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Anisotropy of two-photon absorption in BBO at 264 nm

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Abstract

Femtosecond pulses at $\lambda = 264$ nm were used to measure the two-photon absorption (TPA) coefficient in β -BaB₂O₄ (BBO) crystal. It was found that nonlinear absorption in BBO depends significantly on crystal cut and/or beam polarization. For an ordinary beam propagating along the optical axis ($\parallel c$) and perpendicular to it ($\perp c$) the similar values of TPA coefficient were obtained, namely $(68 \pm 6) \times 10^{-11}$ and $(66 \pm 7) \times 10^{-11}$ cm/W. For an extraordinary beam ($\perp c$) the TPA coefficient is significantly smaller, $(47 \pm 5) \times 10^{-11}$ cm/W. © 2001 Elsevier Science B.V. All rights reserved.

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With the development of high power UV lasers and their applications the knowledge of nonlinear absorption properties of optical media becomes increasingly important. Recently we have shown that the use of femtosecond UV pulses from the newly developed commercial laser system (Twinkle, Light Conversion Ltd., Vilnius, Lithuania) allows to measure the two-photon absorption (TPA) coefficient with high accuracy [1]. This was demonstrated in experiments with three types of commercial fused silica and one crystalline quartz sample. In this paper we report the accurate determination of TPA coefficient in β -BaB₂O₄ (BBO) crystal which is very important for second-harmonic and sum-frequency generation in UV range [2]. Until now, the TPA anisotropy was observed

in sphalerite-type cubic crystals GaAs [3,4], ZnS [4], ZnSe [4,5], ZnTe [4]; in chalcopyrite tetragonal CdGa₂S₄ crystal [6]; in ternary semiconductor AgGaSe₂ [7]; and in orthorhombic KTiOPO₄ crystal [3]. All these findings refer to visible or near IR range measurements. The nonlinear absorption in BBO at 264 and 211 nm was studied recently by two research groups [8–10], however, no TPA anisotropy was observed.

The BBO crystals were grown in the Institute of Monocrystals, Novosibirsk, Russia by TSSG method in Czochralski variant, using Na₂O as a solvent. The samples cut for the experiments were of size $6 \times 6 \times 3$ mm³ with smaller dimension along optical axis (z -cut) or perpendicular to it (x -cut).

In the experiment we used the set-up presented in Fig. 1. The UV radiation ($\lambda = 264$ nm, 27 Hz) from Twinkle laser was directed into the BBO sample. The use of long focal distance (454 mm) fused silica lens allowed the change of the beam

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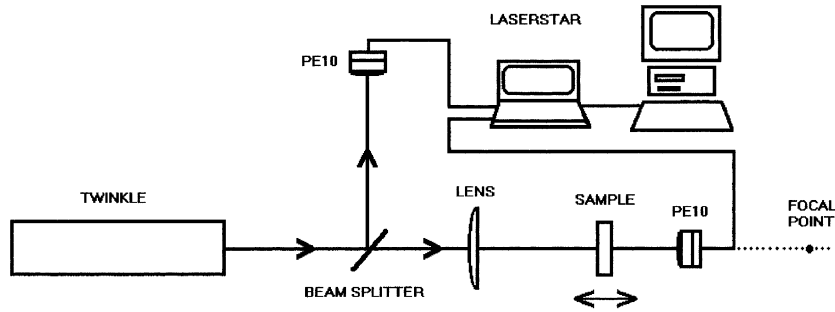


Fig. 1. The scheme of experimental set-up.

cross-sectional area (and hence the beam intensity) by moving the sample along the laser beam axis. However, the position of sample remained well in front of lens focal point, which helped us to avoid self-focusing (see Ref. [1] for details). The incident and transmitted pulse energies were measured by pyroelectric detectors PE10, connected to Laser-Star energy meter system (Ophir Optonics Inc.).

The same set-up was used for the linear absorption measurements with the samples placed in the reference beam possessing much lower intensity values (0.4 GW/cm^2) than the main one ($5\text{--}25 \text{ GW/cm}^2$). The linear absorption coefficient values at 264 nm were evaluated from experimental transmittance values by subtracting Fresnel reflection losses $(1 - R)^2$, where R is the reflectivity of a sample at normal incidence. The reflectivity value is defined through the well-known expression, $R = (n - 1)^2 / (n + 1)^2$, where n is the refractive index of sample corresponding to polarization of laser beam in the crystal. Using published BBO dispersion relations [11], it is easy to calculate the reflectivity values for BBO at 264 nm which equal to 0.07595 and 0.05520 for ordinary and extraordinary polarization, respectively. The experiments done showed that the linear absorption coefficient for ordinary beam (α^o) was $0.04 \pm 0.01 \text{ cm}^{-1}$ when propagating along z -axis and $0.06 \pm 0.003 \text{ cm}^{-1}$ when propagating perpendicular to z -axis. For the extraordinary beam (α^e) in x -cut BBO crystal a noticeably higher value of linear absorption coefficient, $0.1 \pm 0.003 \text{ cm}^{-1}$, was obtained. These data are in good agreement with the linear absorption data, presented in Ref. [12] ($\alpha = 0.04\text{--}0.15 \text{ cm}^{-1}$ at 266 nm). However, they contradict to the data of

Ref. [10] ($\alpha < 0.01 \text{ cm}^{-1}$ at 264 nm, which is unrealistic).

Twinkle laser system generates UV pulses with Gaussian shape in time and in space [13]. If we will choose the following representation for Gaussian beam (similar to that used in Ref. [14])

$$I_{\text{inc}}(r, t) = I_0 \exp \left[-2 \left(\frac{r}{w_0} \right)^2 \right] \exp \left[- \left(\frac{2t}{\tau_p} \right)^2 \right] \quad (1)$$

where I_0 is the maximum on-axis intensity, τ_p is the pulse width at the e^{-1} level, and $w_0/\sqrt{2}$ is the beam radius at e^{-1} level (at full width at half maximum (FWHM) $\tau = \tau_p \sqrt{\ln 2}$, $w = w_0 \sqrt{2 \ln 2}$), then after integration over space and time we will have for pulse energy ε_0

$$\begin{aligned} \varepsilon_0 &= \int_{-\infty}^{+\infty} dt \int_0^{+\infty} I_{\text{inc}}(r, t) 2\pi r dr \\ &= \frac{\pi \sqrt{\pi}}{4} I_0 \tau_p w_0^2 \end{aligned} \quad (2)$$

The energy transmittance through the nonlinear medium of length l in the presence of linear absorption is given by the expression (see, for example, Ref. [1]):

$$\begin{aligned} T &= \frac{\varepsilon_{\text{tr}}}{\varepsilon_0} \\ &= \frac{\alpha T_0}{\sqrt{\pi \beta I_0} (1 - R) (1 - e^{-\alpha l})} \\ &\quad \times \int_{-\infty}^{+\infty} \ln \left\{ 1 + \frac{\beta}{\alpha} I_0 (1 - R) (1 - e^{-\alpha l}) e^{-k^2} \right\} dk \end{aligned} \quad (3)$$

where α is the linear absorption coefficient, β is the TPA coefficient, $T_0 = (1 - R)^2 e^{-\alpha l}$ is the initial transmittance, and $k = 2l/\tau_p$.

In order to account for the change of maximum on-axis intensity due to focussing and focal distance prolongation in sample medium we should replace the I_0 value in previous equation by

$$I_0^* = I_0 \left[\frac{F - S}{F} - \frac{l}{2nF} \right]^{-2} \quad (4)$$

where F is the focal length, S is the distance between the focussing lens and the sample.

It is well known that accurate determination of TPA coefficient requires the precise knowledge of beam diameter and pulse width. The former was found by placing an aperture symmetrically to the axis of the beam and measuring the energy transmittance through it (see, for example, Ref. [15]). It could be shown that for our representation of Gaussian beam the beam diameter could be defined by the expression

$$w_0 = \frac{d}{\sqrt{2 \ln[(1 - A)^{-1}]}} \quad (5)$$

where d is the aperture diameter, A is the transmittance through the aperture. In experiment we used the apertures of four diameters (from 1.6 to 3.1 mm). The weighted average value for beam diameter was 2.56 ± 0.03 mm (3.01 ± 0.04 mm FWHM).

For UV laser pulse width measurement we employed TPA-based autocorrelation set-up [16], presented in Fig. 2. A fused silica wedge (1.5°) was used to divide the UV beam on the three parts. The

transmitted beam served as the pump, the reflected beam from the front surface was used as the probe and the reflected one from the rear surface served as the reference. The pump and probe beams were focussed by plano-convex fused silica lenses with focal distances 930 and 454 mm, respectively. The angle between the pump and probe beams was about 9°. The spatial coincidence of pump and probe beams was accomplished by employing 0.2 mm pinhole (Linos Photonics). The temporal coincidence of pump and probe pulses was achieved by optical delay line based on computer-driven 25 mm motorized micropositioner (Ealing) with resolution 0.02 μ m and unidirectional repeatability 0.1 μ m. The TPA signal was induced in a thin (0.4 mm) fused silica plate and monitored by two pyroelectric detectors PE10 and the LaserStar energy meter. The latter was interfaced to a computer controlling the data acquisition procedure. The sampling rate was 100 laser shots per data point. Only pulses within a particularly defined energy window were accepted in the averaging process, thereby avoiding strong fluctuations of the laser power to be accounted for in the measurement. A typical autocorrelation curve of a pulse at 264 nm is presented in Fig. 3 together with the fitting trace and the deviation between the fitting and measured values. The fitting of a Gaussian pulse was accomplished using a nonlinear least square procedure based on the Levenberg–Marquardt algorithm [17] and taking into account a linearly rising background:

$$A_{\text{fit}}(t) = A_0 \exp \left[- \left(\frac{t - t_{\text{max}}}{\tau} \right)^2 \right] + P_1 t + P_2 \quad (6)$$

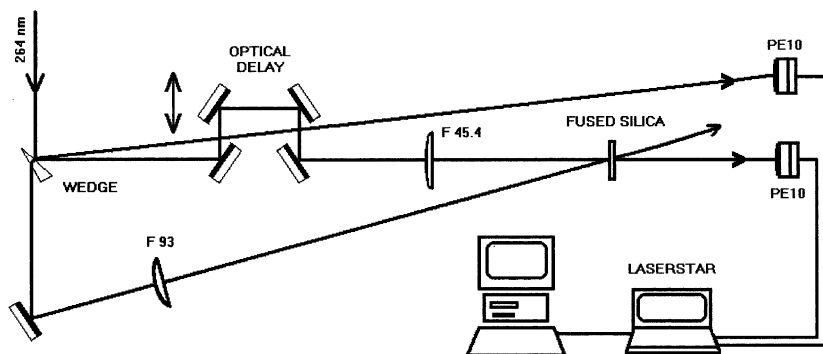


Fig. 2. The TPA-based autocorrelation set-up.

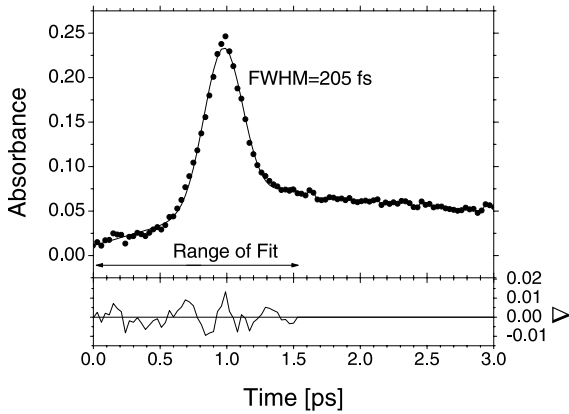


Fig. 3. The typical curve of sample absorbance versus the delay time between the pump and probe pulses. The solid line in upper part represents a Gaussian fit to the measured data according to Eq. (6) using the parameters $A_0 = 0.18$, $t_{\max} = 974 \pm 2$ fs, $\tau = 204.5 \pm 0.3$ fs, $P_1 = (43.9 \pm 1.4) \times 10^{-3}$ fs $^{-1}$, $P_2 = (53.8 \pm 0.9) \times 10^{-3}$ fs $^{-1}$. The lower part of the drawing shows the absolute deviation between calculated and measured values to demonstrate the validity of the fit.

where τ is the corresponding pulse duration at the FWHM, t_{\max} is the temporal position of the pulse maximum. The mean value of pulse duration for a series of four measurements was found to be $\tau = 220 \pm 10$ fs.

In the experiment we have measured the nonlinear transmittance at 264 nm for *z*-cut BBO and *x*-cut BBO for both ordinary and extraordinary beam polarizations. The typical experimental curves are shown in Fig. 4(a)–(c).

In order to fit the experimental results with theoretical transmittance curves described by Eq. (3) we used the software package MATHEMATICA (Wolfram Research Inc.). For each experimental value of nonlinear transmittance the program calculated the TPA coefficient value β_i corresponding to the theoretical curve passing through this point and the initial transmission value, determined from low intensity light experiments described above ($T_0 = 0.845$ for *z*-cut, $T_0 = 0.839$ for *x*-cut, ordinary beam, and $T_0 = 0.867$ for *x*-cut, extraordinary beam). In this procedure the experimental values R and α , mentioned in the text, were used as parameters. The overall β value was determined as arithmetic average of all β_i related to one curve. For each case of crystal cut (and/or

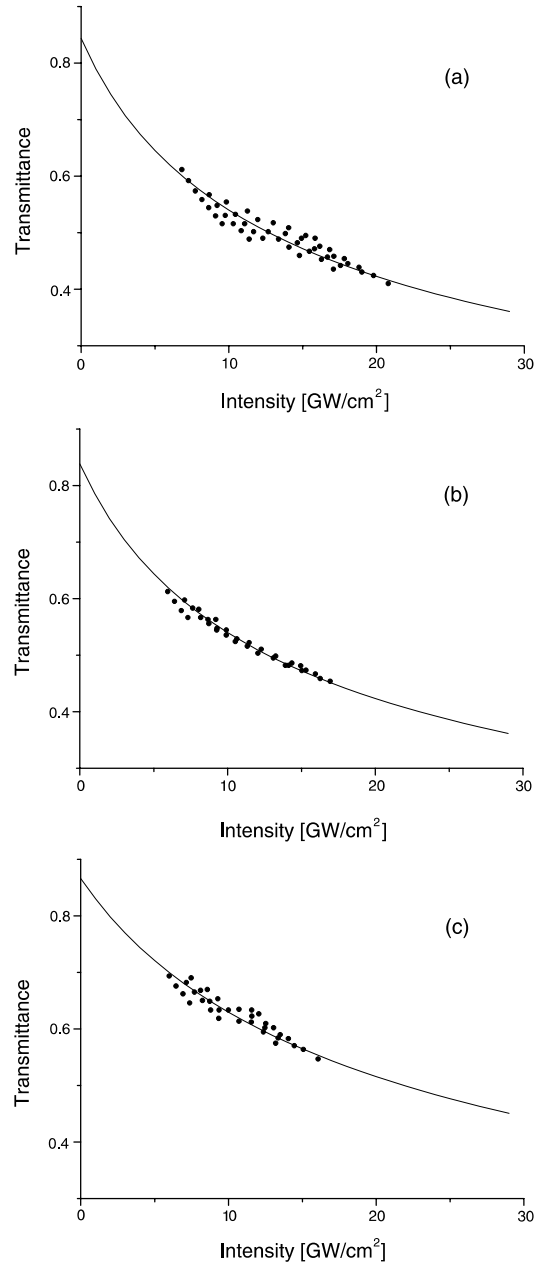


Fig. 4. The typical curves for nonlinear transmission-intensity dependence measured at 264 nm. The fitting gave the following values for BBO TPA coefficient: (a) 72.8×10^{-11} cm/W (*z*-cut), (b) 71.6×10^{-11} cm/W (*x*-cut, ordinary polarization) and (c) 45.0×10^{-11} cm/W (*x*-cut, extraordinary polarization).

light polarization) we made seven series of measurements. In Table 1 the overall TPA coefficients

Table 1
Experimental data on β values for BBO (in 10^{-11} cm/W)

$\parallel c$	$\perp c$	
	Ordinary beam	Extraordinary beam
72.8	54.1	44.2
72.4	60.4	42.3
60.6	59.6	42.3
66.4	71.6	47.0
64.7	70.4	45.0
76.3	66.2	56.5
60.0	76.0	54.5

for each recorded curve are presented. By averaging the overall β values the standard deviation, which corresponds to the relative uncertainty, can be determined. The absolute uncertainty of our β measurements (6%) was mainly determined by the uncertainty of the irradiance value, which combined from inaccuracies in energy calibration (3%), pulse width measurement (4.4%) and laser beam cross-sectional area determination (2.3%). Therefore, total uncertainty of β determination in our experiments was about 10%. The averaged TPA coefficient for an ordinary beam propagating along the optical axis is $\beta = (68 \pm 6) \times 10^{-11}$ cm/W, for an ordinary beam propagating perpendicular to optical axis $\beta = (66 \pm 7) \times 10^{-11}$ cm/W and for an extraordinary beam $\beta = (47 \pm 5) \times 10^{-11}$ cm/W.

From our results it follows that BBO crystal possesses a significant anisotropy of TPA coefficient which was not mentioned before. In Ref. [8] the measured β value for BBO along the optical axis $((90 \pm 20) \times 10^{-11}$ cm/W) agrees with our values for z -cut $((68 \pm 6) \times 10^{-11}$ cm/W) and x -cut, ordinary polarization $((66 \pm 7) \times 10^{-11}$ cm/W). In Ref. [9] the TPA coefficient for BBO at 264 nm was estimated relatively to that of KDP. The BBO crystal was cut at 48° to the optical axis, and the extraordinary polarization of light was used. The obtained value $\beta = 61 \times 10^{-11}$ cm/W (no uncertainty was given) agrees with our value $\beta = (47 \pm 5) \times 10^{-11}$ cm/W. In Ref. [10] the BBO crystal was cut at 30° to the optical axis. The authors used different light polarizations for measurements, however, they had not noticed any difference between β values for ordinary and extraordinary polarizations, probably due to relatively large

uncertainty of their measurements ($\sim 35\%$). The TPA coefficient value reported in that work $((93 \pm 33) \times 10^{-11}$ cm/W) agrees favourably with our findings.

In conclusion, we have measured the TPA coefficient for BBO at 264 nm with higher accuracy than before. It was found that TPA process in BBO depends significantly on crystal cut and/or laser beam polarization.

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