Liquid phase epitaxy of KTa_1-xNb_xO_3(KTN)

Liquid Phase Epitaxy of $KTa_{1-x}Nb_xO_3$ (KTN)

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Abstract: KTN layers of several µm thickness were grown on [100] and [110] orientated $KTaO_3$ substrates from diluted high temperature KF solutions. A versatile apparatus for evaluating new flux systems for liquid phase epitaxy has been developed.

1. Introduction

Thin monocrystalline layers of KTN are of interest for integrated optics due to outstanding electro-optical and nonlinear-optical properties of KTN bulk crystals. The solid solution system of KTN [1] shows a perovskite structure and 3 ferroelectric phase changes between m3m, 4mm, mm2 and 3m [2]. The Curie temperature T_C for the phase change from cubic to tetragonal can be adjusted from 700 K to 10 K through the variation of the chemical composition x. For temperatures near T_C high dielectric constants, large electro-optic and nonlinear-optic effects have been measured for bulk crystals [3].

Liquid phase epitaxy (LPE) allows to work close to the thermodynamic equilibrium at temperatures far below the melting point of pure compounds and in a quasi-stationary flow regime of a solvent. Under these conditions epitaxial layers of good quality can be expected.

2. Experimental

In the course of mapping appropriate growth conditions for LPE, a versatile equipment was developed for in situ molten salt studies, based on visual observation and DTA [4]. Exploratory growth experiments from a $K_2O/Ta_2O_5/Nb_2O_5$/KF/KCl flux gave transparent and blue coloured monocrystalline KTN layers of several µm thickness on small [100] and [110] orientated $KTaO_3$ substrates. For optical applications discolouration is necessary. This was achieved by annealing the as grown layers for 24 h at 950 °C in an O_2-atmosphere (Fig. 1) or by adding small amounts of ZrO_2 or HfO_2 to the flux. Growth parameters for final layers, obtained by an enlarged LPE equipment, are listed in Tab. 1. Para- and ferroelectric KTN layers (300 K) could reproducibly be grown by LPE.

Tab. 1. Growth parameters for LPE

Flux	$KTa_{1-x}Nb_xO_3$ $(0.68 < x < 0.85)$	4 - 8	mole%
	KF or KF/KCl	96 - 92	mole%
Additives for discolouration	ZrO_2, ZrF_4, HfO_2	< 0.5	mole%
Substrates	$KTaO_3$	orientations:	[100] and [110]
	(cubic)	size:	< 8 x 12 mm^2
Atmospheres	N_2, O_2, Ar, air		
Growth temperatures	870 - 930	°C	
Supercoolings	30 - 100	°C	
Substrate rotation	~ 30	rpm	
Growth times	5 - 30	min	
Layer thicknesses	2 - 45	μm	

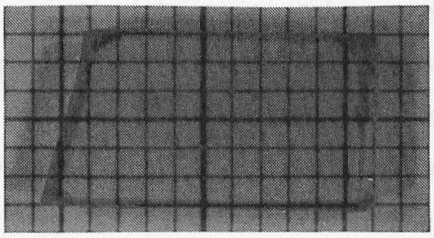

Fig. 1. KTN layer after annealing in O_2

3. <u>Characterization</u>

Scanning electron microscopy (SEM) showed rather flat morphologies for growth on [100] and pronounced facets on [110] orientated substrates. Energy dispersive X - ray spectroscopy (EDX) and electron microprobe analysis traced the Nb content of the layers $(0.27 < x < 0.46)$. Rutherford backscattering spectrometry (RBS) and secondary ion mass spectrometry (SIMS) revealed a constant chemical composition of the layers. Electron channeling (Fig. 2) and 2θ X-ray diffraction indicated monocrystalline KTN layers. Lattice misfits smaller than 0.75 % are found for cubic layers.

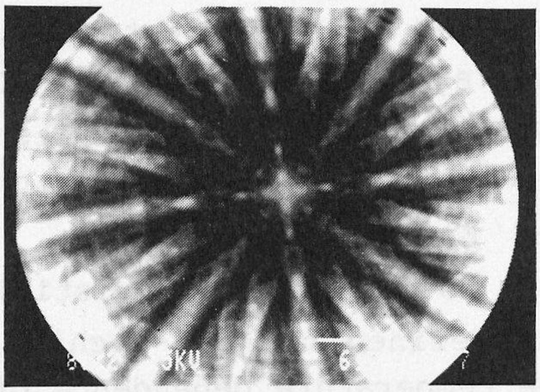

Fig. 2. Electron channeling of a KTN layer on [100] $KTaO_3$.

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