

Quantitative HAADF Imaging of Crystals Containing Heavy Elements: A Comparison with Theory

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Direct comparisons between HAADF simulations and experiments have shown that quantitative agreement can be achieved for single crystalline SrTiO₃ [1, 2]. A previous HAADF comparison study for PbTiO₃, with images acquired on an arbitrary scale, exhibited a significant contrast mismatch relative to simulations [3]. To investigate the possibility of an atomic number dependent contrast mismatch, we present a study of single crystalline PbWO₄, which contains two cations with large atomic numbers ($Z_{\text{Pb}} = 82$ and $Z_{\text{W}} = 74$). An FEI Titan 80-300 kV S/TEM operating at 300 kV was used for HAADF-STEM imaging. The STEM probe forming conditions were set as follows: extraction voltage 4400 kV, gun lens 6, spot size 10, and a convergence semi-angle of 9.4 mrad. The focus was determined using the maximum intensity criterion in both experiments and simulations.

The PbWO₄ crystal was observed down the ideal tetragonal scheelite structure [100] zone axis, see inset of Figure 1 for a projected structure. After normalizing experimental image intensities to the incident probe, we present direct comparisons with simulations. Once spatial incoherence is taken into account, near perfect agreement between simulations and experiments is achieved, shown in Figure 1. The effective source size is demonstrated to be independent of atomic number by comparisons with single crystalline SrTiO₃.

The importance of incorporating accurate Debye-Waller factors for this study will be emphasized. The variation of the image background intensity will be explored to highlight the importance of image simulations to fully appreciate the subtleties of electron scattering by crystals containing heavy elements. In addition, the stability of the Schottky field emission gun will be discussed in the context of the observed finite source size.

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

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

[3] D. O. Klenov and S. Stemmer, *Ultramicroscopy* 106, 889 (2006).

[4] S. S. and J. M. L. acknowledge the NSF for support of this research (grant number DMR-0804631). 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.

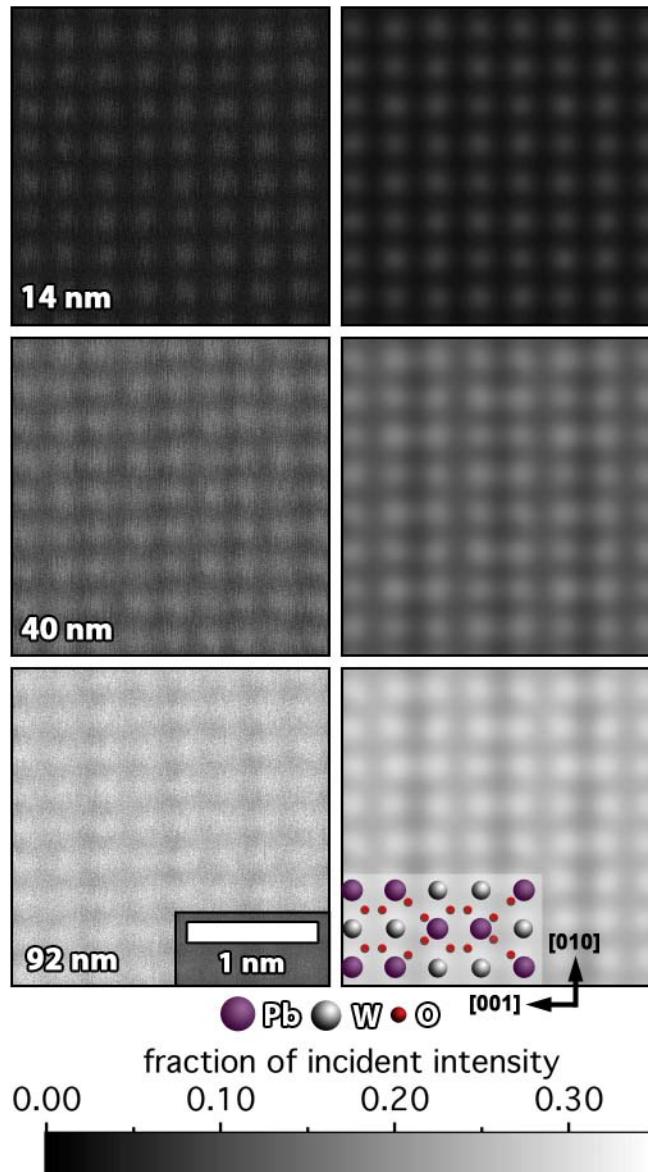


FIG. 1. Experimental (left panels) and simulated (right panels) images for different thicknesses (see labels) of PbWO_4 . The simulated images have the effects of spatial incoherence taken into account.