Direct Imaging of Polarization-induced Charge Distribution and Domain Switching using TEM

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In this talk, I will present two research works in progress, which are: i) mapping of piezoelectric polarization and associated charge density distribution in the heteroepitaxial InGaN/GaN multi-quantum well (MQW) structure of a light emitting diode (LED) by using inline electron holography and ii) in-situ observation of the polarization switching process of an ferroelectric Pb(Zr1-x,Tix)O3 (PZT) thin film capacitor under an applied electric field in transmission electron microscope (TEM). In the first part, I will show that strain as well as total charge density distributions can be mapped quantitatively across all the functional layers constituting a LED, including n-type GaN, InGaN/GaN MQWs, and p-type GaN with sub-nm spatial resolution (~0.8 nm) by using inline electron holography. The experimentally obtained strain maps were verified by comparison with finite element method simulations and confirmed that not only InGaN QWs (2.5 nm in thickness) but also GaN QBs (10 nm in thickness) in the MQW structure are strained complementary to accommodate the lattice misfit strain. Because of this complementary strain of GaN QBs, the strain gradient and also (piezoelectric) polarization gradient across the MQW changes more steeply than expected, resulting in more polarization charge density at the MQW interfaces than the typically expected value from the spontaneous polarization mismatch alone. By quantitative and comparative analysis of the total charge density map with the polarization charge map, we can clarify what extent of the polarization charges are compensated by the electrons supplied from the n-doped GaN QBs. Comparison with the simulated energy band diagrams with various screening parameters show that only 60% of the net polarization charges are compensated by the electrons from the GaN QBs, which results in the internal field of ~2.0 MV cm-1 across each pair of GaN/InGaN of the MQW structure. In the second part of my talk, I will present in-situ observations of the polarization switching process of a planar Ni/PZT/SrRuO3 capacitor using TEM. We observed the preferential, but asymmetric, nucleation and forward growth of switched c-domains at the PZT/electrode interfaces arising from the built-in electric field beneath each interface. The subsequent sideways growth was inhibited by the depolarization field due to the imperfect charge compensation at the counter electrode and preexisting a-domain walls, leading to asymmetric switching. It was found that the preexisting a-domains split into fine a- and c-domains constituting a 90° stripe domain pattern during the 180° polarization switching process, revealing that these domains also actively participated in the out-of-plane polarization switching. The real-time observations uncovered the origin of the switching asymmetry and further clarified the importance of charged domain walls and the interfaces with electrodes in the ferroelectric switching processes.

Keywords: Polarization, Strain, LED, Ferroelectrics, Domain switching