Large nonlinear phase shifts in second-order nonlinear-optical processes

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We show that processes such as second-harmonic generation and subsequent downconversion, and parametric mixing in general, can lead to large field-dependent phase shifts for the input beams under a variety of conditions.

There are many potential applications for materials in which a single (or multiple) incident beam experiences a field-dependent phase shift or local change in refractive index. Typically this has been achieved with an intensity-dependent refractive-index coefficient n_2 ($n = n_0 + n_2 I$) originating from different processes such as electronic $\chi^{(3)}$ susceptibilities. Currently the best materials appear to be organics such as conjugated polymers, which exhibit nonresonant nonlinearities out of the ground state of 10^{-12} cm^2/W in low-loss spectral regions and even larger nonlinearities when virtual transitions out of optically pumped excited states are used.[1,2] In this Letter we show that a widely used phenomenon, second-harmonic generation (SHG), can in certain limits produce large ($>\pi$) nonlinear phase shifts.

SHG was one of the first nonlinear processes investigated and has been discussed extensively in the literature.[3] The emphasis has been on efficient SHG, which requires wave-vector matching of the fundamental to the second harmonic. It is also known, but not widely appreciated, that general formulations of nonlinear optics contain processes that involve the product of second-order nonlinearities and lead to effective third-order nonlinearities.[4–8] To our knowledge the first application of this phenomenon was to produce phase and amplitude distortion in intense pulses in GaAs by means of a combination of the dc field generated by $\chi^{(2)}(0;\omega,-\omega)$ and the electro-optic effect through $\chi^{(2)}(-\omega;\omega,0)$.[9] The existence of an effective n_2 was demonstrated experimentally in CDA and KTP crystals.[7,8] In the KTP case, the $d_{\text{eff}}^{(2)}$ was only a few picometers per volt; and the effective n_2 was of the order of 10^{-14} cm^2/W for a 1-mm length. However, based on the recent development of new ways to use large existing nonlinearities through waveguides and new organic materials with large nonlinearities, we have found that the cascading approach now holds considerable promise for use with phenomena that require a nonlinear phase shift.[9] Initial discussions of the theory leading to an effective n_2 were given in Refs. 7 and 8. Here we discuss more-general features, including the question of whether an n_2 description of this phenomenon is appropriate.

Starting from the wave equation driven by polarization sources including $\chi^{(1)}$ and $\chi^{(2)}$, and using the slowly varying phase and amplitude approximation, one obtains the usual coupled-mode equations that describe SHG,[7,8] namely,

$$\frac{\mathrm{d}}{\mathrm{d}z}a_{2\omega}(z) = -i\kappa(-2\omega;\omega,\omega)a_\omega{}^2(z)\exp(i\Delta\beta z) - \alpha(2\omega)a_{2\omega}(z), \tag{1}$$

$$\frac{\mathrm{d}}{\mathrm{d}z}a_\omega(z) = -i\kappa(-\omega;2\omega,-\omega)a_{2\omega}(z)a_\omega{}^*(z) \times \exp(-i\Delta\beta z) - \alpha(\omega)a_\omega(z), \tag{2}$$

$$\kappa(-2\omega;\omega,\omega) = \frac{\omega d_{ijk}^{(2)}(-2\omega;\omega,\omega)e_i(2\omega)e_j(\omega)e_k(\omega)}{[2n_i(2\omega)n_j(\omega)n_k(\omega)c^3\epsilon_0]^{1/2}}, \tag{3}$$

and a similar expression for $\kappa(-\omega;2\omega,-\omega)$. Here the wave-vector mismatch is $\Delta\beta = 2k_{\text{vac}}(\omega)[n(2\omega) - n(\omega)]$, the complex field amplitudes $a(z)$ are normalized so that $|a(z)|^2$ is the intensity, the e_i's are the field unit vectors, and α is the frequency-dependent linear loss. For frequency-independent loss, far from any material resonances, and in the absence of coupling to other fields, these equations can be solved analytically in terms of Jacobi elliptic functions.[10] We numerically solved coupled-mode Eqs. (1) and (2).

We first examine approximate solutions in the limit of small fundamental depletion, i.e., $|a_\omega(z)| \simeq a_\omega(0)$ and $\alpha(\omega) = 0$.[7,8] Integrating Eq. (1), substituting into Eq. (2), and noting that the imaginary part of the right-hand side of Eq. (2) can be approximately written as $-in_2(z)k_{\text{vac}}|a_\omega(0)|^2 a_\omega(z)$, we get

$$n_2(z) \simeq \mathrm{Im}\left[\kappa(-2\omega;\omega,\omega)\kappa(-\omega;2\omega,-\omega) \times \frac{1 - \cos_c(\psi z) + i\,\sin_c(\psi z)}{k_{\text{vac}}(i\psi)}\right], \tag{4}$$

where $\psi = \Delta\beta - i\alpha(2\omega)$ and the subscripts c refer to trigonometric functions with complex arguments. [The real part of the right-hand side of approximation (4) corresponds to the usual fundamental depletion for $\Delta\beta L \simeq 0$.] Note that, although one can formally define an n_2, it varies with distance z even in this limit of small depletion.

From approximation (4) an effective n_2 can be obtained in different ways. For ω and 2ω far

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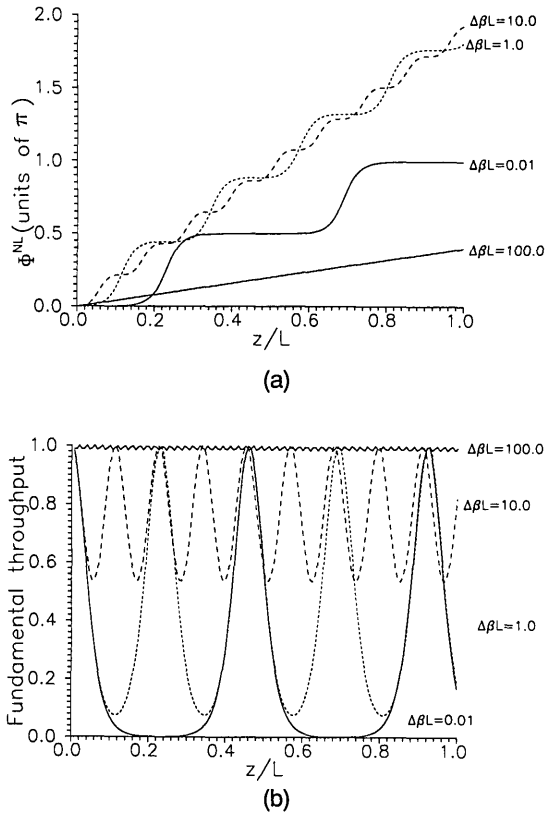

Fig. 1. Variation in (a) the nonlinear phase shift ϕ^{NL} and (b) the fractional fundamental intensity $|a_\omega(z)|^2/|a_\omega(0)|^2$ with z for different detunings $\Delta\beta L = 0.01, 1.0, 10, 100$. Here $\kappa L = 4$ and $|a_\omega(0)|^2 = 25$.

from any material resonance frequency (ω_r) so that $\kappa^{(2)}(-2\omega;\omega,\omega)$ and $\kappa^{(2)}(-\omega;2\omega,-\omega)$ are real or complex conjugates, a wave-vector mismatch $\Delta\beta L \neq 0$ is required for $n_2 \neq 0$.[7,8] When either ω or 2ω is near a resonance, there is an effective n_2, even for $\Delta\beta = 0$. We discuss these two limiting cases.

To examine the simplest limit of approximation (4), we first assume that $\alpha(2\omega) = 0$. When $\Delta\beta L = \pm\pi$ is chosen, n_2 is maximized, with $n_2 = \mp 2|d_{\text{eff}}|^2/n^3 c\epsilon_0 \times L/\lambda$.[8] This effective nonlinearity can be either positive (self-focusing) or negative (self-defocusing), depending on the sign of $\Delta\beta$. These features and the magnitude of n_2 were recently measured by a Z scan in KTP.[8] For a periodically poled polymer waveguide with optimized parameters $d^{(2)} \sim 50$ pm/V,[11] $n \sim 1.8$, $L/\lambda \sim 10^4$, and $n_2 \sim 6 \times 10^{-11}$ cm^2/W. Given that $d^{(2)}$ in excess of 40 pm/V have been reported,[12] this approach to nonlinear phase shifts should prove interesting.

The physical origin of this effective third-order nonlinearity is straightforward. When the SHG process is mismatched, the second-harmonic field propagates with the wave vector $2n(2\omega)k_{\text{vac}}(\omega) \neq 2n(\omega)k_{\text{vac}}(\omega)$. Therefore the product $E_{2\omega}E_\omega{}^*$ in Eq. (2) produces a polarization source term with a component in quadrature with the fundamental and hence slows it down ($\Delta\beta < 0$) or speeds it up ($\Delta\beta < 0$).

We now generalize to include pump depletion. Shown in Fig. 1(a) is the evolution of the nonlinear phase shift ϕ^{NL} of the fundamental beam with the normalized distance z/L for different values of wave-vector detuning $\Delta\beta L$, where L is the sample length. Here ϕ^{NL} is defined by $a_\omega(z) = |a_\omega(z)|\exp[i\phi^{NL}(z)]$, and in terms of n_2 usually by $\phi^{NL}(z) = \int_0^z n_2(z')k_{\text{vac}}|a_\omega(z')|^2 dz'$. The corresponding spatial variation of the normalized fundamental power, $|a_\omega(z)|^2/|a_\omega(0)|^2$, is given in Fig. 1(b).

ϕ^{NL} accumulates in a basically stepwise fashion, one step for every full oscillation in the fundamental (and harmonic) intensity, with a maximum step of $\pi/2$ for $\Delta\beta L \ll 1$. As $\Delta\beta L$ becomes larger, the steps become progressively more smoothed out, smaller in phase change, and more frequent along the propagation. Note that the step-averaged phase change remains linear in κL up to very large phase changes ($>20\pi$ investigated numerically). Because the number of oscillations in the fundamental increases and the step in ϕ^{NL} per oscillation decreases as $\Delta\beta L$ increases, there is a value of $\Delta\beta L$ for which the rate of increase of ϕ^{NL} with κL is an optimum. This optimum $\Delta\beta L$ increases with increasing input intensity.

The variation in $\phi^{NL}(L)$ as a function of input intensity [$|a_\omega(0)|^2$] is shown in Fig. 2(a). A stepwise variation change is also obtained with increasing intensity. For small depletion and phase shifts, ϕ^{NL} is linear in incident intensity. However, its increase with $|a_\omega(0)|^2$ becomes sublinear for high intensities, consistent with the larger intensity increment between the progressively deeper oscillations in the fun-

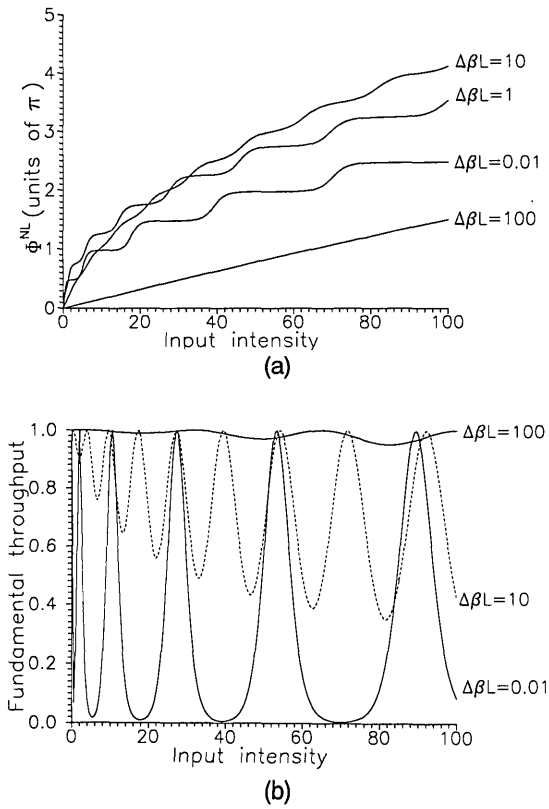

Fig. 2. Variation in (a) the nonlinear phase shift ϕ^{NL} and (b) the normalized fundamental intensity $|a_\omega(L)|^2/|a_\omega(0)|^2$ versus input intensity for various $\Delta\beta L$ and $\kappa L = 4$.

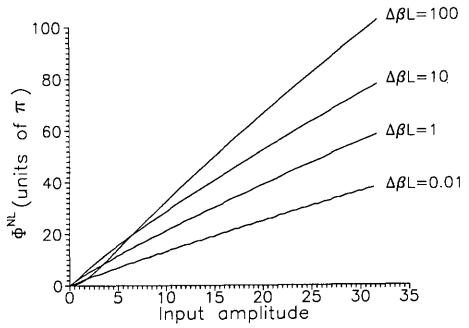

Fig. 3. Variation in the nonlinear phase shift with input amplitude $|a_\omega(0)|$ for large phase shifts and $\Delta\beta L = 0.01$, 1.0, 10, 100. Here $\kappa L = 32$.

damental as a function of input, shown in Fig. 2(b). As Fig. 3 shows, the increase in ϕ^{NL} for large phase changes becomes quasi-linear in input fundamental amplitude. Such large changes might require unrealistically large input intensities.

A number of features are unique to this nonlinear phase change. The first is the stepwise variation with distance and intensity. The distance and intensity increments required for a step phase change increase as $\Delta\beta L \rightarrow 0$, with the steps becoming progressively steeper. Because $d[\phi^{NL}(z)]/dz$ and $d(\phi^{NL})/d|a_\omega|^2 \propto n_2$, the corresponding effective n_2 becomes essentially a series of δ functions. For this reason, and the fact that $\phi^{NL} \propto |a_\omega(0)|$ for large phase shifts, we conclude that an n_2 description for this process is not appropriate and that the key variable is the nonlinear phase change itself (which is frequently the important device parameter).[13]

There are a number of potential problems with using this process for obtaining large nonlinear phase shifts. The oscillation in the fundamental beam power with distance is the principal drawback. If the interaction is terminated at the incorrect length, the effective loss for the fundamental, i.e., SHG conversion, can be large. This is effectively a two-photon loss.[8] Also, there is a finite bandwidth usually associated with phase matching, and this will impose a limit to the bandwidth of usable pulses. Furthermore, a noncentrosymmetric medium is required. Note that the only potential limits to large phase shifts are material damage and the usual problems for efficient SHG.

The only optical beams that experience a nonlinear phase shift are those linked by second-order coefficients. This means that the usual index-change-mediated cross-phase modulation associated with an electronic n_2 process does not exist. However, when three beams of different frequency and appropriate polarization are almost phase matched for sum- or difference-frequency generation, nonlinear phase shifts can occur for all interacting beams. In addition, a probe beam can experience a form of cross-phase modulation through an orthogonally polarized pump beam if both fundamental polarizations are linked by nonzero $d^{(2)}_{ijk}$ to the same SHG wave.

Finally, we note that the $\chi^{(2)}$ can be resonantly enhanced, leading to even larger phase shifts when either ω_1 or ω_2 is near a material resonance, even in the limit of phase-matching $\Delta\beta = 0$. For simplicity we choose $\alpha(2\omega) = 0$. [For a realistic case $\alpha(2\omega)$ must be included, because the same electronic states give rise to both the complex $d^{(2)}$ and $\alpha(2\omega)$.] When an anharmonic oscillator model is used, $d^{(2)}_{eff}(-2\omega; \omega, \omega) = d^{(2)}_{nr}(-2\omega; \omega, \omega)\{\omega_r{}^6/[D(2\omega)D^2(\omega)]\}$, where nr and r refer to nonresonant and resonant, respectively. Here $D(\omega) = (\omega_r{}^2 - \omega^2 = i\omega\Gamma)$, with Γ the linewidth. For $d^{(2)}(-\omega; 2\omega, -\omega)$, $D^2(\omega)$ is replaced by $|D(\omega)|^2$ so that $d^{(2)}(-2\omega)$ and $d^{(2)}(-\omega)$ are no longer complex conjugates. A nonlinear phase shift is obtained whose sign depends on the sign of the frequency detuning from resonance. Decreasing such detuning results in an enhancement of $d^{(2)}$ and consequently in larger ϕ^{NL}.

In summary, macroscopic cascading of second-order nonlinearities leads to a nonlinear phase shift of the fundamental beam under a variety of conditions. These phase shifts can require intensities orders of magnitude lower than required by the usual third-order nonlinearities. Some unique features, specifically the stepwise response, can be useful for devices and will be reported in subsequent publications. Another interesting problem for future work is the trade-off between the enhancement and the loss near resonance.

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