NONLINEAR OPTICAL PHENOMENA

Two-photon absorption in SiO_2- and $(SiO_2 + GeO_2)$-based fibres at a wavelength of 349 nm

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Abstract. **The nonlinear two-photon light absorption coefficients have been measured in an optical fibre with a quartz glass (SiO_2) core and in a fibre with a germanosilicate glass ($SiO_2 + GeO_2$) core. The two-photon absorption coefficient β measured at a wavelength of 349 nm in the ($SiO_2 + GeO_2$)-based fibre (13.7 cm TW^{-1}) multiply exceeds that for the pure quartz glass optical fibre (0.54 cm TW^{-1}).**

Fused silica (quartz glass) is a popular optical material for UV applications. Under high-power laser excitation, along with linear loss, nonlinear two-photon absorption (TPA) in the UV range in fused silica can be comparable with linear absorption. Two-photon absorption in bulk fused silica samples and in SiO_2 fibres was investigated in a number of studies [1–5]. The TPA coefficients in fused silica were measured under coherent interband UV excitation at different wavelengths (see [1] and references therein). The importance of studying TPA in fused silica fibres is related, in particular, to the possibility of forming diffraction gratings in fibres exposed to UV laser radiation; these gratings are used as mirrors or spectral filters in fibre lasers. Two-photon absorption is believed to be one of the processes determining the photosensitivity of SiO_2-based glasses [6].

In [7, 8] we proposed a method for studying and analysing the TPA dynamics in crystals excited by a train of picosecond laser pulses with a gradually varying intensity. In this study we used this method to measure TPA coefficients in optical fibres based on quartz glass. The use of optical fibres, due to the large radiation – medium interaction length, makes it possible to reduce significantly the laser excitation power and thus expand the choice of TPA excitation sources. In addition, in contrast to the excitation of bulk media by focused beams, diffraction divergence of radiation is absent in fibre, and the beam cross section remains constant over the entire radiation – medium interaction length, due to which TPA coefficients can be measured with a higher accuracy.

Diffraction gratings are known to be efficiently recorded in silica fibres with a core doped with germanium dioxide [6]. In view of this fact, we chose two samples of step index multimode fibres for studying TPA. The first sample was a fibre

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with a core based on SiO_2 (KU-1 grade), 205 µm in diameter and 650 cm long, with a reflecting fluorine-doped SiO_2 cladding. The second sample had a core based on germanosilicate glass $SiO_2 + GeO_2$ (5 mol %), 50 µm diameter and 220 cm long, and a reflecting SiO_2 cladding. The TPA coefficients were measured at the wavelength $\lambda = 349$ nm, on the assumption that the necessary condition for interband TPA ($2h\nu > E_g$) is satisfied.

In the case of TPA, the light intensity I changes along the z axis in accordance with the equation

$$\frac{dI}{dz} = -\beta I^2, \tag{1}$$

where β is the TPA coefficient. If the linear absorption coefficient α is nonzero,

$$\frac{dI}{dz} = -\alpha I - \beta I^2. \tag{2}$$

The solution to this equation has the form

$$I(z) = \frac{I_0 \exp(-\alpha z)}{1 + (\beta/\alpha)I_0[1 - \exp(-\alpha z)]}, \tag{3}$$

where I_0 is the light intensity at $z = 0$. Then, the attenuation of light transmitted through a medium of length L can be written as

$$\frac{I_0}{I} = \exp(\alpha L) + \beta \frac{\exp(\alpha L) - 1}{\alpha} I_0. \tag{4}$$

Under TPA conditions, the attenuation of light transmitted through a medium linearly increases with an increase in the input intensity I_0. This is a straight-line dependence with a slope $b = \beta[\exp(\alpha L) - 1]/\alpha$. The coefficient β is determined by measuring this slope.

A schematic of the experiment is shown in Fig. 1. To obtain the attenuation I_0/I as a function of intensity I_0, we used third-harmonic pulses of a picosecond passively mode-locked Q-switched laser based on $YLiF_4$:Nd crystal ($\lambda = 1047$ nm) [7, 8]. The laser generated trains (~100 ns long) of picosecond pulses with a gradually varying amplitude; the pulse duration was 25 ps. The radiation was focused on a fibre end face by means of a quartz lens ($f = 3$ cm) with an antireflection coating.

Oscillograms of radiation pulses at the input and output of optical fibres were recorded in real time within one laser shot. To increase the measurement accuracy, we did not use the regime of signal accumulation over many laser shots. The
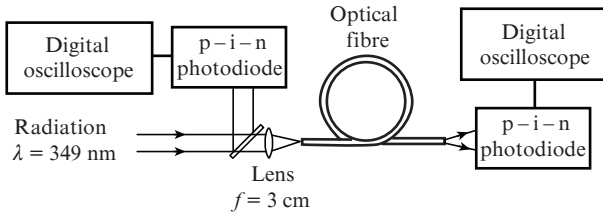
Figure 1. Schematic of the experimental setup.

amplitudes of train pulses were recorded using silicon p–i–n photodiodes and a Tektronix digital oscilloscope with a gain band of 1 GHz. The photodiode installed before the optical fibre was calibrated with respect to the energy of the radiation incident on the fibre end face. The radiation energy was measured by a J3-05 pyroelectric joulemeter (Molectron). For pulsed radiation, formula (4) is transformed into a formula for energy attenuation [9, 10]. Since laser pulses had a Gaussian temporal profile, the pulse energy attenuation can be written as

$$\frac{E_{in}}{E_{out}} \cong \frac{\exp(\alpha L)}{T_{in} T_{out}} + \sqrt{\frac{2\ln 2}{\pi}} \frac{\beta}{S\tau} \frac{1}{T_{out}} \frac{\exp(\alpha L) - 1}{\alpha} E_{in}, \qquad (5)$$

where E_{in}, E_{out} and T_{in}, T_{out} are, respectively, the pulse energies and air/glass interface transmittances at the fibre input and output; $T_{out} = T_{in} = 4n/(n+1)^2$; n is the refractive index of SiO_2; τ is the pulse FWHM; and S is the fibre core cross-sectional area.

The first sample (fibre with a 205-μm quartz glass core) attenuated low-intensity pulses by a factor of 4.83. This means that the linear absorption coefficient of a sample with a length $L = 650$ cm is $\alpha = 0.0024$ cm^{-1}. Figure 2 shows the measured dependence of the attenuation of radiation energy passing through the fibre on the incident pulse energy. The slope of this dependence is $b \cong 0.07$ μJ^{-1}. The energy attenuation increases (due to the enhancement of TPA) with an increase in the incident radiation energy. The formula for calculating the TPA coefficient,

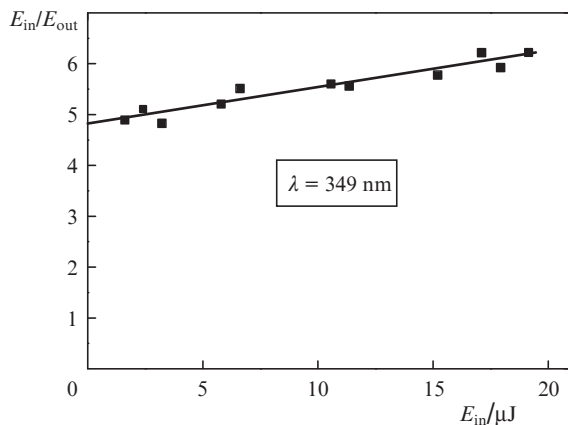$$\beta = \sqrt{\frac{\pi}{2\ln 2}} \frac{\alpha}{\exp(\alpha L) - 1} T_{out} S\tau b \qquad (6)$$

Figure 2. Dependence of the energy attenuation on the output of fibre with a SiO_2 core 205 μm in diameter and 650 cm long on the incident pulse energy.

yields $\beta = 0.54$ cm TW^{-1}. The density power in the fibre core $I_0 = 2.3$ GW cm^{-2} corresponds to an excitation pulse of duration $\tau = 25$ ps and energy $E_{in} = 20$ μJ (Fig. 2). At this intensity the nonlinear additive βI to the linear absorption coefficient α is ~0.001 cm^{-1}. The TPA coefficient in quartz glass at the wavelength $\lambda = 349$ nm is found to be an order of magnitude smaller than the value $\beta = 5$ cm TW^{-1} for a wavelength of 282 nm, which was obtained in [4].

The second sample (with a 50-μm germanosilicate glass) attenuated low-intensity pulses by a factor of about 200. For a sample length $L = 220$ cm, this attenuation corresponds to linear absorption coefficient $\alpha = 0.024$ cm^{-1}. Figure 3 shows a dependence of the attenuation of radiation energy passing through the fibre on the incident pulse energy, obtained by us; this dependence has a slope $b = 155$ μJ^{-1}. Formula (6) yields $\beta = 13.7$ cm TW^{-1}. A power density in the fibre core $I_0 = 4.8$ GW cm^{-2} corresponds to an excitation pulse of duration $\tau = 25$ ps and energy $E_{in} = 2.5$ μJ (Fig. 3). In this case, the nonlinear additive to the linear absorption coefficient is ~0.066 cm^{-1}.
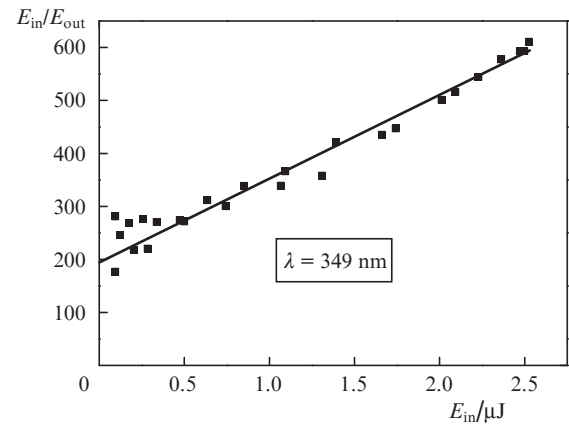

Figure 3. Dependence of the energy attenuation at the output of fibre with a core based on $SiO_2 + GeO_2$ (5 mol %), 50 μm in diameter and 220 cm long, on the incident pulse energy.

According to our measurements, the linear-loss coefficient at the wavelength $\lambda = 349$ nm in the fibre with germanosilicate core exceeds the corresponding parameter of pure quartz glass SiO_2 fibre by a factor of 10. Introduction of germanium oxide into a quartz glass core leads to the occurrence of absorption bands peaking at wavelengths of 240 and 330 nm. These bands are assigned to defects (oxygen-deficient centres) formed in the glass network [11].

The states of defect centres may serve as intermediate levels for two-step photon absorption. The existence of intermediate states may be responsible for the significant difference between the TPA coefficients in the sample with a $SiO_2 + GeO_2$ core and in the sample with a SiO_2 core [4].

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