Crystallography and structural evolution of $LiNbO_3$ and $LiNb_{1-x}Ta_xO_3$ films on sapphire prepared by high-rate thermal plasma spray chemical vapor deposition

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(Received 11 August 2000; accepted 21 May 2001)

The structure and the crystallography of lithium niobate and lithium niobate-tantalate thin films (0.2–1.0 μ m in thickness) with the tantalum composition range of $0 \le x \le 0.5$ grown on (0001) sapphire substrate by thermal plasma spray chemical vapor deposition have been studied by means of cross-sectional high-resolution transmission electron microscopy and x-ray diffraction. The tantalum composition in the films shows a minor effect on the rocking curve full width at half maximum values. The narrowest rocking curve width was obtained for the LiNb_{0.5}Ta_{0.5}O₃ film to be as low as $0.25^{\circ} \theta$. The films are under compressive strain along the c direction; c- and a-axis lattice parameters are correspondingly smaller and higher than those of the bulk single crystal. Under optimized growth conditions, the LiNbO₃ and $\text{LiNb}_{1-r}\text{Ta}_rO_3$ films are 97% *c*-axis oriented. The film out-of-plane orientation changes from the [0001] to the [0112] direction by either decreasing the growth rate or increasing the substrate temperature. Particular attention has been paid to the orientation of individual grains in the partly *c*-axis-oriented films. The results demonstrate that their orientations are not random and specific orientation relationships are preferred for the film nucleation. The surface of as-received sapphire substrate reveals polishing defects with the well-defined surface ledges of 1-2 nm in height with smooth terraces of 25 nm in width. In the case of columnar growth, the terrace width becomes a limiting factor controlling the lateral crystallite size in the film. Finally, the film growth mechanism is discussed.

I. INTRODUCTION

Lithium niobate (LiNbO₃) and lithium tantalate (LiTaO₃) possess excellent ferroelectric, piezoelectric, pyroelectric, and optical properties. Thus they are strongly desired materials as homogeneous thin films grown with good surface morphology and crystallinity on substrates with low index of refraction and high acoustic wave velocity. Over the last decade, LiNbO₃ and LiTaO₃ epitaxial films have been deposited by a large number of various techniques including molecular

beam epitaxy (MBE),¹ chemical vapor deposition (CVD),² metalorganic CVD (MOCVD),^{3–7} ion plating,⁸ liquid-phase epitaxy (LPE),^{9,10} sol-gel^{11,12} and polymeric precursor¹³ methods, excimer laser ablation,¹⁴ pulsed laser deposition (PLD),^{15–18} and rf (radio frequency) magnetron sputtering.^{19–21} The LiNbO₃ and LiTaO₃ films with the best quality have been reported to show optical losses less than 1 dB/cm,^{18,22} rocking curve full width at half maximum values of 0.04° ,^{4–6} and surface rms roughness values of 1.5 nm.²

Thermal plasma spray chemical vapor deposition method using liquid source material²³ has been successfully applied for the deposition of preferentially oriented LiNbO₃ films with the deposition rate as high as 0.1 μ m/min, which was 10–100 times faster than those of other conventional vapor deposition methods. It has been proposed that the main deposition species in the thermal plasma spray CVD are clusters. To meet technological requirements, it is necessary to develop films with large

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surface area, uniform thickness, and little evidence of structural defects. The crystallinity, stoichiometry, and surface roughness of films depend on growth rate, substrate temperature, and orientation. In the case of cluster deposition, however, the possibilities of varying composition, grain size, texture, and morphology of films are largely unknown. Thus, additional studies need to be undertaken.

In the present study, x-ray diffraction (XRD) (θ –2 θ scans, rocking curve measurements), scanning electron microscopy (SEM), and high-resolution transmission electron microscopy (HRTEM) were used to reveal the structure and the crystallography of the LiNbO₃ and LiNb_{1-x}Ta_xO₃ films deposited onto (0001) sapphire substrates.

II. EXPERIMENTAL PROCEDURE

Lithium niobate (LN) and lithium niobate-tantalate (LNT) films were deposited on (0001) sapphire substrates by the thermal plasma spray CVD method²³ for 2-10 min. Lithium-niobium and lithium-tantalum alkoxide metalorganic solutions for dip coatings [LiNb(OR)₆ and LiTa(OR)₆ in 3-methylbutyl acetate as a solvent, which are commercially available at Kojundo Chemical Lab., Sakado, Japan] were used as liquid precursors. The concentration of metals in each individual precursor solution corresponded to 3 wt% LiNbO₃ or LiTaO₃, respectively. These solutions were then mixed in the various molar ratios in order to get source material. As-received mechanically polished (0001)-axis sapphire substrates from Earth Chemicals, Osaka, Japan, were cleaned with acetone and ethanol prior to use. In comparison with our previous study,²³ we used a modified thermal plasma spray CVD apparatus in which the plasma blowing was vertically upward. A liquid solution containing metalorganic precursors was fed into a rf O₂-Ar thermal plasma as a mist from the injection probe. The mist of sprayed solution was co-evaporated and oxidized completely and then deposited onto a substrate. The precursor source materials were introduced into the plasma using a liquid feeder utilizing a pump. The substrate temperature was controlled within the accuracy $\pm 15-20$ °C and was directly measured by detecting infrared radiation from the reverse side of the substrate (covered with golden foil) through an optical fiber and ranged from 640 to 810 °C. The distance from the injection probe to the substrate was varied within the range of 29-35 cm. The power was adjusted to keep the substrate temperature fairly constant during the deposition time. The feeding rate was maintained at 1–2 ml/min. The film growth rates were deduced to vary within the range of 40-130 nm/min from the SEM measurements of the film thickness. The phase composition and the preferential orientation of the films were examined

using the conventional θ -2 θ scan XRD method. The *c*axis (planar) alignment of the films was determined by rocking curve analysis. The rocking curve full width at half maximum (FWHM) value obtained from a sapphire substrate was of 0.04° θ . Thin foils for cross-sectional high-resolution (HR) transmission electron microscopy (TEM) studies were prepared using a standard technique involving sticking of two film/substrate pieces together by epoxy with the films facing each other, cutting slices normal to the interface plane, and then mechanical grinding and dimpling. The specimens were attached by epoxy to a stainless steel ring for reinforcement. Subsequently, they were subjected to ion-milling to perforation at a voltage of 4 kV. The structure of the films was examined in a Hitachi-9000NAR (Hitachi, Tokyo, Japan) transmission electron microscope with the point resolution of 0.19 nm operating at 300 kV. HRTEM images were obtained with the electron beam along either the [2110]_{sub} or [1010]_{sub} zone axis. The morphology and the thickness of the deposited films were determined by a SEM. The composition of the films was determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES).

III. RESULTS AND DISCUSSION

A. Chemical composition

 $LiNb_{1-x}Ta_xO_3$ films were deposited using three different precursor solutions with the Nb:Ta ratios of 1:1, 3:2, and 7:3. Correspondingly, three sets of samples with the tantalum contents of 0.46–0.49, 0.37–0.41, and 0.26–0.30 were prepared as shown in Table I.

B. Lattice parameter

The mean *c*-axis lattice parameters of LNT films were somewhat lower than those in the bulk material (13.85 Å at x = 0.3 and 13.83 Å at x = 0.5) and decreased from 13.79 to 13.76 Å with increasing tantalum content from 0.3 to 0.5 as shown in Fig. 1. The *a*-axis lattice parameters were calculated on the basis of the (110) planes to be correspondingly larger than those in the bulk specimens of similar stoichiometry. This result is believed to be due to the effect of planar tensile stress resulting from the difference in the thermal expansion coefficients of the film and substrate. The fact that

TABLE I. Variation of precursor solution and tantalum contents of LNT films (within the instrumental accuracy of 3%).

Series	Precursor solution, Nb:Ta	Tantalum content (x) of $\text{LiNb}_{1-x}\text{Ta}_x\text{O}_3$ films
LN	1:0	0
LNT1	7:3	0.26-0.30
LNT2	3:2	0.37-0.41
LNT3	1:1	0.46-0.49



FIG. 1. *c*-axis lattice parameter of $LiNb_{1-x}Ta_xO_3$ films as a function of substrate temperature.

the lattice parameters have not changed after annealing at 750 °C for 6 h is further evidence of this assumption. This is in keeping with the experimental data by Lu et al.,⁶ who observed similar trends in the lattice parameter variations of the LN films deposited by MOCVD method. For all films the *c*-axis lattice parameter increased slightly as the substrate temperature was raised, probably due to the relaxation of film stresses.

C. Texture

Figures 2 and 3 show the XRD patterns of LN and LNT3 films obtained at various deposition temperatures. The parameter F describing the degree of c-axis orientation¹⁵ is indicated. It can be seen that their evolution with the deposition temperature was similar. The films deposited at 690 and 710 °C were characterized by the presence of a strong (006) peak indicating that the films were highly textured with the c axis normal to the sapphire substrate. The planar orientation, however, changed from the (006) to the (012) plane when the growth temperature was further raised in keeping with the previous results.^{3,24} The gradual change in texture with deposition temperature could be explained by surface free energy anisotropy. Wu et al.²⁴ have suggested that the (012)plane is the lowest energy plane. At higher temperature, nuclei on the surface can rearrange themselves to minimize total surface energy; thus, (012) orientation rather than (006) is favorable. Since film growth rates in our experiments were an order of magnitude higher than those in the work by Lee and Feigelson,³ the change in texture was observed to shift toward the higher temperatures. Note that under optimized deposition conditions, 97% c-axis oriented LNT films could be grown with the tantalum composition range of 0.3 < x < 0.5.



FIG. 2. XRD spectra of $LiNbO_3$ (LN) films at various deposition temperatures. The subscripts f and s denote film and substrate, respectively. The parameter *F* describes the degree of *c*-axis orientation.

D. Phase composition

Although the LN and LNT films deposited at lower temperatures could show a small peak of the Li-deficient phase $Li(Nb,Ta)_3O_8$, they possessed the best crystalline quality as far as the degree of c-axis orientation is concerned in comparison with other films deposited at higher temperatures where additional small reflections from the (012), (104), (110), etc., planes were visible. It is well-known that the LiM_3O_8 (M = Nb,Ta) phase is frequently observed at higher deposition temperatures^{14,24} that is usually attributed to the re-evaporation of Li from the film surface. Surprisingly, however, that the presence of a Li-deficient phase was not observed in the present study when the deposition temperature was increased. This result can be explained by the cluster deposition²³ that can have a big impact on the film phase composition. Since the LN and LNT films were deposited with high growth rates under a flux of oxygen atoms as high as 10^{18} atoms/(cm² s),²⁵ the Li-deficient clusters

would be formed in plasma before atoms make contact with substrate. In the case of cluster deposition, the partial sputtering yield of a given species could be different and re-evaporation of lithium might be suppressed. Such variations of the sputtering yield of certain material by introducing small concentrations of impurity species are well known.²⁶ When the growth temperature was further raised, the species could diffuse on the surface reducing the compositional inhomogeneity; thus, LN and LNT films were only formed.

E. Crystallinity

FWHM values of LN and LNT films at various deposition temperatures are shown in Figs. 4 and 5. Rocking curve widths were found to exhibit little temperature dependence in the range of 640–720 °C, whereas they became broader as the substrate temperature was further raised. A similar trend has been reported by Lee and Feigelson,⁴ although their MOCVD technique provided considerably lower film growth rates. The increasing of FWHM values with raising the substrate temperature can be explained by the gradual change in texture to the $\langle 01\bar{1}2 \rangle$ direction at higher temperatures. Under optimal growth conditions, the FWHM rocking curve values of the LN and LNT films were almost constant and independent on the tantalum composition. The narrowest rocking curve width was obtained for 0.25-µm-thick LNT3 film to be 0.25° θ , whereas for LN, LNT1, and LNT2 films the best values were in the range of



FIG. 3. XRD spectra of $\text{LiNb}_{0.5}\text{Ta}_{0.5}\text{O}_3$ (LNT3) films at various deposition temperatures. The subscripts f and s denote film and substrate, respectively. The parameter *F* describes the degree of *c*-axis orientation.



FIG. 4. Rocking curve FWHM values for $LiNbO_3$ (LN) and $LiNb_{0.7}Ta_{0.3}O_3$ (LNT1) films as a function of substrate temperature.



FIG. 5. Rocking curve FWHM values for $LiNb_{0.6}Ta_{0.4}O_3$ (LNT2) and $LiNb_{0.5}Ta_{0.5}O_3$ (LNT3) films as a function of substrate temperature.

 $0.25-0.35^{\circ} \theta$. Kawaguchi et al.¹⁰ and Cheng et al.²⁷ reported that the degree of crystallinity in the LNT films degrades when x > 0.3. On the contrary, the formation of LNT films with good crystalline alignment was achieved in the present study when $0 \le x \le 0.5$.

The film crystallinity as a function of growth rate was studied by varying source material input and keeping all the other parameters constant. FWHM values were found to decrease with increasing growth rate. Figure 6 shows XRD spectra of the $\text{LiNb}_{0.6}\text{Ta}_{0.4}\text{O}_3$ films deposited at various growth rates. The substrate temperature was kept constant at 730 °C. It can be seen that when the growth rate was increased from 60 to 130 nm/min, the crystallinity of films was much improved resulting in the narrowing of rocking curve width. This observation can be explained by better crystalline alignment of the films at higher growth rate. When the source input was increased, the nucleation density increased, resulting in a decrease in grain size. Therefore nucleation occurred not only on energetically favorable surface defects, which can reduce surface free energy of nuclei, but also on flat terraces where the activation energy for nucleation is high. Therefore the formation of lowest surface energy (012) texturing was much retarded at high growth rate, which led to improved film crystallinity.



FIG. 6. XRD spectra of $\text{LiNb}_{0.6}\text{Ta}_{0.4}\text{O}_3$ films deposited at various growth rates. The deposition temperature was kept constant at 730 °C. The parameter *F* describes the degree of *c*-axis orientation.

F. Crystallite morphology

Figure 7(a) shows a typical cross-sectional bright-field image of continuous 0.3-µm-thick LN film. The film displayed a columnar structure with a crystallite diameter in the range of $0.25-0.5 \,\mu\text{m}$ and a crystallite height equal to the film thickness. The film growth was also accommodated by the formation of low-angle grain boundaries. In this picture, a thin amorphous intermediate layer of about 2 nm between the film and substrate can be clearly seen. The microstructure also contained a small amorphous island adjoining the interfacial amorphous layer as shown by an arrow. Figure 7(b) shows a HRTEM image of the film/substrate interface viewed along the $[2\overline{1}\overline{1}0]_{\text{film}}/[10\overline{1}0]_{\text{sub}}$ direction. The substrate surface exhibited atomically smooth terraces and steps with the step height equal to one-third of the unit lattice height of sapphire in the [0001] direction as shown by arrows. Figure 7(b) also demonstrates that the substrate surface was mainly covered by a thin amorphous layer. Despite this, the SAED pattern taken from the film/substrate area shows that the LN film was sufficiently well *c*-axis oriented normal to the substrate surface, similar to the previous results.³

LNT1 films deposited with a feeding rate of 2 ml/min showed a much greater variation in grain morphologies; some of them are shown in Figs. 8(a) and 8(b). It can be seen that the most part of the film displayed the columnar growth mode [Fig. 8(a)] although some peculiar morphology was occasionally observed [Fig. 8(b)]. The change in grain morphology can be attributed to the contribution of a number of factors, such as influence of apparatus geometry and deposition conditions,^{30,31} orientation of nuclei on the substrate surface, or growth rate anisotropy effects.³² The c-axis is known to be the fastest growth direction, and if an individual grain is not *c*-axis oriented, the lateral growth rate may be the dominant mechanism. As shown in Fig. 8(a), the crystallite size ranged between 10 and 20 nm in lateral size before the crystallites coalesced, which correlates well with the width of contact area between the individual grain and substrate. Thus, the lateral crystallite size was mainly determined during the nucleation stage. To clarify what is a limiting factor for the lateral crystallite size, a thorough HRTEM examination was done.

Figure 9(a) presents a cross-sectional TEM micrograph showing the interface structure of LNT1 film on sapphire substrate. A number of discontinuous crystallites with a lateral size between 25 and 50 nm are visible. The substrate surface revealed well-defined surface ledges of 1–2 nm in height. These ledges, which are probably polishing defects (scratches, hillocks, etc.), were observed along the $[\bar{2}110]_{sub}$ direction and were not visible along the $[10\bar{1}0]_{sub}$ direction (compare Figs. 7 and 9). The steps on both sides of the ledges are shown in Fig. 9(a) by arrows. These steps provide lower energy



FIG. 7. Cross-sectional (a) bright-field and (b) high-resolution TEM images of as-deposited LiNbO₃ films indicating that an amorphous-like layer is formed along the interface. The insert in (b) is the SAED pattern obtained from both film and substrate. The incident beam direction is $[2\bar{1}\bar{1}0]_{film}/[10\bar{1}0]_{sub}$.

sites for nucleation. Parts of the interface at a higher magnification are shown in Figs. 9(b) and 9(c). The corresponding SAED patterns are inserted. In Fig. 9(b), the incident beam is $[2110]_{sub}$. It can be seen that the lateral crystallite size was inherited from the width of the smooth terrace.

In Fig. 9(b), the interface plane deviated about 3° and 2° from the $(0001)_{sub}$ and $(01\bar{1}\bar{1})_{film}$ planes, respectively, whereas the $(10\bar{1}1)_{film}$ plane was almost parallel to the $(01\bar{1}\bar{1})_{sub}$. The orientation relationship between the film and substrate were deduced to be

$$(0\bar{1}11)_{\text{film}} \approx //(0001)_{\text{sub}};$$
 (OR–I)
 $(10\bar{1}1)_{\text{film}} \approx //(01\bar{1}\bar{1})_{\text{sub}}$.

This result indicates that the topmost atomic plane of the ledge was different from the (0001) sapphire plane. Moreover, the ledge sides were not straight suggesting that the side planes were also different from the $\{11\overline{2}0\}$ planes. Thus, an island nucleating on ledge edge may possess the orientation relationship with substrate different from the epitaxial relationship $(0001)_{film}//(0001)_{sub}$ and $[11\overline{2}0]_{film}//[11\overline{2}0]_{sub}$ even if the substrate is *c*-axis oriented. As a result, the film crystallites with other orientations might nucleate in favor of the [0001] out-of-plane orientation. Note that the surface morphology of substrate can be changed by high-temperature annealing.^{5,29}



FIG. 8. Cross-sectional bright-field TEM images of $LiNb_{0.7}Ta_{0.3}O_3$ (LNT1) films showing various film morphologies.



FIG. 9. Cross-sectional (a) TEM and (b, c) HRTEM micrographs showing the nucleation of LNT crystallites on the ledge terraces of sapphire substrate. The incident beam direction is (b) $[2\bar{1}\bar{1}0]_{sub}$ and (c) $[1\bar{1}00]_{film}//[2110]_{sub}$.

The HRTEM micrograph of Fig. 9(c) shows the atomic structure of the interface between the film and the surface ledge of substrate. In this figure, the film/ substrate interface was almost parallel to the (0001) plane of sapphire. Similar to the previous case, the substrate ledge directly affected the film morphology. The image indicates no amorphous layer between the film and substrate ledge, whereas the areas of film adjoining the ledge appeared to be amorphous. The incident beam direction in Fig. 9(c) is $[1\bar{1}00]_{film}//[\bar{2}110]_{sub}$, and the $11\bar{2}0$ reflection of film is almost on the line drawn through the center and $01\bar{1}2$ reflection of sapphire; thus the OR-II was fulfilled within a few degrees as follows:

$$[1\overline{1}00]_{\text{film}}//[\overline{2}110]_{\text{sub}}$$
 (OR–II)
 $(11\overline{2}0)_{\text{film}}//(01\overline{1}2)_{\text{sub}}$,

G. Film/substrate crystallography

The partially *c*-axis-oriented LN and LNT films deposited on a (0001) sapphire substrate showed several peaks representing the diffraction from planes ($10\overline{1}2$), ($10\overline{1}0$), ($01\overline{1}1$), ($11\overline{2}6$), etc. Thus, the out-of-plane orientation of a number of grains was different from the [0001] orientation. Since the orientation of crystallites with columnar morphology is determined at the nucleation stage, the question arises as to whether any rational orientation relationship exists between the crystallites and substrate or these crystallites are randomly oriented.

The OR-I reported above is the case where the $(01\overline{1}\overline{1})_{\text{film}}$ plane was almost parallel to the (0001) closepacked plane of substrate. This OR was obtained in four cases and was fulfilled with high accuracy. The OR-II was also observed with high accuracy (in five cases), and it corresponds to the case when the $(11\overline{2}\overline{6})_{\text{film}}$ plane was almost parallel to the (0001)_{sub} plane. Although we have not accumulated enough data for other orientations observed, the present results imply that most of the crystallites with an out-of-plain orientation different from the [0001] possessed a rational orientation with the substrate. This is in keeping with the experimental data by Fujimura et al.,²¹ who reported that the epitaxial orientation of LiNbO₃ films could be changed by varying the lithium concentration. The present result is also consistent with the cluster deposition hypothesis mentioned above. Table II shows that the orientation relationships I–II yield a small misorientation between the coincident planes, some of them providing even a better misfit than the epitaxial relationship (0001)_{film}//(0001)_{sub} and $[1120]_{film}/[1120]_{sub}$. Thus, it is expected that low interface energy between the film and substrate could be maintained under different ORs.

H. Growth mechanism

It is well known that there are two basic types of film growth on atomically clean surfaces, which are termed "island growth" (Volmer-Weber mechanism) and "layer growth".³³ The latter is often subdivided into two modes: two-dimensional layer growth (Frank-van der Merve) and layer-plus-island growth (Stranski-Krastanov).³³ In the last case the growth begins as a thin layer but develops islands and particles as growth proceeds.³⁴ The nucleation and coalescence of three-dimensional islands of LN deposited on *c*-plane sapphire substrate by pulsed laser deposition and the MOCVD method have been studied by Veignant et al.^{16,28} and Lee and Feigelson,⁴ respectively. These results demonstrated that the nucleation and growth of LN films proceeded by Volmer-Weber island type growth mode. This contradicts the data by Lee at al.,³⁵ who observed the Stranski-Krastanov growth mode with the critical thickness from two-dimensional to three-dimensional growth at 1 nm. Note that most HRTEM results reported on LN/Al₂O₃ indicate that very sharp interfaces are formed with no intermediate layers.^{2,16,28} The substrate surface structure is usually important for film nucleation and morphology. In the present study film nucleation occurred on the step surface, and island grew both vertically and laterally but did not cross a step edge (Fig. 9). Therefore the surface ledges suspend true layer growth. This result shows that

TABLE II. Lattice misfit between the film and substrate.

Lattice spacing,			Lattice spacing,		
$(hkl)_{\rm film}$	$d_{\rm film}$ (nm)	(hkl) _{sub}	$d_{\rm sub}$ (nm)	$(d_{\rm film}-d_{\rm sub})/d_{\rm film}$	OR
(006)	2.311 ^a	(006)	2.1654 ^b	0.063	Usual OR
(110)	2.576	(110)	2.3795	0.076	
(202)	2.124	(006)	2.1654	-0.019	OR-I
(202)	2.124	(202)	1.9643	-0.075	
(300)	$1.487 \times 3 = 4.461$	(110)	$2.3795 \times 2 = 4.759$	-0.066	OR-II
(110)	$2.576 \times 3 = 7.728$	(012)	$3.4797 \times 2 = 6.9594$	0.099	

^aLiNbO₃, Card No. 20-631, JCPDS.

^bAl₂O₃, Card No. 46-1212, JCPDS.

the terrace width is a limiting factor controlling the lateral grain size under the conditions employed in the present study. Nucleation was also observed to occur on terraces between ledges where either the amorphous or crystalline phase was formed. As the crystallites coalesced, the formation of amorphous phase was almost prevented. The Volmer–Weber particle growth mode implies that an amorphous either layer or patches does not form at all. Thus, in the case under consideration, the film growth type cannot be identified with any of basic types of growth.

IV. CONCLUSIONS

The thermal plasma spray chemical vapor deposition method has been shown to be well suited for the high-rate growth of highly *c*-axis-oriented $\text{LiNb}_{1-x}\text{Ta}_x\text{O}_3$ films with the tantalum composition range of $0 \le x \le 0.5$ on a c-plane sapphire substrate. They were compared with the LiNbO₃ films deposited under similar conditions. The film/substrate interface was thoroughly studied to reveal the morphology and the crystallography of film crystallites on the early stage of growth. The HRTEM analysis of the film/substrate interface showed that the surface of as-received sapphire substrate revealed well-defined polishing defects, ranging from 1 to 2 nm in height with smooth terraces of 25 nm in width. In the case of columnar growth mode, the terrace width was shown to be a limiting factor controlling the lateral crystallite size. Various crystallite morphologies and growth orientations were observed. Under optimal deposition conditions, the LNT films were 97% c-axis oriented. The film crystallographic orientation was strongly affected by the CVD process parameters; the planar orientation of the LN and LNT films changed from the (0001) to the $(01\overline{12})$ plane by either decreasing the film growth rate or increasing the substrate temperature. It was suggested that individual crystallites possess rational orientation relationships with substrate despite their various out-ofplane alignments. Further research is needed to clarify the crystallography of partly *c*-axis-oriented films. Rocking curve FWHM values of LNT films did not appear to depend strongly on the tantalum composition. The LN and LNT films deposited under optimal growth conditions showed FWHM width as low as $0.25^{\circ} \theta$. They were under compressive strain along the c direction; c- and a-axis lattice parameters were correspondingly smaller and higher than those of the bulk material.

ACKNOWLEDGMENTS

D.V.S. and S.A.K. acknowledge the support of the Japan Society for the Promotion of Science (JSPS) during this work. This work was financially supported by JSPS under the program "Research for the Future" (JSPS-RFTF Grant No. 97R15301).

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