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Dielectric relaxation in the $Pb(Yb_{1/2}Nb_{1/2})O_3-PbTiO_3$ solid solution single crystal near the morphotropic phase boundary

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The dielectric properties in $0.47Pb(Yb_{1/2}Nb_{1/2})O_3-0.53PbTiO_3$ (0.47PYN–0.53PT) single crystals near the morphotropic phase boundary (MPB) were investigated in the frequency range from 10 kHz to 1 MHz. Remarkable dielectric relaxation was observed along the polar ⟨001⟩ direction in the tetragonal 0.47PYN–0.53PT single crystal near the MPB. An increase of the dielectric relaxation time was observed as the Curie temperature was approached. The real part and the imaginary part of the complex relative permittivity obeys the Cole–Cole arc law, and the Debye type dielectric dispersion with the polydispersive type among the order-disorder type ferroelectrics was observed.
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Recently much attention has been focused on relaxor-based ferroelectric solid solution single crystals such as $Pb(Zn_{1/3}Nb_{2/3})O_3-PbTiO_3$ (PZN–PT), $Pb(Mg_{1/3}Nb_{2/3})O_3-PbTiO_3$ (PMN–PT), and $Pb(Yb_{1/2}Nb_{1/2})O_3-PbTiO_3$ (PYN–PT) near morphotropic phase boundary (MPB) for applications of ultrasonic, nondestructive testing, and actuator devices.[1–7] Furthermore, dielectric relaxation in relaxor ferroelectrics has also been attracted much attention.[8–12] The dielectric relaxation behavior for relaxor materials such as PMN–PT and PZN–PT was explained in terms of the Vogel–Fulcher law.[8–12] In particular, the dielectric dispersion characteristic in 0.55PYN–0.45PT was also analyzed with the Vogel–Fulcher relation.[5] On the other hand, the study of the complex impedance spectroscopy in 0.68PMN–0.32PT was reported for Debye-like behavior and Cole–Cole type relation from the measurement of ac-conductivity at high temperature range from 200 to 525 °C.[13] In this work, the dielectric dispersion was investigated in the 0.47PYN–0.53PT near MPB. The experimental results was analyzed with Cole–Cole arc law,[14] and the Debye type dielectric relaxation with the polydispersive type for the $0.47Pb(Yb_{1/2}Nb_{1/2})O_3-0.53PbTiO_3$ single crystal was reported in this letter.

Single crystals of the 0.47PYN–0.53PT solid solution near MPB were prepared by a conventional flux method using $PbO-PbF_2-B_2O_3$ flux.[7] Crystal planes were mirror-polished with 1.6 μm Al_2O_3 powder, and silver paste (Dupont No. 7713) was painted on the crystal surfaces and fired at 400 °C for 5 min. The crystal plane used was confirmed to be the (001) plane in the tetragonal phase by the x-ray diffraction technique.[7] The Ti concentration x was estimated to be 0.53 by the inductive charge plasma analysis using reference ceramic samples.[7,15,16] The value of T_C in the 0.47PYN–0.53PT was 404 °C. This value of T_C was slightly higher than that of T_C in single crystals prepared by using the flux method with the addition of barium to enhance the perovskite phase.[17,18] This difference in T_C may be due to the influence on T_C by barium as an impurity.[17,18] The electrical capacitance and dielectric loss tangent tan δ were measured using an LCR meter (HP-4274A) at 10, 40, 100, 400 kHz, and 1 MHz with the probe field weaker than 10 V/cm with heating at a rate of 1 K/min.

The temperature dependence of the real part ϵ_r' of the complex relative permittivity and the dielectric loss tangent tan δ along the polar ⟨001⟩ direction in the tetragonal

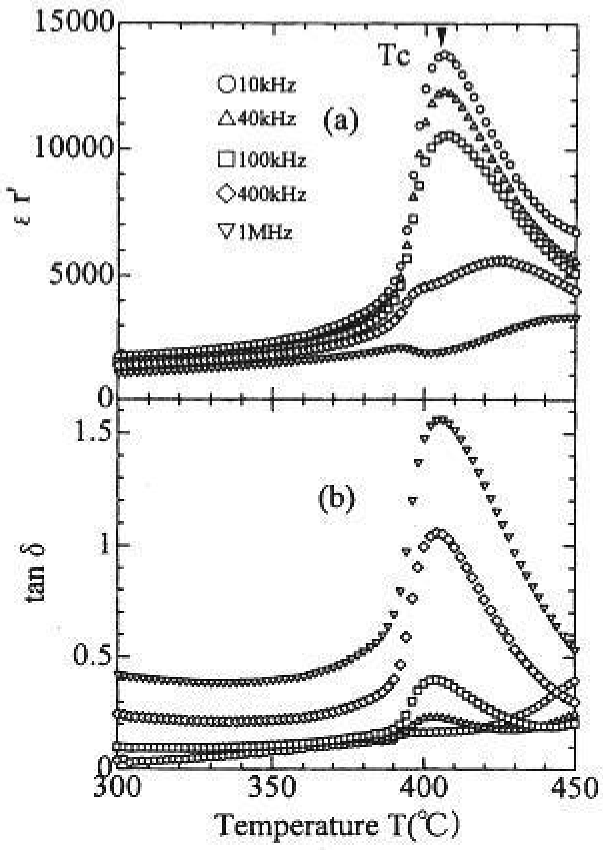

FIG. 1. Temperature dependence of (a) the real part ϵ_r' and (b) the dielectric loss tangent tan δ along the polar ⟨001⟩ axis in the tetragonal 0.47PYN–0.53PT single crystal with the frequency as a parameter.

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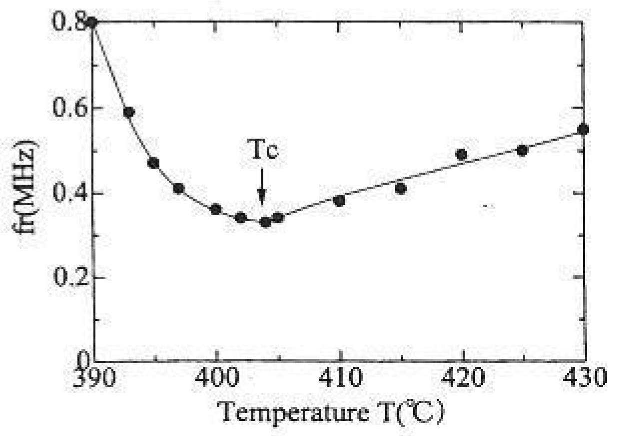

FIG. 2. Temperature dependence of the relaxation frequency f_r.

0.47PYN–0.53PT single crystal are shown in Figs. 1(a) and 1(b), respectively, with the frequency as a parameter. The peak of ϵ_r' appearing at the transition temperature T_C at the lower frequency decreases rapidly with increasing frequency and then gives way to a broad hump at higher frequency at higher temperature. On the other hand, in Fig. 1(b), with increasing frequency the value of tan δ increases at first and successively decreases near the transition temperature T_C. Figure 2 shows the temperature dependence of the dielectric relaxation frequency (the inverse of the relaxation time) f_r which gives the maximum value of the imaginary part ϵ_r'' of the complex relative permittivity in the frequency characteristics of ϵ_r'' replotted from Fig. 1 at the given temperature. The value of f_r decreases monotonically with the change in temperature towards the transition temperature T_C in the paraelectric phase, while decreases rapidly more than in the paraelectric phase, towards T_C in the ferroelectric phase. Thus, as the temperature approaches the transition temperature T_C, the dielectric relaxation time becomes longer.[19] Cole–Cole diagrams in the paraelectric and ferroelectric phase with the temperature as a parameter obtained by a replotting from Figs. 1(a) and 1(b), are drawn as shown in Figs. 3(a) and 3(b), respectively. Data points (ϵ_r', ϵ_r'') at the given temperature in the paraelectric and in the ferroelectric phase in the Cole–Cole diagram lie nearly on circular arc, and the centers of all arcs lie on the straight line below the real axis. Thus, the frequency dependence of ϵ_r' and ϵ_r'' obey the Cole–Cole arc law.[14,19] Two intersections (ϵ_{r0}' and $\epsilon_{r\infty}'$) between the real axis and a circular arc in Fig. 3 give the relative permittivity at zero frequency ϵ_{r0}' which depends on temperature and the relative permittivity at the infinitive frequency $\epsilon_{r\infty}'$ which is essentially independent on temperature and has a value of about 500 in the paraelectric phase and about 450 in the ferroelectric phase, respectively. The parameter β[14,19] estimated by the relation $\beta=1-2\alpha/\pi$ where α is the angle between the real axis and the center of the circular arc (see Fig. 3), representing a measure of width of the distribution of the dielectric relaxation times, is about 0.88 in the paraelectric phase and about 0.84 in the ferroelectric phase, respectively. These indicate the Debye type dielectric dispersion of the polydispersive type. An increase of the dielectric relaxation time was observed, as the Curie temperature was approached. The remarkable dielectric relaxation observed along the polar ⟨001⟩ direction in the tetragonal 0.47PYN–0.53PT single crystal near the MPB is of the Debye type among the dispersive characteristics of the relaxor-PT related materials such PMN–PT and PZN–PT.[5,8–12] A more detailed study analyzed according to the theory for the dielectric dispersion[14,19,20] is now in progress.

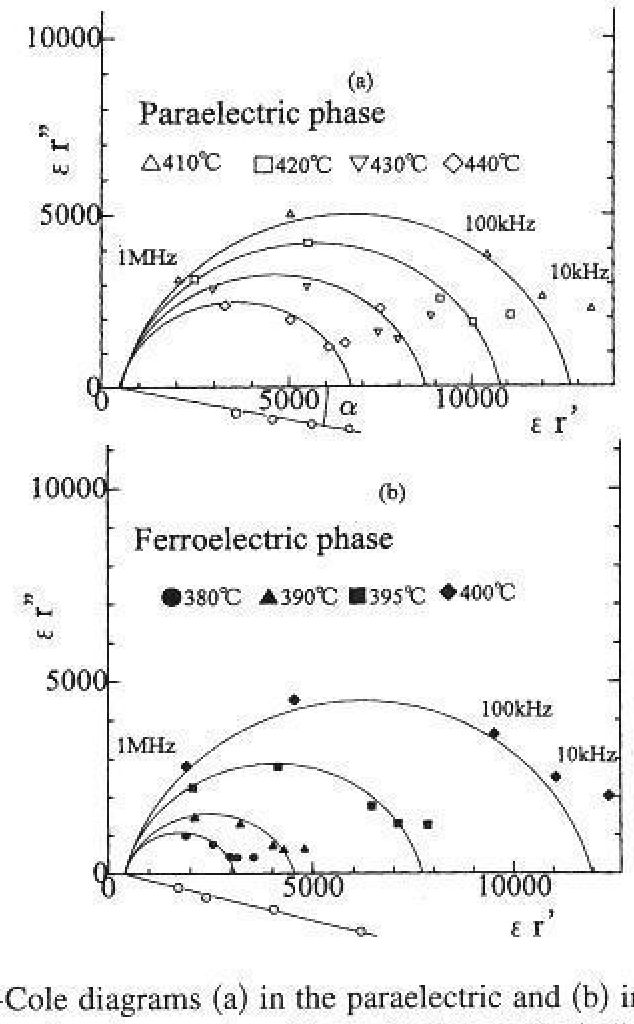

FIG. 3. Cole–Cole diagrams (a) in the paraelectric and (b) in the ferroelectric phase at the given temperature. Open circular marks indicate the centers of the circular arcs.

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