3D Reconstruction of SPM Probes by Electron Tomography

X. Xu¹, Y. Peng¹², Z. Saghi¹, R. Gay¹², B. J. Inkson¹, and G. Möbus¹*

¹Department of Engineering Materials, ²Department of Electronic and Electrical Engineering, University of Sheffield, Sir Robert Hadfield Building, Mappin Street, Sheffield, S1 3JD, UK

E-mail: g.moebus@sheffield.ac.uk

Abstract. Three-dimensional morphological and compositional structures of tungsten tips consisting of layered amorphous oxide shell and crystalline W core are reconstructed by electron tomography using both coherent and incoherent imaging modes. The fidelity of the reconstruction is dependent on three criteria, suppression of unwanted crystal orientation contrast in the crystalline core, nonlinear intensity-thickness relations above a certain thickness limit, and artefacts due to missing angular ranges when acquiring a tilt series of images. Annular dark field (ADF), and EDX chemical mapping are discussed as alternatives to standard bright field (BF) TEM imaging.

1. Introduction
Tungsten tips are one of the most commonly used probes for scanning tunneling microscopy (STM). Recently, they have been used as manipulation probes to measure electrical and mechanical performance of nanomaterials in transmission electron microscopy (TEM) [1-3]. As W tips directly contact samples during measurement, their three-dimensional morphology and composition before and after testing are essential information and have to be well characterized. Electron tomography is a method to reconstruct three-dimensional morphology of materials from their two-dimensional projections [4]. It has been applied to reveal 3-dimensional structures of biological objects and nanomaterials [5-6].

The purpose of this paper is to explore the suitability of various TEM imaging modes for W tips and similar SPM probes, such as bright field (BF), annular dark field (ADF), energy-filtered TEM (EFTEM), and energy dispersive X-ray spectroscopy (EDX). In order for their three-dimensional structures to be reconstructed by electron tomography additional conditions are imposed via the applicability of coherent and incoherent imaging modes [7-8]. Any dependency of the image intensity on other factors than atomic number and thickness would be detrimental for tomography and concerns especially the Bragg diffraction induced scattering contrast in BF-TEM (lattice plane orientation dependence for crystalline areas). In addition, we explore application limits of ADF-STEM and EDX for thickness (projection) mapping as another prerequisite for tomographic reconstructions.

2. Materials
Tungsten wire (99.95%, Goodfellow, UK) with a diameter of 100 µm was etched in 2M NaOH (98.38%, Fisher, UK) electrolyte at room temperature, using a graphite rod as the cathode. The tip etching unit (Nanomagnetics Instruments Ltd., UK) consists of an automatic switch-off control...
working under level function mode. Various current values can be set up as the cut-off point to produce the etched tips with different geometrical apex, see Fig 1. Typical cut-off current is around 0.3 mA in our experiments and the maximum etching current is 7mA. It takes about 15 minutes to etch a tungsten tip by this unit. The etched tips were rinsed thoroughly using deionised water. In order to verify later generated tomograms, we picked up some nanoobjects as reference markers by dipping a W tip into an aqueous solution of gold nanoparticles (Agar, UK), see Fig 2.

![Image](attachment:figure1.jpg)

(a)  (b)  (c)

**Figure 1.** Three W tip morphologies of various etching currents influencing cone-sharpness and ball-shaped tip-ends.

3. Method

For tomographic 3D reconstruction a series of projection images in transmission with a linear intensity-thickness relationship is required across a range of viewing angles as close as possible to $180^\circ$. The SPM-tip samples were directly mounted (without any support grid or carbon film) on a high tilt tomography holder (model 912, Gatan, USA) for ultra-narrow gap lenses and observed on a JEM 2010F TEM (Jeol, Japan) with URP polepiece at 200 kV. Two sets of tilt series of the same tip were recorded by using bright field imaging mode (BF-TEM) and by using STEM mode with annular dark field detector (ADF-STEM) across the range -60˚ to +60˚ at 5˚ angular increment. The tip axis was mounted as close as possible parallel to the rotation axis of the goniometer stage.

4. Results and Discussion

Figure 2(a) shows a BF TEM image of a typical W tip with some gold particles attracted to its surface. The ADF STEM image at 0˚ tilt angle is shown in Figure 2(b). Oxygen mapping by energy filtered TEM (EFTEM) is shown in Figure 2(c) and was obtained via the three-window method using a GIF electron energy loss spectrometer (Gatan, USA). A layer of tungsten oxide produced during the etching procedure was found covering the tip surface. Carbon contaminants were found growing on the tip surface after a period of observation in TEM.

The BF and ADF-STEM tilt series were aligned and reconstructed using IMOD software [9]. The tilt series were primarily aligned using the cross-correlation method. Manual fine adjustment was applied thereafter to fix the sharp apex of the inner W core. Projected views from the reconstructed 3D data volumes of the generated tomograms are shown in Figure 3(a) and (b) using routines of Interactive Data Language IDL (Research Systems, USA). For the online publication a color scheme applies as follows: the inner tungsten cone is highlighted in red and it is covered by an outer layer of oxide in green. The carbon contaminants are shown in blue. For the printed publication, additional elemental markers are added.
A core/shell object consisting of amorphous layers (oxide and carbon) on crystalline core (tungsten, also gold markers) is an ideal test object to evaluate coherent vs incoherent imaging modes in tomography (see also [7-8,10]). BF imaging mode, consisting of diffraction contrast produced by coherent elastic scattering from the crystalline core, does not match the projection requirement where a monotonical relationship is indispensable. On the contrary, ADF-STEM mode is incoherent in nature, where coherent scattering is negligible. Both methods are complementary. BF has the potential (depending on the microscope) of better resolution and crisper edges in the input images and, recovers the outer shape of the tip with little disturbance from the false projections of the crystalline core. Errors due to diffraction contrast in the crystalline core partially cancel over the tilt series. Little errors arise in the amorphous parts of the specimens. ADF on the other hand provides valid thickness information for all parts (W core, W-oxide shell, Au particles) and provides the best overall fidelity.

Unavoidable artefacts due to the missing wedge are illustrated in various viewing directions in Figure 4. In Figure 4(a), a cross-section of the reconstructed ADF-STEM tomogram through the upper gold particle is viewed perpendicular to the Y direction. It clearly shows that the external diameter in Z
direction is apparently larger than that in X direction. Assuming a perfect conic shape of W tip, the
dimension in Z direction, which is parallel to the electrons incident direction in TEM, is distorted by
the missing-wedge effect. The relatively round gold particle also shows an elongation in Z direction.
The isosurface, which has a specific constant value in the same reconstructed volume, are shown in 0˚,
90˚, 180˚, and 270˚ viewing directions in Figure 4(b).

![Image](a) ![Image](b)

**Figure 4.** Residual artefacts of the tomogram due to the missing wedge. (a) Cross section view in the
direction parallel to the rotation axis of the generated tomogram from ADF reconstruction. (b)
Isosurface view of the tip with oxide shell in four viewing directions

Finally in Figure 5, we illustrate the upper thickness limit for tomography in STEM, by means of
nonlinear (and non-monotonous) intensity-thickness relations (absorption, see also [11]). W-cones are
again an ideal test object with the same atomic number Z and nearly linear thickness increase along
their longitudinal axis according to its conic shape revealed by the above mentioned tomographic
reconstruction. Figure 4(a) shows a typical ADF-STEM image of another W tip at lower
magnification than Figure 2(a).

![Image](a) ![Image](b)

**Figure 5.** Image of another tungsten tip than Fig 2. (a) ADF image and line profiles at the positions
labelled as 1 and 2. Width of image ≈ 6.55 µm. (b) Tungsten mapping by EDX (width of image ≈
4.86 µm) with a horizontal line profile along the axis of the tip printed on top.

Two inserted line profiles represent two positions at different thickness labeled as 1 and 2,
respectively. In the profile 2, an anomalous dip in intensity at the center of W tip results from
electrons multiply scattered (“absorbed”), not reaching the detector. This limits linearity in thickness
to about 500 nm. However, EDX mapping shown in Figure 4(b) tolerates the largest thickness. The ADF limit also depends on ADF parameters and A/D converter settings but is intermediate between EDX and EFTEM.

5. Conclusion
In summary, the three-dimensional amorphous morphology of W tips is successfully reconstructed by both BF-TEM and ADF-STEM. However, the crystalline core is better reproduced by ADF-STEM due to the effect of coherent scattering (diffraction contrast) intrinsic to BF. W tips are ideal testing objects to reveal thickness limits of tomographic imaging modes with ADF-STEM reaching the absorption limit much earlier than for EDX mapping. While we emphasized on the use of nanoparticles as markers to support the reconstruction of the SPM tip 3D morphology, we see an important future potential in using the tip (Si tips preferentially due to low Z) itself as a support for any nanoobject attached to it, which then becomes itself the objective of the tomographic reconstruction at higher magnification.

References
[1] Kuzumaki T, Kitakata S, Enomoto K, Yasuhara T, Ohtake N and Mitsuda Y 2004 *Carbon* **42** 2329
[2] Larsson M W, Wallenberg L R, Persson A I and Samuelson L 2004 *Microsc. Microanal.* **10** 41
[3] Bobji M S, Pethica J B and Inkson B J 2005 *J. Mater. Res.* **20** 2726
[4] Frank J (Ed.) 1992 *Electron Tomography: Three-dimensional Imaging with the Transmission Electron Microscope*, (New York, London: Plenum Press)
[5] McEwen B F and Marko M 2001 *The Journal of Histochemistry & Cytochemistry* **49**(5) 553; Marco S, Boudier T, Messaoudi C and Rigaud J L 2004 *Biochemistry* (Moscow) **69**(11) 1219
[6] Kübel C, Voigt A, Schoenmakers R, Otten M, Su D, Lee T C, Carlsson A and Bradley J 2005 *Microsc. Microanal.* **11** 378
[7] Möbus G and Inkson B J 2001 *Appl. Phy. Lett.* **79** 1369; Möbus G and Inkson B J 2003 *Ultramicroscopy* **96** 433
[8] Midgley P A and Weyland M 2003 *Ultramicroscopy* **96** 413.
[9] Kremer J R, Mastronarde D N, McIntosh J R 1996 *J. Struct. Biol.* **116** 71
[10] Friedrich H, McCartney M R and Buseck P R 2005 *Ultramicroscopy* **106** 18
[11] Hillyard S and Silcox J 1993 *Ultramicroscopy* **52** 325

Acknowledgement We would like to thank Dr. D. Mastronarde, University of Colorado, USA, for advice on IMOD. This work was supported by a grant from EPSRC, RA/104868, UK.