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 sections 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.

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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 paper, we propose a formula for the single-differential cross section in the process of one-photon double ionization of an atom. The formula contains a scaling factor that determines the height of the cross-section maximum. Provided the value of this parameter is set to 1, it is shown that the formula yields cross sections that are in agreement with both theoretical and experimental double-photoionization data for He, Li^+, and Li. Furthermore, it is demonstrated how the framework can be generalized to account for triple photoionization of lithium and beryllium.

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 is symmetric with respect to both electrons and dipolelike for each electron independently, can be written on the following heuristic form:

$$H_{\rm int} \propto E(t) z_1 z_2. \tag{1}$$

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 electrons 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, the two 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

$$\Psi_i(\mathbf{r}_1,\mathbf{r}_2) = \psi_{1s}(\mathbf{r}_1)\psi_{1s}(\mathbf{r}_2), \tag{2}$$

where ψ_{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

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wave function is approximated by a symmetrized product of two He^+ continuum states,

$$\Psi_f(\mathbf{r}_1,\mathbf{r}_2)=\frac{1}{\sqrt{N_e!}}[\psi_{E_1}(\mathbf{r}_1)\psi_{E_2}(\mathbf{r}_2)+\psi_{E_2}(\mathbf{r}_1)\psi_{E_1}(\mathbf{r}_2)], \quad (3)$$

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), we obtain for the resulting single-differential cross section for double photoionization of helium

$$\frac{d\sigma}{dE_1} \propto \hbar\omega|\langle\Psi_f|z_1z_2|\Psi_i\rangle|^2 = \frac{4}{N_e!}\hbar\omega|\langle\psi_{E_1}|z|\psi_{1s}\rangle|^2|\langle\psi_{E_2}|z|\psi_{1s}\rangle|^2, \quad (4)$$

with

$$E_1 + E_2 = \hbar\omega - E_b, \quad (5)$$

$\hbar\omega$ being the photon energy and $E_b = 79$ eV the binding energy of helium. Thus 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]

$$\sigma_{He^+} \propto (E - E_{1s})|\langle\psi_E|z|\psi_{1s}\rangle|^2, \quad (6)$$

where E_{1s} is the energy of the He^+ ground state, and σ_{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:

$$\frac{d\sigma}{dE_1} = \frac{C}{N_e!}\frac{\hbar\omega}{4a_0^2}\frac{\sigma_{He^+}(E_1 - E_{1s})}{E_1 - E_{1s}}\frac{\sigma_{He^+}(E_2 - E_{1s})}{E_2 - E_{1s}}, \quad (7)$$

where $E_1 + E_2 = \hbar\omega - E_b$, C is a yet unknown dimensionless constant, and a_0 is the Bohr radius. In application of Eq. (7), the single-electron photoionization cross section of He^+ is multiplied by 2 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 ±2%. Provided we choose the value of the (unknown) constant $C = 1$, the model prediction 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 it 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

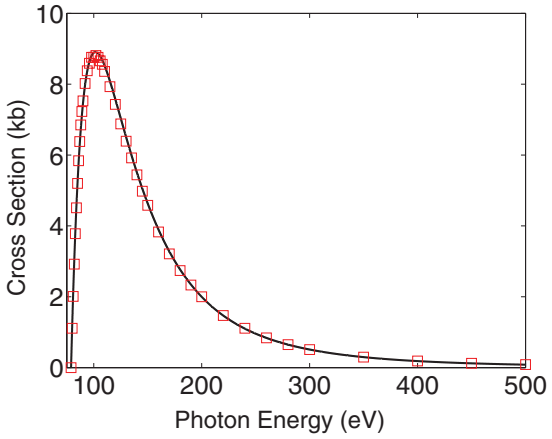

FIG. 1. (Color online) Double-photoionization cross section of helium versus photon energy. Black line: model result Eq. (7) with $C = 1$. Red squares: experimental results of Samson *et al.* [2].

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 to the problem of double ionization of Li^+, with the corresponding photoionization cross sections of Li^{2+} inserted 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:

$$\frac{d\sigma}{dE_1} = \frac{\hbar\omega}{8a_0^2}\frac{\sigma_{Li^+}(E_1 - E_{1s})}{E_1 - E_{1s}}\frac{\sigma_{Li}(E_2 - E_{2s})}{E_2 - E_{2s}}, \quad (8)$$

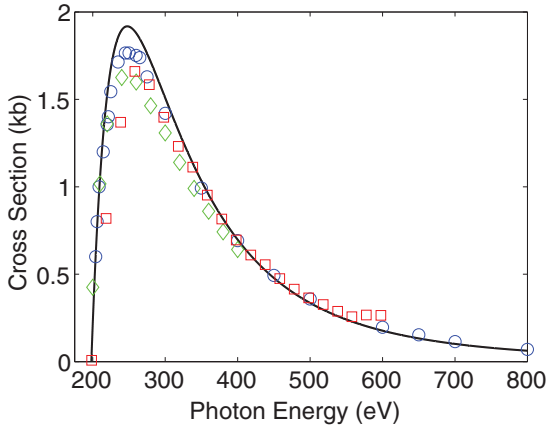

FIG. 2. (Color online) Double-photoionization cross section of Li^+ versus photon energy. Black line: model result. Red squares: theoretical results by van der Hart and Feng [12]. Blue circles: theoretical results by Kheifets and Bray [7]. Green diamonds: theoretical results by Kleiman *et al.* [30].

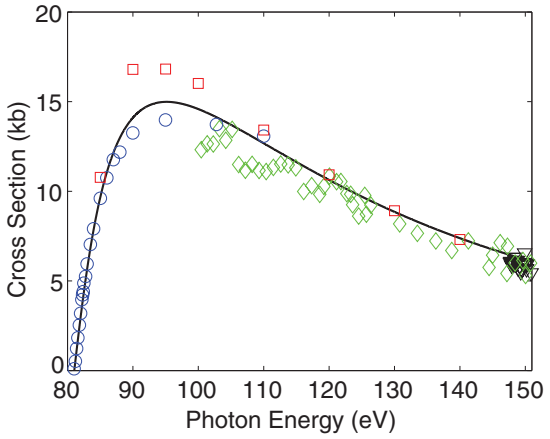

FIG. 3. (Color online) Double-photoionization cross section of lithium versus photon energy. Black line: model result Eq. (8). The absolute one-photon photoionization cross sections of Li and Li^+ are taken from [28]. Red squares: theoretical results by Colgan *et al.* [34]. Blue circles: experimental results by Wehlitz *et al.* [32]. Green diamonds: experimental results by Huang *et al.* [31]. Black triangles: experimental results by Wehlitz and Juranić [33].

where $E_{1s} = -75.6$ 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 σ_{Li^+} and σ_{Li} are the one-photon single-ionization cross sections of Li^+ and Li, respectively [28]. 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, combination of the cross section for the double ionization of Li^+, as obtained in Fig. 2, with the photoionization cross section of neutral lithium, σ_{Li}, according to the rule in Eq. (7), gives the triple photoionization cross section of lithium simply by

$$\frac{d\sigma}{dE_3} = \frac{\hbar\omega}{24a_0^2}\frac{\sigma^D_{Li^+}(E_{12}-E_{Li^+})}{E_{12}-E_{Li^+}}\frac{\sigma_{Li}(E_3-E_{2s})}{E_3-E_{2s}}. \qquad (9)$$

Here $\sigma^D_{Li^+}$ denotes the double-photoionization cross section of 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 (upper panel) and compares well with both experimental [35–37] and theoretical [38–41] data. Interestingly, the results are in close agreement with the predictions of the double-shake-off model for triple photoionization, proposed by Kheifets and Bray [38], in particular for the higher photon energies.

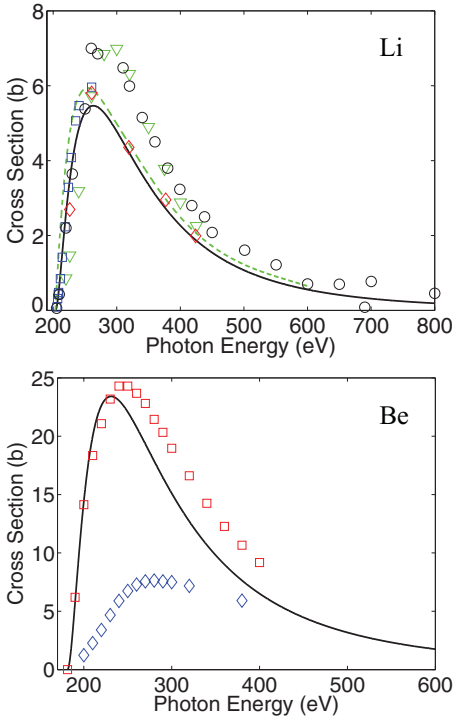

FIG. 4. (Color online) Upper panel: Triple-photoionization cross section of lithium versus photon energy. Black line: model result Eq. (9). Green dashed line: theoretical results by Kheifets and Bray [38]. Green triangles: theoretical results by Colgan *et al.* [39]. Red diamonds, blue squares, and black circles: experimental results by Wehlitz *et al.* [35–37]. Lower panel: Triple-photoionization cross section of beryllium versus photon energy. Black line: model result Eq. (10). Red squares: theoretical results by Kheifets and Bray [38]. Blue diamonds: theoretical results by Colgan *et al.* [39].

The triple-photoionization cross section of beryllium is correspondingly given by

$$\frac{d\sigma}{dE_3} = \frac{\hbar\omega}{24a_0^2}\frac{\sigma^D_{Be}(E_{12}-E_{Be})}{E_{12}-E_{Be}}\frac{\sigma_{Be^{2+}}(E_3-E_{Be^{2+}})}{E_3-E_{Be^{2+}}}, \qquad (10)$$

with σ^D_{Be} being the double-photoionization cross section of Be [as calculated by Eq. (7) with the photoionization cross section of Be^+ inserted], $\sigma_{Be^{2+}}$ 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.* [39] and Kheifets and Bray [38]. Quite interestingly, the prediction of the formula is again in favor of the result of the double-shake-off model by Kheifets and Bray [38], 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 was studied. It was 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, such as, e.g., the process of quadruple ionization of beryllium, which is a problem that is difficult to pursue within the framework of more rigorous *ab initio* methods.

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