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High-order harmonic generation from noble gas clusters

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Summary of the PhD thesis

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Introduction

At modest laser intensities atomic dipole moment is induced with the interaction of light which creates a radiation. The emitted radiation contains low-order harmonics of the laser frequency with exponentially decreasing intensity. At about 10^{13} W/cm^2 light intensity due to ionization processes high-order harmonics can also be generated approximately with the same strength.

In case of the optical high harmonic generation (HHG) a short laser pulse is focused into the gas target resulting in odd high harmonics $(\omega_\mathrm{L}, 3\omega_\mathrm{L}, 5\omega_\mathrm{L}, \ldots)$ in the output beam up to $> 300\,\omega_\mathrm{L}$. This technique is the simplest method to generate coherent extreme ultraviolet (EUV) sources that cover the whole spectral range from visible to keV energy x-ray, even of subfemtosecond (1 fs $= 10^{-15}$ s) pulse durations.

The explanation of the phenomenon is that the atomic Coulomb potential is distorted by the high electric field in atoms, molecules or clusters, so the electrons become quasi free by optical tunnel ionization. They oscillate in the laser field around the ion and then recombine emitting their excess energy in the form of a photon. The photon emission occurs twice during the laser period, close to the positive and negative maximum of the field. This temporal periodicity causes the appearance of discrete spectral lines at the odd multiples of the laser frequency with $2\omega_L$ distance. Hungarian researchers showed at first time that the duration of this broadband $(\Delta\omega)$ radiation can be in the range of $1/\Delta\omega \sim 10^{-18}$ s (1 as) in accordance with the Fourier transformation.

Attosecond physics started with the discovery of high harmonic generation process. It is possible to investigate the different intermolecular and atomic processes on a very short time scale with such pulse. In the past three decades various experimental arrangements have been used to improve the conversion efficiency, which is typically 10^{-7} and limits the applications. For example attosecond pulses in the water window range (2.3-4.4 nm)

- where the biological samples and cells become transparent - provide unique possibilities for spectroscopy and microscopy.

A possible way to produce a laser-gas interaction is the injection of gas through a pulse valve into the vacuum. In case of high backing pressure gas jets cluster particles can be formed. They consist of several thousand-ten thousand atoms with Van der Waals bond between them. Using of noble gases is the most common and clusters are effective targets for the process of HHG.

Scientific background

Attosecond pulses (100-200 as pulse duration) can currently only be produced by HHG and low pulse energy sets a limit for the applications. In case of gas harmonics the main reason is the limited applicable laser intensity, because ionization decreases the efficiency of the HHG process above a certain threshold. Therefore nowadays the main research effort is the generation of high energy and preferably isolated attosecond pulses.

HHG experiments in gas clusters started in the second half of the nineties. There was an assumption that this medium is a perfect nonlinear target, a transition between the solid and gas phases. Harmonics from clusters were assumed to possess the ability of solid state harmonics that there is no ionization limit in the plasma. With the using of clusters higher orders and conversion efficiency should be achieved.

However cluster jets haven't been often used due to several things: The mechanism of HHG from clusters as many particle systems is a complicated physical problem with controversial experimental and theoretical results. Due to the high atomic density of the clusters an electron may recombine to the neighboring ions with a finite probability resulting in incoherent radiation. At the same time it is possible that the wave function is partially delocalized over the whole cluster to which the electrons may coherently recombine.

The different cluster sizes in the medium make the experimental separation of harmonics from atoms and clusters difficult. Moreover, generating of a homogeneous and/or long cluster target from an experimental point of view is challenging.

Neglecting propagation effects the laser-cluster interactions promised higher photon energy and conversion efficiency during the HHG process, however macroscopic properties and thus the experimental implementations haven't confirmed these results. Additionally the laser intensity couldn't be increased to the desired level in cluster media, because the ionization limit is similar to the atomic media. However recent experiments showed that changing the initial gas temperature and thus the size of the clusters, in a certain size range the clusters can really provide higher conversion efficiency.

Clusters - targets for HHG

When the valve is opened the expanding gas jet into the vacuum is an isentropic flow. The gas pressure and temperature rapidly decrease while the gas condenses and clusters are formed. Their size can mainly be changed with the backing pressure or using of gases with different condensation efficiency. Also the use of various valve geometries has a significant effect. It is important to minimize the shot-to-shot fluctuations during the operation of the valve in order to obtain similar density distributions. Namely the nonlinear interactions - such as the high harmonic generation - are very sensitive to a slight change of the initial conditions. This is especially true for cluster media which exhibit a strong nonlinear behavior in strong laser fields.

Clustering can be characterized by one geometrical nozzle parameter. The de Laval geometry - where a converging section before the throat is followed by a divergent section

- is useful in the generation of supersonic gas jets. For conical geometry the diameter of the throat and the expansion half angle are relevant. The simplest case is the cylindrical nozzle, in which the average cluster size is mainly determined by the diameter of the orifice. At certain cases it can be advantageous to use nozzles having a rectangular orifice which can produce a shorter interaction length and can provide a larger cluster size than conical nozzles.

Conical nozzles have been studied in many articles, but the de Laval geometries received less attention. On the one hand this is due to the complicated fabrication which requires an electro-erosion technique, on the other hand in case of de Laval geometry a hollow spatial cluster distribution can be measured in the plane perpendicular to the nozzle axis. However according to simulations even the de Laval nozzles with some geometrical ratio can provide an approximately flat-top density distribution providing an appropriate target for interactions. The nozzles used in different publications have slightly different shapes, which can have a significant influence on the properties of gas flow and high harmonics.

Aims

The significant increase of the conversion efficiency with the use of cluster targets remained a challenge for the future, therefore one of the aims in my work was the detailed experimental analysis of the gas cluster HHG process. My research work was carried out in the hELIos laser laboratory of the Wigner Research Centre for Physics.

The main goal was to investigate the properties of the cluster media as an effective HHG target and to answer the following questions as accurately as possible. What are the advantages of clusters in HHG comparing to atomic media? How high harmonic beams can be used to determine cluster properties? What information can be obtained from the harmonic spectra about cluster behavior in high laser field?

Earlier published experiments did not always provide reproducible results, and also in own experiments we found that different nozzles gave different results. So it was necessary to control accurately the experimental conditions. Therefore my aim was the building of a diagnostic method as well that is convenient for the investigation of the cluster distribution in jets and for the detection of the differences between nozzles.

In the first part of the thesis the properties of the clustering gas jet medium (cluster size and -density) were determined. In the second part the spectral properties and conversion of high harmonic beams from clusters were investigated.

Applied methods

The laser source was a titanium-sapphire amplifier (Legend Elite HE-USP manufactured by Coherent Inc.) providing 40 fs pulses with 4 mJ energy. I actively participated in the construction of the high harmonic generation setup. I have built up the monitoring and delivering of the laser pulses with the using of optical- and optomechanical elements. The vacuum system consists of two main parts, an interaction- and a detection chamber. In the interaction chamber I constructed the gas injection system. The precise characterization of gas jets was performed by the laser Rayleigh scattering diagnostics. A vacuum ultraviolet spectrometer was used to investigate the UV radiation as a result of the high intensity interaction. I fitted the spectrometer to the interaction chamber and improved the method of screening the infrared light from the spectrometer.

Laser Rayleigh scattering diagnostics. I took part in the construction of a laser Rayleigh scattering diagnostics built for the investigation of spatial-temporal distribution of noble gas clusters generated in gas jets. I designed a linear displacement mechanism for the valve that allows the precise analysis of clustering for different distances from the nozzle and for different nozzle geometries. The arrangement can give a picture about the two-dimensional distribution of the cluster sizes in different cross sections of the jet, furthermore we can compare the clustering for the different nozzles.

The diagnostics allows a relatively simple and effective measurement of the cluster formation. A continuous laser beam was focused into the gas jet. The scattered light from clusters was monitored in 90° arrangement with a photomultiplier tube and a CCD camera. Thus it was possible to detect a time-resolved, spatially integrated Rayleigh scattering signal and to investigate the spatial cluster formation.

Vacuum ultraviolet spectrometer. The spectral properties of high harmonic beams are investigated in the spectrometer. A reflective optical grating separates the wavelength components, which are imaged onto the surface of a microchannel plate (MCP) detector. The MCP amplifies the low intensity ultraviolet radiation and converts it to visible light. On its surface the channels work like the photomultiplier tubes allowing the spatial resolution of the radiation. The electron collisions into a fluorescence phosphor screen cause a flash and the spectrum is imaged into a CCD camera.

Nanoplasma model. During the laser-cluster interaction the clusters ionize and nanoplasma is generated. The characteristic time for cluster explosion in the laser field is the function of several parameters including laser intensity. At the used laser intensity and pulse duration it can be assumed that the nanoplasma holds together for the time scale of the interaction. Then this nanoplasma medium determines the macroscopic properties of high harmonic generation. The optical properties of nanoplasma and its dispersion contribution was estimated based on the classical Drude model by J.W.G. Tisch.

I investigated the average cluster size and -density in the interaction volume correlated to the spectral shift of the harmonic orders in the frame of the nanoplasma model. The model can become a diagnostic tool for analyzing the nanoplasma dynamics and can help to create a consistent theory in the topic of HHG from clusters.

Results (list of theses)

T1. The spatial and temporal cluster size distributions was determined with Rayleigh scattering diagnostics. I could show that the spatial density profile of the gas jet depends sensitively on the position of gas jet to the laser axis. Different density modulations were detected in the case of the four investigated nozzles. Thus, I could experimentally verify the simulation results that the de Laval nozzles can have a hollow or a flat-top profile depending on the geometry, from which the flat-top profile is required for the laser-gas interactions. [S4].

T2. I could demonstrate that the cluster sizes scale with different power functions for the different nozzle geometries in the function of the backing pressure. The absolute cluster sizes for the four nozzles could be determined. The material dependence of the cluster formation in argon and xenon gases has interesting features. In the case of certain nozzles the average cluster sizes strongly differed from the values given by the widely used Hagena formulas. I could prove that the simple condensation parameter for the describing of material dependency is insufficient and the parameter can depend on the nozzle geometry, too. Accordingly the Hagena formula and the single parameter for the material dependency is not sufficient for the characterization of a laser-cluster interaction. [S4].

T3.a The conversion efficiency to high harmonics were investigated. Several noble gases with different condensation parameters were used for exploring of the effect of the differing cluster sizes. I could demonstrate that clusters can increase the conversion to high harmonics, but the ionization threshold doesn't increase considerably compared to the atomic gases. These results are in accordance with earlier experiments. [S1, S2].

T3.b The observed significant spectral broadening of the harmonic orders with increasing backing pressure in the largest cluster sized xenon jet is an evidence for the presence of clusters. I have demonstrated that the blueshift and broadening of harmonics can provide a quasi continuosly tunable, coherent EUV radiation, which can also be used as a seed pulse for x-ray free-electron lasers. [S1].

T4. It is shown for the first time that at relatively low laser intensities, when the free electron density is low, an increasing redshift of harmonic orders occurs with increasing backing pressure. This shift is much smaller than the generally dominating free electron induced blueshift, and it could only be observed only in xenon which gives the largest clusters.

This effect can be explained with the help of nanoplasma model, which describes the optical properties of the ionized clusters. Estimating the dispersion of the ionized cluster medium during the propagation of high harmonic beams I showed that this simple model can explain the redshift of harmonic orders with increasing backing pressure, with increasing cluster size. [S2].

T5. The redshift according to the nanoplasma model is the function of the properties of cluster target (cluster size and cluster density). As a consequence of it I could show that the shift allows one to investigate cluster parameters and nanoplasma dynamics in the medium. The possible range of cluster parameters were determined and I could estimate the cluster density in the medium using the cluster size given by the Rayleigh scattering diagnostics. [S3, S4].

Publications related to the thesis

[S1] **M. Aladi**, and I.B. Földes, „High harmonic generation from atom clusters", Journal of Physics: Conference Series 508, 012016 (2014).

[S2] **M. Aladi**, I. Márton, P. Rácz, P. Dombi, and I.B. Földes, „High harmonic generation and ionization effects in cluster targets", High Power Laser Science and Engineering 2, e32 (2014).

[S3] **M. Aladi**, R. Bolla, P. Rácz, and I.B. Földes, „Noble gas clusters and nanoplasmas in high harmonic generation", Nuclear Instruments and Methods in Physics Research B 369, 68 (2016).

[S4] **M. Aladi**, R. Bolla, D.E. Cardenas, L. Veisz, and I.B. Földes, „Cluster size distributions in gas jets for different nozzle geometries", Journal of Instrumentation 12, C06020 (2017).

Conference presentations related to the thesis

[E1] **M. Aladi**, and I.B. Földes, „High Harmonic Generation from atom clusters", Plasma Physics by Laser and Applications, Lecce, 2013.

[E2] **M. Aladi**, and I.B. Földes, „High harmonics generation and ionization effects in cluster targets", Conference on High Intensity Laser and attosecond science in Israel, Tel-Aviv, 2013.

[E3] **Aladi M.**, „Magas harmonikusok keltése ultrarövid lézerimpulzusokkal", Fizikus Doktoranduszok Konferenciája, Balatonfenyves, 2014.

[E4] I.B. Földes, and **M. Aladi**, „High harmonics from noble gas clusters", 17th International Congress on Plasma Physics, Lisbon, 2014.

[E5] **M. Aladi**, R. Bolla, and I.B. Földes, „Noble Gas Clusters and Nanoplasmas in High Harmonic Generation", Photon and fast Ion induced Processes in Atoms, Molecules and Nanostructures, Debrecen, 2015.
[E6] **M. Aladi**, R. Bolla, and I.B. Földes, „The effect of clusters on the generation of high harmonics", 42nd EPS Conference on Plasma Physics, Lisbon, 2015.
[E7] **M. Aladi**, R. Bolla, and I.B. Földes, „High harmonic generation from clusters and monomers", Plasma Physics by Laser and Applications, Frascati, 2015.
[E8] **M. Aladi**, R. Bolla, and I.B. Földes, „Cluster size distributions for different nozzle geometries in noble gas jets", 2nd European Conference on Plasma Diagnostics, Bordeaux, 2017.