Applications of Quantitative STEM to Functional Oxides and Interfaces

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High-angle annular dark-field (HAADF) scanning transmission electron microscopy (STEM) is highly sensitive to both the atomic number (Z) and the Debye-Waller factor of the atom columns. We will briefly discuss the development of quantitative understanding of HAADF imaging in STEM to obtain chemical information along with the atomic structure. We show that because of the strong influence of electron channeling and thermal diffuse scattering, an intuitive interpretation of the image contrast in terms of the atomic numbers present in an atom column is not always possible.

An important method that enabled quantitative STEM is position averaged convergent beam electron diffraction (PACBED). PACBED can reliably measure sample thicknesses to better than 10%, specimen tilts to better than 1 mrad and sample polarity for the same electron optical conditions and sample thicknesses as used in atomic resolution scanning transmission electron microscopy imaging. Illuminating a zone-axis sample with a highly convergent electron probe, as used for STEM imaging, results in a CBED pattern containing overlapping orders of diffracted disks. These overlapping disks generate a complicated interference pattern that is highly dependent on the position of the finely focused (< 0.2 nm) electron probe within the unit cell. Figure 1 shows, as an example, simulations of such patterns for an Ångstrom-size probe focused on different atom columns in a $SrTiO_3$ unit cell.

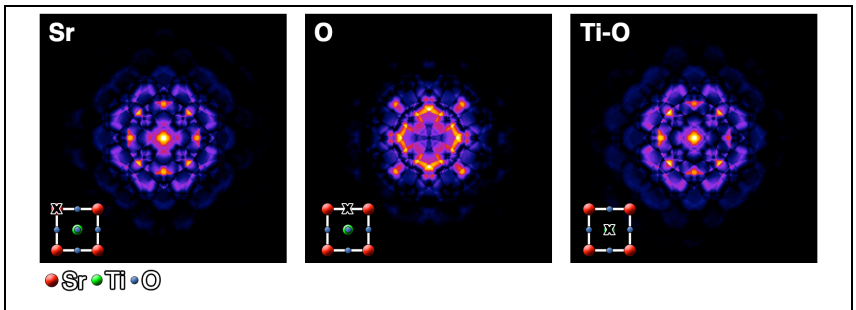

Figure 1. Simulations of coherent CBED patterns for $SrTiO_3$ along <100>, for different positions of the STEM probe within the unit cell (as indicated in the unit cell schematics in the lower left corner).

A PACBED pattern is formed through incoherent averaging of CBED patterns over many probe positions. The fine detail resulting from coherent interference is then lost, but better count rates and a reduced sensitivity to instability are obtained. The

experimental PACBED pattern is highly sensitive to the sample structure, thickness and tilt, which can be quantitatively extracted through comparison with simulated PACBED patterns. The theory for simulating CBED patterns is well established and software packages are available. An analytical depiction of PACBED has also been developed. It has been shown that the PACBED pattern is independent of coherent aberrations of the probe-forming lens and of spatial incoherence. Comparisons between PACBED experiments and simulations allow for determination of the local sample thickness, in regions as small as a few unit cells.

In this presentation, we will discuss important applications of PACBED, in particular the determination of specimen polarity in ferroelectric and polar thin films and of the type and magnitude of octahedral rotations in ultrathin perovskite films with few-unit-cell spatial resolution.

References

[1] J. M. LeBeau, S. D. Findlay, X. Wang, A. J. Jacobson, L. J. Allen, and S. Stemmer, *Phys. Rev. B* **79** 214110 (2009).

[2] J. M. LeBeau, S. D. Findlay, L. J. Allen, and S. Stemmer, *Phys. Rev. Lett.* **100** 206101 (2008).

[3] J. M. LeBeau and S. Stemmer, *Ultramicroscopy* **108** 1653 (2008).

[4] J. M. LeBeau, A. J. D'Alfonso, S. D. Findlay, S. Stemmer, and L. J. Allen, *Phys. Rev. B.* **80** 174106 (2009).

[5] J. M. LeBeau, S. D. Findlay, L. J. Allen, and S. Stemmer, *Ultramicroscopy* **110** 118 (2010).

[6] J. M. LeBeau, A. J. D'Alfonso, N. J. Wright, L. J. Allen, and S. Stemmer, *Appl. Phys. Lett.* **98** 052904 (2011).