Designing ferroelectric domain configurations in epitaxial thin films using strain, electrostatics, and light

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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 difficulties. Notably, when prepared in the form of technologically relevant thin films and heterostructures, achieving a deterministic arrangement of domains – the basic functional entities of ferroelectric materials – is rendered challenging by the competing effects of various interactions, such as strain, electrostatics, and defect chemistry.

In our work, we pave the way toward the design of tailored ferroelectric domain configurations in epitaxial thin films by tracking and controlling domains at the point of their emergence – during thin-film growth. With the direct in-situ access to the functional property of ferroelectrics using non-linear optics, we manage to unravel the mechanisms that drive the formation of domains in prototypical ferroelectric oxides and put forward design strategies to realize highly specialized domain configurations.

In thin films of the archetypal ferroelectrics $PbTiO_3$ and $Pb(Zr_xTi_{1-x})O_3$ (PZT), we discover that tensile epitaxial strain induces the formation of in-plane-oriented domains within an out-of-plane-oriented matrix already during thin-film growth[1]. We further uncover that the epitaxial growth conditions can significantly affect the concentration of charged defects, which determines the electrostatics during growth and, hence, provides us with a handle to control the direction of the polarization[2]. Making use of these in-situ observations, we then tailor the elastic and electrostatic boundary conditions in PZT thin films to obtain a nanoscale domain configuration exhibiting non-binary switching characteristics[3], which is essential for applications in brain-inspired neuromorphic computing. Finally, we explore the use of light as a remote trigger to optically manipulate the ferroelectric polarization and find that above-bandgap optical excitation can strongly alter the charge screening in ferroelectric heterostructures[4]. Depending on the ferroelectric domain configuration, this can prompt a transient modification of the polarization or even non-volatile poling, constituting a major step toward all-optical control of ferroelectricity.

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