Two-photon absorption in THz electro-optical sampling crystals

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Abstract. In this paper we present results of experimental study of two-photon absorption (2PA) induced in electro-optical crystals ZnTe and ZnSe by intensive optical radiation in 450–950 nm wavelength range. These crystals are of interest due to common usage in THz systems. Femtosecond pump-probe spectroscopy with the supercontinuum source helps to induce and detect different types of non-linear processes such as 2PA and to estimate nonlinear part of refractive index.

1. Introduction
Residing in the frequency scale between IR and millimeter spectral ranges, THz radiation provides wide possibilities for studying molecular structures of materials because most vibration and rotation spectra of molecules fit this range [1]. The application of optoelectronic techniques to the generation and detection of THz radiation is now well established. Widespread use of powerful laser systems for ultrashort pulses generation [2] leads to a non-linear electro-optical response of semiconductor crystals. It is important to note that this response is important when we use samples as detectors: arising nonlinearities result in data corruption [3]. For THz emission: photo induced charge carriers screen a significant part of the generated THz radiation in the crystal thereby limiting the conversion efficiency.

Two-photon absorption is one of the main processes influencing on electro-optical sampling of semiconductor crystals. There has been observed reduction of the intensity of light propagated through the crystal and the ultra-fast response [4] of semiconductor materials. In this paper we introduce experimental results of nonlinear processes studied with femtosecond pump-probe supercontinuum spectroscopy. ZnTe [5] and ZnSe wide gap semiconductor crystals used as THz radiation sources and detectors are tested. Using supercontinuum 2PA absorption dynamics and nonlinear part of refractive index for 450–950 nm range is detected.

2. Experimental study of two-photon absorption
To investigate the nonlinear response of ZnTe and ZnSe semiconductor crystals set of experiments on the femtosecond pump-probe spectroscopy setup (Fig. 1) was held. As the result we get dependences of the absorbed radiation incident on crystals on the time delay between pulses. The main feature of
this setup is use of supercontinuum as a probe beam. It provides insight into induced nonlinear properties of media in 350-1400 nm range.

First results using this technique are presented in paper [6]. Pump-probe supercontinuum spectrometer was based on a Ti: Sapphire regenerative amplifier (Avesta Regulas 35F1K), which produces 30 fs pulses with energy up to 2.3 mJ and repetition rate of 1 kHz. Beam was split (BS1) into two parts: one was sent through the delay line and was used as a pump beam, while the second was used for supercontinuum [7] (SC) generation and after that as a probe one. Mirrors $M_1$, $M_2$, installed on a linear translator with stepped motor controlled by a computer, were used as an optical delay line. The maximum delay time between pump and probe pulses was 660 ps with the delay step of 6.6 fs. To produce high-intensity ultra-wide SC in the IR and UV spectral regions a water jet was applied. The supercontinuum generator consisted of $L_2$ lens with 10 cm focus length and 1.8 mm width water jet placed 4 mm before the focus of $L_2$ lens. $L_3$ lens with 5 cm focus length collimated the resulting spectral supercontinuum. Both beams passed through a quartz lens $L_1$ with a focal length of 15 cm and were combined in space on a sample located at distance of 7.5 cm from the lens. The probe beam was registered by spectrometer ASP-100M ($SP1$).

By using pump-probe technique, we recorded pump-probe scans, pumping at 800 nm and probing at different frequencies and power levels of beams. In fig. 2 some results for ZnSe are presented. Dynamics of transmission of probe wavelengths 550, 650 and 900 nm for 50 mW pump pulse are indicated in the graph. As it illustrated there is a peak of 2PA and it changes it’s width and position. Shift of band peak dependence on probe wavelength can be used to quantify refractive index dispersion. Band width at half-maximum wave dependence helps to determinate relaxation of induced excitation in material.
2.1. Characteristics of samples

As it was mentioned above, in this paper we present results of investigation of intrinsic semiconductor crystals ZnTe and ZnSe, plane plates with orientation and width as shown in Table 1. These are crystals with the typical A2B6 band structure and vibrational subsystem.

Table 1. Parameters of ZnSe and ZnTe crystals.

| Orientation | Thickness (μm) |
|-------------|----------------|
| ZnSe        | [100] 560      |
| ZnTe        | [100] 710      |

3. Nonlinear properties of crystals

Using wavelength evolutions of 2PA absorption bands at half-maximum for ZnSe, ZnTe and GaP crystals, extracted from various power dependences, and with knowledge of sample thickness, we are able to obtain nonlinear index of refraction. Dispersion of refractive index for different pump power levels and dispersion under ordinary conditions [8] are shown on Figure 3.

As it can be seen from the refractive index dispersion graph, at the pump power level of the order of 10 GW/cm², the dispersion of the refractive index unalters with slight pump change. When the pump power level increases up to 45 GW/cm², the dispersion of the refractive index changes drastically. This can be explained by the activation of the nonlinear part of the refractive index $n_2$.

We can determine the values of the nonlinear part of the refractive index $n_2$:

$$n_2 = \frac{\Delta n}{\Delta I}$$  \hspace{1cm} (1)

In Table 2 results of estimated values of $n_2$ are presented as well as values obtained in earlier studies [9-11].
Figure 3. Dispersion of refractive index for different pump power levels.

The results are close to each other, which confirms the motivation for the use of the proposed scheme of femtosecond pump supercontinuum probe spectrometer.

Table 2. The values of the nonlinear part of the refractive index $n_2$.

|        | $n_2$, cm$^2$/W |
|--------|-----------------|
|        | 532 nm          | 532 nm, exp. | 780 nm | 780 nm, exp. | 810 nm | 810 nm, exp. |
| ZnSe   | $-4.4 \times 10^{-11}$ [9] | $-1.6 \times 10^{-11}$ | $10 \times 10^{-12}$ [10] | $6.5 \times 10^{-12}$ | -        | -        |
| ZnTe   | -               | -          | $5 \times 10^{-11}$ [11] | $7.4 \times 10^{-12}$ | $1.5 \times 10^{-12}$ [12] | $7 \times 10^{-12}$ |

4. Conclusion

With supercontinuum in a pump-probe spectroscopy system, we investigated the behavior of two-photon resonance at different wavelengths from 450 - 950 nm in commonly usable electro-optic semiconductor crystals ZnTe and ZnSe. Varying pump power, we are able to estimate nonlinear index of refraction, which is significant in THz electro-optical sampling.

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