

The nonlinearity of the refractive index of optical media in the terahertz spectral range

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Introduction

Recently, great progress has been made in the generation of high-intensity terahertz (THz) radiation, which has led to the opportunity to explore non-linear processes in the THz frequency range [1]. A key parameter characterizing the nonlinear response of a material is the non-linear refractive index, denoted as n_2 . It has been predicted theoretically [2] that the THz nonlinear response of crystals is extremely large as a consequence of a strong vibrational contribution. An indirect assessment of n_2 for lithium niobate at THz frequencies was reported in [3], leading to a value of approximately $10^{-11} \text{ cm}^2/\text{W}$. We find that $n_2 = 4.0 \times 10^{-11} \text{ cm}^2/\text{W}$, which is approximately 3 orders of magnitude larger than the value $2.4 \times 10^{-14} \text{ cm}^2/\text{W}$ in the NIR spectral range [4].

The present paper is devoted to theoretical analysis and a direct experimental measurement of n_2 for crystalline ZnSe in the THz spectral range. In this paper, we also consider the special aspects of self-action of single-cycle waves in media with given nonlinearities. It is shown that self-focusing efficiency in transparent dispersive medium decreases when the number of oscillations in input radiation is decreased for few-cycle paraxial waves with the same maximum electric field and transverse size, i.e., with the same ratio of the radiation power to the critical self-focusing power.

Experimental setup

The experimental setup is shown in Fig. 1. We used a femtosecond laser system (Regulas 35f1k, Avesta Project, 30 fs duration, pulse energy 2 mJ, repetition rate 1 kHz) as our radiation source. The radiation was split into two beams; one (pump) beam was used for generating the THz radiation and the other (probe) beam was used for monitoring the THz radiation. Generation of the THz radiation took place in the generator (TERA-AX; Avesta Project), which is based on phase-matched optical rectification in a lithium niobate crystal. The intensity of the THz radiation was regulated by attenuating the intensity of the pump beam through use of a polarization attenuator. Temporal (I) and spectral (II) form of the THz pulse generated by TERA-AX are shown on fig 1 (b). The radiation spectrum extended from 0.1 to 2.5 THz.

The THz pulse was focused and collimated using two lenses ($L1$ and $L2$) each with a focal length of 12.5 mm. The intensity of the radiation at the waist was $8 \times 10^8 \text{ W/cm}^2$. The sample was translated through the beam waist region of the THz beam through use of a translation stage. The ZnSe sample was 0.3 mm thick and oriented in the 100 direction. Initially, the sample was

placed at a distance of 1.5 mm from the waist, which corresponds to the linear propagation regime, and was then scanned through the focal region.

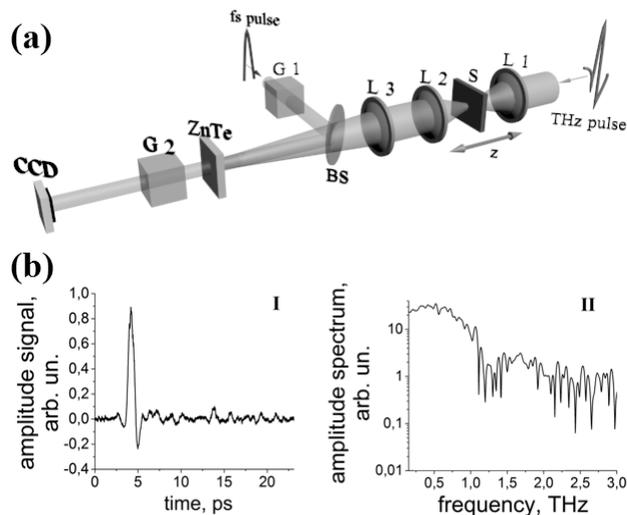


Fig. 1. The experimental setup (a). Temporal (I) and spectral (II) form of the THz pulse generated in the TERA-AX system (b). ($L1$, $L2$, $L3$ are THz lenses; S is the sample; BS is the beam splitter; $G1$, $G2$ are polarizers (Glan prism); CCD is the camera)

We measure the amplitude of the THz radiation by means of the electro-optical effect in a ZnTe crystal that is placed between the crossed polarizers. The THz and fs laser (probe) pulses were made to overlap in space and time at the ZnTe crystal. Initially the THz radiation was overlapped, and the second Glan prism $G2$ oriented so that two prisms are in crossed polarization for the probe beam. Next, we opened the THz radiation and using a delay line we found the maximum intensity of the fs probe radiation (corresponding to the maximum of the amplitude module of the THz pulse) which passes through crossed polarizers and fixed delay position and thus THz pulse position regarding the probe pulse.

Then, as in the conventional Z-scan method, we translated the ZnSe crystal along the z axis. In distinction from the classical Z-scan method, in which one measures the change of energy in the central part of the beam, we fix the value of the change of intensity of probe beam at the CCD camera in the area of maximum of the amplitude modulus of the THz pulse. Image processing with CCD-camera is performed as follows: we select an area in the vicinity of the resulting image. We sum the signal from these pixels, and we take this value to represent the amplitude modulus of the THz signal for this particular location in z .

Results

Z-scan traces for different intensities of the THz radiation are presented in Fig. 2. The vertical axis gives the maximum intensity of the probe fs radiation after passing through the crossed polarizers surrounding the electro-optic crystal. As we see from Fig. 2, translation of the ZnSe crystal along the z-axis leads to a variation of the measured intensity that is characteristic of all Z-scan measurements. We thus use the standard formulas [5] for Z-scan measurements to estimate the non-linear refractive index.

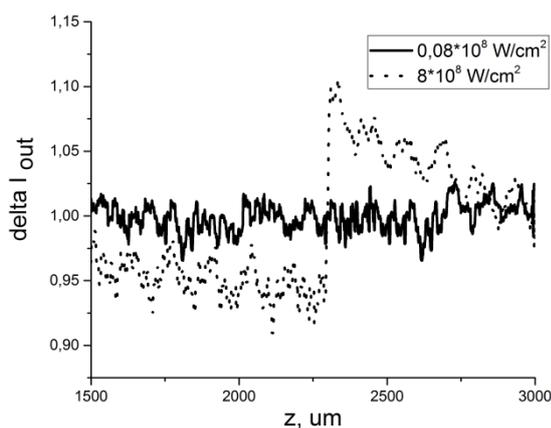


Fig. 2. Z-scan curves for different values of THz radiation intensity

We find that $n_2 = 4.0 \pm 2.5 \times 10^{-11} \text{ cm}^2/\text{W}$.

Summary

We measure an extremely large value of the n_2 coefficient of crystalline ZnSe in the THz spectral regime. This value is orders of magnitude larger than that of typical materials as measured at optical frequencies. Our results are consistent with the prediction of a recent [2] theoretical model that ascribes the origin of THz nonlinearities in crystals to a vibrational response that is orders of magnitude larger than typical electronic responses.

There is a large error in our measured value of $n_2 = 4.0 \pm 2.5 \times 10^{-11} \text{ cm}^2/\text{W}$. We believe that the error is largely methodological, resulting for example from shot-to-shot variations in our laser intensity

In evaluating the coefficient of nonlinear refractive index we used the formula for quasi-monochromatic radiation [5], although our THz pulse was only a few cycles in duration. It is known that the nature of nonlinear effects changes substantially with the decrease in the number of oscillations in the pulse [5]. We are presently in the process of investigating the influence of pulse duration on our results. For example, in Fig. 3 you can see the dependence of self-focusing efficiency on number of oscillations N in input THz radiation. Here the case $N = 0.3$ corresponds initial single-cycle THz radiation.

Nonetheless, our laboratory measurements show that n_2 in the THz spectral range is 3 orders of magnitude larger than the value for the NIR spectral range.

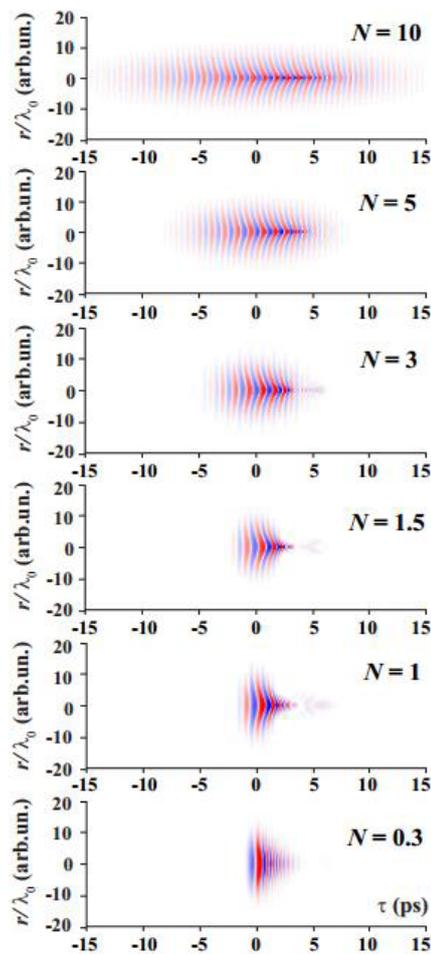


Fig. 3. Dependence of self-focusing efficiency on number of oscillations N in input THz radiation

The results are confirmed by theoretical calculations. A nonlinear response this large holds enormous promise for myriad applications of THz nonlinear optics.

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