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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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 “finger-like” structure for high momenta recently found for ω = 0.055 a.u. 
persists for higher frequencies. At the same time, at ω = 0.187 a.u. a new X-shape 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 non-sequential double ionization is discussed.

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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 [3]: the latter is a three-step 
process where first one electron tunnels to the continuum, 
then it is accelerated and finally 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 the interpretation of the basic strong field 
phenomena, such as ATI (Above Threshold Ionization), 
HHG (High-Order Harmonic Generation) and NSDI 
(Non-Sequential Double Ionization), recent refinements 
in experimental investigations [1, 2], have revealed ad-
ditional structure in the latter, specifically the so-called 
“finger-like” structure (V-shape) in the correlated mo-
menta 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 elec-
tron with the nucleus, while in one version [2], the state 
of the core appears to play a decisive role. At the same 
time, the possible influence of RESI (Recollision Excita-
tion with Subsequent tunneling Ionization) in the finger-
like structure seems to be ruled out according to ref. [1]. 
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 finger-like structure in the momentum distribution of the electrons has been 
gained, it appears that a definitive quantitative interpre-
tation may have to await further work.

In the current letter, we explore the frequency depen-
dence of NSDI. Much of its physical interpretation relies 
on the long wavelength (∼ 800nm) under which essen-
tially all of the experiments have been performed. Al-
though it is understood that under much shorter wave-
length, and comparable intensity, the rescattering mech-
anism 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 finger-
like structure on the wavelength of the radiation while 
keeping the ponderomotive energy constant. We show 
that the finger-like structure for large values of momenta 
recently found for ω = 0.055 a.u. persists for higher fre-
quencies 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 mini-
umon 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 3-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 inter-
actions [6, 7] for which fully quantum calculations en-
tail prohibitive computational complexity. At this time, 
no ab inito, fully 3-dimensional quantum calculation can 
cope with the computational demands it entails for the 
aspects addressed in this letter. Nevertheless, a number 
of judiciously chosen models [8, 9, 10, 11], including some 
classical, 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 ADK for-
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 tra-
jectories 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/r_1 – 
r_2 | + (r_1 + r_2) · E(t) · ẑ, with E(t) the electric field (see [7]) 
linearly polarized along the z-axis. The electric field is a 
cos pulse that is on for 10 cycles and is then switched off.
in 3 cycles with a $\cos^2$ envelope. We note however a difference between our method of propagation and the one used in [2]: 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 finger-like structure depends on the frequency of the radiation we explore the double ionization for three different frequencies 0.055 a.u, 0.11 a.u. 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 tunnel in the field-lowered Coulomb potential to the period of the laser field, the Keldysh parameter $\gamma = \sqrt{T_p/(2U_p)}$ [12], is the same, where $I_p$ is the ionization potential of the He atom. For the frequencies under consideration, the respective intensities $I$, with $I \propto E^2$, are $3 \times 10^{14}$ W/cm$^2$, $1.2 \times 10^{15}$ W/cm$^2$ and $3.47 \times 10^{15}$ W/cm$^2$. In the following, we use the frequency to refer to each case. For the calculations presented, at least $10^8$ 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 $\omega = 0.055$ a.u. with the experimental one for a pulse duration of 40fs, wavelength 800nm and peak intensity $4.5 \times 10^{14}$ W/cm$^2$ [1] shows that we accurately capture the finger-like structure, which according to ref. [1] is due to recoil collisions of the rescattering electron. Specifically, at $\omega = 0.055$ 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 $n = 0, 1, 2, ...$ and $T$ the period of the field, undergoing in addition a collision with the nucleus resulting in its backscattering (recoil collision), with 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 finger-like 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, $155^\circ < \mathbf{p}_{2,af} \cdot \mathbf{p}_{2,be} / |\mathbf{p}_{2,af}||\mathbf{p}_{2,be}| < 180^\circ$, with $\mathbf{p}_{2,be}$ 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. 2b, indeed account for the finger-like structure at $\omega = 0.055$ a.u., also reported in ref [1]. In agreement with ref. [1] 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 details in Fig. 2a 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. 2b are a subset of those in Fig. 2a and that for the remaining trajectories either electron 2 or electron 1 reverses its velocity but with a smaller recoil angle, that is, $90^\circ < \mathbf{p}_{1,af} \cdot \mathbf{p}_{1,be} / |\mathbf{p}_{1,af}||\mathbf{p}_{1,be}| < 150^\circ$. Not evident in Fig. 2a, we find that at the highest frequency the number of trajectories representing $p_1 \wedge p_2 > 2\sqrt{U_p}$ 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 finger-like structure in Fig. 2b 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 finger-like structure, we discuss the imprint of the increasing frequency on the differential probabilities. We note that with increasing frequency the amplitude of excitation of the rescattering electron diminishes. As the frequency changes from 0.055 a.u. 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:

a) a significantly less pronounced double hump in the parallel momentum distribution, see Fig. 3b. For a frequency of $\omega = 0.055$ 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
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 inter-electronic angles of escape are favored, at $\omega = 0.187$ a.u. this is no longer true, see Fig. 3a. As a further check of the compatibility of our calculations with previous work \cite{2}, we have computed the inter-electronic angular distribution for $\omega = 0.055$ a.u. and $I = 1 \times 10^{15}$ W/cm$^2$ and find that a 180$^\circ$ escape is less probable compared to the $I = 3 \times 10^{14}$ W/cm$^2$ case; this is consistent with the fact that with increasing intensity—given that we remain within the non-sequential 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 inter-electronic angles of escape around 90$^\circ$ acquire more prominence; much more so for the highest frequency currently considered as is evident in Figs. 3a and 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$^\circ$ the encounters shift to times $T/2$. For the highest frequency $T/2$ is the most probable escape angle irrespective of the angle of escape. Clearly, Fig. 3b, 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 finger-like 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 finger-like structure at smaller momenta emerges giving rise to a 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 finger-like structure for higher momenta. In a similar way, we identify the trajectories where a second electron undergoes 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 a.u. 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 the finger-like 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-shape or finger-like structure for small momenta in both the first and the third quadrant resulting in an overall X-shape structure that dominates the correlated momenta distribution, see Fig. 1. Interestingly, while for the case of
the finger-like structure for large momenta the two electrons escape with a small angle, we find that for the new figure-like 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-shape 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 Fig. 5a and b we consider trajectories where in addition to the rescattering of electron 2, electron 1 recoils from the nucleus. In Fig. 5a (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. 5b \((\omega = 0.187 \text{ a.u.})\) the time of minimum approach of the two electrons shifts to \(T/2\) and 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 but very importantly to the transfer of energy from the field. For \(\omega = 0.055 \text{ a.u.}\) the transfer of energy to electron 1 takes place through the rescattering of electron 2. At high intensities—while in the non-sequential regime—this transfer of energy mainly takes place through a \((e,2e)\) process while 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-shape structure we find for high frequencies is a new 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.

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

FIG. 5: For frequencies a) \(\omega = 0.055 \text{ a.u.}\), b) \(\omega = 0.187 \text{ a.u.}\) we plot the mean position for electron 1 (solid lines) and electron 2 (dashed lines) for the x component (black) and the z component (grey). Note that these averages are over the trajectories where electron 2 is “born” in the continuum in the negative z direction, that is, the phase of the field when electron 2 tunnels is between \(-\pi/2\) and \(\pi/2\).

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