Electron emission during the switching of ferroelectric lead germanate

G. I. Rozenman, V. A. Okhapkin, Yu. L. Chepelev, and V. Ya. Shur
Leninist Young Communist League Ural Forest-Technological Institute, Sverdlovsk

(Submitted 18 January 1984)
Pis'ma Zh. Eksp. Teor. Fiz. **39**, No. 9, 397–399 (10 May 1984)

A unipolar emission of electrons has been observed during 180° domain orientation reversal in ferroelectrics. An electron image of the surface has been visualized, making it possible to directly observe the motion of domain walls and changes in the domain structure during a switching process.

The 180° polarization reversal which occurs in a ferroelectric is a three-step process involving the formation of a nucleating region of a domain with polarization antiparallel to the original polarization, the growth of this nucleating region, and, finally, the lateral motion of the domain wall.[1] In all three of these steps there is a net charge $2P_s$, which exists for a time on the order of the Maxwellian relaxation time in the region undergoing the switching. This charge creates a strong electric field in a dielectric layer on the surface of the ferroelectric. This field was estimated in Ref. 2 to reach 3×10^6 V/cm, and it could cause a tunneling emission of electrons from the layer into a vacuum. The detection of this emission would be direct proof of the existence of a dielectric gap at the surface of a ferroelectric, and a visualization of the electron flux would make it possible to directly observe the domain-switching process and the motion of domain walls. Methods have been developed previously for directly observing domains in crystals with an optically resolvable domain structure[3] and in collinear ferroelectrics through the use of nematic liquid crystals.[4] The new method which we have developed, which makes use of the observed electron emission, is distinguished by its universal applicability.

For the study of polarization reversal we used single-domain single crystals of lead germanate with dimensions of $10\times 10\times 1$ mm. The electron emission was detected by a set of two microchannel secondary-electron multipliers, which provided a gain on the order of 10^8. A coordinate-sensitive detector of this type makes it possible to measure the integrated emission current density and to form from the emitted electrons a panoramic image of the crystal surface. The electron image is transferred from the surface of the sample to a fluorescent screen by means of the microchannel multipliers. The resulting optical image is photographed by a television camera, displayed on a monitor, and recorded on video tape. One of the electrodes to which the polarization-reversing voltage is applied is a SnO_2 conducting layer: the second is the surface of the microchannel multiplier pressed against the sample.

When a square polarization-reversing pulse is applied, electrons are emitted. The integrated emission current density reaches 10^{-7} A·m^2 at its maximum. It is important to note that this emission occurs only when the negative end of a domain is at the surface of the crystal. Since it is physically impossible for electrons to be emitted from the interior of the ferroelectric across the negatively charged surface, the observation

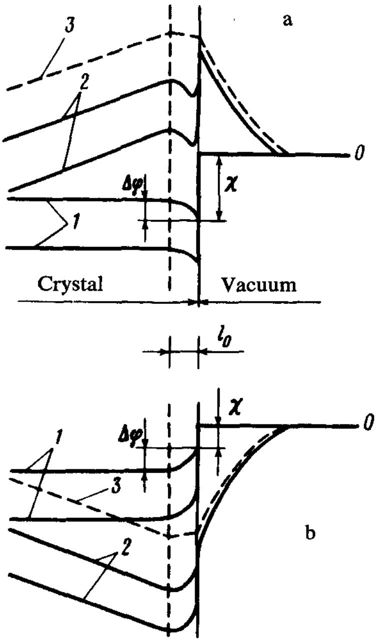

FIG. 1. Potential-energy diagram of an electron near the surface. a: z^+ section. b: z^- section. 1—Initial state of the band structure; 2—state of the band structure after polarization reversal; 3—potential of a free electron in the crystal and outside it after polarization reversal. Here $\Delta\phi$ is the ferroelectric band curvature, χ is the electron affinity, and l_0 is the thickness of the dielectric layer.

of this emission proves unambiguously that there is a layer at the surface of the ferroelectric which does not have ferroelectric properties. The existence of a dielectric gap of this type has been suggested previously.[5–7]

The mechanism for electron emission during polarization reversal of a ferroelectric crystal can be understood with the help of Figs. 1a and 1b, which are potential-energy diagrams of an electron near the surface. The dielectric layer l_0 is a space-charge layer that screens the spontaneous polarization.[8] For the z^+ section of the crystal, perpendicular to the positive direction of the polar axis, there is an energy barrier of height χ and infinite width in the initial state (Fig. 1a). At the time of the switching, an unscreened charge $2P_s$ arises; the potential of this charge alters the energy of an electron in the crystal and—the most important point—deforms the barrier in such a way that a tunneling from the layer l_0 into a vacuum becomes possible (Fig. 1a). The necessary electron density in the conduction band arises because of the field-ionization of impurity centers and the tunneling from the valence band in the field of the charges of the moving domain wall.[9,10] If there is instead a positive domain at the crystal surface, the barrier is much higher, but again infinite in width (Fig. 1b).

Figures 2a–2c show parts of an electron image of a crystal surface obtained by framing photography of the television screen. Just after the polarization-reversing pulse is applied, we see on the screen a bright region which apparently corresponds to a domain of the opposite phase that has formed. This bright spot then begins to

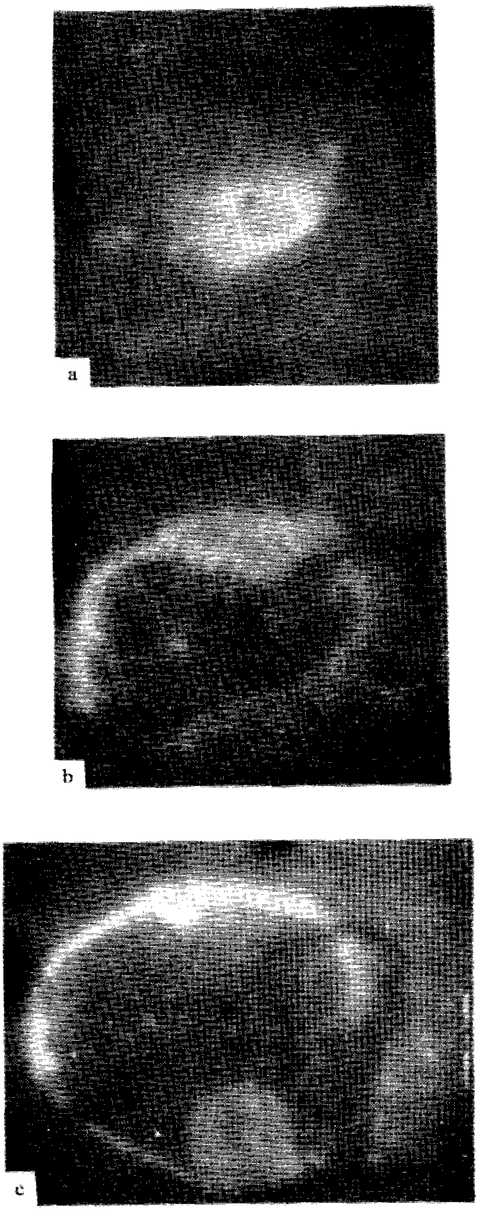

FIG. 2. Fragments of successive emission images of the surface of a crystal undergoing polarization reversal (20×). These photographs of the television screen were taken at six-frame of 0.12-s intervals.

increase in size. The brightness fades at the center of the pattern because of the screening, and the resulting ring expands toward the edges of the crystal (Fig. 2). The time required for the bright region to darken is on the order of 0.05 s, close to the Maxwellian relaxation time in these crystals. The observed growth of the bright ring evidently represents a lateral motion of the domain wall.

[1] G. A. Smolenskiĭ, V. A. Bokov, V. A. Isupov, N. N. Kraĭnik, R. E. Pasynkov, and M. S. Shur, Segnetoélektriki i antisegnetoélektriki (Ferroelectrics and Antiferroelectrics), Nauka, Leningrad, 1971.
[2] R. Miller and A. Savage, J. Appl. Phys. **31**, 662 (1960).
[3] F. Jona and G. Shirane, Ferroelectric Crystals, Pergamon Press, Oxford (1962) (Russ. transl. Mir, Moscow, 1965).
[4] N. A. Tikhomirova, S. A. Dontsova, L. A. Pikin, and L. A. Shuvalov, Pis'ma Zh. Eksp. Teor. Fiz. **29**, 37 (1979) [JETP Lett. **29**, 34 (1979)].
[5] W. Känzig, Phys. Rev. **98**, 549 (1955).
[6] W. Merz, J. Appl. Phys. **27**, 938 (1966).
[7] M. Drougard and R. Landauer, J. Appl. Phys. **30**, 1663 (1959).
[8] V. M. Fridkin, Segnetoélektriki-poluprovodniki (Ferroelectric Semiconductors), Nauka, Moscow, 1976.
[9] A. S. Sidorkin and V. N. Fedosov, Fiz. Tverd. Tela (Leningrad) **23**, 2854 (1981) [Sov. Phys. Solid State **23**, 1667 (1981)].
[10] A. S. Sidorkin, Fiz. Tverd. Tela (Leningrad) **24**, 1542 (1982) [Sov. Phys. Solid State **24**, 884 (1982)].

Translated by Dave Parsons
Edited by S. J. Amoretty