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m-line spectroscopy for optical analysis of thick $LiNbO_3$ layers grown on sapphire substrates by radio-frequency multistep sputtering

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Thick films of lithium niobate ($LiNbO_3$) have been epitaxially grown on sapphire substrates by multistep radio-frequency magnetron sputtering. We have investigated the optical properties of the deposited layers by the *m*-line spectroscopy technique. The relationship between the film-substrate interface, the surface morphology of the $LiNbO_3$ layer, and the optical measurements has been carefully established. Since this optical technique is sensitive to material defects, a detectable degradation of the optical data is observed when the microstructure of the $LiNbO_3$ film changes. In particular, the angular width of the excited optical guided modes ($\Delta\theta$) varies with the film thickness. This agrees perfectly with the evolution of the surface roughness as a function of the film thickness. We report a surface roughness r_{ms} of 2 nm corresponding to an angular width $\Delta\theta$ of 0.1° for the TE_0 mode. The accuracy and the sensitivity of this technique can be advantageously compared to the classical tool of atomic force microscopy. © *2003 American Institute of Physics.*
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I. INTRODUCTION

Lithium niobate ($LiNbO_3$ referred to as LN) has received increasing attention due to its applications in optical communications and wireless systems. $LiNbO_3$ is also widely used in surface acoustic wave devices, sensors, and light modulators. The performance of these devices is strongly affected by the material properties and numerous efforts have been made to improve the quality of $LiNbO_3$ films. Various techniques have been used for the deposition of $LiNbO_3$ films on different substrates, such as sol-gel,[1] pulsed laser deposition,[2] metal-organic vapor deposition,[3] and sputtering.[4,5] Typically, the film thickness is very thin and it is difficult to increase. The main difficulties are the formation of cracks and an important roughness when the film thickness increases. For many applications such as optical waveguides, surface acoustic wave filters, etc., it is imperative to deposit thick films (>0.5 μm). The growth of thick LN films is the first challenge. In order to characterize the morphology of the layers and, in particular, the roughness, the conventional techniques used are scanning electron microscopy (SEM) and atomic force microscopy (AFM). These techniques give fairly accurate results but require special sample preparation. A single measurement technique capable of determining the film thickness, the refractive index, the interface properties,[6] the surface roughness, and the composition[7] with a quick turn-around time can be extremely valuable to any growth program. In this work, optical measurements performed using *m*-line spectroscopy have provided rich information about film microstructures. A correlation is given between the surface roughness obtained by standard AFM and the optical results.

II. FILM DEPOSITION

$LiNbO_3$ films have been grown *in situ* on (0001) sapphire substrates by radio-frequency magnetron sputtering. The structure and the microstructure of the films have been correlated directly with the growth parameters. Structural analysis of the $LiNbO_3$ films using x-ray diffraction (with $\theta/2\theta$ configuration, rocking curve, and φ scans) and transmission electron microscopy (TEM) has been performed. The film morphology was investigated by SEM and AFM analysis, and also by the optical measurements. From the matrix of experiments, we have established possible transfer functions for the deposition of epitaxial $LiNbO_3$ layers. Table I describes the sputtering parameters related to these transfer functions TF_1, TF_2, and TF_3). Note that the substrate temperature during the deposition was fixed to 490 °C and the substrates positioned off axis. The growth parameters (pressure, radio-frequency power, target-substrate spacing) impact the growth deposition rate and the roughness. The growth of an epitaxial thick layer with adequate smoothness presents a very difficult challenge, particularly for $LiNbO_3$ materials. Thickening of the layers (from 120 to 650 nm) using TF_1, TF_2, or TF_3 was accomplished by using a multistep growth process: It is based on the successive growth of $LiNbO_3$ layers and associated with the well known regrowth process currently used in semiconductor technologies. This process is described in recent work.[8]

As observed from SEM analysis, the multistep growth technique improves the surface morphology in comparison to a single growth step of equivalent thickness. The $LiNbO_3$ film has a good density after each step of deposition and no significant degradation of the surface morphology is noticed. AFM investigations have shown that the surface morphology depends mainly on the deposition pressure and the film

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TABLE I. Transfer functions studied for the epitaxial growth of $LiNbO_3$ films on sapphire.

	Pressure (mtorr)	Gas mixture (Ar/O_2)	rf power ($W\ cm^{-2}$)	Distance (mm)	Growth rate (nm/h)
TF_1	10	60/40	1.3	80	30
TF_2	20	60/40	2.36	80	70
TF_3	30	60/40	3	60	120

thickness. Figure 1(a) shows the AFM image of a 120 nm LN film on sapphire for one step of deposition using the TF_1 process. Small grain sizes are around 50 nm and the surface roughness as expressed by r_{ms} is 2.1 nm. Figure 1(b) shows the AFM image of the surface microstructure of a 650 nm thick film $LiNbO_3$ film obtained from a multistep growth process. The grain size is around 110 nm and r_{ms} is 6.9 nm. The above observations suggest that film roughening can be controlled by increasing the layer density if the grain size can be kept small without sacrificing the crystalline quality. Figure 2 describes the evolution of the surface roughness r_{ms} versus the film thickness. Samples are prepared under

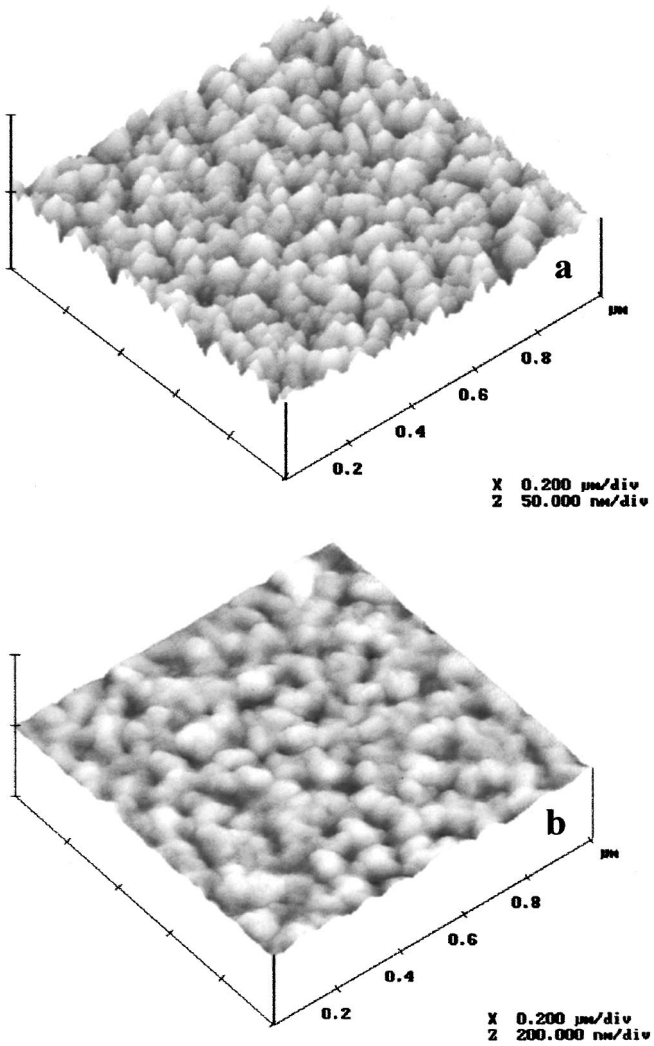

FIG. 1. AFM image of the surface microstructure for a 120 nm $LiNbO_3$ thin film from a single step growth process (a) and for a 650 nm thick $LiNbO_3$ film from a multistep growth process (b).

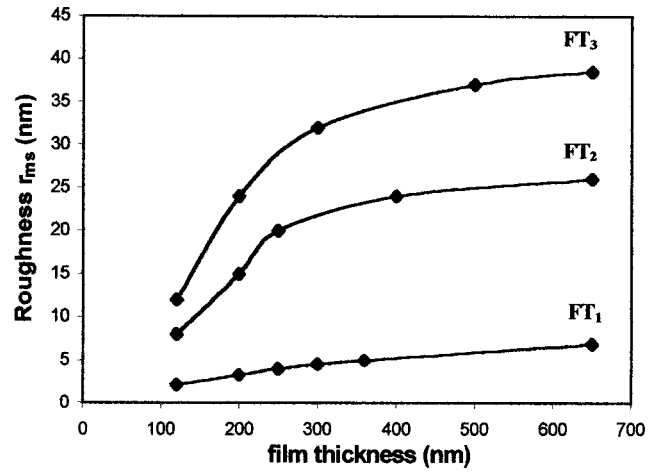

FIG. 2. Evolution of surface roughness r_{ms} with the $LiNbO_3$ film thickness.

TF_1, TF_2, and TF_3 conditions. As is well known, the surface roughness increases with the film thickness. We observe that r_{ms} increases linearly when the thickness is relatively small and is stabilized by application of the multistep process. This trend is observed for TF_2 and TF_3 while TF_1 shows a moderate roughness even for thicker films, probably due to the low growth rate of 30 nm/h. We demonstrate that with a multistep process the surface morphology can be controlled whatever the film thickness. To explain this effect, the $LiNbO_3$ layers could be fully relaxed after each step of deposition. The first LN layer could also be considered as a seed layer which acts as a stress absorber. Controlling the initial growth stage therefore is very important since the grain size can be controlled by changing the sputtering parameters. Fork *et al.*[9] and Lee *et al.*[10] have studied these mechanisms that cause the roughness to increase with the film thickness.

III. OPTICAL MEASUREMENTS

In order to quantify the optical properties of $LiNbO_3$, the prism coupling technique, also referred to as the *m*-line spectroscopy technique, has been widely used. This technique, which is based on optical guided waves, provides information on the thickness and refractive index of various layers in multilayered structures. To illustrate its usefulness, we have studied the case of $LiNbO_3$ films grown by the multistep process. We have analyzed the optical information to qualify the film surface morphology and to compare it with AFM data. This technique, which is described in Ref. 11, is ideal for investigating the main optical characteristics of $LiNbO_3$ layers without destruction of samples. In particular, the film thickness and the optical constants can be determined accurately at any point of the sample surface from optical information. In the experiment setup presented in Fig. 3, a right angle rutile prism (angle $A_p = 45°$ and index $n_{TE} = 2.865$) is used to couple the laser beam (wavelength 632.8 nm) to the guided modes of the structure. The layer is pressed against the base of the prism. By measuring the reflected intensity versus the angle of incidence θ, we draw the guided mode spectrum of the samples. The reflectivity dips in the spectrum are correlated with the incident angles that serve to

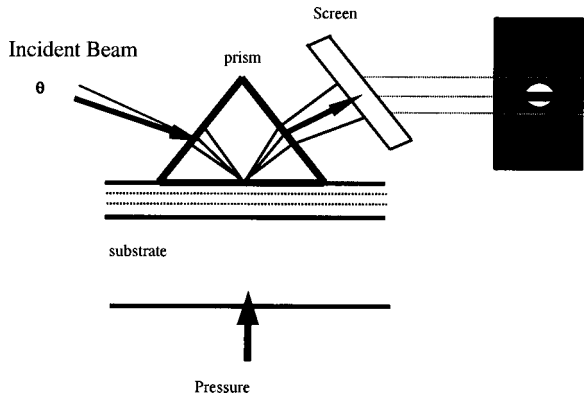

FIG. 3. Schematic view of the *m*-lines spectroscopy setup.

determine the effective mode indices. An example of a reflectivity spectrum is reported in Fig. 4 where three TE guided modes are excited in the 650 nm thick LN layer. Reflectivity dips indicate the excitation of specific guided modes in the LN film and we can see their sharpness. This is a good indication of light confinement in the waveguide. By determining the angular position of the dips in the reflected intensity ($\theta_{TE0}=+19.080°$, $\theta_{TE1}=+7.466$, and $\theta_{TE0}=-7.373°$), the index of refraction and thickness can be calculated simultaneously ($n_0=2.289$ and $n_e=2.206$ at $\lambda=632.8$ nm).

The optical coupling is generally sensitive to a number of factors such as surface and interface roughness. From knowledge of the effective mode indices, the refractive index profile (n) has been determined as a function of the film thickness for either TE or TM modes. The inverse Wentzel-Kramers-Brillouin method has been previously described in the literature by Dogheche *et al.*, for nitride materials.[6] For example, the refractive index profile is reported in Fig. 5. It indicates a steplike index variation, which is synonymous with a good optical homogeneity along the film thickness. The refractive index remains constant within the guiding region and decreases rapidly near the film-substrate interface. In the present study, we remark that the sputtering multistep

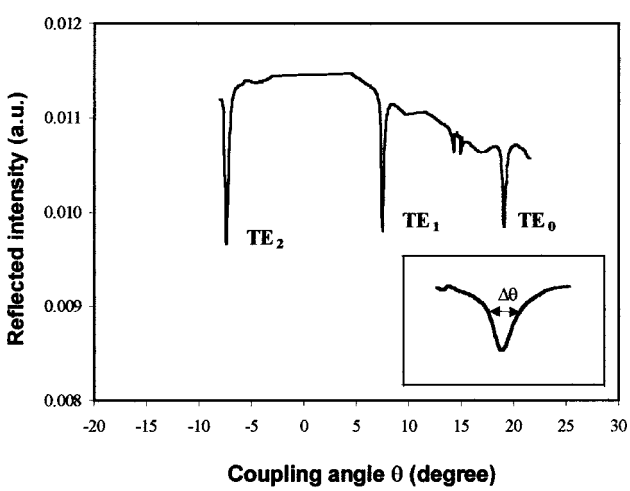

FIG. 4. Guided-mode spectrum obtained by measuring the reflected intensity versus the angle of incidence (transverse electric TE excitation) and definition of the angular width ($\Delta\theta$) of the excited optical guided modes.

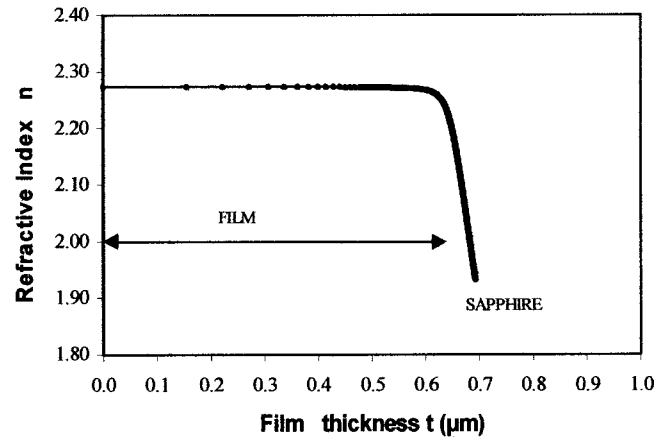

FIG. 5. Reconstruction of the refractive index profile for $LiNbO_3$ using an improved inverse WKB method.

process does not affect the optical properties of the films resulting from different steps, and the index of refraction is not modified in the region near the interface. In previous work, we verified by TEM analysis that there is no interaction between the film and the substrate during the multistep growth process.

Of particular concern is therefore the surface roughness of the films. In fact, the extracted energy from the reflected beam leads to sharp or no dips in the reflectivity curve. This is related to the film morphology. In general, the dip width corresponding to the TE_0 mode is very narrow for smooth films. Different authors have suggested a relationship between the film microstructure and optical properties. In particular, Wessels *et al.*[12] studied the influence of surface roughness on light scattering for ferroelectric oxide films and they indicates that the surface roughness requirements may be 2–4 nm in order to obtain low optical scattering losses of 1–2 dB/cm. Graupner *et al.*[13] explained that the angular width of the dips originates in the whole scattering loss of the LN layer. In this work we have measured the angular extent of the optical guided modes, particularly the full width at half maximum ($\Delta\theta$) of TE_0 guided modes. In particular, we have analyzed the evolution of $\Delta\theta$ as a function of the film thickness. different cases of transfer function have been considered (TF_1, TF_2, and TF_3). By increasing the surface roughness of the layer, the scattering of light by the surface roughness induces considerable change in the width of the TE_0 modes. This behavior is similar to that observed in the AFM analysis (Fig. 2). A correlation can be established between the angular width of the optical modes as expressed by $\Delta\theta$ and the surface roughness r_{ms}. As plotted in Fig. 6, we find that $\Delta\theta$ of 0.1° is related to a roughness r_{ms} of 2 nm. The accuracy for $\Delta\theta$ was estimated to be 10^{-3}. So the prism coupling technique has been used as a sensitive surface probe. The optical method developed can be favorably compared to the AFM with good accuracy.

IV. CONCLUSION

In summary, $LiNbO_3$ films have been deposited on (0001) sapphire substrates by the rf magnetron sputtering. Thickening was achieved by multistep deposition. The

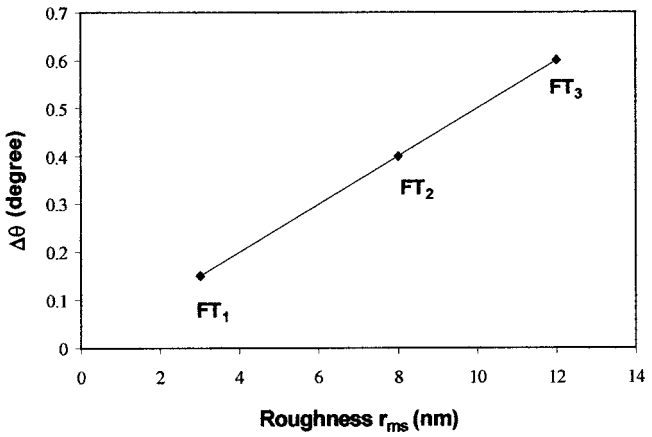

FIG. 6. Relationship between the width of dips ($\Delta\theta$) and the surface roughness r_{ms}.

m-line spectroscopy based on the optical coupling of guided waves by a prism was used for the optical characterizations. The effect of the surface and interface quality on the optical measurements was studied. The film homogeneity and the film-substrate interface were analyzed directly from the experimental optical data. Comparing the morphological investigations by AFM, the optical properties have been found to be very promising. The evolution of the surface roughness (r_{ms}) as a function of the $LiNbO_3$ film thickness is comparable to that of the full width at half maximum ($\Delta\theta$) of the TE_0 guided modes excited inside the layer. We found a close relationship between $\Delta\theta$ and the surface roughness r_{ms}. $\Delta\theta$ of 0.1° is associated with r_{ms} of 2 nm. This might be the main reason for allowing the prism coupling analysis to act as a practical characterization tool in materials growth programs.

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