An atomic-scale view of the structure and electronic properties of manganite thin films and superlattices.¹

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Epitaxial manganite ultrathin films and heterostructures constitute an ideal system to study the nature and effects of inhomogeneity and phase competition in these materials. The presence of interfaces and epitaxial strain may cause new phenomena to arise, and a comprehensive study of the relations between structural, chemical and electronic properties at the atomic scale is needed. The combination of scanning transmission electron microscopy and energy loss spectroscopy represents a very powerful tool to analyze locally such relations. Unlike diffraction techniques, STEM-EELS allows clear separation of electronic valence from structural relaxations. Several examples will be presented, including the study of charge ordering in Bi₅₀.₃₇Ca₆₅.₆₃MnO₃. We find atomically-resolved striping of the Mn L₂,₃ spectra that correlate with two distinct formal charge states (+3/+4). Theoretical results obtained by first-principles density-functional calculations reproduce the structural relaxation that leads to the striping with minimal charge transfer. In contrast, charge transfer may occur across YBa₂Cu₃O₇₋ₓ/La₀.₆₇Ca₀.₃₃MnO₃ interfaces, where extensive charge transfer processes drastically affect the ferromagnetic/superconducting properties. Epitaxial strain can also play a determining role in ultrathin LCMO films, inducing a nanoscale modulation in the Mn+3/Mn+4 ratio which evidences the existence of nanoclusters with different electronic densities.