Self-focusing and self-defocusing by cascaded second-order effects in KTP

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We monitor the induced phase change produced by a cascaded $\chi^{(2)}:\chi^{(2)}$ process in KTP near the phase-matching angle on a picosecond 1.06-μm-wavelength beam using the Z-scan technique. This nonlinear refraction is observed to change sign as the crystal is rotated through the phase-match angle in accordance with theory. This theory predicts the maximum small-signal effective nonlinear refractive index of $n_2^{\text{eff}} \cong \pm 2 \times 10^{-14}$ cm^2/W ($\pm 1 \times 10^{-11}$ esu) for an angle detuning of $\pm 5°$ from phase match for this 1-mm-thick crystal with a measured d_{eff} of 3.1 pm/V. For a fixed phase mismatch, this n_2^{eff} scales linearly with length and as d_{eff}^2; however, for the maximum n_2^{eff} the nonlinear phase distortion becomes sublinear with irradiance for phase shifts near $\pi/4$.

The nonlinear phase distortion that arises from second-order processes in noncentrosymmetric crystals has recently received considerable attention.[1–6] Although the effective $\chi^{(3)}$ that is due to cascading of $\chi^{(2)}(3\omega;\, 2\omega,\, \omega):\chi^{(2)}(2\omega;\, \omega,\, \omega)$ has long been used in generating the third harmonic of laser beams using two crystals, its extension to obtain nonlinear refraction through $\chi^{(2)}(\omega;\, 2\omega,\, -\omega):\chi^{(2)}(2\omega;\, \omega,\, \omega)$ cascading has not been fully utilized. There are two possible consequences of this nonlinearity for the fundamental beam, loss and phase distortion. The loss is well known and is simply due to conversion of the fundamental to the second harmonic. For low conversion efficiency this loss is nearly indistinguishable from two-photon absorption, thus resulting in an effective $\text{Im}[\chi^{(3)}]$. The refractive effect is less well known and usually ignored, occurring only off phase matching where a portion of the frequency-doubled light is downconverted with a shifted phase. Hence the net phase of the fundamental wave is shifted in proportion to the irradiance of the fundamental, which for low irradiance results in a Kerr-like nonlinearity {an effective $\text{Re}[\chi^{(3)}]$}. Using the Z-scan technique,[7] we monitor the self-action of 1.06-μm picosecond pulses as they propagate through a KTP crystal close to the phase-matching angle for type II second-harmonic generation[8] (SHG). It is observed that the sign of the nonlinear phase shift changes from positive (self-focusing) to negative (self-defocusing) on angle tuning the sample from negative to positive phase mismatch. The sign and magnitude of the observed phase change agree with the theoretical results as obtained from the coupled-wave equations. A primary application of a negative, fast (electronic) Kerr-like nonlinearity in the presence of positive group-velocity dispersion is the self-compression of ultrashort pulses that can be achieved during such a cascading process.[3] This is the mechanism responsible for self-compression of the idler pulse during optical parametric oscillation in β-barium borate, described in Ref. 3.

The coupled amplitude equations governing SHG in a noncentrosymmetric crystal as derived from Maxwell's equations in the slowly varying envelope approximation are[9]

$$\frac{\mathrm{d}E_2}{\mathrm{d}z'} = -i\frac{\omega}{2cn_{2\omega}}\chi^{(2)}(2\omega;\, \omega,\, \omega)E_1E_1 \exp(i\Delta kz'), \quad (1)$$

$$\frac{\mathrm{d}E_1}{\mathrm{d}z'} = -i\frac{\omega}{4cn_{\omega}}\chi^{(2)}(\omega;\, 2\omega,\, -\omega)E_2E_1^* \exp(-i\Delta kz'), \quad (2)$$

where Eq. (1) describes the growth of $E_2(2\omega)$ with depth z' in the sample, while Eq. (2) gives the evolution (depletion and phase variation) of the fundamental beam $E_1(\omega)$ during the SHG process. The wave-vector mismatch is $\Delta k = k_{2\omega} - 2k_{\omega} = 2\omega(n_{2\omega}{}^{i} - n_{\omega}{}^{j})/c$, with i and j denoting the polarization directions at frequencies 2ω and ω, respectively. In the absence of loss, the Manley–Rowe relations apply and $\chi^{(2)}(\omega;\, 2\omega,\, -\omega) = 2\chi^{(2)*}(2\omega;\, \omega,\, \omega)$. In order to simplify Eqs. (1) and (2), we define the parameter

$$\Gamma = \frac{\omega d_{\text{eff}}|E_0|}{c\sqrt{n_{2\omega}n_{\omega}}}, \quad (3)$$

where $d_{\text{eff}} = |\chi^{(2)}(2\omega;\, \omega,\, \omega)|/2$ and E_0 is the incident fundamental field. Solving for the fundamental beam by eliminating E_2 and assuming no initial second-harmonic field, we obtain

$$\frac{\mathrm{d}^2E_1}{\mathrm{d}z'^2} + i\Delta k\frac{\mathrm{d}E_1}{\mathrm{d}z'} - \Gamma^2(1 - 2|E_1/E_0|^2)E_1 = 0. \quad (4)$$

For perfect phase matching ($\Delta k = 0$), Eq. (4) yields the well-known $E_1 = E_0\, \text{sech}(\Gamma L)$ solution. Here we concentrate on the non-phase-matched solu-

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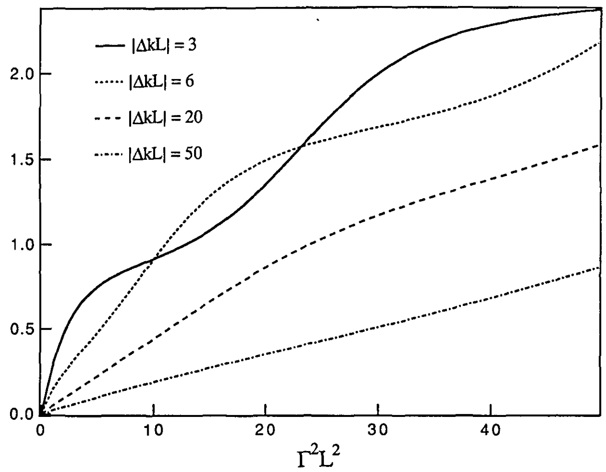

Fig. 1. Induced nonlinear phase shift versus Γ^2L^2 for several values of phase mismatch as calculated by the numerical solution of Eq. (4).

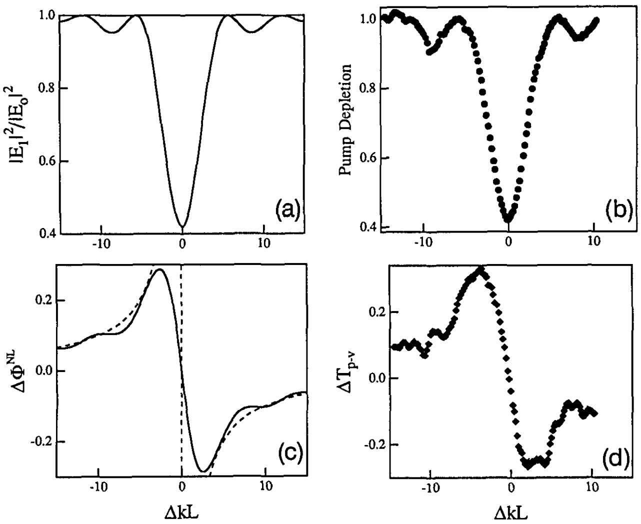

Fig. 2. (a) Calculation of the depletion of the fundamental wave as a function of phase mismatch ΔkL. (b) Experimental measurement of the depletion of the fundamental beam versus ΔkL. (c) Calculation of nonlinear phase shift ($\Delta\Phi^{\mathrm{NL}}$) as a function of ΔkL; the dashed curve is the small conversion efficiency limit, and the solid curve is the exact solution of Eq. (4). (d) Experimental measurement of ΔT_{p-v} as a function of ΔkL as the crystal is rotated through its phase-matching angle.

tion to this equation. In the small conversion efficiency limit, $|E_1| \cong |E_0|$, and hence $E_1(z') = |E_0| \times \exp[-i\Delta\Phi^{\mathrm{NL}}(z')]$ for all z'. From Eq. (4), the nonlinear phase change impressed onto the fundamental beam at the exit surface $z' = L$ is given by

$$\Delta\Phi^{\mathrm{NL}} \cong \frac{\Delta kL}{2}\{1 - [1 + (2\Gamma/\Delta k)^2]^{1/2}\}. \tag{5}$$

It is clear from relation (5) that there is a nonlinear phase distortion, $\Delta\Phi^{\mathrm{NL}}$, on the fundamental beam even though depletion is assumed to be negligible. For large phase mismatch and/or low irradiance (I), $|\Delta k| >> |\Gamma|$, and this nonlinear phase shift varies linearly with irradiance I, similar to an optical Kerr effect,

$$\Delta\Phi^{\mathrm{NL}} \cong -\frac{\Gamma^2L^2}{\Delta kL}, \tag{6}$$

where this phase shift is evaluated at $z' = L$. As the optical Kerr effect is described by $n = n_0 + n_2I$, we can, by analogy, introduce an effective nonlinear index of refraction n_2^{eff}, where $\Delta\Phi^{\mathrm{NL}} = (2\pi L/\lambda)n_2^{\mathrm{eff}}I$ and

$$n_2^{\mathrm{eff}} = -\frac{4\pi}{c\varepsilon_0}\frac{L}{\lambda}\frac{d_{\mathrm{eff}}^2}{n_{2\omega}{n_\omega}^2}\frac{1}{\Delta kL}. \tag{7}$$

Note that this is proportional to the usual figure of merit for $\chi^{(2)}$ materials, ${d_{\mathrm{eff}}}^2/n^3$. For large phase shifts this approximation breaks down, and Eq. (4) must be solved exactly. In Fig. 1 we show the exact dependence of $\Delta\phi^{\mathrm{NL}}$ on Γ^2L^2 as calculated by a numerical solution of Eq. (4) for several values of ΔkL without spatial and temporal averaging. This shows that for large n_2^{eff} the approximation is valid only for small nonlinear phase shifts.

The depletion curve of Fig. 2(a) is a plot of $|E_1(z' = L)|^2/|E_0|^2$ as a function of the phase mismatch with no spatial or temporal integration. The data of Fig. 2(b) are for a 1-mm-thick hydrothermally grown sample of KTP, using 27-ps (FWHM), 1.06-μm pulses, focused to a measured Gaussian waist of 35 μm (half-width at $1/e^2$ of maximum). The result for $I = 9.4$ GW/cm^2 at $\Delta kL = 0$, where the spatial and temporal averaging can be readily performed, gives a value of $\Gamma^2L^2 \cong 4.2$, corresponding to $d_{\mathrm{eff}} \cong$ 3.1 pm/V, which agrees with the results of Ref. 9. Owing to the large depletion observed (>50%), it is important to check that two-photon absorption does not contribute to the depletion. Z-scan measurements of the two-photon absorption coefficient at 532 nm yield a value of 0.1 cm/GW, which gives a depletion much smaller than that due to SHG. In Fig. 2(c), approximate and exact solutions for $\Delta\Phi^{\mathrm{NL}}$ are shown as a function of the phase mismatch ΔkL, again with no space–time integration.

In our initial phase-measurement experiments we performed closed-aperture Z scans at $\phi = \pm 10°$

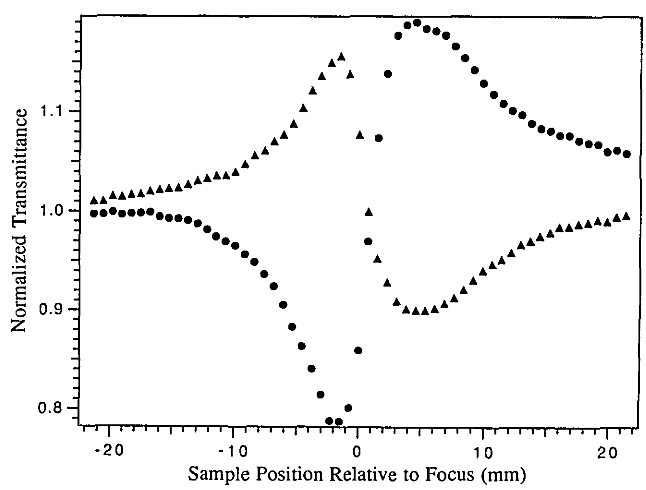

Fig. 3. Z scans showing positive $\Delta\Phi^{\mathrm{NL}}$ (circles) performed at $\Delta kL = -6$ and negative $\Delta\Phi^{\mathrm{NL}}$ (triangles) performed at $\Delta kL = +6$. The positive $\Delta\Phi^{\mathrm{NL}}$ is significantly larger owing to the positive contribution of n_2 (Kerr).

($\Delta kL \cong \pm 2\pi$), which correspond to minima in the SHG signal. These scans,[7] shown in Fig. 3, show a change in sign of $\Delta\Phi^{NL}$ in accordance with the predictions.[6] However, an asymmetry is observed, which indicates that there is noticeably more self-focusing than self-defocusing. This is explained by the presence of the bound electronic Kerr effect, n_2 (Kerr), which adds a positive phase distortion irrespective of Δk. We measured this n_2 (Kerr) to be $\cong(2.4 \pm 0.5) \times 10^{-15}$ cm^2/W by Z scanning with the beam propagating along the crystalline z axis, where $d_{eff} = 0$. This is consistent with n_2 (Kerr) obtained from the asymmetry shown in Fig. 3. We then find that $n_2^{eff} \cong \pm(1.3 \pm 0.3) \times 10^{-14}$ cm^2/W at $I \cong$ 26 GW/cm^2 for $\phi = \pm 10°$ ($\Delta kL \cong \pm 2\pi$), where depletion is minimized. Since at this irradiance, the nonlinearity deviates from the n_2^{eff} approximation, this measured value should be somewhat lower than the small-signal value. The maximum n_2^{eff} should occur at $\phi \cong \pm 5°$ ($\Delta kL \cong \pm 3$) and have a value of $\pm 2 \times 10^{-14}$ cm^2/W ($\pm 1 \times 10^{-11}$ esu).

In order to obtain a plot of $\Delta\Phi^{NL}$ versus phase mismatch we monitored the transmittance through a far-field aperture with $\cong$40% linear transmittance as a function of angle with the sample placed at the position along the beam path that gives minimum transmittance and repeated this with the sample placed at the position of maximum transmittance. As described in Refs. 7 and 10, the difference between the transmittance maximum and minimum is approximately proportional to $\Delta\Phi^{NL}$. The result of this subtraction is shown in Fig. 2(d), where the measured value of n_2 (Kerr) was also subtracted. This curve shows qualitative agreement with the theoretical curve for $\Delta\Phi^{NL}$ shown in Fig. 2(c).

Several conclusions can be drawn from the above observations. While n_2^{eff} for the 1-mm sample of KTP can be approximately as large as that for CS_2, this n_2^{eff} is linearly dependent on the sample thickness, which permits considerably larger values. The maximum n_2^{eff} occurs for a constant value of $\Delta kL \cong \pm 3$, thus we have the linear dependence on L shown in Eq. (7). Also n_2^{eff} scales as the square of d_{eff} such that larger values will greatly enhance n_2^{eff}. Thus values of 10^{-11} cm^2/W ($\cong 10^{-8}$ esu) can be expected for long, high-$\chi^{(2)}$ materials. Clearly organics are of interest here owing to their large $\chi^{(2)}$ values. Conceivably, organics with a d_{eff} of the order of 100 pm/V can lead to ultrafast all-optical switching with low loss to the fundamental beam by using picojoule pulses over an interaction length of a few hundreds of wavelengths. However, because of the saturable nature of $\Delta\Phi^{NL}$, one must use caution when quoting n_2^{eff}, as Fig. 1 clearly illustrates. In reality, it is the phase $\Delta\Phi^{NL}$ that is the more important parameter, and the advantage of using this method of achieving nonlinear refraction will depend on the particular application and the magnitude of $\Delta\Phi^{NL}$ that it requires.

The availability of an ultrafast nonlinearity that can be tuned in sign opens new device possibilities. For example, the fast electronic negative Kerr-like nonlinearity in a cascading process leads to self-compression of ultrashort pulses in the presence of positive group-velocity dispersion. This mechanism recently has been demonstrated in an optical parametric oscillator.[3] Another example of an application of this nonlinearity is mode locking of lasers using the recently reported Kerr mode-locking technique. For example Carruthers and Duling[11] report mode locking a cw Nd:YAG laser using KTP in an antiresonant ring cavity, where the mode locking was achieved for an angle tuning slightly off phase match. The induced self-phase modulation from the cascaded process may be the nonlinearity responsible for this mode locking.

In addition the Z-scan technique yields a new and accurate, absolutely calibrated method to measure d_{eff}, which requires only a measurement of the irradiance and either the loss or phase shift on the fundamental beam.

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