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SHG PHASE MATCHING CONDITIONS FOR UNDOPED AND DOPED LITHIUM NIOBATE

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We measured the refractive indices of undoped and Zn- or Mg-doped lithium niobate in the wavelength range 400 to 1200 nm. The results are described by a generalized Sellmeier equation which takes into account the defect structure of the material. From the generalized Sellmeier equation a simple relation between the phase matching temperature for noncritical type I second harmonic generation (SHG) and the internal critical phase matching angle is derived. Good correspondence with experimental data over a wide composition and fundamental wavelength range is observed.

Key words: $LiNbO_3$, refractive index, nonlinear effects.

1 INTRODUCTION

Lithium niobate can be fabricated with variable Li_2O contents (46–50 mol%) and additional dopants. For Mg and Zn doping an abrupt change of several physical properties at a certain doping level (threshold concentration) is observed.[1,2] Since many optical 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 importance. Here we use a Sellmeier equation with four independent variables—Li content, dopant concentration (Mg or Zn), wavelength and temperature[3]—to derive a simple relation between the phase matching temperature for noncritical type I second harmonic generation and the phase matching angle.

2 EXPERIMENTAL

We measured the refractive indices of undoped and doped lithium niobate by an interferometric technique[4] in a wavelength range from 400 to 1200 nm. Five samples were grown by the Czochralski technique from melts with varying Li content and were characterized by means of their Curie temperature.[5] Stoichiometric material was prepared with K_2O in the melt[6] or by the vapour transport equilibration (VTE) technique.[7] The doped samples were grown from a congruent melt with up to 9 mol% XO (X = Mg, Zn). The Mg concentration in the crystals was determined applying literature data,[8] the Zn content was measured using an electron microprobe.[9]

3 RESULTS

The refractive indices are excellently described by our generalized Sellmeier equation

$$T_{PM} = [\ (1067)^2 + 3*10^6 \cos^2\theta_{PM}\ ]^{1/2} - 1042$$

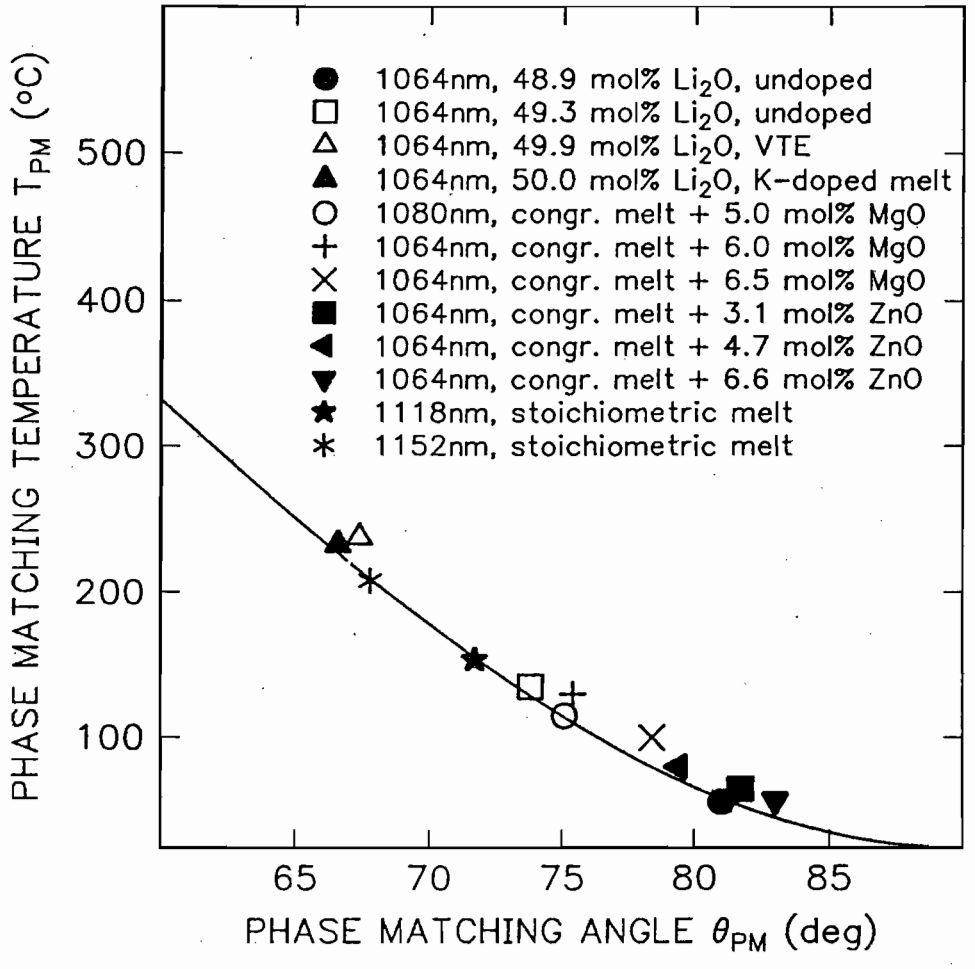

FIGURE 1 Phase matching temperature T_{PM} for noncritical type I second harmonic generation as a function of the internal phase matching angle θ_{PM}. The curve is derived from the generalized Sellmeier equation (see text). Data points represent our measurements and results reported by other authors.[13,14] Fundamental wavelength and crystal composition for each data point are listed in the inset.

which takes into account the defect structure of Li-deficient and Mg- or Zn-doped lithium niobate.

The equation consists of several oscillator terms representing the contribution from the NbO_6-octahedron ($A_{0,i}/(\lambda_{0,i}^{-2}-\lambda^{-2})$) and approximated contributions from the IR and far UV region ($A_{IR,i}\ \lambda^2 + A_{UV}$). The temperature dependence of the resonance wavelength is assumed to be proportional to the temperature dependence of the band gap $\lambda_{0,i}(T) = \lambda_{0,i} + \mu_{0,i}(f(T)-f(T_0))$, where $f(T)$ can be derived by an expression obtained by Manoogian and Woolley.[10] Since the oscillator strength is proportional to the number of oscillators per

volume, we approximate the contributions from Nb antisite defects and from the dopant X (X = Mg, Zn) by $A_{Nb_{Li},i}c_{Nb_{Li}}/(\lambda_{0,i}^{-2}-\lambda^{-2})$ and $A_{X,i}c_X/(\lambda_{0,i}^{-2}-\lambda^{-2})$, respectively. For a detailed derivation the reader is referred to Schlarb and Betzler.[3] The generalized Sellmeier equation is given by

$$n_i^2 = \frac{A_0, i + A_{Nb_{Li},i}c_{NB_{Li}} + A_{X,i}c_X}{(\lambda_{0,i} + \mu_{0,i}F)^{-2} - \lambda^{-2}} - A_{IR,i}^2 + A_{UV} \tag{1}$$

with

$$c_{Nb_{Li}} = \begin{cases} \frac{2}{3}(50 - c_{Li}) - c_X/\alpha_X & \text{for } c_X < \alpha_X\frac{2}{3}(50 - c_{Li}) \\ 0 & \text{for } c_X \geq \alpha_X\frac{2}{3}(50 - c_{Li}) \end{cases}$$

$$\alpha_{Mg} = 5.0;\ \alpha_{Zn} = 6.5;$$

$$F = f(T) - f(T_0), T_0 = 24.5°C;$$

$$f(T) = (T + 273)^2 + 4.0238 \times 10^5[\coth\left(\frac{261.6}{T + 273}\right) - 1].$$

c_X is the dopant concentration given in mol% XO (X = Mg, Zn), c_{Li} denotes the initial Li content, i.e., the ratio $[Li_2O]/([Nb_2O_5]+[Li_2O])$ extrapolated to undoped material (measured in mol% Li_2O). The parameters α_{Mg} and α_{Zn} (which are proportional to the so-called threshold concentration) were estimated from empirical results reported by Schmidt *et al.*[11] and Volk *et al.*[2] for Mg- and Zn-doped material, respectively. The wavelength λ is given in nm, T in °C, and $i = e$ denotes the extraordinary, $i = o$ the ordinary light polarization.

The parameters for equation (1) were calculated by a fit to our measured refractive index data for undoped and Mg- or Zn-doped lithium niobate and temperature dependent literature data.[12,7] The standard deviation was $\Delta n = 2 \times 10^{-3}$. Numerical results for the parameters are listed in Table I.

Table I
Parameters of the generalized Sellmeier equation. For the definition see Eq. (1) in the text.

n_o		n_e	
$\lambda_{0,o}$	$= 223.219$	$\lambda_{0,e}$	$= 218.203$
$\mu_{0,o}$	$= 1.1082 \times 10^{-6}$	$\mu_{0,e}$	$= 6.4047 \times 10^{-6}$
$A_{0,o}$	$= 4.5312 \times 10^{-5}$	$A_{0,e}$	$= 3.9466 \times 10^{-5}$
$A_{Nb_{Li},o}$	$= -7.2320 \times 10^{-8}$	$A_{Nb_{Li},e}$	$= 11.8635 \times 10^{-7}$
$A_{Mg,o}$	$= -7.3548 \times 10^{-8}$	$A_{Mg,e}$	$= 7.2643 \times 10^{-8}$
$A_{Zn,o}$	$= 6.7963 \times 10^{-8}$	$A_{Zn,e}$	$= 1.9221 \times 10^{-7}$
$A_{IR,o}$	$= 3.6340 \times 10^{-8}$	$A_{IR,e}$	$= 3.0998 \times 10^{-8}$
A_{UV}	$= 2.6613$	A_{UV}	$= 2.6613$

The generalized Sellmeier equation allows us to derive a simple relation between the phase matching temperature T_{PM} for colinear noncritical type I second harmonic generation and the internal phase matching angle θ_{PM} measured at room temperature.

Defining $\hat{n}_i = n_i(c_{Li}, c_X, \lambda, T_0)$ and $\tilde{n}_i = n_i(c_{Li}, c_X, \lambda/2, T_0)$ as the refractive indices for the fundamental (λ) and harmonic ($\lambda/2$) wavelengths at room temperature, respectively, the temperature phase matching condition $n_o(c_{Li}, c_X, \lambda, T_{PM}) = n_e(c_{Li}, c_X, \lambda/2, T_{PM})$ can be written as

$$(\tilde{n}_e^2 - \hat{n}_o^2) + F\frac{\partial(\tilde{n}_e^2 - \hat{n}_o^2)}{\partial F}\,|_{F=0} = 0, \tag{2}$$

and the parameter F is approximated with $\coth x \approx 1/x$ yielding

$$F \approx (T_{\mathrm{PM}} + T_s)^2 - (T_s + T_0)^2; T_s = 1042. \tag{3}$$

The angle phase matching condition $\hat{n}_o^{-2} = \tilde{n}_o^{-2}\cos^2\theta_{\mathrm{PM}} + \hat{n}_e^{-2}\sin^2\theta_{\mathrm{PM}}$ can be written as

$$\tilde{n}_e^2 - \hat{n}_o^2 = \cos^2\theta_{\mathrm{PM}}[\hat{n}_o^2\frac{\tilde{n}_e^2}{\hat{n}_o^2} - \hat{n}_o^2] \approx \cos^2\theta_{\mathrm{PM}}[\tilde{n}_e^2 - \tilde{n}_o^2], \tag{4}$$

where θ_{PM} denotes—as already mentioned—the direction of propagation in the crystal with respect to the optical axis. A simple calculation yields

$$T_{\mathrm{PM}} = \sqrt{(T_s + T_0)^2 + \gamma(\lambda)\cos^2\theta_{\mathrm{PM}}} - T_s \tag{5}$$

$$\text{with } \gamma(\lambda) \approx (\tilde{n}_o^2 - \hat{n}_e^2)[\frac{\partial(\tilde{n}_e^2 - \hat{n}_o^2)}{\partial F}]^{-1}\,|_{F=0, c_{Li}=50, c_X=0}$$

In the wavelength range 1000–1200 nm the parameter $\gamma(\lambda)$ varies between 2.8×10^6 and 3.2×10^6, we therefore neglect the dispersion and use $\gamma \approx 3 \times 10^6$. In Figure 1 the relation given in Eq. (5) is depicted and compared with experimental data. The results for a variety of crystal compositions and fundamental wavelengths show that the relation is a good approximation for temperatures up to at least 250°C, a maximum deviation of about 20°C is observed. Eq. (5) therefore allows to estimate the phase matching temperature by simply measuring the more easily accessible phase matching angle.

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