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Zero focusing via competing nonlinearities in BBO crystals

K. Krupa,¹ F. Baronio,² M. Conforti,² S. Trillo,³ A. Tonello,¹ V. Couderc,^{1*}

¹*Institut Xlim, UMR 6172, CNRS, 123 avenue Albert Thomas, 87060 Limoges, France*

²*Dipartimento di Ingegneria dell'Informazione, Università di Brescia, Via Branze 38, 25123 Brescia, Italy*

³*Dipartimento di Ingegneria, Università di Ferrara, Via Saragat 1, 44122 Ferrara, Italy*

* *Corresponding author: vincent.couderc@xlim.fr*

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We investigate nonlinear focusing behavior of light beams propagating in beta-barium-borate (BBO) crystals under mismatched second-harmonic generation. We clearly identify experimentally multiple self-focusing and defocusing regions against the orientation angle, and the condition where competing quadratic and cubic nonlinearities perfectly compensate each other (zero-focusing point).

OCIS codes: 190.2620, 190.4223, 190.3720.

Introduction. Quadratic optical media are usually exploited for frequency conversion applications. However, the energy conversion process is always accompanied by nonlinear phase shifts of the interacting fields, as recognized long ago [1, 2]. This concept remained nearly dormant until the beginning of the nineties when phase shifts produced in quadratic processes attracted a new surge of interest [3–6], also related to their exploitation for controlling optical signals in the spatial, temporal and spectral domains [7–17].

Any three-photon mixing process entail nonlinear phase shifts of all fields which, in the CW limit, are generally described by complicated periodic behavior with distance. However, in the regime of large phase-mismatches, the phase shifts follow a Kerr-like behavior, being nearly proportional to distance and intensity [5]. This regime is also referred to as cascading limit [7] to recall the fact that the phase shifts accumulate through many repeated up- and down-conversion processes. Remarkably, such phase shift can be either self-induced [5] or cross-induced [6], while its sign depends on the sign of mismatch, and can be opposite for the different frequency components [10], thereby adding great flexibility in the applications.

Mostly exploited for its simplicity is the nonlinear phase shift of the fundamental frequency (FF) field occurring via second harmonic generation (SHG) with no seed at second-harmonic (SH). In this configuration the cascaded phase shifts can be controlled through the wavevector mismatch by simply adjusting the orientation or the temperature of the crystal. Particularly interesting is the ability to generate negative phase-shifts which allows to access intriguing regimes (e.g. soliton propagation in normally dispersive bulk media and strong pulse compression and shaping without catastrophic collapse [8, 17]), underlining the most innovative new possibilities [18–20].

However, in general, the net phase shift impressed on a light beam also contain a natural contribution from the cubic (Kerr) nonlinear index $n_2^{(3)}$ existing in any material and usually positive. Therefore, one might think

about cascaded $\chi^{(2)}$ and $\chi^{(3)}$ processes as contributing to an overall effective nonlinear index as $n_2 = n_2^{(2)} + n_2^{(3)}$ [11, 21]. Since the cubic index contribution $n_2^{(3)}$ has negligible dependence on crystal orientation and temperature, it acts as a bias to the total nonlinear index. As a result, multiple self-focusing and self-defocusing regions can exist. For the typical case, when cubic phase shifts are positive, self-defocusing is expected to be possible within a finite range of mismatches (crystal orientations) on one side of phase-matching condition.

The aim of this Letter is to determine theoretically and experimentally such range for beta-barium-borate (BBO) crystals. In particular, by observing the self-focusing and defocusing behavior of standard laser beams in BBO, we are able to clearly show the existence of a critical angle where the quadratic and cubic contributions mutually compensate for a wide range of input beam intensities, remarkably leading to a zero-focusing condition. To the best of our knowledge, compensation of this kind was achieved previously only by employing different crystals in back to back configuration [11]. Our experimental results agree well with theoretical analysis based on the cascading reduction of SHG equations, taking into account the Kerr coefficient of BBO.

Competing nonlinearities in BBO. We consider the propagation of an intense ordinarily polarized beam E_1 at FF $\omega = \omega_0$, together with its SH ($\omega = 2\omega_0$), extraordinarily polarized beam E_2 , generated through a type-I configuration. Since temporal effects over the time scale of the experiment are negligible, the following nonlinear coupled equations can be safely employed to describe SHG in this configuration [21–23]

$$i \frac{\partial E_1}{\partial z} + \frac{1}{2k_1} \nabla_{\perp}^2 E_1 + \chi_2 E_1^* E_2 e^{i\Delta k z} + (\chi_{31}|E_1|^2 + \chi_{32}|E_2|^2) E_1 = 0, \quad (1)$$

$$i \frac{\partial E_2}{\partial z} + \frac{1}{2k_2} \nabla_{\perp}^2 E_2 + \chi_2 E_1^2 e^{-i\Delta k z} + (\chi_{33}|E_1|^2 + \chi_{34}|E_2|^2) E_2 = 0, \quad (2)$$

where $E_{1,2} = E_{1,2}(x, y, z)$, $\nabla_{\perp}^2 = \partial^2/\partial x^2 + \partial^2/\partial y^2$, χ_2

and χ_{3k} are proportional to the relevant elements of the quadratic and cubic susceptibility tensors, respectively (χ_2 refers to *ooe* three wave interaction, χ_{31} to *oooo*, χ_{32} to *eeoo*, χ_{33} to *oeee*, and χ_{34} to *eeee* four wave mixing interactions); z is the propagation distance; $\Delta k = k_2 - 2k_1 = 2[n_e(2\omega_0; \theta) - n_o(\omega_0)]\omega/c$ is the wave-number mismatch, $n_o(\omega_0)$ and $n_e(2\omega_0; \theta)$ being the ordinary and extraordinary refractive index at FF and SH, while θ is the angle of the propagation direction with respect to the optical axis of the crystal.

In Tables 1 and 2, we report the effective nonlinearities for the crystals of class $3m$ to which BBO belongs, as a function of rotation angles (θ, ϕ) by specifying the relative type of interaction. For instance, *ooe* ($o+o \rightarrow e$) indicates the upconversion of two ordinarily polarized photons that generate an extraordinarily polarized photon at SH (for further details see Refs. [24, 25]).

Coefficient Expression	Interaction
$d_0 = -3d_{31} \cos^2 \theta \sin \theta - d_{22} \cos^3 \theta \sin 3\phi$	eee
$d_1 = -d_{22} \cos 3\phi \cos^2 \theta$	eeo, oeo, oee
$d_2 = -d_{31} \sin \theta + d_{22} \cos \theta \sin 3\phi$	ooe, eoo, oeo
$d_3 = d_{22} \cos 3\phi$	ooo

Table 1. Effective quadratic nonlinear coefficients. $d_{22} = 2.2\text{pm/V}$, $d_{31} = -0.04\text{pm/V}$.

Coefficient Expression	Interaction
$c_0 = c_{11} \cos^4 \theta + c_{33} \sin^4 \theta + \frac{3}{2}c_{16} \sin^2 2\theta$	eeee
$-4c_{10} \sin 3\phi \sin \theta \cos^3 \theta$	
$c_1 = \frac{3}{2}c_{10} \cos 3\phi \sin 2\theta \cos \theta$	eeoe, eeeo
$c_2 = -\frac{1}{3}c_{11} \cos^2 \theta + c_{16} \sin^2 \theta$	oeee, eooo
$+c_{10} \sin 2\theta \sin 3\phi$	
$c_3 = c_{10} \cos 3\phi \sin \theta$	ooooe, ooeo
$c_4 = c_{11}$	oooo

Table 2. Effective cubic nonlinear coefficients. $c_{11} = 5.6 \cdot 10^{-22}\text{m}^2/\text{V}^2$, $c_{10} = -0.24 \cdot 10^{-22}\text{m}^2/\text{V}^2$, $c_{16} = -1.4 \cdot 10^{-22}\text{m}^2/\text{V}^2$ [24].

In the cascading regime (high $|\Delta k|$), the leading order equation that can be derived from Eqs. (1) is the following nonlinear Schrödinger equation that governs the evolution the ordinary FF beam E_1 [21, 26]:

$$i \frac{\partial E_1}{\partial z} + \frac{1}{2k_1} \nabla_{\perp}^2 E_1 + (\gamma_2 + \gamma_3) |E_1|^2 E_1 = 0, \quad (3)$$

where cascaded quadratic and cubic Kerr nonlinearities are expressed as $\gamma_2 = -(\frac{\omega_0 d_{eff}}{n_0 c})^2 \frac{1}{\Delta k} [m/\text{V}^2]$ and $\gamma_3 = \frac{3}{8} \frac{\omega_0 c_{eff}}{n_0 c} [m/\text{V}^2]$, where $d_{eff} = d_1$ and $c_{eff} = c_4$ are the effective nonlinear coefficients from Tables 1, 2.

Figure 1 shows the effective nonlinear index $\gamma = \gamma_2 + \gamma_3$, resulting from cascading-quadratic γ_2 , and cubic γ_3 contributions, as function of the angle θ , at fixed $\phi = 90^\circ$. Multiple self-focusing and defocusing regions can be identified. In particular, in the region of positive mismatches ($\gamma_2 < 0$), a zero-focusing point is identified

at $\theta_c = 18.4^\circ$, where the quadratic and cubic contributions exactly balance ($\gamma_2 = -\gamma_3$). While for angles $\theta < \theta_c$ the mismatch increases and hence the Kerr effect prevails leading to self-focusing, for increasing angles $\theta > \theta_c$ the mismatch decreases and leads cascading to dominate thus resulting into self-defocusing, until for θ approaching the phase-matching angle $\theta = 22.9^\circ$, the cascading picture breaks down due to increased efficiency of SHG (shaded region in Fig. 1). For angles greater than the perfect phase-matching angle, γ_2 and γ_3 have the same sign, leading to enhanced focusing.

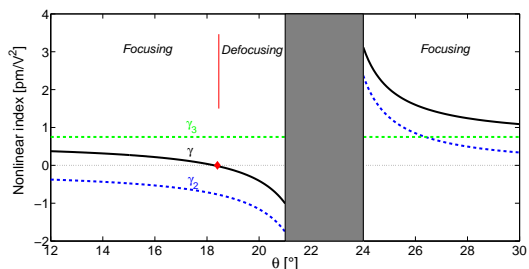


Fig. 1. (Color online) Total effective cubic nonlinearity γ , resulting from both quadratic γ_2 and cubic γ_3 contributions, plotted versus the angle θ . The shaded domain identifies the region where cascading breaks down and SHG enters the regime of efficient conversion.

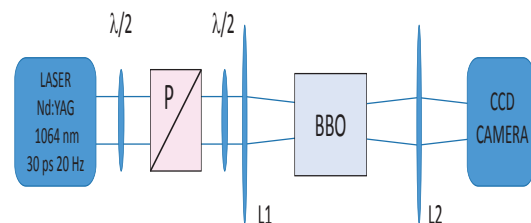


Fig. 2. (Color online) Experimental setup. Nd:YAG laser pump delivering 30 ps pulses, at 1064 nm, 20 Hz repetition rate; L lenses; $\lambda/2$ half wave plates; P polarizer; quadratic uniaxial BBO crystal; CCD camera.

Experimental results. A sketch of our experimental setup is reported in Fig. 2. We employ a sample of BBO crystal with a square aperture of 6×6 mm and a length of 20 mm. The crystal is cut for frequency doubling at 1064nm under type-I (*ooe*) phase-matching, at the angles $\theta = 22.9^\circ$ and $\phi = 90^\circ$ to the principal axes. The crystal faces are coated for antireflection at 1064 nm. The crystal is pumped using a Q-switched, mode-locked Nd:YAG laser which delivers 30 ps pulses at 1064 nm at a repetition rate of 20 Hz, with energy up to 1.23 mJ.

A polarizer in between half-wave plates are used to adjust the energy and the polarization of the pump pulses. The pump light is collimated using a lens of 140 mm focal length. The average beam diameter at the input is around $140\mu\text{m}$ FWHM. The spatial beam patterns at the output of the crystal are imaged, with magnification $G = 4.6$, onto a CCD camera and analyzed. Under such experimental conditions, we characterize the spatial dynamics of an ordinary FF beam E_1 at the output of the BBO crystal, at different crystal orientation angles and input energies.

In Fig. 3 we summarize the dependence of the beam diameter (FWHM) at the output of the BBO crystal versus crystal orientation angle θ , for different input energies. First notice that diffraction has by itself negligible impact due to the large beam size (see curve relative to the linear limit in Fig. 3), while it becomes obviously effective, in the nonlinear regime, to transform the self-induced spatial phase modulation into beam narrowing (self-focusing) or broadening (self-defocusing). The data indicate, as expected from theory, the existence of different nonlinear focusing and defocusing regions. In particular in the region of negative phase mismatches ($\theta > 24^\circ$ in Fig. 3), the cascading effect always adds up to the Kerr coefficient leading to self-focusing (beam narrowing). Conversely for positive mismatches we observe opposite behavior across the critical angle $\theta_c = 18.6^\circ$, at which the beam travels nearly undistorted due to mutual balance of Kerr and cascading nonlinear index. While for $\theta < 18.6^\circ$, the FF beam self-focuses due to dominant positive Kerr index, for $18.6^\circ < \theta < 22.5^\circ$ the cascading nonlinear index overwhelms the Kerr effect causing the beam to defocus. Remarkably, while increasing the input energy (intensity) results into an enhanced nonlinear effect, the point of zero-focusing remains the same with good accuracy regardless of the beam intensity. In order

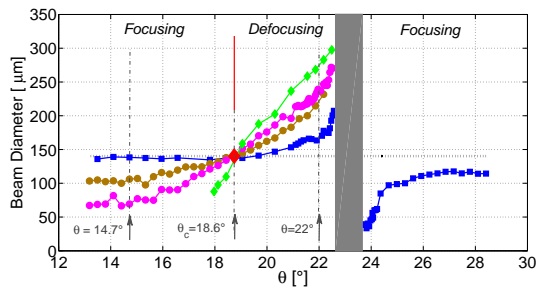


Fig. 3. (Color online) Beam diameter (FWHM of intensity) at FF measured at BBO crystal output vs. crystal rotation angle θ , for different energy of the pulse: linear regime (dotted line); 0.08 mJ (blue squares); 0.34 mJ (gold circles); 0.54 mJ (magenta circles); 1.23 mJ (green diamonds).

to show that the beams undergo self-focusing or defocusing without substantial distortion, we also display in Fig. 4 the beam profiles obtained for fixed input energy (0.54 mJ) for three characteristic values of the angle θ across

the critical value θ_c (vertical lines in Fig. 3). In particular the defocusing behavior in Fig. 4(a,d), obtained for $\theta = 22^\circ$ above the critical value θ_c , is contrasted with the focusing behavior in Fig. 4(c,f), obtained below the critical value $\theta = 14.7^\circ < \theta_c$, while Fig. 4(b,e) corresponds to the critical angle that give rise to the undistorted propagation.

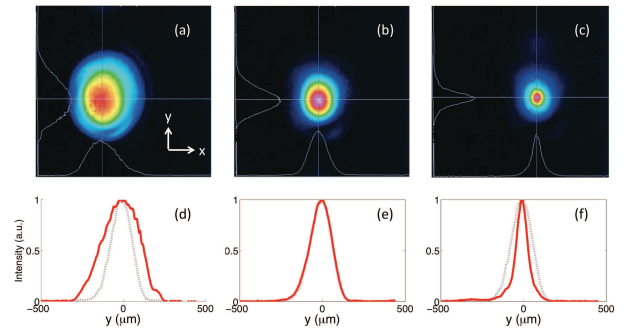


Fig. 4. (Color online) (a-c) measured 2D output beam shapes and (d-f) corresponding beam profiles vs. y ($x = 0$ section), compared with the input. (a,d) $\theta = 22^\circ$; (b,e) $\theta = \theta_c = 18.6^\circ$; (c,f) $\theta = 14.7^\circ$. Here the input energy is 0.54 mJ.

Conclusions. We have considered the effect of nonlinear phase shifts impressed on light beams during propagation in BBO crystals, showing evidence for competing quadratic and cubic contributions. We have identified theoretically and have demonstrated experimentally the existence of multiple overall self-focusing and defocusing regions, and the existence for a critical value of orientation which yields a compensation of cascading and cubic contributions.

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