Pulsed-laser deposition and optical properties of completely (001) textured optical waveguiding $LiNbO_3$ films upon SiO_2/Si substrates

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Completely (001) textured $LiNbO_3$ films have been fabricated upon thermally oxidized SiO_2/Si substrates by *in situ* application of a low electric field in a pulsed-laser deposition system. The biased voltage was 70–75 V on parallel electrodes separated by 7.0 mm. Li-enriched crystal and ceramic targets were used. The achieved films were nearly stoichiometric and have smooth surfaces. Waveguiding performance of the films was demonstrated by a prism-coupling method. The refractive indices were calculated on the basis of isotropic and anisotropic waveguide theories. A low-light propagation loss of 1.9 dB/cm in the films was achieved. It is expected that the films can be used in integrated-optical devices that require d_{33}, γ_{33}, ϵ_{33}, and n_{33} elements. © 1996 Optical Society of America

Lithium niobate ($LiNbO_3$; LN) has pronounced piezoelectric, pyroelectric, electro-optic, acousto-optic, nonlinear-optic, and photorefractive properties, which make it one of the key materials used in integrated optics.[1] Waveguide structures are essential for many integrated-optic devices. Therefore there are stringent requirements imposed on waveguide films. Imperfections such as porosity, refractive-index inhomogeneity, and surface roughness, if they are present to a sufficient degree, can scatter light out of the waveguide.[2] Thin-film deposition can prepare sharp step-index profiles and multilayer structures and, importantly, suits the existing semiconductor processing technology.[3,4] In particular, integration of LN films upon Si wafers with SiO_2 coating is commercially important. (001) epitaxial LN films have been grown upon sapphire at higher temperatures,[5] and (001) textured LN films upon (111) Si by sputtering, but polycrystalline LN films were obtained upon Si substrates with or without SiO_2 coating by pulsed-laser deposition (PLD).[6] Recently low-loss (104) textured LN films were sputtered upon SiO_2/Si substrates.[4] The refractive index is not yet homogeneous in these films because the normal of the (104) plane is not the optical axis of LN. In this Letter we report the preparation of (001) textured LN films upon SiO_2/Si by the PLD method. The (001) textured growth was induced by an *in situ* applied electric field during film deposition, which can align the spontaneous polarization of LN grains in the same direction. (001) textured LN films can create single coefficient elements such nonlinear and piezoelectric (d_{33}) and electro-optic ones (γ_{33}), permittivity (ϵ_{33}), and refractive index (n_{33}), which is essential for integrated-optic devices.

First a 1-μm-thick SiO_2 coating was grown by thermal oxidization of Si substrate to isolate the waveguide mode from Si substrate of $N = 3.85$.[7] Amorphous SiO_2 has a lower refractive index, $n = 1.46$, and is thermally stable. LN waveguide films were grown by a PLD method. The setup was described earlier.[8] We designed two large parallel planar electrodes in the deposition chamber, separated at 7.0 mm, to apply a low electric field. The substrate was placed upon the bottom electrode, which was electrically grounded, and rotation was set to ensure attainment of uniformity in film thickness and substrate temperature. A biased voltage Vb was applied to the top electrode, which consisted of a copper grid at the center region to allow for transmission of the laser-induced plume. A uniform electric field was built up perpendicular to the substrate and the film.[8] The deposition conditions were optimized and are listed in Table 1.

Considering the volatility of Li, we used two kinds of target: crystal plate grown from 20% Li-enriched LN melt and 10% Li-enriched ceramic disk. Without application of an electric field, the film is typically polycrystalline without preferential texture, as shown in Fig. 1. For simplicity in describing the influences of the electric field on the (001) textured growth, we introduced texture degree f in the form

$$f = (I - I_0)/(1 - I_0), \tag{1}$$

where I is the relative diffraction intensity of (006) peak normalized by the total peaks in the x-ray diffraction $\theta-2\theta$ scans and takes the value I_0 for the polycrystalline ceramic without preferential orientation.[9] $f = 0$ and $f = 1$ correspond to the typical polycrystal

Table 1. PLD Conditions

Parameter	Value
Laser type	Lambda Physik LPX205i
Excimer	KrF at 248 nm and 30 ns
Laser fluence	2.5 J/cm^2
Repetition rate	5 Hz
O_2 pressure	30–35 Pa
Deposition temperature	600 °C
Target	LN crystal and ceramics
Target substrate distance	3.5 cm
Biased voltage	$Vb = 0$–150 V

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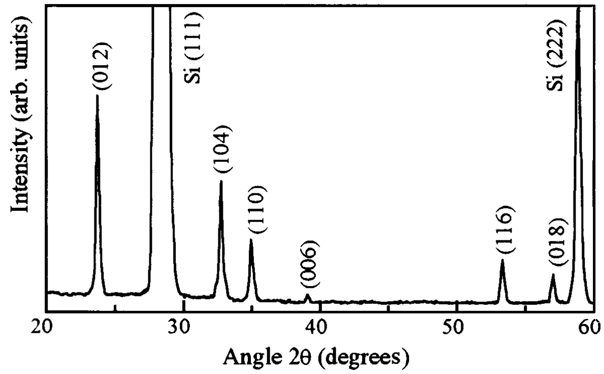

Fig. 1. X-ray diffraction $\theta-2\theta$ scan of LN film upon SiO_2/Si without application of an electric field. The film was polycrystalline.

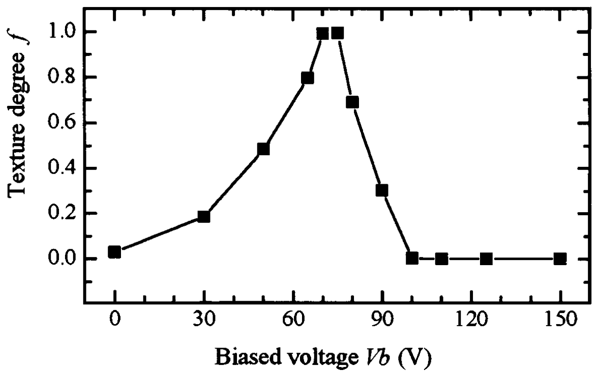

Fig. 2. Evolution of texture degree f of (001) orientation versus the biased voltage Vb. At $Vb = 70-75$ V, completely (001) textured LN films were obtained.

and complete (001) texture cases, respectively. $f < 1$ represents partial texture. Figure 2 shows the evolution of f versus Vb. At $Vb = 70-75$ V, f approached unity, and complete (001) texture was obtained. One of the x-ray diffraction θ-2θ scans appears in Fig. 3, which shows that a higher-biased voltage will deteriorate (001) textured growth of LN films. In the (001) textured LN film the refractive index is spatially homogeneous because (001) is the optical axis, $n_{11} = n_{22} = n_o = 2.29$, and $n_{33} = n_e = 2.20$.[4] This will lead to a lower propagation loss if light is confined in the waveguiding films.

The following points should be noted: (1) The (001) textured growth of LN films was induced by application of a biased electric field during deposition. As the unit cells of LN have spontaneous polarization at the deposition temperature, the low electric field can align the LN cells one by one with their polarization parallel to the electric field, forming the complete (001) texture.[8] This method is largely different from the conventional poling technique for ferroelectric ceramics, in which a high electric field must be applied to reverse or change the polarization direction of the ferroelectric domains as a whole, and it often leads to the breakdown of the ferroelectric materials. (2) This low-biased voltage cannot trigger glow discharge in the deposition chamber and is also applicable to other ferroelectric film-depostion techniques. (3) In the growth of LN films the epitaxial and amorphous nominal temperatures were 720 and 500 °C, respectively, in the present deposition conditions. The actual substrate temperatures were 50–100 °C lower than the indicated values because the thermocouple was positioned beneath the substrate stage. (001) textured growth achieved at the indicated temperature of 600 °C was in practice compatible with the semiconductor technology.

The stoichiometry was determined by x-ray photoelectron spectroscopy. The atomic ratio of Li to Nb to O was determined as 1.01 to 1.00 to 3.64. This result indicates that by using Li-enriched targets we achieved

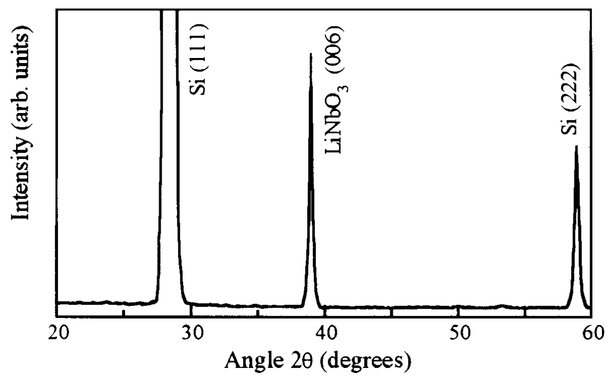

Fig. 3. X-ray diffraction $\theta-2\theta$ scan of LN film upon SiO_2/Si deposited by *in situ* application of a low electric field at the biased voltage of $Vb = 75$ V. Only the (006) peak is present.

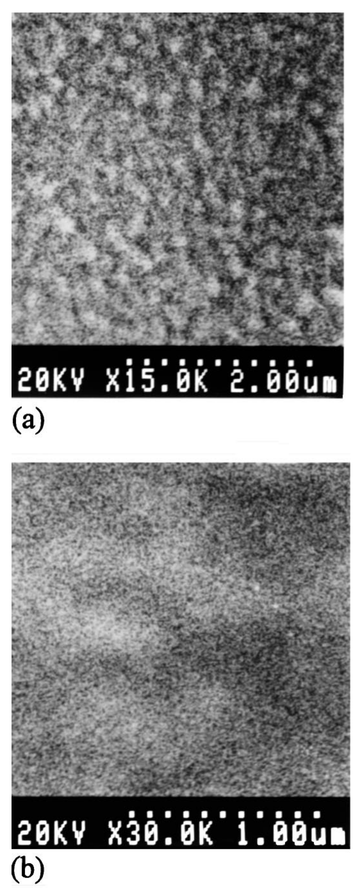

Fig. 4. Surface morphology of LN films deposited in the electric field at (a) $Vb = 0$ and (b) 75 V with the Li-enriched crystal target.

Table 2. Refractive Indices of $LiNbO_3$ Films Calculated from the Measured Coupling Angles of TE_0, TE_1 and TM_0, TM_1

Sample	Isotropic Theory[a]		Anisotropic Theory[b]	
	n_{TE}	n_{TM}	n_o	n_o
1	–	–	2.247	2.185
2	2.201	2.203	–	–
3	–	–	2.218	2.164
4	2.186	2.181	–	–

[a]Ref. 10.
[b]Ref. 11.

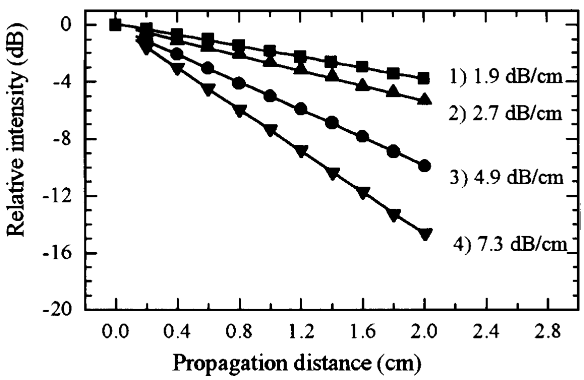

Fig. 5. Propagation attenuation of LN waveguide films upon SiO_2/Si and their propagation losses for the TE_0 mode. Samples 1 and 2 were (001) textured and polycrystalline films, respectively, with a crystal target. Samples 3 and 4 were (001) textured and polycrystalline films, respectively, with a ceramic target.

nearly stoichiometric LN films. A little higher O concentration was caused by the absorbed O_2 from the base atmosphere. We observed surface morphology with a scanning electron microscope. In (001) textured LN films the surface was much smoother, as shown in Fig. 4(b), than the polycrystalline films shown in Fig. 4(a).

The waveguide performance, refractive index, and propagation attenuation of the achieved LN films were demonstrated by a prism–film-coupling method. The TE and TM multimodes were excited by a He–Ne laser beam at a wavelength of 0.628 μm. We compiled two BASIC programs to calculate the refractive index and the film thickness from the measured coupling angles of TE_0, TE_1 and TM_0, TM_1 based on isotropic and anisotropic waveguide theories, respectively. The former suits isotropic polycrystalline LN films, and n_{TE} and n_{TM} were obtained[10]; the latter suits (001) textured LN films, and n_o and n_e were obtained.[11] The results are listed in Table 2. Four samples were measured and calculated. Sample 1 was (001) textured, and sample 2 was polycrystalline LN films, prepared by use of the crystal target. Sample 3 was (001) textured, and sample 4 was polycrystalline LN films, with a ceramic target.

From the observed bright-light propagation line the propagation attenuation was recorded and the propagation loss was fitted by the linear regression method. The results for the TE_0 mode are shown in Fig. 5. Films prepared from the crystal target had a lower propagation loss than those from the ceramic target. In particular, in sample 1 the propagation loss is as low as 1.9 ± 0.2 dB/cm. Evidently, the optical quality of the films depends greatly on the target quality and the degree of film texture. Scanning electron microscopy observation showed that (1) films deposited with crystal targets were much smoother, with negligible particulates, than those with ceramic targets and (2) grains of 0.2–0.3 μm were obvious on the polycrystalline films. On the other hand, the surfaces of (001) textured films were much smoother, and no grains could be discerned in the present scanning electron microscope observation.

In conclusion, we have successfully prepared (001) textured LN thin films upon amorphous SiO_2/Si substrates by applying *in situ* a low electric field at a biased voltage of 70–75 V, which was low enough not to trigger glow discharge in the pulsed-laser deposition system. The films were nearly stoichiometric at this low deposition temperature of 600 °C. A crystal target led to higher-optical-quality films than did a ceramic target. In the (001) textured LN film prepared from the crystal target the propagation loss was as low as 1.9 dB/cm. The method used is advantageous for fabricating (001) textured LN films used in integrated-optic devices that require specific physical coefficient elements.

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References

1. R. S. Weis and T. K. Gaylord, Appl. Phys. **37,** 191 (1985).
2. J. M. Mir and J. A. Agostinelli, J. Vac. Sci. Technol. A **12,** 1439 (1994).
3. S. Miyazawa, J. Appl. Phys. **50,** 4599 (1979).
4. C. J. Huang and T. A. Rabson, Opt. Lett. **18,** 811 (1993).
5. Y. Shibata, K. Kaya, K. Akashi, M. Kanai, T. Kawai, and S. Kawai, Appl. Phys. Lett. **61,** 1000 (1992).
6. S. B. Ogale, R. N. Dikshit, S. J. Dikshit, and S. M. Kanetkar, J. Appl. Phys. **71,** 5718 (1992); T. A. Rost, H. Lin, T. A. Rabson, R. C. Baumann, and D. L. Callahan, J. Appl. Phys. **72,** 4337 (1992).
7. S. Sriram, W. D. Partlow, and C. S. Lin, Appl. Opt. **22,** 3664 (1983).
8. W. S. Hu, Z. G. Liu, and D. Feng, Solid State Commun. **97,** 481 (1996).
9. Joint Committee on Powder Diffraction Standards, PDF 20-631.
10. P. K. Tien, Appl. Opt. **10,** 2395 (1971).
11. S. Yamanoto, Y. Koyamada, and T. Makimoto, J. Appl. Phys. **43,** 5090 (1972).
12. E. van de Riet and U. K. P. Biermann, Thin Solid Films **241,** 134 (1994).