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Rocking-Beam Variable Coherence Electron Microscopy: An Alternative Approach to Fluctuation Electron Microscopy

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Fluctuation electron microscopy (FEM) is a novel technique to characterize the "medium-range order" (MRO) in amorphous materials [1-2]. MRO is the order that extends beyond the second and third atomic shell (~1.5-2 nm) but cannot be identified with high-resolution transmission electron microscopy (HRTEM). MRO is believed to affect diffusive, mechanical, optical and electronic properties of amorphous materials. FEM provides a measure of MRO through analysis of the normalized variance, $V(\boldsymbol{k},Q)$, of the image intensity, $I(\boldsymbol{r})$, in a series of dark-field (DF) TEM images,

$$V(k,Q) = \frac{\langle I^2(r,k,Q)\rangle}{\langle I(r,k,Q)\rangle^2} - 1 \quad ,$$

where the statistical average is over the real-space coordinate $\boldsymbol{r}$ in a given image, $\boldsymbol{k}$ is the scattering vector, and Q is the diameter of the objective aperture at which the image was acquired [1-2]. Equivalently, the data may be acquired in scanning transmission electron microscopy (STEM) mode with a series of diffraction patterns, $I(\boldsymbol{k})$, with Q representing the diameter of the probe-defining aperture [3]. In the typical case where the variance is measured as a function of $\boldsymbol{k}$ at constant Q, FEM is alternatively named variable coherence electron microscopy (VCEM). Traditionally, FEM is performed in TEM mode with annular hollow-cone dark field (HCDF) illumination to form an incoherent average image over all azimuths at a given $k = |\boldsymbol{k}|$ and a one-dimensional $V(k)$ profile.

We have been exploring an alternative data collection strategy for FEM, where a series of TEM DF images are acquired at distinct scattering vectors along a two-dimensional grid $\boldsymbol{k} = (k_x, k_y)$, as shown schematically in Fig. 1. This rocking beam (RB) VCEM can be usefully regarded as a four-dimensional extension of hyperspectral imaging, with two real-space and two reciprocal-space coordinates. We have found that the DF images formed in RB-VCEM mode have greater contrast than those acquired with HCDF illumination, which may help to explain the systematically lower values of $V(\boldsymbol{k},Q)$ observed in TEM relative to those acquired in the equivalent STEM mode [3]. RB-VCEM also provides advantages for study of MRO during the early stages of the amorphous-to-crystalline transition, since nanocrystalline precipitates can be identified by their characteristic spot diffraction patterns and removed prior to calculation of $V(\boldsymbol{k},Q)$ for the adjacent amorphous matrix.

As a proof-of-principle, we have performed RB-VCEM on an amorphous tungsten (a-W) film sputtered onto a holey carbon grid. Series of DF images were acquired with a Philips CM200FEG STEM/TEM operated at 200 kV in "rocking beam" mode, ~10 nA beam current, "parallel" incident illumination, 135kX mag, and an objective aperture diameter $Q = \sim 2.4\ \text{nm}^{-1}$. The beam orientation was varied manually on a 10x10 grid with a step size of 1 nm^{-1} with an EMiSPEC Vision integrated acquisition system, in synchronicity with a Gatan slow-scan CCD camera, 2x binning, 10 s exposure operated in Acquire Series mode in DigitalMicrograph on a 15 s interval. The rocking-beam channeling pattern, formed with the STEM bright field detector centered on the optic axis, is shown in Fig. 1B. A false-color map of the corresponding $V(k_x,k_y)$ data is shown in Fig. 2. The peak in the $V(\boldsymbol{k})$ map at $k \sim 4.2\ \text{nm}^{-1}$ and a value of 0.07 – 0.095 coincides with the corresponding peak in the diffraction pattern. The influence of beam damage and energy filtering has also been studied [4].

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[2] J.M. Gibson et al., Ultramicrosc. 83 (2000) 169.
[3] P.M. Voyles and D.A. Muller, Ultramicrosc. 93 (2002) 147.
[4] Research at the SHaRE User Facility was sponsored by the Office of Basic Energy Sciences, U.S. Department of Energy, under contract DE-AC05-00OR22725 with UT-Battelle LLC.

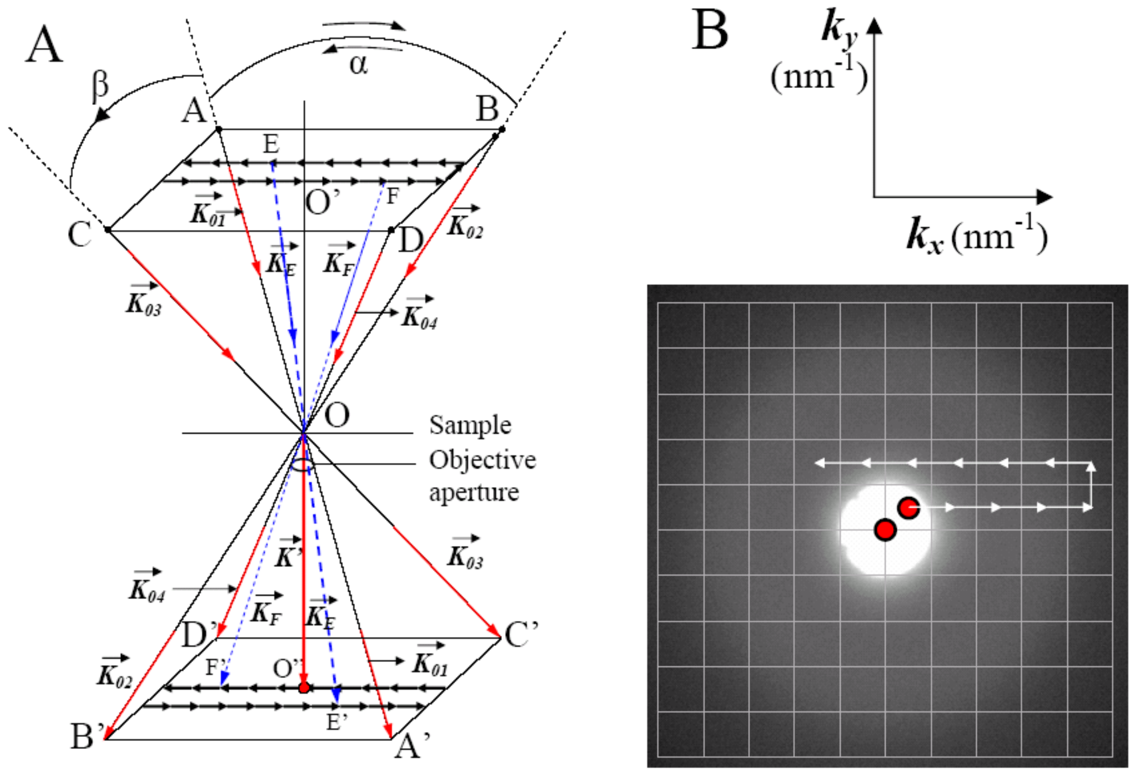

Fig. 1. A. Schematic representation of rocking-beam variable coherence electron microscopy (RB-VCEM). Incident beam angles α and β are varied in two dimensions to form a space defined by the "four-fold prism cone" ABDCO. ***K'*** is the diffracted beam wave vector which is collected by a centered objective aperture to record TEM DF image. The blue arrows show two examples in TEM rocking-beam mode for the incident wave vectors $\boldsymbol{K_E}$, $\boldsymbol{K_F}$ along EOE', FOF' directions, respectively. B. Rocking-beam channeling pattern and grid over which RB-VCEM images were acquired.

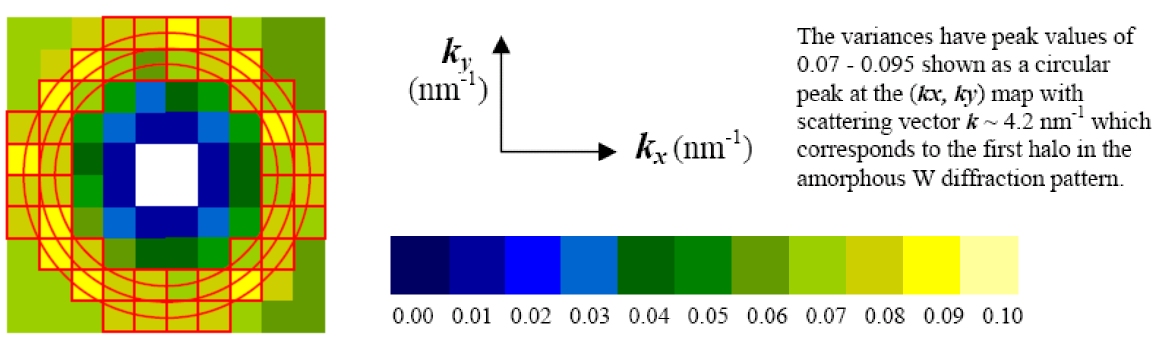

Fig. 2. Variance map $V(\boldsymbol{k})$ of a-W as the function of two dimensional scattering vector $\boldsymbol{k} = (k_x, k_y)$.

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