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Direct Electron Detection for Atomic Resolution *in situ* EELS

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Spectroscopic mapping by STEM-EELS has emerged as a powerful technique for determining the structure and chemistry of a wide range of materials and interfaces with atomic resolution [1-3]. At the same time, developments in instrumentation and data processing have opened the door to microscopy experiments across a broad phase space of materials physics, including *in situ* heating and cooling, electrical biasing, and mechanical straining [4]. Uniting these two fields of microscopy offers the potential to explore fundamental phenomena down to the unit cell response. Dedicated side-entry holders provide the flexibly of applying a range of external stimuli to the sample, but often suffer from significantly reduced stage stability. *In situ* cryogenic cooling using side-entry holders, for example, can result in significant sample drift even after a two-hour holder settle period. This is particularly detrimental for atomic-resolution spectroscopic mapping due to stringent stability requirements over minutes-long acquisitions. As such, atomic-resolution *in situ* cooling EELS experiments have remained largely out of reach. However, recently developed direct electron detectors (DED) count individual electrons without intermediate photon conversion, offering improved detective quantum efficiency (DQE), narrower point spread function (PSF), and superior signal-to-noise ratio (SNR) as compared to traditional CCD cameras [5, 6]. These improvements combined with a faster readout speed enable atomic resolution EELS experiments previously prevented by reduced stage stability or by other limitations to SNR such as a material's low tolerable electron dose.

Here, we demonstrate the use of a Gatan K2 Summit DED for variable temperature atomic-resolution spectroscopic mapping. Using a short dwell time of 2.5 ms/pixel, we overcome the significant reduction in mechanical stability of the cryogenic side-entry holder to acquire atomically resolved spectroscopic maps of a $La_{0.8}Sr_{0.2}MnO_3$ / $SrTiO_3$ interface near liquid nitrogen temperature with high enough signal and spectral dispersion to preserve detailed O-K edge fine-structure (Fig. 1). Beyond drift limitations, many other experiments are dose-limited due to beam induced sample damage: the improved SNR of the direct EELS detector allows us to obtain atomic resolution maps of a beam-sensitive Sr_2RuO_4 / $NdGaO_3$ sample using a low beam current of 20 pA and 5 ms/pixel dwell time (Fig. 2). The DED's ability to yield higher SNR data in less overall time than a standard CCD detector operated under identical experimental conditions (Fig. 3) will be critical for acquisition- and stability-limited *in situ* EELS experiments. [7]

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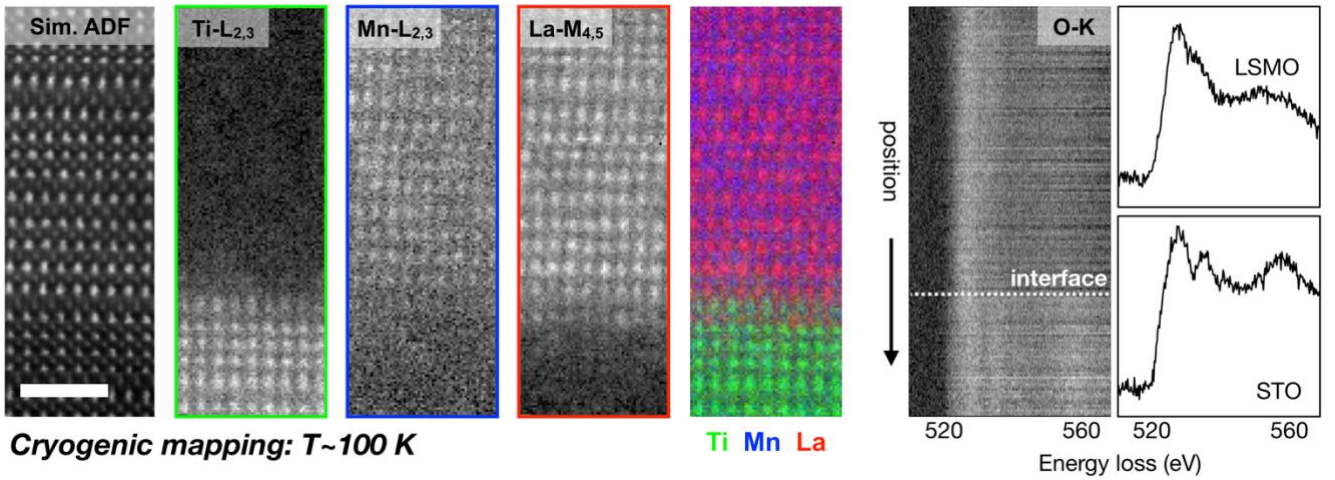

Fig. 1 Atomic resolution elemental maps acquired at cryogenic temperature using a side-entry liquid nitrogen sample holder. The spectrum image is acquired in only 32 sec. A dispersion of 0.5 eV/ch covers an energy range of 1855 eV while preserving enough signal to clearly resolve the O-K edge fine-structure in the LSMO film and STO substrate. Dwell time 2.5 ms/px, beam current ~150 pA. Scale bar 2 nm.

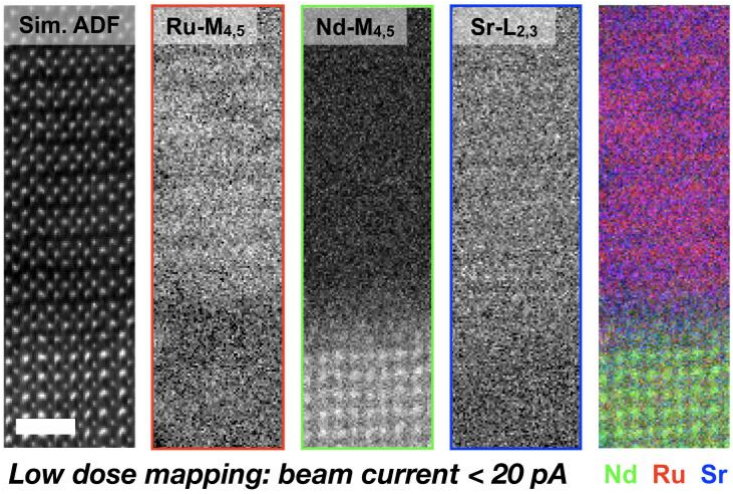

Fig. 2 (above) Improved SNR of the DED enables spectroscopic mapping of the beam-sensitive Sr_2RuO_4 film using a beam current <20 pA. Edges ranging from the slow onset Ru-$M_{4,5}$ at 279 eV to the Sr-$L_{2,3}$ at 1940 and 2007 eV are captured simultaneously without the need for dual EELS acquisition. Scale bar 2 nm.

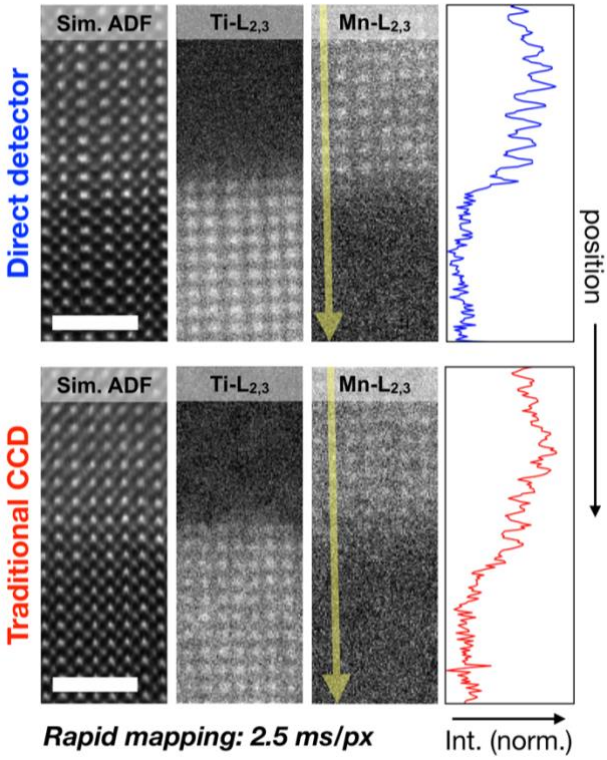

Fig. 3 (right) Identically acquired spectrum images of an LSMO / STO interface using a DED and a traditional CCD. Normalized line profiles (far right) through a single row of Mn atoms from each map (yellow arrows) clearly show the increase in atomic column fringe contrast by up to 40% resulting from the improved SNR of the DED. Simultaneous ADF images show that both maps were acquired under comparable probe conditions. Each data set was the same number of pixels and used the same 2.5 ms/px dwell time, but the total acquisition time on the CCD was 25% longer than on the DED due to increased readout (non-active) time. Scale bar 2 nm.

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