Probing Intertwined Interactions in Strongly Correlated Material Systems Using Transmission Electron Microscopy

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As a first-year graduate student, I started a research project of studying emergent phenomena arising from complicated interactions among charge, orbital, spin and lattice in doped manganites in John Spence's group at Arizona State University. We synthesized a few Ca-doped LaMnO₃ bulk crystals (Ca doping level x = 1/3, 1/2 and 2/3) and measured their physical properties. The crystal structures and electronic structures, i.e., charge ordering (CO) of Mn³⁺ and Mn⁴⁺ ions and orbital ordering (OO) of e_g electrons (Fig. 1a)), at low temperatures were characterized using temperature-dependent transmission electron microscopy (TEM) (Fig. 1b) - 1d)). A novel CO phase at the nanoscale was discovered at the x = 1/3 material sample (Fig. 1b)) [1].

This project has turned out to be a longstanding research work in my life. With various scenarios at different length scales, doped manganites seem to be paradigm systems to explore by taking advantages of TEM techniques. Strong couplings between electrons and the lattice counterpart in manganites give rise to electronic phase separations and gigantic secondary effects such as colossal magnetoresistance effect. In situ TEM techniques offer local probes which interact very strongly with nanoscale electronic phases at controllable temperatures. Hence the inhomogeneously distributed electronic structures can be visualized in real-space (Fig. 2a)) [2] with the nature of the phase revealed (Fig. 2b)) [3]. To this date, doped manganites we synthesized at John's group are still under exploration with interesting findings using advanced TEM methods.

The manganite project has also shaped the way I conduct research on multiple correlated materials, such as dichalcogenides with charge density wave and magnetites with structural phase transitions. Since electron beams are sensitive to both valence electrons and nuclei during scattering processes [4], I pay special attention to the entanglement between electrons and the lattice that often plays a key role in the materials' functionalities, where electron diffraction and TEM techniques can provide unique observations compared to other experimental tools. I will never forget that John led me to the research area and will keep my feet to get going on the road.

References

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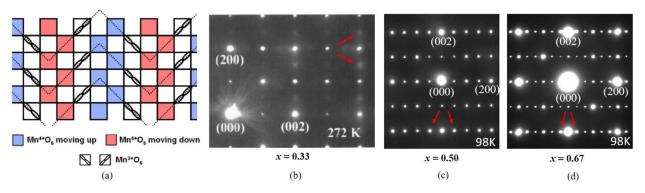


Fig. 1. (a) A schematic drawing showing a charge ordering (CO) of Mn^{4+} and Mn^{3+} with an orbital ordering (OO) of e_g electrons in Mn^{3+} ions in $La_{1/3}Ca_{2/3}MnO_3$. (b) A short-range CO phase was discovered in $La_{2/3}Ca_{1/3}MnO_3$ with the diffuse scattering pointed out by the arrows in the electron diffraction pattern. (c) and (d) Electron diffraction patterns at low temperatures obtained from $La_{1/2}Ca_{1/2}MnO_3$ and $La_{1/3}Ca_{2/3}MnO_3$ show long-range CO structures pointed out by the arrows.

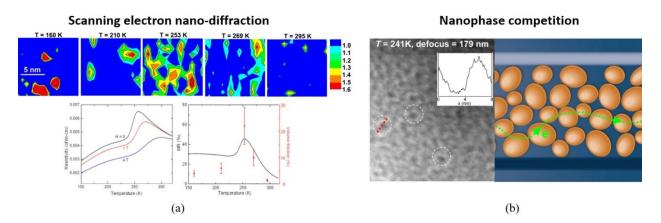


Fig. 2. (a) Top: nanoscale CO phases in a $La_{0.55}Ca_{0.45}MnO_3$ sample were mapped in real-space using scanning electron nano-diffraction technique. Bottom: physical properties measured from the material show colossal magnetoresistance (CMR). The visualization of the CO phases establishes a structural-property relationship to reveal the role of the nanoscale CO in CMR. (b) A Lorentz TEM image shows some magnetic component in the nanoscale CO phases (left) which suggests a new mechanism of the CMR effect (right).