Contributions to Acoustic Loss in Langasite, Langatate, and Catangasite Resonators at High Temperatures

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Summary:
Piezoelectric crystals in the langasite family can serve as a basis for resonant acoustic sensors that operate at temperatures exceeding the range of conventional piezoelectric resonators, but their performance is limited by acoustic loss at elevated temperatures. This paper presents an overview of what is currently known and not known about physical contributions to temperature-dependent acoustic loss in langasite, langatate, and catangasite [1].

Keywords: acoustic loss, attenuation, high temperature, langasite, langatate, catangasite, piezoelectric resonators

Background

Applications of resonant piezoelectric sensors have traditionally been limited to temperatures below several hundred degrees Celsius, because phase transitions and/or material degradation occur at higher temperatures in common commercially available piezoelectric materials [2]. However, over the past four decades, substantial research has focused on growing, characterizing, and optimizing innovative piezoelectric crystals that can be used in resonant sensors at temperatures exceeding 1000 K [3]. These piezoelectrics include crystals with the structure of langasite ($La_3Ga_5SiO_{14}$, LGS).

A critical factor in the performance of resonators is the quality factor Q and corresponding acoustic loss Q^{-1}. This paper is focused on contributions to Q^{-1} in LGS and two piezoelectrics with similar crystal structure, langatate ($La_3Ga_{5.5}Ta_{0.5}O_{14}$, LGT) and catangasite ($Ca_3TaGa_3Si_2O_{14}$, CTGS).

LGS

Figure 1 shows measurements of Q^{-1} acquired at the National Institute of Standards and Technology (NIST, U.S.A.) and Clausthal University of Technology (TUC, Germany) on two Y-cut crystals grown by different manufacturers [2,4]. The combined data in this figure extend over an exceptionally broad temperature range and thereby provide a unique illustration of typical contributions to the temperature-dependent loss in LGS and other crystals in the langasite family. The NIST measurements were performed with noncontacting electrodes in vacuum, and the TUC measurements were performed with Pt surface electrodes in air.

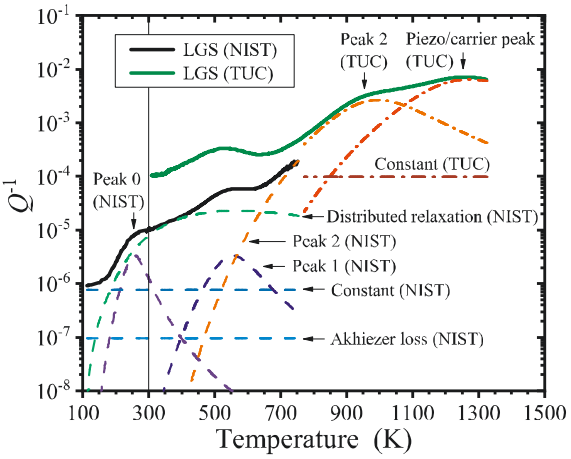

Fig. 1. Acoustic loss Q^{-1} of two LGS crystals measured at NIST and TUC. Resonant frequencies were 6.05 MHz (NIST) and 5.0 MHz (TUC) at ambient temperature. Dashed and dashed-dotted lines are contributions to Q^{-1} of the NIST and TUC crystals, respectively, determined from least-squares fits.

The three peaks below 1100 K in Fig. 1 are consistent with anelastic point-defect relaxations with a characteristic Debye dependence on temperature T and angular frequency ω [5]:

$$Q^{-1} = (\Delta/T)\omega\tau/(1+\omega^2\tau^2) , \qquad (1)$$

where the relaxation strength Δ is proportional to defect concentration. The relaxation time τ has an Arrhenius dependence on T,

$$\tau = \gamma\exp(U/kT) , \qquad (2)$$

where *U* is an activation energy on the order of an electron volt and *k* is Boltzman's constant.

Another peak appears in the TUC data near 1260 K, and this is associated with piezoelectric/carrier relaxation, involving the motion of charge carriers in acoustically generated piezoelectric fields [2,4]. In LGS, this peak is expected to have Debye form, but without *T* in the pre-factor [4]. An additional contribution in all crystals is intrinsic loss from phonon-phonon interactions (Akhiezer loss) [6], which is expected to be proportional to ω and weakly dependent on *T* above 100 K [4].

The data from NIST in Fig. 1 (112 K to 752 K) were fit to a function that includes three point-defect relaxations, Akhiezer loss approximated as independent of *T*, a constant background, and a broad relaxation consisting of a continuous set of Debye functions with a log-normal distribution of activation energies. The last term was found necessary to accurately fit the data [4]. Piezoelectric/carrier loss was not included, because initial analysis indicated it to be a minor contribution at all measured temperatures. The results of this fit at 6 MHz, simultaneously performed on data from two additional harmonics, are shown in the figure.

A fit of the TUC data (Fig. 1) accurately matches the data without including a distributed relaxation term. To simplify the figure, contributions to this fit are only plotted above the range of the NIST data and Peak 1 is not included. The piezoelectric peak position is consistent with predictions based on the measured temperature-dependent conductivity and dielectric constant [7]. The constant term in the fit is 1.0×10^{-4}, two orders of magnitude greater than the constant in the fit of the NIST data. This difference may be due to greater mechanical contact and/or anelasticity in the Pt electrodes.

LGT and CTGS

Similar point-defect and piezoelectric/carrier contributions have been reported in LGT and CTGS at resonant frequencies in the low megahertz range [4,7,8]. Two point-defect peaks are typically observed between 300 K and 1000 K. Lower conductivity and corresponding τ_c of CTGS, relative to LGS, lead to lower piezoelectric carrier loss over the measured ranges of temperature, since the peak maximum is shifted to higher temperatures [7].

In an LGT crystal measured at NIST, no broad temperature-dependent contribution similar to that observed in LGS (NIST, Fig. 1) was detected [4]. The fact that dislocation density was much lower in this LGT crystal supports the hypothesis that the physical mechanism responsible for this contribution in LGS involves anelastic motion of kinks in dislocations [4].

Suhak *et al*. [7] found evidence for a broadly temperature-dependent background loss in Y-cut CTGS and employed a simpler Arrhenius form to fit this background. They also found the constant term in the fitting function to be much greater for a CTGS crystal with Pt electrodes than for a different CTGS crystal without electrodes.

Conclusions

LGS, LGT, and CTGS display similar features in the temperature dependence of acoustic loss, although the magnitudes and peak positions of loss contributions vary. The general form of the piezoelectric/carrier contribution and its dependence on electrical conductivity is well understood. However, specific defects responsible for point-defect peaks have not been identified. Such identification may be less than straightforward because correlations of peaks with impurity concentrations can be indirect. In particular, defect symmetries and associated relaxations can depend on charge states that change with dopant levels. The physical mechanisms responsible for observed contributions with broad temperature dependence are also not established. The combination of results from multiple crystals suggests that these mechanisms include effects that are both internal and external to the piezoelectric material.

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[2] Fritze, H., High-temperature piezoelectric crystals and devices, *J. Electroceram.* 26, 122-161 (2011); doi: 10.1007/s10832-011-9639-6

[3] Johnson, W., Acoustic and Electrical Properties of Piezoelectric Materials for High-Temperature Sensing Applications, *Proc. SENSOR 2015*, 384-389 (2015); doi: 10.5162/sensor2015/C3.1

[4] Johnson, W., Kim, S. A., Uda, S., and Rivenbark, C. F., Contributions to anelasticity in langasite and langatate, J. Appl. Phys. 110, 123528 (2011); doi: 10.1063/1.3672443

[5] A. S. Nowick and B. S. Berry, *Anelastic Relaxation in Crystalline Solids* (Academic, NY, 1972).

[6] Akhiezer, A., On the absorption of sound in solids, *J. Phys. USSR 1*, 277-287 (1939).

[7] Suhak et al., Electromechanical properties and charge transport of $Ca_3TaGa_3Si_2O_{14}$, Solid State Ionics 317, 221-228 (2018); doi: 10.1016/j.ssi.2018.01.032

[8] Johnson, W. L., High-Temperature Electroacoustic Characterization of Y-Cut and Singly-Rotated $Ca_3TaGa_3Si_2O_{14}$ Resonators, *IEEE T. Ultrason. Ferr.* 61, 1433-1441 (2014); doi: 10.1109/TUFFC.2014.3052