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Complex Elastic Properties and Piezoelectric Activity in Stoichiometric and Congruent LiNbO₃ Single Crystals

IWONA FRANKE

Institute of Physics
University of Silesia
ul. Uniwersytecka 4, 40-007 Katowice, Poland

JEAN-PAUL SALVESTRINI
MARC FONTANA

MOPS Laboratory, FRE CNRS 2304
University of Metz and Supelec
2 rue E. Belin, 57-078 Metz, France

KRYSTIAN ROLEDER

Institute of Physics
University of Silesia
ul. Uniwersytecka 4, 40-007 Katowice, Poland

*Piezoelectric effect has been measured for stoichiometric ($Li/Li + Nb = 50$ mole%) and congruent ($Li/Li + Nb = 48.6$ mole%) lithium niobate crystals. Observations of the piezoelectric resonance allowed to investigate the temperature dependence of the complex elastic coefficient s^*_{11} . Distinct local maxima on the $s''_{11}(T)$ dependencies have been found.*

Keywords Piezoelectricity; complex elastic constant; lithium niobate

Introduction

Progress made in crystal-growth techniques enables a better control of chemical composition of LiNbO₃ crystals [1] which are widely used in electro-optics [2] and which properties strongly depend on their stoichiometry. In the electrooptical effect the piezoelectricity plays an essential role [3]. Temperature behaviour of piezoelectric deformation can however be influenced by crystal imperfections. By piezoelectric resonance observations the elastic properties of samples could be directly determined.

Address correspondence to Iwona Franke, Institute of Physics, University of Silesia ul., Uniwersytecka 4, Katowice 40-007, Poland. E-mail: ifranke5@us.edu.pl

Experimental Conditions

The absolute admittance $|Y_c|$ and phase angle θ were measured in the function of frequency by means of a Hewlett-Packard 4192A Impedance Analyser. The $|Y_c|$ was fitted with the relation

$$|Y_c(\omega)| = \sqrt{G_c^2(\omega) + B_c^2(\omega)}, \quad (1)$$

in which the mathematical form of the total conductance $G_c = |Y_c|/\cos\theta$ and susceptance $B_c = |Y_c|/\sin\theta$ has been derived basing on the theory of forced damped harmonic oscillator with complex amplitude. Under the action of the external electric field of frequency ω the $G_c(\omega)$ and $B_c(\omega)$ near the piezoelectric resonance obey the following relations:

$$G_c(\omega) = A \cdot \frac{\omega^2 \Gamma}{(\omega_0^2 - \omega^2)^2 + \omega^2 \Gamma^2} + G \quad B_c(\omega) = A \cdot \frac{\omega \cdot (\omega_0^2 - \omega^2)}{(\omega_0^2 - \omega^2)^2 + \omega^2 \Gamma^2} + \omega C'_d \quad (2)$$

In which A is a constant value connected with sample geometry, amplitude of external electric field and piezoelectric and elastic coefficients. The G and C'_d are related with dielectric properties of the sample far behind the piezoelectric resonance. Example of a fit representing model applicability is shown in Fig. 1. Having the resonance frequency $f_0 = 2\pi/\omega_0$, damping $\gamma_0 = 2\pi/\Gamma$, length l and density ρ of a sample the complex elastic coefficient $s = s' - is''$ could be calculated from the relation $s^E = s'^E - is''^E = \frac{1}{4\rho l^2 f_0^2} - i \frac{1}{4\rho l^2 f_0^2} \left(\frac{\gamma}{f_0}\right)$.

The stoichiometric and congruent LiNbO₃ were grown by the TSSG and Czochralski method respectively [1]. According to geometrical conditions needed for coefficient s_{11}^E assignment, samples of bar shape were cut from single domain single crystals. The largest surfaces perpendicular to c-axis were electroded by gold evaporation. Temperature dependence of the complex elastic constant has been measured in the temperature range 20–180°C.

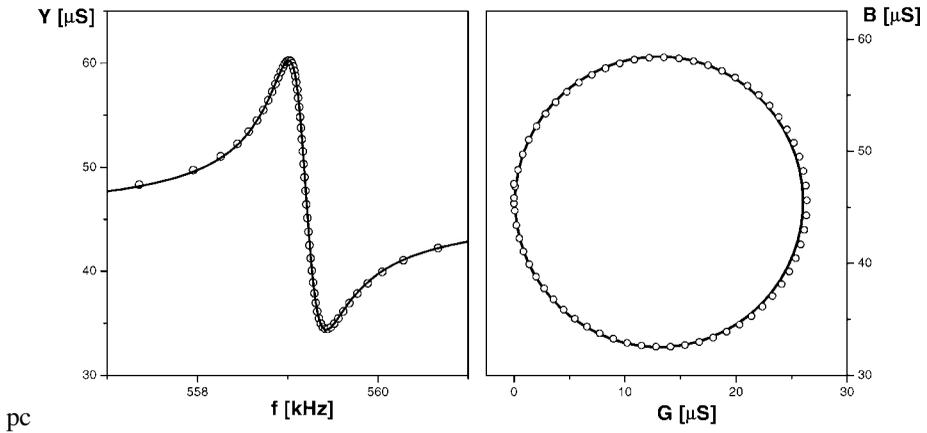


FIGURE 1 Simultaneous fit with theoretical functions (1) and (2) to the experimental run $|Y_c|/(\omega)$ at room temperature. Separate fits for the $G_c(\omega)$ and $B_c(\omega)$ are shown in complex plane. Elastic constant s_{11}^E and piezoelectric module $|d_{31}|$ calculated from the data were equal respectively to $5, 6 \cdot 10^{-12} \text{ m}^2/\text{N}$ and $0.84 \cdot 10^{-12} \text{ m/V}$.

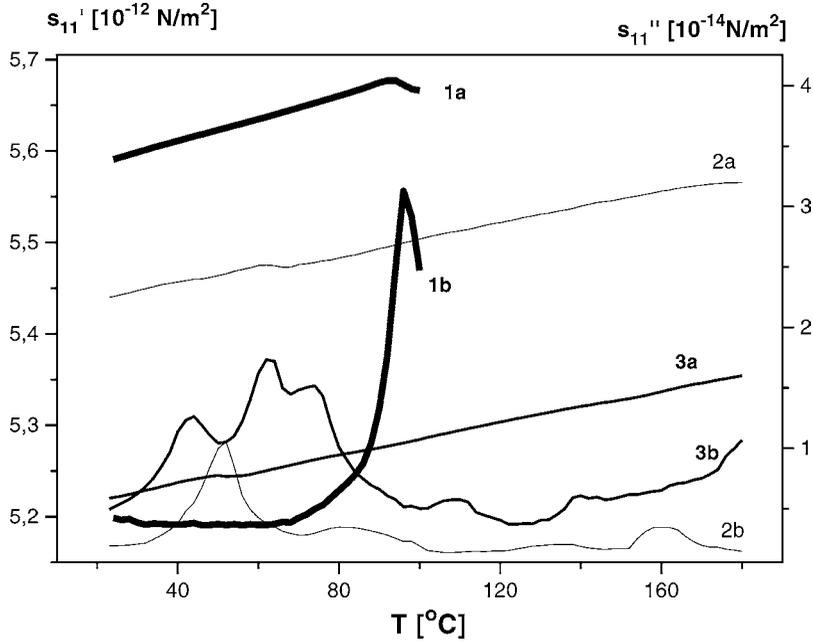


FIGURE 2 Temperature behaviour of real $s_{11}^{\prime E}$ (curves “a”) and imaginary $s_{11}^{\prime\prime E}$ part (curves “b”) of the elastic coefficient for two stoichiometric (curves 1, 2) and one congruent (curves 3) LiNbO_3 single crystals.

Results and Conclusions

Changes of the real and imaginary part of s_{11} with temperature are shown in Fig. 2. While real parts (curves 1a, 2a, 3a) smoothly increased the $s_{11}^{\prime\prime}(T)$ runs (curves 2a, 2b, 3b) revealed distinct local maxima at particular temperatures. The number and intensity of those maxima depended on crystal stoichiometry and number of thermal cycles (heating-cooling). It was found that the cycles made between 25°C and 70°C did not change the elastic properties of the samples: in one of the stoichiometric crystals there was only single, but distinct, local maximum of the $s_{11}^{\prime\prime}(T)$ function (curve 1b). If final temperature on heating was higher than 70°C the observed maxima in both samples shifted towards lower temperatures with increasing number of thermal cycles. Since imaginary part of elastic tensor is related with damping of lattice vibrations induced by external forces the appearance of the $s_{11}^{\prime\prime}(T)$ peaks might be related with local absorption of elastic energy due to defects. It is suggested that the effects observed in lithium niobate were caused by defect migration because of the exo-diffusion of Li^+ ions as considered by Ridah et al. [4].

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