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# Inverse hysteresis of field induced elastic deformation in the solid solution 90 mol % $Pb(Mg_{1/3}Nb_{2/3})O_3$–10 mol % $PbTiO_3$

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Elastic deformations induced by high electric fields in the solid solution system $Pb(Mg_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ have been measured using a strain gauge method at temperatures in the relaxation range. Unusual inverse hysteretic curves have been observed in the relation between the deformation and electric field typically in 90 mol% Pb-$(Mg_{1/3}Nb_{2/3})O_3$–10 mol % $PbTiO_3$ at temperatures outside the range where they would be of practical use in electrostrictive displacement control.

## 1. Introduction

Dielectric properties and electrostriction of ceramics in the solid solution system $Pb(Mg_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ have been reported in our recent papers [1–4]. In pure $Pb(Mg_{1/3}Nb_{2/3})O_3$ and in $Pb(Mg_{1/3}Nb_{2/3})O_3$-rich compositions, a most unusual inverse hysteresis in the relation between the weak-field permittivity and the cyclic bias field has been observed at temperatures in the relaxation range.

Uchida and Ikeda [5] reported rather similar bias characteristics in the weak-field permittivity of Pb(Zr, Ti)$O_3$ based ceramics. They explained the effects qualitatively by making specific assumption as to the behaviour of 180° domain reversals and non 180° rotations of domains as a function of the applied field [6]. On the other hand, they observed normal hysteresis in the biasing field characteristics of elastic deformation for $BaTiO_3$ and PZT families as are predicted by their theoretical model.

Concerning $Pb(Mg_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ ceramics, however, we observed also the peculiar inverse hysteresis in the bias characteristics of strain. We describe in this paper preliminary experimental data of temperature and frequency dependence of longitudinal and transverse strain measured in 90 mol %$Pb(Mg_{1/3}Nb_{2/3})O_3$–10 mol % $PbTiO_3$ ceramics, which reveals the largest inverse hysteresis effect among samples containing less than 20 mol% $PbTiO_3$.

## 2. Sample preparation

Ceramic specimens were prepared from reagent grade PbO, MgO, $Nb_2O_5$ and $TiO_2$. The constituent oxides were mixed in appropriate proportions, ball-milled in alcohol, then dried and calcined in air at 800° C for 15 h in a closed alumina crucible. The resulting calcine was ground and refired for two additoinal 15 h periods to ensure complete reaction.

For the transverse strain measurement, samples were prepared by cold pressing into discs and firing on platinum setters in air at 1000° C for 2 h. Gold electrodes were sputtered onto the faces. For the longitudinal effect, internally electroded multilayer samples were prepared by standard tape casting techniques using calcined $Pb(Mg_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ powder and a commercial doctor blade media (Cladan Inc., San

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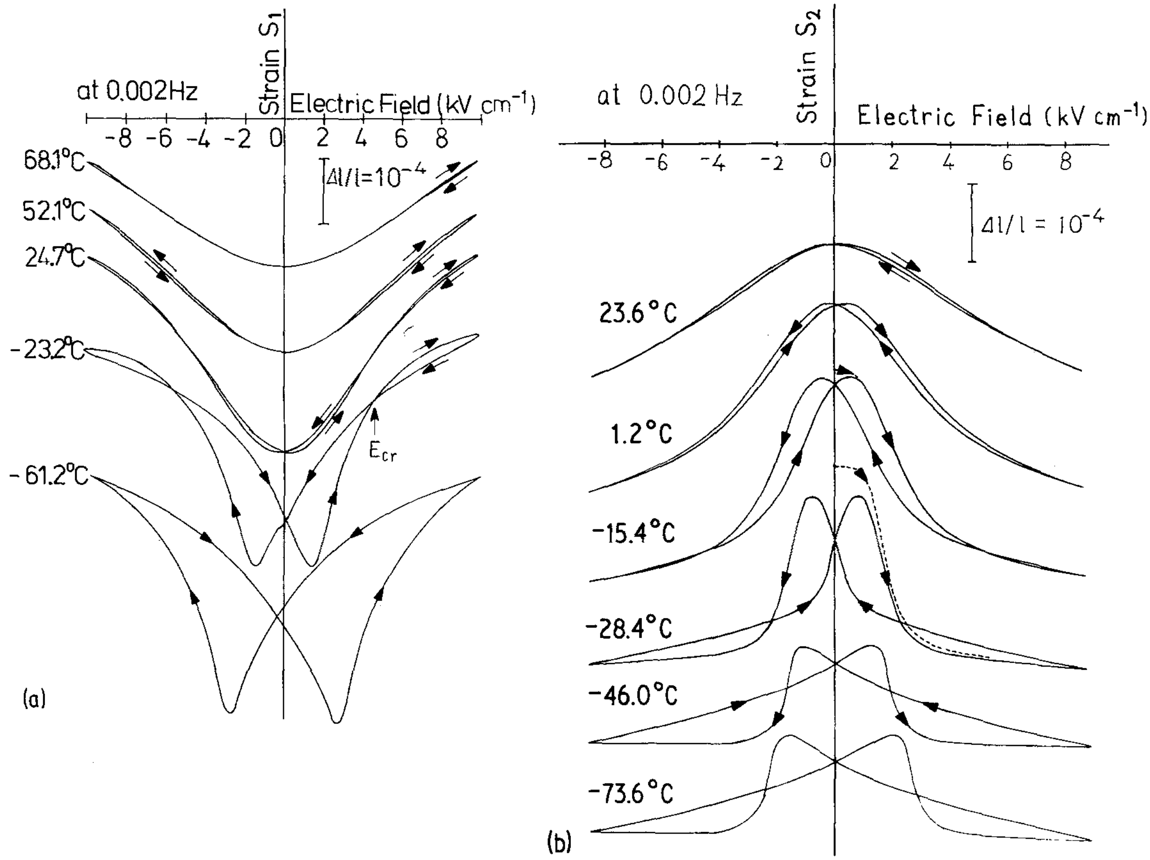


*Figure 1* (a) longitudinal electrostriction and (b) transverse electrostriction as a function of electric field (0.002 Hz) at various temperatures for 90 mol% $Pb(Mg_{1/3}Nb_{2/3})O_3$ – 10 mol% $PbTiO_3$.

Diego, CA, type B42) [7]. Internal electrodes were applied by screen printing platinum ink (Englehard Industries, East Newark, NJ, type E-305-A) onto the dried cast tape. Ten layer devices with a total thickness of 2.5 mm were prepared by firing under the same conditions as described above.

The field-induced strain was measured by a strain gauge method. A polyimide foil strain gauge (Kyowa, KFR-02-C1-11) was bonded with a cement (Kyowa, PC-6) on the electroded face of the sample for the tranverse measurement, or on the edge face of the multilayer sample for longitudinal measurements.

## 3. Experiments and discussions

Measurements were carried out by a d.c. method using a double bridge technique. The longitudinally ($S_1$) and transversely ($S_2$) induced strains were measured as a function of the applied electric field (0.001 ~ 0.1 Hz) at various temperatures (– 80 to 70° C).

### 3.1. Temperature and field dependence of strain

Figs. 1a and b show the dependence of the longitudinal ($S_1$) and transverse strains ($S_2$) on a cyclic electric field at 0.002 Hz for 90 mol% $Pb(Mg_{1/3}Nb_{2/3})O_3$–10 mol% $PbTiO_3$ ceramics at various temperatures. In the longitudinal effect the inverse hysteresis can be observed even above the mean Curie temperature (~ 25° C). Below that temperature, the normal hysteresis is dominant and the inverse hysteresis is observed only under high electric fields, and gradually disappears with decreasing temperature. In the transverse effect, the normal hysteresis is dominant at all temperatures. Below – 10° C, however, the inverse effect appears at high electric field. It is notable that the magnitude of the strain change decreases

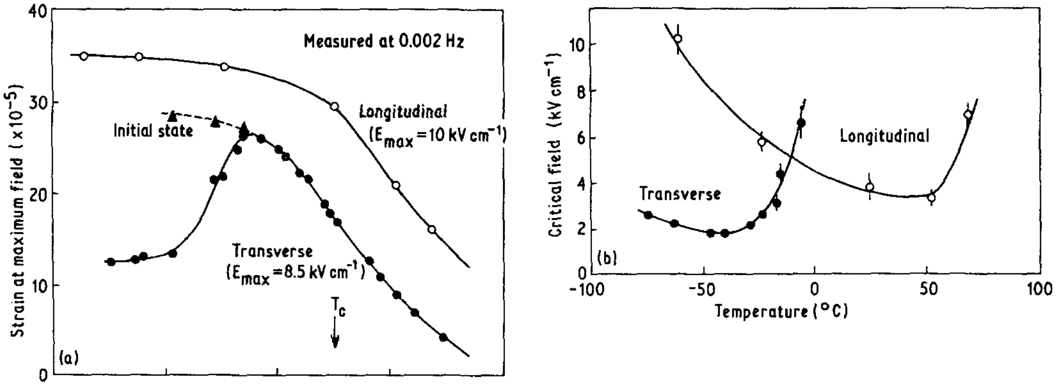


*Figure 2* (a) maximum longitudinal electrostriction at $E = 10\,\text{kv cm}^{-1}$ and maximum transverse electrostriction at $E = 8.5\,\text{kv cm}^{-1}$ as a function of temperature. Initial strains are also plotted (dashed lines); (b) critical field of the inverse hysteresis as a function of temperature.

with decreasing temperature below $-10°$ C. The initial strain curves are also shown in Fig. 1b, by dashed lines.

Fig. 2a shows the maximum longitudinal strain at $E = 10\,\text{kV cm}^{-1}$ and the maximum transverse strain at $E = 8.5\,\text{kV cm}^{-1}$ plotted as a function of temperature. The initial strains of the transverse effect are also plotted for comparison. The temperature dependence curves are very similar to the curve for the polarization. Compared with the value of the transverse strain, the longitudinal strain is relatively smaller than the expected value ($S_1 \sim -3S_2$), which can be attributed to the effect of the multilayer configuration due to two separate electrode systems and insulated electrode edges reducing the measured displacement [7]. The large strain difference between the initial and cyclic states in the transverse effect suggests that part of the non 180° rotations of domain switching may be quenched by the initial field application and does not subsequently contribute to the cyclic domain switching. This quenching effect is not observed in the longitudinal effect.

Fig. 2b shows the plots of the critical electric field for inverse hysteresis as a function of tem-

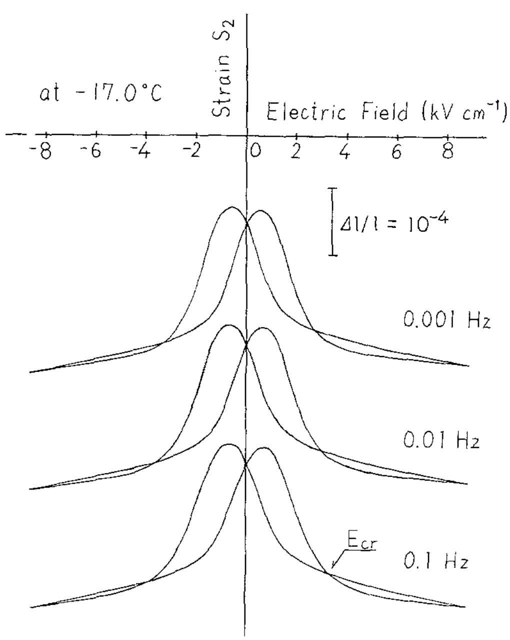


*Figure 3* Frequency dependence of the transverse electrostriction curve measured at $-17.0°$ C.

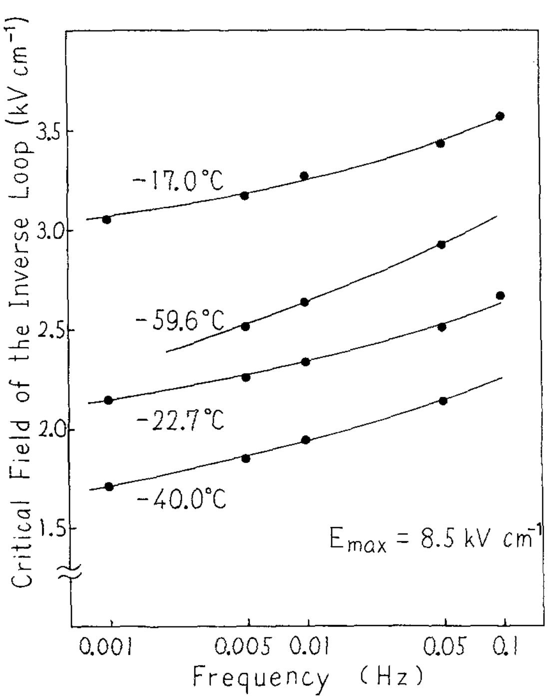


*Figure 4* The dependence of the critical field of the inverse hysteresis on frequency and temperature.

perature. The critical field shows a minimum near the mean Curie temperature.

### 3.2. Frequency dependence of strain

Fig. 3 shows the frequency dependence of the transverse strain ($S_2$) curve measured at $-17.0^\circ$ C. The critical field $E_{cr}$ required for inverse hysteresis increases with increasing frequency, as is clearly evident in Fig. 4. It is notable that the slope of the $E_{cr}$–$\log f$ curve tends to increase with decreasing temperature.

It is not possible to explain the inverse hysteresis in the elastic strain by the model proposed by Uchida and Ikeda [6]. It appears probable that consideration must be given to the possible interactions between 180° and non 180° reorientations which are ignored in the Uchida–Ikeda model. Another possibility is the effect of the superposed giant quadratic electrostriction usually observed in relaxor ferroelectrics [3, 4]. Further investigation will be necessary to expand these possible models for the inverse hysteresis effect.


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