High-intensity THz pulse generation by TW laser radiation in ionized gas and nonlinear crystals

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Abstract. Intense electromagnetic fields can be obtained now in THz frequency range. THz pulses with electric field intensity above 1 MV/cm are now requested for strong-field applications like particle acceleration, short electron bunches measurements, controlling the state of the matter. The advantage of a THz pulse in comparison with the visible frequency range is the almost non-oscillating field that interacts with charged particles more efficiently. The advantage over a direct field is the ability to remotely focus this radiation in space and in time. Except for the huge free-electron lasers facilities, the only way to get THz pulses of MV/cm intensity is the conversion of high-power femtosecond laser pulses. We compare two ways of generating intense THz pulses using the radiation of TW and GW laser systems operating at 800 nm. They are two-color filamentation in gas and optical rectification in nonlinear crystals. The prospects for scaling of THz generation methods for a petawatt laser are discussed.

1. Introduction

Besides applications in nonlinear optics, strong THz pulses can serve as a tool to instantly and remotely modify the state of the matter without thermal damage. For example, coherent lattice excitation (phonon modes) by short THz pulses is a promising route towards ultrafast control of phase transitions and associated changes in material properties.

If we do not consider free-electron laser (FEL) facilities, where THz pulse energy is much stronger [1], the maximum THz field strength (\(>\) 10 MV/cm) is obtained in laser-irradiated nonlinear crystals [2,3]. In this case, despite the limitations on the available aperture and damage threshold of the crystal (typically it is below 50 mJ/cm\(^2\)), a relatively high energy conversion efficiency from laser to THz–10\(^{-3}\) can be obtained. But generated spectra bandwidth is limited from above by the crystal transparency region and the phase-matching condition. In this case, the frequency of the maximal field amplitude is 1–2 THz. Unfortunately, this generation method is difficult to scale into petawatt laser pulses due to the breakdown and insufficient size of
nonlinear crystals [4] and due to the lack of suitable crystals for the 800 nm wavelength of existing petawatt laser systems. The strongest THz pulses in crystals are obtained not for 800 nm wavelength lasers, but 1200–1500 nm femtosecond lasers because of better phase-matching in highly nonlinear crystals like OH1 or DSTMS there, although 800 nm pulses can be much more powerful due to Ti:Sapphire crystal outstanding properties.

On the other hand, THz pulse generation via two-color laser-induced plasma is free from damage threshold and phase-matching drawbacks, although due to the low conversion efficiency of \(10^{-5}\), it does not provide strong THz field with a laser pulse with a power lower than a few TW. In this two-color method, ultrashort amplified laser pulses are focused in the air (or any gas) to create gaseous plasma via tunneling ionization. To obtain two colors the fundamental wave (\(\omega\)) passes through a thin nonlinear crystal and is partly converted into its second harmonic (2\(\omega\)). Both pulses propagate together before focusing into the area, where plasma and THz generation occurs. A photoelectric current model was proposed to explain the principle of THz wave generation from two-color laser-induced gas plasma [5–8]. This model operates with the directional drift of ionized electrons in the asymmetric two-color laser field. The current occurs on the subpicosecond timescale and emitted radiation falls into the THz frequency range.

The region of THz radiation generation can be extended by forming a laser filament [5–8] in an ambient air serving as an elongated plasma converter. It is preferable to stay in a monofilamentation regime for the best efficiency of THz yield. This obliges to limit the power and pressure product to a certain value [9]. When applying terawatt powers of a laser pulse, it is necessary to decrease the pressure of the gas in which filamentation occurs [10] and increase the focusing length, then this monofilamentation regime can be scaled further [9] to a petawatt power level. Thus we can increase THz pulse energy \(E\) considerably. Another advantage of THz generation in plasma is the substantially greater spectral bandwidth and accordingly shorter duration of the THz pulse \(\tau\). Thus we can increase THz peak field amplitude \(A\) considerably, since \(A \sim \sqrt{E/\tau}\).

At present, for the filamentation at 35 mJ laser pulses pumping of 800 nm (\(\omega\)) and 5% of pump power at 400 nm (2\(\omega\)), the achievement of THz pulse energy of \(E = 2 \mu J\) has been reported in [6]. In [7], at 10 mJ pumping, \(E = 2.7 \mu J\) THz pulse was obtained with an estimated peak field of \(A = 16 \text{ MV/cm}\). In earlier paper [8] 7 \(\mu J\) THz pulses have been obtained from 2 TW laser pulses. All these experiments were carried out in the ambient air.

As far as we know, with TW pulses in a low-pressure gas, two-color filamentation experiments were not carried out to generate THz. In this study, we experimentally demonstrated that decreasing the pressure to a proper value can essentially increase THz yield. When working with a pressure other than atmospheric one or with another gas, a vacuum chamber is necessary (figure 1). The entrance thick window of such a chamber limits the available energy of a laser pulse due to phase self-modulation in the window material, therefore in this work, we limited ourselves to a laser pulse energy of 80 mJ with an incoming laser beam diameter of 7 cm.

2. Experiment on THz generation in a gas
For a generation in LiNbO\(_3\) crystal as well as in atmospheric plasma, we used conventional kHz rate 2 mJ Ti:Sapphire amplifier laser system. In all other experiments, we used the radiation of the subpetawatt Ti:Sapphire laser system “Pulsar 200 TW”, installed in the NRC “Kurchatov Institute”. The laser system can generate 25 fs pulses with an energy up to 7 J and a 10 Hz repetition rate. An essential element of the THz generation scheme in the two-color filament is the second harmonic generation (SHG) (2\(\omega\)) crystal, see figure 1, since the presence of the second harmonic increases the THz generation efficiency by several orders of magnitude. Usually, a beta barium borate (BBO) crystal is used [6, 7] for frequency doubling, but it is difficult to obtain thin BBO crystal with a large aperture, which is necessary for safe intensity level when working with terawatt pulses. For the laser pulses with an energy of hundreds of mJ, a unique thin SHG
crystal of a potassium dihydrogen phosphate (KDP) $20 \times 20 \times 0.15$ mm was produced in Institute of Crystallography of Russian Academy of Science. This crystal withstands the laser intensity up to 0.2 TW/cm$^2$ (which for the used laser parameters corresponds to 2 cm beam diameter).

To adjust the distance “$z$” between the THz generation area and SHG generation point, the KDP crystal was positioned on a moving carriage controlled by magnets from the outside of the vacuum chamber, see figure 1(b). Laser radiation after filamentation was blocked by a silicon small-aperture filter. The exit window of the vacuum chamber was a 4 mm thick polypropylene block, transparent enough for frequencies below 5 THz. To detect higher THz frequencies we replaced this plastic window onto the 3 mm high resistivity Si one. A polyethylene window installed in the Golay cell attenuates the high-frequency part of THz spectra, so in presented data; we deal with the low-frequency part of the THz spectrum, below 10 THz. To focus laser radiation, a large aperture spherical mirror with a focal length of 2.5 meters was used. The second harmonic crystal was located at a distances of $z$ from 30 to 75 cm from the geometrical focus, depending on the laser energy used. The fraction of second-harmonic radiation in our case was 2–5% of the fundamental radiation power. The visual length of the filament was about 15 cm for 20 mbar (air or nitrogen) pressure.

We investigated what maximal energy of THz pulses can be obtained in such a scheme in the laser power range below 80 mJ. The optimization of the second harmonic crystal position, the gas pressure and the pulse duration were performed (the data in figure 2). In our case, the pressure decrease provides almost two orders of magnitude gain in the THz generation efficiency. Filling the vacuum tube with air, nitrogen or argon does not significantly change the obtained THz signal. Optimal pulse duration for pulse energy below 40 mJ is the minimal one. As a
result, for a 40 mJ, 35 fs pulse, 10 mbar of nitrogen pressure, \( z = 40 \text{ cm} \) to the KDP crystal we obtained a THz pulse of about 2 \( \mu \text{J} \) (4500 mV peak amplitude on the Golay cell). From the energy dependence in figure 2(c) the saturation of THz yield is observed above 40 mJ even for rather low gas pressure, although SHG yield still increases. Excessive ionization is unlikely to be the main reason for the observed saturation because it happens also at low pressure, even below monofilamentation regime. Another possible reason for THz saturation is the pulse distortion in the entrance window, however it is not confirmed by the SHG data in figure 2(c). Pulse duration dependence in figure 2(b) for the 50 mJ case demonstrates, that saturation is coursed not by pulse energy (it is fixed), but by pulse peak power (governed by pulse duration). This saturation should be investigated and, perhaps, overcome before the further increase of the incoming laser energy becomes reasonable. Distinct fluctuations in the terahertz energy as the gas pressure increases in figure 2(a) are associated with a phase slippage between \( \omega \) and \( 2\omega \) radiation [10]. It is possible to tune to the maximum generation of THz by either shifting the SHG crystal toward the laser focus (while still avoiding the breakdown threshold) or by adjusting the pressure.

A significant advantage of low-pressure case is the increase of the effective length at which \( \omega \) and \( 2\omega \) are in the right phase inside filament, for long filaments this is important, otherwise the useful THz generation will not be from the entire plasma region. However, the approach of using THz generation in a long filament results in the formation of a long and diverging source which is not convenient enough for most applications. To improve THz radiation collimation, we placed a filament into a specially designed waveguide for THz range [11]. The waveguide consists of a ring of polypropylene capillaries, the filament propagates 10 cm in its central air core with a diameter of 1.5 mm. The THz signal collected in this way was strong enough, only 2 times smaller than in the case without a waveguide. The signal decrease is due to the limited bandwidth of this structure [11]. Nevertheless, for frequencies inside the transparency range of this waveguide (0.7–1.5 THz and 2.5–3 THz) generated radiation is confined inside a 1.5 mm area of the central part. This radiation can be refocused to the application point providing strong intensity there.

### 3. Experiment on THz generation in crystals

Using the same laser source (“Pulsar 200 TW”) and detector, the efficiency of the generation of THz in nonlinear crystals of ZnTe and GaSe was measured. For 6 mJ of pumping, values of 30 and 10 mV were obtained from the Golay cell THz detector for ZnTe and GaSe accordingly, figure 3(b). Beam size on the crystals (4–6 mm in diameter) was optimized for maximal THz yield but below the damage threshold. For the 40 mJ case, we had to increase beam diameter and use another GaSe crystal with a transverse size of 6*4 cm to avoid optical damage. For this case we manage to convert 40 mJ of laser energy into 40 mV of THz signal, but still, it is not so efficient as plasma generation or ZnTe crystal.

Disadvantages of ZnTe material are strong two-photon absorption for the visible radiation (that prohibits to apply powerful pulses to it) and a low frequency of phonon absorption (at 5.4 THz) [12]. The advantage of ZnTe for 750–850 nm laser radiation is the exact phase-matching for the THz generation process. Here phase-matching means that THz radiation phase velocity coincides with the group velocity of pumping pulse. Thus ZnTe is the best material for sub-mJ Ti: sapphire laser systems and conversion into THz range.

The advantage of GaSe crystal is high transparency in THz range and high birefringence in both visible and THz ranges. This allows us to obtain phase-matching for a desirable THz frequency by tilting the crystal [13]. Optimal crystal thickness is determined by the smallest value among the coherence length (determined by phase-matching in a particular THz frequency range) and absorption length (determined by absorption coefficient in both THz and visible ranges). For frequencies below 3 THz the optimal thickness for ZnTe and GaSe crystals is about 1 mm.
Another possibility to obtain intense THz pulse is to use wide-aperture crystals with a high damage threshold, such as lithium triborate (LiB<sub>3</sub>O<sub>5</sub> or LBO), or with high nonlinearity such as LiNbO<sub>3</sub>. One should use small crystal thickness (50 µm), because of phase mismatch for the THz generation process. In most such crystals useful length for THz generation is small (100 µm) and efficiency of conversion into THz is below 10<sup>−5</sup>. Still with subPetawatt laser pulses, one can obtain hundreds of microJoules energy in THz pulse [3].

To calibrate the THz detector in the 10 Hz repetition rate, THz signals were compared with the THz radiation obtained in the scheme of a tilted pulse front in lithium niobate at 1.5 mJ, 100 fs of Ti:Sapphire laser pumping (it does not withstand the higher input power). For that purpose, we used commercially available high power THz generator TERA-AX-2 (Avesta Project Ltd), based on the tilted front technique. It contains triangular prism made of MgO:LiNbO<sub>3</sub> crystal that can be cooled down to the cryogenic temperatures to enhance optical-to-THz conversion efficiency [14], cooling decreases strong THz absorption in LiNbO<sub>3</sub> material. Standard scheme with the 1200 mm<sup>−1</sup> grating and biconvex F = 150 mm lens is used to create the tilted front. It could provide us up to 2 µJ of THz pulses with 1.5 mJ of input optical pump radiation at a 1 kHz repetition rate. The spectrum of the source, measured with the THz-TDS technique (with 1 mm thick ZnTe detector), is depicted in figure 3(a). With the help of two 50 mm aperture, 50 mm focal length off-axis parabolic mirrors we can reach THz fields above 0.5 MV/cm. THz spectra obtained in LiNbO<sub>3</sub> and two-color filament are compared in figure 3(a), obtained THz energy for different emitters is summarized in figure 3(b).

So far, the received THz pulse energy in two-color plasma (4500 mV signal with 40 mJ pumping) is almost the same as that from lithium niobate (5000 mV signal with 1.5 mJ pumping). If the peak field of the THz pulse is above 0.1 MV/cm, a nonlinear transmission increase can be observed in the open aperture z-scan measurements [15].

When generating THz in a two-color filament with a 40 mJ laser pulse, we experimentally observed a 12% induced (nonlinear) transparency increase in silicon (figure 4). In this case, the maximum radiation spectra were estimated around 1 THz. From literature data [15], it is known that induced transparency in Si wafer with the used resistivity requires field strength above 0.1 MV/cm. For 2 µJ THz pulse it can only be in the case of sub-ps duration and mm focusing. This way we prove the possibility of good focusing of THz radiation in space and time for the case of two-color generation in a gas. Note that spatial focusing can be further improved
Figure 4. Transmission of THz radiation in p-doped silicon (with conductivity 4 Ohm cm, thickness 670 µm) depending on the offset from the beam waist of the THz beam: (a) the case of THz generation in lithium niobate; (b) the case of THz generation in a two-color filament.

by THz confinement in a hollow-core waveguide. For the case of THz generation in LiNbO3 with 1.5 mJ laser energy and known THz energy of 1 µJ, the induced transparency was 30%. For this case, the maximum pulse spectra were estimated at around 1.5 THz. Used p-doped silicon with a conductivity of 4 Ohm cm and a thickness of 0.67 mm was the same for two different THz generation experiments. Since doped silicon better transmits high frequencies, the value of the background transmittance was 0.27 for the plasma case and 0.25 for LiNbO3 case. For plasma generation, the field strength value of 300 kV/cm is obtained by comparing the measured energy (2 µJ), duration (0.5 ps) and beam-waist diameter (1 mm).

4. Conclusion

For 800 nm fs pulses of energy below 1 mJ, ZnTe crystal is a good, simple THz emitter but limited to a 2.5–4 THz range. Narrowband THz emission at 5–10 THz can be obtained in GaSe crystal with its proper orientation, due to layered structure GaSe can be in the form of large-area thin (flexible) plates. The later can be implemented for the conversion of TW-level laser pulses conversion into THz, yet the efficiency is lower than for optimized plasma generation.

LiNbO3 with tilted pulse front scheme up to now provides the highest THz field values for mJ 800 nm laser pulses, but damage threshold limits further increase in THz yield, while the spectra bandwidth is limited to 3 THz. Two-color plasma can generate THz fields comparable to that in LiNbO3 (MV/cm), but it requires 20 times higher laser energy (40 mJ) for that. The advantage of plasma generation is larger THz bandwidth (5–10 THz) and correspondingly shorter pulses. The further progress is expected due to mitigating of self-modulation, excessive ionization, and elongated radiation source. The aim is to keep the same conversion efficiency while the power of laser pulses will far exceed tens of TW. Then a much more powerful (and short) THz pulse than in the nonlinear crystals could be generated. This will be enough to work on THz nonlinear optics and the effect of powerful THz pulses influence on the matter.

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