Recoil collisions as a portal to field-assisted ionization at near-uv frequencies in the strong-field double ionization of helium

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(Received 26 May 2008; published 19 August 2008)

We explore the dependence of the double ionization of the He atom on the frequency of a strong laser field while keeping the ponderomotive energy constant. As we increase the frequency we find that the remarkable "fingerlike" structure for high momenta recently found for $\omega=0.055$ a.u. persists for higher frequencies. At the same time, at $\omega=0.187$ a.u., a new X-shaped structure emerges for small momenta that prevails in the correlated momenta distribution. The role of this structure as a signature of the frequency dependence of nonsequential double ionization is discussed.

DOI: 10.1103/PhysRevA.78.023411 PACS number(s): 32.80.Rm, 31.90.+s, 32.80.Fb, 32.80.Wr

The double ionization of the helium atom driven by an infrared laser field at intermediate intensities of $10^{13}-10^{15}$ W/cm^2 has attracted considerable interest over the last few years as a prototype system for the study of the correlated emission of two electrons in a driven atom. In this range of parameters, double ionization proceeds via the rescattering mechanism [1]: the latter is a three-step process where first one electron tunnels to the continuum, then it is accelerated, and finally it is driven back by the laser field to its parent ion, where it transfers energy and liberates the still bound electron.

Although the rescattering model has worked well in providing an interpretation of the basic strong-field phenomena, such as above-threshold ionization, high-order harmonic generation, and nonsequential double ionization (NSDI), recent refinements in experimental investigations [2,3], have revealed additional structure in the latter, specifically the so-called fingerlike structure (V shape) in the correlated momenta of the outgoing electrons, suggesting the presence of an underlying layer of effects. Their interpretation so far rests on a further interaction of the rescattering electron with the nucleus, while in one version [3] the state of the core appears to play a decisive role. At the same time, the possible influence of recollision excitation with subsequent tunneling ionization (RESI) in the fingerlike structure seems to be ruled out according to Ref. [2]. Moreover, work at 390 nm radiation seems to suggest that the presence of the laser influences NSDI beyond the recollision [4,5]. Thus, although considerable insight into the basic underlying mechanism for a fingerlike structure in the momentum distribution of the electrons has been gained, it appears that a definitive quantitative interpretation may have to await further work.

In the present paper, we explore the frequency dependence of NSDI. Much of its physical interpretation relies on the long wavelength ($\sim$800 nm) under which essentially all of the experiments have been performed. Although it is understood that under much shorter wavelength, and comparable intensity, the rescattering mechanism eventually ceases to be valid, the transition from low to higher frequency remains an unexplored question. Our aim is to explore the dependence, if any, of the fingerlike structure on the wavelength of the radiation while keeping the ponderomotive energy constant. We show that the fingerlike structure for large values of momenta, recently found for $\omega=0.055$ a.u., persists for higher frequencies as well. At the same time a surprising X-shape-like structure prevails for high frequencies. We find that this structure is related to a shift of the time of minimum electron-electron approach (recollision time) from $(2/3+n)T$ for small frequencies to $T/2$ for higher ones. In contrast to smaller frequencies, we find that for higher frequencies the target electron is significantly affected by the field and moves away from the nucleus before the rescattering electron reaches the nucleus.

Our approach is quasiclassical, but fully three dimensional. That alone would not be a sufficient justification, if it were not for the fact that it has proven quite useful in providing insight into problems of photon-atom interactions [6,7] for which fully quantum calculations entail prohibitive computational complexity. At this time, no *ab initio*, fully three-dimensional quantum calculation can cope with the computational demands entailed for the aspects addressed in this paper. Nevertheless, a number of judiciously chosen models [8–11], including some classical ones, have proven quite useful in their interpretative and often predictive power.

The quasiclassical model we use entails one electron tunneling through the field-lowered Coulomb potential with a quantum tunneling rate given by the Ammosov-Delone-Krainov (ADK) formula [12]. The longitudinal momentum is zero while the transverse one is given by a Gaussian distribution [7]. The remaining electron is modeled by a microcanonical distribution [13]. For the evolution of the classical trajectories we use the full three-body Hamiltonian in the laser field, that is, $H=p_1^2/2+p_2^2/2-Z/r_1-Z/r_2+1/|\mathbf{r}_1-\mathbf{r}_2|+(\mathbf{r}_1+\mathbf{r}_2)\cdot E(t)\hat{\mathbf{z}}$, with $E(t)$ the electric field (see [7]) linearly polarized along the z axis. The electric field is a cosine pulse that is on for ten cycles and is then switched off in three cycles with a $\cos^2$ envelope. We note, however, a difference between our method of propagation and the one used in [7]: we employ regularized coordinates [14] (to account for the Coulomb singularity) which we believe result in a faster and more stable numerical propagation.

To explore how the fingerlike structure depends on the frequency of the radiation we explore the double ionization for three different frequencies 0.055, 0.11, and 0.187 a.u. In all three cases the ponderomotive energy $U_p=(E^2/(4\omega^2)$ is the same. Thus, the ratio of the time the electron needs to

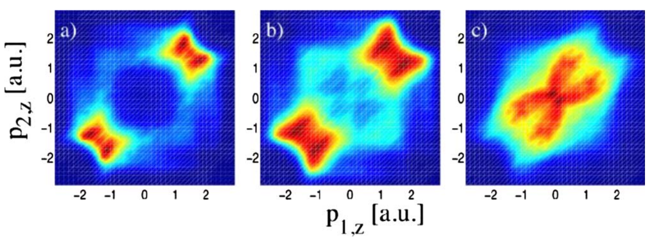

FIG. 1. (Color online) Correlated momenta parallel to the field polarization for ω=(a) 0.055, (b) 0.11, and (c) 0.187 a.u.

tunnel in the field-lowered Coulomb potential to the period of the laser field, the Keldysh parameter $\gamma=\sqrt{I_p/(2U_p)}$ [15], is the same, where I_p is the ionization potential of the He atom. For the frequencies under consideration, the corresponding intensities I, with $I \propto E^2$, are 3×10^{14}, 1.2×10^{15}, and 3.47×10^{15} W/cm^2. In the following, we use the frequency to refer to each case. For the calculations presented, at least 10^5 double-ionization events are obtained, rendering our results quite accurate. Double-ionizing trajectories are propagated even after the electric field is switched off until asymptotic values are reached.

In Fig. 1 we show the correlated momenta of the two electrons for the three different frequencies. A comparison of our result for ω=0.055 a.u. with the experimental one for a pulse duration of 40 fs, wavelength 800 nm, and peak intensity 4.5×10^{14} W/cm^2 [2] shows that we accurately capture the fingerlike structure, which according to Ref. [2] is due to recoil collisions of the rescattering electron. Specifically, at ω=0.055 a.u., this implies that the rescattering electron (denoted as electron 2) impacts the other electron (denoted as electron 1) at times $(2/3+n)T$, with $n=0,1,2,\ldots$. and T the period of the field, undergoing in addition a collision with the nucleus resulting in its backscattering (recoil collision), mostly reversing the direction of its velocity. The above times of recollision are also obtained in our calculation, through the examination of the average potential energy of the electron-electron interaction term and the identification of its maxima.

As a further check of our model, we show now that the fingerlike structure we obtain (Fig. 1) is indeed due to recoil collisions. To this end, we identify the recollision time (the time of minimum approach of the two electrons) through the maximum in the electron pair potential energy. Further, we select those trajectories for which electron 2 backscatters from the nucleus, inverting the direction of its velocity. That is, $155° < \mathbf{p}_{2,\text{aft}}\cdot\mathbf{p}_{2,\text{bef}}/|p_{2,\text{aft}}p_{2,\text{bef}}| < 180°$, with $\mathbf{p}_{2,\text{bef/aft}}$ the momentum of electron 2 just before and after the recollision time. The correlated momenta of the thus selected trajectories, as can be seen in Fig. 2(b), indeed account for the fingerlike structure at ω=0.055 a.u., also reported in Ref. [2]. In agreement with Ref. [2] we find that this structure extends beyond the $2\sqrt{U_p}$ maximum momentum limit (1.6 a.u. in our case). Note first that this structure persists for all three frequencies. In somewhat more detail, in Fig. 2(a) we show the structure for correlated momenta with at least one of the two momenta having magnitude greater than $2\sqrt{U_p}$. We note that the trajectories shown in Fig. 2(b) are a subset of those in Fig. 2(a) and that for the remaining trajectories either elec-

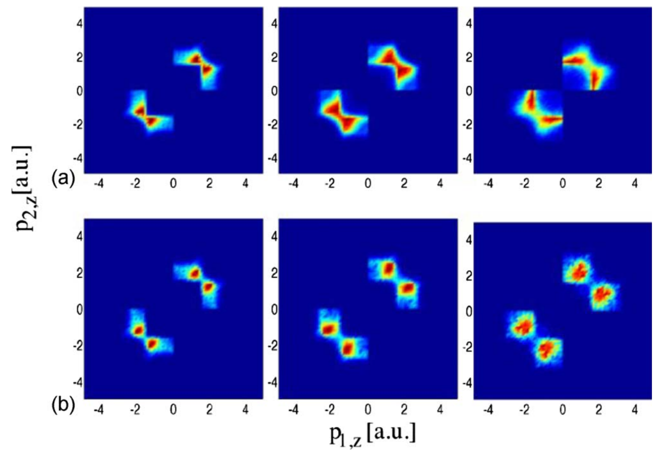

FIG. 2. (Color online) For frequencies from left to right ω =0.055, 0.11, and 0.187 a.u. we plot (a) the correlated momenta using only the trajectories where $p_1 \vee p_2 > 2\sqrt{U_p}$; (b) same as the top panel except that in addition electron 2 is backscattering with $155° < \mathbf{p}_{2,\text{aft}}\cdot\mathbf{p}_{2,\text{bef}}/|p_1p_2| < 180°$.

tron 2 or electron 1 reverses its velocity but with a smaller recoil angle, that is, $90° < \mathbf{p}_{i,\text{aft}}\cdot\mathbf{p}_{i,\text{bef}}/|p_{i,\text{aft}}p_{i,\text{bef}}| < 150°$. Not evident in Fig. 2(a), we find that at the highest frequency the number of trajectories representing $p_1 \vee p_2 > 2\sqrt{U_p}$ decreases and, moreover, the number of trajectories representing "backscattering" in the sense of large recoil angle also decreases; suggesting a reduction of recoil collisions. It is worth noting that we obtain the fingerlike structure in Fig. 2(b) for electrons escaping asymptotically with a very small angle, almost parallel to each other. To a smaller extent, the strong interaction with the nucleus also results in backscattering of either electron 2 or electron 1 with the two electrons escaping at a large angle, resulting in related structure in the second and fourth quadrants of the correlated momenta.

Having established that the interaction of the rescattered electron with the nucleus is responsible for the fingerlike structure, we discuss the imprint of the increasing frequency on the differential probabilities. We note that with increasing frequency the amplitude of excursion of the rescattering electron diminishes. As the frequency changes from 0.055 to 0.187 a.u., we note the following major changes: (a) for increasing frequency the time of closest electron-electron approach shifts from $(2/3+n)T$ to $T/2$, when the velocity of the rescattered electron due to the field is nearly zero; (b) the examination of the average potential energy of electron 2 for the highest frequency reveals an increased effect of the nucleus.

The signatures of increasing frequency that appear to emerge are the following.

(a) There is a significantly less pronounced double hump in the parallel momentum distribution; see Fig. 3(b). For a frequency of ω=0.055 a.u., it is known that a less pronounced double-hump structure results from an increased significance of the RESI mechanism versus the $(e,2e)$ one [16,17]. For that frequency, in both mechanisms the main electron-electron encounters take place at a zero of the field, at times $(2/3+n)T$. However, while the release of the second

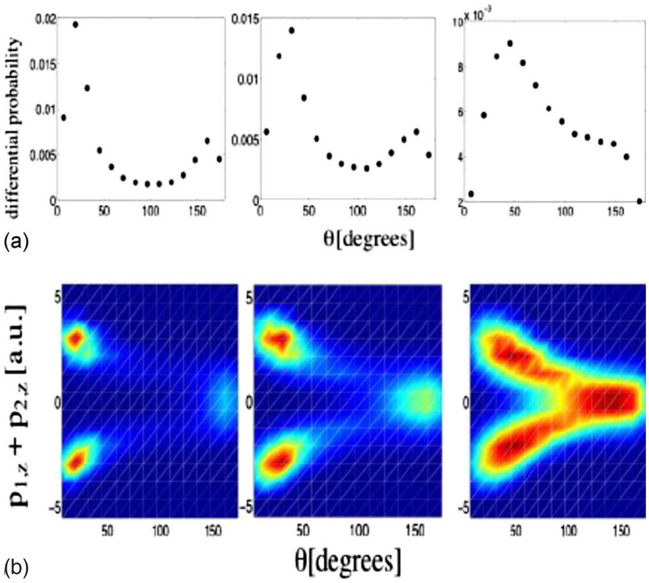

FIG. 3. (Color online) For frequencies from left to right $\omega=0.055$, 0.11, and 0.187 a.u., we plot (a) the distribution of the interelectronic angles of escape binned in 14 intervals, $180°(l-1)/14<\theta<180°/14\times l$ with $l=1,\ldots,14$. Note that the interelectronic angular distribution is plotted per P^{2+}, that is, divided by the double ionization probability. (b) Sum of the parallel momenta as a function of the interelectronic angle of escape.

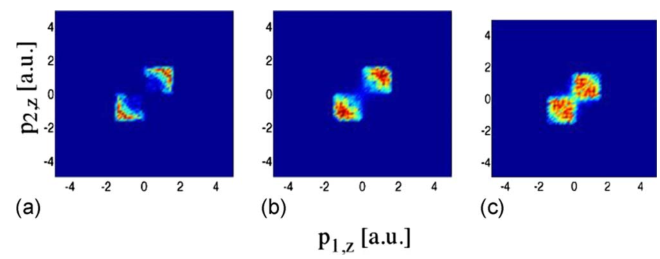

FIG. 4. (Color online) For frequencies $\omega=$(a) 0.055, (b) 0.11, and (c) 0.187 a.u., we plot the correlated momenta with electron 1 recoiling.

electron happens at the same time as rescattering for the $(e,2e)$ process, for RESI it happens later, at a maximum of the field. As a result, in the RESI process the electrons are released with smaller energy filling in the "valley" between the two humps. Is it then possible that for the higher frequency RESI becomes more pronounced? On physical grounds, that might seem reasonable, since the ponderomotive energy responsible for the recollision excitation is the same while the photon energy is bigger. At this stage this is a conjecture that remains to be confirmed.

(b) While for the small frequency $\omega=0.055$ a.u. small interelectronic angles of escape are favored, at $\omega=0.187$ a.u. this is no longer true [see Fig. 3(a)]. As a further check of the compatibility of our calculations with previous work [3], we have computed the inter-electronic angular distribution for $\omega=0.055$ a.u. and $I=1\times10^{15}$ W/cm^2 and find that a 180° escape is less probable compared to the $I=3\times10^{14}$ W/cm^2 case; this is consistent with the fact that with increasing intensity—given that we remain within the nonsequential range—it is more likely that the second electron is ionized through an $(e,2e)$ process. For the higher frequencies, already at $\omega=0.11$ a.u. it appears that interelectronic angles of escape around 90° acquire more prominence; much more so for the highest frequency currently considered, as is evident in Figs. 3(a) and 3(b). Note that, already at $\omega=0.11$ a.u., while for small angles of escape the electron-electron encounters take place at the same times as for $\omega=0.055$ a.u., for angles around 90° the encounters shift to times $T/2$. For the highest frequency $T/2$ is the most probable electron-electron encounter irrespective of the angle of escape. Clearly, Fig. 3(b) (bottom panel), even for larger frequencies when the two electrons escape almost parallel to each other, we still find that the sum of the momenta components parallel to the field is around its maximum possible value of $4\sqrt{U_p}$. For increasing angles of escape the sum of the momenta components decreases significantly for the highest frequency.

Summarizing the results so far, we find that for $\omega=0.11$ a.u. the fingerlike structure for momenta greater than $2\sqrt{U_p}$ becomes more pronounced. However, for the frequency of 0.187 a.u., while the above structure is still present, somewhat unexpectedly a fingerlike structure at smaller momenta emerges, giving rise to an X-like pattern (see Fig. 1). We have already discussed how we identify the trajectories where, in addition to recollision, electron 2 backscatters from the nucleus giving rise to the fingerlike structure for higher momenta. In a similar way, we identify the trajectories where, in addition to electron 2 undergoing a recollision, now it is electron 1 that backscatters from the nucleus for both electron momenta smaller than $2\sqrt{U_p}$. Using the latter trajectories we obtain the correlated momenta shown in Fig. 4. While for frequencies of 0.055 and 0.11 a.u. the trajectories with the additional feature of electron 1 backscattering from the nucleus merely complement the lower part of the fingerlike structure previously discussed for large momenta, this is not the case for $\omega=0.187$ a.u. For the highest frequency, these trajectories give rise to a V-shaped or fingerlike structure for small momenta in both the first and the third quadrants, resulting in an overall X-shaped structure that dominates the correlated momenta distribution (see Fig. 1). Interestingly, while for the case of the fingerlike structure for large momenta the two electrons escape with a small angle, we find that for the new figurelike structure the electrons escape with larger angles. Thus, it is the increased contribution of trajectories with angles of escape around 90° that are responsible for the prevailing X-shaped structure for small momenta.

If one were to single out a major difference in behavior at the higher frequency, it is perhaps encapsulated in Fig. 5 which shows the relative position of the two electrons as a function of time. In both Figs. 5(a) and 5(b) we consider trajectories where in addition to the rescattering of electron 2, electron 1 recoils from the nucleus. In Fig. 5(a) (small frequency) the position of electron 1 does not significantly change until electron 2 reaches the minimum distance from electron 1 which is practically the time of arrival at the nucleus. This happens around a zero of the field at $(2/3+n)T$. On the contrary, in Fig. 5(b) ($\omega=0.187$ a.u.) the time of minimum approach of the two electrons shifts to $T/2$, and

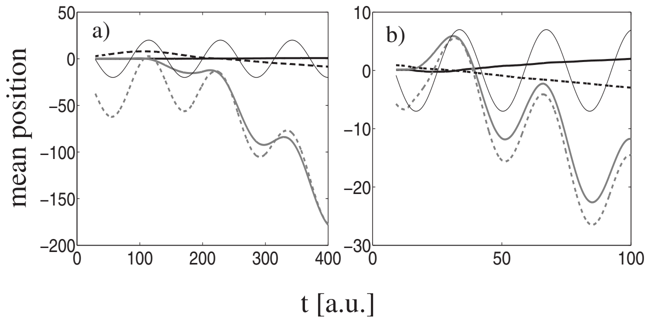

FIG. 5. For frequencies ω=(a) 0.055 and (b) 0.187 a.u. we plot the mean positions for electron 1 (solid lines) and electron 2 (dashed lines) for the x (black) and z (gray) component. Note that these averages are over the trajectories where electron 2 is created in the continuum in the negative z direction, that is, the phase of the field when electron 2 tunnels is between $-\pi/2$ and $\pi/2$.

while electron 2 is still approaching the nucleus, electron 1 is already moving away from the nucleus. It is clearly seen that after the time of minimum electron-electron approach electron 1 responds both to the energy transferred from electron 2 and very importantly to the transfer of energy from the field. For ω=0.055 a.u. the transfer of energy to electron 1 takes place through the rescattering of electron 2. At high intensities—while in the nonsequential regime—this transfer of energy mainly takes place through an $(e,2e)$ process but for smaller intensities through an excitation and subsequent ionization from the field. However, at high frequency the motion of electron 1 is significantly influenced by the field before the return of electron 2 close to the nucleus.

In conclusion, the prevailing X-shaped structure we find for high frequencies is an additional feature of NSDI that will motivate future experiments in this so far unexplored regime of frequencies. Future theoretical work will focus on better understanding the interplay of an $(e,2e)$ collision and the effect of the field and how the increased influence of the field on the target electron before the approach of the rescattering electron to the nucleus is imprinted on the prevailing X-shape-like structure.

I am indebted to Peter Lambropoulos for suggesting the problem and for all the great discussions I had with him throughout this work.

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