Engineering competing nonlinearities

Ole Bang, Carl Balslev Clausen, and Peter L. Christiansen

Department of Mathematical Modelling, Technical University of Denmark, DK-2800 Lyngby, Denmark

Lluis Torner

Laboratory of Photonics, Department of Signal Theory and Communications, Universitat Politecnica de Catalunya, Gran Capitan UPC-D3, Barcelona ES-08034, Spain

Received June 11, 1999

Weak modulation of a quasi-phase-matching (QPM) grating opens possibilities for engineering both the average quadratic nonlinearity and the incoherent average cubic nonlinearity induced by QPM. The relative strength of the average quadratic and effective (intrinsic plus induced) cubic nonlinearity is studied for $LiNbO_3$. We show how the induced average cubic nonlinearity can be engineered to dominate the intrinsic material cubic nonlinearity and how doing so will allow the intensity at which the quadratic and cubic nonlinearities balance and thus compete to be decreased to a few gigawatts per square centimeter.

OCIS codes: 070.4340, 050.2770, 060.4080, 190.5530.

Quasi-phase-matching (QPM) by electric-field poling in ferroelectric materials, such as $LiNbO_3$, is promising for the engineering of photolithographic masks and thus of QPM gratings (see Ref. 1 for a review). Engineering a QPM grating by breaking its periodicity introduces additional degrees of freedom for light control. An appropriate design of the longitudinal grating structure allows for distortion-free temporal pulse compression,[2] soliton shaping,[3] broadband phase matching,[4] multiwavelength second-harmonic generation[5] (SHG), and an enhanced cascaded phase shift.[6] Transverse patterning can be used for beam tailoring,[7] broadband second-harmonic generation,[8] and soliton steering.[9]

At lowest order the effect of QPM is to eliminate the phase mismatch and to average the quadratic [or $\chi^{(2)}$] nonlinearity, resulting in an effective $\chi^{(2)}$ nonlinearity experienced by the slowly varying (on the scale of the coherence length) averaged field, which is reduced by a factor of $\pi/2$. At the next order, QPM induces cubic nonlinear self-phase-modulation (SPM) and cross-phase-modulation terms in the equations for the averaged field.[10] This induced nonlinearity is a result of incoherent or non-phase-matched coupling between modes[11] and is of a nature that is fundamentally different from the intrinsic material Kerr nonlinearity. Thus its effect can be significantly different from that of material $\chi^{(3)}$ nonlinearity on SHG[12] and $\chi^{(2)}$ solitons.[11] It has been shown how the induced $\chi^{(3)}$ nonlinearity affects the amplitude and phase modulation of cw waves[13] while still supporting solitons.[10] However, in the materials in which QPM has been demonstrated, the cubic corrections were small. Here we show how the average $\chi^{(2)}$ and $\chi^{(3)}$ nonlinearities can be engineered by modulation of the QPM grating, e.g., to make their effects equally strong and important.

We consider a linearly polarized electric field $\mathbf{E} = \hat{e}[E_1(z)\exp(ik_1 z - i\omega t) + E_2(z)\exp(ik_2 z - i2\omega t) + \text{c.c.}]/2$, propagating in a lossless QPM $\chi^{(2)}$ medium, for which the dynamic equations take the form

$$i\partial_z E_1 + G(z)\chi_1 E_1^* E_2 \exp(-i\Delta k z) = 0\,, \tag{1}$$

$$i\partial_z E_2 + G(z)\chi_2 E_1^2 \exp(i\Delta k z) = 0\,, \tag{2}$$

where $\partial_z = \mathrm{d}/\mathrm{d}z$, $\chi_j = \omega d_{\text{eff}}/(n_j c)$, $E_1(z)$ is the slowly varying envelope of the fundamental wave with frequency ω and wave vector k_1, and $E_2(z)$ is the second harmonic with wave vector k_2. The $\chi^{(2)}$ coefficient $d_{\text{eff}} = |\chi^{(2)}|/2$ is given in MKS units, and $\Delta k = 2k_1 - k_2$ is the wave-vector mismatch. The total intensity $I = \eta_0(n_1|E_1|^2 + n_2|E_2|^2)/2$ is conserved, where $n_j = n(j\omega)$ is the refractive index at frequency $j\omega$ ($j = 1, 2$) and $\eta_0 = \sqrt{\epsilon_0/\mu_0}$ is the specific admittance of vacuum.

The modulation of the $\chi^{(2)}$ susceptibility is described by the periodic grating function $G(z)$ with unit amplitude, and Fourier series $G(z) = \sum_n g_n \exp[inf(z)]$, where $g_n = 0$ for n even and $g_n = 2/(i\pi n)$ for n odd, are the coefficients of the unperturbed square grating. We take $f(z) = \kappa_0 z + \epsilon_2 \sin(\kappa_2 z)$ and consider weakly modulated QPM gratings with $L_2/\epsilon_2 \gg L_0$, where $L_2 = 2\pi/\kappa_2$ is the modulation period and $L_0 = \pi/\kappa_0$ is the unperturbed domain length. Such gratings correspond to those of a square grating with a slowly varying local domain length given by $L_d(z) \approx \pi/\partial_z f = \pi/[\kappa_0 + \epsilon_2\kappa_2 \cos(\kappa_2 z)]$, as illustrated in Fig. 1 for $\epsilon_2 = 1.2$ and $L_2 = 10L_0 = 40\ \mu$m.

We consider first-order QPM with slow modulation of the short domain length, i.e., $L_0 \sim L_c \ll L$ and $L_0 \ll L_2$, where L is the crystal length and $L_c = \pi/\Delta k$ is the coherence length. Then we expand E_j in a Fourier series in wave number κ_0, $E_1 = \sum_n w_n(z)\exp(in\kappa_0 z)$, and $E_2 = \sum_n v_n(z)\exp(in\kappa_0 z)$, where the coefficients are slowly varying on the L_0 scale, i.e., $|\partial_z w_n| \ll |\kappa_0 w_n|$ and $|\partial_z v_n| \ll |\kappa_0 v_n|$. Following the approach of Ref. 10, we then obtain the averaged equations for $\tilde{w} = w_0$ and $\tilde{v} = iv_0$:

$$i\partial_z \tilde{w} + \eta_1 D_+ \tilde{w}^* \tilde{v} \exp(-i\beta_0 z) + (\gamma_2|\tilde{w}|^2 - \gamma_1|\tilde{v}|^2)\tilde{w} = 0\,, \tag{3}$$

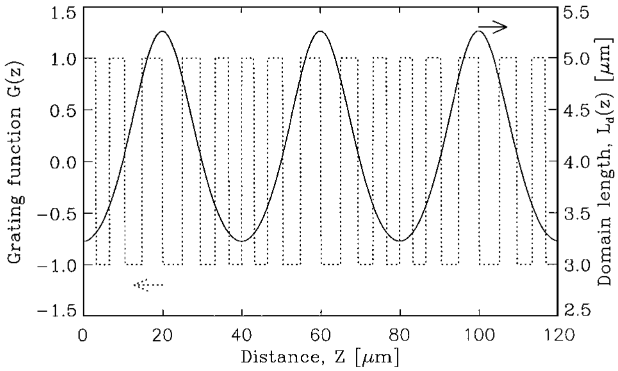

Fig. 1. Modulated QPM grating function $G(z)$ (left y axis, dotted line) with the corresponding local domain length $L_d(z)$ (right y axis, solid line) for $L_2 = 10L_0 = 40$ μm and $\epsilon_2 = 1.2$.

$$i\partial_z \tilde{v} + \eta_2 D_- \tilde{w}^2 \exp(i\beta_0 z) - 2\gamma_2 |\tilde{w}|^2 \tilde{v} = 0\,, \quad (4)$$

where $\beta_0 = \Delta k - \kappa_0 \ll \kappa_0$ is the small residual mismatch, $\eta_j = 2\chi_j/\pi$, and $\gamma_j = \chi_1\chi_j(1 - 8/\pi^2)/\kappa_0$. The difference between Eqs. (3) and (4) and the equations obtained in Ref. 10 is the periodic function $D_\pm = D_\pm(z) = \exp[\pm i\epsilon_2 \sin(\kappa_2 z)]$, which can be expanded in the Fourier series

$$D_\pm(z) = \sum_n d_{\pm n} \exp(in\kappa_2 z)\,, \qquad d_n = J_n(\epsilon_2)\,, \quad (5)$$

where $J_n(\epsilon_2)$ is the Bessel function of the first kind of order n. Thus $d_{-n} = (-1)^n d_n$.

Analytical treatment is possible in two cases: In the adiabatic limit when $D_\pm(z)$ varies much more slowly than $\tilde{w}(z)$ and $\tilde{v}(z)$, the field can be assumed to follow adiabatically the variation of $D_\pm(z)$.[2,3] Here we consider the opposite limit, i.e., that when the modulation period is short compared with the length of the crystal, $L_2 \ll L$. The spectrum of $G(z)$ for such a grating is decomposed into separate nonoverlapping blocks about the peaks of the unperturbed spectrum, as shown in Fig. 2.

We now expand $\tilde{w}(z)$ and $\tilde{v}(z)$ in a Fourier series in wave number κ_2, $\tilde{w} = \sum_n \tilde{w}_n(z)\exp(in\kappa_2 z)$ and $\tilde{v} = \sum_n \tilde{v}_n(z)\exp(in\kappa_2 z)$, where the coefficients vary slowly on the L_2 scale, $|\partial_z \tilde{w}_n| \ll \kappa_2 \tilde{w}_n$ and $|\partial_z \tilde{w}_n| \ll \kappa_2 \tilde{v}_n$. Again following the approach of Ref. 10, we obtain the equations for the average fields $w = \tilde{w}_0$ and $v = \tilde{v}_0$:

$$i\partial_z w + \eta_{1m} w^* v \exp(-i\beta_m z) + (\gamma_{2m}|w|^2 - \gamma_{1m}|v|^2)w = 0\,, \quad (6)$$

$$i\partial_z v + \eta_{2m} w^2 \exp(i\beta_m z) - 2\gamma_{2m}|w|^2 v = 0\,, \quad (7)$$

where $\beta_m = \beta_0 - m\kappa_2 = \Delta k - \kappa_0 - m\kappa_2 \ll \kappa_2$ is the effective mismatch for matching to the mth peak next to the κ_0 peak, as illustrated in Fig. 2, right. The nonlinearity coefficients are given by

$$\eta_{jm} = \chi_j[2J_m(\epsilon_2)/\pi]\,, \quad (8)$$

$$\gamma_{jm} = \chi_1\chi_j[(\pi^2 - 8)/\kappa_0 - 4S_m(\epsilon_2)/\kappa_2]/\pi^2\,, \quad (9)$$

where $S_m = -S_{-m} = \sum_{n\neq 0} J_{n+m}{}^2/n$. For $m = 0$ we obtain $S_0 = 0$. Thus $\gamma_{j0} = \gamma_j$ of the unperturbed grating, as it should. Using the recurrence and addition formulas for Bessel functions, we obtain $S_1 = -2J_0(\epsilon_2)J_1(\epsilon_2)/\epsilon_2$. Closed-form analytical expressions for S_m are progressively more difficult to obtain for higher orders, $m \geq 2$. The effective equations (6) and (7) for the averaged fields are easily extended to incorporate higher-order QPM and diffraction in the transverse x and (or) y direction.[10]

In Fig. 3 we show the normalized average nonlinearity coefficients η_{1m}/η_1 and γ_{2m}/γ_2 versus the modulation parameter ϵ_2 for different orders of phase matching and the same QPM grating as in Fig. 2. The right-hand ordinate axis gives the corresponding values for bulk $LiNbO_3$ of the average $\chi^{(2)}$ coefficient $d_{\text{eff}}^{\text{qpm}}$, defined as $\eta_{1m} = \omega d_{\text{eff}}^{\text{qpm}}/(n_1 c)$, and the induced average cubic SPM coefficient $\chi_{\text{qpm}}^{(3)}$, defined as $\chi_{\text{qpm}}^{(3)} = 4n_1\lambda_1\gamma_{2m}/(3\pi)$. We have used a fundamental wavelength of $\lambda_1 = 1.064$ μm, for which $d_{\text{eff}} = 30$ pm/V, $n_1 \simeq n_2 \simeq 2.2$, and the nonlinear refractive index is $n_{\text{ref}} \simeq 50 \times 10^{-14}$esu for $LiNbO_3$.[14] From Fig. 3 we see that by matching to the $m = 1$ peak we can increase the strength of the induced $\chi^{(3)}$ nonlinearity by a factor of 23 (to 44×10^3 pm^2/V^2 in $LiNbO_3$) by choosing a sufficiently weak modulation ($\epsilon_2 \ll 1$). In comparison, the material SPM nonlinearity in bulk $LiNbO_3$ is $\chi_{\text{spm}}^{(3)} = 3 \times 10^3$ pm^2/V^2, and thus the induced $\chi^{(3)}$ nonlinearity can actually be made dominant by the modulation. However, for $\epsilon_2 \ll 1$ the effective $\chi^{(2)}$ nonlinearity is reduced (averaged out) to nearly zero for $m \geq 1$. Choosing the correct modulations is thus a matter of optimization for the specific design purpose. For example, if the aim is efficient uniform multiwavelength SHG, $\epsilon_2 \simeq 1.7$ should be chosen to

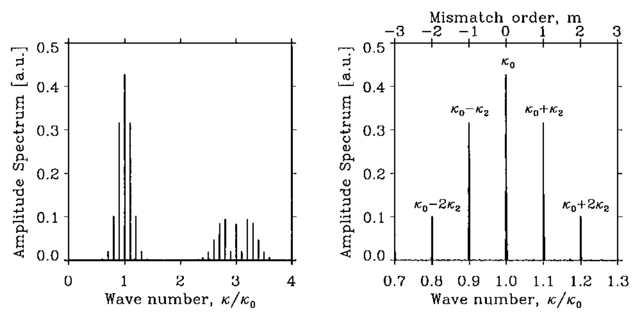

Fig. 2. Left, amplitude spectrum of $G(z)$ for $\epsilon_2 = 1.2$ and $L_2 = 20L_0 = 100$ μm. Right, block structure near $\kappa = \kappa_0$, showing order m of effective mismatch β_m.

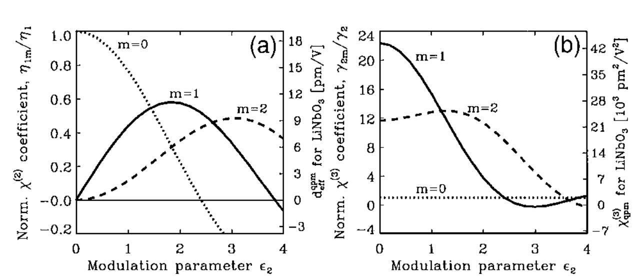

Fig. 3. (a) Normalized average quadratic nonlinearity η_{1m}/η_1 and (b) induced average cubic nonlinearity γ_{2m}/γ_2 versus modulation parameter ϵ_2 for $L_2 = 20L_0 = 100$ μm. The right-hand ordinate axis shows the actual strength for bulk $LiNbO_3$.

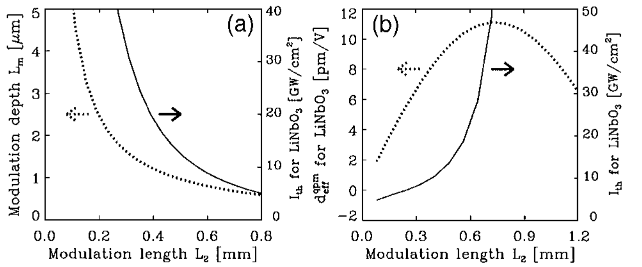

Fig. 4. (a) Modulation depth L_m and threshold intensity I_{th} versus L_2 for bulk $\mathrm{LiNbO_3}$ with $\epsilon_2 = 1.2$ and $L_0 = 10\ \mu\mathrm{m}$. (b) Effective averaged $\chi^{(2)}$ coefficient $d_{\mathrm{eff}}^{\mathrm{qpm}}$ and I_{th} versus L_2 for bulk $\mathrm{LiNbO_3}$ with $L_m = 1\ \mu\mathrm{m}$ and $L_0 = 10\ \mu\mathrm{m}$.

have a constant value of the effective $\chi^{(2)}$ nonlinearity for all three peaks $m = 0, 1, 2$.

Introducing normalized dimensionless coordinates $w(z) = \alpha\phi_1(\zeta)$ and $v(z) = \alpha\phi_2(\zeta)$ into Eqs. (6) and (7), where $\alpha = [2I/(n_1\eta_0)]^{1/2}$ and $\zeta = z/L$, we measure the relative strength of the average $\chi^{(2)}$ and $\chi^{(3)}$ nonlinearities as the ratio of the coefficients in front of the cubic SPM term for the fundamental wave and the $\chi^{(2)}$ term. Equating this intensity-dependent ratio to unity gives the threshold intensity $I_{\mathrm{th}} = (n_1\eta_0/2)[\eta_{1m}/(\gamma_{2m} + \gamma_{\mathrm{SPM}})]^2$, where we have included the intrinsic cubic SPM nonlinearity $\gamma_{\mathrm{SPM}} = 3\pi\chi_{\mathrm{SPM}}^{(3)}/(4n_1\lambda_1)$. For intensities above (below) I_{th} the $\chi^{(3)}$ ($\chi^{(2)}$) nonlinearity is dominant on average.

In Fig. 4 we show how the threshold intensity and effective $\chi^{(2)}$ nonlinearity are changed by the modulation given by period L_2 and depth $L_m = \pi/(\kappa_0 - \epsilon_2\kappa_2) - \pi/(\kappa_0 + \epsilon_2\kappa_2)$. We chose the unperturbed domain length $L_0 = 10\ \mu\mathrm{m}$ (because I_{th} decreases with L_0), corresponding to the coherence length at $\lambda_1 = 1.5\ \mu\mathrm{m}$ in $\mathrm{LiNbO_3}$. From Fig. 4(a) with fixed $\epsilon_2 = 1.2$ (i.e., fixed $d_{\mathrm{eff}}^{\mathrm{qpm}} = 12$ pm/V) we see that for reasonable modulation depths the threshold intensity can be decreased to a few gigawatts per square centimeter. From Fig. 4(b) with fixed $L_m = 1\ \mu\mathrm{m}$ we see that the threshold intensity can be further decreased but at the expense of a significant reduction in the effective $\chi^{(2)}$ nonlinearity. The singularity at $L_2 = 1$ mm (and at $L_2 = 1.5$ mm) is due to the induced cubic nonlinearity's becoming equally as strong as the material nonlinearity, but negative. Between $L_2 = 1$ mm and 1.5 mm the effective (material plus induced) cubic SPM nonlinearity for the fundamental wave is thus defocusing.

In conclusion, we have shown that weak modulation of a QPM grating permits engineering of average $\chi^{(2)}$ and induced average $\chi^{(3)}$ nonlinearities. We have shown how the induced average $\chi^{(3)}$ nonlinearity can dominate the intrinsic material nonlinearity, thereby decreasing the intensity at which the $\chi^{(2)}$ and $\chi^{(3)}$ effects balance to a few gigawatts per square centimeter. This result opens a range of new possibilities that arise when $\chi^{(2)}$ and $\chi^{(3)}$ nonlinearities compete on an equal footing. Such possibilities include attainment of engineered bandwidth for parametric wave mixing and frequency generation, cascading phase shifts, and solitons. In particular, the averaged model can have a self-defocusing cubic nonlinearity, which can dominate the self-focusing nonlinearity that is intrinsic to typical $\chi^{(2)}$ materials such as $\mathrm{LiNbO_3}$ and might therefore support stable dark vortex solitons.[15]

This research is part of project 9800400 supported by the Danish Technical Research Council. Balslev Clausen acknowledges support from the Danish National Research Council under grant 9600852. O. Bang's e-mail address is ob@imm.dtu.dk.

References

1. M. M. Fejer, in *Beam Shaping and Control with Nonlinear Optics*, F. Kajzar and R. Reinisch, eds. (Plenum, New York, 1998), p. 375.
2. M. A. Arbore, O. Marco, and M. M. Fejer, Opt. Lett. **22,** 865 (1997). M. A. Arbore, A. Galvanauskas, D. Harter, M. H. Chou, and M. M. Fejer, Opt. Lett. **22,** 1341 (1997).
3. L. Torner, C. Balslev Clausen, and M. M. Fejer, Opt. Lett. **23,** 903 (1998).
4. K. Mizuuchi and K. Yamamoto, Opt. Lett. **23,** 1880 (1998).
5. S. Zhu, Y. Zhu, Y. Qin, H. Wang, C. Ge, and N. Ming, Phys. Rev. Lett. **78,** 2752 (1997).
6. M. Cha, Opt. Lett. **23,** 250 (1998).
7. G. Imeshev, M. Proctor, and M. M. Fejer, Opt. Lett. **23,** 673 (1998).
8. P. E. Powers, T. J. Kulp, and S. E. Bisson, Opt. Lett. **23,** 159 (1998).
9. C. Balslev Clausen and L. Torner, Phys. Rev. Lett. **81,** 790 (1998); Opt. Lett. **24,** 7 (1999).
10. C. Balslev Clausen, O. Bang, and Y. S. Kivshar, Phys. Rev. Lett. **78,** 4749 (1997).
11. O. Bang, Y. S. Kivshar, A. V. Buryak, A. De Rossi, and S. Trillo, Phys. Rev. E **58,** 5057 (1998).
12. Y. Zhao, G. Town, and M. Sceats, Opt. Commun. **115,** 129 (1995); R. Maleck Rassoul, A. Ivanov, E. Freysz, A. Ducasse, and F. Hache, Opt. Lett. **22,** 268 (1998).
13. A. Kobyakov, F. Lederer, O. Bang, and Y. S. Kivshar, Opt. Lett. **23,** 506 (1998).
14. R. DeSalvo, A. A. Said, D. J. Hagan, E. W. Van Stryland, and M. Sheik-Bahae, IEEE J. Quantum Electron. **32,** 1324 (1996).
15. T. J. Alexander, A. V. Buryak, and Y. S. Kivshar, Opt. Lett. **23,** 670 (1998).