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THE TWIN STRUCTURES AND THE PIEZOELECTRIC EFFECT IN FERROELECTRIC CERAMICS

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Experimental evidence show that the piezoelectric effect in ferroelectric ceramics cannot be accounted for by the average of the intrinsic piezoelectric effect of the oriented ferroelectric domains. The measured values of piezoelectric moduli d_{ij} are usually 60–70% larger than the calculated average values at room temperature.[1,2,3] The difference as well as the absolute values of d_{ij} become smaller when the system is cooled,[4] which suggests that reversible ferroelectric/ferroelastic domain wall movement may be the cause for the discrepancy, because the walls are more mobile at higher temperatures.

The movements of defect-free walls and walls which contain high defect concentration are controlled by different mechanisms. Collective motion of domains are involved for the coherent defect free walls, but ion hopping occurs for the movement of a defect wall. In either cases, it is conceivable that the response time is longer for the domain wall process compared with the intrinsic process (polarization induced unit cell distortion). Therefore, the relaxation of the domain wall process occurs at relatively lower frequencies ($\sim 10^9$ Hz). This has been confirmed by experiments on both $BaTiO_3$ and PZT systems.[5] It is found that the difference between the values of d_{ij} at low and microwave frequencies differ by as much as 60–70%, which agrees with the theoretical estimates mentioned above. From these arguments it is concluded that the piezoelectric effect in ferroelectric ceramic contains substantial extrinsic contributions, i.e., resulting from the reversible movement of non-180° domain walls.

In order to provide further support for this claim, we have investigated the nonlinearity of the piezoelectric moduli by driving a PZT system in large electric fields. Experimental results indicate that the increase of nonlinearity is accompanied by the increase of loss.[6] This implies that the piezoelectric nonlinearity is mainly generated by the movement of domain walls since the wall movement is known to be responsible for the generated loss.

As a major contribution to the observed piezoelectric effect in ferroelectric ceramics, the domain wall movements have not been given enough attention. There remain some fundamental questions despite the previous efforts mentioned above. From the physical point of view there are two stages in addressing the domain wall problem: the static and the dynamic. In the first stage, questions to be addressed include:

(1) What are the detailed lattice positions in a twin structure, especially inside the domain wall region?
(2) What is the energy stored in a domain wall?
(3) What are the roles of defects in the formation of a twin and the displacement of a domain wall?
(4) What are the relative movements experienced by the lattices in domain wall displacements?
(5) What factors control the density of domain wall formation?

We have recently constructed a Landau-Ginzburg model which could provide some answers to some of these questions.[7] It is found that a twin boundary has a finite width which is determined by the strength of nonlocal coupling of the polarizations. The crystal structure experiences a gradual change across a twin boundary. In the case of a 90° tetragonal twin, the symmetry at the center of the twin boundary is quasi-orthorhombic. It is also proved that positive energy is stored in a twin boundary. The twin structure is stabilized by the internal and/or external stress fields which are provided either by defects or by intergranular coupling. Any changes of the stress distributions will therefore introduce spatial instabilities to the domain walls, and vice versa.

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