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A GENERALIZED SELLMEIER EQUATION FOR THE REFRACTIVE INDICES OF LITHIUM NIOBATE

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Abstract A temperature dependent generalized Sellmeier equation is proposed which takes into account the defect structure of stoichiometrically varying lithium niobate. The physical assumptions leading to the equation are briefly discussed. On the basis of this generalized Sellmeier equation all refractive index dependent effects in lithium niobate can be calculated in the wavelength range 400 - 1200 nm, the composition range 46 - 50 mol% Li content and the temperature range 50 - 600 K. The phase matching conditions for nonlinear effects like second harmonic generation and optical parametric oscillation are calculated. Excellent agreement is found with the respective experimental values.

INTRODUCTION

Although commonly referred to as $LiNbO_3$, lithium niobate can be fabricated over a relatively wide composition range. Since many applications depend on the refractive indices, a precise description of the refractive indices of $LiNbO_3$ as a function of the composition is of great interest. Here we propose a Sellmeier equation being a function of the three independent parameters composition, wavelength and temperature. With this generalized temperature dependent Sellmeier equation it is possible to calculate the conditions for technologically important effects like second harmonic generation and optical parametric oscillation.

EXPERIMENTAL

For the measurement of the refractive indices we applied an interferometric technique consisting of a monochromatically illuminated Michelson-type interferometer with a parallel-plate sample in one arm.[1] The sample is rotated around an axis parallel to the c-axis of the crystal and perpendicular to the incident beam, causing therefore a rotation angle dependent shift in the optical pathlength difference. The interferogram is evaluated with appropriate numerical fit procedures, yielding an accuracy in the absolute refractive index of about $\Delta n = 5 \times 10^{-4}$ for samples of good optical quality.[2]

We measured six samples with compositions ranging from 46.9 to 49.9 mol% Li_2O in a wavelength range from 400 to 1200 nm.

GENERALIZED SELLMEIER EQUATION

In contrast to several other authors who give a description of the refractive index only at a fixed composition [3, 4, 5] we propose a generalized Sellmeier equation which takes into account the defect structure of Li-deficient $LiNbO_3$. This leads to four oscillator terms:

1. Nb on Nb site, or more precisely the NbO_6 octahedron, governs the optical properties in the near UV region.[6] According to Abrahams and Marsh [7] the occupancy of the Nb site decreases with decreasing Li content.
2. Nb replaces each missing Li atom on its site in Li deficient $LiNbO_3$.[7] The corresponding oscillator term depends therefore linearly on the Li deficit.
3. Reststrahl absorption contributes to the refractive index in the IR region.[5] This small term is assumed to be independent of composition.
4. Plasmons in the 13 to 25 eV region[8] represent a contribution which is independent of polarization because the optical anisotropy in $LiNbO_3$ primarily arises from transitions below 10 eV.[6]

Here only the first two oscillator terms are assumed to be temperature dependent. Since the corresponding transitions are excitations from upper valence band states into lower conduction band states,[8] the temperature variation of the resonance wavelengths can be assumed to be proportional to the temperature dependence of the energy gap.[11, 12]

The refractive indices of $LiNbO_3$ can therefore be excellently described by a generalized Sellmeier equation of the form [13]

$$n_i^2 = \frac{50 + c_{\mathrm{Li}}}{100} \frac{A_{0,i}}{(\lambda_{0,i} + \mu_{0,i}F)^{-2} - \lambda^{-2}} + \frac{50 - c_{\mathrm{Li}}}{100} \frac{A_{1,i}}{(\lambda_{1,i} + \mu_{1,i}F)^{-2} - \lambda^{-2}} - A_{IR,i}\lambda^2 + A_{UV}$$

$$F = f(T) - f(T_0) \; ; \qquad f(T) = (T+273)^2 + 4.0238 \times 10^5 [\coth(\frac{261.6}{T+273}) - 1] \; ;$$

c_{Li} in mol% Li_2O ; λ in nm ; T in °C ; $T_0 = 24.5\,$°C ; $i = e, o$.

TABLE 1: Parameters of the generalized Sellmeier equation.

n_o	n_e
$A_{0,o} = 4.5312 \times 10^{-5}$	$A_{0,e} = 3.9466 \times 10^{-5}$
$\lambda_{0,o} = 223.219$	$\lambda_{0,e} = 218.203$
$A_{1,o} = 2.7322 \times 10^{-5}$	$A_{1,e} = 8.3140 \times 10^{-5}$
$\lambda_{1,o} = 260.26$	$\lambda_{1,e} = 250.847$
$A_{IR,o} = 3.6340 \times 10^{-8}$	$A_{IR,e} = 3.0998 \times 10^{-8}$
$A_{UV} = 2.6613$	$A_{UV} = 2.6613$
$\mu_{0,o} = 2.1203 \times 10^{-6}$	$\mu_{0,e} = 7.5187 \times 10^{-6}$
$\mu_{1,o} = -1.827 \times 10^{-4}$	$\mu_{1,e} = -3.8043 \times 10^{-5}$

Here $i = e$ refers to the extraordinary, $i = o$ to the ordinary refractive index. The room temperature parameters $\lambda_{0,i}$, $\lambda_{1,i}$, $A_{0,i}$, $A_{1,i}$, $A_{IR,i}$ and A_{UV} are fitted to our measured refractive index data. The coefficients for the temperature dependence of the refractive index $\mu_{0,i}$ and $\mu_{1,i}$ are obtained by a fit to temperature dependent literature data for congruent[4] and stoichiometric[3] $LiNbO_3$. The numerical results for all parameters are given in table 1.

NONLINEAR EFFECTS IN LITHIUM NIOBATE

Spontaneous noncolinear frequency doubling. In this technique[9, 14] an ordinary polarized intense laser beam (wavelength λ_1, k-vector $\vec{k}_1$), its Rayleigh scattered light (λ_1, $\vec{k}'_1$) and the extraordinary polarized second harmonic light ($\lambda_2 = \lambda_1/2$, $\vec{k}_2$) which forms an elliptic cone around the incident beam must obey the vectorial phase matching condition $\vec{k}_2 = \vec{k}_1 + \vec{k}'_1$. Defining φ_{cryst} as the angle between $\vec{k}_1$ and $\vec{k}_2$ and in a plane normal to the optic axis this condition can be written as $\cos\varphi_{cryst} = n_e(\lambda_1/2)/n_o(\lambda_1)$. Applying Snellius law one obtains the respective angle φ_{air} outside the crystal.

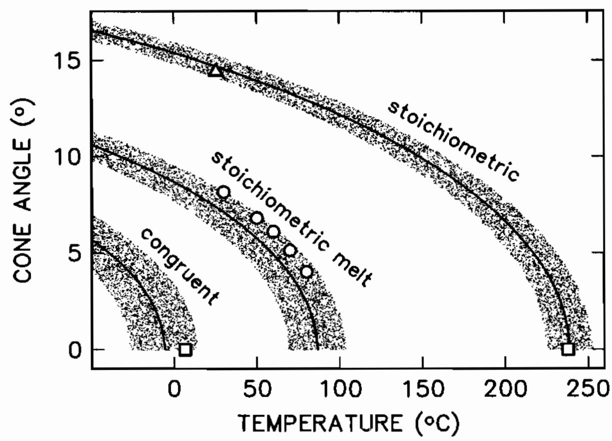

FIGURE 1: Cone angle for spontaneous noncolinear frequency doubling as a function of the temperature. The angle is measured outside the crystal in a plane normal to the optic axis. The curves are calculated from the generalized Sellmeier equation (see text) for crystals grown from congruent melt (about 48.4 mol% Li_2O),from stoichiometric melt (solid composition about 48.9 mol%) and VTE equilibrated material (about 49.9 mol%). The grey areas correspond to a variation of ±0.1 mol% Li_2O in the composition. The data points represent our measurement (△) and results from Bates [9] (○) and Byer *et al.*[10] (□).

In figure 1 the cone angle φ_{air} for a fundamental wavelength of $\lambda_1 = 1064$ nm is calculated as a function of temperature for several crystal compositions. For a cone angle of 0° the condition reduces to $n_e(\lambda_1/2) = n_o(\lambda_1)$, the corresponding temperature at this point is therefore identical to the phase matching temperature for type I noncritical second harmonic generation. Within an uncertainty in the composition of ±0.1 mol% Li_2O our measurement and the results of other authors[9, 10] agree well with these calculations.

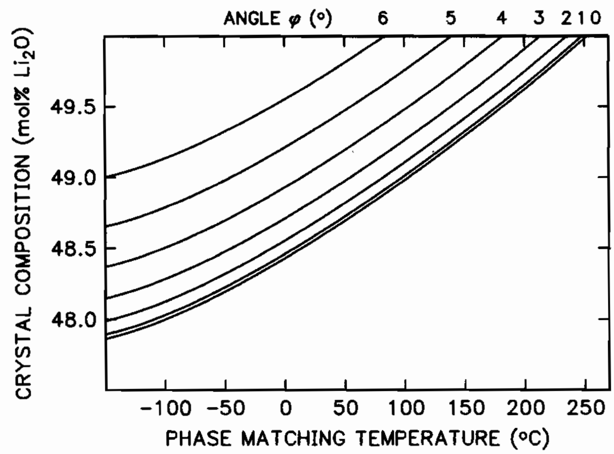

FIGURE 2: Calibration curves for the crystal composition of lithium niobate as a function of the phase matching temperature T^{INCFD} for induced noncolinear frequency doubling. The two fundamental beams are inclined to each other under the angle 2φ inside the crystal.

Induced noncolinear frequency doubling. The method of induced noncolinear frequency doubling (INCFD) uses two intense laser beams which are crossed inside the crystal under an angle $2\varphi_{cryst}$. This newly presented method[14] has the advantage that the interaction volume within the sample is limited in all three spatial direction. This allows to perform a three dimensional topographic inspection of the crystal by the measurement of the noncolinear phase matching temperature T^{INCFD} at any position in the crystal.[14] In figure 2 the calibration curves for the crystal composition as a function of the phase matching temperature T^{INCFD} are calculated for a fundamental wavelength of $\lambda_1 = 1064$ nm and several angles φ_{cryst}.

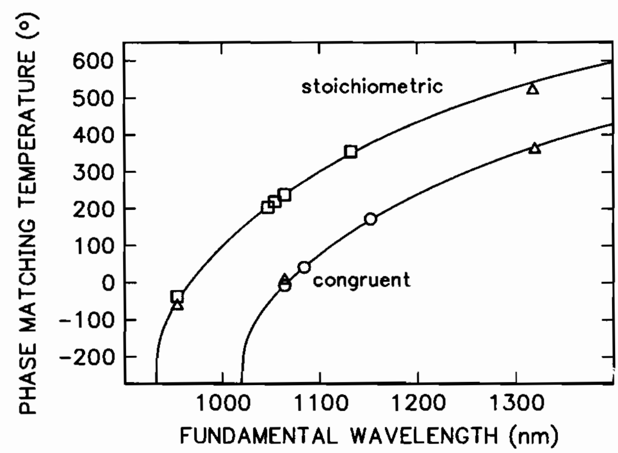

FIGURE 3: Phase matching temperature for noncritical type I second harmonic generation in lithium niobate as a function of the fundamental wavelength. The curves are calculated from the generalized Sellmeier equation (see text) for congruent (about 48.4 mol% Li_2O) and stoichiometric (about 49.9 mol%) material. The data points represent results from Bordui *et al.*[10] (△), Luh *et al.*[15] (□), and Byer *et al.*[16] (○).

Phase matching temperature. The application of the Sellmeier equation on the phase matching condition $n_e(\lambda_1/2) = n_o(\lambda_1)$ for colinear noncritical type I second harmonic generation allows for a given Li content to compute the phase matching temperature T_{PM} as a function of the fundamental wavelength. This is done for congruent and stoichiometric $LiNbO_3$ in figure 3. The experimental results of several other authors reveal excellent correspondence within an extremely wide temperature range from about -100 to at least 400 °C.

Parametric oscillation and difference frequency mixing. As a further test for our generalized Sellmeier equation, we calculated the phase matching temperatures for parametric oscillation in vapour transport equilibrated $LiNbO_3$ (49.9 mol% Li_2O). A comparison with the experimental results from Jundt *et al.*[3] is shown in figure 4. Edward and Lawrence [4] measured the phase matching temperature for difference frequency generation in almost congruent $LiNbO_3$ (about 48.6 mol% Li_2O) for a pump wavelength of 488 nm. The comparison with our calculations in figure 4 for the respective experimental configuration reveals execellent correspondence also.

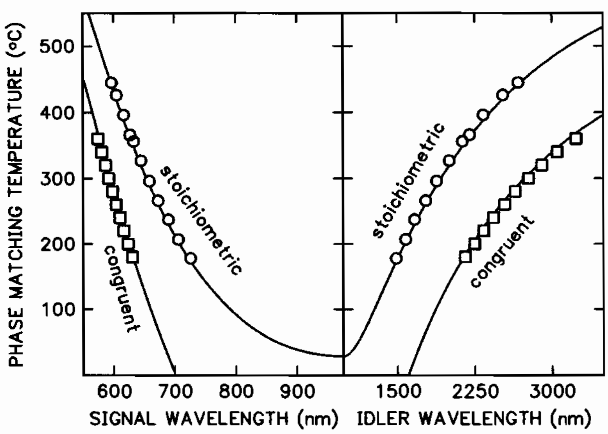

FIGURE 4: Phase matching temperature for optical parametric oscillation as a function of the signal and idler wavelength. The curves are calculated from the generalized Sellmeier equation (see text) for almost congruent (about 48.6 mol% Li_2O) and stoichiometric (about 49.9 mol%) material for a pump wavelength of 488nm. The data points represent results from Edwards and Lawrence[4] (□) and Jundt *et al.*[3] (○).

CONCLUSION

We propose a generalized Sellmeier equation which describes the refractive indices of lithium niobate as a function of composition, wavelength, and temperature. The equation consists of two approximated oscillator terms for infrared and plasmonic contributions and two terms representing Nb on Nb site and Li site. Only the latter two terms are composition and temperature dependent. The temperature variation is assumed to be proportional to the shift of the UV absorption edge with temperature. The parameters of this equation were fitted to our refractive index measurements and literature data. The error in the calculated refractive indices is less than 0.005 in the composition range from 47 to 50 mol% Li_2O , in the wavelength range from 400 to 1200nm and for temperatures between 50 and at least 600K. For room temperature the error is less than 0.001.

With this Sellmeier equation the phase matching conditions for nonlinear effects like second harmonic generation and parametric oscillation can be calculated with excellent accuracy.

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REFERENCES

1. M.S.Shumate. Applied Optics **5**, 327 (1966).
2. U.Schlarb and K.Betzler. Ferroelectrics **126**, 39 (1992).
3. D.H.Jundt, M.M.Feijer, and R.L.Byer. IEEE J. Quantum Electron. **26**, 135 (1990).
4. G.J.Edwards and M.Lawrence. Optic. Quant. Electron. **16**, 373 (1984).
5. M.V.Hobden and J.Warner. Phys. Lett. **22**, 243 (1966).
6. E.Wiesendanger and G.Güntherodt. Solid State Commun. **14**, 303 (1974).
7. S.C.Abrahams and P.Marsh. Acta Cryst. **B42**, 61 (1986).
8. A.M.Mamedov. Opt. Spectrosc. (USSR) **56**, 645 (1984).
9. H.E.Bates. J. Opt. Soc. Am. **61**, 904 (1971).
10. P.F.Bordui, R.G.Norwood, D.H.Jundt, and M.M.Fejer. J. Appl. Phys. **71**, 875 (1992).
11. A.Manoogian and J.C.Woolley. Can. J. Phys. **62**, 285 (1984).
12. D.Redfield and W.J.Burke. J. Appl. Phys. **45**, 4566 (1974).
13. U.Schlarb and K.Betzler. to be published, (1993).
14. A.Reichert and K.Betzler. Ferroelectrics **126**, 9 (1992).
15. Y.S.Luh, M.M.Fejer, R.L.Byer, and R.S.Feigelson. J. Cryst. Growth **85**, 264 (1987).
16. R.L.Byer, J.F.Young, and R.S.Feigelson. J. Appl. Phys. **41**, 2320 (1970).