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FOURIER TRANSFORM "DARKFIELD" TECHNIQUES

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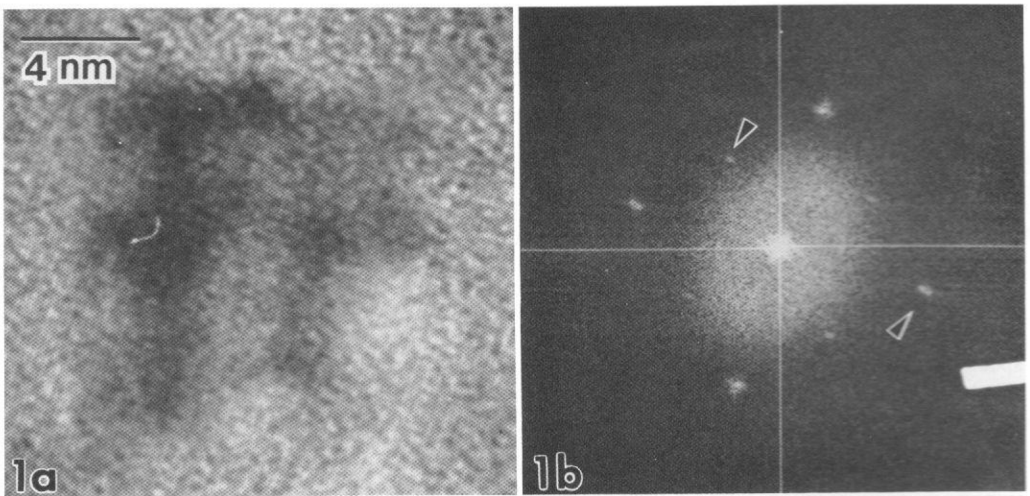
The standard approach to localizing single crystal regions in a transmission electron microscope (TEM) is to form tilted-illumination darkfield images with a diffracted beam down the microscope axis, and other electrons cut off by a centered objective aperture. This capability for localizing periodicities is powerful and shared by few other diffraction techniques. High resolution TEM's with point resolution and information limit below the spacing between atoms (e.g. 0.2 nm) are capable of putting information on all of the large crystal periodicities directly into phase contrast images. The refinement of techniques for finding and characterizing the crystals associated with spots, corresponding to the diffracted beams, in power spectra from such images has thus become a regular subject in the microscopy literature [e.g. 1-3]. Here, we illustrate that such techniques are useful for more than localizing crystallite boundaries. We also suggest one strategy for improving on the useful, but *ad hoc*, strategies now in hand.

In addition to highlighting crystallite boundaries, TEM darkfield imaging is useful for: (i) localizing regions (e.g. included phases or compositional zones) in which lattice parameter varies only slightly, (ii) finding crystals of like orientation in a polycrystalline field, and (iii) associating individual periodicities with single crystals in a polycrystalline field to allow measurement of interspot angles. Given suitable images from suitable materials, all of these things can be done by inspection. In the frequent cases when inspection fails, Fourier "darkfield" techniques can help with these tasks as well. Fig. 1a contains image of an inclusion in silicon (cf. [4]) whose lattice parameter differs from that of the matrix by only about 3%. The presence of two spacings is shown by the splitting of Si [110] spots in the power spectrum (Fig. 1b). Simple removal by filtering of all periodicities outside of the inner spots revealed an image with the matrix periodicities absent, showing that the larger periodicities in fact reside in the inclusion. Similar analysis of spacing variations across the breadth of a 20 nm ruthenium pyrochlore crystal indicated a trend to larger spacings at the particle exterior [5]. Fig. 2 shows an image of tungsten oxide which exhibits "powder rings" in its power spectrum. These images are excellent candidates for Fourier "darkfield" analysis. Such a "powder" analysis was used, for example, in reference [6] to measure the angle between 0.2 nm planes in a 2 nm presolar diamond crystal.

A goal of Fourier "darkfield" imaging is to determine the strength of a periodicity of given spatial frequency and phase, as a function of position in an image. This involves consideration of non-stationary behavior in two dimensions, for which Fourier power spectra are not optimum [7]. The eventual solution may require putting prior information here into forms suitable for Bayesian spectrum analysis.

[1] A. F. De Jong, W. Coene & D. Van Dyck, *Ultramicroscopy* **27** (1989) 53.
[2] G. Y. Fan, in: *Proc. 45th EMSA Meeting* (1987) 106-107.
[3] M. A. O'Keefe and M. L. Sattler, in: *Proc. 46th EMSA Meeting* (1988) 834-835.
[4] P. Fraundorf, H.-D. Chiou & G. K. Fraundorf, *J. Cryst. Growth* **76** (1986) 383.
[5] T. R. Felthouse, P. Fraundorf, R. M. Friedman and C. L. Schosser, *Abstracts 11th N. Amer. Meeting of the Catalysis Soc.* **B17** (1989) Detroit, Michigam
[6] P. Fraundorf, G. K. Fraundorf, T. Bernatowicz, R. Lewis and T. Ming, *Ultramicroscopy* (1989) in press.
[7] E. T. Jaynes, in: *Proc. Third Workshop on Maximum-Entropy and Bayesian Methods* (D. Reidel, 1983).



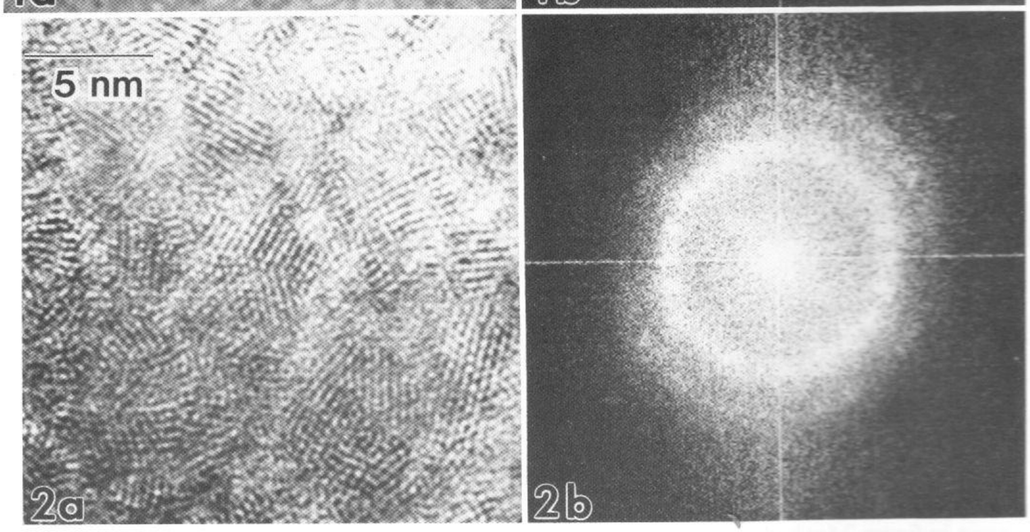


Fig. 1: (a) Digitized high resolution TEM image of precipitate in <100> silicon taken with an EM430ST at 300 keV, showing fringes in the silicon matrix as well as in the precipitate body. (b) Two-dimensional power spectrum of the image in 1a, showing strong 0.20 nm spots just inside of the Si 0.19 nm spots, and 0.28nm spacings diagonal to these. Removal of the outer periodicities from the image cleanly removed all of the matrix periodicities.

Fig. 2: (a) Digitized high resolution TEM image of a polycrystalline tungsten oxide specimen. (b) Power spectrum of image in 2a, showing multiple "powder diffraction" rings. Note: The fact that these rings are not a result of spherical aberration "zeros" in the contrast transfer function follows from our knowledge of the CTF, but also from the fact that cross-fringes corresponding to ring periodicities frequently show spatial correlations as though they come from single crystals. The angle between such correlated fringes can be checked against single crystal models of the structure for consistency.