L.8: Maxwell-Wagner relaxation driven high-dielectric constant in TiO_2/Al_2O_3 nanolaminates

Current approach towards scaling the metal–insulator–metal (MIM) capacitors is to replace the conventional SiO_2 by high-k dielectric layers to achieve higher capacitance density, reduced feature size, and low standby power. Multi-layered nanolaminates (NLs) of two dielectrics with conductivity contrast such as $Al_2O_3/TiO_2/Al_2O_3$ (ATA) containing alternatively stacked ultrathin layers of TiO_2 and Al_2O_3 have emerged as an attractive candidate due to the high dielectric constant owing to Maxwell-Wagner (M-W) relaxation, excellent thermal stability, and low leakage.

Amorphous ATA NLs were deposited by pulsed laser deposition on TiN/Si and Si substrates at 300 °C in O_2 pressure of 0.01 mbar using KrF excimer laser (248 nm) at laser fluence of ~ 0.5 J/cm^2. The sublayer thicknesses were reduced from ~2 nm to 0.7 nm in different NLs while keeping the total thickness of ~ 60 nm. The TiN/ATA/TiN capacitors were fabricated by depositing TiN top electrodes (thickness ~50 nm, diameter ~200 nm) by RF-magnetron sputtering using shadow mask. X-ray reflectivity plots of these NLs with intense Bragg peaks, as shown in Figure L.8.1, confirmed the superlattice structures with uniform layer thickness and chemically as well as physically sharp interfaces. Cross-sectional TEM image of the ATA NL with sublayer thicknesses of ~ 2 nm (inset of Figure L.8.1) revealed well-defined layer structure with ~3.94 nm periodicity, abrupt interface and long-range thickness uniformity of sublayers.

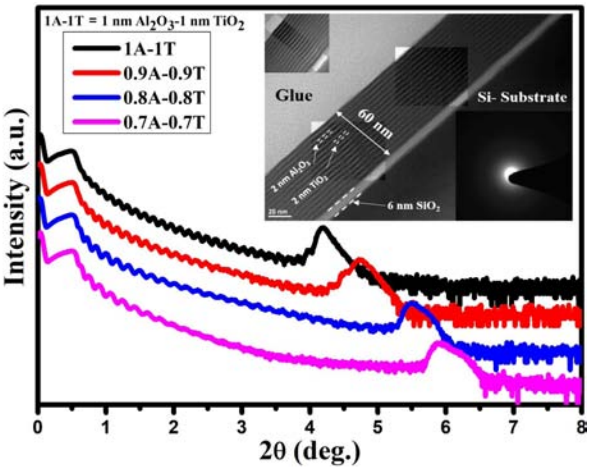

Fig. L.8.1. XRR plot for ATA NLs of different sublayer thickness. Inset shows HRTEM image of 2 nm ATA NL.

The dielectric constant and loss of ATA NLs studied through impedance spectroscopy is shown in Figure L.8.2. It can be seen that the dielectric constant of ATA NLs increased from ~60 to 670 with decreasing thicknesses of sublayers from ~2 nm to 0.8 nm and then reduced to ~380 for sublayer thicknesses of ~0.7 nm. The highest dielectric constant of ~ 670 obtained for ATA NL with 0.8 nm sublayer thickness (0.8A-0.8T) is significantly larger than that of both Al_2O_3 (K~10) and TiO_2 (K ~ 40). The dielectric loss in all ATA NLs were <1 as shown in inset of Figure L.8.2.

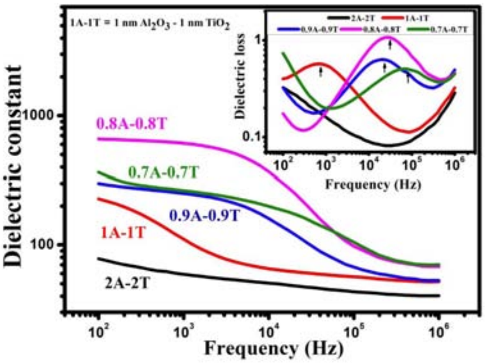

Fig. L.8.2: Dielectric constant and dielectric loss (inset) of ATA NLs as a function of frequency.

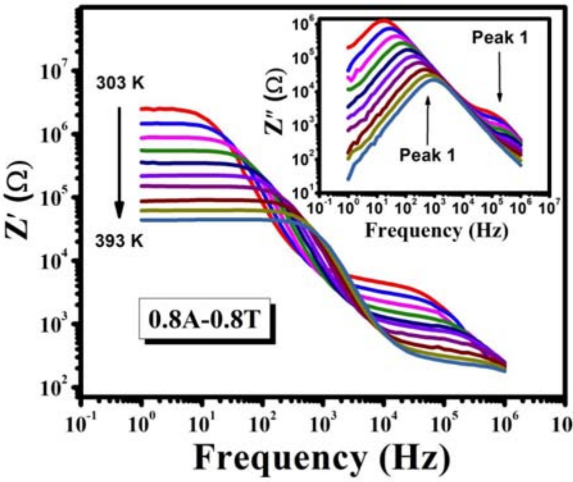

Fig. L.8.3: Variation of Z' and Z" of ATA NL with frequency and temperature.

The origin of high dielectric constant in ATA NLs was proposed to be due to M-W type relaxation caused by space charge polarization across the interfaces. When ac signal is applied across ATA NLs, surface charges accumulate at interfaces resulting in M-W relaxation, which is a typical external contribution to the dielectric constant. Temperature dependent dispersion in real and imaginary impedance (Z' and Z") of 0.8A-0.8T NL, as shown in Figure L.8.3, clearly indicated two sets of relaxations, both being thermally activated, confirming interfacial M-W relaxation. The 0.8A-0.8T NL showed high cut-off frequency of ~10^5 Hz along with low loss of ~ 0.17, and low leakage current density of ~ 10^{-5} A/cm^2, which further reduced to ~ 0.03 at 100 Hz and ~10^{-7} A/cm^2 at 1 V by sandwiching ATA NL between ~ 3 nm thick Al_2O_3 layer. A high capacitance density of 19 fF/μm^2, low EOT of ~1 nm, small quadratic voltage coefficient of capacitance α ~ 85 ppm/V^2 and linear coefficient β of ~263 ppm/V, together with a temperature coefficient of 81 ppm/°C and breakdown field of 1.13 MV/cm make ATA NL based MIM capacitors as promising candidate for next generation energy and data storage applications.

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