Nonsequential Two-Photon Double Ionization of Atoms: Identifying the Mechanism

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We develop an approximate model for the process of direct (nonsequential) two-photon double ionization of atoms. Employing the model, we calculate (generalized) total cross sections as well as energy-resolved differential cross sections of helium for photon energies ranging from 39 to 54 eV. A comparison with results of ab initio calculations reveals that the agreement is at a quantitative level. We thus demonstrate that this complex ionization process is fully described by the simple model, providing insight into the underlying physical mechanism. Finally, we use the model to calculate generalized cross sections for the two-photon double ionization of neon in the nonsequential regime.

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Correlated dynamical processes in nature poses unique challenges to experiments and theory. A prime example of this is the double ionization of helium by one-photon impact, which has been studied for more than 40 years. However, it is only during the last 15 years or so, that advances in theory, modeling and experiment have enabled scientists to gain a deeper insight into the role of electron correlations in this ionization process [1–5]. The corresponding problem of two-photon double ionization of helium, in the photon energy interval between 39.4 and 54.4 eV, is an outstanding quantum mechanical problem that has been, and still is, subject to intense research worldwide, both theoretically [5–19] and experimentally, employing state-of-the-art high-order harmonic [20–22] and free-electron (FEL) light sources [23–24]. Despite all the interest and efforts that have been put into this research, major fundamental issues remain unresolved. What characterizes this particular three-body breakup process is that the electron correlation is a prerequisite for the process to occur, i.e., it depends upon the exchange of energy between the outgoing electrons, and as such it represents a clear departure from an independent-particle picture.

In this Letter, we present a novel approximate model for the direct or nonsequential two-photon double ionization process in helium, sketched in Fig. 1 (a). We show that the simple model predicts the essential features of the process, even at a quantitative level, which is quite surprising given the very high complexity of the problem. In particular, we find very good agreement between the model predictions and the results obtained by solving the time-dependent Schrödinger equation from first principles, regarding (generalized) total cross sections as well as energy-resolved differential cross sections for the process. The proposed model may be generalized to account for direct double ionization processes in multi-electron atoms. We demonstrate this by calculating the generalized cross section for nonsequential two-photon double ionization of neon.

Few-photon multiple ionization of noble gases beyond helium have been studied experimentally in some detail [23,26], but to the best of our knowledge, the cross section for the nonsequential two-photon double ionization process has not yet been obtained. Therefore, we hope that our results will encourage further investigation of nonsequential double ionization processes in various noble gases.

Reducing a complex quantum mechanical problem to a simple and transparent model problem, while retaining the essential physics, is very useful in order to access the underlying physics [4,27,28]. With such a goal in mind, we will now outline a possible physical mechanism for the nonsequential two-photon double ionization process in an atom, and then proceed to construct a simple quantum mechanical model which implements these ideas. The idea behind the model is that the electrons are considered to be distinguishable particles that can absorb one photon each. However, in order to include the effect of the first emitted electron on the second one, we impose the additional but important constraint that the absorption of the second photon, by the second electron, can only occur after the first photon absorption. In this way, and according to the principle of conservation of energy, the first electron may transfer energy to the second electron as it is emitted, allowing for the nonsequential ionization process to take place.

The starting point of our model is the single-active electron approximation (SAE) where both electrons are considered to be independent particles and treated differently in that they are both assumed to move in their respective ionization potentials. That is, the ‘outer’ electron moves in an effective potential set up by the nucleus of charge Ze (e is the elementary charge), the ‘inner’ electron and the Z – 2 other electrons. The inner electron sees a corresponding screened potential given by the nucleus and the Z – 2 remaining electrons. We will label these two different cases simply by ‘A’ and ‘B’, respectively. Following this procedure, the wave function of the ground state may be approximated by the product ansatz

\[ \Psi(r_A, r_B) = \psi_A(r_A)\psi_B(r_B), \] (1)

where \( \psi_A \) and \( \psi_B \) refer to the one-electron wave function of electron A and B, respectively.

Now, the first ionization event in the direct two-photon double ionization process can be represented by the one-electron dipole coupling between the ground state wave function of either A or B, i.e., the state |A, E_A^0\rangle or |B, E_B^0\rangle, and their respec-
FIG. 1. (color online). a) Sketch of the direct two-photon double ionization process in helium. The abbreviation SI and DI stands for single and double ionization continuum, respectively, whereas the arrows illustrate the photons that are absorbed by the system. b) Sketch of the model process for two-photon double ionization (see text for details). c) Matrix representation of the model Hamiltonian, for the case where the outer electron is emitted before the inner electron (see text for more details). Atomic units (a.u.) are used in the figure (1 a.u. of energy corresponds to 27.2 eV).

At the instant of ionization of electron $A$, electron $B$ remains unaffected. However, once electron $A$ has absorbed its photon, we allow for the possibility that electron $B$ (but not $A$) can be hit by a second photon. This secondary process is included into the model by introducing additional dipole couplings between the $B$ ground state and its corresponding one-electron continuum states in the following way:

$$\langle B, E_B^0 | - eE(t) \cdot r_B | B, E_B^0 \rangle \delta(E_A, E_A')$$

(4)

Note here that there are only non-vanishing couplings between SAE states (system $A$) of the same energy, i.e., the resulting coupling matrix attains a very simple structure, as shown in Fig. 1 with typically only a few hundred different couplings. The same procedure may also be followed with $A$ and $B$ interchanged, however, this will necessarily yield the same result, and therefore need not explicitly be considered.

The couplings (2) and (4) and the mentioned constraints, along with the corresponding diagonal energies, constitute the entire model that we propose. To this end, we would like to add that all excited, bound states have been left out of the model, as they play no role in the present context. As a matter of fact, despite the extremely simple form of the model matrix elements, with no explicit presence of the correlation potential, it actually allows for the possibility that the two electrons exchange energy in the excitation process. Thus, both electrons may be emitted into the continuum even though the energy of the secondary photon may not itself be sufficient to eject the inner electron into the continuum.

Applying second order perturbation theory to the resulting...
A more detailed exposition of the model and a derivation of the perturbation theory expression for the cross section in direct two-photon double ionization processes. 

For instance, for helium all these parameters are well known. The model result is obtained using the time-dependent Schrödinger equation of helium from first principles. The model result is obtained using ab initio results as the values obtained for the cross section for the reaction may differ by as much as an order of magnitude. On the theoretical side, the great discrepancies that remain between different approaches are usually ascribed to the different ways electron correlations are handled in the final state. To this end, we hope that the predictions of the present model study may shed new light on this controversy.

Having justified the validity of our simple approach, we now turn to a more complex problem, namely the process of nonsequential two-photon double ionization of neon. Inserting, in Eq. (5), the correct first and second ionization energies of neon, i.e., 21.6 and 40.9 eV, as well as experimental values for the photoionization cross sections of Ne [30] and Ne+ [32], obtained using synchrotron radiation, the resulting model prediction for the double ionization cross section is shown in Fig. 4 (upper panel). The lower panel shows the corresponding electron energy distribution at three selected photon energies. Interestingly, at lower photon energies, the energy distribution exhibits a maximum (negative concavity) when both electrons are emitted with the same energy, while at higher photon energies the distribution is U-shaped. In sharp contrast to this trend, for helium, the model yields a U-shaped energy distribution for all photon energies (see Fig. 3).

In conclusion, we have implemented an approximate and very simple model to study the two-photon double ionization process of helium in the direct regime, i.e., at photon energies below 54.4 eV where the sequential ionization process is energetically inaccessible. We have investigated the validity of the model by calculating generalized total cross sections and energy-resolved differential cross sections and compared the model results with corresponding results obtained by accurate ab initio calculations. Quantitative agreement between model results and the full results was achieved in all considered cases, demonstrating the general validity of the model for the two-photon double ionization process. Finally, we have obtained the cross section for nonsequential two-photon double ionization of neon, demonstrating that the model has a great potential to be used in studies of nonsequential multiphoton multiple ionization processes in more complex atomic systems. This is an avenue of research we plan to pursue in the future.
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