STEM imaging of the third dimension using Laue zone scattering at atomic resolution

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We have recently shown that scattering into higher order Laue zone (HOLZ) rings can be separated from the rest of the high angle scattering in a STEM dataset using a fast pixelated detector to record diffraction patterns at every scan point [1]. This can then be used as a signal in STEM imaging to reveal the spatial variation of a specific periodic ordering along the beam direction, thus in principle allowing 3 dimensional local crystallography to be mapped from a single projection. However, in our previous work, the beam was too broad for atomic resolution, due to the difficulties with working with a detector on a fixed mount in the 35 mm camera position on our microscope. In this work, the newly commissioned JEOL Grand ARM300F at the ePSIC facility was used with a Medipix detector mounted in a fixed bottom mount position, allowing easier microscope tuning before using the pixelated detector. The fast readout of the detector was performed using a Merlin system (Quantum Detectors Ltd., Harwell, UK) and specialized scripts were used for the data handling and live imaging: https://www.gla.ac.uk/schools/physics/research/groups/mcmp/researchareas/pixstem/
In Figure 1a, it is clearly shown that atomic resolution imaging has now been achieved using pixelated STEM on a bilayer of $La_{0.7}Sr_{0.3}MnO_3$ on $LaFeO_3$ on a $SrTiO_3$ substrate [2] - in this case the diffraction patterns have simply been processed to produce a straightforward HAADF contrast. Unsurprisingly, the two La-containing layers both show similar brightness, on account of the similar atomic number of the A-site atoms. In Figure 1b, the intensity in the innermost HOLZ ring is plotted. This innermost HOLZ ring is only present in the $LaFeO_3$ and arises from a doubling of the unit cell along the beam direction. In contrast to Figure 1a, the main peaks of intensity are even more on the A-site atoms, and there is very little intensity around the B-site positions. Such imaging reveals the spatial variation across the unit cell of the atomic modulations that lead to period doubling, and demonstrates a method whereby the three dimensional crystallography can be both imaged qualitatively and investigated quantitatively on a length scale of individual unit cells.

References

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[2] I Hallsteinsen *et al.*, Phys. Rev. B, **94** (2016) 201115.

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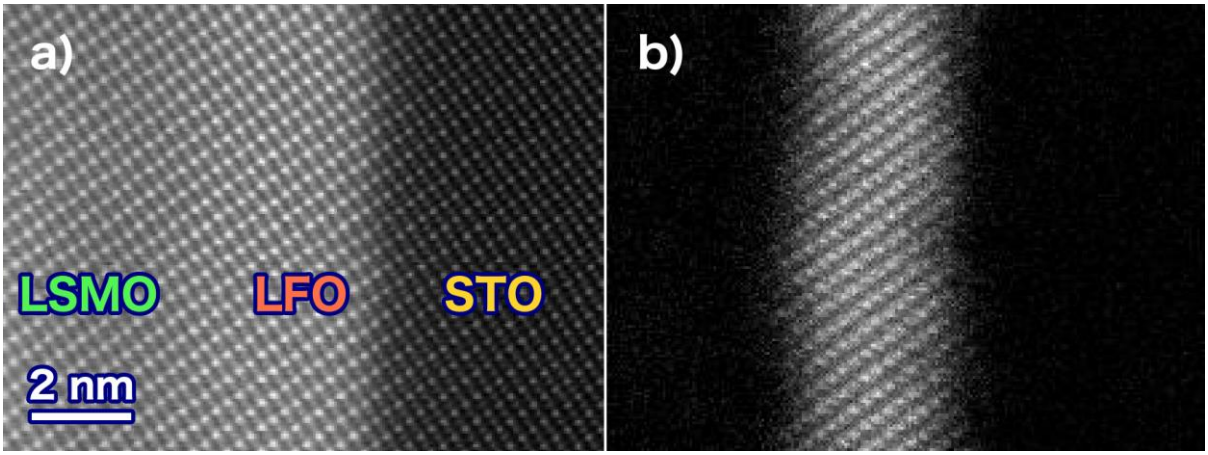

Figure 1: Atomic resolution pixelated STEM imaging: a) HAADF image; b) image from the intensity in the innermost HOLZ ring.