Functional Response of Monolithic and Hetero-Layered Ferroelectric Thin Films

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Ferroelectric (FE) materials show a spontaneous polarization, reversibly switchable with an electric field, in addition to showing large pyroelectric and piezoelectric response, making them attractive for fulfilling multiple functionalities in MEMS devices as sensors, actuators, and energy harvesting units. Traditionally, lead zirconate titanate ($Pb[Zr_{1-x}Ti_x]O_3$, PZT) has been used in such devices. In bulk form, relaxor-FE single crystals, such as $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3$-$xPbTiO_3$, (1-x)PMN-xPT, exhibit even larger electromechanical response than ceramic PZT, when cut and poled along the (001) direction with compositions on the rhombohedral side of the morphotropic phase boundary (x~0.32).

However, these compositions are typically challenging to process in thin film form, with substantially smaller processing windows and higher crystallization temperatures than PZT. Additionally, films thicker than ~600 nm are often susceptible to cracking, when processed on Si substrates. In this work, hetero-layers of 0.7PMN-0.3PT and $Pb[Zr_{0.52}Ti_{0.48}]O_3$ are studied as an alternative approach to increase selective functional properties of the thin films.

All films were deposited on platinized Si substrates through chemical solution deposition. 0.4M precursor solutions of PZT and PMN-PT were prepared through a 2-methoxyethanol route and deposited onto the substrate via spin coating. A seed layer prepared using a 0.1 M PZT solution was used to induce (100) crystallographic orientation in the subsequently grown films. Hetero-layer thin films were created by depositing layers of either material in varying sequences. All films were pyrolyzed at ~ 400 °C for 1 to 5 minutes and annealed together in stacks of up to 3 pyrolyzed layers at temperatures ranging from 700 °C to 740 °C. Thin film thicknesses were varied between 200 and 1000 nm.

X-ray diffraction (XRD) analysis of the films confirmed a strong (100) perovskite orientation in all films. Functional measurements of the hetero-layered thin films were compared to pure PMN-PT samples, all of about 300 nm in thickness. Saturation effective longitudinal piezoelectric response $d_{33,f,sat}$ showed up to a 15% increase in films with a PZT bottom layer and PMN-PT top (PZPMN) layer composition in a 1:1 ratio. Conversely, films with a flipped architecture (PMNPZ, bottom layer of PMNPT and top layer of PZT) showed a decrease of up to 35% in $d_{33,f,sat}$. Dielectric permittivity values showed up to 30% decrease in PZPMN films, and up to 35% decrease in PMNPZ films, without any substantial change in the dielectric loss. Both hetero-layer thin film architectures showed increase in saturated polarization of up to 25% with respect to pure PMNPT samples of similar thickness. Polarization curves of all films indicated an existence of internal bias. Effects of the relative content of PMNPT to PZT as well as the effects of bottom layer chemistry choice on functional response will also be discussed.