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Elastic Anomaly of Uniaxial Ferroelectric Strontium Barium Niobate with Very Weak Random Fields

微弱なランダム場のある一軸性強誘電体ニオブ酸ストロンチウムバリウムの弾性異常

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1. Introduction

Ferroelectric materials with tetragonal tungsten bronze (TTB) structure are technologically important for optical applications such as electro-optic, nonlinear optic, and photorefractive devices. Since the direction of spontaneous polarization is only along the *c*-axis of TTB structure, they are called uniaxial ferroelectrics. The structural formula of TTB ferroelectrics is expressed by $(A1)_2(A2)_4(C)_4$-$(B1)_2(B2)_8O_{30}$ with corner sharing distorted BO_6 octahedra as shown in **Fig. 1**. In the case of ferroelectric $Ba_2NaNb_5O_{15}$, the A1 and A2 sites are completely occupied by Ba^{2+} and Na^{1+} ions. Thus belongs to so called the filled TTB ferroelectrics, which undergo a normal ferroeletric sharp transition [1].

On the other hand, in $Sr_xBa_{1-x}Nb_2O_6$ (SBN100*x*), the A1 sites are occupied only by Sr^{2+} ions and the A2 sites are occupied by both Ba^{2+} and Sr^{2+} ions. Since 1/6(A1+A2) sites remain unoccupied, it belongs to the unfilled TTB ferroelectrics. The randomly distributed vacancies at A1 and A2 sites are the main source of quenched random fields (RFs) due to the immanent charge disorder. They enhance the diffusivity of a broad phase transition. With the increase of the Sr content, the lattice entropy at A1 and A2 sites [2] increases as shown in **Fig. 2**, and the Curie temperature, T_C decreases. Consequently, the diffusive nature increases due to the increase of the strength of RFs, which are believed to play a dominant role for the relaxorness. Thie is because quenched random fluctuations of RFs induce polar nanoregions (PNRs) which have a dominant contribution to the precursor phenomena of the ferroelectric phase transition [3,4].

In the present study, the effect of very weak RFs on the elastic properties of SBN26 (x = 0.26) single crystals was investigated using Brillouin scattering spectroscopy. The observed result of SBN26 is compared with that of SBN80 (x = 0.80) with strong RFs.

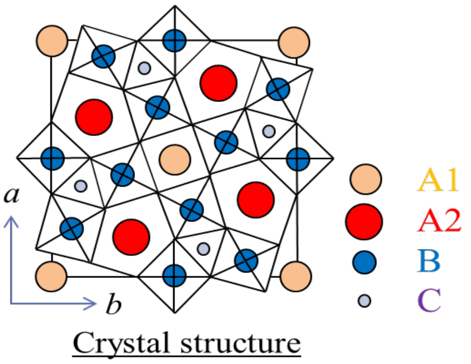

Fig. 1 Crystal structure of TTB ferroelectrics along the *c*-axis.

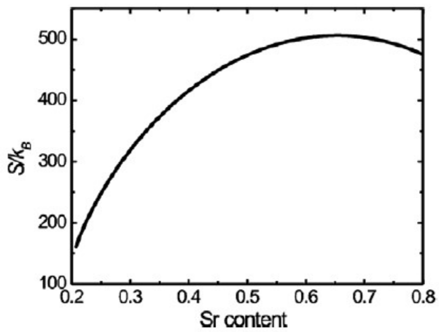

Fig. 2 Lattice entropy of strontium barium niobate with TTB structure [2].

2. Experimental procedure

$Sr_xBa_{1-x}Nb_2O_6$ single crystals were grown by the Czochralski method for the nominal compositions, x = 0.26 (SBN26) and x = 0.80 (SBN80). Single crystal plates were cut perpendicularly to [001] (*c*-plate) with optically polished 5 mm × 5 mm surfaces and 1 mm thickness. Brillouin scattering spectra were

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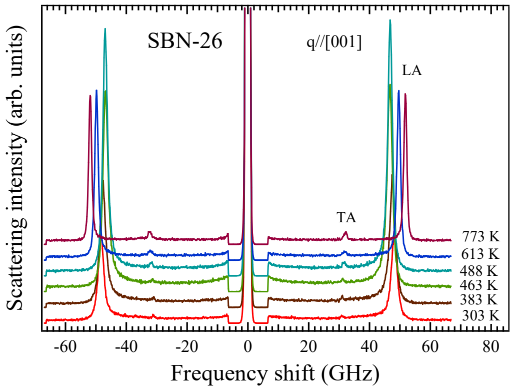

Fig. 3 Brillouin scattering spectra of the *c*-plate of a SBN26 crystal with very weak RFs.

measured at the back scattering geometry using a high-contrast 3+3 passes tandem Fabry-Perot interferometer with a free spectral range of 75 GHz for longitudinal acoustic (LA) and transverse acoustic (TA) modes and 300 GHz for the central peak [5]. The exciting source was a diode-pumped solid state (DPSS) laser with a wavelength of 532 nm. The specimen temperature was controlled by a cooling/heating stage (Linkam THMS600) with a stability of ± 0.1 °C.

3. Results and Discussion

The temperature dependence of Brillouin scattering spectra of *c*-plate of the SBN26 crystal was measured as shown in **Fig. 3**. The doublets of LA and TA modes were observed. Since the intensity of TA mode is very weak and the temperature variation is very small, we analyzed only the LA mode which propagates along the *c*-axis. The mode frequency, ν_{LA}, and the full width at half maximum, FWHM, of a LA mode are shown

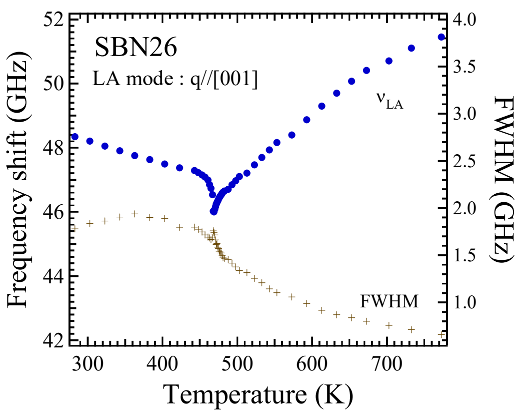

Fig. 4 Temperature dependence of the frequency of the LA mode along the *c*-axis of a SBN26 crystal.

as a function of temperature in **Fig.** 4. The LA mode frequency shows the remarkable decrease in the vicinity of T_C = 463 K. While the FWHM shows the gradual increase from the high temperature and the maximum appears at T_C. In the ferroelectric phase, a further increase is observed by the increase of scattering of LA modes at frozen PNRs.

These elastic anomalies of SBN26 with very weak RFs were compared with those of SBN80 with strong RFs. In SBN80, the temperature dependence of the LA mode frequency in the vicinity of T_C = 289 K, is diffusive and no remarkable change near T_C is observed. In comparison with a recent study of SBN40 [6], the elastic anomaly of SBN26 is more remarkable. These differences can be caused by the difference of the strength of RFs which suppress the remarkable changes in the vicinity of T_C.

4. Conclusion

The elastic anomalies of uniaxial relaxor ferroelectric SBN with strong RFs were well studied, while those of SBN with very weak RFs were not yet studied. The elastic anomaly of SBN26 (x = 0.26) with very weak RFs was studied by Brillouin scattering spectroscopy. The temperature dependences of mode frequency and FWHM of the LA modes along the ferroelectric *c*-axis show a remarkable change in the vicinity of T_C = 463 K. In contrast, those of SBN80 (x = 0.80) with strong RFs show only a diffusive broad anomaly in the vicinity of T_C = 289 K. This marked difference between the observed results of SBN26 and SBN80 can be attributed to the different number density of PNRs caused by RFs.

Acknowledgments

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