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Electric field effect of relaxor ferroelectric (1-*x*)Pb($Mg_{1/3}Nb_{2/3}$)O_3-*x*$PbTiO_3$ crystals near MPB composition probed by Brillouin scattering

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1. Introduction

Lead based relaxor ferroelectric single crystals having the ABO_3-type perovskite structure such as (1-*x*)Pb($Mg_{1/3}Nb_{2/3}$)O_3-*x*$PbTiO_3$ (PMN-*x*PT) with the morphotropic phase boundary (MPB) have been studied last several decades due to its excellent piezoelectric and electromechanical response [1,2]. The PMN-PT materials demonstrate a great variety of physical properties and, hence, is also very interesting from the technological point of view. The dynamics of polar nanoregions (PNRs) which appears at a certain characteristic temperature so called Burns temperature, T_B, has been thought to play an essential role in the occurrence of high dielectric constant and piezoelectric coefficient. Consequently, many experimental and theoretical efforts have been paid to clarify the role of PNRs in relaxor ferroelectrics. However, there is still a long way to understand the dynamical behavior of PNRs. The different phases near the MPB composition have very close energy states. Therefore, different phases may coexist in a crystal, and can switch from one phase to another via polarization switching/rotations by the application of a dc electric field. As a result many investigations have been conducted on the field induced changes in PMN-PT single crystals near the MPB compositions. Very recently, intermediate monoclinic and orthorhombic phases at MPB between rhombohedral and tetragonal phases of PMN-PT have been revealed and their influence on the acoustic phonon is currently under intensive investigations [3,4]. Due to the coupling between the polarization fluctuaions and strain, acoustic properties shows anomalous behaviors during phase transions. Brillouin scattering spectroscopy is a very powerful tool to investigate the acoustic phonon behaviors in the gigahertz frequency range. Therefore, in this study, we investigated the PMN-30PT (*x*= 0.30) single crystals under the zero filed and the externally applied dc electric field by using broadband Brillouin scattering spectroscopy over a wide temperature range of 30-500 ºC during both the cooling and heating processes. Electric field dependent measurements at the constant temperature 31 ºC also have been carried out.

2. Experimental procedure

The (1-*x*)Pb($Mg_{1/3}Nb_{2/3}$)O_3-*x*$PbTiO_3$ (*x* = 0.3, PMN-30PT) single crystal with a (001)-oriented plate of 7x7 mm^2 size and 0.5 mm thickness was used for measurements. Gold plate electrodes were coated on larger faces for the application of dc electric field along the [001] direction. Brillouin spectra were measured in a backward scattering geometry using a tandem Fabry-Perot interferometer (JRS TFP-1) in combination with a reflection optical microscope (Olympus BX-60) and a single frequency green Yttrium aluminium garnet (YAG) laser (Coherent Compass 315M-100) with a wavelength of 532 nm [5]. For the temperature variation, the sample was placed inside a heating/cooling stage (Linkam HTMS600) in the temperature range from room temperature up to 600 ºC with a free spectral range of 75 GHz.

3. Results and discussion

Figure 1 shows the typical Brillouin scattering spectra of the PMN-30PT single crystal at selected temperatures at VO scattering geometry upon zero filed heating (ZFH). These spectra consist of a longitudinal acoustic (LA) phonon at ***k*** ~ 0, and a central peak (CP). The Brillouin components and the CP were fitted using Voigt functions, where a width of a Gaussian function was fixed as an instrumental function. We obtained the Brillouin shift (ν_B) and the full width at half maximum (FWHM denoted as Γ_B) as a function of temperature as shown in **Fig. 2.** The ν_B is linearly proportional to the longitudinal sound velocity, and

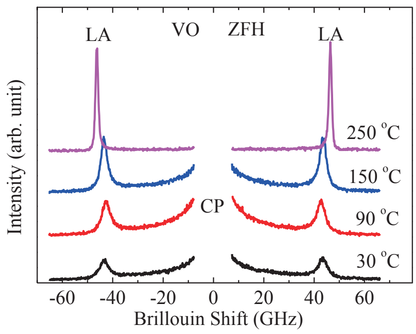

Fig. 1 Brillouin scattering spectra of the PMN-30PT single crystal at selected temperatures under ZFH.

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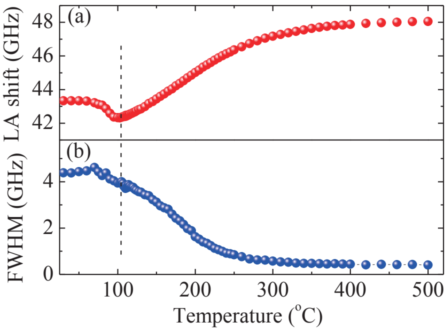

Fig. 2 Temperature dependences of (a) the Brillouin shift and (b) FWHM on ZFH.

the Γ_B is related to the acoustic attenuation coefficient of a sound wave [6]. A noticeable thermal hysteresis was observed in ν_B between ZFH and zero field cooling (ZFC) processes. The ν_B in ZFH exhibits a much sharper minimum at ~103 °C. Similar acoustic hysteresis have been observed from other relaxor ferroelectrics [6,7,8].

Figure 3 shows the temperature dependence of the Brillouin scattering spectra under 0.5 kV/cm dc electric field along the [001] direction on cooling. The interesting feature on cooling process at constant filed is that the LA mode exhibits splitting below a specific temperature at ~78 °C and that an additional weak transverse acoustic (TA) mode appears near 29 GHz. It clearly indicates that the PMN-30PT undergoes a ferroelectric phase transition at ~78 °C under the field of 0.5 kV/cm. If the field increases up to 1 kV/cm (not shown), the LA mode splitting disappears. The splitting of LA mode indicates that the crystal consists of two states, one is the ferroelectric macrodomain with the high-frequency LA mode (HLA) and the other is the nanodomain state caused by the random field with low-frequency LA mode. At higher field the crystal possesses single domain with HLA [6].

The electric field dependence of the Brillouin scattering spectra of an unpoled c-plate crystal just after ZFC was studied at 31 °C. The LA velocity was determined from the ν_B as shown in **Fig. 4.** In the ferroelectric phase, upon the application of an electric field to the ZFC crystal along the ferroelectric c-axis, a discontinuous transition of the LA velocity from a nonequilibrium state (A) of nanodomains induced by the random field to an intermediate state (B) with coexisting nano- and macrodomains was observed at 1.05 kV/cm. With a further increase in the electric field, the state (B) changes into the equilibrium state (C) at 2.6 kV/cm, which mainly comprises a single domain due to the complete switching of the nonequilibrium nanodomains to the equilibrium macrodomains or single-domain states.

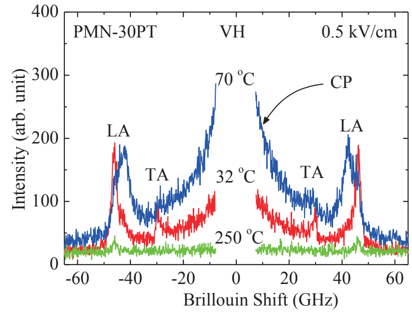

Fig. 3 Brillouin spectra measured under dc electric field of 0.5 kV/cm along [001] direction on FC.

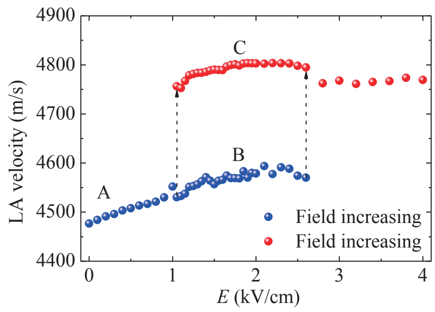

Fig. 4 Electric field dependence of LA velocity at 31 °C.

4. Conclusions

The significant thermal hysteresis in LA shift was observed between ZFH and ZFC reflecting the relaxor nature. The effect of the electric field applied along the [001] direction was observed. Under the moderate electric field, LA mode splitting was observed which indicates a mixed state consists of nanodomains and macrodomains. Under the sufficiently high electric filed, the nanodomain state changes into a single domain state.

Acknowledgments

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