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# The Effect of Clusters on the Generation of High Harmonics

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# The effect of clusters on the generation of high harmonics

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## 1. Introduction

High-harmonics as sources of attosecond pulses can be efficiently generated from cluster targets [1]. The problem of the use of cluster targets is that even for clusters ionization sets an upper limit for the applicable laser intensity [1,2] and thus, for the total harmonic energy. On the other hand, the generated high harmonics may become appropriate diagnostic tools for the internal structure of clusters.

The mechanism of high harmonic generation from clusters is still a subject of intense debates. A recent model of Ruf et al [3] suggests tunnel ionization from a partly delocalized electron wave function and recombination to this wave function, i.e. to the cluster itself.

Experiments were carried out using different noble gases from commercial valves with 1 to 12 bar backing pressures. He and Ne provided atomic gas targets, Xe was a source of clusters and Ar was intermediate depending on the pressure. The observation that in the presence of clusters the intensity of harmonics increases steeper with increasing pressure is in agreement with previous experiments [1] and it is a demonstration of the presence of clusters which is confirmed by Rayleigh scattering. High harmonics spectra show several interesting properties. Increasing the laser intensity results in a blue shift of the harmonics due to propagation effects both for atomic gases and for clusters. This is large enough for generating a continuously tuneable coherent light source in the EUV [2,4,5].

At modest intensities however - when free electrons in the interaction domain are rare – a spectral shift of opposite signature could be observed which increased with increasing pressure. Whereas the above-mentioned blue shift is attributed to the free electrons in the interaction region, this red shift is attributed to the nanoplasmas inside the clusters [5]. From the observed red shift we can even estimate the size and the density of the nanoparticles, thus it may even serve as a diagnostics of the clusters.

## 2. Experimental

A Ti:sapphire laser system was used delivering 4 mJ pulse energy of 40 fs pulse duration centered at a wavelength of 805 nm at 1 kHz repetition rate. The initial beam diameter was

9 mm. The linearly polarized laser beam was focused into the vacuum chamber using a plano-convex spherical lens of 30 cm focal length. Gas jets using a commercial valve (Parker series 9) were used with an additional nozzle. The density was characterized earlier using x-ray shadowgraphy [6]. The valve had an orifice of 0.99 mm diameter and it was opened for 1 ms duration. The backing pressure could be varied between 1 and 12 bar and the laser beam propagated 1 mm in front of the conical nozzle. The nozzle was situated some millimeters in front of the focal plane, thus the generating gas medium was in the diverging beam, preferring the short trajectories of electrons for the generation of harmonics.

A toroidal holographic grating (Jobin Yvon, 550 lines $mm^{-1}$) imaged the target onto the cathode of a microchannel plate (MCP) with a phosphor screen and the spectra were taken by a CCD. It is to be noted that the reflectivity of the grating decreases when the wavelength is shorter than 30 nm.

### 3. Results and Discussion

High harmonics were observed up to the $45^{th}$ order with laser intensity increasing up to $1\text{-}2 \cdot 10^{14}$ W/cm$^2$ when saturation occurs due to the ionization of the media. Note that due to the relative high total gas density at the interaction range it occurs somewhat earlier than for low density gases. This saturation occurs both for atomic gases as He and Ne and for cluster targets as Xe. Fig. 1 illustrates the pressure dependence of the efficiency of harmonics in a typical case of 25ω.

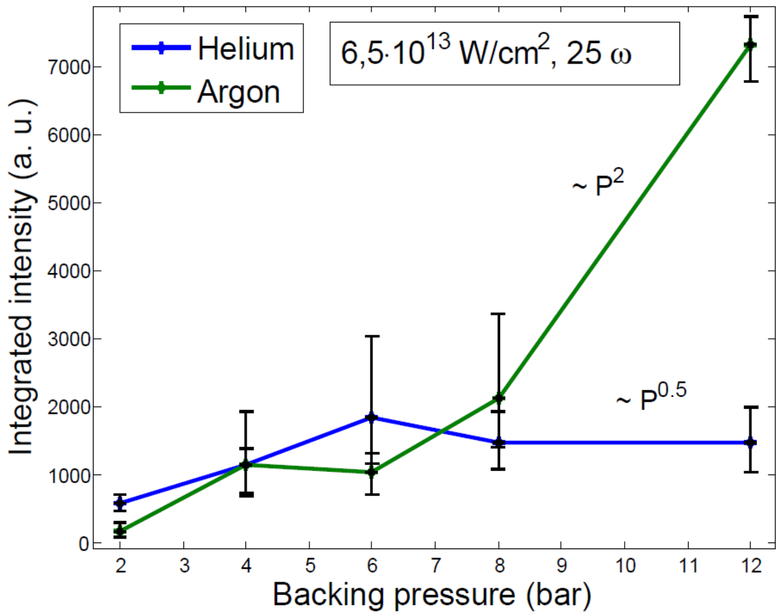


Fig. 1. Pressure dependence of the 25th harmonic in He and Ar for an intensity of $6.5\times10^{13}$ W/cm$^2$.

The conversion to harmonics shows that whereas in the atomic He the increase of harmonics intensity with pressure is modest and it saturates probably due to the increasing ionization, in the case of Ar a sharp increase sets in when – according to the Hagena-scaling [7] the clusters will consist of more than 1000 particles. This sharp increase is a well-known feature of harmonics generation from clusters [1].

An interesting phenomenon was observed in the case of relatively low, $6.5\times10^{13}$ W/cm$^2$ intensity in which case there are still not many free electrons in the interaction range. Harmonics spectra show a red shift of the harmonics, especially for Xe, in which case the clusters are relatively large. Increasing the pressure and consequently increasing the size of the clusters, this frequency shift reaches $\Delta\omega/\omega\approx10^{-2}$ for backing pressures above 8 bar. This corresponds – according to the Hagena-scaling [7] – to clusters consisting of more up to $10^5$ atoms. The more recent data of Dorchies et al [8] suggests – especially for the largest applied pressures - a slightly modified scaling which gives clusters consisting of $\sim6\times10^4$ atoms.

We can even use the observed wavelength shift to the independent estimation of the size and density of the clusters. According to Tisch [9] the phase mismatch due to the nanoplasmas in the clusters can be estimated as

$$\Delta k_{nanoplasma}=\frac{q\cdot 2\pi}{\lambda_\omega}\left(\sqrt{1-\frac{4\pi n_e r^3 n_{cl}}{3n_{qcrit}-n_e}}-\sqrt{1-\frac{4\pi n_e r^3 n_{cl}}{3n_{crit}-n_e}}\right).$$

Here $q$ is the order of the given harmonic, $\lambda_\omega$ is the laser wavelength, $n_e$ the electron density, $n_{cl}$ the cluster density, $r$ is the radius of the cluster (or nanoparticle), $n_{crit}$ and $n_{qcrit}$ are the critical densities for the laser and the given harmonic, respectively.

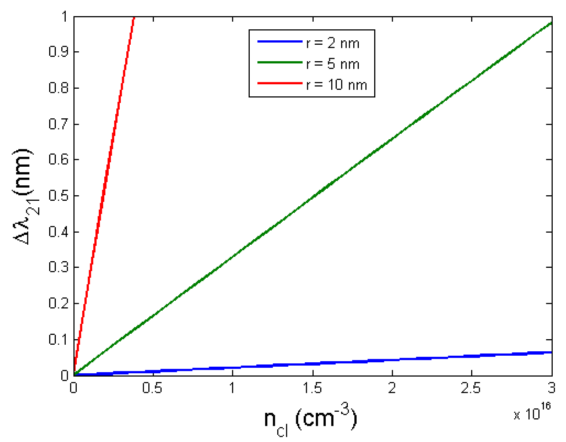


Fig. 2 Calculated wavelength shift of the 21st harmonic radiation as a function of cluster density with different assumed cluster radii..

Figure 2 compares the calculations with the experimental results as the wavelength shifts are illustrated depending on the density and radius of the cluster. We compare the calculations with the experimental results obtained for Xe with 12 bar backing pressure. The experiments gave a red shift of *0.5 nm* for the 21st harmonic in this case. Clearly, a very small radius of *~2 nm* would require very high cluster density to reach the observed wavelength shift. In the case when the radius of the cluster is *10 nm*, then it consists of - taking account the van der Waals binding length for it to be 0.273 nm [10] – 60000 atoms in agreement with the scaling law [8]. In this case the cluster density is $1.9 \times 10^{15}\ cm^{-3}$ which corresponds to the earlier observed total atomic density of $10^{19}\ cm^{-3}$ [6]. If however a cluster radius of *5 nm* is assumed then the cluster consists of ~6000 atoms which is an order of magnitude less.

Our preliminary Rayleigh-scattering measurements using a 530 nm laser beam gave similarly, only some thousands of particles for the cluster sizes. Summarizing the results it can be concluded that measuring the detailed spectral properties of the generated harmonics gives not only a proof for the existence of nanoplasmas in the clusters but it also serves as an additional diagnostics for their structure.

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