DEPENDENCE OF TWO-ELECTRON CORRELATED DYNAMICS ON THE RELATIVE PHASE OF TWO-COLOR ORTHOGONAL LASER PULSE

HUYNH VAN SON[*], TRUONG DANG HOAI THU[**],
TRAN HOANG HAI YEN[***], VO THANH LAM[****], PHAM NGUYEN THANH VINH[*****]

ABSTRACT

In this paper, the correlated dynamics between two ionized electrons under the influence of the orthogonal two-color laser pulse consisting of 800-nm and 400-nm fields were analyzed. Trajectory analysis indicates that the moment of double ionization and the repulsive force between two ionized electrons are responsible to the strong modification of the two-electron momentum distribution in the direction parallel to the polarization axis of 800-nm field with respect to the variation of the relative phase of the pulse. The out-of-plane effect is also considered to explain the dependence of He^{2+} yield on the relative phase.

Keywords: nonsequential process, double ionization, classical ensemble model, orthogonal two-color laser pulse, relative phase.

TÓM TẮT

Sự phụ thuộc của động lực học tương quan giữa hai electron vào pha tương đối của xung laser hai màu trực giao

Trong bài báo này, quá trình động lực học tương quan giữa hai electron dưới tác dụng của xung laser hai màu trực giao bao gồm trường 800nm và 400nm được phân tích. Phép phân tích quỹ đạo chỉ ra rằng thời điểm ion hóa kép và lực đẩy giữa hai electron ion hóa chính là nguyên nhân gây ra sự thay đổi mạnh trong phổ động lượng tương quan của hai electron đó theo phương song song với trục phân cực của trường 800-nm khi pha tương đối của laser được thay đổi. Hiệu ứng ngoại phẳng cũng được xem xét để giải thích sự phụ thuộc của tín hiệu He^{2+} vào pha tương đối.

Từ khóa: quá trình không liên tiếp, ion hóa kép, mô hình tập hợp cổ điển, laser hai màu trực giao, pha tương đối.

1. Introduction

When an atom or a molecular is exposed to an oscillating laser pulse, its electron can be ionized. The ionized electron is first accelerated, then decelerated and driven back as the laser pulse reserves its direction to recollide with the parent ion. The

[*] M.Sc. Student, University of Science Ho Chi Minh City; *Email: sonhuynh_23@yahoo.com.vn*
[**] M.Sc. Student, Ho Chi Minh City University of Education
[***] Student, Sai Gon University
[****] Ph.D., Sai Gon University
[*****] Ph.D., Ho Chi Minh City University of Education

recollision process is the root of the strong-field induced nonlinear dynamics of current interests such as the generation of high-order harmonic [1, 2], above-threshold electron emission [6], double or multiple ionization [5, 8]. Among them, nonsequential double ionization (NSDI) process is scrutinized as a tool to comprehensively study the electron-electron (*e*-*e*) correlation toward the recollision process [8]. In addition, how to control the motion of the ionized electronic wave packets in time domain with attosecond resolution is a hot topic in recent years. Orthogonally polarized two-color (OTC) laser pulses are considered to be a powerful tool for this problem since they allow us to establish an attosecond time scale in the polarization plane of both the emitted and recolliding wave packets [4]. The OTC laser fields are widely used in attosecond physics such as interrogating atomic and molecular orbital structure via high harmonic radiation [9], steering electrons in laser induced electron diffraction [7] and double ionization [14]. Numerically, there are two well-known approaches to the problem of NSDI. The first one is TDSE (Time Dependent Schrödinger Equation) method providing the exact solutions. However, this consideration is extremely tedious by means of computational demand, and can only grant the final output. Therefore it is difficult to deeply understand the underlying dynamics beneath the results using this method. For implementing TDSE, the classical ensemble model is proved to give results which are in good consistency to those using quantum consideration provided that the laser intensity is sufficiently high [3] since the electron is propagated solely under the influence of the oscillating laser field after being ionized [1]. The advantages of the classical calculation over the full-quantum consideration were stated in [3].

Recently, we have been aware of several studies regarding the NSDI process induced by OTC such as the investigation of NSDI of Ne close to the saturation regime [13] and for a wide range of laser intensities [12]. The correlated electron dynamic in NSDI process of He is also controlled by the variation of the relative phase $\Delta\varphi$ of the OTC pulses [14]. The investigation in case of He, however, is restricted to the polarization plane of the OTC pulses, thus omits the out-of-plane effect. Therefore, the dependence of He^{2+} yield on $\Delta\varphi$ as well as the peculiar butterfly-like shape in the correlated two-electron momentum distribution (CTEMD) along the polarization direction of the major field (800-nm field) are still vague. Hence this is deserved to deeper consider the NSDI of He induced by OTC laser pulse.

In this work, we extend the investigation in reference [14] by using classical model for full three-dimensional space, thus it is possible to investigate the behavior of the momentum distributions in the direction perpendicular to the polarization plane of the OTC pulse where there is no external force exerting on the ionized electrons. These momentum distributions are called transverse momentum distributions (TMDs) which contain rich information of the returning wave packet as well as the atomic or molecular shape. We use the OTC laser pulse consisting of 800-nm and 400-nm laser fields whose polarization axes are perpendicular to each other at intensity of 5.0×10^{14}

W/cm^2. By varying the relative phase $\Delta\varphi$, we figure out that the He^{2+} yield has maxima around $\Delta\varphi = n\pi/4$ and minima around $\Delta\varphi = (n+0.5)\pi/4$ with $n \in \mathbb{Z}$. Although there is no experimental data for He to compare with, the similarity of this behavior in case of He to that of Ne observed in both experiment and simulation [12, 13] validates our result. In this paper, we concentrate on the evolution of the CTEMD along the polarization axis of 800-nm field as the relative phase $\Delta\varphi$ varies since the correlated dynamic between two ionized electrons can be observed obviously in this direction [14]. By using back trajectory technique [3], we indicate that the delay in double ionization process plays vital role in forming the drift momenta of two ionized electrons. Moreover the *e-e* repulsive force is figured out to be the root of the butterfly-like shape in the CTEMD along the polarization axis of the 800-nm field at $\Delta\varphi = 0.35\pi$. These features are also embedded in the TMD as expected.

The paper is organized as follows. In section 2, we briefly introduce the classical ensemble model used to consider the NSDI process under the influence of OTC laser pulse. In section 3, we present and discuss the numerical results for the dependence of He^{2+} yield on the relative phase of the OTC laser pulse as well as the *e-e* correlated dynamic resulting in the behavior of CTEMD along the polarization direction of 800-nm field. Section 4 concludes the paper.

2. Three-dimension classical ensemble model

In the classical model, the evolution of the two-electron system is determined by the explicitly classical equations of motion (unless otherwise stated, atomic units are used throughout this paper)

$$\frac{d^2x}{dt^2} = -\frac{2x_i}{\left(x_i^2+y_i^2+z_i^2+a\right)^{3/2}} + \frac{\left(x_i-x_j\right)}{\left[\left(x_i-x_j\right)^2+\left(y_i-y_j\right)^2+\left(z_i-z_j\right)^2+b\right]^{3/2}} - E_x(t), \quad (1a)$$

$$\frac{d^2y}{dt^2} = -\frac{2y_i}{\left(x_i^2+y_i^2+z_i^2+a\right)^{3/2}} + \frac{\left(y_i-y_j\right)}{\left[\left(x_i-x_j\right)^2+\left(y_i-y_j\right)^2+\left(z_i-z_j\right)^2+b\right]^{3/2}} - E_y(t), \quad (1b)$$

$$\frac{d^2z}{dt^2} = -\frac{2z_i}{\left(x_i^2+y_i^2+z_i^2+a\right)^{3/2}} + \frac{\left(z_i-z_j\right)}{\left[\left(x_i-x_j\right)^2+\left(y_i-y_j\right)^2+\left(z_i-z_j\right)^2+b\right]^{3/2}}. \quad (1c)$$

Here *a* and *b* are the softening parameters which are chosen as 0.75 and 0.01, respectively, in order to avoid autoionization [10]. $E_x(t)$ and $E_y(t)$ are the *x* and *y* components of the OTC laser pulse taken the explicit forms as $E_x(t) = E_0\cos(\omega t)$ and $E_y(t) = E_0\cos(2\omega t + \Delta\varphi)$, respectively. The intensities of both fields are set to be 5.0x10^{14} W/cm^2. To obtain the initial condition, the ensemble is populated starting from a classically allowed position for the helium ground-state energy of -2,9035 a.u. The

available kinetic energy is distributed between two electrons randomly in momentum space. Then the electrons are allowed to evolve a sufficiently long time (200 a.u.) in the absence of the laser field to obtain stable position clustering around the core locating at the origin (see figure 1) and stable momentum distribution [10]. Having this initial condition, we numerically solve equation (1) for individual atom in the influence of the laser field by using well-known Runge-Kutta method [11]. Then the energies of two ionized electrons of each atom are analyzed at the end of the pulse. The atom is considered to be double ionized only if the energies of both electrons are positive [3, 10] (read [10] for more details). We note that in the framework of the classical model, no tunneling ionization occurs, both ionized electrons are set free via over-the-barrier mechanism. Indeed the laser intensity used in our consideration is sufficiently high to suppress the atomic potential so that the electron can transfer to the continuum state by over-the-barrier ionization. In order to obtain stable results, we use ensemble sizes as two millions of atoms.

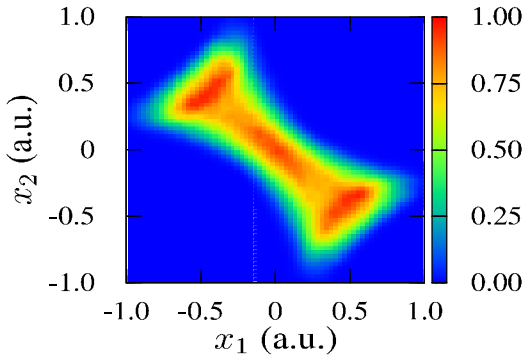

Fig 1. *Spatial distribution of two bounded electrons in x axis along the polarization axis of 800-nm laser pulse*

3. Numerical results and discussion

We proceed to discuss the NSDI process of He by the OTC laser pulse whose parameters are indicated in section 2. Firstly, the dependence on the relative phase $\Delta\varphi$ of the He^{2+} yield is illustrated in figure 1. The yield is normalized in such a way that the maximum value is equal to unity. Note that at the laser intensity used in our calculation, the signals of He^{2+} are mostly associated with NSDI process, the SDI process is more considerable at higher intensity. In addition, the results are presented only for relative phase chosen to be in the interval $0 \le \Delta\varphi \le \pi/2$ due to the periodicity of the laser pulse. Obviously, He^{2+} yield exhibits strong dependence on $\Delta\varphi$, the maxima occur around $\Delta\varphi = n\pi/4$ while the minima locate around $\Delta\varphi = (n+0.5)\pi/4$, here $n \in$. Another interesting feature can be observed in figure 2 is the knee structures at some intermediate relative phases such as around $\Delta\varphi = 0.05\pi$ and 0.3π. Although there is no experimental result relating to this structure, we still strongly believe that it is

reasonable since the similar trend has been observed experimentally [13] and studied theoretically [12] for Ne^{2+}.

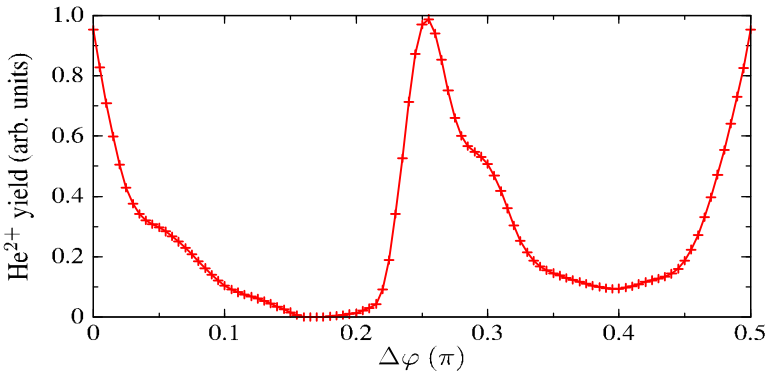

Fig 2. *Dependence of He^{2+} yield on the relative phase $\Delta\varphi$ of the OTC laser pulse*

To understand the transition behavior mentioned above, it is instructive to present the correlated momentum distribution of two ionized electrons along *x* and *z* directions in figure 3 for three representative values of the relative phase $\Delta\varphi$. Note that the polarization plane of the OTC pulse is confined to *x*–*y* plane. The momentum distributions along *z* axis, which are called TMD, are not affected by the pulse, thus they provide a pure signature of the ionized wave packet just after ionization process. The relative phase dependence of the correlated momenta in the polarized plane of laser pulse is deferred to the following discussion, here we focused on the behavior of TMD to investigate strong dependence of He^{2+} yield on $\Delta\varphi$ shown in figure 2. For cases of $\Delta\varphi = n\pi/4$ with $n \in \mathbb{Z}$, the TMDs of two ionized electrons cluster around the origin implying the fact that the evolution of recolliding electron is confined to the polarized plane of the OTC pulse. Hence the possibility to recollide with the parent ion increases resulting in the peak in He^{2+} yield. While for some intermediate values of the relative phase such as 0.35π, the correlated TMD spreads out to cluster around the secondary diagonal. In this case two ionized electrons fly out into full three-dimensional space, thus the revisiting probability is low. This is the root of the minima observed in He^{2+} yield (see figure 2). Especially, there is no double ionization event for $\Delta\varphi$ around 0.15π. Back analysis [3] shows that the first ionized electron strongly diffuses to the perpendicular direction in this case, therefore it cannot return to revisit its parent ion for the next ionization step. The analysis discussed above is the out-of-plane effect which was omitted in reference [14]. Another interesting feature associated with the relative phase dependence of He^{2+} yield is the knee structure obviously observed for $\Delta\varphi$ around 0.3π. In order to explain this feature, the travelling time defining as the time duration between the first ionization and recollision events is also considered since this is another vital factor affecting the NDSI process [12]. We found that the mean travelling time of the recolliding electron in this case is considerably smaller than that in cases

where He^{2+} yield has maximum values, thus increasing the possibility of recollision. Meanwhile the diffusion of this electron is still large. This issue lessens the probability of revisiting process. Hence the knee structure can be straightforwardly explained by the competition between the concentration and mean-travelling-time effects [12]. In conclusion, the out-of-plane effect is noticeable for some intermediate relative phases, thus it is indispensable for analyzing the NSDI of He induced by the OTC laser pulse.

We now discuss the evolution of the CTEMD along the polarization axis of the 800-nm field. For $\Delta\varphi = 0\pi$, the CTEMD exhibits strong correlated relationship, meaning two electrons emit in the same direction after being ionized. While for $\Delta\varphi = 0.25\pi$, a strong anti-correlated behavior reveals, the distribution clusters around the secondary diagonal implying that two electrons have opposite drift momenta. The most interesting feature can be observed in the left column of figure 3 is the butterfly structure appearing in the CTEMD in case of $\Delta\varphi = 0.35\pi$, i.e. neither correlated nor anti-correlated behavior occurs. For the explanation of these behaviors in CTEMD, reference [14] gave an assumption that two electrons are ionized simultaneously after the recollision process happens so that the repulsive force between them is sufficiently strong, which is doubtable. The delay time between recollision and second ionization events always exists. Thus it is a vital factor having to be taken into account in consideration of NSDI process.

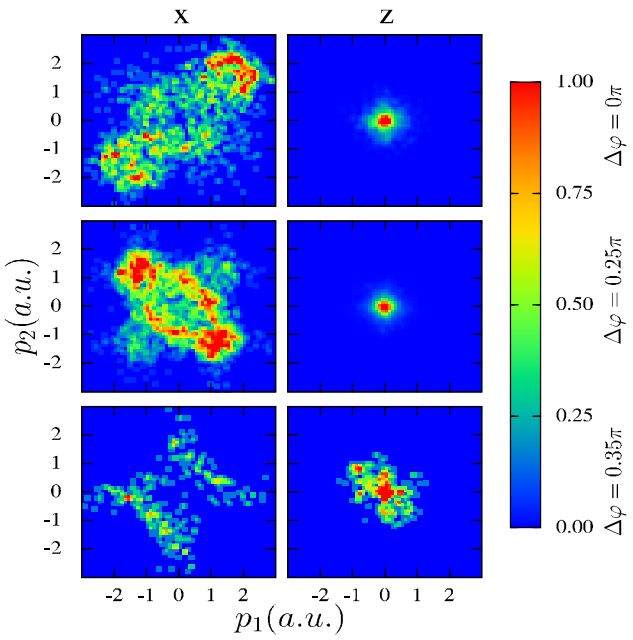

Fig 3. *Correlated two electron momentum distribution for three representative values of the relative phase* $\Delta\varphi = 0\pi$, 0.25π , 0.35π *in x and z axes which are along the polarization axis of 800-nm field and perpendicular to the polarization plane of the OTC pulse, respectively*

In order to explain the evolution of CTEMD observed in figure 3 as relative phase varies, we present in figure 4 the counts of DI events versus the 800-nm laser's phase at recollision and delay times in the left and right panels for similar three representative relative phases as in figure 3, respectively. For $\Delta\varphi = 0\pi$, recollisions that finally leads to the NSDI occurs in a wide range of the laser pulse. While for relative phases different from zero, the recollisions are focused on a much narrower time window. This result means that the OTC pulse is a powerful tool to resolve the NSDI process with much higher temporal resolution. From that figure, this is straightforward to understand the switch from correlated to anti-correlated behavior as $\Delta\varphi$ changes from 0π to 0.25π. Note that the final momentum of ionized electron can be classically estimated as $v = v_0 - (\mathrm{E}_0/\omega)\sin(\omega t_0)$ where v_0 is the initial momentum at the moment of ionization t_0. Back trajectory analysis assists us to indicate that in the first two cases where $\Delta\varphi = 0\pi$ and 0.25π both electrons ionized with almost zero momenta. Thus it is the laser pulse that plays dominant role in controlling the emission dynamic of these two electrons. For $\Delta\varphi = 0\pi$, the first ionized electron recollides with its parent ion when the electric field varies from $-E_0/2$ to $E_0/2$ and the delay time is about $0.1T_1$, hence the second ionization event happens in similar half-cycle part of the pulse leading to parallel emission of these two ionized electrons. While in case of $\Delta\varphi = 0.25\pi$, the delay time is almost equal to that in $\Delta\varphi = 0\pi$ case, however the recollision occurs just before the peak of 800-nm pulse. Therefore the second electron is ionized in different half-cycle part of the pulse resulting in the anti-correlated behavior in CTEMD. For $\Delta\varphi = 0.35\pi$, the recollision occurs while the laser pulse has intermediate field strength and the delay time is very small, around $0.01T_1$. This fact is an evidence of the simultaneous exit of these two ionized electrons.

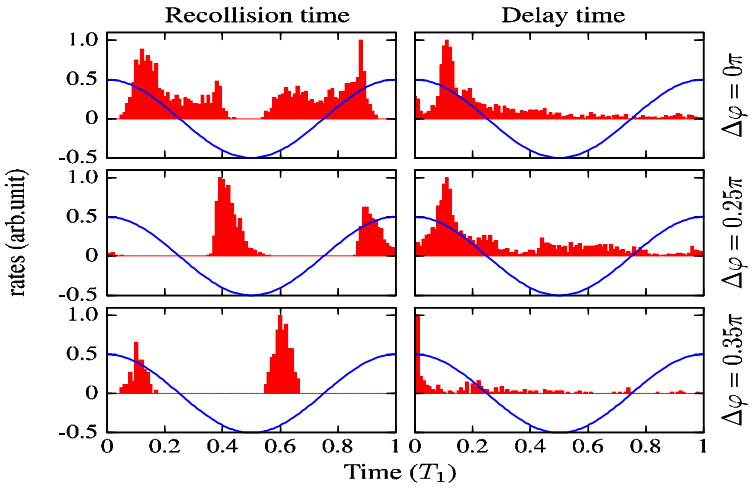

Fig 4. *Counts of DI events versus 800-nm field phase at recollision and delay time. The solid blue curve represents the electric field of 800-nm pulse. T_1 is the optical cycle of this pulse*

The information provided in figure 4 is a hint for explanation of the butterfly structure in CTEMD in case of $\Delta\varphi = 0.35\pi$. The simultaneous emission of two ionized electrons implies the fact that they exert strong repulsive force on each other. This is the dominant effect to form such interesting structure instead of the asymmetric energy sharing (AES) process as claimed in reference [14]. This repulsion effect also imprints in the TMD (see the spreading of the momentum in the right bottom panel in figure 3). In order to confirm that expectation, we operate two further considerations. Firstly the AES effect is considered, the signals are picked up only for the case in which the energy difference between two electrons just after DI events is $\Delta E > 1$ a.u., the result is presented in the left panel of figure 5. Obviously, the butterfly structure still remains. Note that this result is insensitive to the variation of the critical energy difference ΔE from 1 to 2 a.u. which is not shown here. This fact absolutely opposes the conclusion made in reference [14]. In addition we perform second consideration that the final state *e-e* interaction $V_{ee}^{'}(r_1, r_2) = 1/\sqrt{(r_1 - r_2)^2 + b}$ is replaced by the screening Yukawa potential as

$$V_{ee}^{'}(r_1, r_2) = \frac{1}{\sqrt{(r_1 - r_2)^2 + b}} \exp\left(-\lambda\sqrt{(r_1 - r_2)^2 + b}\right), \qquad (2)$$

here the screening factor $\lambda = 5$ is large enough to make the repulsive interaction between two ionized electrons rapidly decreases [10]. The spectrum associated with screening effect is illustrated in the right panel of figure 5. The butterfly structure is faded and the distribution now clusters around to main diagonal. In conclusion, with the parameters of OTC pulse used in this paper, the repulsive interaction between two electrons is dominant over the AES process in forming the butterfly structure observed in CTMED along the polarization axis of 800-nm pulse in case $\Delta\varphi = 0.35\pi$. We still believe that at higher laser intensity, the AES emerges to be more important in constructing such structure since the recolliding electron gains more energy and passes through the core so quickly, thus it leaves smaller portion of energy to the bounced electron for being ionized.

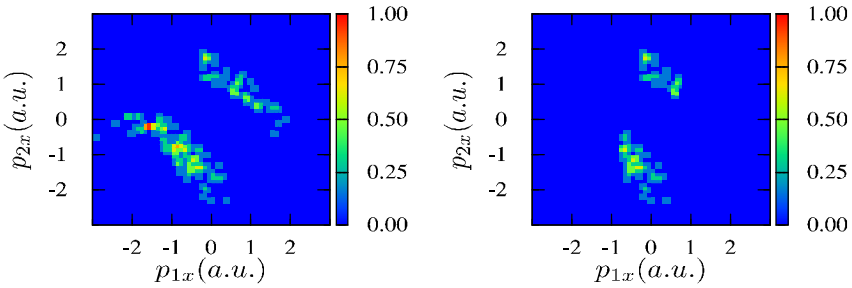

Fig 5. *Correlated electron momentum distribution in x axis for $\Delta\varphi = 0.35\pi$. The left and right panel corresponds to the asymmetric energy sharing and interacting screening effect, respectively.*

4. Conclusion

In conclusion, we have provided a comprehensive explanation of the correlated dynamics of two ionized electrons as the relative phase of the OTC phase varies. The results show that the yield of He^{2+} as a function of the relative phase can be straightforwardly explained using the out-of-plane effect which was omitted in reference [14]. Furthermore, the recollision and delay times determining the moment of double ionization are responsible to the correlated and anti-correlated behaviors in CTEMD for $\Delta\varphi = 0\pi$ and $\Delta\varphi = 0.25\pi$, respectively. Meanwhile in case of $\Delta\varphi = 0.35\pi$, the butterfly structure is originated by the strong repulsion between two electrons just after ionization since they escape from the parent ion almost at the same time. This feature also imprints in the TMD. Experimentally, it is effortless to adjust the relative phase between two electric fields in OTC pulse. Thus our analyses are vital for experimental sake.

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