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Measurements of Acoustical Physical Constants and Their Temperature Coefficients for Langasite Family Crystals
ランガサイト系結晶の音響関連物理定数と温度特性の測定

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1. Introduction

Langasite family crystals are promising materials for temperature and pressure sensors operating under high temperature environment as well as high stability oscillators for future communication applications 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. Their acoustical physical constants and temperature coefficients are of fundamental for designing acoustic devices, so it is important to develop a method for determining constants and temperature coefficients accurately. The authors have been proposing a method for determining accurate constants by combining ultrasonic micro-spectroscopy (UMS) technology,[1-3] which can determine accurate constants around room temperature,[4] with the resonance method, which is possible to measure temperature coefficients at wide temperature range, taking $La_3Ta_{0.5}Ga_{5.3}Al_{0.2}O_{14}$ (LTGA) single crystal.

In this paper, we extended the constant determination method to $La_3Ga_5SiO_{14}$(LGS) and $Ca_3NbGa_3Si_2O_{14}$(CNGS) belonging to langasite family crystals and discussed merits and problems of this method.

2. Experiments

2-1. Specimens

We prepared five specimens for measurements by the UMS system(*X*-, *Y*-, *Z*-, and two rotated *Y*-cut), two specimens for measurements of dielectric constants(*X*- and *Z*-cut), five specimens for measurements of resonance and antiresonance frequencies (4 kind of *X*-cut rotated *Y*-bar and *Y*-cut), and two specimens for measurements of thermal expansion coefficients (*X*- and *Z*-bar) from each ingot of LGS, LTGA, and CNGS.

2-2. Measurements

Using the UMS system, we measured longitudinal wave velocities at 50-450 MHz and shear wave velocities at 40-200 MHz at 20, 23, and 26°C. 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.

We obtained dielectric constants from the capacitance measurements in a temperature range from -30 to +80°C. Thermal expansion coefficients were measured in a range from -100 to +600°C with heating rate of 3°C/min by using the thermo-mechanical analysis (TMA) equipment. Density was measured at 23°C based on the Archimedes method and its temperature coefficient was determined from the thermal expansion coefficients obtained. Resonance and antiresonance frequencies were measured each about 10°C in a temperature range from -30 to +80°C.

3. Determining constants

According to the procedure of the constant determination,[4] we obtained acoustical physical constants at 20, 23, and 26°C from the velocities of longitudinal and shear waves measured by the UMS system. Each constant varies linearly in the range of 20-26°C. The results of absolute values at 23°C (converted to elastic compliance constants s_{ij} and piezoelectric strain constants d_{ij}) and temperature coefficients for LTGA and CNGS were shown in Table I. We determined signs of piezoelectric constants according to the IEEE Standard.[5]

Next, we determined acoustical physical constants from the resonance and antiresonance frequencies measured in a range from -30 to 80°C according to refs. 5 and 6. Temperature coefficients normalized at 23°C for LTGA and CNGS were shown in Table I.

4. Discussion

To confirm the accuracy of the constants determined by the UMS, we compared the measured velocities of leaky surface acoustic wave (LSAW) obtained by the line-focus-beam ultrasonic material characterization system[1,2] and calculated ones using the determined constants at 23°C, resulting in good agreement within -3.0 to 2.9 m/s for all propagation directions. In Table I, comparing the temperature coefficients around room temperature obtained from the UMS and those of first-order obtained from the resonance method, most results were relatively good agreement with each other although significant differences were observed in $s^E{}_{13}$, $s^E{}_{33}$, d_{11}, and d_{14}.

Longitudinal and shear waves propagating along *Z*-axis for crystals of point group 32 can be directly excited and measured by the UMS technology regardless of piezoelectric constants, such as $c^E{}_{33}=\rho V_{Zl}{}^2$ and $c^E{}_{44}=\rho V_{ZS}{}^2$. However, these propagation modes cannot directly be excited by the resonance method, resulting in determination from the complex equations of several propagation modes related with several constants. Because $s^E{}_{33}$ and $s^E{}_{13}$ are deeply concerned with $c^E{}_{33}$, determination errors for these constants become large in the case of the resonance method. In the same manner, errors for $s^E{}_{44}$ and $s^E{}_{14}$ deeply concerning with $c^E{}_{44}$ become large. $s^E{}_{13}$ and $s^E{}_{33}$ among elastic constants and d_{14} among piezoelectric constants are less sensitive to oscillation of length-extensional modes for *X*-cut rotated *Y*-bar. We observed that signs of the temperature coefficients for $s^E{}_{33}$ and d_{14} obtained by the resonance method in Table I differed from the results for the UMS, it is considered to be a common problem in the resonance method. It will be improved by employing a pulse-echo method having external excitation source for measurements of velocities of *Z*-propagating longitudinal wave and rotated *Y*-propagating *X*-polarized shear wave. Using the UMS technology, the absolute constants are guaranteed and the temperature coefficients around room temperature can be obtained as a reference for confirmation of the accuracy of the results obtained by the resonance method.

5. Summary

We investigated a method for determining accurate acoustical physical constants at wide temperature range by combining the resonance method with the UMS technology extending to LGS and CNGS as well as LTGA. Using the UMS technology, we demonstrated that we can obtain accurate constants and temperature coefficients around room temperature.

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Table I. Acoustical physical constants and temperature coefficients of langasite family crystals.

		LTGA				CNGS			
		UMS		Resonance method		UMS		Resonance method	
		Absolute value at 23°C	$\frac{1}{x}\frac{\partial x}{\partial T}$ [/°C]	$\frac{1}{x}\frac{\partial x}{\partial T}$ [/°C]	$\frac{1}{2x}\frac{\partial^2 x}{\partial T^2}$ [/°C^2]	Absolute value at 23°C	$\frac{1}{x}\frac{\partial x}{\partial T}$ [/°C]	$\frac{1}{x}\frac{\partial x}{\partial T}$ [/°C]	$\frac{1}{2x}\frac{\partial^2 x}{\partial T^2}$ [/°C^2]
Elastic	$s^E{}_{11}$	9.049	-3.91E-5	-3.22E-5	1.77E-7	9.060	1.19E-4	1.17E-4	-2.27E-8
constant	$s^E{}_{12}$	-4.503	-1.93E-4	-2.10E-4	1.37E-7	-3.443	1.89E-4	6.92E-5	-1.74E-7
10^{-12} [m^2/N]	$s^E{}_{13}$	-1.741	1.50E-4	3.82E-5	-5.54E-6	-1.903	1.58E-6	2.03E-4	-5.34E-7
	$s^E{}_{14}$	-3.605	-4.73E-4	-4.66E-4	1.20E-6	-0.120	1.19E-2	7.06E-3	-3.31E-6
	$s^E{}_{33}$	5.168	1.30E-4	-2.50E-4	-1.11E-5	5.706	5.66E-5	-1.18E-4	-6.33E-7
	$s^E{}_{44}$	21.42	-7.59E-5	-6.05E-5	2.59E-7	24.66	-1.81E-4	-1.27E-4	3.11E-7
Piezoelectric	d_{11}	6.624	5.05E-5	-1.45E-5	1.45E-6	4.367	1.03E-3	8.05E-5	3.56E-7
constant [pC/N]	d_{14}	-4.643	6.36E-4	-1.50E-3	6.03E-6	7.567	-1.43E-2	-3.97E-4	6.39E-7
Dielectric	$\varepsilon^T{}_{11}/\varepsilon_0$	19.6	2.08E-5	2.69E-5	3.38E-8	16.7	-2.87E-4	-2.40E-4	7.36E-7
constant	$\varepsilon^T{}_{33}/\varepsilon_0$	74.6	-1.38E-3	-1.44E-3	1.98E-6	27.9	-4.40E-4	-4.36E-4	7.57E-7
Thermal	α_{11}			5.61E-6	8.01E-9			7.37E-6	1.59E-9
expansion	α_{33}			3.85E-6	4.28E-9			5.93E-6	3.25E-9
Density [kg/m^3]	ρ	6108.8		-1.51E-5	-2.01E-8	4119.3		-2.07E-5	-6.43E-9