Dynamics and radiation of atomic and free electrons interacting with an intense laser pulse

Summary of Ph.D. thesis

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Introduction

Real-time studies of ultrafast processes in atoms, molecules and solids can now be routinely performed in the leading laboratories of the world [1, 2], and attosecond physics has become one of the fastest developing fields of physics [3, 4]. At the time of writing this thesis, those phenomena are considered ultra-fast processes whose characteristic time scale is in the order of hundred attoseconds (as). These processes, which in most cases are related to the dynamics of the electron system in the atom or molecule, currently represent the lower limit of measuring dynamic processes.

The state-of-the-art measurement method based on light-matter interaction is the so-called pump-probe experiment having the finest time resolution, i.e. dynamic processes of 50 to 100 as can be resolved [5, 6]. An essential part of this method is the generation of attosecond light pulses, usually in the XUV – soft X-ray spectral range, typically by high-order harmonic generation on noble gas atoms [7, 8, 9]. However, due to the characteristics of the secondary radiation source, i.e. the noble gas sample, this method has its own limitations in terms of the intensity and pulse length of the emitted light pulse. On the other hand, it is a well-known fact that the carrier-envelope phase difference (CEP) of the few-cycle, femtosecond laser pulse, involved in most of these pioneering experiments, affects various processes in atomic or molecular systems on this time scale. Recently, it has been proven that the phase difference of CEP of the attosecond light pulses is also crucial in these pump–probe experiments [10, 11].

The strong-field ionization of atoms plays a fundamental role in attosecond physics. A sufficiently strong laser pulse enables an electron to escape from its atomic bound state into the continuum; this is usually assumed to happen by tunnelling, which is the first step of the very successful three-step model [12, 13]: (i) the electron is released from the Coulomb potential of the atomic ion core by tunnelling, (ii) as a classical particle it gains kinetic energy from the laser field, (iii) the electron returns to the vicinity of the parent ion core, and under appropriate conditions they recombine while emitting ultraviolet radiation (photon). This model underlies much of our understanding in this area. Currently, the problems of tunnelling time and exit momentum are of outstanding importance regarding both quantum theory and attosecond metrology [14].

Objectives

The objective of the work presented in this PhD thesis was to examine the open questions related to the theoretical models and the generation of attosecond light pulses, and to attempt to answer some of these questions.

As mentioned in the previous section, the method most frequently used for the generation of attosecond XUV pulses has its own physical limitations due to the properties of noble gas atoms, i.e. secondary light sources. Therefore, we were scientifically motivated to look for a new, alternative method. One of the main problems of high-harmonic generation on noble gas atoms is that the maximum intensity of the generated attosecond light pulse is limited: the intensity of the driver laser pulse must not exceed a threshold value, because a very intense laser field would ionize the gas, and therefore prohibit recombination and the emission of ultraviolet radiation.

It has been long known that when electromagnetic radiation is scattered on a free, relativistic electron (this phenomenon is called Thomson or Compton scattering depending on the energetic conditions), the scattered radiation also contains components with much higher frequencies than the central frequency of the laser field [15, 16, 17, 18]. It is also known that under appropriate experimental conditions an electron bunch containing many particles is capable of emitting coherent radiation, which was previously used to produce mono-energetic radiation [19, 20]. Based on these two findings, as well as pioneering simulations [21, 22] and experiments [23, 24] related to electron nanobunches, we aimed to investigate whether the interaction between an electron-bunch with appropriate parameters and a suitable laser pulse can emit attosecond light pulses that can be detected macroscopically and used in experiments.

The optical ionization of atoms or molecules is an interesting phenomenon due to the associated fundamental physical questions. In addition, it is an important process not only for the production of attosecond light pulses with noble gas atoms, but also for the interpretation and evaluation of measurement results obtained with modern devices. These measurement methods can detect individual ionization events based on electrons, ions and other fragments.

Our objective was to investigate the quantum and classical dynamics of ionization in a strong laser field as a time-dependent phenomenon, and to compare the two dynamics in the phase space. We examined under what initial conditions classical dynamics approximates quantum dynamics derived from the solution of the Schrödinger equation. Furthermore, an important and far from trivial question in such a time-dependent quantum mechanical process is the following: from which point of the phase space can dynamics be considered classical? Based on the comparison of classical and quantum dynamics and using the deterministic nature of classical dynamics, we also examined the possibility of reconstructing the starting point of the classical trajectory that best models quantum dynamics from measurable physical quantities.

Methods

From among the theoretical models most frequently used for describing light-matter interactions, we opted for the classical light and classical material or the semi-classical description due to the peculiarities of the studied physical problems.

In the case of relativistic Thomson scattering, any change in electron energy is neglected compared to the change in light energy. During the discussion of this problem, we considered the electron as a classical source of electromagnetic radiation and the scattering laser field as a classical electromagnetic wave. Thus, the electron moves under the influence of the laser field according to the classical Newton-Lorentz equations, and in a sufficiently distant point in space the spectral distribution of the far-field radiated by the electron can be obtained using the well-known formula based on Liénard-Wiechert potentials. Although numerical methods for dealing with similar problems are becoming more common today, we limited ourselves to an exact, analytical solution of the equation of motion, and we used numerical methods to calculate the emitted field radiated by the electron or by the electron bunch only. When we calculated the radiation field of a so called electron bunch consisting of a large number of particles instead of a single electron, we assumed that the electrons travel along the same trajectory, they differ only in the initial trajectory coordinates. Therefore, when we calculated the electric field emitted by the electron bunch, the spectrum of the radiation field from a single emitter was multiplied by the coherence factor, to obtain the radiation field of the "ideal" electron-bunch having the assumed parameter.

Strong-field ionization is a more complex problem in the sense that we aimed to describe the electron of the hydrogen atom (which served as a model) from both the classical and quantum dynamics perspectives. Thus, when we considered the electron as a classical particle, we confined ourselves to the non-relativistic form of the previously mentioned Newton-Lorentz equation and its partly analytical and partly numerical treatment. When solving the time-dependent Schrödinger equation, we used dipole approximation for the interaction of a single active electron atom with the classical electromagnetic field in the length gauge. The assumed linearly polarized laser pulse excited the electron from its atomic ground state. As the electron's wave function does not depend on the azimuth angle around the polarization axis, we were able to write the three-dimensional time-dependent Schrödinger equation in cylindrical coordinates. Then we used a hybrid Crank – Nicolson method based on operator splitting [25]. In order to achieve better comparability with classical physics, we investigated the quantum mechanical problem in the phase space with the Wigner function and with the so-called quantum momentum function.

Scientific results

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

1. The possibility of generating an attosecond light pulse by Thomson scattering on a suitable electron bunch [T1,T2]

I have given a particular solution to the relativistic trajectory of a point charge that interacts with a laser pulse described by a sine-squared envelope and often used in practice. Using the analytical trajectory of the electron, I calculated the spectral distribution and temporal dependence, and investigated the spatial dependence of electromagnetic radiation emitted by a single electron and a monoenergetic electron nanobunch during the Thomson scattering of an intense, linearly polarized, single-cycle, near-infrared laser pulse. I have found that an electron bunch with suitable properties can emit a light pulse having a pulse length of 16 attoseconds (full width at half maximum) and an energy of 99 nJ in the extreme ultraviolet–soft X-ray spectrum without any spectral filtering, due to the collective nature of the radiation. The spectrum of the attosecond light pulse includes the so called “water window” ranging from 2.33 nm to 4.37 nm, which is of great importance for life sciences.

2 Proposal for the dynamic control of the carrier-envelope phase difference of an attosecond light pulse [T2]

I investigated the dependence of the electric field radiated by the electron nanobunch on the carrier-envelope phase difference of the driving laser pulse during relativistic Thomson scattering. I found that there is a simple linear relationship between the carrier-envelope phase difference of the driving laser pulse and the carrier-envelope phase difference of the emitted attosecond light pulse. This property of the attosecond light pulse generation process makes the carrier-envelope phase difference dynamically controllable.

3. Proposal for generating a high pulse energy attosecond light pulse [T3]

I found that any increase in the intensity of the laser pulse scattered on the electron bunch only lightly increases the length of the attosecond light pulse generated during the scattering process. However, the intensity of the light pulse increases considerably and non-linearly up to a certain saturation value, which depends on the parameters of the interaction. Furthermore, assuming a chirped incoming laser pulse I found that a properly chosen negative chirp modifies the spectral distribution of the emitted field without significantly affecting the pulse shape, but the energy of the attosecond light pulse can be increased up to the μJ energy range.

4. Introduction of a new type of classical trajectory for the approximate description of strong-field ionization [T4]

For strong-field ionization I suggested classical trajectories with improved initial conditions for the classical description of the electron liberated from the atomic potential. The initial conditions of such a trajectory are provided by the outermost inflection point of that stationary phase space trajectory of the instantaneous potential which intersects the quantum momentum function at the inflection point. When comparing the Wigner function of the escaped wave packet and the quantum momentum function, I found that a suitable set of such trajectories with different starting times gives an apparently good approximation of the quantum dynamics of the ionized wave packet, and such a set also reflects the temporally blurred feature of the liberation process. Furthermore, I found that the initial energy of the classical electron travelling on the trajectory falls in the range of over-the-barrier ionization, which solves and explains the problem of the non-zero initial momentum in models assuming traditional tunnelling.

5. Reconstruction procedure for determining the classical initial parameters of a directly ionized electron [T4]

Using the trajectories I introduced earlier, I proposed an iterative method to reconstruct the starting time and position, and then the starting momentum, of the classical trajectory from the measured momentum of a directly ionized electron (i.e. ionized without rescattering). I tested the proposed method using a numerical experiment for the range of possible starting times for different laser intensities and carrier-envelope phase difference values. I found that the method reconstructs the starting time of the classical trajectory with a maximum deviation of about 2 a.u. (ca. 50 as), however, for the more probable trajectories the deviation is less than 5 as, and for the initial position it is less than one tenth of the Bohr radius (5 pm).

Refereed research papers related to the thesis:

[T1] Sz. Hack, S. Varró and A. Czirják, „Interaction of relativistic electrons with an intense laser pulse: HHG based on Thomson scattering”, *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **369**, 45-49 (2016)

[T2] Sz. Hack, S. Varró and A. Czirják, „Carrier-envelope phase controlled isolated attosecond pulses in the nm wavelength range, based on coherent nonlinear Thomson-backscattering”, *New Journal of Physics* **20**, 073043 (2018)

[T3] Sz. Hack, Z. Tóth, S. Varró and A. Czirják, „Isolated attosecond pulses of μJ energy via coherent Thomson-backscattering, driven by a chirped laser pulse”, *The European Physical Journal D* **73**, 77 (2019)

[T4] Sz. Hack, Sz. Majorosi, M. G. Benedict, S. Varró and A. Czirják, ”Quantum interference in strong-field ionization by a linearly polarized laser pulse, and its relevance to tunnel exit time and momentum” *(submitted);* arXiv:2103.12699

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