

Two-photon absorption in zinc tellurite glass of composition 70TeO₂–30ZnO

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Abstract. The coefficient of nonlinear two-photon absorption in zinc tellurite glass of composition 70TeO₂–30ZnO is measured at a wavelength of 523 nm; its value amounts to 3.1 cm GW⁻¹.

Keywords: two-photon absorption, zinc tellurite glass.

Some of the relatively new optical nonlinear materials are TeO₂–ZnO zinc tellurite glasses having a wide transparency range from ~0.4 to 7 μm [1], which, in combination with good optical-mechanical properties, makes them attractive for quantum electronics. Previously, the measurement of the intensity of Raman spectra in the glasses of this system and their comparison with the spectra of silica glasses was reported in [2]. Recently, a report has been published on laser generation in thulium-doped zinc tellurite glass [3]. In the present study, the two-photon absorption coefficient in this glass was measured using the express technique proposed by us in [4, 5]. The technique is based on the measurement of the transmission nonlinearity in the excitation of glass by trains of picosecond laser pulses with smoothly varying intensity, which are emitted by a passively mode-locked repetitively pulsed solid-state laser.

To measure nonlinear absorption in 70TeO₂–30ZnO glass we used the second harmonic of the Nd:YLF laser, which has a radiation wavelength of 523 nm. The short-wavelength boundary of the transparency range of the glass is near the wavelength $\lambda_g \approx 400$ nm, so that the necessary condition for interband two-photon absorption ($2h\nu > E_g$, E_g is the bandgap width) is well satisfied. The duration of the laser pulses was 25 ps; the radiation was focused into the sample by a lens with a focal length $f = 20$ cm. The diameter of the laser spot in the focus was measured with a CCD camera and was 60 μm at half-height in intensity, and the beam radius w_0 in the lens focus at the 1/e level was 36 μm. The

length of the Gaussian beam caustic was $l_{\text{caus}} = 4\pi w_0^2/\lambda \approx 3.0$ m. The length of our sample, $L = 2.4$ mm, is many times smaller than the caustic length, so that the cross section of the laser beam can be considered constant along the length of the medium.

The oscillograms of radiation incident on the sample and passing through it (Fig. 1) were photographed in a single laser shot. The oscillogram clearly shows the nonlinearity of absorption: the attenuation of more ‘intense’ pulses is greater than that of less intense ones. The pulse amplitudes in the train were recorded using high-speed silicon pin photodiodes and a digital oscilloscope. The photodiodes were calibrated for the energy of radiation incident on the sample and passed through it, so that for each picosecond pulse we knew the energies E_{in} and E_{out} . The radiation energy was measured by a Molektron J3-05 pyroelectric detector.

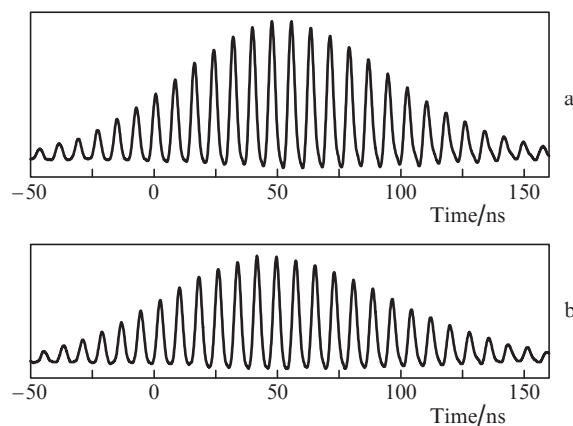


Figure 1. Oscillograms of pulsed radiation at (a) the input and (b) output of a sample of 70TeO₂–30ZnO glass.

With two-photon absorption, the change in the intensity I of light along the propagation axis z is given by

$$\frac{dI}{dz} = -\alpha I - \beta I^2, \quad (1)$$

where α is the coefficient of linear absorption; and β is the two-photon absorption coefficient. The solution of this equation has the form

$$I(z) = \frac{I_0 \exp(-\alpha z)}{1 + (\beta/\alpha) I_0 [1 - \exp(-\alpha z)]}, \quad (2)$$

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Received 13 December 2017; revision received 25 May 2018
Kvantovaya Elektronika 48 (8) 715–716 (2018)
Translated by I.A. Ulitkin

where I_0 is the light intensity at $z = 0$. Then the attenuation of light transmitted through a medium of length L is described by expression

$$\frac{I_0}{I} = \exp(\alpha L) + \beta \frac{\exp(\alpha L) - 1}{\alpha} I_0. \quad (3)$$

With two-photon absorption, the coefficient of attenuation of the light transmitted through the medium increases linearly with increasing input intensity I_0 . This dependence represents a straight line with a slope

$$b = \beta \frac{\exp(\alpha L) - 1}{\alpha}.$$

We estimate the two-photon absorption coefficient β by measuring this slope.

For pulsed radiation, formula (3) transforms into a formula for energy attenuation [6, 7]. In the case of a pulse with a Gaussian time profile, the attenuation as a function of energy E_{in} at the input to the sample is described by expression

$$\frac{E_{in}}{E_{out}} \approx \frac{\exp(\alpha L)}{T_{in} T_{out}} + \frac{1}{\sqrt{2}} \frac{2}{\sqrt{\pi}} \sqrt{\ln 2} \frac{\beta}{S_{eff} \tau} \frac{1}{T_{out}} \frac{\exp(\alpha L) - 1}{\alpha} E_{in}, \quad (4)$$

where T_{in} , T_{out} are the transmittance coefficients of the air/glass interface at the input and output of the glass sample; τ is the FWHM pulse duration; and $S_{eff} = 2\pi w_0^2$ is the effective cross-sectional area of the beam.

Figure 2 shows the dependence of the attenuation coefficient of the radiation energy passing through a sample of length $L = 2.4$ mm on the energy of the incident pulses. Since the laser generates a train of picosecond pulses with a smoothly varying intensity, we measure this dependence in one laser shot. The amplitudes of low-intensity pulses decrease by a factor of ~ 1.4 . Consequently, assuming the linear losses in the medium volume to be small ($\alpha = 0$), we find the transmission of the interface between the media: $T_{in} = T_{out} \approx 0.85$. The coefficient of energy attenuation increases due to the growth of two-photon absorption with increasing energy E_{in} of incident radiation. It is important to note that the pulses of the second (decreasing) half of the train have a smaller transmission than pulses with the same energy from the first (increasing) half of the train (Figs 1 and 2). This effect results from additional linear absorption in the medium, which is induced by two-photon absorption. Two-photon absorption of optical radiation in solids causes the excitation of electronic states in the conduction band. Such excitations accumulate with each subsequent pulse of laser radiation, which causes the growth of additional absorption from excited states. This explains the characteristic hysteresis in the dependence of Fig. 2. To determine the two-photon absorption coefficient, we measure the slope of the dependence in Fig. 2, using only pulses from the first half of the train. In our experiments, the slope is $b \approx 0.26 \pm 0.03 \mu\text{J}^{-1}$.

The calculated formula for the two-photon absorption coefficient

$$\beta = \sqrt{2} \frac{\sqrt{\pi}}{2} \frac{\tau}{\sqrt{\ln 2}} S_{eff} \frac{\alpha}{\exp(\alpha L) - 1} T_{out} b \quad (5)$$

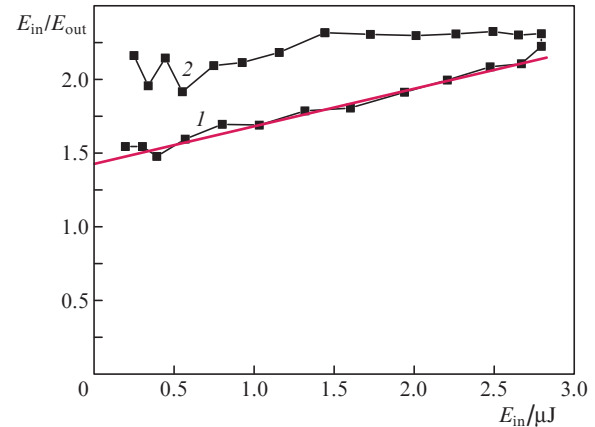


Figure 2. Dependence of the attenuation coefficient of the pulsed radiation energy E_{in}/E_{out} on the energy of the incident radiation E_{in} for a sample of $70\text{TeO}_2-30\text{ZnO}$ glass with a length $L = 2.4$ mm. The curves correspond to (1) increasing and (2) decreasing halves of the train. The straight line is a linear approximation.

yields $\beta \approx 3.1 \pm 0.4 \text{ cm GW}^{-1}$. This value is three times the same factor we measured for arsenic sulfide-based glass [8], as well as the values of β measured in oxide crystals, i.e. tungstates and molybdates [9]. Doping of glasses with an admixture of Cu^{2+} up to 2400 ppm [1] did not lead to a significant change in the coefficient β .

Acknowledgements. This work was supported by the Russian Foundation for Basic Research (Grant No. 16-02-00338).

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