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V. Ya. Shur, A. L. Gruverman, V. P. Kuminov and N. A. Tonkachyova

Ural State University, Sverdlovsk, 620083, USSR

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DYNAMICS OF PLANE DOMAIN WALLS IN LEAD GERMANATE AND GADOLINIUM MOLYBDATE

V. YA. SHUR, A. L. GRUVERMAN, V. P. KUMINOV
and N. A. TONKACHYOVA

Ural State University, 620083 Sverdlovsk, USSR

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With the help of original methods the peculiarities of sidewise motion of plane domain walls in ferroelectric-semiconductor lead germanate and improper ferroelectric-ferroelastic gadolinium molybdate are investigated. It has been found that the behaviour of domain walls in these materials are qualitatively similar. The observed peculiarities of sidewise motion are explained in terms of external and bulk screening of depolarization field in shortcircuit ferroelectric capacitor with surface dielectric layers. It is supposed that domain wall velocity is determined by the internal electric field strength on the wall. The results of calculation are in good agreement with experimental data. It is assumed that proposed model is applicable to other ferroelectrics.

INTRODUCTION

It is known that optical methods of studying of domain structure dynamics are most effective and informative. These methods provide high time and space resolution but are applicable for the investigation of crystals with optically distinguished domains only. Among such crystals with quite simple 180° domain structure lead germanate (LG) PbGe₃O₁₁ and gadolinium molybdate (GMO) Gd₂(MoO₄)₃ are the most well studied.

The detail studying of domain wall motion in constant electric field in LG and GMO shows that there are some peculiarities of sidewise motion observed for domains of any shape: the irregularity of the motion and spontaneous return of domain structure to the initial state after the external field is switched off. It must be noted that the character of showing of this peculiarities depends upon the prehistory of the sample, the switching field strength, temperature and the value of the domain wall shift from initial position. In order to carry out the detail investigation of this effects, to improve measurement precision and to simplify the calculation in the present work the motion of plane domain walls has been studied.

Experimental Method

The LG samples with carefully polished surfaces covered by the transparent electrodes of Sn and In oxides were used. Due to the high quality of sample surfaces the process of arising of new domains during the switching was suppressed in comparatively wide field region. Before the measurement the strip-like domain of 50–100 μm width and about 1–2 mm long was formed by electric field in the LG sample.

In the rectangular plate of GMO also covered by the same transparent electrodes
the bidomain structure with single plane domain wall parallel to the shorter plate side was formed by mechanical stress. Before the carrying out of the measurements the domain wall was held for a few hours in the middle part of the sample.

The polarization in both the samples was switched by the applying of square alternating-sign pulses of electric field (front duration 0.2 μs). In order to observe the domain structure during polarization reversal stroboscope or pulse enlightenment was used (light pulse duration is less than 1 μs). The measurement accuracy of domain wall position was about 1 μm.

Due to the single plane domain wall in GMO sample it was possible to measure the relative wall velocity by the registering of switching current with the help of memory oscillograph. The value of switching current was proportional to the domain wall velocity, because the length of domain wall during its motion was constant.

The measurements show that domain wall motion under the electric field in LG and GMO is quite nonuniform (Figure 1). It has been found out that at once after the applying of field pulse the wall velocity reaches its maximum value, then it decreases up to constant value. In weak fields the wall stops at some distance from initial position. After the field pulse is switched off the wall spontaneously returns at the initial position. The backward motion is also nonuniform.

The Influence of Bulk Screening

To explain the observed peculiarities let’s consider the more common problems of the motion of phase boundary. The domain walls dynamics is a special case of this problem. It is known that the growth velocity of new phase is determined by the oversaturation degree on the phase boundary. In our case the oversaturation degree corresponds to the electric field strength, so the velocity of domain wall motion is determined (at the given walls orientation and crystal inhomogeneity) by the magnitude of electric field on the boundary of growing domain (plane domain wall).

Let’s consider the investigated samples as the ferroelectric capacitors of thickness d with surface dielectric layers (dielectric gaps) of thickness L and dielectric permittivity \( \varepsilon_L \). At the polarization reversal the depolarization field, produced by the bound charges in the sample bulk, quickly decreases due to the current in the external circuit changing the charges on the electrodes. This process we shall name as “external screening.” The screening velocity depends upon the time constant of external circuit RC. Due to the presence of surface dielectric layers in the sample bulk after the finishing of external screening nevertheless exists residual depolarization field:

\[
E_{dr} = 2LP_r/\varepsilon_L \varepsilon_0 d
\]

Rather long waiting at constant conditions leads to the “bulk screening” of depolarization field as a result of redistribution of carriers and formation of bulk charges bound to traps.

Let’s consider the ferroelectric plate cutted normal to the polar axis and covered by the metal electrodes. Let this plate contain two domains, separated by the plane (parallel to Y axis) noncharged 180° domain wall which can move in the middle part of the sample (end effects don’t take into account) (Figure 2). The sample is
FIGURE 1  The time dependences of domain wall shift from initial position at different external fields: a—in LG; b—in GMO.

held in this state for a long time which is enough for completion of bulk and external screening processes. As a result forming in the bulk sample the screening field fully compensates the residual depolarization field: \( E_0 = E_s + E_{dr} = 0 \). The screening bulk charges can be considered as charged planes localized at the distance from the surfaces equal to the \( L_r \)-effective screening lengths (Figure 2). It must be noted, that due to the symmetry of the problem in the infinitive bidomain plate at the complete screening on the domain wall \( E_{dr} = 0 \) and \( E_s = 0 \).

While considering the wall shifting we take into account that time constant of external circuit \( \tau_{cs} \) in our experiment is less than \( 10^{-5} \) s. So we can state that the
FIGURE 2  The schematic drawing, illustrating the process of external and bulk screening of depolarization field during the domain wall shift: $a - t = 0; b - \tau_{\text{es}} < t < \tau_{\text{bs}}; c - t > \tau_{\text{bs}}$.

External screening is finished immediately after the wall has stopped. On the other hand measuring time constant of the bulk screening $\tau_{\text{bs}}$ for the crystals, used in our work, are much more than switching time and field pulse duration (at $T = 300$ K $\tau_{\text{bs}}$ is equal to $10^2$ s in LG and to $10^4$ s in GMO). So the distribution of screening bulk charges during the wall shifting is not changed.

In this case the internal field on domain boundary $E_{ib}$ depends upon the value
of wall shift $X$ from equilibrium position. It is easy to show that field produced on domain boundary by the pair of half-planes of screening bulk charges is equal to the field, produced by the pair of charged strips of width $X$ with surface charge density $2P$,:

$$E_s = E_{dr} * F(X/2L_s)$$  \hspace{1cm} (2)

where $F(A) = \frac{1}{\pi} [2\arctg A + A \ln(1 + 1/A^2)]$.

It is clear that at $X = 0$: $E_s(0) = 0$, and at $X >> L_s$: $E_s(X) = E_{dr}$.

It is obvious that long (comparative with the time of bulk screening) standing of domain wall in shifting position or multiple reversal in alternating electric field (forming) must lead to the change of $E_s(X)$. This can be accounted by inventing the reduction factor $k$ ($-1 < k < 1$), which characterizes the screening degree.

Thus, while applying electric field $E_{ex} = Ud$ the internal field at the domain wall is defined by the following way:

$$E_{ib}(X) = E_{ex} + kE_{dr} * F(X/2L_s)$$  \hspace{1cm} (3)

It is known that field dependence of wall velocity in weak field for LG and GMO is as follows:

$$V = \mu(E_{ex} - E_{st}) = \mu(E_{ib} - E_{th})$$  \hspace{1cm} (4)

where $\mu$-domain wall mobility; $E_{th}$-threshold field.

Then, domain wall moving in electric field stops on condition that

$$E_{ib}(XM) - E_{th} = 0$$  \hspace{1cm} (5)

where $XM$-maximum shift of domain wall from equilibrium position.

The field dependence of $XM$ can be obtained from equation:

$$E_{ex} + kE_{dr} * F(XM/2L_s) - E_{th} = 0$$  \hspace{1cm} (6)

Experimental data for the field dependence of maximum domain wall shift for LG and GMO and theoretical results obtained by the minimizing on two parameters: screening length $L_s$ and reduction factor $k$ (the other parameters are defined from independent experiments) are compared on Figure 3.

The Field Dependence of Velocity of Sidewise Domain Wall Motion

Following to proposed model at the measuring of field dependence of sidewise wall velocity we must take into account that internal field at the domain wall $E_{ib} = E_{ex} = Ud$ only at $X = 0$. But making of such measurement is impossible because in order to define sidewise velocity we must measure the time of wall shifting of finite value. The first decades of microns the domain wall moves in changeable field and it is difficult to interpret the experiment data of “initial” velocity.

The most accuracy is the measuring of terminal (independent on the wall position) velocity $V$, when the wall shift is essentially more than $L_s$. At such shift ($X >> L_s$) in completely screening sample ($k = 1$) $E_{ib} = E_{ex} - E_{dr}$.

It must be noted that this consideration is correct only for stopping domain wall
after the finishing of external screening. For moving wall it must be taken into account the existence of depolarization field which is not fully compensated by the external screening. This field can be considered as a train behind the wall. The width of the train $\Delta X_{tr}$ is determined by the velocity of domain wall motion and by the time constant of external screening: $\Delta X_{tr} = V^* RC$. Then in first-order approximation the depolarization field on the wall which decreases the wall velocity and arising as a result of wall motion can be defined by the following expression:

$$E_{tr}(V) = P_s/\varepsilon \varepsilon_0 F(V RC/L)$$

(7)

Thus, the expression for internal field, determined the motion of plane domain wall is as follows:

$$E_{ib}(X) = E_{ex} + E_s(X) - E_{tr}(V)$$

(8)

The field dependence of terminal wall velocity is described by the law:

$$V_t = \mu [E_{ex} - E_{dr} - P_s/\varepsilon \varepsilon_0 F(V RC/L) - E_{ib}]$$

(9)

From the best fit of this relation with experimental data the values of mobilities and threshold fields for LG and GMO are obtained (Table I). The other parameters are determined from independent experiments.

The time dependence of domain wall shift from equilibrium position calculated

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**TABLE I**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Sample thickness $d$, mm</th>
<th>Surface layer thickness $L$, $\mu$m</th>
<th>Screening length $L_s$, $\mu$m</th>
<th>Threshold field $E_{th}$, $10^5$ V/m</th>
<th>Mobility $\mu$, m$^2$/V·s</th>
</tr>
</thead>
<tbody>
<tr>
<td>LG</td>
<td>1.50</td>
<td>0.15</td>
<td>31.0</td>
<td>1.05</td>
<td>4.5 x 10$^6$</td>
</tr>
<tr>
<td>GMO</td>
<td>0.47</td>
<td>1.60</td>
<td>11.2</td>
<td>1.34</td>
<td>2.2 x 10$^6$</td>
</tr>
<tr>
<td>GMO</td>
<td>1.73</td>
<td>2.85</td>
<td>14.2</td>
<td>1.39</td>
<td>2.1 x 10$^6$</td>
</tr>
</tbody>
</table>

---

**FIGURE 3** The field dependences of maximum shift of plane domain wall from initial position in LG and in GMO. Solid curves—results of calculation.
Spontaneous Backward Motion of Domain Walls

According to proposed model after the external field is switched off the sign of internal field on domain wall is changed and the spontaneous backward switching must be observed.

FIGURE 4  The comparison of experimental data on the time dependences of domain wall shift from initial position and result of calculation (dotted line): a—in LG; b—in GMO.

from obtained expressions are in good agreement with experimental data (Figure 4).
In this case it is possible to study the wall motion at different time constants of external circuit. It is seen that velocity of backward motion in GMO quickly reaches the constant value and only while approaching to the initial position its monotone decreases up to zero (Figure 5). In order to explain the peculiarities of backward motion let's define the correlation between the reversal wall velocity $V_r$ and the resistance of external circuit $R$.

The change of charge density on the covers of ferroelectric capacitor during the switching is as follows:

$$\frac{d\sigma}{d\tau} = \frac{dP}{dt} - j_{ex} \quad (10)$$

At the motion of single plane domain wall we have:

$$\frac{dP}{dt} = 2bP, V_r = 2\mu bP_e(E_s - E_{r t} - E_{th}) \quad (11)$$

where $b$ — the wall length, equal to the width of the sample.

The current in external circuit is described by the expression:

$$j_{ex} = \Delta U/R = E_{r t} d/R \quad (12)$$

For the terminal wall motion $d\sigma/dt = 0$ and hence:

$$E_{rt} = (1 + d/2\mu bP_e R)^{-1}*(E_s - E_{th}) \quad (13)$$

Then let's define the dependence of terminal velocity of spontaneous backward motion upon the resistance of external circuit:

$$V_{rt} = \mu_0 (1 + BR)^{-1}*(E_s - E_{th}) \quad (14)$$

where $B = 2 \mu_0 bP_e /d; \mu_0$ — the domain wall mobility at $RC << t_s$.

There is an agreement of theoretical dependence and experimental data in LG and GMO (Figure 6). The obtained from regression analysis value of constant $B$ is equal to the value obtained from independently determined parameters (Table I).

FIGURE 5. The switching current during the spontaneous backward motion of plane domain wall in GMO at different resistance of external circuit (base time 200 \(\mu\)sec/sm). $R$, Ohm: $a$ — $6 \times 10^6; b$ — $15 \times 10^6$. 
FIGURE 6  The dependence of terminal velocity of spontaneous backward wall motion upon the resistance of external circuit: a—in LG; b—in GMO.

CONCLUSION

Thus, on example of dynamics of plane domain wall it is shown that taking into account the existence of dielectric gap and bulk screening of residual depolarization field makes it possible to explain the peculiarities of sidewise domain wall motion
in ferroelectrics and improper ferroelectric-ferroelastics. It was obtained a good agreement of calculation with experimental results on domain wall motion under applied external field and during the spontaneous backward switching. The same calculations may be provided for uncharged 180° domain walls of other shape. This model is applicable to another ferroelectrics with taking into account the values of bulk conductivity, spontaneous polarization, dielectric permittivity and parameters of dielectric gap.

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