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Measurements of Acoustical Physical Constants and Their Temperature Coefficients of $La_3Ta_{0.5}Ga_{5.3}Al_{0.2}O_{14}$ Single Crystal

$La_3Ta_{0.5}Ga_{5.3}Al_{0.2}O_{14}$ 単結晶の音響関連物理定数と温度係数の測定

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1. Introduction

$La_3Ta_{0.5}Ga_{5.3}Al_{0.2}O_{14}$ (LTGA), one of Langasite crystal family, is a promising material for temperature and pressure sensors operating under high temperature environment, such as automobile engine and turbine, because of 4-times larger electromechanical coupling factor, comparable temperature coefficients and aging properties as compared to α-quartz, and no phase transition up to melting point around 1500°C. However, its acoustic properties and homogeneities have not sufficiently been investigated, so it is necessary to evaluate such information.

We have been working on development and application of the ultrasonic micro-spectroscopy (UMS) technology in which the line-focus-beam ultrasonic-material-characterization (LFB-UMC) system plays a central role[1, 2]. We have verified usefulness of this technology through evaluating the homogeneity and accurately determining acoustical physical constants for such as $LiNbO_3$, $LiTaO_3$, α-quartz, ZnO [3, 4]. However, it is difficult to measure the acoustic properties at high temperature which are important information for such materials aiming for operation at high temperature.

In this paper, we propose and demonstrate a method for determining precise constants covering desired temperature range by combining our technology with the resonance method [5] which is possible to measure temperature coefficients at wide temperature range taking LTGA single crystal.

2. Experiments

(a) Specimens: We prepared total six specimens for measurements of acoustic wave velocities by the UMS system, two *Z*-cut, *X*-cut, *Y*-cut, (052) (=29.14°*Y*-cut), and (502) (=150.86°*Y*-cut) specimens, from an LTGA single crystal ingot (54 mm in diameter and 55 mm in length) grown along the crystallographic *Z*-axis direction by the Czochralski technique. These specimens were about 2-mm thick except *X*-cut (4 mm) and prepared with both surfaces optically polished. We also prepared *X*- and *Z*-cut substrates with dimensions of 20×20×0.5 mm^3 for measurements of dielectric constants. We prepared four *X*-cut rotated *Y*-bar (-30°*Y*, -15°*Y*, 0°*Y*, +30°*Y*) and *Y*-cut specimens with dimensions of 20×1.5×0.5 mm^3 for measurements of resonance and antiresonance frequencies. In addition, *X*- and *Z*-bar specimens with dimensions of 1×1×20 mm^3 were prepared for measurements of thermal expansion.

(b) LSAW velocity: Measurement principle of leaky surface acoustic wave (LSAW) velocity by the LFB-UMC system is described in detail in [1]. Through the measurements of LSAW velocity distributions for each specimen at 225 MHz, we observed velocity distributions of 2.5 m/s at maximum for *Z*-cut specimen due to core around central axis of the ingot.

(c) Bulk wave velocity: Using the PW-UMC system [2], we measured longitudinal wave velocities at 50-450 MHz and shear wave velocities at 40-200 MHz at temperatures of 20, 23, and 26°C avoiding the core position. We could not observe any velocity dispersions in these frequency ranges for all results. Temperature coefficients for all measured velocities were almost linear around room temperature.

(d) Others: We obtained dielectric constants from the capacitance measurements for the *X*- and *Z*-cut plates in a range from -30 to +80°C. Thermal expansion coefficients were measured in a range from -100 to +600°C with the thermo-mechanical analysis equipment. Density was measured at 23°C based on the Archimedes method using the *X*-cut specimen for acoustic wave measurement and its temperature coefficient was determined from the thermal expansion coefficients obtained. The results were shown in **Table I**.

3. Constant Determination

According to the procedure of the constant determination [3], we obtained acoustical physical constants at 20, 23, and 26°C from the results of bulk wave velocities in section 2(c). The results at 23°C were shown in Table I. Each constant varies linearly in the range of 20-26°C. To confirm the accuracy of the determined constants, we compared the measured LSAW velocities and calculated ones using the determined constants at 23°C, resulting in

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good agreement within -3.0 to 1.1 m/s for all propagation directions.

Next, we obtained acoustical physical constants from the resonance and antiresonance frequencies measured in a range from -30 to 80°C. The obtained constants at 23°C differed from those determined by the UMS technology by -37 to 13% in elastic constants and by -60 to 8% in piezoelectric constants. Significant differences in temperature coefficient were observed especially for c_{33}^E as shown in **Fig. 1**. Correcting the temperature coefficients for each constant measured by the resonance method to adjust the linearly approximated slope in the range of 20-26°C to those determined by the UMS technology, we obtained the temperature coefficients in the range from -30 to 80°C as shown in Table I.

4. Discussion

For the crystal of point group 32, longitudinal and shear waves propagating along *Z*-axis directly relate to c_{33}^E and c_{44}^E. However, these propagation modes cannot directly be excited by the resonance method because these do not couple with piezoelectricity, resulting in determination from the complex equations of several propagation modes related with several constants. Significant difference in temperature coefficient in Fig. 1 arises from a stack of the slight errors in those for the other constants. Differences between absolute constants at 23°C determined by the UMS technology and the resonance method might be suggested some problems in the calibration of the equipment of the resonator measurement because the measured resonance (or antiresonance) frequency tended to upward.

5. Summary

We proposed a method for determining accurate acoustical physical constants at wide temperature range by combining the resonance method with the UMS technology taking LTGA single crystal. Obtaining the accurate constants and their temperature coefficients by the UMS technology even for narrow temperature range around room temperature, we demonstrated that we were able to calibrate the resonance method and to conduct more reliable determination of constants.

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Table I. Acoustical physical constants of LTGA single crystal and their temperature coefficients.

	Absolute values 23°C	Normal. temp. coefficients	
		$\frac{1}{x}\frac{\partial x}{\partial T}$	$\frac{1}{2x}\frac{\partial^2 x}{\partial T^2}$
Elastic constant	10^{11}[N/m^2]	10^{-6}[/°C]	10^{-8}[/°C^2]
c_{11}^E	1.888	-138	256
c_{12}^E	1.077	-150	254
c_{13}^E	0.999	-78.3	462
c_{14}^E	0.136	-52.2	-5.76
c_{33}^E	2.608	-290	77.3
c_{44}^E	0.513	-0.424	-23.0
Piezoelectric constant	[C/m^2]	10^{-6}[/°C]	10^{-8}[/°C^2]
e_{11}	-0.474	-14.3	19.6
e_{14}	0.057	189	-115
Dielectric constant		10^{-6}[/°C]	10^{-8}[/°C^2]
$\varepsilon_{11}^S/\varepsilon_0$	18.8	252	246
$\varepsilon_{33}^S/\varepsilon_0$	74.6	-1.03×10^5	1.47×10^4
Thermal expansion		10^{-6}[/°C]	10^{-8}[/°C^2]
α_{11}		5.61	0.801
α_{33}		3.85	0.428
Density	[kg/m^3]	10^{-2}[/°C]	10^{-4}[/°C^2]
ρ	6108.8	-9.20	-1.23

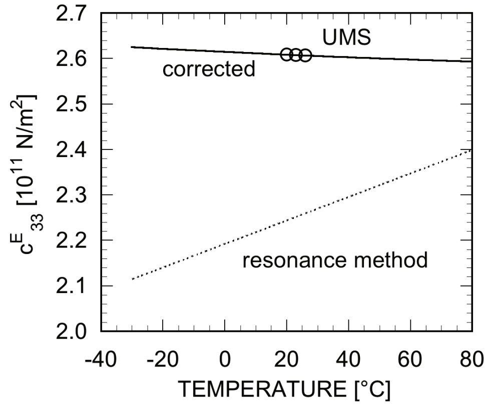

Fig. 1. Temperature dependences of c_{33}^E.

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