Enhanced Sensitivity of Atomic-Resolution Spectroscopic Imaging by Direct Electron Detection

David J. Baek¹, Berit H. Goodge², Di Lu³, Yasuyuki Hikita⁴, Harold Y. Hwang^{4,5}, and Lena F. Kourkoutis^{2,6}

¹ School of Electrical and Computer Engineering, Cornell University, Ithaca, NY, USA.

². School of Applied and Engineering Physics, Cornell University, Ithaca, NY, USA.

³ Department of Physics, Stanford University, Stanford, CA, USA.

⁴ Stanford Institute for Materials and Energy Sciences, SLAC, Menlo Park, CA, USA.

^{5.} Department of Applied Physics, Stanford University, Stanford, CA, USA.

⁶ Kavli Institute at Cornell for Nanoscale Science, Cornell University, Ithaca, NY, USA.

With recent technical advances in spectroscopic imaging using electron energy loss spectroscopy (EELS) performed in an aberration-corrected STEM, we are now able to study the chemical and electronic structures of materials with atomic-resolution [1]. Challenges, however, remain; particularly for applications that require short acquisition times or those that are limited to low electron dose such as the mapping of beam sensitive materials. Improving the signal-to-noise ratio (SNR) of spectra recorded under low-dose conditions is, therefore, imperative for a range of spectroscopic applications. Here, we demonstrate the use and benefits of a direct electron detector (DED) for atomic-resolution spectroscopic mapping. Compared to traditional indirect detectors, these DEDs use active pixel sensor technology for direct detection of electrons without the use of an intermediate scintillator. As a result, DEDs offer improved detective quantum efficiency (DQE), narrow point spread function (PSF), and superior SNRs in dose-limited imaging applications [2-4].

We perform spectroscopic mapping at 300 kV on an aberration-corrected FEI Titan Themis equipped with a 965 GIF Quantum ER and a Gatan K2 Summit operated in electron counting mode. To test the performance of the set-up, a 150x150 pixel map was recorded on the perovskite oxide Nd_{0.5}Sr_{0.5}MnO₃ (NSMO) with a dwell time of 5 msec/pixel and a beam current of 100 pA. The benefit of low read-out noise and narrow PSF are visible in the electron energy loss spectrum shown in Figure 1a, which is the O-K edge background subtracted spectrum summed over the full map (total dwell time <2min). The low read-out noise ensures that even at a dwell time of 5 msec/pixel, a high SNR is observed across the full spectrum, including minor edges such as the Mn-L₁ and Nd-M₃ edges. In addition, the Sr-L edge at an energy loss of ~1940 eV is also clearly identified. Due to the size of the chip (*i.e.*, 3710 channels), an energy range of >1800 eV is available at a moderate dispersion of 0.5 eV/ch, revealing all elements in a single spectrum in the case of NSMO. Importantly, the near edge fine structure remains visible because of the narrow PSF of the detector (Figure 1b). Finally, with greatly reduced read-out noise, standard powerlaw background subtraction can be performed accurately and atomic-resolution elemental maps can be extracted reliably including those at high energy losses such as the Sr-map (Figure 1c). The enhanced detector sensitivity in EELS is expected to play a large role for applications that require low electron dose or short dwell times such as low temperature experiments [5].

[1] DA Muller et al, Science 319 (2008), p.1073-1076.

- [2] RS Ruskin, Z Yu, and N Grigorieff, J. Struct. Biol. 184 (2013), p. 385.
- [3] G McMullan, AR Faruqi, D Clare, R Henderson, Ultramicroscopy 147 (2014), p. 156.
- [4] JL Hart, AC Lang, C Trevor, R Twesten, ML Taheri, Microsc. Microanal. 22 (S3) (2016), p. 336.

[5] This work was supported by the Cornell Center for Materials Research with funding from the NSF (DMR-1120296 and DMR-1429155).



Figure 1. (a) EELS edges from all elements present in $Nd_{0.5}Sr_{0.5}MnO_3$ are recorded simultaneously using a 965 GIF Quantum ER plus K2 Summit in counting mode at a dispersion of 0.5 eV/ch. The large number of pixels available in the dispersive direction (*i.e.* 3710 pixels) gives access to an energy range of 1855 eV at 0.5 eV/ch. (b) O-K, Mn-L, and Nd-M edges from (a) after background subtraction. (c) Atomic-resolution elemental maps (150x150 pixels) obtained from the Mn-L, Nd-M, and Sr-L edges marked by dotted squares in (a). Due to the high signal-to-noise ratio in counting mode, even a short dwell time of 5 msec/pixel allows the extraction of the high-energy Sr-map at ~1940 eV with clear atomic contrast. (d) False colored images constructed from elemental maps in (c). Far-right map of Nd and Sr reveals local variations in relative doping across the sample.