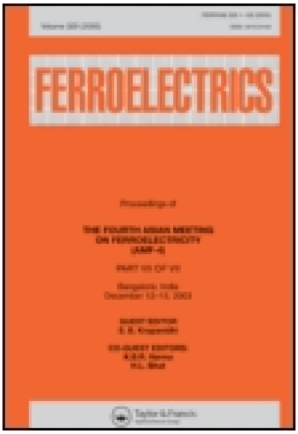

Ferroelectrics
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gfer20

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Available online: 26 Oct 2011

Ferroelectrics, 2001, Vol. 252, pp. 273-280
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Growth of $LiNbO_3$ Crystal with Periodic Ferroelectric Domain Structure by Current-Induction and its Acoustic Application

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(Received June 2, 2000)

In this report, Y-doped lithium niobate (LN) crystal with periodic ferroelectric domain structure (PFDS) was grown by the current-induction technique. The distribution of dopant (Y) along the growth direction was measured by an electron-probe microscopic analyzer. The periodic variation of the dielectric constant in the crystal was revealed by the light diffraction. When a radio-frequency electric field was applied, a 340 MHz ultrasonic wave was excited in the sample by the crossed-field scheme, which implies that the LN with PFDS is suitable for constructing the high-frequency bulk-wave acoustic devices.

Keywords: Lithium niobate; Current-induction; Periodic ferroelectric domain structure; Ultrasonic wave

INTRODUCTION

LN has long been a research topic because of its outstanding nonlinear

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optic, electro-optic and acoustic properties. In recent years, the LN crystal with periodic ferroelectric domain structure (PFDS) has attracted great research interest due to its applications in quasi-phase-matched (QPM) nonlinear optical frequency converter[1-4] and high-frequency bulk-wave acoustic devices.[5-7] Some effective techniques, such as the patterned electric-field poling technique[8], the off-center technique[4,9,10] and current-induction technique[11] have been developed to fabricate the PFDS.

In this work, a current-induction technique was proposed for the growth of LN crystals with PFDS during the Czochralski process. Large size LN crystals with the P_s perpendicular to the domain wall were successfully grown. The microstructure and acoustic properties of the as-grown crystal were characterized.

CRYSTAL GROWTH

As we know, if an electric current is applied through the solid-liquid interface (SLI) during the crystal growing process, the segregation coefficient of dopant in the melt will be affected.[12,13] Thus while the applied current is periodically varied, a periodic distribution of dopant along the growth direction would be induced in the crystal. The periodic concentration distribution of dopant may cause a periodic space-charge field and then the periodic ferroelectric domain is written in the crystal when the ferroelectric phase transition takes place.[14,15] In our experiment, 0.5 wt% yttrium was selected as the dopant. The period of the current pulse is about 6~20 s. The peak-value of current density at the SLI is around 15 mA/cm^2. The crystal was grown along the z-direction with a pulling rate of 3~6 mm/h. In order to avoid the influence of growth striation due to crystal rotation, we kept the rotation axis static. The crystal seed was also located at the center of the temperature field to achieve the uniformity of the crystal quality. Using this method, LN crystals with the dimension of 30 mm in diameter and 35 mm in length were successfully grown. Figure 1 is a scanning electron microscope (SEM) photograph of the y-face of an as-grown crystal etched in a HF:HNO_3 mixture. The PFDS was finely built through out the bulk crystal with the period of 10.3 μm. The continuous period number of the PFDS is over 400, with a period fluctuation of domain less than 4%.

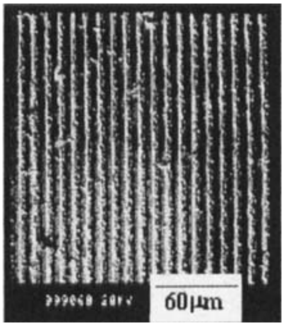
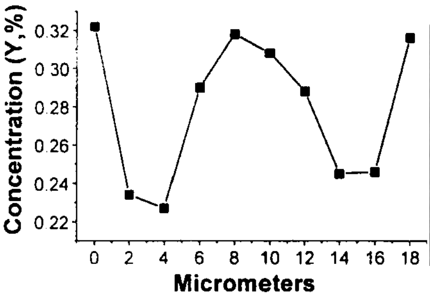


FIGURE 1 SEM photograph of the etched y-face of a LN crystal with PFDS structure grown by current-induction technique.

DISTRIBUTION OF DOPANT

The concentration distribution of dopant in LN or $LiTaO_3$ (LT) crystal with PFDS grown by off-center technique was studied by energy dispersive X-ray analyzer in the scanning electron microscope and found to be periodic along the growth axis.[14-16] In current-induction technique, the periodic current pulse would also result in a periodic concentration fluctuation of dopant in the as-grown LN crystal. To confirm this postulation, a JXA-8800M electron-probe microscopic analyzer was used to measure the concentration of dopant Y in the LN crystal with PFDS by current-induction technique. The y-cut sample was taken from a z-grown crystal. After being polished and etched, a thin carbon-coat was deposited on the y-face of the sample in order to avoid the congregation of electron during the measuring process. The concentration of dopant Y was measured, at a uniform interval of 2 μm, point by point, along a line normal to the domain boundaries on the y-face of the sample. In the figure 2 (a), the squares denote the ten measured data. The start and end positions of measurement were specified in the figure 2 (b). From figure 2, we can find that the period of concentration variation of dopant Y is equal to that of ferroelectric domain structure with a value of 12 μm.



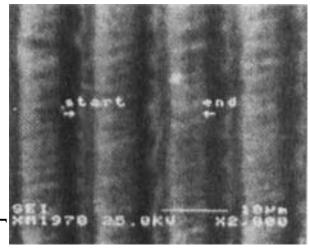


FIGURE 2 (a) Measured concentration of dopant Y, point by point, at a uniform space of 2 μm, along a selected line. (b) The specified positions of measurement on the morphology of PFDS.

DIFFRACTION EXPERIMENT

As we know, the even-rank tensor and corresponding physical properties are homogeneous in the LN crystal with PFDS. Since the refractive index is equal to the square root of the dielectric constant that is a second-rank tensor, the linear optical properties of a LN with PFDS should be uniform in the whole crystal. However, the dopant concentration is not uniform in a Czochralski grown LN with PFDS and in turn the dielectric constant will also fluctuate along the crystal growing direction. For the PFDS grown by the off-center technique, Lu *et. al.* demonstrated that there was really a periodic dielectric constant distribution associated with the periodic dopant fluctuation and the periodic domains.[17] For the current-induced PFDS, the investigation of the relationship between the domain structure and the dielectric constant distribution is also interesting and necessary.

To study the dielectric constant distribution, a light diffraction experiment was employed. A LN crystal with PFDS grown by the current-induction technique with the modulation period of 10.3 μm was selected for the experiment. The crystal was cut into an $8\times10\times2$ mm^3 ($z\times x\times y$) sample. A He-Ne laser beam with the wavelength of 6328 Å was shot into the sample along its y-axis and a white screen was put behind the sample to record the light spots. After passing through the sample, the light was diffracted into several beams and then several diffraction light spots were observed on the screen. The light diffraction picture shown in Figure 3 indicates that a periodic dielectric constant fluctuation exists in the sample. The 1st-order diffraction angle of 3° 24′ was measured and in turn the modulation period of the dielectric constant 10.6 μm is calculated. The value agrees well with the period of domain structure. Since the dielectric constant fluctuation is caused by the periodic dopant, we can conclude that its period is the same to the period of concentration of dopant.

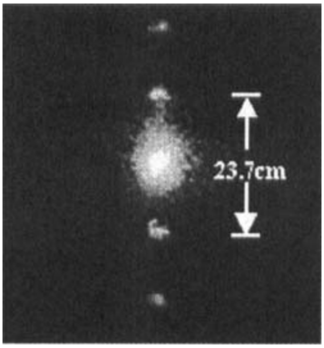


FIGURE 3 The diffraction picture of a He-Ne laser beam passing through a LN with PFDS structure. The distance between the sample and the screen is 200 cm and two 1st-order diffraction spots are 23.7 cm apart.

ULTRASONIC WAVE EXCITATION

Because of the different signs of piezoelectric coefficient in positive domains and negative domains, the domain boundaries in LN crystal with PFDS could be viewed as sound sources under the excitation of an alternating external electric field.[18] The ultrasonic waves excited in these sound sources will interfere with each other. As a result, those that satisfy constructive interference will lead to the appearance of resonance. The unique features of PFDS for acoustic application are low insertion loss and high working frequency that is determined by the period of domain.[5] There are two different schemes for the excitation of ultrasonic wave. One is "in-lined field" and another is "crossed field". The diagram of the "crossed field" excitation scheme is shown on the right corner of Figure 4. The electric field is applied on the y-face of LN to excite the ultrasonic wave propagating along the z-direction. Theoretically the main resonance is given by $f_n = nf_0$ (n=1,2,3...) with the fundamental frequency $f_0 = \frac{\upsilon}{a+b}$,[7] where a, b is the thickness of positive domain and negative domain, respectively and υ is the velocity of shear wave propagating along z-axis with a value of 3600 m/s.

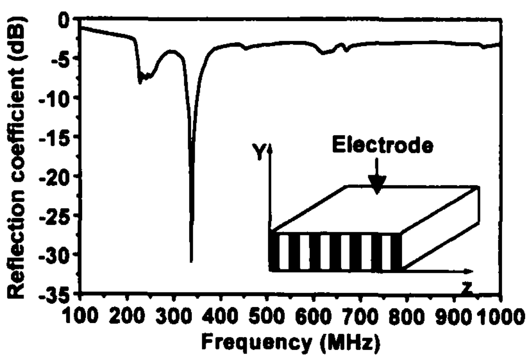


FIGURE 4 The measured reflection coefficient of a PFDS with the modulation period of 10.3μm. Right corner is the schematic of "crossed field" excitation of ultrasonic wave propagating along z-axis. The alternating electric field is applied along y-axis.

To testify this theoretical prediction, Ag electrodes were deposited on both sides of y faces of the LN sample with PFDS. Using a HP8510 network analyzer, the reflection coefficient of the sample was measured and shown in Figure 4. The resonant frequency locates at 340 MHz, which is very close to the theoretical value 346 MHz. The slight difference between them might result from the measurement error of the modulation period. The insertion loss of a transducer based on this sample that is determined by $IL = -20\log(1 - R^2)$.[19] The value of insertion loss is about 0 dB near the resonant peak and is very advantageous.

CONCLUSION

In conclusion, the LN crystal with PFDS was successfully grown by pulsed current induction during the crystal growth process. The distribution of dopant Y in the crystal was measured by an electron-probe microscopic analyzer. The periodic variation of the dielectric constant along the growth axis was revealed by the laser beam diffraction. A 340 MHz ultrasonic wave was excited by applied a radio-frequency electric field, which implies that the LN fabricated by this technique is suitable for constructing the high-frequency bulk-wave acoustic devices.

Acknowledgements
This work is supported by the State Key Program of Basic Research of China, the National Natural Science Foundation Project of China (contract 69708007), and the National Advanced Materials Committee of China. The technical supports from Dr. Yi-jun Feng and Wen-lan Zhang are also acknowledged.

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