

Phase-matched nonlinear diffraction

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We report on a new (to our knowledge) configuration incorporating both birefringence and quasi-phase-matching, enabling efficient phase-matched nonlinear diffraction in one-dimensional periodically poled nonlinear crystals. We demonstrate the method experimentally, showing an efficient nonlinear diffraction to the first few orders in two types of crystals, MgO doped congruent lithium niobate and congruent lithium niobate, and characterize its efficiency dependence on the fundamental power, the propagation angle, and the crystal temperature. This configuration can increase efficiencies observed in nonlinear diffraction experiments, enables ferroelectric domain characterization by nonlinear microscopy, and can be used to determine the duty cycles of periodically poled nonlinear crystals. © 2011 Optical Society of America

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It is well known that when a light beam passes through a periodic structure, it will diffract in several well defined angles. An interesting case occurs when only the second order nonlinear coefficient is periodically modulated, thereby leading to nonlinear diffraction (NLD): the beam at the fundamental frequency (FF) will remain in its original direction, but its second harmonic (SH) will diffract in specific angles. Whereas the first demonstration of NLD was made by Freund back in 1968 [1], there was a renewed interest in this effect and its applications [2–6] in recent years, due to the wide availability of periodically poled ferroelectric crystals. In the most common scheme used to realize NLD, the FF propagates along the optical axis (i.e., the crystallographic z axis) of a periodically poled crystal. There are two options for partially satisfying the momentum conservation law in such a scheme, the Raman–Nath [4] and the Cerenkov [3] phase-matching conditions. In the Raman–Nath case, also referred to as transverse quasi-phase-matching (QPM), the phase-matching condition for the m th diffraction order is defined by $\sin \alpha_m = mG_0/k_2$, where α_m is the diffraction angle, G_0 is the grating vector, and k_2 is the wave vector of the SH beam. In the Cerenkov case, also referred to as longitudinal QPM, the phase-matching condition is defined by $k_2 \cos \theta = 2k_1$, where θ is the angle of emission and k_1 and k_2 are the wave vectors of the FF and SH waves, respectively. The disadvantage of both schemes is that the full vectorial phase-matching is not fulfilled, and hence the resulting conversion efficiency is quite small in comparison with that achieved in the case of collinear phase-matching.

It has been recently shown that if both of the above conditions are fulfilled simultaneously, as in the case of nonlinear Bragg diffraction [1], the respective diffraction order is much more intense than the neighboring SH diffraction orders [4]. In the configurations examined so far, in order to satisfy both conditions for the first diffraction order, the poling period of the crystal had to be very small. For example, when working with periodically poled lithium niobate with an FF wavelength of $1.0645 \mu\text{m}$, the desired period at 100°C for an o–oo interaction is $0.8 \mu\text{m}$ [7]. Because of this limitation, experimental results have been available only for high diffraction orders, for example, $m = 13$ [2] or $m = 18$

[8]. An additional limitation of an experimental setup based on light propagating along the optical axis is short interaction length, typically around 0.5 mm [2,4]. These are the main reasons for the low conversion efficiencies reported so far. A different configuration was proposed recently in which light propagates perpendicular to the optical axis [5]. Such a scheme allows for a longer interaction length, but the reported result indicated that the conversion efficiency is still small, since only the transverse phase-matching condition was fulfilled.

In this Letter we study a new configuration for both transverse and longitudinal phase-matched NLD for the first diffraction order. Moreover, the interaction length can be much longer than 0.5 mm . This configuration combines the natural birefringence of the crystal with QPM, resulting in poling periods that can be easily fabricated. In this configuration, the ordinary polarized FF beam propagates perpendicular to the optical axis (along the y axis) and generates an extraordinary polarized SH beam. For example, when working with MgO doped congruent lithium niobate (MgO:CLN), with an ordinary polarized FF input beam at a wavelength of $1.0645 \mu\text{m}$, the required period at a crystal temperature of 60°C is $7 \mu\text{m}$ [9].

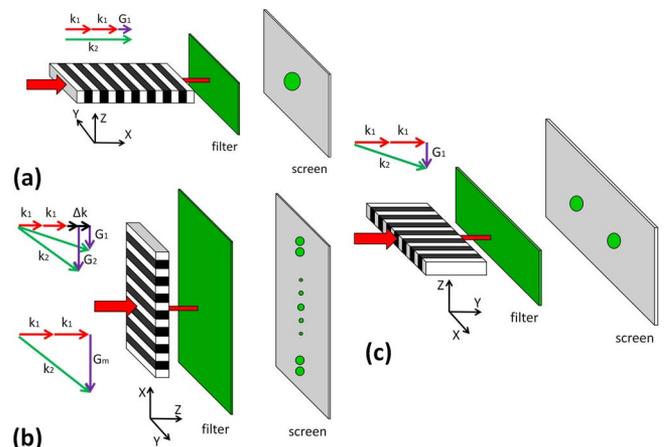


Fig. 1. (Color online) Phase-matching configurations and their corresponding wave vector diagrams: (a) collinear phase-matching, (b) Raman–Nath and Bragg NLD with the FF propagating along the optical axis, (c) suggested NLD scheme with the FF propagating perpendicular to the optical axis.

Table 1. Comparison between Predicted and Measured Diffraction Order Parameters in the MgO:CLN and CLN Crystals

Crystal	Diffraction Order	Prediction		Measurement	
		T [°C]	External Diffraction Angle [mrad]	T [°C]	External Diffraction Angle [mrad]
MgO:CLN	1	54.3	27.3	99	28.8
	3	61.2	82.1	115	84.5
	5	74.7	137	145	136.4
CLN	2	64.6	135.2	57	134.8
	3	145.8	203.5	125	200.7

Figure 1 summarizes the phase-matching schemes in one-dimensional periodically poled crystals discussed in this Letter: 1(a) shows standard collinear phase-matching with light propagating along the x axis, 1(b) shows Raman–Nath and Bragg NLD with light propagating along the optical axis (the z axis), and 1(c) shows the configuration proposed in this Letter satisfying the phase-matching condition for the first diffraction order with light propagating along the y axis.

In our experiment we examined two periodically poled crystals, MgO:CLN and congruent lithium niobate (CLN). The crystals were set in the schematic arrangement shown in Fig. 1(c). The length of the MgO:CLN crystal along the propagation direction was 2.4 mm, and the poling period was $19.48\ \mu\text{m}$. The length of the CLN crystal along the propagation direction was 4 mm, and the poling period was $7.9\ \mu\text{m}$. The FF source was a Nd:YAG laser producing 10 ns pulses at a 2 kHz repetition rate at a wavelength of $1.0645\ \mu\text{m}$. A half-wave plate was used to set the FF to be ordinary polarized. The laser beam was focused with a lens, creating a waist radius of approximately $50\ \mu\text{m}$ at the center of the crystal.

We have clearly observed the odd diffraction orders $m = 1, 3, 5$ in the MgO:CLN crystal at different crystal temperatures, as indicated in Table 1. For the CLN crystal, the first diffraction order could not be observed, since its calculated phase-matching temperature of 4.8°C was below the available operating temperatures of the crystal mount, but we have observed the next two diffraction orders, $m = 2, 3$. Table 1 summarizes the predicted and experimental results for the diffraction orders in both crystals. The interaction measured in both crystals was e-oo. For both crystals, the measured and calculated external diffraction angles are in good agreement. The measured temperatures deviate from prediction by 45° up to 70° in the different diffraction orders of MgO:CLN and by a smaller deviation of 7.5° up to 20° degrees in the case of CLN. We believe that the shift in temperature, in particular for MgO:CLN, might be due to possible inaccuracies in the thermal dependent part of the Sellmeier equation [9].

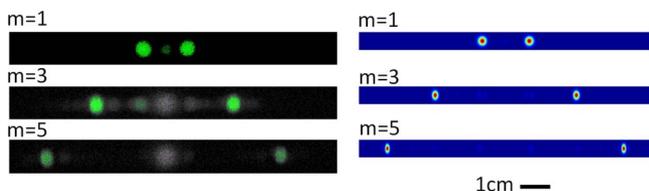


Fig. 2. (Color online) Comparison between measured (left) and simulated (right) diffraction patterns on a screen 29.5 cm away from the MgO:CLN crystal.

Measuring the diffraction efficiency of several diffraction orders enables one to determine the duty cycle of the pattern [10]. The intensity of each diffraction order is proportional to the corresponding square value of the Fourier coefficient, $[\sin(m\pi D)/mD]^2$, where m is the diffraction order and D is the duty cycle of the crystal. For MgO:CLN the experimentally derived ratio of these coefficients for the three odd orders of $1:0.088:0.024$ can be fitted well with a duty cycle of 55%. In a similar way, for CLN the experimentally derived ratio of the coefficients for the second and third diffraction orders ($1:0.66$) can be fitted with a duty cycle of 58%.

Figure 2 shows a comparison between the experimentally obtained and simulated NLD patterns, as seen on a screen placed 29.5 cm away from the crystal, for the $m = 1, 3, 5$ diffraction orders in MgO:CLN. Numerical simulations were performed based on the split-step Fourier method, with physical parameters identical to the ones in the experiment and assuming the following value for the element of the quadratic susceptibility tensor: $d_{31} = 4.4\ \text{pm/V}$ [11].

A detailed comparison between experimental and predicted results for the first diffraction order in the MgO:CLN crystal is presented in Fig. 3. We first checked the angular dependence of the doubling efficiency. From a geometrical point of view the phase-mismatch

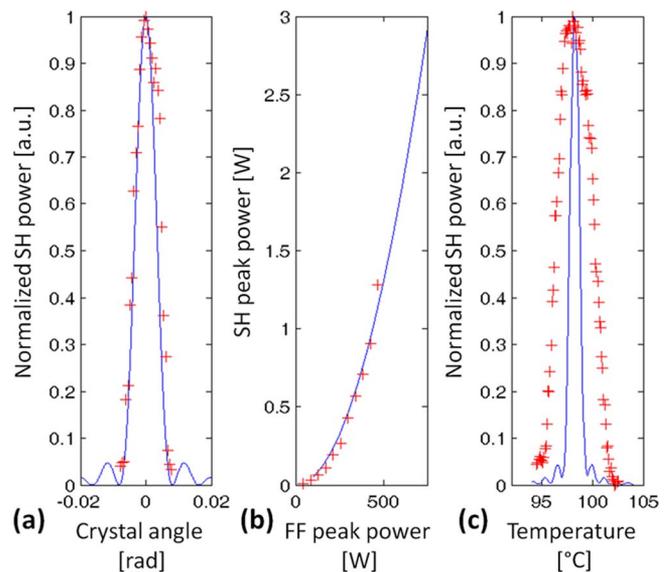


Fig. 3. (Color online) Comparison between measured (plus sign curves) and predicted (solid curves) results: (a) output power dependence on the rotation angle of the crystal, (b) output power dependence on input power, (c) output power dependence on temperature.

Table 2. Comparison between Predicted and Measured Conversion Efficiencies in the MgO:CLN and CLN Crystals

Crystal	Diffraction Order	Prediction [%W ⁻¹]	Measurement [%W ⁻¹]
MgO:	1	5.1×10^{-4}	7.55×10^{-4}
CLN	3	3.99×10^{-5}	3.96×10^{-5}
	5	7.62×10^{-6}	4.42×10^{-6}
CLN	2	1.95×10^{-5}	1.3×10^{-5}
	3	1.47×10^{-5}	1.51×10^{-5}

originating from changing the angle is equal to $G_0 \sqrt{2 - 2 \cos \varphi}$, where G_0 is the grating vector and φ is the rotation angle of the crystal. The predicted dependence of the output power on the rotation angle, assuming undepleted pump and plane-wave interaction, is given by $\text{sinc}^2(G_0 \sqrt{2 - 2 \cos \varphi} L / 2)$, where L is the length of the crystal along the propagation direction. A good agreement between the measured and the predicted result can be seen in Fig. 3(a). The expression for angular dependence is identical to the one expected for collinear phase-matching [Fig. 1(a)].

We examined the conversion efficiency of the first diffraction order in MgO:CLN by measuring the output power for different input powers. A comparison between simulated and measured results is presented in Fig. 3(b). A good agreement was found between the simulated and the measured conversion efficiencies of the examined modes for both crystals; the results are summarized in Table 2.

We also checked the temperature dependence of the nonlinear process. A comparison between measured and simulated results is presented in Fig. 3(c). The simulated result was shifted by 44.7 °C according to Table 1. The measured dependence is wider in comparison with the simulated one, and we believe that this can also originate from inaccuracies in the temperature dependent part of the Sellmeier equation for MgO:CLN [9]. A similar comparison between measured and simulated temperature dependence for the third diffraction order in CLN showed a better agreement. The measured and simulated FWHM were 7.3 °C and 8 °C, respectively. For comparison, in the case of collinear phase-matching, the FWHM is expected to be 2 °C.

In this Letter we presented results of phase-matched NLD for two specific crystals, but the same concept can be utilized for different wavelengths and different poling periods and in different materials. An additional material that can be examined is KTiOPO_4 (KTP); for example, if looking at an o-oe interaction with FF beams at 1.0645 μm , the required period for a crystal temperature of 80 °C is 8 μm [12].

Figure 4 presents the poling periods required to satisfy the phase-matching condition for either the first or the second diffraction order for a range of FF wavelengths for CLN, MgO:CLN, and KTP; in the first two materials the examined interaction is e-oo, and in KTP the interaction is o-oe. All the results are for 50 °C. For CLN we chose to show the second diffraction order, since for the same range of poling periods, the first diffraction order is phase-matched at much lower temperatures.

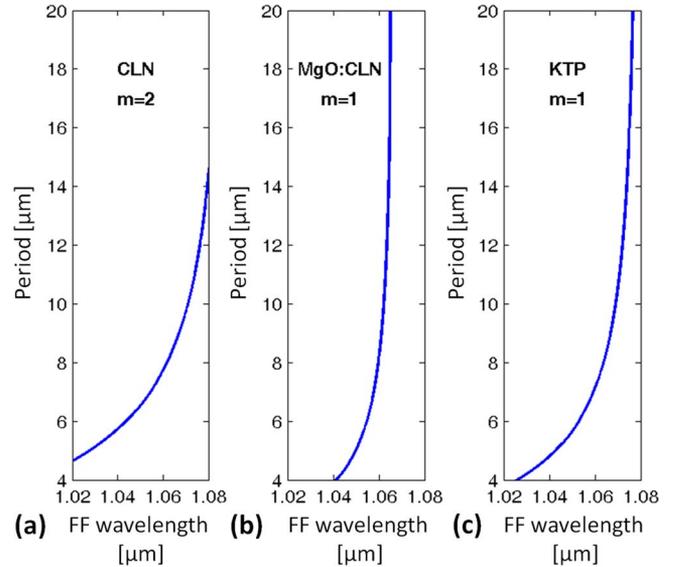


Fig. 4. (Color online) Required poling periods for phase-matched NLD for different FF wavelengths at 50 °C: (a) second diffraction order in CLN, (b) first diffraction order in MgO:CLN, (c) first diffraction order in KTP.

In conclusion, we have suggested and experimentally demonstrated a phase-matched NLD configuration resulting in efficient conversion of FF to SH in the first few diffraction orders. The basic concept is to incorporate both the natural birefringence of the crystal and QPM to allow for a complete fulfillment of the vectorial phase-matching condition. This configuration can increase efficiencies observed in diffraction experiments such as NLD from a virtual beam [8], can be beneficial in nonlinear microscopy [6], and can be used to characterize duty cycles of periodically poled nonlinear crystals.

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