Mechanisms of Anelastic Loss in Langasite at Temperatures from 113 K to 1324 K

Ward L. Johnson[1], Yuriy Suhak[2], and Holger Fritze[2]
[1]Applied Chemicals and Materials Division, National Institute of Standards and Technology, 325 Broadway St., MS 647, Boulder, CO 80305, USA
[2]Clausthal University of Technology, Am Stollen 19B, Goslar 38640, Germany
ward.johnson@nist.gov

Summary:

Synthetic piezoelectric crystals with the structure of langasite (LGS) are being pursued for resonant acoustic sensors that can operate at temperatures exceeding the range of conventional piezoelectric materials. The optimization of these crystals is currently focused primarily on minimization of acoustic loss, which degrades signal strength and resolution of sensors. This paper presents analysis and discussion of two sets of measurements of loss of LGS with a combined temperature range of 113 K to 1324 K. Physical mechanisms for the loss include intrinsic phonon-phonon interactions, multiple point-defect relaxations, piezoelectric/carrier loss, contact loss, and, perhaps, dislocation relaxations [1].

Keywords: acoustic loss, piezoelectric sensors, high temperatures, langasite, LGS, langatate, LGT catangasite, CTGS, quartz

Introduction

Traditional piezoelectric sensors are limited to operation at temperatures below several hundred degrees Celsius, because crystal transformations or degradation occur at higher temperatures in common commercially available piezoelectrics [2]. However, substantial research in recent decades has focused on synthesizing and optimizing innovative piezoelectric crystals that can be used in resonant sensors at temperatures exceeding 1000 K [3], including crystals with the structure of langasite ($La_3Ga_5SiO_{14}$, "LGS"), often termed members of the "langasite family."

The performance of resonators in sensing applications is limited by acoustic loss Q^{-1}. Within the langasite family of piezoelectric crystals, LGS has not been found to have the lowest Q^{-1}. For example, resonators of langatate ($La_3Ga_{5.5}Ta_{0.5}O_{14}$, "LGT") and langanite ($La_3Ga_{5.5}Nb_{0.5}O_{14}$, "LGN") at room temperature are reported to have lower loss than similarly manufactured LGS resonators [4]. Crystals of LGT [5] and catangasite ($Ca_3TaGa_3Si_2O_{14}$, "CTGS") [6] have also been found to have lower Q^{-1} at elevated temperatures (*e.g.*, above 200 °C) than that of any reported LGS specimen.

Despite the less-than-stellar quality factor of LGS, the available data on this material currently provide unique information on physical mechanisms that contribute to loss in crystals in the langasite family. Specifically, the range of temperatures over which Q^{-1} in LGS has been measured in the low megahertz range is exceptionally broad, enabling identification of contributions to the loss ranging from the small intrinsic loss associated with phonon-phonon interactions to conductivity-related loss five orders of magnitude larger at elevated temperatures. Analysis and discussion of these LGS data are the focus of this paper.

Results and Discussion

Figure 1(a) shows measurements of Q^{-1} of two Y-cut LGS crystals grown by different manufacturers [2,5]. Measurements on one crystal in vacuum were acquired from 113 K to 752 K with noncontacting electrodes at the National Institute of Standards and Technology (NIST, U.S.A.). Measurements on the other crystal in air were acquired from 309 K to 1324 K with Pt surface electrodes at Clausthal University of Technology (TUC, Germany).

This figure also shows the maximum Q^{-1} at 10 MHz reported for LGS at room temperature with noncontacting electrodes [4]. This Q^{-1} is an order of magnitude smaller than that measured near room temperature on the LGS specimen at NIST, indicating the presence of greater material loss in the NIST specimen. Q^{-1} of the specimen measured at TUC is an additional order of magnitude greater near room temperature. For the purpose of characterizing non-intrinsic contributions to the loss, the relatively high Q^{-1} of the NIST and TUC specimens is advantageous.

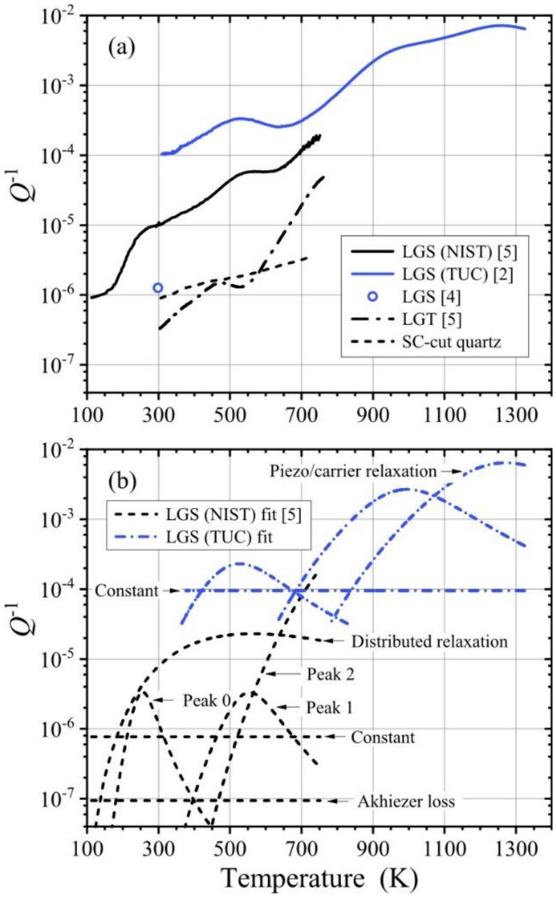

Fig. 1: (a) Q^{-1} of two LGS crystals measured at NIST [5] and TUC [2], an LGS crystal with lowest reported loss [4], an LGT crystal [5], and a swept SC-cut quartz crystal. The resonant frequencies near ambient temperature are, respectively, 6.1 MHz [5], 5.0 MHz [2], 10.0 MHz [4], 6.0 MHz [5], and 10.0 MHz. (b) Contributions to Q^{-1} of the NIST and TUC LGS crystals, determined from least-squares fits.

For comparison, Figure 1(a) also includes data on LGT from 302 K to 759 K [5] and data on swept SC-cut quartz from 306 K to 717 K, all obtained with noncontacting electrodes at NIST.

The LGS data from NIST in Fig. 1(a), along with simultaneously acquired data from two additional harmonics of this crystal, were fit to a function that includes intrinsic phonon-phonon (Akhiezer) loss (approximated as proportional to frequency and independent of temperature) [5], three anelastic point-defect relaxations [7], a constant frequency-independent background, and a broad relaxation consisting of a continuous set of Debye functions [7] with a log-normal distribution of activation energies. The physical mechanism responsible for the last term is hypothesized as arising from dislocations [5], consistent with the fact that no such term is required to fit the data in Fig. 1(a) for LGT, which has much lower dislocation density [5].

Results of fitting of the TUC data at the single measured harmonic are consistent with the NIST results for LGS, with respect to the temperatures of Peaks 1 and 2, considering the difference in resonant frequency. They reveal an additional large peak with a maximum near 1260 K, consistent in form with an expected relaxation involving the motion of charge carriers in acoustically generated piezoelectric fields [5,6]. The fit accurately matches the data without a broadly temperature-dependent term (*e.g.*, distributed relaxation). The constant term, which is inseparable from the Akhiezer term in the absence of measurements of additional harmonics, is two orders of magnitude greater than that determined for the LGS specimen at NIST. This difference is attributed primarily to greater mechanical contact.

Conclusions

Analysis of LGS data from 113 K to 1324 K reveals a number of anelastic loss mechanisms, including intrinsic loss, point-defect relaxations, piezoelectric/carrier relaxation, a constant background, and a broad background that may arise from dislocations. Similar effects have been reported in LGT and CTGS, even when the crystals are state-of-the-art, such as the LGT in Fig. 1(a). Despite the identification of the general nature of loss contributions, the optimization of these innovative piezoelectric materials for applications at elevated temperatures is far from complete. This situation is contrasted, here, with that of swept SC-cut quartz, which shows no evidence for point-defect relaxations over a more limited range of measured temperatures.

[1] This manuscript is a contribution of the National Institute of Standards and Technology and is not subject to copyright in the United States.

[2] Fritze, H., High-temperature bulk acoustic wave sensors, *Meas. Sci. Technol.* 22, 12002 (2011); doi: 10.1088/0957-0233/22/1/012002

[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] Smythe, R. C., Langasite, langanite, and langatate bulk-wave Y-cut resonators, IEEE T. Ultrason. Ferr. 47, 355 (2000); doi: 10.1109/58.827420

[5] 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

[6] 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

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