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RESPONSE OF FERROELECTRIC CERAMICS TO UNIAXIAL STRESS

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Abstract

Changes in electrical polarization of ferroelectric ceramics subjected to non-hydrostatic stress can be caused by rotation of ferroelectric domains, or, in the case of materials near ferroelectric to antiferroelectric phase boundaries, by a stress-induced phase transformation. Both processes have been observed in mechanical measurements made on PZT 95/5. The results may have important implications for the operation of shock-actuated ferroelectric power supplies.

Introduction

Ferroelectric materials with stress-induced ferroelectric (FE) to antiferroelectric (AFE) phase transitions are of interest because of their use in shock-pulse actuated power supplies [1-3]. At present, only a limited understanding of the operation of such devices has been developed. A major factor limiting the understanding of stress-wave propagation in FE's is the fact that a correct theory must be quite complex, both because a number of important physical processes must be included, and because these processes are highly nonlinear, rate-dependent and coupled to one another [4,5]. Measurements of electrical and mechanical properties under conditions of statically applied stress can provide information that is helpful in elucidating many of the properties of FE ceramics [6-8]. Although the stress states and time scales of static-stress experiments are not the same as under shock-pulse loading, the physical processes that occur should be qualitatively similar.

In this paper some recent experiments on PZT 95/5 ceramics will be reviewed. Results of mechanical measurements, made on unpoled material only, will be presented. Much of the interesting stress response can be seen in data from such studies. Experiments under hydrostatic-pressure conditions will also be briefly discussed.

Experiments

The chemical formula of the niobium doped PZT 95/5 used in the experiments is $Pb_{0.99}Nb_{0.02}(Zr_{0.95}Ti_{0.05})_{0.98}O_3$. Most of the studies were done on material which was prepared by chemical coprecipitation followed by hot pressing, although some work has been done on low-density sintered material [6]. The hot-pressed material typically withstood uniaxial stress levels up to ~ 0.6 GPa, whereas the sintered material usually broke at ~ 0.4 GPa.

Experimental techniques used to obtain electrical and mechanical data were standard and will not be discussed in detail. Mechanical strains were measured by small strain gauges cemented to the rectangular sample faces. Measurements of longitudinal sound velocities were made by a standard pulse-superposition technique at 10 MHz.

Results and Discussion

The hydrostatic pressure data will be discussed first. PZT 95/5 undergoes a pressure-induced FE-AFE transition at a pressure of 0.2 GPa under hydrostatic conditions. The transition can be seen in Fig. 1, which is a plot of the volume strain $x_V = (V - V_o)/V_o$ as a function of pressure for an unpoled sample. Here V is the volume at pressure p and V_o is the initial volume. With increasing pressure, the transition from FE to AFE is marked by the rather sharp downward break in the curve at 0.2 GPa. The plot shows significant curvature up to ~ 0.8 GPa, whereafter the strain becomes relatively linear with pressure. As a function of decreasing pressure, the strain exhibits hysteretic behavior, and the reverse transformation is only partially complete at atmospheric pressure. The overall change in strain due to the transition is 0.8%.

The ultrasonic data as a function of pressure, again for an unpoled sample, are shown in Fig. 2. The variable f(p) is a measured pulse repetition rate, and is proportional to the longitudinal sound velocity. From Fig. 2 it is seen that the FE-AFE transition is accompanied by an anomalous increase in velocity of ~ 14%. Such an increase is expected, as the transition is to a denser, and hence mechanically stiffer, phase.

Under conditions of applied uniaxial stress, it is to be expected that anomalies similar to those shown in Figs. 1 and 2 should be observed if the phase transition occurs. However, there is a complication because domain reorientation processes also are expected [7,8]. Domain reorientation occurs because the crystalline unit cell is elongated along the polarization axis. Under the influence of an applied stress, the domains whose polar axes are aligned close to the stress direction will tend to switch to a configuration with the polar axes more nearly perpendicular to the stress direction. With increasing stress, the strain x_3 parallel to the stress direction should exhibit an anomalous decrease, as the domains switch, whereas the strain x_1 should have an anomalous increase. These anomalies are with respect to the normal elastic strains produced by the applied stress, and they occur because of the elongation of the unit cell along the polarization direction. No anomaly in the volume strain $x_V = 2x_1 + x_3$ is expected. The ultrasonic velocity perpendicular to the stress direction should show an anomalous decrease due to switching, because the ferroelectric perovskites are softer along their polarization axis than perpendicular to it.

Strain gauge data for PZT 95/5 under uniaxial stress are given in Fig. 3. The three curves are for the strain x_1 normal to the stress, the volume strain x_V and the strain x_3 parallel to the stress. The dashed lines represent the purely elastic contribution, as predicted from elastic constant data. The response is elastic only for very low stress levels (< 0.05 GPa). In the range of ~ 0.1-0.3 GPa x_1 has an anomalous increase and x_3 an anomalous decrease, as would be expected for a domain reorientation process. However, x_V also has an anomaly in the 0.05-0.3 GPa range, indicating that some of the crystallites may actually be transforming beginning at ~ 0.05 GPa. From ~ 0.3 GPa up to the highest stresses attained, the data show that the phase transformation is occurring. This can be seen clearly from the cross-contraction in x_1 and the downward inflection in x_v. The three curves do not indicate elastic behavior even at the highest stress levels of the experiment, and it is concluded that the FE-AFE phase transformation is not complete by 0.6 GPa.

Ultrasonic data as a function of uniaxial stress are shown in Fig. 4, which shows relative velocity changes, as a function of stress, for waves propagating perpendicular to the stress direction. These data support the above conclusion that both domain reorientation and the FE-AFE transition occur in the experimental stress range. Between 0 and 0.15 GPa the velocity increases slightly. If the explanation proposed above for the behavior of the strains between 0.05 and 0.3 GPa (partial transformation, accompanied by domain rotation) is correct, then the small change in velocity up to 0.15 GPa is the net effect of the two processes, which have opposite effects on the velocity. Between 0.15 and 0.23 GPa the velocity decreases, indicating that domain reorientation dominates the response. Above 0.23 GPa the velocity rises rapidly with increasing stress, consistent with the large increase associated with the transition as illustrated by Fig. 2.

Conclusion

The data discussed above show that the response of unpoled 95/5 ceramic to uniaxial stress is determined by a competition between domain switching processes and the FE-AFE transformation. The same is undoubtedly true of the response of the material to shock-wave loading, although direct quantitative comparison is not possible, because the stress state is different (uniaxial strain conditions) and because rate effects appear to be important [9]. There are, in fact, some published data which indicate that the effects observed under uniaxial stress and shock-wave loading may be similar. One of the most striking features of the uniaxial stress data presented above is the broad spreading of the FE-AFE phase transition compared to the hydrostatic-pressure case. Similar behavior was found by Lysne [4] in a study of shock-induced depoling of PZT 95/5 in the "normal mode" configuration (shock wave and polarization directions perpendicular). Lysne found that stresses greater than 1.6 GPa were necessary for complete depoling. This may be a larger stress than is needed to complete the transition in an unpoled sample under uniaxial stress conditions, but a somewhat larger transformation stress is expected in the shock-wave case, because of the stress being perpendicular to the polarization direction, and because a larger stress is required to produce a given volume change [10].

In conclusion, it appears that a complete theoretical description of shock-loaded FE's will require taking a number of factors into account, including rate-dependent switching and transformation processes, and the anisotropic nature of the material.

References

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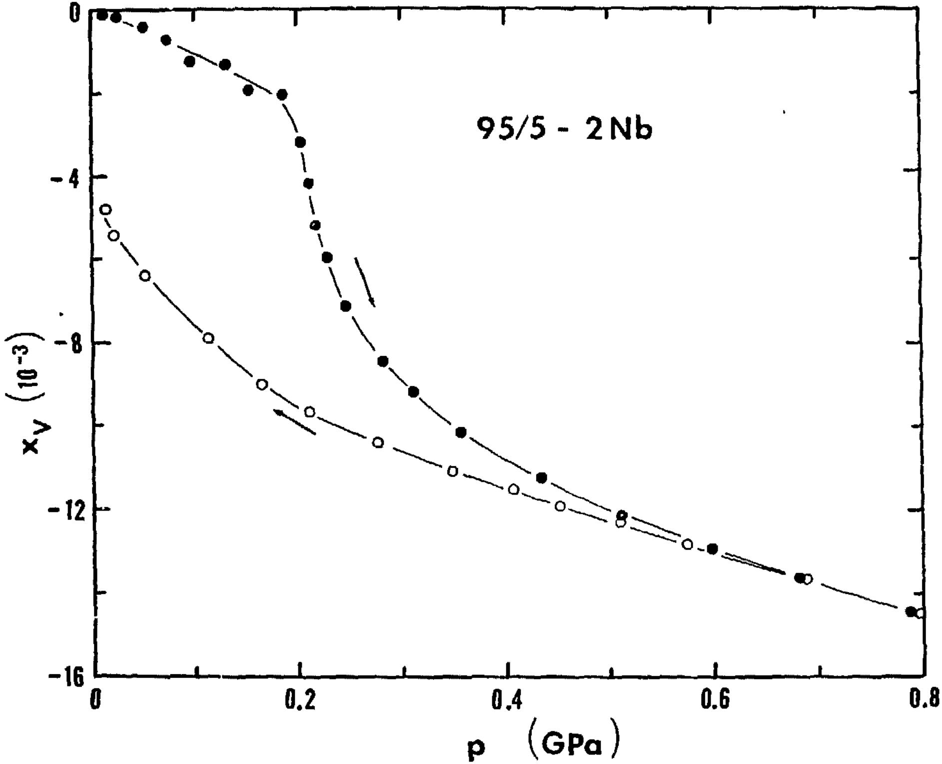
Figure Captions

Fig. 1 Volumetric strain as a function of hydrostatic pressure for PZT 95/5. Sharp drop at ~ 0.2 GPa is due to the FE-AFE phase transition.

Fig. 2 Reduced ultrasonic pulse repetition rate versus pressure for PZT 95/5.

Fig. 3 Axial (x_1,x_3) and volumetric (x_V) strains versus uniaxial stress (σ_3) for PZT 95/5. Dashed lines represent elastic behavior predicted from ultrasonics. Deviations from elastic behavior are due to domain switching and phase transition, as explained in text.

Fig. 4 Ultrasonic data versus uniaxial stress for PZT 95/5. Data are for longitudinal waves propagating normal to the stress direction. Velocity decrease is due to domain rotation, large increase is due to FE-AFE transition.



Fritz
Fig. 1

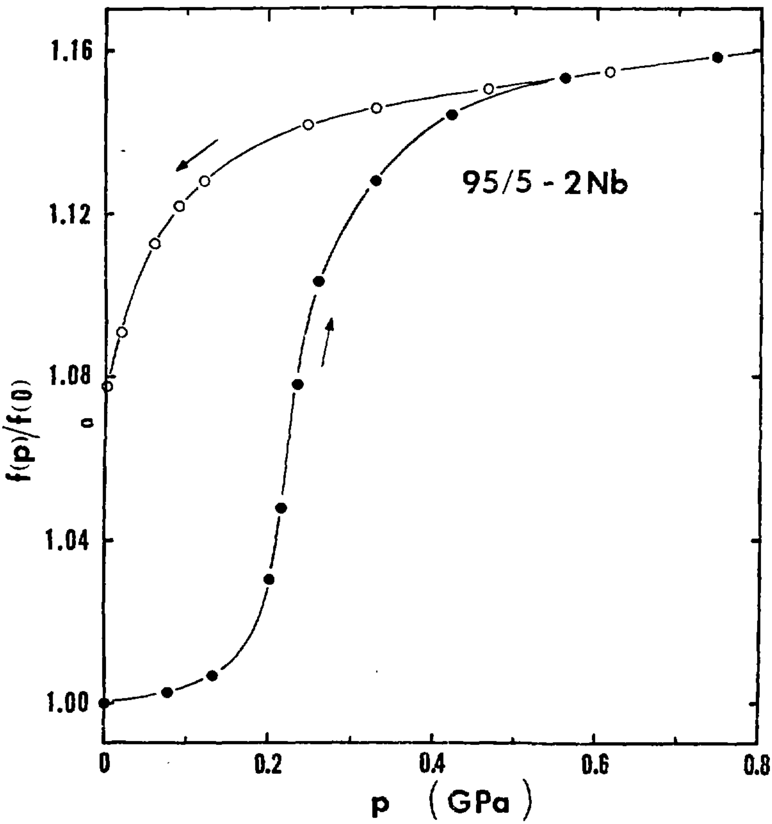


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Fig. 2

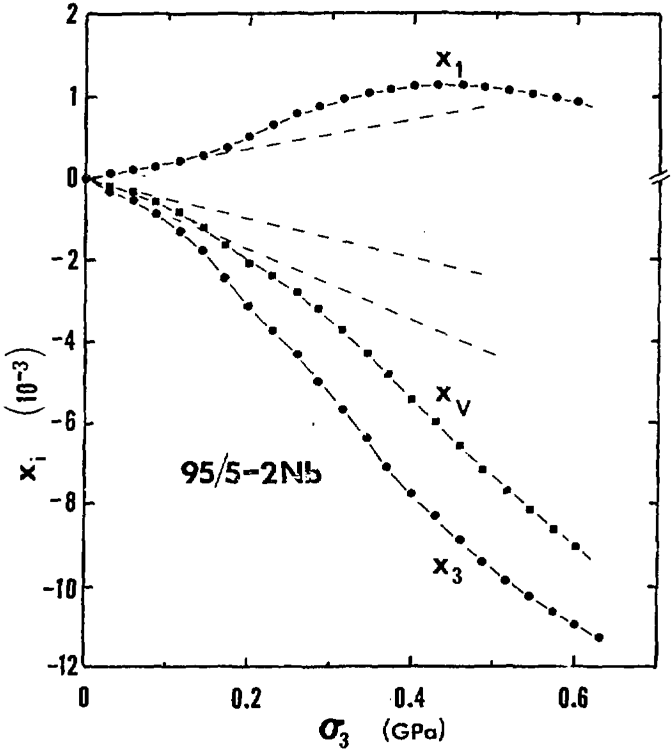

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Fig. 3

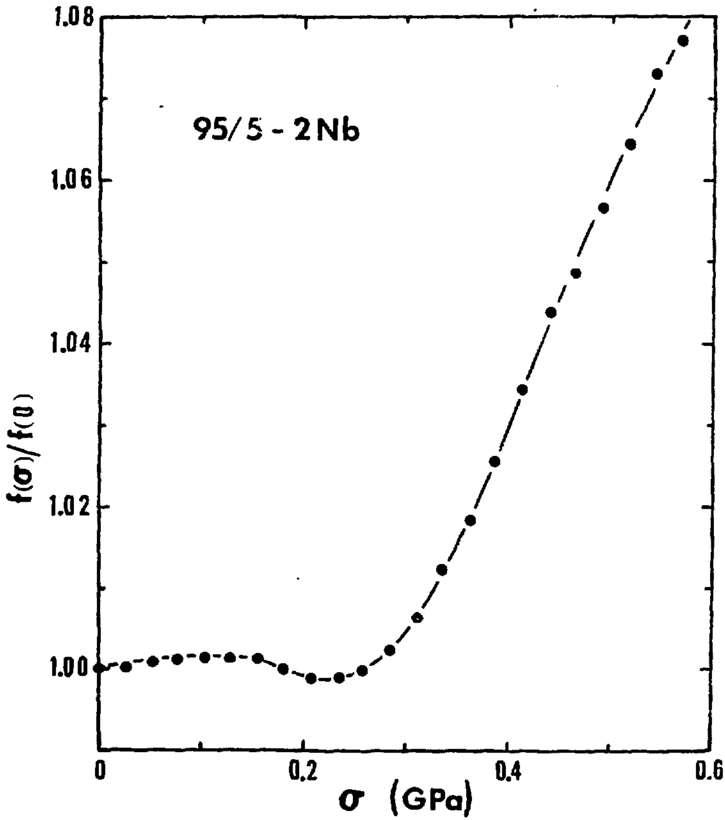

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Fig. 4