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Evaluation of Piezoelectric and Electrooptic Properties in Reverse-Proton-Exchanged X-cut LiNbO$_3$ Optical Waveguides
逆プロトン交換 X カット LiNbO$_3$ 光導波路の圧電・電気光学特性

Yoshio Sakai[‡] and Shoji Kakio (Univ. of Yamanashi)
酒井 香雄[‡]，垣尾 省司 (山梨大院・医工)

1. Introduction

Reverse proton exchange (RPE) has been proposed as a method of exchanging lithium ions and protons in a proton-exchanged (PE) layer fabricated on LiNbO$_3$ (LN) or LiTaO$_3$.[1] Kakio, one of the authors, and colleagues clarified that an RPE layer fabricated on a Z-cut LN substrate by the RPE method has the same functionalities, such as electrooptic and piezoelectric properties, as those of bulk LN,[2] and are investigating the application of such an RPE layer to an optical modulator[2] and a substrate structure for the loss reduction of leaky surface acoustic waves (LSAWs).[3,4] However, when an RPE layer was fabricated on X-cut LN (X-LN) to reduce the loss of a longitudinal-type LSAW, there was a problem that the recovery of piezoelectricity due to the RPE process was insufficient on almost the entire area of the sample.[5]

On the other hand, Ti-diffused optical modulators using X-LN have attracted attention owing to their long-range transmission because they can realize a zero-chirp characteristic owing to the symmetrical application of voltage to the optical waveguide (WG).[6] Furthermore, the combination of an RPE WG and X-LN is expected to suppress the dc drift phenomenon because it can be fabricated at a lower temperature than a Ti-diffused WG. Therefore, it is necessary to establish fabrication conditions under which the functionalities are recovered sufficiently after the RPE process is applied to X-LN.

In this study, the piezoelectric and electrooptic properties in an RPE WG fabricated on X-LN were evaluated by measurement of the SAW property and by applying the phase-modulation method[7] to an optical guided wave, respectively.

2. Piezoelectric Property of RPE X-LN

Samples with an RPE layer formed on an X-LN substrate were fabricated under two different conditions, and the propagation characteristics of a Rayleigh-type SAW (R-SAW) were measured in the Z-propagation direction. First, a PE layer was formed by immersing the substrate in a solution of benzoic acid diluted by lithium benzoate (Li 1.0 or 2.0 mol%) at 250 °C. The PE time for Li 1.0 and 2.0 mol% was set to 5 and 35 h, respectively, so that an initial depth d_{PE} of approximately 1.7 μm was obtained.

g11me017@yamanashi.ac.jp

Table I   Measured R-SAW properties.

	Virgin	Li 1.0 mol%		Li 2.0 mol%	
		as PE	RPE	as PE	RPE
d_{PE}/Λ, d_{RPE}/Λ	—	0.088	0.072	0.085	0.069
IL (dB)	15.3	20.0	17.8	19.4	16.1
K^2 (%)	2.5	1.2	1.7	1.4	2.3

Next, an RPE layer with a depth d_{RPE} of approximately 1.4 μm was formed by immersing each sample with the initial PE layer in an equimolar mixture of LiNO$_3$-NaNO$_3$-KNO$_3$ at 350 °C for 5.2 h. Interdigital transducers (IDTs) with wavelength Λ=20 μm, split electrodes with 8 finger pairs, and a propagation path length of 100 Λ were formed using an Al film. Samples without an RPE layer (as PE) were also fabricated.

The frequency response and admittance characteristics of the R-SAW were measured using a network analyzer. **Table I** shows the minimum insertion loss IL and coupling factor K^2. The depths of the PE layer and RPE layer normalized by the wavelength are also shown in Table I. The values of IL and K^2, which were deteriorated by the PE process, were recovered by the RPE process. Furthermore, it was found that the RPE sample fabricated with Li 2.0 mol% has closer values of IL and K^2 to those of the virgin sample than the RPE sample fabricated with Li 1.0 mol%. From these results, it is considered that greater recovery of the piezoelectric property can be obtained when the dilution of Li$^+$ in the initial PE process is increased. However, these properties include the properties of the RPE layer, buried PE layer, and LN substrate, and it is difficult to correctly evaluate the piezoelectric constant in the RPE layer.

In the following section, an optical guided wave is confined to an RPE channel WG whose electrooptic constant has the same (third) order tensor as the piezoelectric constant, and its electrooptic property is evaluated.

3. Electrooptic Property of RPE X-LN

Figure 1 shows the configuration of the phase modulators used to evaluate the electrooptic constant. First, a PE layer with d_{PE} of 1.8 or 2.7 μm was formed on the whole surface of an X-LN substrate. Next, RF-sputtered SiO$_2$ masks with a width of 2.8 or 3.0 μm were formed by a lift-off

Table II Fabrication conditions and parameters of phase modulators.

Sample	PE				RPE			WG width (μm)	Length of electrode l (mm)	r^S_{13} (pm/V)
	Temp. (°C)	Li (mol%)	Time (h)	Depth (μm)	Temp. (°C)	Time (h)	Depth (μm)			
RPE1	240	1.0	20	2.7	300	40	~1.0	3.0	11.3	1.7
RPE2	240	1.0	20	2.7	350	3.6	~1.0	2.8	10.5	1.3
RPE3	250	2.0	26	1.8	350	60.5	~1.0	3.0	19.7	4.3
Ti:LN	Ti diffusion at 950 °C for 9 h in Ar and 1 h in O$_2$							2.8	19.7	8.6
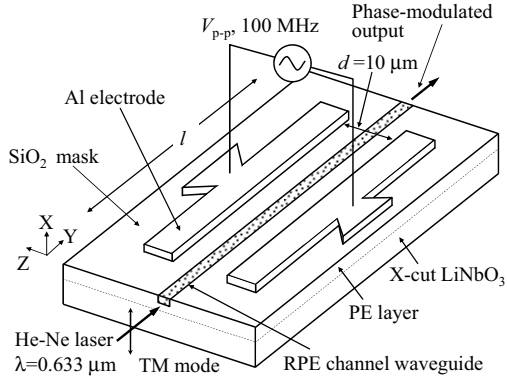
Fig. 1 Configuration of phase modulator.

Fig. 2 Measured phase modulation characteristics.

technique. Then, an RPE channel WG with d_{RPE} of approximately 1.0 μm was formed. Finally, after polishing the end face, two parallel strip electrodes with a gap d of 10 μm were formed using an Al film. Phase modulators were fabricated under three different conditions, as shown in **Table II**. A similar phase modulator with a Ti-diffused WG on X-LN was also fabricated as a reference sample.

A He-Ne laser beam with an optical wavelength of λ=0.633 μm was focused by an object lens onto the end face of a fabricated sample so that a TM-like single-mode guided wave was excited. An RF voltage with a frequency of 100 MHz was applied between the electrodes. The spectrum of the modulated output beam was detected using a photomultiplier and was observed with an oscilloscope. The electrooptic constant determined from the guided wave in the RPE waveguide is r^S_{13} in this measurement system.

The plots in **Fig. 2** show the phase modulation property for each sample at different applied voltages. The vertical axis is the ratio of the measured power in the carrier to that in the first sideband. The power ratio is given as the square of the ratio of the Bessel functions of the zeroth and first orders.[7]

$$\frac{\text{Power of first sideband}}{\text{Power of carrier}} = \left(\frac{J_1(u)}{J_0(u)}\right)^2 \quad (1)$$

Here, u is the phase modulation index,

$$u = \frac{\pi n_o^3 r_{13}^S l}{\lambda d} \Gamma V_{p-p}/2 \quad (2)$$

where n_o is the ordinary refractive index and l is the length of the electrodes. Γ is the correction factor
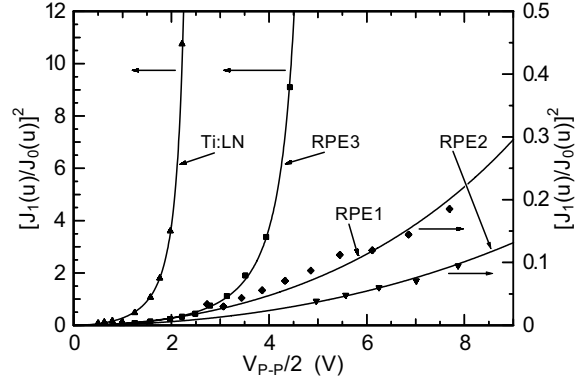
for the applied electric field, which was determined to be 0.94 from the phase modulation bulk value (8.6 pm/V). The values of r^S_{13} for RPE1-RPE3 were determined so that the theoretical curve (solid line in Fig. 2) could be fitted to the measured values.

The determined values of r^S_{13} are shown in Table II. When the proton source was diluted by Li 1.0 mol%, the values of r^S_{13} for RPE1 and RPE2 were less than 1/5 of the bulk value. On the other hand, when the proton source was diluted by Li 2.0 mol%, r^S_{13} for RPE3 recovered to half of the bulk value.

4. Conclusions

The piezoelectric and electrooptic properties in an RPE layer fabricated on X-LN were evaluated. It was found that greater recovery of K^2 and r^S_{13} due to the RPE process was obtained when the dilution of the proton source was increased from Li 1.0 to 2.0 mol%. The electrooptic constant r^S_{13} of the RPE WG was measured to be half of the bulk value for the dilution of Li 2.0 mol%. Further investigation will be needed to obtain piezoelectric and electrooptic constants closer to the bulk values.

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