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Terahertz generation optimization in an OH1 nonlinear organic crystal pumped by a Cr:forsterite laser

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We present the results of experimental investigations of terahertz radiation generation conversion efficiency in an OH1 nonlinear organic crystal pumped by femtosecond laser pulses at 1240 nm wavelength. An influence of OH1 crystal thickness on the terahertz generation by optical rectification method was studied. It is shown that the optimal crystal thickness for the maximum conversion efficiency is 1 mm, which agrees with the previously made theoretical estimates.

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Introduction. Recently, a significant increase has been observed in a number of studies of interaction of subpicosecond pulses of terahertz radiation with various materials. Rapid expansion of terahertz technologies became possible due to intensive research and development of terahertz sources; in particular, based on laser generation methods. Optical rectification is one of the methods widely applied. Generation of the terahertz radiation by optical rectification of picosecond laser pulses has been obtained in a nonlinear lithium niobate crystal [1]. However, difficulties with phase-matching conditions hindered practical implementation of this technique. At present, lithium niobate crystal is widely used being pumped by radiation of titanium–sapphire femtosecond lasers with a tilted wavefront to obtain high-energy terahertz pulses [2–6]. A record-breaking energy of a terahertz pulse of 1.4 mJ with a conversion efficiency of 0.7% has been achieved recently in lithium niobate crystal with an electric field strength of more than 6.4 MV/cm [7]. Other nonlinear inorganic crystals such as ZnTe [8] and GaP [9,10] are also used to generate terahertz radiation through optical rectification, but the obtained conversion efficiency is much lower. In recent years, OH1 [11,12], DAST [13,14], DSTMS [11], HMQ-TMS [15], and HMB-TMS [16] organic nonlinear crystals with high coefficient of second-order nonlinearity [17,18] have been widely applied. They are pumped by infrared laser radiation with a wavelength between 1.2 and 1.6 μm. A significant advantage of these organic crystals is high conversion efficiency of approximately a few percent, while the drawback lies in a relatively low pump energy density that damages the crystal.

Radiation of titanium–sapphire lasers with a wavelength of 800 nm converted by an optical parametric amplifier (OPA) into the required spectral range is commonly used for pumping such crystals. The disadvantage of OPA is the heterogeneity of the spatial distribution and low energy stability. BNA, in contrast to the organic crystals listed above, can be pumped by 800 nm radiation but its damage threshold is several times lower [19]. A femtosecond Cr:forsterite laser with a wavelength of 1240 nm is an alternative source for pumping organic crystals. Applying such a laser made it possible to obtain terahertz pulse energy of 0.9 mJ in DSTMS [20] crystal, as well as to demonstrate efficient conversion in DAST and OH1 with a high quality of terahertz beam spatial distribution [21]. Compared with DAST and DSTMS, OH1 crystal has a higher figure of merit [17]. The phase-matching condition is satisfied well for pump wavelengths ranging from 1200 nm to 1460 nm resulting in generation of radiation up to 3 THz. There is also no absorption in the vicinity of 1 THz, in contrast to DAST and DSTMS crystals. However, OH1 has significant absorption around 3 THz compared with HMQ-TMS and HMB-TMS crystals, which demonstrates lower absorption up to 5 THz. The optimal phase-matching conditions for an OH1 crystal below 3 THz are satisfied when pumping with a wavelength of 1240 nm is used, in contrast with the HMB-TMS crystal, where good matching is satisfied at longer wavelengths, starting from 1300 nm [16]. Thus, OH1 crystal seems to be the most efficient generator of terahertz radiation among organic ones in the frequency range below 3 THz pumped by a Cr:forsterite laser.

The thickness of nonlinear crystal is an important parameter for efficient generation of terahertz radiation, since it determines the electric field strength of a pulse obtained in a crystal. The thickness affects also the spectral intensity of the radiation if phase-matching conditions are not satisfied. In addition, modulation of spectral intensity of the terahertz pulse can be associated with absorption of the radiation in the crystal. Thus, utilization of very thin nonlinear crystal makes it possible to obtain a terahertz pulse with a wide smooth spectrum, which is limited by the duration of the pump pulse only. Conversion efficiency will, however, be low. The use of a relatively thick crystal leads to a high conversion efficiency when the phase-matching

condition is satisfied. However, modulation in the spectrum of the terahertz radiation appears, depending on the absorption coefficient at the generated frequencies [18]. The terahertz radiation obtained via different frequency generation methods was studied in [22] for OH1 crystals with thicknesses varying from 0.25 mm to 1.05 mm. In Ref. [23], generation of high-power single-cycle terahertz pulses by optical rectification methods in OH1 crystals was considered for only two thickness values of 0.4 mm and 0.93 mm. The reported data [22,23], however, did not allow us to determine the optimal thickness corresponding to the maximum of conversion efficiency.

In this Letter, we present the results of experimental studies of generation efficiency and spectral intensity of terahertz radiation obtained by optical rectification of femtosecond laser pulses with a wavelength of 1240 nm in nonlinear organic OH1 crystals of different thicknesses in the range of 0.4 mm to 1.6 mm.

Methods. Nonlinear OH1 organic crystals [2-(3-(4-hydroxystyryl)-5,5-dimethylcyclohex-2-enylidene)malononitrile)] manufactured by Swiss Terahertz GmbH with a thickness from 0.4 to 1.6 mm and a diameter of 2 mm on a sapphire substrate have been used in experiments. The 3,5,5-trimethyl(cyclohex-2-enylidene)-malononitrile was prepared by means of Knoevenagel condensation of malononitrile (100 mmol) and isophorone (100 mmol). Both compounds were dissolved in N,N-dimethylformamide (500 ml). The solution was stirred for 12 hours at room temperature in the presence of a catalyst. A yellow precipitate was obtained from the resulting dark-yellow solution after partial evaporation of the solvent. The product was recrystallized from 95% ethanol. The title compound was prepared according to a published procedure [24] from 3,5,5-trimethyl(cyclohex-2-enylidene)malonodinitrile (1.86 g, 10 mmol) and 4-hydroxybenzaldehyde (1.22 g, 10 mmol) in a 150 ml trichloromethane solution with continuous stirring for two days at room temperature using a catalyst. The orange solution was purified by column chromatography on silica gel. The orange precipitate was recrystallized. Crystals were grown by slow evaporation from methanol.

The nonlinear organic crystal OH1 was pumped by a femtosecond Cr:forsterite laser [25]. The central radiation wavelength was 1240 nm, the pulse duration was 100 fs, and the repetition rate was 10 Hz. The pump beam diameter was 1.5 mm, and the pulse energy varied from 20 μJ to 300 μJ. Laser pump beam and terahertz radiation were polarized along the *c* axis of the organic crystal OH1.

Terahertz pulse energy was measured by calibrated an acousto-optical Golay cell. A broadband terahertz cutoff filter (LPF8.8-47, Tydex) was used to cut radiation at wavelengths shorter than 34 μm. The optical pump energy density was adjusted by a polarization attenuator consisting of a polarizer and a half-wave plate. Attenuators (ATS-5-50.8, Tydex) were used to adjust the terahertz pulse energy. All the experiments were carried out in a dry air box with a humidity of 2% at a temperature of 23° C.

Results. Figures. 1 and 2 demonstrate the energy of a terahertz pulse and the conversion efficiency as a function of the optical pump fluence for OH1 crystals with different thicknesses. It can be seen that an increase in the crystal thickness results in a growth of the terahertz pulse energy. However, a significant energy decrease is observed for crystal thickness of 1.63 mm. The maximum conversion efficiency (Fig. 2) is achieved for all crystal thicknesses at a pump energy density of ~ 8 mJ/cm^2; it reaches 2.5% for OH1 crystal 1 mm thick. Despite the fact

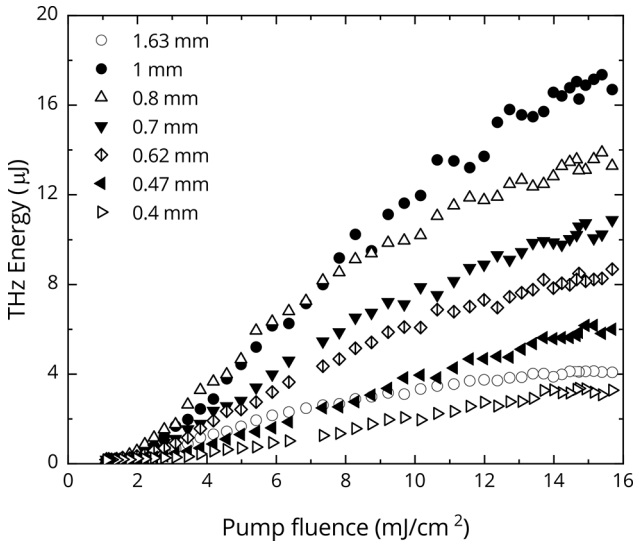

Fig. 1. Energy of terahertz pulses versus pump fluence for various OH1 crystal thicknesses.

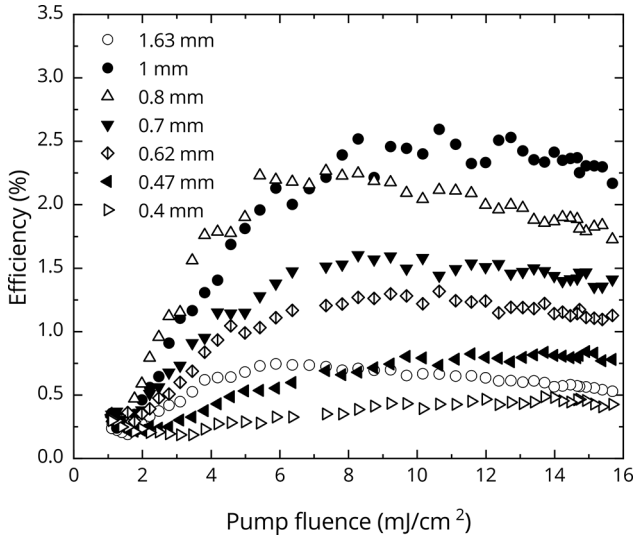

Fig. 2. Conversion efficiency as function of pump fluence for various OH1 crystal thicknesses.

that the conversion efficiency does not change or even slightly decreases for pump fluences greater than 8 mJ/cm^2, the output energy of the terahertz pulse continues to increase (Fig. 1). Relative errors of measurements were 5% for pump fluence and 10% for terahertz pulse energy. The maximum energy density in the experiments was limited to 16 mJ/cm^2 to prevent damage to the OH1 crystal. This is because the damage threshold of the OH1 crystal at a wavelength of 1240 nm does not exceed 20 mJ/cm^2.

Figure 3 shows the experimental dependence of the terahertz pulse energy on the thickness of the OH1 crystal at the maximum pump fluence of ~16 mJ/cm^2.

Using Ref. [26] in the approximation of plane wave, non-depleted pump, negligible optical and terahertz absorption, the dependence of the generated electric field $E_{THz}(\omega, L)$ at the angular terahertz frequency $\omega_{THz} = \omega$ on the crystal thickness L can

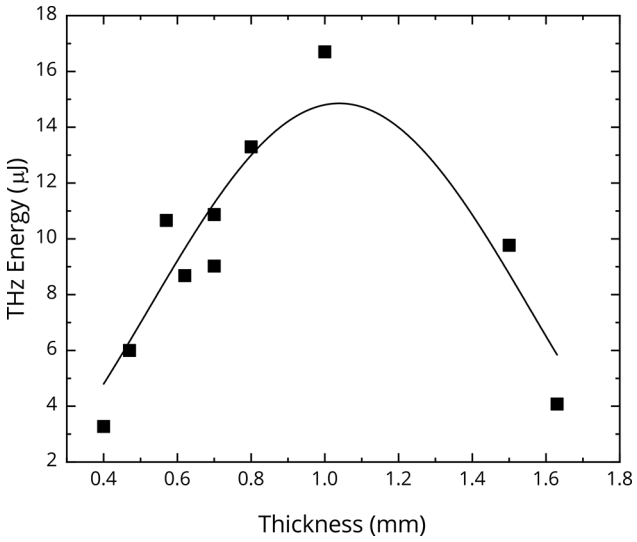

Fig. 3. Energy of the terahertz pulse as a function of the OH1 crystal thickness at the maximum pump fluence of 16 mJ/cm^2. The dots are experimental data, and the line is the approximation by Eq. (3).

be written as

$$E_{THz}(\omega, L) \propto \mathrm{sinc}\left(\frac{\pi L}{2l_c(\omega,\lambda)}\right) L, \tag{1}$$

where $l_c(\omega, \lambda)$ is the coherence length of generation of terahertz radiation by optical rectification, λ is the central wavelength of optical pulse. The coherence length determines the maximum thickness of the crystal, which still leads to an increase in the generated field [17,18]. The corresponding intensity of the generated terahertz wave $I(\omega, L)$ is given by [17]

$$I(\omega, L) = \frac{1}{2}\varepsilon_0 c n_{THz} |E_{THz}(\omega, L)|^2, \tag{2}$$

where n_{THz} is the refractive index of the crystal at frequency ω. Using Eq. (1), Eq. (2) can be transformed, as follows:

$$I(\omega, L) \propto \mathrm{sinc}^2\left(\frac{\pi L}{2l_c}\right) L^2 = \frac{4l_c^2}{\pi^2} \sin^2\left(\frac{\pi L}{2l_c}\right). \tag{3}$$

Approximation of the experimental data by Eq. (3), which defines the dependence of the terahertz intensity on the crystal thickness, shows that the optimal crystal thickness at the optical pump wavelength $\lambda = 1240$ nm is around 1 mm. It should be noted that the obtained experimental data are in a good agreement with previously published calculations [17,21].

Measurements of the terahertz radiation spectrum generated in the OH1 crystals with different thicknesses were carried out using an electro-optical detection scheme with a GaP crystal 200 μm thick. Figure 4 shows the influence of the crystal thickness on the waveforms and the spectral characteristics of terahertz radiation.

The maximum amplitude of the emission spectrum lies in the frequency range of 1.2–1.3 THz.

As expected, the terahertz spectrum narrows with increasing crystal thickness, and in the vicinity of the frequency of 1.5 THz, modulation appears due to absorption in the OH1 crystal [26]. The depth of modulation increases as crystal thickness grows. An important difference between the OH1 and DAST/DSTMS

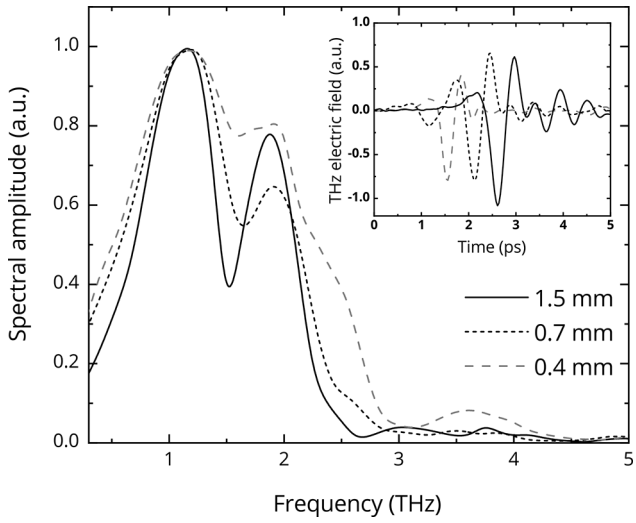

Fig. 4. Normalized terahertz spectra measured for nonlinear organic crystals with different thicknesses. Inset: terahertz pulse waveforms.

spectra is the absence of a strong drop at a frequency of 1.1 THz associated with a strong optical phonon vibration in DAST and DSTMS crystals [27]. The absolute amplitude of this drop at a frequency of 1.5 THz is much smaller than that in DAST and DSTMS crystals at a frequency of 1.1 THz [21].

Conclusion. The optimal thickness of ~ 1 mm of nonlinear OH1 organic crystal for generating terahertz radiation by optical rectification of femtosecond pulses with a wavelength of 1240 nm was determined. Increasing the crystal thickness leads to a significant increase in absorption at a frequency of 1.5 THz. For a thickness of 1.5 mm, the spectrum modulation reaches 50%, but for the optimal crystal thickness it does not exceed 20%.

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Disclosures. The authors declare no conflicts of interest.

Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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