A novel type of quasi-phasematching for the second harmonic generation

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Abstract. We propose a novel type of quasi-phasematching for the second harmonic generation in periodically-poled nonlinear crystals. In contrast to the conventional quasi-phasematching where one (or few) quasi-wavevector(s) of periodical poling compensate for the momentum mismatch between a pair of the fundamental photons and the SHG one, with the proposed mechanism the momentum mismatch between several pairs of fundamental and SHG photons is compensated with one quasi-wavevector of periodical poling.

1. Introduction
Generation of the second harmonic (SHG) of infrared (IR) light offers one of the most preferred ways for realization of compact visible laser sources with a number of cutting-edge applications in microscopy, spectroscopy, biophotonics and photomedicine. Availability of compact, efficient and cost-effective IR laser diodes in the near-IR range of 0.8 - 1.5 μm supports this trend both from the market and technical sides [1]. However, the efficient SHG is possible only with simultaneous photon energy $E_{\lambda} = 2E_{2\lambda}$ and momentum conservation $k_{\lambda} = 2k_{2\lambda}$ (here and below the subscript $2\lambda$ denotes the fundamental and $\lambda$ SHG wavelength). The last requirement of the photon momentum conservation (or “phase-matching”) is difficult to achieve due to dispersion of the refractive index in the nonlinear crystal. Without phase-matching, the generated second harmonic grows and decays as the fundamental and SHG waves go in and out of phase over each coherence length $L_c = \frac{\lambda}{|n_\lambda - n_{2\lambda}|}$, where $n_\lambda$ and $n_{2\lambda}$ are the refractive indexes at SHG and fundamental wavelength correspondingly [2].

Since its first introduction in 1960s [2,3], the preferred approach for the phase-matching between interacting harmonic and fundamental waves is the periodical poling (or “quasi-phase-matching” - QPM) of the ferroelectric nonlinear crystals. This is normally achieved by periodically reversing the crystals polarization under large electric field. The proper phase relationship between the propagating waves is maintained with the poling period being double the coherence length $\Lambda = 2L_c$. Under this condition, the SHG efficiency is maximized because of momentum conservation by the quasi-wave-vector of the periodical poling $k_{\lambda} = \frac{2\pi}{\Lambda} = \Delta k = k_{2\lambda} - 2k_{2\lambda}$ as shown schematically in figures 1(a) and (b). Tuning of the conversion wavelength is possible by introduction of multiple gratings or variable poling period [4], Fibonacci or Fourier-constructed quasi-periodical poling [5,6], shifting dispersion in the nonlinear crystal with temperature [7] or by introduction of the multimode waveguide [8,9].
As shown by Fejer at al. [10], it is also possible to achieve QPM in the case of the high-order periodical poling that dramatically extends the spectral tuning range of SHG. With the high-order poling, the period is a multiple of the doubled coherence length $\Lambda=2L_c$, where the natural number $m=2, 3, 4...$ is the order of poling. With the $m^{th}$ order poling, in contrast to the first-order poling, it is necessary to ‘utilize’ not one, but $m$ quasi-wave-vectors of the periodical poling for compensation of the momentum mismatch $mk_\Lambda=\Delta k=k_\lambda-2k_{2\lambda}$, as shown schematically in figures 1(c) and (d). Generally speaking, this process is very similar to the high-order diffraction with the conventional diffraction grating.

2. Fractional-order periodical poling for the second harmonic generation

In this paper, we propose a fractional order of poling period of nonlinear crystal. In contrast to the higher-order poling with $m$ quasi-wave-vectors of the periodical poling compensating the momentum mismatch between a pair of fundamental photons and SHG one, fractional order poling enables momentum compensation for $p$ pairs of fundamental and $p$ SHG photons with one quasi-wave-vector of the periodical poling: $mk_\Lambda=\Delta k=p(k_\lambda-2k_{2\lambda})$. The effective coherence length in this case of the multi-photon momentum compensation reduces $p$-fold $L_c=\pi/\Delta k$ leading to the corresponding decrease of the fractional-order poling period $\Lambda=2\pi/\Delta k=2\pi/p(k_\lambda-2k_{2\lambda})^{-1}$ being a ‘fraction’ of the first-order one. Also, it is possible to generalize this approach by merging both the high- and the fractional-order poling: $mk_\Lambda=\Delta k=p(k_\lambda-2k_{2\lambda})$. In other words, momentum mismatch between $p$ pairs of the fundamental and $p$ SHG photons can be compensated with $m$ quasi-wave-vectors of the $m/p$-order periodical poling as shown schematically in figures 1(e) and (f).

Now, it is important to understand the law for the spatial evolution of the SHG intensity with the fractional-order poling. With the conventional and higher-order poling, the rate of
growth of the slowly varying SHG field amplitude in the one-dimensional periodically-poled nonlinear crystal is proportional to the square of the amplitude of the fundamental wave, spatially-modulated nonlinear coefficient and the phase term [10]:

\[
\frac{dE_2}{dz} \sim E_1^2 d(z) \exp(-i\Delta k z)
\]  

(1)

**Figure 2** Spatial evolution of the SHG field amplitude and intensity for the first-order poling (a,b), third-order poling (c,d) and half-order poling (e,f).
This simple expression is valid under assumption of low conversion efficiency, loose focusing and absence of losses at fundamental and SHG wavelengths. Integration of (1) yields the spatial evolution of the SHG field amplitude:

\[ E(z) = E_0 \int_0^z d(x) \exp(-i\Delta k x) dx \]  

(2)

where \( x \) is the integration variable and \( z \) is the coordinate along the direction of propagation of the fundamental and SHG waves in the periodically-poled nonlinear crystal. In the case of an ‘ideal’ first-order poling, with the period of poling corresponding to the momentum mismatch \( \Lambda = 2\pi/\Delta k = 2\pi/(k_0 - 2k_2) \) and the module of material nonlinearity always having the maximal value \( d(z) = \pm d_{\text{eff}} \) (corresponding to the meander-like modulation with negligibly thin inverted domain interfaces) integration of (2) yields the classical spatial evolution of the SHG field amplitude represented in figure 2(a). Figure 2(b) shows the spatial evolution of the SHG intensity being a square of the module of the field amplitude.

Taking the third-order poling as an example of the higher-order poling, one should increase the period of poling 3-fold: \( \Lambda = 6\pi/\Delta k \) and keep the momentum mismatch value \( \Delta k = k_0 - 2k_2 \). With meander-like modulation and thin domain interfaces, integration of (2) and its square provides one with the spatial evolution of the SHG field amplitude and intensity as shown in figures 2(c) and 2(d). Comparing figures 2(a,b) and 2(c,d) correspondingly it is easy to note significant decrease of the conversion efficiency with the third-order poling due to uncompensated oscillations of the SHG field in the first two thirds of each half-period of poling.

In contrast to a higher-order poling, a fractional poling period becomes a ‘fraction’ of the first-order one \( \Lambda = 6\pi/\Delta k \) not due to ‘artificial’ denomination by the fraction value \( p \) but because of increase of the effective momentum mismatch \( \Delta k = p(k_0 - 2k_2) \) which in turn is due to compensation of momentum mismatch between \( p \) pairs of fundamental and \( p \) SHG photons with one quasi-wave-vector of the periodical poling as stated in the first paragraph of this section. Figures 2(e) and (f) show the spatial evolution of the SHG field amplitude and intensity for the simplest case of the \( 1/2 \) period poling, with periodically-poled grating quasi-wave-vector compensating for the momentum mismatch of two pairs of fundamental and two SHG photons. One can note striking similarity between the figures 2(e,f) and 2(a,b) which differ only with the period of ‘ripples’ caused by the SHG and fundamental waves coming in and out of phase. This should be attributed to an ‘ideal’ compensation of the multi-photon momentum mismatch with the shortened period of poling.

3. Conclusion
In summary, we have discussed a novel mechanism of quasi-phasesmatching for the second harmonic generation in periodically-poled nonlinear crystals. The proposed mechanism is based on compensation of the momentum mismatch between several pairs of fundamental and SHG photons with one quasi-wavevector of periodical poling. This is the ‘opposite’ to the conventional quasi-phasesmatching where momentum mismatch between two fundamental and one SHG photon is compensated with the one (or few) quasi-wavevector(s) of periodical poling.

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