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REFRACTIVE INDICES OF ZN/IN–CO–DOPED LITHIUM NIOBATE

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Abstract We measured the refractive indices of Zn/In-co-doped lithium niobate in the wavelength range of 400 to 1200 nm, using samples with 5.5 mol% ZnO and various additional concentrations of In_2O_3. The results can be well described by a generalized Sellmeier equation which takes into account the defect structure of the material. The proposed Sellmeier equation allows the treatment of nonlinear optical effects; calculated phase-matching conditions for second-harmonic generation are in good agreement with experimental results.

INTRODUCTION

Lithium niobate is presently one of the most important materials for optical devices. Usually crystals with a distinct index of refraction are required; often applications are hampered by the photorefractive effect, also known as optical damage [1]. Both the refractive index and the sensitivity to light illumination are influenced by the crystal composition or - to be more specific - by the number of Nb antisites in the crystal [2]. Besides the techniques which improve the stoichiometry without any doping [3, 4, 5], dopants like Mg [1], Zn [6], In [7] and Sc [8] are known to reduce the optical damage. As each of these dopants is known to increase the birefringence of lithium niobate (thus the applicability for nonlinear optics) only up to a certain threshold concentration, it is important to study the effect of co–doping with more than one material.

EXPERIMENTAL DETAILS

For our investigations we choose lithium niobate co–doped with Zn and In, as both of these dopants allow growing crystals of excellent optical quality. The crystals were grown by the Czochralsky technique from a congruent melt (48.5 mol% Li_2O and 51.5 mol% Nb_2O_5) to which ZnO and In_2O_3 was added. Crystals with a fixed ZnO concentration of 5.5 mol% (slightly lower than the threshold value) and In concentrations in the range 0 – 3 mol% were investigated.

For the refractive index measurements we used an interferometric method, recently described in detail [9, 10], which yields an accuracy of about $\Delta n = 5 \times 10^{-4}$, provided the optical homogeneity of the sample is sufficient. Homogeneity tests by means of spatially resolved second harmonic measurements show that the composition varies less than 0.05 mol %, a value well below the composition uncertainty.

RESULTS AND DISCUSSION

Both the ordinary and the extraordinary index of refraction are shown in Fig. 1 for five selected wavelengths in the range 400 - 1200 nm as a function of the additional In content in the crystal. The results can be well described by a generalized Sellmeier equation which takes into account the defect structure of Li-deficient $LiNbO_3$ with a di- or trivalent dopant (Zn or In). For details of the derivation the reader is referred to Schlarb and Betzler [11].

The generalized Sellmeier equation is defined as

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

with

$$\begin{aligned} c_{\mathrm{Nb_{Li}}} &= \begin{cases} \frac{2}{3}(50 - c_{\mathrm{Li}}) - c_{\mathrm{Zn}}/\alpha_{\mathrm{Zn}} - c_{\mathrm{In}}/\alpha_{\mathrm{In}} & \text{if} > 0 \\ 0 & \text{else} \end{cases} \\ \alpha_{\mathrm{Zn}} &= 6.5 \qquad \alpha_{\mathrm{In}} = 1.5 ; \\ F &= f(T) - f(T_0) , \quad T_0 = 24.5°\mathrm{C} ; \\ f(T) &= (T + 273)^2 + 4.0238 \times 10^5 [\coth(\frac{261.6}{T + 273}) - 1] . \end{aligned}$$

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).

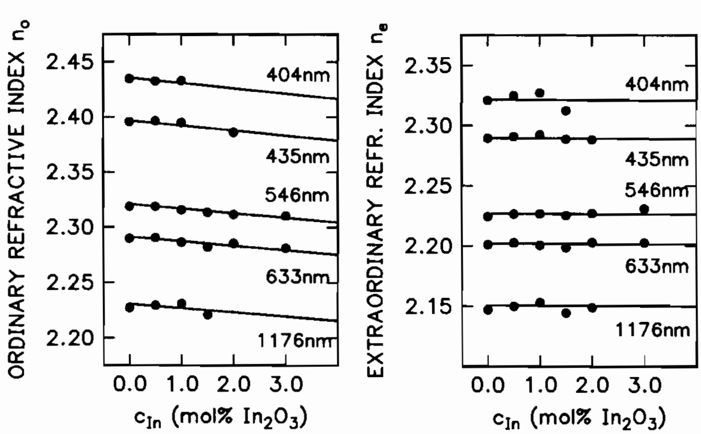

FIGURE 1: Variation of the ordinary (n_o) and extraordinary (n_e) refractive indices of congruently melted $LiNbO_3$ doped with 5.5 mol % ZnO and various In concentrations for selected wavelengths at room temperature. The lines are calculated with the generalized Sellmeier equation, experimental values are represented by dots.

Since all other parameters can be adopted from Refs. 11 and 12, it was only necessary to determine the parameters $A_{In,i}$ by a fit to our refractive index data as a function of wavelength and In concentration at room temperature. The standard deviation is about 1.8×10^{-3}. The numerical results for all parameters of the generalized Sellmeier equation are listed in Table 1.

n_o	n_e
$\lambda_{0,o} = 223.219$	$\lambda_{0,e} = 218.203$
$\mu_{0,o} = 1.1082 \times 10^{-6}$	$\mu_{0,e} = 6.4047 \times 10^{-6}$
$A_{0,o} = 4.5312 \times 10^{-5}$	$A_{0,e} = 3.9466 \times 10^{-5}$
$A_{Nb_{Li},o} = -7.2320 \times 10^{-8}$	$A_{Nb_{Li},e} = 11.8635 \times 10^{-7}$
$A_{Zn,o} = 6.7963 \times 10^{-8}$	$A_{Zn,e} = 1.9221 \times 10^{-7}$
$A_{In,o} = -5.0307 \times 10^{-7}$	$A_{In,e} = -2.3178 \times 10^{-8}$
$A_{IR,o} = 3.6340 \times 10^{-8}$	$A_{IR,e} = 3.0998 \times 10^{-8}$
$A_{UV} = 2.6613$	$A_{UV} = 2.6613$

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

The phase matching for all nonlinear effects is governed by the values of the refractive indices. This allows us to calculate the respective data from the generalized Sellmeier equation. The results for the phase matching temperatures for colinear second harmonic generation and the phase matching angles for colinear and noncolinear second harmonic generation are depicted in Figs. 2 and 3 together with the measured values. The good agreement between calculated and experimental data is obvious.

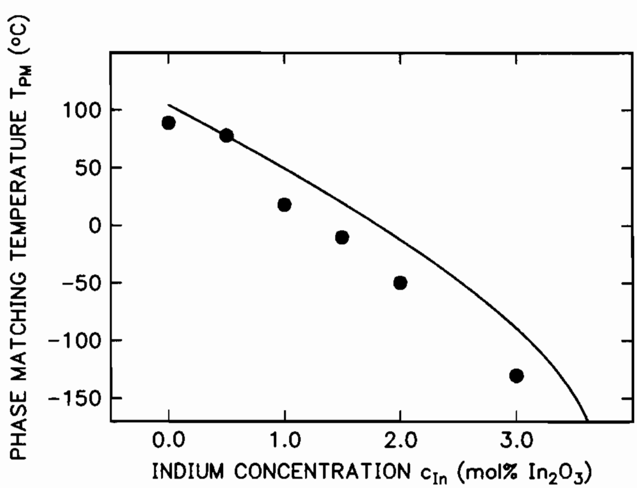

FIGURE 2: Phase-matching temperature of $LiNbO_3$:Zn,In as a function of the indium content in the melt.

As can be derived from the data the additional In doping does not result in a further increase of the optical birefringence but instead has an effect similar to doping beyond the threshold value with only a single dopant.

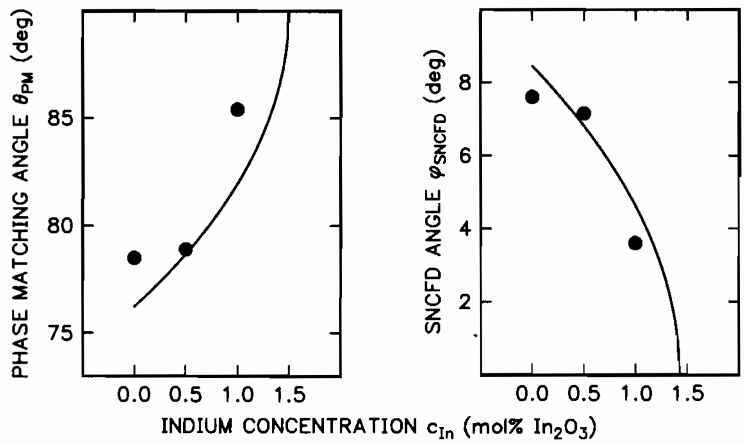

FIGURE 3: Phase-matching angle (left) and conus angle for SNCFD (right) for $LiNbO_3$:Zn,In as a function of indium concentration in the melt.

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