A two-dimensional, two-electron model atom in a laser pulse: 
exact treatment, single active electron-analysis, 
time-dependent density functional theory, classical calculations, 
and non-sequential ionization

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Owing to its numerical simplicity, a two-dimensional two-electron model atom, with each electron 
moving in one direction, is an ideal system to study non-perturbatively a fully correlated atom 
exposed to a laser field. Frequently made assumptions, such as the “single active electron”-approach 
and calculational approximations, e.g. time dependent density functional theory or (semi-) classical 
techniques, can be tested. In this paper we examine the multiphoton short pulse-regime. We 
observe “non-sequential” ionization, i.e. double ionization at lower field strengths as expected from 
a sequential, single active electron-point of view. Since we find non-sequential ionization also in 
purely classical simulations, we are able to clarify the mechanism behind this effect in terms of 
single particle trajectories.

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I. INTRODUCTION

Several theoretical approaches were able to reproduce experimentally observed ion yields in multi-electron ionization, 
at least qualitatively (see e.g. [1]). Most of them are based on a “single active electron” (SAE) point of view [2]. A new impact on the research in this field had the discovery of the so-called “knee” or “shoulder” in the ionization 
yields of helium exposed to a laser pulse [4]. This means that double ionization occurs many orders of magnitude 
more strongly at intensities where, according to a sequential SAE scenario, almost no He$^{++}$ should be present. Early 
after the experimental observation of this non-sequential ionization (NSI), two possible mechanisms were suggested in 
order to explain it. Corkum proposed a rescattering scenario [5] where the first electron revisits the core and ionizes 
the second electron collisionally. Fittinghoff et. al. suggested a “shake off” effect [4] where the second electron ionizes 
due to the sudden loss of screening of the core by the first electron. Walker et. al. [6] concluded by analyzing their 
experimental data that a rescattering process is not able to explain the observed yields. Their arguments are based 
on the absence of a rigorous threshold in the He$^{++}$ yields. Instead they propose “that NSI occurs via a simultaneous 
two-electron ejection either through a shake off or threshold mechanism involving some form of electron correlation”. 
Recently, the NSI mechanism has been clarified within the intense-field many-body $S$-matrix theory [7]. It was shown 
“that the dominant mechanism behind the observed large probability of laser-induced double escape is a quantum 
mechanical process of absorption of photon energy by one of the electrons which is shared cooperatively with the 
other electron through the Coulomb correlation”. This mechanism for the NSI process was independently deduced 
from 1D He-studies where the model atom had been exposed to a low frequency, short pulse laser field [8]: “[...] 
before the outer electron disappears completely, the inner electron is already sufficiently strongly excited so that it 
leaves the atom within a short time interval later. It is during this time interval that the correlated double ionization 
takes place.” Simulations where the outer electron is calculated in the SAE way but the inner one feels (in a second 
computer run) the time dependent potential created by the outer one, succeeded in reproducing the NSI-“knee” [9]. 
This result is also a strong indication that the suggested mechanisms as quoted above are, indeed, the correct ones. 
However, there is no detailed physical picture how this energy sharing between the outer and the inner electron takes 
place.

Our calculations were performed for a relatively high frequency ($\omega = 0.4$ a.u.) and a very short pulse duration (6 
optical cycles) while in [8] a low frequency short pulse was used. Since we are rather in the multiphoton-regime than 
in the tunneling domain, the occurrence of NSI might be surprising at all. Indeed, in our calculations NSI is relatively 
weak compared to the many orders of magnitude effect for ionization of helium in strong low frequency laser light. 
However, with the help of our additional classical simulations we are able to provide (i) a detailed physical picture 
how NSI takes place in terms of one-particle trajectories, and (ii) a proof that NSI, in its essence, is not a quantum 
mechanical effect.
Because the full quantum mechanical numerical simulation of helium exposed to a laser field is an extremely demanding task \[10\], approximate approaches are desirable. Among these, Hartree-Fock- \[11\], \[13\], time-dependent density functional- (TDDFT) \[16\], \[18\] and semi-classical molecular dynamics-calculations \[24\], \[28\] are most frequently used. Especially the latter method succeeded in reproducing the “knee” \[21\]. On one hand the molecular dynamics calculations are very appealing and instructive since particle trajectories and the single particle energies can be traced. On the other hand the additional “Heisenberg-force” which must be introduced in order to avoid instabilities where one electron falls into the “black hole” (i.e. the nucleus) while the other one ionizes, is somewhat artificial and may evoke objections against the results produced by this method.

The time-dependent Hartree-Fock method was found to be problematic in the framework of multi photon ionization \[13\], \[15\]. Results from TDDFT, in principal an exact approach, depend on the choice of the effective exchange-correlation potential \[24\]. Another disadvantage of this procedure is that only the total electron density \[13\]–\[15\], \[24\] is calculated and the single particle orbitals \(\phi_i(r,t)\) are physically meaningless in a rigorous sense.

The study of systems where the motion of each electron is reduced to one spatial dimension has a relatively long tradition. Potentials of the form \(-Z/x^2 + \epsilon\), so-called “soft core Coulomb potentials”, provide an energetic Rydberg-like scaling \[20\] and lead to results, qualitatively similar to those from full 3D calculations. Two 1D electrons are a two-dimensional system which is tractable with nowadays computers. Two 1D electron-systems are used to study non-perturbatively autoionization \[26\], ionization of a negative ion \[27\], validity of time-dependent Hartree-Fock theory for the multiphoton ionization of atoms \[13\], \[15\], and, most recently, two-electron effects in harmonic generation and ionization \[8\].

This paper is organized as follows. In Section \[\text{II}\] the model system is introduced. In Section \[\text{III}\] results from the 2D quantum calculations are presented. Section \[\text{IV}\] is devoted to an analysis of the results in terms of a SAE-approach. In Section \[\text{V}\] we present the results from a time-dependent density functional-calculation and in Section \[\text{VI}\] we discuss our classical particle simulations within which the NSI scenario can be clarified. Finally, we summarize and conclude in Section \[\text{VI}\].

II. THE 1D HELIUM MODEL

The two 1D electrons with coordinates \(x\) and \(y\) interact with the core and with each other through a “soft core”-interaction, i.e. \(-2/\sqrt{x^2 + \epsilon}\) and \(1/\sqrt{(x-y)^2 + \epsilon}\), respectively, and with the field \(E(t)\) through the dipole term \((x+y)E(t)\) (atomic units (a.u.) will be used throughout this paper). Thus the total Hamiltonian reads

\[
H(x, y, t) = -\frac{1}{2} \frac{\partial^2}{\partial x^2} - \frac{1}{2} \frac{\partial^2}{\partial y^2} - \frac{2}{\sqrt{x^2 + \epsilon}} - \frac{2}{\sqrt{y^2 + \epsilon}} + \frac{1}{\sqrt{(x-y)^2 + \epsilon}} + (x+y)E(t).
\]

The desired ground state energy can be tuned by varying \(\epsilon\). We used

\[\epsilon = 0.55\]

in our calculations which leads to the ground state energy

\[\mathcal{E}_0 = -2.897 \text{ a.u.}\]

on our numerical grid. \(\mathcal{E}_0\) is approximately the ground state energy for the real 3D helium atom which is \(-2.902\).

One may prefer thinking in terms of one 2D particle which moves in the somewhat peculiar 2D potential

\[
V(x, y, t) = \frac{2}{\sqrt{x^2 + \epsilon}} - \frac{2}{\sqrt{y^2 + \epsilon}} + \frac{1}{\sqrt{(x-y)^2 + \epsilon}} + (x+y)E(t)
\]

instead of the two electrons interacting with each other. The potential \(V\) and the ground state energy level are shown in Figure \[\text{II}\] for the three constant fields \(E = 0.0, 0.1, \text{ and } 0.616\). The electric field \(E\) tilts the fieldfree potential around the axis \(y = -x\).

In order to estimate at what fieldstrengths strong single and double ionization should occur, it is advantageous to calculate the classical critical fields. However, the commonly used method of equating the initial ground state energy level to the maximum of the barrier formed by the atomic potential and the external field leads to an unphysically small critical field \(E_\text{crit}^- = 0.009\) for single ionization. In our numerical simulations in the next sections we will find a negligible ionization probability for such low field strengths. Like in the case of hydrogen-like ions, the electron is not
able to move beyond the point where the energy level touches the lowest point of the energetic barrier \[28\]. Therefore the “real” critical field is higher. On the other hand, total ionization should be strong at latest when the ground state energy level exceeds the electron-electron repulsion ridge in \(x = y\)-direction. This is the case at \(E^{++} = 0.616\) (see Figure 1c).

### III. QUANTUM CALCULATION

We used a spectral method \[29\] to solve the time-dependent Schrödinger equation (TDSE)

\[
i \frac{\partial}{\partial t} \Psi(x, y, t) = H(x, y, t) \Psi(x, y, t)
\]

with the Hamiltonian \(H\). In order to keep the numerical effort as small as possible we chose a rather high laser frequency

\[\omega = 0.4\]

and a very short pulse covering 6 cycles,

\[T = 6 \frac{2\pi}{\omega} = 94.248\,\text{.}\]

The pulse envelope was \(\sin^2\)-shaped, thus

\[E(t) = \hat{E} \sin^2(\frac{\pi}{T}t) \sin \omega t\]  

(3)

for \(0 < t < T\). With the frequency chosen and \(\hat{E}\) not greater than 1 we are in the multiphoton-domain since the Keldysh-parameter

\[\sqrt{\frac{|E_0|}{2U_p}}\]

is not much less than unity over the whole intensity region of interest. \(U_p\) is the ponderomotive potential \(\hat{E}^2/(4\omega^2)\), i.e. the mean quiver energy of a free electron in the laser field. A spatial grid spacing \(\Delta x = \Delta y = 0.4\) and a temporal one \(\Delta t = 0.1\) was found to be sufficient. We chose the grid size large enough so that almost no probability density reaches the boundaries within the pulse time \(T\). The ground state wave function was determined by propagating a Gaussian seed function in imaginary time. A contour plot of the ground state probability density is shown in Figure 2. Its energy is \(E_0 = -2.897\).

If we assume that one electron is already ionized we are left with the 1D version of hydrogen-like helium \(\text{He}^+\). The ground state energy of this system,

\[i \frac{\partial}{\partial t} \Psi^+(x, t) = \left( -\frac{1}{2} \frac{\partial^2}{\partial x^2} - \frac{2}{\sqrt{x^2 + \epsilon}} \right) \Psi^+(x, t),\]  

(4)

was determined to be \(E^+ = -1.920\). Therefore, the ionization energy for the “outer” electron is \(E_0 - E^+ = -0.977\). The removal energy for the outer electron in real 3D \(\text{He}\) is 0.9 a.u.

The probability density \(|\Psi(x, y, t)|^2\) during the pulse for the peak field strength \(\hat{E} = 0.3\) is shown in Figure 3. One looks perpendicularly from above onto the illuminated, logarithmically scaled probability density.

Density flowed along the \(\pm x\)- or \(\pm y\)-axis corresponds to single ionization while double ionization happens when both \(|x|\) and \(|y|\) are significantly greater than the width of the ground state. We use this simple picture to define our ionization probabilities at the end of the laser pulse for total, double and single ionization \(P\), \(P^{++}\) and \(P^+\), respectively,

\[P = 1 - \int_{-a}^{a} \int_{-a}^{a} |\Psi(x, y, T)|^2 dx \, dy,\]  

(5)

\[P^{++} = 1 - \int_{-\infty}^{\infty} \int_{-a}^{a} |\Psi(x, y, T)|^2 dx \, dy - \int_{-a}^{a} \int_{-\infty}^{\infty} |\Psi(x, y, T)|^2 dx \, dy + \int_{-a}^{a} \int_{-a}^{a} |\Psi(x, y, T)|^2 dx \, dy,\]  

(6)
\[ P^+ = P - P^{++}. \]  

We chose \( a = 5 \). In Figure 3 the integration areas corresponding to \( P, P^{++} \) and \( P^+ \) are shown.

Of particular interest is the evolution of the probability density when the double ionization regions \( |x| > 5 \) and \( |y| > 5 \) are occupied for the first time. In Figures 3 and 4 these time intervals are shown for \( \hat{E} = 0.2 \) and 0.7, respectively. The delay between the snapshots is a quarter optical cycle in each case.

Let us first analyze the \( \hat{E} = 0.2 \)-case in Figure 3. Till \( n = 2.75 \) optical cycles mainly single ionization has occurred, i.e., the probability density is still located along the axes. At time \( n = 3.00 \) two density jets enter the region \( x, y > 0 \) (indicated by two arrows). They are clearly separated by the Coulomb repulsion ridge along \( y = x \). These density jets represent states where both electrons are on the same side of the nucleus. Therefore the Coulomb repulsion is relatively high and the jets tend to flow back towards the single ionization channels. This reflux of probability density is supported by the electric field which has its maximum a quarter of a cycle later, at \( n = 3.25 \). However, some density passes the single ionization channels and appears in the regions \( x > 0, y < 0 \) and \( x < 0, y > 0 \), respectively (see arrows in the \( n = 3.25 \)- and in the \( n = 3.5 \)-plot). Now, the two electrons represented by this density are on opposite sides of the nucleus, and both are ionized. So we conclude that although the electric field amplitude \( \hat{E} \) is not sufficiently strong to double ionize by simply tilting the \( xy \)-plane, Coulomb repulsion and electric field can lead to double ionization by acting together constructively. Therefore we propose Coulomb repulsion assisted laser acceleration to be responsible for NSI.

At higher intensities (\( \hat{E} = 0.7 \), Figure 4) we also observe the two density jets entering regions of the \( xy \)-plane where \( x \) and \( y \) have equal signs (see \( n = 2.00 \) till \( n = 2.25 \)). However, reflux of these jets is obviously not essential to stimulate subsequent emission of probability density into regions \( x > 0, y < 0 \) and \( x < 0, y > 0 \), respectively. This can be seen in plot \( n = 2.25 \) where already bursts of density leave the single ionization channels although the jets are not flown back yet. Here, double ionization is mainly due to the strongly tilted \( xy \)-plane.

Note that these backflowing jets which support double ionization are of course absent in any SAE model while they are expected to be included in TDDFT since there both electrons are allowed to respond to the field simultaneously. Furthermore, purely classical simulations should show a similar NSI scenario as the quantum density current in the \( xy \)-plane does, i.e., electrons which collide while moving in the same direction (the two jets) followed by a subsequent turn of one electron which crosses the origin and finally leaves in the opposite direction. All these presumptions will be confirmed in the next sections.

We performed several runs varying the peak fieldstrength \( \hat{E} \). In Figure 4 the ionization degrees \( P, P^+ \), and \( P^{++} \) are plotted vs. the intensity \( I = \hat{E}^2 \). There are also shown the ionization probabilities \( P_{\text{sequ}} \) and \( P \cdot P_{\text{sequ}} \) which results from solving (4) in the external laser field. As long as \( P^{++} \) is small \( P \cdot P_{\text{sequ}} \) would be the probability for He\(^{++} \) production if double ionization occurs purely sequential, i.e., the first electron does not interact with the residual ion anymore and the second electron remains non-excited after the emission of the first one. Obviously this completely sequential scenario cannot explain the correct ionization degree \( P^{++} \). Double ionization occurs earlier than predicted by \( P \cdot P_{\text{sequ}} \). This corresponds to the experimentally observed “knee” in the He ion yields, although our He\(^{++} \)-curve does not show such a pronounced shape due to the fact that we are in the multiphoton and not in the tunneling regime as in 8. We have also performed some runs using \( \omega = 0.2 \) and saw an increasing deviation from the SAE He\(^{++} \)curves, i.e., an increasing “knee”. The \( P_{\text{sequ}} \) and \( P \cdot P_{\text{sequ}} \) curves cross the correct \( P^{++} \) result since depletion of the He\(^+ \) ions is not taken into account.

IV. SINGLE ACTIVE ELECTRON ANALYSIS

In the previous section we have already compared the correct ionization degree \( P^{++} \) with the product \( P \cdot P_{\text{sequ}} \) for the rate when the second electron ionizes from its He\(^+ \) ground state without interacting with the electron already freed. The ionization degree \( P \cdot P_{\text{sequ}} \) was found to be too small in the intensity region of interest \( 0.02 < I < 0.07 \), as depicted in Figure 6.

Now, we may try to describe the ionization probability \( P^+ \) in the single active electron approach. In order to do this we have to find an appropriate \( Z_{\text{eff}} \) and to solve the equation

\[ i \frac{\partial}{\partial t} \Psi^0(x, t) = \left( -\frac{1}{2} \frac{\partial^2}{\partial x^2} - \frac{Z_{\text{eff}}}{\sqrt{x^2 + \epsilon}} \right) \Psi^0(x, t). \]  

The first electron has an ionization potential \( \mathcal{E}_0 - \mathcal{E}^+ = -0.977 \). We found that \( Z_{\text{eff}} = 1.117 \) yields such a binding energy. In Figure 7 the resulting ion yields \( P^+ \) are shown and compared with the exact result \( P^+ \). The ionization
degree calculated from (8) is too high over the whole region where mainly single ionization takes place. As soon as double ionization occurs the curve differs also qualitatively from the exact one: there is a dip in the exact $P$ and $P^+$ curves around $I = 0.03$ which is absent in the $\tilde{P}^+$ result. We conclude that the second electron shares some energy with the escaping first electron which leads to a decreased single ionization probability. Since in a similar work for a very low frequency [8] the SAE curves are found to fit well, the deviations we observe might be mainly due to the relatively high frequency we have chosen. For higher frequency laser fields the “cracking up” of the initial ground state wave function into an “inner” and an “outer” electron may be too slow to be well described by an SAE ansatz for the “outer” electron.

One may object that although tuning $Z_{\text{eff}}$ in order to fit the binding energy of the “outer” electron leads to an over-estimation of the ion yields, there might be a certain combination of $Z_{\text{eff}}$ and an effective soft-Coulomb-parameter $\epsilon_{\text{eff}}$ which does both: providing the correct binding energy and the right ionization probabilities. However, we tried $Z_{\text{eff}} = 1$ and $\epsilon_{\text{eff}} = 0.398$ which leads also to the desired binding energy $-0.977$. The resulting curve for the single ionization yields are also shown in Figure 8. It overestimates the single ionization too.

V. DENSITY FUNCTIONAL THEORY

Density functional theory (DFT) is a powerful tool in determining multielectron atomic structures (see e.g. [16,30] for an overview). It has been shown [31] that a Hohenberg-Kohn-type theorem exists also for time-dependent phenomena. Therefore the existence of an effective potential which transforms the problem of $N$ interacting electrons to that of $N$ non-interacting ones is proved. The non-interacting electrons move in an effective potential which is a functional of the total electron density only. The problem reduces to finding an appropriate effective potential which includes all relevant exchange and correlation effects.

There is some recent work on the field of time-dependent density functional theory (TDDFT) applied to laser ionization of atoms [16–19].

In the case of a singlet two-electron-system like our model atom of helium, there is only one occupied Kohn-Sham-orbital $\varphi(x,t)$. The total electron density is

$$n(x,t) = 2|\varphi(x,t)|^2. \tag{9}$$

There are no exchange contributions, and neglecting correlation effects leads to the time-dependent Hartree equation

$$i \frac{\partial}{\partial t} \varphi(x,t) = \left( -\frac{1}{2} \frac{\partial^2}{\partial x^2} - \frac{2}{\sqrt{x^2 + \epsilon}} + \int \frac{|\varphi(x',t)|^2}{\sqrt{(x-x')^2 + \epsilon}} \, dx' + xE(t) \right) \varphi(x,t). \tag{10}$$

Hartree’s independent electron-model was used by Geltman already in 1985 to analyze experimental results in the multiple ionization of atoms [14–19].

The ground state energy we obtained by solving (10) in imaginary time is $E_{\text{KS}}^0 = -2.878$.

In Figure 8 the comparison between the TDDFT-results and the exact ones are made. The ionization yields for He$^+$ and He$^{++}$ are observables, of course. Thus they are functionals of the density $n(x,t)$ and, due to the simple relation (6), explicit functionals of the Kohn-Sham-orbital density $|\varphi(x,t)|^2$. We want do adopt the simple “integration-over-areas” picture in order to calculate the ionization, as described in Section II. According to Ullrich et. al. [18] we proceed, for the time being, as follows: with

$$P_{\text{KS}} := 1 - \int_{-a}^{a} |\varphi(x,t)|^2 \, dx \tag{11}$$

the probability for neutral helium is the product of the probabilities for each orbital to be non-ionized. Thus

$$P_{\text{KS}}^0 = (1 - P_{\text{KS}})^2. \tag{12}$$

For the single and double ionization

$$P_{\text{KS}}^+ = 2P_{\text{KS}}(1 - P_{\text{KS}}), \tag{13}$$

$$P_{\text{KS}}^{++} = P_{\text{KS}}^2. \tag{14}$$
follows and the total ionization clearly is

$$P_{\text{KS}}^{\text{tot}} = 1 - P_{\text{KS}}^0.$$  \hfill (15)

The three curves corresponding to $P_{\text{KS}}^{\text{tot}}$, $P_{\text{KS}}^+$ and $P_{\text{KS}}^{++}$ are shown in Figure 3. The agreement is quite bad. The total and single ionization degree are overestimated as in the SAE calculation.

However, the total ionization $P_{\text{KS}}$ fits well the exact $P$ if one avoids assigning any physical relevance to the Kohn-Sham-orbital $\varphi(x,t)$ and proceeds instead as follows: $P_{\text{KS}}$ as defined in (11) is the probability to find any of the two electrons outside the interval $[a, a]$ since the physical total electron density is $n(x,t) = 2|\varphi(x,t)|^2$. Therefore the probability for total ionization should be simply

$$P_{\text{KS}}^{\text{tot}} = P_{\text{KS}},$$  \hfill (16)

and

$$P_{\text{KS}}^0 = 1 - P_{\text{KS}}.$$  \hfill (17)

$P_{\text{KS}}^{\text{tot}}$ according (16) is also depicted in Figure 3. The agreement between $P_{\text{KS}}$ and $P$ is excellent. The dip around $I = 0.03$ is well reproduced. Since the dip is absent in the SAE results, the onset of NSI seems to be included in the TDDFT although only the simple Hartree effective potential is taken.

There is no simple way to deduce $P_{\text{KS}}^+$ and $P_{\text{KS}}^{++}$ without claiming physical significance of the Kohn-Sham-orbital $\varphi(x,t)$ although the existence of purely density dependent functionals $P_{\text{KS}}^+[n]$ and $P_{\text{KS}}^{++}[n]$ are proved by the Hohenberg-Kohn-theorem. Eqs. (12–13) would be valid if the correct wave function $\Psi(x,y,t)$ was the product of the Kohn-Sham-orbitals, $\varphi(x,t)\varphi(y,t)$. A plot of $|\varphi(x,T)|^2|\varphi(y,T)|^2$ for $\hat{E} = 0.3$ is shown in Figure 14. Clearly, there is a gridlike pattern imprinted due to the construction of the wavefunction as a pure product. This leads to a totally different angular distribution in the $xy$-plane. A similar behaviour is observed in time-dependent unrestricted Hartree-Fock calculations [15].

However, for higher field strengths both electrons are ionized rapidly and subsequently behave as free and almost independent electrons. Thus the total wave function should develop a grid-like pattern. This is shown in Figure 11 where the peak field strength $\hat{E} = 0.7$ was chosen. The TDDFT result is also plotted.

### VI. CLASSICAL SIMULATIONS

We solved the classical equations of motion according the Hamiltonian (1) and the electric field (3) for a microcanonical ensemble of the two electrons. We traced the one-particle-energies

$$E_x(x,y) = \frac{1}{2} \dot{x}^2 - \frac{2}{\sqrt{x^2 + \epsilon}} + \frac{1}{\sqrt{(x-y)^2 + \epsilon}}.$$  \hfill (17)

$$E_y(x,y) = \frac{1}{2} \dot{y}^2 - \frac{2}{\sqrt{y^2 + \epsilon}} + \frac{1}{\sqrt{(x-y)^2 + \epsilon}}.$$  \hfill (18)

The total energy is

$$E(x,y,t) = E_x(x,y) + E_y(x,y) - \frac{1}{\sqrt{(x-y)^2 + \epsilon}} + (x+y)E(t).$$

Each electron is considered to be ionized when $E_x(x,y) > 0$ or $E_y(x,y) > 0$, respectively. There is no unique way in defining the single particle energies (17,18) since the electron-electron term may be shared between the two electrons in various ways (24). However, this has little influence on the ionization degrees since in the case of single or double ionization the distance between the two electrons is normally large at the end of the pulse.

The initial conditions were chosen to meet the quantum mechanical ground state energy $E_0 = -2.897$. Fortunately, the resulting ion yields were not sensitive to the choice of the ensemble of initial conditions. Instead of taking several initial positions and momenta we started with one "mother"-configuration at $t = 0$ and vary the time $t_{\text{on}}$ where the laser pulse sets in. We tried several mother-configurations to ensure insensibility of the resulting ion yields.

We would like to mention that a classical treatment of an 1D model helium has also been undertaken in [34] in the framework of stabilization of multielectron atoms.
The results are shown in Figure 12. The single ionization is strongly overestimated. This is due to the fact that the ionizing electron gains energy at the expense of the still bound electron which occupies a state of quantum mechanically forbidden low energy, i.e. an energy below $-1.920$. This is shown in Figure 13 where the two one-particle-energies $E_x$ and $E_y$ are plotted for a representative single ionization event at $E = 0.1$.

This behavior could be prevented if one introduces a velocity dependent “Heisenberg”-potential [20]. However, for our purpose to study the NSI-mechanism this is not necessary. We have calculated also the classical SAE single ionization process in the potential $V(x) = -Z_{\text{eff}}/\sqrt{x^2 + \epsilon}$ with $Z_{\text{eff}} = 1.117$ as in Section IV. The resulting yields are lower and show a rapid increase at the critical intensity which is 0.05 for the potential used. This is the typical behavior in pure classical simulations. Note that double ionization is already observed where according the SAE approach even no classical single ionization occurs.

The result of the SAE calculation $\text{He}^+ \rightarrow \text{He}^{++}$ was multiplied with the probability for $\text{He}^+$-production from the full 2D run. Below $I = 0.25$, the classical critical intensity for sequential $\text{He}^{++}$-production the probability vanishes, as expected. Thus the intensity region $0.03 < I < 0.2$ is the classical NSI regime we are particularly interested in. We examined each double ionization event in that region. The ionization dynamics of two representative examples in terms of single-particle-energies and trajectories is depicted in Figures 14 and 15.

For the purpose of comparing our classical results with the quantum mechanical probability density current in the $xy$-plane we look at the electron trajectories in Figure 15. One observes that the electron which leaves first is accompanied by the other electron moving in the same direction (corresponding to density flowing into regions of the $xy$-plane where $x$ and $y$ have equal sign). The Coulomb interaction is strong within that half cycle. Then, as the first electron ionizes, the second one turns around and moves in the opposite direction (corresponding to probability density passing one of the $xy$-plane’s axes). Thus, the second electron in NSI leaves the atom approximately half laser cycle after the first one. The dynamics of the classical test particles corresponds to the temporal and spatial evolution of the quantum probability density as described in Section II.

In Figure 14 we show a representative example for double ionization at a higher field strength ($\hat{E} = 1.0$). At this field strength the sequential pathway is more probable than NSI. The temporal delay between the ionization of the two electrons is greater (1–3 cycles) but still small since our pulse is ramped strongly over 3 cycles only. In longer pulses the temporal delay between the ejection of the two electrons would be even greater.

VII. CONCLUSIONS

In this paper we have confirmed the recently proposed mechanism for the NSI process, namely the ionization of the second electron by Coulomb-interacting with the outer partner. We have traced the NSI scenario by observing the quantum mechanical probability density as it evolves in space and time. We have identified the Coulomb repulsion assisted laser acceleration of the inner electron to be responsible for NSI. Our classical simulations have supported this point of view and contributed a detailed picture of how NSI happens in terms of one particle energies and trajectories. Moreover, we have shown that “non-sequential” means “within half an optical cycle” and that NSI is not an essentially quantum mechanical effect. We showed that in the frequency and pulse duration regime under consideration, SAE ionization yields are in poor agreement with the exact results. TDDFT reproduces the total ionization probability very well, even for the simplest effective potential available, namely the Hartree-potential. TDDFT fails if one claims physical relevance for the Kohn-Sham-orbitals by separately constructing single and double ionization yields. However, since generally only the knowledge of the total electron density is necessary, e.g. for the calculation of high harmonics generation, TDDFT should produce very accurate results [17].

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The 2D potential \( V(x, y) \) for \( \hat{E} = 0.0 \) (a), \( \hat{E} = 0.1 \) (b) and \( \hat{E} = 0.616 \) (c). At \( \hat{E} = 0.1 \) the initial ground state level cuts the effective potential within the single ionization channels. For \( \hat{E} = 0.616 \) the ground state level even exceeds the potential ridge along \( x = y \). At latest at that field strength strong double ionization is expected.

Contour plot of the ground state probability density. The energy is \( \mathcal{E}_0 = -2.897 \). The electron-electron repulsion along \( x = y \) clearly leaves its fingerprint on the wave function (butterfly-shape). Such an asymmetric shape is absent in corresponding Hartree-Fock \( ^{[14]} \) or simple DFT ground states.
The probability density after \( n \) optical cycles for \( \hat{E} = 0.3 \). One looks perpendicularly from above onto the illuminated \( xy \)-plane and the logarithmically scaled probability density \( |\Psi(x, y, t)|^2 \). Till \( n = 2 \) mainly single ionization takes place (probability density along the axes). Afterwards also regions \( |x|, |y| > 5 \) are occupied which corresponds to double ionization. The Coulomb-repulsion ridge along \( y = x \) can be clearly identified at later times.

FIG. 4.
The areas to be integrated over in order to calculate the probabilities for (from left to right) total, double and single ionization are shaded. The parameter \( a \) was chosen to be 5 a.u.

FIG. 5.
The probability density for \( \hat{E} = 0.2 \) at \( n = 2.5, 2.75, 3, 3.25, 3.5, 3.75 \) optical cycles. At time \( n = 3 \) two probability density jets are emitted into the region \( x, y > 0 \). At \( n = 3.25 \) they are partly flown back and density appears in regions \( x < 0, y > 0 \) and \( y < 0, x > 0 \).

FIG. 6.
The probability density for \( \hat{E} = 0.7 \) at \( n = 1.5, 1.75, 2, 2.25, 2.5 \) optical cycles. Double ionization is mainly due to the strongly tilted \( xy \)-plane.

FIG. 7.
The 2D calculation results for total, single and double ionization (bold, +). The SAE result for the \( \text{He}^+ \rightarrow \text{He}^{++} \) is also drawn (\( \bigcirc \)). Multiplication with \( P \) (see text) leads to the expected sequential double ionization degree (\( \ast \)) as long as \( P^{++} \ll P^+ \). The \( P_{\text{seq}} \) and \( P \cdot P_{\text{seq}} \) curves intersect the correct \( P^{++} \) result since depletion of the \( \text{He}^+ \) ions is not taken into account.

FIG. 8.
SAE results for the outer electron compared with the exact 2D yields (bold, +). The curve plotted with connected \( \ast \) was calculated using \( Z_{\text{eff}} = 1.117 \) and \( \epsilon_{\text{eff}} = \epsilon = 0.55 \). For the \( \bigcirc \)-curve \( Z_{\text{eff}} = 1 \), \( \epsilon_{\text{eff}} = 0.398 \) was chosen. In both cases the SAE ionization degrees are too high.

FIG. 9.
Comparison of the TDDFT results with the exact 2D solutions. The total ionization degree \( P_{\text{KS}} \) (\( \bigtriangleup \)) matches nicely the exact curve (bold, +). If one claims physical relevance of the Kohn-Sham orbitals one gets the \( \bigcirc \)-curves for \( P_{\text{KS}}^{\text{tot}} \) (total), \( P_{\text{KS}}^+ \) (single) and \( P_{\text{KS}}^{++} \) (double) ionization which agree poorly with the exact probabilities (see text for a discussion).

FIG. 10.
The TDDFT probability density at the end of the pulse for \( \hat{E} = 0.3 \). The gridlike pattern is due to the construction as a pure product of the Kohn-Sham orbitals. There is no displacement of probability density along \( y = x \), of course.

FIG. 11.
The probability densities at the end of the laser pulse for \( \hat{E} = 0.7 \). In plot (a) the exact density, in (b) the TDDFT result is presented. The exact density (a) develops a gridlike pattern since both electrons were ionized rapidly and subsequently behaved like almost free electrons, i.e., the wave function becomes more and more a pure product of single particle wave functions. However, the agreement with the product of the Kohn-Sham-orbitals (b) is poor even at those high fieldstrengths.

FIG. 12.
The classical yields (bold, ∗) for \( P_{\text{cl}} \) (total), \( P_{\text{cl}}^+ \) (single) and \( P_{\text{cl}}^{++} \) (double) ionization. The single ionization is strongly overestimated (compare with the exact quantum mechanical results, drawn dashed) due to the classical effect discussed in the text. The classical SAE results for the outer \( (\tilde{P}_{\text{cl}}^+, \Box) \) and the inner electron \( (\tilde{P}_{\text{cl}}^{++}, \triangle) \) are also plotted. There is classical NSI at \( I = 0.04 \) even when no sequential single ionization should occur!

**FIG. 13.**

A representative example for classical single ionization in terms of the single particle energies (17, 18). The inner electron (solid) drops below the quantum mechanical He\(^+\) binding energy \(-1.920\). This leads to an overestimation of the single ionization probability since the outer electron (dashed) can gain more energy during Coulomb collisions (see peaks in the energy-curves) with the inner electron as allowed quantum mechanically.

**FIG. 14.**

Two representative classical NSI scenarios in terms of the single particle energies (17, 18).

**FIG. 15.**

The particle trajectories corresponding to Figure 14. The electrons become free within approximately \( \frac{1}{2} \) an optical cycle. Before both electrons ionize they move together in the same direction for a quarter of a cycle until one electron turns and finally vanishes in the opposite direction. This has to be compared with the quantum dynamics in Figure 5.

**FIG. 16.**

A representative example of classical sequential double ionization at \( \hat{E} = 1.0 \). The temporal delay between the ejection of the two electrons is 3 half cycles and would be even greater in a more adiabatically ramped pulse.
Figure 1 ("A two-dimensional, two-electron ..." by D. Bauer)
Figure 2 (“A two-dimensional, two-electron...” by D. Bauer)
Figure 3 ("A two-dimensional, two-electron ..." by D. Bauer)
Figure 4 ("A two-dimensional, two-electron ..." by D. Bauer)
Figure 5 ("A two-dimensional, two-electron ..." by D. Bauer)
Figure 6 ("A two-dimensional, two-electron ..." by D. Bauer)
Figure 7 ("A two-dimensional, two-electron ..." by D. Bauer)
Figure 8 ("A two-dimensional, two-electron ..." by D. Bauer)
Figure 9 ("A two-dimensional, two-electron ..." by D. Bauer)
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Figure 11 ("A two-dimensional, two-electron ..." by D. Bauer)
Figure 12 ("A two-dimensional, two-electron ..." by D. Bauer)
Figure 13 ("A two-dimensional, two-electron ..." by D. Bauer)
Figure 14 ("A two-dimensional, two-electron ..." by D. Bauer)
Figure 15 ("A two-dimensional, two-electron ..." by D. Bauer)
Figure 16 ("A two-dimensional, two-electron ..." by D. Bauer)