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Interferometric Measurement of Refractive Indices of $LiNbO_3$

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Abstract An interferometric method for the determination of refractive indices is described which uses the shift in the optical path length when a parallel plate sample is rotated in one arm of a Michelson-type interferometer. High precision is achieved by including appropriate numerical fit procedures in the evaluation of the experimental results. We could prove that an accuracy of about $2 \cdot 10^{-4}$ can be reached with the method. Sample nonidealities (slight wedge or lens shape) have only moderate effects on the determination of the refractive index. Measurements on $LiNbO_3$ show that not only the extraordinary index of refraction varies with the Li/Nb ratio — which is in good agreement to earlier results —, but also the ordinary one.

INTRODUCTION

For the measurement of the refractive indices of transparent optical materials several methods are applicable. Some of them are restricted to materials with low refractive indices (e. g. Abbe and Pulfrich type refractometers), others exhibit only low accuracy (reflectivity, Bernard, Duc de Chaulnes). Most commonly used for high index materials is the minimum deviation method which is by far the most accurate, but requires the fabrication of appropriate large-aperture prisms.

When — due to other reasons — the fabrication of prisms is not possible or not desirable, the interferometric method can be used. This method measures the shift of the optical pathlength when a plane-parallel sample is rotated in one arm of a two-beam interferometer[1, 2]. Here we describe the application of the method on the measurement of the refractive indices of various $LiNbO_3$ samples. Considerable improvement in the accuracy of the method was achieved by including a numerical fit procedure in the evaluation of the experimental results.

FUNDAMENTALS OF THE METHOD

To measure optical pathlength shifts, two-beam interferometers — either of Mach-Zehnder or of Michelson type — are the most suitable devices. When a sample is inserted in one of the interferometer arms, however, the resulting pathlength shift cannot be measured

directly, as the interference order changes by a large amount. So the method applied here doesn't determine the optical pathlength through the sample but measures the increase in the interference order when the sample is rotated away from normal incidence. For a Michelson type interferometer, the excess interference order $m(\theta)$ as a function of the rotation angle θ is described by

$$m(\theta) = \frac{2dn_a}{\lambda}(\sqrt{n^2/n_a^2 - \sin^2\theta} - \cos\theta - n/n_a + 1) - m_0 \,, \qquad (1)$$

where d is the sample thickness, λ the wavelength used, n and n_a the refractive indices of the sample crystal and the surrounding medium (usually air), respectively, and m_0 the (constant) fractional part of the interference order. This function is shown in fig. 1 for several values of n.

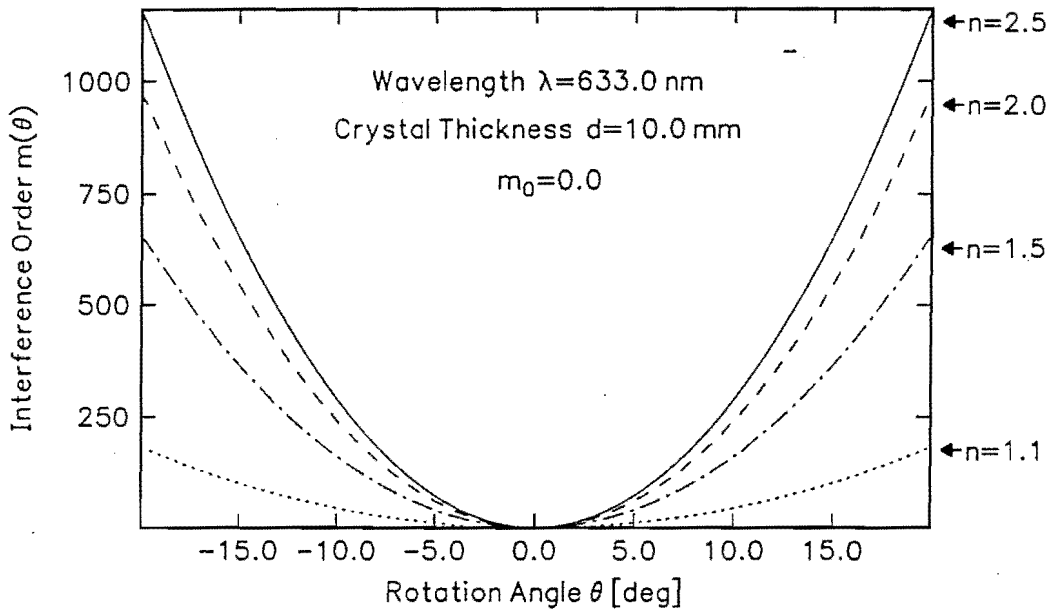

FIGURE 1: Fringe shift function $m(\theta)$ for various refractive indices.

There are two main possibilities to determine the refractive index by this interferometric method:

1. The interferometer is adjusted such that $m_0 = 0$. Then from eq. 1 an expression for n can be derived [1]. The rotation angle for a certain fringe count is measured, from these data n can be calculated.

2. Eq. 1 can be used — as it is — as a basis for a numerical fit procedure using n and m_0 as the fit parameters.

The first method depends very much on the accuracy of adjusting $m_0 = 0$ and on the exact determination of one fringe and the respective angle but has the advantage of

simplicity. The second method requires no additional adjustment. Furthermore, a large number of detected fringes may be included in the fit algorithm thus improving the accuracy. In spite of the drawback of the fit algorithm to be added, we used the second method because of the obviously higher accuracy.

EXPERIMENTAL

For our measurements we used a Michelson type interferometer as it yields twice the path-length inside the sample compared to a Mach-Zehnder type and it reverts lateral beam shifts caused by the sample. As light source served a Helium-Neon laser. The sample was rotated by means of a motor-driven rotation table, the rotation angle was measured by a highly accurate incremental encoder with an angular resolution of about $5 \cdot 10^{-4}\,°$. The output signal of the interferometer was detected by a silicon photodiode, this signal and the encoder signal were sampled by a personal computer. With this setup a very precise measurement of the fringe intensity function was possible. From this function, the maxima and minima were derived using a third order polynomial fit in the respective region. All fringe maxima and minima must be uniquely described by eq. 1, so the best values for n and m_0 can be found by minimizing the quadratic error

$$\sigma^2 = \sum_{i=1}^{N} \left[\frac{2dn_a}{\lambda} \left(\sqrt{n^2/n_a^2 - \sin^2\theta_i} - \cos\theta_i - n/n_a + 1 \right) - m_0 - m_i \right]^2 \qquad (2)$$

m_i, θ_i are the experimentally determined index–angle pairs for the fringe maxima and minima. The function σ^2 for a typical measurement is sketched in fig. 2.

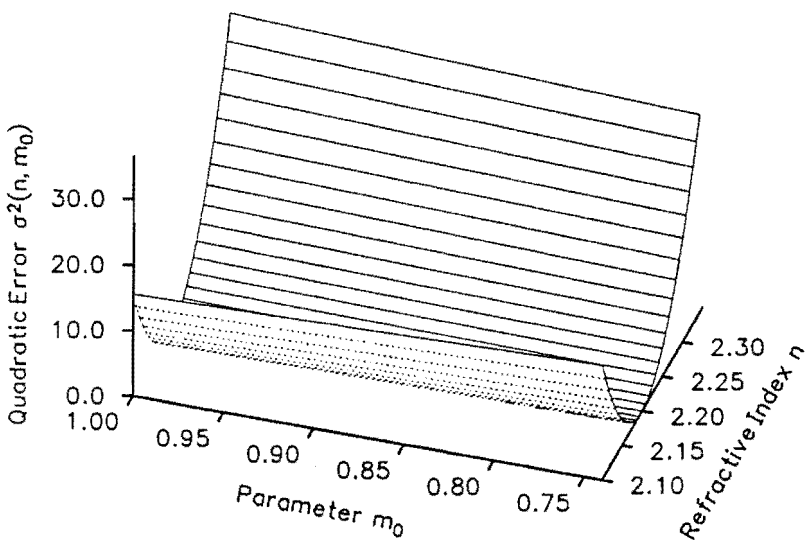

FIGURE 2:
Three-dimensional plot of the quadratic error as a function of the fit parameters n and m_0. The experimental values used were measured on a $LiNbO_3$ crystal with 48.85 % Li_2O.

The simplicity of the function σ^2 allows that practically any standard algorithm may be used to find the minimum. We transformed the problem to a one-dimensional one by eliminating m_0 from eq. 2. The minimum of the resulting function — and the corresponding value of n — can then be quickly found using a first derivative method.

One of the limitations of the method was found to be the uncertainty in the measurement of the sample thickness d. To avoid this limitation, we tried to handle d as an additional fit parameter. This improved the results in theoretical simulations but not in practical measurements which indicates that d and n are too much interdependent (optical pathlength).

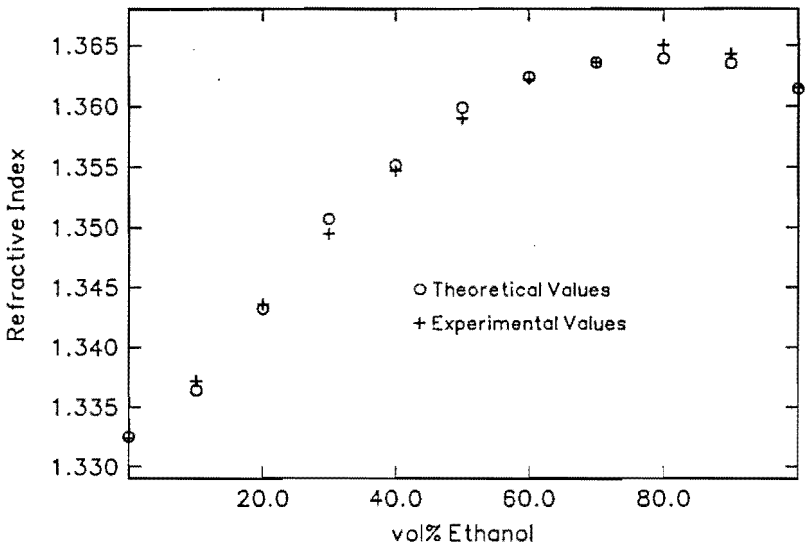

FIGURE 3: Refractive indices measured(+) and calculated(o) for water-ethanol mixtures. The parabolic dependence is due to the volume changes.

We tested our method on water-ethanol mixtures which were measured in a plane-parallel cuvette. The refractive indices of such mixtures can be calculated with good accuracy from the refractive indices of the two constituents taking into account volume changes [3]. The results are shown in fig. 3, excellent agreement is found between calculated and measured refractive indices. The differences correspond to the uncertainties in the values for the volume changes. From these results a maximum error of the method of $5 \cdot 10^{-4}$ for low refractive index materials can be estimated. The mean error is less than $2 \cdot 10^{-4}$. These are the typical errors for single measurements, multiple measurements yield a further error reduction.

RESULTS AND DISCUSSION

The described method was applied to measure the refractive indices of $LiNbO_3$ as a function of the Li content. Various samples with Li contents between 46.9 and 48.8 % [4] were included in the measurements. The crystals had a typical size of 10*10*10 mm^3, the sample thickness was measured mechanically with an accuracy of about 1 μm. As rotation axis the polar axis of the samples was chosen, thus — depending on the light polarization — n_o or n_e could be determined. The results are shown in fig. 4. Both refractive indices vary with Li content. The extraordinary one decreases with increasing Li content by about 0.02 in the concentration range considered, the ordinary one shows an increase by about 0.004.

Taking into account all experimental errors as well as the abberations brought in by a nonideal sample shape we could estimate the maximum uncertainty of the refractive

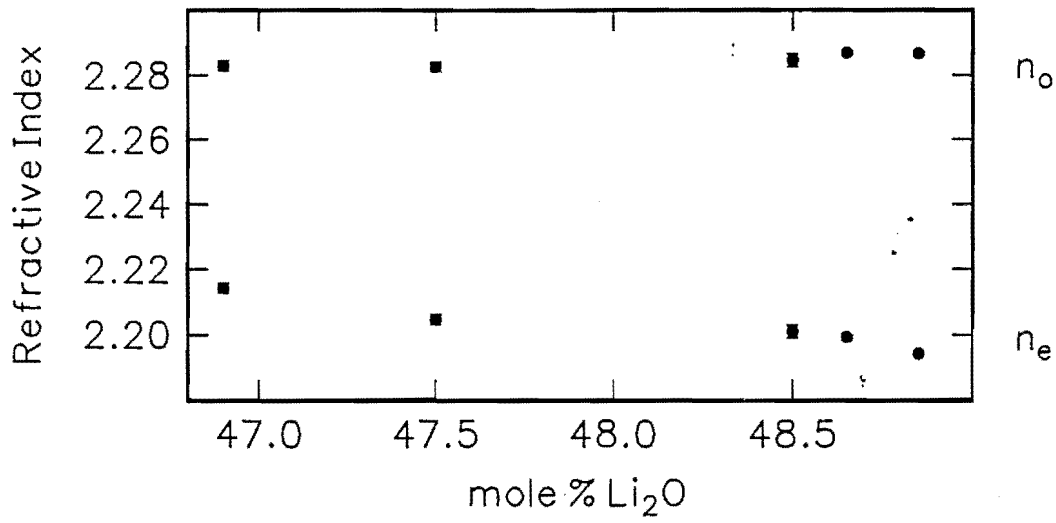

FIGURE 4: Ordinary (n_o) and extraordinary (n_e) refractive index of $LiNbO_3$ as a function of the Li content (λ = 632.8 nm, T = 17 °C).

index values to be about 0.0005 [5]. With an accuracy like that, the extraordinary refractive index can be used as an easy-to-apply measure for stoichiometry determinations of $LiNbO_3$ crystals. The method would yield a sensitivity of about 0.05 % in the Li content. This sensitivity is less than that of second-harmonic-generation (SHG) methods [6, 7], yet the method can be applied in the whole concentration range, whereas the simple SHG methods fail at Li contents less than about 47.5 %.

The difference of the refractive indices yields the birefringence, a crystal parameter which commonly can be measured very much easier and more accurate than each of the

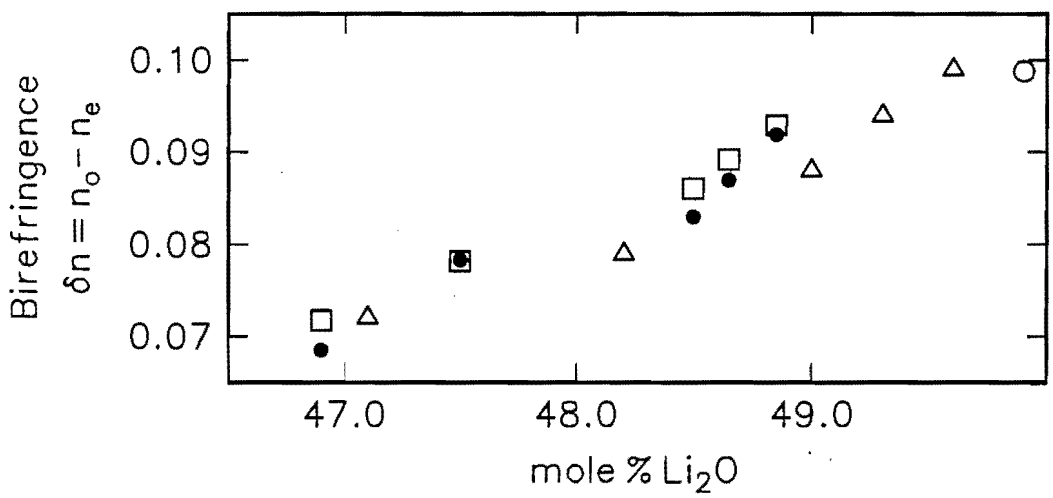

FIGURE 5: Birefringence of $LiNbO_3$ as a function of the Li content (λ = 632.8 nm, T = 17 °C). •: difference of the refractive index values from fig. 4, □: Hofmann [8], △: Carruthers et al. [9], ○: Jundt et al. [10].

refractive indices. Fig. 5 shows our birefringence values in comparison to the directly measured ones obtained from other authors. In spite of our values being the difference of two larger numbers, the agreement is excellent, a fact which expresses the accuracy of the method applied.

CONCLUSION

Using an interferometric method in combination with a numerical fit procedure, the refractive indices of $LiNbO_3$ were measured with considerable accuracy. Both refractive indices vary with the Li content in a characteristic way. The extraordinary index — which is the stronger varying one — can be applied as a sensitive measure for the stoichiometry of $LiNbO_3$. An accuracy of 0.05 % can be achieved for stoichiometry checks. This characterization method may serve as a complementary to the SHG methods in concentration regions where the latter ones aren't applicable.

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