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Monitoring of Sound Velocity and Attenuation in $SrTiO_3$ across the Phase Transition at Low Temperatures using Picosecond Ultrasonics

ピコ秒超音波法を用いた $SrTiO_3$ の低温相変態における音速と減衰のモニタリング

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1. Introduction

$SrTiO_3$ shows a cubic perovskite structure at room temperature, and it is widely used as a substrate for thin-films deposition in many fields. It has been pointed out for five decades that $SrTiO_3$ has several phases at low temperature region[1)]. Near 105 K, it shows a cubic-tetragonal phase transition as shown in **Fig. 1**, and moreover, it shows anomalies below 70 K. For example, the lattice constant decreases linearly with cooling to 100 K, and it shows a very sharp peak at 10 K[1)]. Behaviors of the elastic constants between 70 K and 37 K cannot be explained in the normal lattice anharmonicity[2)], and the permittivity shows a broad peak in low frequency range, showing frequency dependence[3)].

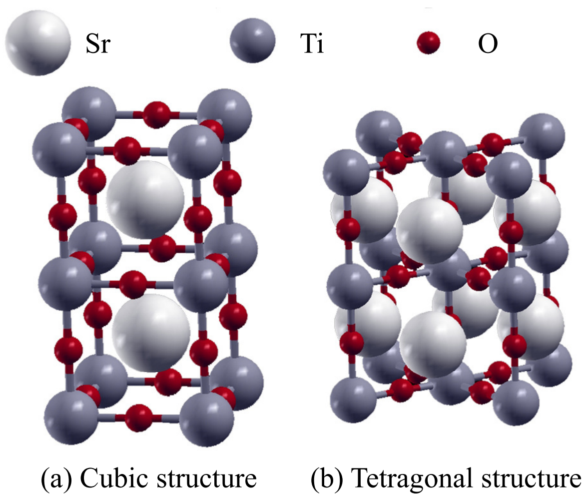

Fig. 1 The structures of $SrTiO_3$. (a) is cubic perovskite the structure(above 105K) and (b) is tetragonal the structure(below 105 K).

In this work, we monitored $SrTiO_3$'s phase transitions by ultrasonic measurements. We used picosecond ultrasonics spectroscopy which can generate longitudinal-ultrasound wave of very high frequency without contacts. We adopt 800 and 400 nm light pulses for pumping and probing coherent phonon pulses. Because $SrTiO_3$ is transparent for wavelength from 400 to 800 nm, attenuation of the signal reflects ultrasonic attenuation which is sensitive for revealing a phase transition. In the past, the relationship between $SrTiO_3$'s phase transition and ultrasonic attenuation has been investigated in the frequency region from 30 MHz to 300 MHz[4)]. However, a sub-terahertz-order ultrasonic measurement has never attempted to $SrTiO_3$, and it is important because such anomalies are expected to be sensitive to high frequencies because they are highly related with high-frequency phonons.

Moreover, few reports appear for attenuation measurements using the picosecond ultrasonics technique, and there is no report for monitoring phase transition using it due to difficulties of measurements at various temperatures. Thus, this work demonstrates the importance of picosecond ultrasonics at low temperatures for studying phase transformations of nano-scale materials

2. Experiment

Picosecond ultrasonics uses two ultrafast light pulses to generate an ultrashort strain pulse and to detect it. Firstly, the pump light pulse of 800 nm wavelength irradiates a 17-nm Pt thin film deposited on the (100) $SrTiO_3$ specimen. The film expands and shrinks in a very short time (~ 200 fs), which generates the ultrasonic pulse through lattice anharmonicity. The strain pulse makes the $SrTiO_3$'s dielectric constants change due to the piezo-elastic effect, leading to the change in the refractive index. Secondly, the time delayed probe light pulse of 400 nm wavelength is reflected and transmitted at the Pt thin film. The refractivity change in $SrTiO_3$ diffracts the transmitted light backward following the Bragg's condition of diffraction. The diffracted light interferences with the reflected light from the surface and the oscillating signals can be observed, whose frequency f is approximately given by

$$f = \frac{2nv}{\lambda} \quad (1)$$

where n is the refractive index of $SrTiO_3$ at the wavelength of the probe light (400 nm), v is the longitudinal-wave velocity in the specimen, and λ is

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the wavelength of the prove light in vacuum. This is called Brillouin oscillation. We measured *n* by the ellipsometry measurement, so the sound velocity can be determined by measuring the Brillouin-oscillation frequency. This technique leads to accurate measurements of the sound velocity within 1% error[5]. Furthermore we developed optics to perform picosecond ultrasonics between 20 K and 300 K, where we used a cryostat with a glass window as the specimen holder as shown in **Fig. 2**.

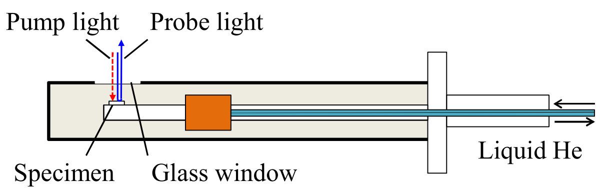

Fig. 2 The schematic of a cryostat specimen holder.

3. Results and Discussion

We have succeeded in monitoring the cubic-tetragonal phase transition through the velocity and attenuation as shown in **Figs. 3** and **4**. Toyoda *et al.* reported that the refractive index of $SrTiO_3$ is decreased with temperature near 400 nm between 350 and 700 K[6], and we assumed the similarly decrease to the cryogenic temperature and considered the temperature dependent of the refractive index.

Their behaviors are almost equivalent with cooling and heating up processes. However, the temperature dependence of the sound velocity is not consistent with Ref. 2. This may be due to the behavior of the refractive index at low temperature. Ang *et al.* reported that permittivity of $(Sr_{1\text{-}1.5x}Bi_x)TiO_3$ increased below 70 K and showed anomalies in the frequency range from 1 kHz to 220 MHz[3]. So refractive index at 400nm would more increase at cryogenic temperatures.

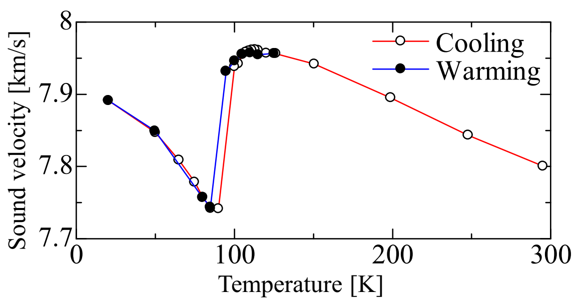

Fig. 3 The behavior of longitudinal-wave velocity.

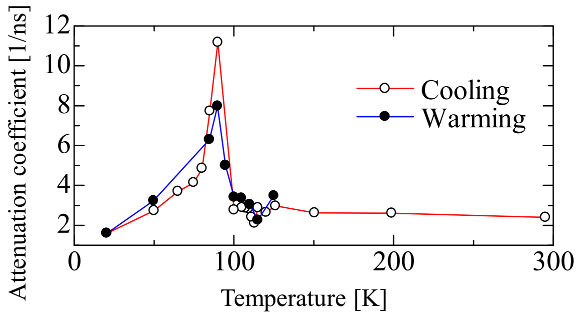

Fig. 4 The behavior of attenuation coefficient.

On the other hand, attenuation is not affected by the change in the refractive index, and we can compare our results to other measurements. Attenuations are very small at the room temperature and it remains unchanged before the phase transition. However, it shows a sharp peak at the cubic-tetragonal phase transition temperature, and it gradually decreased after that. No anomalies appeared in the low temperature and that the anomalies of $SrTiO_3$ below 70 K don't appear in the sub-terahertz frequency ultrasounds.

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