Effects of cubic nonlinearity on frequency doubling of high-power laser pulses

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The effects of the phase mismatch due to cubic nonlinearity in the equations for second-harmonic generation are investigated. We show that the phase mismatch induced by the nonlinear refractive index of a doubling crystal can dramatically reduce the conversion efficiency of high-peak-power laser pulses. Simple, analytic expressions are derived for the conversion efficiency of cw radiation and for the estimation of the dispersion in the nonlinear refractive index of the doubling crystal, which is quite important in the determination of the magnitude of the nonlinear effects on maximum conversion. The consequences that these nonlinearities have on the frequency doubling of ultrashort (≤100 fs) pulses, including the additional effects of group-velocity walk-off between the pulses, are then numerically calculated.

1. INTRODUCTION

The phenomenon of second-harmonic generation in a nonlinear crystal has been extensively studied since its first observation in 1961.[1] The theory of three-wave interactions in a material with a quadratic nonlinearity has been well developed, and the exact solutions to this problem for second-harmonic generation by plane waves were advanced by Armstrong *et al.*[2] in 1962. The influence of the cubic nonlinearity on harmonic conversion was first addressed with the advent of high-power pulsed lasers. The effect was first studied by Akhmanov and Khokhlov[3] and has since been examined by a number of groups.[4,5] Telegin and Chirkin[6] reported the first experimental evidence of higher-order nonlinearities in second-harmonic frequency conversion. Recently, the exact solutions of the plane-wave conversion equations in the presence of a cubic nonlinearity have been worked out by Choe *et al.*[7] and McKinstrie and Cao.[8]

With the proliferation of high-peak-power, short-pulse lasers and the desire to convert their output, the effects of the higher nonlinearities on short-pulse frequency conversion need to be taken into account. In this paper we examine the intensity-dependent nonlinear phase mismatch resulting from the cubic nonlinearity and discuss its effects on the conversion of short pulses. The nonlinear phase mismatch can be compensated for a specific intensity, but the resulting shape of the second-harmonic pulse will be altered.[9] Furthermore, the inclusion of group-velocity walk-off, which can be a major effect for pulses of 100 fs or less,[10] can act to decrease the deleterious effects of the cubic nonlinearity and prevent reconversion from the intensity-induced phase mismatch.

2. ANALYTIC ANALYSIS OF SECOND-HARMONIC GENERATION WITH A $\chi^{(3)}$ NONLINEARITY

To understand the essential effects of a cubic nonlinearity on frequency doubling of laser pulses we start with the scalar wave equations governing the propagation of the drive wave and its second harmonic through a nonlinear crystal (in cgs units):

$$-\nabla^2 E_i(\mathbf{x}, t) + \frac{\varepsilon_i}{c^2}\frac{\partial^2}{\partial t^2}E_i(\mathbf{x}, t) = -\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}P_i^{\mathrm{NL}}(\mathbf{x}, t), \quad (1)$$

where the subscript i is equal to 1 or 2 and denotes the first- or second-harmonic field, respectively, ε_i is the dielectric constant for each of the two fields E_i, and P^{NL} is the nonlinear polarization term. The nonlinear polarization terms to the third order in the electric fields are

$$P_1^{\mathrm{NL}}(\mathbf{x}, t) = 4d_{\mathrm{eff}}E_2E_1^* + 3\chi^{(3)}(\omega_1, -\omega_1, \omega_1)|E_1|^2E_1 + 6\chi^{(3)}(\omega_2, -\omega_2, \omega_1)|E_2|^2E_1 + \mathrm{c.c.}, \quad (2a)$$

$$P_2^{\mathrm{NL}}(\mathbf{x}, t) = 2d_{\mathrm{eff}}E_1^2 + 3\chi^{(3)}(\omega_2, -\omega_2, \omega_2)|E_2|^2E_2 + 6\chi^{(3)}(\omega_1, -\omega_1, \omega_2)|E_1|^2E_2 + \mathrm{c.c.}. \quad (2b)$$

The relation of the effective nonlinear susceptibility d_{eff} to the second-order polarizability tensor χ_{ij} is determined by the second-harmonic crystal-point group and the doubling geometry (type I or type II).[11] For type I doubling in KDP, for example, it is given by $d_{\mathrm{eff}} = (1/2)\chi_{36}^{(2)}\sin\theta$, where θ is the phase-matching angle.

If we assume that the fields are infinite plane waves in the transverse (x, y) directions and ignore the effects of pulse broadening due to group-velocity dispersion, the use of the slowly varying envelope approximation yields equations describing second-harmonic production:

$$\frac{\partial a_1}{\partial z} = i\alpha a_2 a_1^* \exp(-i\Delta k z) + i(\beta_{11}|a_1|^2 a_1 + \beta_{21}|a_2|^2 a_1), \quad (3a)$$

$$\frac{\partial a_2}{\partial z} + \eta\frac{\partial a_2}{\partial t} = i\alpha a_1^2 \exp(i\Delta k z) + i(\beta_{22}|a_2|^2 a_2 + \beta_{12}|a_1|^2 a_2). \quad (3b)$$

Here, a_1 and a_2 are the slowly varying envelopes of the fundamental and the second-harmonic fields, respectively, and $\Delta k \equiv 2k_1 - k_2$ is the phase mismatch between the two fields. The expression $\eta = 1/\nu_{g1} - 1/\nu_{g2}$

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is the rate of the group-velocity walk-off between the fundamental pulse and its second harmonic. This term can be quite significant for 100-fs pulses that are doubled in crystals of only a few millimeters. It is, for example, equal to 77 fs/mm for a pulse with a wavelength of 800 nm in KDP.[12] In addition, α is the coupling coefficient between the fundamental and the second harmonic and is given by $\alpha \equiv 8\pi\omega_1^2 d_{\text{eff}}/c^2 k_1$, where these symbols have their usual definitions. The final terms on the right-hand side of each expression in Eqs. (3) represent an intensity-dependent phase shift. These terms, in principle, add an additional phase mismatch even when $\Delta k = 0$. The coefficients are

$$\beta_{ij} \equiv 2^{|i-j|}\,\frac{6\pi\omega_j^2 \chi^{(3)}(\omega_i, -\omega_i, \omega_j)}{c^2 k_j}.$$

To examine the physical effect of the nonlinear phase terms in Eq. (1) in the absence of group-velocity walk-off and to estimate the effect on the pulse frequency conversion efficiency, it is convenient to rewrite the equations in Hamiltonian form.[9] First we introduce the normalized propagation distance, $z = s/\alpha$, and phase mismatch, $\Lambda = \Delta k/\sqrt{2}\alpha$. Because we are concerned only with the intensities of the two fields, we can introduce an arbitrary phase to the field envelopes and renormalize them. We now define the new fields[8]: $b_1 = (1/\sqrt{2})a_1 \exp(i\Lambda s - i\delta \int_0^s |a_2|^2\,\mathrm{d}s)$ and $b_2 = (1/2)a_2 \exp(-i2\delta \int_0^s |a_2|^2\,\mathrm{d}s)$. And we further define δ such that $\delta \equiv [\beta_{21} - (1/2)\beta_{12}]/\alpha$. The equations for b_1 and b_2 then become

$$\frac{\partial b_1}{\partial s} = 2ib_2 b_1^* + i\Lambda b_1 + i(T_{11}|b_1|^2 b_1 + T_{21}|b_2|^2 b_1), \quad (4a)$$

$$\frac{\partial b_2}{\partial s} = ib_1^2 + i(T_{22}|b_2|^2 b_2 + T_{12}|b_1|^2 b_2), \quad (4b)$$

where the nonlinear phase coefficients are $T_{11} = 2\beta_{11}/\alpha$, $T_{12} = T_{21} = 2\beta_{12}/\alpha$, and $T_{22} = 4\beta_{22}/\alpha - 8\delta$. Equations (4) represent a Hamiltonian system that can be represented by the relations

$$\frac{\partial b_1}{\partial s} = i\,\frac{\delta H}{\delta b_1^*}, \qquad \frac{\partial b_2}{\partial s} = i\,\frac{\delta H}{\delta b_2^*}. \quad (5)$$

The spatially invariant Hamiltonian is

$$H = b_2 b_1^{*2} + b_1^2 b_2^* + \Lambda|b_1|^2 + \frac{1}{2}T_{11}|b_1|^4 + T_{12}|b_1|^2|b_2|^2 + \frac{1}{2}T_{22}|b_2|^4. \quad (6)$$

Next, we turn to the Manley–Rowe relations and the initial conditions to calculate the value of H. For the renormalized fields this relation can be written as

$$|b_1|^2 + 2|b_2|^2 = \text{const}. \quad (7)$$

The initial condition is taken to be such that there is no second harmonic initially incident on the crystal face:

$$b_1(x = 0) = b_0, \quad (8a)$$

$$b_2(x = 0) = 0. \quad (8b)$$

Consequently

$$H = \frac{1}{2}T_{11}|b_0|^4 + \Lambda|b_0|^2, \quad (9)$$

$$|b_1| = \sqrt{|b_0|^2 - 2|b_2|^2}. \quad (10)$$

At this point it is convenient to introduce the magnitude of the field amplitudes and the field phases:

$$b_1 = |b_1|\exp[i\phi_1], \qquad b_2 = |b_2|\exp[i\phi_2],$$

where $\phi \equiv \phi_2 - 2\phi_1$. The equation for the evolution of $|b_2|$ is

$$\frac{\partial|b_2|}{\partial s} = |b_1|^2 \sin\,\phi. \quad (11)$$

Using Eqs. (10) and (11) and the fact that H is conserved, we can write

$$2|b_2|\,|b_1|^2\cos\,\phi + \Lambda|b_1|^2 + \frac{1}{2}T_{11}|b_1|^4 + \frac{1}{2}T_{22}|b_2|^4 + T_{12}|b_1|^2|b_2|^2 = \Lambda b_0^2 + \frac{1}{2}T_{11}b_0^4, \quad (12)$$

which yields a complete system for the evaluation of the maximum conversion. From Eq. (11) we expect that b_2 is maximized when $\partial b_2/\partial s = 0$, or, namely, when $\phi = 0$ or $\phi = \pi$. From this it follows that $\cos\,\phi = \pm 1$, with the sign chosen such that the conversion efficiency does not exceed unity. For the case of high conversion, we can treat the nonlinearities as a small perturbation and use perturbation theory to estimate the maximum of b_2. If we assume that the zeroth-order solution is $|b_2|^2 = b_0^2/2$ we have

$$|b_2|^2_{\max} \approx \frac{1}{2}b_0^2 - \frac{1}{\sqrt{2}}\left|\left(\frac{1}{4}T_{22} - T_{11}\right)b_0^3 - 2\Lambda b_0\right|. \quad (13)$$

In the case of no phase mismatch ($\Lambda = \Delta k/\sqrt{2}\alpha = 0$) and no dispersion in the nonlinear index of refraction, then $T_{22} = 4T_{11}$ and we have a 100% conversion efficiency ($|b_2|^2 = b_0^2/2$). This analytically derived result was observed recently in the numerical simulations of Chien *et al.*[13] without explanation. The fact that 100% conversion can be achieved in the presence of a nonlinearity with no wavelength dispersion is due to the fact that, although the nonlinear phase mismatch grows as the driving laser propagates through the doubling crystal, as energy is transferred to the second harmonic the mismatch changes signs and cancels the phase slip caused by the fundamental. As the conversion approaches 100% the phase accumulated by the second harmonic approaches that already accumulated by the fundamental field, and the efficiency asymptotically approaches unity. In general, however, the nonlinear refractive index will exhibit some dispersion and the conversion efficiency will be limited.

From Eq. (12) we see that it is possible to compensate for the nonlinear phase mismatch by deliberately introducing a mismatch such that

$$\Lambda = \frac{1}{2}\left(\frac{1}{4}T_{22} - T_{11}\right)b_0^2. \quad (14)$$

In this case, the conversion efficiency asymptotically approaches 100% for a single incident intensity. This is

an exact result derived from Eq. (9), which implies that 100% conversion is possible for an arbitrarily large incident field. This result was derived previously in more cumbersome calculations by Choe *et al.*,[7] as well as by McKinstrie and Cao.[8] For the case of a pulse with a temporally varying envelope it is possible to compensate the nonlinear effects for only a single intensity in the pulse, as was pointed out by Chien *et al.*[13] In the absence of group-velocity walk-off effects the pulse can be considered as a set of independent slices in time. If the nonlinear phase mismatch is removed for one intensity slice, the others still experience a drop in their conversion efficiencies, and reconversion will commence at different points in the pulse. Although 100% conversion is possible for a flattop pulse if the crystal phase mismatch is properly tuned, the conversion efficiency for a time-varying pulse will go through a distinct maximum. It is important to point out that a time-varying pulse will experience an intensity-dependent phase mismatch, resulting in the growth of temporal gradients in the profile of the fundamental and the second harmonic. Group-velocity walk-off effects can then become important even for long pulses.

3. NUMERICAL CALCULATION OF THE NONLINEAR PHASE MISMATCH AND MAXIMUM CONVERSION EFFICIENCY

It is instructive to investigate numerical calculations for the second-harmonic conversion efficiency for parameters of interest to the doubling of current high-power laser systems. For this reason, we have examined the doubling of 800-nm light in a KDP crystal because this is a situation that is of most interest for the frequency conversion of ultrashort pulses amplified in Ti:sapphire. These calculations illustrate the relative magnitudes of the effects described above. For these calculations we take for KDP, $d_{\mathrm{eff}} = 8.7 \times 10^{-10}$ esu. To calculate the magnitude of the nonlinear phase mismatch we require an estimate of the dispersion of $\chi^{(3)}$ between the first- and the second-harmonic wavelengths in the doubling crystal (between 800 and 400 nm). Recent measurements of the nonlinear refractive index of a variety of wide-band-gap crystals at wavelengths of 1064, 532, and 355 nm by Adair *et al.*[14] suggest that the dispersion of $\chi^{(3)}$ can be quite substantial (nearly a factor of 2 between 1- and 0.3-μm light in fused silica, for example.) We expect then that the nonlinear phase mismatch can be important for pulse conversion.

Because no reliable data exist, however, on the values of $\chi^{(3)}$ for common doubling crystals (such as KDP), we require a reasonable estimate for the variation of $\chi^{(3)}$ between 800 and 400 nm. To do this we resort to a simple model that permits us to estimate the self- and cross-phase modulation contributions of $\chi^{(3)}$ for a crystal of known linear refractive index. To evaluate the perturbation-series summation for $\chi^{(3)}$ we use the simple technique of Bebb and Gold.[15] A simplified version of their theory was shown by Adair *et al.*[14] to be in reasonable agreement with the measured dispersion of the n_2 of a number of optical materials.

The $\chi^{(3)}$ of the doubling crystal is calculated by an explicit summation of the perturbation series, but with the assumption that all intermediate states are centered at an effective energy of $\hbar\omega_0$. When the summation is performed the $\chi^{(3)}$ term, representing the nonlinear phase shift of one field with an arbitrary frequency ω_i, on a second with a frequency ω_j can be expressed as where

$$\chi^{(3)}(\omega_i, -\omega_i, \omega_j) = \frac{e^4 N}{6\hbar^3\omega_0^3}\left(\langle r^4\rangle\left\{\frac{4}{(1-x_i^2)(1-x_j^2)} + \frac{2}{(1-x_i)(1-x_j)[1-(x_i-x_j)]}\right.\right.$$
$$+ \frac{2}{(1-x_i)(1+x_j)[1+(x_i-x_j)]} + \frac{2}{(1+x_i)(1+x_j)[1+(x_i+x_j)]}$$
$$+ \frac{2}{(1-x_i)[(1-x_j)(1-(x_i-x_j)]} + \frac{2}{(1-x_j)[(1-x_j)^2-x_i^2]} + \frac{2}{(1+x_j)[(1+x_j)^2-x_i^2]}$$
$$\left.\left. + \frac{2}{(1-x_i)[(1-x_i)^2-x_j^2]} + \frac{2}{(1+x_i)[(1+x_i)^2-x_j^2]}\right\} - \langle r^2\rangle\frac{4[2-(x_i^2+x_j^2)]}{(1-x_i^2)^2(1-x_j^2)^2}\right), \tag{15}$$

$x_{i,j} = \omega_{i,j}/\omega_0$. For simplicity we have assumed that the dipole matrix elements r_{ab} are equal for any direction of field polarization (which is not necessarily true for a birefringent crystal.) Following Adair *et al.*,[14] we have defined $\langle r^4\rangle = \sum_{a,b,c} r_{ga}r_{ab}r_{bc}r_{cg}$ and $\langle r^2\rangle = \sum_a r_{ga}r_{ag}$, where g denotes the ground state and a, b, and c denote the upper states. The value of the effective energy level $\hbar\omega_0$ can be inferred from known values of the linear refractive index of the crystal if the refractive index is fitted to the Mossotti formula:

$$\frac{n(x_i)^2+2}{n(x_i)^2-1} = \frac{3\hbar\omega_0}{8\pi e^2 N\langle r^2\rangle}(1-x_i^2). \tag{16}$$

To derive specific values of $\chi^{(3)}$ we exploit the fact that the Kurtosis, defined as $\langle r^4\rangle/2\langle r^2\rangle - 1$, is approximately equal to 0.5 for most wide-band-gap crystals.[16] The use of Eqs. (15) and (16), combined with published values for the linear refractive index and the nonlinear refractive index of the crystal at 1 μm, allow us to make estimates for the $\chi^{(3)}$ terms in Eqs. (2). It should be emphasized that this model gives us estimates only for the actual values of $\chi^{(3)}$ and probably underestimates the value, particularly at shorter wavelengths ($\lambda < 500$ nm).

We calculate the value of n_2 measured[16] at 1064 nm (which is 7.2×10^{-14} esu for ordinary polarization and 7.8×10^{-14} esu for extraordinary polarization) and Eq. (15) (with $\omega_0 = 1.7 \times 10^{16}$ s^{-1} for KDP) to find values for the $\chi^{(3)}$ components for the doubling of 800-nm light. We use values of $\chi^{(3)}(\omega_1, -\omega_1, \omega_1) = 1.2 \times 10^{-14}$ esu, $\chi^{(3)}(\omega_2, -\omega_2, \omega_1) = 1.4 \times 10^{-14}$ esu, $\chi^{(3)}(\omega_2, -\omega_2, \omega_2) = 1.8 \times 10^{-14}$ esu, and $\chi^{(3)}(\omega_1, -\omega_1, \omega_2) = 1.5 \times 10^{-14}$ esu,

(i.e., $T_{11} = 1.2 \times 10^{-8}$, $T_{21} = 2.9 \times 10^{-8}$, $T_{22} = 3.4 \times 10^{-8}$, and $T_{12} = 5.9 \times 10^{-8}$, respectively). It is also convenient to relate these coefficients to the material nonlinear index of refraction defined by the relation $\Delta n = \gamma I$, where Δn is the intensity-induced change in the index of refraction. Therefore, the nonlinear coefficients are $\gamma_{11} = 2.2 \times 10^{-20}$ m^2/W, $\gamma_{21} = 5.5 \times 10^{-20}$ m^2/W, $\gamma_{12} = 6.1 \times 10^{-20}$ m^2/W, and $\gamma_{22} = 3.0 \times 10^{-20}$ m^2/W.

It is important to note that Eq. (15) predicts that the dispersion in n_2 between 1054- and 527-nm light is significantly less than that between 800- and 400-nm light. It is therefore reasonable to expect that the nonlinear effects on doubling will be more severe for Ti:sapphire lasers operating at 800 nm than for Nd:YAG or Nd:glass lasers operating with a fundamental wavelength of approximately 1 μm. For example, our model predicts that the dispersion between the nonlinear refractive index for the fundamental at 800 nm and the second harmonic at 400 nm is $\chi^{(3)}(\omega_2, -\omega_2, \omega_2)/\chi^{(3)}(\omega_1, -\omega_1, \omega_1) = 1.5$, whereas the dispersion between 1054- and 527-nm light is predicted to be a factor of $\chi^{(3)}(\omega_2, -\omega_2, \omega_2)/\chi^{(3)}(\omega_1, -\omega_1, \omega_1) = 1.3$. As Eq. (13) illustrates, the drop in the conversion efficiency scales roughly linearly with the magnitude of this nonlinear index dispersion. The size of the dispersion predicted by our model for conversion of 1054-nm light is slightly lower than the value used by Chien *et al.*[12] to match their data. They empirically determined that using a dispersion of $\chi^{(3)}(\omega_2, -\omega_2, \omega_2)/\chi^{(3)}(\omega_1, -\omega_1, \omega_1) = 1.5$ seemed to fit their experimental data. This is consistent with our estimate that our model will underestimate the value of $\chi^{(3)}(\omega_i, -\omega_i, \omega_j)$ at shorter wavelengths. However, our prediction of the dispersion is roughly comparable with the value found by Chien *et al.*,[13] suggesting that our simple model is useful for making estimates when no data exist for the dispersion of n_2 in a doubling crystal of interest (at 800 and 400 nm in KDP, for example).

Using these numbers, we have calculated the maximum conversion efficiency by numerically solving Eq. (12). This solution is plotted in Fig. 1 as the solid curve. The approximate solution of Eq. (13) is also shown (dashed curve) to illustrate the validity of the perturbation formula. From Fig. 1 it is evident that, for incident intensities above 100 GW/cm^2, the maximum possible conversion efficiency begins to fall below 90%. The frequency conversion of current short-pulse laser systems is typically achieved with incident drive intensities of 100–500 GW/cm^2.[13] In this regime, the nonlinear phase mismatch can be an important factor in the determination of the conversion efficiency. With incident intensities approaching 500 GW/cm^2, the nonlinear phase can have very deleterious effects on the pulse frequency conversion,[13] lowering the maximum conversion by as much as 30%.

4. SIMULATIONS OF SHORT-PULSE FREQUENCY DOUBLING

To further investigate the effects of high incident intensities, we have numerically solved the coupled equations (3a) and (3b) for the frequency conversion of an 800-nm pulse. Figure 2 illustrates the energy conversion efficiency of a pulse that is flat in time, with an intensity of 500 GW/cm^2 in the absence of group-velocity walk-off. The calculated conversion efficiencies, both with and without the inclusion of the $\chi^{(3)}$ nonlinearity, as functions of the distance propagated in the KDP crystal are compared. In the absence of the nonlinearity nearly 100% conversion is achieved in <2 mm of KDP, whereas the conversion efficiency rolls over at 82% after the light has propagated 1.1 mm in the crystal when the nonlinear phase mismatch is included. Furthermore, the conversion efficiency drops down and the second harmonic undergoes complete reconversion to the fundamental at 2.2 mm into the crystal. This reconversion is similar to that observed when a geometric phase mismatch is introduced.

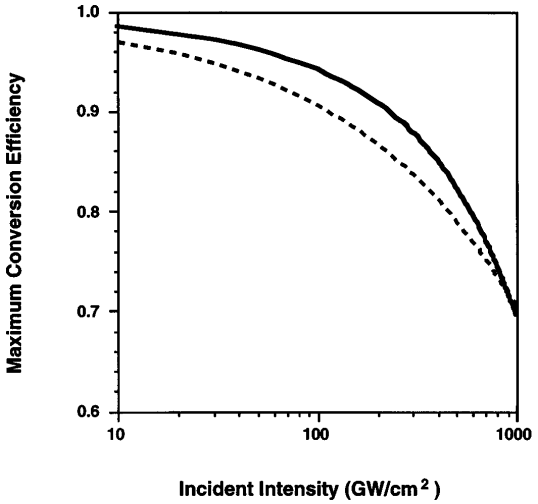


Fig. 1. Numerical solution of Eq. (12) for the maximum conversion efficiency for 800-nm light in KDP (solid curve). The approximate perturbation-theory formula, Eq. (13), is also shown for comparison (dashed curve).

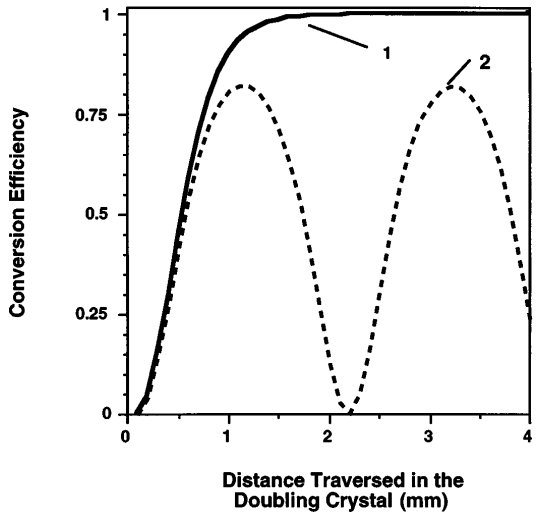
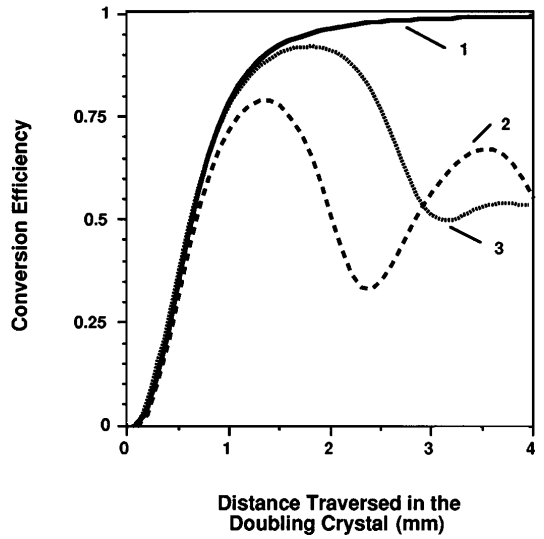
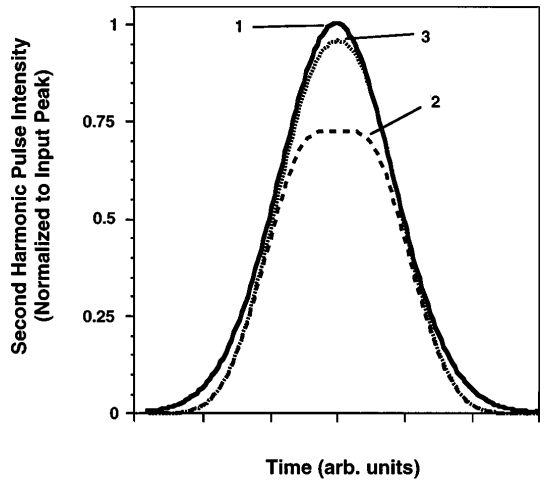


Fig. 2. Energy conversion efficiency calculated for a square pulse as a function of the distance propagated in a KDP crystal in the absence of group-velocity walk-off ($\eta = 0$). The incident intensity was 500 GW/cm^2, and the pulse wavelength was 800 nm. Curve 1 shows the case for harmonic conversion with no cubic nonlinearities ($\beta_{ij} = 0$), and curve 2 represents the efficiency when nonlinearities are included.



(a)



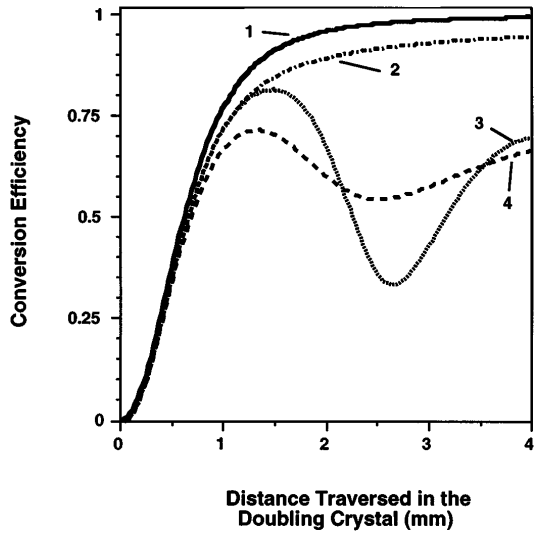
(b)

Fig. 3. Results for a Gaussian pulse shape: (a) The calculated conversion efficiency as a function of distance in a KDP crystal for a Gaussian pulse envelope with no group-velocity walk-off. The peak intensity of the incident pulse is 500 GW/cm^2. Curve 1 (solid) represents the case of no cubic nonlinearity, curve 2 (dashed) represents the case with the nonlinear phase mismatch included, and curve 3 (dotted) represents the case with the nonlinear phase mismatch and the inclusion of additional phase mismatch to compensate for the cubic phase slip at the peak intensity of the pulse. (b) Pulse envelopes of the second-harmonic pulse for the three cases calculated in (a), shown at the peak of the pulse-conversion curves. These positions are, for curve 1, $z = \infty$; for curve 2, $z = 1.3$ mm; and for curve 3, $z = 1.7$ mm.

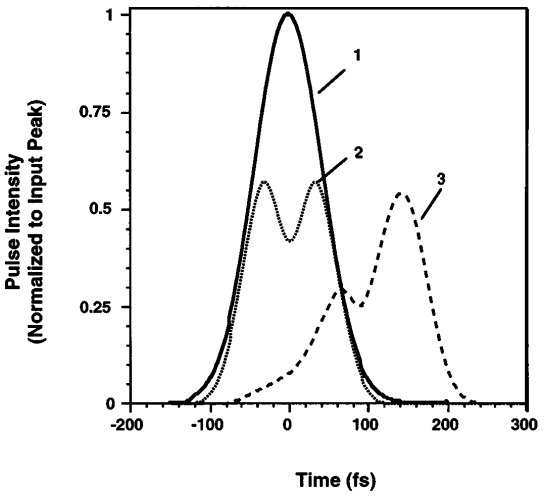
The results for a Gaussian pulse shape are shown in Fig. 3. The conversion efficiency for a pulse with an initial peak intensity of 500 GW/cm^2 in the absence of group-velocity walk-off (appropriate for a pulse of $\geq$1 ps) is shown in Fig. 3(a). The solid curve represents the calculated conversion for the pulse without the cubic nonlinearity. The dashed curve represents the conversion with the inclusion of the nonlinear phase for an initial value of $\Delta k = 0$, and the dotted curve represents the conversion curve when a phase mismatch is deliberately induced such that Eq. (14) is satisfied for the peak intensity. The total conversion efficiency can be increased from 78% to 93% if the crystal is simply retuned to compensate for the nonlinear phase at the peak of the pulse. The resulting second-harmonic pulse profiles are shown in Fig. 3(b) for the respective points along the conversion curves corresponding to the maximum in conversion efficiency for each case. The pulse with the initial value of $\Delta k = 0$ exhibits a distinctly flat-top profile, resulting from the reconversion that occurs at the more intense pulse peak earlier in the crystal. The pulse with the phase-mismatch compensation does not exhibit this reconversion at the pulse peak and has a resulting peak intensity that is nearly equal to that of the pulse converted in the absence of the cubic nonlinearity. This near equality illustrates the great advantage gained in generating the maximum peak power in the second-harmonic pulse by the introduction of an initial compensating phase mismatch.

It is also important to note that the point along the propagation path through the crystal corresponding to the greatest integrated-energy conversion efficiency for the uncompensated pulse conversion does not correspond to the point along the propagation of the greatest peak power of the second-harmonic pulse. The maximum total energy conversion actually occurs at a distance within the crystal that is reached after the peak of the second-harmonic pulse has begun to reconvert, and the pulse wings, where the nonlinear effects are lower, are still increasing in intensity. The dramatic increase in conversion efficiency is due to the fact that the peak of the pulse does not begin to reconvert until further into the crystal, permitting a more complete harmonic conversion in the pulse wings. This effect can be seen in Fig. 3(b), where the pulse without Δk compensation (dashed curve) shows a nearly complete conversion in the wings, with a flat profile in the center resulting from reconversion at the higher intensity. This effect permits some flexibility in the choice of the output-pulse shape. A shorter crystal yields high peak powers and shorter pulse widths, whereas a slightly longer crystal yields a greater conversion efficiency with a pulse that is somewhat flat at its peak.

The maximum achievable conversion is actually slightly increased for some instances in which group-velocity walk-off is important. Figure 4(a) shows the conversion curves for a 100-fs, 800-nm pulse with a peak intensity of 400 GW/cm^2. The case with no walk-off and no nonlinearity (curve 1) is compared with the nonstationary case without a nonlinearity (curve 2), the stationary case with the inclusion of a cubic nonlinearity (curve 3), and the case in which both effects are included (curve 4). The most important result is that, when the second-harmonic pulse walks off the fundamental pulse, the reconversion that we see for the stationary case is not as dramatic. Inclusion of the group-velocity walk-off term in Eq. 1(b) makes the solution more robust, lessening the large reconversion that results from the phase slip at the highest intensities that is caused by the cubic terms. The consequences of this large reconversion can be seen when the second-harmonic pulse shapes are considered at a point after the roll-over of the pulse conversion (at a distance of 2 mm into the crystal), as shown in Fig. 4(b). The walk-off of the second-harmonic pulse peak prevents the severe reconversion that is seen at the peak of the pulse for the stationary case. This pre-



(a)



(b)

Fig. 4. (a) Calculated conversion efficiency for a 100-fs Gaussian pulse with a peak intensity of 400 GW/cm^2 plotted as a function of the distance traversed in KDP. Curve 1 represents the case with no walk-off and no cubic nonlinearity. Curve 2 represents the case with a group-velocity walk-off ($\eta = 77$ fs/mm) but no nonlinear phase. Curve 3 represents the case with no walk-off but with the nonlinear phase included. Curve 4 illustrates the effects of both group-velocity walk-off and nonlinear phase. (b) Curve 1 represents the pulse envelope of the incident fundamental pulse for the calculation from (a). Curve 2 represents the second-harmonic pulse envelope after a distance travelled of 2 mm into the crystal when cubic nonlinearities are included but no group-velocity walk-off is present, and curve 3 represents the pulse shape when both effects are included.

vention occurs because the second-harmonic light moves away from the peak of the drive pulse, where the nonlinear phase mismatch becomes worse as the second harmonic grows. The two-peak structure evident in the pulse shape of curve 3 in Fig. 4(b) was observed under similar circumstances in an experimental study and in numerical modeling of short-pulse harmonic conversion with group-velocity walk-off and a geometric phase mismatch by Noordam *et al.*[18] The temporal structure observed in our calculation is from the same interference effect detailed in Ref. 18. However, the phase mismatch in our calculation is itself time dependent (through the intensity), arising from the cubic nonlinearity.

This behavior is, in general, only true when the group-velocity walk-off and cubic-nonlinearity terms are of roughly equal importance. If we define a dimensionless parameter,

$$\zeta = \frac{\beta |a_0^{(p)}|^2 \tau_p}{\eta}, \tag{17}$$

where τ_p is the laser pulse width, β is the maximum of the β_{ij} values, and $a_0^{(p)}$ is the value of the incident pulse peak field strength, then ζ is a measure of the relative importance of these two perturbations. If $\zeta \gg 1$, then the nonlinear phase mismatch dominates the conversion dynamics, and, if $\zeta \ll 1$, then the nonlinear phase is unimportant. For the calculation illustrated in Fig. 4(a), curve 4, $\zeta \approx 3.5$. Therefore both effects are roughly equally important in this case. Because ζ is proportional to $|a_0|^2 \tau_p$, it is therefore dependent on only the input-laser fluence. The consequences of this single dependence are that, for a given energy fluence, the relative importance of these two effects remains unchanged for any pulse width. For frequency doubling of 800-nm pulse in KDP, the fluence at which $\zeta = 1$ is approximately 10 mJ/cm^2. When ζ is large, the conversion dynamics are similar to those shown in Fig. 4(a), curve 3, and when ζ is small the conversion curve is similar to that of Fig. 4(a), curve 2. It follows, then, that it is desirable to keep the input fluence at or just below the level at which ζ is equal to 1 so as to avoid the reconversion that can accompany frequency doubling when the nonlinear mismatch dominates.

The conversion efficiency as a function of the peak incident intensity for an 800-nm, 100-fs pulse is shown for three crystal lengths, 1, 2, and 3 mm, in Fig. 5. The maximum conversion efficiencies are 70%, 75%, and 78%, respectively. The roll-over point in conversion efficiency occurs at a lower intensity for a longer crystal. This effect can be understood when one considers Eq. (12), which predicts that the maximum conversion efficiency achievable, under the assumption that a crystal of appropriate

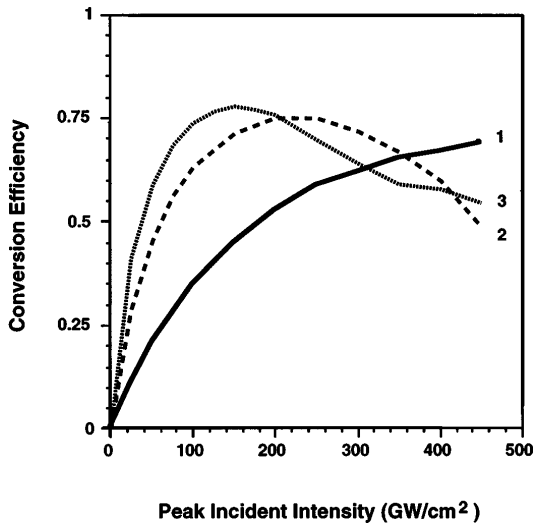


Fig. 5. Calculated conversion efficiency for an 800-nm, 100-fs pulse in KDP plotted as a function of the initial peak intensity for crystal lengths of 1 mm (curve 1), 2 mm (curve 2), and 3 mm (curve 3).

length has been chosen to reach that maximum, is a function of the incident intensity only. Consequently, longer crystals, which permit high conversion with lower drive intensity, have a higher maximum conversion. In other words, using a lower drive intensity implies that a higher total conversion efficiency is possible [from Eq. (12)], as long as a suitable crystal length is chosen to achieve this theoretical maximum conversion efficiency. This theory suggests that, in the regime of $\zeta \sim 1$, to achieve highest conversion efficiency it is advantageous to operate at lower drive intensities with longer crystals. It also emphasizes the importance of the correct choice of crystal length when the nonlinear phase mismatch becomes important.

5. CONCLUSION

In conclusion, we have investigated the effects of the nonlinear phase in the second-harmonic generation of ultrashort pulses and have shown that the phase mismatch that is due to n_2 of the doubling crystal can significantly lower the maximum possible conversion efficiency. By examining the second-harmonic equations we have derived an estimate for the net drop-off in conversion efficiency for a given input intensity and have derived the expression for compensating the nonlinear phase mismatch with an initial phase mismatch in a straightforward way.

We have also shown that, in the absence of any dispersion in the nonlinear refractive index of the doubling crystal, the nonlinear phase from the first and second harmonics cancel, and the third-order nonlinearity is unimportant. In general, however, these effects can be quite important for parameters that are common to the doubling of high-peak-power laser pulses. Numerical simulations have shown that the effects of the nonlinear phase coupled with group-velocity walk-off for ultrashort pulses can also alter the second-harmonic pulse shape quite significantly. We have also found that some dispersive walk-off can actually ameliorate the nonlinear phase mismatch effects.

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