Influence of In Doping on the Refractive Indices of Lithium Niobate

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(Received February 9, 1998; accepted February 24, 1998)

Introduction  Lithium niobate is presently one of the most important materials for electrooptic and nonlinear optical applications. Its properties can be tuned by varying its composition and by adding specific dopants like Mg [1], Zn [2] or Sc [3]. Here we present first measurements and a theoretical description of the influence of In on the refractive indices of lithium niobate. We can show how this dopant can be used to tune the phase matching properties for nonlinear optical applications.

Refractive Indices  Lithium niobate crystals doped with up to 2 mol % indium in the crystal [4] were grown by the Czochralski method from a congruent melt. The refractive indices of the samples were measured using an interferometric technique [5].

The experimental data of the refractive indices \( n \) can be described by a generalized Sellmeier fit which takes into account the defect structure of the material. It is assumed that certain defects in Li-deficient LiNbO\(_3\) mainly affect the concentration of Nb antisite defects \( c_{\text{NbLi}} \), which decreases linearly with increasing doping concentration [6]. The Sellmeier formula thus can be expressed as

\[
n^2 = \frac{A_0 + A_{\text{NbLi}} c_{\text{NbLi}} + A_{\text{In}} c_{\text{In}}}{(\lambda_0 + \mu_0 f(T) - f(T_0))^2 - \lambda^2} + A_{\text{UV}} - A_{\text{IR}} \lambda^2; \tag{1}
\]

with

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

\[
c_{\text{NbLi}} = \begin{cases} \frac{2}{3} (5 - c_{\text{Li}}) - c_{\text{In}} / \alpha_{\text{In}} & \text{if } c_{\text{In}} > 0, \\ 0 & \text{else}; \end{cases}
\]

\( T_0 = 24.5; \quad A_{\text{UV}} = 2.6613; \quad \alpha_{\text{In}} = 1.5; \)

where the wavelength \( \lambda \) is to be given in nm, the temperature \( T \) in °C and the concentration of In

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Parameters for the Sellmeier equation (1)</th>
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</thead>
<tbody>
<tr>
<td>( \lambda_0 )</td>
<td>223.219</td>
</tr>
<tr>
<td>( \mu_0 )</td>
<td>1.1082 \times 10^{-6}</td>
</tr>
<tr>
<td>( A_{\text{IR}} )</td>
<td>3.6340 \times 10^{-8}</td>
</tr>
<tr>
<td>( A_0 )</td>
<td>4.5312 \times 10^{-5}</td>
</tr>
<tr>
<td>( A_{\text{NbLi}} )</td>
<td>-7.2320 \times 10^{-8}</td>
</tr>
<tr>
<td>( A_{\text{In}} )</td>
<td>-2.4 \times 10^{-7}</td>
</tr>
</tbody>
</table>
as percentage in the crystal. The Li-content \( c_{Li} \) has to be extrapolated to undoped \( \text{LiNbO}_3 \) according to 
\[
\frac{\text{Li}_2\text{O}}{\text{Li}_2\text{O} + \text{Nb}_2\text{O}_5}
\]

The ordinary or extraordinary index of refraction is chosen by selecting the corresponding parameters from Table 1. Except for the new In-specific \( A_{In} \) all other parameters correspond to those determined for other compositions and dopants (for more details on the Sellmeier fit see e. g. [7] and [8]).

The measured refractive indices are – together with the theoretical data from the Sellmeier fit – depicted in Figure 1.

Using the Sellmeier fit (eq. (1)), phase matching conditions for various nonlinear optical processes can be derived. This was tested for the phase matching temperature for second harmonic generation where excellent agreement with experimental results was found.

Conclusions  In a similar way as Mg, Zn, Sc, the addition of In to lithium niobate can be used to taylor the optical properties of the material. Indium doping is especially useful for tuning the phase matching conditions for nonlinear optical processes as e.g. the phase matching temperature for second harmonic generation or optical parametric oscillation.

References