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Phonon Dynamics of $Pb(Sc_{1/2}Ta_{1/2})O_3$ Ceramics Studied by Brillouin Scattering Spectroscopy

ブリルアン散乱分光による $Pb(Sc_{1/2}Ta_{1/2})O_3$ セラミクスのフォノンダイナミクスの研究

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1. Introduction

Relaxor materials have drawn attentions since they have superior dielectric and piezoelectric properties, which contribute to the development of numerous applications. One of the charactaristic fuetures of relaxor is the diffusive dielectric anomaly in contrast to the sharp anomaly in normal ferroelectric materials such as $PbTiO_3$ [1]. The diffusive nature of relaxor is attributed to the compositional heterogeneity [1]. $Pb(Sc_{1/2}Ta_{1/2})O_3$ (PST) has been often used as an ideal model for the research of relaxor properties because the macroscopic properties of PST depend substantially on the degree of B-site order. PST belongs to the group of relaxor ferroelectrics with $A(B'_{1/2}B''_{1/2})O_3$ perovskite-type structure. The degree of B-site ordering of PST, which is represented by *S* parameter, can be controlled by heat treatment. Well-ordered PST (PST-O) shows a normal ferroelectric properties, whereas disordered PST (PST-D) exhibits relaxor behavior [2].

Brillouin scattering is one of the useful methods to study the elastic properties and associated relaxation dynamics of ferroelectric materials. There are several Brillouin scattering reports on PST single crystals [3-5], but the data of ceramics is not enough. Moreover, in the previous Brillouin studies on PST-D, only disoredered PST with Pb vacancies were examined. In the present study, we peformed Brillouin spectroscopy on PST-O and PST-D ceramics without Pb vacancy to investigate the anomalous behavior of acoustic phonon and central peak.

2. Experimental

Samples were translucent PST-O (S=0.89) and PST-D (S=0.09) ceramics without Pb vacancies. The degree of B-site ordering was determined by X-ray diffraction. Brillouin scattering measurement was performed by using a tandem Fabry-Perot interferometer in a backscattering geometry. The sample was excited by a diode-pumped solid state (DPSS) laser with a wavelength of 532nm. The sample temperature was controlled by a heating and cooling stage for microscope (THMS600) from 78 K to 773 K.

3. Result and Discussion

Figure 1 shows Brillouin spectra of PST-O and PST-D with a free spectral range (FSR) of 75 GHz at various temperatures. An inelastic peak which arises from longitudinal acoustic (LA) mode was observed at around 45 GHz in both PST-O and PST-D, and the spectra were fitted with the Voigt function. **Figure 2** shows the temperature dependences of the Brillouin shift and the full width

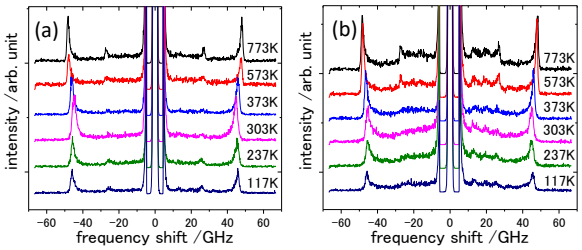

Fig. 1 Brillouin spectra of PST-O (a) and PST-D (b) observed with a free spectral range (FSR) of 75 GHz at various temperatures.

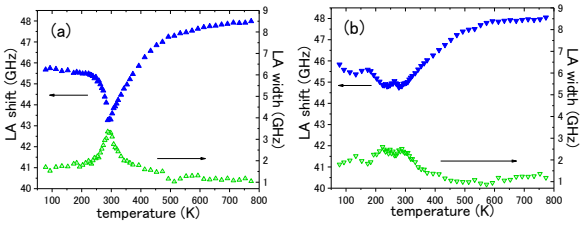

Fig. 2 Temperature dependence of the Brillouin shift and the FWHM of the LA mode of (a) ordered and (b) disordered PST ceramics.

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at half maximum (FWHM) of the LA mode of PST-O and PST-D. The width of the LA mode of PST ceramics is approximately 1.5 times as broad as that of single crystals [3-5], which may be attributed to weak multiple refractions and reflections at the grain boundaries [6]. PST-O ceramics shows a sharp acoustic anomaly at around *T*c, similarly to normal ferroelectrics, while PST-D exhibits a broad diffused anomaly being consistent with previous results [3-5]. This result clearly shows that 1:1 order is important for the formation of a long-range ferroelectric order at low temperature region.

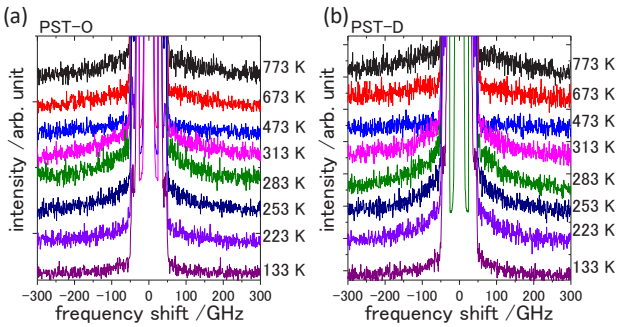

Fig. 3 Brillouin spectra of PST-O (a) and PST-D (b) observed with a free spectral range (FSR) of 375 GHz at various temperatures.

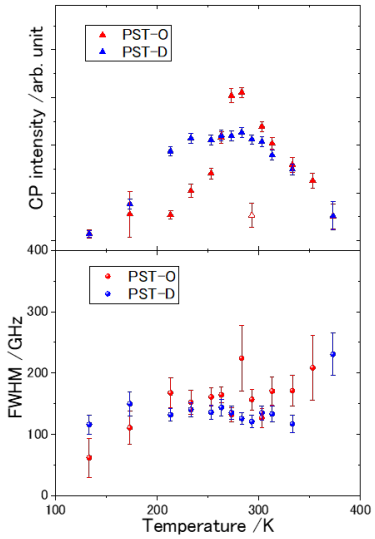

Fig. 4 Temperature dependence of the integrated intensity and the FWHM of the central peak of PST-O and PST-D.

Brillouin spectra with the FSR of 375 GHz at various temperatures are shown in **Fig. 3**. A central peak (CP) appears in both PST-O and PST-D ceramics at low temperatures. The CP of relaxors is important to investigate the relaxation dynamics related to the polarization fluctuations. **Figure 4** shows the temperature dependences of the integrated intensity and the FWHM of the CP. The integrated intensity of PST-O shows a sharp anomaly at around 283 K and that of PST-D shows a diffusive anomaly. These behaviors are similar to the temperature dependence of the LA mode. The anomaly of the CP intensity may indicate the increase of the polarization fluctuations corresponding to the ferroelectric instability near phase transition. In contract to the CP intensity, the CP FWHM of *both* PST-O and PST-D monotonically decreases and approaches to a certain value on cooling. This indicates that the relaxation time of the polarization fluctuations is independent of the degree of the B-site ordering in PST.

4. Conclusion

Sharp behaviors in the LA shift and the integrated intensity of the CP were observed in PST-O, while PST-D showed a broad anomalies. The temperature dependence of the integrated intensity of the CP showed a similar behavior to that of the LA shift. In contrast, the temperature dependence of the FWHM of the CP showed similar behaviors in both PST-O and PST-D regardless of the different degree of the B-site ordering. These results indicate that the B-site ordering is not responsible for the relaxation time of the polarization fluctuation in PST, whereas it is indispensable for the formation of a long-range ferroelectric order.

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