Stabilization phenomenon revisited in attosecond regime: Applicability of a regularized potential
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A B S T R A C T
We present the results of numerical simulations performed for a realistic model of the hydrogen atom as well as for a model with a smoothed core, both in the presence of the strong linearly polarized laser field in the attosecond regime. These results generally confirm the existence of stabilization in 3D ionization, although there are some significant quantitative discrepancies in comparison to earlier reports, which may be important in the context of possible future experiments. We have also checked the applicability of the smoothed model potential and the rectangular pulse shape for the photoionization process in the strong field and ultra-short time regime. Both simplifications strongly influence the numerical effort necessary in \textit{ab initio} simulations and thus its duration. In contrast to the 1D case, in which the results obtained using the potential with a smoothed core are essentially different from those for a singular potential, in the 3D case removing the singularity does not lead to such a strong discrepancy: the stabilization is visible though the depth and range of the stabilization window are different. Thus it may serve as a useful model of some aspects of the real hydrogen atom, which allows one to significantly shorten 3D computations.

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1. Introduction

Due to a great progress in generation of short and strong laser pulses [1], investigations of the dynamics of atomic systems in ultra-strong fields are of a vivid interest. In particular it is important to discover the process’ dependence on various parameters and the possibilities to control it. One of such important effects is stabilization against photoionization [2–4], i.e. a non-monotonic behavior of the ionization probability as a function of the laser intensity. This effect has been discussed and numerically simulated since late 80s of the past century [2,5] but our knowledge of its details and the conditions in which it occurs is still not complete, especially in the context of very short pulses, which are available since recently [1,6]. Also the experimental data are limited to those obtained in mid of the 90s by one group from Amsterdam [7]. Moreover, in these experiments in order to lower the ionization energy the atom initial state was not the ground state. The new kind of lasers developed nowadays, namely attosecond lasers [6] and free-electron lasers, allows one to have hope that new attempts to experimental examination of the stabilization will be make soon [8]. The attosecond lasers produce so-called attosecond pulse train [6]. In this context the series of papers reporting studies of stabilization phenomena in such periodically kicked systems should be noted [9].

The adiabatic stabilization, which is a particular subject of our interests, is usually described in the language of the Kramers–Henneberger model [10], which quantitatively describes this phenomenon as the atom population trapping in the potential created by averaging over time the rapidly oscillating binding potential seen from the electron’s reference frame (Kramers frame). Another, but closely related model is high-frequency Floquet theory developed by Gavrila [3]. The natural assumption in both cases is high laser frequency corresponding to extreme ultra-violet or soft X-ray. However, lately Gavrila generalized its model to lower frequencies provided that laser intensity is high enough. Similar effort was made also by Miyagi and Someda [11].

In 80s and early 90s only one-dimensional simulations were possible for a long-range potential with a smoothed core [12], which are much less numerically demanding compared to the case of a potential containing singularity. Those simulations allowed one to study ionization, including the stabilization phenomenon, but in rather artificial conditions. The ionization in one dimension must differ from that in two and three dimensions: the electron freed from the atom and oscillating in the rhythm of the external electric field cannot pass the nucleus without a disturbance; its backscattering is unavoidable. This discrepancy has been first observed in classical simulations performed by Grochmalicki et al. [13] and Gajda et al. [14] using...
the phase-space averaging method, who have shown that the stabilization in 1D may be described in classical terms and that this phenomenon is significantly reduced when the number of dimensions increases; lack of stabilization in 3D was observed, at least for the laser frequency equal to unity [13,14]. Moreover, in one dimension the potential with a singularity does not allow the electron to pass the nucleus at all. Thus, because the influence of the binding potential, necessary to prevent the wavepacket from spreading out, is stronger in one dimension, it is in this case that the stabilization was considered to be better pronounced than in two and three dimensions. In order to confirm this conviction, two- and three-dimensional quantum-mechanical simulations began to be performed in the turn of the centuries [15–17], also for a real hydrogen atom. We want to refer particularly to the calculations of that kind performed by Kundliya and Mohan [17]. In fact we have performed similar simulations for analogous laser pulse shapes and strengths using a different numerical method and limiting ourselves to the field intensity range in which the nonrelativistic approach makes sense and the dipole approximation holds (cf. [18]). Their general conclusion agrees with ours and is against the predictions obtained by classical simulations: there is stabilization in three-dimensional hydrogen atom. However, some important details and quantitative results of the two papers are different. These differences may be important in the context of experiment, which could possibly be made for super-strong laser pulse in the attosecond regime.

But our aim is not only to verify the earlier results, but also mainly to check whether the smoothed model potential, similar to those widely used at the beginning of one-dimensional computer investigations of strong-field phenomena, may be useful in three dimensions. A smoothed model potential may be used to significantly decrease the number of spatial nodes of the grid used in the calculations and therefore to shorten them. Lately we have revised the usefulness of such model potentials in 1D and clarified the role of the singularity [19]. We have also studied its influence on recombination in one, two and three dimensions [20].

The additional motivation for such tests comes from a recently published paper of Kaestner et al. [21], which concerns using softened potentials for $H^*_2$ molecules. They investigate the photoionization probabilities and HHG (high-order harmonic generation) spectra, but not the stabilization, in ultrastropical fields, and they find essential discrepancies between the results for soft-core one-dimensional models and those for true three-dimensional ones. A similar problem has also recently been addressed by Gordon et al. [22], who investigated the influence of the singularity on HHG spectra. Their results suggest that the presence of the potential singularity is even more important than the difference of the dimensionality of the system, which is against the intuition, at least in the context of the stabilization phenomenon. Thus our aim was also to check the importance of the dimensionality on the stabilization phenomenon.

2. Numerical method

Fully three-dimensional numerical simulations of an atomic system interacting with an arbitrarily polarized laser field are still a hard task, even for current computers, especially if the atom is described by the Coulomb potential with the singularity, which requires a dense spatial grid to represent it numerically, and the evolution in any dimension may not be reduced. However, if the laser is linearly polarized one can perform the calculations using a two-dimensional spatial grid. The details of the algorithm have been described in our paper concerning the recombination process [20]. Here we shortly recapitulate its essential points. We describe the atomic system interacting with a strong laser field using the hamiltonian in the length gauge (atomic units, abbreviated as a.u., are used throughout this paper):

$$\hat{H} = -\frac{1}{2} \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \right) + V(x,y,z) + z \cdot \hat{a}(t).$$

(1)

The potential is given by following formula:

$$V(x,y,z) = \frac{1}{\sqrt{x^2+y^2+z^2}}.$$ \hspace{1cm} (2)

The parameter $a$ denotes the width of the potential core. For $a=0$ a.u. this is the standard singular hydrogen atom potential and for $a=1$ a.u.—a model atom with a smoothed core, analogous to the one-dimensional potential introduced by Su and Eberly in the early 80s of past century [12].

The laser electric field $\hat{a}(t) = \hat{a}_0(t) \hat{\Theta}(t) \hat{\Theta}(t_f - t) \cos(\omega t - \pi)$ is directed along the $z$-axis and due to the dipole approximation does not depend on the spatial coordinates. The symbol $\hat{\Theta}$ denotes here the Heaviside function. The laser is switched on at $t=0$ and switched off after the time $t_p$. The laser field frequency $\omega$ has been always taken equal to $\omega = 1$ a.u. (thus the optical period is equal to $T = 2\pi$ a.u. = 152 as). We have performed the simulations for the laser field amplitude $\hat{a}_0$ in the range between 1 and 5 a.u.

We have examined two kinds of laser pulses: the rectangular one, for which $f(t) = 1$, lasts maximally $t_f = 16T$ and the smooth one, for which

$$f(t) = \begin{cases} \sin^2(\omega t/12), & 0 < t < 3T \\ 1, & 3T < t < 11T \\ \sin^2(\omega(t_f-t)/12), & 11T < t < 14T \end{cases}$$

(3)

lasts for $t_f = 14T$ and was switched-on and switched-off during the three optical cycles.

The spherical symmetry of the potential together with the assumption of the linear polarization of the external field leads to the axial symmetry of the whole system, which allows one to separate the angular function $\exp(i\rho \phi)$ (the cylindrical coordinates $\{s,\rho,\phi\}$ are used). This reduces by one the dimensionality of calculations without any approximation, which makes the simulations of 3D systems much easier. A similar reduction was made for example by Kulander [23]. Unfortunately the price for the dimensionality reduction is a new form of the Laplace operator (in the cylindrical coordinates) containing a singular term in the variable $\phi$. One can remove this term by a substitution for the part of the wavefunction dependent on $s$ and $\phi$: $\Psi = \Psi' / \sqrt{s}$. This transformation causes the that the boundary condition for $\Psi'$ at $s=0$ is such that the wavefunction $\Psi$, and not its derivative, should vanish. Moreover, for the transformed function the Jacobian becomes equal to 1, which means that three-dimensional simulations may be performed using the code prepared for 2D calculation in Cartesian coordinates. However, such transformation introduce an additional potential-like term $-1/(8s^2)$ to the hamiltonian, which may be incorporated to the sort of an effective potential (see [24] for its more extensive discussion). Finally we solve numerically the following Schrödinger equation using the two-dimensional grid:

$$i \frac{\partial}{\partial t} \Psi(s,\phi, t) = -\frac{1}{2} \left( \frac{\partial^2}{\partial s^2} + \frac{\rho^2}{s^2} \right) \Psi(s,\phi, t)$$

$$+ \left[ -\frac{1}{\sqrt{\rho^2 + s^2 + z^2}} + \frac{1}{2} \frac{m^2 - 1/4}{s^2} + z \cdot \hat{a}(t) \right] \Psi(s,\phi, t).$$

(4)

The initial state of the system is its ground state. The quantum number $m$, which is connected with the rotation around the $z$-axis, is conserved during the whole process; thus the simulations have to be performed separately for each value of $m$. For the
ground state $m = 0$, therefore we have limited our simulation only to this value of $m$.

To calculate the populations of the bound states it is also necessary to obtain the exact wavefunctions of those states for both Coulomb and smoothed potentials. The former ones may be calculated analytically. The latter ones we have obtained numerically using our implementation of the Davidson method [25] with parameters listed in [20]. However, the Davidson method failed if we tried to calculate the eigenstates of the Kramers–Henneberger (KH) well (i.e. the binding potential moving in the electron’s reference frame averaged over one optical cycle). In that case we used the One Electron Diatomic states method described in [26], which was designed to solve the eigenproblems for double-centered potentials.

The dynamics of the ionization has been examined by a numerical integration of the time-dependent Schrödinger equation (4). As described earlier, the 3D simulations (in the dipole approximation) require 2D calculations which can be efficiently performed using the alternate-direction implicit (ADI) algorithm [27]. Because of the singular term in the pseudo-potential numerical requirements of the calculations do not depend so crucially on whether the singularity is also present in the binding potential (cf. Section 3.4 for its discussion). This is why we used the same space grid step for both potentials: In the case of the soft-core potential we used the spatial grid with $2048 \times 4096$ nodes covering the range of $[0,200]$ in $s$ and of $[-200:200]$ in $z$ direction, while for the Coulomb potential both the number of nodes and the space range were twice as large. The time step was equal to $1/1000$.

It should be also noted, that other approaches to 3D simulations of atoms interacting with strong laser fields were developed. An example is the algorithm of Geltman [28], extensively used by Hansen et al. [29]. It is the close-coupling algorithm, in which the dynamics of the wavefunction is calculated using the basis of eigenstates of the binding potential. The usefulness of this method is thus limited to systems in which both the bound states and the continuum states are explicitly known. For other algorithms see [30] and [31] and the references therein.

3. Results

3.1. Stabilization for singular and regularized 1D potentials

It is known that the influence of the potential singularity on the stabilization phenomenon in photoionization by an XUV attosecond laser pulse is essential in 1D (see [19] for our report on that subject). It is clearly visible not only in the dependence of the total population of the bound states on the laser field intensity (Fig. 1), but also in the evolution of the wavefunction discussed in more detail in [19]. Fig. 1 shows the results of 1D numerical simulations for the Coulomb potential (upper plot) and the soft-core potential (lower plot) for the smooth pulse envelope given by (3). The nonmonotonicity of the final total population of the bound states (survival probability), i.e. the stabilization phenomenon, is not visible in the former case. In fact for laser intensities larger than 0.5 a.u. after the end of pulse the system are fully ionized due to the packet tearing apart. In contrast to that, the same quantity calculated for the soft-core potential exhibits a well visible stabilization window, i.e. the range of laser intensities $\epsilon_0$, for which the bound states’ final total population is a growing function of $\epsilon_0$. In this case the packet conserves its integrity and its shape corresponds to the eigenstate of the KH well. In the case of the rectangular pulse (not shown), the stabilization window is well developed already after 3T in the case of soft-core potential and the atom is fully ionized (for $\epsilon_0 > 0.5$) in the case of the potential with the singularity.

3.2. Stabilization in 3D

In the 3D case the general observation is that the potential shape looses its importance, which can be attributed to the simple fact, that the potential singularity can be more easily passed by, thus its presence is not so important. Instead, the shape of the pulse envelope becomes crucial. Figs. 2 and 3 show the final total population of 15 bound states with lowest energies (the convergence was carefully checked, see also Ref. [20] for details) in the presence of the Coulomb potential (upper plots) and soft-core
the bound states for the smooth pulse envelope occurs for the shape changes while it fits to the KH potential). Which the wavepacket remains in principle undivided (of course level is much larger than in the case of the soft-core potential, for into small parts leaving the vicinity of the nucleus, the ionization similarities for the model atom and for the real hydrogen atom. The shape of the plots obtained for both examined pulse shapes, for both pulse shapes is larger than for the soft-core model atom. The appearance and the extent of the stabilization, is similar for the model atom and for the real hydrogen atom. However, the level of ionization depends rather on the potential shape than on the shape of the pulse. For the Coulomb potential, for which the singularity more efficiently tears the wavepacket into small parts leaving the vicinity of the nucleus, the ionization level is much larger than in the case of the soft-core potential, for which the wavepacket remains in principle undivided (of course its shape changes while it fits to the KH potential).

Potential (lower plots) for the ionization in 3D by a laser with a rectangular and a smooth pulse envelope. The general picture is that the stabilization is visible for longer rectangular pulses (t ≥ 8τ), but is more prominent in the case of a smooth pulse envelope. In the latter case, for the Coulomb potential the stabilization is well visible already after three optical cycles. The influence of the pulse shape seems to be stronger than that of the singularity in the potential, although it must be noted that the ionization efficiency in the case of the Coulomb potential for both pulse shapes is larger than for the soft-core model atom. The shape of the plots obtained for both examined pulse shapes, and thus the appearance and the extent of the stabilization, is similar for the model atom and for the real hydrogen atom. However, the level of ionization depends rather on the potential shape than on the shape of the pulse. For the Coulomb potential, for which the singularity more efficiently tears the wavepacket into small parts leaving the vicinity of the nucleus, the ionization level is much larger than in the case of the soft-core potential, for which the wavepacket remains in principle undivided (of course its shape changes while it fits to the KH potential).

For both potentials the minimum of the total population of the bound states for the smooth pulse envelope occurs for the intensity ε₀ equal to 1–1.5 a.u. (our precision is 0.5 a.u.) and the stabilization window appears for intensities up to 3 a.u. In the case of the Coulomb singular potential the upper limit of the stabilization window increases with time.

The upper plot from Fig. 3 corresponds to Fig. 2 from the paper [17]. However, we have limited ourselves to the intensities for which nonrelativistic approach is applicable and the dipole approximation holds. One can see the nonmonotonicity of the total population of the bound states of the hydrogen atom, but in our case the stabilization window is located rather for intensities between 1 and 5 a.u. while in Ref. [17] it occurs for higher intensities and it is not as deep as ours. The discrepancy might be due to the phases of the pulses in the two works, which differ by π/2. Note, that both these results disagree with those obtained in the classical calculations [13,14], which predicted no stabilization

An effect often observed in strong-field ionization is the so-called slow drift [32,33], which consists in oscillations of the wavepacket much slower than the optical oscillations and due to the interaction of an almost free electron with the nucleus. However, here this effect is not visible in the survival probabilities during the ionization process, at least for laser intensities larger than ε₀ = 1. This is in contrast to the case of recombination [20]. Therefore the results, especially the total bound states populations, are not sensitive to the exact time of calculating the projection onto the bound states—the dependence of the total bound states populations after full cycles on the laser intensity (main plots of Figs. 2 and 3) are very similar to those calculated at the times at which the total population reaches its maximum, that is at the moment at which the wavepacket turns in its oscillatory motion (the insets in the Figs. 2 and 3, cf. [20]). The exceptions are the bound states’ total populations in the case of both types of potentials, calculated for a pulse initially smooth but not realistically abruptly cut. One can observe the absence of the slow drift directly at the plots of the wavefunction (not shown)—the maximum of the electron density is located near its initial position after an entire number of cycles. Thus it should not be a surprise, that also the plots of the total population of the KH wells as functions of the laser field amplitude are very similar to those for true bound states (not shown). In the case of a smooth pulse and regularized potential the slow drift can be noticed when one observes the maxima of the probability density, but its effect on the bound states’ final population is small.

For small field intensities (ε₀ ≤ 1) the slow drift is relatively better pronounced. It causes that the maximum occupation of the bound states does not occur exactly after an entire number of full optical cycles, when the packet still has a nonnegligible velocity. This explains why value of the total population of the bound states after the full optical cycles (see main plots of Figs. 2 and 3) does not tend to unity while the laser intensity goes to zero. The insets of Figs. 2 and 3 show maximal values of total bound states population are much closer to unity, although still lower. This is because the maximal value of various bound states populations do not occur simultaneously.

3.4. Usefulness of the soft-core model potential in 3D calculations

The fully 3D simulations, performed for example for an arbitrary laser polarization or a more complicated potential, are still the challenge taking into account the current capabilities of modern computers. Thus any possibilities to make them faster are welcome. We have in particular decided to check whether the model atom potential with a smoothed core, similar to that proposed for 1D calculation by Eberly and Su [12], given by Eq. (2) with a = 1, may be useful in simulations. While we have been mainly concerned with the cases of a = 0 and a = 1 we have checked that all our results change smoothly with the value of a (no phase transition symptoms are noticeable).

Using a regularized potential instead of the singular one in fully 3D calculations would allow one to significantly decrease the number of grid nodes (by at least an order of magnitude in each dimension) and therefore to speed up such calculations. Note however that, this is not the case of our particular calculation, in which the reduction of the dimensionality introduces an additional singular term independently on the potential. Nevertheless, it could be crucial for problems in which no reduction of the dimensionality of any kind may be made, for example if the laser field is not linearly polarized. The only condition is that the results obtained using the model potential should well imitate the
results obtained using the Coulomb potential, which can be already checked in our simulations.

One can thus draw the following conclusion: it is possible to use the soft-core model to obtain qualitative results (for example the presence of the stabilization), but some properties of the system (e.g. the detailed level of the survival probability) are dependent on the potential details.

3.5. Comparison of ionization and recombination

We have recently checked that the counterpart of the stabilization phenomenon in the recombination process occurs for soft- and hard-core hydrogen atom models in 1D, 2D and 3D [20]. It is worth to compare the results of the simulations of ionization presented in this paper and those obtained earlier for recombination in 1D and 3D. Let us note than in 1D recombination the barrier created by the singular core of the potential, impenetrable for the wave packet, is less important because the wavepacket remains mainly at this side of the nucleus at which it has been located at the beginning of the recombination process; the slow drift may additionally push the oscillating wavepacket toward the barrier or to temporarily pull it away. On the contrary, in 1D ionization [19], in which the initial wavepacket represents the ground state of the atom (which for 1D Coulomb potential is twice degenerate with respect to parity, see Ref. in [19]), it is divided into two parts with an equal probability of finding the electron on both sides of the singularity. Thus the singularity influences the wavepacket stronger than during the recombination process.

In the recombination process the phenomenon analogous to the stabilization against photoionization, namely a nonmonotonic behavior of the recombination probability as a function of the field amplitude and its dependence on the presence of the potential singularity can be shortly described as follows. For 1D the discrepancy of the results for the total population of the atom bound states, obtained for different shapes of the potential was strongest. The analogue of the stabilization was clearly visible in the case of a softened Coulomb potential and was absent for the potential with a singularity. In contrast to that, for 2D and 3D this effect could be observed rather in the presence of the singularity and was only weakly pronounced for the soft-core model potential. Surprisingly the dependence of the recombination level on the electric field intensity was not much sensitive to the pulse shape, as it is in the ionization process.

Nonmonotonical dependence of the total bound states population on laser intensity (i.e. the stabilization) appears in 1D simulations of both ionization and recombination processes in the case of the model potential with smoothed core only. It is not visible for Coulomb singular potential. The results of ionization simulations in 3D show that presence of the singularity is not very important (especially comparing to the influence of the pulse envelope shape). In the case of 3D recombination we have checked only the influence of the potential details for the rectangular pulses. Weak indications of stabilization are visible for real Coulomb atom while there is no stabilization for model atom.

4. Conclusion

The results concerning the effectiveness of ultrastrong field ionization, presented in this paper, show that in the 3D case using the softened potential as a model of the real Coulomb potential can be justified if qualitative results are expected. This is in contrast to the 1D case, in which the results strongly depend on the presence of the singularity and thus the results obtained for a softened Coulomb model essentially differ from those obtained for the potential with a singularity. In 3D the wave form of attosecond pulse, both its envelope and phase, strongly influences the ionization probabilities. In particular the stabilization is better pronounced for smooth pulses for both types of the potential, while the ionization level is higher for a singular potential for both smooth and rectangular pulses.

In the context of many simulations performed in 80s and 90s of the past century, it is important to notice that the results obtained for 1D soft-core model (Fig. 1, lower) are even qualitatively similar, for our parameters of the laser field and the potential, to those obtained using 3D Coulomb potential (Fig. 3, upper). In both cases the stabilization is clearly visible, the stabilization window has similar properties (depth and range) and the slow drift is rather unimportant. This confirms the common belief that a soft core one-dimensional model may be used to test the behavior of a real atom during ionization.

In the 3D case the discrepancy between the results for the two classes of the binding potential is less pronounced in the ionization than in the recombination. Instead, the differences due to a different pulse envelope, which were not very important in the recombination, seem to be essential in the case of the ionization process.

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