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Atomic-resolution spectroscopy of quantum materials at cryogenic temperatures

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Recent developments in cryogenic scanning transmission electron microscopy (cryo-STEM) has enabled direct visualization and quantification of functional structural order parameters with picometer precision, advancing our understanding of emergent order in quantum materials such as charge-ordered phases [1]. Coupled with electron energy loss spectroscopy (EELS) it has also allowed measurements of electronic structure changes and charge transfer associated with emergent electronic behavior at interfaces [2].

Atomic-resolution spectroscopic imaging is, however, largely limited to room temperature as standard side-entry cryo-holders suffer from increased sample drift and instabilities due to cryogen bubbling. In imaging, sufficiently short pixel dwell times can be chosen so that drift can be accounted for by averaging a series of rapid acquisitions. To go beyond high-resolution cryogenic STEM imaging to spectroscopic mapping is significantly more challenging. A spectrum with sufficient signal-to-noise ratio requires a dwell time of a few milliseconds, orders of magnitude larger than typical imaging dwell times. Thus, low temperature EELS experiments have focused on collecting area-averaged spectra or line profiles [2,3]. However, the tendency of emergent states towards nanoscale inhomogeneity and spatial modulation requires atomically-resolved spectroscopic maps at low temperatures.

In addition to ongoing improvements in holder stability, more efficient detectors hold great promise for EELS mapping at cryogenic temperatures. Here, we demonstrate atomic-resolution spectroscopic mapping near liquid nitrogen temperature enabled by a Gatan K2 Summit direct electron detector (DED) for spectroscopy. In addition to improvements in detector quantum efficiency, point spread function, and increased signal-to-background [4], the reduced readout time is critical for current cryo-experiments or application that require rapid acquisitions. Even at a relatively short dwell time (2.5 msec/pixel), the signal-to-noise ratio of the spectra recorded using a DED camera remains high, allowing, for instance, atomic-resolution elemental mapping of a $La_{0.8}Sr_{0.2}MnO_3/SrTiO_3$ interface near liquid nitrogen temperature (Fig. 1) [5]. In this talk, I will discuss how these new capabilities have enabled us to map fine structure changes associated with orderings of charge and lattice in low temperature quantum phases.

So far, our low temperature atomic-resolution EELS experiments have been limited to the temperature of the cryogen used, i.e, liquid nitrogen (LN_2) in our case. Combining a LN_2 cryo-holder with local sample heating using a MEMS chip (HennyZ dual-tilt LN_2 MEMS holder) gives access to intermediate temperatures. First results show stable atomic resolution imaging in the range of ~100K to room temperature (Fig. 2). Drift rates can be greatly reduced by continuously controlling the cryogen level during the experiment. Further developments of stable cryogenic stages that extend the range of accessible phases will play an important role in understanding quantum materials. STEM based imaging and spectroscopy across a range of temperatures will enable real-space, atomic-resolution measurements of emergent, spatially heterogeneous electron-lattice correlations in a broad range of electronic phases and will help disentangle the complex couplings and emergent states in quantum materials [6].

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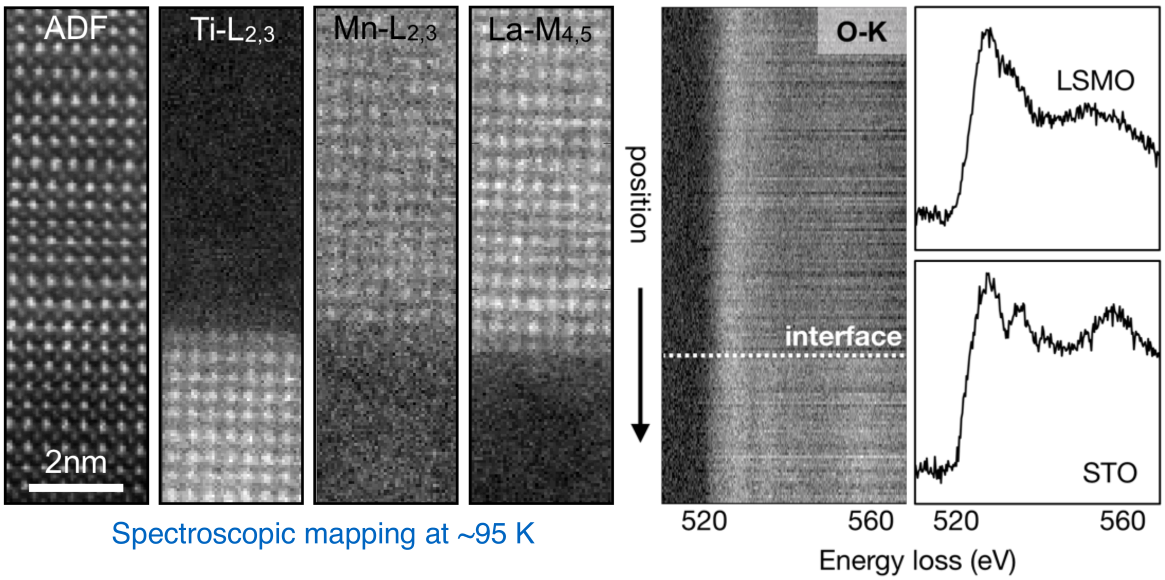

Figure 1. Atomic-resolution spectroscopic mapping of a $La_{0.8}Sr_{0.2}MnO_3/SrTiO_3$ interface near liquid nitrogen temperature. The data is acquired using a Gatan K2 Summit direct electron detector camera, which allows rapid acquisition while maintaining high signal-to-background [5].

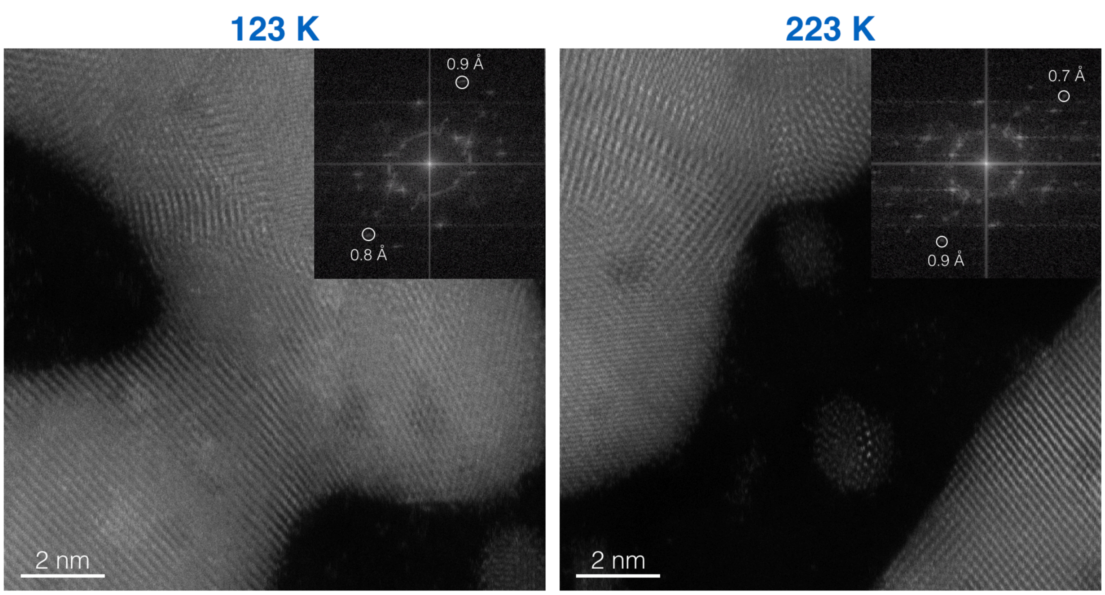

Figure 2. Cryogenic HAADF STEM imaging of gold nanoparticles on carbon support imaged at intermediate sample temperatures using the HennyZ dual-tilt variable-temperature LN_2 cryo-holder. To demonstrate stability, no image registration was performed.

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