Tailoring Domain and Domain-Wall Properties of Epitaxial Ferroelectric and Ferroelastic Thin Films Using Strain, Electrostatics, and Light

Martin F. Sarott[1,2]

1) Zernike Institute for Advanced Materials, University of Groningen, Nijenborgh 3, 9747AG Groningen, The Netherlands
2) Groningen Cognitive Systems and Materials Center (CogniGron), University of Groningen, Nijenborgh 3, 9747AG Groningen, The Netherlands

m.f.sarott@rug.nl

The pursuit for new materials to form the basis for future energy-efficient electronics has brought forward transition-metal oxides that offer a broad range of potentially coexisting functionalities. Ferroelectric oxides distinguished by the presence of a non-volatile electric polarization that can be switched by an electric field are especially promising for the development of novel electronic devices with reduced power consumption. Despite having found countless commercial applications owing to their concomitant piezo- and pyroelectric properties, the integration of ferroelectric oxides into next-generation memory and logic devices is, however, still facing several difficulties. Notably, when prepared in the form of ultrathin films and heterostructures, achieving a deterministic arrangement of domains – the basic functional entities – is rendered challenging by the competing effects of various interactions, such as strain, electrostatics, and defect chemistry.

In our work, we design tailored ferroelectric and ferroelastic domain and domain-wall configurations in epitaxial thin films with bespoke nanoscale and macroscopic properties by systematically elucidating the roles of the elastic and electrostatic boundary conditions. In ferroelectric thin films, we track the formation and evolution of domains *in situ* – during thin-film growth – using non-linear optics. This in-situ access to the functional properties enables us to design specialized nanoscale ferroelectric domain configurations in films of the archetypal ferroelectrics $PbTiO_3$ and $Pb(Zr_xTi_{1-x})O_3$ that feature (i) analog switching characteristics[1] and (ii) allow for a remanent optical control of the ferroelectric polarization.[2] Knowledge of the domain-formation process and its dependence on strain further enables us to address another major challenge for the device integration of epitaxial ferroelectrics: their compatibilization with silicon substrates. Typically, strain engineering of oxides on silicon is strongly limited due to the large structural mismatch and thermal strain arising during cooldown that often result in an interior interfacial quality and degraded switching characteristics. By introducing a strain-mediating pseudo-substrate buffer layer, we manage to overcome this limitation and accomplish single-crystalline ferroelectric $BaTiO_3$ thin films on silicon that exhibits imprint-free switching, a low coercivity and a high remanent polarization.[3] Finally, we show that extending these strain-engineering strategies to ferroelastic materials further allows us to evoke emergent domain and domain-wall phases with new properties that cannot be found in bulk. Specifically, we find that epitaxial shear strain imposed on thin films of WO_3 stabilizes a polar triclinic phase that features periodic in-plane polarized stripe domains. Remarkably, the stripe-domain walls display an enhanced electrical conductivity along with a reduction of all inherent structural distortions, pointing toward *anti-distortive polaronic* conduction.[4]

[1] Sarott et al. Nat. Commun. **13**, 3159 (2022)
[2] Sarott et al. Adv. Mater. **36**, 2312437 (2024)
[3] Zhao,...,Sarott et al. Nano Lett. **26**, ,764−772 (2026)
[4] Sarott*, van der Veer* et al. arXiv:2505.11021v2 (2025)