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Elevated cryogenic permittivity of epitaxial $SrTiO_3$ films on silicon by stoichiometry and thickness control

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Strontium titanate ($SrTiO_3$, STO) stands out as a promising material for various electronic applications thanks to its exceptional dielectric properties. Its intrinsic parameters allow a direct growth on silicon, it shows nonlinear optical behaviour and importantly, its bulk dielectric permittivity of several hundred at room temperature is very high and increases further to ~10^4 at cryogenic temperatures thanks to its quantum paraelectric behaviour [1].

Molecular beam epitaxy (MBE) is one of the few techniques which allows epitaxial growth of STO directly on industry-relevant Si substrates. The large lattice mismatch between STO and Si (-39.1%) can be reduced tremendously to only few percents when the STO lattice rotates 45° with respect to Si, i.e., (001) $[100]_{STO}$ // (001) $[110]_{Si}$, allowing direct heteroepitaxy [2]. However, maintaining precise stoichiometry and high crystalline quality in this process remains a significant challenge. Establishing this is essential to obtain STO with bulk-like dielectric properties and to minimize leakage current and optical absorbance. In this study, the importance of cationic stoichiometry and the effect of thickness are investigated for STO thin films epitaxially grown on (001)-oriented silicon substrates.

During epitaxy, the Sr molecular beam is gradually disturbed by the high oxygen environment generally conduct to a Ti-rich STO layer. We employed real-time reflection high-energy electron diffraction (RHEED) as a feedback loop mechanism to counteract Sr source oxidation and maintain a constant flux. Additionally, high-temperature post-growth annealing treatments in O_2 were investigated to promote layer relaxation and reduce oxygen vacancy concentration, thereby improving the physical, electrical, and optical properties of stoichiometric STO. As a result, high-quality STO thin films exceeding 100 nm were successfully fabricated featuring a bulk-like out-of-plane lattice parameter and refractive index, as well as rocking curve full width at half maximum (FWHM) below 0.2°, smooth surface ($R_q < 0.2$ nm) and a leakage current density below 1E-7 A/cm^2 [3].

This epitaxy process for MBE growth of high-quality thick STO layers on silicon (001)-oriented substrates is essential for optimizing dielectric properties, such as the dielectric permittivity. By establishing a correlation between cationic stoichiometry, crystallinity, and STO thickness, we achieve significant enhancement of the effective permittivity at cryogenic temperatures, reaching value of over 2,500 for our stoichiometric 105 nm STO film. To our knowledge, this is the highest reported permittivity for STO thin films on silicon. This study paves the way for using STO thin films as active materials in advanced devices for various applications, including energy storage and quantum information technology.

References:

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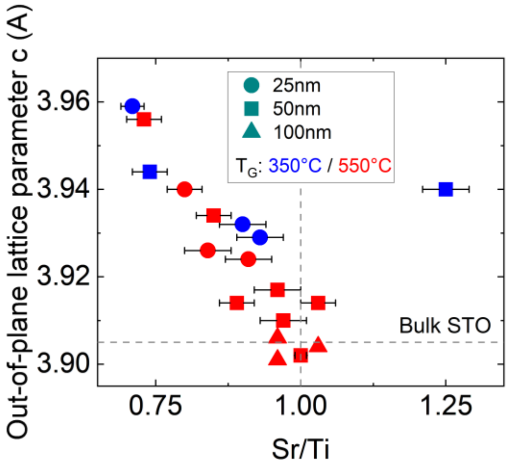

Figure 1: Out-of-plane lattice parameter (c) as a function of Sr/Ti ratio for STO films with varying thickness and growth temperature T_G. For obtaining a bulk-like lattice parameter, perfect cationic stoichiometry, high growth temperatures and layers > 50 nm need to be fulfilled.

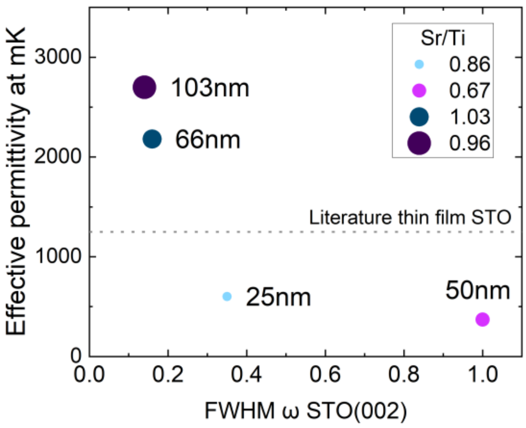

Figure 2: Effective permittivity ε_{eff} near 0 K as a function of the STO (002) rocking curve FWHM. Cationic stoichiometry (Sr/Ti = 1) and increasing STO thickness result in lower FWHM and higher effective permittivity.