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Optical Sectioning with Atomic Resolution Spectroscopy

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Aberration correction allows the use of larger probe forming apertures in scanning transmission electron microscopy (STEM). Larger apertures provide a reduced depth of field that can be exploited through optical sectioning to provide three-dimensional structural and compositional information. The depth of field is roughly the wavelength of the electron divided by the square of the aperture convergence angle [1]. So, for example, an instrument operated at 200 kV with a 22 mrad convergence angle has a depth of field of approximately 5 nm. Such nm scale depths of field have previously been utilized to determine the three dimensional locations of individual dopant atoms and to image the structure of dislocations with annular dark field (ADF) imaging.

ADF imaging can provide compositional information through the approximately $Z^{1.7}$ dependence of the intensity, where Z is the atomic number. However it remains difficult to differentiate heavy elements with similar atomic numbers. In such situations electron energy loss spectroscopy (EELS) is often employed. EELS provides robust compositional analysis and with aberration correction two dimensional atomic resolution elemental mapping.

Atomic lateral resolution turns out to be the key to longitudinal resolution. Due to the large missing cone in the STEM 3D optical transfer function, a nanometer scale depth of field does not necessarily result in nanometer longitudinal resolution [1]. The longitudinal resolution varies with the lateral spatial frequency. In regions relevant to lateral resolutions achievable today, the missing cone causes the longitudinal resolution to vary as approximately d/α where d is the characteristic spacing of the object and α is the convergence angle. If the highest spatial frequency resolved is for example the width of a 3 nm nanoparticle, the depth resolution will be around 136 nm with a 22 mrad convergence angle. If instead we were able to resolve atomic columns with a spacing of 0.3 nm, the depth resolution improves to 13.6 nm for a 22 mrad convergence angle. This can be improved further by using a larger convergence angle, for instance a 30 mrad convergence angle provides a 10 nm depth resolution for the same lateral spacing. This relationship between lateral spatial frequency transfer and depth resolution applies both to ADF and EELS imaging. However, although theoretical simulations suggested it was possible to perform optical sectioning with EELS, it had not previously been demonstrated experimentally.

We will present EELS optical sectioning which displays clear depth sensitivity. Recently we used EELS optical sectioning to reveal the presence of a network of yttria-stabilized zirconia (YSZ) islands buried beneath strontium titanate (STO). Using a Nion UltraSTEM 100 operated at 100 kV with a 30 mrad convergence angle we acquired successive spectrum images from the same area of the sample, but with the probe focused to different depths (Figure 1). An ADF image is also acquired simultaneously with

each spectrum image. Due to channeling effects the clearest images are typically formed when the probe is focused at the entrance surface of the material. This is certainly true for the ADF images. At the entrance surface the ADF contrast appears STO like, while 18 nm into the depth of the sample a region of YSZ like contrast appears. The Sr and Ti elemental maps also appear clearer at the entrance surface. The Zr maps exhibit the opposite dependence on depth. When the probe is focused at the entrance surface, Zr map appears nebulous and no atomic columns are resolved. When the probe is refocused 18 nm into the depth of the sample however, a Zr lattice comes into focus. The Zr columns have minimum transverse separation of about 0.28 nm, corresponding to a depth resolution of about 9 nm as calculated from the 3D contrast transfer function. The Ti-Ti and Sr-Sr spacings are larger at 0.39 nm and therefore are transferred with a slightly poorer depth resolution of approximately 11 nm. We will also present further experimental developments in spectroscopic optical sectioning and image simulations [2].

References:
[1] G. Behan *et al,* Phil. Trans. R. Soc. A **367** (2009), p. 3825.
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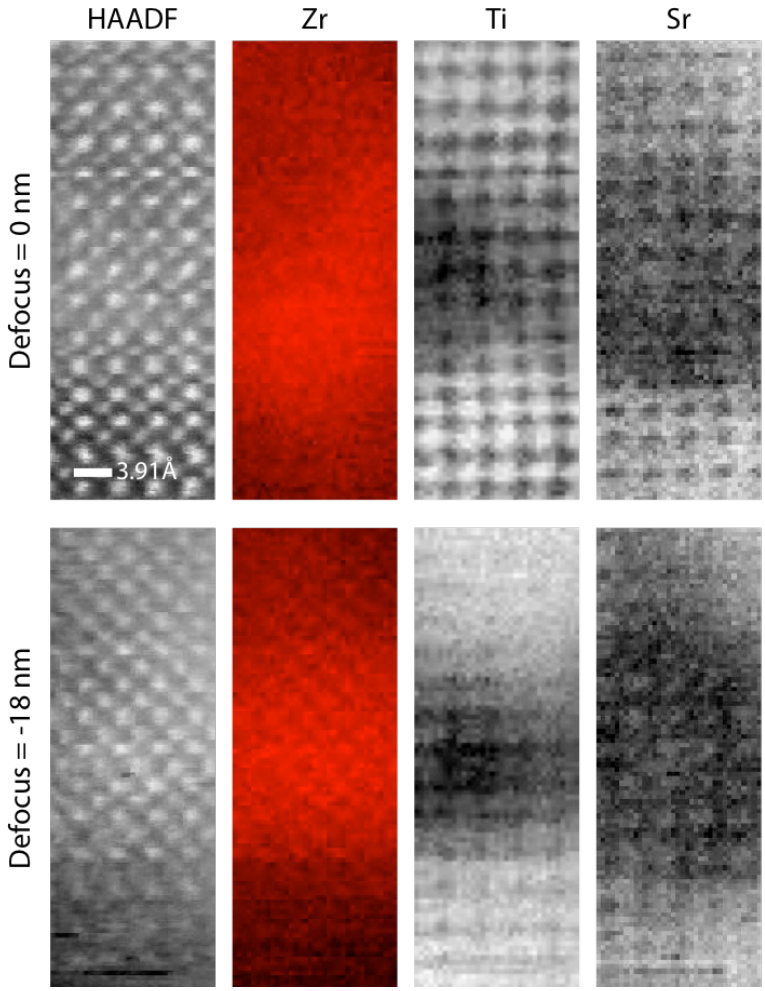

Figure 1. Optical sectioning with atomic resolution spectrum imaging. The Sr (M-edge), Ti (L-edge) and Zr (M-edge) maps have been denoised with principle component analysis, and are shown alongside the simultaneously acquired high angle annular dark field (HAADF) images. The images in the top row were acquired with the probe focused to the entrance surface of the sample, while those in the bottom row were acquired from the same area but with the probe focused 18 nm deeper into the material. The Zr lattice appears in focus only when the probe is focused deeper within the sample, indicating it is buried under STO.

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