Summary of the PhD thesis

Macroscopic study and control of high-order harmonic and attosecond pulse generation in noble gases

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Introduction

The main driving force of science is our inexhaustible ambition to understand and control the world around us. People are always trying to design new tools, or push the boundaries of the existing ones to study, explain or predict yet unobserved or unexplained phenomena. As the most natural observing instruments available to us are our eyes and ears, the phenomena first studied by mankind were those that happen on the time scales these “instruments” can detect.

However, our curiosity goes way beyond the scales what we can directly observe with our primary senses. This curiosity has lead our way in designing tools enabling us to reveal the hidden world of objects too small, too fast, too slow, or simply too distant to be perceived directly. In general, recording and visualizing a fast event can be done by taking still pictures of it in its different phases, then, viewing these pictures in sequence gives the illusion of motion due to the phi phenomenon.

The time resolution of mechanical and purely electronic devices is limited to the microsecond (1 μs = 10^{-6} s) and nanosecond (1 ns = 10^{-9} s) scale. To record faster events new ways of taking still pictures were developed. Recording a picture over a relatively long time in complete darkness but illuminating the target by a very short light pulse ensures that the detector sees and records only a single phase of the event – while it is illuminated by the light pulse. By taking advantage of the fact that ultrashort electromagnetic pulses can be transmitted through air or vacuum, detectors based on light pulses initiated the next breakthrough in the available time-resolution. In this case the time-resolution is limited by the length of the light pulse, and by the precision this can be synchronized with the event in question—after all, no matter how good the picture we are taking is, if nothing is happening while it is taken.

Ultrashort light signals became available to researchers after the invention of lasers in the middle of the 20th century. The evolution of laser technology made possible the generation of light pulses with picosecond (1 ps = 10^{-12} s) and then even with femtosecond durations (1 fs = 10^{-15} s). As even light only travels about 0.3 μm in one fs, the processes to be studied with such high resolution involve minute particles that move very fast, like atoms under the force of electric fields. These ultrafast detection methods can thus be applied to study and control processes on the time scale of molecular reactions and they initiated the rise of a whole new research field, called femtochemistry.

Although this field only started by researchers trying to measure the time needed for a molecule's vibrational energy to redistribute between different vibrational modes, it led to the understanding of such complex processes like the human vision. In fact, in his Nobel lecture in 1999, Ahmed Zewail (who is also known as the father of femtochemistry) stated that he believes “every time we improve the time resolution by a factor of even a hundred or a thousand, we must be able to see new phenomena that we did not even think of”.

Improving the time resolution further leads us to the time domain of attoseconds (1 as = 10^{-18} s), and also to smaller and faster particles interacting with these pulses and/or evolving in this time domain: the electrons. Like in femtochemistry, the study of attosecond processes promises a new, deeper level of understanding of nature and one of its most fundamental processes, the interaction of light with matter. As the formation of all molecular compounds is also governed by the Coulomb force acting on the nuclei and the surrounding electrons, it is unquestionable that the understanding of atomic and molecular processes in general also relies on understanding the motion of electrons.

Obviously, to directly study the time-evolution of these processes one needs signals which are in this time domain as well. Light pulses significantly shorter than 1 femtosecond are called attosecond pulses and, to date, they are the shortest experimentally demonstrated and controllably reproducible coherent light pulses available to researchers.

The main laboratory source of attosecond pulses today is the generation of high-order harmonics in gases by intense, femtosecond infrared laser pulses. The result of this process is a broad spectrum of XUV to x-ray radiation ending in a cutoff and containing odd harmonics of the fundamental field. The special phase-relation of these harmonics enables the synthesis of attosecond pulses by filtering out the low order harmonics from the radiation. As this process is highly nonlinear and relatively inefficient, the main focus of research today is the optimization of the generation efficiency, the isolation of a single attosecond pulse from a pulse-train, and the generation of the shortest possible attosecond pulse.

The aim of this work, which started in 2010, was to investigate high-order harmonic and attosecond pulse generation in gases by near-infrared laser pulses, with special focus on the effects of weak, long-wavelength assisting fields, and on the macroscopic processes involved in the generation.

Scientific background

It can be deduced from the Fourier transform that an attosecond pulse can be synthesized only from very broadband radiation, spanning over ~tens of eVs. Thus attosecond pulse generation requires the use of at least UV, but more preferably XUV or x-ray frequencies, a wide bandwidth and also control over spectral phase. The two most widespread candidates to achieve this goal are high-order harmonic generation in solid surfaces and in gases.

Generation of high-order harmonics on solid surfaces has the advantage that it has not got a known upper limit to the applicable laser intensity, therefore it is a possible source of very bright x-rays. This process is under intense investigation, and it might, one day, be a widespread method for attosecond pulse generation. However, because of the large intensities required to produce these harmonics, the large divergence of the generated radiation, the technical difficulties risen by the need for “fresh” solid surface at every laser shot and the challenges in stabilization of the beam reflected from the moving surface, these methods of x-ray generation are still not widespread, and limited to smaller repetition rates.

Today, attosecond pulses are most widely generated by the process of high-order harmonic generation (HHG) in gas targets. This mechanism produces XUV to x-ray photon energies with very wide bandwidth and excellent temporal and spatial coherence. It is demonstrated that this process works at very high repetition rates (~MHz) and can produce attosecond pulses, although with relatively low efficiency. Due to this, it is actively researched not just for its direct applications in attophysics, but also for its possible use in generating seed pulses for some of the more efficient schemes which are able to produce very intense radiation, like free-electron- or x-ray-lasers.

High-order harmonic generation in gases

In this scheme an intense, femtosecond, infrared (IR) laser pulse is focused into a gas cell or jet placed inside a vacuum chamber. The vacuum is needed to prevent: 1) the destruction of the pulse before the focus by nonlinearities in air, and 2) the absorption of the generated radiation. The gas jet (or cell) contains a gas with high ionization potential (in most cases a type of noble gas), that can withstand intense laser radiation without complete ionization. Through interaction of the laser pulse with gas particles, high-order harmonic generation takes place inside the gas cell, and the unconverted laser pulse and the harmonic beam leave the target gas together. At this point, the spectrum is dominated by the low-order harmonics, hence the duration of the pulses is in the femtosecond domain. After leaving the target cell, a thin metallic foil (Aluminium, Zirconium etc.) blocks the IR beam and also the low-order harmonics while letting the XUV beam through. An aperture is usually used to block the divergent part of the beam. The remaining harmonic beam – now collimated and free of low-order harmonics – contains a train of attosecond pulses, which can be manipulated with specialised XUV optics and used in applications.

Single-atom model

The first successful model that explained the HHG process in an intuitive manner used an approach from plasma physics: it assumed that the laser field forces the electrons into continuum by tunnelling ionization, and there they move in the field of the laser, unaffected by the Coulomb field of the parent ion.

This approach then gained the name of classical or three step model of HHG. This model is based on the fact that the laser electric field strengths used in HHG are already comparable with the Coulomb field acting on the outermost electron. Together they create a potential barrier through which electrons can tunnel into the continuum, and quickly departing from the ionic core they become unaffected by the Coulomb field so their movement in the laser field can be treated classically. As they oscillate in the laser electric field, some of them re-encounter the parent ion, recombining and releasing their energy in form of a high-harmonic photon. In this simple model a few assumptions are made:

1) the electrons appear in the continuum very close to the nucleus with 0 initial velocity,
2) after ionization, the motion of the electrons in the continuum is governed by Newton's equations, while the classical laser electric field is the only force acting on them,
3) if the electrons re-encounter the core, they emit a photon, whose energy is the sum of the electron's kinetic energy at the moment of recombination and the ionization energy.

Depending on the instant the electron "appears" in the continuum, it may drift away, producing above threshold ionized (ATI) electrons, or return to the nucleus, recombining and emitting a harmonic photon. The highest kinetic energy at the moment of first return is calculable to be 3.17 U_p, which explains both the presence and position of the sharp cutoff observed in the experiments, and shown to be at $I_p + 3.2U_p$.

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A more accurate method to calculate the generated high-order harmonic (HOH) spectrum was developed in the early years of HHG research. The ferivation is based on the strong-field approximation, and results the Lewenstein integral, which produces the dipole moment of an atom with a single active electron, placed in a strong laser field. Using this model the time-dependent dipole moment can be obtained. From this one can calculate the dipole acceleration, which is the source of the produced radiation. The Fourier transform of this gives us the harmonic spectrum, which, after spectral filtering, can be used to calculate the produced attosecond pulses.

Macroscopic processes

To accurately describe HHG in gases the entire macroscopic generation process has to be modelled. Because HHG is a highly nonlinear process, it is very sensitive to the shape and intensity of the laser field. A focused laser beam, however, has a spatial intensity distribution through the cross-section of the beam, and also different phase velocities on and off-axis around the focus. There are also propagation effects arising as the laser pulse propagates through the gas cell, like absorption and dispersion, and nonlinear interactions appear at high intensities, like self-focusing and plasma generation. These effects create different conditions for HHG in different parts of the gas cell.

Because HHG is a coherent process, perfect constructive interference from all radiating sources in the cell can cause the harmonic intensity to increase quadratically with the number of interacting particles, but destructive interference can eliminate the harmonic radiation altogether at the detection target. The phase difference between the atomic sources depends not only on the phase with which harmonics are generated, but also on their phase velocities in the generation medium. These processes altogether define the characteristics of the harmonic beam, and they can also significantly alter the intensity and structure of the attosecond pulses.

Phase-matching

To achieve constructive interference from the elementary radiating sources, the conditions of phase-matching have to be fulfilled. Phase matching, as its name suggests, describes how well the phases of harmonics generated at different parts of the cell matches each-other. The conditions for phase-matching – along the propagation axis of the fields – are fulfilled when the phase-velocity of the polarization generated by the laser field matches the phase-velocity of the harmonic field. This can be achieved by optimizing the focusing geometry, gas pressure and intensity of the leaser pulse. The latter is important because it defines the ionization rate, which strongly affects the phase velocity of the laser field.

In general phase matching is achievable only at moderate laser intensities. This limits the highest harmonic photon energy obtainable in HHG.

Quasi-phase-matching

At photon energies where conventional phase-matching is hard to achieve, quasi-phase matching (QPM) schemes are often used to increase harmonic yield. QPM is a powerful tool when conventional phase matching is not possible, thus phase-mismatch arises, resulting an oscillating harmonic intensity along the propagation axis. The zones where harmonic intensity increases/decreases are called zones of constructive/destructive interference. The basic idea of QPM is to eliminate harmonic emission in destructive zones, or switch these into constructive zones, thus increasing the harmonic yield over longer propagation distances.

As in HHG the traditional QPM schemes based on birefringence (achieved by periodic poling of the nonlinear crystal) are not possible other methods have been proposed. These are based on some type of periodic modulation along the propagation axis, which includes atomic density, driving field intensity, or modulation caused by a secondary periodic field, which is either static, or propagating in another direction than the driving field. QPM methods employing low-intensity assisting fields are based on the fact that the phase-shift induced by the assisting electric field scales linearly with its amplitude in the limit when that is much weaker than the amplitude of the generating field, and the shape of the phase-modulation resembles that of the assisting field.

The result of the phase-modulation is the lengthened constructive and shortened destructive zones. Due to this, the intensity of the generated harmonic increases approximately quadratically with the length of the cell as, with only slight sub-coherence-length oscillations around the parabola. The intensity in optimal QPM conditions might increase until it reaches the absorption limit. Whereas without the phase-modulation, the peak intensity is reached at half of the coherence length, severely limiting the achievable photon number in macroscopic media.

Periodic assisting fields that can induce QPM can be of many types: to date periodic static electric fields, counter-propagating (to the IR) quasi-cw laser fields and pulse trains and sawtooth-shaped fields been proposed or used.

Three-dimensional model

For an accurate description of the macroscopic generation process, a complete, three-dimensional model has to be used which takes into account the propagation effects affecting both the laser and harmonic fields.

The high-order components of the nonlinear polarization are orders of magnitude weaker than the low-order components (perturbative response). Thus, the propagation of the laser field can be described independently of HHG. The nonlinear response of the medium affecting the laser field can be calculated using the standard perturbative description but including the effect of plasma dispersion, while the generation of high-order harmonics and their propagation is described separately.

At the entrance of the gas cell we describe the laser field assuming a simple Gaussian beam. From there on, the electric field can no longer be described by an analytical expression, due to the distortion effects, so it is numerically propagated: a spatial grid is defined over the interaction region, and on every grid point the laser field is calculated by propagating it using the wave-equation.

From the propagated laser field, the produced harmonic field can be calculated in each spatial grid point using the previously mentioned Lewenstein integral. The total harmonic field leaving the cell is calculated by propagating the harmonic field through the medium, using the calculated dipole radiation as a source term in the wave equation.

Results

T1.a I have analysed high-order harmonic generation in the presence of strong THz fields, and I have shown that: THz pulses can cause a large extension of the cutoff with reduced GDD, and they can redistribute the amplitude of electron trajectories, making the shorter trajectory class more dominant. Besides the different trajectory lengths, the increased field strength at the moment of ionization (due to the shifted ionization times) also contributes to the stronger yield from short trajectory radiation.

T1.b I have studied how in a macroscopic environment the generation process differs significantly from the single-atom results, and shown that even in cases when longer laser pulses are used (8, 10 or 12 fs) and the single atom response would yield multiple attosecond pulses, propagation effects can eliminate the contribution from certain sets of trajectories, yielding an isolated attosecond pulse at the exit of the gas cell. The large bandwidth of these pulses greatly decreases their transform limit.

I have also shown that the long-trajectory components are cleaned from the surviving pulse during propagation, resulting in an effective decrease of pulse duration, making the technique promising for obtaining a reliable source of short, isolated attosecond pulses with good contrast and low divergence. By careful adjustment of the parameters, such as gas pressure and peak intensity of the laser pulse, and by adequate spectral filtering short SAPs can be produced in a straightforward manner (without post-compression).

T2.a I have analysed the importance of focusing geometry on phase matching and harmonic yield in HHG when the IR pulse is assisted by a THz pulse, using experimentally verified parameters. I have shown that, despite the limited THz pulse energy, the most powerful SAP can be produced by relatively loose focusing. I attributed this to the deteriorated phase-matching conditions under strong focusing of the long-wavelength fields.

T2.b I have shown that the assisting field can be used to compensate phase mismatch that arises during harmonic generation and the selection of the short or long trajectory components (defining the sign of the resulting SAP's chirp) can be achieved by varying the delay between the THz and IR pulses.

T3.a I have worked out a model suitable for further optimization, which is based on the single atom response calculation with limited temporal integration. I have shown with macroscopic HHG modelling that the Lewenstein integral is able to partially predict a good approximation of the macroscopic behaviour of the attosecond pulse generation process. I have interpreted the two distinct ways (consecutive half-cycle or short and long trajectory radiation in a single half-cycle generation) of the double attosecond pulse generation.

T3.b I have modelled attosecond pulse generation by the selected driver waveforms with a 3D macroscopic model and shown that the pulse shortening achieved by the optimization remains robust in a macroscopic environment. I have also shown that in case of Gaussian generating beams, tight spatial filtering of the harmonic beam is required for the pulse duration to come close to the ones obtained in the optimization. Moreover, I predict that double attosecond pulses generated from short and long trajectories are only reproducible macroscopically in very inefficient generating conditions.

T4.a I have described a method to calculate the phase-modulation of harmonics by a weak assisting field in terms of the generating laser pulse's parameters, depending on the two fields' relative wavelength and the length of the electron trajectory in question. I have discussed the relationship between the simplest case of a counter-propagating, same wavelength assisting field (analytical treatment), to the case when a different wavelength assisting field is used, and showed that the two can be related through a wavelength-dependent correction factor.

T4.b I have analysed the bandwidth of QPM methods and formulated an approximate expression to calculate it. I have shown that the dependence of phase-modulation amplitude on the harmonic order is not limiting the QPM bandwidth significantly when the method is applied to a large number of coherence periods. Therefore, in these cases the bandwidth does not depend on the shape of the applied assisting field. On the other hand, I have shown that trajectory interferences can significantly change the fine-structure of the QPM efficiency. I have also discussed the optimal field profile of assisting fields for short and long trajectory components for efficient QPM, and I have found that short trajectories have the advantage of requiring the same profile for driver and assisting beams.

T4.c Using a 1D model I have predicted that HHG can be enhanced by perpendicularly propagating long-wavelength fields at photon energies above the phase-matching limit. I calculated the amplitude of the optimal THz field that was used in 3D numerical calculations which verified that these fields are able to induce QPM. In a case study, I have found an increase in efficiency of more than two orders of magnitude around the region of the single-atom cutoff. A chirped THz field, matching the coherence length of the generated harmonics was found to further increase the generation efficiency.

T5 I participated in an experimental campaign measuring the variation of attosecond group delays with the pressure present inside the generation cell. We found that by increasing the pressure the group delay of the attosecond pulse train decreases, in agreement with a one-dimensional propagation model. Due to this, for the stability of attosecond pump-probe measurements to be maintained, not just the optical path lengths, but the generation gas pressure also has to be kept constant.

Publications

Published papers related to the thesis:

[T1.a] E. Balogh, J. A. Fülöp, J. Hebling, P. Dombi, Gy. Farkas, K. Varjú, *Application of High Intensity THz Pulses for Gas High Harmonic Generation*, Central European Journal of Physics **11**, 1135 (2013)

[T1.b] E. Balogh, K. Kovacs, P. Dombi, J.A. Fulop, G. Farkas, J. Hebling, V. Tosa, and K. Varju, *Single attosecond pulse from terahertz-assisted high-order harmonic generation*, Physical Review A **84**, 023806 (2011)

[T2] Emeric Balogh, Katalin Kovács, Valer Toșa and Katalin Varjú, *A case study for terahertz-assisted single attosecond pulse generation*, J. Phys. B: At. Mol. Opt. Phys. **45**, 074022 (2012)

[T4] Katalin Kovács, Emeric Balogh, János Hebling, Valer Tosa, and Katalin Varjú, *Quasi-phase-matching high-harmonic radiation using chirped THz pulses*, Physical Review Letters **108**, 193903 (2012)

[T5] D. Kroon, D. Guénot, M. Kotur, E. Balogh, E. W. Larsen, C. M. Heyl, M. Miranda, M. Gisselbrecht, J. Mauritsson, P. Johnsson, K. Varjú, A. L'Huillier, and C. L. Arnold, *Attosecond pulse walk-off in high-order harmonic generation*, Optics Letters **39,** 2218, (2014)

Conference proceedings related to the thesis:

[T4] K. Kovács, E. Balogh, J. Hebling, V. Toşa, and K. Varjú, *Quasi-phase-matching high-harmonics with THz assistance*, AIP Conference Proceedings **1462**, pp. 41-44 (2012)

Papers under review:

[T3] E. Balogh, B. Balazs, V. Tosa, E. Goulielmakis, P. Dombi, and K. Varju. *Genetic optimization of attosecond pulse generation in light-field synthesizers*, submitted

[T4] E. Balogh and K. Varjú. *Quasi-phase-matched high-order harmonic generation by low-intensity assisting fields: field strength scaling and bandwidth*, submitted