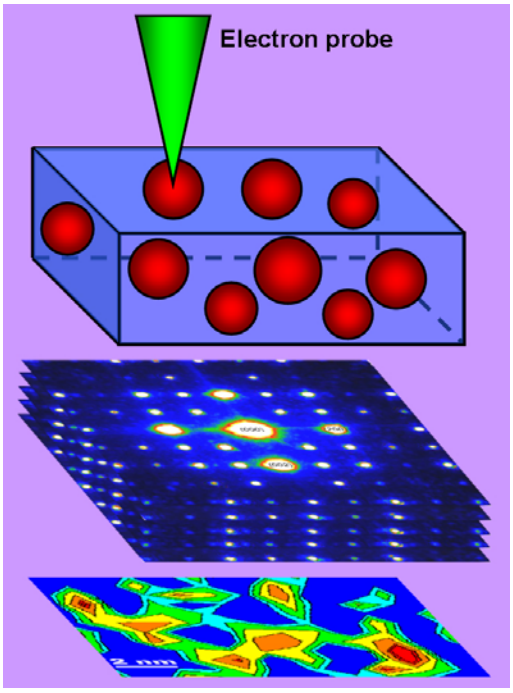
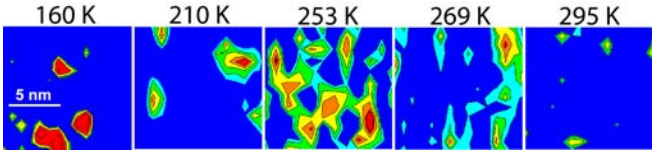
**Direct Imaging of Structural Phase Separation at Nanoscale: a Key Phenomenon underlying the Colossal Magnetoresistance Effect in Manganites**

Without direct observation, convincing evidence is often missing to link the structural inhomogeneity to material properties, such as the colossal magnetoresistance (CMR) effect in doped manganites. Using a newly developed technique, scanning electron nanodiffraction (SEND), we were able to map in real space the distribution of the charge-ordered (CO) phase, ~3-4 nm in size, with its characteristic structure as a function of temperature in $La_{0.55}Ca_{0.45}MnO_3$. The direct observation enables us to quantify the volume fraction of the nanoscale phase and establish its relationship with the CMR effect through the phase transition. The data further establish that the nanoclusters are not due to chemical inhomogeneities, suggesting the CMR effect be an intrinsic property of the material. Our research opens the door to better understanding of the nature of complex functional materials.




SEND intensity maps of the superlattice reflections upon warming in a single crystal domain in $La_{0.55}Ca_{0.45}MnO_3$ (LCMO). Warm color is corresponding to the nanoscale CO phase. The flat blue area has the intensity of the superlattice reflections below the noise level. Although each image is not necessarily in precise registration with any other, the change of the density of the CO nanoclusters is evident through the phase transition. The temperature with maximum density of the CO nanoclusters (~ 253 K) coincides with the CMR peak temperature in LCMO. The volume fraction of the CO nanoclusters was quantitatively estimated from the SEND maps to be significant enough to contribute to the CMR effect.

Scanning Electron Nanodiffraction: Using a small electron probe (green) ~ 1.7 nm in size, local structure at nanoscale can be obtained in electron nanodiffraction (END) patterns. A stack of END patterns can be acquired, each corresponding to every point in the probe-scanning area. In nanoscale phase separated LCMO, only when the electron probe is located on the CO phase (red) do superlattice reflections appear in the END pattern. The measured intensities of the superlattice reflections allow us to map the real space distribution of the nanoscale CO phase.

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