RAMAN SPECTRA OF MULTIPLE CRYSTALLINE PHASES IN PROTON-EXCHANGED LiTaO₃ WAVEGUIDES

S.M. Kostritskii*, Yu.N. Korkishko, V.A. Fedorov

MPTE Dept., Moscow Institute of Electronic Technology, Moscow 124498, Zelenograd, Russia; E-mail: skostritskii@optolink.ru

Keywords: lithium tantalate, waveguide, phonon spectrum, electro-optic effect

Abstract: Proton-exchanged LiTaO₃ waveguides, containing multiple HₓLi₁₋ₓTaO₃ phases, were characterized by Raman scattering spectroscopy. Electro-optic coefficients of individual crystalline phases were determined using Raman data on electron-phonon interaction.

Lithium Tantalate is an attractive host material for use in integrated optics due to its large electro-optic and nonlinear coefficients. Currently, the Annealed Proton Exchange (APE) technique is used almost exclusively for the fabrication of waveguides in LiTaO₃, owing to its simplicity and the fact that it can be performed at temperatures below the Curie point. Proton exchange (PE) is known [1-3] to produce multiple crystalline phases in LiTaO₃ and severely degrade the electro-optic effect in as-exchanged waveguides. Subsequent annealing causes phase transformations and can restore electro-optics in APE waveguides. Each phase has its own specific dependence of refractive index on proton concentration, which is proportional to the lattice strain εᵢ₃ of the proton-exchanged layer [2]. Therefore, it is important to provide a detailed analysis of the electro-optic properties of all the HₓLi₁₋ₓTaO₃ phases. Such an analysis can be a guiding tool in developing advanced LiTaO₃ devices with high electro-optic efficiency.

It has been established [4] that the electro-optic coefficients of many materials, including LiTaO₃, can be determined from Raman spectroscopy data with accuracy comparable to that of standard direct measurements. Advantageously, the process of data acquisition with Raman spectroscopy is more tolerant to possible imperfections in the material under test. Raman spectroscopy was performed on the samples with the aid of a Renishaw Ramanoscope spectrometer. In the setup, a linearly polarized laser beam was focused on the optical-grade polished endface of the sample under study. Electro-optical coefficients were determined for multiple HₓLi₁₋ₓTaO₃ phases by using data on Raman spectra of the different waveguides.

By equating the electro-optic polarization to the sum of lattice and electronic contributions, we get the following relationship [4] between the electro-optic nᵢ²rıₖnᵢ², Raman αᵢⱼₖᵐ and infrared βₖᵐ oscillator parameters,

\[ nᵢ²rıₖnᵢ² = αᵢⱼₖᵐβₖᵐ + ξᵢⱼₖ \]  (1)

where the summation index m denotes the number of a phonon mode; ξᵢⱼₖ = 4dᵢⱼₖ is the pure electronic contribution to the electro-optic effect with dᵢⱼₖ. From SHG experiments, ξᵢⱼₖ was estimated [4] to be less than 10% of the lattice contribution in the case of LiTaO₃ and could therefore be neglected. As such, only the dominating lattice contribution (first term in the right-hand side of Eq. (1)) was retained and used in determining the electro-optic properties of the HₓLi₁₋ₓTaO₃ phases.

Using the data on frequencies and Raman scattering efficiencies of TO and LO phonons, as plotted in Fig.1, the values of ᵣ₃₃ were estimated for the fabricated samples and are presented in Table 1. The data clearly show a strong degradation of electro-optics in the βᵢ phases (due to a drastic decrease of the electron-phonon contribution to the linear response) and thus render these phases unsuitable for use in electro-optic devices.
Fig. 1. Micro-probe Raman spectra measured in near-surface layer of (1) LiTaO$_3$ substrate, (2) $\alpha$-phase waveguide with strain $\varepsilon_{33}=0.93\times10^{-3}$ (see Table 1), and (3) $\beta_1$-phase waveguide. Scattering geometry is $x(zy)$.  

Table 1. Primes and 0s are used to denote $r_{33}$ values corresponding to PE waveguides and virgin LiTaO$_3$, respectively.

<table>
<thead>
<tr>
<th>Phase:</th>
<th>$\alpha$</th>
<th>$\kappa$</th>
<th>$\beta_1$</th>
<th>$\beta_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Strain $\varepsilon_{33} \times 10^3$:</td>
<td>0.46</td>
<td>0.93</td>
<td>1.27</td>
<td>2.82</td>
</tr>
<tr>
<td>$r_{33}/r_{33}^0$:</td>
<td>0.97</td>
<td>0.9</td>
<td>0.63</td>
<td>0.48</td>
</tr>
</tbody>
</table>

It is worth noting that direct measurements [3] have suggested a non-monotonic dependence of $r_{33}$ on hydrogen concentration $x$ and strain $\varepsilon_{33}$ ($\varepsilon_{33} \sim x$)$^3$ with a minimum $r_{33}$ reached at intermediate values of $x$ that correspond to the $\kappa$ and $\beta_1$ phases. On the contrary, the data in Table 1 show that the electro-optic effect monotonically decreases as the strain increases. The discrepancy may be ascribed to a significant disordering of crystalline layers and its effect on the $r_{33}$, as the Raman data allow the evaluation of a local response, while direct measurements reflect the superposition of the contributions from many local oscillators. Obviously, the lattice disorder would reduce the coherency of local oscillators. Our data provide direct evidence of a high degree of such disordering. Indeed, Fig.1 shows a marked broadening of the Raman lines in waveguides with $\varepsilon_{33} \geq 0.7\times10^{-3}$ (see Fig.1). Observed differences are discussed and point to better accuracy obtained with the present approach.

References: