DOI 10.5162/SMSI2025/A5.2

Electro-acoustic Properties of Li(Nb,Ta)O_3 Solid Solutions in Harsh Environments

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Summary:
The prospects of lithium-niobate-tantalate solid solutions ($LiNb_{1-x}Ta_xO_3$, LNT) for high-temperature sensors and actuators are presented from a physical and materials science perspective. The focus lies on defect mechanisms that are relevant for the acoustic losses that determine potential applications of such crystals at high temperatures. Further, tailoring of the materials properties by variation of the niobium-tantalum ratio is demonstrated.

Keywords: Lithium-niobate-tantalate solid solutions, ionic and electronic conductivity, high-temperature, oxygen partial pressure, defect models

Introduction and Motivation

Resonant sensors offer advantages for multiparameter in-situ monitoring and control of industrial processes. In particular, there is an increasing need in sensitive, robust and cost-effective sensors for gas composition, temperature and pressure for the application in harsh environments. Thereby, piezoelectric crystals offer the advantage that they can be directly excited to oscillate by applying a voltage of appropriate frequency. Changes in environmental conditions are directly transferred to frequency changes, which is feasible at very high temperatures with materials such as lithium niobate-lithium tantalate (LNT).

Moreover, LNT represents a piezoelectric material which combines high piezoelectric coefficients, low acoustic loss, and thermal stability which are crucial for the development of high-temperature sensors and actuators.

Objectives

LNT solid solutions are used as model system with the aim of exploring the correlation of defect structure, electronic and ionic transport, and electromechanical properties in polar oxides.

The specific aim of this presentation is to present and interpret the high-temperature electromechanical properties and, in particular, the acoustic losses in bulk LNT at high temperatures. Further, materials data required for modelling such as the temperature dependent piezoelectric coefficient should be determined.

Piezoelectric Resonators

LNT crystals grown by the Czochralski technique [1] are cut into circular disks, polished and, if required, coated with Pt-based electrodes. Such a device and a schematic profile of resonant mechanical displacement is shown in Fig. 1.

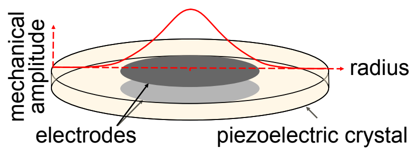

Fig. 1: Piezoelectric resonator with metal electrodes and displacement profile.

Results and Discussion

Crucial high-temperature properties of LNT solid solutions such as bulk conductivity as well as acoustic losses were determined as a function of temperature and oxygen partial pressure (p_{O2}) and correlated with the atomistic transport processes.

Above 400°C, the acoustic losses are governed by the relaxation of piezoelectrically excited charge carriers and thus the electrical conductivity [2]. Fig. 2 shows the related loss peak at about 850 °C.

Below this temperature, the losses decrease and reach values that correspond to that of phonon scattering. The electronic conductivity tends to be suppressed by high Ta contents, which becomes apparent above 600°C and allows a reduction in losses. High mechanical resonance frequencies also lead to a reduction in losses, so that small structures or even thin films are desirable.

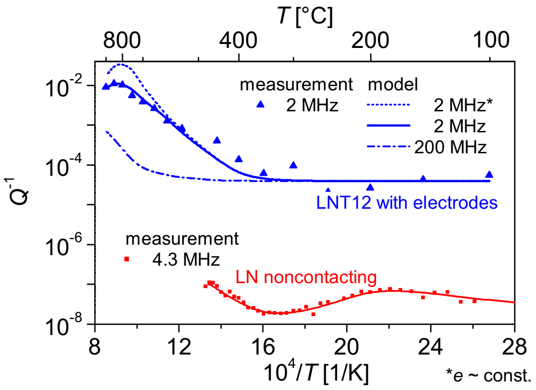

Fig. 2: Total losses of LN and $LiNb_{88}Ta_{12}O_3$ (LNT12) resonators with different composition determined with and without contacting metal electrodes (points) and modelling including variation of the operating frequency (lines).

To enable modeling of the losses, for example, the temperature dependent piezoelectric coefficient has been determined, see Fig. 3.

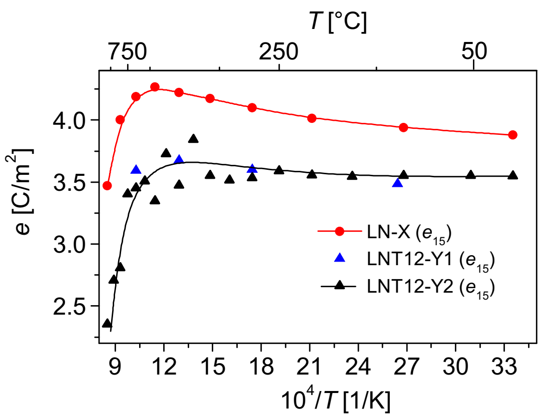

Fig. 3: Piezoelectric coefficient e_{15} for LN and $LiNb_{88}Ta_{12}O_3$ (LNT12) as a function of temperature [3].

Further, the p_{O2} dependence of the conductivity has been measured. It can be explained by a defect mechanism that is not linked to the unwanted evaporation of Li_2O. Fundamental findings, such as the unexpectedly strong change in the activation energy of the electrical conductivity at the transition between the ferroelectric and paraelectric phase, are now also discussed [4,5].

Conclusions

Extraction of materials data using a one-dimensional physical model for piezoelectric resonators together with atomistic models enables to identify the piezoelectric/carrier relaxation as the dominating loss mechanism at high temperatures.

Based on the results, the LNT system is seen as a platform for the development of novel high-tech components for resonant sensors, micro-actuators, integrated acoustics and photonics as well as quantum technologies even at high temperatures. The results can be transferred to other material systems such as multiferroics and perovskite-related materials.

Acknowledgements

The Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) is gratefully acknowledged for financial support within the research unit FOR5044 *Periodic low-dimensional defect structures in polar oxides*.

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