High harmonic generation from atom clusters

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Abstract. High harmonics generation driven by 40 fs laser pulses is investigated in three different noble gases, both in gas and cluster regime depending on gas type and backing pressure. Pressure dependence of the harmonic intensity shows different scaling laws in case of cluster formation compared with monomer gas. Both the higher harmonic yield and the stronger spectral broadening of harmonics in xenon gas show the presence of clusters in the jet. The central wavelength of harmonics has a blue shift increasing with increasing laser intensity which is a consequence of free electron contribution to the phase matching.

1. Introduction
One of the main objects in the process of high-order harmonic generation (HHG) is to increase the conversion efficiency. In most of the cases atomic gas is used as target in form of gas jet or static gas cell. Gas jets can be generated by commercial solenoid valves in which case clusters may be formed in this high backing pressure jet due to the rapid cooling by the adiabatic expansion into vacuum. Clusters are promising targets for HHG and in our experiments we investigate characteristics of cluster harmonics compared with harmonics from monomer gases. In the case of gas harmonics the laser intensity limit is a considerable problem, because ionization mitigates the HHG process. Clusters are considered as suitable medium for HHG, because this medium forms a transition between gas and solid and it gives a higher conversion efficiency to harmonics. Ionization of the clusters however sets a limit to their application, too. The purpose of the present work is to determine the limits of applicability of cluster targets compared with atomic medium.

2. Experimental condition
High harmonics can be produced simply by focusing laser light into a gas target. We investigate the intensity of harmonics depending on laser intensity and backing pressure in the case of three noble gases.

2.1. Experimental setup
In the experiment a Ti:sapphire laser beam was used with 800 nm central wavelength, 1 kHz repetition rate. The pulse duration is 40 fs with 4 mJ pulse energy. The gas jet target used in the present experiment was characterized earlier [1]. The generated harmonic beam is characterized using a toroidal grating, which images the target onto a microchannel plate detector (MCP). The spectrum appears on the phosphor screen of the MCP and it is imaged onto a normal CCD.
2.2. Noble gas as cluster source

Noble gases as helium, argon, and xenon were used as targets. In a gas-cluster medium, the average number of atoms in one cluster can be featured by means of the semi-empirical Hagena-parameter:

\[ \Gamma^* = k_h \cdot P_0 \cdot (0.74 \cdot d)^{0.85} \cdot T_0^{-2.29} \]

with \( N = 33 \cdot \left( \frac{\Gamma^*}{1000} \right)^{2.35} \),

where \( k_h \) is the empirical condensation parameter depending on the gas, \( P_0 \) is the backing pressure, \( d \) is the nozzle diameter, and \( T_0 \) is the initial gas temperature. For xenon \( k_h = 5500 \), for argon \( k_h = 1650 \), and for helium \( k_h = 4 \). Therefore, xenon is the most convenient gas for cluster formation, whereas in helium no clusters are expected and argon is an intermediate gas for cluster presence.

3. Observations

3.1. Intensity dependence

Odd harmonics were observed up to the 45th order. The intensity of harmonics was investigated depending on the laser intensity whereas the laser energy was kept constant and the position of the gas jet was varied relative to the focal plane. In Figure 1, we can see that the 15\( \omega \) harmonic yield in xenon is nearly an order of magnitude higher than that in the other gases at 6 bar backing pressure. It can also be seen that the conversion to harmonics is stronger in argon than in helium. Increasing the intensity above \( 10^{14} \) W/cm\(^2\), the conversion saturates and then decreases. It is similar for each gas, independently from the eventual cluster formation. Although the conversion efficiency is the highest for xenon where clusters are formed, on the other hand, due to the lower ionization potential for xenon - independently from the cluster formation - the conversion starts to decrease even at lower laser intensity than for argon. Thus it is confirmed that ionization limit of harmonics generation is independent from cluster formation.

![Figure 1. Laser intensity dependence of 15\( \omega \) harmonic signal at 6 bar backing pressure.](image1)

![Figure 2. Backing pressure dependence of 27\( \omega \) harmonic signal at 6.5 \( \cdot 10^{13} \) W/cm\(^2\) laser intensity.](image2)

3.2. Backing pressure dependence

Back pressure dependence of the harmonic intensity at fixed laser intensity shows the effect of cluster formation. Figure 2 illustrates it for argon for a special example of the 27\( \omega \) harmonics, in which case at lower pressures when no clusters are formed the conversion efficiency is low. A different scaling law can be seen above 6 bar due to cluster formation. For helium, the conversion efficiency slowly increases with pressure, whereas for argon, where clusters are expected an approximately square-law dependence can be observed. This dependence is different from some earlier observations,
where $P_0^2$ dependence was observed for atomic gas and $P_0^3$ dependence for clustered gas [2]. In the present experiment just cluster targets show a $P_0^2$ dependence. Note that in the early experiments of Donnelly et al. [2] a longer, 160 fs pulse duration was used, which duration exceeds the characteristic time for Coulomb explosion of the cluster.

3.3. Spectral broadening
Our observation confirms the earlier results that the spectral widths of harmonics are larger for clusters than for monomer gases. Figure 3 illustrates that at 12 bar backing pressure harmonic components are significantly broader in xenon than in helium. This spectral broadening is increasing with increasing pressure.

![Figure 3](image1.png)

Figure 3. Spectral broadening of harmonic spectrum at 12 bar backing pressure in the case of helium (top) and xenon (bottom).

3.4. Spectral blueshift
Harmonics spectra are illustrated at three different laser intensities for helium and xenon. In Figures 4 and 5 the backing pressure was 2 bar. Clearly the harmonic yield is much higher in xenon (Figure 5) than in helium (Figure 4). Argon with 12 bar backing pressure shows similar spectra than xenon at 2 bar.

![Figure 4](image2.png)

![Figure 5](image3.png)

Figure 4. Integrated spectrum at 2 bar backing pressure for helium target at three different laser intensities.

Figure 5. Integrated spectrum at 2 bar backing pressure for xenon target at three different laser intensities.

It is evident from these spectra that the central wavelength of harmonics has a frequency shift toward shorter wavelengths. This ionization effect is known as frequency blueshift [3], which is effect of the free electrons onto the propagation and thus onto the phase matching of the harmonics with the incoming beam. The blueshift is illustrated quantitatively in case for the $21\omega$ radiation in Figure 6.
4. Conclusion

It was demonstrated through a series of experiments that clusters may enhance the conversion efficiency to high harmonics in a limited range of laser intensities, below the ionization limit. On the other hand separating the contributions from monomers and clusters is not simple [4] because the order of magnitude difference between the clustered xenon and the monomer helium is only partly caused by cluster formation, partly by the generally higher efficiency for xenon. On the other hand the steep increase of harmonics conversion efficiency in argon when cluster formation starts at higher pressures gives a clear evidence for the beneficial effect of clusters on harmonics generation. Additionally the spectral broadening of harmonic spectrum can refer to the existence of clusters in the gas jet as well. On the other hand the observed spectral blueshift is caused by the effect of free electrons onto the phase matching, i.e. not a cluster-effect. This blueshift gives a possibility for tuning the wavelengths of harmonics with the laser intensity.

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