Multiphoton Rabi Oscillations of Correlated Electrons in Strong Field Nonsequential Double Ionization

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With quantum calculations, we have investigated the multiphoton nonsequential double ionization of helium atoms in intense laser fields at ultraviolet wavelengths. Very surprisingly, we find 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 profound understandings of electronic correlations and complicated multiphoton phenomena and is expected to be a new tool for broad applications, such as quantum coherent control.

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Electronic correlations are of fundamental importance to the dynamics of many phenomena such as high-temperature superconductivity in solid states. 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 in both experiment [1–5] and theory [6–9] in the past few decades. Because of possessing rich information, the correlated electron momentum spectra [2–5] have revealed a great many physical pictures of electron-electron correlation in NSDI under recollision mechanism. These physical pictures are well-described by the classical recollision model [3, 10, 11]. The correlated electron momentum spectra from quantum mechanical calculations of NSDI of He at extreme ultraviolet (XUV) wavelengths [12], as well as visible and ultraviolet (UV) wavelengths [13, 14], 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 into continuum states [12]. 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 recollision threshold [15]. Such a 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 [16, 17].

Another fundamental effect in nonlinear light-matter interaction is optical Rabi oscillations, which are of general importance to quantum optics and have extensive applications in many fields such as quantum coherent control in atomic clocks [18, 19] and especially in quantum computing [20–22]. In this Letter, we demonstrate 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, 13, 23]. We use the split-operator spectral method [24] to numerically solve the two-electron time-dependent Schrödinger equation (in atomic units)

$$-i\frac{\partial}{\partial t}\Psi(z_1,z_2,t)=H(z_1,z_2,t)\Psi(z_1,z_2,t), \qquad (1)$$

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

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

$E(t)$ is the electric field of a laser pulse. Following Ref. [7], the two-dimensional space is partitioned into two outer 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 [25]. At the end of the propagation,

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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 10 optical cycles respectively. A very large grid size of 2500 × 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 large grid provides sufficiently dense continuum states [13] to yield highly accurate two-electron momentum and energy spectra. The initiate 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 [Fig. 1(a)] and 0.4 PW/cm^2 [Fig. 1(b)], which differs from the single-circle structure in previous works [12–15]. These concentric circles satisfy $(p_1^2 + p_2^2) = constant$, which is the signature of a resonant double-ionization process [14]. 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)[14]. Comparing Figs. 1(a) with 1(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 undistinguishable in the correlated spectrum (not shown in this Letter). 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 Fig. 2. The energy separation between adjacent doublets is constant and equals the photon energy, whereas the energy separation between each doublet is also constant for one laser intensity but becomes larger with the increasing intensity. At 0.2 PW/cm^2, 0.3 PW/cm^2 and 0.4 PW/cm^2, they are, on average, 0.013 a.u., 0.030 a.u. 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. The population of doubly excited states is examined by monitoring the population in region 1: {7 a.u.< $|z_1|$ <10 a.u., 7 a.u.< $|z_2|$ <10 a.u. } [26] and we define region 2: {$|z_1|$ >20 a.u., $|z_2|$ >20 a.u.} as the region of doubly ionizing wavepackets. 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 the case when the double-circle structure dominates the correlated spectra, which is elaborated below. Fig. 3 shows the population of doubly excited states and the flux of double ionization as functions of time for the laser parameters of 198 nm, 0.3 PW/cm^2 [Figs. 3(a) and 3(b)] and 198 nm, 0.4 PW/cm^2 [Figs. 3(c) and 3(d)]. The population of doubly ex-

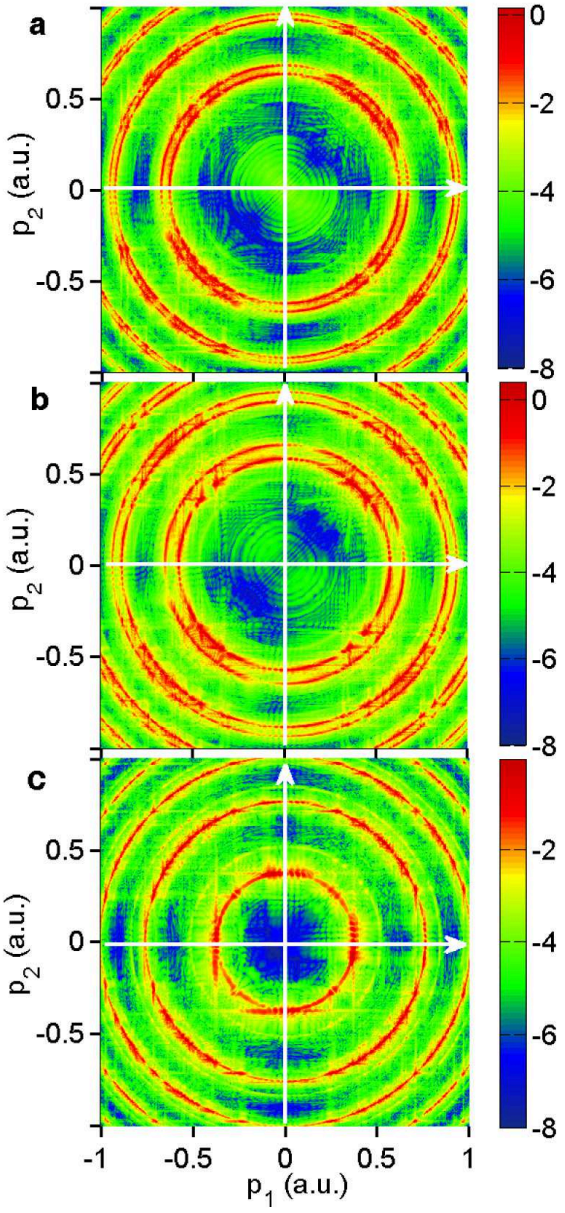

FIG. 1: (color online) Log plot of the correlated electron momentum spectrum for double ionization of He by laser pulses at (a) 198 nm, 0.3 PW/cm^2, (b) 198 nm, 0.4 PW/cm^2 and (c) 208 nm, 0.3 PW/cm^2. The units are arbitrary.

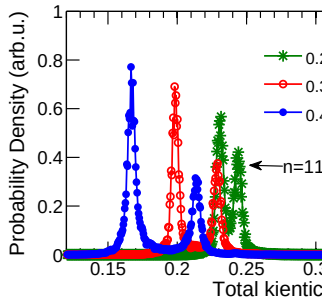

FIG. 2: (color online) Photoelect
trum of two ionized electrons frc
198 nm laser pulses at different

cited states and the flux of
synchronously and damply. N
riods of the population of dc
the flux of double ionization
intensities, the oscillating peri
oscillation damping becomes
average, 16.2, 7.64 and 4.92
pulses with intensities of 0.2 I
0.4 PW/cm^2, respectively. T
frequencies are 0.014 a.u., 0.0
very good agreement with ene
responding doublets.

Evidently, the above oscillat
oscillations that occurs when t
the two-electron ground state
cited state. Both the two stat
energy states separated in ene
[27]. This is the physical mec
double-circle structure in the c
tra. Because the two-electroı
and the doubly excited state
single and double ionization,
damped strongly depending o
Rabi frequency is given by [28

$$\Omega = \sqrt{(m\omega - \omega_0}$$

where μ is the transition dipo
amplitude of the laser pulse,
quency, ω is the photon frequ
of photon that resonance requ
that the change of ω_0 is negligi
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 a.u., 0.038 a.u. and 0.044 a.u. at 198 nm, 200 nm and 202 nm, respectively. According to Eq. (3), we determine $m = 6$.

Now it is very obvious that the double-cycle structure essentially reveals two stages of the multiphoton double ionization process in which the two electrons are

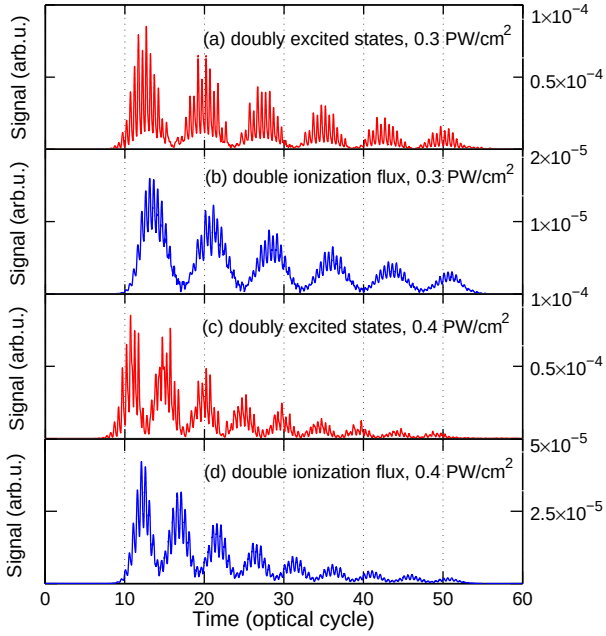

FIG. 3: (color online) (a, c) Population of doubly excited

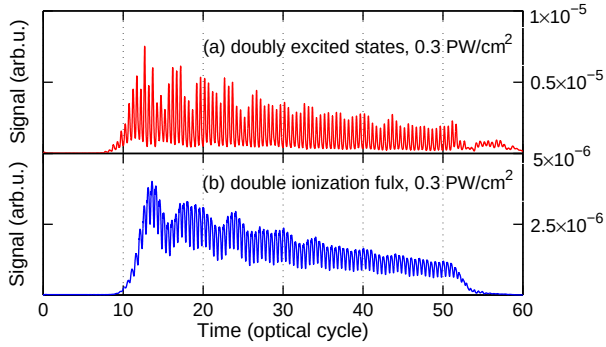

FIG. 4: (color online)(a) Population of doubly excited states and (b) double ionization fluxes as functions of time. The laser parameters are 208 nm, 0.3 PW/cm^2.

strongly correlated. Firstly, both electrons resonantly absorb a number of photons, transiting from the two-electron ground state into a certain doubly excited state. Then they emit photons, transiting into the ground state, or are emitted by resonantly absorbing additional 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 [29], the total kinetic energy of the doubly ionized elec-

trons can be appro

E

n is the total numb
trons. $I_p = 2.238$ a
of He atom in our
$U_p = E_0^2/(4\omega^2)$. Tl
is not included in I
timation of the mi
example, for n=11
and 0.188 a.u. at
PW/cm^2 respectiv
a.u. and 0.190 a.u
Stark effect for the
in Rabi oscillations

Further varying
appears rapidly. F
oscillations of the p
of the flux of doub
shown in Fig. 4.
doubly excited sta
at 208 nm are muc
that doubly excited states can not be populated when the intermediate resonance is broken and population in region 1 arises mostly from contribution of doubly ionizing wavepackets. However, the double-circle structure [see Fig. 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 structure directly coming 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 Fig. 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 have a deep insight into electronic correlations and complicated multiphoton phenomena. Our study, fundamentally important to quantum optics and many-body physics, can advance exploring 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 broad applications, such as direct quantum coherent control in atomic clock [18, 19], quantum information processing [20–22] and chemical reactions [30].

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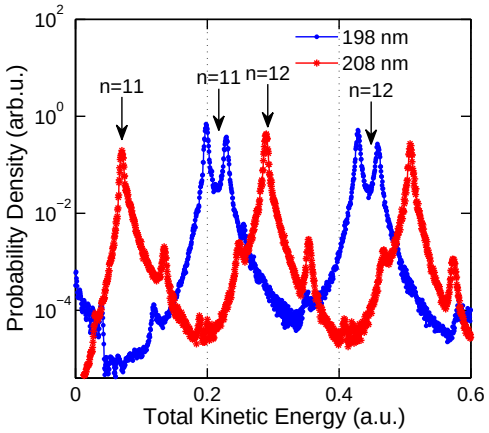

FIG. 5: (color online) Photoelectron total-kinetic energy spectra of two ionized electrons from double ionization of He by 0.3 PW/cm^2 laser pulses at two different wavelengths.

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