Domain structure kinetics in ultrafast polarization switching in lead germanate

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Experimental observations have been made of a new mechanism of ferroelectric domain kinetics in ultrafast switching: this is the lateral motion of domain boundaries due to the formation of chains of new domains near a domain boundary and the existence of a delay time in nucleus formation.

The change in the direction of polarization in ferroelectrics under the action of an external electric field is a complicated process of rearrangement of the domain structure. Domains with the favorable polarization direction are formed and they increase in size. This phenomenon may be regarded as an example of the kinetics of growth of a phase during a phase transition.

There are many ways to render visible the slow kinetics of the domain structure of a ferroelectric,[1] but for ultrafast switching only optical methods are suitable. For a model crystal we chose the uniaxial ferroelectric lead germanate $Pb_5Ge_3O_{11}$, which has a comparatively simple domain structure that can be discerned in transmitted light.[2,3] In the phase transition at 450 K ($C_{3h} \rightarrow C_3$) the reflection plane in lead ger-

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manate is eliminated, and in the ferroelectric phase there exist only antiparallel domains separated by 180° walls. The samples we studied were plates about 0.5 mm thick, cut perpendicular to the polar axis. After careful mechanical treatment, transparent indium and tin oxide electrodes were deposited on them. With the use of short pulses of polarized light passing along the polar axis one can detect with good time and spatial resolution the instantaneous domain configurations directly during the switching.

The light source was an LGI-21 pulsed nitrogen laser, the radiation from which was converted into green light with the use of rhodamine 6G. The duration of the light pulse was less than 10 ns. The direction of the polarization in the sample was cyclically switched by alternating-sign rectangular voltage pulses. Under these conditions it was found that in each cycle the domain structure changed reproducibly, so that it was possible to use stroboscopic illumination.

It is well known that the kinetics of the domain structure in lead germanate

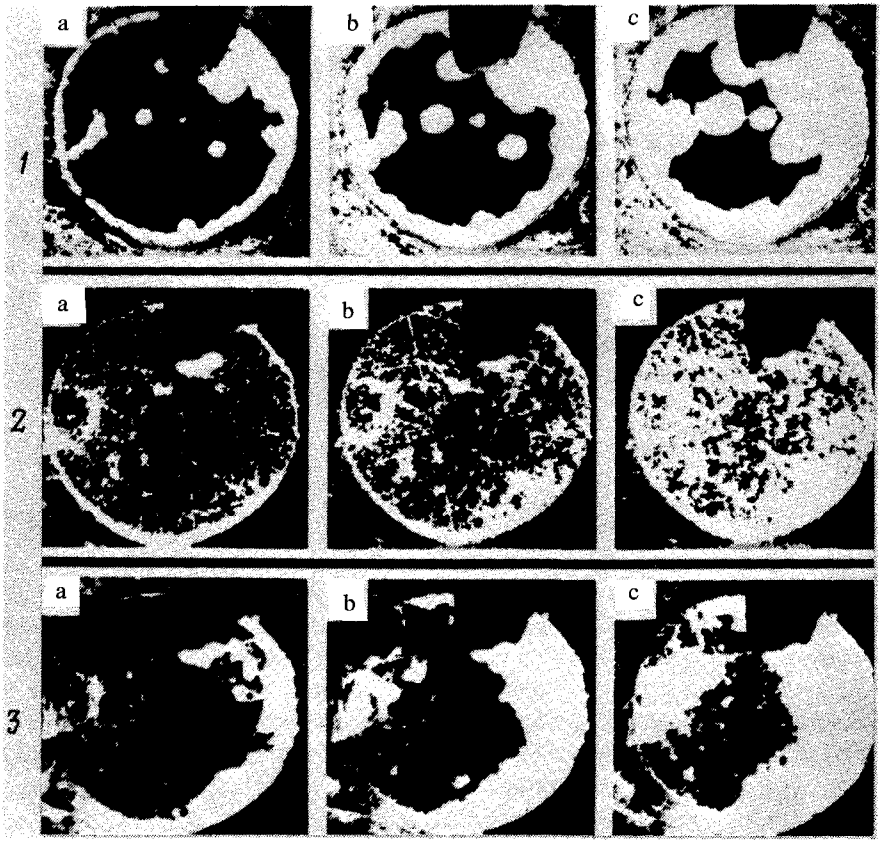

FIG. 1. Change in domain structure in various fields E, in units of 10^5 V/m: 1) 1.8; 2) 9.5; 3) 17.9. Time interval from the application of the field, in μs: (1) a) 2×10^4; b) 4×10^4; c) 7×10^4; (2) a) 160; b) 320; c) 480; (3) a) 6; b) 10; c) 18.

depends strongly on the electric field strength.[3] In weak fields (lower than 2×10^5 V/m) a few new domains of a regular hexagonal shape are formed. These domains grow via the lateral motion of the domain walls (Fig. 1.1). In strong fields (above 3×10^5 V/m) domains of small diameter, which pass through the sample, appear along the length of the entire sample surface, and are formed by the coalescence of formless domains (Fig. 1.2). The change in mechanism was accounted for by a change in the size of the nuclei that control the switching process.[4] It was assumed that in weak fields one-dimensional nuclei are formed at steps in the domain walls, and this process then leads to the layer-by-layer growth of the domains. In strong fields three dimensional nuclei are formed, and these form new domains, while two-dimensional nuclei form on the domain walls, leading to their isotropic growth. As the field strength increases, there is an increase in the number of domains that participate in the switching.

In ultrahigh fields (over 1.5×10^6 V/m) lateral motion of the domain boundaries is observed again, as it is in weak fields (Fig. 1.3). However, a detailed examination reveals that the mechanism for the motion of the domain walls in this case is qualitatively different. A large number of new through domains and domains that do not extend through the sample appear near the domain wall, forming a distinctive chain (Fig. 2). The domains that are formed expand and merge with an initial domain, after which the chain is formed again. This process is repeated many times.

Hence, the particular features of the ultrafast switching involves a decrease in the number of new domains that participate in the switching, and also involves their formation only in the form of chains along the domain boundaries. The first circumstance may be a consequence of a delay time—an interval between the time that the electric field is applied and the time a critical nucleus is formed.[5] It is clear that this delay increases with the size of the nuclei. Therefore, it is possible that the time of action of the field (the duration of the field pulse or the switching time) is less than the delay time for nuclei of a certain size, and so these nuclei simply will not be

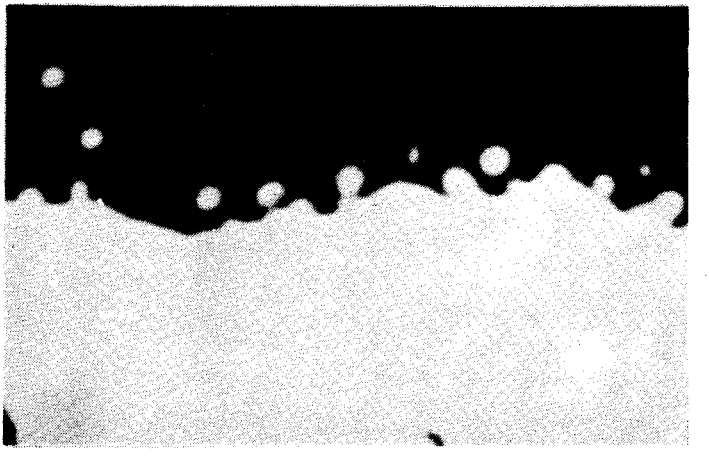

FIG. 2. Formation of domains near a moving domain wall in an ultrahigh field.

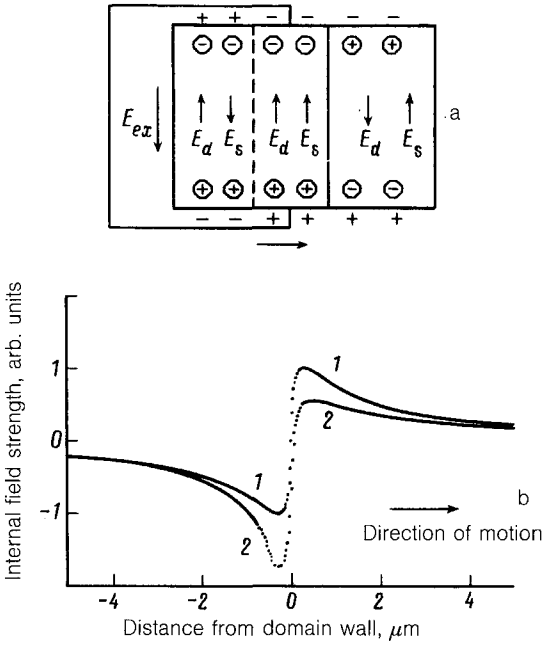

FIG. 3.(a) Distribution of charges and fields in a ferroelectric capacitor. (b) Calculation of the distribution of the internal field near a moving domain wall. Wall velocity, m/s: 1) 0.1; 2) 5.

formed. Evidently this condition is satisfied for the three-dimensional nuclei in ultrahigh fields.

But why do the new domains form near a domain wall? To answer this question, one must recall that the probability of nucleus formation is governed by the local internal field strength. Therefore, it is necessary to calculate the spatial distribution of the internal field in a ferroelectric capacitor near a domain wall (Fig. 3a). The field is determined not only by the difference in potentials between the electrodes, but also by the sum of the depolarizing field E_d and the screening field E_s (Ref. 6). Here it should be taken into consideration that a so-called dielectric gap exists near the surface of a ferroelectric.[7]

The results of the calculation of the spatial distribution of the internal field is shown in Fig. 3b. It can be seen that near a moving domain wall the depolarizing field is not completely screened, and the internal field has a maximum. This behavior also explains the phenomenon of the correlation of the nucleation and the resulting appearance of chains of domains in an ultrahigh field.

It should be noted that this analysis is wholly applicable not only to other ferroelectrics, but also to the kinetics of phase transitions in other materials. For example, it has been observed experimentally that in sufficiently strong pulsed electric fields in some magnetic films microdomains are nucleated ahead of the moving domain wall.[8] One of the explanations of this phenomenon is based on the effect of the fringing field near the domain wall.[9]

In summary, the qualitatively new mechanism of domain structure kinetics involved in ultrafast switching of the polarization may be explained by the particular

features of the spatial distribution of the internal fields and the fact that a finite interval of time is required for the formation of three-dimensional domains.

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