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 85287, USA
[2] Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons, 52425 Juelich, Germany
[3] Characterization Science Department, Corning Inc, Corning, NY, USA
[4] Florida A&M University-Florida State University, College of Engineering, Tallahassee, FL 32310

Fluctuation Electron Microscopy (FEM) is 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 reasons for this discrepancy are still a mystery. We present a preliminary report on what we know.

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

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

$I(k_x, k_y)$ is the diffraction pattern intensity into scattering vector (k_x, k_y). Angular brackets designate the diffraction pattern average from different sample volumes. The last term corrects for Poisson noise. For a random sample under perfect kinematical scattering conditions V, should equal a baseline value of 1, rising above 1 if medium-range order (MRO) is present. However, experimental normalized variances are often two or more orders of magnitude lower than expected. By comparison, the Stobbs effect for image *intensity* is only a factor of two or three lower than theory.

Initially, we thought that the variance suppression arose from spatial *incoherence* in the illumination. It is clear now that displacement *decoherence*, arising from changes in the scattering during the data acquisition time, is likely responsible. Rezikyan et al [5] explored the role of beam damage and found that variance suppression was equivalent to average relative 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 shorter-exposure experiments might mitigate some of the decoherence from low-frequency disturbances.

Rapid-acquisition FEM explore 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 V 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. We applied an additional normalized-readout-noise correction term $-cN/\langle I(k_x, k_y)\rangle^2$, where N is the number of patterns, and c is a fitted constant. This exacerbates the negative variance. 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 giving a ratio near 4. Monitoring the diffraction intensity over time using a Nion UltraSTEM at 60kV [7], with a 1.8-nm probe diameter (Fig. 4) reveals strong fluctuations in intensity at the 1 msec time scale. Clearly, faster acquisition times are essential, pushing the limits of current CCD technology.

We conclude that non-Poisson detector noise contributions are significant at low signal strengths.

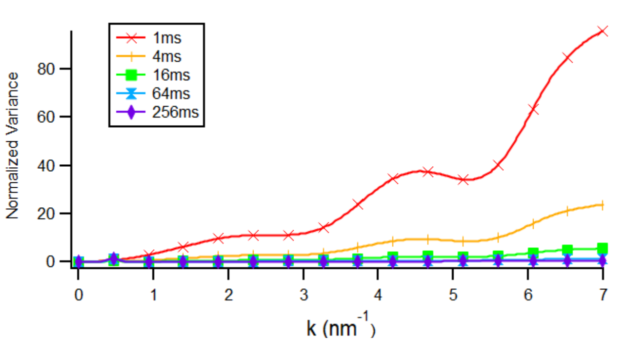

Figure 1: The uncorrected Normalized Variance

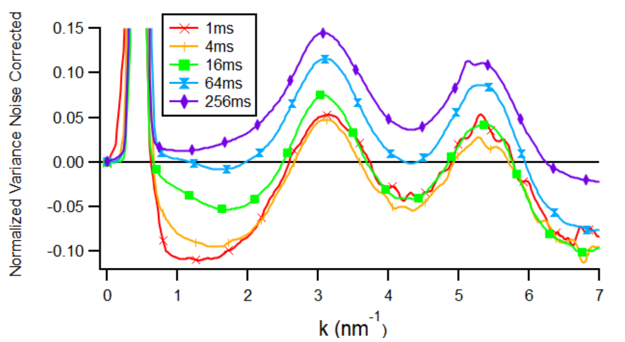

Figure 2: Noise-corrected Normalized Variance

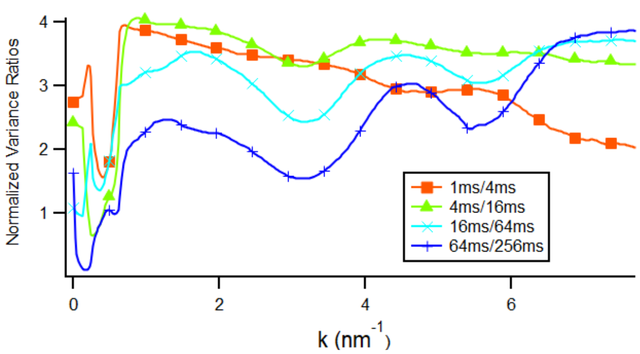

Figure 3: The ratios of the normalized variances.

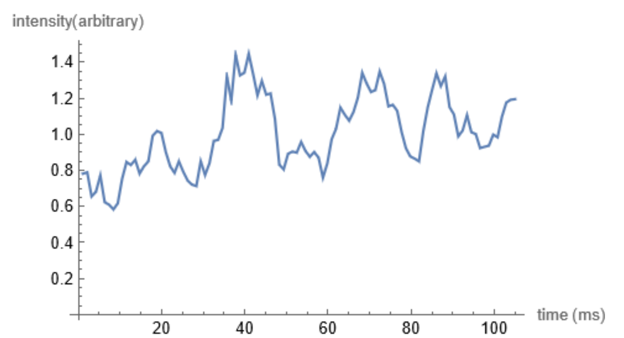

Figure 4: Diffracted intensity fluctuations at k sampled every millisecond.

References

[1] M.M.J. Treacy et al., *Rep. Prog. Phys.* **68** (2005) p. 2899–2944.
[2] J.M. Gibson and M.M.J. Treacy, *Phys. Rev. Lett.* **78** (1997) p. 1074–1077.
[3] P.M. Voyles and J.R. Abelson, *Solar Energy Materials and Solar Cells* **78** (2003) p. 85–113.
[4] M.M.J. Treacy et al., *J. Non-Cryst. Solids* **231** (1998) p. 99–110.
[5] A. Rezikyan et al. *Microscopy and Microanalysis*, **21** (2015) p. 1455–1474
[6] A. Zjajo et al. *Microscopy and Microanalysis* **27** (2021) p. 1776-1777.

[7] The authors thank Toma Susi and Clemens Mangler of the University of Vienna.