Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Detangling Extrinsic and Intrinsic Hysteresis for Detecting Dynamic Switch of Electric Dipoles using Graphene on Ferroelectric Gates

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The dielectric constant and loss of the epitaxial 500nm PLZT thin films were characterized in capacitor form as function of electric voltage across the device, as shown in Figure S1. The butterfly shape of the dielectric constant and loss curves are characteristic of ferroelectric thin films. The dielectric constant peaks locate almost symmetrically with respect to zero voltage, at ~1.6V and -1.75 V, respectively. However, the peak values of 670 and 830 differ considerably. This asymmetry is likely due to asymmetric top and bottom electrode of the capacitor as consequence of the strain gradient on PLZT lattice initiated at the PLZT and the bottom Nb:$SrTiO_3$ electrode (and also substrates) interface due to their lattice mismatch and different work function. The dielectric loss in the PLZT thin film capacitors is as low as 0.03. As report earlier, the dielectric constant decreases with frequency, following the logarithmic relationship in the frequency range of 100 Hz-100 kHz.[1]

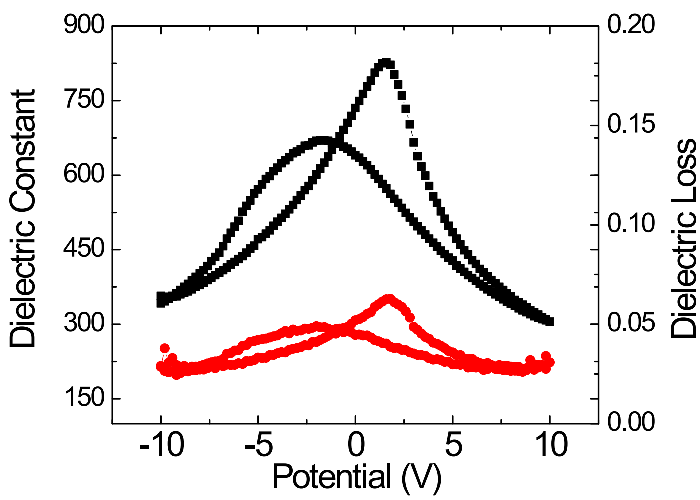

Figure S1. Dielectric constant (black) and loss (red) of PLZT thin film of 500 nm in thickness measured in capacitor form as a function of electric potential across the capacitor.

Figure S2a includes three current-voltage (*I-V)* loops measured by cycling the bias voltage between -0.5 V and +0.5 V (black), -1.0 V and +1.0 V (blue) and -4.0 V and 4.0 V (red), respectively, at a scan rate of 1000V/s with a 760E Bipotentiostat (CHI Instruments) across the PLZT film using a parallel-plate capacitor geometry. The polarization-electric field (*P-E*) curves can be derived by integrating the area under the *I-V* loop and then dividing the scan rate, as shown in Figure 4d.[2, 3] Both the E_c and P_r increase with increasing maximum applied field (E_{max}), as earlier report, due to the fact that a larger applied field can induce the increase of domain wall movement and better electrical dipole alignment in the PLZT thin films.[1] The observed slim *P-E* hysteresis loop is determined by the nature of PLZT material (low coercive field and remnant polarization) and anticipated for ferroelectric materials, which will certainly affect the I_D of the GFET/PLZT-gate devices through alteration aerial charge density. The observed low coercive field E_c is about 0.12 V/100 nm (or 12 kV/cm) under the maximum applied voltage of 1V. This means the dipole switching and the maximum dielectric constant can be reached at a much smaller applied electrical field across the PLZT gate, which is advantageous for low-power operation of the GFET/PLZT-gate devices. Figure S2b-f depict schematically the electric dipole alignment and switch as function of the electric field amplitude in different ranges defined in the

P-E hysteresis loop shown in Figure 4d.[4] Red arrow in the figure represents the direction of electric dipole in a single domain. A ferroelectric single crystal, when grown, has multiple ferroelectric domains in an effort to minimize energy.[4] Specifically, Figure S2b corresponds to the very low field range AB in Figure 4d below the E_c at point B. In this range, P increases monotonically with the increasing E field as a consequence of dipole alignment along the electric field direction, in other words, the single domain, in which the polarization is the same as the applied electric field, grows larger through domain wall motion. When the applied electric field is further increased above the E_c to the range of BC, electrical dipole switch begins from the opposite direction of polarization to the direction of electric field, increasing the measured displacement charge density nonlinearly as schematically illustrated in Figure S2c. When the applied electric field is decreased from Point C, the dipoles back-switch to opposite direction of the E field will be initiated as depicted schematically in Figure S2e, while a positive P (in the direction of the E-field) is maintained even when the applied electric field decreases to zero at Point D due to the remnant polarization. Figure S2d,f therefore represent two extreme cases when the maximum number of electric dipoles (or the maximum polarization P_{max}) are aligned with the electric field applied in the positive (at point C) and negative (at point E) directions, respectively.

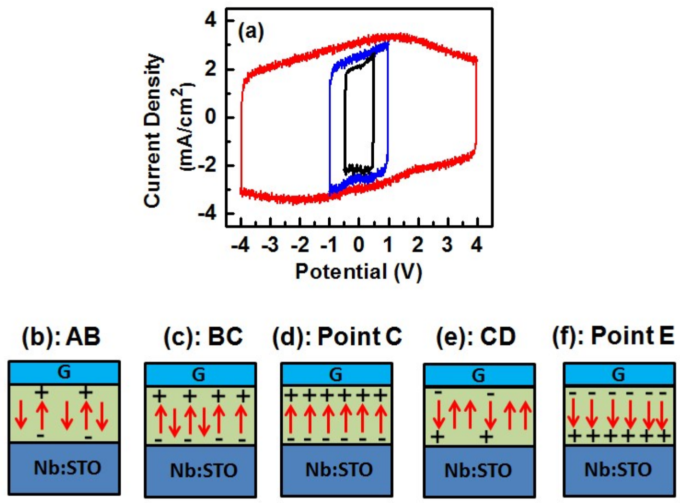

Figure. S2. a) The current density as function of applied electric voltage measured on a PLZT thin film in parallel-plate capacitor geometry. b-f) Schematic diagrams for the electrical dipole alignment and switch in the different ranges of the applied electric field as indicated in *P-E* loop in Figure 4d.

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