Temperature Dependence of Acoustic Property for Ca₃Ta(Ga,Al)₃Si₂O₁₄ Single Crystals

Ca₃Ta(Ga,Al)₃Si₂O₁₄単結晶の音響特性の温度依存性

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1. Introduction

Ca₃TaGa₃Si₂O₁₄ (CTGS), one of the langasite-type single crystals, is an attractive material for high stability oscillators as well as pressure sensors operating under high temperature environment [1, 2]. There have been some reports on the effects of Al substitution for the Ga site, i. e., the Al substitution improved the piezoelectric constant and electrical resistivity of the langasite-type single crystals [3, 4]. Recently, we have successfully grown Al-substituted CTGS, $Ca_{3}Ta(Ga_{1x}Al_{x})_{3}Si_{2}O_{14}$ [CTGAS(x)] with x=0, 0.25, 0.50, and 0.75, by Czochralski technique [5]. And we have measured Al content dependence of acoustic properties for CTGAS(x), resulting in linear relationship between acoustic properties and Al content [6]. However, temperature dependence of the acoustic properties was not clear. Temperature coefficient of acoustic property is one of the most important parameter for piezoelectric device applications.

In this paper, we examined dependences of temperature coefficient of acoustical properties on Al substitution for CTGAS(x) single crystals using the ultrasonic micro-spectroscopy (UMS) technology [7].

2. Specimens

We grew CTGAS(x) single crystals with different compositions of x=0, 0.25, and 0.50 by Czochralski method pulling along Y-axis [5]. All crystals are about 1 inch^{Φ}×60 mm^L. We prepared three specimens of X-, Y-, and Z-cut with 2-mm thickness from each crystal ingot.

3. Experiments

Using the plane wave ultrasonic material characterization (PW-UMC) system [7], which is one of the main system of the UMS technology, we measured longitudinal wave velocities at 50-300 MHz and shear wave velocities at 50-200 MHz at around room temperature. We could not observe



Fig. 1 Temperature dependence of Z-propagating longitudinal wave velocities for CTGAS(x).



Fig. 2 Temperature dependence of Z-propagating shear wave velocities for CTGAS(x).

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any velocity dispersions in these frequency ranges for all results. Figs. 1 and 2 are one of the results of longitudinal and shear velocities for Z-cut specimen as a function of temperature. Bulk wave velocities for all propagation directions linearly changed with temperature.

4. Discussion

From the measured results including Figs. 1 and 2, we found that bulk waves linearly change with around room temperature at temperature. Therefore, we plotted the temperature coefficient of the bulk wave velocities for each propagation direction as a function of Al substitution content x. Results are shown in Fig. 3. Al contents were measured by electron probe microanalyzer (EMPA). Temperature coefficients for X-, Y-. and Z-propagating bulk wave velocities linearly changed with Al substitution content. All of the results for longitudinal velocity in Fig. 3(a) were negative temperature coefficient. On the other hand, only for the Z-propagating shear velocity, the temperature coefficient was positive in Fig. 3(b). We can suggest that there exists propagation where the temperature coefficient direction becomes zero among rotated Y-axes for shear wave velocity. However, in the range over x=0.75, it is not necessarily true because the temperature coefficient for Z-propagation becomes negative value over x=0.75. Further investigation will be needed.

5. Conclusion

We examined dependence of temperature coefficient of acoustic properties for CTGAS on Al substitution content. Through measurements of bulk wave velocities for X-, Y-, and Z-prop. by using the PW-UMC system at around room temperature, we experimentally demonstrated that longitudinal and shear waves velocities for CTGAS linearly change with temperature, and the temperature coefficient of the velocities also linearly change with Al content for all propagation directions. In addition, we suggested that there exists propagation direction where the temperature coefficient becomes zero among rotated Y-axes for shear wave velocity.

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Fig. 3 Al substitution content dependence of temperature coefficient of bulk wave velocities for CTGAS(x).

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