

Nonlinear Phase Shifts Using Second Order Nonlinearities

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A continuing problem in nonlinear optics has been to find third order materials with large, fast nonlinearities in spectral regions of low linear and nonlinear absorption. Typical applications of such materials require large nonlinear phase shifts. It has been known, but not widely appreciated, that second order nonlinear interactions lead to effective third order nonlinearities and nonlinear phase shifts in the fundamental beam. While such phenomena are implicitly included in the standard equations governing, for example, secondharmonic generation (SHG) and have been theoretically predicted previously,¹ only recently has the phase distortion been studied in detail experimentally and theoretically.^{2,3}

Physically, the nonlinear refraction at the fundamental arises when the second harmonic is down-converted back to the fundamental with a shifted phase (*i.e.*, a polarization is produced at 90° to the fundamental). Such a phase shift is a result of the wavevector mismatch Δk between the two beams and, therefore, its sign can be varied from positive to negative by controlling the orientation (or temperature) of the crystal going through a null at phase match. The figure shows this phase shift, $\Delta \Phi$, measured as a function of ΔkL in an L=1 mm thick KTP sample along with the depletion of the fundamental at 1 μ m.²

At low irradiance, $\Delta\Phi$ is linear in irradiance, making the discussion of this phenomena as an effective $\chi^{(3)}$ reasonable. This process can be simply viewed as obtaining nonlinear refraction via the cascading of $\chi^{(2)}(\omega; 2\omega, -\omega)$ with $\chi^{(2)}(2\omega; \omega, \omega)$. The resulting effective nonlinear refractive index is proportional to the usual figure-of-merit for $\chi^{(2)}$ materials, d^2_{eff}/n^3 and, for a fixed phase mismatch, to L (sample thickness).² For $\Delta\Phi$ larger than $\simeq \pi/4$, this approximation breaks down and the coupled equations must be solved exactly. Performing this numerical integration, we find for high irradiance that $\Delta\Phi$ grows in steps of maximum value $\pi/2$ and scales effectively as \sqrt{I} . Also, the maximum change of phase occurs at the positions of maximum depletion (maximum SHG).

The solid lines in the figure are the theoretical results for KTP using a d_{eff} of 3.1 pm/V. Here the approximation of a n^e is nearly valid and has a maximum value of $n^{e}_{2} \simeq \pm 2 \times 10^{-1}$ ¹ cm²/W for the 1 mm crystal. Clearly, organic Materials are of interest due to their large second order nonlinearities, (orders of magnitude larger than that of the KTP).² However, one must use caution when quoting n^{eff} for the reasons given above and because it is $\Delta \Phi$ that is the more important parameter. The advantage of using this method of achieving phase shifts will depend on the particular application and the magnitude of $\Delta \Phi$ required. The advantages that come with long propagation lengths and precise control over the phase mismatch indicate that quasi-phasematched waveguides may prove to be the best media for devices based on this effect. Furthermore, the possibility of inducing down-chirp (negative n₂) in the presence of normal (positive) GVD can be promising for soliton propagation and



(a) Peak nonlinear phase shift versus the phase mismatch measured for 1mm thick KTP at λ =1.06 µm with a peak irradiance of ~10 GW/cm². (b) The corresponding normalized depletion of the beam due to SHG process. The solid lines are the calculated results using d_{arr}=3.1 pm/V.

pulse compression. Lastly, due to the coherent nature of the second order process, phase and amplitude of a weak SH seed beam will strongly affect the fundamental output.⁴

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