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Infrared reflection spectroscopy and optical constants of $LiNbO_3$ films on crystal substrates

N.N. Novikova[1], V.A. Yakovlev[1,*], and P.G. Medaglia[2]

[1]Institute for Spectroscopy RAS, 108840 Moscow, Troitsk, Russia
[2]Dipartimento di Ingegneria Industriale, Università di Roma Tor Vergata, Rome, Italy

Abstract. We have measured infrared reflectivity spectra of thin lithium niobate films of nanometer thickness, grown by a pulsed laser deposition technique using KrF-excimer laser (λ=248 nm) on the single crystalline substrates (sapphire, MgO, NdGaO3 and SrTiO3). Using the dispersion analysis technique, we have calculated thicknesses and optical constants of the films. The phonon parameters of the substrates and films are obtained.

Electro-optical and optical photorefractive crystal of lithium niobate ($LiNbO_3$) is one of the most widely currently used ferroelectric materials in piezotechnique, quantum opto-and acoustoelectronics. Currently, active development of optical devices is based on the films of lithium niobate. The study of optical properties and obtaining the main parameters of films of lithium niobate deposited on different types of substrates, may be very promising for practical applications, pointing out ways to improve the efficiency of management of physical characteristics of this unique material. Epitaxial films of lithium niobate were grown by pulsed laser deposition on single crystal substrates of sapphire, magnesium oxide, $SrTiO_3$ and $NdGaO_3$, using a KrF (λ=248 nm) excimer laser [1, 2].

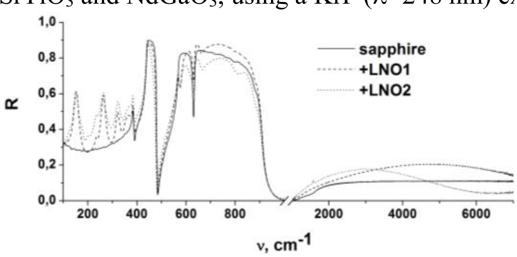

Fig. 1. Reflectivity of sapphire substrate and $LiNbO_3$ films 1 and 2.

The laser beam was focused (3 mm^2) on the surface of the target ($LiNbO_3$ single crystal), with a pulse energy of 170 to 220 mJ. Typical growth parameters – pressure of O_2 is from 0.5 to 1 mbar and the substrate temperature is from 600°C to 700°C. The distance target-substrate was about 4.5 cm. After synthesis, the samples were rapidly cooled in argon atmosphere to room temperature.

Reflectance of the infrared (IR) radiation is widely used to study the vibrational spectra of bulk materials and coatings on them. This non-destructive technique allows to study the phonon spectra and to obtain optical constants and thicknesses of films. Reflection spectra in the IR region (30-7500 cm^{-1}) with a resolution of 2 cm^{-1} were registered on a Fourier-transform spectrometer Bruker IFS66v with the reflectance unit at the angle of incidence of 16 degrees. Grid polarizers on KRS-5 and polyethylene were used in the mid and the far IR regions, respectively. Some spectra are shown in Fig. 1. The optical constant of the substrates and films were obtained by dispersion analysis of reflection spectra [3,4] using the SCOUT software (Windows).

The measured IR reflection spectrum was compared with the spectrum calculated using the model of damped harmonic oscillators for the dielectric constant of the film and

* Corresponding author: yakovlev@isan.troitsk.ru

substrate. $\varepsilon = \varepsilon_\infty + \Sigma\ f_i^2/(\nu_{TOi}^2 - \nu^2 - i\gamma_i\nu)$. The film thickness and the parameters of the oscillators (the frequency - ν_{TOi}, the oscillator strength f_i and the damping γ_i) were varied to minimize the differences of the spectra. At first, this procedure was applied to spectra of uncoated substrates. The parameters of the substrates were obtained. They are close to the literature data (e.g. [3]). Then the film spectra were fitted taking into account the reflection from the two interfaces of the film. The film parameters were obtained. Table 1 presents the parameters of the optical phonons of the films and of the $NdGaO_3$ substrate, obtained from the reflection spectra. For comparison, the parameters of a single crystal $LiNbO_3$ from the Ref. [4] are given too.

Table 1. Parameters of the optical phonons of the films and of the $NdGaO_3$ substrate.

Film No	LNO1	LNO2	LNO3	LNO4	LNO5	LNO6	Substrate $NdGaO_3$	Crystal $LiNbO_3$ E	Crystal $LiNbO_3$ A
d, nm	256	432	682	830	485	1080			
ε_{oo}	4	4.27	4.5	4.48	4.83	5.45	4.08	5	4.6
ν_{TO1}, cm^{-1}	152.7	154.9	152.1	151	155.9	169.1	170.8	152	248
f_1, cm^{-1}	664	559	643	409	545	498	433	713	992
γ_1, cm^{-1}	19	20	13	23	18	17	7.6	2	5
ν_{TO2}, cm^{-1}	234.1	235.4	234.4	242	235.6	239	243.3	236	274
f_2, cm^{-1}	217	247	176	66	346	62	169.4	211	274
γ_2, cm^{-1}	12	11	10	7.7	11	8	7.6	3	4
ν_{TO3}, cm^{-1}	262.2	261.5	261.6	266.4	258.4	266.4	272.9	265	307
f_3, cm^{-1}	563	532	611	284	717	511	553.9	621	123
γ_3, cm^{-1}	14	15	12	15	13	9	10	3	8
ν_{TO4}, cm^{-1}	320.5	320.2	320.9	325.9	313.9	274.9	292.1	322	628
f_4, cm^{-1}	387	393	393	148	552	691	503.2	478	1000
γ_4, cm^{-1}	14	15	8	8	40	30	11.6	3.5	22
ν_{TO5}, cm^{-1}	361.8	360.2	362	404.4	321.6	318.5	338.7	363	692
f_5, cm^{-1}	454	474	727	655	236	250	553.6	551	250
γ_5, cm^{-1}	41	41	42	32	8	12	18	12	34
ν_{TO6}, cm^{-1}	426.9	418.3	432	443.5	362.9	343.9	442.3	431	
f_6, cm^{-1}	511	505	237	558	438	648	91.6	183	
γ_6, cm^{-1}	40	42	23	41	23	44	6.2	5	
ν_{TO7}, cm^{-1}	565.1	565.8	556	558.7	433.3	584	521.8	586	
f_7, cm^{-1}	891	923	289	912	197	724	124.5	1065	
γ_7, cm^{-1}	24	56	30	57	21	59	22.6	21	
ν_{TO8}, cm^{-1}	585.4	576.2	561.8	600.7	591.7	621.9	594.3	670	
f_8, cm^{-1}	501	291	859	307	1106	320	232.3	300	
γ_8, cm^{-1}	7	32	55	30	60	23	22.9	31	
ν_{TO9}, cm^{-1}	668.7	670.1	670.1	674.2	655.5	668.6			
f_9, cm^{-1}	176	325	325	340	347	316			
γ_9, cm^{-1}	22	69	69	71	69	88			
Substrate	Al_2O_3	Al_2O_3	MgO	MgO	$SrTiO_3$	$NdGaO_3$			

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