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Characterization of LiNbO$_3$ Single Crystal Substrates Irradiated by Electron
電子線照射 LiNbO$_3$ 単結晶基板の評価

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Introduction

The lithium niobate (LiNbO$_3$) is a ferroelectric material. The LiNbO$_3$ crystal exhibits interesting piezoelectric, pyroelectric, electrooptic, and non-linear optical properties. Therefore, LiNbO$_3$ has various advantages for opto-electronic and acousto-optic applications including waveguids, modulators, second harmonic generators and surface acoustic wave (SAW) devices.[1] However, large number of defects in LiNbO$_3$ are generated during crystal growth process, slicing process from ingots and wafer polishing. Thus, introduced defects are induced a large influence on several devices.[2]

The electron irradiation will introduce the primary defect due to the atomic displacement damage in the material not influenced easily by the radiolysis. E. R. Hodgson and F. Agullo-Lopez reported that high-energy electron irradiation of stoichiometric LiNbO$_3$ induces F and F$^+$ centers.[3] They describes that the oxygen vacancies also play an important role in LiNbO$_3$. However, there are few articles discussing at the polished surface and the non-polished surface.

In this study, we characterized the defects in electron irradiated LiNbO$_3$ single crystal substrates evaluated by photoacoustic spectroscopy (PAS), Fourier transform infrared (FT-IR) spectroscopy and Raman spectroscopy.

Experiments

The samples used were commercially available LiNbO$_3$ single crystal grown by conventional Czochralski method. The samples were cut out of $18 \times 12 \times 0.5$ mm^3 from a 128°-Y-cut substrate 50 mm diameter and one surface was mirror-polished. The electron irradiation with 1 MeV was carried out at JAEA, Takasaki. The samples were irradiated with electron at fluencies between 1×10^{16} and 1×10^{17} /cm^2 at a

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fluence rate of 7.78×10^{12} /cm^2/sec. The electrons were irradiated onto the mirror-polished surface. To avoid sample heating up, samples were put on a water-cooled metal plate during electron irradiation. Thus, sample temperature was about 317 – 323 K during electron irradiation in this experiment.

The UV-VIS-IR absorption spectra of the samples were obtained with a gas-microphone photoacoustic spectroscopy (PAS) system,[4] operated in the 400-1600 nm range. Raman scattering spectra of samples were measured by a JASCO NRS-1000 micro laser-raman spectroscopy system. The wavelength of excitation laser is 532 nm using Nd:YVO$_4$. The infrared transmission spectra of the samples were obtained with a Fourier transform infrared (FT-IR) spectroscopy (JASCO FT/IR-610). All samples were measured at room temperature.

Results and Discussion

Photoacoustic (PA) spectrum of all samples are very broad. We normalized the spectrum based on the peak intensity of H$_2$O. Figure 1 shows the normalized photoacoustic (PA) signal intensity at 540 and 750 nm as a fuction of electron fluence. The peak of 540 nm and the peak of 750 nm show the electron colar centers and O^{2-}-hole centers, respectively.[5] The PA signal intensity at 540nm is not change by electron irradiation conditions. However, the PA signal intensity at 750 nm is increasing with increasing electron electron fluence. These results indicate that electron irradiation of our experimental range is affected for oxgen atoms.

IR transmission spectra around 3480 cm^{-1} are shown in Fig. 2. The peak in this region indicates the OH$^-$ absorption. The peak at around 3465 cm^{-1} and the peak at around 3480 cm^{-1} show the stoichiometric LiNbO$_3$ and Li-poor LiNbO$_3$, respectively.[6] Moreover, the OH$^-$ absorption spectra are usually used to examine the LiNbO$_3$'s defecte structure.[7] The shape of the spectrum is about the same compared with experiment condition at 5x10^{17} /cm^2. However, strength of the peak increases only in the experimental conditions at 5x10^{17} /cm^2. These
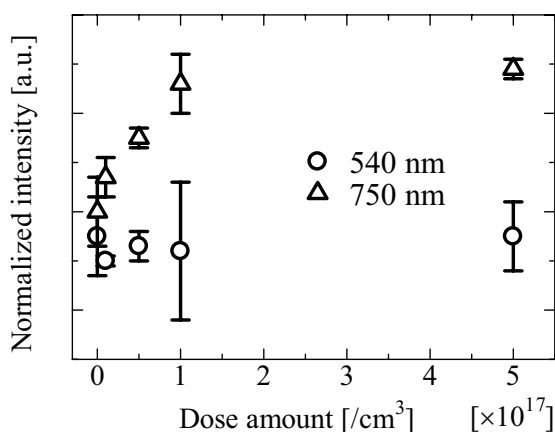
Fig. 1. PA signal intensity at 540 nm and 750 nm. The peak of 540 nm and the peak of 750 nm show the electron colar centers and O^{2-}-hole centers, respectively.
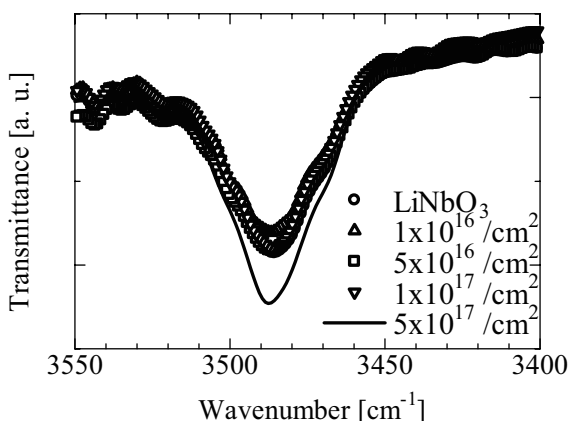

Fig. 2. FT-IR transmittance spectra related to the OH$^-$ vibration. These spectra normalized by Nb-O vibration at around 1745 cm^{-1}.

results shows that defects in LiNbO$_3$ increase over 5x10^{17} /cm^2.

Raman spectroscopy is sensitive to the lattice straine and existance of point defects. Figure 3 shows the high energy (A$_1$ mode) Raman spectra of LiNbO$_3$ samples. There were no change in the Raman spectra on mirror-polished LiNbO$_3$ surface side (Fig. 3 (a)). However, all peak intensity are decreased with increasing electron irradiation conditions from non-polished LiNbO$_3$ surface (Fig. 3 (b)). We confirm that the mirror-polished side has less lithium (Li) than the non-polished side in X-ray photoelectron spectroscopy. Therefore, it is considered that the Li vacancy weakens the effect of the electron beam irradiation. Further work is underway to clarify electron irradiation efects for LiNbO$_3$, especially between surface composition and irradiation condisions.
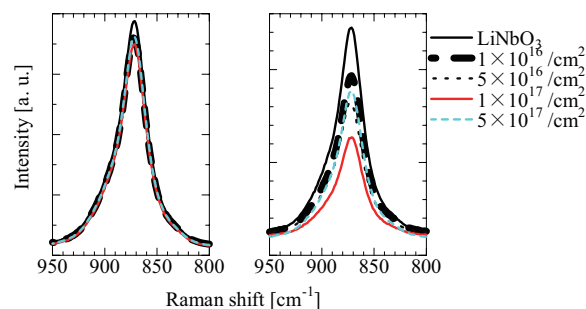

Fig. 3. Raman spectra of LiNbO$_3$ crystal irradiated onto mirror-polished surface. (a) The signal from mirror-polished surface, (b) non-polished surface.

Conclusions

We discuss about defects induced by electron irradiation in LiNbO$_3$. We understood that the electron beam irradiation introduced a defect, especially oxgen atom, in LiNbO$_3$ and mirror-polished surface, which has poor Li composition, is resists from electron irradiation effects.

Acknowledgements

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