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Electron Tomography of Nanoscale Materials – Opportunities and Challenges Ahead

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Over the past 10 years electron tomography in the physical sciences has grown from a niche pursuit to a mature technique regarded now as a cornerstone tool for microscopists investigating nanoscale materials. The primary use of electron tomography is to record series of images, typically using HAADF STEM or BF TEM, each of which gives an approximation to a projection of a 3D structure, to reveal detail about an object's internal morphology. In this paper I will give a personal perspective on how the electron tomography field may expand and diversity in the next few years, to outline opportunities that have arisen with new hardware and software, and to highlight the challenges that lie ahead.

In recent years there has been an increasing desire to quantify tomograms and extract meaningful and robust information including volume fractions, surface areas and particle distributions. This has led to a development of improved algorithms for segmentation and volumetric analysis and new reconstruction methods that include 'prior information', which may be regarded as any known information (beyond the images themselves) that constrains the reconstruction and improves its fidelity. A number of techniques incorporating prior information have emerged (or re-emerged) recently, two of which are discrete electron tomography [1] and compressed sensing electron tomography (CS-ET) [2]. In both cases prior knowledge of the object being reconstructed is used to improve the reconstruction, in essence by reducing the artefacts that arise from the limited data set. In discrete electron tomography, the object to be reconstructed is assumed to be 'discrete' in terms of the number of grey levels present in the reconstruction. Grey levels for reconstructions of nanoscale materials may correspond for example to different atomic numbers or mass-densities. In CS-ET the constraint placed on the reconstructed object is that it can be described as being 'sparse' in some domain. As an example, images of individual single-species nanoparticles when transformed into a gradient domain would be 'sparse' – the only non-zero pixels would be at the particle surface. This 'sparsity' can be used as a powerful constraint in the reconstruction process to improve the fidelity of the tomogram. For objects which are not discrete, for example a 3D map of slowly-varying composition, these may still be regarded as sparse but in a different domain – for example a wavelet domain [3].

The great advantage of incorporating prior information into reconstruction algorithms is that, in general, far fewer images are needed to obtain high fidelity tomograms. The acquisition time for a tomographic data set may be reduced and/or enables the use of more sophisticated imaging and spectroscopy techniques, which generally need a longer acquisition per pixel, but lead to remarkably rich data sets. One area that will grow rapidly is the 3D mapping of composition, especially via STEM-EDX spectrum-imaging, building on early work [4]. Although EELS- and EFTEM-based tomography are possible [4-6], the introduction of larger, more efficient, x-ray detectors (with ca. 1sr solid angle) offers the possibility of routine 3D mapping of nanoscale materials. EDX spectrum-imaging can yield 2D compositional maps in minutes and, by using prior information in the reconstruction process, high fidelity 3D tomograms may be possible with only 10 spectrum-images. From such rich 4D data sets, 3D compositional maps can be extracted [7] and more sophisticated PCA/ICA/NMF techniques used in a more routine fashion. A rather different 4D data set is available through time-resolved electron tomography. Pioneering work by Zewail [8] has shown how through time-resolved (femtosecond) pulsing, recording a series of images across a time interval, repeating at successive tilts, unique insights can be made into, for example, the 3D vibrational behaviour of nano-materials. The use of time-resolved microscopy, and tomography, should grow rapidly as commercial instruments become available.

Another area ripe for further work is that of electron tomography of physical properties. One example is the combination of holographic (or other phase-sensitive) methods and electron tomography, pioneered by Tonomura and co-workers [9], with more recent examples concentrating on mapping the built-in potential at p-n junctions [10,11]. Extending this further, there has been some key work recently in developing vector tomography in the electron microscope with both theoretical and experimental papers showing the 3D mapping of magnetic induction **B** of nanoparticles and nanoscale films [12,13]. Vector quantities (fields) can be mapped in a tomographic way but more than one tilt series is needed and often an extra constraint (prior information), such as the field being divergence-free, is needed to complete the reconstruction. Electric fields induced by the electron beam (or another source) can be probed with low loss EELS. In particular, for metal nanoparticles there is now huge interest in mapping surface plasmons, whose character reflects that of the induced electric field, in two and three dimensions. Recent work has shown that for highly symmetric structures (cubes) 3D maps can be obtained by scalar tomography [14] but for more complex nanoparticles, vector tomography will be needed.

Lastly, is it possible to map tensor quantities, such as strain, in 3D? First attempts have been published recently [15] analysing the displacements of atom images in high resolution tomographic slices. However, it should be possible also to extract 3D strain information from more conventional diffraction contrast images of, say, a dislocation. A series of images (with a range of diffraction conditions) at successive tilts has encoded within it a wealth of information regarding local strain - new reconstruction approaches are required though to extract that information in a robust and meaningful fashion [16].

References:

[1] KJ Batenburg *et al*, Ultramicroscopy **109** (2009) p. 730.
[2] Z Saghi *et al*, Nano Lett. **11** (2011) p. 4666.
[3] R. Leary *et al* (2013), these Proceedings.
[4] G Möbus *et al,* Ultramicroscopy **96 (**2003) p.433.
[5] M Gass *et al* Nano Lett. **6** (2006) p.376
[6] K Jarausch *et al* Ultramicroscopy **109** (2009) p.326
[7] K Lepinay *et al* Micron (2013) in press.
[8] OH Kwon and AH Zewail Science **328** (2010) p.1668
[9] G Lai *et al* J. Appl. Phys 75 (1994) p.4593.
[10] AC Twitchett-Harrison *et al,* Nano Lett. **7** (2007) p. 2020.
[11] D Wolf *et al* Ultramicroscopy **110** (2010) p.390
[12] SJ Lade *et al* Optics Comm. **253** (2005) p.392
[13] C Phatak *et al* Phys Rev. Lett. **104** (2010) p.253901
[11] O Nicoletti *et al* (2013), submitted.
[12] B Goris *et al,* Nat. Mater. **11** (2012) p. 930.

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