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Incoherence, Decoherence and Speckle Statistics in Fluctuation Microscopy

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Fluctuation Electron Microscopy (FEM) examines the scattering statistics from small volumes of thin amorphous materials to detect the presence of medium-range order (MRO) [1]. By now it has been thoroughly demonstrated, by both modeling and simulations, that FEM is extraordinarily sensitive to MRO, much more than high-resolution diffraction and high-resolution imaging. Experiments have confirmed this sensitivity. In particular, FEM has shown that most forms of amorphous silicon are not ideal continuous random networks [2, 3], as had been believed. Instead, they contain high densities of paracrystallites, which are small nanometer-sized regions of material that have a strained cubic silicon topology[4]. There are two key contributing factors to this sensitivity. First, FEM examines the *variance* of the scattering statistics, which is the second moment of the intensity distribution. Essentially, FEM examines the speckliness of the diffraction data. This means that it examines four-body correlations, whereas diffraction alone examines the first moment of intensity, and examines pair-correlations. The four-body terms are much more sensitive to medium-range correlations. The second key factor, which in this era of aberration-corrected microscopy may come as a surprise, is that it is a *low-resolution technique*. The sensitivity of FEM to MRO is maximized when the probed resolution is comparable to the length scale of the MRO.

Although FEM is successful as a qualitative technique – it can disclose unambiguously and sensitively the signature of MRO in a sample – it is not yet truly quantitative. There are two main reasons for this state of affairs. The most difficult barrier is that we do not know how to invert, analytically, four-body diffraction data. There has been significant progress in bypassing this issue by use of experimentally constrained reverse Monte Carlo methods. In this approach, diffraction and FEM data, as well as a potential energy cost function, are used as constraints while constituent atoms in a model are moved around randomly [5-7]. This method shows great promise, but it has revealed a huge discrepancy between simulated variance and experimental variance; the experimental variance is usually a factor of 10–100 less than the calculated values.

This discrepancy between experimental variance and computed variance was masked in early FEM studies by the fact that illumination incoherence was used as an adjustable experimental parameter. The most common form of such *variable coherence microscopy* was hollow-cone dark-field imaging in a TEM. It was generally assumed that the incoherence could be modeled as the sum of the scattering from a set of incoherent sources. A model of such incoherence was developed [8], where it was assumed that if there were m such sources, then the speckle intensity probability distribution in a tilted dark-field image would be

$$P(I) = \frac{m^m}{(m-1)!}\frac{I^{m-1}}{\bar{I}^m}\exp\left(-\frac{mI}{\bar{I}}\right),$$

where $\bar{I}$ is the mean intensity. This formula is obeyed remarkably well for data obtained from tilted dark-field TEM images of amorphous carbon (Figure 1). Fits to that data give $\bar{I} \approx 100$ and $m \approx 30$, yet we know experimentally that the illumination is highly coherent, suggesting $m \approx 1$. This result strongly suggests that we have been misinterpreting the meaning of the m-value in this formula. The formula imposes two constraints on the intensity distribution; the mean intensity $\bar{I}$ must be held fixed; and the second constraint that governs m is *not* coming from the illumination incoherence.

This surprise result points us towards an alternative source for m – decoherence of the scattering within the sample. In essence, this is a type of diffuse scattering, but it is induced by beam damage in the sample, which subtly alters the underlying structure being probed. In turn, this causes the speckle intensity to fade when speckle is time-averaged over the several seconds needed to record the data. If this new interpretation is correct, it would suggest that rapid data acquisition will help remedy the problem, and will restore the ideal data back to an m=1 negative exponential distribution. Reducing the sample temperature may also be beneficial. This may assist efforts at quantitative modeling. Ironically, because FEM is so sensitive to such fluctuations, it is therefore more strongly affected by decoherence than are simple diffraction and imaging. The authors are presently exploring ways to model such decoherence in FEM as a diffuse scattering term.

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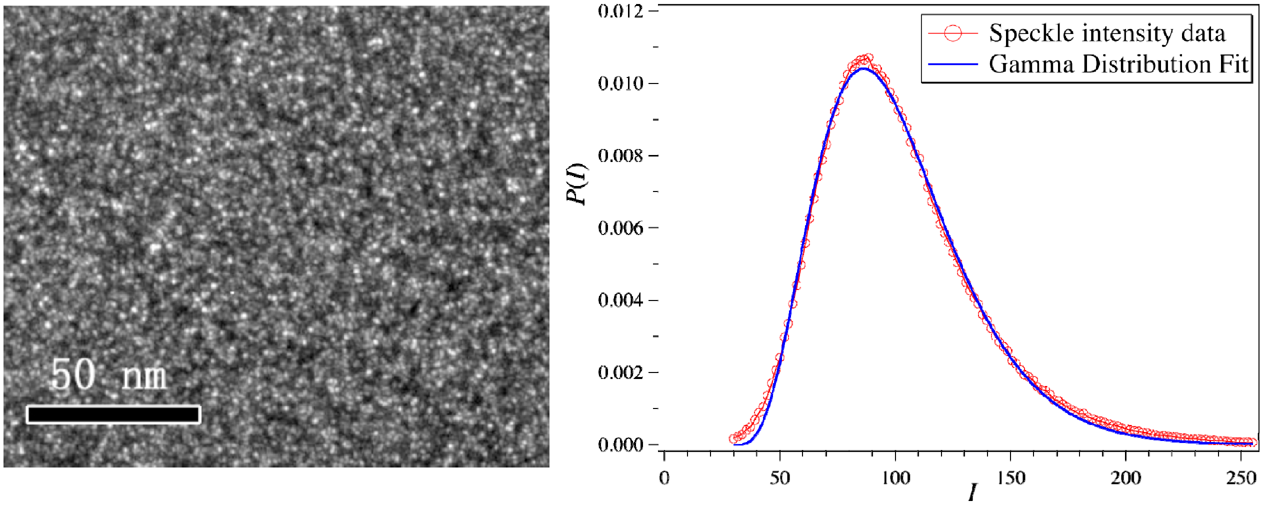

FIG 1. Left: Typical tilted dark-field image of a 20-nm thick amorphous carbon illustrating the speckle distribution observed when the microscope resolution is 1.0 nm. (Taken by a JEOL 2010F at 200 kV, and a beam tilt of ~4 nm^{-1}.) Right: The intensity histogram of this image, and the fit to a gamma distribution with $m = 30$ and $\bar{I} = 100$.