Study of the domain memory in ferroelectric crystals by a sound-generation method

G. P. Morozova, O. Yu. Serdobol'skaya, and N. A. Tikhomirova
M. V. Lomonosov State University, Moscow; Institute of Crystallography, Academy of Sciences of the USSR

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The generation of sound by domain walls in a ferroelectric crystal disappears well after the domain structure disappears as the crystal is put in a single-domain state by an electric field.

The application of an alternating electric field to a ferroelectric crystal can cause a system of periodic domain walls to effectively generate elastic waves if the period of the structure is equal to the wavelength. When an alternating field $E_z = E\sim e^{j\omega t}$ is applied to a trigonal ferroelectric crystal with a Z polar axis, in which a system of N plane domain walls running parallel to the zy plane has been produced, the change in the sign of the piezoelectric modulus e_{31} can cause neighboring domains to undergo contraction and expansion of different signs. The equation for the generation of a longitudinal wave along the X axis can be written

$$\frac{\partial^2}{\partial x^2} u_1 + k^2 u_1 = \frac{E^{\sim}}{c_{\parallel}} \frac{\partial}{\partial x} [e_{31}(x)],$$

where u_1 is the displacement amplitude, $c_{\parallel}$ is the elastic modulus, and k is the wave number. The excitation function on the right side of this equation is a sequence of δ-functions. Under optimum conditions, the coefficient of the conversion of electrical energy into elastic energy is $A = [e_{31}^2/(c_{\parallel}\epsilon_{33})]N$ $(N \gg 1)$. In the present letter we report a study of the kinetics of the disappearance of the sound generation during the decay of the domain structure in a lead germanate crystal. In this crystal, as was shown in Ref. 1, domain sound transducers with a conversion coefficient on the order of 30 dB ($N \sim 10$) can be produced.

A periodic domain structure is produced in a lead germanate sample, previously put in a single-domain state, by applying a static electric field E_z^0 to a system of electrodes shaped as stripes with a period equal to the sound wavelength λ. For efficient sound generation, the inhomogeneities of the domain walls must be significantly smaller than λ; the effect is to impose a limitation on the strength of the field E_z^0 and on the time interval over which is it applied, τ. For the excitation of sound, the periodic electrodes are replaced by solid electrodes of SnO_2 on glass; where necessary, a field E_z^M, to produce a single domain, can also be applied to these electrodes. Transparent electrodes make it possible to monitor the state of the domain structure in polarized light directly during the acoustic measurements.

The results show that while the sample is being put in a single-domain state, the sound generation disappears well after the domain structure disappears. The level of the sound generation from the domain "memory" and the temporal characteristics of this generation depend on both the structural relaxation conditions and the procedure by which the sample is put in a single-domain state. Figure 1 shows some typical time evolutions of the acoustic signal excited by the domain memory (beginning at the time at which the visible temporal characteristics of the acoustic signal for $E_z^{01} = 2.5$ kV/cm and $\tau_1 = 1$ s; the dashed lines correspond to $E_z^{02} = 8$ kV/cm and $\tau_2 = 10$ ms. These results can be described well by a function $u_1 \sim \exp[\alpha_i (E_m - E_t^m t$, where $E_t^m = 2$ kV/cm is the threshold field for conversion of the sample to a single domain. In

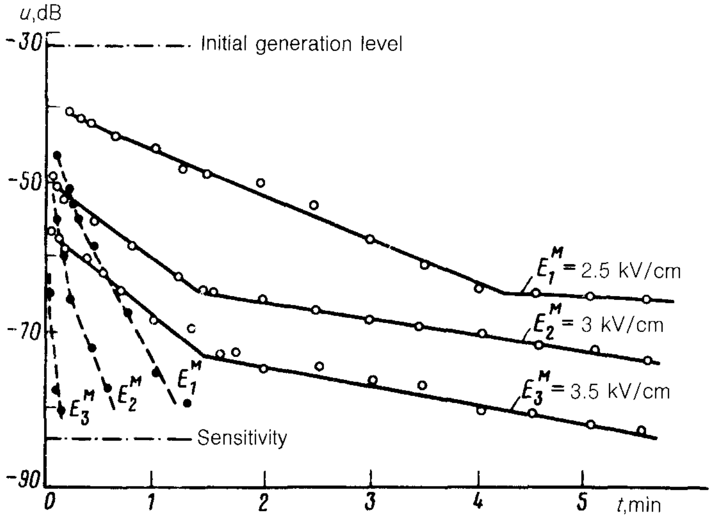

FIG. 1. Temporal characteristics of the sound generation by the domain "memory" for various strengths of the field which puts the sample in a single-domain state.

Fig. 1 we clearly see two relaxation times, corresponding to constants $\alpha_1' = 0.07$ cm/(kV·s) and $\alpha_1'' = 0.015$ cm/(kV·s) for the establishment field E_z^{01}. In the second case, the domain memory disappears much more rapidly $[\alpha_2 = 0.035$ cm/(kV·s) $(E_z^0 = E_z^{02})]$.

Can we explain these experimental results? In ferroelectric semiconductors one observes a screening of the spontaneous polarization which results from a capture of charge carriers by local active centers near the polar surface of the crystal, and possibly, near domain walls.[2,3] The so-called bound internal field which forms in the process has a strong effect on polarization reversal in the crystal, and it also stabilizes the domain structure in the absence of an external field. The strength and spatial distribution of this internal field are determined by the conditions under which the domain structure is formed, the temperature, and the defects in the sample.[2–4] In particular, if the relaxation time of the domain structure is shorter than the Maxwellian relaxation time τ^*, the internal field will not have time to form completely; this circumstance is apparently responsible for the difference between the domain-memory characteristics shown in Fig. 1. In the first case, the structural relaxation time satisfies $\tau_1 \gg \tau^*$, while in the second case it is $\tau_2 \sim \tau^*$, and the domain memory does not have time to form completely, although the establishing field is much stronger in the second case.

It is interesting to note the changes in the temperature characteristics of the domain memory up to the point of the phase transition (177 °C). Figure 2 shows the temperature dependence of the amplitude of the acoustic output signal generated by the domain walls. The generation level falls off sharply with increasing temperature near the phase transition, because of both the disappearance of the piezoelectric effect

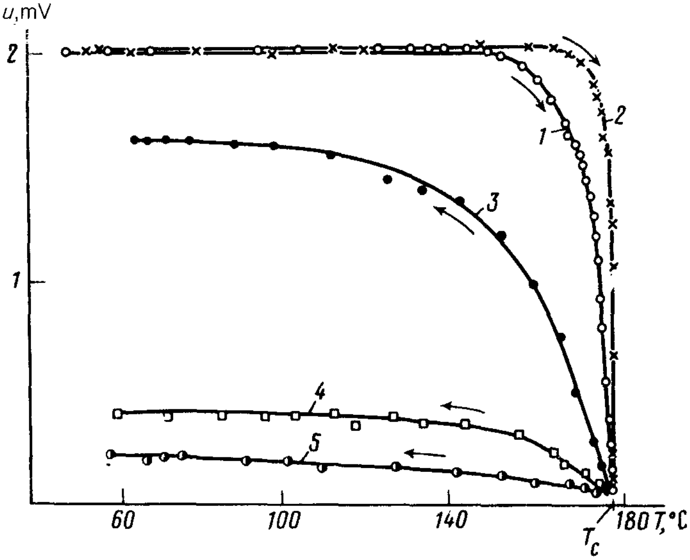

FIG. 2. Temperature dependence of the amplitude of the acoustic signals generated by domain walls (1,3–5) and by a plane face of the sample (2).

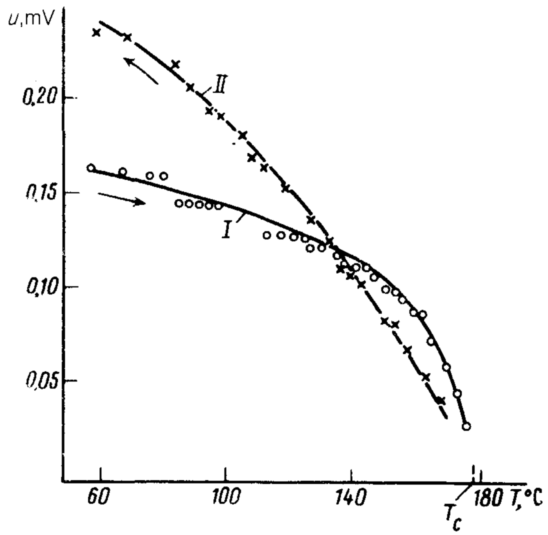

FIG. 3. Temperature dependence of the amplitude of the acoustic signal generated by the domain memory.

in the paraelectric phase and an increase in the attenuation of the sound near the transition, on the one hand, and a possible erosion of the domain structure, on the other. Shown for comparison, by curve 2, is the temperature dependence of the emission of sound by a plane *zy* face of the crystal. In this case the change in the signal near the transition results exclusively from the temperature dependence of e_{31}^2/ϵ_{33} and the attenuation of sound. During cooling, the regular domain structure is partially restored before the domain memory, and an acoustic signal appears. The level of this restored generation depends on the temperature reached during the heating and on the relaxation conditions for the domain structure (curves 3–5).

Figure 3 shows the temperature dependence of the sound generation by the domain memory. During heating (curve 1), the acoustic signal decreases more smoothly than during generation by the domain structure. This result shows that the generation of sound also decreases because of a dissipation of the memory with the temperature and an increase in the conductivity of the crystal. During cooling (curve 2) the generation level exceeds the original level of the generation by the domain memory, since the periodic domain structure is partially restored through the memory, as is observed visually.

Clearly, these results are evidence that the domain memory is localized not only near the surface but also in the interior of the crystal, near the positions of former domain walls. This circumstance is responsible for the generation of volume sound waves by the domain memory. Further evidence for the existence of a volume component of the memory may come from the fact that the memory and the sound generation do not disappear when a surface layer some hundreds of microns thick is removed. The memory forms over times much shorter than diffusion times and is apparently related exclusively to a redistribution of charges among local states, rather than to a motion of defects toward the surface and toward domain walls in the crystal.

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