DOI: 10.1017/S1431927608082263
Microsc Microanal 14(Suppl 2), 2008

# Quantitative HAADF-STEM and EELS

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High angle annular dark-field (HAADF) in scanning transmission electron microscopy (STEM) has proven to be a technique that is atomic number sensitive and directly interpretable in terms of the atomic column positions. However, previous attempts to quantify the image contrast have been limited by the lack of an absolute intensity scale. Quantified image contrast could be used for a host of applications where atomic resolution composition mapping is desired. Recently, it was reported that a mismatch between experimental and simulated image contrast exists for HAADF STEM [1]. There are two possible explanations: an overestimated atomically resolved signal [2] or an underestimated background (the intensity between the atomic columns) [1]. Without an absolute measure of image intensities, one cannot distinguish between these two possibilities. Using an annular dark field detector that has single electron sensitivity and an output voltage that is directly proportional to intensity, we report a method to measure the incident probe intensity. For this study, an FEI Titan 80-300 STEM/TEM equipped with a super-twin lens ($C_s \sim 1.2$ mm) operating at 300 kV was used.

In this presentation, we discuss the practical aspects of achieving quantified HAADF imaging. Focus will be placed on characterization of the detector and acquisition/quantification of the image intensities. Limitations of the current generation of HAADF detectors, such as scintillator heating and intensity saturation, will be discussed in the context of the probe intensity measurements. By normalizing the atomically resolved signal to the incident probe, we demonstrate quantified HAADF imaging of a $SrTiO_3$ single crystal. Figure 1 displays an experimental image from a region ~ 200 Å thick and a corresponding Bloch wave image simulation. As can be seen, a quantitative match exists between simulations and experiments after taking into account spatial incoherence.

The electron energy loss spectroscopy (EELS) log-ratio method [3] was used for determination of the local sample thickness. Figure 2 shows EEL spectra that exhibit strong surface effects that lead to an overestimation of thickness by the log-ratio method. We will discuss how thickness determination by EELS can be combined with information from the HAADF background signal to provide improved estimates of the thickness.

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[4] S. S. and J. M. L. acknowledge the U.S. DOE for support of this research (DE-FG02-06ER45994). L. J. A. acknowledges support by the Australian Research Council. S. D. F. is supported as a Japan Society for Promotion of Science (JSPS) fellow.

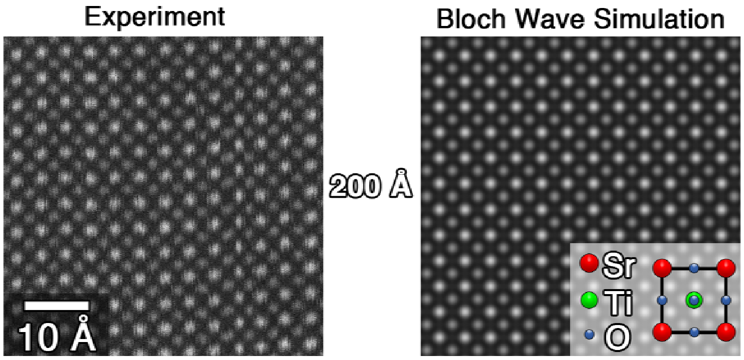

FIG. 1. Experimental HAADF image of $SrTiO_3$ along <100> for a region approximately 200 Å thick (left) compared with the corresponding Bloch wave simulation (right). Spatial incoherence is included in the simulation by convolution using a Gaussian envelope with a FWHM of 0.8 Å. Note that the image intensities in both cases are over the same range and normalized to the incident beam intensity.

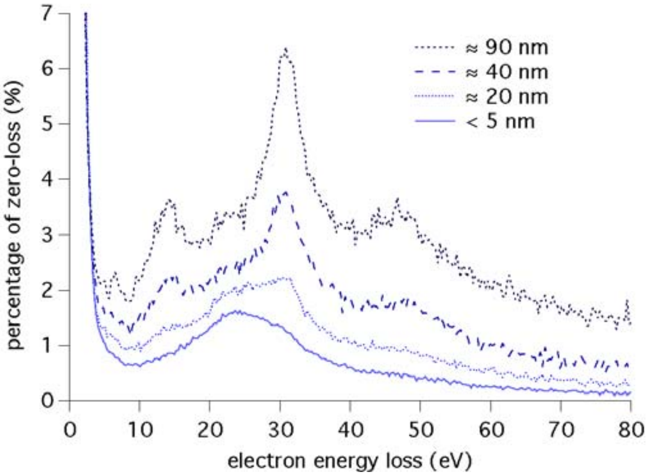

FIG. 2. EELS low-loss spectra normalized to the zero-loss peak for a range of specimen thicknesses. The sample has been etched in HF to remove surface damage from ion milling. As is evident in the very thin region (< 5 nm thick), a strong surface plasmon is observed at ~ 24.3 eV. The existence of the strong surface plasmon peak causes a significant overestimate of the thickness determined from the log-ratio method, $\ln(I_t/I_0) = 0.27$. As the thickness increases, bulk plasmon and interband transition losses are readily observed.