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Multiphoton Rabi oscillations of correlated electrons in strong-field nonsequential double ionization

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Abstract. With quantum calculations, we have investigated the multiphoton nonsequential double ionization of helium atoms in intense laser fields at ultraviolet wavelengths. Very surprisingly, we found a so-far unobserved double-circle structure in the correlated electron momentum spectra. The double-circle structure essentially reveals multiphoton Rabi oscillations of two electrons, which are strongly supported by the oscillating population of a certain doubly excited state and by the oscillating double ionization signals. This two-electron multiphoton Rabi effect provides a profound understanding of electronic correlations and complicated multiphoton phenomena and is expected to be a new tool for broad applications, such as quantum coherent control.

Electronic correlations are of fundamental importance for the dynamics of many phenomena such as high-temperature superconductivity in solid state. Nonsequential double ionization (NSDI) of atoms and molecules by short intense laser pulses can provide one of the basic examples for studies of dynamical electron correlations, and thus has been investigated extensively both experimentally [1–5] and theoretically [6–10] in the last few decades. Because they possess rich information, the correlated electron momentum spectra [2–5] have revealed a great many physical pictures of electron–electron correlation in NSDI under the recollision mechanism. These physical pictures are well described by the classical recollision...
model [3, 11, 12]. The correlated electron momentum spectra from quantum mechanical calculations of NSDI of He at extreme ultraviolet (XUV) wavelengths [13], as well as visible and ultraviolet (UV) wavelengths [14, 15], exhibit a circle (or circular arc) structure with energy separation of the photon energy. This structure reveals a resonant double-ionization process in which the correlated electrons simultaneously absorb and share energy in integer units of the photon energy, transiting from the ground state to continuum states [13]. Even at near-infrared wavelengths there is also a resonant double-ionization process dominating NSDI of He after doubly excited states populated via recollision below the recollision threshold [16]. Such an NSDI process has been observed in recent experiments on double ionization of He and Ne by strong free-electron laser pulses at vacuum UV wavelengths [17, 18].

Another fundamental effect in nonlinear light–matter interaction is optical Rabi oscillations, which are of general importance for quantum optics and have extensive applications in many fields such as quantum coherent control in atomic clocks [19, 20] and especially in quantum computing [21–23]. In this paper, we demonstrate the multiphoton Rabi effect of two strongly correlated electrons in NSDI of He by strong laser fields at UV wavelengths. By numerically solving the two-electron time-dependent Schrödinger equation, we obtained the correlated electron momentum spectra from NSDI. A ‘one-plus-one’-dimensional model of a helium atom with soft Coulomb interactions, where the motion of both electrons is restricted to the laser polarization direction, is employed. This model has been able to reproduce many NSDI features [7, 14, 24]. We use the split-operator spectral method [25] to numerically solve the two-electron time-dependent Schrödinger equation (in atomic units)

\[-i \frac{\partial}{\partial t} \Psi(z_1, z_2, t) = H(z_1, z_2, t) \Psi(z_1, z_2, t),\]

where \(z_1\) and \(z_2\) are the electron coordinates. \(H(z_1, z_2, t)\) is the total Hamiltonian and reads

\[H(z_1, z_2, t) = -\frac{1}{2} \frac{\partial^2}{\partial z_1^2} - \frac{1}{2} \frac{\partial^2}{\partial z_2^2} - \frac{2}{\sqrt{z_1^2 + 1}} - \frac{2}{\sqrt{z_2^2 + 1}} + \frac{1}{\sqrt{(z_1 - z_2)^2 + 1}} + (z_1 + z_2) E(t).\]

\(E(t)\) is the electric field of a laser pulse. The laser field has no effect on the relative motion of the electrons, which means that the two electrons behave like a doubly charged single-particle concerning the interaction with the laser field. The wave function \(\Psi(z_1, z_2)\) is symmetric with respect to exchange of the two electrons because the two electrons are indistinguishable. Following [7], the two-dimensional (2D) space is partitioned into two regions: (A) \(|z_1| < a\) or \(|z_2| < a\) and (B) \(|z_1|, |z_2| \geq a\) with \(a = 150\) a.u. The final results are insensitive to the choice of \(a\) ranging from 100 to 200 a.u. In region A, the wave function is propagated exactly in the presence of combined Coulomb and laser field potentials. In region B, which corresponds to double ionization, all the Coulomb potentials between the particles 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 [26, 27]. There is a broad overlapping region between regions A and B. The overlapping region is sufficiently far from the nucleus so that the Coulomb field can be neglected, compared with the strong laser filed. More details of the splitting procedure can be seen in [26]. At the end of the propagation, the wave function in region B yields the two-electron momentum and energy spectra from double ionization.

Our calculations use trapezoidally shaped laser pulses with a total duration of 60 optical cycles, switched on and off linearly over ten optical cycles, respectively. A very large grid size of \(2500 \times 2500\) a.u. with a spatial step of 0.15 a.u. is used, while the time step is 0.1 a.u. The very
Figure 1. Log plot of the correlated electron momentum spectrum for double ionization of He by laser pulses at (a) 198 nm, 0.3 PW cm$^{-2}$, (b) 198 nm, 0.4 PW cm$^{-2}$ and (c) 208 nm, 0.3 PW cm$^{-2}$. The units are arbitrary.

A large grid provides sufficiently dense continuum states [14] to yield highly accurate two-electron momentum and energy spectra. The initial wave function is the two-electron ground state of He obtained by imaginary-time propagation. After the end of the pulse, the wave function is allowed to propagate without laser field for an additional time of 40 optical cycles. The final results do not change any more even though the wave function propagates without laser field for a longer additional time.

Figure 1 displays the resulting correlated electron momentum spectrum from double ionization of helium atoms. Very surprisingly, a double-circle structure is prominent in the momentum spectra for double ionization at 198 nm, 0.3 PW cm$^{-2}$ (figure 1(a)) and 0.4 PW cm$^{-2}$ (figure 1(b)), which differs from the single-circle structure in previous works [13–16]. These concentric circles satisfy ($p_1^2 + p_2^2$) = constant, which is the signature of a resonant double-ionization process [15]. In this process, the two electrons simultaneously absorb an integer
number of photons and share the excess energy in integer units of the photon energy. This process has been called nonsequential double-electron above-threshold ionization (DATI) [15]. Comparing figures 1(a) and (b), we find that the separation of each doublet becomes larger when increasing the laser intensity and keeping the wavelength unchanged. At 198 nm, 0.1 PW cm\(^{-2}\), the separation becomes indistinguishable in the correlated spectrum (not shown in this paper). However, the separation between each doublet is much less than that between adjacent doublets. The relations between these circles manifest themselves in the corresponding total kinetic energy spectra of two ionized electrons, as shown in figure 2. The energy separation between adjacent doublets is constant and is equal to the photon energy, whereas the energy separation between each doublet is also constant for one laser intensity but becomes larger with increasing intensity. At 0.2, 0.3 and 0.4 PW cm\(^{-2}\), they are, on average, 0.013, 0.030 and 0.048 a.u., respectively.

In order to gain insight into the physical mechanism responsible for the double-circle structure in the correlated momentum spectra, we investigate the time evolution of the population of doubly excited states and the flux of double ionization. Following [28], we define region 1: \(|z_1| < 7 \text{ a.u.}, \ |z_2| < 10 \text{ a.u.}\) and region 2: \(|z_1| < 3 \text{ a.u.}, \ |z_2| < 3 \text{ a.u.}\) as the regions of doubly excited states and the two-electron ground state, respectively. Region 3: \(|z_1| > 20 \text{ a.u.}, \ |z_2| > 20 \text{ a.u.}\) is defined as the region of doubly ionizing wavepackets. The populations of doubly excited states, the two-electron ground state and double ionization are calculated by

\[
P_1(t) = \int_{|z_1|<10} \int_{|z_2|<10} \, dz_1 \, dz_2 \, |\Psi(z_1, z_2, t)|^2,
\]

\[
P_2(t) = \int_{|z_1|<3} \int_{|z_2|<3} \, dz_1 \, dz_2 \, |\Psi(z_1, z_2, t)|^2,
\]

\[
P_3(t) = \int_{|z_1|>20} \int_{|z_2|>20} \, dz_1 \, dz_2 \, |\Psi(z_1, z_2, t)|^2,
\]

respectively. \(dP_3(t)/dt\) gives the double-ionization flux. We must emphasize the fact that the doubly ionizing wavepackets may contribute to the population in region 1. However, if the doubly ionizing wavepackets arise dominantly from doubly excited states, the contribution from doubly ionizing wavepackets to population in region 1 can be neglected. This is verified for
Figure 3. (a, d) Population of doubly excited states, (b, e) double-ionization flux and (c, f) population of the two-electron ground state as functions of time. The laser parameters are 198 nm, 0.3 PW cm$^{-2}$ for (a–c) and 198 nm, 0.4 PW cm$^{-2}$ for (d–f), respectively.

the case when the double-circle structure dominates the correlated spectra, which is elaborated below. Figure 3 shows the population of doubly excited states, double ionization flux and population of the two-electron ground state as functions of time for the laser parameters of 198 nm, 0.3 PW cm$^{-2}$ (figures 3(a)–(c)) and 198 nm, 0.4 PW cm$^{-2}$ (figures 3(d)–(f)). The population of doubly excited states and the flux of double ionization oscillate synchronously in time, but antisynchronously with the oscillation of the population of the ground state. Note that the periods of oscillation of the population of doubly excited states and of the flux of double ionization are equal. For higher laser intensities, the period of oscillation becomes smaller and the oscillation damping becomes larger. The periods are, on average, 16.2, 7.64 and 4.92 optical cycles for 198 nm pulses with intensities of 0.2, 0.3 and 0.4 PW cm$^{-2}$, respectively. Thereby, the corresponding frequencies are 0.014, 0.030 and 0.047 a.u., in very good agreement with energy separations of the corresponding doublets.

For a two-level atom exposed to a strong laser field, if the resonance happens, the resonance level may become populated. Meanwhile, both the initial level and the resonance level are split into two quasi-energy levels each with equal populations, which are separated in energy by the
Rabi frequency \[29\]. The population also oscillates between the initial level and the resonance level with a period of \(2\pi/\Omega\), where \(\Omega\) is the Rabi frequency. Evidently, the above oscillations are the so-called Rabi oscillations that occur when there is resonance between the two-electron ground state and a certain doubly excited state. This is the physical mechanism responsible for the double-circle structure in the correlated momentum spectra. Because the two-electron ground state population and the doubly excited state population is depleted by single and double ionization, the Rabi oscillations are damped strongly depending on the laser intensity. The Rabi frequency is given by \[30\]
\[
\Omega = \sqrt{(m\omega - \omega_0)^2 + (\mu E_0)^2},
\]
where \(\mu\) is the transition dipole moment, \(E_0\) is the field amplitude of the laser pulse, \(\omega_0\) is the transition frequency, \(\omega\) is the photon frequency and \(m\) is the number of photons that resonance requires. With the assumption that the change of \(\omega_0\) is negligible relative to that of \(m\) at the condition of varying the laser wavelength slightly and keeping the laser intensity constant, the value of \(m\) can be determined. For 0.3 PW cm\(^{-2}\) pulses, the Rabi frequency is 0.03, 0.038 and 0.044 a.u. at 198, 200 and 202 nm, respectively. According to equation (3), we determine \(m = 6\).

Now it is obvious that the double-cycle structure essentially reveals two stages of the multiphoton double-ionization process in which the two electrons are strongly correlated. Firstly, both electrons resonantly absorb a number of photons, transiting from the two-electron ground state to a certain doubly excited state. Then they emit photons, transiting to the ground state, or are emitted by resonantly absorbing additionally a number of photons and sharing excess energy in integer units of the photon energy. In the whole process, the two electrons behave in the same way as one electron. Therefore, analogous to the kinetic energy of photoelectrons resulting from above-threshold ionization by ultrashort pulses [31], the total kinetic energy of the doubly ionized electrons can be approximately predicted by
\[
E_n = n\hbar\omega - I_p - 2U_p.
\]
\(n\) is the total number of photons absorbed by the two electrons. \(I_p = 2.238\) a.u. is the double-ionization potential of the He atom in our model. The ponderomotive energy \(U_p = E_0^2/(4\omega^2)\). The ac-Stark shift of the ground state is not included in equation (4). Equation (4) gives a very good estimation of the middle position of the double peak. For example, for \(n = 11\), equation (4) gives 0.242, 0.215 and 0.188 a.u. at 0.2, 0.3 and 0.4 PW cm\(^{-2}\), respectively, and figure 2 gives 0.238, 0.214 and 0.190 a.u., respectively. This implies no ac-Stark effect for the population of the two states involved in Rabi oscillations.

Further varying the wavelength, the Rabi effect disappears rapidly. For a 208 nm, 0.3 PW cm\(^{-2}\) pulse, the oscillations of the population of doubly excited states and of the flux of double ionization are blurred severely, as shown in figure 4. In addition, both the population of doubly excited states and the flux of double ionization at 208 nm are much lower than those at 198 nm. Note that doubly excited states cannot be populated when the intermediate resonance is broken and population in region 1 arises mostly from the contribution of doubly ionizing wavepackets. Meanwhile, the periodic oscillation of the population of the ground state also disappears. This implies that the resonance between the two-electron ground state and the doubly excited state is broken down in this case. However, the double-circle structure (see figure 1(c)) can be still distinguished in the logarithmic plot of the correlated momentum spectrum, but with probability of about two orders of magnitude lower than the single-circle
Figure 4. (a) Population of doubly excited states, (b) double-ionization flux and (c) population of the two-electron ground state as functions of time. The laser parameters are 208 nm, 0.3 PW cm$^{-2}$.

Figure 5. Photoelectron total-kinetic energy spectra of two ionized electrons from double ionization of He by 0.3 PW cm$^{-2}$ laser pulses at different wavelengths.

structure coming directly from the unsplit ground state. In the corresponding total kinetic energy spectrum, we find the primary peak shifted from the middle position of the double peak due to the ac-Stark effect of the ground state, as shown in figure 5. Thereby the ac-Stark shift at 208 nm, 0.3 PW cm$^{-2}$ is determined to be 0.012 a.u.. At shorter wavelengths (about 142 nm) or longer wavelengths (about 228 nm), again we found the multiphoton Rabi oscillations of the two correlated electrons.
In summary, we have demonstrated the multiphoton Rabi oscillations of strongly correlated electrons in strong-field NSDI by quantum mechanical calculations. The demonstration of the two-electron multiphoton Rabi effect both in time domain and in frequency domain enables one to gain a deep insight into electronic correlations and complicated multiphoton phenomena. Our study, of fundamental importance for quantum optics and many-body physics, can advance the exploration of the physical mechanisms of many effects in nature governed by electronic correlations. The optical Rabi effect involving two correlated electrons is expected to be a new tool for direct quantum coherent control.

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