Nearly-Noncritical Phase Matching in $MgO:LiNbO_3$ Optical Parametric Oscillators *

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We have proposed and demonstrated a nearly-noncritical phase-matched (NCPM) optical parametric oscillation (OPO) in an $MgO:LiNbO_3$ crystal with 5 mol% MgO by temperature tuning. By giving up perfect NCPM, the practical tuning range for the OPO is increased by two times. For the crystal, the operated temperature decreases with the phase-matching angle at degeneracy. With a cutting angle of 82° instead of the noncritical case of 90°, the tuning range was increased. In order to obtain a sufficiently high output pulse energy for both signal and idler throughout the entire tuning range, five sets of mirrors were used for the resonator. The tuning range of the OPO was $800-1700$ nm with temperatures tuning from 83°C to 224.2°C. The output energy was about 6.45 mJ with a conversion efficiency of nearly 13%. The bandwidth of the output was $1.0-1.1$ nm.

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Lithium niobate ($LiNbO_3$) is one of the most important nonlinear crystals for nonlinear frequency conversion due to its large nonlinearity. Moreover, large-sized crystals of optical quality can be grown. However, the $LiNbO_3$ crystal has its drawbacks, such as large photorefractive effect and low damage threshold. The large photorefractive effect is due to a change of the refractive indices under the effect of laser radiation. In 1980, Zhong *et al.*[1] suggested that $LiNbO_3$ could be doped with magnesium oxide (MgO) to reduce the photorefractive damage. Bryan *et al.*,[2] Sweeney *et al.*[3] and Volk *et al.*[4] also reported similar results leading to the same conclusions. Since then, $MgO:LiNbO_3$ has been used for second-harmonic generation (SHG) and optical parametric oscillation (OPO) extensively. Using a 12 mm long $MgO:LiNbO_3$ crystal we obtained an SHG conversion efficiency of 45% at a fundamental peak-power density of 140 MW/cm^2 without any damage.[5]

A number of papers have discussed OPO with the $MgO:LiNbO_3$ crystal. In order for NCPM to be achieved, temperature tuning is necessary. The temperature changes the refractive indices, which alters the phase-matching condition and the output wavelengths. Kozlovsky *et al.*[6,7] and Nabors *et al.*[8] reported a cw doubly-resonant (DR) OPO at the temperature range of $107-110$°C. The tuning range was $1000-1130$ nm and a conversion efficiency of $34-40$% was achieved. Using both 1064 and 532 nm pumpings, Yang *et al.*,[9] Schiller *et al.*[10] and Gerstenberger *et al.*[11] obtained similar results. Using 30 ps pulses as the pumping source, He *et al.*[12] obtained a wide tuning region of $700-2000$ nm, albeit with a smaller conversion efficiency of 5.5%. Dmitriev *et al.*[13] presented a good summary of results on OPO with $MgO:LiNbO_3$. In most cases of noncritical phase-matching (NCPM) temperature tuning, the OPOs pumped by the fundamental at 1064 nm and the second harmonic at 532 nm of the Nd:YAG laser have relatively narrow tuning regions of around $100-200$ nm. We have reported an OPO with high output energy and wide tuning region without any damage. With nanosecond pulses, a singly-resonant OPO (tuning region $738.9-1032.2$ nm) and a doubly-resonant OPO (tuning region $844.1-1411.3$ nm) with an energy conversion efficiency of 10.4% could be obtained.[14-17]

The aim of this letter is to find a way to obtain a wide tuning range for the $MgO:LiNbO_3$-OPO device with reasonable efficiency. In calculations for the device with the tuning range of $800-1700$ nm, angle tuning needs $24-65$° at room temperature (20°C). If tuning ±7° for every $MgO:LiNbO_3$ crystal it will require three crystals at least to be used. Thus it will be more complex for adjusting and changing crystals, and it will also introduce more loss, to increase the threshold and to decrease the conversion efficiency when the crystal tilts from the normal position. Thus, for a wide tuning range, temperature tuning is a good method.

In this letter, we report on a study of the nearly-NCPM case and show that a wider tuning range can be achieved with reasonable efficiency. By relaxing the conditions on the phase-matching angle, it is possible to extend the wavelength tuning range for the same temperature range of the OPO.

In order to calculate the tuning curve of the $MgO:LiNbO_3$-OPO, we need to obtain the temperature and wavelength dependence of the ordinary and extraordinary indices of refraction. Obviously, the indices of refraction are related to the doping concentration of MgO in $MgO:LiNbO_3$ crystals. The typical

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MgO:LiNbO$_3$ is grown in a congruent melt doped with 5 − 7 mol% of MgO. The Sellmerier equations for the MgO:LiNbO$_3$ crystal at room temperature were obtained experimentally, i.e.,

$$n_o^2 = 4.9130 + \frac{1.173\times10^5 + 1.65\times10^{-2}T^2}{\lambda^2 - (2.12\times10^2 + 2.7\times10^{-5}T^2)^2} - 2.78\times10^{-8}\lambda^2, \quad (1)$$

$$n_e^2 = A + 2.605\times10^{-7}T^2 + \frac{0.97\times10^5 + 2.7\times10^{-2}T^2}{\lambda^2 - (2.01\times10^2 + 5.4\times10^{-5}T^2)^2} - 2.24\times10^{-8}\lambda^2, \quad (2)$$

where T is in units of K and wavelength λ is in nanometres. These equations are valid for the wavelength range from 400 to 4000 nm and the temperature range from 273 to 673 K. Notice that the first-order term for n_e is represented by a parameter A. We assume that it changes in the refractive indices of ordinary (o) and extraordinary (e) waves due to the fact that the doping is of the first order, i.e., n_o is constant and n_e is dependent on the doping concentration. The exact value of A therefore is different for different crystals and has to be determined for each sample.

In order to determine the constant A, we consider type I-NCPM from fundamental wave (1064 nm) to second-harmonic wave (532 nm) with the MgO:LiNbO$_3$ crystal. In this case the phase matching condition is given by

$$n_o(1064\text{ nm}, T_{\text{ncpm}}) = n_e(532\text{ nm}, T_{\text{ncpm}}, A). \quad (3)$$

From Eqs. (1)−(3) we can obtain the relation between T_{ncpm} and A. This relationship is shown in Fig. 1 (dots). The points can be fitted by the following formula:

$$A = 4.5667 - 2.1432\times10^{-4}T_{\text{ncpm}} - 4.07\times10^{-7}T_{\text{ncpm}}^2. \quad (4)$$

Thus, in order to determine A, we can simply measure the NCPM temperature T_{ncpm}. In our experiments, two MgO:LiNbO$_3$ samples, with dimensions of $4\times4\times35$ mm and $5\times5\times25$ mm, were used. The NCPM temperature T_{ncpm} was measured to be 85°C and 110°C for SHG from 1064 to 532 nm. Therefore we could obtain that A equals to 4.5455 and 4.5382 for these two samples, respectively. In order to check the accuracy of the measured SHG phase-matching temperature, we also measured the signal and idler waves for an OPO operated at 85°C and pumped by the SHG of the Nd:YAG. The signal and the idler waves were measured to be 1062.5 and 1064.4 nm respectively. These values are very close to the degenerate case.

Using Eqs. (1) and (2) and values of A, we calculated curves of n_o and n_e versus wavelength at different temperatures as shown in Fig. 2. From this figure it is obvious that: firstly, n_o is almost independent of temperature; secondly, n_e is temperature dependent; thirdly, the difference between n_o and n_e increases with the increasing temperature. The phase-matching angle θ is 90°, and the walk-off angles are zero for NCPM. Whereas for the MgO:LiNbO-OPO, NCPM operation always leads to a narrow temperature tuning range because of practical temperature limits. Here we have shown that a nearly-noncritical situation could lead to a much wider wavelength tuning range. One can plot the temperature T_{dp} of the degeneracy point and obtain the operating temperature for an OPO. The degeneracy case is the same as type I SHG for the Nd:YAG laser. The condition is given by

$$n_o(1064\text{ nm}, T) = n_e(532\text{ nm}, T, \theta). \quad (5)$$

The phase-matching temperature for a fixed cutting angle θ is calculated and plotted in Fig. 3. This SHG phase-matching temperature is the same as T_{dp}. By calculation, $T_{\text{dp}} = 109$°C for NCPM ($\theta = 90°$). The tuning curve of an OPO under the exact NCPM condition is shown in Fig. 4. The lowest temperature for tuning is T_{dp} which is 109°C in this case. Thus the tuning range for this OPO is limited by the high-temperature requirement for tuning.

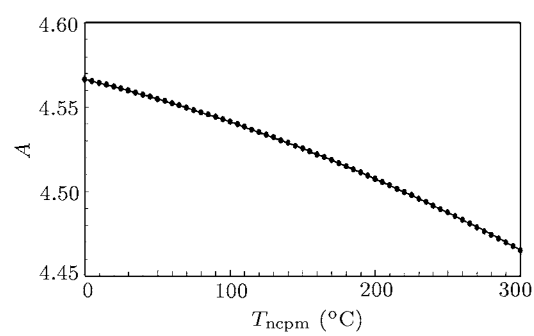

Fig. 1. Corrected constant A versus noncritical phase-matching temperature T_{ncpm}.

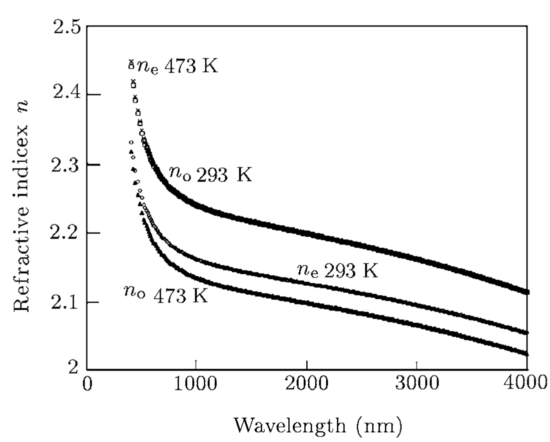

Fig. 2. Refractive indices n_o and n_e versus wavelength at different temperatures.

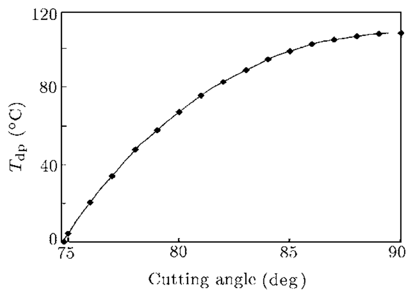

Fig. 3. Degeneracy temperature T_{dp} versus cutting angle θ at $\lambda = 1064$ nm.

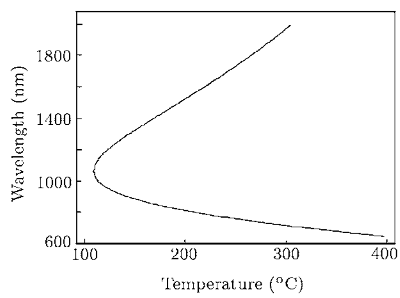

Fig. 4. Temperature tuning curve of OPO pumped by a 532 nm laser at noncritical phase matching at $T_{dp} = 109°$C.

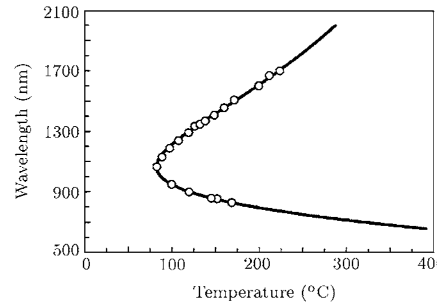

Fig. 5. MgO:$LiNbO_3$ OPO temperature tuning curve: solid, calculated; dots, experimental.

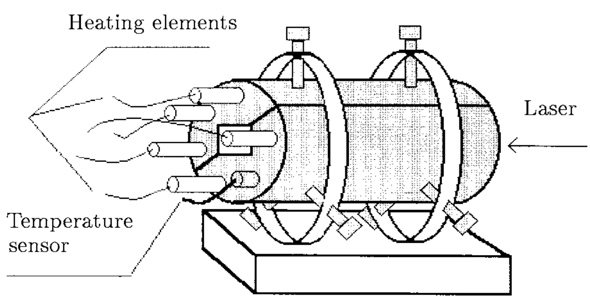

Fig. 6. Heating device for temperature tuning of OPO.

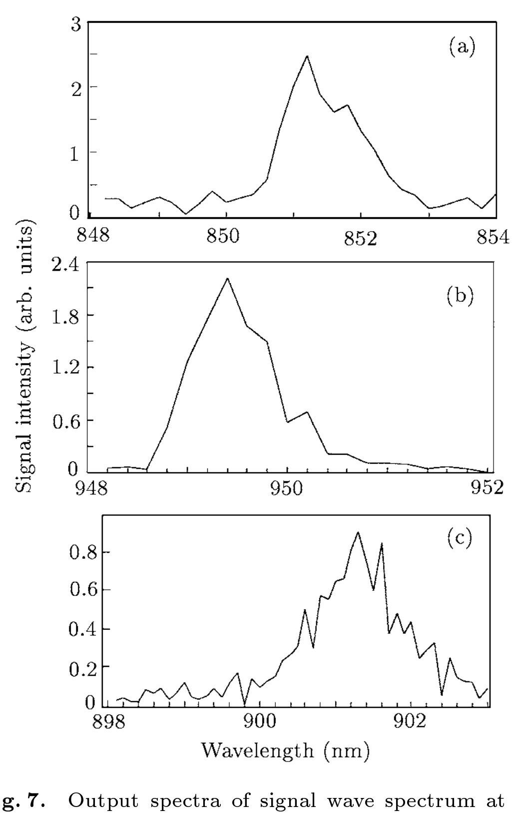

Fig. 7. Output spectra of signal wave spectrum at (a) 100°C, (b)120°C and (c)150°C.

Now, if the cutting angle of the crystal deviates slightly from the NCPM angle of $\theta = 90°$, for example, $\theta = 80°$, T_{dp} becomes 67.2°C which is much lower than the case for $\theta = 90°$. This means that the operating temperature of an OPO can be lower and hence the tuning range of the OPO can also be much broader. Here we should consider conversion efficiency, walk-off angle and wide tuning range simultaneously in designing the OPO. A nearly-NCPM is suggested. This is a compromise in the beam walk-off in order to decrease the operating temperatures. In our design, we used a cutting angle θ of 82° which corresponds to $T_{dp} = 83°$C. The tuning curve for this OPO can be calculated using Eqs. (1) and (2) and is shown in Fig. 5. The tuning range from 800 to 1700 nm can be obtained with temperatures from 83°C to 224.2°C. This temperature range is easy to realize and is quite practical. The calculation shows that the walk-off angles are $1 - 2.5°$ through the tuning range ($800 - 1700$ nm, temperature tuning from 83°C to 225°C) for the case of $\theta = 82°$ phase matching, and the difference of an effective nonlinear coefficient between the angle tuning and the temperature tuning is less than 4%.

The $LiNbO_3$ crystal used was doped with more than 5 mol% MgO with dimensions of $8(\pm 0.1) \times 8(\pm 0.1) \times 34.5$ mm. For this crystal, the constant A in the Sellmeier equations has a value of 4.5455. This crystal was cut at $\theta = 82° \pm 5'$ along the negative $y - z$ plan for nearly-NCPM. The pumping source was a Q-switched and frequency-doubled Nd:YAG laser with a pulse width of $5 - 8$ ns, at a repetition rate of $1 - 10$ Hz. The maximum output energy of the second harmonic at 532 nm was 50 mJ, with a bandwidth of less than 1 nm and a divergence of less than 0.9 mrad.

The OPO resonator had a flat–flat configuration. The cavity length was 70 mm. There was no coating on one of the end faces of the MgO:$LiNbO_3$ crystal. To obtain a sufficiently high output pulse energy for both signal and idler, five sets of mirrors were used for the wavelength ranges of 700−950, 950−1200, 1200−1300, 1300 − 1450 and 1400 − 1700 nm, respectively. The reflectivity of the end mirrors of the resonator were always higher than 99%. The output coupler reflectivity was around 50%. Such combinations guarantee a sufficient high output pulse energy for both signal and idler waves. The crystal was placed in a high-temperature oven for temperature tuning as shown in Fig. 6. An REXC-100 temperature control device was employed. The temperature can be increased to 400°C with a high-temperature control precision of ±0.1°C. We kept the temperature at less than 225°C. Figure 5 shows the experimental tuning curve (dots). These data points fit the theoretically obtained tuning curve (solid line) exactly, indicating that the parameters used in the calculation were reasonable. From Fig. 5, it is seen that a wide wavelength tuning from 800 to 1700 nm was obtained for the corresponding temperature range of 83 − 193.7°C to 83 − 224.2°C. This wavelength tuning range is the largest reported for the temperatures indicated. It is the direct result of giving up slightly on the NCPM condition.

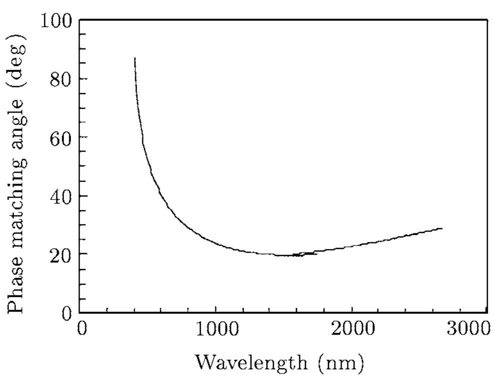

Fig. 8. BBO-SHG phase matching angle versus wavelength.

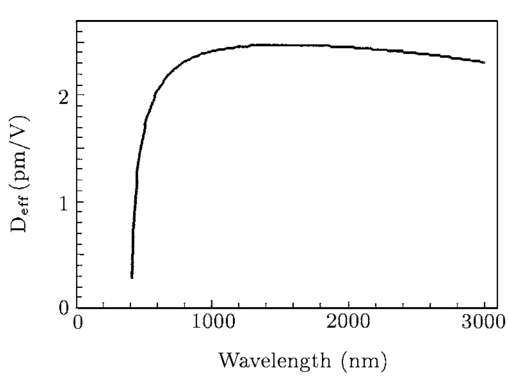

Fig. 9. BBO-SHG effective nonlinear optical coefficient (type I phase matching) versus wavelength.

The output energy of the signal and idler waves of the OPO was larger than 2 mJ in the whole tuning region of 800 − 1700 nm and larger than 5 mJ at 1128 − 1300 nm. The typical output energy at 1128 nm (89.1°C) was 6.45 mJ which corresponds to a conversion efficiency of nearly 13%. This efficiency is quite high and comparable to the doubly resonant NCPM cases. The output spectra of the signal wave at 100°C, 120°C and 150°C were shown in Figs. 7(a)−7(c), respectively. It can be seen that the bandwidth varies slightly from 1.0 to 1.1 nm. The tuning range of this OPO can be easily extended by SHG. Using a typical BBO crystal as an SHG device, we can extend it to the wavelength region of 400 − 850 nm. Figures 8 and 9 show the type I phase-matching angles and the effective nonlinear coefficient for BBO-SHG. Experimentally, a piece of BBO with $\theta = 22.8°$, $\varphi = 0°$ and dimensions of $8 \times 8 \times 7$ mm was used. The SHG output energy of 1 mJ could be obtained, which corresponds to a conversion efficiency of 17%. The final bandwidth of the SHG was 1 nm.

In summary, we have proposed and demonstrated the application of nearly-NCPM for OPO. The idea is to remain a little on the beam walk-off and NCPM condition for reducing the operating temperature. In the present case of $MgO:LiNbO_3$, the tuning range of the OPO was extended by more than two times for the practical temperature range of 83-225°C. We have constructed and demonstrated this OPO with a good efficiency and high output energies. We have also demonstrated that this OPO can be extended further in tunability by SHG with a BBO crystal. This device should find many applications in spectroscopy and pollution monitoring.

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