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Names and affiliations of applicants (* indicates experimentalists):

Dr. J. Kräußlich *1)
Dr. O. Wehrhan *1)
PhD U. Zastrau *1)
Dr. C. Dubs 2)
DP A. Lorenz 2)
Prof. A. Tünnermann 2)

Friedrich-Schiller-University of Jena
1) Institute for Optics and Quantum Electronics
2) Institute of Applied Physics
Max-Wien-Platz 1
07743 Jena
Gemany

Report:

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In-plane lattice parameter determination of Zn:$LiNbO_3$ thin films epitaxially grown on x-cut $LiNbO_3$ substrates using X-ray diffraction methods

J. Kräußlich, O. Wehrhan, U. Zastrau, C. Dubs, A. Tünnermann

The aim of the experiment has been to characterize Zn:$LiNbO_3$ thin films epitaxially grown on $LiNbO_3$ substrates by means of high resolution x-ray diffraction methods. Data concerning the characteristics of the epitaxial growth, the crystallographic orientation of the thin film in respect to the substrate as well as the numerical evaluation of the in-plane lattice parameter $\mathbf{c} = \mathbf{d}_{(000\ 0)}$ and $\mathbf{d}_{(01\text{-}1\ 0)}$ of the epitaxially grown $Li_{1-x}Zn_xNbO_3$ thin films are of special interest for the thin film producer to optimize the technological growth process.

$LiNbO_3$, due to its outstanding optical and electro-optical properties, is a suitable material to optical waveguide devices such as modulators and switches and to second-harmonic generation devices for integrated optical applications [1, 2, 3]. Optical waveguides demand a well-defined modification of the local refractive index Δn. For this purpose, Zn-substituted stoichiometric $Li_{1-x}Zn_xNbO_3$ thin films with a thickness until 4 µm were grown on conventional x-cut $LiNbO_3$ single crystal substrates by means of the liquid phase epitaxy (LPE) method [4, 5].

X-ray diffraction pattern taken up with symmetric reflexes in the Θ/2Θ-mode permit to determine the lattice parameter differences of the grown thin film in respect to the substrate only perpendicularly to the sample surface, however with high precision ($\Delta d_\perp/d_\perp < 10^{-5}$) (Fig. 1).

It particularly results for the sample LNBV-1kt-2 (thin film without Zn):
$(\Delta d/d)_{\perp} = 8.3 \cdot 10\text{-}4 \pm 2\cdot10^{-5}$
$a_{\perp\text{hin film}} = 0.51478\text{nm} \pm 2\cdot10^{-5}\text{nm}$
with respect to $a_{\perp\text{substrate}} = 0.51522\text{ nm} \pm 2\cdot10^{-5}\text{nm}$

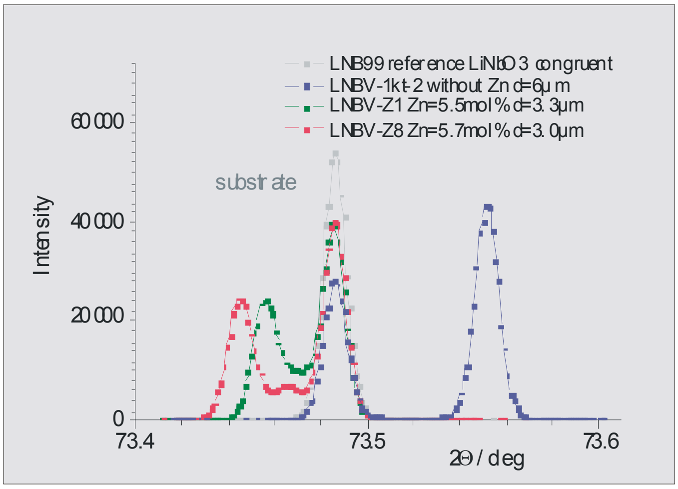
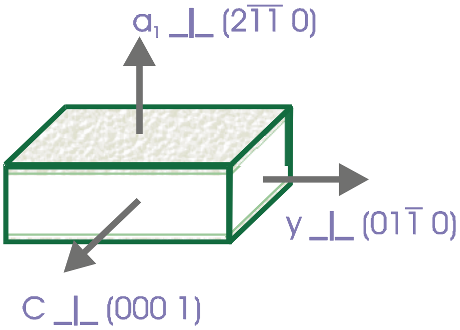


Fig. 1: Θ/2Θ-scan taken up with the symmetric 4-2-2 0 -reflection of the LiNbO3 thin film epitaxially grown on x-cut $LiNbO_3$ substrate



From two-dimensional ´reciprocal space maps´ taken up by x-ray diffraction with asymmetric reflexes, data can be won both to the lateral and normal lattice parameters of the grown thin film in respect to the substrate, although with somewhat small precision ($\Delta d_{\parallel}/d_{\parallel} < 2\cdot10^{-4}$).

The rsm´s (Fig. 2 and 3) reveals for the two different lateral directions c ⊥ (000 1) and y ⊥ (01-1 0) pseudomorphous growth of the thin film in respect to the substrate, despite the existing different crystallographic symmetry.

Using generalized Hooke' law in matrix way of writing, the relaxed lateral and normal lattice parameters as well as the deformation and tension components of the grown thin film can be computed.

$$\boldsymbol{\sigma_m = C_{mn}\varepsilon_n} \qquad m,n = 1,\dots,6$$

with $\sigma_2 \neq 0;\ \sigma_3 \neq 0;\ \sigma_1 = \sigma_4 = \sigma_5 = \sigma_6 = 0;$
and $\varepsilon_1 \neq 0;\ \varepsilon_2 \neq 0;\ \varepsilon_3 \neq 0;\ \varepsilon_4 = \varepsilon_5 = \varepsilon_6 = 0;$

σ_m - tension components
ε_n - deformation components
C_{mn} - elastic stiffnesses / 10^9 Pa

$$\boldsymbol{0 = C_{11}\varepsilon_1 + C_{12}\varepsilon_2 + C_{13}\varepsilon_3}$$
$$\boldsymbol{\sigma_2 = C_{12}\varepsilon_1 + C_{11}\varepsilon_2 + C_{13}\varepsilon_3}$$
$$\boldsymbol{\sigma_3 = C_{13}\varepsilon_1 + C_{13}\varepsilon_2 + C_{13}\varepsilon_3}$$

and assuming that $\varepsilon_2 = \varepsilon_3$

↳ $\varepsilon_1 = - (C_{12} + C_{13}) / C_{13}\varepsilon_2$

$\varepsilon_1 = - 0.64\ \varepsilon_2$ (← transversal contraction)

C_{11} [10^9 Pa]	C_{12} [10^9 Pa]	C_{13} [10^9 Pa]	C_{14} [10^9 Pa]	C_{33} [10^9 Pa]	C_{44} [10^9 Pa]
203	55	75	8.8	244	60

Table 1: Elastic stiffness modulus [6] of $LiNbO_3$

From this follows the numerical calculation of the **relaxed lattice parameters a_{relax} und c_{relax}** for the stoichiometric $LiNbO_3$ thin film coherently grown on congruent $LiNbO_3$ substrates

with the measured values
especially for the sample LNBV-1kt-2 (thin film without Zn):

$a_{\|} = 0.51522$ nm
$a_{\perp} = 0.51478$ nm
$c_{\|} = 1.3867$ nm

and with $(a_{\|} - a_{relax})/a_{relax} = \varepsilon_{\|} = \varepsilon_2$
$(a_{\perp} - a_{relax})/a_{relax} = \varepsilon_{\perp} = \varepsilon_1$
$\varepsilon_1 = - 0.64\ \varepsilon_2$

↳ $a_{relax} = (a_{\|} - \varepsilon_2/\varepsilon_1 \cdot a_{\perp})/(1 - \varepsilon_2/\varepsilon_1) = 0.5149 \text{ nm} \pm 0.0005 \text{ nm}$

$\varepsilon_1 = - 3.49(1) \cdot 10^{-4}$

$\varepsilon_2 = 5.45(1) \cdot 10^{-4}$

and with $(c_{\|} - c_{relax})/c_{relax} = \varepsilon_{\|} = \varepsilon_3\ (= \varepsilon_2)$

↳ $c_{relax} = 1/(1 + \varepsilon_3) \cdot c_{\|} = 1.386 \text{ nm} \pm 0.002 \text{nm}$

$\varepsilon_3 = 5.45(1) \cdot 10^{-4}$

As final result arises now the ´in-plane´ tension components of the grown thin film, in direction of the lateral crystallographic axes y and c, respectively

↳ $\sigma_2 = 1.32(1) \cdot 10^8$ N/m^2

$\sigma_3 = 1.48(1) \cdot 10^8$ N/m^2

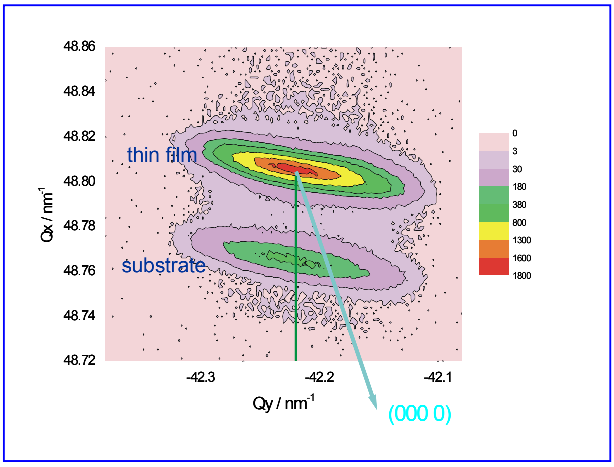


Fig 2: rsm: asymmetric 4-51 0-reflection

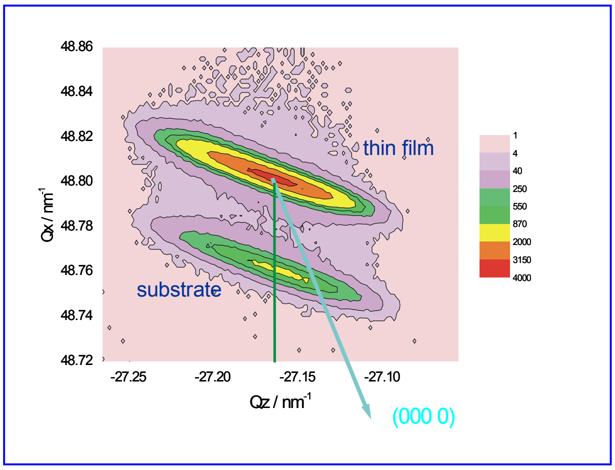


Fig 3: rsm: asymmetric4-2-2 6-reflectio

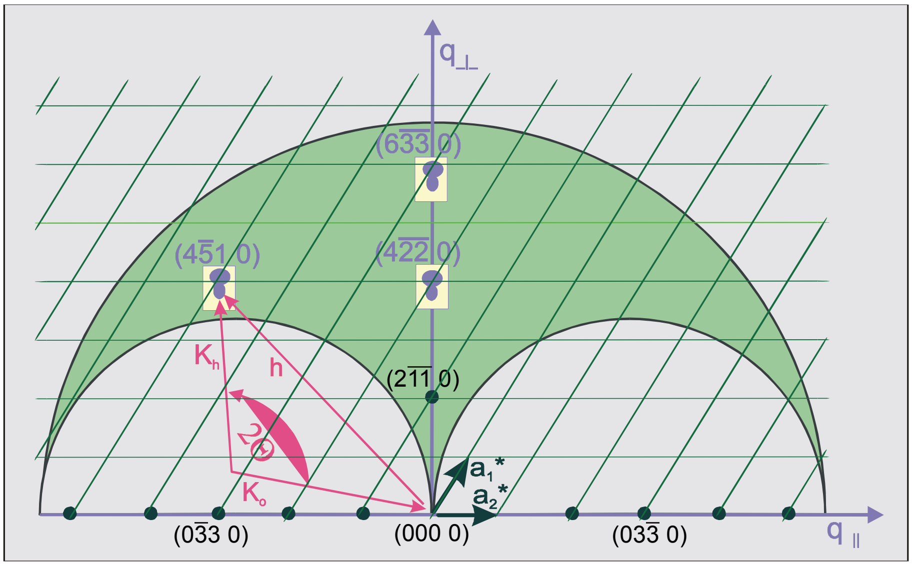


Fig. 4: Reciprocal lattice of $LiNbO_3$ in (000 1) cross-section projection for the two-dimensional taking up ´reciprocal Space maps´ with asymmetric reflections to determine the lateral thin film lattice parameters in relation to the substrate.

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