

TERAHERTZ RADIATION

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A source of THz radiation with electric field strength of more than 1 MV cm^{-1} on the basis of 100-Hz femtosecond Cr : forsterite laser system

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Abstract. **A source of terahertz (THz) radiation with a bandwidth of 0.5–2.5 THz and a pulse repetition rate of 100 Hz has been developed. The THz generation is based on optical rectification of femtosecond pulses (generated by a Cr : forsterite laser system) in a nonlinear organic crystal OH1. An optical scheme of a multipass amplifier for the Cr : forsterite laser system is developed to increase the pulse energy to 2.2 mJ. The conversion efficiency for pump laser radiation at a wavelength of 1.24 μm into THz radiation is found to be to 2%. An electric field strength of 3.5 MV cm^{-1} is attained when focusing a THz beam into a near diffraction-limited spot.**

Keywords: *multipass amplifier, femtosecond laser pulse, optical rectification, terahertz radiation.*

1. Introduction

Currently, there are several laser-based methods for generating terahertz (THz) radiation in the spectral range of 0.5–10 THz, which can be used to obtain high-energy pulses [1]; focusing this radiation, one can implement high electric field strengths. One of the methods for generating THz pulses of picosecond duration is optical rectification of a femtosecond laser pulse with an oblique wavefront. This method is efficient for generating high-energy pulses both without cooling a $LiNbO_3$ crystal [2, 3] and under cryogenic cooling conditions [4, 5]. In particular, a laser-to-THz conversion efficiency at a level of $\sim 10^{-3}$ was obtained in [3], and focusing 1-THz radiation into a spot ~300 μm in diameter provided a field strength of 1.2 MV cm^{-1}. An increase in the laser-to-THz conversion efficiency upon cooling a $LiNbO_3$ crystal was demonstrated in [4]. Vicario et al. [5] obtained 45-μJ pulses and estimated the possibility of attaining an electric field strength of 2 MV cm^{-1} when generating radiation with a centre frequency of 0.4 THz. THz pulses with an energy of 186 μJ and a centre frequency of 0.25 THz were obtained in [6] as a result of cooling a $LiNbO_3$ crystal to 23 K; the electric field strength was as high as ~0.7 MV cm^{-1}. Since the spectral range of THz radiation generated in a $LiNbO_3$ crystal is generally below 1.5 THz, it is rather difficult to obtain high electric field strengths because of the relatively large focused-beam diameter (which is determined by the radiation wavelength). In addition, focusing causes additional difficulties since the THz beam divergences in the vertical and horizontal planes differ. To compensate for this effect, special optical schemes of $LiNbO_3$ crystal pumping and THz radiation focusing are applied [7].

The THz radiation obtained when forming laser plasma [8–11] allows one to generate pulses with energies up to several microjoules in the frequency range of 1–100 THz; however, it is very difficult to implement higher pulse energies because of plasma instability. THz radiation sources based on electron accelerators provide pulses with energies higher than 100 μJ and electric field strengths up to 20 MV cm^{-1} at frequencies above 10 THz [12]. Drawbacks of these sources are low repetition rate of THz pulses and complexity of a synchronising electron accelerator with an external optical laser; as a result, some problems arise when carrying out time-resolved (pump–probe) experiments. The method of difference-frequency generation is also widely applied to generate THz radiation in the spectral range of 1–10 THz. In this case, crystals with high second-order nonlinearity (e.g., $LiNbO_3$) are used. However, it is difficult to obtain a high laser-to-THz conversion efficiency because of the complexity of implementing phase matching between the pump and THz beams [13].

An efficient way to generate high-energy THz pulses by means of optical rectification is to apply nonlinear organic crystals pumped by femtosecond pulses generated in a Cr : forsterite laser system [14–16]. This laser pumping made it possible to obtain pulses with energies up to 900 μJ [14] and generate broadband THz radiation in OH1, DAST, and DSTMS crystals [15] and narrow-band THz radiation in an OH1 crystal with a tunable central frequency [16] and high (up to 3%) conversion efficiency. The use of a Cr:forsterite laser system with a radiation wavelength $\lambda = 1.24$ μm yields a much more uniform spatial beam distribution [15] than that provided by an optical parametric amplifier [17]. Uniform spatial distribution improves significantly the THz beam focusing. However, the THz radiation source pumped by a Cr : forsterite laser system had a pulse repetition rate of 10 Hz; when using this source for experimental studies, the signal-to-noise ratio was much lower than that for an optical parametric amplifier operating at frequencies of 100 Hz or 1 kHz.

To obtain pump radiation uniformly distributed in space, which is converted with high efficiency into THz radiation in nonlinear organic crystals, it was proposed [18] to apply stimulated Raman scattering of Ti : sapphire laser radiation in gas; this technique provided femtosecond pulses with a centre wavelength of 1.28 μm and a repetition rate of 100 Hz. However, there are no data on applying this radiation to generate THz pulses.

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The simplest and most promising way to obtain pump radiation for generating THz pulses in nonlinear organic crystals is to develop a multipass amplifier for a Cr:forsterite laser system operating with a pulse repetition rate of 100 Hz or higher. Despite the fairly low gain of the Cr:forsterite active medium in comparison with Ti:sapphire, it was shown in [19] that this medium may provide femtosecond pulses with an energy up to 90 mJ and a pulse repetition rate of 10 Hz. Data on generating 1-mJ femtosecond pulses with a pulse repetition rate up to 50 Hz were reported in [20]. A multipass amplifier with an optical scheme based on spherical mirrors was applied in that study to generate high-energy pulses. A drawback of the proposed scheme is the fixed size of the laser beam in the active medium that impedes generation of high-energy pulses.

Thus, the problem of generating THz pulses with a high conversion efficiency, high energy, uniform spatial distribution, and high repetition frequency remains urgent.

In this paper, we report the results of experiments on the generation of THz pulses with high energy and repetition rate of 100 Hz using optical rectification of femtosecond pulses produced by a Cr:forsterite laser system with $\lambda = 1.24$ μm in a nonlinear organic crystal OH1 with high conversion efficiency. To obtain a desired pulse fluence for this system, we developed and implemented a scheme of a multipass laser amplifier (MLA) without any fundamental limitations on the pulse energy. A possibility of using the same laser to pump both the active medium of regenerative amplifier (RA) and the MLA active element is demonstrated.

2. Multipass laser amplifier

The MLA was pumped by radiation of a Cr:forsterite femtosecond laser system (Avesta Project, Russia), operating in the chirped-pulse amplification regime and consisting of a master oscillator, a stretcher, a regenerative amplifier, and a time compressor. This laser system provided output pulses with duration of 95 fs, energy of 350 μJ, and repetition rate of 100 Hz. The RA was pumped by a repetitively pulsed, Q-switched Nd:YAG laser (LQ629-100 Solar LS) with $\lambda = 1.064$ μm, a pulse energy of 200 mJ and a pulse duration of 10 ns. A specific feature of the proposed MLA scheme is the possibility of using one laser with one output optical pulse to pump both the RA active medium and the MLA active element. The case in point is that the energy necessary to pump the active RA medium is no more than 30 mJ, and the energy required to pump the MLA active medium does not exceed 170 mJ. To implement efficient amplification of a chirped laser pulse, the optical pump pulse must be absorbed in the active medium immediately before the transmission of the amplified signal, because in this case the population inversion in the active medium is maximal, and the gain is directly proportional to inverse population. When the same laser is used to pump both the RA and MLA, optimal conditions for amplification are implemented in only the RA active medium, because a chirped pulse with $\lambda = 1.24$ μm arrives at the MLA only after ~500 ns (the chirped-pulse amplification time in the RA). For this time, because of the finite lifetime of the upper laser level (~3 μs [21]), the inverse population in the MLA active medium decreases, thus reducing the gain.

In the experiments aimed at studying the MLA amplification parameters, the active element was a Cr:forsterite crystal, cut in the form of a parallelepiped measuring $20 \times 5 \times 5$ mm along the crystallographic axes a, b, and c, respectively. The pump beam was directed along the a axis and polarised in the direction of the b axis. The end faces of the active element had a dielectric coating, which reduced the reflection of radiation with a wavelength of 1.24 μm from each surface to 0.5%. The crystal length was chosen to make the absorption of pump radiation with $\lambda = 1.064$ μm be at a level of about 90%; the absorption of radiation with $\lambda = 1.24$ μm did not exceed 10%. The measured absorption coefficients for the pump beam and the radiation with $\lambda = 1.24$ μm were, respectively, $\alpha_{1.06} = 1.15$ cm^{-1} and $\alpha_{1.24} = 0.05$ cm^{-1}. Thus, the crystal figure of merit

$$\mathrm{FOM} = \alpha_{1.06}/\alpha_{1.24} \quad (1)$$

amounted to 23.

The active-element housing was made of brass, with holes for coolant circulation. The coolant temperature was maintained at a level of 18.5 °C using a chiller.

To estimate the efficiency of amplifying a pulse of radiation with $\lambda = 1.24$ μm in the MLA with one pump laser (in order to form an inverse population in the RA and MLA active media), we measured the dependence of gain on the pump fluence. The amplification of a weak signal is described by the expression

$$E_{\mathrm{out}} = E_{\mathrm{in}}\exp[(g_0 - \alpha)L], \quad (2)$$

where E_{in} and E_{out} are, respectively, the energies at the input and output of the active element; L is the crystal length; and g_0 and α are, respectively, the gain and absorption coefficient of the amplified signal.

To obtain a more uniform distribution of inverse population in the active medium [22], the pump beam was split using a dielectric mirror with equal (50%) reflectance and transmittance into two beams and directed (by means of dielectric mirrors) to the active element from two sides. As was shown in [22], the maximum pump fluence corresponding to gain saturation is ~3 J cm^{-2} (at the end face of the active element). To provide this fluence, a positive lens with a focal length of 1 m was installed in each pump beam. The distance between the lens and active element was chosen to make the beam diameter on the crystal surface be 1.6 mm at a level of $1/e^2$. With this beam diameter and energy of 30 mJ for the pump pulse incident on the crystal end face, the desired pump fluence was obtained in each arm. The beam diameter for 1.24-μm radiation was 1 mm at a level of $1/e^2$. Figure 1 shows an experimental dependence of the gain of 1.24-μm radiation on the pump fluence. It can be seen that maximum gain is $g_0L = 0.68$, which corresponds to an increase in the pulse energy per pass through the active element by a factor of 1.96; however, because of the absorption of 1.24-μm radiation in the crystal, the energy increases by a factor of 1.78.

The MLA optical scheme is shown in Fig. 2. It includes plane mirrors, which provide fourfold transmission of a pulse of radiation with $\lambda = 1.24$ μm through the active element. To provide the optimal beam size in all passes, the divergence of the 1.24-μm beam was corrected (using a 1.25$^{\times}$ telescope) to make, with allowance for the thermal lens formed in the active medium as a result of pump beam absorption, the fluence obtained by the fourth pass close to the saturation fluence. In this case, the effect of pump pulse energy instability on the stability of the output Cr:forsterite laser energy is much lower.

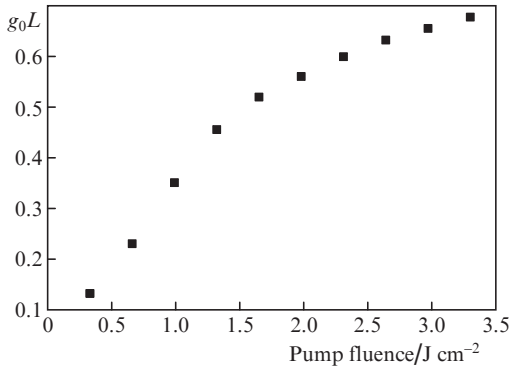

Figure 1. Dependence of the MLA active-element gain per pass on the pump fluence.

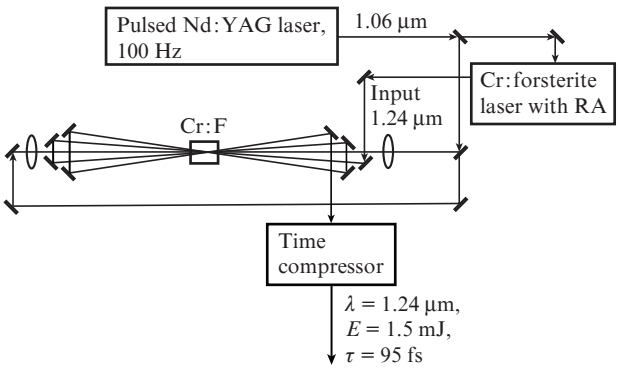

Figure 2. Optical scheme of a femtosecond Cr:forsterite laser system with a multipass laser amplifier.

To determine the optimal number of passes, we measured the active-element gain g_0 and the pulse energy after each pass (Table 1).

Table 1.

Number of pass	Energy/mJ	Gain/cm^{-1}
1	0.91	0.34
2	1.35	0.25
3	1.8	0.19
4	2.2	0.12

As follows from Table 1, the gain decreases by the fourth pass by a factor of almost 3 to a value of 0.12 cm^{-1}, and the pulse energy at the MLA output reaches 2.2 mJ; with a further increase in the number of passes, the pulse energy increases only slightly.

After amplification in the MLA, the radiation pulse was compressed by a time compressor, before which a 4$^{\times}$ telescope was installed; the latter served to increase the laser-beam diameter to 5 mm in order to limit the beam fluence on the diffraction grating at a level less than 100 mJ cm^{-2} (the destruction threshold for the diffraction grating coating) and to correct the beam divergence. The efficiency of the time compressor (having one diffraction grating) was ~70%, and the femtosecond pulse energy reached 1.5 ± 0.05 mJ. Figure 3 shows the measured autocorrelation function of femtosecond laser pulse, corresponding to a pulse FWHM of 95 ± 5 fs, and the pulse spectrum.

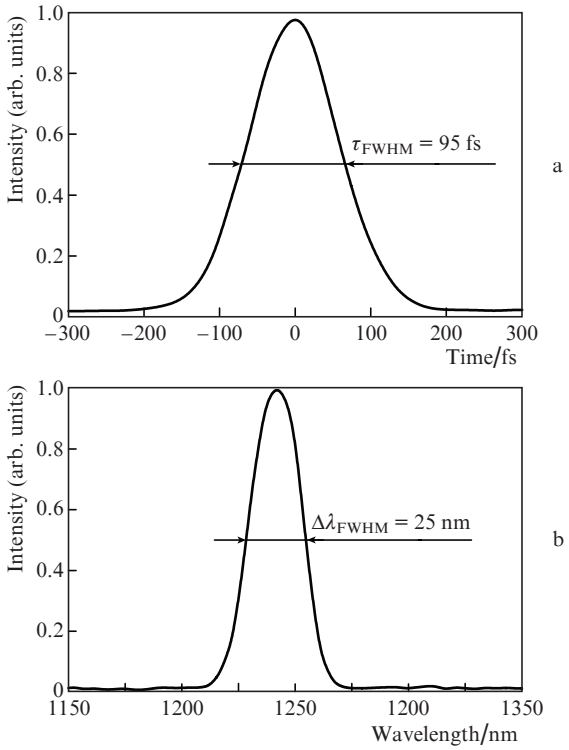

Figure 3. (a) Autocorrelation function of a femtosecond pulse from the Cr:forsterite laser system with MLA and (b) the spectrum of femtosecond laser pulse.

3. Generation of THz radiation in a nonlinear organic crystal

THz pulses with a repetition rate of 100 Hz were generated using optical rectification of femtosecond pulses from a Cr:forsterite laser system in a nonlinear organic crystal OH1 [(2-(3-(4-Hydroxystyryl)-5,5-dimethylcyclo-hex-2-enylidene) malononitrile]. A schematic of the experimental setup for generating THz pulses is presented in Fig. 4.

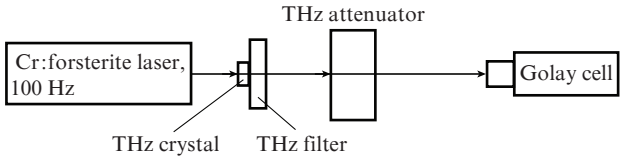

Figure 4. Schematic of the experimental setup for generating THz radiation.

The radiation from a Cr:forsterite laser system with λ = 1.24 μm was directed to an OH1 crystal with a diameter of 4 mm and a thickness of 420 ± 10 μm (Rainbow Photonics, Switzerland). A THz filter LPF8.8-47 (Tydex, Russia) was placed behind the organic crystal to cut off the pump radiation. A THz attenuator was applied to reduce the radiation energy when measuring the THz pulse parameters. To determine the optimal pump fluence for the crystal, we measured the dependences of the THz conversion efficiency and pulse energy on the pump fluence (Fig. 5). In the experiments, the THz pulse energy was measured using a calibrated Golay cell (optoacoustic detector GC-1D, Tydex).

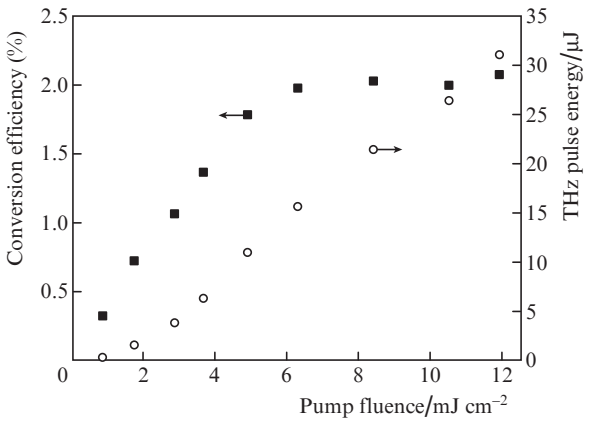

Figure 5. Dependences of the conversion efficiency and THz pulse energy in an OH1 crystal on the pump fluence.

It can be seen in Fig. 5 that the maximum conversion efficiency is ~2.1%; this value, obtained at a pump fluence of 6–7 mJ cm^{-2}, barely varies with a further increase in the pump fluence. At the same time, the THz pulse energy increases almost linearly up to the maximum pump fluence (12 mJ cm^{-2} for our experimental setup), at which the maximum THz pulse energy reaches 30 μJ.

Note that the organic crystal OH1 is damaged when the pump fluence exceeds 25 mJ cm^{-2}. Thus, the organic crystal OH1 used in the experiments provides even higher pulse energy at a higher pump fluence.

The use of a collimated laser beam for generating THz radiation in nonlinear organic crystals makes it possible to form a symmetric spatial intensity distribution, due to which the radiation can be focused well and high electric field strengths can be implemented. In experiments with focusing radiation by an off-axis parabolic mirror with a focal length of 50.8 mm, the THz beam radius, measured by the knife-edge method [23], turned out to be 410 μm at a level of 1/e^2 of maximum intensity. To achieve the minimum spot size when focusing, the THz beam was expanded using a telescope consisting of two off-axis parabolic mirrors with focal lengths of 15 and 152.4 mm, which increased the beam diameter from 4 to 40 mm.

To determine the THz pulse spectrum, we measured the pulse waveform by the method of electro-optic detection. A 200-μm-thick GaP(110) crystal was used as an electro-optic element, and a pulse with a wavelength of 1.24 μm was applied as a probe one. Figure 6 shows the THz pulse waveform and the corresponding spectrum. The pulse FWHM is ~0.5 ps, and the emission spectrum lies in the range of 0.3–2.5 THz. To exclude absorption of radiation by water vapour, all measurements of THz pulse parameters were performed in a housing filled with dry air (having a relative humidity less than 1.5%).

The electric field strength in the focal plane of focusing parabola, determined by measuring the THz pulse energy, the beam size in the focus, and the pulse width [24], was found to reach 3.5 MV cm^{-1}.

4. Conclusions

We demonstrated highly efficient generation of single-period THz pulses in the spectral range of 0.3–2.5 THz with a repetition rate of 100 Hz by means of optical rectification of femtosecond pulses from a Cr:forsterite laser system in a nonlinear organic crystal OH1. The conversion efficiency of laser pump energy into THz radiation was ~2.1%, and the pulse energy reached 30 μJ. Focusing a THz pulsed beam provided an electric field strength as high as 3.5 MV cm^{-1}. Using other organic crystals, such as DAST or DSTMS, one can expand the spectral range of THz source to 6 THz [15].

A relatively simple optical scheme for amplifying pulses from a Cr:forsterite laser system, operating with a pulse repetition rate of 100 Hz, was proposed. An advantage of this scheme over other schemes and methods is that it can be used to obtain higher energies at the corresponding laser pump energy and active element sizes.

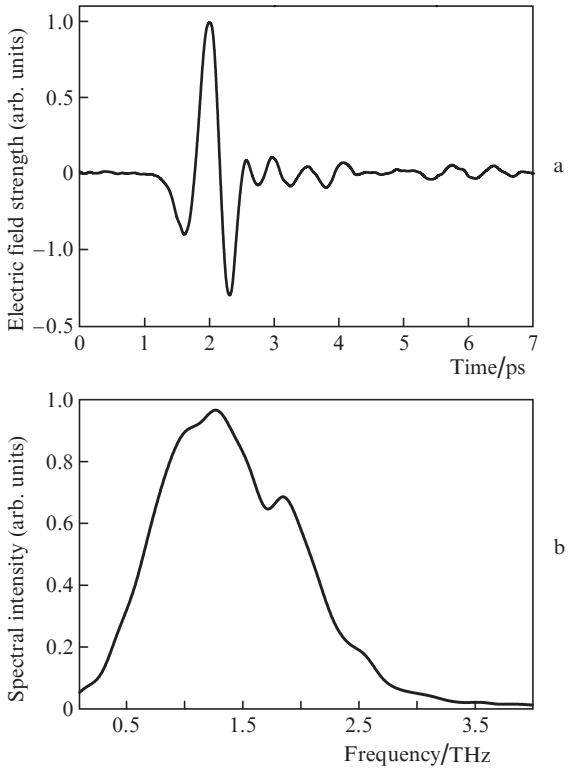

Figure 6. (a) Waveform and (b) spectrum of a THz pulse.

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