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The Kerr nonlinearity of the beta-barium borate crystal

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A popular crystal for ultrafast cascading experiments is beta-barium-borate (β-BaB2O4, BBO). It has a decent quadratic nonlinear coefficient, and because the crystal is anisotropic it can be birefringence phase-matched for type I (oo → e) second-harmonic generation (SHG). For femtosecond experiments BBO is popular because of low dispersion and a high damage threshold. The main attractive property of ultrafast cascading is that the induced cascading nonlinearity \( n_{2,\text{casc}} \) can be negative, i.e. generate a self-defocusing Kerr-like nonlinearity. However, the material Kerr nonlinearity \( n_{2,\text{Kerr}} \) is self-focusing and competes with the cascading nonlinearity. Therefore, precise knowledge of its strength is crucial. We perform an experiment measuring the main \( c_{11} \) tensor component, and together with literature experimental data [1], we propose a \( c_{11} \) value composed of 14 different data points.

Figure 1. (a) Experimental spectra recorded with 50 fs@1030 nm and 200 GW/cm² transform-limited pulses, (b) the spectral bandwidth@-10 dB vs. \( \Delta k \). (c) Summary of the experimental data from the literature corresponding to the \( c_{11} \) nonlinear susceptibility coefficient \( n_{2,\text{Kerr}} = 3c_{11}/4n_2^2\varepsilon_0 \). The data are corrected by us for cascading contributions. The shaded areas “σ” and “2σ” represent one and two standard deviations.

BBO is a negative uniaxial crystal in the point group 3m. When the pump is \( \sigma \)-polarized, the nonlinear susceptibility component that accounts for the Kerr self-phase modulation (SPM) is \( c_{11} = \chi^{(3)}_{XXX} = \chi^{(3)}_{YYY} \). We pumped a 25 mm BBO cut for oo → e SHG with 50 fs 1030 nm pulses from a commercial OPA. By tuning the phase mismatch away from zero cascading set in as a Kerr-like SPM nonlinearity with the nonlinear index \( n_{2,\text{casc}} = -2\omega_1d_{\text{eff}}^2/e_0\varepsilon_0n_2^2\Delta k \), where \( d_{\text{eff}} \) is the effective quadratic nonlinearity, and \( n_1 \) and \( n_2 \) the FW and SH linear indices. Fig. 1(a) shows that for low \( \Delta k \) the self-defocusing cascading dominates leading to strongly modulated and broadened spectra. At some critical point the cascading exactly cancels the material Kerr nonlinearity \( n_{2,\text{casc}} + n_{2,\text{Kerr}} = 0 \). Such a zero SPM nonlinearity should leave the spectra invariant with intensity. We found this point by observing the -10 dB bandwidth crossing of two different intensities, see Fig. 1(b). Then, by using the well-known quadratic nonlinearities for BBO we can calculate \( n_{2,\text{Kerr}} = -n_{2,\text{casc}} = 4.9 \pm 0.4 \cdot 10^{-20} \) \( \text{m}^2/\text{W} \), which corresponds to \( c_{11} = 4.7 \pm 0.4 \cdot 10^{-22} \) \( \text{m}^2/\text{V}^2 \). In the literature other experiments have measured the Kerr nonlinearity in BBO [1]. We have done a careful analysis of these to (a) clarify which tensor components were excited, (b) ensure consistent definitions of the Kerr nonlinearity and (c) correct for the deterministic contribution from cascaded SHG [2]. The summary is shown for the most important \( c_{11} \) component in Fig. 1(c). The values agree surprisingly well with the two-band model, originally derived for wide-gap semiconductors. We confirmed that Miller’s delta, calculated as \( \Delta_{\text{eff}} = c_{11}/n_2^2(\lambda_p - 1)^2 \), was nearly constant over all data at various pump wavelengths \( \lambda_p \) (except the UV measurements below 400 nm that were not used). Therefore we could calculate a weighted mean over 14 data points as \( \Delta_{\text{eff}} = 52.3 \pm 7.7 \cdot 10^{-21} \) \( \text{m}^2/\text{V}^2 \), which corresponds to \( c_{11} = 4.78 \cdot 10^{-22} \) \( \text{m}^2/\text{V}^2 \) and \( n_{2,\text{Kerr}} = 4.93 \cdot 10^{-20} \) \( \text{m}^2/\text{W} \) at 1030 nm. Our experiment agrees very well with this average. We finally analyze literature data [3] to obtain the 3 other tensor components \( c_{10}, c_{16} \) and \( c_{33} \) as well.

References

