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Effective nonlinearity and multi-wavelength second-harmonic generation in modulated quasi-phase-matching gratings

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Abstract: Quasi-phase-matching gratings induces Kerr effects in quadratic nonlinear materials. We show analytically and confirm numerically how modulating the grating changes the effective quadratic and cubic nonlinearities and allows for multi-wavelength second-harmonic generation.

Quasi-phase-matching (QPM) by electric-field poling in ferroelectric materials, such as LiNbO3, is promising due to the possibilities of engineering the photolithographic mask, and thus the QPM grating, without also generating a linear grating. A proper design of the longitudinal grating structure allows for distortion free temporal pulse compression [1], soliton shaping [2], broad-band phase matching [3], multi-wavelength second-harmonic generation (SHG) [4], and an enhanced cascaded phase shift [5]. Transverse patterning can be used for beam-tailoring [6], broad-band SHG [7] and soliton steering [8].

At lowest order the effect of QPM is to eliminate the phase mismatch and 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- and cross-phase modulation terms in the equations for the averaged field [9]. This induced nonlinearity is a result of non phase-matched coupling between the average field and higher order modes [10] and of a fundamentally different nature than the intrinsic material Kerr nonlinearity.

The first order averaged equations with the induced cubic terms provide an elegant tool to study the effect of QPM. It has been shown both analytically and numerically that the induced $\chi^{(3)}$ nonlinearity significantly affects the amplitude and phase modulation of cw waves [11], as well as the properties of solitons [9]. However, in conventional materials with single period QPM, such as LiNbO3, the cubic corrections to the effective quadratic nonlinearity are typically small. Here we show analytically and verify numerically how the average $\chi^{(2)}$ and $\chi^{(3)}$ nonlinearities can be engineered by modulating the QPM grating, such as, e.g., to make their effects equally strong and important.

We consider a linearly polarized electric field $\vec{E} = \vec{E}(z) = E_1(z) \exp(i k_1 z - i \omega t) + E_2(z) \exp(i k_2 z - i 2 \omega t) + c.c.$, propagating in a lossless QPM $\chi^{(2)}$ medium under conditions for type I SHG. Then the dynamical equations take the form

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

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

where $\partial_z = d/dz$, $\chi_1 = \omega d_{eff}/(n_1 c)$, $E_1(z)$ is the slowly varying envelope of the fundamental wave (FW) with frequency $\omega$, refractive index $n_1$, and wavevector $k_1$, and $E_2(z)$ is the second harmonic (SH) with refractive index $n_2$ and wavevector $k_2$. The $\chi^{(2)}$ coefficient $d_{eff} = |\chi^{(2)}|/2$ is given in MKS units, and $\Delta k = 2k_1 - k_2$ is the wavevector mismatch. The modulation of the $\chi^{(2)}$ susceptibility is described by the periodic grating function $G(z)$ with unit amplitude and Fourier series $G(z) = \sum n g_n \exp[imf(z)]$, 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 $f(z) = \kappa_0 \xi + \xi \sin \kappa_0 \xi$ and consider weakly modulated first order QPM gratings with $L_2/e_2 \gg L_0$, where $L_0 = 2\pi/\kappa_0$ is the modulation period and $L_0 = \pi/\kappa_0$ is the unperturbed domain length. This corresponds to a square grating with a slowly varying local domain length given by $L_2(z) \approx \pi/\partial_z f = \pi/[\kappa_0 + e_2 \kappa_2 \cos(\kappa_0 z)]$, as illustrated in Fig. 1(left).

Following the approach of [9] we obtain the equation for the average fields on the shortest scale $L_0 \ll L_2$. 

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with induced cubic nonlinear terms and a modulated effective $\chi^{(3)}$ nonlinearity that depends periodically on $z$ (its Fourier coefficients are Bessel functions of the first kind, $J_n$). Assuming that the modulation length is still much shorter than the crystal length, $L_2 \ll L$, the spectrum of $G(z)$ has the block structure shown in Fig. 1(center), which allows us to do a second independent averaging on the $L_2$-scale. Thus we obtain the final equations for the average fields $w=(E_1)$ and $v=(E_2)$, which includes the induced cubic terms:

$$i\partial_z u + \eta_1 w u e^{-i\beta_m z} + \gamma_1 |w|^2 u = 0, \quad i\partial_z v + \eta_2 w^2 e^{i\beta_m z} - 2\gamma_2 |w|^2 v = 0,$$

(2)

where $\beta_m = \Delta k - \kappa_0 - m\kappa_2 \ll \kappa_2$ is the effective mismatch for matching to the $m$th peak next to the $\kappa_0$ peak, as illustrated in Fig. 1(right). The nonlinearity coefficients are given by

$$\eta_{1m} = \chi_3 [2 J_m(\epsilon_2)]/\pi, \quad \gamma_{1m} = \chi_1 \chi_3 [(\pi^2 - 8)/\kappa_0 - 4S_m(\epsilon_2)/\kappa_2]/\pi^2,$$

(3)

where $S_m = -S_{-m} = \sum_{n \neq m} J_n^2 / n$. For $m=0$ we obtain $S_0=0$. Thus $\gamma_1=\gamma_2$ of the unperturbed grating, as it should be. 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 Eqs. (2) for the averaged fields are easily extended to incorporate higher order QPM and diffraction in the transverse $x$ and/or $y$ direction [9].

In Fig. 2 we show the normalized average nonlinearity coefficients $\eta_{1m}/\eta_1$ and $\gamma_{2m}/\gamma_2$ versus the modulation parameter $\epsilon_2$ for different orders of phase matching, and the same QPM grating as in Fig. 1. The right ordinate axis gives the corresponding values for bulk LiNbO$_3$ of the average $\chi^{(3)}$ coefficient $\chi_{\text{eff}}^{(3)}$, defined as $\eta_{1m} = \omega \chi_{\text{eff}}^{(3)}/(n_1 \epsilon_2)$, and the induced average cubic SPM coefficient $\chi_{\text{ind}}^{(3)}$, defined as $\chi_{\text{ind}}^{(3)} = 4n_1 \lambda_1 \gamma_2 m/(3\pi)$. We have used a fundamental wavelength of $\lambda_1=1.064\mu m$, for which $d_{\text{eff}}=30\mu m/V$, $n_1 \approx n_2 \approx 2.2$, and the nonlinear refractive index is $n_{\text{eff}}=50 \times 10^{-14} \epsilon_0 u$ for LiNbO$_3$ [13]. From Fig. 2 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 $44 \times 10^3$
pm²/V² in LiNbO₃) by choosing a sufficiently weak modulation (ε₂<<1). In comparison, the material SPM nonlinearity in bulk LiNbO₃ is χ⁽⁽⁽⁾⁾(3)non ≈ 3·10⁻¹³ pm²/V², and thus the induced χ⁽⁽⁽⁾⁾ nonlinearity can actually be made dominant by the modulation. However, for ε₂<<1 the effective χ⁽⁽⁽⁾⁾ nonlinearity is reduced (averaged out) to nearly zero for m≥1. Choosing the right modulation is thus a matter of optimization for the specific design purpose. For example, if the aim is efficient uniform multi-wavelength SHG, then ε₂≈1.7 should be chosen to have a constant value of the effective χ⁽⁽⁽⁾⁾ nonlinearity for all three m=0,1,2 peaks.

In conclusion we have shown analytically that modulation of a QPM grating allows to engineer the effective χ⁽⁽⁽⁾⁾ and induced average χ⁽⁽⁽⁾⁾ nonlinearities. We have shown how the induced average χ⁽⁽⁽⁾⁾ nonlinearity can even be made to dominate the intrinsic material one, thereby decreasing the intensity at which the χ⁽⁽⁽⁾⁾ and χ⁽⁽⁽⁾⁾ effects balance.

A whole new range of possibilities arise when χ⁽⁽⁽⁾⁾ and χ⁽⁽⁽⁾⁾ nonlinearities compete on equal footing. Such possibilities include, e.g., engineered bandwidth for parametric wave mixing and frequency generation, cascading phase-shifts, or solitons. In particular, the averaged model can have a self-defocusing cubic nonlinearity, which can dominate the self-focusing one intrinsic to typical χ⁽⁽⁽⁾⁾ materials like LiNbO₃, and might therefore support stable dark vortex solitons [14]. This will be the topic of a separate contribution.

In this contribution we will further present numerical verification of the average two-period QPM model (2) and the multi-wavelength SHG spectrum shown in Fig. 1. Such multi-channel wavelength conversion is of considerable technical interest and was recently investigated and confirmed experimentally in LiNbO₃ waveguides [15]. The average model provides an elegant tool to analytically calculate the spectrum and the bandwidth for SHG at the different peaks. We will also present these calculations and confirm them numerically.

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