Controlling the Intrinsic Polarization State in RF Sputtering Grown Ferroelectric Ultrathin Films

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Ferroelectric ultrathin films are a technologically important field of study, proposed as potential memory devices, memristors, or as a platform for novel integrated electronics using domain wall conduction. Crucial to all of these applications is the intrinsic polarization state of the thin film. This intrinsic polarization results from the interplay of the depolarizing field, stemming from the bound charges at the interfaces of the film, which destabilizes the uniform polarization configuration[1,2], and of the built-in field, which favors one polarization direction over the other[3]. The former must be compensated by screening either by external free charges from metallic electrodes or ions from the atmosphere, or by internal mobile charges from within the large-band-gap semiconducting ferroelectric itself to maintain a uniform polarization configuration. The latter results from an asymmetry in this screening or from internal sources, such as trapped charges or strain gradients leading to flexoelectricity.

We show that we can manipulate both these fields, acting on the electrostatic boundary conditions via the use of dielectric spacer layers to increase the depolarizing field[2,3], or modulating the built-in field through changes in the growth temperature of $PbTiO_3$ thin films, allowing full control over the intrinsic polarization state (monodomain up vs. polydomain vs. monodomain down).

Another important issue for applications is optimizing both the quality and production cost of samples. Off-axis radio frequency (RF) magnetron sputtering has been successfully used for over a decade to grow epitaxial oxide thin films and heterostructures. However, the relatively low cost of such a system compared to other thin film deposition techniques such as molecular beam epitaxy (MBE) or pulsed laser deposition (PLD) traditionally brings the disadvantage of lower control over the sample quality. Here, using $PbTiO_3$ as a model system, we present a slow kinetics intermittent sputtering (SKIS) approach to RF sputtering which greatly enhances the sample surface, crystalline and functional quality for a wide range of growth parameters, opening a pathway towards an affordable method to grow high quality epitaxial thin films.

[1] Ferroelectricity in ultrathin-film capacitors, C. Lichtensteiger et al, Ch. 12 in Oxide Ultrathin Films, Science and Technology, Wiley (2011) (**arXiv:1208.5309v1** **[cond-mat.mtrl-sci])**

[2] Tuning of the depolarization field and nanodomain structure in ferroelectric thin films, C. Lichtensteiger et al, NanoLetters 14(8) 42025 (2014)

[3] Built-in voltage in thin ferroelectric $PbTiO_3$ films: the effect of electrostatic boundary conditions, C. Lichtensteiger et al, New Journal of Physics 18(2016)04 3030