One-photon double ionization of helium: a heuristic formula for the cross section

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Without a formal derivation, we propose a formula for the total and single-differential cross in the problem of one-photon double ionization of an atom. The formula is benchmarked against accurate experimental data for the total cross section of helium. Furthermore, a direct comparison with ab initio calculations for the double ionization of Li$^+$ suggests that the framework is valid for the entire helium isoelectronic sequence. To this end, we introduce a formula for the double ionization of lithium, as well as for the triple ionization of lithium and beryllium.

PACS numbers: 32.80.Rm, 32.80.Fb, 42.50.Hz

Double photoionization of helium by a single photon has been studied for a period of more than 40 years since the pioneering work of Byron and Joachain [1], who pointed out the importance of electron correlation in the process. Such correlated processes pose many challenges, and it is only during the last 15 years or so that quantitative agreement between experiment [2] and theory [3–16] for the total cross section was obtained, and a rather complete understanding of the breakup process in helium has emerged [17–25]. A simple analytical formula for the shape of single-photon multiple ionization cross sections was proposed by Pattard [26]. This shape function contains two parameters, the position and height of the cross section maximum.

In this work, we propose a formula for the single-differential cross section in the problem of one-photon double ionization of an atom. The formula contains a scaling factor that determines the height of the cross section maximum. The present work is partly motivated by the idea behind a recent model for direct (nonsequential) two-photon double ionization of helium [27], i.e., that the explicit form of the electron-electron interaction is not of crucial importance in order to obtain a qualitative description of the electrons’ route to the double continuum, as far as the total and single-differential cross sections are concerned. It is here argued that the corresponding one-photon double ionization event is similarly dictated by the electrons’ electric dipole couplings to their respective single-particle continua, rather than the Coulombic interaction between the electrons. As such, the assumption is that the electron-electron interaction merely plays the role of distributing the excess energy between the ejected electrons in the excitation process, assuring that the total energy of the system is conserved. Keeping in mind that the electrons are emitted more or less simultaneously (in coincidence) in the double ionization process, we simply assume that there is essentially no time for the electrons to explore the explicit geometric form of the repulsive potential at the instant of ionization, which again suggests that the electron-electron interaction can be handled in a simple and approximate way. The simplest possible approximate model interaction that allows for double ionization by photon impact (to first order in perturbation theory), controls the energy given to each electron, and which is symmetric with respect to both electrons and dipole-like for each electron independently, can be written on the following heuristic form

$$H_{int} \propto E(t)z_1z_2.$$  \hspace{1cm} (1)

Here $E(t)$ is the electric field modeling the laser pulse, which for simplicity is assumed to be z-polarized, and $z_1$ and $z_2$ are the z-coordinates of electron 1 and 2, respectively. Although we give no formal proof of the assertion Eq. (1), we will nevertheless make use of it in the following to derive an explicit formula for the single-differential cross section in the process of double photoionization of helium and compare it with accurate experimental data.

However, before proceeding, we would like to emphasize that the model interaction in Eq. (1) is heuristic in nature and should be used with caution. For example, it fails completely in describing the evolution of the system in the time after the electrons have been emitted into the continuum, and as such it would in general yield incorrect angular distributions. On the other hand, by construction the interaction allows for the possibility that the electrons can absorb the photon as a unified system, concordant with the model of Førre et al. [27].

In the next step of approximation, both electrons are considered to be independent particles and the ground (initial) state wave function of the helium atom is simply approximated by the product ansatz

$$\Psi_f(r_1, r_2) = \psi_{1s}(r_1) \psi_{1s}(r_2),$$  \hspace{1cm} (2)

where $\psi_{1s}$ refers to the ground state of the He$^+$-ion. It should be noted that the overlap between this approximate wave function and the real ground state wave function of helium is more than 90%. As such, the assumption is that most of the essential features relevant for the double ionization process in helium are captured in the simplified wave function. This is a crucial point in the model presented here. Likewise, the final state wave function is approximated by a symmetrized product of

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two He\(^+\) continuum states,
\[ \Psi_f(r_1,r_2) = \frac{1}{\sqrt{N_e!}} \left[ \psi_{E_1}(r_1)\psi_{E_2}(r_2) + \psi_{E_2}(r_1)\psi_{E_1}(r_2) \right], \]
with \( N_e = 2 \) being the number of electrons involved in the ionization process.

Applying lowest order perturbation theory to the resulting system, with the interaction defined in Eq. (1), and the initial and final states defined in Eqs. (2) and (3), the resulting single-differential cross section for double photoionization of helium takes the form
\[ \frac{d\sigma}{dE_1} \propto \hbar\omega \left| \langle \psi_f | z_1 z_2 | \psi_i \rangle \right|^2 \]
\[ = \frac{4}{N_e!} \hbar\omega \left| \langle \psi_{E_1} | z | \psi_{1s} \rangle \right|^2 \left| \langle \psi_{E_2} | z | \psi_{1s} \rangle \right|^2, \]
with
\[ E_1 + E_2 = \hbar\omega - E_b, \]
\( \hbar\omega \) being the photon energy and \( E_b = 79 \text{ eV} \) the binding energy of helium. As such, the total binding energy of the system is not considered a free parameter in the present work. The coupling elements in Eq. (4) are related to the well known one-photon (one-electron) photoionization cross section of He\(^+\) [28] via the relation [29]
\[ \sigma_\text{He}^+ \propto (E - E_{1s}) \left| \langle \psi_{E} | z | \psi_{1s} \rangle \right|^2, \]
where \( E_{1s} \) is the energy of the He\(^+\) ground state, and \( \sigma_\text{He}^+ \) is the photoionization cross section of He\(^+\).

Combining Eqs. (4) and (6), we propose the following formula for the single-differential cross section in the process of one-photon double ionization of helium,
\[ \frac{d\sigma}{dE_1} = \frac{C}{N_e!} \frac{\hbar\omega}{4a_0^2} \frac{\sigma_\text{He}^+ (E_1 - E_{1s})}{E_1 - E_{1s}} \frac{\sigma_\text{He}^+ (E_2 - E_{1s})}{E_2 - E_{1s}}, \]
where \( E_1 + E_2 = \hbar\omega - E_b \), \( C \) is a yet unknown dimensionless constant, and \( a_0 \) is the Bohr radius. Applying Eq. (7), the single-electron photoionization cross section of He\(^+\) is multiplied by two to account for the statistical weight of having two identical electrons in the 1s orbital initially. Furthermore, the presence of the energy factors in the denominators is related to the fact that the photon is absorbed simultaneously by both electrons via a nonresonant transition to the final state of each electron [27].

The one-photon double ionization process of helium has been investigated in length in both theoretical and experimental studies, resulting in close quantitative agreement in the total cross sections. In Fig. 1 we compare the total (integrated) cross section obtained with Eq. (7), choosing \( C = 1 \), with the accurate experimental data of Samson et al. [2], who stated the accuracy of their results to be within \( \pm 2\% \). Provided we choose the value of the (unknown) constant \( C = 1 \), the model predictions is, within the experimental uncertainty, in almost exact agreement with the experimental data over the entire interval of photon energies considered. The value of the parameter \( C \) is now determined and will no longer be considered a free parameter in the rest of this work. Finally, we would like to note that formula (7) also yields single-differential cross sections that are in good agreement with calculated and measured data for helium.

Figure 2 shows the result of formula Eq. (7) when applied on the problem of double ionization of Li\(^+\), inserting the corresponding photoionization cross sections of Li\(^{2+}\) into the equation. Theoretical results by Kheifets and Bray [7], van der Hart and Feng [12], and Kleiman et al. [30] are included.
for comparison, and the formula seems to be consistent with the calculated data, suggesting that it is valid for the entire helium isoelectronic sequence.

In order to test the validity of the theoretical framework further, we now turn to the more challenging case, namely the double photoionization of lithium. The assumption is that it is the outer weakly bound 2s electron and one of the tightly bound 1s electrons that are emitted. This suggests the following formula for the single-differential cross section,

\[ \frac{d\sigma}{dE_1} = \frac{\hbar \omega}{8a_0^2} \frac{\sigma_{2s}^J(E_1 - E_{1s}) \sigma_{L_i}^J(E_2 - E_{2s})}{E_1 - E_{1s} E_2 - E_{2s}}, \]  

where \( E_{1s} = -7.56 \) eV and \( E_{2s} = -5.4 \) eV are the effective (single-electron) energies (the negative of the ionization potential) of the 1s (inner) and 2s (outer) electrons, and \( \sigma_{2s}^J \) and \( \sigma_{L_i}^J \) are the one-photon single-ionization cross sections of \( \text{Li}^+ \) and \( \text{Li} \), respectively. Figure 3 depicts the results for the total cross section for double ionization of lithium, as obtained by integrating Eq. (8), and a comparison with previously obtained experimental [31–33] and theoretical data [34]. It turns out that the formula yields results that are in good agreement with the experimental measurements, which is somewhat surprising given the high complexity of the problem.

Finally, we consider the problem of triple ionization of lithium and beryllium. We do this to show that it is relatively straightforward to generalize the framework to consider multiple ionization processes. As a matter fact, combining the cross section for the double ionization of \( \text{Li}^+ \), as obtained in Fig. 3 together with the photoionization cross section of neutral lithium, \( \sigma_{Li} \), according to the rule in Eq. (7), the triple photoionization cross section of lithium is simply given by

\[ \frac{d\sigma}{dE_3} = \frac{\hbar \omega}{24a_0^2} \frac{\sigma_{Li}^J(E_{12} - E_{Li^+}) \sigma_{L_i}^J(E_3 - E_{2s})}{E_{12} - E_{Li^+} E_3 - E_{2s}}. \]  

Here \( \sigma_{Li}^J \) denotes the double photoionization cross section of \( \text{Li}^+ \) (Fig. 2), \( E_{Li^+} = -198 \) eV is the total energy of the two bound 1s (inner) electrons, \( E_{2s} = -5.4 \) eV is the energy of the 2s (outer) electron, \( E_{12} = E_1 + E_2 \), and \( E_{12} + E_3 = \hbar \omega + E_{Li^+} + E_{2s} \) is the total excess energy shared by the electrons in the continuum.

The result for triple ionization of lithium is shown in Fig. 4.
The triple photoionization cross section of beryllium is correspondingly given by

$$\frac{d\sigma}{dE_3} = \frac{\hbar \omega}{24a_0^3} \frac{\sigma_{Be}^{D}(E_{12} - E_{Be})}{E_{12} - E_{Be}} \frac{\sigma_{Be^{2+}}(E_{3} - E_{Be^{2+}})}{E_{3} - E_{Be^{2+}}},$$

(10)

with $\sigma_{Be}^{D}$ being the double photoionization cross section of Be (as calculated by Eq. (7) inserting the photoionization cross section of Be$^{+}$), $\sigma_{Be^{2+}}$ the one-photon single-ionization cross section of Be$^{2+}$, $E_{Be} = -27.5$ eV the total energy of the two (outer) bound 2s electrons, and $E_{Be^{2+}} = -153.9$ eV the effective (single-electron) energy (the negative of the ionization potential) of the (active) 1s electron. Furthermore, $E_{12} = E_{1} + E_{2}$, and $E_{12} + E_{3} = \hbar \omega + E_{Be} + E_{Be^{2+}}$ is the total excess energy shared by the three electrons in the continuum.

The lower panel in Fig. 4 depicts the result for beryllium together with the theoretical calculations by Colgan et al. [36] and Kheifets and Bray [35]. Quite interestingly, the prediction of the formula is again in favor of the result of the double shake-off model by Kheifets and Bray [35], as far as the total ionization yield is concerned, but further theoretical and experimental investigations are required in order to settle the problem definitely.

In conclusion, without a formal derivation, we have proposed a formula for the single-differential cross section in the problem of one-photon double ionization of He, Li$^+$ and Li. The corresponding function contains an unknown (dimensionless) constant that dictates the height of the cross section maximum. The value of the constant was determined in helium by fitting with the experimental data of Samson et al. [2], and the same value for the constant was used in the other systems. Provided a qualified guess for the initial state is taken, the resulting parametrization is shown to yield results in near quantitative agreement with experimental and theoretical data for all considered cases. Finally, the problem of triple photoionization of lithium and beryllium is studied. It is demonstrated that agreement with experimental and theoretical results can be obtained. Furthermore, although not shown here, our results are consistent with the general shape function proposed by Pattard [26] and the results of the half-collision model by Pattard and Burgdörfer [25, 42].

As a final remark, we would like to add that it is relatively straightforward to generalize the framework to account for multiple ionization processes involving more than two or three electrons, like e.g. the process of quadruple ionization of beryllium, which is a problem that is difficult to pursue within the framework of more rigorous \textit{ab initio} methods.

Acknowledgments

This work was supported by the Bergen Research Foundation (Norway). The author gratefully acknowledges discussions with Peter Lambropoulos, Ladislav Kochbach, Sølve Selstø and Aleksander Simonsen, and would like to thank Ralf Wehlitz, Hugo W. van der Hart, Emmanuel Foumouo and Bernard Piraux for sending their data in numerical form.

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