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Influence of Sc substitution on acoustic properties of $Ca_3Ta(Ga_{0.9}Sc_{0.1})_3Si_2O_{14}$ single crystal

$Ca_3Ta(Ga_{0.9}Sc_{0.1})_3Si_2O_{14}$単結晶の Sc 置換による音響特性への影響

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1. Introduction

Langasite-type single crystals have been studied as piezoelectric materials since the 1980's, and ordered langasite-type single crystals were discovered around 2000. $Ca_3TaGa_3Si_2O_{14}$ [CTGS] which is one of the ordered langasite-type single crystals has the properties of low impedance and zero temperature coefficient of resonant frequency, so we have found the possibility of application to small and low power consumption oscillators with stable temperature characteristics. Furthermore, the growth and evaluation of piezoelectric properties of $Ca_3Ta(Ga_{1-x}Al_x)_3Si_2O_{14}$ [CTGAS(x)] single crystal[1], which is CTGS substituted by Al with smaller ionic radius into the Ga site were carried out. As a result, we clarified that the piezoelectric constant e_{11} and the electromechanical coupling coefficient k_{12} systematically increase[2]. On the other hand, $|e_{14}|$ decreases systematically as the amount of Al substitution increases, and it is expected that Al substitution works disadvantageously for devices using vibration mode with large influence of $|e_{14}|$.

In this paper, we examined influence of acoustical properties on Sc substitution for $Ca_3Ta(Ga_{0.9}Sc_{0.1})Si_2O_{14}$ [CTGSS(0.1)] single crystal, substitution of Sc with larger ionic radius into the Ga site by 10%, using the ultrasonic micro-spectroscopy (UMS) technology[3-5].

2. Specimens

CTGSS(0.1) single crystal was grown by Czochralski method pulling along *Y*-axis[6]. All crystals are about 1 inch$^{\Phi}$×45 mm^{L}. We prepared specimen of *X*-cut with 2-mm thickness from the crystal ingot. In addition, specimens of *X*-cut, with 0.5mm thickness were also prepared for the measurement of dielectric constant.

3. Experiments and results

Leaky surface acoustic wave (LSAW) velocities were measured for the 2-mm-thick specimen using the line-focus-beam ultrasonic material characterization (LFB-UMC) system[3, 4]. The result is shown in Fig. 1 with the data of CTGS and CTGAS(0.1)[2]. In Fig. 1, we found that LSAW velocity decreased with Sc substitution contrary to effect of increasing with Al substitution.

Lattice parameters of the grown crystals were calculated from the XRD patterns resulting in a = 8.110 [Å] and c = 4.995 [Å]. Density was also measured at 23°C based on the Archimedes method.

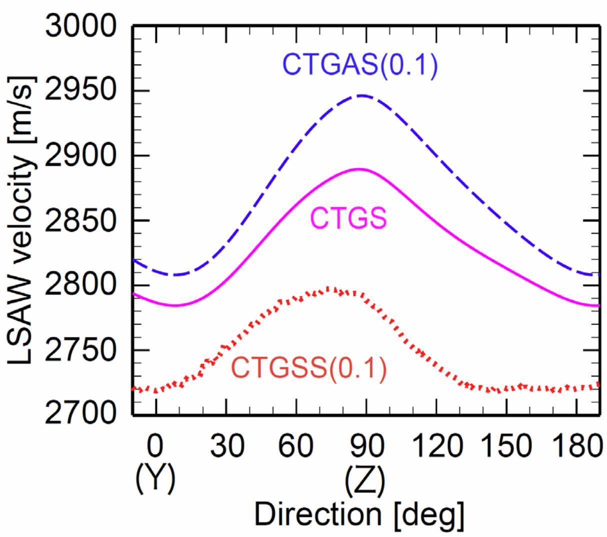

Fig. 1 Angular dependences of LSAW velocities for *X*-cut CTGSS(0.1), CTGS, and CTGAS(0.1).

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Measured density was 4587 kg/m^3 and this value showed smaller than that of CTGS and larger than that of CTGAS(0.1)[2]. Relative permittivity $\varepsilon_{11}{}^{T}/\varepsilon_0$ and resonance/anti-resonance frequencies (f_r, f_a) of the CTGSS(0.1) were measured using impedance analyzer and piezoelectric constants d_{11} was calculated from these constants. We obtained following constants; $\varepsilon^{T}{}_{11}/\varepsilon_0$ = 16.86, d_{11} = 3.7 pC/N. These values also showed change in the opposite direction as compared with substitution of Al for CTGS[2].

4. Discussion

In general, the sound velocity increases when the density decreases. In Fig. 1, however, the LSAW velocity decreases in spite of decreasing density. This suggests that the elastic constants for CTGSS(0.1) act to decrease the LSAW velocity. We calculated the influence of each elastic constant on change in LSAW velocity for X-cut CTGS. The result was summarized in Table I. In order to decrease the LSAW velocity, the elastic constants are required to change in decreasing direction for c_{12} and c_{13} and increasing direction for the rest. These changes were contrary to the effect of Al substitution [2]. In piezoelectric constant, d_{11} decreased due to the effect of Sc substitution. This change is opposite direction to the Al substitution. In the same manner, the dielectric constant showed the opposite change from that of Al substitution. These results suggest that $|e_{14}|$ increases due to Sc substitution.

Table I. Change of elastic constants with velocity change of LSAW propagating on the surface of *X*-cut CTGS.

LSAW velocity	c_{11}	c_{12}	c_{13}	c_{14}	c_{33}	c_{44}
increase	-	+	+	-	-	-
decrease	+	-	-	+	+	+

5. Conclusion

We examined influence of Sc substitution on acoustic properties of CTGSS(0.1). LSAW velocity of CTGSS(0.1) was lower than that of CTGS and it was found to show change in the direction opposite to Al substitution. The piezoelectric constant d_{11} and the dielectric constant showed change in the opposite direction to Al substitution. In addition a change in the elastic constant was suggested since the density was reduced by Sc substitution and the change direction was also expected to be opposite to Al substitution. From the above, it is suggested that the Sc substitution causes increase in $|e_{14}|$ which decreased with Al substitution.

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