NONLINEAR SUSCEPTIBILITY AND SECOND HARMONIC GENERATION IN PROTON-EXCHANGED LITHIUM NIOBATE WAVEGUIDES

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Lithium niobate (LiNbO$_3$) is one of the most commonly used optical crystals because of its excellent electro-optic and nonlinear optical properties. Ease of waveguide fabrication and amenability to periodic poling has made lithium niobate highly attractive for quasi-phase-matched guided wave interactions such as second harmonic generation (SHG) and optical parametric amplification/oscillation [1]. Currently, the annealed proton exchange (APE) technique has become a very convenient way of fabricating waveguides in LiNbO$_3$. Proton exchange (PE) in LiNbO$_3$ is an attractive technique for nonlinear frequency conversion, because PE LiNbO$_3$ waveguides have high photorefractive damage threshold. Our works, reviewed and summarized in [2], have shown that the seven different crystallographic phases $\text{H}_x\text{Li}_{1-x}\text{NbO}_3$, the $\alpha$, $\kappa_1$, $\kappa_2$, $\beta_1$, $\beta_2$, $\beta_3$ and $\beta_4$ phases, can be realized in PE LiNbO$_3$ layers depending on exchange and annealing conditions. However, the relationships between the phase composition and the resulting nonlinear optical properties of PE LiNbO$_3$ waveguides were not well known. For the design, fabrication and optimization of nonlinear optical devices it is essential to determine the nonlinear optical susceptibility of PE waveguides consisting of the different $\text{H}_x\text{Li}_{1-x}\text{NbO}_3$ phases and correlate it to the processing conditions and their optical properties.

PE waveguides were formed on X-cut lithium niobate substrates. The fabrication conditions were chosen to provide the various phase compositions consisting of all the possible $\text{H}_x\text{Li}_{1-x}\text{NbO}_3$ phases. The effective refractive indices (ERI) of the waveguide modes were measured at a wavelength of 633 nm with the prism-coupling method. The profiles of ERI across the depth of the multimode single-phase waveguides were evaluated with the WKB method [2]. The strained state of the crystal structure was determined from X-ray rocking curves obtained with a DRON-3 diffractometer. Then, using the measured values of the strains $\epsilon_{33}$ and the ERI increments $\Delta n_e$, we determined the phase composition of the waveguides from the phase structural diagram [2]. The IR-reflection spectra were measured with a Bruker VERTEX 80v spectrophotometer.

Transmission spectra in the visible and near-UV ranges were taken with a SF-2000 UV/VIS spectrophotometer. The radiation was incident normally to the specimens. Therefore, the transmission spectra taken from the specimens with the PE waveguides represented the superposition of the spectra from the crystal plate and thin (0.5–4.3 μm) near-surface $\text{H}_x\text{Li}_{1-x}\text{NbO}_3$ layer. Thus, the contribution of the surface layers can be separated out from the integral spectrum if the partial spectra of lithium niobate and the $\text{H}_x\text{Li}_{1-x}\text{NbO}_3$ phase are considerably different. The accuracy of discrimination is the highest when the transmission is measured near the fundamental absorption edge, where the optical density of the specimens is extremely high. The optical absorption spectroscopy data in visible and near UV ranges were used to evaluate the nonlinear optical susceptibility of proton-exchanged waveguides in LiNbO$_3$ crystals, as the shift of fundamental absorption edge has been related to the changes of spontaneous polarization $P$ and coefficient of nonlinear susceptibility $d_{33}$ [3]. It has previously been established [3], that the nonlinear susceptibility coefficients of LiNbO$_3$ can be determined from optical absorption spectroscopy data with accuracy comparable to that of standard direct measurements. Advantageously, the process of data acquisition with optical spectroscopy is more tolerant to possible imperfections in the material under test, which typically manifest themselves as optical and electrical inhomogeneities. Such an analysis can help quantify the contribution of individual phases to the net nonlinear susceptibility and be a guiding tool in developing advanced high-throughput PE/APE LiNbO$_3$ devices with high optical nonlinearity.

Reflection second-harmonic (SH) generation from the polished waveguide end face was used to investigate the second-harmonic generation efficiency of different waveguides in congruent LiNbO$_3$ crystals: as-exchanged and annealed proton-exchanged (APE) waveguides with different $\text{H}_x\text{Li}_{1-x}\text{NbO}_3$ phases. The reflection SHG measurements have been performed from the polished waveguide end face using experimental setup early proposed [4]. 1.064 μm light polarized parallel to the Z-axis of LiNbO$_3$ from a mode-locked and Q-switched Nd:YAG laser was focused using lens onto the polished end face of the waveguide, mounted on a piezo-electrically driven translator permitted to vertically scanning both the waveguide and the bulk region to compare the waveguide nonlinearity with its substrate value. The reflected fundamental beam and the generated reflection-SH beam were separated using beam-splitters and an interference filter and detected with a germanium detector and a photomultiplier tube, respectively. The reflected IR intensity, which was used to determine the position of the air/sample interface, was monitored using a lock-in amplifier, and the SH signal which intensity is proportional to the square of the second-order nonlinearity was analyzed with a gated integrator.

The spectra taken from the $\text{H}_x\text{Li}_{1-x}\text{NbO}_3$-containing specimens and from the pure lithium niobate plates are depicted in Fig. 1. In the former case, the energy of the fundamental absorption edge markedly decreases according to the phase formed. In the specimens containing any of the phases, this energy shift is different for the ordinary and extraordinary polarizations. According to the theory of energy band shifts in ferroelectrics with oxygen-filled octahedrons [5], a shift of the band gap energy implies the significant decrease in the spontaneous polarization $P$ in the $\text{H}_x\text{Li}_{1-x}\text{NbO}_3$ phases in comparison with that in pure LiNbO$_3$. It is known [3,5] that a change in the spontaneous polarization by a value of $\Delta P$ causes a band gap shift $\Delta E^{se}$ for the extraordinary polarization, that is defined by the associated component of the effective polarization potential tensor $\text{t}_{11}$:

$$\Delta E^{se} = \text{t}_{11} \Delta P \times P$$

Thus, using Eq.(1), one can determine the spontaneous polarization $P_t$ ($P_t = P_0 + \Delta P$) in the phase studied from the band gap shift. The relative nonlinear susceptibilities $d_i$ in the various phases were estimated by the following relation [3]:

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\[ \frac{dI}{dI_0} = \left(t_0 \left(P_0 - \Delta P \right) \beta \right)^3 \left( t_0 \left(P_0 \right) \beta \right)^3 \]  

where \( d_0 \) and \( t_0 = t_{11} \) are related parameters for pure lithium niobate.

\[ \text{Fig.1. Optical spectra of PE waveguides, containing different Li}_{0.5}\text{H}_{0.5}\text{NbO}_3 \text{ phases: } \alpha, \kappa_1, \text{ and } \beta_2 \text{ (it are noted from left to right side at optical density level of 1.25). LiNbO}_3 \text{ substrate spectrum coincides with the spectrum of sample with } \alpha-\text{phase waveguide.} \]

Table 1 lists the nonlinear susceptibilities for the \( \text{H}_{1.0}\text{Li}_{0.5}\text{NbO}_3 \text{ phases that were calculated under the assumption that the effective polarization potential remains constant (} t_0 = t_{11} \text{). It should be noted that the above values of } P/P_0 \text{ and } d/d_0 \text{ far exceed our data obtained from IR reflection spectra for various phases of lithium niobate [6] and also by directly measuring the second harmonic generation efficiency. Recall, however, that the above calculations allow the determination of a local response, while direct SHG measurements reflect the superposition of the contributions from many local oscillators. Obviously, the lattice disorder in the protonated phases would reduce the coherency of local oscillators; consequently, the integral response that is determined by directly measuring } d_0 \text{ may diminish and even disappear. In fact, the lattice disorder is significant in the proton-exchanged \( \kappa_1, \kappa_2 \) and \( \beta_1 \text{, H}_{1.0}\text{Li}_{0.5}\text{NbO}_3 \text{ phases as follows from our data for IR-reflection spectra [3,6]. Therefore, the values of } d_0 \text{ evaluated from the band-gap shift } \Delta E \text{ have been corrected on basis of IR-data on lattice disorder in accordance with the approach given in Ref.3.} \]

Table 1. Relative values of nonlinear susceptibility coefficient \( d_{31} \) evaluated with data on band gap shift in optical absorption spectra \( d/d_0 (\Delta E) \), band-gap and IR-reflection \( d/d_0 (\Delta E, \text{IR}) \), and direct measurements of SHG efficiency \( d/d_0 \text{(SHG)} \).

<table>
<thead>
<tr>
<th>H(<em>{1.0}\text{Li}</em>{0.5}\text{NbO}_3 \text{ phase}</th>
<th>( \alpha )</th>
<th>( \kappa_1 )</th>
<th>( \kappa_2 )</th>
<th>( \beta_1 )</th>
<th>( \beta_2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( d/d_0 (\Delta E) )</td>
<td>1.0</td>
<td>0.924</td>
<td>0.809</td>
<td>0.8</td>
<td>0.793</td>
</tr>
<tr>
<td>( d/d_0 (\Delta E, \text{IR}) )</td>
<td>1.0</td>
<td>0.73</td>
<td>0.28</td>
<td>0.12</td>
<td>0.5</td>
</tr>
<tr>
<td>( d/d_0 \text{(SHG)} )</td>
<td>1.0</td>
<td>*</td>
<td>*</td>
<td>0.1</td>
<td>0.55</td>
</tr>
</tbody>
</table>

*- any quantitative evaluation of \( d_1 \) is impossible due to strong scattering of the pump and SHG beams.

Note, that the step-like \( \beta_2 \)-phase PE LiNbO\(_3 \) waveguides with large \( \Delta n \) and still significant nonlinearity have prospects for efficient SHG due to superior mode confinement that is in contrast to graded-index \( \alpha \)-phase waveguides.

The absorption spectroscopy method may be regarded as an attractive alternative to direct measurement, particularly for newly developed materials, available samples of which are often unsatisfactory in size or quality for any but crudest direct measurements. Besides, the optical requirements on a material for quantitative optical absorption data acquisition are often much less stringent than those for direct measurements of the SHG. Thus, our study opens the new possibilities for fast and easy estimation of performance of any newly developed LiNbO\(_3\)-based material for application in nonlinear integrated-optical devices.