Classical Trajectory Diagnosis of Finger-Like Pattern in the Correlated Electron Momentum Distribution for Helium Double Ionization

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With a semiclassical quasistatic model we identify the distinct roles of nuclear Coulomb attraction, final state electron repulsion and electron-field interaction in forming the finger-like (or V-shaped) pattern in the correlated electron momentum distribution for Helium double ionization [Phys. Rev. Lett. 99, 263002; ibid. 263003 (2007)]. The underlying microscopic trajectory configurations responsible for asymmetric electron energy sharing after electron-electron collision have been uncovered and corresponding sub-cycle dynamics are analyzed. The correlation pattern is found to be sensitive to the transverse momentum of correlated electrons.

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Nonsequential double ionization (NSDI) of atoms subject to ultrashort intense laser pulses attracts constant interests because it is a prototype model for the study of three-body Coulomb problem intervened by the highly unperturbed interaction of the electrons with the strong laser field. Until recently it has been consensus that rescattering is the dominant mechanism for NSDI [1]. In this three-step mechanism, the first electron is freed by a quasi-static tunneling ionization, and is driven back to its parent ion and imparts part of energy to dislodge a second electron.

The electron recollision picture as a cornerstone of the rescattering mechanism inspires the further investigations that achieve insight into the microscopic dynamics of the ionization process on the timescale of subfemtosecond. The advent of experimental techniques, represented by the sophisticated Cold Target Recoil Ion Momentum Spectroscopy (COLTRIMS), combined with higher-repetition-rate lasers, has to a large extent facilitated this type of study. For example, the observed double hump structure in the ion momenta [2] and the electron momenta correlation [3] parallel to the field gave solid evidence of the time delay introduced by the rescattering process and the emission time of both electrons close to the zero crossing of the oscillating field.

Despite the great success of this picture, a comprehensive understanding of the microscopic dynamics in this recollision process is far from being complete. Indeed, new high resolution and high statistics COLTRIMS experiments on double ionization of helium are performed independently by two groups and a striking finger-like (or V-shaped) structure is observed [1, 3] in the correlated electron momenta parallel to the laser polarization, in qualitative accordance with the prediction of S-matrix approach [2] and quantum mechanical calculation [4]. These high resolution and high statistics COLTRIMS experiments of double ionization (DI) provide benchmark data for comprehensive theoretical treatments.

In this letter, by exploiting an ab initio 3D semiclassical model, we have reproduced essentially all the experimental characteristics, and identified the distinct roles of nuclear Coulomb attraction, final state electron repulsion and electron-field interaction in the formation of the finger-like structure. Furthermore, classical trajectory (CT) analysis facilitates to unveil the sub-cycle microscopic dynamics behind the finger-like structure.

Compared with other approximate approaches extensively employed in strong field double ionization, e.g. the one dimensional quantum model [10], strong field S-matrix calculation [6, 7, 8, 11] and simplified classical methods [12], our semiclassical model has the advantage that all the effects determining the DI ionization process, such as the quantum tunneling, the effective interactions between particles and the laser field as well as the Coulomb focusing effect, can be fully included, while keeping the computational capacity still accessible. The model has achieved great success in explaining various DI phenomena [13], including the excessive DI yield, the recoil momentum distribution of doubly ionized ions, momentum correlation between two emitted electrons, and the energy spectra and angular distribution of photoelectrons.

For simplicity, we just briefly present the theoretical methodology here. We consider a helium atom interacting with an infrared laser pulse. When the laser field is strong enough, one electron is released at the outer edge of the suppressed Coulomb potential through quantum tunneling with a rate given by the ADK formula [14]. The tunneled electron has a Gaussian-like distribution on transverse velocity and zero longitudinal velocity [13]. For the bound electron, the initial position and momentum are depicted by single-electron micro-canonical distribution (SMD) [15]. The subsequent evolution of the two electrons with the above initial conditions are governed by Newton’s equations of motion: $\frac{d^2 r_i}{dt^2} = \epsilon(t) - \nabla r_i (V_{ne} + V_{ee})$. Here the index i denotes the two different electrons, $V_{ne} = -\frac{2}{r_1}$ and $V_{ee} = \frac{1}{r_1 - r_2}$.
cates the classical limit of the tunneled electron momentum. The most important experimental feature, i.e., the finger-like postcollision electron-electron interaction supposed to be at the time of emission is assumed to be small and the finger-like structure is striking and contradicts to the trajectory of oscillating electric field. The cycle averaged quiver energy of a free electron in an weak. Therefore, the parallel momentum \( p' \) is softened.

The above Newtonian equations are solved by employing the standard 4-5th Runge-Kutta algorithm and DI events are identified by energy criterion. In our calculations, more than 10^7 weighted (i.e., by the tunneling rate) classical two-electron trajectories are traced until one electron moves to such a position that \( r_1 > 200a.u. \). This results in more than 10^4 DI events for statistics.

The resulting electron momentum distribution, calculated with this semiclassical model for the same parameters as in the experiment [3], is shown in Fig. 1(a). The calculation reproduces many key features observed in the experiment, including the emission of the two electrons primarily into the same hemisphere, the small circular accumulation around the zero momentum surrounded by four elliptical hard-to-reach regime, and more importantly, the finger-like structure beyond the limit of 2\( \sqrt{U_p} \). Here \( U_p = \frac{e^2}{(4\omega^2)} \), the ponderomotive energy, refers to the cycle averaged quiver energy of a free electron in an oscillating electric field.

The off-diagonal and beyond-limit properties of the finger-like structure is striking and contradicts to the traditional scenario on DI, in which the electron momentum at the time of emission is assumed to be small and the postcollision electron-electron interaction supposed to be weak. Therefore, the parallel momentum \( k_{1,2}^\parallel \) of each electron results exclusively from the acceleration in the optical field: \( k_{1,2}^\parallel = 2\sqrt{U_p}\sin\omega t_{\text{ion}} \) with ionization time \( t_{\text{ion}} \).

We now proceed to explore the physical effects that give rise to this peculiar finger-like structure. In the context of strong field double ionization, there are essentially three major effects that may play significant role in the double electron emission dynamics: electron-laser field interaction that occurs throughout the DI process, electron-nuclear Coulomb interaction in the post-collision duration and the inter-electron Coulomb repulsion which becomes significant when both electrons get close. Below we investigate all three interactions and clarify their distinct roles in the formation of the finger-like structure.

The first step is to check the role of the external laser field and an additional calculation is thus performed, in which the laser field is intentionally removed and the tunneled electrons are replaced by a beam of projectile electrons; (c) the electron-electron Coulombic interaction is replaced with a Yukawa potential; (d) the nuclear Coulomb interaction is softened.

FIG. 1: (Color online). (a) Distribution of correlated electron momenta along the laser polarization for Helium DI irradiated by 800nm, 4.5 \times 10^{14} \text{W/cm}^2 laser pulses. The black box indicates the classical limit of the tunneled electron momentum. (b) The laser field is removed and the tunneled electrons are replaced by a beam of projectile electrons; (c) the electron-electron Coulombic interaction is replaced with a Yukawa potential; (d) the nuclear Coulomb interaction is softened.

Last but not the least procedure is to justify the role of the electron-nuclear interaction, which is commonly believed to be the main reason for the recoil collision in field-free (e, 2e) process. This interaction was also suggested to be the very ingredient for the field-assisted recoil collision in the context of intense field DI of atomic helium [4]. Accordingly, an additional calculation, in which we soften the nuclear Coulomb attraction by em-
the prominent finger-like structure, i.e., the regimes of $|k_i^j| > 2 \sqrt{U_p}$, and at DI moment (d). Here, the statistics is only collected for the DI trajectories that contribute to the finger-like structure and trace back their origin, i.e., the regimes of $1.5$ a.u. $< |k_i^j| < 2.0$ a.u. and $2.5$ a.u. $< |k_i^j| < 3.0$ a.u., where $i, j = 1, 2, i \neq j$. The dashed curves represent the laser field for guiding eyes.

Physically, the shielding of nuclear potential would to a great extent diminish the Coulomb focusing effect that have significant effects upon both electrons. Clearly, a Coulombic potential would attract the tunneled electron more dramatically when it moves near the atomic core. Such strong attraction may unambiguously bring the tunneled electron to share more kinetic energy with the bound one. For the bound electron, after achieving considerable transferred momentum upon collision, it may elastically backscatter from the Coulombic core on its way out of the atom. This double scattering process is coined as recoil collision [17] and was routinely found in traditional electron impact ionization experiments, especially when the projectile electron possesses the energy of only a few times of the binding energy of the inner one. The result shown in Fig. 1(d) indicates that Coulomb focusing effect is decisive for the production of the electrons with high energy, and thus for the finger-like structure.

The dynamics behind the finger-like structure observed in the experiments was supposed to be related with the recoil collision in the presence of the external laser field [1]. This conjecture has been justified by a 2-body 3D quantum mechanical simulation of reduced dimensionality [18]. However the dynamical details of such collision is hardly explored in the quantum mechanical treatment. In the remaining part of this letter, we proceed to unveil the collision dynamics of the DI electrons, especially those that contribute to the finger-like structure beyond the limit $2 \sqrt{U_p}$. With the CT approach, this becomes possible by back-analyzing the history of the DI events of interest.

It has been recognized that the characteristics of the DI trajectory can be well represented by the recollision and DI time [12]. We thus provide such an information for the trajectories that contribute to the finger-like structure in Fig. 2(c) and (d). It is found that, in Fig. 2(c), the electron pairs contributing to the finger-like structure tend to encounter right at zero field. Within re-scattering picture, this can be understood as that these trajectories include the most energetic collisions for which the tunnelled electrons are released at the laser phase of about $17^\circ$, and return around the zero crossing of the electric field at $270^\circ$ with maximal energy of $3.17 U_p$ [16, 19].

Upon recollision, the bound electron may be directly freed, a process termed as collision ionization (CI), or be excited and subsequently ionized by the next field maximum, known as collision-excitation ionization (CEI) [20]. The smaller and larger peaks in Fig. 2(d) correspond to these two mechanisms, respectively. The smaller ones around zero field represents the electron pairs emitted after a very short thermalization process ($\sim$ attosecond) [21]. While the larger peaks correspond to CEI events that are ionized after a few optical cycles delay leading to a $0.3 \pi$ phase difference from the collision phase peaks [22]. The above calculation indicates that both CI and CEI types of DI events contribute to the finger-like structure. The time delay in CEI could lead to an off-diagonal momentum distribution apart from $k_i^j = k_i^j$, but, nevertheless, cannot account for the excess momentum that spills over the limit of $2 \sqrt{U_p}$.

To further unveil the microscopic mechanism underlying this unusual pattern, we collect all trajectories that constitute the finger-like structure and trace back their dynamical evolution at collision and post-collision. With the CT diagnosis we sketch two types of trajectory configurations that are responsible for the asymmetric electron energy sharing after electron-electron collision and leads to the finger-like structure beyond the limit of $2 \sqrt{U_p}$ (see Fig. 2(a) and (b), correspond to type-I and II configurations, respectively). While the tunneled electron is driven back to the nucleus by the laser field, the field strength reduces to zero and the collision is essentially a field free three-body system under the pure Coulomb potential. For type-I trajectories, as shown in Fig. 2(a), the electron-electron collision near nucleus could lead to following consequences: the second electron acquires considerable momentum from the returned electron and emits in the forward direction, while the returned electron is slowed down. Under the influence of the nuclear attraction, the latter is transferred to a hyperbolic orbit around the nucleus with a large scattering angle. In this way, the returned electron reverses its direction in a time scale of attoseconds after the collision. Notice that meanwhile the laser field changes its direction. As the returned electron has nonzero residual momentum parallel to the instantaneous field direction and is further accelerated by the field, its final longitudinal momentum is expected to be above the limit $2 \sqrt{U_p}$. For the second
electron, after collision its initial momentum is opposite to the instantaneous field direction, one expects that its final longitudinal momentum is below the limit $2\sqrt{U_p}$. In Fig. 2(b) where a type-II trajectory is schematically shown, the situation is similar except that the roles of the two electrons have exchanged: Under assistance of nuclear Coulomb attraction, the second electron acquires a nonzero momentum parallel to the instantaneous field and therefore emits with a longitudinal momentum larger than $2\sqrt{U_p}$. Although the returned electron is slowed down by electron-electron repulsion, it still has a residual momentum opposite to the instantaneous field, resulting in a final longitudinal momentum below $2\sqrt{U_p}$. Our statistics reveals that the DI events in the finger-like structure consists of 30% type-I configuration and 70% type-II configuration.

The above analysis reveals that the electron-electron collision assisted by the nuclear attraction is crucial for the emergence of finger-like structure. In certain cases that the collision is strong, the finger-like structure should be more prominent. To test this idea, we impose an additional confinement on our statistics to observe the variation of the correlated momentum patterns with respect to the relative perpendicular momentum between two electrons. The results are presented in Fig. 3.

It is shown in Fig. 3(d) that the relative perpendicular momentum $k_{12}^\perp$ for all DI events distributes over an interval of $[0,1.4]$ and exhibits a notable accumulation around 0.5 a.u.. Moreover, the finger-like pattern is more prominent for the case of small relative perpendicular momentum (see Fig. 3(a)). As we increase the value of $k_{12}^\perp$ the two finger patterns start to merge (Fig. 3(b)) and finally totally disappear (Fig. 3(c)). The above result is in agreement with the experimental observations [22], and provides a good explanation for the 1D quantum calculation in [10]. Due to dimensional restriction, the quantum calculation made there greatly magnifies the electron-electron collision effects. As a result, a butterfly-like structure similar to Fig. 3(a) emerges (see Fig. 1 of [10]).

In summary, with a semiclassical quasistatic model we have made the CT diagnosis on the finger-like structure observed in recent experiments, and unveiled the microscopic mechanism behind the striking pattern. Our results suggest that the finger-like structure is generated by the interplay of backscattering of the returning electron and the electron-electron Coulomb repulsion.

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