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Effect of electric field on uniaxial relaxor ferroelectric $Sr_xBa_{1-x}Nb_2O_6$ with intermediate random fields studied by Brillouin scattering

Md Aftabuzzaman[1,2‡] and Seiji Kojima[2]
[1]Dept. of Phys., Pabna Univ. of Sci. and Tech., Pabna, 6600, Bangladesh; [2]PAS, Univ. Tsukuba, Tsukuba, Ibaraki 305-8573, Japan

1. Introduction

Relaxor ferroelectrics (REFs) have been extensively studied during the last several decades owing to their outstanding piezoelectric and electromechanical properties [1], which make them very promising materials for industrial applications. Generally, REFs can be catagorised into cubic perovskite and uniaxial tetragonal tungsten bronze (TTB) type relaxors. Perovskite relaxors such as $Pb(Mg_{1/3}Nb_{2/3})O_3$, $Pb(Zn_{1/3}Nb_{2/3})O_3$ are Pb-based, and have been extensively investigated. While TTB relaxors such as $Sr_xBa_{1-x}Nb_2O_6$ (SBN), $Ca_xBa_{1-x}Nb_2O_6$ (CBN) are Pb-free, and uptodate the research on these REFs is inadequate, and the understanding of their insight mechanism is still unclear. Recently, owing to the emerging demand in green sustainable technology, the interest of the research on Pb-free materials has immensely been enhanced. Uniaxial REFs with the TTB structure such as SBN are very attractive materials in technological applications owing to their markedly high dielectric, piezoelectric, pyroelectric, and photorefractive properties [2-4]. Due to these excellent physical properties, SBN are the potential candidates for modern applications such as sensors, data storage, lasers, and holography. Therefore, the unique combination of excellent physical properties and Pb-free nature make SBN single crystals crucial materials for research. The structure of TTB relaxors is represented by $(A1)_2(A2)_4(C)_4$-$(B1)_2(B2)_8O_{30}$ which create the corner sharing distorted BO_6 octahedra. In case of SBN, the A1 sites are occupied only by Sr^{2+} ions and the A2 sites are occupied by both Ba^{2+} and Sr^{2+} ions, while the C sites and 1/6 of A1 and A2 sites remain unoccupied. These unoccupied A1 and A2 sites are thus the main source of quenched random fields (RFs) in the TTB relaxors. With the increase of a Sr content, the Curie temperature, T_C decreases. Consequently, the diffusive nature of the phase transition increases owing to the increase of the strength of RFs, which are believed to play a vital role in the relaxor nature. Vacancies at A1 and A2 sites are distributed ramdomly inside the crystal and create RFs which induce polar nanoregions (PNRs). PNRs have the dominant contribution to the precursor phenomena of the ferroelectric phase transition. Upon cooling from a high temperature, RFs induce dynamic PNRs at the Burns temperature, and these dynamic PNRs become frozen into a nonequilibrium nanodomain state upon cooling below T_C. This nonequilibrium nanodomain state show very sensitivity to an external electric field by which this state gradually switches into a metastable macro- or even a single domain state. However, the role of PNRs in the states above and below T_C is still a unsolved issue of materials science. In the present study, the effects of external electric field on the acoustic properties of SBN61 (x = 0.61) single crystals with intermediate RFs were investigated using the Brillouin scattering spectroscopy to explore the critical nature and the related functionality of PNRs in domain states of uniaxial REFs with the TTB structure.

2. Experimental procedure

The Czochralski method was employed to grow $Sr_xBa_{1-x}Nb_2O_6$ (x = 0.61, SBN61) single crystals. Single crystal plates were cut along [001] direction (c-plate) with optically polished 5 mm × 5 mm surfaces and 1 mm thickness. For the application of a dc electric field along [001] direction, silver paste electrodes were coated on larger surfaces of the crystal with a hole of 1 mm radius on one of the surfaces. Brillouin spectra were collected at the back scattering geometry by a high-contrast 3+3 passes tandem Fabry-Perot interferometer with a free spectral range of 75 GHz. A diode-pumped solid state (DPSS) laser with a wavelength of 532 nm was used as an exciting source. The sample temperature was controlled by a cooling/heating stage (Linkam THMS600) with a stability of ± 0.1 °C.

3. Results and discussion

Figure 1 shows Brillouin scattering spectra of a SBN61 (c-plate) single crystal at some selected temperatures measured at $c(a,a+b)\bar{c}$ scattering geometry on zero field heating (ZFH). These spectra consist of longitudinal acoustic (LA) and transverse acoustic (TA) phonon modes. Very weak TA peak persists in all temperature range which indicates that SBN61 single crystals have tetragonal

‡ azamanphy@gmail.com

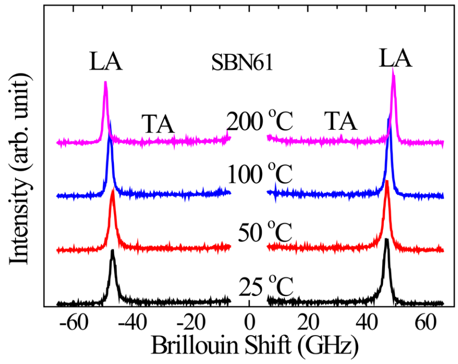

Fig. 1 Brillouin scattering spectra of the SBN61 single crystal at some selected temperatures on ZFH.

structure both in ferroelectric and paraelectric phases. Brillouin spectra were fitted using the Voigt functions, a convolution of Lorentzian and Gaussian functions at which the width of Gaussian function was fixed as an instrumental function, to obtain the Brillouin shift, ν_B, the full width at half maximum (FWHM), and the peak intensity of the phonon modes. The ν_B is linearly proportional to the longitudinal sound velocity, and the FWHM is related to the acoustic attenuation coefficient of a sound wave [5]. The acoustic properties were studied under zero and externally applied *dc* electric field along [001] direction.

In **Fig. 2**, a noticeable thermal hysteresis was observed below T_C (= 74 ºC on ZFH) in the LA shift between ZFH and zero field cooling (ZFC). It indicates that the SBN61 single crystal undergoes a diffused phase transition which is related to the incomplete alignment of nanodomains induced by quenched RFs. On field heating (FH) under 1.0 kV/cm, the LA shift starts to increase rapidly and a relatively sharp increase was observed around 62 ºC owing to the complete alignment of nanodomain to a field induced macrodomain state, and after the maximum a remarkable decrease of LA shift was observed towards T_C. By application of external electric field, the long-range ferroelectric order was enhanced and hence, T_C shifted towards the higher temperature (~85 ºC) at which a sharp minimum of LA shift was observed. On subsequent continuous field cooling (FC) and reheating processes, the observed minima of LA shift were same as that of previous FH process, while the anomaly around 62 ºC was absent. Consequently, a remarkable increase of the LA shift and a decrease of diffuseness were observed in the ferroelectric phase owing to the complete suppression of nanodomains by an external electric field during the previous FH process. Similar behavior was also observed in SBN crystals with weak and strong RFs where this acoustic anomaly was very sharp near T_C [6,7]. In addition, a broad and weak anomaly was also reported in the low temperature region of SBN with weak RFs owing to the incomplete alignment of nanodomains caused by the interaction among PNRs [6].

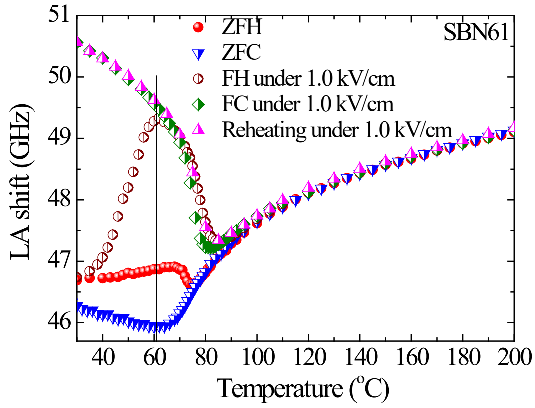

Fig. 2 Temperature dependences of the LA shift under zero and 1.0 kV/cm electric field on heating and cooling.

4. Conclusions

The effects of an electric field and the temperature dependence of acoustic properties of SBN61 single crystals with intermediate RFs were investigated using the broadband Brillouin scattering spectroscopy. A remarkable thermal hysteresis was observed below T_C in the LA shift between ZFH and ZFC, which attributes to the incomplete alignment of nanodomains induced by quenched RFs. The effect of an electric field along the [001] direction was clearly observed. Under the external electric field, a remarkable decrease of diffuseness and an enhancement of the long-range ferroelectric order were observed below T_C. On FH under 1.0 kV/cm, the complete alignment of a nanodomain to a macrodomain state around 62 °C was observed. On FC and reheating, the anomaly around 62 °C was absent indicating the complete alignment of nanodomains during the previous FH process.

Acknowledgments

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