Temperature-dependent Sellmeier equation for the index of refraction, ne, in congruent lithium niobate

Article *in* Optics Letters · November 1997

DOI: 10.1364/OL.22.001553 · Source: PubMed

1 author:

Dieter Jundt
Gooch & Housego

Temperature-dependent Sellmeier equation for the index of refraction, n_e, in congruent lithium niobate

Dieter H. Jundt

Crystal Technology, Inc., Palo Alto, California 94303

Received July 14, 1997

A Sellmeier equation for the extraordinary index of congruent lithium niobate is derived. The source data for the fit contain previously published data [Opt. Commun. **17,** 322 (1996); erratum **20,** 188 (1997); J. Appl. Phys. **45,** 3688 (1974)] and new measured tuning data for an optical parametric oscillator using periodically poled lithium niobate. Phase-matching predictions are accurate for temperatures between room temperature and 250 °C and wavelengths ranging from 0.4 to 5 μm. © 1997 Optical Society of America

A commonly cited Sellmeier equation[1] for congruent lithium niobate is based on refractive-index measurements at room temperature for wavelengths ranging from 404.6 nm to 3.39 μm (Refs. 2 and 3) as well as on measurements of the temperature dependence of the optical length at two He–Ne wavelengths, 632.8 nm and 3.39 μm.[2] Since electric-field-poled lithium niobate was first demonstrated,[4] interest in using periodically poled lithium niobate (PPLN) for optical applications has been growing steadily. This material offers a wide range of phase-matching possibilities[5] and high nonlinearities. Among the numerous applications are second-harmonic generation (SHG) to blue[6,7] and green[8] wavelengths, IR light generation by difference-frequency generation[9] (DFG), and optical parametric oscillation to produce tunable mid-IR wavelengths.[10–13] It has been observed[13,14] that the Sellmeier equation derived in Ref. 1 is inadequate for predicting phase matching at wavelengths beyond 3.3 μm, particularly at elevated temperatures. This is not surprising since the data used to derive the Sellmeier equation do not extend beyond 3.39 μm. As discussed in Ref. 15, the multiphonon absorption in oxide materials leads to an exponential increase in absorption at wavelengths near the IR cutoff of the transparency range. For lithium niobate, the absorption coefficient at 4 μm is 0.08 cm^{-1} and that at 5 μm is 0.94 cm^{-1}.[13] As the temperature increases, the absorption edge moves toward shorter wavelengths and therefore has an even greater effect on the refractive index in the mid-IR spectral region. The potential applications for trace-gas sensing and countermeasures using mid-IR light generation in PPLN justify a revision of the Sellmeier equations to predict more accurately refractive indices at long wavelengths.

The index of refraction, n_e, in the mid-IR region was measured indirectly by measurement of the signal wavelength of an optical parametric oscillator (OPO) with electric-field-poled lithium niobate as the nonlinear medium. A lamp-pumped, pulsed Nd:YAG laser emitting at 1.064 μm was used to pump a two-mirror cavity with a flat input coupler and a 50-mm radius-of-curvature output coupler. The cavity resonated the signal, which could be tuned from 1.35 to 1.6 μm by changes in the domain grating period and the temperature. The pump beam was focused to a radius of 90 μm at the location of the input coupler to mode match to the cavity modes. Three PPLN crystals, each 19 mm long, with domain grating spacings ranging from 25.5 to 30.2 μm were used for the experiment. The crystals were manufactured from congruently grown lithium niobate with a composition of 48.38 ± 0.01 mol.% Li_2O.[16] Each crystal was placed at the center of 53-mm long cavity. A single layer of SiO_2 on the end faces of the crystals provided losses of less than 2% over the signal-tuning range. The input coupler had a reflectivity exceeding 98% in the wavelength range spanned by the signal, and the output coupling ranged from a minimum of 14% at 1.45 μm to 22% at 1.6 μm. Oscillation identified by the emerging red beam owing to the (non-phase-matched) sum frequency of the signal and the pump beams was easily achieved. The crystal temperature was controlled with 0.1-K stability and absolute accuracy of better than 1.5 K. The wavelength of the peak of the red sum frequency, λ_{red}, as well as the pump wavelength, λ_p, was measured with a 1-m grating spectrometer and a silicon p-i-n diode. A gated integrator with a signal averager processed the signal current from the diode. Data were gathered at six different temperatures ranging from room temperature to 250 °C for all the gratings that yielded signal wavelengths shorter than 1.6 μm. The spectrometer was calibrated with a series of cw lasers that were cross calibrated to a wavemeter with absolute accuracy of 5 parts in 10^6. The absolute wavelength accuracy of the red sum-frequency signal and the pump-wavelength measurements was 0.02 nm. From these measurements the signal and the idler wavelengths can be determined, since they are related to the pump wavelength according to the equations

$$1/\lambda_{\text{red}} = 1/\lambda_p + 1/\lambda_s, \qquad 1/\lambda_p = 1/\lambda_s + 1/\lambda_i\,. \quad (1)$$

The highest conversion occurs at the center of the phase-matching peak, where the additional condition

$$\Delta k = 2\pi\left(\frac{n_p}{\lambda_p} - \frac{n_s}{\lambda_s} - \frac{n_i}{\lambda_i} - \frac{1}{\Lambda}\right) = 0 \qquad (2)$$

applies. n_p is the extraordinary refractive index at the pump wavelength, and n_s and n_i are the corresponding quantities for the signal and the idler waves,

0146-9592/97/201553-03$10.00/0

respectively. The domain grating period is given by Λ. The published index data[2,3] give good estimates of the refractive indices for both the pump and the signal wavelengths. The grating period is lithographically defined and is accurate to 2 parts in 10^6 at room temperature. At elevated temperatures, the crystal expands in the propagation direction, and the grating period increases correspondingly. The thermal-expansion coefficients[17] α and β describe the crystal length l at temperature T normalized to the length of 25 °C, $l_{25^\circ\mathrm{C}}$:

$$l = l_{25^\circ\mathrm{C}}[1 + \alpha(T - 25\,^\circ\mathrm{C}) + \beta(T - 25\,^\circ\mathrm{C})^2], \qquad (3)$$

where $\alpha = 1.54 \times 10^{-5}\ \mathrm{K}^{-1}$ and $\beta = 5.3 \times 10^{-9}\ \mathrm{K}^{-2}$. Thus, by measuring the pump and the signal wavelengths, one can get an estimate of the refractive index at the idler wavelength in the mid IR from Eq. (2). The experimentally determined idler wavelengths as a function of grating period are shown in Fig. 1 for three of the measured temperatures. The values predicted with the Sellmeier equation of Ref. 1 are shown as dashed curves.

The Sellmeier equation of Ref. 1 describes the IR absorption contribution to the refractive index with a single term proportional to the square of the wavelength, an approximation that is valid only for wavelengths that are much shorter than the absorption edge. To describe accurately the measured data, the proposed Sellmeier equation has an added oscillator at wavelength a_5 in the mid-IR spectral region. The temperature dependence of the multiphonon absorption is approximated with the term $b_4 f$:

$$n_e^2 = a_1 + b_1 f + \frac{a_2 + b_2 f}{\lambda^2 - (a_3 + b_3 f)^2} + \frac{a_4 + b_4 f}{\lambda^2 - a_5^2} - a_6 \lambda^2 . \qquad (4)$$

The temperature parameter f is the square of the absolute temperature in degrees Kelvin, with an added offset to make it vanish at the reference temperature $T_0 = 24.5°$. For temperatures T expressed in degrees Celsius, f is given by

$$\begin{aligned} f &= (T - T_0)(T + T_0 + 2 \times 273.16) \\ &= (T - 24.5\,^\circ\mathrm{C})(T + 570.82). \end{aligned} \qquad (5)$$

The OPO data measured in the experiment are not sufficient to permit us to determine all the parameters a_i and b_i. Additional data were included to do this. The refractive-index data at room temperature were taken from Refs. 2 and 3. The temperature dependence of the refractive index at 0.632 and 3.39 μm was calculated from the polynomial expression given in Ref. 2 for temperatures between 0 and 500 °C. To consolidate all the data from different sources, we performed a Levenberg-Marquardt[18] nonlinear fit on all the data simultaneously. This method adjusts the fit parameters a_i and b_i to minimize the sum of the normalized residuals given by

$$\chi^2 = \sum_{n=1}^{N} \left(\frac{y_n - y_{\mathrm{fit}}}{\sigma_n} \right)^2 . \qquad (6)$$

The sum runs over all N data points. y_n is the experimentally determined value of the quantity used in the fit function, y_{fit} is the value of the function numerically determined by the fit parameters, and σ_n is the experimental uncertainty in the value of y_n that is due to the various uncertainties in the measured quantities such as wavelength and temperature. The value of σ_n determines how much an individual data point is weighed in the fitting. Table 1 shows the different forms of the fit function y that have been used to accommodate the various types of measurement. For the case of refractive-index measurements, the indices and the errors used were published in Refs. 2 and 3. To minimize the effect of a potential systematic error in the temperature-dependent index data, we normalized the index to the value at 20 °C. The uncertainty for those data was calculated to be 2.4×10^{-4}, based on the index uncertainty of 5×10^{-4} in Ref. 2. The OPO tuning data as well as three data points for the

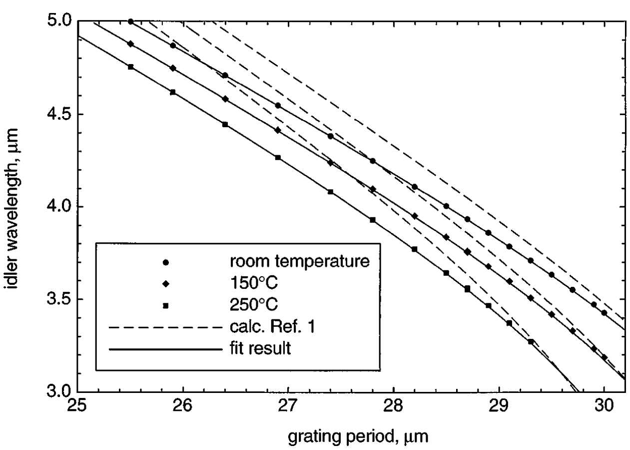

Fig. 1. Idler wavelength tuning of Nd:YAG pumped OPO in PPLN. The dashed curves are calculated with the Sellmeier equation of Ref. 1, and the solid curves use the Sellmeier equation described in this Letter.

Table 1. Fit Function *y* and Data Sets

Data Set	$y(x)$	No. of Points	Source
Refractive-index data at	$n_e(20\,^\circ\mathrm{C}, \lambda)$	5	Ref. 2
room temperature	$n_e(24.5\,^\circ\mathrm{C}, \lambda)$	30	Ref. 3
Normalized index as a function of temperature	$n_e(T, \lambda)/n_e(20\,^\circ\mathrm{C}, \lambda)$	50	Calculated from Ref. 2
OPO tuning data	$\Delta k(T, \lambda_p, \lambda_s, \Lambda_{25^\circ\mathrm{C}})$	104	This Letter
Degenerate OPO data	$\Delta k(T, \lambda_p, 2\lambda_p, \Lambda_{25^\circ\mathrm{C}})$	3	Refs. 19–21

Table 2. Fitted Parameters for Eq. (4)

Parameter	Value
a_1	5.35583
a_2	0.100473
a_3	0.20692
a_4	100
a_5	11.34927
a_6	1.5334×10^{-2}
b_1	4.629×10^{-7}
b_2	3.862×10^{-8}
b_3	-0.89×10^{-8}
b_4	2.657×10^{-5}

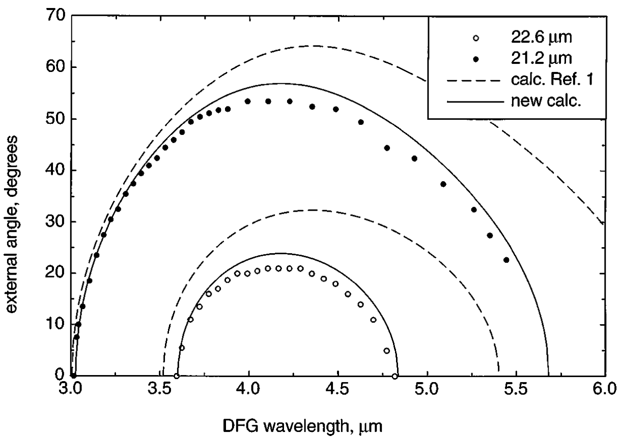

Fig. 2. Angle tuning of PPLN according to Ref. 9. The dashed curves are calculated with the Sellmeier equation of Ref. 1, whereas the solid lines use Eq. (4) with the parameters given in Table 2.

degenerate operation of OPO's from the literature were fitted by use of Eq. (2). The fit target for these points is $y = 0$, and the errors σ were calculated from the measurement uncertainties, which were assumed to be randomly distributed and independent of one another.

For the fit, the parameters of Eq. (4) were allowed to be optimized. It was found that the strength a_4 of the IR pole was not well defined, as there were many different parameter sets with almost identical χ^2 values. We thus kept a_4 constant at 100 μm^2. The resulting χ^2 was 89 for the 192 points fitted, indicating that the data are internally consistent and accurately fitted by the assumed form of the Sellmeier equation. The resulting parameters are shown in Table 2. Tuning curves for a 1.064-μm pumped OPO using the fit results of Table 2 are shown as solid curves in Fig. 1. The agreement is significantly improved over the prediction using the parameters of Ref. 1. To test the usefulness of this new Sellmeier equation at long wavelengths, we recalculated angle-tuning data as measured by Goldberg and Burns[9] at a temperature of 21 °C. In the experiment two angle-tuned PPLN samples with periods of 22.6 and 21.2 μm were used, and DFG between a tunable Ti:sapphire laser and a Nd:YAG laser was performed. Figure 2 shows the original data from Ref. 9 as circles, the calculated phase-matching curves using the Sellmeier equation of Ref. 1 as dashed curves, and the curves calculated with the results of Table 2 as solid curves. The agreement with experimental data is much improved, even at wavelengths as long as 5.4 μm.

In summary, using measurements involving a PPLN OPO and previous refractive-index data,[2,3] the Sellmeier equation was modified to allow for more-accurate refractive-index predictions for n_e in lithium niobate at long wavelengths.

We thank Walt Bosenberg for suggesting this study and supplying the OPO mirrors, Mitch LaBuda for fabrication of the antireflection coatings, Loren Eyres and Mark Abore for help with the experiment, and Lew Goldberg for supplying a copy of his DFG data.

References

1. G. J. Edwards and M. Lawrence, Opt. Quantum Electron. **16,** 373 (1984).
2. D. S. Smith, H. D. Riccius, and R. P. Edwin, Opt. Commun. **17,** 332 (1976); erratum **20,** 188 (1977).
3. D. F. Nelson and R. M. Mikulyak, J. Appl. Phys. **45,** 3688 (1974).
4. M. Yamada and K. Kishima, Electron. Lett. **27,** 828 (1991).
5. M. M. Fejer, G. A. Magel, D. H. Jundt, and R. L. Byer, IEEE J. Quantum Electron. **28,** 2631 (1992).
6. J. Webjörn, V. Pruneri, P. S. J. Russell, and J. R. M. Barr, Electron. Lett. **30,** 894 (1994).
7. L. Goldberg, R. W. McElhanon, and W. K. Burns, Electron. Lett. **31,** 1576 (1995).
8. J. Webjörn, V. Pruneri, P. St. J. Russell, and D. C. Hanna, Electron. Lett. **31,** 669 (1995).
9. L. Goldberg and W. K. Burns, Appl. Phys. Lett. **67,** 2910 (1995).
10. L. E. Myers, R. C. Eckardt, M. M. Fejer, R. L. Byer, W. R. Bosenberg, and J. W. Pierce, J. Opt. Soc. Am. B **12,** 2102 (1995).
11. W. R. Bosenberg, A. Drobshoff, J. I. Alexander, L. E. Myers, and R. L. Byer, Opt. Lett. **21,** 1336 (1996).
12. W. R. Bosenberg, A. Drobshoff, J. I. Alexander, L. E. Myers, and R. L. Byer, Opt. Lett. **21,** 713 (1996).
13. L. E. Myers, R. C. Eckardt, M. M. Fejer, R. L. Byer, and W. R. Bosenberg, Opt. Lett. **21,** 591 (1996).
14. G. Hansson and D. D. Smith, in *Advanced Solid State Lasers*, C. R. Pollock and W. R. Bosenberg, eds., Vol. 10 of Topics in Optics and Spectroscopy Series (Optical Society of America, Washington, D.C., 1997), p. 238.
15. M. E. Thomas, R. I. Joseph, and W. J. Tropf, Appl. Opt. **27,** 239 (1988).
16. P. F. Bordui, R. G. Norwood, C. D. Bird, and G. D. Calvert, J. Cryst. Growth **113,** 61 (1991).
17. Y. S. Kim and R. T. Smith, J. Appl. Phys. **40,** 4637 (1969).
18. W. H. Press, B. P. Flannery, S. A. Teukolsky, and W. T. Vettering, *Numerical Recipes* (Cambridge U. Press, Cambridge, 1986).
19. L. E. Myers, R. C. Eckardt, M. M. Fejer, R. L. Byer, and J. W. Pierce, Electron. Lett. **13,** 1869 (1995).
20. L. E. Myers, G. D. Miller, R. C. Eckardt, M. M. Fejer, and R. L. Byer, Opt. Lett. **20,** 52 (1995).
21. G. D. Miller, R. G. Batchko, W. M. Tulloch, D. R. Weise, M. M. Fejer, and R. L. Byer, in *Conference on Lasers and Electro-Optics*, Vol. 11 of OSA Technical Digest Series (Optical Society of America, Washington, D.C., 1997), p. 58.