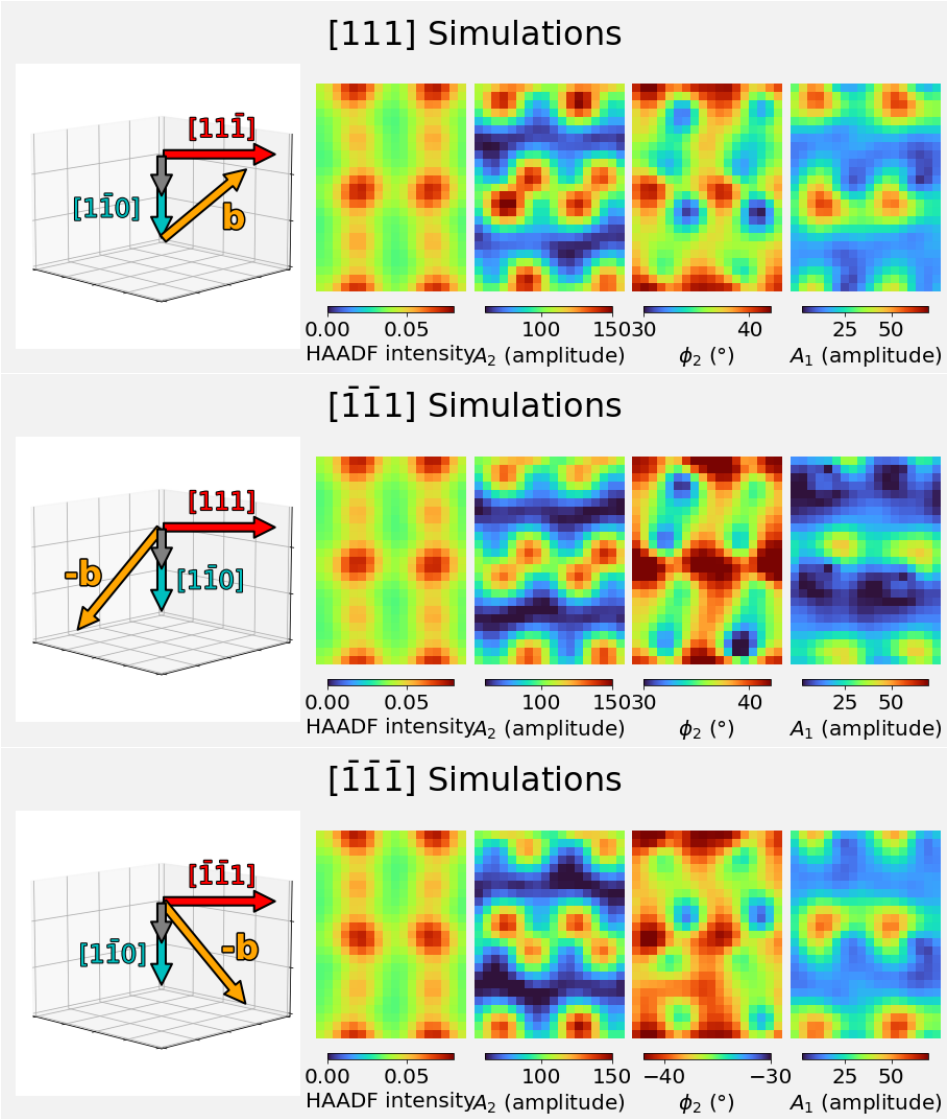
Atomic resolution imaging of 3D crystallography in the scanning transmission electron microscope

Ian MacLaren[1], Aurys Silinga[1], Juri Barthel[2], Josee Kleibeuker[3,4], Judith L MacManus-Driscoll[3], Christopher S Allen[5,6], Angus I Kirkland[5,6]

1. School of Physics and Astronomy, University of Glasgow, Glasgow G12 8QQ, UK
2. Ernst Ruska-Centre (ER-C 2), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany
3. Department of Materials Science & Metallurgy, University of Cambridge, 27 Charles Babbage Road, Cambridge CB3 OFS, UK
4. DEMCON high tech systems Eindhoven BV, Kanaaldijk 29, 5683 CR Best, The Netherlands
5. electron Physical Science Imaging Centre, Diamond Light Source Ltd., OX11 0DE, UK
6. Department of Materials, University of Oxford, Parks Road, Oxford OX1 3PH, UK

Demonstration that this mapping works consistently for $\langle 111 \rangle_{monoclinic}$ directions



Figs S1-S3: Fitting results for 4DSTEM simulations for three beam directions representative of all $\langle 111 \rangle_{monoclinic}$ directions. Detailed commentary given in the text.

Simulated 4DSTEM datasets were calculated for several different $\langle 111 \rangle_{monoclinic}$ directions corresponding to the main distinct ones in this monoclinic structure. It is in a standard setting with $\beta \neq 90°$, thus, there is basically no difference between $[uvw]$ and $[u\bar{v}w]$. There is, however, a significant difference between $[uvw]$ and $[uv\bar{w}]$ because the **a** and **c** axes are not orthogonal. Thus, $[111]$ and $[\bar{1}\bar{1}1]$ represent the two distinct types of $\langle 111 \rangle_{monoclinic}$ directions. Including $[\bar{1}\bar{1}\bar{1}]$ demonstrates the effect looking along a direction in the opposite sense.

The first main point to notice is that very similar features come out in all maps. Note, the ϕ_2 is measured clockwise from the vertical direction in all maps. This is reversed (as is the colour map) in Fig S3 where the "dumbbell" structure is pointing anticlockwise from vertical. So this parameter always tracks with the appearance of the maps. The really important point is that the direction in which the **b** axis (or -**b**) direction points up out of the sample plane is always in the direction from the A_1 peaks to the HAADF peaks.

There is a minor difference between $[111]$ and $[\bar{1}\bar{1}1]$ analyses in that the variations in the ϕ_2 and A_1 maps are slightly stronger for the latter, presumably slight differences in the angle of the displacement vector, **d**, to these directions causes a stronger effect in one case than the other.

A note on the 3D vector plots – these are done to roughly indicate the 3D direction of the vectors (although are done using a pseudocubic approximation of the axis lengths). To allow visibility of the grey Beam direction vector, these are tilted down by 11° so are not quite along the same direction as the Beam direction for the 4DSTEM datasets but are there to show the relative orientation of the key vectors in each image. In each case, the correct indices for the horizontal right direction (in monoclinic vectors) are given in red, and those for the vertical down direction in cyan. Beam directions are calculated with the normal convention for electron microscopy as being vertical upwards from the page, antiparallel to the direction of electron motion and parallel to the direction of current flow.

Large area mapping of same film (not at atomic resolution)

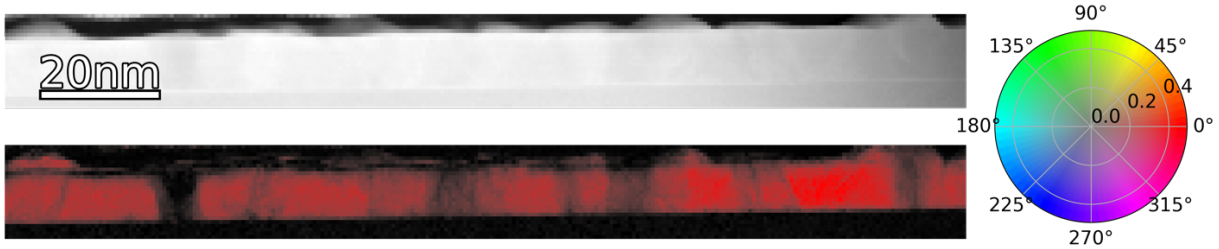


Figure S4: Upper: HAADF image of the large area of this film with carbon to the top, film in the middle, and LSAT substrate at the bottom. Lower: large area mapping of the direction of the **d***-axis for this film of LCMO on LSAT. All colour mapping of directions referenced to the image axes.* ***d*** *consistently lies close to in-plane suggesting that the* ***b*** *axis is in-plane for much of the film.*

Applying the method of this paper to a large area 4DSTEM dataset from the same film results in the following map in Figure S4. This shows that almost all areas have **d** laying in-plane at some angle to the specimen plane, suggesting the **b** lays preferentially in-plane. Some areas are dark in this map, which indicates regions where the inner FOLZ disappears and must correspond to other crystal orientations (separate evidence from low convergence angle nanobeam electron diffraction suggests these are [010] oriented domains). There is no evidence in the whole dataset for any area where the **b**-axis does not lie in the film plane.