Investigations on Ferroelectric/Multiferroic tunnel junctions for multifunctional applications

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A ferroelectric tunnel junction (FTJs) consists of a few unit cells of a ferroelectric material sandwiched between two electrodes where interplay of ferroelectricity and electron tunneling occurs[1]. This is possible since ferroelectricity can be retained in perovskite oxides films with thickness of the order of a few nanometers[2-3]. FTJ may exhibit a tunneling electroresistance (TER) effect associated with the polarization switching of the ferroelectric barrier layer, leading to a change in resistance of the junction[4]. FTJ devices have the potential to function as nanometer scale non-volatile memory devices, operating with low power consumption. We demonstrate room temperature polar switching and tunneling in $PbZr_{0.52}Ti_{0.48}O_3$ (PZT) ultra-thin films of thickness 3-7 nm, sandwiched between platinum metal and ferromagnetic $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) layers, which also shows magnetic field dependent tunnel current switching in Pt/$PbZr_{0.52}Ti_{0.48}O_3$/$La_{0.67}Sr_{0.33}MnO_3$ heterostructures. The oxide PZT/LSMO heterostructure films were synthesized on $(LaAlO_3)_{0.3}(Sr_2AlTaO_6)_{0.7}$ (LSAT) using a pulsed laser deposition (PLD) technique. The epitaxial nature, surface quality and ferroelectric switching of heterostructured films were examined with the help of X-ray diffraction (XRD) patterns, atomic force microscopy (AFM), and piezoresponse force microscopy (PFM), respectively. The capacitance versus voltage graphs show butterfly loop above the coercive field (> ±3 V) of PZT for small probe area (~16 μm^2). The effect of ferroelectric switching was observed in current density versus voltage curves with a large variation in high-resistance/low-resistance (HRS/LRS) ratio (2:1 to 100:1), however, these effects were more prominent in the presence of in-plane external magnetic field.

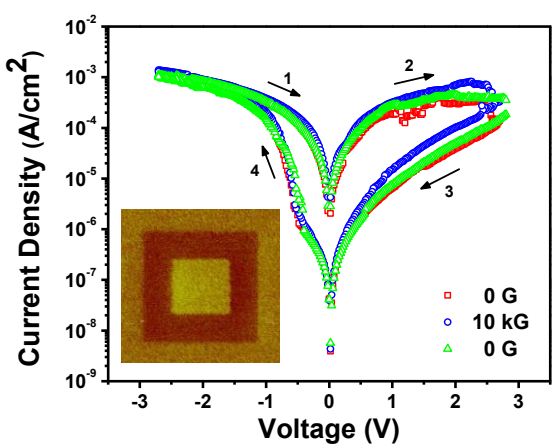

Figure 1 Current density versus voltage curve of the sample 7 nm showing the effect of applied 10 kG magnetic field. Inside picture on left down show the switching polarizations.

We show the effect of magnetic field along the in-plane direction; it was observed when 10 kG was applied to the junction during current versus voltage measurements, the resistance is decreased only in the positive voltage range (controlled by the LSMO electrode), while no significant change was observed in the negative voltage range (controlled by the Pt electrode). This is reasonable considering that LSMO becomes more metallic in its ferromagnetic state. For this junction the resistance switching between LRS and HRS became sharper, and the HRS/LRS ratio values at zero bias was between ~60 (at 0 G) and 110 (at 10 kG); the maximum value of HRS/LRS ratio was obtained in presence of a magnetic field (see Figure 1). The conductance is fitted with Brinkman's model, and the parabolic conductance upon bias voltage implies electron tunneling governs the transport[5].

Another interesting topic is multiferroic tunnel junction (MFTJ) that consists of metal or ferromagnetic electrodes separated by a ferroelectric (FE) or single-phase multiferroics barrier. MFTJ device displays four non-volatile resistance states due to the coexistence of tunneling magnetoresistance and tunneling electroresistance effects, which makes them very promising for the application in memory devices. $Pb(Zr_{0.53}Ti_{0.47})_{0.60}(Fe_{0.5}Ta_{0.5})_{0.40}O_3$ (PTZFT) is a single-phase multiferroic material, exhibiting remanent polarization of ~14 μC/cm^2 in bulk, remanent magnetization ~0.024 emu/g and, low loss ~ 0.06-10 kHz and a magnetoelectric coupling coefficients ~ 1x10^{-7} s/m at room temperature[6-7]. In order to study thickness effect on electrical and magnetic properties from thicker to ultrathin films, we have grown films with thicknesses from 4 to 80 nm of PZTFT on LSMO/$(LaAlO_3)_{0.3}(Sr_2AlTaO_6)_{0.7}$ (LSMO/LSAT) (001) substrate deposited by pulsed laser deposition technique. The X-ray diffraction patterns of the heterostructures show only (00*l*) reflections corresponding to the LSAT substrate, PZTFT and LSMO layers. The Atomic force microscopy of PZTFT/LSMO/LSAT heterostructures showed that the average surface roughness decreases from 1.5 to 0.4 nm for PZTFT films with thickness from 80 to 4 nm respectively. Well saturated ferroelectric loops were observed for PZTFT films with a remanent polarization of 60, 35 and 10 μC/cm^2 for films with thicknesses of 80, 50 and 20 nm respectively (see Figure 2). An enhanced saturated magnetization (M_s) was observed with increased of PZTFT layer thickness in PZTFT/LSMO structures. The average M_s values for PZTFT/LSMO heterostructures were 65, 50, and 40 emu/cm^3 for 80, 50, and 20 nm respectively, at 300 K.

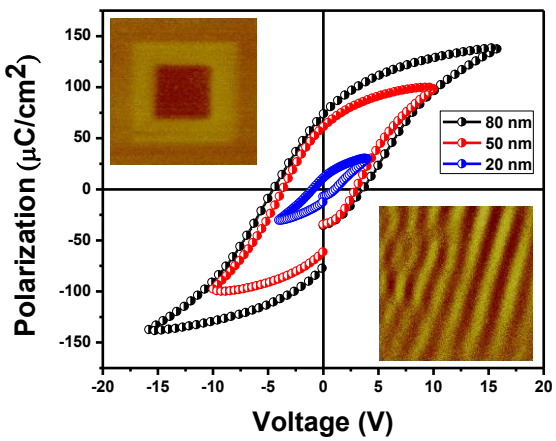

Figure 2 Well saturated ferroelectric loops were observed for PZTFT films. Inside pictures on left up and on right down show the ferroelectric and magnetic nature of PZTFT-7 nm.

Enhancement in magnetization with increase in PZTFT thickness may be due to the interface effect between PZTFT/LSMO layers. Piezo force microscopy measurements for 4, 5, and 7 nm ultrathin PZTFT films showed a clear and reversible out-of-plane phase contrast above ± 4 V, which indicates the ferroelectric character of ultra-thin films. Magnetic force microscopy shows a clear magnetic domain patterns. Stripe domains were present, typical of systems with perpendicular magnetic anisotropy. The effect of PZTFT film thickness on temperature dependent dielectric properties will be discussed.

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