Terahertz emission during interaction of ultrashort laser pulses with gas cluster beam

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Abstract. We present the results of experimental study of terahertz (THz) generation in gas cluster beam excited by intense femtosecond laser pulses. Cluster beam was produced by partial condensation of pure Ar and mixestures CF₂Cl₂+He, Ar+He during their expansion through a conical nozzle into vacuum. There were used two excitation schemes in our experiments: single color and two color (fundamental frequency mixed with its second harmonic). We have studied how THz signal scales with various control parameters such as laser pulse duration, gas backing pressure and laser pulse energy. Simultaneously we measured intensity of X-Ray emission which originates from laser-cluster interaction. We found that in a single color scheme energy of THz pulses from Ar cluster beam strongly decreases in the region of minimum laser pulse duration while X-Ray power is maximal under these conditions. Both in single- and two color excitation regimes THz signal demonstrated growth without saturation with increasing of optical pulse energy up to its peak value of 25 mJ.

1. Introduction.
Clusters are aggregations of atoms or molecules consisting of 10…10⁷ particles [1]. Interaction of intense laser pulses with clusters is now of great interest due to the possibility of usage in various practical applications and fundamental problems of behavior matter under intense laser fields. There were observed various nonlinear phenomena during interaction of intense laser pulses with clusters: optical harmonic generation [2], self-focusing [3], X-Ray emission [4-6], stimulated Raman scattering [7]. One of the most convenient methods to produce clusters is partial condensation of gas flow during its adiabatic expansion through a nozzle [8]. Cluster beam, obtained by this technique, combines advantages of solid and gaseous media. Cluster inherits high local density and this implies high value of nonlinear response. At the same time cluster beams do not expose to ablation and renew their properties before each act of interaction with the laser pulse. It was shown theoretically and experimentally [6, 9, 10] that absorption of laser radiation in cluster beam can reach high values (up to 95%) which is related to linear (Mie) and nonlinear resonance interactions. Resonance absorption of pulse energy results in efficient production of X-Rays and fast charged particles.

In some recent articles [11-13] intense THz generation in cluster beam excited by ultrashort laser pulses was reported. Authors of [12] observed more than two orders of magnitude enhancement of THz pulse intensity in Ar cluster jet compared that in gaseous Ar with equal average atomic density. Generation of THz pulses in laser produced plasma of gases and solids was widely studied in past two
decades [14-17]. Plasma source allows to obtain intense and broadband terahertz pulses. Maximum bandwidth of subpicosecond pulse can reach up to ~100 THz [16] and is limited only by laser pulse duration. In work [14] two color scheme was introduced where the fundamental laser frequency at $\omega$ is mixed with its second harmonic at $2\omega$. Two-color scheme allows to increase THz yield by 2…3 orders of magnitude and at the present time this scheme is widely used for THz generation in gaseous media. Nevertheless, optical to terahertz conversion efficiency in laser-plasma generation method is still low. Also there was observed saturation of THz yield in a two-color scheme at high excitant pulse energy which originates from THz absorption in dense plasma [17]. Cluster target seems to be attractive to solve these problems. As for the possible mechanism of THz emission in clusters, authors of the article [13] proposed that low-frequency radiation originates from time-varying electrical quadrupoles which are produced by ponderomotive induced charge separation in plasma. Ponderomotive force arises from radial gradient of the electric field in Gaussian laser beam and its value is proportional to $VE^2$. Authors of [13] related the enhancement of THz intensity to high local plasma density in clusters. However, complete explanation of nature of low-frequency emission from the clustered plasma is a matter of research.

The subject of the present work is experimental study of THz generation in gas cluster jets both in single- and two color excitation regimes. Simultaneously we study X-Ray emission from clustered plasma to obtain information about laser-cluster interaction.

2. Experimental setup.

The scheme of our experiment is shown in figure 1. CPA laser system with Ti-Sapphire multipass amplifier was used as the source of high-energy optical pulses for the experiment. Our laser system delivered pulses with maximum energy of 25 mJ at repetition rate of 10 Hz. The central wavelength was 810 nm, the beam diameter 1.5 cm and the quality factor $M^2$=1.6. Pulse duration could be changed in a range of 50…1000 fs. Laser radiation was focused into a cluster jet by lens L1 (F = 20 cm) at the distance 6 mm below the nozzle edge. Peak laser intensity in a focal plane was estimated to be $\sim10^{17}$ W/cm$^2$ in vacuum. Nanosecond and picosecond contrast of laser pulse was $\sim10^8$ and $\sim10^7$ respectively. As it follows from our evaluation, influence of the prepulse on the laser-cluster interaction can be neglected in our experiments. Entrance window of the vacuum chamber and focusing lens L1 were made of MgF$_2$ which has low value of nonlinear refraction index and therefore the self-interaction effects were minimized. Second harmonic generation in a two-color scheme was obtained by 300 um-thick I-type BBO crystal. Nonlinear crystal was placed inside the vacuum chamber between the entrance window and the laser beam focus.

![Figure 1. Schematic of experimental setup: (a) single color-, (b) two color excitation scheme.](image-url)
For cluster beam production we used conical nozzle with input diameter \( d_{\text{in}} = 0.75 \text{ mm} \), output diameter \( d_{\text{out}} = 4 \text{ mm} \), half opening angle \( \alpha = 5^\circ \) and 20 mm length. The nozzle was connected to the high-pressure chamber with a pulsed electromagnetic valve which operated at repetition rate 1.25 Hz and was synchronized with the laser pulse. Delay time between laser pulse and the opening of the valve could be tuned in a timing module to provide the ability to control and optimize cluster formation process. Opening duration of the valve was 400 us to reach stationary jet flow. Working pressure in vacuum chamber was not above 5 mTorr. For cluster beam production we used pure Ar gas and mixtures: \( \text{CF}_2\text{Cl}_2+\text{He} \), \( \text{Ar+He} \) with various partial concentrations of the components in mixture. Maximum value of gas backing pressure was 4 MPa. Vacuum chamber was equipped with polypropylene windows with transmission band 0…3.5 THz. Terahertz pulse energy was measured with a liquid helium - cooled bolometer (LN-6C type, Infrared Laboratories, Inc). The input THz filter of the bolometer transmitted radiation in the spectral range of 0.1…1.5 THz. In a single-color scheme THz beam was collected and focused by pair of off-axis parabolic mirrors: PM1 (\( F = 5 \text{ cm}, D = 2.4 \text{ cm} \)) and PM2 (\( F = 15 \text{ cm}, D = 5 \text{ cm} \)). Optical axis of PM2 was placed at 30° angle from the laser propagation direction in horizontal plane. There was found previously that spatial structure of THz pattern which is generated in cluster jet has four-lobed structure [13]. In our experiments PM1 collected THz radiation in one of the significant peaks of power angular distribution which is located at \( \approx 30^\circ \) direction to laser beam axis. In a two color scheme THz beam was collected by TPX lens L2 (\( F = 10 \text{ cm}, D = 5 \text{ cm} \)) in forward direction. X-Ray emission of clustered plasma was measured by photomultiplier tube (PMT) equipped with NaI scintillator. Beryllium window of vacuum chamber transmitted X-Ray radiation with quanta energy above 2 keV. PMT was installed in front of the beryllium window at the angle of 135° from laser propagation direction.

3. Results and discussion.

Energy dependences obtained in a single- and two color excitation regimes are shown in the figure 2 and figure 3 respectively. We have observed that THz radiation intensity scales by a power function (“\( \omega + 2\omega \)” scheme) or linearly (“\( \omega \)” scheme) with increasing of laser pulse energy up to maximum available pulse energy of 25 mJ. Note that THz signal on graphs in figure 1 and figure 2 cannot be directly compared due to the difference in conditions of THz beams collection. Also we found that in a two color scheme usage of mixture Ar + He at concentration ratio (1:10) results in \( \approx 10 \)-fold enhancement of THz yield compared to pure Ar jet.

**Figure 2.** Energy dependence in a single color scheme, Ar cluster jet (backing pressure 2 MPa)

**Figure 3.** Energy dependence in a two color scheme, Ar + He (1:10) cluster jet (backing pressure 1.8 MPa)

In a single color scheme we observed an interesting phenomenon while we studied influence of laser pulse duration on THz emission intensity. Compressor of our laser system produced positively or
negatively chirped pulse with variable duration \( \tau \) which was defined by value of displacement of diffraction grating. Terahertz and X-Ray intensity scaling with pulse duration \( \tau \) for positively and negatively chirping are shown in a Fig. 4 (argon cluster beam at a backing pressure of 2 MPa).

**Figure 4.** THz and X-Ray yield from Ar cluster jet (backing pressure 2 MPa) as a function of laser pulse duration for positively/negatively chirping. Laser pulse energy \( E = 22.7 \) mJ.

As can be seen, THz yield strongly decreases in the region of minimum \( \tau \) whereas X-Ray intensity demonstrates maximum values under these conditions. We suggested that THz radiation can be absorbed in the dense cluster plasma since laser intensity takes the maximum values in the region of minimum pulse duration. For further investigation we carried out similar measurement at different laser pulse energies (figure 5). Nevertheless, all curves obtained with various laser pulse energies had the same shape. Influence of pulse duration on X-Ray yield in laser-cluster interaction was studied earlier. X-Ray power scaling with pulse duration, represented in figure 4, was observed in [6] and was explained by nonlinear resonance absorption process which involves energetic electron oscillations back and forth through the cluster.

Also we have studied terahertz generation in molecular clusters of Freon gas (CF\(_2\)Cl\(_2\)) using mixture CF\(_2\)Cl\(_2\) + He at concentration ratio (1:9). Adiabatic condensation of such mixture leads to formation of large clusters with higher atomic density compared to noble gas clusters. There was shown earlier that usage of molecule clusters allows to sufficiently increase X-Ray emission [5, 6]. Peak amplitude of THz signal from CF\(_2\)Cl\(_2\) + He jet in our experiment was of the same order that from Ar jet. Figure 6 demonstrates influence of laser pulse width on THz and X-Ray generation in CF\(_2\)Cl\(_2\) clusters (at mixture backing pressure of 25 atm and laser pulse energy 22.7 mJ). As can be seen, THz signal decreases in the region of minimum \( \tau \) values as well as in the case of Ar clusters. Power of X-Ray emission also decreases since laser pulse has the shortest duration and this differs from previous experiment with Ar cluster jet (figure 4). A possible reason of this phenomenon may be self-interaction effects which occur inside cluster jet excited by intense laser pulses. The self-interaction of laser radiation is strong in (CF\(_2\)Cl\(_2\) + He) cluster beam since molecular cluster acquires high internal density of electrons during ionization [5].

**Figure 5.** THz yield from Ar cluster jet (backing pressure 2 MPa) as a function of laser pulse duration for positively/negatively chirping at different pulse energies.
4. Conclusion.
We have experimentally investigated terahertz generation in cluster beam excited by single- and two color intense laser pulses. We found that amplitude of THz signal reaches minimum values when laser pulse has the shortest duration and this effect occurred both in atomic (Ar) and molecular (CF$_2$Cl$_2$ + He) cluster beams in a single color scheme. Scaling of terahertz radiation power with pulse duration represents dynamics of laser-cluster interaction and there is an actual problem to find theoretical explanation of the observed phenomenon. Both in single- and two color excitation regimes we observed growth of THz yield without saturation with increasing of laser pulse energy up to its maximum value of 25 mJ. We found that in a two color scheme usage of mixture Ar + He at concentration ratio (1:10) results in ~10-fold enhancement of THz pulse energy compared that in pure Ar cluster jet.

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