Size effects in LiNbO3 thin films fabricated by pulsed laser deposition

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Abstract. This paper shows the results of the study of size effects of electro-physical parameters in nanostructured LiNbO3 thin films fabricated by pulsed laser deposition. Obtained results shown that with an increase in the number of laser pulses from 50 000 to 150 000, the maximum value of the amplitudes (psi) increases from 36 to 47, and phase shift (delta) varies from 160 to 175. It was found that with increasing of the film thickness from 45.3 to 192.9 nm, charge carrier concentration in the films increasing from 4.1·1012 to 8.75·1012 cm−3. An increase in the grain diameter from 118 to 172 nm causes a gradual increase in charge carriers mobility from 125.714 to 505.841 cm2/V·s.

1. Introduction
Currently, lithium niobate (LiNbO3) bulk crystals are widely used in acousto-optic [1] and piezoelectric devices as well as waveguide structures with low losses [2, 3]. Due to its physical properties LiNbO3 can be considered as the backbone of modern integral electronics. Thus, the formation of LiNbO3 thin films on a silicon substrate will allow combining optical and electronic components on the same chip [4].

LiNbO3 thin films can be fabricated by the following growth techniques: molecular-beam epitaxy [5], sol-gel method [6], magnetron sputtering [7], and pulsed laser deposition (PLD) [8, 9]. The latter is the most suitable for complex oxides since it has many technological parameters, as well as the ability of preserving the stoichiometric composition of the target [10]. Moreover, PLD allows obtaining films in a wide range of electro-physical and optical properties, which in turn depend on the morphological parameters of the growing films. Therefore, the purpose of this paper is studying the influence of size effects in LiNbO3 films on their electro-physical and optical properties.

2. Experiment
In order to fabricate LiNbO3 films nanotechnological facility NANOFAB NTK-9 (NT-MDT, Russia), comprising PLD module Pioneer 180 (Neocera Co., USA) was used. The LiNbO3 target was ablated
by excimer KrF laser (\(\lambda=248\) nm) (Coherent Inc., USA) with energy density on target surface about 1.5 J/cm\(^2\). The number of laser pulses varied from 50 000 to 200 000 at repetition rate of 10 Hz. The argon pressure in the growth chamber varied from 10\(^{-3}\) to 10\(^{0}\) Torr. Films with a thickness from 45 to 180 nm were deposited at a temperature from 300 to 600 °C on Si and SiO\(_2\) substrates.

The morphology of obtained films was studied by atomic force microscopy (AFM) in semi-contact mode using Ntegra probe nanolaboratory (NT-MDT, Russia). Electro-physical parameters of the films have been studied by measuring the Hall electromotive force at Ecopia HMS-3000 system (Ecopia Co., Korea). The dependences of the optical characteristics were studied on spectral ellipsometer M-2000X (Woollam J.A. Co, USA) under beam angle of 65° in the wavelength range from 240 to 1000 nm with a 10-nm pitch. Since the ellipsometry is indirect technique, the values of the optical constants can be estimated by applying the model to recalculate measured values of the amplitudes (psi) and the phase shift (delta) into the optical parameters of samples. Elemental composition of obtained films was studied by energy dispersive X-ray (EDX) spectroscopy using Nova Nanolab 600 scanning electron microscope (SEM) (FEI Co., The Netherlands).

3. Results and discussion
Figure 1 shows AFM images of LiNbO\(_3\) films fabricated under different amounts of laser pulses on Si substrates.

![AFM images of LiNbO\(_3\) films](image)

**Figure 1** (a, b). AFM images of nanostructured LiNbO\(_3\) films on Si fabricated under different number of laser pulses: 50 000 (a) and 100 000 (b).

With increasing number of pulses from 50 000 to 100 000, the roughness of nanostructured LiNbO\(_3\) films increased from 7.764 to 8.20 nm, which may be associated with a longer thermal effect on the film obtained under 100 000 pulses compare to the film obtained under 50 000 pulses. Wherein, in both cases formation of large droplets on the film surface with the diameter of 180 – 220 nm were observed.

Figure 2 shows cross-section of LiNbO\(_3\) thin film on SiO\(_2\) and the dependence of LiNbO\(_3\) films roughness on Ar pressure fabricated on Si and SiO\(_2\) substrates. The SiO\(_2\) layer was grown on the Si surface by chemical vapor deposition method. The thickness of the layer was about 100 nm.
Figure 2 (a, b). Cross-section of LiNbO$_3$ thin film on SiO$_2$ (a), and the dependence of surface roughness on Ar pressure (b).

For LiNbO$_3$ films fabricated over a wide range of growth conditions on Si substrates, the formation of large droplets is typical as Figure 3 (a) shows.

Figure 3 (a, b). Elemental mapping: electron micrograph region of droplet (a), and the distribution of niobium element (b).

The results of EDX study shows the formation of niobium oxide clusters on the LiNbO$_3$ film surface fabricated on Si substrate under an O$_2$ pressure of $10^{-2}$ Torr. Using of SiO$_2$ as substrates and addition of Ar on the film forming step are allowed to reduce the number of the droplets as Figure 2 (a) shows.

Figure 4 shows the dependencies of the electro-physical parameters of nanostructured LiNbO$_3$ films on grain size and films thickness. It was found that increase in the film thickness from 45.3 to 192.9 nm results in increasing of charge carrier concentration from $4.1 \times 10^{12}$ to $8.75 \times 10^{12}$ cm$^{-3}$. Increasing in grain size from 118 to 172 nm causes a gradual increase in the charge carrier mobility from 125.714 to 505.841 cm$^2$/V-s.
Figure 4 (a, b). Dependences of charge carriers concentration on LiNbO$_3$ films thickness (a) and mobility of charge carriers on the grain size (b).

The increase in the mobility of charge carriers can be associated with enhancing of the crystal structure of the films, as well as decreasing of LiNbO$_3$ film roughness due to increasing of grain size. This fact is confirmed by the results of AFM and SEM studies.

Figure 5 shows the spectral dependences of psi and delta for films with different thicknesses. It has been shown that increases in the number of laser pulses from 50 000 to 150 000 results in changing of psi maximum value from 36 to 47, and delta from 160 to 175.
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Figure 5 (a, b). Dependences of amplitude (psi) and phase shift (delta) on the wavelength for samples fabricated under different number of laser pulses: 50 000 (a) and 150 000 (b).

The refractive index value ($n$) calculated on the basis of the measured psi and delta in the visible part of the spectrum was 2.01 – 1.97, while the value of extinction coefficient ($k$) did not exceed 0.05.

4. Conclusion
Obtained results shows that increasing in the number of laser pulses from 50 000 to 150 000 results in increasing the maximum value of psi from 36 to 47, and delta from 160 to 175. It was established that increasing of film thickness from 45.3 to 192.9 nm results in increasing of charge carriers concentration from $4.1\cdot10^{12}$ to $8.75\cdot10^{12}$ cm$^{-3}$. Increasing in grain size from 118 to 172 nm causes gradual increasing in charge carriers mobility from 125.714 to 505.841 cm$^2$/Vs. The increasing in charge carriers mobility can be associated with enhancing of crystal quality of films, as well as a decreasing in film roughness and the number of grains due to an increase of their diameter. This fact is confirmed by the results of AFM and SEM studies.

Obtained results can be used as a physical and technological basis for the development and manufacture of integrated acousto-optic devices, as well as sensitive elements of sensors using various effects of surface acoustic waves.

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