Angular Correlation in Strong-Field Double Ionization under Circular Polarization

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Using a classical ensemble approach, electrons detached sequentially by short circularly polarized laser pulses are predicted to be correlated in their emission directions. The correlation is introduced by the laser pulses. By changing the laser intensity, the angle between the two emissions can be controlled continuously, from 0° (parallel) to 90° (perpendicular) to 180° (antiparallel). The effect on the resultant ion momentum distribution is discussed.

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Electron correlation in strong-field ionization has been extensively studied for the past two decades since the discovery of nonsequential double ionization [1,2], which cannot be understood by assuming two independent ionization processes.

Various theories have been proposed for the underlying mechanism of this mutual interaction (a review is given in Becker *et al.*, [3]). In the wavelength regime of midinfrared and longer, it has been widely accepted that recollision [4] is the dominant mechanism responsible for electron correlations in strong-field double ionization.

Electron correlations can thus be controlled by controlling the recollision process, which depends on the laser parameters. Extensive attempts have been made, both experimentally and theoretically, by using intensities below the recollision threshold intensity [5–10], or using wavelengths other than the fundamental Ti:sapphire laser wavelength [11–14], or using few-cycle pulses [15,16], or using a configuration of two pulses [17], to control recollision and electron correlation and new physical effects have been observed or predicted.

However, recollision depends critically on laser polarization and a slight ellipticity is capable of eliminating most recollision events [18,19]. No correlation between the two electrons would be expected if they are emitted *sequentially* under a circularly polarized (*CP*) laser pulse without involving recollision [20].

In this Letter, we will show simulation results that contradict the above expectation. The two electrons ionized sequentially by a short *CP* pulse are predicted to be strongly correlated in their emission angles. Further, we will show that the correlation angle can be controlled continuously from 0° (parallel) to 90° (perpendicular) to 180° (antiparallel), by just changing the laser intensity. The electron angular correlation will also have a measurable effect on the resultant ion momentum distribution.

A similar topic has not been considered as far as we are aware of. The only related work we know addressing electron correlations in sequential double ionization (SDI) is a recent experiment by Fleischer *et al.* [21] (theory can be found in Ref. [22]), in which a linearly polarized pulse is used to ionize the first electron and a *CP* pulse is used to ionize the second electron. The two electrons are shown to be angularly correlated. However, we want to point out that the *CP* pulse is a tool to investigate the preferred emission angle of the second electron, instead of introducing any correlation of the electrons.

The asymptotic angle between the two emissions is noted as θ and illustrated in Fig. 1 using a typical double ionization trajectory under circular polarization.

Figure 2 shows simulation results for the distribution of the angle θ under three different laser intensities, namely 5.7, 7.7, and 11.0 PW/cm^2. The pulse is a short *CP* pulse with a sine-squared shape and a full duration of 5 cycles, without carrier-envelope phase stabilization. For $I = 5.7$ PW/cm^2, θ is centered at 180°, meaning that the two electrons are most likely to be emitted into opposite directions. For $I = 7.7$ PW/cm^2, the two electrons are most likely to be emitted to perpendicular directions. For $I = 11.0$ PW/cm^2, the two electrons are most probably emitted into the same direction.

Let us briefly introduce the method that we have used to obtain the above results. Details of the classical ensemble method have been explained in Ref. [23]. We start from a microcanonical ensemble of classically modeled atoms, each member of the ensemble includes two electrons and has the same total energy in the absence of laser fields

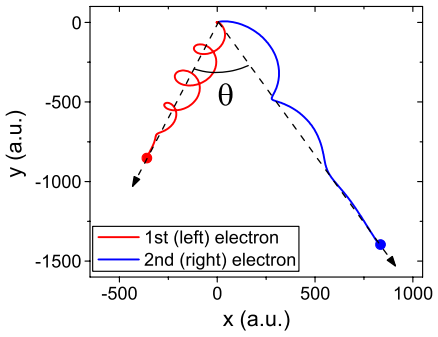

FIG. 1 (color online). Illustration of the angle between the two emissions, using a typical classical SDI trajectory under circular polarization.

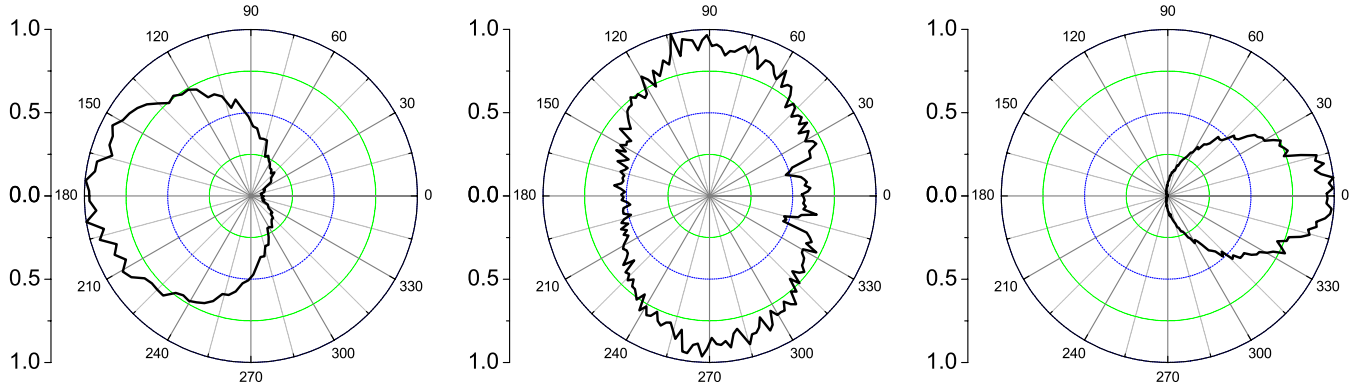

FIG. 2 (color online). Distribution of the angle θ for three laser intensities: 5.7 PW/cm^2 (left), 7.7 PW/cm^2 (center), and 11.0 PW/cm^2 (right).

$$E_{\mathrm{tot}} = \sum_{i=1}^{2}\left(\frac{\mathbf{p}_i^2}{2} - \frac{2}{\sqrt{\mathbf{r}_i^2 + a^2}}\right) + \frac{1}{\sqrt{|\mathbf{r}_1 - \mathbf{r}_2|^2 + b^2}}, \quad (1)$$

where $\mathbf{p}_i$ and $\mathbf{r}_i$ are the momentum and the position of the ith electron. Atomic units have been used. E_{tot} is set to be the negative sum of the first two ionization potentials of Ar, so $E_{\mathrm{tot}} = -1.6$ atomic units (a.u.). The Coulomb potentials have been softened [24,25] with parameter a between the ion core and the electrons and with parameter b between the electrons. Parameter a is chosen to be 1.5 a.u. to avoid autoionization and parameter b is chosen to be 0.1 a.u. to avoid computational singularity. Given E_{tot}, the positions and momenta of the two electrons are randomly assigned. The ensemble contains 100 000 members (model atoms) for the results shown in Fig. 2.

Then a *CP* pulse is turned on. The electric field can be written as

$$\mathbf{E}(t) = E_0 f(t)[\hat{\mathbf{x}}\sin(\omega t + \phi) + \hat{\mathbf{y}}\cos(\omega t + \phi)], \quad (2)$$

where $f(t) = \sin^2(\pi t/\tau)$ is the pulse envelope, $\omega = 0.0584$ a.u. corresponding to a wavelength of 780 nm, and ϕ is randomly chosen within $(0, 2\pi)$ for each pulse.

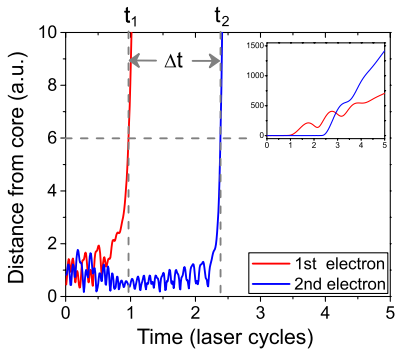

FIG. 3 (color online). A typical SDI trajectory for a *CP* pulse. An electron is regarded as ionized once it reaches a distance of 6 a.u., and the corresponding time is defined as the ionization time, noted as t_1 and t_2 for each electron. The difference is noted as Δt. The inset shows the same trajectory, but in a much larger space scale.

The motion of the electrons is governed by Newtonian mechanics via numerically integrating the Hamiltonian equations of motion

$$\frac{d\mathbf{r}_i}{dt} = \frac{\partial H}{\partial \mathbf{p}_i}, \qquad \frac{d\mathbf{p}_i}{dt} = -\frac{\partial H}{\partial \mathbf{r}_i}, \quad (3)$$

where $H = E_{\mathrm{tot}} + (\mathbf{r}_1 + \mathbf{r}_2)\cdot\mathbf{E}(t)$ is the effective Hamiltonian of each two-electron atom.

The positions and the momenta of the two electrons are recorded step by step during the entire pulse. A typical trajectory is demonstrated in Fig. 3 showing the distance of each electron from the ion core during the pulse.

We have defined 6 a.u. as the ionization criterion: an ionization event is achieved once an electron reaches this distance from the ion core. The corresponding times are defined as the electron emission times, denoted as t_1 and t_2 for the two electrons respectively. The difference between the two emission times is denoted as Δt, as illustrated in Fig. 3. We will show that the electron correlation shown in Fig. 2 is related closely to this time difference.

A simple analytical theory is sufficient to understand the electron correlations induced by the *CP* pulse. The theory starts from the time when an electron is ionized and ignores the Coulomb attraction afterward. The initial velocity of this electron is assumed to be zero. This theory was first

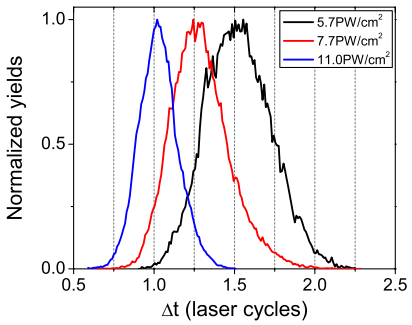

FIG. 4 (color online). Distributions of the time difference Δt between the two emissions for three different intensities: 5.7 PW/cm^2 (black line), 7.7 PW/cm^2 (red line), and 11.0 PW/cm^2 (blue line).

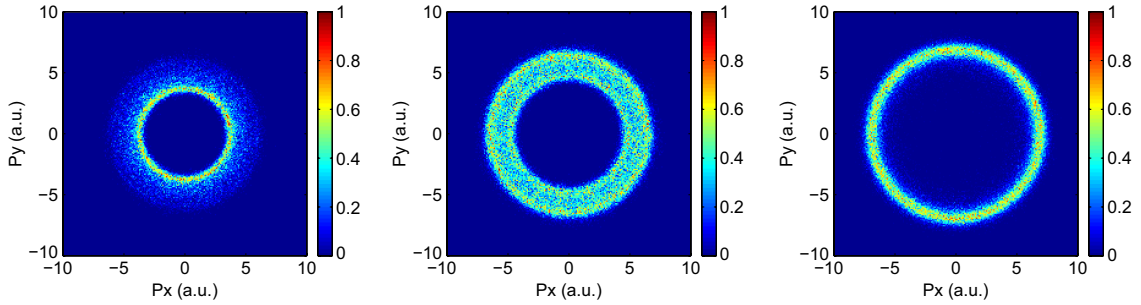

FIG. 5 (color online). Momentum distributions of the resultant doubly charged ions, for the same three intensities of Fig. 2.

used by van Linden van den Heuvell and Muller over twenty years ago and was given the name "Simpleman theory" [26]. Although extremely simple, the Simpleman theory has been very effective for intuitive understanding of many strong-field phenomena.

It is straightforward to derive that an electron emitted at time t_1 with zero momentum (velocity) will gain these x and y momenta at the end of the pulse:

$$p_{1x} = -\frac{E_0}{\omega} f(t_1) \cos(\omega t_1 + \phi) = -\frac{E_y(t_1)}{\omega}, \quad (4)$$

$$p_{1y} = +\frac{E_0}{\omega} f(t_1) \sin(\omega t_1 + \phi) = +\frac{E_x(t_1)}{\omega}. \quad (5)$$

Therefore the final emission angles θ_1 and θ_2 are determined by t_1 and t_2:

$$\theta_1 = \pi - (\omega t_1 + \phi), \quad (6)$$

$$\theta_2 = \pi - (\omega t_2 + \phi). \quad (7)$$

So the angle between the two electrons is determined by the time difference between the two emissions

$$\theta = \theta_1 - \theta_2 = \omega(t_2 - t_1) = \omega \Delta t. \quad (8)$$

If the time difference is an integer number of optical cycles (i.e., $1, 2, 3, \ldots$), then the two electrons are most likely to be emitted into the same direction; if the time difference is an odd multiple of a half cycle (i.e., $1/2, 3/2, 5/2, \ldots$), then the two electrons are most likely to be emitted into opposite directions; if the time difference is an odd multiple of a quarter cycle (i.e., $1/4, 3/4, 5/4, \ldots$), then the two electrons are most likely to be emitted perpendicularly to each other.

The recorded time differences corresponding to the three cases shown in Fig. 2 are shown in Fig. 4. For $I = 5.7$ PW/cm^2, Δt peaks at 1.5 cycles, so the two electrons are more likely to be emitted into opposite directions. For $I = 7.7$ PW/cm^2, Δt shifts to the left and peaks at 1.25 cycles, so the two electrons are more likely to be emitted perpendicularly to each other. For $I = 11.0$ PW/cm^2, Δt shifts further to the left and peaks at 1.0 cycle, so the two electrons are most probably emitted into the same direction.

It is natural to expect that the momentum distribution of the resultant doubly charged ion will reflect the correlation between the two electrons. The net momentum of the ion is expected to be larger if the two electrons are emitted into the same direction than if the two electrons are emitted into opposite directions. The concept is analogous to the non-Z and Z trajectories found with linear polarization [27]. This point is confirmed by the momentum distribution of doubly charged ions, as shown in Fig. 5, for the same three intensities. For $I = 5.7$ PW/cm^2, the two electrons are emitted into opposite directions, so that the inner part of the ring structure is more populated than the outer part, indicating a relatively small net ion momentum. In the other end, for $I = 11.0$ PW/cm^2, the two electrons are emitted into the same direction, so that the outer part of the ring structure is more populated (the inner part can barely be seen in this case), indicating a relatively large net ion momentum. For the middle panel with intensity 7.7 PW/cm^2, the whole ring structure is relatively evenly populated.

Note that although the emission directions of the two electrons are strongly correlated, the absolute emission direction is random. This point can be seen from the circularly symmetric feature shown in all three ion momentum distributions and it is a direct consequence of the nonstabilized laser phase. If phase-stabilized pulses are used, this circular symmetry will be broken. However, the relative angle θ, or the angular correlation between the two electrons, will not be affected.

The correlation angle is not limited to the above-mentioned three special values. In fact, any value is possible and the relation between the correlation angle and the emission time difference is given in Eq. (8). For example,

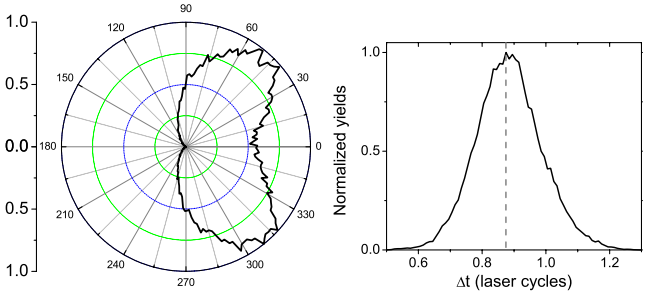

FIG. 6 (color online). The distributions of θ and of Δt for intensity 15.0 PW/cm^2.

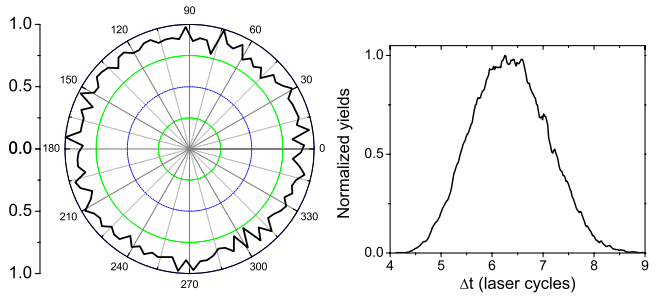

FIG. 7 (color online). The distributions of θ and of Δt for a 20-cycle pulse with intensity 5.7 PW/cm^2.

Fig. 6 shows the distributions of the emission angle and of the time difference for intensity 15.0 PW/cm^2. The time difference Δt is centered at 0.875 laser cycles and a correlation angle of 45° is expected, which is confirmed by the distribution of θ.

The existence of this angle correlation depends critically on the pulse duration. As shown in Fig. 4, Δt must be localized to be roughly within 1 optical cycle to generate the directional correlation. The shorter and the stronger the pulse, the better the localization of Δt. That is why the correlation for 11.0 PW/cm^2 is stronger than that for 5.7 PW/cm^2, albeit the same pulse duration has been used. A long pulse can destroy the correlation completely. For example, Fig. 7 shows the angular correlation and the distribution of Δt for a 20-cycle pulse with intensity 5.7 PW/cm^2. Almost all angular correlations are lost because Δt spans several optical cycles.

In summary, we have shown that the two electrons emitted in sequential double ionization by short circularly polarized pulses are correlated in their emission directions. The correlation is shown to be introduced by the external laser field, even though the two emissions may be dynamically independent.

We have further shown that the correlation can be continuously controlled by changing the laser intensity. The two electrons can be controlled to be emitted to the same direction, to perpendicular directions, to opposite directions, or even to any arbitrary direction one might want. The correlation between the two emission directions will have a direct impact on the resultant doubly charged ion, the momentum of which has been shown to have a ring structure. If the two emissions are parallel, only the outer edge of the ring structure is populated; if the two emissions are antiparallel, only the inner edge of the ring structure is populated. This makes short circularly polarized pulses, which can be routinely generated in laboratories, a potentially useful tool to control electron correlations and ion momentum distributions.

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