CONTRIBUTION TO THE THEORY OF FERROELECTRIC POLARIZATION CURVES

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The principles of a polarization theory for single- and many-domain ferroelectric crystals possessing one or two Curie points are developed for weak, strong, and medium fields at various temperatures. In contrast to the magnetization-curve theory previously developed, it is assumed that rotation can be neglected in the first approximation. The various refinements can be introduced in a way similar to that employed in the magnetization-curve theory.

THE theory of electrification curves for ferroelectrics should be formulated with account of the results of the theory of magnetization curves of ferromagnets.[1] Sometimes, however, one begins with the theory formulated by Becker in 1930, in which inversion processes (shifting of the domain boundaries) are not taken into account. According to this theory, if one starts with an S-shaped curve (Fig. 1), the Barkhausen jump should not occur anywhere except at points C or C′. In this case the coercive force E'_C is determined by the segment DC′. In 1931 — 1933[1] we showed that this theory is inadequate in principle and in practice. Actually (as indicated, for example in reference 1) the Barkhausen jump can occur at any point δ or in the segment RC (or R′C′), which indeed determines the value of E_C. This is due to the existence of inversion (displacement of the domain boundaries), in addition to rotation processes and the paraprocess.

If these concepts are carried over to the theory of ferroelectric electrification curves it becomes necessary to develop a new theory of coercive force and its temperature dependence, differing in principle from the single-domain theory (in spite of the attractiveness of this theory because of its great simplicity).[2] In solving this problem, we are able at the same time to formulate a theory of susceptibility in weak and strong fields. Owing to the existence of the rotation and inversion processes, and of the paraprocess we have accordingly for the susceptibility $\kappa = \kappa_r + \kappa_i + \kappa_p$, with κ_r small for Rochelle salt. To calculate κ_i and κ_p at various values of T, we start with the relation we have derived for the temperature dependence of the electrification curve[3]

$$E = -a(T-\Theta_1)^{\nu}(\Theta_2-T)^{\mu}P + BP^3 + CP^5, \qquad (1)$$

where P and E are the polarization and the electric field, Θ_i are the Curie points, a and B are parameters, and ν and μ are integers. In the case of a single Curie point, $\mu = 0$.

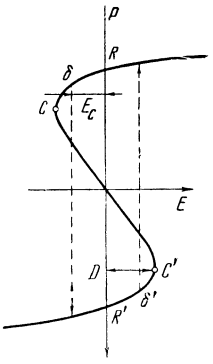

FIG. 1

This formula can be obtained most simply in the following manner:[3] we expand the work of electrification into a series (even) in powers of P

$$\int_0^P EdP = \tfrac{1}{2}AP^2 + \tfrac{1}{4}BP^4 + \tfrac{1}{6}CP^6 + \ldots \qquad (2)$$

The quantity A is expanded in powers of the differences $T-\Theta_1$ and Θ_2-T

$$A = A_0 + C_1(T-\Theta_1)^{\alpha} + C_2(\Theta_2-T)^{\beta} - a(T-\Theta_1)^{\nu}(\Theta_2-T)^{\mu} + \ldots \qquad (2')$$

But A = 0 at the Curie points for here $(P)_{E=0} = 0$. Equation (1) follows from (2) and (2′).

In addition, we should know the dependence of the domain energy U on the positions of the boundaries between domains. Let x be the displacement of the boundary from the equilibrium position. Then, expanding in powers of x and P, we have

$$U_i = x^2P(c_1\sigma + c_0P), \qquad (3)$$

where c_0 is the sum of two terms, one linear and

the other quadratic in σ, the internal elastic stress. A formula of this type is applicable to various types of domain energy (electrostriction, spontaneous electrification, and energy of the domain boundaries). Consequently, we obtain an analogous formula for the total energy, too. In the general case we should also take into account the types of energy which are proportional to other powers of P. In the first approximation it is enough, however, to restrict oneself to Eq. (3).

Taking into account the energy of the external field ($U_a = -2PEx$), we obtain for the equilibrium condition, $\partial (U_i + U_a)/\partial x = 0$, the following equation

$$E = x(c_0 P + c_1 \sigma). \tag{4}$$

Let n be the number of plane domains per cubic centimeter. We then obtain for the initial electric susceptibility

$$\varkappa_i = 2nxP/E. \tag{5}$$

From (4) and (5) we get

$$\varkappa = 2nc_0', \quad c_0' = P/(c_0 P + c_1\sigma), \tag{6}$$

i.e., the susceptibility of inversion at $\sigma \to 0$ is independent of the temperature. The situation is different with the susceptibility of the paraprocess. We consider first fields considerably weaker than the coercive force. We then obtain for the point E = 0

$$\varkappa_p = dP/dE = 1/2A. \tag{7}$$

Thus we have from (2), (6), and (7), for the total susceptibility in weak fields at $\nu = \mu = 1$,

$$\varkappa = \varkappa_0 + 1/2a(T - \Theta_1)(\Theta_2 - T), \tag{8}$$

where

$$\varkappa_0 = \varkappa_i + \varkappa_r.$$

According to (1), we have for the case of strong fields at $\nu = \mu$ (for $\Theta_1 < T < \Theta_2$ and $c = 0$)

$$\varkappa' = P/E = 2\sqrt{(T - \Theta_1)(\Theta_2 - T)}/E(\Theta_2 - \Theta_1), \tag{9}$$

where κ' is the susceptibility in the absence of rotation and inversion.

At the Curie points themselves, P depends strongly on E. Indeed, we get from (1) and (2)

$$P = (E/B)^{1/3}. \tag{10}$$

Supplementing (9), we readily find by the method of successive approximation the value of P for temperatures near Θ_1 and Θ_2. As a result we obtain a curve of type III (Fig. 2).

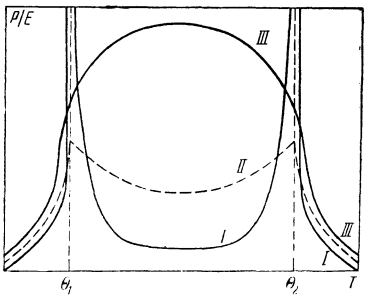

FIG. 2

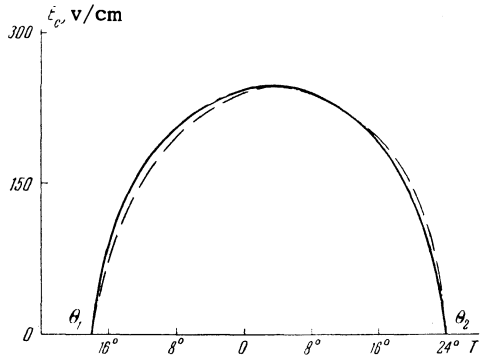

FIG. 3. Solid line– theoretical data; dotted–experimental data.

The types of susceptibility curves obtained in weak fields (I) and in strong ones (II) are also shown in Fig. 2. The experiment yields a family of curves of this type.

Let x_c be the critical boundary displacement corresponding to the Barkhausen jump. We then obtain for the coercive force, from (4),

$$E_c = c_0 x_c P, \tag{11}$$

i.e., according to (2) and (1), we have (for $\nu = \mu$)

$$E_c = 2E_{st}\sqrt{(T - \Theta_1)(\Theta_2 - T)}/(\Theta_2 - \Theta_1), \tag{12}$$

where E_{st} pertains to the point $T = (\Theta_1 + \Theta_2)/2$. As can be seen from Fig. 3, Eq. (4) is in good agreement with the experimental data of Bradford[3] (reference 4, p. 576). We assume that the discrepancy between the experimental data of various authors is due to insufficient stabilization of the temperature, and also to the presence of dielectric viscosity.

[1] N. S. Akulov, Z. Physik **81**, 790 (1933).

[2] W. P. Mason, Piezoelectric Crystals and their Application to Ultrasonics, (Russ. Transl.) IIL, 1952, p 221, Van Nostrand, N. Y., 1950.

[3] N. S. Akulov, Сб. Применение ультраакустики к исследованию вещества (Anthology, Application of Ultrasonics to Investigations of Matter) No. 2, p. 279, 1958.

[4] W. C. Cady, Piezoelectricity, (Russ. Transl.) IIL, 1950, p. 546, Fig. 137, McGraw-Hill, New York, 1946.

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