Crossover from 180-to-90 degree domains in ferroelectric thin films

A. Vlooswijk[1], A. Janssens[2], G. Rijnders[2], D. Blank[2], B. Noheda[1]
[1] *University of Groningen, Laboratory of Solid-State Chemistry*
[2] *University of Twente, Inorganic Materials Science Group, The Netherlands*

We have grown thin films of the classical tetragonal perovskite ferroelectric $PbTiO_3$ on *DyScO*3 substrates. Due to the minuscule mismatch between $PbTiO_3$ and *DyScO*3 at the growth temperature (< 0, 4% at 570◦*C*), high-quality paraelectric thin films can be grown, in which periodic ferroelectric domains form upon cooling down. The thinnest of these films (d< 8*nm*) display 180◦ periodic domains due to the large depolarizing fields, whereas the thicker films (d> 28*nm*) consist of 90˚ domain patterns determined by the elastic energy. For intermediate thicknesses, we have observed for the first time the crossover from 180˚-to-90˚domain walls. For this crossover, we propose a model that combines the elastic and electrical boundary conditions, giving rise to ferroelectric closure-like domains.

The observed domain periodicity (Λ) versus film thickness (d) correlation, has revealed the energetics of domain wall formation. For d< 8*nm*, 180domains form with periodicity in accordance with Kittel's Law applied to ferroelectric domains ($\Lambda \sim d^{1/2}$). Taking into account that during the cooling process, the domain walls can 'freeze in'[2], we have measured up to T=200℃ and did not observe any changes, showing that the 180˚ domain 'freezing' occurs above this temperature. Fitting the vs. d data provides domain wall energy between 120 and 132mJ/m^2 for freezing temperatures (T_f) between 200◦and 440◦C, respectively, which is in good agreement with ab initio calculations for free-standing $PbTiO_3$ [1].

The 90˚ domains in thicker films (d> 28*nm*), obey Roytburd's Law for 90˚ domains, with the same $d^{1/2}$ dependence and domain wall formation energy between 10 (T_f = 440◦*C*) and 65mJ/m^2 (T_f =25◦*C*). Therefore, 90˚ domain walls are likely to be mobile down to room temperature [4]. This result, which is in good agreement with ab initio calculations [1] for freestanding $PbTiO_3$, is somewhat surprising for epitaxially grown films and is most likely due to the elastic properties of *DyScO*3 [3].

[1] B. Meyer and D. Vanderbilt, *Phys. Rev.* B 65(10), 104111 (2002).

[2] M.L. Mulvihill et al., *J. Am. Ceram. Soc.* 80(6), 1462-68 (1997).

[3] S. Venkatesan et al., *J. Appl. Phys.* 102(10), 104105 (2007).

[4] A.H.G. Vlooswijk et al., *Appl. Phys. Lett.* 91(11), 112901 (2007).