Influence of target-substrate distance during pulsed laser deposition on properties of LiNbO$_3$ thin films

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Abstract. This paper shows the results of studying the influence of target-substrate distance during pulsed laser deposition on electrical properties of LiNbO$_3$ thin films. It has been shown that changing the target-substrate distance we can obtain thin films with a different composition. EDX spectra indicate that the LiNbO$_3$ thin films fabricated under 120 mm target-substrate distance depleted Nb (0.7 atm. %) compare to LiNbO$_3$ films fabricated under 20 mm target-substrate distance (11.0 atm. %). Varying target-substrate distance in the range from 20 mm to 120 mm charge carrier mobility increasing from 24 cm$^2$/V·s to 39 cm$^2$/V·s and concentration of charge carrier decreasing from $3 \cdot 10^{13}$ cm$^{-3}$ to $1 \cdot 10^{12}$ cm$^{-3}$. Obtained results can be used under development and fabrication of integral acousto-optic and surface acoustic wave (SAW) devices.

1. Introduction

Lithium niobate (LiNbO$_3$) is the material which is widely used for fabrication of SAW devices due to its piezo-, pyroelectrics, acousto-optics, and electro-optics properties [1]. Development of integral acousto-optic cell for radio engineering processors of ultra-wideband microwave signals is one of the prospective areas of acousto-optic. Currently, the task of obtaining of LiNbO$_3$ thin films with low optic losses by traditional microelectronic techniques is most important. LiNbO$_3$ thin films could be fabricated by different techniques: molecular beam epitaxy (MBE), RF-sputtering, sol-gel process, pulsed laser deposition (PLD). PLD is a prospective technique for fabrication LiNbO$_3$ thin films due to a numerous of technological parameters and the ability to keep stoichiometric composition of a target [2, 3].

Besides, LiNbO$_3$ films are multicomponent oxides and its properties (crystallographic orientation, resistance, concentration and mobility of charge carriers, optical and piezoelectric properties, and surface roughness) depend on the exact observance of the stoichiometric composition and structure, which in turn depends on the method and technological modes. In that light research influence of technological modes on properties of LiNbO$_3$ thin films in order to fabricate the films with target
properties is important. Since target-substrate distance is one of the key parameters of the PLD [4], the task of this work is researching the influence of target-substrate distance during PLD on LiNbO₃ thin films electrical properties.

2. Experiment
In order to fabricate of LiNbO₃ thin films, we used nanocluster NANOFAB NTK-9 (NT-MDT, Russia), comprising PLD equipment Pioneer 180 (Neocera Co., USA). The LiNbO₃ target was ablated by excimer KrF laser (λ=248 nm) (Coherent Inc., USA). Energy density on target surface was maintained at 2.0 J/cm². The quantity and frequency of laser pulses were 50.000 and 10 Hz, respectively. Target-substrate distance had been varied from 20 mm to 120 mm. LiNbO₃ films were fabricated on Al₂O₃ substrates (1 cm x 1 cm). All substrates were subjected to preliminary purification in inorganic solvents to remove surface contamination. The morphology of obtained film was studied by scanning electron microscopy (SEM, Nova Nanolab 600 with EDX system (FEI Co., Netherlands)) and atomic-force microscopy in semi-contact mode (AFM, Ntegra probe nanolab (NT - MDT, Russia)). Electrical properties of the LiNbO₃ films were measured by Ecopia HMS-3000 equipment (Ecopia Co., Republic of Korea). Optical properties of obtained films (refractive index n, absorption coefficient k) were studied by spectral ellipsometry technique on spectral ellipsometer M-2000X (Wollam J.A. Co, USA) in the range of wavelength from 240 nm to 1000 nm.

3. Results and discussion
Figure 1 shows AFM images and dependencies of LiNbO₃ thin films morphological properties on target-substrate distance.

![AFM images and dependencies of LiNbO₃ thin films morphological properties on target-substrate distance.](image-url)
Increase target-substrate distance from 20 mm to 110 mm surface roughness of obtained films decrease from 447.5 nm to 69.8 nm. Further increase target-substrate distance from 110 mm to 120 mm increase surface roughness to 144.6 nm. Wherein, increase target-substrate distance from 20 mm to 80 mm results in reducing of grain size from 471 nm to 27.4 nm. Further increase of target-substrate distance from 80 mm to 120 mm results in increasing grain size up to 612 nm. Such behaviour of the dependences may be linked to additional heating from plasma plume (in case of target-substrate distance from 20 mm to 60 mm). The subsequent increase of grain size (target-substrate distance > 80 mm) may be related to deficiency of Li in the films and Nb deposition in the form of a solid phase.

Figure 2 shows dependencies of LiNbO$_3$ thin films electrical properties on target-substrate distance.

(a)

(b)
Figure 2 (a, b, c). Dependencies of concentration (a), mobility (b), and resistivity (c) of charge carrier on target-substrate distance.

Varying target-substrate distance in the range from 20 mm to 120 mm charge carrier mobility increasing from 24 cm²/V·s to 395 cm²/V·s and concentration of charge carrier decreasing from $3 \times 10^{13}$ cm⁻³ to $1 \times 10^{12}$ cm⁻³. According to the results of morphological study this fact indicates enhancing of crystalline quality of the films. The resistivity of the films was changed insignificantly (from $8.3 \times 10^{3}$ Ohm·cm to $1.27 \times 10^{4}$ Ohm·cm).

Figure 3 shows the results of EDX analysis of LiNbO₃ films fabricated under different target-substrate distance. However, the EDX of the matrices could not show the presence of lithium due to its low atomic number. EDX of the matrices showing the presence of the niobium (Nb) and oxygen (O) in obtained LiNbO₃ films.

Figure 3 (a, b). The results of EDX analysis of LiNbO₃ films fabricated under different target-substrate distance: 20 mm (a), and 120 mm (b).

The results of EDX analyses shows that LiNbO₃ thin films obtained under target-substrate distance 120 mm contain less Nb (0.7 atm. %) compare to the same films obtained under target-substrate distance 20 mm (11.0 atm. %). Presence of Al peak is related to Al₂O₃ substrate.
Figure 4 shows spectral dependence of refractive index $n$ and absorption coefficient $k$ of the LiNbO$_3$ film fabricated under target-substrate distance 80 mm.

![Spectral dependence of refractive index and absorption coefficient](image)

**Figure 4.** Spectral dependence of refractive index $n$ and absorption coefficient $k$ of the LiNbO$_3$ film fabricated under target-substrate distance 80 mm.

It was established, that the results of optical study didn’t depend on orientation of samples. This fact shows isotropic character of the optical properties. In the visible wavelength range the refractive index decreased from 3.03 (350 nm) to 1.95 (800 nm).

### 4. Conclusion

Obtained results show that morphological, electrical, and optical properties of LiNbO$_3$ strongly depend on the target-substrate distance. EDX analysis indicate that LiNbO$_3$ thin films fabricated under 120 mm target-substrate distance depleted Nb (0.7 atm. %) compare to LiNbO$_3$ films fabricated under 20 mm target-substrate distance (11.0 atm. %). The ability of formation LiNbO$_3$ thin films with target properties and different composition by PLD has been shown. The results can be used under development and fabrication of integral acousto-optic and SAW devices.

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