Comment on "Abrupt Appearance of the Domain Pattern and Fatigue of Thin Ferroelectric Films"

In their paper [1] Bratkovsky and Levanyuk have revisited an old problem [2–5] of the energy of a lamellar 180° ferroelectric domain structure in a plane capacitor with layers of linear dielectric ("passive" layers) between the ferroelectric and the electrodes. As a central result the authors have presented an expression for small signal dielectric susceptibility of the structure, $\varepsilon_{\rm eff}$, also obtained in Ref. [3]. This expression can be written as

$$\varepsilon_{\rm eff} = \varepsilon_g \frac{L}{d}, \tag{1}$$

where L, d, and ε_g are the thickness of the sandwich structure, the total thickness of the dielectric, and its dielectric susceptibility, respectively. Based on this equation the authors arrive at the principal conclusion that a growing passive layer might be the main reason for polarization fatigue in ferroelectric thin films. In this Comment I criticize the following points. First, the authors have overlooked a simple physical interpretation of Eq. (1) and ignored the knowledge developed in the field. Second, the calculations performed in Ref. [1] do not justify the conclusion concerning the aforementioned link between fatigue and the passive layer.

(1) Equation (1) can easily be obtained by considering the sandwich structure as a series connection of a ferroelectric and a linear dielectric capacitor. Using the formula for the capacitance of two in-series connected capacitors one readily finds

$$\frac{L}{\varepsilon_{\rm eff}} = \frac{L-d}{\varepsilon_f} + \frac{d}{\varepsilon_g}, \tag{2}$$

where ε_f is the dielectric permittivity of the ferroelectric. In the model treated by Bratkovsky and Levanyuk, the dielectric susceptibility of the ferroelectric is infinity as being controlled by the contribution of unpinned domain walls. Thus, setting in Eq. (2) $\varepsilon_f = \infty$ one arrives at Eq. (1), which exactly corresponds to the capacitance of the sandwich equal to that of the passive layer. Treating the problem this way, one can explain why the dielectric response of the sandwich structure, calculated in Ref. [1], is independent of the values of spontaneous polarization of the ferroelectric, which actually plays the role of the "coupling constant" between the domain structure of ferroelectrics and the field. The point is that, being identically equal to zero, the restoring force of nonpinned domain walls provides an infinite dielectric response of the ferroelectric capacitor for any value of the coupling constant.

(2) The modification of a hysteresis loop caused by introducing a passive layer or changing its thickness has been treated earlier [6,7] using the above approach of in-series capacitors. In these papers, it has been shown that the principle manifestation of the passive layer is a tilt of the loops. Bratkovsky and Levanyuk [1] claim that their model leads to the same conclusion. However, this claim has no grounds since the equilibrium approach used in Ref. [1] is inapplicable for the description of nonequilibrium hysteresis phenomena. Thus, the results obtained in this paper have no implication on the hysteresis behavior of ferroelectric thin films.

(3) Bratkovsky and Levanyuk suggest that the tilt of polarization loops, which usually accompanies polarization fatigue, attests to the growing passive layer scenario. This possibility has already been critically analyzed [8]. It has been shown that the growing passive layer model, though able to describe the progressive loop tilt during the fatigue cycling, at least for $\mathrm{Pt/Pb(Zr,Ti)O_3/Pt}$ capacitors studied in Ref. [8], cannot satisfactorily describe the evolution of other parameters of the loop. Another problem is that the progressive loop tilt can originate not only from the growing passive layer, but just a blocking of ferroelectric switching under a growing fraction of the electrode area also results in a loop tilt [9]. As has been shown in Ref. [9], it is a comprehensive analysis of the evolution of several parameters of the loop that enables identification of the latter scenario, whereas the information contained in the loop tilt only does not suffice to distinguish between the two scenarios. All that does not mean that a growing passive layer should be excluded from possible fatigue scenarios; however, the tilt loop analysis mentioned by Bratkovsky and Levanyuk cannot be treated as a reliable one for identification of the origin of ferroelectric fatigue.

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