Jpn. J. Appl. Phys. Vol. 35 (1996) pp. 4040-4041
Part 1, No. 7, July 1996

Short Note

Surface Steps Observed in a Dry Annealed *z*-Cut $LiNbO_3$ Substrate

Hirotoshi NAGATA* and Kaori SHIMA

New Technology Research Laboratory, Central Research Laboratories, Sumitomo Osaka Cement Co., Ltd., 585 Toyotomi-cho, Funabashi-shi, Chiba 274, Japan

(Received March 8, 1996; accepted for publication April 30, 1996)

Surface reformation is observed in an OH-reduced *z*-cut $LiNbO_3$ single-crystal substrate after annealing at 1000∞C in a dry oxygen atmosphere. Atomic force microscopic observation reveals a step-like texture with step height of 0.7 nm height, corresponding to half of the *z*-axial length of $LiNbO_3$.

KEYWORDS: $LiNbO_3$ crystal, atomic force microscopy, surface, step

Because Ti:$LiNbO_3$ optical waveguides are fabricated at high temperatures, near 1000°C, surface coarsening of the $LiNbO_3$ substrate due to outdiffusion of Li must be suppressed to reduce optical propagation loss. In order to achieve this, a moist gas is usually supplied during the Ti-indiffusion treatment.[1)] However, for OH-reduced $LiNbO_3$ substrates, the surface degradation by Li evaporation is reported to be slight, even after annealing in a dry atmosphere.[2,3)] Here, the surface morphology of such OH-reduced $LiNbO_3$ was investigated on an atomic scale using an atomic force microscope (AFM), revealing the occurrence of surface step formation rather than surface coarsening.

A commercial *z*-cut OH-reduced $LiNbO_3$ substrate (*Nihon Kessho Koogaku Co., Ltd.,* see ref. 2) was enclosed in a platinum box and annealed in a tube furnace at 1000°C for 10 h, while dry O_2 with a dew point of less than -70°C was introduced into the furnace at a flow rate of 500 cm^3/min. The temperature increase and cooling times were 2 h and about 3 h, respectively. The residual OH ion concentration was measured using a Fourier transform infrared spectrometer and found to be 2.4×10^{18} cm^{-3} for the unannealed substrate and 2.6×10^{18} cm^{-3} for the annealed substrate. The unannealed and annealed substrates were cut into small pieces for AFM observation of their -*z* faces. In order to prevent charging of the sample and the Si_3N_4 AFM probe, due to the pyroelectric effect of $LiNbO_3$, the sample was surrounded by silver paste and earthed. The observation was carried out in an ordinary room atmosphere.

Figure 1 shows an AFM image of the as-received $LiNbO_3$ substrate before annealing. The machachemically polished optically flat surface had a roughness of 1-2 nm. No particular texture was found within the observed 10×10 μm^2 area.

Figure 2 shows a similar image for the dry annealed substrate in which the growth of a step-like texture with mutually parallel edges is seen. About 22 steps were observed in the 16.2 nm slope-rise, along the diagonal of the 10×10 μm^2 AFM image, suggesting that the polished surface was slightly inclined from the *z*-axis normal. The step height was measured to be 0.74 ± 0.05 nm and was close to half of the *z*-axial length of $LiNbO_3$, 1.3862 nm × (1/2) = 0.6931 nm. A few steps about 0.46 nm high were also observed.

The typical step structure is shown in greater detail in Fig. 3 which is a 2×2 μm^2 image. Steps ~ 0.70 and ~ 0.46 nm high were observed. A terrace with a smaller height appeared to emerge from the neighboring step wall. The terrace faces were smooth compared with the observed vertical steps, except for the existence of a few small pits.

The results indicated that the -*z*-face of $LiNbO_3$ was thermally reformed on the atomic scale. The surface coarsening due to Li-outdiffusion during annealing in a dry atmosphere seemed to be suppressed for the OH-reduced $LiNbO_3$ substrate. The height of the steps, corresponding to half of the *z*-axial length, can be explained by the fact that it is equivalent to the shortest unit consisting of the $[LiO]^{-1}$, $[NbO_2]^{+1}$ and cation vacancy layers, in which the electric charge is neutralized.

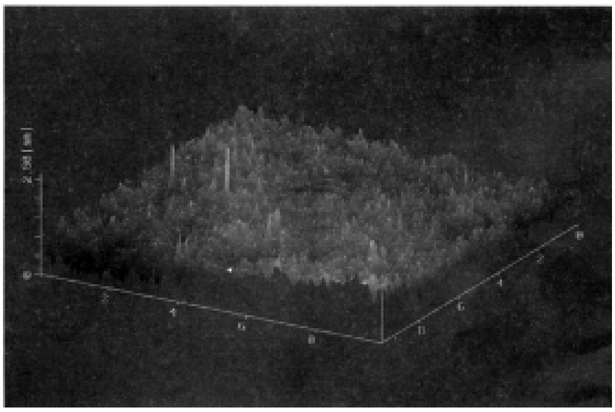

Fig. 1. AFM image of the unannealed -*z*-$LiNbO_3$ substrate (10×10 μm^2 area).

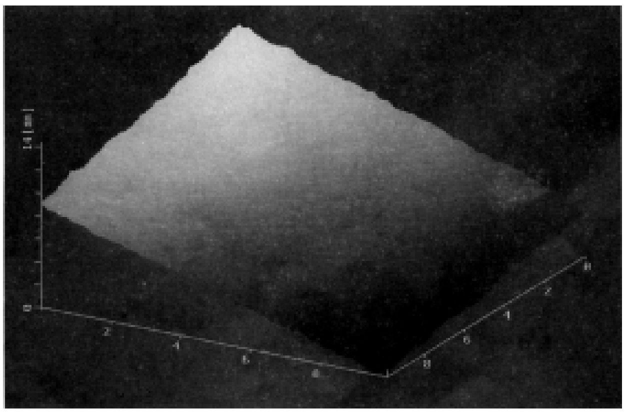

Fig. 2. AFM image of the annealed -*z*-$LiNbO_3$ substrate (10×10 μm^2 area)

*E-mail: LDT00246@nifty.ne.jp

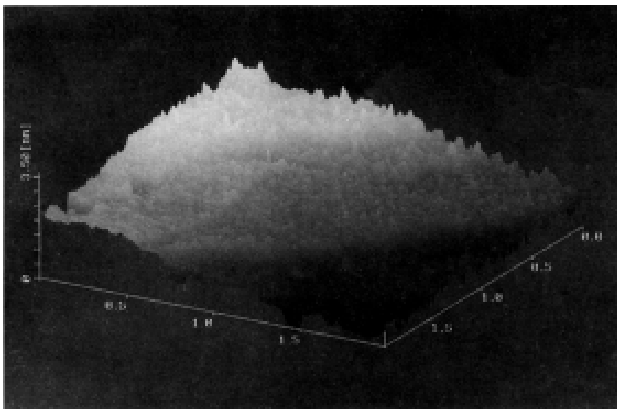

Fig. 3. AFM image of the annealed -*z*-$LiNbO_3$ substrate (2×2 μm^2 area). Terraces with ~ 0.7 and ~ 0.46 nm step height are observed.

Similar step formation by annealing has been reported for other simple oxide crystals, such as $SrTiO_3$ and Al_2O_3.[4,5] The steps 0.46 nm high are considered to be due to stacking faults in the $LiNbO_3$ substrate, 1.3862 nm × (2/6) = 0.4621 nm, because the $LiNbO_3$ consists of 6 layers along the *z*-axis. Further work is now in progress to investigate step orientation and the effects of annealing temperature, etc.

1) J. L. Jackel, W. Ramaswamy and S. P. Lyman: Appl. Phys. Lett. **38** (1981) 509.
2) A. Koide, T. Shimizu and T. Saito: Jpn. J. Appl. Phys. **33** (1994) L957.
3) H. Nagata, T. Sakamoto, H. Honda, J. Ichikawa, E. M. Haga, K. Shima and N. Haga: to be published in J. Mater. Res. **11**, No. 8.
4) M. Yoshimoto, T. Maeda, K. Shimozono, H. Koinuma, M. Shinohara, O. Ishiyama, and F. Ohtani: Appl. Phys. Lett. **65** (1994) 3197.
5) M. Yoshimoto, T. Maeda, T. Ohnishi, H. koinuma, O. Ishiyama, M. Shinohara, M. Kubo, R. Miura and A. Miyamoto: Appl. Phys. Lett. **67** (1995) 2615.