Appl. Phys. B 85, 97–100 (2006)
DOI: 10.1007/s00340-006-2382-z

Applied Physics B
Lasers and Optics

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Quasi-phase matching in $LiNbO_3$ using nonlinear coefficients in the XY plane

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Received: 21 February 2006/Revised version: 11 June 2006
Published online: 14 July 2006 •

ABSTRACT We report quasi-phase-matched second-harmonic generation in periodically poled lithium niobate (PPLN), where both fundamental and second-harmonic waves are ordinary waves. It provides a lower-limit value for d_{22} of 1.1 pm/V. The measured temperature and wavelength bandwidth of the second-harmonic signal are in good agreement with the theoretical predictions. Since the $d_{22} = d_{YYY}$ nonlinear coefficient of $LiNbO_3$ changes its sign as a result of electric field periodic poling along the Z direction, we deduce that all tensor components of the second-order susceptibility $\chi^{(2)}$ of trigonal $3m$ crystals are reversed, thereby expanding the quasi-phase-matching possibilities in these crystals. Furthermore, it enables the realization of all-optical processes based on the nonlinear coefficients in the XY plane, such as all-optical polarization rotation in PPLN, as well as multipartite entanglement experiments based on simultaneous phase matching using different elements of $\chi^{(2)}$ in a single $LiNbO_3$ crystal.

PACS 42.65.-k; 42.65.Ky; 42.65.Lm

1 Introduction

Quasi-phase matching (QPM) is a widely used method today in designing ferroelectric crystals for nonlinear frequency conversion. QPM is usually achieved by periodically reversing the electrical dipole of the crystal along the Z direction. It is well known that this also reverses the sign of nonlinear coefficients that have Z dependence, for example $d_{33}(= d_{ZZZ})$ and $d_{32}(= d_{ZYY})$ [1, 2]. It is very interesting to investigate nonlinear processes using the other non-zero coefficients of the second-order nonlinear optical tensor, the XY plane nonlinear coefficients, namely d_{YYY} and $d_{YXX} = -d_{YYY}$. These coefficients allow us to expand the quasi-phase-matching possibilities in nonlinear processes. Furthermore, sign reversal of the XY plane nonlinear coefficients allows realization of second-order cascading processes, which are difficult to implement with only Z-dependent nonlinear coefficients. For example, the process of all-optical polarization rotation of a single frequency by two cascaded second-order nonlinear-optical processes [3] requires us to use $d_{22}(= d_{YYY})$. In addition, the sign reversal of the XY plane nonlinear coefficients can be useful in quantum information applications, e.g. for the realization of multipartite continuous-variable entanglement [4].

Most of the quasi-phase-matching experiments to date concentrated on periodic inversion of the largest nonlinear coefficient, d_{33}. Assuming that this was done using a stack of inverted plates [5] made from $LiNbO_3$, we can rotate all even plates by 180° either around the crystal X axis or around the Y axis. Both cases will lead to inversion of the d_{33} coefficient, but only the first will also invert the d_{YYY} coefficient (the axes maintain the right-hand Cartesian coordinate system). In the commonly used case of electric field poling along the Z direction, the domain inversion is equivalent to a rotation of 180° around the X axis [6]; hence d_{22}, as well as all other components of $\chi^{(2)}$, are inverted. As far as we know, there is a single qualitative observation of QPM second-harmonic generation (SHG) using the d_{22} coefficient in $LiNbO_3$, where the poling was performed by laser-heated pedestal growth [7]. Neither the conversion efficiency nor estimates of d_{22} are reported in [7]. Furthermore, we are not aware of QPM experiments using d_{22} in crystals poled by electric field poling.

In this article we present an experimental investigation of the d_{22} nonlinear coefficient behavior in periodically poled lithium niobate (PPLN). The measurements enable us to set a lower limit on the value of d_{22} in $LiNbO_3$, and provide an estimate of the accuracy of the Sellmeier equations for ordinary waves in $LiNbO_3$. In addition, we analyze all optical polarization rotation, based on ordinary-wave QPM processes.

2 Experimental setup and results

In order to study QPM processes based on d_{22} in electric field poled $LiNbO_3$, we have performed SHG experiments with a Y-polarized pump in two PPLN samples. The experimental setup (Fig. 1) includes either a tunable cw diode laser (ANDO AQ4321D), lasing in the range of 1520–1580 nm, and amplified by an Erbium-Doped Fiber Amplifier (EDFA) with a 100 mW saturation power, or an un-amplified tunable cw diode laser (New Focus 6262) emitting 5 mW in the range of 1506–1586 nm. The Y-polarized beam was focused into a waist radius of 22 μm in the middle of a ∼ 6-mm-long PPLN crystal. The poling length inside the crystals was 4.18 mm, and the poling periods 16 ± 0.01 μm and 15.48 ± 0.01 μm. The Y-polarized second-harmonic light was measured using a silicon photodetector, which is insensitive to the pump wavelength.

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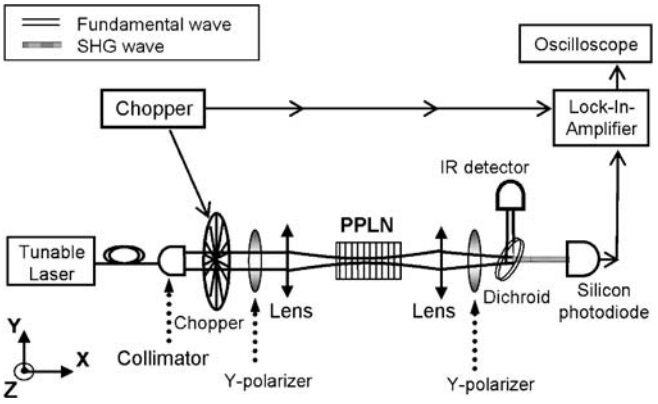

FIGURE 1 Experimental setup for QPM SHG using the d_{22} coefficient in PPLN

We have observed a phase-matched second-harmonic signal with both crystals. For the crystal with poling period 16 μm, the highest conversion efficiency is obtained at a wavelength of 1536.6 nm and a crystal temperature of 27 °C. By measuring the second-harmonic power as a function of the pump power, as shown in the inset of Fig. 2, we have deduced an external conversion efficiency coefficient of $3.15 \times 10^{-4}\,\%\,\mathrm{W}^{-1}$. Taking into account the Frensel reflection losses of the fundamental and second-harmonic waves (14.2% and 14.9%, respectively), and the poling length (4.18 mm), the normalized internal conversion efficiency is $0.12\,\%\,\mathrm{W}^{-1}\,\mathrm{m}^{-1}$. By comparing this measurement to the calculated conversion efficiency of a focused Gaussian beam [8] with a Boyd–Kleinman parameter $h = 0.754$, we have derived a lower-limit value $d_{22} = 1.1$ pm/V. This is a lower-limit value, since it assumes that the PPLN crystal has a perfect 50% duty cycle throughout the entire poling length of the sample. This value is almost twice lower than the previously commonly referenced value, $d_{22} = 2.1$ pm/V, for the fundamental wavelength 1.064 μm [9].

We have also measured the SHG power versus the pump wavelength (Fig. 2) and compared it to a theoretical calculation, based on the conversion efficiency equation for Gaussian beams [8]. Edwards and Lawrence [10] predicted that the phase-matched wavelength should be slightly lower than that measured, i.e. 1535.6 nm instead of 1536.6 nm. Note that the theoretical prediction assumes an optimal phase-mismatch value of $\sim 400\,\mathrm{m}^{-1}$ [8]. In Fig. 2 we have added a fixed offset to the calculated wave-vector difference in order to obtain the theoretical and experimental peaks at the same wavelength. This implies that the experimental refractive-index difference $n_{o2\omega} - n_{o\omega}$ is larger by 0.16% with respect to the one calculated based on [10], i.e. 0.04800 instead of the nominal calculated value of 0.04793. By using a different set of Sellmeier coefficients [11], which is given only at a fixed temperature of 21 °C, we obtain a slightly larger difference of 0.22%. The widths of the experimental and theoretical curves in Fig. 2 are essentially identical, thus indicating that the phase matching is obtained throughout the entire poled length.

The temperature dependence of the SHG power at a pump wavelength of 1541.4 nm, shown in Fig. 3, is compared to a theoretical curve, based on [8]. Edwards and Lawrence [10]

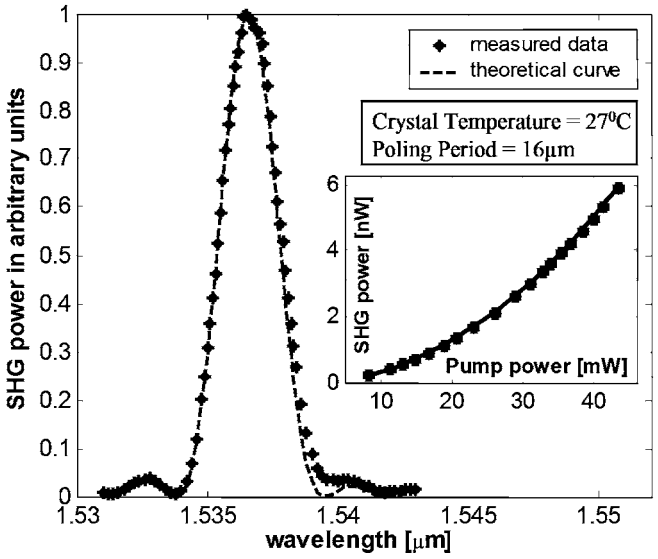

FIGURE 2 Measured and theoretical SHG power versus wavelength. Theoretical curve was shifted up by 1.0 nm in order to coincide with the measured curve. The *inset* is the measured and calculated second-order fit of SHG power versus fundamental power

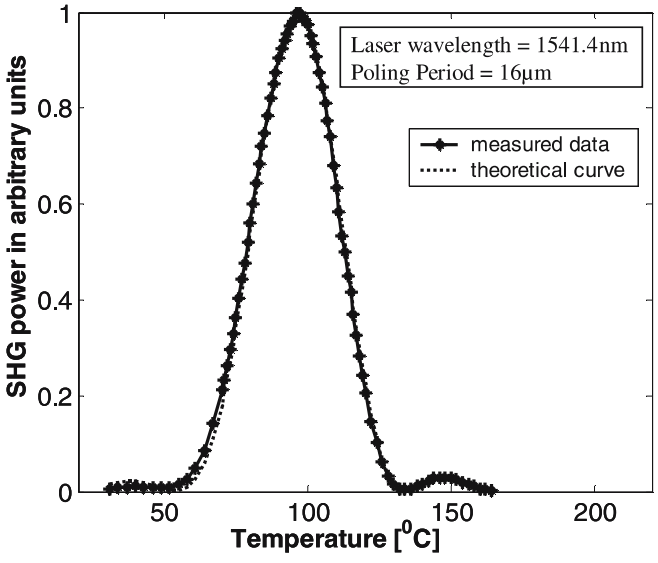

FIGURE 3 Measured and theoretical SHG power versus crystal temperature. Theoretical curve was shifted down by 16 °C in order to coincide with the measured curve

predicted that the phase-matched temperature should be slightly higher than that measured, i.e. 113 °C instead of 97 °C. By applying again the fixed correction of 0.16% to the refractive-index difference $n_{o2\omega} - n_{o\omega}$ as found in the previous experiment, we observe the theoretical and experimental peaks at the same temperature. The widths of the experimental and theoretical curves are essentially identical.

The phase-matched wavelength versus temperature was measured for both crystals (Fig. 4). Compared to theoretical curves based on [8, 10], there is a ~ 1.5 nm shift at pump wavelengths near 1520 nm from the measurement on the 15.48-μm PPLN crystal, and a $\sim$ 1-nm shift at pump wavelengths near 1540 nm from the measurement on the 16-μm PPLN crystal. By applying the fixed correction of 0.16% to the refractive-index difference $n_{o2\omega} - n_{o\omega}$, the theoretical

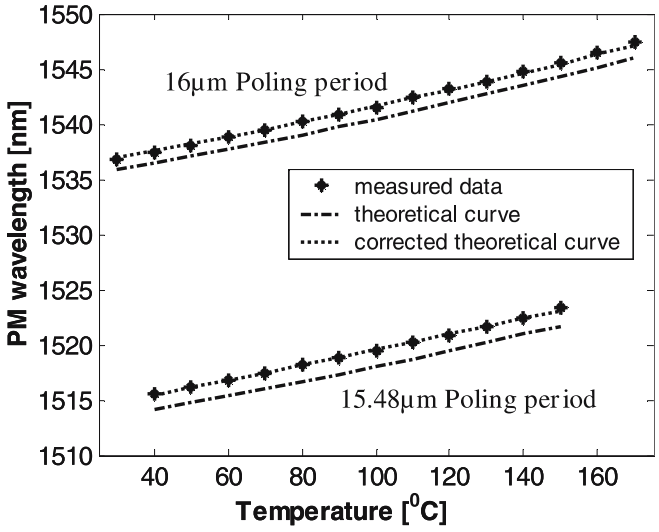

FIGURE 4 Measured and theoretical ([10] and corrected by 0.16%) phase-matched wavelength versus temperature in both PPLN samples

and experimental curves coincide. In fact, the same correction factor of 0.16% leads to a good agreement between the measurements and theoretical predictions in the three different experiments we have performed.

3 Discussion

The phase-matched process that was observed relies on periodic reversal of the d_{22} coefficient. Moreover, according to the second-order susceptibility $\chi^{(2)}$ spatial symmetry [12], $d_{21}\,(=d_{YXX}) = d_{16}\,(=d_{XYX}) = -d_{22}\,(=d_{YYY})$ in $LiNbO_3$, and consequently it is reasonable to conclude that the sign of d_{21} and d_{16} is also periodically reversed in the case of periodic poling. Furthermore, the d_{33} and d_{31} nonlinear coefficients are also reversed in PPLN. Hence, all non-zero PPLN tensor components of $\chi^{(2)}$ are reversed when periodically poling in the Z direction.

Quasi-phase matching using the d_{22} coefficient can be used for example in $\chi^{(2)}:\chi^{(2)}$ cascaded cross polarized wave (XPW) generation. Presently this process is realized, for example, in BaF_2 by a $\chi^{(3)}$ four-wave-mixing process, and has a direct and important application for contrast enhancement of femtosecond pulses in petawatt femtosecond laser systems [13, 14]. Efficiencies of XPW generation in the order of 10% require an extremely high intensity level of 400 GW/cm^2 at a pulse duration of 50 fs. With cascading in quadratic crystals, one can achieve the same efficiency with much less power density because it is well known that the effective cubic nonlinearity based on second-order cascading can exceed significantly the nonlinear effects based on the bulk cubic nonlinearity $\chi^{(3)}$. Furthermore, the efficiency of the cascaded $\chi^{(2)}:\chi^{(2)}$ process in the condition of double phase matching is proportional to the fourth power of the crystal length, contrary to a single-step process, which is proportional only to the square of the crystal length. The two steps required for cascaded single quadratic crystal XPW generation are: generation of a Y-polarized second harmonic with a Y-polarized fundamental wave using the $d_{22}\,(=d_{YYY})$ coefficient, followed by difference-frequency mixing based on the $d_{32}\,(=d_{ZYY})$ coefficient of the Y-polarized second-harmonic and Y-polarized fundamental waves, generating in this way a XPW at the fundamental frequency. To estimate the advantage of the cascaded approach, we calculated the ratio of the efficiencies of the cascaded XPW generation process in $LiNbO_3$ and the direct XPW generation process in BaF_2 in approximation of non-depleted pump and with the same input intensity [15, 16]:

$$\frac{\eta\,(\text{XPW-cascaded})}{\eta\,(\text{XPW-direct})} = \left[\frac{128 n_{\omega,\mathrm{BaF_2}}}{3\pi n_{\omega,\mathrm{LN}} n_{2\omega,\mathrm{LN}} m_1 m_2}\,\frac{d_{YYY} d_{ZYY}}{\chi^{(3)}_{\mathrm{BaF_2}}}\,\frac{L}{\lambda}\right]^2 . \tag{1}$$

Using $n_{\omega,\mathrm{BaF_2}} = 1.474$, $n_{\omega,\mathrm{LN}} = 2.21$ and $n_{2\omega,\mathrm{LN}} = 2.26$ (refractive indices at $\lambda = 1.536\,\mu$m and at room temperature), $\chi^{(3)}_{\mathrm{BaF_2}} = 160\,\mathrm{pm^2/V^2}$ [17], $d_{YYY} = 1.1$ pm/V, $d_{YYZ} = 4.3$ pm/V, $L \sim 4$ mm, and assuming that the product of the QPM orders for the two cascading steps $m_1 m_2 = 1$ we obtain that the direct process is $\sim 95\,000$ times weaker. Since the efficiency of the process is proportional to the square of the pump intensity, for the same XPW generation efficiency the $\chi^{(2)}:\chi^{(2)}$ cascaded process requires ~ 310 times less intensity than the direct four-wave-mixing process.

Another application in the field of quantum information is multipartite entanglement based on simultaneous phase matching in a single crystal of several optical parametric oscillator channels, as recently proposed for a KTP crystal [4]. However, in contrast to KTP crystals, where there are only three possible second-order interactions, in $LiNbO_3$ there are four possible second-order interactions for an x-cut sample: ZZ-Z; YY-Z; YZ-Y and YY-Y.

4 Summary

In this article, we have studied QPM interactions using nonlinear coefficients in the X–Y plane of electric field poled $LiNbO_3$. The measurements set a lower limit on the magnitude of d_{22} of 1.1 pm/V. Furthermore, we conclude that all components of the PPLN electro-optic and nonlinear tensors will have periodic sign reversal. In addition, we deduce that other trigonal $3m$ uniaxial ferroelectric crystals behave in a similar fashion, and in particular $LiTaO_3$. This enables new and interesting possibilities, for all-optical processing and multipartite entanglement, based on nonlinear and Pockels electro-optic effects in periodically poled $LiNbO_3$ and $LiTaO_3$ using Y- and X-polarized components of the $\chi^{(2)}$ tensor or the electro-optic tensor.

ACKNOWLEDGEMENTS This work was supported by the Israeli Ministry of Science and by the Israel Science Foundation, Grant No. 960/05. S.M. Saltiel is thankful for the hospitality of Tel-Aviv University, Department of Physical Electronics and for the support of the Gordon Center for Energy Studies and the Bulgarian National Science Fund (Grant No. F1201/2002).

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