Laser-assisted two-color two-photon double ionization of helium atoms

Zhizhen Zhu1 · Kai Liu1 · Xiaofan Zhang1 · Ye Li1 · Feng Wang1 · Meiyan Qin1 · Zhe Wang1 · Qing Liao1

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Abstract
Correlated momentum and kinetic energy distributions of two photoelectrons in laser-assisted two-color two-photon double ionization of helium are investigated by numerically solving a one-plus-one dimensional time-dependent Schrödinger equation (TDSE). We find that the weak assisting laser field can act as an energy transferring field, resulting in burst of double ionization triggered by the intense extreme ultraviolet (XUV) pulse. More importantly, the participation of the laser photon into the double ionization reshapes the correlation patterns in the momentum and kinetic energy distributions. The laser photon can be absorbed by any one of the two electrons, providing two channels that induces destructive interference in the correlated momentum and kinetic energy distributions, which is never found in previous work.

Keywords Strong field laser physics · Photoionization

1 Introduction
Electron correlation plays a very important role in nonsequential double ionization of atoms and thus has received extensive investigations in the past few decades (Schwarzkopf et al. 1993; Schwarzkopf and Schmidt 1996; Bräuning et al. 1998; Fittinghoff et al. 1992; Walker et al. 1994; Weber et al. 2000; Li et al. 2021). As the simplest multi-electron system, helium atoms have showed great advantages to explore electron correlation dynamics. In spite of that, the underlying physical mechanisms in double ionization of helium atoms are still complicated, since they depend strongly on the parameters of the driven laser pulses. While single-XUV-photon double ionization of helium atoms have been well studied both in experiment (Bräuning et al. 1998) and in theory (Maulbetsch and Briggs 1993; Briggs and Schmidt 2000; Parker et al. 2001; Avaldi and Huetz 2005; Jiang et al. 2013), the investigations of two-XUV-photon and few-XUV-photon double ionization are

✉ Kai Liu
12005010008@stu.wit.edu.cn

1 Hubei Key Laboratory of Optical Information and Pattern Recognition, Wuhan Institute of Technology, Wuhan 430205, China
challenging experimentally (Rudenko et al. 2008) and some details remain incomprehensible theoretically.

In two-color two-photon double ionization, the double ionization process can be divided into two steps, if the double ionization is triggered by absorption of one XUV photon and the subsequent correlation dynamics of the two photoelectrons is perturbed by the weak laser field through additional one-laser-photon absorption or emission. By isolating the photoexcitation process from the perturbation process, we can better understand the correlation dynamics according to the features in the correlated momentum and kinetic energy distributions. In this paper, we present the quantum-mechanically calculated momentum and kinetic energy distributions of the two photoelectrons in double ionization of helium atoms driven by an intense XUV pulse and a weak laser field. The correlation patterns change with the central photon energy of the XUV pulse. By analyzing the features of the correlation patterns, we draw different photoelectron-laser-photon interaction pictures.

2 Numerical Model

We use a one-plus-one dimensional TDSE model (Lein et al. 2000; Liao and Lu 2010; Liao et al. 2012, 2017; Liu et al. 2018) to simulate the double ionization of helium atoms driven by a linearly polarized XUV pulse and an assisting laser electric field with the same polarization direction. Limited by this model, the motion of the two electrons is restricted to the direction of laser polarization. However, the model has provided reasonable double ionization processes and successfully reproduced many double ionization mechanisms that are able to explain experimental results (Lein et al. 2000; Liao and Lu 2010). The numerical solution of the two-dimensional model can provide wave function of two related electrons in coordinate space and momentum space with time evolution. The TDSE for two electrons subject to their mutual interaction and their interactions with the nucleus and the external electric fields is

\[-i \frac{\partial}{\partial t} \Psi(x, y, t) = H(x, y, t)\Psi(x, y, t).\]  

Here \(x, y\) are the coordinates of the two electrons, respectively. Atomic units are used throughout this paper unless otherwise stated. The total Hamiltonian is

\[H = H_0 + V_{\text{int}},\]  

where the field-free Hamiltonian

\[H_0(x, y, t) = T_x + T_y + V_0\]

\[= -\frac{1}{2} \frac{\partial^2}{\partial x^2} - \frac{1}{2} \frac{\partial^2}{\partial y^2} - \frac{2}{\sqrt{x^2 + a^2}}\]

\[= \frac{2}{\sqrt{y^2 + a^2}} + \frac{1}{\sqrt{(x - y)^2 + b^2}},\]  

includes electron-nuclear attractive potentials and electron-electron repulsive potential. The Coulomb interactions between the particles are thus modeled by softened potentials, which is a way of allowing in 1D for the possibility that the particles pass by each other without probing a Coulomb singularity. The softcore Coulomb parameters \(a\) and \(b\) are chosen to be 1 in this paper. The interaction potential is

\[\Psi(x, y, t) = H(x, y, t)\Psi(x, y, t).\]
\[ V_{int} = (x+y)[E_{XUV}(t) + E_L(t)], \]

including the interactions between the two electrons and the external XUV and laser electric fields in the dipole length gauge. We assume that both the XUV and laser electric fields have a sine squared temporal envelope,

\[ E_a(t) = \begin{cases} E_{0,a} \sin^2 \left( \frac{\pi t}{T_a} \right) \cos(\omega_a t), & 0 \leq t \leq T_a, \\ 0, & t > T_a, \end{cases} \]

where the index \( a \) stands for XUV or L and \( E_{0,a} \), \( T_a \) and \( \omega_a \) denote the electric-field amplitudes, pulse lengths and frequencies of the two pulses, respectively. \( E_t(t) \) is the electric field of a 400 nm laser pulse with a total duration of 200 optical cycles. We solve equation (1) by numerical wave-function propagation (Table 1) on the \( x, y \) numerical grid, repeatedly applying the split time-evolution operator over equally spaced small time steps \( \Delta t \).

\[ \Psi(x, y, t + \Delta t) = e^{-iH_0(\Delta t/2)}e^{-iV_{int}\Delta t}e^{-iH_0(\Delta t/2)}\Psi(x, y, t) + O(\Delta t^2). \]

For this helium atom model, the two-electron ground state has been obtained by imaginary-time propagation under the field-free hamiltonian and its energy \( E_0 = -2.24 \) (\(- 61 \) eV). The first ionization potential is 0.44. The two-electron wave function propagates in a large box of \( 480 \times 480 \) with a spatial step of 0.23 and a time step of 0.1. Following Ref. Liao and Lu (2010), the two dimensional coordinate space is partitioned into two outer regions, (A) \( |x| < d \), or \( |y| < d \) and (B) \( |x|, |y| \geq d \), with \( d = 150 \). The final results are insensitive to the choice of \( d \) ranging from 100 to 200. The wave function in region A propagates under the action of the total hamiltonian. In region B, which corresponds to double ionization, all the Coulomb interactions are neglected and the time evolution of the wave function can be performed simply by multiplications in momentum space. The two regions are smoothly divided by a splitting technique (Liao and Lu 2010). At the end of the propagation, the wave function in region B yields the two-electron momentum and energy distributions. After the end of the pulse, the wave function is allowed to propagate without external fields for a long time such that the final results do not change anymore. The momentum distributions of the two photoelectrons are calculated according to the following formula

\[ P(p_1, p_2) = 2 |\Psi(p_1, p_2)|^2, \quad (7) \]

\( \Psi(p_1, p_2) \) is two-electron wave function of the final state in momentum space. Multiplying by 2 means that the two electrons cannot be distinguished.

**Table 1** Real-time propagation of the wave functions from time \( t \) to \( t + \Delta t \)

| \( t \) | \( \Psi_1(x, y, t) \) | \( \Psi_2(x, y, t) \) | \( \Psi_3(x, y, t) \) | \( \Psi_4(x, y, t) \) | \( \Psi(x, y, t + \Delta t) \) |
|------|----------------|----------------|----------------|----------------|----------------|
| 1    | \( e^{-iT_e(\Delta t/2)}\Psi_1(x, y, t) \) | \( e^{-iT_e(\Delta t/2)}\Psi_2(x, y, t) \) | \( e^{-iV_{int}\Delta t}\Psi_3(x, y, t) \) | \( e^{-iV_{int}\Delta t}\Psi_1(x, y, t) \) | \( e^{-i(T_e + V_{int})\Delta t}\Psi_4(x, y, t) \) |
| 2    | \( e^{-iV_{int}\Delta t}\Psi_1(x, y, t) \) | \( e^{-iV_{int}\Delta t}\Psi_2(x, y, t) \) | \( e^{-iT_e(\Delta t/2)}\Psi_3(x, y, t) \) |
| 3    | \( e^{-iV_{int}\Delta t}\Psi_1(x, y, t) \) | \( e^{-iV_{int}\Delta t}\Psi_2(x, y, t) \) |
| 4    | \( e^{-iV_{int}\Delta t}\Psi_1(x, y, t) \) |
| 5    | \( e^{-iV_{int}\Delta t}\Psi_1(x, y, t) \) |
3 Results and Discussion

First, we present the correlated momentum and kinetic energy distributions for single-photon double ionization, as shown in Fig. 1. The circle momentum distributions is a consequence of the sharing of excess kinetic energy between the two photoelectrons. In the energy distributions, it means the sum of the kinetic energies of the two photoelectrons $E_1 + E_2 = \omega_{\text{XUV}} + E_g$. Since both the two electrons are restricted by our simulation model to move in one dimension, they can only be emitted side-by-side or back-to-back. However, our model reproduces the correlated features qualitatively with full dimensional TDSE (Guan et al. 2008; Liu and Thumm 2014, 2015), such as the forbidden emission for equal energy sharing in both back-to-back and side-by-side emission ($|p_1| = |p_2|$) because of the selection rule B2 discussed by Briggs and Schmidt (2000) and (Maulbetsch and Briggs 1995). As the excess energy increases (by increasing the central XUV photon energy), the energy sharing ratio between the two photoelectrons becomes more asymmetric.

When the double ionization process is exposed to a weak laser field, the weak laser field can largely influence the double ionization probability and the correlation patterns in the momentum and kinetic energy distributions, as shown in Fig. 2 for a central XUV photon.

![Fig. 1 Logarithmic plot of the correlated momentum (left plots) and kinetic energy (right plots) distributions of the two escaping electrons in single-photon double ionization of He. $p_1$, $p_2$, are the final momenta of the two photoelectrons, and $E_1$, $E_2$ the final kinetic energies, respectively. The XUV pulse has a peak intensity of $5 \times 10^{14}$ W/cm$^2$ and a time duration of one hundred optical cycles. The central photon energies are 64 eV (top plots) and 67 eV (bottom plots), respectively. The units are arbitrary.](image)
energy of 67 eV and a 400 nm laser field. As the laser field intensity increases from $1 \times 10^{11}$ W/cm$^2$ (top row), to $1 \times 10^{12}$ W/cm$^2$ (bottle row), the double ionization yield is enhanced by one order in magnitude for $1 \times 10^{12}$ W/cm$^2$ (middle row), but then enhanced by a little for a higher intensity of $1 \times 10^{13}$ W/cm$^2$ (bottle row). Furthermore, the energy sharing ratio becomes more extreme for side-by-side emission and back-to-back emission gradually dominates. The additionally emerging rings in Fig. 2a, c, e are results of absorption or emission of one laser photon by the two photoelectrons. When the two photoelectrons absorb or emit one laser photon, the selection rule B2 is broken for equal energy sharing and we can see the
appearance of the two photoelectrons with the same energy in back-to-back emission (Liu and Thumm 2014, 2015). For emission of one laser photon, the total excess energy become less and the probability with any energy sharing ratio is almost the same. While the absorption of one laser photon increases the total excess energy and the two photoelectrons intend to share the excess energy more asymmetrically.

To gain more insights into the dependence of the electron-electron correlation in laser-assisted double ionization of helium atoms on the XUV photon energy, i.e., the total excess energy, we change the XUV central photon energy while keeping the laser intensity unchanged. For low XUV photon energy at 64 eV, the absorption probability of one laser photon by the two photoelectrons is much larger than the emission probability of one laser photon, see Fig. 3a, b. This is well understood since the emission of one laser photon (3 eV) decreases the excess energy near to 0, making the electron that first absorbs one XUV photon much difficult to kick out the other through energy transferring. As the XUV photon energy increases to 67 eV, the emission probability of one laser photon becomes approaching to the absorption probability, see Fig. 3c, d. The asymmetric energy sharing ratio is preferable for the laser-photon absorption channel. As the XUV photon energy further increases to 77 eV, the asymmetric energy sharing ratio is preferable for the laser-photon emission channel. In contrast, the equal energy sharing ratio is preferred for the laser-photon absorption channel, see Fig. 3f. More surprisingly, forbidden emission of the two photoelectrons appears in some asymmetric energy sharing regions for the laser-photon absorption channel, which is never found in previous work.

The above dependence of the correlated energy distributions on XUV photon energy and the weak laser field can be explained based on the kick-out double ionization mechanism. In kick-out double ionization mechanism, one electron absorbs one XUV photon (we refer to this electron as the first electron) and then transfers energy to the other one (the second electron) through Coulomb repulsion, kicking the other one out from the nucleus. When the two electrons are exposed to a weak laser field, another energy transferring channel is open. In this channel, the first electron emits one laser photon into the laser field, while the second one absorbs one laser photon from the laser field. Here the laser field acts as an energy transferring field. No net laser photon participates into the double ionization process, but the additional energy transferring channel enhances double ionization probability dramatically.

When the first electron additionally absorbs one laser photon and the other does not absorb or emits one laser photon, see Fig. 4a, the final kinetic energy of the first electron will be larger than that of the second one. Conversely, see Fig. 4b, the difference in final kinetic energies between the two electrons becomes small and they can have the same kinetic energy. For the case of the laser photon emission, see Fig. 4c, d, the similar explain also works. There are two channels for the laser-photon absorption. One channel is that the first electron absorbs one laser photon, the energy of the second electron \( E_2 \in [0, E_{2,\text{max}}] \), where \( E_{2,\text{max}} \) is the maximum kinetic energy, less than half the total excess kinetic energy. The other channel is that the second electron absorbs one laser photon, the energy of the second electron \( E_2 \in [\omega_L, E_{2,\text{max}} + \omega_L] \). There is an overlap energy region for the two channels if \( E_{2,\text{max}} > \omega_L \). The two double ionization channels interfere in the overlap energy region. If \( E_{2,\text{max}} \) is much larger than \( \omega_L \), fully destructive interference happens.
4 Conclusion

We investigated two-color two-photon double ionization of helium atoms driven by a linearly polarized XUV pulse and an assisting laser electric field with the same polarization direction. By numerically solving a one-plus-one dimensional TDSE model, we calculated the correlated momentum and kinetic energy distribution of the two photoelectrons. The

![Logarithmic plot of the correlated momentum (left plots) and kinetic energy (right plots) distributions of the two escaping electrons in laser-assisted single-photon double ionization of He. The XUV pulse has a peak intensity of $5 \times 10^{14}$ W/cm$^2$ and a time duration of one hundred optical cycles. The XUV central photon energies are 64 eV (top plots), 67 eV (middle plots), and 77 eV (bottom plots), respectively. The 400 nm laser pulse has a peak intensity of $5 \times 10^{12}$ W/cm$^2$. The units are arbitrary.](image-url)
weak assisting laser field provides an energy transferring channel, increasing double ionization probability remarkably for single-XUV-photon double ionization. The participation of the laser photon into the double ionization dramatically changes the correlation patterns in the momentum and kinetic energy distributions. Such changes depend on the XUV photon energy, the laser intensity and how the laser photon interacts with the two electrons. Our work provides instructions in revealing dynamics details of single-photon double ionization and manipulating the electronic correlation in double ionization.

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