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ACOUSTIC EMISSION STUDIES OF DISPLACIVE TRANSFORMATIONS

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ABSTRACT

When ferroelectric crystals undergo their displacive transformation they produce marked acoustic emission; in contrast, single crystals of indium alloys emit acoustically well below their martensitic phase transition, probably due to twin boundary movement. Domain boundary movement as the crystal becomes polarised is shown to produce pronounced acoustic emission in ferroelectric crystals.

Crystals undergoing phase transitions could be clearly defined sources of acoustic emission. Phase transitions in solids can be classified into two main groups : (i) those in which the transition is associated with individual movement of the atoms, (ii) the displacive, structural transformations which take place by cooperative atomic movements. In studies of the nature of AE a reproducible source, which can be used repeatedly, is desirable. Crystals undergoing diffusionless displacive transitions could be such a source - a fact recognised by a number of workers (1-4) who have studied AE generated during martensitic transformations. Our aim is to compare and contrast AE from materials which undergo displacive phase transitions; work has been performed on both martensitic (indium alloys) and ferroelectric materials (Rochelle salt, TGS, LATGS, $BaTiO_3$ and $Pb_5Ge_3O_{11}$).

One factor which has influenced the choice of phase transitions is the requirement that there should not be large volume changes at T_c. Thus several of the transitions studied are essentially second order in character. Such phase transitions are usually accompanied by softening of a phonon mode; as T_c is approached the soft mode energy tends towards zero and the number of thermally excited soft mode excitations grows until it reaches an essentially unlimited value and the phase transition occurs. In the martensitic transitions studied here, soft acoustic mode behaviour is evidenced by the approach to zero of an elastic constant (5). Ultrasonic methods allow direct observation of the mode softening and thus of $\partial\omega/\partial\underline{q}$ near T_c. One ultimate aim of the present work is to seek a relationship, if any, between the acoustic mode softening and acoustic emission.

Indium and its disordered alloys with cadmium and thallium undergo a soft acoustic mode, martensitic transition from a f.c.c. (high temperature) to a f.c.t. (low temperature) phase. The acoustic mode which softens is that polarised $[1\bar{1}0]$ for a wave vector along $[110]$ and is manifested as the tendency for the elastic stiffness modulus $\frac{1}{2}(C_{11}-C_{12})$ to go towards zero at T_c(6). There is a peak at T_c in the ultrasonic attenuations (7) which provides a monitor of T_c and its thermal hysteresis. A typical example is illustrated in Figure 1 for an In-4.43 at.%Cd sample which is in a banded twin tetragonal form below, and a single f.c.c. crystal above T_c. The hysteresis is small - however it does indicate that the phase transition is first order, although the latent heat and the volume changes at T_c are negligible; the behaviour is close to that expected of a second order transition. Figure 1 shows that there is no direct correlation between T_c, as observed by ultrasonic techniques, and the AE produced. At room temperature this sample is in the banded twin form; as the temperature is raised, there is a steady but relatively small amount of AE. At 380K the sample undergoes the transformation by passage of a single (110) type interface, and becomes a single crystal, without an obvious surge of AE. As the temperature is lowered through T_c (370K), the single crystal transforms back into a banded twin with no detectable AE. However, on further cooling AE onsets well below T_c and the count increases dramatically by an order of magnitude greater than that measured on the heating cycle. If a banded twin sample is heated up towards, but not through T_c, it does not show the massive emission on cooling shown by samples which have undergone the transition. Plausibly, AE produced during cooling is due to twin movement (rather than twin production at T_c) as equilibrium rearrangement of the banded twins occurs to minimise strain induced at the transformation.

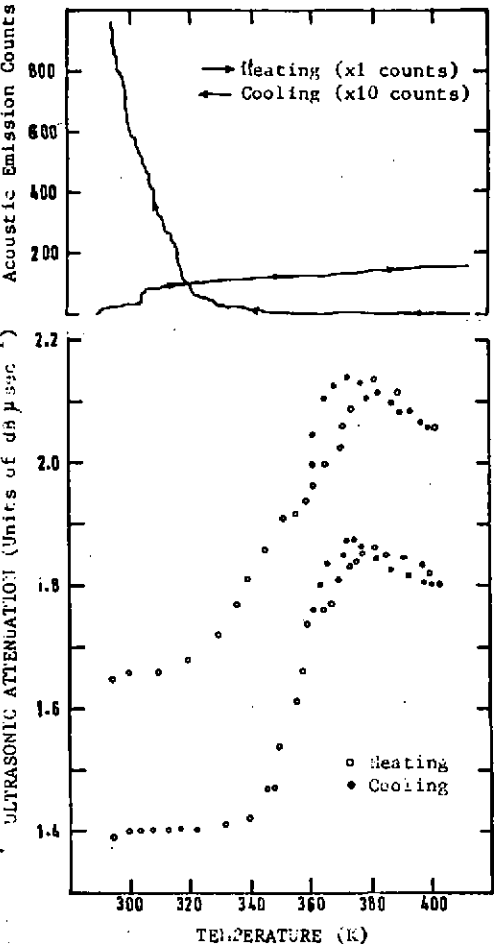

Fig.1. AE and ultrasonic attenuation in the vicinity of the martensitic phase transition in In-4.43 at.%Cd alloy.

Previously, Buchman (8) has observed AE from ferroelectric $BaTiO_3$ both as a function of the applied electric field in the polar phase and when the crystal goes through the tetragonal to cubic phase transition in the region of 390K. We have observed AE as Rochelle salt (Figure 2) and TGS undergo their phase transitions. In contrast to the indium alloys, these ferroelectrics exhibit marked AE activity only at the phase transition; the levels of emission are similar both on the heating and the cooling cycles. The hysteresis confirms the slight first order character of the transition in Rochelle salt.

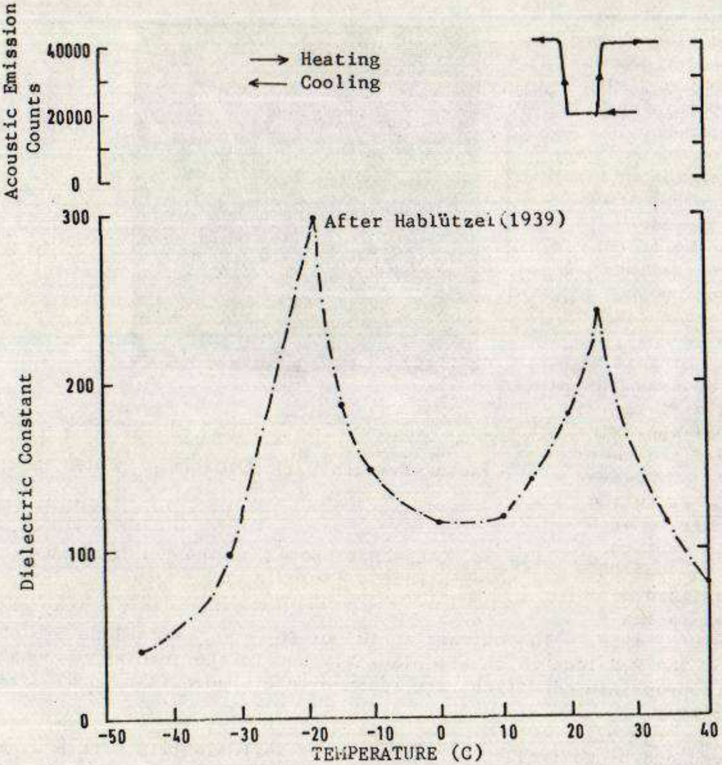

Fig.2. AE at the Upper Curie point of Rochelle salt. The dielectric constant data (9) shows the Curie temperatures.

As ferroelectrics are reversibly polarised, domain movement takes place. To examine the question of whether such domain activity produces AE, crystals of TGS, LATGS, $BaTiO_3$ and $Pb_5Ge_3O_{11}$ have been taken round the hysteresis loop whilst the acoustic activity is monitored. Several interesting features have become apparent. The period of the AE envelope is twice that of the AC field applied across the polar axis of the crystal. There is a difference in the peak intensity of AE activity which is roughly proportional to the value of the saturation polarisation for the particular ferroelectric. Thus $BaTiO_3$ appears most active and Rochelle salt the least. The intensity and duration of the emission produced during one cycle of the applied field is also dependent upon the instantaneous position on the P-E loop (Figure 3). The maximum signal is observed for electric field strengths (or induced polarisation) just below the saturation value, corresponding to the position on the look at which the extinction occurs of the final few domains misoriented relative to the applied field.

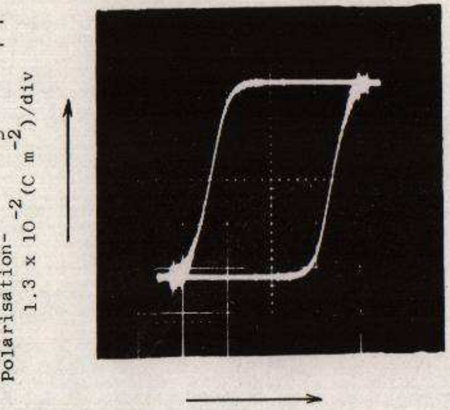

Fig.3. AE superimposed on the P-E hysteresis loop for a 50 Hz electric field in TGS.

Smaller but well defined AE is also observed at a point corresponding to the field at which domain nucleation commences. No emissions have been observed in TGS or $Pb_5Ge_3O_{11}$ along the parts of the loop corresponding to constant polarisation. In this region the domains remain aligned and do so until nucleation of antiparallel domains occurs at a field strength of approximately half the coercive field. However, in $BaTiO_3$ saturation is difficult to attain due in part to the existence of 90° domains aligned perpendicular to the tetragonal 'c' axis. In contrast to TGS or $Pb_5Ge_3O_{11}$ emissions are observed throughout the P-E loop which are plausibly due to the removal of such domains when the appropriate conditions are reached to overcome this local stress induced anisotropy.

AE produced by controlled polarisation of ferroelectric crystals has been found to be more reproducible than that from martensitics.

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