Engineering materials at the atomic scale for energy efficient application

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The successful integration of functional oxides into energy-efficient nanoscale devices relies on our ability to engineer materials properties in the application relevant ultrathin regime. In films of only a few unit cells in thickness, however, the increasing impact of defect chemistry, surfaces and interfaces contributions on the materials response obstructs a direct implementation into technological application. I will share our most recent insights into the engineering of thin films properties towards deterministic functional behaviours down to the single unit cell regime. Our work is enabled by a unique approach based on in-situ and real-time monitoring schemes bringing direct access to the emergence and evolution of the physical property of the films during the film's epitaxial deposition. I will showcase a few examples related to ferroelectric materials, i.e. established candidates for beyond complementary metal-oxide semiconductor (CMOS) technology, owing to their non-volatile spontaneous electrical polarization.

Using a combination of nonlinear optics and electron beam in-situ metrology, we visualize in real time the impact of the films processing on the physical properties. It expedites the identification of key parameters such materials off-stoichiometry and interface chemistry towards the stabilization of a polarization from the very first unit cell deposited. Taking most debated ferroelectric materials $BaTiO_3$, $PbTiO_3$ as model systems, we revolutionize the electrostatic-boundary-condition control in ferroelectric multilayers and create composite materials exhibiting coexisting ferroelectric and long-range magnetic orders. Finally, the use of unconventional ways to manipulate multifunctional phase coexistence and polarization states with light in thin films will be discussed. Hence our work highlights the strategic need to develop in-situ and operando materials characterization approaches to harness the complex chemistry and physics of materials interfaces to facilitate their technological integration.