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Annular Bright Field Scanning Transmission Electron Microscopy – Direct and Robust Atomic-Resolution Imaging of Light Elements in Crystalline Materials

Scott D. Findlay[1]

[1.] School of Physics and Astronomy, Monash University, Melbourne, Australia

Light elements are key constituents of many advanced materials and energy management technologies – oxygen in dielectrics and superconductors, lithium in battery materials, hydrogen in hydrogen storage materials – making their reliable imaging an important goal for atomic-resolution structure analysis. Annular dark field (ADF) imaging, the paragon of directly interpretable atomic resolution imaging in scanning transmission electron microscopy (STEM), is dominated by strongly scattering heavy elements, making weakly scattering light elements difficult to detect. However, it was demonstrated six years ago that both light and heavy elements are directly and robustly visible in at atomic resolution STEM images recorded from an annular detector in the outer area of the bright field region [1,2]. This tutorial will present an overview of this technique, dubbed annular bright field (ABF) imaging, describing its implementation, conceptual underpinnings, limitations, and successful applications.

Two distinct image-forming mechanisms contribute to the form of ABF images: elastic scattering that tends to forward-focus the electron probe when placed on columns of light elements (see left side of figure 1) and thermal scattering that tends to scatter the electron probe to high angles when placed on columns of heavy elements [2,3]. Both mechanisms reduce intensity in the outer area of the bright field region, and hence both light and heavy atomic columns appear as dark spots in ABF images, as in the example in the upper right of figure 1. Moreover, this proves to be true over a wide range of thicknesses and a moderate range of defocus values – lower right in figure 1 – meaning ABF images are not only directly interpretable but also robust, a highly desirable property for exploring unknown specimens.

ABF has been successfully applied to visualizing both lithium [4,5] and hydrogen [6,7] within crystalline environments. However, for these lightest of elements the imaging remains challenging: direct interpretation may apply only over a restricted range of thicknesses, the signal can be of similar magnitude to the noise level, and the specimens tend to damage under the beam. Further limitations of the technique, such as contrast inversion with defocus in very thin specimens [8,9] and incorrect apparent inter-column spacing in materials with very small inter-column spacing [3], will be discussed. Intriguingly, ABF appears tolerant to modest distortions of atomic columns, enabling light element columns to be imaged in defect structures like grain boundaries [10].

Drawing on experience from both experimental and theoretical explorations, practical aspects such as implementation and optimum experimental geometry will be discussed, as will related techniques. For instance, using a detector complementary within the bright field region to an ABF detector can image light elements with improved spatial resolution [11] and taking the difference between this and the ABF signal can enhance the signal-to-noise [12]. A brief selection of applications to which ABF has been applied will be presented [13].

References:

[1] E. Okunishi *et al*, Microscopy and Microanalysis **15(S2)** (2009), p. 164.
[2] S.D. Findlay *et al*, Appl. Phys. Lett. **95** (2009), p. 191913.
[3] S.D. Findlay *et al*, Ultramicroscopy **110** (2010), p. 903.
[4] Y. Oshima *et al*, J. Electron Microsc. **59** (2010), p. 457.
[5] R. Huang *et al*, Angew. Chem. Int. Ed. **50** (2011) p. 3053.
[6] S.D. Findlay *et al*, Appl. Phys. Expr. **3** (2010) p. 116603.
[7] R. Ishikawa *et al*, Nature Materials **10** (2011) p. 278.
[8] S. Lee *et al*, Ultramicroscopy **125** (2013) p. 43.
[9] P.J. Phillips and R.F. Klie, Applied Physics Letters **103** (2013) p. 033119.
[10] S.D. Findlay et al, Ultramicroscopy **111** (2011) p. 285.
[11] M. Ohtsuka *et al*, Ultramicroscopy **120** (2012) p. 48.
[12] S.D. Findlay *et al*, Ultramicroscopy **136** (2014) p. 31.
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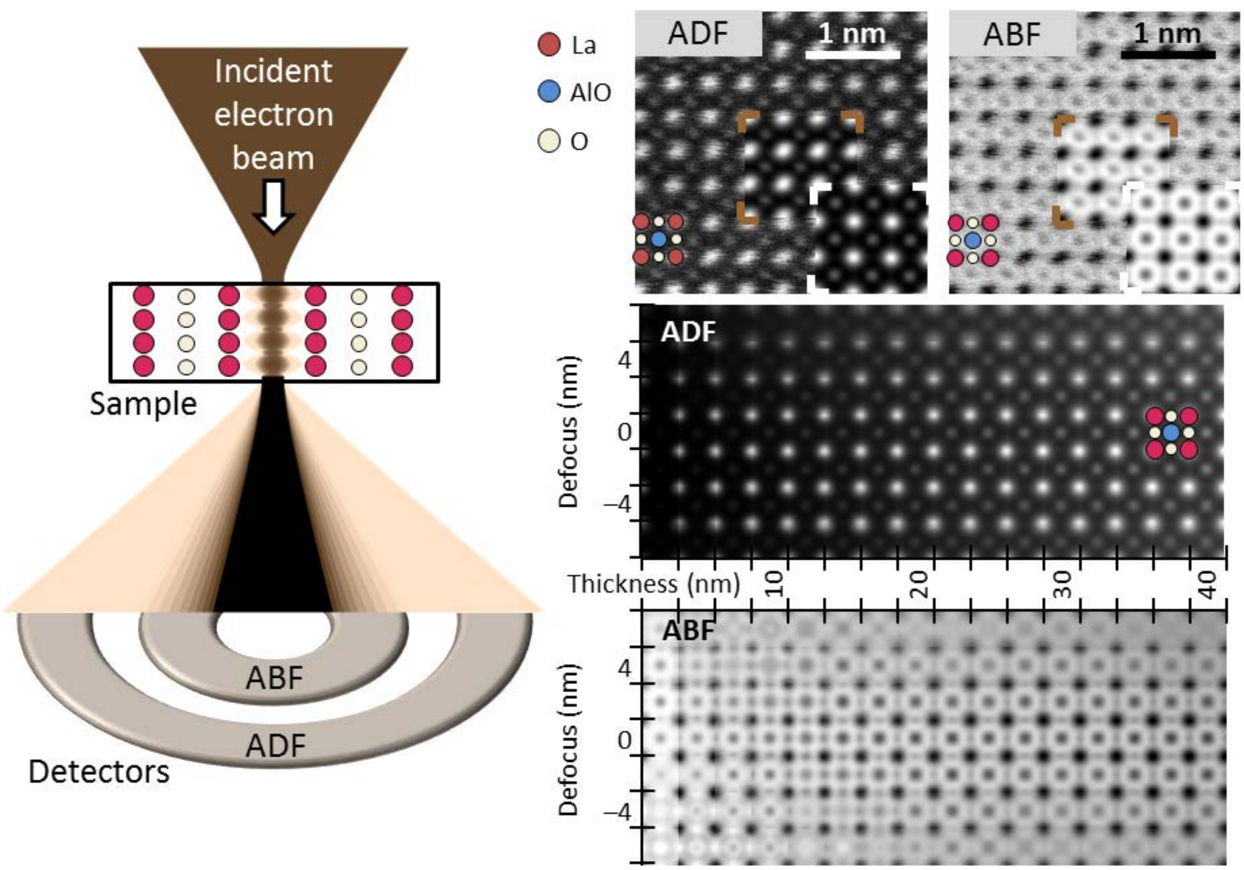

Figure 1. *Left*: schematic of a STEM probe scattering through a crystal and the ADF and ABF detector geometry. *Right upper*: Experimental ADF and ABF images of $LaAlO_3$ [001] for 200 keV electrons and a 23 mrad probe-forming aperture semiangle (courtesy of A/Prof. N. Shibata). The dark-cornered overlay shows the repeat-unit-averaged experimental image. The white-cornered overlay shows the corresponding simulation. *Right lower*: Simulated tableaux showing that ABF images are directly interpretable over a range thickness and defocus values almost as wide as that of ADF.