Elastic Constants and Piezoelectric Coefficients of Langasite Single Crystal at Low Temperatures

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1. Introduction
Because of its good electromechanical coupling coefficients and relatively low sound velocities, langasite (La₃Ga₅SiO₁₄) is a candidate material for future applications of small and/or low power devices. As well as the engineering perspective, low temperature elastic constants $C_{ij}$ and piezoelectric coefficients $e_{ij}$ are also of our interest to fully understand the fundamental lattice dynamics from the acoustic point of view. Up to now, variety of measurement techniques(1-2) have been proposed to determine $C_{ij}$ and $e_{ij}$. Among them, resonant ultrasound spectroscopy (RUS)(3) is one of the attractive techniques since (i) it enable us to determine $C_{ij}$ and $e_{ij}$ simultaneously, and (ii) measurement accuracy for resonance frequency is extremely high (inaccuracy is approximately 10⁻⁶). In this study, we have developed the tripod-type low-temperature RUS equipment and determine $C_{ij}$ and $e_{ij}$ of langasite single crystal from ambient temperature down to 5.5 K.

2. Experiment procedure
The material used in this study is a rectangular parallelepiped shape langasite single crystal. Mass density is determined by the Archimedean method (see Table 1). Langasite belongs to the 32-point-group trigonal system, having six independent elastic constants ($C_{11}$, $C_{33}$, $C_{12}$, $C_{14}$, $C_{44}$), two piezoelectric coefficients ($e_{11}$, $e_{14}$) and two dielectric constants ($H_{11}$, $H_{33}$),

$$C_{ij} = \begin{bmatrix} C_{11} & C_{12} & C_{13} & C_{14} & 0 & 0 \\ C_{11} & C_{13} & -C_{14} & 0 & 0 & 0 \\ C_{13} & 0 & 0 & 0 & 0 & 0 \\ C_{14} & 0 & 0 & 0 & 0 & 0 \\ C_{44} & 0 & 0 & 0 & 0 \\ C_{66} & \end{bmatrix}, \quad (1)$$

$$e_{ij} = \begin{bmatrix} e_{11} - e_{11} & 0 & 0 & 0 \\ 0 & -e_{14} & -e_{11} & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ \end{bmatrix}, \quad (2)$$

$$\varepsilon_{ij} = \begin{bmatrix} \varepsilon_{11} & 0 & 0 \\ 0 & \varepsilon_{11} & 0 \\ \varepsilon_{33} & \varepsilon_{33} & \varepsilon_{33} \end{bmatrix}. \quad (3)$$

Where $C_{66} = (C_{11} - C_{12})/2$. In this study, we employed the dielectric constants, $\varepsilon_{11}/\varepsilon_{0} = 19.04$, $\varepsilon_{33}/\varepsilon_{0} = 50.51$ obtained from average of literatures at room temperature (4-6).

Free vibration resonance frequencies of a specimen depend on dimensions, mass density, and all independent material constants. We can therefore inversely determine $C_{ij}$ and $e_{ij}$ from experimentally obtained resonance frequencies $f_i$ when we know $H_{ij}$. For $f_i$ measurement, we employed a tripod transducer which consists of two piezoelectric needle transducers and a supporting needle. The specimen is mounted on the tripod transducer. One needle transducer excites vibrations in the specimen, and the other detects the vibrational amplitude as a function of the input frequency. Then we successively obtain a resonance spectrum from a frequency sweep around 1 MHz. Resonance frequencies are determined by the Lorentzian-function fitting. Note that the transducers require no coupling agents and no external forces except for the specimen’s weight.

Table 1. Dimension and mass density of the langasite specimen.

<table>
<thead>
<tr>
<th>Dimension (mm)</th>
<th>Density (g/cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$L_1$</td>
<td>$L_2$</td>
</tr>
<tr>
<td>9.810</td>
<td>8.030</td>
</tr>
<tr>
<td>$L_3$</td>
<td>$\rho$</td>
</tr>
<tr>
<td>6.021</td>
<td>5.71</td>
</tr>
</tbody>
</table>

Figure 1 shows the low temperature RUS measurement system developed in the present study. The specimen and tripod transducer are set in a cryogenic chamber. The chamber is fulfilled by...
He gas (about 1.4 atm) and connected with the cold-head cylinder. This system controls temperature of the specimen down to 5 K with an accuracy of 0.1 K.

Figure 1. Schematic drawing of the low temperature cryostat.

3. Results and discussion

Figure 2 shows resonance spectra of the langasite single crystal obtained by the RUS measurements. Unfortunately, the number of resonance peaks decreases with decreasing in temperature, which is caused by the mechanical vibration due to the cold head cylinder. However, more than 30 peaks can be detected throughout the measurements; they are sufficient to determine eight independent variables (two $e_{ij}$ and six $C_{ij}$).

Figure 2. Temperature dependence of resonance spectra for the langasite crystal obtained by RUS.

Table 2 summarizes ambient and low temperature $C_{ij}$, $e_{ij}$ and the Debye characteristic temperature $\Theta_D$ obtained from the langasite single crystal. Surprisingly, $\Theta_D$ keeps almost constant even at 5.5 K. Another notable feature is that $C_{66}$ shows weak but finite unusual temperature dependence which would be due to the effect of negative mode Grüneisen parameter.

Table 2. Elastic constants and piezoelectric coefficients of langasite at room temperature (R.T.) and 5.5K. The units are GPa for $C_{ij}$, C/m² for $e_{ij}$ and K for $\Theta_D$.

<table>
<thead>
<tr>
<th></th>
<th>$C_{11}$</th>
<th>$C_{12}$</th>
<th>$C_{13}$</th>
<th>$C_{14}$</th>
<th>$C_{33}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>R.T.</td>
<td>189.5</td>
<td>105.0</td>
<td>97.16</td>
<td>14.25</td>
<td>262.6</td>
</tr>
<tr>
<td>5.5K</td>
<td>191.8</td>
<td>108.2</td>
<td>99.63</td>
<td>15.51</td>
<td>269.1</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>$C_{44}$</th>
<th>$C_{66}$</th>
<th>$e_{11}$</th>
<th>$e_{14}$</th>
<th>$\Theta_D$</th>
</tr>
</thead>
<tbody>
<tr>
<td>R.T.</td>
<td>53.5</td>
<td>42.09</td>
<td>-0.397</td>
<td>0.203</td>
<td>414.5</td>
</tr>
<tr>
<td>5.5K</td>
<td>53.90</td>
<td>41.82</td>
<td>-</td>
<td>-</td>
<td>413.0</td>
</tr>
</tbody>
</table>

4. Conclusions

In summary, we developed the tripod type low temperature RUS measurement system. Elastic constants and piezoelectric coefficients for langasite single crystal have been successively obtained from ambient temperature to 5.5 K. Some resonance frequencies and $C_{66}$ showed unusual temperature dependence while acoustic Debye temperature $\Theta_D$ kept almost constant throughout the present temperature range.

References

6. J.Bohm, et al.: “*Czochralski growth and characterization of piezoelectric single crystals with langasite structure: La₃Ga₅SiO₁₄(LGS), La₃Ga₅.5Nb₀.5O₁₄(LGN) and La₃Ga₅.5Ta₀.5O₁₄ (LGT)*” II. *Piezoelectric and elastic properties*