

# Corrections to refractive index data of stoichiometric lithium tantalate in the 5–6 $\mu\text{m}$ range

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We propose corrections to the coefficients in the published Sellmeier equation for stoichiometric  $\text{LiTaO}_3$  [Opt. Lett. **28**, 194 (2003)] that allow the extension of the wavelength range within the region of midinfrared absorption edge up to 6  $\mu\text{m}$ . The required extraordinary refractive index data for this range were obtained using single-pass optical parametric fluorescence measurements with a pump wavelength of 1064.4 nm. We also observed efficient parasitic second-harmonic generation that could affect some quasi-phase-matching interactions. The corrected Sellmeier equation improves the accuracy of poling period calculations where the idler wavelength is within the region. © 2006 Optical Society of America

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Periodic poling of ferroelectric second-order nonlinear optical crystals has produced a variety of quasi-phase-matching (QPM) devices capable of generating, via efficient optical parametric conversion, a wide range of wavelengths across the visible and midinfrared.<sup>1,2</sup> While up to now periodically poled lithium niobate (PPLN,  $\text{LiNbO}_3$ ) has usually been the material of choice, its susceptibility to damage caused by photorefractive and green-induced infrared absorption<sup>3</sup> has limited its use at high average powers, especially for visible wavelengths. Doping with MgO and operation at elevated temperatures increase the threshold for photorefractive damage by increasing the material conductivity, although some residual photorefractivity remains. Periodically poled stoichiometric lithium tantalate<sup>4,5</sup> (PPLSLT,  $\text{LiTaO}_3$ ) is an emerging material that appears more suitable for high average power applications because of its reduced photorefractivity due to lower defect levels and the elimination of Li deficiency. Additionally,  $\text{LiTaO}_3$  has somewhat lower absorption at wavelengths about 5.5–6  $\mu\text{m}$  compared with  $\text{LiNbO}_3$ ,<sup>6</sup> which could be favorable for generation in this range. It has been demonstrated that even PPLN can provide useful infrared powers at wavelengths as long as 6.6  $\mu\text{m}$  in spite of the strong absorption.<sup>6</sup>

Both undoped<sup>5,7</sup> and MgO-doped<sup>8</sup> near-stoichiometric  $\text{LiTaO}_3$  have been used for QPM devices. MgO doping at 1.0 mol. % was suggested<sup>8</sup> to further reduce the photorefractivity. Importantly, it has been shown<sup>9</sup> that at room temperature this doping level has little effect on the extraordinary refractive index of near-stoichiometric  $\text{LiTaO}_3$ . A similar result was also reported<sup>10</sup> for stoichiometric  $\text{LiNbO}_3$  (MgO doping up to 4.6 mol. %), in contrast with the more common congruent crystal exhibiting significant variation with MgO doping. Accurate knowledge of the refractive index and its dispersion described by the Sellmeier equation is required for precise calculation of the poling period for any particular QPM interaction. Reliable QPM calculations for MgO-doped SLT at temperatures close to 200 °C were performed<sup>8</sup>

using the Sellmeier equation for undoped SLT,<sup>11</sup> suggesting that the observed near independence of the refractive index on doping concentration is not restricted to room temperature and, therefore, the same temperature-dependent Sellmeier equation could apply for undoped as well as up to 1 mol. % doped SLT crystals. The existing Sellmeier equation<sup>11</sup> is accurate within the wavelength region 0.39–4.1  $\mu\text{m}$ . Attempting to use it for QPM calculations for an optical parametric process with the idler wavelength within the midinfrared absorption edge around 5–6  $\mu\text{m}$ , we found considerable deviation between the actual idler refractive index values and the theoretical prediction. This deviation was large enough to cause an error in the calculated poling period of 0.7  $\mu\text{m}$ . The resulting shift in the desired signal (idler) wavelength was too large to be compensated by temperature tuning, thus rendering the QPM device useless for the intended interaction.

To extend the applicability range of the existing Sellmeier equation towards longer wavelengths, we used a set of six periodically poled, 1.0 mol. % MgO-doped, near-stoichiometric  $\text{LiTaO}_3$  crystals (Li/Ta ratio 0.996–0.999) obtained from two suppliers: SWING Corp. of Japan and HC Photonics Corp. of Taiwan. The poling periods were 21.9, 23.2, 23.4, 24.1, 24.7, and 30.2  $\mu\text{m}$ . The crystals were mounted in a temperature-controlled oven and irradiated in a single pass with the pump at 1064.4 nm as shown in Fig. 1. The pump beam consisted of 13 ps pulses at a 1.5 MHz repetition rate generated by a long-cavity mode-locked Nd:YVO<sub>4</sub> laser<sup>12</sup> coupled to a Nd:YVO<sub>4</sub> amplifier capable of producing average power in excess of 45 W.<sup>13</sup> A few watt average power pump beam

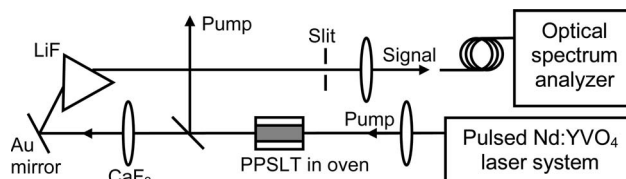


Fig. 1. Experimental setup.

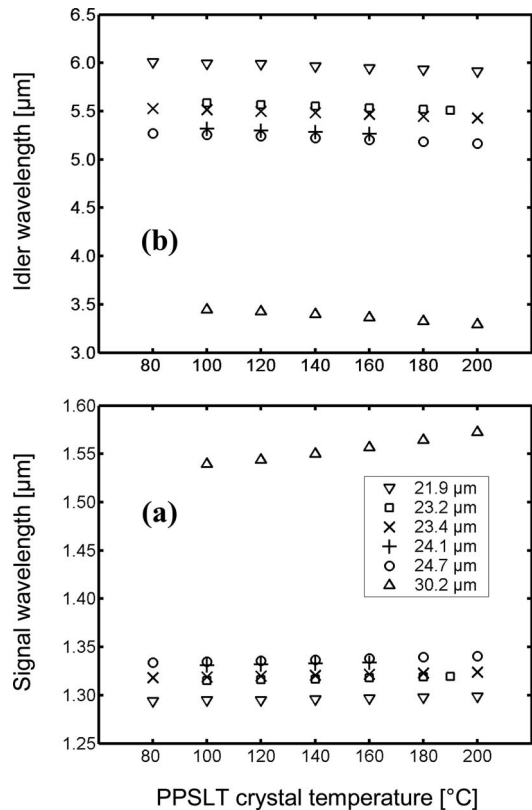


Fig. 2. (a) Experimentally obtained values for the signal wavelength at different temperatures using six crystals with poling periods according to the legend. The pump wavelength was 1064.4 nm. (b) Corresponding idler wavelengths.

was focused in the crystal to a spot with a diameter around 150  $\mu\text{m}$ , achieving peak intensities up to  $9 \times 10^8 \text{ W/cm}^2$ . The amplified parametric fluorescence from the output of the crystal was collimated, passed through a LiF prism and a slit to discard any residual pump and parasitically generated wavelengths, and coupled into a multimode optical fiber connected to an ANDO AQ6317B optical spectrum analyzer. Figure 2(a) shows the measured central wavelengths of the signal for the six crystals at different temperatures; Fig. 2(b) shows the calculated corresponding idler wavelengths.

Obviously, for the wavelengths of interest (5–6  $\mu\text{m}$ ) the temperature dependence is not strong, and hence temperature tuning of the QPM device is not very effective, making the poling period calculation accuracy critically important. The idler wavelength produced using the 30.2  $\mu\text{m}$  poling period crystal was much shorter and well within the range where the existing Sellmeier equation was correct. The crystal was essentially used as a reference to verify the applicability of the equation for our MgO-doped material. This was also confirmed via direct room temperature measurement of the extraordinary refractive index at 633, 810, and 1064 nm, using the prism coupling technique. Therefore it was justified to assume that the existing Sellmeier equation was correct for the signal and pump wavelengths in our QPM experiments; hence we were able to calculate the extraordinary refractive index for the idler

wavelengths from the results of these experiments. The refractive index data for the 5–6  $\mu\text{m}$  range, obtained in this way, were then used to correct the coefficients in the temperature-dependent Sellmeier equation<sup>11</sup>:

$$n_e^2(\lambda, T) = A + \frac{B + b(T)}{\lambda^2 - [C + c(T)]^2} + \frac{E}{\lambda^2 - F^2} + \frac{G}{\lambda^2 - H^2} + D\lambda^2. \quad (1)$$

A standard least-squares approximation algorithm was employed, keeping the changes affecting the visible and near-infrared regions negligible. The general form of the equation and the temperature-dependent coefficients,  $b(T)$  and  $c(T)$ , were not altered. Corrected coefficients are presented in Table 1 in comparison with the original coefficients. The main difference is in significantly increased strength of the low-energy oscillator close to 7  $\mu\text{m}$  accompanied by a slight blueshift in the position of this oscillator. A graphical comparison between refractive index values calculated using the two sets of Sellmeier coefficients at 140 °C is presented in Fig. 3. For wavelengths below 4  $\mu\text{m}$ , both sets of coefficients produce essentially identical results. Importantly, the point corresponding to the 30.2  $\mu\text{m}$  period reference crystal (at  $\sim 3.5 \mu\text{m}$ ) agrees well with the original Sellmeier equation. As is evident from Figs. 3 and 4, some deviations between the experimental results and calculations still remain. One possible reason could be that a simplified equation such as Eq. (1), generally adequate for the transparency region, could have limited application where strong absorption with complex wavelength dependence exists. Slight variations in the stoichiometry of the crystals used could have also affected the fit. This is very difficult to address, since the complexity of current technological processes often leads to minor compositional differences in near-stoichiometric crystals produced by different vendors or even from different batches from the same vendor. However, from a practical point of view, the new set of coefficients results in considerably in-

Table 1. Values of the Coefficients in Sellmeier Eq. (1) for SLT

Coefficient	Original <sup>a</sup>	Corrected <sup>b</sup>
$A$	4.502483	4.528254
$B$	0.007294	0.012962
$C$	0.185087	0.242783
$D$	-0.02357	-0.02288
$E$	0.073423	0.068131
$F$	0.199595	0.177370
$G$	0.001	1.307470
$H$	7.99724	7.061878
$b(T)$	$3.483933 \times 10^{-8}$ $(T+273.15)^2$	$3.483933 \times 10^{-8}$ $(T+273.15)^2$
$c(T)$	$1.607839 \times 10^{-8}$ $(T+273.15)^2$	$1.607839 \times 10^{-8}$ $(T+273.15)^2$

<sup>a</sup>Ref. 11.

<sup>b</sup> $T$  Temperature-dependent coefficients were not changed.

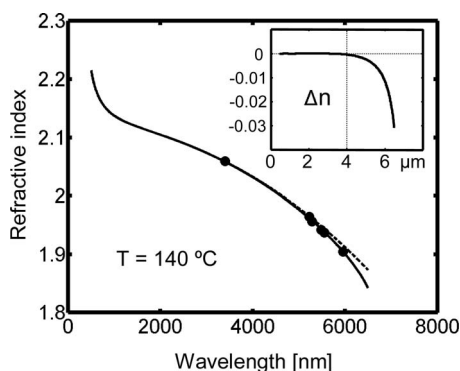


Fig. 3. Comparison between the values calculated for the refractive index by using the Sellmeier equation [Eq. (1)] with the original (dashed curve) and corrected (solid curve) coefficients. The dots represent experimental points. The inset shows the difference in the refractive index calculated by using the two sets of coefficients.

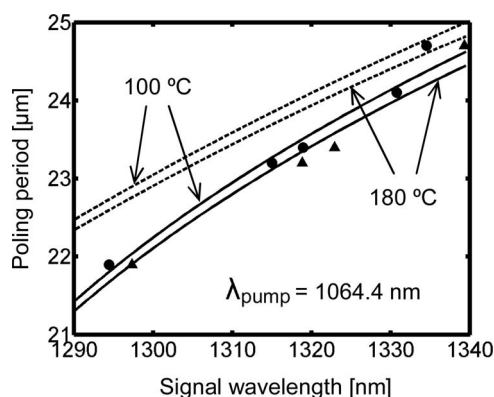


Fig. 4. Poling period calculated using original (dashed curves) and corrected (solid curves) Sellmeier coefficients at  $100^\circ\text{C}$  and  $180^\circ\text{C}$  as a function of the design signal wavelength. Our experimental results are represented by dots at  $100^\circ\text{C}$  and triangles at  $180^\circ\text{C}$ .

creased accuracy in calculations of the poling period required for optical parametric interactions when the idler wavelength is within the region of the midinfrared absorption edge. In most cases, the errors in poling period estimates calculated using corrected coefficients could simply be compensated by temperature tuning.

During our experiments, we observed relatively strong second-harmonic generation (SHG) of the pump, especially for the crystals with 23.2 and 23.4  $\mu\text{m}$  poling periods. For the 23.4  $\mu\text{m}$  crystal, maximum conversion efficiency in excess of 50% was observed close to  $140^\circ\text{C}$  at a pump intensity of  $\approx 8 \times 10^8 \text{ W/cm}^2$ . Calculations using both Sellmeier equations predict a period of  $\approx 23.36 \mu\text{m}$  for third-order QPM SHG at  $140^\circ\text{C}$ , which is in good agreement with the experiment. This parasitic SHG is a process that competes with parametric generation and might interfere with parametric devices producing wavelengths close to 5.5  $\mu\text{m}$ . In addition, we frequently observed laser damage at the output face of the crystal as the temperature was increased but was still significantly below the QPM temperature for SHG, and this had little or no correlation with the amount of generated second harmonic. Furthermore,

when the crystal was operated at temperatures above the QPM temperature for SHG, no damage occurred even at much higher laser powers. This suggested that the damage was caused by the onset of self-focusing of the pump beam by the cascaded nonlinearity<sup>14,15</sup> that has been discussed extensively in the context of generation of spatial solitons. This process should not occur in MgO:PPLN because third-order QPM SHG requires a substantially different poling period than parametric conversion to the 5–6  $\mu\text{m}$  region.

In conclusion, we proposed corrections to the coefficients of the Sellmeier equation for SLT. The corrected equation allows higher accuracy in poling period calculations for QPM interactions when one of the generated wavelengths is within the 5–6  $\mu\text{m}$  absorption edge. Additionally, we observed interesting damage behavior that was attributed to cascaded nonlinear processes close to the third-order QPM condition for SHG of the pump.

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## References

1. A. C. Chiang, Y. C. Huang, Y. W. Fang, and Y. H. Chen, *Opt. Lett.* **26**, 66 (2001).
2. S. Izumi, M. Sato, J. Suzuki, T. Taniuchi, and H. Ito, *Jpn. J. Appl. Phys. Part 2* **37**, L1383 (1998).
3. Y. Furukawa, K. Kitamura, A. Alexandrovski, R. K. Route, M. M. Fejer, and G. Foulon, *Appl. Phys. Lett.* **78**, 1970 (2001).
4. K. Kitamura, Y. Furukawa, K. Niwa, V. Gopalan, and T. E. Mitchell, *Appl. Phys. Lett.* **73**, 3073 (1998).
5. T. Hatanaka, K. Nakamura, T. Taniuchi, H. Ito, Y. Furukawa, and K. Kitamura, *Opt. Lett.* **25**, 651 (2000).
6. M. Sato, T. Hatanaka, S. Izumi, T. Taniuchi, and H. Ito, *Appl. Opt.* **38**, 2560 (1999).
7. D. J. Bamford, S. J. Sharpe, D. J. Cook, A. Tracy, and C. A. López, in *Technical Digest of Conference on Lasers and Electro-Optics, Quantum Electronics and Laser Science Conference*, Baltimore, Maryland (2005), Paper CFL6.
8. N. E. Yu, S. Kurimura, Y. Nomura, M. Nakamura, K. Kitamura, Y. Takada, J. Sakuma, and T. Sumiyoshi, *Appl. Phys. Lett.* **85**, 5134 (2004).
9. M. Nakamura, S. Higuchi, S. Takekawa, K. Terabe, Y. Furukawa, and K. Kitamura, *Jpn. J. Appl. Phys. Part 2* **41**, L465 (2002).
10. M. Nakamura, S. Higuchi, S. Takekawa, K. Terabe, Y. Furukawa, and K. Kitamura, *Jpn. J. Appl. Phys. Part 2* **41**, L49 (2002).
11. A. Bruner, D. Eger, M. B. Oron, P. Blau, M. Katz, and S. Ruschin, *Opt. Lett.* **28**, 194 (2003).
12. V. Z. Kolev, M. J. Lederer, B. Luther-Davies, and A. V. Rode, *Opt. Lett.* **28**, 1275 (2003).
13. B. Luther-Davies, V. Z. Kolev, M. J. Lederer, N. R. Madsen, A. V. Rode, J. Giesekeus, K.-M. Du, and M. Duering, *Appl. Phys. A* **79**, 1051 (2004).
14. R. DeSalvo, D. J. Hagan, M. Sheik-Bahae, G. Stegeman, E. W. Van Stryland, and H. Vanherzeele, *Opt. Lett.* **17**, 28 (1992).
15. G. I. Stegeman, M. Sheik-Bahae, E. Van Stryland, and G. Assanto, *Opt. Lett.* **18**, 13 (1993).