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Experimental Estimate of the Nonlinear Refractive Index of Crystalline ZnSe in the Terahertz Spectral Range

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Abstract—A modification of the *Z*-scan technique for measuring nonlinear refractive index n_2 in the terahertz spectral region is proposed. Measurements are made at a broadband terahertz radiation intensity of 0.8×10^9 W cm^{-2}. Coefficient $n_2 = 2.5 \times 10^{-11}$ cm^2 W^{-1} is estimated for semiconductor crystalline ZnSe.

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INTRODUCTION

Considerable progress has recently been made in developing systems for the generation of the high-intensity terahertz (THz) radiation [1, 2], allowing us to study nonlinear phenomena for waves of this spectral range [3–6]. The most important parameter characterizing the nonlinearity of a material's response in the field of intense waves is the nonlinear part of refractive index n, usually denoted as n_2 [7]. Its characteristic is determined using the relation

$$n = n_0 + n_2 I, \quad (1)$$

where n_2 is the nonlinear refractive index; n_0 is its linear part; and I is the intensity of radiation, which in relation (1) is assumed to be monochromatic. It was predicted earlier that the nonlinear response of crystals in the THz radiation field could theoretically be very strong, due to the great contribution from nonlinearity of an oscillatory nature [8]. It was shown experimentally in [6] that for the continuous THZ radiation of the FELIX free-electron laser, coefficient n_2 of some optical crystals does indeed differ substantially from the values obtained for them in the visible and IR spectral regions. However, the n_2 values themselves were not given in [6]. In [5], an indirect experimental estimate of n_2 at THz frequencies was given for a lithium niobate crystal; this also turned out to be substantial at around 10^{-11} cm^2 W^{-1}. In this work, we present the results from direct experimental measurements of the nonlinear refractive index of a ZnSe crystal in the THz spectral range. We estimate from the experiment that $n_2 = 2.5 \times 10^{-11}$ cm^2 W^{-1}. This value is about three orders of magnitude higher than 2.4×10^{-14} cm^2 W^{-1} for this crystal in the near infrared spectral range [9].

† Deceased.

EXPERIMENTAL

The optical scheme of our experimental setup for measuring the nonlinear refractive index of materials in a field of THz radiation in the spectral range is shown in Fig. 1a. we used femtosecond laser system *FA*, based on a regenerative amplifier, as our source of radiation (duration, 30 fs; pulse energy, 2 mJ; frequency of repetition, 1 kHz). The radiation was divided into two beams using beam splitter *BS*. One beam (pumping) was used to generate THz waves; the other beam (trial) was used to control their parameters. The THz radiation was generated and collimated in generator *TG* (TERA-AX, Avesta Project), based on the phase-optical rectification of femtosecond pulses in a lithium niobate crystal. The intensity of the THz radiation was adjusted by reducing the intensity of the pump beam with a polarizing attenuator *PA*. The temporal shape of field E (I) and the spectrum (II) of the generated THz wave is shown in Fig. 1b. It can be seen from the figure that the THz radiation was only one complete oscillation of the wave field, the spectrum of which ranged from 0.1 to 2.5 THz.

The THz pulse was focused and collimated with two parabolic mirrors (*PM*1 and *PM*2) using a focal length of 12.5 mm. The intensity of radiation in the caustic of the THz beam reached 0.8×10^9 W cm^{-2}. Sample *S* was moved along the caustic region using a motorized linear translator. The polarization of the THz radiation was linear and vertical. The ZnSe sample was 0.3 mm thick and had orientation 100. The sample was initially placed at a distance of 1.5 mm from the waist, which corresponded to the linear mode of propagation, and then scanned along the z axis through the focal region of the THz beam.

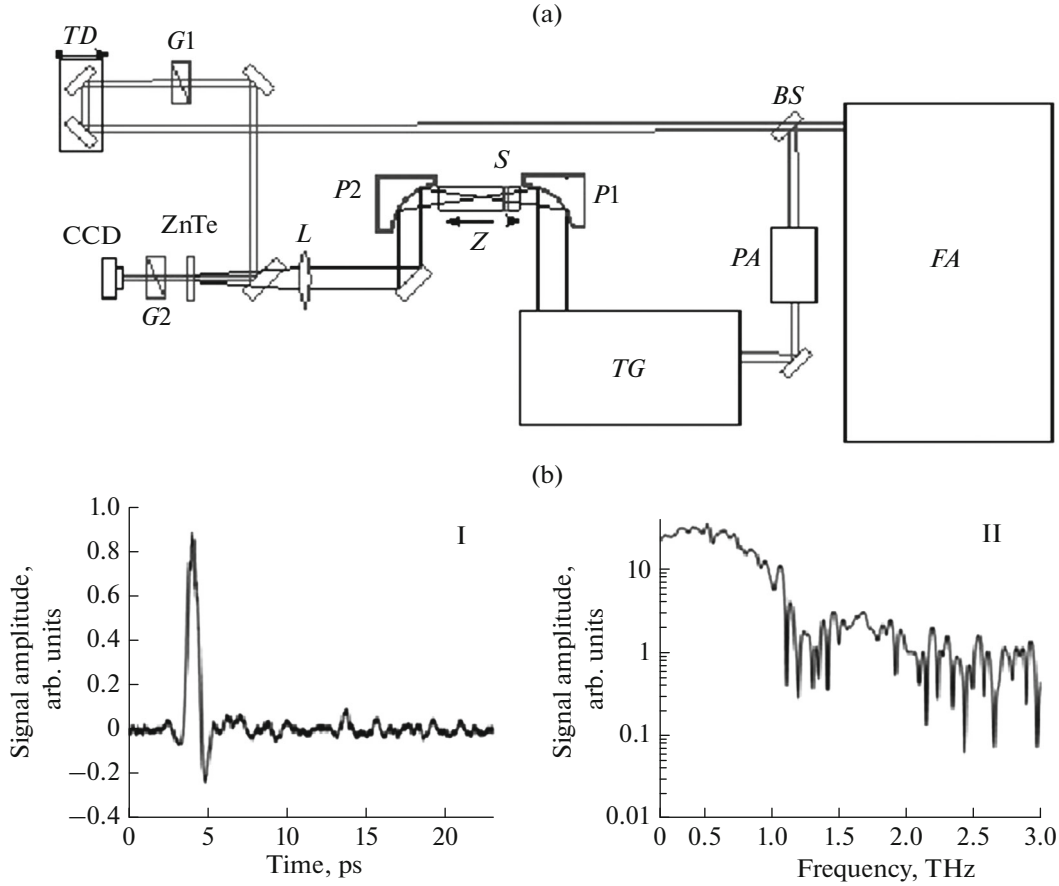

Fig. 1. (a) Optical scheme of the experimental setup: *FA* is the femtosecond amplifier; *BS* is the beam splitter; *PA* is the polarizing attenuator; *TG* is the THz generator; *P*1, *P*2 are the parabolic mirrors; *L* is the THz lens; *S* is a sample; *TD* is the adjustable time delay; *G*1, *G*2 are the polarizers (Glan prisms); CCD is the CCD chamber. (b) Temporal shape (I) and spectrum (II) of a THz pulse generated by the TERA–AX system.

We measured the amplitude of the THz radiation using the electro-optical effect in a ZnTe crystal positioned between crossed polarizers *G*1 and *G*2. The THz and femtosecond (trial) pulses overlapped in space and time on a ZnTe crystal. The THz radiation was initially blocked, and the second Glan *G*2 prism was oriented so that the two prisms were crossed for the test beam. we then switched on the THz radiation and, using delay line *TD* in the test beam, found the maximum intensity of the test radiation (corresponding to the maximum modulus of the THz pulse amplitude) passing through the crossed polarizers. we simultaneously fixed the position of the delay line and thus the position of the THz pulse in time with respect to the test pulse. As in a conventional *Z*-scan, we moved the ZnSe crystal along the z axis. Unlike a classical *Z*-scan, where the change in energy in the central part of the beam is measured, we determined the change in intensity for a femtosecond (trial) beam on a CCD camera in the region of the maximum modulus of the amplitude of the THz pulse. The image from the CCD camera was processed by selecting a region near the maximum of the image and summing the pixel signal contained in this area. This value was then taken as that of the THz signal amplitude with respect to the z axis.

RESULTS AND DISCUSSION

The *Z*-scan curves for the different intensities of THz radiation are shown in Fig. 2. The vertical axis shows the change in the deviation of the maximum intensity: $\Delta I = 8 \times 10^8$ and 8×10^6 W cm^{-2} of the trial femtosecond radiation after it passed through the crossed polarizers surrounding the electro-optical crystal as the ZnSe crystal moved along the z axis along the waist of the THz wave beam.

It can be seen from Fig. 2 that the crystal's movement along the z axis notably altered the measured intensity of the test beam, which is typical of the ones obtained using known *Z*-scan techniques. This is due to the different divergences of the radiation at different positions in the caustic of the crystal plate in which a nonlinear lens is induced by the field of the THz radiation. We used the standard formulas of the *Z*-scan

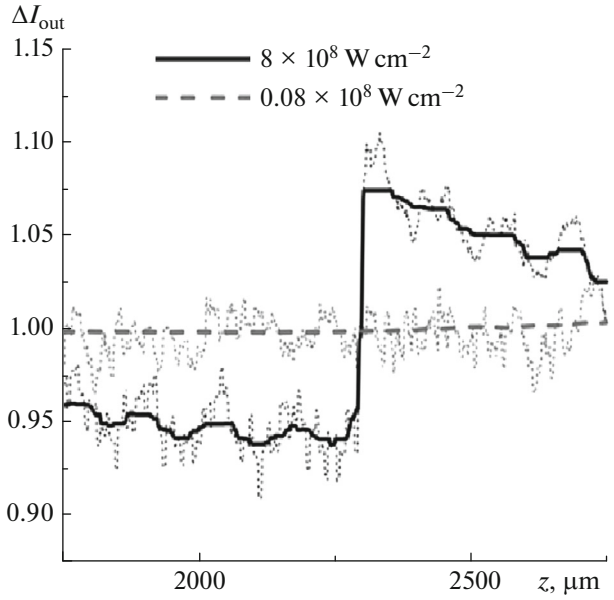

Fig. 2. Z-scan curves for two different values of THz radiation peak intensity. Solid and dashed lines give the averages for individual measurements represented by points.

technique in [9, 10] to estimate the n_2 of the ZnSe crystal from our measurements shown in Fig. 2:

$$n_2 = \frac{\Delta T}{0.406 I_{in}} \cdot \frac{\sqrt{2}\lambda}{\left(2\pi L_\alpha (1-S)^{0.25}\right)}, \qquad (2)$$

where ΔT is the difference between maximum and minimum transmittance; S is the aperture linear transmittance; L is the sample thickness; $L_\alpha = \alpha^{-1}[1 - \exp(-\alpha L)]$ is the effective length of interaction; α is the absorption coefficient ($\alpha = 0.85$ см$^{-1}$); λ is the wavelength; and I_{in} is the intensity of the input radiation. The result from our estimate using formula (2) was $n_2 = (2.5 \pm 1.0) \times 10^{-11}$ cm^2 W^{-1}.

CONCLUSIONS

Analysis of our experimental results shows we obtained a fairly high value of coefficient n_2 for crystalline ZnSe in the THz spectral region. This value was orders of magnitude greater than that of typical materials measured at optical frequencies. Our experimental estimates were consistent with the theoretical prediction of [8], which attributes the origin of THz nonlinearities in crystals to their vibrational response, which is orders of magnitude greater than typical electronic responses in the visible range. However, the authors of that work notably referred to their results only as experimental estimates of n_2, and not the results from measuring this quantity. Formula (2) is, after all, strictly valid only for quasi-monochromatic radiation, while the THz radiation in our experiments was a pulse from just one complete oscillation of the radiation field with a very wide spectrum. For such waves with very few oscillations, the effects of self-action differ considerably from the usual effects for quasi-monochromatic waves [12]. A theory of the Z-scan technique for waves from very few oscillations has yet to be developed. At the same time, however, we emphasize that the importance of the results of this work is, in our opinion, that they convincingly demonstrate the possibility of observing the effects of THz radiation's self-action at an intensity on the order of 5×10^8 W cm^{-2}. Such intensities of THz radiation have already been achieved in many laboratories.

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