

Aberration-Corrected STEM/EELS at Cryogenic Temperatures

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Today's aberration-corrected scanning transmission electron microscopes (STEM) routinely focus high-energy electrons down to a spot smaller than 1 Å in diameter to perform scattering experiments that allow us to study the atomic-scale structure of materials and devices. When combined with electron energy loss spectroscopy analysis of the inelastically scattered electrons, these narrow probes can also provide atomic-scale information about the composition and local electronic structure of bulk materials, defects and interfaces [1, 2].

Most efforts, so far, have focused on room temperature measurements where stable imaging with sub-Å resolution is routinely achievable. However, a range of materials including complex oxides and transition-metal dichalcogenides (TMDs) exhibit exotic functionalities below room temperature. Correlating the atomic level structure and electronic properties at cryogenic temperatures is therefore an important step in realizing the goal of understanding and controlling emergent phenomena in these materials. Charge density waves (CDWs) and the coupled periodic lattice distortions (PLD), for example, govern the electronic properties in many layered TMDs. In particular, 1T-TaS₂ can undergo a phase transition from a conducting to an insulating state upon cooling as the PLD becomes commensurate with the crystal lattice. Using an uncorrected FEI Tecnai F20 equipped with cryo-blades we have directly imaged PLDs in thin exfoliated 1T-TaS₂ using atomic resolution cryo-STEM [3].

Aberration-corrected cryo-STEM on a FEI Titan Themis allows imaging at sub-Å resolution and atomic tracking with picometer precision. This, however, requires acquisition optimization to account for stage drift and instabilities during imaging at cryogenic temperature. A series of rapid acquisitions with sufficiently large SNRs to allow for effective rigid registration of frames has proven to be successful demonstrating an information transfer below 1 Å (Figure 1). Using this technique we have mapped the evolution of PLD stripes in Bi_{1-x}(Sr,Ca)_xMnO₃, a model charge-ordered system with T_c ~300K. We observe temperature-dependent inhomogeneities in the stripe order such as shear deformations and topological defects, and the emergence of phase coherence well below T_c. Atomic resolution spectroscopic mapping, which has more stringent stability requirements, is successfully performed using a direct electron detector (DED) – a Gatan K2 Summit operated in electron counting mode (Figure 2). The detector's superior SNR for low-dose applications enables spectroscopic mapping at reduced dwell times. Real space visualization of strongly correlated materials by aberration-corrected cryo-STEM/EELS promises a deeper understanding of these complex systems [4].

[1] D. A. Muller *et al.*, Science **319** (2008), p. 1073.

[2] J. Mundy *et al.*, Nat. Commun. **5** (2014), p. 3464.

[3] R. Hovden *et al.*, Proc. Natl. Acad. Sci. U.S.A. **113** (2016), p. 11420.

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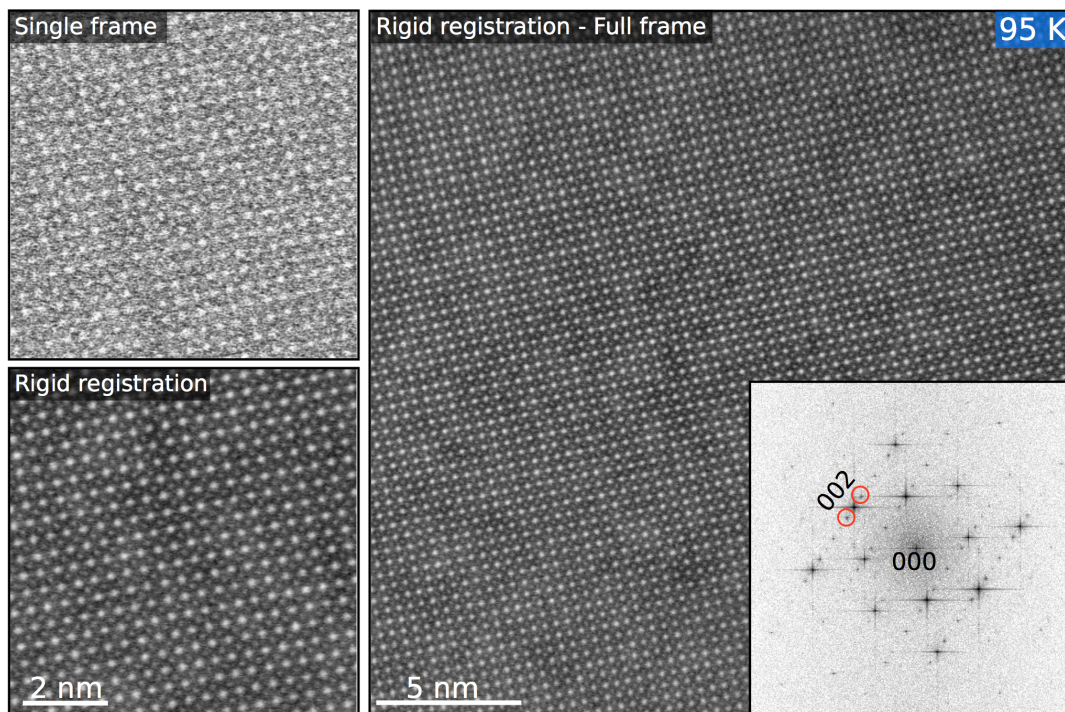


Figure 1. Aberration-corrected cryo-STEM of a stripe ordered manganite. (*top left*) Small section of a 2kx2k single frame recorded at a dwell times of 0.5 s/pixel. (*bottom left*) Same section after rigid registration of all frames. (*right*) Large field of view HAADF cryo-STEM image (frame averaged) of the manganite oxide $\text{Bi}_{1-x}(\text{Sr,Ca})_x\text{MnO}_3$. The FFT shows PLD peaks (two marked with red circles) decorating the Bragg peaks.

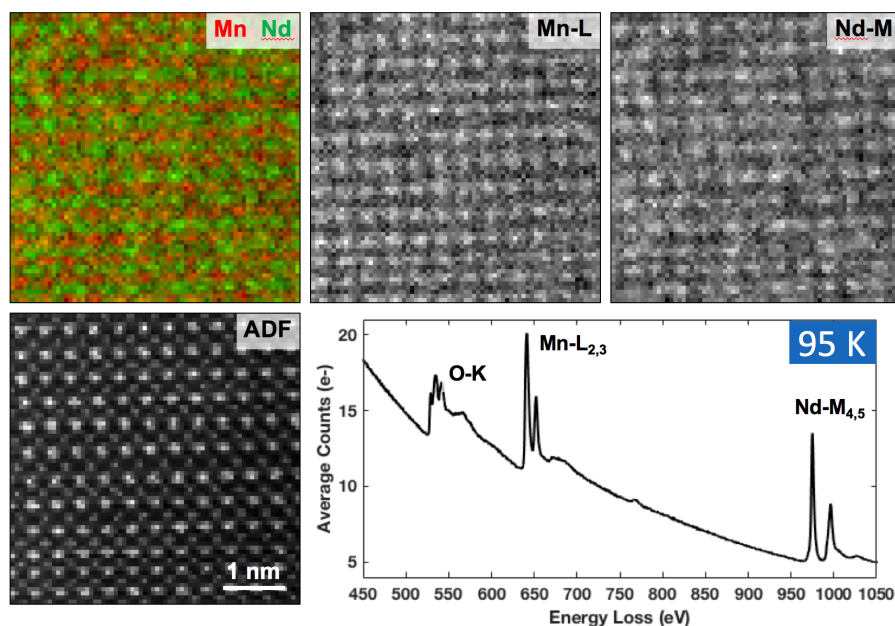


Figure 2. Atomic-resolution spectroscopic mapping of $\text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ at cryogenic temperature. The 70x70 pixel map was recorded in ~13s on a customized FEI Titan Themis with 965 GIF Quantum ER, K2 Summit in counting mode and a LN_2 double-tilt cryo-holder. Compared to traditional indirect detectors, the low read-out noise ensures that even at short dwell times (2.5 msec/pixel) the SNR allows mapping of the Mn-L and Nd-M edges. Rapid mapping enables spectroscopic studies at low temperatures.