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Citation: Appl. Phys. Lett. **68**, 2642 (1996); doi: 10.1063/1.116267

View online: http://dx.doi.org/10.1063/1.116267

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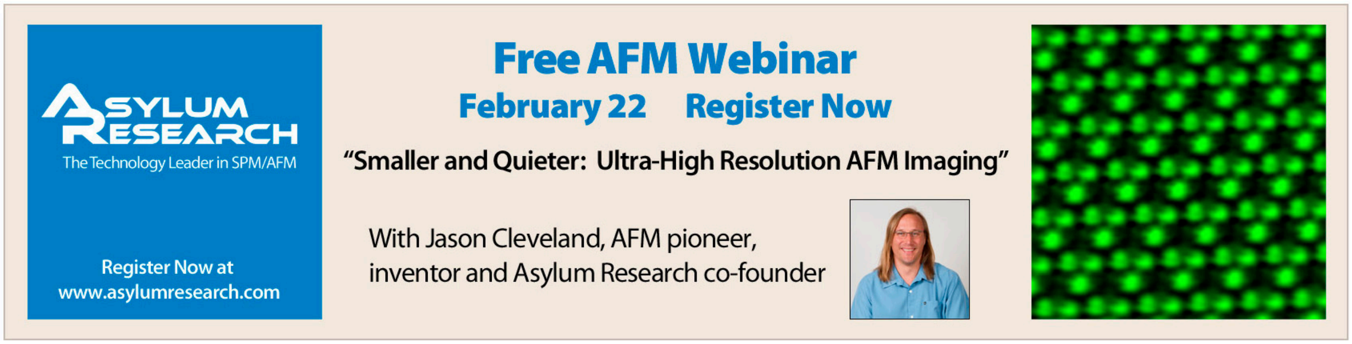

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Formation mechanism for ferroelectric domain structures in a $LiNbO_3$ optical superlattice

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(Received 16 October 1995; accepted for publication 27 February 1996)

Yttrium solute concentration distribution over periodic domains in a $LiNbO_3$ optical superlattice was measured with x-ray energy dispersive spectrum analysis (EDS). A critical concentration gradient mechanism has been proposed in which it is the concentration gradient which determines the configuration of periodic ferroelectric domains. An islandlike domain in periodic domains has been proved to originate from a solute aggregation during crystal growth. © *1996 American Institute of Physics.* [S0003-6951(96)01219-3]

Compact visible sources based on frequency conversion of diode lasers are of interest for applications in optical storage and optical communication. A $LiNbO_3$ (LN) optical superlattice (OSL) (i.e., $LiNbO_3$ crystal with periodic ferroelectric domain structures) allows quasiphase matched[1] (QPM) second-harmonic generation (SHG) over a wide range of fundamental wavelengths. The OSL also allows QPM optical parametric oscillation[2] (OPO), by appropriate choice of the modulation period according to corresponding coherence length. Several methods have been used to create a bulk LN OSL. The laser-heated pedestal growth[3] method has disadvantages of domain curvature and nonuniformities in the periodic domain structures. Some field-induced methods have permitted $LiNbO_3$ crystal with a domain inverted depth to about 0.5 mm,[4–7] but the incomplete reversal of the domains limited the effective interaction length, and the thickness of 0.5 mm is still insufficient for angle tuning in QPM OPO. The Czochralski method[8,9] has been used to prepare LN OSL with practical sample dimensions, and with a modulation period ranging from 2.0 μm to over 15.0 μm.[10] Efficient SHG for generating light from violet to green wavelengths have been achieved,[11] and direct frequency doubling of an 810 nm diode laser has been performed[12] in the crystal. However, there are also some disadvantages in the Czochralski method. They include the islandlike domains in periodic domain structures, the thickness inequality between the positive and the negative domain lamina, and the period fluctuations along the growing direction. These disadvantages severely limit the effective interaction length obtainable in these crystals when used for nonlinear optical applications. Therefore, careful studies of the origin of the above irregularities are important.

For the formation mechanism of periodic ferroelectric domain structures, Ming[13] found that there is a one-to-one correspondence between the growth striations and the domain configuration, and obtained the result that the formation of domain structures depends on the doped yttrium concentration gradient. Chen[14] has shown the same result in $LiTaO_3$ OSL. They have shown that domain walls are always situated at the place where the gradient of the Y solute concentration changes its sign from plus to minus or vice versa. The domain wall situated at the concentration's minima is very sharp, and the domain wall situated at the maxima is diffusive.

In this letter, a critical concentration gradient mechanism for periodic domain formation is proposed by measuring the correspondence between the growth striations and the domain configuration. The origin of the thickness inequality between the positive and the negative domain lamina, and the origin of the islandlike domains in periodic domain structures were interpreted from this mechanism.

LN OSL crystals doped with 0.5 wt % yttrium were grown along the *a* axis by the Czochralski method.[8] The as-grown crystals were first acid itched and then examined by an optical microscope so as to ascertain the periodicity of the domains. The Y solute concentration distribution over the growth striations was measured with an x-ray energy dispersive spectrum (EDS) attached to a scanning electron microscope (SEM). Two samples, one of which, sample A, has its positive domain thickness nearly equal to that of the negative, and sample B, with its positive domain thickness which is about two times larger than that of the negative, were chosen for our measurement. Figure 1 shows a photograph of the periodic domains of sample A which has a modulation period of about 8 μm. The one-to-one correspondence between Y solute concentration distribution over the periodic domain was also shown in Fig. 1. The curve is nearly symmetric. The sharp domain wall is situated at the concentration minima. The vertical dotted line in the figure shows the position of the diffuse domain wall which is near the maxima of the curve. The result is comparable with the previous work.[13,14] Figure 2 shows the photograph and the result obtained from sample B with a modulation period of about 7.5 μm. The distribution curve is still symmetric. The sharp domain wall is situated at the concentration's minima whereas the diffuse domain wall is situated in the region where the gradient is larger than zero and deviated strongly from the concentration's maxima. This result is different from that

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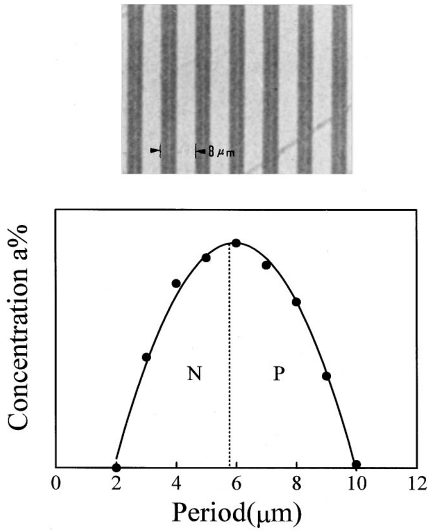

FIG. 1. Photograph of $LiNbO_3$ optical superlattice and corresponding Y concentration distribution (in the case of nearly equal domain laminae thickness).

presented by Ming and Chen.[13,14] This phenomenon shows that there is a critical concentration gradient which determines the position of the diffuse domain wall, thus the reversal of ferroelectric domains needs a critical space-charge field strength to act as a driving force. It should be stressed here that the LN OSL crystals are grown off-axis so as to impose a periodic temperature variation on the solid–liquid interface (SLI).[9] The periodic space-charge field is built up by the periodic solute concentration distribution which is introduced by this periodic temperature variations on SLI. It is the periodic space-charge field which determines the reversal of ferroelectric domains when para- to ferroelectric phase transition takes place. From Figs. 1 and 2, we found that sample A has a large periodic solute concentration fluctuation. The defined critical concentration gradient is a plus but close to the solute concentration's maxima. Thus the thickness of the positive domain laminae is nearly equal to that of the negative. Whereas in sample B, a low periodic fluctuation makes the defined critical concentration gradient deviate from the maxima by a large amount. Thus, the thickness inequality is large in sample B. The periodic concentration fluctuation relies on the periodic temperature fluctuation on the SLI, which was determined by the off-axis temperature system designed for the growth of LN OSL. Thus with a carefully designed temperature field system, this thickness inequality can be kept to a low level. A typical photograph of a LN OSL with a modulation period of 6.0 μm, in which the positive domain thickness is nearly equal to that of the negative, is shown in Fig. 3(a).

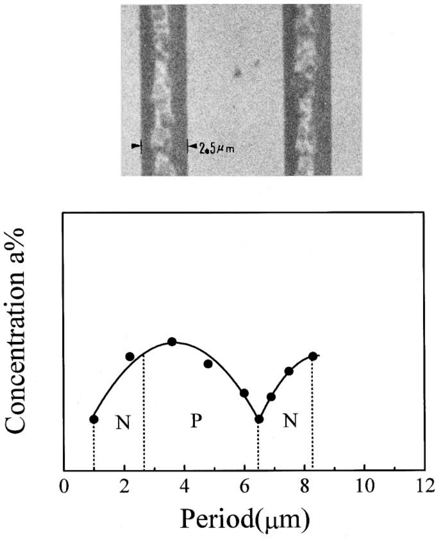

FIG. 2. Photograph of $LiNbO_3$ optical superlattice and corresponding Y concentration distribution (in the case of severely unequal domain laminae thickness).

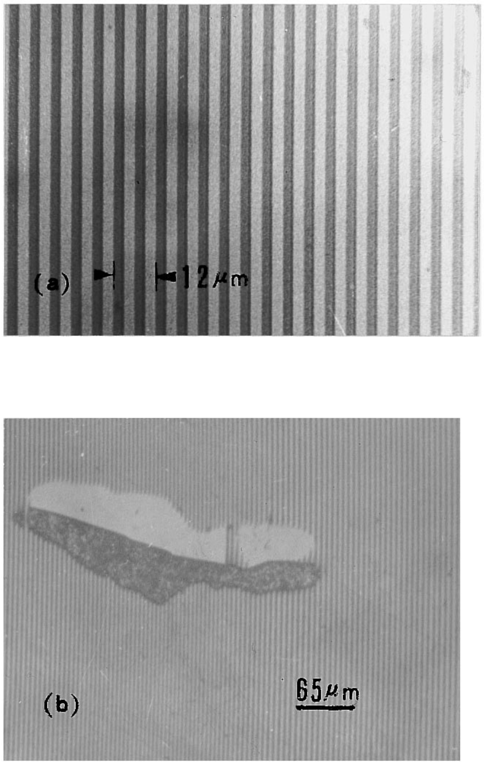

FIG. 3. Photographs of a perfect $LiNbO_3$ optical superlattice (a) and an islandlike domain in periodic domain structures (b).

Islandlike domain is one of the most destructive factors in the periodicity of a LN OSL. A typical islandlike domain is shown in Fig. 3(b). The Y solute concentration distributions both in the region of periodic domain (A–A) and inside the islandlike domain (B–B) were shown in Fig. 4. The enlarged photograph of the measured area was also shown in this figure. The A–A solute concentration curve is periodic and is in agreement with the previous result. Whereas, the B–B curve is not periodic and has a larger average solute concentration than that of curve A–A, this shows that there is a solute aggregation in this islandlike domain. Inside this islandlike domain the periodic concentration variation is suppressed by this larger Y solute aggregation. There is no point on the B–B curve where the gradient is larger than our defined critical concentration gradient. Thus periodic domain structures cannot be produced inside the islandlike domain. Such a solute aggregation originates from random temperature fluctuations on the SLI, which was introduced by the instability of the temperature system. So a carefully designed temperature field system is required to suppress this tempera-

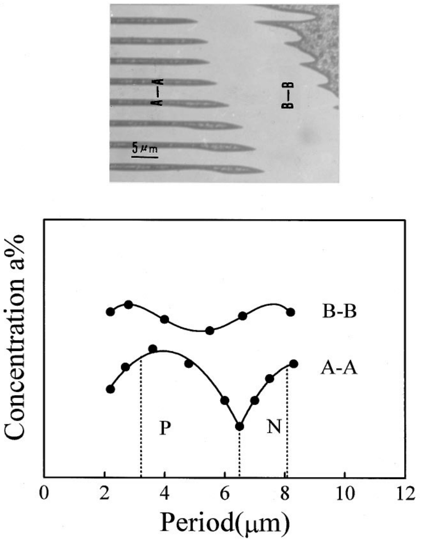

FIG. 4. Y solute concentration distributions in the region of periodic domain (A–A) and inside the islandlike domain (B–B).

ture instability. In our experiments, the number of islandlike domains in the periodic domain structures can be controlled by adjusting the off-axis temperature system. Crystals with a modulation period number over 300 and no islandlike domains on the crystal's etched $+b$ or $-b$ surface were obtained.

In conclusion, we have measured the Y solute concentration distribution across the growth striations by x-ray energy dispersive spectrum analysis. The sharp domain wall of periodic domains is determined by solute concentration gradient's sharp variation (at the solute concentration's minima), whereas the diffuse domain wall is determined by a defined critical concentration gradient near the concentration's maxima, which also determines the domain thickness inequality. Solute aggregations have been proved to be the origin of islandlike domains in periodic domain structures. The suppression of random temperature fluctuations on SLI is needed for eliminating such islandlike domains.

This work is supported by Ke-Li Fellowship.

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