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Estimation of film-electrode contact resistance and domain switching time from ferroelectric polarization-voltage hysteresis loops

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ABSTRACT

The large resistance of bottom oxide electrodes in epitaxial/polycrystalline ferroelectric thin films and the existence of interfacial passive layers between the film and top/bottom electrode can affect domain switching speed as well as coercive-voltage estimation from polarization-voltage (*P-V*) hysteresis loops. With the measurements of *P-V* hysteresis loops at different frequencies, we estimated the intrinsic ferroelectric coercive voltage as well as the contact resistance either in epitaxial Au/Pb(Zr,Ti)O_3/Fe_2O_3 thin-film capacitors with the poor conductance of Fe_2O_3 bottom electrodes or in polycrystalline Pt/Al_2O_3/Pb(Zr,Ti)O_3/Ir capacitors with random grain orientations where the ultrathin Al_2O_3 amorphous layer can mimic the interfacial-layer behavior. The averaged domain switching times at different frequencies were also obtained in both films from *P-V* hysteresis loops without the assistance of traditional square pulse characterization.

1. Introduction

Ferroelectric control of the spin polarization has been reported to occur at interfaces between ultrathin ferroelectrics and magnetic semiconductors, such as Fe, Co_2MnSi, or Fe_3O_4 with epitaxial growth [1–4]. These ferroelectric tunnel junctions with ferromagnetic electrodes below a few tens of nanometers demonstrate local large and non-volatile control of carrier spin polarization by electrically switching of ferroelectric polarization [5]. Otherwise, the thicker films show a much rougher surface to deviate from atomic layer-by-layer growth in quick deterioration of the film electrical quality [6]. Unfortunately, these ultrathin ferromagnetic electrodes unlike metal electrodes usually have poor electrical conductance, which adversely affect the accurate determination of the intrinsic ferroelectric domain switching field E_c from polarization-voltage (*P-V*) hysteresis loops. Presently, how to determine E_c is not only crucial for tailoring device driving force but also important for the study of domain switching kinetics.

Both theoretical and experimental evidences coincidently prove the existence of interfacial passive layers ("dead layers") between top/bottom electrode and the film, which are induced by space–charge screening length, stress–field coupling with spontaneous polarization, gradient polarization termination near surfaces, compositional and microstructural inhomogeneities [7–12]. The interfacial layers have much smaller dielectric permittivity and spontaneous polarization than those within the bulk [7–9], and the field across them is extremely high during domain switching time. If the interfacial layers are highly insulating, the *P-V* hysteresis loop is tilted with a reduced apparent coercive voltage $V_c^{(a)}$ [13]. Otherwise, the layers can be equivalently regarded as a nonlinear electrical resistor under a sufficiently high field (6–17 MV/cm) with a nonlinear Schottky emission current equal to domain switching current, where the *P-V* hysteresis loop becomes more squared with an enlarged $V_c^{(a)}$ [14]. Unfortunately, the latter case ubiquitously occurs in most thin films, and none can estimate the nonlinear resistance from *P-V* hysteresis loops directly.

Since 1950s, the positive-up-negative-down (PUND) technique has been widely applied to estimate domain switching time/speed under a series of square short pulses, while *P-V* hysteresis loops have been complimentarily adopted in characterization of ferroelectric spontaneous polarization and coercive voltage at a comparatively lower frequency or a longer time [15–18]. With the two techniques, a large number of data were obtained and explained on the basis of modified Kolmogorov–Avrami–Ishibashi (KAI) models (domain nucleation and growth) generally in the form of either $p(t) = 1 - \exp[-(t/t_0)^n]$ or $p(E) = 1 - \exp[-f^{-n}\Phi(E)]$, where p is the volume fraction of switched domains under the applied field E, n is the dimension of the domain growth, t is time, t_0 is domain switching time, f is the frequency for measuring *P-V* hysteresis loops, and Φ is the function of the volume density of the nuclei and domain forward growth speed [19–22]. Both t_0 and Φ depend on E. However, t_0 is not directly seen from *P-V* loop measurements. The further analysis is required to overcome this difficulty. Moreover, the circuit parasitic effects can also affect both domain switching time and apparent coercive voltage in the two techniques, for example, the poor electrode conductance, the resistance of the interfacial passive layers, the rise time of input pulse, and the capacitor area, besides the applied voltage [23]. All data without these corrections cannot truly reflect domain switching kinetics.

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In this article, we show the method to estimate the electrode-film contact resistance R_c and intrinsic domain switching coercive voltage V_c immune to the above parasitic effects from *P-V* hysteresis loops. Finally, the domain switching times at different frequencies are also calculated.

2. Experiments

For the study of the electrode resistance effect on domain switching, we firstly deposited 200 nm-thick $Pb(Zr_{0.4}Ti_{0.6})O_3$ (PZT) films on a 50 nm-thick Fe_2O_3 layer buffered $SrTiO_3$ (100) substrates using pulsed laser deposition technique, where the Fe_2O_3 layer has the poor resistance and is widely used for the investigation of interfacial ferroelectric–magnetic polarization coupling. The films were deposited at a flowing O_2 pressure of 30 Pa at 650 °C with the laser fluence of 1.5 J/cm^2. Next, we deliberately deposited a 6 nm-thick Al_2O_3 amorphous layer on a 300 nm-thick polycrystalline PZT with random grain orientations on $Ir/SiO_2/Si$ substrates to mimic the interfacial passive-layer effect on domain switching. The two layers were prepared using the atomic layer deposition and the metal–organic chemical vapor deposition separately, as reported elsewhere [24]. The film structure was inspected by X-ray diffraction (XRD) patterns (Bruker X-ray Diffractometer D8) using Cu$K_{\alpha1}$ and $K_{\alpha2}$ lines. Finally, top electrodes of Au and Pt dots with the areas of 3.6×10^{-5} and 6.1×10^{-4} cm^2, respectively, were deposited on the two films through metal shadow masks by electron beam evaporation with bottom electrode exposure after chemical solution etching. *P-V* hysteresis loops were checked using the Aixacct TF-2000 ferroelectric analyzer and Radiant Precision Materials analyzer with a triangular wave form at frequencies of 1–10 kHz in the virtual ground mode. The internal resistance of the two testers is $r = 100\ \Omega$. The triangular wave is configured in step-by-step voltage increase/decrease of many square pulses.

3. Results and discussion

Fig. 1(a) shows the sketch of Au/PZT/Fe_2O_3 capacitors using the two top electrodes separated with the distances of l and $2l$ ($l = 200$ μm), respectively, for the measurements of *P-V* hysteresis loops at different frequencies. The XRD patterns in Fig. 1(b) show the main reflections of PZT(001)//Fe_2O_3(202)//$SrTiO_3$(001) lattice matching, besides others due to the imperfection of single-crystal $SrTiO_3$ substrates, where Fe_2O_3 is tetragonal (space group $P4_32_12$) with a = b = 8.346 Å and c = 25.034 at room temperature. The consequent *P-V* hysteresis loops are shown in Fig. 1(c) with electrode separation of l. The apparent coercive voltage increases quickly over than 150% with the enhanced frequency from 50 to 2 kHz, far larger than 6% of the similar capacitor using the excellent conductance of Pt bottom electrodes. Fig. 2 (a) shows the equivalent circuit description of the setup, where R_c shows the resistance of the bottom electrode in series with an ideal ferroelectric capacitor C_f.

To elucidate the R_c effect on the domain switching, we sketched the triangular wave form in Fig. 2(a) with the voltage amplitude of V_m. If $R_c >> r$, the voltage drop across R_c is $V(t) - V_c$ during domain switching time, and the domains switching current is $J_{sw}(t) = \frac{V(t)-V_c}{R_c}$ [23], i.e.,

$$J_{sw}(t) = \frac{V_m 4ft - V_c}{R_c}, \quad \left(t_c < t < t_c^{(a)}\right), \tag{1}$$

where t_c is the beginning time of domain switching, and $t_c^{(a)}$ is time to complete the half polarization reversal. At $t_c^{(a)}$, the voltage across both R_c and C_f is $V_c^{(a)}$ in the loop. In combination with the equation of

$$P_r S = \int_{t_c}^{t_c^{(a)}} J_{sw} dt, \tag{2}$$

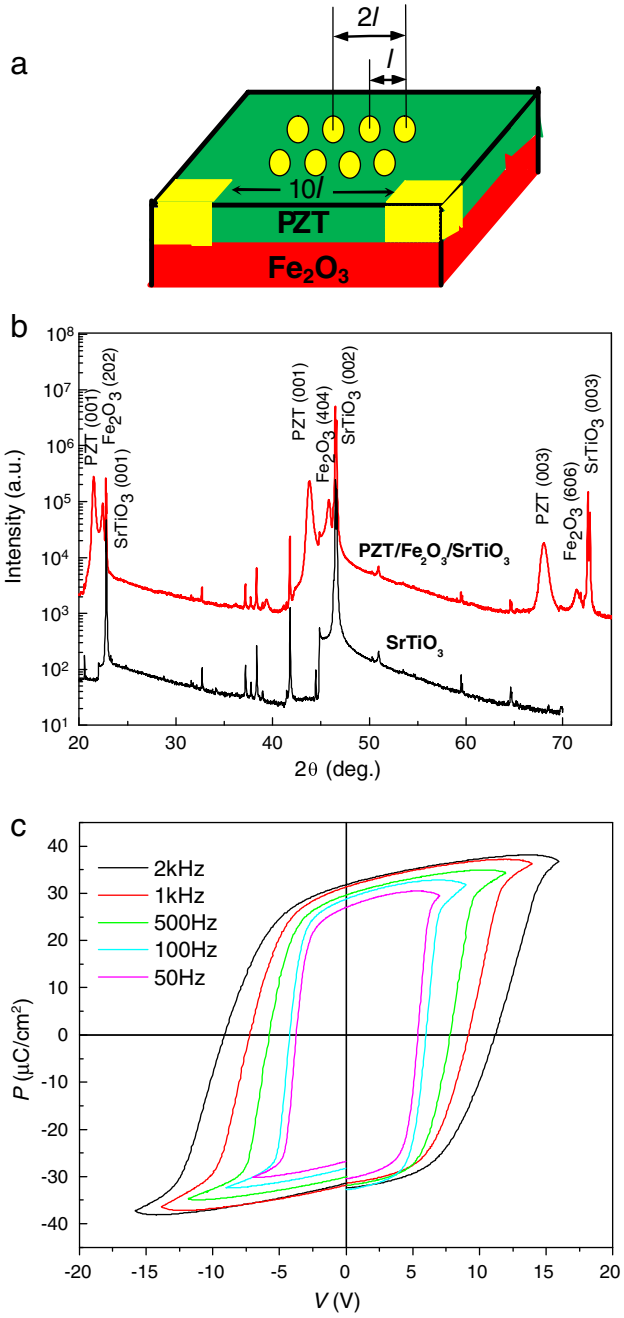

Fig. 1. (a) The sketch of Au/PZT/Fe_2O_3 thin-film capacitors with two top electrodes separated with the distances of l and $2l$ for the measurements of *P-V* hysteresis loops. (b) XRD patterns for both the PZT/Fe_2O_3/$SrTiO_3$ thin film and $SrTiO_3$ substrate. (c) *P-V* hysteresis loops measured with two top electrodes separated with l at different frequencies.

we have

$$V_c^{(a)} = V_c + \sqrt{8R_c P_r S V_m f}, \tag{3}$$

where S is the electrode area. The validity of Eq. (3) can be proved from the linearity of the $V_c^{(a)}$–$(V_m f)^{1/2}$ plot in Fig. 2(b). The plot is linear to obey Eq. (3), as shown by the solid-line fitting of the data, and its interception with the $V_c^{(a)}$ axis is $V_c = 4.1$ V, independent of the two top electrode distances. From the slopes of the two linear plots in Fig. 2(b), we further estimated bottom electrode resistance of 6.2 or 14.2 kΩ with electrode separation of l or $2l$. This R_c estimation