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Importance of structural irregularity on dielectric loss in $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3-(x)PbTiO_3$ crystals

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The dielectric properties of $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3-(x)PbTiO_3$ (PMN–PT) crystals have been investigated over a temperature range of 4 to 450 K at various frequencies. At low temperatures, an unusual frequency dependent plateau region in the absorption was observed between 75 and 175 K. At both higher and lower temperatures, the absorption was frequency independent. Analysis of the relaxation time constant revealed power-law divergence, typical of fractal behavior in disordered magnetic systems. The results demonstrate the importance of structural irregularities on the dielectric loss mechanism in poled oriented PMN–PT crystals. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1482791]

⟨001⟩-oriented $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3-(x)PbTiO_3$ (PMN–PT) and $(1-x)Pb(Zn_{1/3}Nb_{2/3})O_3-(x)PbTiO_3$ (PZN–PT) single crystals are currently under investigation due to high electromechanical properties in the poled condition.[1,2] Investigations have focused on compositions close to the morphotropic phase boundary (MPB). Dielectric investigations of poled and thermally annealed conditions have shown significant differences.[3] Annealed specimens have relaxor ferroelectric characteristics with a single diffuse peak, whereas poled specimens have sharper ferroelectric transformations and two anomalies.

Relaxor ferroelectrics have glasslike features, similar to dipolar and spin glasses.[4,5] A Vogel–Fulcher-type freezing of polarization fluctuations was observed by analysis of dielectric relaxation. Long-range order does not appear on cooling in the absence of an applied electric field.[4–6] However in the field-cooled condition, long-range polar order is locked in at temperatures below that of polarization freezing (T_f).[4] Compositions close to the MPB exhibit sharp ferroelectric phase transformations in the field-cooled condition.[3] However, reciprocal phase space mapping investigations have demonstrated significant mosaicity.[7] Structural nonuniformity within a poled condition on the nanometer scale can be caused by microheterogeneity. Recent investigations of polarization dynamics in MPB compositions of PMN–PT have shown that switching occurs by heterogeneous nucleation in the vicinity of random-fields around microheterogeneity.[8,9]

Clearly, microheterogeneity is important in oriented PMN–PT and PZN–PT crystals. Structural irregularities exist within the poled condition. Previous investigations of loss mechanisms in ferroelectric crystalline solutions have focused on domain-wall contributions, and ignored the possibility of significant contributions from excitation of irregularities within domains. In this letter, we will present evidence that a significant portion of the dielectric loss is due to structural irregularity, in PMN–PT.

Large PMN–PT single crystals of MPB composition (68/32) were synthesized by using a conventional flux method and a suitable ratio of the PMN–PT: PbO flux. ⟨001⟩-oriented seed crystals were used to initiate crystal growth. The large single crystal was diced into slices oriented along ⟨001⟩ direction, with dimensions of $2.6\times2.5\times1$ mm^3. The samples were electroded by gold sputtering. Dielectric measurements were performed from 120 Hz to 100 kHz using a Keithley 3330 LCZ Meter. Great attention was paid to the accuracy of the dielectric loss which can be affected by capacitance value, measurement frequency and voltage level, and cable lengths. A four-terminal pair configuration with twisted coaxial cables extending all the way to the sample was used. The current and voltage contacts were geometrically arranged such that the effect of contact resistance was minimized.

Figures 1(a)–1(c) show the dielectric response as a function of the temperature for a ⟨001⟩-oriented PMN–PT crystal in the as-grown condition. Data shown in each figure for the four frequencies were 1.2×10^2, 10^3, 10^4 and 10^5 Hz. Figure 1(a) shows the real component taken over the temperature range of 125 to 450 K. This data is identical to that previously reported for this composition in the poled condition,[3] demonstrating that the as-grown condition has a significant degree of poling. Figure 1(b) shows the real component taken over the temperature range of 4 to 300 K. Some dielectric relaxation is evident in Fig. 1(b) due to the smaller scale of the y axis. However, on cooling, the response became nondispersive below ~50 K. Figure 1(c) shows the dielectric loss factor taken over the temperature range of 4 to

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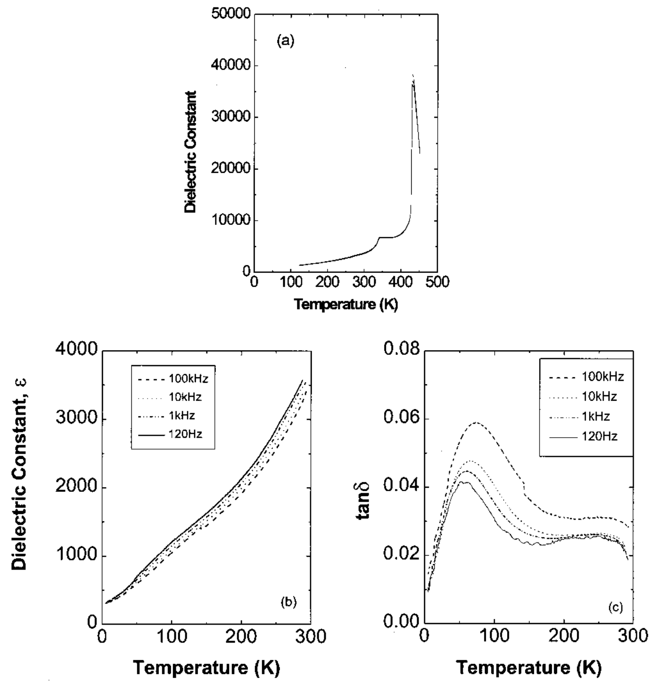

FIG. 1. Complex dielectric response as a function of temperature at various measurement frequencies for a ⟨001⟩-oriented PMN–PT crystal in the as-grown condition. (a) the real component taken over the temperature range of 125 to 450 K, (b) the real component taken over the temperature range of 4 to 300 K, and (c) the dielectric loss factor taken over the temperature range of 4 to 300 K.

300 K. A strong peak in tan δ was observed near 75 K, which was frequency dependent. With increasing frequency, the maximum in tan δ was shifted to higher temperatures. At lower temperatures, tan δ was frequency independent, similar to that observed in the real component. At higher temperatures, frequency dispersion persisted until near the temperature of the maximum in the dielectric constant.

The imaginary component of the dielectric constant (K''), known as the dielectric absorption, was calculated from the product of the dielectric constant and the loss factor. Figure 2 shows K'' as a function of temperature between 4 and 300 K. At low temperatures, an unusual frequency dependent plateau region in the absorption was observed between 75 and 175 K. At both higher and lower temperatures, the absorption was frequency independent and strongly temperature dependent. Accordingly, non-Krammers–Kroning-

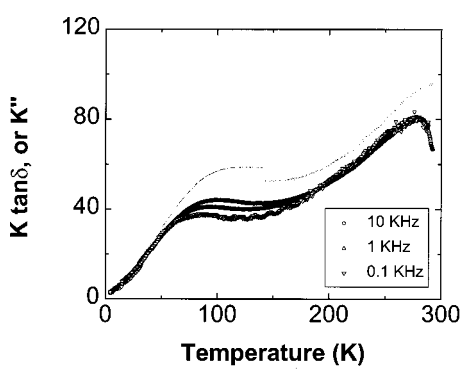

FIG. 2. Imaginary component of the dielectric response (or the dielectric absorption) as a function of temperature for a ⟨001⟩-oriented PMN–PT single crystal in the as-grown condition.

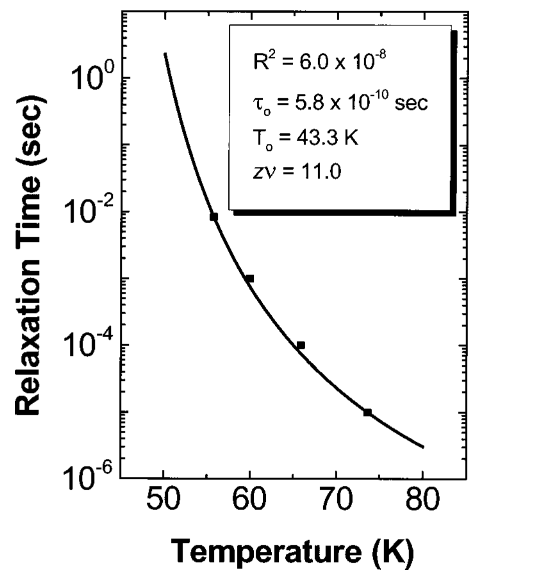

FIG. 3. Power-law fitting of relaxation time constant as a function of temperature for PMN–PT single crystal.

type behavior is observed only in the temperature range of the dielectric plateau.

The results in Fig. 2 can be explained using the fractal cluster model commonly used in disordered magnetic systems.[10,11] In this model, the cluster size s_ξ is related to the characteristic correlation length ξ and to the clusters fractal dimensionality D by the relationship $s_\xi \propto \xi^D$.[12] Dynamic scaling then relates the critical relaxation time τ to the ξ as $\tau \sim \xi^z$. Since ξ diverges with temperature as $\xi \sim [T/T - T_0]^v$, a power-law divergence for the relaxation time is obtained, as given in Eq. (1);

$$\tau = \tau_0 \left(\frac{T}{T - T_0} \right)^{zv}, \qquad (1)$$

where the exponent zv is called the dynamical exponent, T_0 is the freezing temperature determined by the maximum in the absorption. The characteristic relaxation time τ is related to s_ξ by the relationship $\tau = \tau_0 s_\xi^x$, where $x = z/D$. The value of the dynamical coefficient zv lies between 4 and 12.

Svedlindh *et al.*[13] suggested that the average relaxation time can be obtained from the inflection points of dielectric absorption as a function of temperature. This simple result is valid for the limit of low field and allows a straightforward determination of the critical exponents. Continentino and Malozemoff[14] extended this analysis and showed that for the case where zv is large that the inflection point of χ'' corresponds to the condition $\omega\tau = 1$. Accordingly, the relaxation time at any temperature (τ, T) can be approximated as $(1/\omega, T_{max})$, where T_{max} is the temperature of the maximum of K'' in the plateau region or the maximum in the peak of tan δ.

Figure 3 shows a plot of the pairs $(1/\omega, T_{max})$. The data in Fig. 3 were analyzed using Eq. (1), where the fitting is shown as a solid line. The fit parameters are shown in the inset of Fig. 3. A good fitting with the divergent power-law equation can be seen, which have reasonable values of the constants τ_0, T_0, and zv. The value of zv was ~11 and that of the freezing temperature was ~45 K. These results are consistent with the models of disordered magnetic systems.[15–17] The results demonstrate the importance of fractal cluster contributions to the dielectric loss mechanism of poled PMN–PT single crystals.

Dielectric loss in ferroelectrics has generally been attributed to domain-wall contributions. However, the results of this investigation show that the losses are dominated by structural irregularities (or fractal clusters of lower symmetry) inside of normal micron-sized domains. Fractal clusters are excited by application of electric field, resulting in dielectric absorption. On cooling, the average relaxation time of the fractal cluster undergoes a power-law divergence.

In summary, this letter reports the importance of fractal cluster contributions to the dielectric loss mechanism of oriented poled PMN–PT crystals. Structural irregularities within domains control the dielectric loss, rather than the motion of domain walls.

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