

Geometry-driven dielectric enhancement in SrTiO₃/BaTiO₃ superlattices

L. Yin¹, T. Ricart^{2,3}, F. Ducroquet³, A. Cantaluppi¹, A. Lahmar¹, A. Sylvestre² and N. Lemée^{1*}

¹Laboratory of Condensed Matter Physics, UR 2081, University of Picardie Jules Verne, 80039 Amiens, France

²University Grenoble Alpes, CNRS, Grenoble INP, G2Elab, 38000 Grenoble, France

³University Grenoble Alpes, CNRS, Grenoble INP, CROMA, 38000 Grenoble, France

***Email:** nathalie.lemee@u-picardie.fr

Abstract: Ferroelectric/dielectric superlattices constitute ideal systems to investigate the impact of nanoscale domains on dielectric properties at low dimensionality [1, 2]. These superlattices offer great flexibility in terms of design, and their dielectric properties depend on their geometry, defined by the number of periods and the thickness ratio between the ferroelectric and dielectric layers. Enhanced dielectric permittivity of superlattices is attributed to polarization topologies, lattice distortions or interfacial Maxwell-Wagner effects [1 - 4]. Here, we have investigated geometry-driven dielectric and structural evolution in SrTiO₃/BaTiO₃ (ST/BT) superlattices grown on (001) SrTiO₃ substrates. XRD results demonstrate the successful fabrication of single-phase epitaxial ST/BT superlattices with varying numbers of periods and ST/BT thickness ratios. Reducing the ST layer thickness progressively weakens the satellite peak sharpness and induces strain relaxation. The temperature- and frequency- dependent dielectric behavior of the superlattices as a function of the number of periods and the ferroelectric/dielectric ratio was studied. Increasing the number of periods at fixed composition leads to a pronounced enhancement of dielectric constant (ϵ_r) while preserving low dielectric loss tangent ($\tan\delta$), illustrated by a (ST₁₂BT₄)₅₀ superlattice composed of 12/4 unit-cell ST/BT layers with 50 repetitions, exhibiting $\epsilon_r \approx 481$ and $\tan \delta \approx 0.018$ at 1 kHz. Further optimization of the superlattice geometry by adjusting the ST/BT thickness ratio enables an even higher dielectric constant, with an (ST₄BT₄)₇₇ superlattice reaching $\epsilon_r \approx 732$ while maintaining a moderate loss tangent ($\tan \delta \approx 0.045$) at 1 kHz. Maxwell-Wagner relaxation, a phenomenon commonly encountered in materials with high dielectric constants, causes a more or less significant drift in dielectric properties depending on frequency and temperature, which is a hindrance for applications such as supercapacitors using superlattices. Adapting the geometry of our superlattices grown on SrTiO₃ substrates with a minimum SrTiO₃ layer thickness (~ 4 unit cells) is required to suppress Maxwell-Wagner relaxation. These findings highlight the essential role of superlattice geometry in controlling the dielectric response of ST/BT superlattices and achieving optimal performance in the fabrication of low-loss high-k dielectric capacitors.

Keywords: ferroelectric superlattices, strain relaxation, dielectric constant, dielectric loss,

References

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