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Meeting-report

Rapid-Acquisition FEM – Grappling the Noise

A. Zjajo[1], H. Du[2], R. E. Dunin-Borkowski[2], A. Rezikyan[3], J. M. Gibson[4], and M. M. J. Treacy[1]

[1]*Dept. of Physics, Arizona State University, Tempe, AZ, United States*
[2]*Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons, Juelich, Germany*
[3]*Characterization Science Department, Corning Inc, Corning, NY, United States*
[4]*Florida A&M University-Florida State University, College of Engineering, Tallahassee, FL, United States*

Fluctuation Electron Microscopy (FEM) has proven to be a versatile technique for detecting subtle traces of ordering in amorphous and glassy materials [1–4] However, quantitative results remained elusive, mainly because experimental variance data disagree with theory by several orders of magnitude. The precise reasons for this discrepancy are still a mystery. Here, we present a preliminary report on what we know.

FEM detects Medium Range Order (MRO) using the normalized intensity variance given by

$$V(k_x,\, k_y) = \frac{\langle I^2(k_x,\, k_y)\rangle}{\langle I(k_x,\, k_y)\rangle^2} - 1 - \frac{1}{\langle I(k_x,\, k_y)\rangle}. \tag{1}$$

$I(k_x, k_y)$ is the diffraction pattern intensity, and angular brackets designate the average over many different patterns, taken from different sample volumes. The last term is the correction for Poisson noise. Under perfect kinematical scattering conditions, the normalized variance, V, should be near a value of 1 for a randomized sample and rise above 1 if medium-range order (MRO) is present. However, experimental normalized variances are often two orders of magnitude or more, lower than expected. For comparison, the Stobbs effect for image intensity is a factor of two or three lower than theory. It has been a mystery why the normalized variance of experimental data is so low.

Initially, we assumed that the suppressed variance arose from spatial *incoherence* in the illumination, but it is clear that the suppression arises from displacement *decoherence* arising from changes in the scattering itself during the data acquisition time. Rezikyan et al explored the role of beam damage and found that variance suppression was equivalent to average atomic motions of ~1.5 Å during acquisition [5]. These motions are clearly too big for individual atoms, but could arise from collective rotations of regions of the sample after beam damage events. Alternatively, small beam tilts arising from transient charging effects may be contributing. This suggests that short exposure experiments should mitigate some of the decoherence from low-frequency disturbances.

Our rapid-acquisition FEM explores the impact of decreasing exposure time, reducing the total fluence on each probed volume, while increasing the number of probed regions to maintain signal-to-noise. Modeling shows that this idea works under ideal conditions [6].

Experiments were carried out on a ChemiSTEM at 80 kV, with a 1 nm probe collecting diffraction patterns out to 7.2 nm^{-1} on a 128x128-pixel EMPAD detector, which allows acquisition times as short as 1 msec. We examined amorphous silicon (obtained from SPI, Inc) at three thicknesses, $t = 5$, 9, and 15 nm. Five sampling conditions were used; 256 patterns at 256 msec, 1024 patterns at 64 msec, 4096 patterns at 16 msec, 16384 patterns at 4 msec, and 65536 patterns at 1 msec. The total signal was the same in each experiment. Fig. 1 shows that noise variance dominates at high k. The average noise should be the same for pure Poisson noise and a perfect detector (detective quantum efficiency, DQE = 1). We observe a problematic increase in non-Poisson noise at the shortest exposure times, resulting in negative V over most of the range of k (Fig. 2). Thus, the Poisson noise correction in Eq. (1) seems to be insufficient for the shortest exposure times, below 256 msec, with the EMPAD detector.

Several modifications to (1) were tested. Detector readout noise, while low, is not Poisson distributed and may be a significant contribution when large numbers of short-exposure patterns are acquired. To explore the non-Poisson noise characteristics, we compared the ratios of the variance traces (Fig. 3), 4 msec/1 msec; 16 msec/4 msec etc. For the shortest exposures at low k, the ratio is 4 as expected, but decreases to 2 at high k for the 4 msec/1 msec ratio. This suggests that about half of the noise contribution is unaccounted for at the lowest intensities. Poisson noise dominates when the signal is strong k giving a ratio near 4.

Monitoring the diffraction intensity using an UltraSTEM at 60kV, with an 18Å probe diameter, k￼ value over time (Fig. 4) reveals strong fluctuations in detected intensity at the 1 msec time scale. Clearly faster acquisition times are essential.

We applied an additional normalized-readout-noise correction term $-cN/I(k_x,\, k_y)^2$, where N is the number of patterns, and c is a fitted constant, but this term does not correct the negative variance.

We conclude that non-Poisson detector noise contributions are important at low signal strengths. We are currently looking into the origin of the additional noise [7].

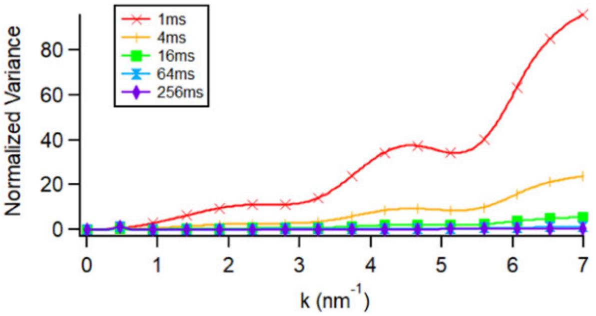

Fig. 1. The uncorrected Normalized Variance

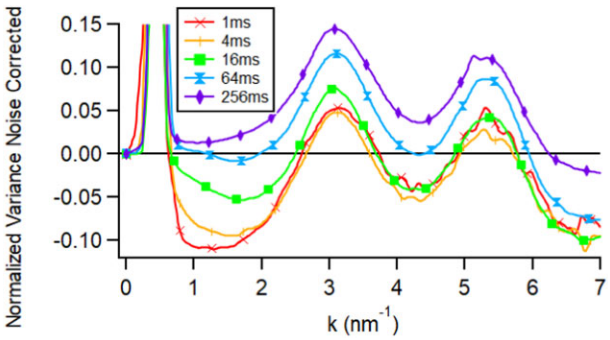

Fig. 2. Noise-corrected Normalized Variance

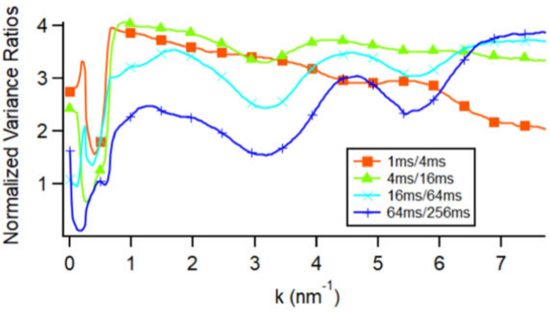

Fig. 3. The ratios of the normalized variances.

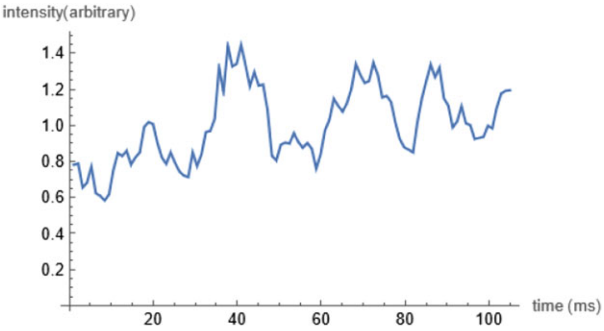

Fig. 4. Diffracted intensity at a single *k* value sampled every millisecond. Strong intensity fluctuations confirm the scattering is changing

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