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Ferroelectric domain reversal: The role of domain wall conduction[1]

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Ferroelectric domain reversal is a vast research area relevant to the fundamental science and applications. Here, the general feature is that the coercive field E_c is orders of magnitude smaller than the characteristic depolarizing field $E_{\rm d}^0 = 4\pi P_s/\varepsilon_{zz}$, where P_s is the spontaneous polarization. The real reversal process is viewed as nucleation and growth of numerous microscopic counter-domains [1, 2]. While compensation of the arising bound charge $\pm 2P_s$ occurs at electrodes, it is not generally allowed at domain walls (DWs) inside the crystal. This leads to the generation of depolarizing field $E_{\rm d}$ ranging from 0 to $E_{\rm d}^0$, i.e., to an apparent inconsistency of the reversal concept. To overcome it, counter-domains are assumed to be needle-like [1–4]. This assumption is satisfactory only for an initial stage of the reversal. Moreover, there are documented cases [5–7] where $E_d \gg E_c$ and the reversal concept not including the charge compensation experiences serious difficulties. This is relevant to both capacitor and AFM experimental configurations.

We claim that the DW conduction, which is now detected in many ferroelectrics [8–10], has to be regarded as a crucial and general ingredient of the domain reversal processes. Its importance is in providing an automatic compensation of typically huge depolarizing electric fields. The presence of DW conduction modifies the basics of domain reversal processes. Concerning AFM applications, domain reversal theories have to include injection models from conductive tip electrodes. We provide some primary results relevant to the basics of DW conduction mediated domain reversal. For simplicity, we consider uniaxial ferroelectrics where the spontaneous polarization is parallel to the z axis and acquires the values $\pm P_s$.

The values of domain formation energy $\delta\mathcal{E}$ are crucial for domain reversal [1–4]. The main contributions to $\delta\mathcal{E}$ are the surface and electrostatic ones. The surface contribution is given by the integral over the DW surface, $\delta\mathcal{E}_s = \int w\, dS$, where w is a positive surface density. As DW is typically charged, w must depend on the angle θ between the DW surface normal and the z axis. We model this by the relation $w = w_0 + w_1 \cos\theta$ with $w_1/w_0 \gg 1$ leading to $\delta\mathcal{E}_s = w_0 S + w_1 S_\perp$, where S is the domain surface and $S_\perp$ its maximal cross-section.

The electrostatic contribution $\delta\mathcal{E}_{\rm el}$ crucially depends on the charge compensation assumptions. In the absence of DW charge compensation, we obtain the classical relation of [4] leading to unrealistically large values of $\delta\mathcal{E}$ and thus to practically forbidden reversal process. Admission for DW conduction means that the dielectric boundary conditions (BCs) must be replaced by the metal BCs for the electrostatic potential, $\varphi(\mathbf{r}_{\rm DW}) = U$, where U is the applied voltage. The actual values of $\delta\mathcal{E}_{\rm el}$ can be substantially smaller here facilitating the domain formation.

Figures 1a, b illustrate the dependence of $\delta\mathcal{E}$ on the applied electric field E_0 in the capacitor configuration and on the transverse and longitudinal domain sizes ($l_\perp$ and l_z) for a half-spheroidal domain shape. We have employed parameters relevant to lithium niobate (LN) crystals: $P_s = 70\,\mu{\rm C/cm}^2$, $\varepsilon_{zz} = 30$, $\varepsilon_\perp = 85$ and representative values $w_0 = 3$, $w_1 = 15\,{\rm erg/cm}^2$. One sees from Fig. 1a that for $E_0 = 4\,{\rm kV/mm}$, which is representative for E_c in LN crystals, the maximal in l_z values of $\delta\mathcal{E}$ are about 1 eV. Figure 1b shows that increase of E_0 causes a rapid decrease of the values of l_z and $\delta\mathcal{E}(l_z)$ relevant to the maximum of $\delta\mathcal{E}(l_z)$. The predictions of Figure 1 are beneficial for the domain reversal as compared to those relevant to the absence of the DW charge compensation.

Consider now the effect of DW conduction charge compensation in the case of lateral domain growth in the AFM configurations. Experiments with application of U, τ voltage pules show that the inverted domain radius $r_0(U,\tau)$ exceeds $1\,\mu$m in LN crystals for $U \approx 100\,$V and $\tau \approx 10^3$s [6, 7]. This is much larger than the conductive tip radius. In the absence of charge compensation, this would lead to the existence of depolarizing fields

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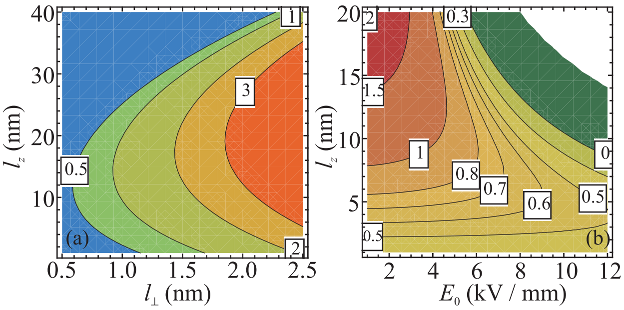

Fig. 1. (Color online) The domain formation energy $\delta\mathcal{E}$ (in eV) versus E_0, $l_\perp$, and l_z in the presence of DW conduction for the LN parameters, $w_0 = 3$ erg/cm^2 and $w_1 = 15$ erg/cm^2. (a) – Contour lines $\delta\mathcal{E}(l_\perp, l_z) = \mathrm{const}$ for $E_0 = 4\,\mathrm{kV/mm}$. (b) – Contour lines $\delta\mathcal{E}(E_0, l_z) = \mathrm{const}$ for $l_\perp = 1\,\mathrm{nm}$

$E_\mathrm{d} \approx 3$ MV/mm in non-electroded area exceeding the coercive field E_c by about three orders of magnitude. The charge compensation can indeed be explained by the presence of free surface charges injected from the tip, such that $\varphi(r) = U$ for $r \le r_0$ and the field component $E_z(r)$ beneath the front surface acquires the sign necessary for the domain reversal.

However, such an extended electrode model is insufficient to explain the data accumulated in the AFM experiments on the lateral domain expansion [6, 7]. The point is that the the data on $r_0(U, \tau)$ can be nicely fitted using the empirical Merz law $v \propto \exp[-\mathrm{const}/E_z(r_0)]$ for the lateral expansion velocity and, additionally, the assumption $E_z(r_0) \propto 1/r_0$. The latter has no visible physical grounds. Moreover, within the extended electrode model, i.e., for metal disk of radius r_0, the component $E_z(r) \to \infty$ for $r \to r_0$ [11].

We have realized that the presence of such an edge singularity corresponds to infinite gradients of free charge concentration at $r = r_0$. Account for diffusion of free carriers is necessary here. It is shown unambiguously that this leads just to the necessary dependence $E_z(r_0) \propto 1/r_0$. The data on the lateral domain expansion acquire thus qualitative and quantitative explanations.

In conclusion, physical models providing a strong charge compensation during the ferroelectric domain reversal, especially for AFM configurations, are crucial for development of the domain engineering. Domain wall conduction can be regarded as a general ingredient for such a compensation and for the explanation of numerous accumulated experimental data. Particular models are presented to demonstrate the positive impact of this conduction on nucleation and growth of ferroelectric counter-domains. The prospects for further development of DW conduction related models of the domain reversal are outlined.

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