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Short Communication

Piezoelectric Characteristics of $LiNbO_3$ Thin-film Heterostructures via Piezoresponse Force Microscopy

D.A. Kiselev[1,*], S.V. Ksenich[1], R.N. Zhukov[1], A.S. Bykov[1], M.D. Malinkovich[1], V.V. Shvartsman[2], D.C. Lupascu[2], Yu.N. Parkhomenko[1]

[1] *National University of Science and Technology "MISiS", 4, Leninskiy Pr., 119049 Moscow, Russian Federation*
[2] *Institut für Materialwissenschaft, and Center for Nanointegration Duisburg-Essen (CENIDE), Universität Duisburg-Essen, 15, Universitätsstr., 45141 Essen, Germany*

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Electro-optic $LiNbO_3$ thin films were deposited on Si(100) and Si(111) substrates using a radio-frequency magnetron sputtering process. The piezoelectric properties of the $LiNbO_3$ films were investigated using the scanning probe microscopy in the piezoresponse mode. The obtained results show the high degree of grains orientation in polycrystalline structure. The piezoelectric modulus (d_{zz}) was estimated to be 16 pm/V (for $LiNbO_3$/Si(100)) and 22 pm/V (for $LiNbO_3$/Si(111)) and the polarization about of 0.37 $C \cdot m^{-2}$. These values are larger than those reported previously for $LiNbO_3$ films.

Keywords: $LiNbO_3$ thin film, Piezoresponse force microscopy, Domain structure.

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1. INTRODUCTION

Among the most interesting and promising directions in the physics of optics and materials science is the study of the synthesis and characterization of thin film gradient optical structures [1]. The results already obtained in this area are of great practical importance. In particular, created by ultra-wide range (for a visible, near and mid-IR range) and deep modulation on the beam splitter, reflector less optical phase shifters, compressors femtosecond optical pulses that work in non-prism mode transient tunneling narrow-band filters in the visible and near – infrared regions of the spectrum etc. [2].

As the next step in the development of this area is important to develop a new class of optical elements - gradient optical "transformers" on the basis of gradient thin film coatings, i.e. the gradient of thin-film multilayer structures, the characteristics of which may vary considerably during their work by changing the optical thickness of a certain number of sub-elements. For example, the reflection of a certain wavelength range will change his passing, etc. This will greatly simplify the optical circuit devices, in some cases, remove the multiple channels to reduce the weight and dimensions. This is possible when creating one-layers in multilayer structures of electro-optic materials, such as lithium niobate. However, this procedure requires the synthesis of dielectric substrates oriented both crystallographic direction and the polarization vector by fine film of lithium niobate with 100 nm thickness.

Oriented polycrystalline lithium niobate ($LiNbO_3$, LN) thin films are also of interest for a variety of electro-optical and acousto-optical applications, including integrated device structures containing micro- and opto-electronic components. The ferroelectric properties of LN films were investigated by piezoresponse force microscopy (PFM), which allows identifying the polarization direction and the local domain distribution [3]. Previously [4-6], this method has already been used to visualize ferroelectric domains in LN films deposited on conductive silicon substrates.

In this article we report on direct investigation of domain structures and local piezoelectric properties of $LiNbO_3$ thin films by the PFM method.

2. EXPERIMENTAL DETAILS

The $LiNbO_3$ films were deposited on n-type Si (100) and Si (111) substrates ($\rho = 2\ \Omega \cdot$cm) by RF magnetron sputtering of the single-crystalline target in Ar / O = 1 atmosphere and under the working pressure of 0.6 Pa. The thickness of the $LiNbO_3$ thin films was about 100 nm. After sputtering process the films were annealed in a furnace at 700 °C for two hours. The piezoelectric properties of the LN thin films were characterized by the piezoresponse force microscopy using commercial scanning probe microscopes MFP-3D (Asylum Research) and NTEGRA Prima (NT-MDT). Out-of-plane PFM images of the samples were obtained by applying AC voltage (5V peak-to-peak) with the frequency of 5 kHz. To address the DC voltage dependence of the local piezoresponse so-called remnant piezoelectric loop (REM hysteresis) were measured by applying a sequence of DC voltage pulses with cycling magnitude and collecting the PFM signal after each pulse. The measurements were done in a DC voltage range – 40 V $\leq V_{DC} \leq$ + 40 V. Both DC voltage pulse duration and measuring time were 1 sec.

3. RESULTS AND DISCUSSIONS

The XRD was performed with a $\theta - 2\theta$ configuration and the results for the samples are summarized in Fig. 1.

The XRD spectra show several peaks from 20° to 60° for polycrystalline $LiNbO_3$ films, that correspond to the diffraction contribution from the (012), (104), (006), (116).

* dm.kiselev@misis.ru

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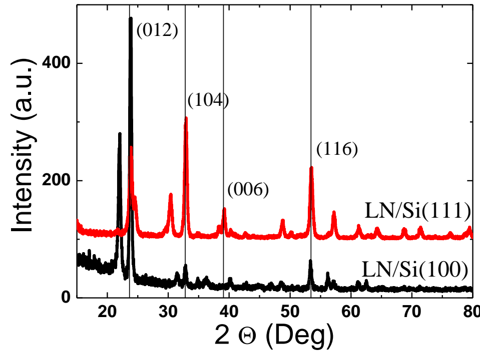

Fig. 1 – XRD spectra and phase analysis for $LiNbO_3$ thin films

The surface morphologies of the $LiNbO_3$ films synthesized on silicon substrates with (100) and (111) orientations observed by PFM as shown in Fig 2a, d.

The surface roughness was about 6 nm and 8 nm for the $LiNbO_3$/Si(100) and $LiNbO_3$/Si(111) heterostructures, respectively. This result meets the demands for practical wave guiding devices. Lateral relief correspondence grain sub-structure of the LN films with average grain diameter of 50 nm and 75 nm for the films on Si(100) and Si(111) substrates, respectively.

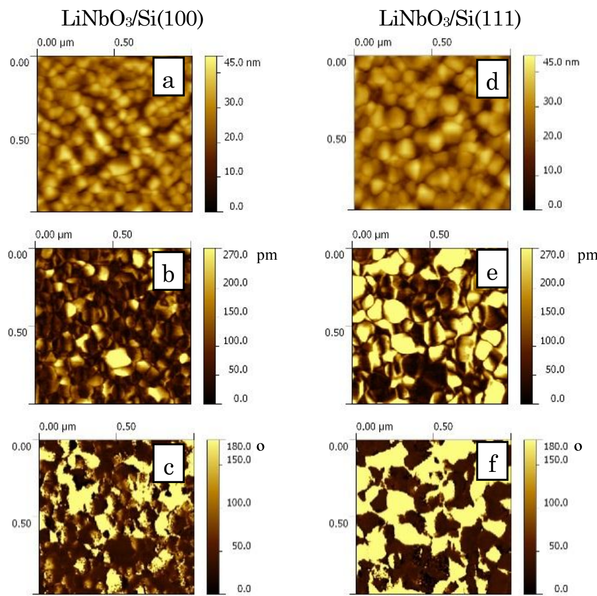

Fig. 2 – Simultaneously obtained topographic (a, d), amplitude (b, e) and phase (c, f) PFM images for LN thin films, sputtered on (100) and (111) oriented silicon substrates

PFM was used to characterize the polarization states of the LN films and to determine the piezoelectric constants. Figs. 2b, e and c, f show the amplitude and phase PFM images, respectively, obtained simultaneously with the topography images. The amplitude of the PFM signal corresponds to a local deformation of the sample under an alternating electric field i.e. provides values of the local piezoelectric coefficient d_{zz}. Phase images show grains with «bright» and «dark» contrast, which correspond the orientation of the polarization vector up and down relative to the film plane.

Fig. 3a-c represent the PFM amplitude images obtained at different ac probing voltage values 3 V, 7 V and 11 V (scan size 1 × 1 μm). It is clear that the piezoresponse amplitude increases with the increasing magnitude of the AC voltage. Fig. 3d shows the corresponding histograms of the amplitude PFM signal for the PFM image obtained by ac voltage varied from 1 to 11 V. In both cases this histograms can be identified as Gaussian distributions with a single narrow maximum.

The position of this maximum corresponds to a mean piezoresponse signal for a given image. The AC voltage dependences of this mean piezoresponse a for LN film are shown in Fig. 4. As expected, the piezoresponse amplitude increases linearly with the voltage. The mean d_{zz} piezoelectric coefficient can be evaluated from the slope of these dependences (Fig. 4) according the formula [7]:

$$d_{zz} = \frac{A}{V_{AC}}, \quad (1)$$

where A (pm) is the piezoresponse amplitude and V_{AC} (V) is the amplitude of the applied voltage. The best fit yields for $LiNbO_3$/Si(111) heterostructures the value of $d_{zz} \approx 22$ pm/V and for $LiNbO_3$/Si(100) we obtained a slightly smaller $d_{zz} \approx 16$ pm/V. The measured d_{zz} values are close to the d_{33} value for LN single crystal [8], and nanocrystalline particles based on $LiNbO_3$ [9].

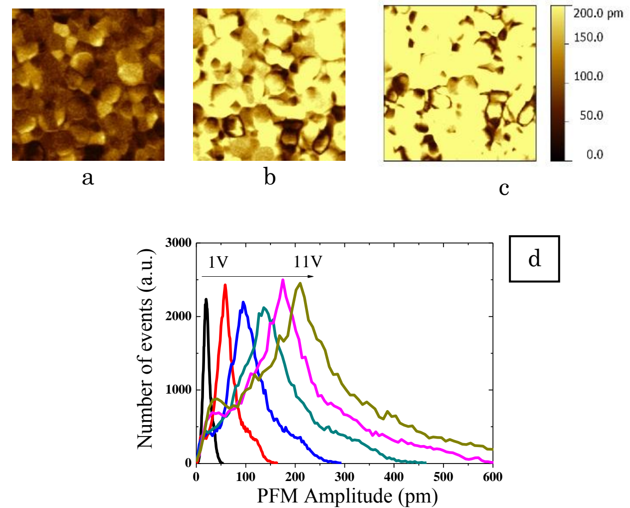

Fig. 3 – Out-of-plane PFM amplitude images taken at 3 V (a), 7 V (b), and 11 V (c), for $LiNbO_3$ thin film, and corresponding histograms (d) extracted from PFM amplitude images

On the other hand, the piezoelectric coefficient is proportional to the polarization, P:

$$d_{zz} = 2Q\varepsilon\varepsilon_0 P, \quad (2)$$

where Q is the electrostriction coefficient ($Q \approx 0.095$ m^{4}C^{-2} for bulk LN [10]), ε_0 is the permittivity of vacuum, ε is the relative dielectric permittivity. Taking the relative dielectric permittivity of the LN to be about 30 at low frequency, the polarization for $LiNbO_3$/ Si(111) heterostructure can be estimated to be ~ 0.37 C·m^{-2}, which is relatively large in comparison with the value reported previously for LN films [11, 12].

To get information about the anisotropy of the lon-

gitudinal piezoelectric coefficient d_{zz} of LN thin films, the remnant piezoelectric loops were measured shown in Fig. 5.

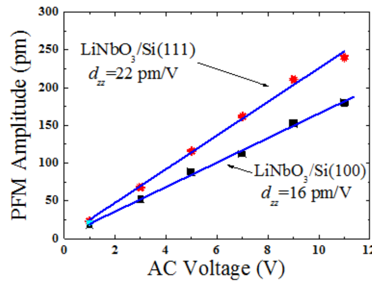

Fig. 4 – The mean PFM amplitude signals versus AC voltage for the LN thin film

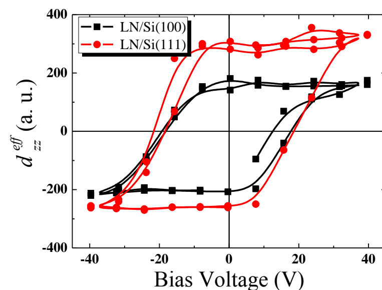

Fig. 5 – REM piezoelectric hysteresis loops for the $LiNbO_3$ films with different orientations of Si substrate

It is clear that the remnant piezoelectric response for the LN films synthesized on Si (111) is larger than for the films synthesized on Si (100). Accordance with to the imaging principle of the PFM, the value of d_{zz} for grains with the polarization vector perpendicular to the film surface is higher than that for grains with the polarization vector deviating from the direction of the surface normal [13]. Therefore, the piezoelectric results shown in Fig. 4 and 5 suggest that the presence of differences in the values of piezoelectric modules (polarization vector) for LN thin films synthesized by varying the substrate orientation.

4. CONCLUSIONS

Piezoresponse force microscopy technique was used to investigate the local piezoelectric properties of the electro-optic $LiNbO_3$ thin films. The piezoelectric response of the films depends on the crystallographic orientation of the silicon substrate. The piezoelectric coefficient (d_{zz}) was determined to be 16 pm/V and 22 pm/V for $LiNbO_3$/Si(100) and $LiNbO_3$/Si(111) heterostructures, respectively. The polarization was estimated to be ~ 0.37 $C \cdot m^{-2}$. These values are higher than previously reported values for LN films. The results indicate that one of the problems the solution of which is necessary for the creation of electro-optic lithium niobate thin films on dielectric layers, namely the high crystallographic orientation of the structure can be solved by means of RF sputtering technology.

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REFERENCES

1. O.D Volpian., A.I. Kuzmichev, *Negative Refraction of Waves. Keeping to the Physics and Technology of Electromagnetic Metamaterials* (Kiev: Avers: 2012).
2. O.D. Volpian, S.I. Shkatula, M.D. Malinkovich, *The Defense Industry - Scientific and Technical Progress Russia* No4, 22 (2012).
3. V. Bornand, B. Gautier, P. Papet, *Mater. Chem. Phys.* **86**, 340 (2004).
4. R.N. Zhukov, A.S. Bykov, D.A. Kiselev, M.D. Malinkovich, Yu.N. Parkhomenko, *J. Alloy. Compd.* **586**, S336 (2014).
5. R.N. Zhukov, D.A. Kiselev, M.D. Malinkovich, Yu.N. Parkhomenko, E.A. Vygovskaya, O.V. Toropova, *Russ. Microelectronics* **41**, 459 (2012).
6. D.A. Kiselev, R.N. Zhukov, A.S. Bykov, M.D. Malinkovich, Y.N. Parkhomenko *PIERS Proceedings, 520* (August 19 – 23: Moscow: Russia: 2012).
7. K.Ch. Chiu, T.H. Yang, J.M. Wu, *Nanotechnology* **24**, 225602 (2013).
8. M.P. Shaskol'skaya, *Acoustic Crystals: A Handbook*. (Moscow: Nauka: 1982).
9. D. Mohanty, G.S. Chaubey, A. Yourdkhani, S. Adireddy, G. Caruntu, J.B. Wiley, *RSC Advances* **2**, 1913 (2012).
10. *Piezoelectric Actuators and Ultrasonic Motors, Electronic Materials: Science & Technology, **1*** (Ed. K. Uchino) (1996).
11. D.G. Lim, B.S. Jang, S.I. Moon, C.Y. Won, J. Yi, *Solid-State Electron.* **45**, 1159 (2001).
12. S.W. Jung, Y.S. Kim, S.H. Jeong, Y.I. In, K.H. Kim, K. No *J. Korean Phys. Soc.* **42**, 1386 (2003).
13. O. Auciello, A. Gruverman, H. Tokumoto, S.A. Prakash, S. Aggarwal, R. Ramesh, *MRS Bull.* **23**, 33 (1998).