Infrared corrected Sellmeier coefficients for congruently grown lithium niobate and 5 mol.% magnesium oxide –doped lithium niobate

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Infrared corrected Sellmeier coefficients for congruently grown lithium niobate and 5 mol. % magnesium oxide–doped lithium niobate

David E. Zelmon and David L. Small

Materials Directorate, Wright Laboratory (WL/MLPO), Wright-Patterson Air Force Base, Ohio 45433-7707

Dieter Jundt

Crystal Technologies Corporation, 1040 East Meadow Circle, Palo Alto, California 94303

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The growth in the uses of lithium niobate for infrared applications has created a need for knowledge of its optical characteristics in the infrared spectral region for the purpose of designing phase-matched or quasi-phase-matched devices. We report measurements of the refractive indices of congruently grown lithium niobate and lithium niobate doped with 5 mol. % magnesium oxide. We use these results to predict the tuning curve of a room-temperature multigrating optical parametric oscillator in each material. © 1997 Optical Society of America [S0740-3224(97)00612-7]

1. INTRODUCTION

Lithium niobate ($LiNbO_3$) has been used extensively in a wide variety of optical frequency-conversion devices.[1–5] Design of these devices and prediction of their behavior depend critically on precise knowledge of the indices of refraction throughout the spectral region of interest. Although the refractive indices of $LiNbO_3$ have been studied for many years,[6–12] most of the data have been limited to wavelengths in the UV, visible, and near-IR spectral regions. $LiNbO_3$ is now being considered for infrared applications,[13–19] so there is a need for improved knowledge of its refractive indices at wavelengths throughout the transmission range of this material. We have measured the refractive indices of congruently grown $LiNbO_3$ from 0.4 to 5.0 μm and calculated new Sellmeier coefficients that accurately predict refractive indices in this spectral range. With the improved Sellmeier coefficients, we have been able to predict the signal and idler wavelengths for an optical parametric oscillator (OPO) fabricated from periodically poled $LiNbO_3$ that conforms well to experimental observations throughout the spectral region of interest.

$LiNbO_3$ is subject to photorefractive changes in its optical properties, which is intolerable in many applications. Devices have therefore been fabricated with $LiNbO_3$ doped with various amounts of magnesium oxide (MgO). Only limited refractive-index data of any kind are available for this material, and the doping level of the samples and the spectral range over which the data exist vary widely.[20–23] We have measured the refractive indices of $LiNbO_3$ doped with 5 mol. % MgO from 0.4 to 5.0 μm and have calculated new Sellmeier coefficients for this material.

2. EXPERIMENTS

We measured dispersion for both pure and MgO-doped $LiNbO_3$ using the minimum-deviation technique. Crystal prisms were manufactured from poled Czochralski-grown boules of approximate dimensions 80 mm in diameter by 45 mm in length. The growth direction was the ferroelectric Z axis. The melt composition that was used to grow the undoped crystal was congruent (48.38 mol. % lithium oxide),[24] resulting in a crystal of the same composition. The MgO-doped crystal was pulled from a melt of congruent Li/Nb ratio with MgO added to achieve 5.0 mol. % MgO in the melt.[25] The melt fraction crystalized was below 30% in both cases, ensuring high-purity and strain-free optical-quality material.

The prisms were x-ray oriented, cut, and polished by standard techniques. The base faces were Z faces with a 2 min. tolerance. The polished faces were perpendicular to these Z faces. When the prism was aligned for minimum deviation, the light propagation was along the X direction, permitting the measurement of both n_e and n_o. The optical apertures were 15 mm by 13.7 mm (Z). The apex angle was measured optically and was 44.941° for the undoped prism and 44.980° for the MgO-doped prism.

A Gaertner L-124 precision spectrometer was used for the minimum-deviation measurements. We calibrated the spectrometer by measuring indices of a calcium fluoride prism several times and determining the standard deviation. Our data were within 2×10^{-5} of the published values.[26] An Oriel Hg-Xe lamp source was coupled to a Digikrom L 240 monochromator to permit selection of wavelength at which the index was to be measured. The near-IR measurements were made with an Electrophysics

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hand-held IR viewer. For measurements from 2.5 to 5 μm the quartz optics of the spectrometer were replaced by a single ZnSe collimating lens. An IR imaging camera (Cincinnati Electronics IRRIS 160 LN) was used in place of the imaging optics to detect the refracted beam. Five separate runs were made throughout the entire spectrum, and the standard deviation of the data at any wavelength was less than 2×10^{-4}. The temperature at which the measurements were taken was 21 °C.

3. RESULTS

The refractive indices of undoped congruently grown $LiNbO_3$ are shown in Fig. 1, in which the refractive index for both ordinary (n_o) and extraordinary (n_e) waves is plotted versus wavelength. We also measured the refractive indices of congruently grown $LiNbO_3$ that had been doped with 5-mol. % MgO. The results of these measurements are shown in Fig. 2, in which the refractive index for both ordinary and extraordinary waves is plotted versus wavelength.

In order to use the refractive index for phase-matching or quasi-phase-matching calculations, the index data are fit to a Sellmeier equation to predict refractive indices at the wavelengths pertinent to the optical process that was being investigated. In most cases a single oscillator of the form $A\lambda^2/(\lambda^2 - B)$ was used, and we incorporated other oscillators into the Sellmeier equation by assuming that their wavelengths were sufficiently far from the spectral region of interest that they could be approximated by a constant (far-UV) and a term proportional to λ^2 (IR). However, it has been pointed out[27] that these approximations lead to significant deviations from the refractive indices found experimentally, even in the short-wavelength end of the visible spectrum in $LiNbO_3$, and we have found that similar difficulties arise as one approaches the edge of the transmission range in the IR. As a result, we have chosen to use a three-oscillator model to represent our index data.

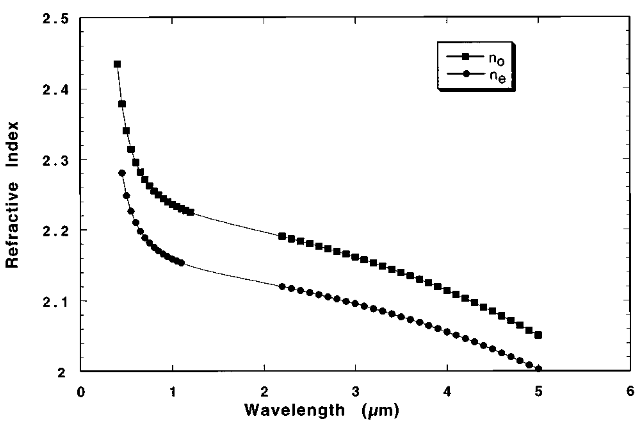

Fig. 1. Refractive indices of undoped congruently grown $LiNbO_3$ as a function of wavelength. The solid curves represent the three-oscillator Sellmeier fit to the data.

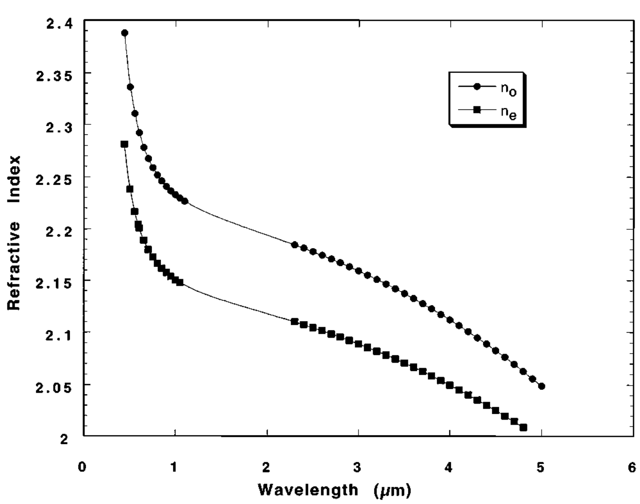

Fig. 2. Refractive indices of congruently grown $LiNbO_3$ doped with 5-mol. % MgO as a function of wavelength. The solid curves represent the three-oscillator Sellmeier fit to the data.

Table 1. Sellmeier Coefficients for Congruently Grown $LiNbO_3$

Coefficient	n_e	n_o
A	2.9804	2.6734
B	0.02047	0.01764
C	0.5981	1.2290
D	0.0666	0.05914
E	8.9543	12.614
F	416.08	474.6

Table 2. Sellmeier Coefficients for Congruently Grown $LiNbO_3$ Doped with 5-mol. % MgO

Coefficient	n_e	n_o
A	2.4272	2.2454
B	0.01478	0.01242
C	1.4617	1.3005
D	0.05612	0.05313
E	9.6536	6.8972
F	371.216	331.33

Multioscillator models for the refractive index of oxide ferroelectrics such as $LiNbO_3$ have been discussed extensively in the literature.[11,12,27,28] On the basis of UV reflectance data[27,29] Uchida[27] proposed a model for oxide ferroelectrics using two UV oscillators corresponding to transitions from the oxygen $2p$ valence band to the $d\varepsilon$ and $d\gamma$ transitions at approximately 5 and 10 eV, respectively. Schlarb and Betzler[11,12] proposed a different model in which they used two closely spaced oscillators in the near UV, which they attributed to the defect structure of the $LiNbO_3$, and a far-UV oscillator, which they approximated as a constant term because the oscillator wavelength was far away from the spectral regions of interest. They used the usual restrahl approximation for the behavior of the refractive index in the IR region. We have found that, because of the wide spectral range of our data, neither of these models is adequate to reproduce the data within the limits of our experimental error. We used a three-oscillator Sellmeier equation of the form[30]

$$n^2 - 1 = A\lambda^2/(\lambda^2 - B) + C\lambda^2/(\lambda^2 - D) + E\lambda^2/(\lambda^2 - F).$$

This equation incorporates two UV oscillators and also an IR oscillator to account for the refractive-index behavior near the IR edge of the transmission range.[31,32] The curve fit parameters are shown in Tables 1 and 2 for the undoped congruent and 5% MgO-doped materials, respectively. The fit that we obtained predicts the data within experimental error throughout the spectral range. The wavelengths of the UV oscillators calculated by the fitting routine are consistent with the transitions observed by Uchida and Mamedov.[26,28] However, the position of the UV oscillators should be considered only approximate, as our data do not extend into the UV region. In the IR the oscillator wavelength and strength are fitting parameters made necessary by the closeness of our data to the IR transmission edge of the material.

The results of the Sellmeier fits for the undoped material are shown in Table 1, and those for the doped material are shown in Table 2. The fit of the calculated indices to the measured indices was excellent, with differences of less than 2×10^{-4} for both ordinary and extraordinary indices at all wavelengths. These Sellmeier coefficients were used in all the subsequent calculations.

4. DISCUSSION

To test the validity of the new Sellmeier equations, we performed calculations to predict the results of a room-temperature frequency-conversion experiment performed on a congruently grown $LiNbO_3$ device. We simulated an experiment performed by Myers *et al.*[16] in which a room-temperature (25 °C) multigrating OPO, fabricated from periodically poled $LiNbO_3$, was pumped at 1.064 μm and the signal was tuned from 1.35 to 1.92 μm by translating the pumping beam over a series of gratings whose periods ranged from 26 to 32 μm. We predicted the tuning curve for phase matching of this process, and the results are shown in Fig. 3, in which we plot the output wavelengths of the OPO, using our Sellmeier equations, and compare them with the experimental observations of Ref. 16. The agreement is excellent between the theoretical predictions and the experimental results throughout the spectral range. The largest deviations occur near the degeneracy point of the OPO because of the extreme sensitivity of the phase-matching curve to small ($<1 \times 10^{-4}$) changes in the refractive index.

There have been few frequency-conversion experiments performed at room temperature on MgO-doped $LiNbO_3$, and most of those have involved doubling of near-IR wavelengths.[33] Although our data predict the results of these experiments well, our study emphasizes the IR spectral region, and almost all the frequency-conversion experiments on MgO-doped $LiNbO_3$ have been performed at an elevated temperature. Although some temperature-dependent refractive-index data on 5% MgO-doped $LiNbO_3$ are available,[15] they are limited mostly to wavelengths in the visible where thermo-optic data are strongly wavelength dependent, making it impossible to extrapolate to longer wavelengths without significant errors. The only other discussion of which we are aware of thermo-optic coefficients near the IR is from Edwards and Lawrence,[10] but because the form of the Sellmeier equation used by them is different from the one that we used it is not obvious how to apply their thermo-optic data to our Sellmeier equation. In the absence of accurate thermo-optic data in the IR, we calculated the grating periods required for a device similar to the one described in Ref. 16 but fabricated from 5% MgO-doped congruently grown $LiNbO_3$. The results of these calculations are shown in Fig. 4. The shape of the tuning curve is similar to that from Ref. 16, but the grating periods are all smaller by ~4 μm.

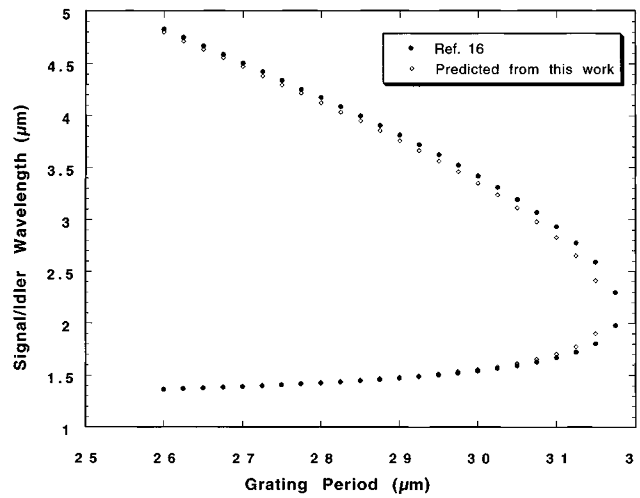

Fig. 3. Signal and idler wavelengths of a 1.064-μm-pumped OPO in periodically poled $LiNbO_3$ as a function of grating period.

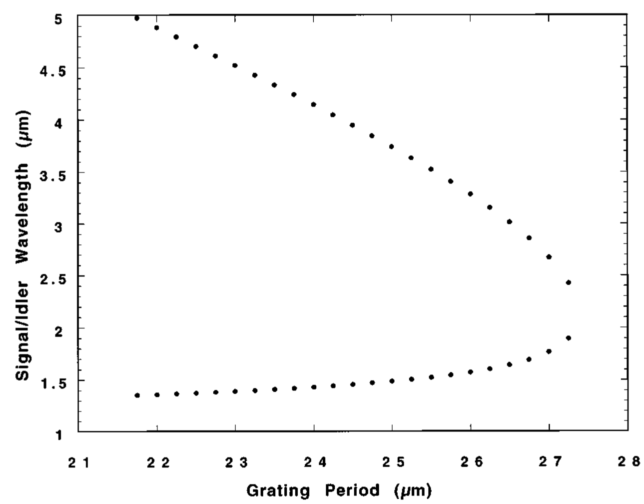

Fig. 4. Predicted signal and idler wavelengths of a 1.064-μm-pumped OPO in 5-mol. % MgO-doped periodically poled $LiNbO_3$ as a function of grating period.

5. CONCLUSION

We have measured the refractive indices of congruently grown $LiNbO_3$ and 5-mol. % MgO-doped $LiNbO_3$ from 0.4 to 5 μm. The modified Sellmeier equations derived from these data give an accurate prediction of experimental results for IR frequency-conversion processes in undoped periodically poled $LiNbO_3$. The data will be useful to designers of devices that will use these materials for IR frequency-conversion devices.

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