Epitaxial Lithium Niobate Thin Films Grown by Chemical Beam Epitaxy on Sapphire

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Lithium Niobate ($LiNbO_3$) is a versatile material with a number of remarkable qualities. It finds application in optical modulators because of its electro-optic properties. Nonlinearity opens its use in bio-physical applications where particles or wires of $LiNbO_3$ can be used as highly localized optical probes. Optical frequency conversion is another possible use, as well. One of the current commercial applications of the material is in optical modulators in telecomunication devices. Nowadays bulk crystals of the material are used. However, in order to make devices more compact and affordable it is necessary to be able to produce $LiNbO_3$ films on suitable substrates with sufficient crystalline and optical quality.

Here we present films deposited by means of chemical beam epitaxy (CBE). The films were produced in a high vacuum chemical vapour deposition reactor using lithium tert-butoxide ($Li(OBu^t)$) and niobium tetra-ethoxy di-methyl-amino-ethoxide ($Nb(OEt)_4(dmae)$) as sources of Li and Nb atoms. No additional oxygen was used. Deposition took place at 650°C. The reactor is designed for deposition on up to 6” wafers, however smaller (10x20 mm2) sapphire (0001) substrates were used to facilitate the research. The choice of the substrate was determined both by its suitability for epitaxial deposition, mechanical properites, availability and preferable optical properties (transparency and sufficient refractive index difference for possible waveguiding applications). Optimization of the Li/Nb content ratio was performed using gradients of precursors in a combinatorial way and based on the feedback from X-ray diffraction (XRD) and Raman spectroscopy.

The optimized samples reveal high crystallinity and are highly textured in the {0001} direction. The rocking-curve measurements on the (0006) peak confirmed a high degree of orientation; the broadening of the peak being only 0.03°. The pole-figure XRD measurement confirmed that the film is highly {0001} oriented in the vertical direction and also in plane. Six discreet (012) reflections have been observed instead of three expected for the perfect $LiNbO_3$ structure. This was explained by growth of two equivalent variations of the $LiNbO_3$ trigonal structure rotated by 60°. Additionally, $LiNbO_3$ crystalline quality and phase purity was confirmed by Raman spectroscopy.

TEM observation revealed a very thin transition layer of few nanometres at the film-substrate interface. The formation of the layer is probably due to the large lattice and/or thermal expansion coefficient mismatch between $LiNbO_3$ and sapphire.

The roughness of the film is one of the key factors in the fabrication of high quality, low optical loss devices. The roughness of the deposited films was determined by AFM measurements and the RMS value is ~5 nm for 200 nm thick films.

A second harmonic generation signal was detected, when the film was irradiated with 800 nm light from a femtosecond Ti:sapphire laser, proving the non-linear properties of the films.

Thus, the ability to deposit epitaxial $LiNbO_3$ films with high crystalline quality and low surface roughness using chemical beam epitaxy has been demonstrated and an understanding of the deposition process has been gained. Further optical characterization of the films is in process.