Control of high harmonic generation on noble gas atoms and clusters

Summary of the Ph.D. thesis

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Antecedent research and motivation

As laser technology emerged in the 1960s, a compact, intense coherent, easily controllable light source became available that gave base to immense progress in science and technology in the past six decades. The increase of laser intensities catalysed not only industrial applications, but a new, prospering branch of science appeared, nonlinear optics [1].

The signal emerging from classical nonlinear processes usually scales with the nth power of intensity, where n is the order of nonlinearity, which means orders of magnitude signal increase with more powerful light sources. Further developement and the invention of pulsed lasers created new potential, giving birth to laser induced strong-field physics. The definition of this regime is that the electric field of the laser exceeds the field defined by the atomic potential and the strength of the electron-electron interaction.

Higher order harmonic generation

The high harmonic generation (HHG) phenomenon is studied since the end of the '80s. Infrared or visible light generates its higher order harmonics while focusing it into a gas (or solid) medium. It was proposed a few years later, that with this technique attosecond pulses can be created [2]. During gas HHG experiments, mostly noble gases are used, but some other research also applies molecular systems, e.g. CO_2, steam or organic molecules.

The semi-classical three step model [3] gives a simple picture of the mechanism of HHG:

- an electron leaves the atom with tunnel emmission induced by the strong laser field
- the electron accelerates in the electric field away from the parent ion and back when the sign of the field swaps

- the electron returns to the ion and recombines, and its kinetic energy is converted to the generation of an XUV photon

One of the most popular quantum mechanical models of HHG is called strong-field approximation (SFA) [4, 5], describing a single atom (without any interaction) and its emitted electron, also known as the single atom response. The model uses more approximations, e.g. calculating with only one electron leaving the ground state (that never depletes). The electron interacts with the laser field only (neglecting the effect of the ion) and recombines when arriving back to the parent ion. The model considers only the electrons that do return.

There are other possible ways to calculate the HHG process, e.g. the time-dependent Scrödinger equation, that uses less approximations, and follows the electron wave packet in vacuum. While providing more precise results [6], this method needs more processor time and memory space to run. I used the SFA because of faster calculations that could be performed with personal computers.

To study HHG experimentally, one needs a vacuum chamber, since the generated XUV light is absorbed and chirped in air. Then, we need to focus the laser beam close to a gas jet or cell, where the process described above occurs. The generated XUV beam leaves the medium in the same direction as the infrared with orders of magnitude lower intensity: a typical HHG conversion efficiency is around 10^{-6} [7]. To avoid damaging the detector, a common practise is to attenuate the infrared beam with metallic filters, a thin layer of aluminium absorbs most of the photons below 20 eV energy.

The result of HHG is usually a single attosecond pulse (SAP), or a train of atto-pulses. Out of these two the case of isolated pulses has more potential, since it is difficult to reconstruct the effect of each atto-pulse in a train upon excitation of the studied system. A common method to generate SAPs is to use a short

enough driving pulse.

Light-wave synthesizer

The so-called light-wave synthesizer device [8], that is able to generate sufficiently short and well shaped laser pulses, opened up new possibilities in femtosecond laser technology and attoscience. To synthesize laser pulses, an input pulse is used that is spectrally broadened in a capillary, including frequency components from the near infrared through the visible regime until near ultraviolet as a supercontinuum white light. This pulse is split to spectral ranges with the use of beam splitters. These spectral channels are separately modified, and then recombined to form a short pulse at the end. With this method, a much shorter laser pulse duration is achievable than with chirped mirrors or optical grating compressors. The output has a frequency range of more than 1.5 octaves, which means that the device is capable of generating pulses shorter than the optical period. This was eventually proven by attosecond streaking measurements [9].

The developers of the device are using four spectral channels, that made it possible to generate an optical pulse as short as 380 as that they used to study the bound states of krypton atoms [10].

The short duration of the output pulse and its easy power scalability makes it a potential driver pulse for high harmonic generation. I aimed to study the question, what kind of attosecond pulses can be created using the light-wave synthesizer device. Toward achieving this idea, I prepared a genetic optimization program code and used it to search for different attopulse shape goals within the parameter space of the synthesizer.

Summing up the details about the synthesizer device and HHG process, we can conclude, that the device is capable of creating SAPs. Additionaly, I studied, what are the properties and limitations of this possibility.

I was inspired by pump-probe experiments to use this framework to study the possibility of creating double attosecond pulses. The first pulse excites the system under study, while the second one probes the system with any spectroscopic or particle detection method. To have full control as in a pump-probe measurement, my goal was to make the time delay between the pulses tunable. I aimed to use the synthesizer model to generate these double pulses and study their properties.

During my simulations I also investigated SAPs that has tunability in the central frequency. The simplest case is to keep the parameters of the synthesizer constant and replace a filtering multilayer XUV mirror to achieve tuning by selecting different parts of the HHG spectrum for creating an attopulse. I tried also to generate SAPs with different central frequency when running the simulation with a fixed setting of the beamline.

HHG with cluster as medium

The first studies in physics about clusters dates back to the '40s and received special attention since the '80s. A cluster means atoms (or molecules) attached to each other with some physical bonds. It is also possible to have different particle species in a cluster.

Because of the size of clusters, they are on the limit of atomic (or molecular) and bulk solid materials, which represents a motivation for research, namely to find the transition between microscopic and macroscopic properties. Clusters are usually metastable, which means that they decompose by themselves to smaller clusters or atomic/molecular constituents. The characteristic decomposing times depend on the material and the type of bonding.

The atoms of noble cases bond with van der Waals forces and noble gas clusters are potential media for the HHG process. It was published that cluster media are more efficent than mono-

mers when generating XUV light using the same backing pressure values [11–13].

Some studies claim that the cutoff energy (the highest XUV photon energy generated, from where the spectrum decays exponentially) is increased also when using clusters instead of monomer gases [12, 14], while other sources claim the opposite [15, 16], therefore this phenomenon is not clarified yet. There is a numeric result that claims the cutoff value to be $I_P + 8U_p$ [17], where I_P is the ionization potential (the amount of energy to ionize the electron closest to vacuum level) and $U_p = \frac{e^2 E_{\mathrm{dr}}^2}{4 m_e \omega_{\mathrm{dr}}^2}$ is the so-called ponderomotive potential (e and m_e are the charge and the mass of the electron, E_{dr} and ω_{dr} are the amplitude and the angular frequency of the driving field), but this has not been clearly verified by experiments [12, 14].

For cluster media, the cutoff frequency increases in a steeper way than for monomer gases [18].

Taking these studies into account, clusters are not only alternative media for gas HHG, but has clear advantages against monomers through the properties of the generated XUV light that represents the motivation to study this subject further. The mechanism of the process is not explained either; in this regard the main question is, where the electron can recombine. The literature lists three possible mechanisms of the process [19]. We call atom-to-itself recombination when the electron can recombine on the parent ion only. If the charge state delocalizes that increases the potential volume to recombine (the *neighbours* are added), then we call this atom-to-neighbour recombination. If this delocalization happens to occur for the whole cluster, even circularly polarized light could generate higher harmonics; this is then called cluster-to-itself recombination.

To acheive the goal of investigating the recombination question, I had a setup at my disposal suitable for high harmonic generation containing an amplified femtosecond laser system and

a vacuum chamber including an XUV spectrometer with a toroidal grating and an XUV detector. I experimentally measured the XUV signal decay depending on the ellipticity of the driver field for different cluster sizes (gas and backing pressure). I determined the characteristic decay length ($\varepsilon_{\mathrm{th}}$ threshold ellipticity) value for each cluster size. I aimed at finding out the mechanism of cluster HHG recombination based on these experimental results.

Results

By studying the above presented problems within the physics of high harmonic generation, in line with my goals, I claim to have achieved the following new scientific results:

1 I created the theoretical model of an existing light-wave synthesizer device to study its capability of generating attosecond pulses. I developed a unique genetic algorithm for the optimization of 14 parameters. I proved that with this method the XUV radiation can be optimized and the simulation can generate double attosecond pulses with a tunable delay as result.

2 I improved the genetic algorithm and fine-tuned it for the high harmonic generation process to reach a convergence at least three times faster than the standard algorithm. I defined new, non-trivial fitness functions that led to the generation of the shortest single attosecond pulses. Using these, I determined the length and expected properties of the shortest achiavable attosecond pulse created with the light-wave synthesizer device.

3 I proved that it is possible to generate a wide enough XUV spectrum using the light-field synthesizer device to generate attosecond pulses with tunable central frequency between 100 and 177 eV by proper spectral filtering.

4 I experimentally studied the mechanism of higher order harmonic generation on noble gas clusters. I determined the XUV decay as a function of driver (infrared laser pulse) ellipticity for several cluster sizes. By analysing these experimental data, I showed that the positive charge of the parent ion does not delocalize, therefore the electron can recombine only on the parent ion.

Thesis related publications

[T1] E. Balogh*, B. Bódi*, V. Tosa, E. Goulielmakis, K. Varjú, and P. Dombi, "*Genetic optimization of attosecond-pulse generation in light-field synthesizers*," Phys. Rev. A **90**, 023855 (2014).
*Equal contribution of authors

[T2] B. Bódi, E. Balogh, V. Tosa, E. Goulielmakis, K. Varjú, and P. Dombi, "*Attosecond pulse generation with an optimization loop in a light-field-synthesizer*," Opt. Express **24**, 21957 (2016).

[T3] B. Bódi, M. Aladi, P. Rácz, I. B. Földes, and P. Dombi, "*High harmonic generation on noble gas clusters*," Opt. Express **27**, 26721 (2019).

Additional publications

[P1] P. Dombi, P. Rácz, and B. Bódi, "*Surface plasmon enhanced electron acceleration with few-cycle laser pulses*," Laser Part. Beams **27**, 291 (2009).

[P2] N. Tsatrafyllis, B. Bergues, H. Schroeder, L. Veisz, E. Skantzakis, D. Gray, B. Bódi, S. Kühn, G. D. Tsakiris, D. Charalambidis, and P. Tzallas, "*The ion microscope as a tool for quantitative measurements in the extreme ultraviolet*," Sci. Rep **6** 21556 (2016).

[P3] A. Nayak, I Orfanos, I Makos, M. Dumergue, S. Kühn, E. Skantzakis, B. Bódi, K. Varjú, C. Kalpouzos, H. I. B. Banks, A. Emmanouilidou, D. Charalambidis, and P. Tzallas, "*Multiple ionization of argon via multi-XUV-photon absorption induced by 20-GW high-order harmonic laser pulses*," Phys. Rev. A **98** 023426 (2018).

[P4] P. A. Carpeggiani, B. Bódi, E. S. Skantzakis, D. Charalambidis, P. Tzallas, M. Arnold, L. Chipperfield, J. W. G. Tisch, J. P. Marangos and A. Zaïr, "*Temporal characterization of high harmonics emitted from the long quantum path*," New J. Phys, prepared for submission (2020).

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