Photoelectric domains in para- and ferroelectrics

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(Submitted 18 December 1984)
Pis'ma Zh. Eksp. Teor. Fiz. **41**, No. 5, 180–182 (10 March 1985)

The formation of photoelectric domains similar to those which have been discovered in ruby is predicted in para- and ferroelectrics. It is shown on the basis of experimental data that this phenomenon can also occur in crystals of the KDP group.

It has recently been discovered[1] that laser bombardment causes a ruby crystal to break up into domains, with electric fields which alternate between opposite directions from domain to domain but which are equal in modulus. A theory for this phenomenon[2–4] is based on the assumption that a photovoltaic current $J(E)$ arises in a weak electric field E, that this current is directed opposite the field, and that it exceeds the conduction current σE in modulus. Figure 1a shows the field dependence of the resultant current, $j(E) = J(E) + \sigma E$, which follows from this assumption. For a functional dependence $j(E)$ of this sort, the resistance of the crystal in a zero electric field is negative, so that a domain instability occurs. The conclusions of this theory agree completely with the experimental results of Refs. 5–7. The reason for this functional dependence $J(E)$ has not yet been completely resolved, however.

0021-3640/85/050216-05$01.00

In this letter we work from experimental data to predict a photoelectric domain instability in high-resistivity para- and ferroelectrics (e.g., in crystals of the KDP group). The specific functional dependence $J(E)$ that causes this instability occurs because this current is proportional to the field-dependent electric polarization.

The basic assumption of Refs. 2–4 can be written

$$\left(\frac{dJ(E)}{dE}\right)_{E=0} < 0, \qquad \left|\left(\frac{dJ(E)}{dE}\right)_{E=0}\right| > \sigma. \tag{1}$$

We will show that these conditions hold in high-resistivity ferroelectrics. During bombardment, a photovoltaic current arises parallel to the C polar axis in the ferroelectric phase, and this current gives rise to an anomalous photovoltage if the sample is in an open circuit.[8] Since this current can arise because of the absence of an inversion center, it is proportional to the polarization $\mathbf{P}(\mathbf{E})$, which depends on the electric field. In various materials, under various bombardment conditions, this current will be directed equally frequently parallel to and antiparallel to the vector $\mathbf{P}(\mathbf{E})$ (Refs. 8 and 9). In the latter case, which holds in KDP, in particular, we can write $J(E) = -JP(E)/P_s$. Here the field E is assumed to be parallel to the C axis, P_s is the spontaneous polarization and J is the photovoltaic current in a single-domain ferroelectric with $E=0$ and $P=P_s$. The minus sign means that this current is directed antiparallel to the polarization $P(E)$ and thus antiparallel to the field. After incorporating this expression, condition (1) reduces to the inequality $J_\chi/P_s < \sigma$, where $\chi = (dP(E)/dE)_{E=0}$ is the electric susceptibility. For χ and J/σ we have the estimates $\chi \sim P_s/E_c$, $J/\sigma \sim E_s$, where E_c is the coercive field, and E_s is the saturation field in the anomalous photovoltage effect. These estimates reduce conditions (1) to the requirement $E_s > E_c$.

According to experimental data[8,10] on KDP, we have $E_s = 3\times 10^4$ V/cm and $E_c \approx 1.5\times 10^3$ V/cm, so that conditions (1) hold with a wide margin.

We now consider a ferroelectric in its paraelectric phase. For simplicity, we assume that the ferroelectric is a uniaxial crystal with a symmetry center. Without an electric field, a photovoltaic current would not be possible by virtue of symmetry considerations. In a field E parallel to the crystal axis C, however, a polarization $P(E)$ arises, as does a photovoltaic current $J(E)$, proportional to this polarization. If this current is directed opposite the polarization, it will also be directed opposite the field that causes the polarization. Condition (1) can thus also be satisfied in the paraelectric phase, if the medium has a sufficiently high resistivity.

Since the conductivity σ increases with the temperature T, the susceptibility χ is anomalously high at a temperature near the point of the phase transition, T_0, and the current is proportional to the bombardment intensity I; whether condition (1) is satisfied depends in a critical way on I and T. For KDP, this condition holds at a bombardment intensity 10^6 times lower than that for ruby.

The satisfaction of condition (1) means that the media under discussion here, like ruby, should have a negative resistance during bombardment, and this negative resistance will lead to an electrical instability. In the paraelectric phase the field dependence of the resultant current, $j(E)$, should be of the same form as in ruby (Fig. 1a). The theory of Refs. 2 and 3, which is based on this dependence, predicts the following:

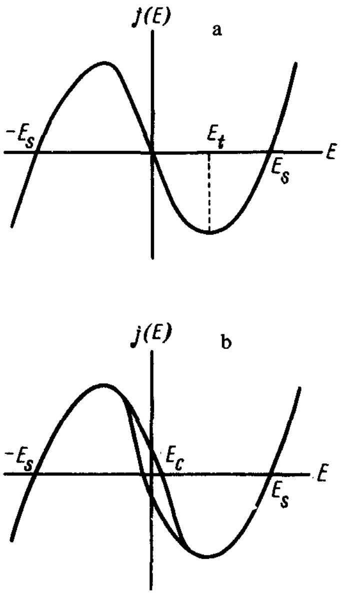

FIG. 1. Schematic diagram of the resultant current as a function of the field, $j(E)$. a—In ruby and paraelectrics; b—in ferroelectrics.

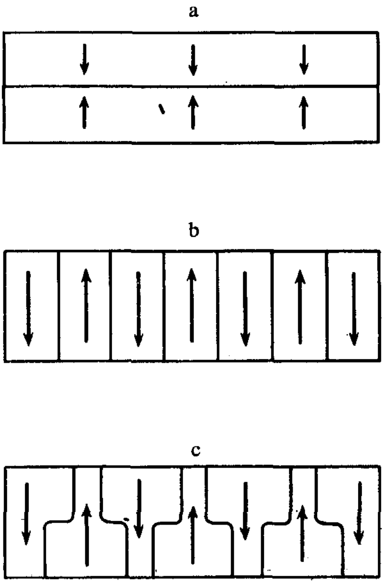

FIG. 2. Schematic diagram of the steady-state domain structure. a—Electric-field distribution during bombardment (photoelectric domains); b, c—*polarization distribution in ferroelectrics (ferroelectric domains)*; b—before bombardment; c—during bombardment in the case $E_s \gtrsim E_c$. The arrows show the directions of the field (a) or the polarization (b,c).

During bombardment in a short-circuited sample, cut in the form of a plate perpendicular to the C axis, the domain structure in Fig. 2a forms. The fields in the $\pm E_s$ domains are determined by the condition $E_s = J(E_s)/\sigma$ (Fig. 1a). After the bombardment, the domain structure dissipates over a time on the order of the Maxwellian relaxation time τ_M.

Figure 1b shows the dependence $j(E)$ in the ferroelectric phase. This dependence is determined by the hysteresis of the polarization, $P(E)$, which is characteristic of ferroelectrics. At $E_s \gg E_c$, the dependence $j(E)$ is similar to $j(E)$ in the paraelectric phase. During the bombardment of a sample with preexisting ferroelectric domains (Fig. 2b), the instability should then give rise to photodomains with fields $\pm E_s$, of the same type as in the paraelectric field. The ferroelectric domains should be destroyed. It can be seen from Fig. 2, a and b, that the photoelectric domain structure differs substantially from the ferroelectric domain structure.

At $E_s \gtrsim E_c$, the bombardment of a sample with ferroelectric domains may lead to the electric field distribution shown in Fig. 2a. Figure 2c shows a possible distribution of the polarization in this case: The ferroelectric domains are preserved, but their walls are deformed by the fields $\pm E_s$.

In some experiments[8,10] on KDP, the sample, already in an electric field $E_0 \sim E_c$, was bombarded at $T < T_0$. A current antiparallel to the field E_0 was observed. If the field E_0 exceeded the field E_t corresponding to the minimum on the $j(E)$ curve (Fig. 1a), there should be a homogeneous stable state with a current $J(E_0)$. If instead the condition $E_0 < E_t$ holds, the homogeneous state will be unstable, and a domain structure will form in a time $\tau \sim \tau_M$; the current will decrease to zero.[2,3] The oscilloscope traces of the current given in Refs. 8 and 9 were observed for too short a time, $\tau < \tau_M$, so that they do not reveal which of these two situations prevailed.

The collective interaction which gives rise to the ferroelectric phase transition is not important for the existence of the phenomenon described here. It can also occur in paraelectrics in which there is no such transition, e.g., dipole glasses. The orientational mechanism for polarizability is important. Since the photovoltaic current is related to the asymmetry of the elementary processes, it will be far lower in ordinary dielectrics, where this asymmetry results from a deformation of electron wave functions, than in paraelectrics, in which the field orients the dipole moments of the atomic order. The ratio of the currents in these two cases is determined by the small parameter $T/\mathscr{E}$, where T is the temperature in energy units, and $\mathscr{E}$ is the energy of the atomic order.

The general phenomenological features of the media in which photoelectric domains are possible can apparently be described as follows: These media may have an inversion center on the whole, but they contain polar elements, (ferroelectric domains in the case of a ferroelectric or impurities in various positions in the case of ruby). Groups of such elements differing in inversion give rise to large photovoltaic currents which cancel out exactly. In an electric field, this balance is disrupted, and a difference current arises along the direction of the field or opposite the field. In the latter case, the effect described here can also occur.

I wish to thank S. A. Basun, A. A. Kaplyanskiĭ, and S. P. Feofilov for useful discussions. I especially wish to thank M. I. D'yakonov for useful advice.

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Translated by Dave Parsons