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Atomic-scale Smoothing and Structural Analysis of $LiNbO_3$ Surface

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1. Introduction

$LiNbO_3$ single crystal has a variety of application as optical and piezoelectric devices. The wide use of $LiNbO_3$ is based on diverse physical properties: a large optical nonlinearity (that is important for optical wave-length modulators), large electro-optic effects (that is applied to electric field sensors, switches, and amplitude modulators), and high electromechanical coupling efficiency (needed for filters and acceleration sensors). In addition, $LiNbO_3$ crystal is a representative substrate material for the SAW (surface acoustic wave) devices such as RF and IF (intermediate frequency) filter that are also based on the piezoelectricity.

The above high electro-optic and electromechanical constants give to the practical devices high performance such as low insertion loss, high frequency, small size and lightweight. For the piezoelectric device, the single crystal has a higher merit than other sintered materials because of the large and stable dielectric polarization.

All of these devices involve an interface on the $LiNbO_3$ surface in the form of electrode, waveguide, or buffer layers: the essential part of the optical modulators is an optical waveguide consisting of a higher refractive index. This element is a few microns in thickness at the crystal surface, made by diffusion or crystal growth. In case of the SAW devices, the interdigital transducer (IDT) is formed by metal deposition and etching process. However, the interface structure is not taken care enough, despite the importance of the role for the device property.

Usually, the $LiNbO_3$ wafer is cut from the CZ (Czochralski) crystals that are supplied widely. The wafer is treated by mechano-chemical process to provide a mirror-polished surface. However, the mirror-polished surface is not atomically flat and gives many grain boundaries to the film grown on the surface. The structure with the grains gives damages to the film under high frequency condition, due to migration. Since the SAW device property is seriously affected by the quality of the electrode film [1], this damage is critical for the higher frequency devices that need smaller fine IDT electrode.

In this paper, we present a trial for obtaining the atomically flat surface of the $LiNbO_3$ single crystal. After obtained the atomically flat surface, we analyzed the surface structure by use of coaxial-impact collision ion scattering spectroscopy (CAICISS) in order to check the influence of the treatment. It is important because the crystal structure relates to the dielectric polarization that is strongly linked to the optical property of devices [2].

2. Experiments and Results

A single crystal of $LiNbO_3$ (0001) wafer with size of 15 $\times$10$\times$0.5 mm was supplied from Earth pharmaceutical Co.,Ltd. The Z-cut crystal is practically used for the optical waveguide devices by forming a layer by diffusion or epitaxial growth on the substrate, while the 64 ° cut substrate is used for the SAW devices. The sample was rinsed at room temperature in methanol and aceton successively for 20min each with an ultrasonic cleaner. After dried, the sample was annealed at 1000℃ for 1 h in air, using the muffle furnace. The effectiveness of annealing process in air has been reported for other some kinds of oxide surface, although it depends on the materials [3].

Figure 1 (left) shows the atomic force microscope (AFM, SPI-3800 system by SEIKO Corp.) image of the as-supplied $LiNbO_3$ (0001) crystal. The surface has many irregular corrugations from 0.4 to 1.2 nm. The surface after annealing is shown in the right part of Fig.1. The AFM confirms the presence of atomically flat 300 nm wide terraces and 0.22 nm height steps. This step height is well accords to the periodicity of a set of (Nb,Li,O) layers, which is 0.231 nm for illmenite structure of $LiNbO_3$.

The results of the annealing treatment were dependent on the temperature of about ±100℃ and not to the during time of about 1 hour. It suggests that this treatment is dominated by a finite activation energy. From viewpoint of the application, it is worth to note that this condition to obtain the atomically flat $LiNbO_3$ surface was wirhout any other process such as ion bombardment.

A merit of the atomically flat surface of the $LiNbO_3$ crystal is not limited to the SAW devices, because the good uniformity and crystallinigy are generally required for layers grown on the substrate in many devices. In addition, the atomically flat surface will provide other possibilities: a base plate to fabricate a 2-dimensional defect-free film not only the epitaxial layers, or a substrate for high-resolution AFM observation [4], or a template for nanowire fabrication [5].

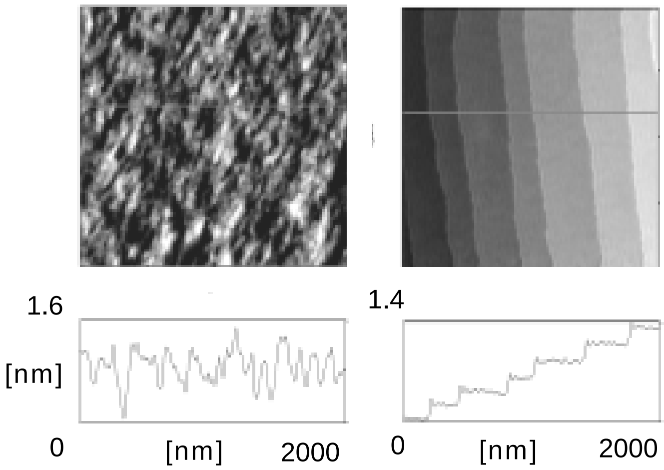

Fig. 1. AFM images of $LiNbO_3$ (0001) crystal, $2\times2\,\mu m^2$ for both: (left) as-supplied, (right) annealed at 1000℃ for 1 hour. The cross-sectional profile was taken along the horizontal line indicated in the image.

Next, we analyzed the surface topmost layers structure of the annealed $LiNbO_3$(0001) surface by CAICISS to verify the influence of the treatment. All experiments with CAICISS (TALIS 9700, Shimadzu Corp.) were carried out in an ultra high vacuum chamber with a base pressure of about 1×10^{-7} Pa. The He ion beam (2KeV, beam diameter of 3mm) was introduced to the sample and a time-of-flight energy analyzer was set coaxially to obtain a scattering angle of 180°.

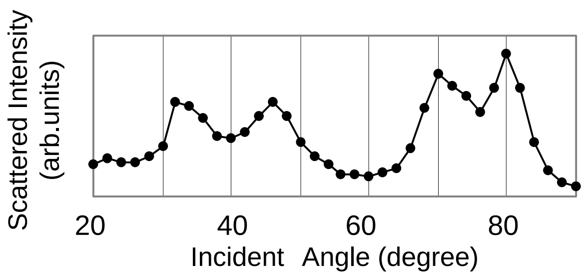

Fig. 2. The incident angle dependence of scattered yield of He ion from the Nb atoms on the annealed (0001) surface (1000℃ for 1h) measured by CAICISS. Ion beam is along [$\bar{1}$100] direction. Appearance of a clear peak of the Nb signal at 32 degrees represents the shift of Li atoms from the original site.

A series of CAICISS spectra were taken by scanning the incident (θ) or azimuth (ϕ) angle and compared with simulation spectra. The spectra for as-supplied sample showed a difference between (0001) and (000$\bar{1}$) surface, refletcting a distinction of dielectric polarity. After annealing the sample in air, θ scan data showed a new peak only for (000$\bar{1}$) surface, as shown in Fig.2. Considering the atomic geometry, it was found that this peak represents a shift of Li atoms in surface region. Since this peak did appear for (0001) surface before and after annealing, this change suggests the inversion of dielectric polarity by annealing process [6].

3. Conclusions

We have achieved an atomic-scale smoothing of surface of the $LiNbO_3$ single crystal and obtained an appropriate condition. Next, we analyzed the surface structure of the atomically flat surface by CAICISS. A change was found after annealing, which suggest the inversion of dielectric polarization from (+) phase to (-) phase. It will give a clue to know the influence of the smoothing process to the optical property, because the spacial control of the dielectic polarization is important part for the waveguide formation in the opto-electrical devices.

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