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Electric field effect on polar nanoregions of uniaxial ferroelectric $Sr_xBa_{1-x}Nb_2O_6$ with weak random fields studied by Brillouin scattering

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1. Introduction

Relaxor ferroelectrics (REFs) are very attractive materials for industrial applications because of their excellent piezoelectric and electromechanical properties [1], and have been extensively studied during the last several decades. REFs can be classified into cubic perovskite and uniaxial tetragonal tungsten bronze (TTB) type relaxors. Most of the 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 intensively been explored. While most of the TTB relaxors such as $Sr_xBa_{1-x}Nb_2O_6$ (SBN), $Ca_xBa_{1-x}Nb_2O_6$ (CBN) are Pb-free, and till now the study of these REFs is insufficient and the understanding of their insight mechanism is still unclear. Nowadays, the immense interest of the research on Pb-free materials has been grown due to their emerging demand in green sustainable technology. Uniaxial REFs with TTB structure such as SBN are technologically important materials owing to their remarkably high dielectric, piezoelectric, pyroelectric, and photorefractive properties [2-4]. These excellent physical properties make them potential candidates for modern applications such as sensors, data storage, lasers, and holography. The unique combination of physical properties and lead free nature make SBN single crystals crucial materials for research. The structural formula of TTB relaxors is expressed by $(A1)_2(A2)_4(C)_4$-$(B1)_2(B2)_8O_{30}$ with corner sharing distorted BO_6 octahedra. In 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(A1+A2) sites are unoccupied and are thus the main source of quenched random fields (RFs). By increasing the Sr content, the Curie temperature, T_C decreases. Consequently the diffusive nature increases due to the increase of the strength of the RFs, which are believed to play a dominant role in the relaxor nature. Ramdomly distributed vacancies at A1 and A2 sites create RFs and induce polar nanoregions (PNRs) which have the dominant contribution to the precursor phenomena of the ferroelectric phase transition. Dynamic PNRs induced by RFs appear at the Burns temperature, which freeze into a nonequilibrium nanodomain state upon cooling below T_C. This nanodomain state is very sensitive to an external electric field by which it gradually switches into a metastable macro- or even single domain state. However, the role of PNRs in the states above and below T_C is still a puzzling issue of materials science. In the present study, the effects of external electric field on the elastic properties of SBN40 (x = 0.40) single crystals were investigated using the Brillouin scattering spectroscopy to clarify the critical nature and the related functionality of PNRs and domain state in the uniaxial REFs with TTB structure.

2. Experimental procedure

$Sr_xBa_{1-x}Nb_2O_6$ (x = 0.40, SBN40) single crystals were grown by the Czochralski method. Single crystal plates were cut along [001] (c-plate) with optically polished 5 mm × 5 mm surfaces and 1 mm thickness. Silver electrodes were coated on the larger surfaces of the crystal with a hole of 1 mm radius on one of the surfaces for the application of a dc electric field along the [001] direction. Brillouin spectra were 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. 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 Brillouin scattering spectra of the SBN40 (c-plate) single crystal were measured at $c(a,a+b)\bar{c}$ scattering geometry. In order to obtain the Brillouin shift, ν_B, the full width at half maximum (FWHM), and the peak intensity of the phonon modes, the measured 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. The

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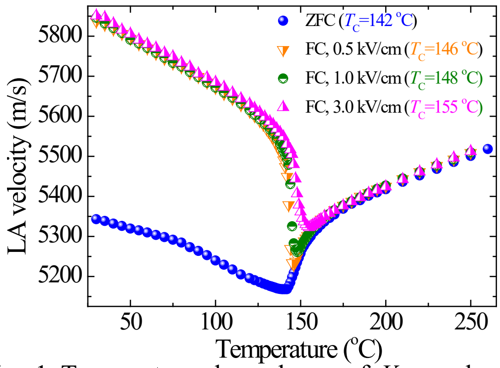

Fig. 1 Temperature dependence of V_{LA} under constant electric field along the [001] direction on cooling.

LA velocity (V_{LA}) was determined from the ν_B using the equation $V_{LA} = \lambda \nu_B/2n$, where λ is the wavelength of the incident laser light (532 nm at the present case) and n is the ordinary refractive index of the sample at λ. Figure 1 shows the temperature dependence of V_{LA} on zero field cooling (ZFC) and field cooling (FC) processes. Upon FC, the external electric field suppresses quenched RFs and aligns PNRs along the field direction. As a result, a remarkable increase of V_{LA} was observed below T_C. The FC states of high V_{LA} below T_C actually attributes to the field induced macro/single domain state. By increasing the strength of external electric field, T_C shifts towards the high temperature region. It signifies the enhancement of the long range order in the ferroelectric phase. After poling the crystal

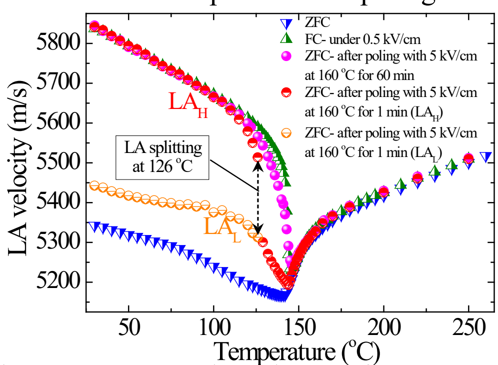

Fig. 2 Temperature dependences of V_{LA} on cooling after poled the crystal with 5 kV/cm at 160 ºC for 60 and 1 min.

with 5 kV/cm for 60 min at 160 ºC (i.e. below the intermediate temperature, T^* =185 ºC), the V_{LA} during ZFC was comparable to that under FC with 0.5 kV/cm (Fig. 2). Poling the crystal with such a high electric field for 60 min is sufficient to align all static PNRs along the field direction. Therefore, V_{LA} becomes similar to that under FC. By decreasing the poling time to 1 min, some of the PNRs switch back to their initial direction, and therefore a splitting of the LA mode was observed below T_C (Fig. 2). The high-frequency LA mode (LA_H) corresponds to macrodomains induced by the external electric field, while the low-frequency LA mode (LA_L) corresponds to nanodomains induced by RFs. Below T_C, upon application of the electric field at constant

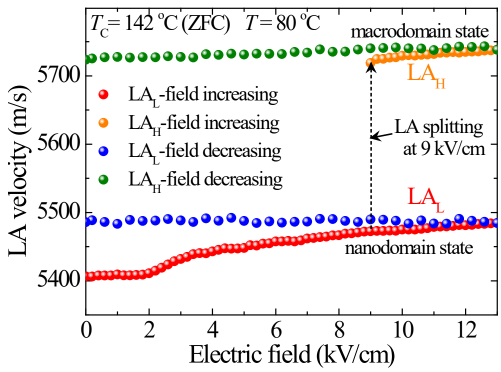

Fig. 3 Electric field dependence of V_{LA} at constant temperature.

temperature (Fig. 3), a coexisting state of nano- and macrodomains was also observed at about 9.0 kV/cm. This mixed state persists up to 13 kV/cm due to the incomplete switching of the nanodomains into the macro- or single domain state. Below T_C, PNRs are strongly correlated with each other, hence the switching of nanodomains by the external electric field becomes interrupted. The mixed state exists even after removing the external electric field. This strong memory effect of electric field can be removed by short circuiting the electrodes above T^* for more than 15 min.

4. Conclusions

The effect of electric field along the [001] direction was clearly observed. On FC, the alignment of PNRs and the enhancement of the long-range ferroelectric order were observed below T_C. After poling the crystal with high electric field for 60 min above T_C, the alignment of all static PNRs along the field direction was observed. By decreasing the poling time to 1 min, back switching of PNRs was also observed, which results the LA mode splitting below T_C. In the field dependent measurement below T_C, a mixed state consisting of macro- and nanodomains was observed. This mixed state persists up to a very high electric field because of the incomplete switching of nanodomains due to the correlation among PNRs.

Acknowledgments

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