

Quantum entanglement and advanced numerical modeling of strong-field ionization

Summary of the Ph.D. thesis

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Introduction

Pioneering experiments with attosecond light pulses [1–6], based on high-order harmonic generation (HHG) in noble gases [7, 8], have been revolutionizing our view of fundamental atomic, molecular and solid state processes in this time domain [9]. A key step in gas-HHG is the tunnel ionization of a single atom and the return of the just liberated electron to its parent ion due to the strong, linearly polarized femtosecond laser pulse driving this process [10, 11]. Recent developments in attosecond physics revealed that the accurate description of this single-atom emission is more important than ever, especially for the correct interpretation of the experimental data obtained by attosecond metrology.

Although an intuitive and very successful approximate analytical solution [12] and many of its refinements exist [13], the most accurate description of the single-atom response is given by the numerical solution of the time-dependent Schrödinger equation (TDSE). The peculiarity of this problem is due to the electric field strength of the laser pulse, which has its maximum typically in the range of 0.05-0.1 atomic units, i.e. it enables the tunneling of the electron through the time-dependent potential barrier formed by strongly distorting the atomic potential, but this effect is weak during the whole process. On the other hand, this small part of the wave function outside the barrier extends to large distances and in fact this is the main contribution to the time-dependent dipole moment, which is the source of the emitted radiation. Thus, a very weak effect needs to be computed very accurately, and these requirements get even more severe, if the model goes beyond the usually employed single-active-electron and dipole approximations.

For linearly polarized pulses, the main dynamics happens along the electric field of the laser pulse which underlies the success of some one-dimensional (1D) approximations. These typically use various 1D model potentials to account for the behavior of the atomic system. However, the particular model potential chosen heavily influences the 1D results and their comparison with the true three-dimensional (3D) results is usually nontrivial. Therefore, a

more elaborate connection between the 3D problem and its 1D model is necessary to allow the 1D simulation of strong-field processes physically as correctly as possible.

Although strong-field ionization is widely used as a standard procedure for high-order harmonic generation, it is very little known that this process generates also quantum entanglement between the liberated electron and its parent ion-core. Quantum entanglement is a fundamental feature of quantum theory which enables strong correlations without classical counterpart between constituents of a quantum system. Despite the fact that its discussion dates back to the early days of quantum theory [14], the features of continuous variable quantum entanglement [15] are still much less explored and utilized than those of discrete variables systems. Entanglement between the fragments of an atomic system due to a light-induced break-up process, like photoionization and photodissociation, was studied by Fedorov and coworkers [16, 17] in the framework of Gaussian states. However, this latter approach is not suitable to deal with the problem of quantum entanglement during the strong-field ionization of an atom, which motivated us to perform an accurate numerical investigation of the problem.

Objectives

We adopted the usual semiclassical description of the light-matter interaction for the modeling of strong-field phenomena described in the Introduction, making also use of the dipole approximation and choosing the length gauge [18]. In dipole approximation, the spatial variations of the electromagnetic field are neglected in the spatial domain of the atomic processes which is true in the frequency range relevant for the phenomena treated in this thesis. We aim to investigate the effects of a linearly polarized few- or single-cycle femtosecond laser pulse, which has a near-infrared carrier frequency. Our motivation is that this type of laser pulse is short enough to give a more complex response than a simple plane wave. The form of the Hamiltonian suggests the solution of the 3D time-dependent Schrödinger equation in cylindrical coor-

dinates z and ρ , where the z -axis coincides with the polarization of the laser pulse. The resulting time evolution preserves the magnetic quantum number m and the initial axial symmetry of the wave function if given.

Our first objective was to construct a novel numerical method that is capable to compute the time propagation of the electron's wave function by solving this cylindrical TDSE with high-order accuracy, incorporating also the singularity of the atomic Coulomb potential. Our aim was to construct an algorithm that scales approximately linearly with the total number of spatial gridpoints and that supports parallelization, whenever its possible.

In our earlier work [A1], we modeled a single-active-electron atom as a two-body system in one spatial dimension, using the Dirac-delta potential as the atomic model potential. We verified a strong correlation between the shape of the laser pulse and the oscillations of the quantum entanglement, most importantly, the local maxima of the quantum entanglement coincide with the zero crossings of the laser electric field. A straightforward question is, whether such correlations are also present in the strong-field ionization of a real atom? In accordance with this, our second objective was the analysis of the quantum entanglement properties of a single-electron atom in 3D under strong-field ionization as also mentioned in the Introduction. In this description we modeled a single-electron atom as an interacting two-body system of the electron (e) and the ion-core (c) subsystems. Even though the numerical solution can be carried out, the computational cost of calculating the true electron – ion-core entanglement is prohibitive. We aimed to give an approximate solution for this problem by a directionally separable approximation, and this way to gain insight also into the intricacies and the structure of the pair correlations between the electron's and ion-core's coordinates $z_e - z_c$ and $x_e - x_c$.

Our third objective was to find a one-dimensional quantum mechanical model that is capable of providing quantitatively good agreement with the true 3D solution of strong-field processes, reduced to the z axis. The one-dimensional description should use only the z Cartesian coordinate, with the very same laser electric field term in the time-dependent Schrödinger equa-

tion as in 3D, but the atomic potential should be replaced with a suitable time-independent $V_{0,M}^{1D}(z)$ atomic model potential. We aimed to derive an analytic form for this model potential based on a clear physical principle, and to compare it to well-known existing model potentials like e.g. the 1D soft-core Coulomb model potential $V_{Sc}^{1D}(z) = -1/\sqrt{z^2 + 2}$. We also aimed to test the improved model potential rigorously, focusing especially on the particular system's strong-field ionization features.

Methods

For the numerical solution of the time-dependent Schrödinger equation, we use a combination of various methods. For the spatial derivatives, we use the high-order finite difference formulation [19] with equal grid spacing Δz , $\Delta \rho$ in cylindrical coordinates. For the time evolution algorithm, we perform steps in small time intervals Δt using the short-time approximation of the time evolution operator $e^{-i\Delta t H_k}$ with the second-order effective Hamiltonian $H_k^{(2)}$ at the k th time step. We also utilize the second-order Padé-approximation of the exponential operator $e^{-i\Delta t H_k^{(2)}}$, which is the usual Crank-Nicolson method [20]. The latter is an implicit method, which means that it involves the solution of a systems of linear equations at every time step, the coefficient matrix of which includes the spatially discretized Hamiltonian matrix. The great advantage of this method is that it also allows us to incorporate any boundary condition into the implicit equations, thus allowing high-order spatial accuracy for a wide variety of problems. For gaining the speed that we need, we also utilize the split-operator methods [21]: by splitting the effective Hamiltonian as $H_k^{(2)} = H_A + H_B$, these methods factorize the above exponential operator into parts that are easy to solve. The most famous of this formula is the second-order accurate symmetric splitting $U_2(t + \Delta t, t) \approx e^{-i\frac{\Delta t}{2} H_B} e^{-i\Delta t H_A} e^{-i\frac{\Delta t}{2} H_B}$. The main advantage is that we can achieve linear scaling with the number of gridpoints (if we split kinetic energy terms in the Hamiltonian directionwise). The disadvantage is that it is not possible to split the Hamiltonian operator this way, if it is singular or

“very sharp” at a gridpoint. The final relevant splitting method is of Bandrauk and Shen [22], they give a formulation how it is possible to achieve high-order Δt convergence by performing a series of back-and-forth substeps with any properly constructed second-order accurate short-time evolution operator.

Deriving the formulas of the numerical method is hardly enough to determine its actual accuracy, rigorous testing is necessary in most cases. Our method of testing the spatial discretization involves computing the eigenenergy of the ground state and at least one excited state of known problems and comparing these values to other accurate numerical or analytical solutions. By changing the discretization parameter Δz (or $\Delta \rho$) we can determine the convergence order of the discretization scheme. These are straightforward typically for systems that have an analytic solution, like the harmonic oscillator and (most importantly) the Coulomb problem. However, analytical solution is not available for most of the time-dependent problems, thus we compare the results to a converged numerical solution that has orders of smaller numerical errors than the setup that we are investigating. The most straightforward method is to compare time-dependent mean values of these two solutions, which is an efficient way of error determination dependent on Δt , but it can be also used to test the spatial accuracy with a given Δz (and $\Delta \rho$) in these situations.

The problem of the interacting two-body quantum system, consisting of the electron (e) and the ion-core (c) as subsystems, is conventionally solved by transforming the system into the center-of-mass reference frame, where the wave function becomes separable in center-of-mass and relative coordinates. We describe the center-of-mass part as a free localized Gaussian wave packet, and the relative part involves the strong-field simulations of the relative particle with coordinates z, ρ . The form of governing Schrödinger equation is not changed, only the particle mass is replaced by the reduced mass. During the calculations of physical quantities we also utilize axial symmetry around z axis, which means that dynamics is the same in the x and y directions for each particle.

For the quantification of the quantum entanglement between the electron

and ion-core motion, the usual way requires the density matrix of either the electron or the ion-core by performing partial trace of the other degrees of freedom, assuming the whole quantum system is in pure state. Then the lack of purity of the density matrices is an indicator of quantum entanglement, and can be quantified by calculating the Neumann entropy – amongst other types of quantum entropies – of either subsystem’s density matrix. If we can perform this at every time instant we can see the time-dependent entropy dynamics of two particles’ entanglement, which is unfortunately a feasible computation in one dimension only. In three dimensions, this type of bipartite approach can be applied to the cylindrical relative coordinates z, ρ , by performing the partial trace on one of these coordinates, which gives interesting information about the directional nonseparability of the strong-field process. However, the composite system of these two particles has six degrees of freedom in 3D, which implies a more complex correlation structure between their individual coordinates $z_e - z_c$ and $x_e - x_c$. We investigated the nature of these correlations using recent results of quantum information theory [23]. One can quantify these directional pair correlations generally using the so-called $S(e : c, t)$ quantum mutual entropy, and investigate the behavior using the $S(e|c, t)$ or $S(c|e, t)$ quantum conditional entropies, which latter characterize the remaining entropy of one subsystem if the other one is measured. In the classical limit these two satisfy the relations in classical information theory, however quantum entanglement introduces nonclassical values to them.

For the low-dimensional modeling of an atom, we used the elements of the density functional theory [24]. We derived our model potential in analogy with the the exact calculation of the Kohn-Sham potential of a helium atom with a single Kohn-Sham orbital: knowing the correct reduced (single-particle) density one can invert the Schrödinger equation to determine the Kohn-Sham potential which ensures that the resulting Hamiltonian’s ground state has the correct reduced density. In this way one can model the ground state of the system physically as accurately as it is possible with a single orbital.

Scientific results

In the following, I present a brief summary of my new scientific results discussed in the thesis which are collected in five thesis points. The publications connected to my statements are listed at the end of this booklet and cited in each title.

T1. The hybrid splitting algorithm for the solution of the three-dimensional time-dependent Schrödinger equation with Coulomb singularities [P1]

To treat the Coulomb singularities in the 3D Schrödinger equation using cylindrical coordinates, I derived the formula of the boundary condition of the Coulomb potential in the axially symmetric case, which is a Robin type boundary condition at the $\rho = 0$ axis. I also gave the discretized formulation of this boundary condition using one-sided finite difference formulas.

Based on the 4th-order finite difference discretization of the Hamiltonian in the Crank-Nicolson method, I created the method of hybrid splitting. This uses partial directional splitting of the short-time evolution operator based on the splitting of $H_k^{(2)} = H_A + H_B$ both according to spatial directions and spatial domains: the H_A near $\rho = 0$ is left intact to retain spatial numerical accuracy near the boundary, while in the outer domain ($\rho \gtrsim 1$) the z -component of the kinetic energy is moved into H_B .

I created an optimized algorithm to solve the special block pentadiagonal system of linear equations that is provided by the Crank-Nicolson approximation of the central exponential operator $e^{-i\Delta t H_A}$. This algorithm reduces the computation costs by N_ρ^2 where N_ρ is the number of the discretization points along the ρ direction.

I verified that the discretization scheme is 4th-order accurate in spatial steps Δz and $\Delta \rho$ by computing the eigenvalues of the Coulomb and the harmonic oscillator Hamiltonians. Using the time-dependent analytic solution of the forced harmonic oscillator, I verified that the hybrid splitting method is Δt^2 accurate, and that it can be successfully combined with the 4th-order

approximation of the evolution operator to become Δt^4 accurate in temporal steps.

T2. Quantification of the electron – ion-core quantum entanglement during strong-field ionization [P2]

I showed that the computation of the Neumann entropy in a 3D simulation of strong-field ionization of the electron – ion-core system is not feasible due to the prohibitively large numerical load of the problem. Therefore, I created the following procedure to characterize the quantum entanglement of the electron – ion-core system in a 3D strong-field simulation.

First I reduced the 3D dynamics along spatial directions (parallel and perpendicular to the polarization of the laser pulse) by partial tracing of the respective density matrices. Then I performed the transformation in each direction to electron and ion-core coordinates and reduced these density matrices to single-particle density matrices by tracing over the respective particle coordinates. Using known relations of quantum entropies, I showed that the type of the correlation between the same coordinates of these two particles in each direction is dominantly quantum entanglement. I quantified this directionwise quantum entanglement via the average directional mutual entropy. I verified that the time evolution of the average mutual entropy along the polarization direction is very similar to that of the exact quantum entanglement entropy obtained by my former 1D model simulations.

Using these density matrices as building blocks, I approximated the quantum state of the 3D electron – ion-core system by the product of the spatially reduced two-particle density matrices, in accordance with my experience that the strong-field dynamics along the above spatial directions is weakly coupled.

Based on this, I defined the approximate entanglement entropy of the 3D electron – ion-core system by adding up the directional average mutual entropies individually, which thus can be efficiently computed based on the entropies of the building block density matrices. I showed via simulations that

this approximated entanglement entropy satisfies the strong subadditivity relation, which is an analytic property of the exact entanglement entropy.

T3. Features of the electron – ion-core quantum entanglement during strong-field ionization [P2]

Since the spatially reduced subsystems described in **T2** are in mixed states, their correlations are more complicated than pure state entanglement, thus I identified the physical meaning of the several kinds of entropies involved in the above procedure and I made the following observations.

I analyzed the entanglement entropy relations in each of the directions specified in **T2** and I found that their local maxima almost coincide with the zero crossings of the electric field of the laser pulse. The entanglement along the directions parallel and perpendicular to the laser polarization are very similar to each other if the process stays in the tunnel ionization regime. However, in the over-the-barrier ionization regime, I found entropy increase along the parallel direction but a surprising entropy decrease in the perpendicular directions which causes also the total electron – ion-core entanglement entropy to decrease.

I investigated the dependence of these proposed measures of entanglement dynamics on the strength and the carrier-envelope phase of the driving laser pulse. I found many features of quantum entropies that do not depend on these parameters, like the electron – ion-core entanglement has local maxima always near the zero crossings of the laser pulse. I found that while the intensity of the field governs the dynamics as a whole, the carrier-envelope phase changes the subcycle dynamics of the strong-field ionization.

T4. One-dimensional density-based model potentials: comparison of the 1D and 3D results [P3]

I derived the analytic formula of a 1D atomic model potential that by definition has the same ground state probability density as the probability density of the 3D Coulomb problem integrated over ρ . I determined that

through asymptotics this 1D system preserves the 3D ground state energy if the source 3D problem has a long-range Coulomb form. The density-based potential consists of a 1D regularized Coulomb potential plus a kinetic energy correction.

I realized that the regularized Coulomb potential part of the density-based model potential suggests the value of $\frac{1}{2}Z$ for the effective ion-core charge in 1D. Based on this, I proposed improved formulas of the 1D soft-core Coulomb potential and the 1D regularized Coulomb potential by applying the effective charge $\frac{1}{2}Z$ while maintaining that their ground state energies equal to that of the 3D Coulomb problem.

I showed by direct comparison of simulation results of typical time-dependent strong-field processes, driven by a linearly polarized near-infrared laser pulse, that both these new and the proposed improved 1D model potentials exhibit an impressive enhancement in the accuracy of the low frequency response of typical strong-field processes by capturing the essence of the real 3D dynamics. These tests also showed that the best model potential quantitatively is the improved soft-core Coulomb potential.

I also computed the dipole power spectrum in a wide variety of cases and I observed that the structure of the spectra based on the density-based and improved 1D model potentials is remarkably similar to those based on the 3D simulations. The match of the corresponding spectral phases is also very good, especially in the higher frequency range, which is of fundamental importance for the generation of isolated attosecond pulses. I gave a simple frequency-dependent scaling function that proved to be capable to convert the 1D spectra to the corresponding 3D spectra in all of the tested cases, thus it enables to compute the dipole power spectra via 1D simulations.

T5. Improved numerical method of constructing discrete model potentials in one dimension: improved simulation accuracy [P3]

I suggested a formula to acquire an improved discretized representation of 1D model potentials, based on the potential's ground state and ground state energy, by inverting the discretized time-independent Schrödinger equation. The resulting discretized potentials have the numerically exact ground state and ground state energy. I showed that if the exact 1D potential is not differentiable at some spatial point (e.g. like a regularized 1D Coulomb potential) then the resulting discretized Hamiltonian is still Δz^4 accurate, if the finite difference formulas of the partial derivatives is also at least Δz^4 accurate, even during simulations of strong-field ionization phenomena.

I showed that the application of this method is possible also for the 1D Dirac-delta potential. The same inversion formula as above gives a nonsingular discretized model potential. I tested the results using convergence tests of strong-field simulations, the numerical errors at $\Delta z = 0.2$ were comparable to the correct method used in [A1]. I came to the conclusion that this nonsingular method of discretization converges to the true solution, and it shows Δz^2 numerical accuracy.

Publications

Refereed research papers related to the thesis:

- [P1] Szilárd Majorosi and Attila Czirják. Fourth order real space solver for the time-dependent Schrödinger equation with singular Coulomb potential. *Computer Physics Communications*, 208: 9–28, 2016; doi:10.1016/j.cpc.2016.07.006.
- [P2] Szilárd Majorosi, Mihály G. Benedict, and Attila Czirják. Quantum entanglement in strong-field ionization. *Physical Review A*, 96(4): 043412, 2017; doi: 10.1103/PhysRevA.96.043412.
- [P3] Szilárd Majorosi, Mihály G. Benedict, and Attila Czirják. Improved one-dimensional model potentials for strong-field simulations. *Physical Review A*, 98(2): 023401, 2018; doi: 10.1103/PhysRevA.98.023401.

Additional refereed research papers and conference proceedings:

- [A1] Attila Czirják, Szilárd Majorosi, Judit Kovács, and Mihály G. Benedict. Emergence of oscillations in quantum entanglement during rescattering. *Physica Scripta*, 2013(T153): 014013, 2013. doi:10.1088/0031-8949/2013/T153/014013.
- [A2] Attila Czirják, Szilárd Majorosi, Judit Kovács, and Mihály G. Benedict. Build-up of quantum entanglement during rescattering. In *AIP Conference Proceedings*, 1462(1): 88-91. AIP, 2012; doi: 10.1063/1.4736766.

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