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Acoustic properties PIN-PMN-PT relaxor ferroelectric single crystal studied by micro-Brillouin spectroscopy

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1. Introduction

Relaxor ferroelectics have been receiving great attention due to their outstanding electromechanical properties and wide application fields. (1-x)Pb($Mg_{1/3}Nb_{2/3}$)O_3- $x$$PbTiO_3$ (PMN-xPT) is one of typical relaxors. However, there are some limitations of PMN-xPT because this relaxor ferroelectrics suffer a relatively low rhomohedral-tetragonal transition temperature T_{RT} (lower than 100 °C) [1]. The ternary compound xPb($In_{1/2}Nb_{1/2}$)O_3-(1-x-y)Pb($Mg_{1/3}Nb_{2/3}$)O_3-$y$$PbTiO_3$ (PIN-PMN-PT) has not only higher T_{RT} but also higher coercive field (~15 kV/cm)[2], which may be very attractive from the viewpoint of application. However, many physical properties have not yet been investigated in detail for this ternary compound.

In this work, we investigated dynamical behaviors of a [001]-oriented PIN-PMN-PT (x=0.26, y=0.28) single crystal by micro-Brillouin light scattering. The acoustic properties of this system have been examined in a wide temperature range between 77 and 873 K on both cooling and heating precesses.

2. Experimental

The [001] oriented plates of PIN-PMN-PT crystal with the size 5 x 1 x 1 mm^3 grown using the modified Bridgeman technique are provide by TRS technologies, Inc. The Brillouin spectrum of PIN-PMN-PT was measured by using a micro-Brillouin scattering system consisting of a 6-pass tandem Fabry-Perot interferometer (FPI) combined by a microscope. All spectra were obtained at a backward scattering. The sample was put inside a cryostat cell placed on the stage of an optical microscope. By using two free spectral ranges, we could investigate a longitudinal acoustic (LA) mode in a smaller spectral range of ± 69 GHz and a quasielastic central peak (CP) in a wider spectral range of ± 563 GHz.

3. Results and Discussion

The temperature dependence of the Brillouin spectra was measured from 873 to 77 K.

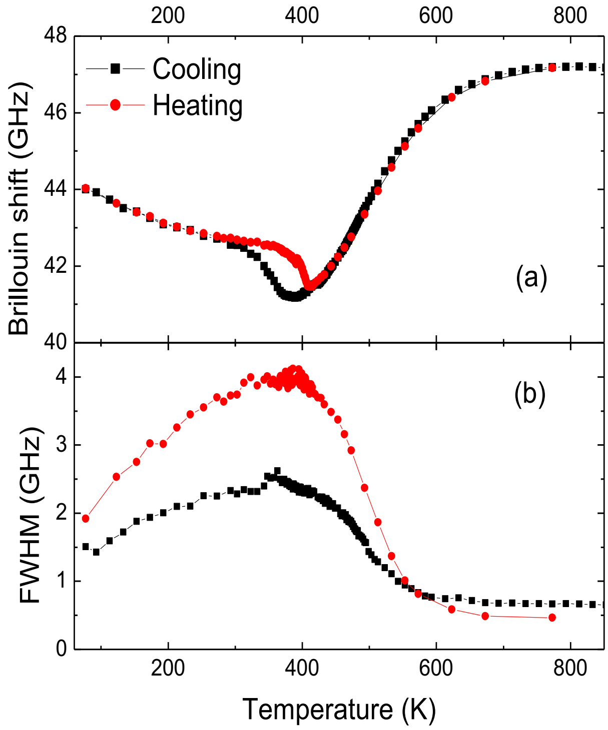

Fig. 1. Brillouin shift (a) and the FWHM (b) of PIN-PMN-PT measured upon cooling and heating processes.

The Brillouin doublet was fitted by using a response function of a damped harmonic oscillator, from which the Brillouin frequency shift (ν_B) and the full width at half maximum (FWHM) were obtained.

The dielectric constant of this PIN-PMN-PT shows a typical relaxor behavior with a substantial frequency dispersion. The ν_B and the FWHM of PIN-PMN-PT measured at both cooling and heating process are plotted in **Fig. 1** as a function of temperature. The ν_B of PIN-PMN-PT, which corresponds to the elastic stiffness coefficient c_{11}, measured on cooling exhibits almost a constant value at high temperatures, a mild softening upon cooling and a broad minimum near ~393 K. This behavior has also been observed in other typical relaxor ferroelectric crystals. The Brillouin shift and the related sound velocity normally increase upon cooling due to usual lattice anharmonicity. In case of relaxors, the formation of nano-sized polar

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clusters or polar nanoregions (PNRs) at the Burns temperature induces electrostrictive coupling between the square of the local polarization and the strain caused by the acoustic waves. This coupling is responsible for the deviation of the elastic behavior from the normal lattice anharmonicity.

The ν_B and the FWHM show different behaviors between the cooling and heating processes near the dielectric maximum temperature. The observed thermal hysteresis is analogous to PZN-4.5%PT[3] but is more pronounced. The minimum of ν_B becomes sharper on heating compared to that on cooling. Except near the dielectric maximum temperature, the ν_B does not exhibit any difference between the two processes. In addition, the FWHM exhibit a very large difference between the cooling and the heating processes. The maximum of FWHM measured on heating is almost twice compared to that measured on cooling. This kind of substantial hysteresis in FWHM has not been observed in typical relaxors and reflects differences in the kinetics of the diffuse phase transition.

This phenomenon may be explained by two-stage model based on the intrinsic compositional inhomogeneities[4]. According to this model, PNRs appear at the Burns temperature on the first stage and their size and the number increase upon cooling toward the phase transition temperature. The kinetics of the phase transition is influenced by the number of PNRs at the onset of the ferroelectric fluctuations at the second stage which are dependent on the phase transition temperature. The PNRs cannot be collapsed easily due to, for example, the local quenched random fields, and the number of PNRs near the phase transition temperature is larger during cooling process. Therefore, the kinetics is expected to become slower at lower temperatures, which may be responsible for the broader, more sluggish aspect of ν_B measured on cooling. On the other hand, the phase transition becomes sharper upon heating due to the fewer number of polar clusters that would allow the ferroelectric nuclei to grow faster. This larger polar clusters and enhanced ferroelectric fluctuations are mainly responsible for the much larger hypersonic damping reflected in the temperature dependence of FWHM.

The temperature evolution of dynamics of PNRs is correlated with anomalous changes in both LA mode and the quasielastic CP in relaxor ferroelectrics[5]. The polarization fluctuations induced by the flipping motions of PNRs are directly related to the formation of CP. The half width of CP is inversely proportional to the relaxation time of the PNRs dynamics. The CP appears at around 500 K and its intensity becomes stronger upon cooling reflecting the increasing size and density of PNRs. The spectral feature of quasielastic CP was fitted by using a response function of a single Debye relaxator. The half width of CP decreased upon cooling in the ergodic relaxor phase. This is correlated with the slowing down of the PNRs dynamics since their size is expected to increase and their flipping motion will be slower upon cooling. The slowing down of PNRs dynamics is also responsible for the enhanced coupling between the order parameter and the acoustic waves resulting in the substantial softening of the mode frequency as well as the remarkable increase in the hypersonic damping.

4. Conclusions

High-resolution micro-Brillouin scattering technique was used to investigate the acoustic anomaly and CP in a [001]-oriented PIN-PMN-PT single crystal. The LA mode exhibited a substantial softening that is correlated to the temperature evolution of PNR. A significant thermal hysteresis was observed the ν_B and the FWHM between the cooling and the heating processes, which was attributed to the different kinetics that is controlled by the number of PNRs. The softening of the LA mode was accompanied by the growth of CP. The half width of CP decreased upon cooling toward the dielectric maximum temperature, which reflected the slowing down behavior of PNRs.

Acknowledgments

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