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Study of uniaxial relaxor ferroelectrics $Sr_xBa_{1-x}Nb_2O_6$ using Brillouin scattering

ブリルアン散乱法による一軸性リラクサー強誘電体 $Sr_xBa_{1-x}Nb_2O_6$ の研究

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1. Introduction

Strontium barium niobate, $Sr_xBa_{1-x}Nb_2O_6$ (SBN) is one of the technologically important ferroelectrics owing to its remarkablly large electro-optic and pyelectric coefficients useful for applications such as sensorics and data storage. SBN with tetragonal tungsten bronze structure (TTB) is a uniaxial ferroelectric with point groups 4mm and 4/mmm, respectively, below and above the Curie temperature, T_C. In TBB, A2 sites are occupied by Sr and Ba, while A1 sites are occuppied only by Sr owing to smaller ionic radius as shown in Fig. 1. 1/6 of all A sites (A1+A2 sites) are vacancies, which are the origin of random fields (RFs). With increasing Sr/Ba ratio, SBN transforms from normal to relaxor ferroelectrics [1]. The relaxor nature is attributed to the strength of random electric fields (RFs) due to the unfilled structure [2]. The compositional dependence of the lattice entropy in SBN was calculated as a function of the Sr/Ba ratio by a simple statistical model as shown in Fig. 2 [1]. It shows that relaxor nature strongly appears in the Sr rich region. In the present study, the temperature dependences of the elastic properties of SBN ($0.61 \leqq x \leqq 0.80$) were investigated by Brillouin scattering.

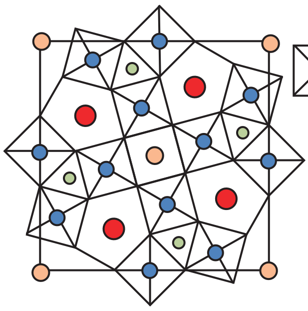

Fig. 1 Crystal structure of a unit cell of the tetragonal tungsten bronze (TTB) projected along the *c*-axis.

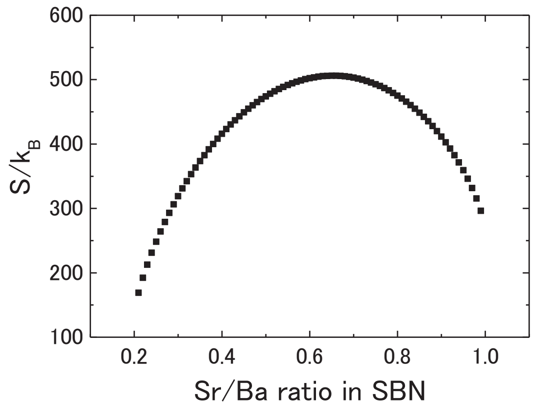

Fig. 2 The number of ways of distribution as a function of the Sr/Ba ratio *x* for 100 unit cells [1].

2. Experimental

$Sr_xBa_{1-x}Nb_2O_6$ (x=0.61, 0.70, 0.80) single crystals (SBN61, SBN70, SBN80) were grown by the Czochralski method [3]. Single crystals of high-quality were cut perpendicularly to their *c* and *a*-axes into parallelepipeds sized 5mm×5mm×1mm and polished. Brillouin scattering spectra were measured using a 3+3 pass tandem Fabry-Perot interferometer (FPI) with a single frequency green YAG laser at the wavelength of 532nm [4]. A free spectral range (FSR) of 75GHz was used to measure the frequency shifts. Brillouin scattering spectra were fitted by a Voigt function to analyze frequency shift and width of the LA mode. The temperature dependence was measured between -180°C and 300°C using a heating/cooling stage.

3. Results and discussion

Brillouin scattering spectra of from a SBN61 *c*-plate at selected temperatures are shown in Fig. 3. The longitudinal acoustic (LA) peaks were clearly observed at all temperatures, while transverse acoustic (TA) peaks appeared upon heating.

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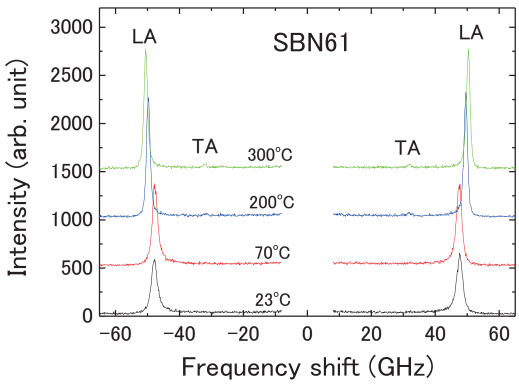

Fig. 3 Brillouin scattering spectra of SBN61 observed in backward scattering of a *c*-plate at a few selected temperatures.

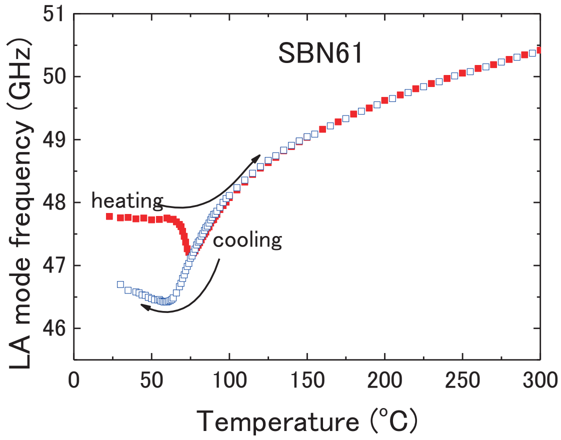

Fig. 4 Softening towards T_C and thermal hysteresis of LA mode frequency of SBN61 along the *c*-axis.

The temperature dependence of LA mode frequency along the *c*-axis of SBN61 is shown in Fig. 4. The remarkable softening towards $T_C \approx 75^oC$ [2] an be attributed to the interaction between the LA mode and polar nanoregions (PNRs). Thermal hysteresis was clearly observed. Upon heating a sharp minimum appears at about 75°C, while upon cooling the broad minimum appears around 60°C. Below T_C, the RF controlled metastable nanodomain state may enhance the difference upon cooling from the stable state behavior upon heating [6].

The temperature dependences of the LA mode frequencies of SBN61, 70, and 80 crystals on cooling are shown in Fig. 5. It is known that T_C decreases as the Sr/Ba ratio increases [7]. The temperature dependence of the LA mode frequency around T_C becomes more diffusive as the Sr/Ba ratio increases. These results are in good agreement with dielectric measurements [7]. The variation of diffusivity in a ferroelectric phase transition of SBN cannot be explained only by a simple approximation of distribution shown in Fig. 2. It was discussed that cations at A2 sites affect polar properties stronger than those at A1 sites [1]. The elastic anomaly along the *a*-axis was also studied as a function of the Sr/Ba ratio.

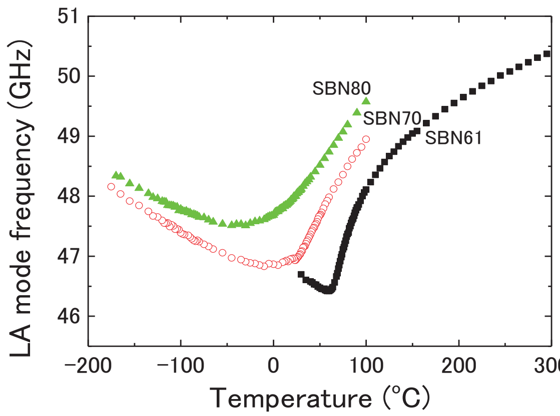

Fig. 5 Temperature dependences of LA mode frequency of SBN61, SBN70, and SBN80 observed in a *c*-plate upon cooling.

4. Conclusion

Elastic properties of SBN61, 70, and 80 crystals have been studied by Brillouin scattering in the temperature range between -190°C and 300°C. The LA mode frequency with the propagation along the *c*-axis shows a softening around T_C. A remarkable thermal hysteresis was clearly observed in SBN61 owing to the non-equilibrium state induced by RFs. The diffusive nature of elastic properties around the ferroelectric phase transition is enhanced with the increase of the Sr/Ba ratio owing to the increase of the strength of RFs.

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