

THE FORMATION OF DOMAIN STRUCTURE DURING JUMP MOTION OF A PHASE FRONT IN THIN PbTiO_3 MONOCRYSTALS

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A description is given of conditions in which ordered 180° wedge-shaped domain configuration may be formed in PbTiO_3 monocrystals.

The domain structure of crystals of lead titanate has been described in detail in [1–8]. The particular feature of PbTiO_3 crystal domains, as distinct from BaTiO_3 , is that they form fairly large monodomain regions blocked on the side of the “head” P_s by 180° wedge-shaped domains. The polarisation of such wedge-shaped domains is opposite to that of the remaining volume of the domain. These peculiarities in domain structure are closely related to the conditions in which phase transition and the formation of domain structure take place, i. e.

- high conductivity at the Curie point ($\sigma_{T_c} \sim 10^{-3} \text{ (ohm cm)}^{-1}$)
- large change in spontaneous polarisation $\Delta P_s \sim 40 \text{ } \mu\text{C/cm}^2$
- high elastic stresses.

Phase transition in PbTiO_3 crystals is similar to the martensite transformation [9, 10], i. e. the ferro- and paraelectric phases are separated by a sharp boundary (the phase front), and in the phase of lower symmetry 90° twins are formed. The angles between the normal to the phase front n and the crystal axes (Fig. 1) have the values $\omega_x = 89^\circ 6'$, $\omega_y = 55^\circ 30'$, $\omega_z = 34^\circ 30'$, which correspond to crystallographic planes of indices (320). Calculated from LWR equations [7, 9] the angle values are in good agreement with those above. Symmetry allows the occurrence of 24 crystallographically equivalent planes of phase division. All these 24 planes may be obtained in the temperature-gradient field by orientating the gradient in such a way that the position of the isothermal surfaces approximates to a plane of type (320). In report [10] it was shown that, depending on crystal orientation relative to direction of temperature gradient, various types of phase fronts may be obtained. Each phase front corresponds to a different 90° domain structure formed after the

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phase transition. Further it was ascertained that, depending on the velocity of the isothermal surface, both the type of phase front motion and the 180° structure formed behind the front are changed.

In particular it was observed that in a certain velocity range the phase front progressed in jumps. In the present report results are a presentation of detailed investigations on jump motion of the phase front and the formation of domain structure under these conditions.

PbTiO_3 crystals were obtained from the $\text{PbO-TiO}_2\text{-B}_2\text{O}_3$ system by the technique described in report [3]. Monocrystal samples of dimensions 2×3 mm and thickness from

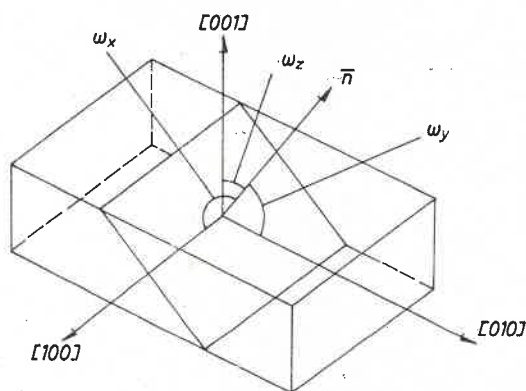


Fig. 1. Position of phase separation plane

20 to 200 μm were cut for testing. The samples were placed in a gradient furnace equipped with an optical channel making possible observations of phase transition in a polarisation microscope. In each case the samples were placed so that the temperature gradient was along the direction [010], corresponding to the situation presented schematically on Fig. 1. All measurements were performed with the same and constant temperature gradient of $\Delta T/\Delta x = 6^\circ \text{C/mm}$. Under such conditions, shift velocity of the phase boundary depends only on the rate of the temperature changes ranging from 0.15 to 3°C/s . The mean value of the shift velocity of the phase boundary was estimated from measurements of the shift distance and the suitable time intervals. It was shown that the values of the shift velocity coincide with the values of the isothermic surface velocity, which may be calculated from:

$$v = - \frac{R}{g},$$

where R is the rate of temperature changes, and g is the constant temperature gradient.

The domain structure was brought out by etching sections of the plane (100) and (001). The crystals were cut, polished and then etched for 3–5 sec by a mixture of hydro-fluoric and hydro-chloric acids. It appeared that the boundaries of the antiparallel domains in these sections, evidently due to the deviations of the plane secant from the normal to (001), etch intensively, revealing the domain pattern.

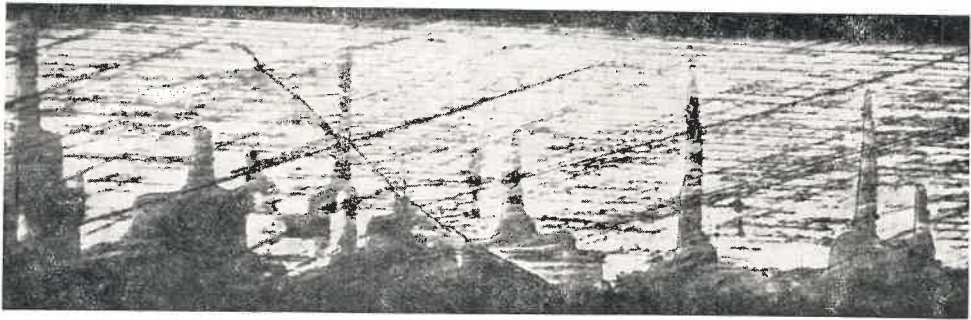


Fig. 2a. Etching figures of section through plane (100)



Fig. 2b. Etching figures of section through plane (001)

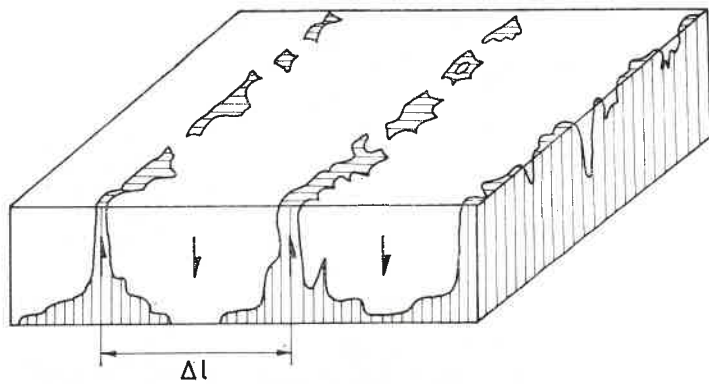


Fig. 2c. Diagram of 180° domain structure formed

Fig. 2 shows the figures obtained from etching a section of a sample through plane (100). Distinct wedge-shaped reciprocal domains may be seen, their apices located at approximately equal intervals. The mean distance between apices is equal to the mean length of jump determined by an optical method. The etching figures for section through plane (001), made at about 30 μm below the upper crystal surface, are shown in Fig. 2b. Darker etching figures, corresponding to the positive ends of the spontaneous polarisation vector, may be seen regularly distributed over the whole surface of the crystal. They correspond to the wedges which also etched as dark lines in Fig. 2a. On the basis of these two sections the conclusion may be drawn that in the phase front jump motion process an ordered wedge-shaped domain structure is formed, for which spatial scheme is shown on Fig. 2c.

Tests conducted on phase front motion in crystals of thickness $d = 20 \div 200 \mu\text{m}$ showed that when jumps occur in a phase front the velocity range is changed. In this way

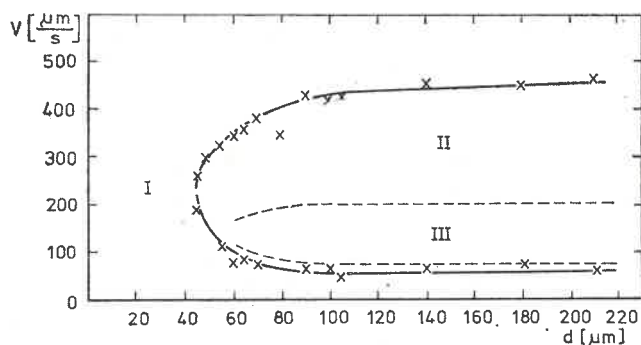


Fig. 3. Diagram showing relation between jump region and crystal thickness

regions corresponding to the various types of phase front motion were determined as shown on the diagram $V-d$ (Fig. 3) in which may be distinguished:

- I. a region of velocities at which the front progresses smoothly,
- II. a region of transition or of unstabilised jumps. Irregular jumps interspersed with sections of smooth motion may be observed,
- III. a region of regular jumps. In this velocity interval the phase front progresses by regular jumps of approximately equal length. In this case only a regular 180° wedge-shaped domain structure, as described above, is formed.

It is noteworthy that with crystal thickness, from about 80 μm the region of jumps grows smaller and at thickness $d \leq 45 \mu\text{m}$ no jumps were observed throughout the whole tested phase front velocity range. It was also observed that there is a relation between mean length of jump and sample thickness, and in particular that when sample thickness is reduced from 100 μm to 60 μm the jump length is correspondingly reduced from 70 to 20 μm . From this it may be concluded that ordered 180° wedge-shaped domain configurations can only be formed in crystals of thickness $d \geq 60 \mu\text{m}$.

In this way it was shown that the ordered a 180° wedge-shaped domain configuration has been precisely associated with jump motion of the phase front and may only be formed of thickness $d \geq 60 \mu\text{m}$.

REFERENCES

- [1] E. G. Fesenko, R. V. Kolesova, *Kristallografiya* **4**, 62 (1959).
- [2] M. Tanaka, T. Yatsuhashi, G. Honjo, *J. Phys. Soc. Jap. Suppl.* **28**, 386 (1970).
- [3] E. G. Fesenko, V. G. Gavrilachenko, R. J. Spinko, M. A. Martynenko, E. A. Grigoreva, A. D. Feronov, *Kristallografiya* **17**, 153 (1972).
- [4] E. G. Fesenko, V. G. Gavrilachenko, *J. Phys. Suppl.* **33**, C2-169 (1972).
- [5] E. G. Fesenko, V. G. Gavrilachenko, M. A. Martynenko, A. F. Semenchov, Y. P. Lapin, *Kristallografiya* **18**, 1014 (1973).
- [6] E. G. Fesenko, V. G. Gavrilachenko, M. A. Martynenko, A. F. Semenchov, Y. P. Lapin, *Ferroelectrics* **6**, 61 (1973).
- [7] E. G. Fesenko, M. A. Martynenko, V. G. Gavrilachenko, A. F. Semenchov, *Ferroelectrics* **7**, 309 (1974).
- [8] M. Tanaka, *Acta Crystallogr.* **A31**, 59 (1975).
- [9] M. S. Wechler, D. S. Liberman, T. A. Read, *Trans. Amer. Inst. Mining Met. Energ.* **197**, 1503 (1953).
- [10] E. G. Fesenko, M. A. Martynenko, V. G. Gavrilachenko, A. F. Semenchov, *Izv. Akad. Nauk SSSR, Ser. Fiz.* **39**, 762 (1975).