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Measurement of the nonlinear index n_2 of BSO crystals

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Photorefractive materials are now widely studied for their large nonlinear optical properties. Large gain values in two-beam coupling or degenerate four-wave mixing experiments are easily available using a low-power continuous laser beam, making them attractive materials for such uses as real-time holography,¹ reconfigurable optical connections,² and gain media for self-oscillating laser cavities.³

For these crystals, the photoinduced variation of the refraction index can arise from two different origins. The first one is the photorefractive effect (PRE), the second is a nonlinear optical Kerr effect related to the third-order susceptibility tensor. In the case of the PRE the index variation results from the building of a space charge field created by a separation of positive and negative charges inside the crystal. Often considered as slow materials, response times in the nanosecond time scale are now demonstrated for BSO⁴ crystals. Even BaTiO crystals and doped photorefractive semiconductors exhibit response time in the picosecond range. On the contrary, in these materials, the n_2 component of the refraction index ($n = n_0 + n_2 I$, where I is the incident light intensity and n_0 is the linear component of the index) is believed to arise from electronic effects and is expected to have a response time in the femtosecond time scale. Unfortunately, the magnitude of the nonlinear coefficient n_2 is not well determined. We present the results of measurement of n_2 for undoped BSO crystals in the femtosecond domain.

To measure the n_2 coefficient we use a classical optical Kerr effect setup described in Fig. 1. In this configuration, an intense laser beam linearly polarized induces a variation of the refraction index in the direction of the optical field. We detect the index variation by measuring the transmission of a weak probe through the crystal placed between polarizer and analyzer. The probe beam is polarized at 45° from the pump beam. The pump and probe beams are produced by a colliding pulse mode-locked dye laser with internal Brewster prisms for group velocity compensation and amplified by a frequency-doubled Nd:YAG laser pumping a chain of dye amplifiers. That produces single amplified pulses of 500- μ J energy at a 620-nm wavelength and a 10-Hz repetition rate. At the exit of the amplifier the pulse duration is ~ 150 fs. The two beams are focused on the sample to an area of $\sim 3 \times 10^{-4}$ cm². The temporal coincidence between the two pulses can be adjusted with 15-fs accuracy by means of an optical delay line. The analyzer is a Glan prism with antireflection coatings, allowing one to obtain a very high extinction ratio. The crystal thickness is 1 mm, and the two beams propagate

along the (001) axis. The signal is detected by an optical multichannel analyzer placed at the exit of a monochromator to suppress all the noised light. The signal is obtained after an averaging over 100 successive laser shots.

The use of femtosecond optical pulses to determine the n_2 value presents many advantages. First, we have high-intensity levels and a high temporal resolution. The wavelength is weakly absorbed by the sample ($\alpha = 0,1$ cm⁻¹), thus allowing one to neglect variation of the sample transmission as a function of the pump beam intensity. Furthermore, on this time scale, the contribution of the PRE via optical beam coupling can be neglected.

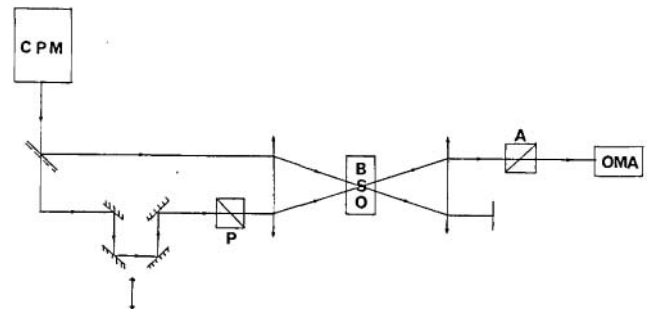


Fig. 1. Experimental setup. A, analyzer; P, polarizer.

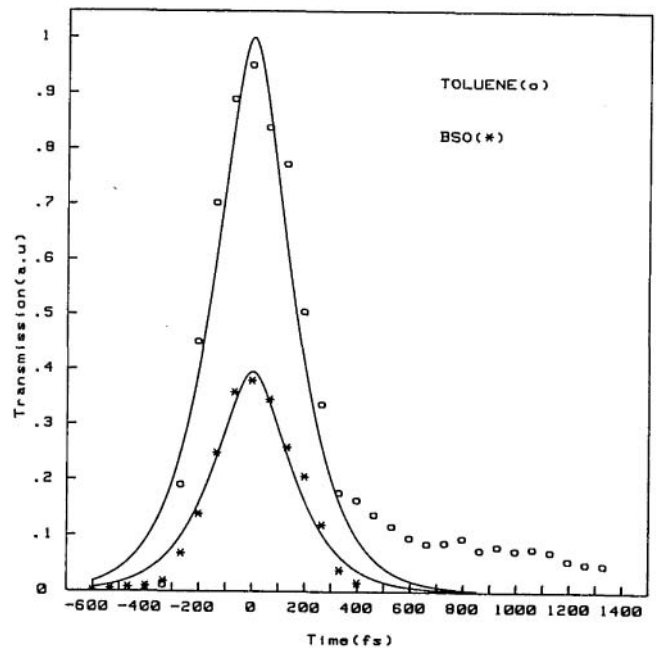


Fig. 2. Comparison between BSO (*) and toluene (O) samples. Toluene exhibits two different relaxation times. Solid line, the third-order correlation function of the pulse (supposed to be bi-exponential).

The experimental results are given in Fig. 2. The temporal response is found to fit well the third-order correlation function of the pump pulse temporal profile (supposed to be bi-exponential), thus indicating, as expected, an instantaneous response time for this nonlinearity. The magnitude of n_2 is obtained by comparison with the n_2 coefficient of the toluene. The temporal resolution of our experiment allows us to separate clearly the two components of the toluene response: the electronic response (instantaneous) and the molecular contribution (response time, 1.8 ps). If we compare the BSO response to the electronic component of toluene we obtain $n_2 = 5 \times 10^{-13}$ esu. Nevertheless, these materials exhibit a large rotary power ($22^\circ/\text{mm}$ at 620 nm). The n_2 measurement is not affected, because at each point inside the crystal the difference between the two directions of polarization is always equal to 45° ; furthermore, we have verified that the n_2 value does not depend on the polarization orientation with respect to the crystals axes. The n_2 value has already been estimated by means of a classical degenerate four-wave mixing method.⁷ The estimated value was deduced from a comparison with the CS₂ response. Unfortunately, this experiment used a 30-ps laser pulse, and the authors cannot resolve the different components of the liquid response. Thus in their case it is hazardous to compare the two results, due only to electronic contribution for BSO and resulting from both electronic and molecular effects in CS₂.

Summarizing: we have measured the n_2 coefficient for BSO crystals in the transparency region and in the femtosecond domain. The n_2 coefficient is of the same order of magnitude as for alkali halides such as NaCl, KCl, KBr, and transparent glasses such as BK7.⁸ It is interesting to notice that using femtosecond optical pulses, index variations as large as those observed using the PRE can be easily obtained with an instantaneous response time.

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Thermal expansion data for Zerodur from 247 to 373 K

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Zerodur is a material with a very low expansion coefficient of approaching 10^{-8} K⁻¹, which is widely used where maximum mechanical stability is of prime importance. Consequently, the performance of the material, in terms of thermal expansion, has been investigated over a wide range of temperatures. These investigations¹⁻⁶ have shown that isothermal length changes, some of which are permanent, occur when the material is thermally cycled over the 200–310 and 400–600 K temperature ranges.

There is a high industrial interest in the performance of this material over the 245–370 K temperature range, which includes only one region of known length instability. However, previous investigations have not been specifically confined to this range, and, therefore, we have investigated the thermal expansion reproducibility and length stability of three pieces of Zerodur over this range of temperatures.

We used the NPL automatic interferometric dilatometer,⁷ which has an uncertainty of $\pm 1 \times 10^{-8}$ K⁻¹, to make our measurements. These showed that the length of each piece of Zerodur was stable and reproducible at temperatures of >273 K, while for those temperatures of <273 K the length of the material was found to be critically dependent on its previous thermal history, and at 247 K isothermal length contractions were observed.

Three cylindrical pieces of Zerodur, 25 mm in diameter and 50 mm long, were used for this investigation. Each piece was contacted to a Zerodur base plate located inside the evacuable chamber of the dilatometer. Thermal expansion coefficients were subsequently determined by measuring interferometrically the movement of the upper face of the specimen as the temperature of the dilatometer chamber was varied. The variation of the sample temperature with time is shown in Fig 1.

Two samples were initially cooled from room temperature (point A in Fig. 1) to ~ 247 K (point B) in 24 h. The mean expansion coefficient ($\bar{\alpha}$) was subsequently determined as the sample was heated to 373 K in four consecutive temperature intervals of ~ 30 K with each set point temperature

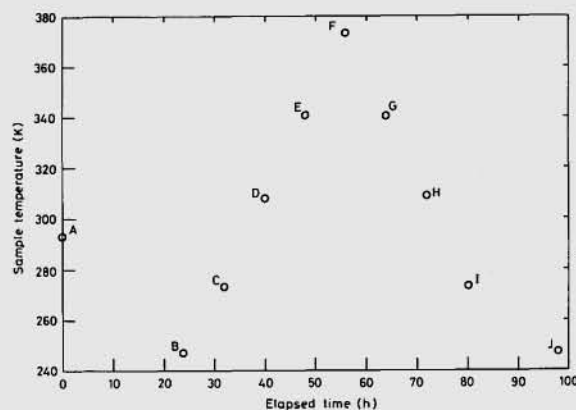


Fig. 1. Zerodur temperature cycle.