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Fabrication of acoustic superlattice $LiNbO_3$ by pulsed current induction and its application for crossed field ultrasonic excitation

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An acoustic superlattice $LiNbO_3$ crystal with periodic ferroelectric domain structure was fabricated by introducing a periodic electric current through the solid–liquid interface during the crystal growing process. The domain morphology of an as-grown crystal was observed with a scanning electron microscope, and was found to be of good periodicity. A light diffraction experiment indicated that there was a periodic fluctuation of the dielectric constant along the crystal's growing direction. Using the ''crossed field'' scheme, a 340 MHz ultrasonic was excited in the crystal, which means that the acoustic superlattice is suitable for constructing high-frequency bulk-wave acoustic devices. © *2000 American Institute of Physics.* [S0003-6951(00)05038-5]

Lithium niobate (LN) has long been a research topic because of its outstanding nonlinear optic, electro-optic, and acoustic properties. In recent years, the LN single crystal with periodic ferroelectric domain structure (PFDS) has attracted great research interest due to its applications in quasiphase-matched (QPM) nonlinear optical frequency converter[1–4] and high frequency bulk-wave acoustic devices.[5–7] According to its different usage, the PFDS may be termed as optical superlattice (OSL) or acoustic superlattice (ASL). In this work, we will focus on the latter one, i.e., ASL LN. In order to fabricate the PFDS, some effective techniques have been developed, which may be sorted into two major kinds. The first one is fabricating the PFDS in an as-grown single domain crystal. The patterned electric-field poling technique[8] is an example. This technique is now widely used because of its accurate control of domain period. However, the sample thickness of about 0.5 mm might limit its application in some practical devices.[9] Furthermore, this technique can only fabricate the PFDS with the spontaneous polarization P_s parallel to the domain walls, which is commonly used as OSL. Another PFDS configuration is characterized by P_s perpendicular to the domain walls and has important acoustic applications, but it is impossible to be fabricated by this technique. For LN, fabricating the PFDS directly during the crystal growing process is another major effective way. Up to date, LN crystals with different PFDS configurations and with various dopants have been fabricated with the growth striation technique, which is realized by designing a special asymmetrical temperature field. Because of practical dimension of the sample, this technique is attracting more and more attentions.[4,9,10] However, one disadvantage of this method is that the periodic domain is worse formed in the center of crystal because of the smaller temperature variation, which affects the quality of domain structure. Thus it is very beneficial to find a technique that may fabricate the PFDS structure in LN with both large size and good quality.

In this letter, a current induction technique was proposed for fabricating the ASL LN with PFDS during the Czochralski crystal growing process by applying a periodic pulsed current between the crystal seed and the crucible. Large size ASL LN crystals with the P_s perpendicular to the domain walls were successfully fabricated. The microstructure and acoustic properties of an as-grown crystal were characterized.

As we know, if an electric current is applied through the solid–liquid interface (SLI) during the crystal growing process, the segregation coefficient of impurity in the melt will be affected.[11] Thus if the applied current is periodically varied, a periodic distribution of dopant along the growth direction should be induced in the crystal. This periodic impurity fluctuation is similar to the growth striation by crystal rotation, but their origins are different. Furthermore, the concentration fluctuation of the impurity exists even in the center of the crystal while there is no change of impurity concentration in the crystal's center area for the growth striation technique. Since the growth striation may cause a periodic space-charge field and then makes the periodic ferroelectric domain be written in the crystal when the ferroelectric phase transition takes place.[12] The current-induced periodic impurity distribution should also be able to make the PFDS be produced. In our experiment, 0.5 wt % yttrium was selected as the dopant. The crystal seed was used as the positive pole and the crucible as the negative pole for applying the periodic electric current. The period of the current pulse is 10 s, with 5 s duration of positive pulse, and 5 s of zero current. The current density in the SLI for the positive current is about $15\,mA/cm^2$. The crystal was grown along the z direction with a pulling rate of 3.5 mm/h. In order to avoid the influence of growth striation due to crystal rotation, we kept the rotation axis static in the experiment. The crystal seed was also put at the center of the temperature field to keep 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 after being etched in a $HF:HNO_3$ mixture to reveal its domain morphology. In this figure of PFDS, the positive domain thickness is almost equal to that of the

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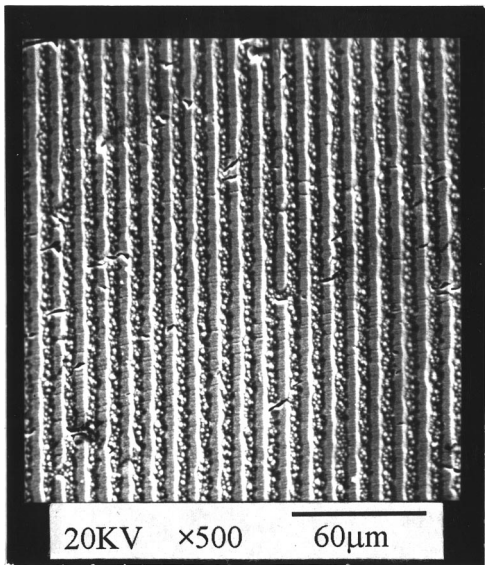

FIG. 1. SEM photograph of the etched y face of a LN crystal with PFDS structure fabricated by pulsed current induction technique.

negative domain. The PFDS was finely built both in the outer region and in the center through out the bulk crystal. The continuous period number of the PFDS is over 400, with a period fluctuation of domain less than 4%.

As we know, the nonlinear optical coefficient and piezoelectric coefficient were modulated from positive to negative domain periodically in LN with PFDS, thus the QPM and high-frequency ultrasonic excitation may be realized. In fact, other third-rank tensors such as the electro-optic coefficient[13] are also modulated periodically, while the even-rank tensor and corresponding physical properties are homogeneous in the crystal. 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 throughout the crystal. However, for a Czochralski grown LN with PFDS, the impurity concentration is not uniform, thus the dielectric constant will also fluctuate along the crystal growing direction. Although this fluctuation is not a consequence of the PFDS, the period is equal to the modulation period of the ferroelectric domain structure. For the PFDS that was fabricated by the growth striation technique, Lu *et al.* demonstrated that there was really a periodic dielectric constant distribution associated with the periodic impurity fluctuation and the periodic domains.[14] For the current-induced PFDS, investigating the relation between the domain structure and the dielectric constant distribution is also interesting and necessary.

To study the dielectric constant distribution, a simple light diffraction experiment was employed. An ASL LN crystal fabricated 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 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 Fig. 2 indicates that a periodic dielectric constant fluctuation exists

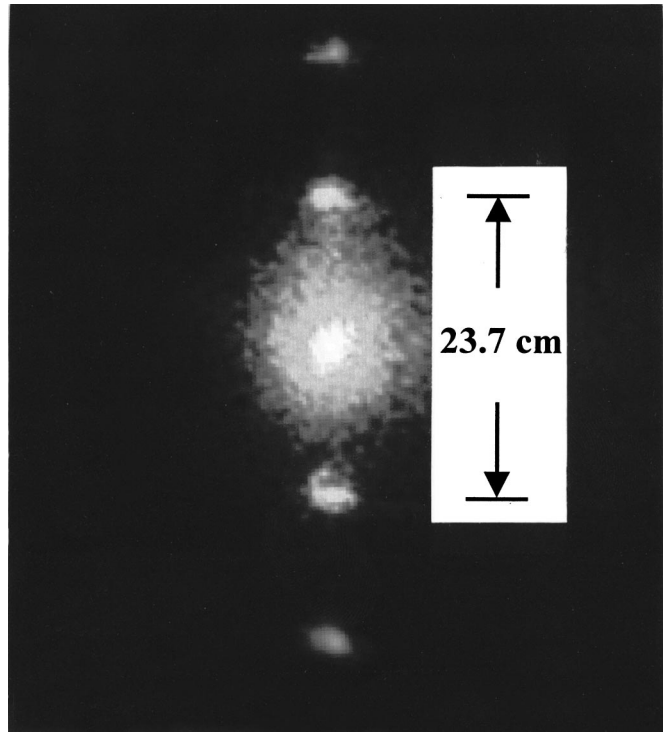

FIG. 2. The diffraction picture of a He–Ne laser passing through an ASL LN with PFDS structure. The distance between the sample and the screen is 200 cm and two first-order diffraction spots are 23.7 cm apart.

in the sample. The first-order diffraction angle was measured to be 3° 24′, thus the calculated modulation period of the dielectric constant is 10.6 μm, which agrees well with the period of the PFDS. Since the dielectric constant fluctuation is caused by the periodic impurity, we could conclude that there is also a periodic yttrium distribution in the crystal, which results in the PFDS.

As an ASL, the ultrasonic excitation effect was also studied in the same sample. Because of the different signs of piezoelectric coefficient in positive domains and negative domains, the domain boundaries could be viewed as sound sources under the excitation of an alternating external electric field.[15] 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 and thus the energy of electric field is converted to the elastic energy. The excitation of ultrasonic can also be treated as coupling between vibration of the superlattice and the electromagnetic waves, in which the ASL is considered as a 1D ionic-type phononic crystal.[16] The unique features of ASL for ultrasonic applications 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. One is ''in-lined field'' and the other is ''crossed field.'' Figure 3 shows the diagram of the crossed field excitation scheme. The electric field is applied on the y face of ASL LN to excite the ultrasonic propagating along the z direction. Theoretically there are two types of resonance, the main resonance and the satellite-like resonance.[7] The main resonance is given by

$$f_n = n f_0 \quad (n=1,2,3...) \tag{1}$$

with the fundamental frequency

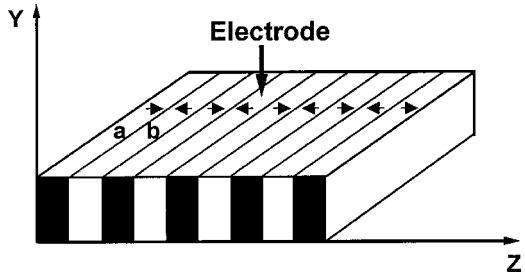

FIG. 3. Schematic of crossed field excitation of ultrasonic propagating along z axis. The arrows represent the orientation of the spontaneous polarization of LN and the alternating electric field is applied along y axis.

$$f_0 = \frac{v}{a+b}, \tag{2}$$

where a and b is the thickness of positive domain and negative domain, respectively, and v is the velocity of shear wave propagating along z axis. Satellite-like resonance is

$$f_m = f_n \pm \frac{m}{2N} f_0 (m = \pm 1, \pm 3, \pm 5 \ldots), \tag{3}$$

where N is the number of domain periods. For testifying the theoretical prediction above, a pair of Ag electrodes were deposited on the y faces of the ASL LN sample for the experiment. Using a HP8510 network analyzer, the reflection coefficient of the sample was measured and was shown in Fig. 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 domain walls not being exactly perpendicular to the z axis may influence its resonance frequency as well. From this resonance testing, no satellite-like resonance is observed, which shows that the positive domain thickness is almost equal to thickness of the negative domains.[7] The insertion loss of a transducer based on this sample is determined by[17]

$$IL = -20 \log(1 - R^2). \tag{4}$$

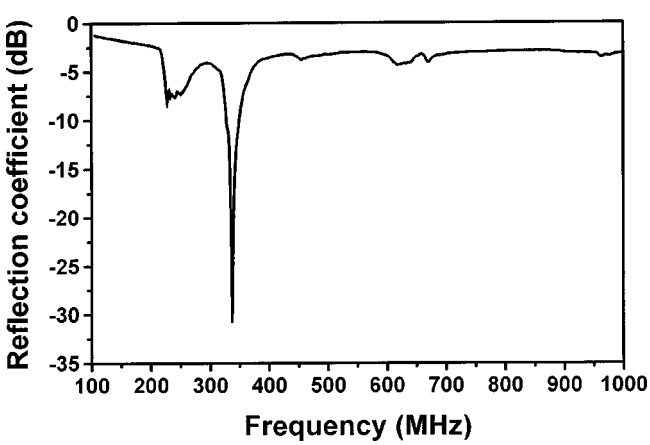

FIG. 4. The measured reflection coefficient of an ASL LN with the modulation period of 10.3 μm.

If we ignore the dissipative losses in measurement, the value of insertion loss is 0 dB near the resonant peak and is very advantageous. As we know, the elastic properties, which are related to the even ranks, should be identical in positive domain and in negative domain. As a consequence, ASL LN is acoustically homogenous and there is no additional propagation loss resulted from the periodic domain structure. In addition, a contiguous piece of LN that integrates ASL LN for wave-exciting and single domain LN for wave traveling can be fabricated by our current-induction technique. Thus, by adopting the crossed field scheme, one can devise an attractive kind of acoustic device in which the path of acoustic wave is not obstructed by bonds or electrodes.[18]

In conclusion, the ASL LN with PFDS was successfully fabricated by periodic pulsed current induction during the crystal growth process. Through a simple light diffraction experiment, the periodic variation of the dielectric constant along the growth axis caused by concentration variation of impurities was revealed. A 340 MHz ultrasonic was excited by an applied radio frequency electric field on the y face of the sample, which implies that the ASL LN fabricated by this technique is suitable for constructing the high frequency bulk-wave acoustic devices.

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[1] G. A. Magel, M. M. Fejer, and R. L. Byer, Appl. Phys. Lett. **56**, 108 (1990).
[2] M. Houé, and P. D. Townsend, J. Phys. D: Appl. Phys. **28**, 1747 (1995).
[3] V. Bermúdez, J. Capmany, J. García Solé, and E. Diéguez, Appl. Phys. Lett. **73**, 593 (1998).
[4] J. J. Zheng, Y. Q. Lu, G. P. Luo, J. Ma, Y. L. Lu, N. B. Ming, J. L. He, and Z. Y. Xu, Appl. Phys. Lett. **72**, 1808 (1998).
[5] Y. Y. Zhu and N. B. Ming, Appl. Phys. Lett. **53**, 1381 (1988).
[6] S. D. Cheng, Y. Y. Zhu, Y. L. Lu, and N. B. Ming, Appl. Phys. Lett. **66**, 291 (1995).
[7] Y. Y. Zhu, S. N. Zhu, and N. B. Ming, J. Phys. D: Appl. Phys. **29**, 185 (1996).
[8] L. E. Myers, R. C. Eckardt, M. M. Fejer, R. L. Byer, W. R. Bosenberg, and J. W. Pierce, J. Opt. Soc. Am. B **12**, 2102 (1995).
[9] J. Capmany, V. Bermúdez, and E. Diéguez, Appl. Phys. Lett. **74**, 1534 (1999).
[10] V. Bermúdez, M. D. Serrano, P. S. Dutta, and E. Diéguez, J. Cryst. Growth **203**, 179 (1999).
[11] A. Räuber, Mater. Res. Bull. **11**, 497 (1976).
[12] J. Chen, Q. Zhou, J. F. Hong, W. S. Wang, N. B. Ming, D. Feng, and C. G. Fang, J. Appl. Phys. **66**, 336 (1989).
[13] Y. Q. Lu, J. J. Zheng, Y. L. Lu, N. B. Ming, and Z. Y. Xu, Appl. Phys. Lett. **74**, 123 (1999).
[14] Y. L. Lu, T. Wei, F. Duewer, Y. Q. Lu, N. B. Ming, P. G. Schultz, and X. D. Xiang, Science **276**, 2004 (1997).
[15] H. E. Bommel and K. Dransfeld, Phys. Rev. **117**, 1245 (1960).
[16] Y. Q. Lu, Y. Y. Zhu, Y. F. Chen, S. N. Zhu, N. B. Ming, and Y. Y. Feng, Science **284**, 1822 (1999).
[17] E. K. Sittig, in *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1972), Vol. 10, p. 231.
[18] H. Gnewuch, N. K. Zayer, C. N. Pannell, G. W. Ross, and P. G. R. Smith, Opt. Lett. **25**, 305 (2000).