Comparative study of low-order harmonic generation in gas and cluster media at Ti:Sa femtosecond laser intensity up to 2000 TW/cm²

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Abstract. In comparative experiments, it was found that the maximum efficiency of third harmonic generation (THG) in cluster nanoplasma was ~9x10⁻⁵ and was approximately 4 times less effective than the case of THG in gas under similar experimental conditions. This result is consistent with theoretical calculations and show that THG amplitude in a gas is about an order of magnitude higher than in a cluster medium. The discrepancy between the THG efficiency in gas and clusters obtained theoretically and experimentally is discussed.

1. Introduction

Low order harmonics of intense femtosecond laser in gas and cluster media is of interest for development efficient coherent UV sources with duration of 10⁻¹³-10⁻¹⁴ s. The laser energy conversion efficiency to harmonics in gas depends on its number and for the third and the fifth harmonics it can reach values of ~10⁻³-10⁻⁴ [1]. These sources open the possibility of studying ultrafast intermolecular processes under photoexcitation and photodissociation [2]. Optimization of low order harmonics parameters (efficiency, spatial properties, polarization state, etc.) is important problem for various applications [3]. For instance, fifth harmonic can be amplified in Kr₂ and Xe₂ excimer media which have wide amplification bands and relatively high saturation energies [4]. It should be noted that the photon energy at the frequency of the Ti:Sa laser fifth harmonic coincides with the energy of the 229mTh state that can be used for nuclear isomer excitation [5]. On the other hand, atomic-molecular cluster media are of potential interest due to the following reasons. Clusters of atoms is a perspective nonlinear medium for the generation of short-wavelength radiation as a result of harmonic generation [6]. Theoretical calculations [7] show that the single cluster emits low-order harmonics approximately 2 times more efficiently in comparison a with single atom response. Thus, it can be expected that a cluster jet may have an advantage in the efficiency of generating low harmonics compared to a gas at a
comparable average density of atoms. In the case of gas harmonics, the laser intensity clamping at the level of about 100 TW/cm² is a known problem, since the appearance of electrons due to ionization leads to phase mismatch and restricts the process of generating harmonics. Clusters can be considered as a suitable nonlinear medium for generating harmonics, since this medium is a transition between a gas and a solid and can provide efficient conversion to harmonics, including due to the removal of restrictions on laser intensity. Future studies will determine the actual efficiency of the cluster harmonic yields at higher intensities and the phase matching properties of a dense medium of clusters.

The subject of this work is a comparative experimental and theoretical study of the laser energy conversion efficiency of the THG induced by femtosecond Ti:Sa laser in Ar gas and Ar cluster medium at intensity up to 1PW/cm².

2. Experimental setup and results

The experimental setup is shown in figure 1. The experiment is performed with pulse duration 300 fs (FWHM), 800 nm Ti:Sa laser, which operated at a repetition rate 10 Hz and single pulse energy of 5mJ. The laser beam is focused into the interaction chamber by a fused silica lens (L1) with a focal length of 30 cm. Back pressure inside the vacuum chamber is of 10 mbar.

Argon clusters were produced by the technique based on supersonic gas expansion through the conical nozzle (850 um orifice, 5° half expansion angle) into vacuum. The argon backing pressure was adjusted in the range of 5-30 bars. The estimates made using the Hagena formulas [8] for the given conditions show that the cluster sizes varied within the range of 16-50 nm. The laser beam was focused about 1.5 mm below the nozzle edge. The pulse duration of 300 femtoseconds was taken for two reasons. On the one hand, this allowed avoiding phase self-modulation on the laser beam propagation path. On the other hand, the conditions of resonant nonlinearity in a cluster nanoplasma were realized in a similar way to those that occur for argon clusters with an average size of about 30 nm [9].

At the first stage of our investigations, THG in a cluster argon jet was studied as a function of the laser intensity in the range I≈100–2000 TW/cm². Laser intensity control and THG optimization were provided by moving laser focus relatively to the cluster jet boundary. The cluster jet boundaries were determined by registration the Rayleigh scattering signal of a probing diode laser (λ = 445 nm) on the CCD matrix. The argon backing pressure was 30 bars. It was found that the maximum THG efficiency reach ~9x10⁻⁵ acting laser intensity I≈200 TW/cm². The increase in the laser intensity up to I>1000 TW/cm² under interaction cluster jet is accompanied by the formation of filament (extended plasma channel) and a decrease in the THG efficiency by a factor of ~4. We believe, that the nanoplasma mode of THG turned out to be less effective due to the small number of clusters in the jet (in the argon jet, only ~20% of atoms are in clusters). We found that in case expanding from the supercritical state of the Widom’s delta, the concentration of clusters in the jet (using the example CO₂ clusters) can increase 5-10 times compared to subcritical conditions (to be published). This result allows us to expect that in the case of generation of low harmonics in cluster jets obtained from the supercritical state, the mode of nanoplasma nonlinearity will be most effective. At the second stage of our research, the dependence of the THG efficiency on the argon backing pressure in range of 5-30 bar was measured at optimal laser intensity I≈200 TW/cm². As a result, it turned out that the THG efficiency depends linearly on the argon backing pressure.

In the case of a gas cell filled with argon of pressure 50-300 mbar it was found that the efficiency of THG in the range up to 150 mbar pressures (corresponds to 3x10¹⁸ cm⁻³) varies according to the quadratic law, which is consistent with the calculation results and indicates the presence of phase matching conditions. With a further increase in the pressure in the gas cell, the phase matching was disrupted and the efficiency of THG corresponded to linear growth. The maximum conversion efficiency of THG in the experiment was 5x10⁻⁵ at a gas pressure of 300 mbar (corresponds to 6x10¹⁸ cm⁻³) in the interaction chamber. It should be noted that under interaction of intense near IR picosecond laser pulses with laser induced extended plasma was obtained conversion to THG a comparable efficiency of 10⁻⁴ [10]. The pressure and temperature in the cluster jet is very different.
from that of a gas, so the comparison is made by atomic density (figure 2). In the conducted comparative experiments, it was found that the maximum efficiency of THG in cluster nanoplasma was approximately 4 times less effective than in the case of THG in gas under comparable experimental conditions (intensity and average density of particles per unit volume).

2.1. The interference model
We also carried out comparative theoretical studies of THG in Ar gas and cluster media. Numerical calculations of the THG were performed within the framework of the interference model [11] in which the nonlinear gas medium is simulated as a 1D chain of atoms oriented along the propagation direction of laser field. The single atom responses were calculated in the frame of non-perturbative approach [12]. In the case of a cluster medium, the atoms were grouped in segments corresponding to the average diameter of the clusters, which were separated by vacuum. Comparison of the results of TH calculations with experimental data is shown in figure 2. Experimental and numerical results of THG as a function of the atomic density demonstrate quadratic growth in the low gas pressure region up to $3\times10^{18} \text{ cm}^{-3}$ as a result of phase matching process. Calculations show that a further increase in pressure in the gas cell leads to a drop in the THG amplitude due to destructive interference of THG by different atoms in media. The 1D interference model doesn’t take into account the contributions of atoms located at the periphery of the laser beam, which leads to a discrepancy between experiment and theory. In the case of a cluster jet, numerical calculations of the THG for a cluster jet with an average cluster size in the range of 10-100 nm show that the THG yield corresponded to a power function with an exponent of 1.4. Theoretical calculations show that the THG amplitude in a gas is about an order of magnitude higher than in a cluster medium. The discrepancy between the calculated TH intensity and the experimental data in a cluster jet can be explained by the contribution of non-clustered monomers, which was not taken into account in the calculations.

3. Conclusions
In comparative study, it was found that the maximum efficiency of THG in a cluster jet was $9\times10^{-5}$ and was approximately 4 times less effective than in the case of third harmonic generation in gas under comparable experimental conditions. In the case of a gas cell, the THG efficiency up to $3\times10^{18} \text{ cm}^{-3}$ changes according to the quadratic law as a result of phase matching process. With an increase in pressure, phase matching was broken and the efficiency of the THG corresponds to a linear increase, reaching a value of $\sim 5\times10^{-4}$ at $6\times10^{18} \text{ cm}^{-3}$.

It was observed that in the case of a cluster jet, the THG efficiency increased linearly with backing pressure up to $\sim 9\times10^{-5}$ at laser intensity of 1–200 TW/cm². The increase in the laser intensity up to
I>1000 TW/cm$^2$ is accompanied by the formation of filament in cluster jet and a decrease in the THG efficiency to 2x10$^{-5}$.

The interference model was applied for describing the interaction of laser radiation with clusters within the framework of which a comparative numerical calculation of the THG in gas and cluster media was carried out. The efficiency of THG in a gas cell changes according to a quadratic law up to 3x10$^{18}$ cm$^{-3}$, which is consistent with experiment. Further increase in pressure, phase matching was broken due to destructive interference if the third harmonics generated by different atoms in media and a drop in the THG amplitude was observed.

In the case of a cluster jet, numerical calculations of the THG for a cluster jet with an average cluster size in the range of 10-100 nm show that the THG yield corresponded to a power function with an exponent of 1.4. Theoretical calculations show that the THG amplitude in a gas is about an order of magnitude higher than in a cluster medium.

A possible way of low order harmonics efficiency enhancement in cluster media may be in using of two-color laser field instead of single color one. The advantages of multicolor laser field for harmonics generation in gases was demonstrated in [13].

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