Fringe-covariance "fingerprinting" of nanoparticle lattice images

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Individual diffraction patterns only provide 2nd-moment (e.g. atom-atom correlation) data on specimens, while HRTEM and z-contrast images provide information on higher order correlations as well. Variable coherence-width microscopy [1], and high-tilt lattice-parameter determination [2], involve ways to quantitatively characterize higher order (e.g. pair-pair) correlation by examining data from more than one image or diffraction pattern. Here we discuss a simple strategy for quantifying information of this sort found in a single lattice-fringe image of multiple nano-particles. The theory we discuss addresses the kinds of correlations expected if the nanoparticles are randomly oriented.

On the experimental end, the strategy is simple. Find all nanoparticles showing cross-fringes in the image, and draw one or more pairs of x's on a plot of interspot angle versus lattice spacing. The angles and spacings might be measured directly, or perhaps better from a small power spectrum superposed on each particle of interest. Each cross-fringe pair will result in marks at two spacings (one for each spot in the power spectrum) and at a common interspot angle value. Examples of such patterns, for 10 nm particles in a WC1-x plasma enhanced CVD film (Fig. 1) and TiO2 aerosol catalyst particles wet deposited on a holey carbon film (Fig. 2).

The data for the foregoing figures was obtained manually. Nonetheless, the figures offer a scatter diagram characteristic of both the primary zones expected for randomly-oriented particles of each crystal type, and the range of spacing errors resulting from both foreshortened projection (more significant for smaller particles) and measurement error. Automated analysis may become practical in the future, for example by tiling the image on a size scale characteristic of the grain size of interest, and then using power spectrum peak analysis to determine what to plot where.

What if we now wish to generate a theoretical fingerprint for a given crystal type? To this end, we consider maps of angular covariances, i.e. the average product of power spectrum intensity at two reciprocal spacings, separated in the three dimensional reciprocal-lattice by an interspot angle of $\Delta\alpha$. Suitable normalization, with means and standard deviations, allows one to thereby map a kind of angular correlation coefficient across the space of scattering probabilities. A plot of such correlations in the (hk0) plane of a simple cubic lattice is shown in Fig. 3. The maps for comparison with a given set of data will depend on both crystallite size (e.g. fringe foreshortening effects are more prevalent in smaller crystals) and the microscope's instrument response function (e.g. even single zone axis patterns will show more fringe pairs with better contrast transfer). Links to angular covariance maps for some commonly encountered structures will made available on the web [3].

References

[1] M. M. J. Treacy, J. M. Gibson, *Ultramicroscopy* **52** (1993) 31.
[2] W. Qin and P. Fraundorf, *Ultramicroscopy* **94** (2003) 245.
[3] http://www.umsl.edu/~fraundor/fringecov.html

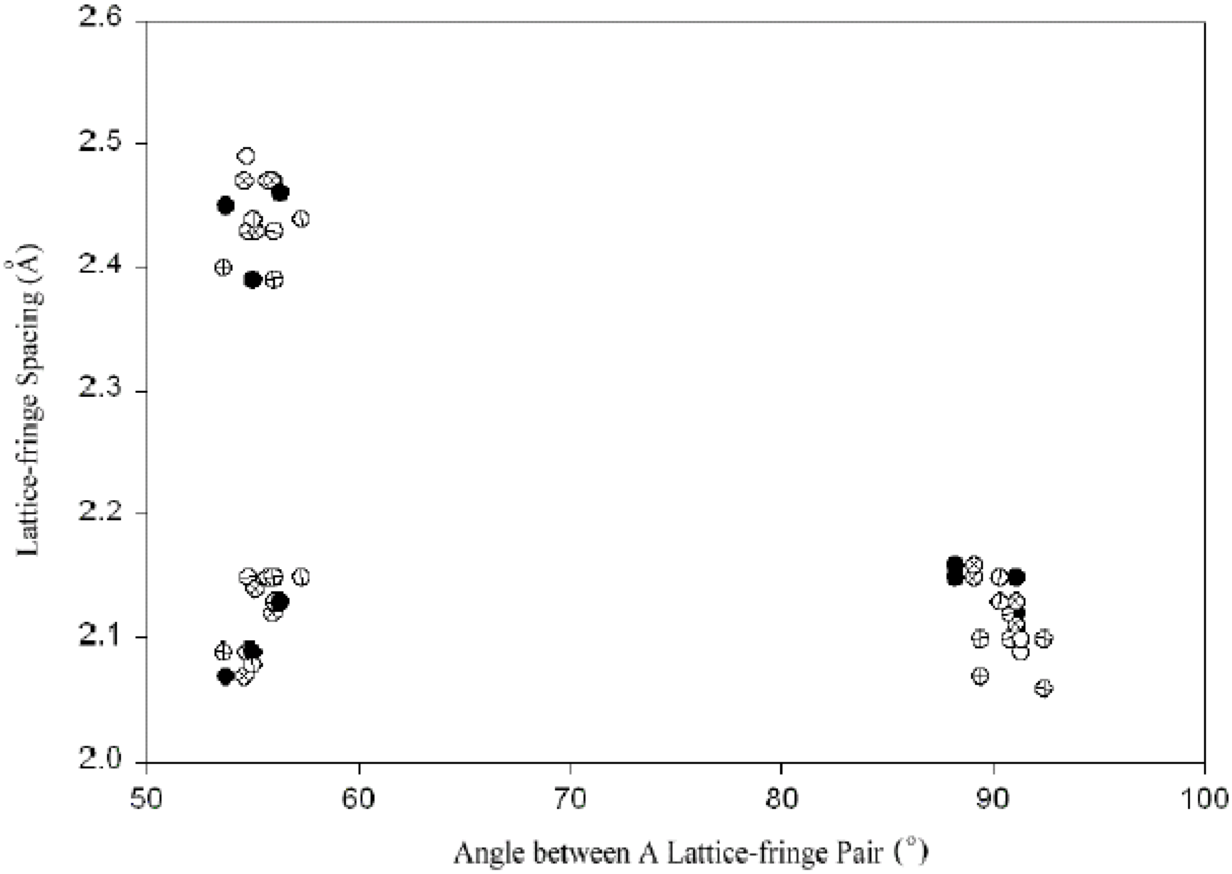
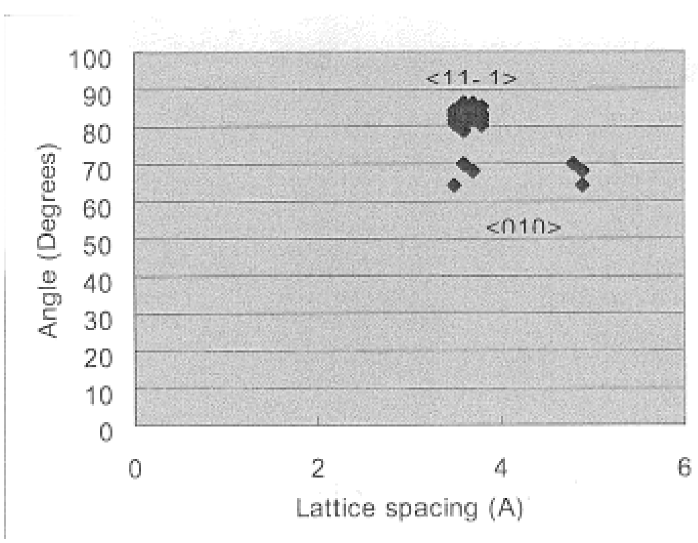


FIG. 1. Spacings and interplanar angles measured from cross-fringes of 23 10nm WC_{1-x} crystals [4].



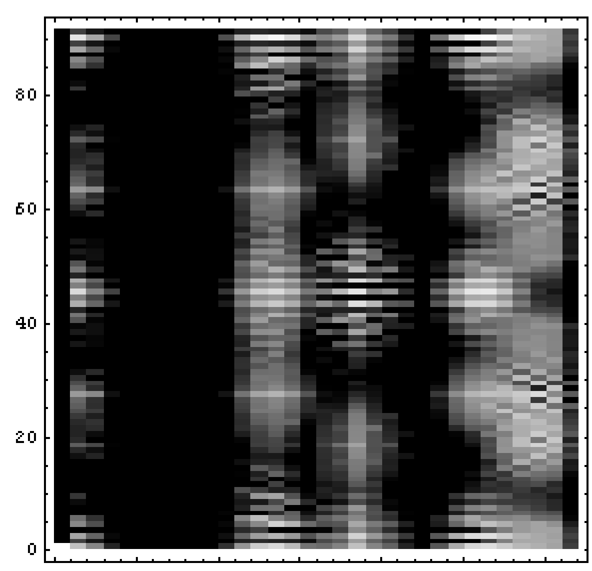


FIG. 2. (left) Angles and spacings from cross-fringe images of 10 nm anatase nanocatalyst particles.
FIG. 3. (right) Angular covariance map for spacing pairs in the (hk0) plane of a simple cubic lattice, with angular lag in degrees on the vertical axis, and g-vector on the horizontal axis.