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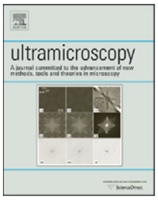

Seeing the atoms more clearly: STEM imaging from the Crewe era to today

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ABSTRACT

This review covers the development of scanning transmission electron microscopy from the innovations of Albert Crewe to the two-dimensional spectrum imaging in the era of aberration correction. It traces the key events along the path, the first atomic resolution Z-contrast imaging of individual atoms, the realization of incoherent imaging in crystals and the role of dynamical diffraction, simultaneous, atomic resolution electron energy loss spectroscopy, and finally the tremendous impact of the successful correction of lens aberrations, not just in terms of resolution but also in single atom sensitivity.

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1. The Crewe era

Albert Crewe and his coworkers laid the foundations for the modern-day scanning transmission electron microscope (STEM), with their realization that brightness is everything. Their use of a cold field emission source and an annular detector produced the first atomic-resolution images of single atoms and the first simultaneous electron energy loss spectroscopy (EELS). This review traces these and subsequent developments that have led to the development of modern aberration corrected instruments, bringing sub-Angstrom resolution, the imaging of individual light atoms, and two-dimensional atomic-resolution spectroscopic imaging. This is a succinct version of a much longer historical account of the development of STEM that was published recently [1].

Although Crewe is thought of as the father of STEM, it was actually conceived not long after the development of the first TEM by Knoll and Ruska [2]. Baron Manfred von Ardenne [3,4] developed the STEM, placing the imaging lens before the specimen instead of after the specimen as in the Ruska TEM design. He realized that the transmitted electrons would not need to be focused to form a high resolution image, merely detected, and so the STEM optics would avoid any chromatic aberration due to energy losses suffered during transmission through the specimen. While this was a sound idea in principle, he did not use a field emission source, and although he achieved a resolution of 10 nm the images were very noisy. He quickly abandoned the STEM in favor of the Ruska-style TEM design [5,6].

Almost 30 years pass before the STEM is taken up again by Crewe, who realized the necessity of using a high brightness cold field emission gun [7] to achieve sufficient beam current in a small probe. The first machine produced a resolution of 30 Å [8]. Over the next few years a new design produced a resolution stated to be about 5 Å, [9,10] but was actually closer to 2.5 Å (see [11]). This resolution allowed the first imaging of individual atoms in an electron microscope, using molecules stained with uranium and thorium atoms. The images were formed from the ratio of the elastic signal collected by the annular detector to the inelastic signal collected by the spectrometer. The cross section ratio is approximately proportional to atomic number Z and so they called the ratio image a “Z-contrast” image [12]. Wall et al., [13] showed line traces of the annular detector signal across individual atoms demonstrating unequivocally a probe size of 2.5 Å, and also images of small crystallites, as shown in Fig. 1.

With this microscope phase contrast images were also obtained by using a small axial collector aperture [9]. The images were similar to those obtained with a small axial condenser aperture of the TEM, thus demonstrating the principle of reciprocity [14,15]. Significant effort was also put into the development of EELS, which revealed its great promise as a microanalytical technique [16], spurring the development of modern analytical electron microscopy.

2. Z-contrast for materials science

The spectacular results generated by the Crewe group led to the establishment of the first commercial manufacturer of a dedicated STEM, VG Microscopes [17], and the emergence of the STEM as a high resolution analytical microscope [18]. Materials, however, unlike the biological systems of primary interest to the Crewe group, are typically crystalline, with strong diffraction effects. Attempts to use the Crewe ratio method for Z-contrast imaging were unsatisfactory because diffraction contrast tended

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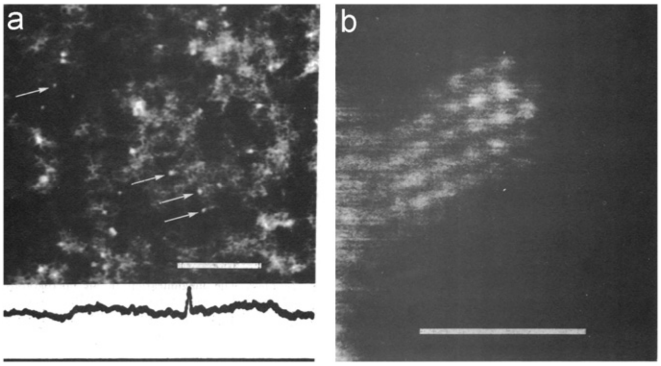

Fig. 1. (a) Annular dark field (ADF) image of a sample of mercuric acetate showing individual atoms obtained with the Crewe STEM. The line trace across a single atom shows a full width half maximum of 2.5 ± 0.2 Å. Scale bar is 50 Å. (b) ADF image of small crystallites containing uranium and thorium atoms. Scale bar is 20 Å. Reproduced from Ref. [13] with permission.

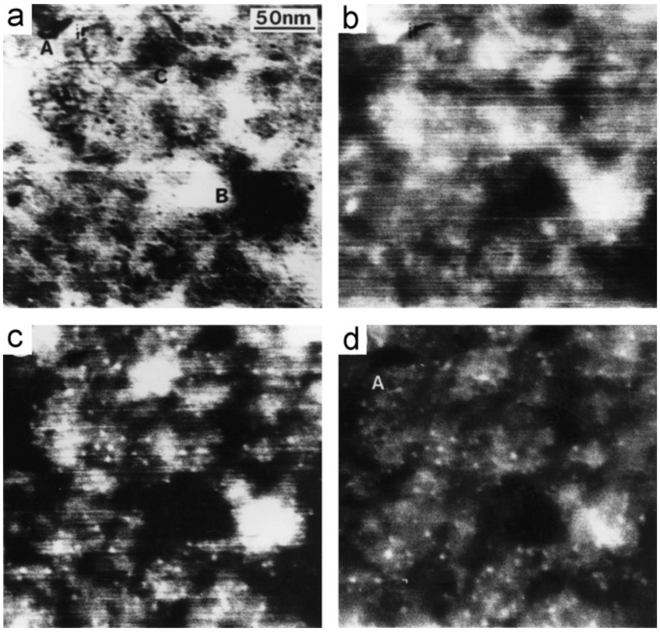

Fig. 2. Images of Pt particles on γ-alumina recorded in (a) bright field, (b) low angle ADF, (c) HAADF and (d) the ratio of high angle to low angle ADF signals. Particle contrast is highest in the HAADF image, reproduced from M.M.J. Treacy, PhD thesis, University of Cambridge, 1979, with permission.

to dominate any Z-contrast that might be present, making interpretation ambiguous [19]. More success was had imaging catalyst particles [20], leading to the idea of a high angle annular dark field (HAADF) detector where coherent diffracted beams would be replaced by thermal diffuse scattering, otherwise known as Rutherford scattering, with a scattering cross section approaching Z^2 [21]. Such a signal should therefore give enhanced Z-contrast compared to the Crewe ratio method, with minimal diffraction effects. The idea was tested by [22], and a comparison with the bright field, Crewe-style wide angle annular detector and a ratio image is shown in Fig. 2. The HAADF image shows the best contrast.

The technique was used to image dopant profiles in ion-implanted single crystal Si, where the suppression of diffraction contrast in the HAADF signal is quite apparent, see Fig. 3 [23]. Such results naturally led to the question of the ultimate resolution of the Z-contrast signal in a crystalline material. Clearly, if the crystal were a monolayer raft of atoms then atomic resolution would be expected with a sufficiently small probe, like in the Crewe images. And since Rutherford scattering is at high angles, it is generated close to the nucleus of each atom, and each atom can be considered an independent source. The situation is precisely analogous to the self-luminous source in light optics which produces an incoherent image [24]. An incoherent image shows no contrast reversals as focus is changed, like the image in a camera, and also shows better resolution than a coherent phase contrast image, as was appreciated by the Crewe group. However, the role of dynamical diffraction in thicker crystals was not known. Cowley [25] actually published an image demonstrating the improved resolution of the ADF image over the bright field phase contrast image, but surprisingly made no mention of any incoherent characteristics associated with the ADF image. Incoherent characteristics had been predicted theoretically for single atoms by Engel [26] and for weak phase objects by Misell [27], but the issue was controversial for high resolution [28] and for atoms in a column [29].

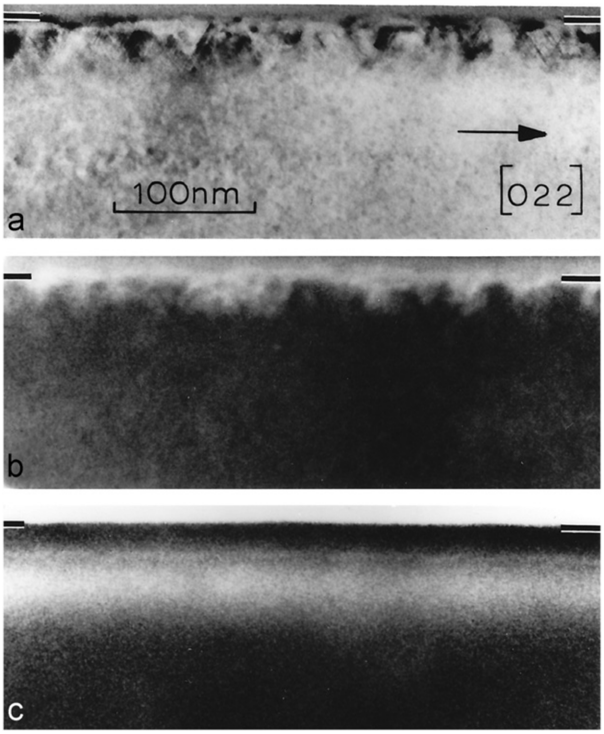

Fig. 3. Cross section images of Sb implanted Si. (a) TEM diffraction contrast image showing defects near the surface and end of range damage, (b) Low angle ADF image also dominated by diffraction contrast, (c) HAADF image revealing the Sb distribution. Reproduced from Ref. [23].

Incoherent characteristics were observed in HAADF images from crystals of the high temperature superconductors $YBa_2Cu_3O_{7-\delta}$ and $PrBa_2Cu_3O_{7-\delta}$, using a VG Microscopes HB501UX with a high resolution pole piece producing a theoretical probe size of 0.22 nm [30,31]. The contrast showed the expected dependence on atomic number and freedom from contrast reversals with thickness and objective lens focus. Fig. 4 shows images of the semiconductors Si and InP viewed along the $\langle 110 \rangle$ zone axis [32]. In this orientation the atoms form closely spaced pairs of columns referred to as dumbbells. Although the Si dumbbells spaced 0.136 nm apart are not resolved, they are clearly elongated, indicating the presence of the two columns. In the case of the InP the high Z of the In column dominates the image contrast and circular features are seen instead. Besides the lack of contrast reversals, a more surprising result was the apparent lack of any evidence of dynamical diffraction with increasing specimen thickness, the image intensity simply rising with increasing sample

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