_1) = (2\pi)^{-3/2} e^{-ik_1 \cdot \mathbf{r}_1}$), equation (9) takes the simplified form

$$P_{\alpha} = \frac{|\langle \phi_{\alpha} | \Psi_0(\mathbf{r}_1 = 0, \mathbf{r}_2) \rangle|^2}{\langle \Psi_0(\mathbf{r}_1 = 0, \mathbf{r}_2) | \Psi_0(\mathbf{r}_1 = 0, \mathbf{r}_2) \rangle}. \quad (10)$$

Equation (10) reveals the quantum mechanical nature of the shake-off process since it is expressed as an overlap of the initial bound state wavefunction and the final continuum state wavefunction.

Although equation (10) was derived in the high-energy limit, we assume that the primary electron absorbs the photon on the nucleus for all excess energies; that is, we adopt the PEAK approximation as in the knockout case. To find the double-escape probability P_{SO}^{++} we then integrate over all possible energies of the shaken electron in the continuum:

$$P_{\text{SO}}^{++}(E) = \int_0^E P_{\epsilon} d\epsilon. \quad (11)$$

We further simplify the evaluation of the shake-off probability in equations (10), (11) by taking the initial state to be the symmetrized wavefunction:

$$\Psi_0(\mathbf{r}_1, \mathbf{r}_2) = N_1 \left(\phi_{1s}^{Z_{\text{so}}}(\mathbf{r}_1) \phi_{2s}^{Z_{\text{so}}}(\mathbf{r}_2) \pm \phi_{1s}^{Z_{\text{so}}}(\mathbf{r}_2) \phi_{2s}^{Z_{\text{so}}}(\mathbf{r}_1) \right) \quad (12)$$

for the singlet and triplet states, respectively, with N_1 a normalization constant. The initial state correlations are accounted for only through the effective charges. We next assign the same set of effective charges Z_{SO}^1 and Z_{SO}^2 for both the triplet and the singlet states as follows. The asymptotic ratio (high-energy limit) of double-to-single ionization is found very accurately in [2] to be $R_\infty = 0.009\,033/0.003\,118$ for the singlet/triplet states, where in our model R_∞ is given by

$$R_\infty = P_{\text{SO}}^{++}(E \rightarrow \infty)/(1 - P_{\text{SO}}^{++}(E \rightarrow \infty)), \quad (13)$$

and

$$P_{\text{SO}}^{++}(E \rightarrow \infty) = \int_0^\infty P_\epsilon \, d\epsilon = 1 - \sum_n P_n. \quad (14)$$

Using equations (13), (14), and the symmetrized wavefunctions in equation (12) we find the sets of charges that match both asymptotic ratios R_∞ for the singlet and the triplet states. We then select that set of charges for which the shake-off double-ionization probability as a function of the excess energy, obtained using the simple wavefunctions given in equation (12), is closest to the one obtained using the fully correlated Hylleraas wavefunctions given in [20]. The set of charges thus found is $Z_{\text{SO}}^1 \approx 1.757$ and $Z_{\text{SO}}^2 \approx 1.728$. The reason we do not use the Hylleraas wavefunctions given in [20] is that they do not reproduce the accurate asymptotic ratios obtained in [2] using highly accurate Pekeris-type wavefunctions. We emphasize though that one does not need to use the approximate wavefunctions in equation (12) to compute the double-ionization probability; highly accurate wavefunctions that reproduce the correct asymptotic ratios could be used instead.

For the shake-off probability, P_ϵ in equation (10) already gives the differential double-ionization probability. Despite the symmetrization in equation (12) we have lost the indistinguishability of the electrons by identifying one electron as the primary one which absorbs the photon. Thus we need to symmetrize again in the final state with respect to the equal-energy-sharing point $\epsilon = E - \epsilon = E/2$ [6]. That is,

$$\frac{dP_{\text{SO}}^{++}}{d\epsilon} = \frac{1}{2}(P_\epsilon + P_{E-\epsilon}). \quad (15)$$

4. Results and conclusions

4.1. Photoionization ratios

According to equation (1) $\sigma^{++} = \sigma_{\text{abs}}(P_{\text{KO}}^{++} + P_{\text{SO}}^{++})$ and $\sigma^+ = \sigma_{\text{abs}} - \sigma^{++}$. Thus, the double-to-single-ionization ratio is given by

$$\sigma^{++}/\sigma^+ = P^{++}/(1 - P^{++}), \quad (16)$$

where $P^{++} = P_{\text{KO}}^{++} + P_{\text{SO}}^{++}$. In figure (1) we compare the double-to-single ratio for the $2^{1,3}\text{S}$ helium excited states with the results obtained by Kheifets *et al* [4] using the convergent close-coupling method and show that there is a very good agreement. The agreement is better for the 2^3S state. We find that the deviation occurs, particularly for the 2^3S state, at photon energies where the contributions of knockout and shake-off mechanisms are comparable. At these energies any interference between the knockout and shake-off mechanism would have its largest effect. So, it may be that the deviations that we see are due to that interference effect that we do not account for in our calculation, since we add the knockout and shake-off contributions incoherently. For the 2^1S state a maximum of $\approx 2.84\%$ is reached at 14 eV above the double-ionization threshold of the 2^1S state. For the 2^3S state a maximum of $\approx 0.69\%$ is reached at 60 eV above the ionization threshold of the 2^3S state. In figure 1 we see that at high

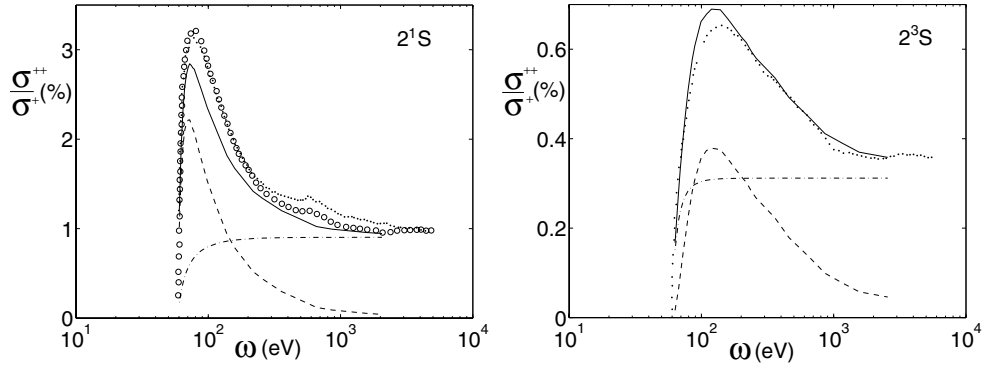


Figure 1. Double-to-single-ionization ratio as a function of the photon energy. Dots/open circles indicate the results of Kheifets *et al* [4] in the velocity/acceleration gauge. For the triplet state, Kheifets results in the two gauges are indistinguishable to the scale of the figure. Our results are indicated by a solid curve for the total, by a dashed curve for the knockout, and by a dashed-dot curve for the shake-off double-to-single ratio.

energies the knockout contribution goes to zero as expected, and the shake-off contribution dominates and reaches the asymptotic limit of 0.009 033/0.003 118 for the singlet/triplet states. Stronger correlation effects for the singlet symmetry ($r_1 = r_2$ is not forbidden as is the case for the triplet) result in a much higher double-to-single-ionization ratio compared to the triplet case. From figure (1), for the $2^{1,3}S$ states, and [6], for the helium ground state $1S$, we see that as we go along $1S \rightarrow 2^1S \rightarrow 2^3S$ the electron–electron correlation becomes smaller. That is, for the $2^{1,3}S$ states, the $1s$ and $2s$ electrons are better separated than the $1s^2$ electrons in the $1S$ state. In addition, in the 2^3S state the electron separation is even stronger because of the Pauli exclusion principle. Thus, as we go along $1S \rightarrow 2^1S \rightarrow 2^3S$, the energy sharing becomes more asymmetric with one fast and one slow electron leaving the atom favouring the shake-off over the knockout mechanism at smaller excess energies.

4.2. Single differentials

To compute the single-differential probabilities for the helium excited states we use

$$\frac{dP^{++}}{d\epsilon} = \frac{dP_{\text{KO}}^{++}}{d\epsilon} + \frac{dP_{\text{SO}}^{++}}{d\epsilon}. \quad (17)$$

In addition we compute the single-differential cross sections using

$$\frac{d\sigma^{++}}{d\epsilon} = \sigma_{\text{abs}} \frac{dP^{++}}{d\epsilon}, \quad (18)$$

where for σ_{abs} we use the results for the total photoionization cross section for the 2^1S and the 2^3S states given in [4]. In figure 2, we compare our results for $d\sigma^{++}/d\epsilon$ with the results obtained very recently by Colgan *et al* [5] using the time-dependent close-coupling method for four values of the excess energy at 10, 20, 30, and 40 eV. We see that our results for the single-differential cross section as a function of the ejected electron energy normalized by the excess energy are smaller for the 2^1S state, while there is an excellent agreement for the 2^3S state. Again as we go along $1S \rightarrow 2^1S \rightarrow 2^3S$ the single-differential cross sections become more U-shaped for the same excess energy. The reason is again that the electron–electron correlation decreases, thus favouring the ejection of one fast and one slow electron—that is, favouring unequal energy sharing.

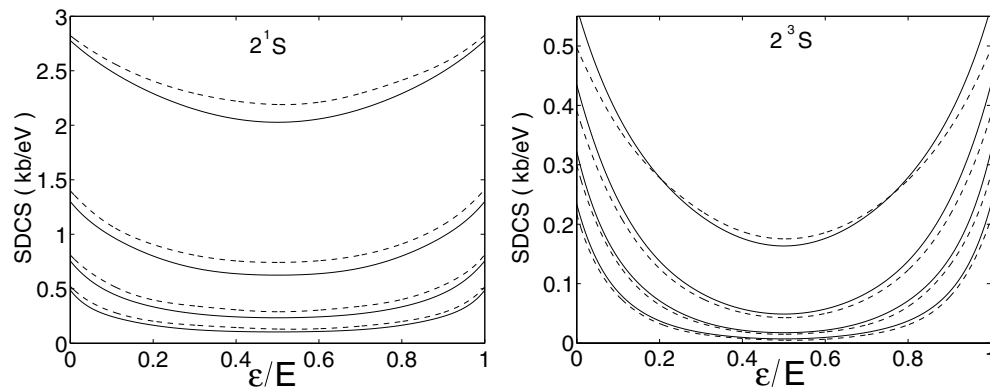


Figure 2. Absolute single-differential cross sections, at excess energies 10, 20, 30, and 40 eV from top to bottom, as a function of the electron ejected energy scaled by the excess energy. The dashed curves are the results given by Colgan *et al.* Our results are indicated by solid curves.

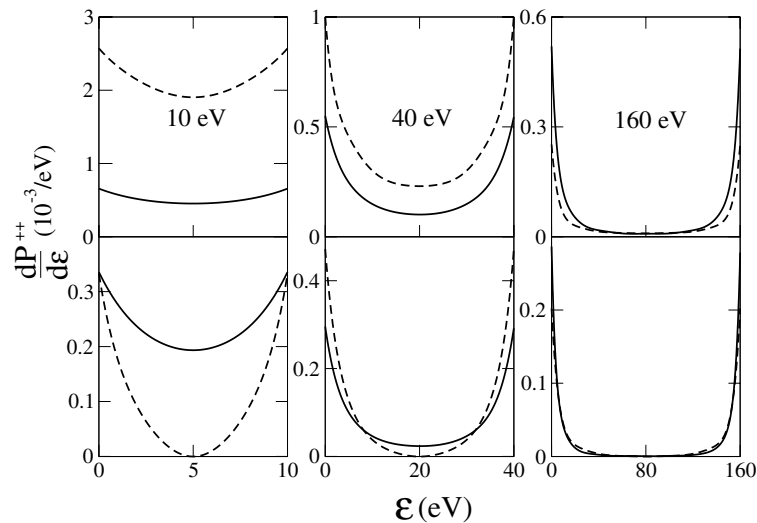


Figure 3. Absolute single-differential probabilities as a function of the ejected electron energy. The knockout contribution is indicated by dashed curves while the shake-off is indicated by solid curves. The top panel is for the 2^1S state while the bottom one is for the 2^3S state.

In figure (3), using equation (17), we show the separate contribution of knockout and shake-off to the single-differential probabilities, $\frac{dP^{++}}{d\epsilon}$, for the $2^{1,3}S$ helium states for excess energies 10, 40, and 160 eV. For the singlet state the knockout contribution dominates at small excess energies, 10 and 40 eV, while as the excess energy is increased to 160 eV the shake-off contribution begins to dominate regions of unequal energy sharing. For the triplet case the shake-off mechanism is already significant at small excess energies. Note, that the knockout contribution for the triplet case is zero at the equal-energy-sharing point, $\epsilon = E - \epsilon$, because of the symmetrization with respect to the two identical electrons; see equation (7).

4.3. Conclusions

In conclusion, we have shown that the double ionization from the $2^{1,3}S$ states can be accurately described by separate formulations and calculations of the knockout and shake-off mechanism at any excess energy. In comparison to the helium ground state [6], this presents a harder problem because we have to account for the different symmetries of the singlet and triplet states. The success of this simple model for describing double ionization from the helium ground state as well as the helium excited states is proof of its validity. In the future, we plan to use this simple model to describe triple-photoionization cross sections.

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