Improving the SNR of STEM EELS & EDX Mapping while Reducing Beam-damage using Non-rigid Spectrum-image Averaging

Lewys Jones*[1], Richard Beanland[2] Sergio Lozano-Perez[1], Karim Baba-kishi[3] and Peter D. Nellist[1]

1. Department of Materials, University of Oxford
2. Department of Physics, University of Warwick
3. Department of Applied Physics, Hong Kong Polytechnic University

Background

Spectrum imaging in the scanning transmission electron microscope can deliver compositional information about materials down to the atomic scale. However, with EELS cross-sections around 100 smaller than those for imaging, and those for EDX being 10,000 times smaller, spectrum images often exhibit low SNR and with long collection times show significant imaging artefacts (Figure 1). When scan deviation fluctuates with the slow-scan direction at around 0.1 – 5 Hz, this is known as scanning distortion [1]. This scan-distortion corrupts the lattice parameters and angles in atomic resolution imaging and composition mapping and limits the signal-noise ratio of data.

Aiming to improve SNR we could increase the dwell-time, though this would worsen the appearance of scanning-distortions, stage-drift or focal-drift. Impasse. The operator could increase beam-current, but this may lead to beam damage [2], increase carbon contamination, or worsen the resolution.

Alternatively here, instead of increasing either probe-current or dwell-time, signal is accumulated by recording multiple spectral images. Image drift and scan-distortion is measured from the simultaneously acquired ADF images, but applied through the spectral volumes.

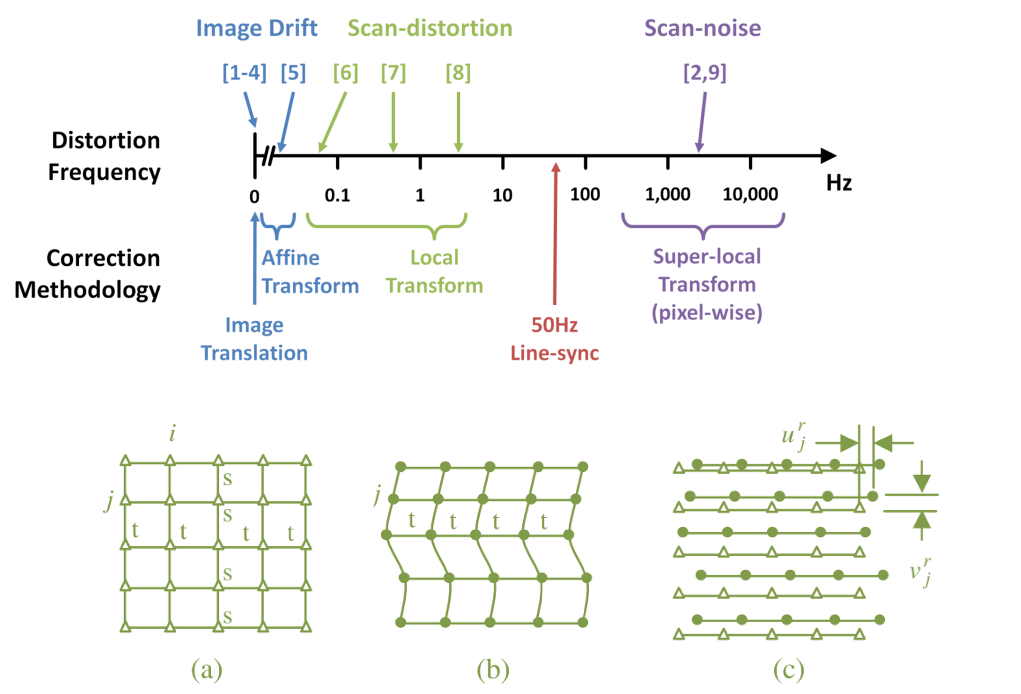


Figure 1. Top: Summary of frequencies of different image distortion types (from [1]). Bottom: Comparison of ideal and experimental rasters: a) a perfect scan-grid over equally spaced rows, s, and times, t; b) In the presence of environmental distortions the fast-scan spacing t remains approximately constant but the offsets between rows may become corrupted. c) Comparing the differences between a) and b) then describes the environmental disturbance, but also the transformation required to undistort the data back to a true scan. Schematic modified from [3].

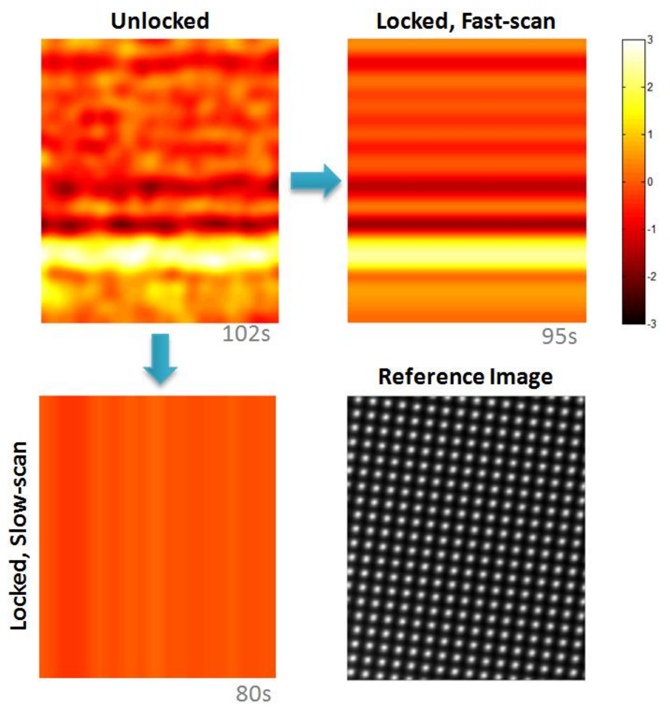
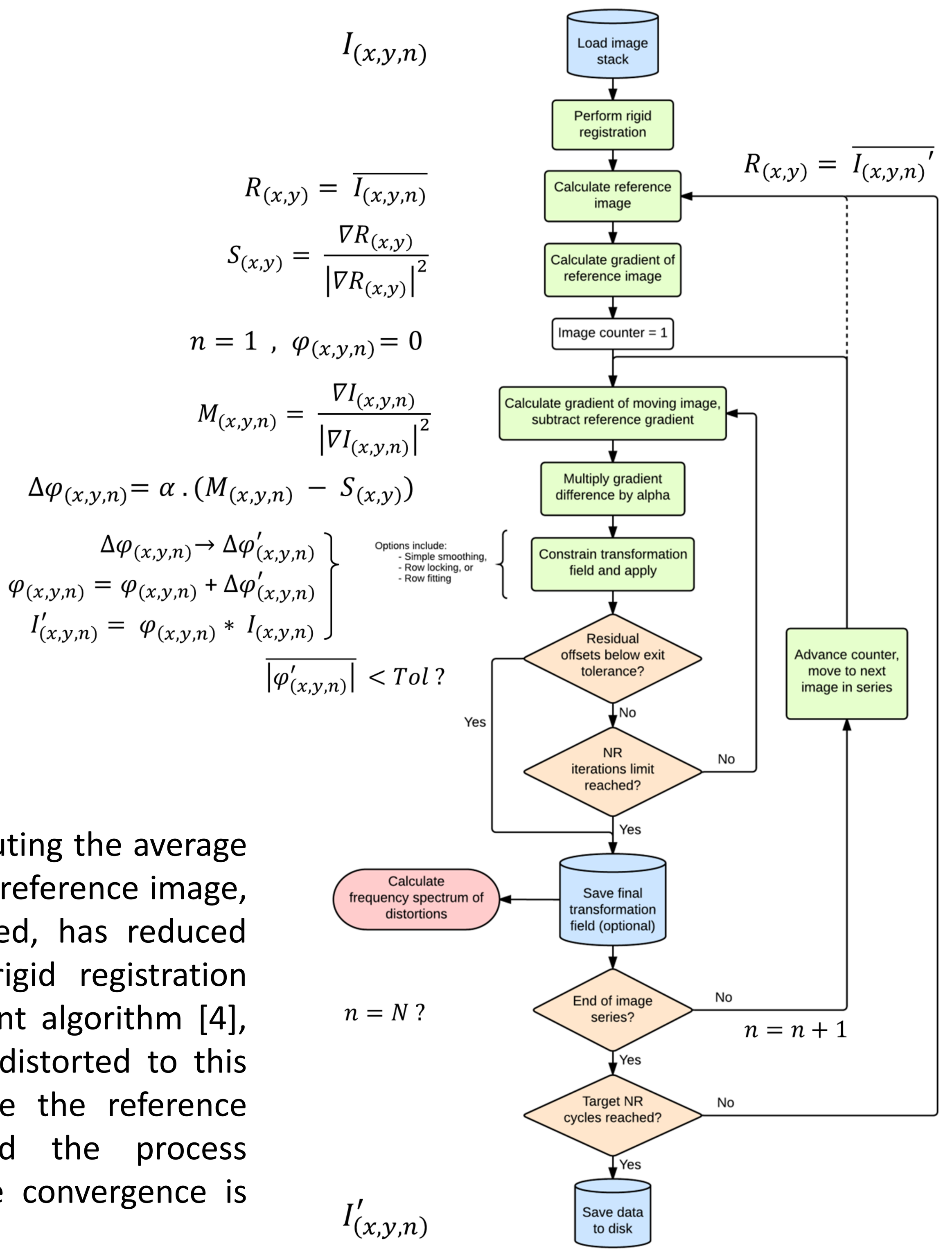


Figure 2. Experimentally measured transformation field from an ADF image series of [100] MgO (y direction shown only). Top-left: so called un-locked field, where motion is unconstrained. Top-right, the same filed but constrained by the prior knowledge of the STEM scan system [1].

The Smart Align Algorithm

Scan distortion manifests as a bending of what should be regular lattice planes. We could enforce this prior knowledge and geometry, but this would destroy potentially valuable strain information. Instead by recording a time-series stack of images we can observe the consistent (genuine sample) and time varying (scan-distortion) information. The separation of varying and constant image detail is the task of the Smart Align software, the core algorithm of which is represented in Figure 3.

Figure 3. Key elements from the core algorithm of the Smart Align software; full details and symbol definitions in [1].

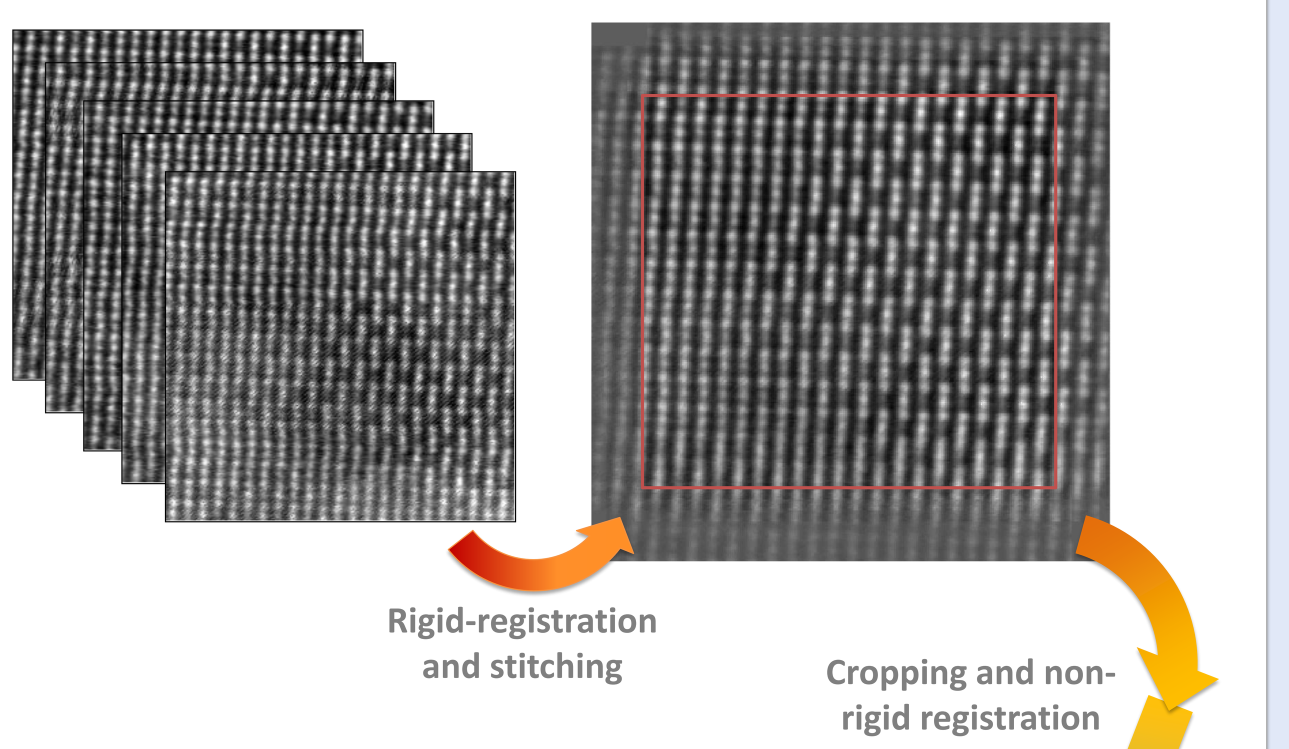


This is based on first computing the average of all images to use as the reference image, R. This average, while blurred, has reduced distortion. Next using non-rigid registration based on the gradient descent algorithm [4], the individual frames are undistorted to this reference image. From these the reference image is recalculated, and the process repeated until an acceptable convergence is reached.

Spectrum Image Quality Improvement

To test our new approach two data-sets were recorded (Figure 1); a single 256x256 spectrum image with a dwell-time of 0.01 s/pix, and for comparison five separate 0.002s/pix spectrum images (same total electron dose). The sample used was Pb_2ScTaO_6 , which exhibits incomplete long-range ordering on the Sc:Ta sublattice. Simultaneous ADF images and hardware-synced spectra were recorded using a Gatan Quantum GIF and an Oxford Instruments X-max 80mm^2 detector near an anti-phase boundary. To produce maps the EELS data were de-noised using principle component analysis before edge extraction. For the EDX maps characteristic x-rays were integrated after subtracting a linear background.

Figure 4. Rigid registration is used to coarse realign frames (here 5 images with 256x256pix) before the common areas is cropped (225x214 pixels) and the new trimmed image stack is sent for non-rigid registration. On a standard desktop PC this takes around 10 minutes, up to several hours for 2048x2048x40 frames.



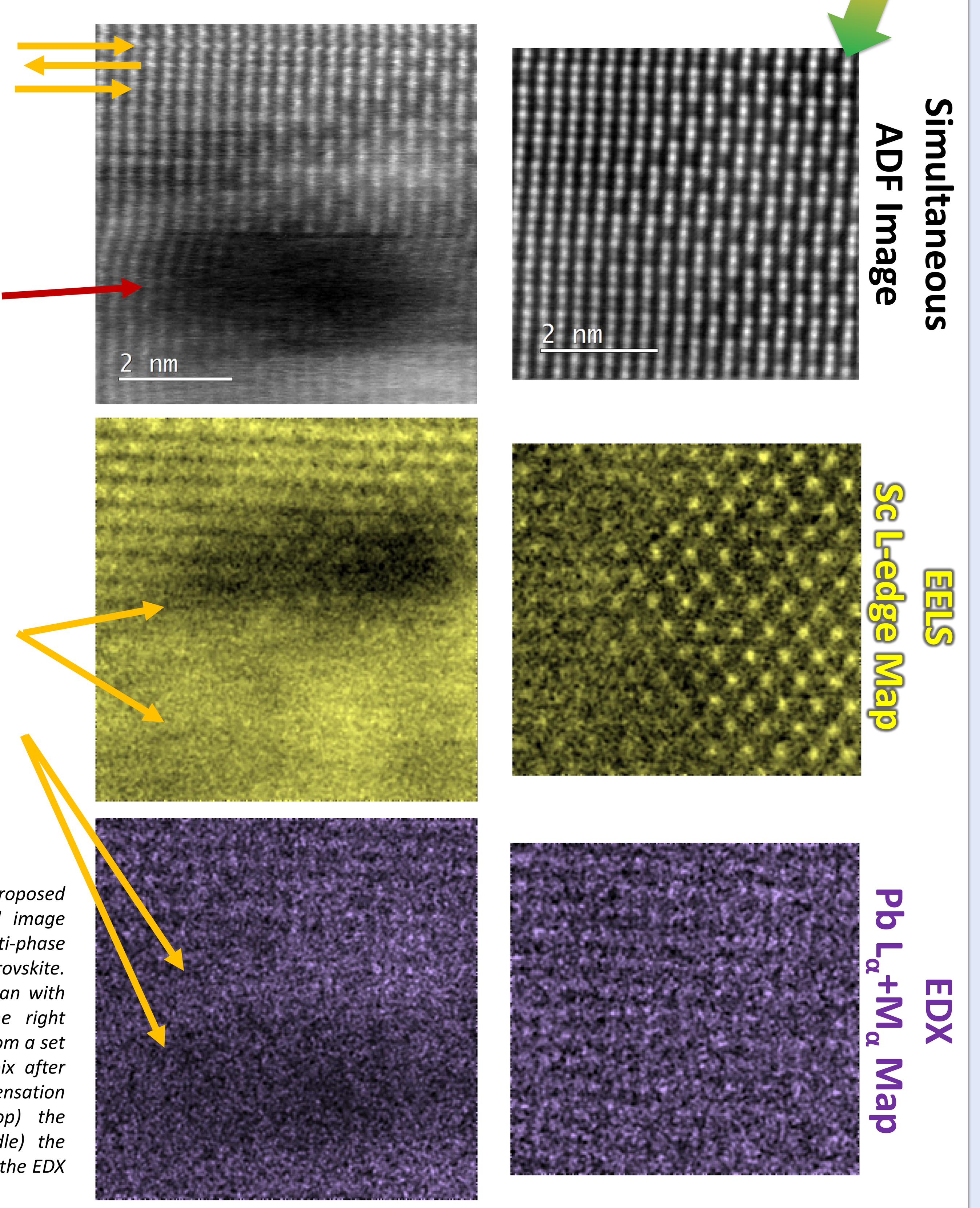


Figure 5. Conventional and proposed approaches to acquiring spectral image data. Example shows an anti-phase boundary region in a Pb_2ScTaO_6 perovskite. The left column shows a single scan with 256x256 pixels at 0.01s/pix. The right column shows the restored data from a set of 5 frames scanned at 0.002s/pix after non-rigid scan-distortion compensation and averaging. Rows show (top) the simultaneous ADF images, (middle) the EELS Sc L-edge map, and (bottom) the EDX Pb Lα+Mα map.

The slower scan speed ADF shows scanning-distortions and significant beam-damage. The fast-scanned, distortion-corrected, and averaged ADF image however shows improved SNR and straight lattice planes as expected. The location of the anti-phase boundary is now clearly resolved. The SNR of the Sc L-edge EELS maps show an improvement from 2.1 to 5.1 with the Sc positions relating directly with the ADF image. EDX maps are less clear but in the multi-frame average spectrum image the Pb lattice begins to become apparent. As sample damage is reduced so drastically, more frames could be recorded to improve SNR further if desired. This was not done here to allow a fair comparison at fixed total dose conditions.

Conclusions

By sharing the same electron dose across several fast frames we have produced compositional maps with distortion free crystal lattices, reduced beam-damage and improved SNR. This new approach allows for more beam-sensitive samples to be mapped, SNR to be increased and/or field-of-view and sampling to be improved.

A demo version of the image registration software (requires MatLab) is available free of charge for academic / non-commercial use from **www.lewysjones.com** .

References

1) L. Jones et al., "Smart Align – A new tool for Robust Non-rigid Registration of Scanning Microscope Data," *Adv. Struct. Chem. Imaging*, **1**, (2015).
2) R. Egerton, P. Li, and M. Malac, *Micron* **35** (2004), p.399–409.
3) Y. Sun and J. H. L. Pang, "AFM image reconstruction for deformation measurements by digital image correlation.," *Nanotechnology*, **17**, p.933–9, (2006).
4) P. Cachier, X. Pennec, and N. Ayache, "Fast Non Rigid Matching by Gradient Descent: Study and Improvements of the 'Demons' Algorithm," (1999).

Acknowledgments

The authors would like to acknowledge ESTEEM2 (EU-FP7 grant agreement 312483) for financially supporting this work.

* lewys.jones@materials.ox.ac.uk

Copies of this poster, the software described and the associated manuscript are available online at: **www.lewysjones.com**