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# Z-scan measurements of the nonlinear refractive indices of novel Yb-doped laser crystal hosts

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**ABSTRACT** We report on the z-scan characterization of the nonlinear refractive indices of three borate crystals (GdCOB, YCOB, and BOYS), which are new promising Yb-doped laser hosts. The results indicate the possibility of substantial Kerr-lensing and self-phase modulation effects in femtosecond oscillators and amplifiers. High values of the nonlinear refractive indices suggest possible KLM operation of such lasers.

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## 1 Introduction

$Yb^{3+}$-ion doped solid-state laser materials are very attractive for the development of efficient, diode-pumped mode-locked femtosecond oscillators. This is due to a combination of their broad gain bandwidths, high emission cross sections, small quantum defects [1] and potential for efficient diode pumping using commercially available, high-power laser diodes in the 900–980 nm spectral range. Recently, several promising new Yb-doped borate laser crystals have been introduced [$Ca_4GdO(BO_3)_3$ (GdCOB), $Ca_4YO(BO_3)_3$ (YCOB) and $Sr_3Y(BO_3)_3$ (BOYS)]. Efficient operation of Yb-doped GdCOB, YCOB and BOYS laser crystals was demonstrated both in the continuous wave [2–4] and femtosecond pulse regimes [5, 6] using semiconductor saturable absorber mirrors (SESAM) [7] for passive mode locking. However with all of these materials, the minimum pulse durations, as predicted by the available gain bandwidths, were not achieved. Therefore, continuing research into the relevant optical properties of these new laser-active media is needed in order to optimize the mode locking process to further reduce the pulse duration.

In this paper we report on the characterization of the nonlinear refractive indices of the laser crystal hosts GdCOB, BOYS and YCOB. The nonlinear refractive index, $n_2$, has a strong influence on the mode locking dynamics through the process of self-phase modulation (SPM), which effectively contributes to the total positive intracavity dispersion. Thus, an accurate knowledge of the nonlinear refractive index would allow for a more precise evaluation of the amount of positive dispersion needed for compensation. The amount of effective positive dispersion introduced by SPM (in $fs^2$) can be approximately evaluated by differentiating the power spectrum $I(\omega)$ of a pulse [8]:

$$-\frac{d^2\phi(\omega)}{d\omega^2} = -\frac{d^2}{d\omega^2}\left(\frac{\omega}{c} L n_2 I(\omega)\right), \qquad (1)$$

where the bracket term on the right side of (1) is the phase shift (in frequency domain) acquired by pulse due to SPM (where $L$ is the crystal's length). In addition, the magnitude of the nonlinear refractive index can be viewed as a measure of the crystal's potential for efficient Kerr-lens mode locking (KLM) [9]. Although KLM poses more demands on the alignment requirements for the laser, it generally offers shorter pulses and a more cost-effective solution when compared to SESAM-based mode locking. Since Yb-doped laser materials are prone to Q-switching instabilities [7, 10] due to their relatively long excited upper state lifetimes ($\sim$ 1–3 ms [6]), and the strength of Kerr-lensing is directly proportional to the magnitude of the nonlinear index of refraction, characterization of $n_2$ is critical in determining the stability limits of continuous-wave mode-locked regime against the Q-switched mode-locked, or Q-switched regimes. So far for Yb-based lasers, KLM under direct diode-pumping has only been realized with Yb:KYW material. Pulses as short as 71 fs with 120 mW average output power were generated [11]. Finally, the value of the nonlinear refractive index is also important in determining the overall KLM stability as well as limits of multipulse operation [12].

## 2 Experimental setup

Nonlinear characterization of the crystals was carried out using the well-known, single-beam, z-scan technique [13], which can accurately measure both the magnitude and sign of $n_2$. The samples of GdCOB, YCOB and BOYS were cut and polished such that measurements with the two orthogonal polarizations parallel to the appropriate optical axes of the crystals were possible. The thickness of samples was 4 mm, 1 mm, and 1 mm, respectively. For the experiments we used $\sim$ 1 ps, $\sim$ 1 μJ energy pulses at 1300 nm from an

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optical parametric amplifier pumped by a chirped-pulse amplified Ti:sapphire laser with a repetition rate of 1 kHz. The detailed description of the experimental set-up can be found elsewhere [14]. Using a 175 mm focal length lens the laser radiation was focused to the spot size of approximately $\sim 54\ \mu m$ in diameter, which resulted in a maximum, on-axis peak intensity in the range of $\sim 80\ GW/cm^2$.

## 3 Results and discussion

An example of the experimental z-scan trace is presented in Fig. 1, where the vertical axis plots normalized transmission and the horizontal axis plots distance normalized to the Rayleigh range $z_0$ of the beam (with far-field aperture transmission rate of ∼ 13%). The theoretical fit (shown as a solid curve) was calculated according to the formalism derived in [15] assuming a Gaussian pulse shape and beam profile. The estimated uncertainty of our measurements is ±25% which is in line with other z-scan measurements [15]. The validity of our results was ensured by performing control z-scan measurements with a silica-lead SF-59 glass reference sample [14, 16].

The main results of the z-scan measurements, along with the data for previously characterized relevant crystal hosts, are presented in Table 1. All crystals showed a positive refractive nonlinearity. No two-photon absorption was observed in the experiments. The values of the nonlinear indices of refraction can be compared with the $\sim 3.1 \times 10^{-7}\ cm^2/GW$ value for Ti:sapphire laser crystal [19, 20] which is often used for efficient KLM operation. Therefore, our results indicate the possibility of appreciable Kerr-lensing and self-phase modulation effects in femtosecond oscillators and amplifiers based on these Yb-doped laser crystals. No significant change in the $n_2$ values is expected in the 1000–1300 nm wavelength range, since the absorption edges of GdCOB, YCOB and BOYS crystals lie in the UV region. Such wavelength dependence of the refractive nonlinearity was well predicted by theoretical models [21] and later confirmed experimentally [19]. For example, our previous results with Yb:KGW crystal [14] showed only slight increase (∼ 5%) in the $n_2$ values in the 1100–1600 nm wavelength region. Doping of crystals with Yb-ions would probably result in a similar increase in the $n_2$ values, since the dopant concentrations typically used are not high. The conversion between CGS (esu) and MKS ($m^2/W$) units can be done using a simple relation, $n_2[10^{-13}\ esu] = (cn/40\pi)n_2\ [cm^2/GW]$, where $n$ is the linear refractive index and $c$ is the speed of light (in meters per second).

| Host | Polarization | Direction | $n$ | $n_2$, $\times 10^{-7}\ cm^2/GW$ |
|---|---|---|---|---|
| GdCOB | $E \parallel Z$ | $k \parallel Y$ | 1.71 | 9.8 |
| [$Ca_4GdO(BO_3)_3$] | $E \parallel X$ | | 1.68 | 11.4 |
| YCOB | $E \parallel Z$ | $k \parallel X$ | 1.71 | 9.0 |
| [$Ca_4YO(BO_3)_3$] | $E \parallel Y$ | | 1.70 | 7.4 |
| BOYS | $E \perp c$ | $k \perp c$ | 1.74 | 6.6 |
| [$Sr_3Y(BO_3)_3$] | $E \parallel c$ | | ∼ 1.74 | 8.8 |
| KYW [a] [$KY(WO_4)_2$] | $E \parallel a$ | – | ∼ 2.00 | 8.7 |
| KGW [b] | $E \parallel X$ | $k \parallel Z$ | 1.98 | 15.0 |
| [$KGd(WO_4)_2$] | $E \parallel Y$ | | 2.00 | 20.0 |
| YAG [c] [$Y_3Al_5O_{12}$] | – | – | 1.82 | 6.2 |
| $Y_2O_3$ [c] | – | – | 1.92 | 11.6 |

[a] Ref. [17], [b] Ref. [14], [c] Ref. [18]

**TABLE 1** Results of the z-scan measurements

Yb-doped laser materials are prone to Q-switching instabilities owing to their long excited upper state lifetimes [7, 10]. Therefore, the implementation of a fast (instantaneous) saturable absorber (in this case KLM) as a passive mode locker should lead to a better discrimination between pure mode locking and Q-switched mode-locked regime. Furthermore, the use of a fast saturable absorber would result in the generation of shorter pulses because the absorber would be able to shape both the leading and trailing edge of the generated pulse. The use of symmetric KLM resonators [22] operating in the middle of the cavity stability limits, regenerative mode locking [23] to initiate ultrashort pulse generation, or negative feedback control loop circuits [24] to suppress Q-switching instabilities might be appropriate solutions to overcome these unwanted effects. On the other hand, experiments with KLM Yb:glass laser (∼ 1 ms upper state lifetime) with diffraction-limited pump beam did not report on any occurrence or problems with Q-switching instabilities [25].

Reliable KLM operation should be more easily achieved in Yb:KYW/KGW laser crystals because of their shorter excited upper state lifetimes ($\sim 350\ \mu s$ [26]) and higher stimulated emission cross sections. Nonetheless, shorter laser pulses would be possible with GdCOB/YCOB/BOYS materials that have much broader amplification bandwidths [6]. Finally, it is worth noting that high $n_2$ values are beneficial for diode-pumped KLM laser configurations since the minimum pump spot size inside the crystal is limited by the poor beam quality of laser diodes.

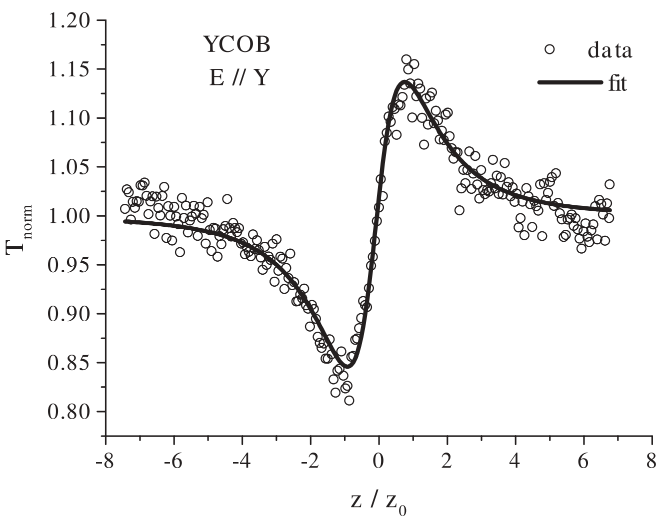


**FIGURE 1** Sample closed-aperture z-scan trace showing fit to theoretical curve

## 4 Conclusions

In conclusion, we have characterized the nonlinear refractive indices of three promising borate crystal hosts

(GdCOB, YCOB, BOYS) for Yb-doped laser gain media. The results indicate the possibility of substantial Kerr-lensing and self-phase modulation effects in high-power femtosecond oscillators or amplifiers based on these laser crystals. High values of the nonlinear refractive indices also suggest possible KLM operation of such lasers.

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## REFERENCES

1 L.D. DeLoach, S.A. Payne, L.L. Chase, L.K. Smith, W.L. Kway, W.F. Krupke: IEEE J. Quantum Electron. **QE-29**, 1179 (1993)
2 F. Mougel, K. Dardenne, G. Aka, A. Kahn-Harari, D. Vivien: J. Opt. Soc. Am. B **16**, 164 (1999)
3 A. Aron, G. Aka, B. Viana, A. Kahn-Harari, D. Vivien, F. Druon, F. Balembois, P. Georges, A. Brun, N. Lenain, M. Jacquet: Opt. Mater. **16**, 181 (2001)
4 S. Chénais, F. Druon, F. Balembois, P. Georges, R. Gaumé, P.H. Haumesser, B. Viana, G.P. Aka, D. Vivien: J. Opt. Soc. Am. B **19**, 1083 (2002)
5 F. Druon, F. Balembois, P. Georges, A. Brun, A. Courjaud, C. Hönninger, F. Salin, A. Aron, F. Mougel, G. Aka, D. Vivien: Opt. Lett. **25**, 423 (2000)
6 F. Druon, S. Chénais, P. Raybaut, F. Balembois, P. Georges, R. Gaumé, G. Aka, B. Viana, S. Mohr, D. Kopf: Opt. Lett. **27**, 197 (2002)
7 U. Keller, K.J. Weingarten, F.X. Kärtner, D. Kopf, B. Braun, I.D. Jung, R. Fluck, C. Hönniger, N. Matuschek, J. Aus der Au: IEEE J. Sel. Top. Quantum Electron. **2**, 435 (1996)
8 J.-M. Hopkins: Compact, low-threshold femtosecond lasers, Ph.D. Thesis, University of St. Andrews, United Kingdom (1999)
9 D.E. Spence, P.N. Kean, W. Sibbett: Opt. Lett. **16**, 42 (1991)
10 C. Hönniger, R. Paschotta, F. Morier-Genoud, M. Moser, U. Keller: J. Opt. Soc. Am. B **16**, 46 (1999)
11 H. Liu, J. Nees, G. Mourou: Opt. Lett. **26**, 1723 (2001)
12 V.L. Kalashnikov, E. Sorokin, I.T. Sorokina: IEEE J. Quantum Electron. **QE-39**, 323 (2003)
13 M. Sheik-Bahae, A.A. Said, E.W. Van Stryland: Opt. Lett. **14**, 955 (1989)
14 A. Major, I. Nikolakakos, J.S. Aitchison, A.I. Ferguson, N. Langford, P.W.E. Smith: Appl. Phys. B **77**, 433 (2003)
15 M. Sheik-Bahae, A.A. Said, T.-H. Wei, D.J. Hagan, E.W. Van Stryland: IEEE J. Quantum Electron. **QE-26**, 760 (1990)
16 I. Nikolakakos, A. Major, J.S. Aitchison, P.W.E. Smith: Broadband characterization of the nonlinear optical properties of common reference materials, In IEEE. J. Sel. Top. Quantum Electron. (to be published)
17 K.V. Yumashev, N.N. Posnov, P.V. Prokoshin, V.L. Kalashnikov, F. Mejid, I.G. Poloyko, V.P. Mikhailov, V.P. Kozich: Opt. Quantum Electron. **32**, 43 (2000)
18 R. Adair, L.L. Chase, S.A. Payne: Phys. Rev. B **39**, 3337 (1989)
19 R. DeSalvo, A.A. Said, D.J. Hagan, E.W. Van Stryland, M. Sheik-Bahae: IEEE J. Quantum. Electron. **QE-32**, 1324 (1996)
20 A. Major, F. Yoshino, I. Nikolakakos, J.S. Aitchison, P.W.E. Smith: Opt. Lett. **29**, 602 (2004)
21 M. Sheik-Bahae, D.C. Hutchings, D.J. Hagan, E.W. Van Stryland: IEEE J. Quantum Electron. **QE-27**, 1296 (1991)
22 G. Cerullo, S. De Silvestri, V. Magni: Opt. Lett. **19**, 1040 (1994)
23 D.E. Spence, J.M. Evans, W E. Sleat, W. Sibbett: Opt. Lett. **16**, 1762 (1991)
24 T.R. Schibli, U. Morgner, F.X. Kärtner: Opt. Lett. **26**, 148 (2001)
25 V. Petrov, U. Griebner, D. Ehrt, W. Seeber: Opt. Lett. **22**, 408 (1997)
26 A. Major, L. Giniûnas, N. Langford, A.I. Ferguson, D. Burns, E. Bente, R. Danielius: J. Mod. Opt. **49**, 787 (2002)