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Fluctuation Cepstral STEM for Imaging Disordered Materials

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Materials research is often confounded by nanoscopic heterogeneity and disorder. For example, in battery technology the electrochemical environment induces non-uniform degradation of materials components [1]. A key impediment to understanding such disorder is the availability of characterization methods with sufficient spatial resolution and large enough field-of-view to visualize and classify distinct phase information contained within the material. In this regard, characterization methods based on electron diffraction have been developed over the years to decode crystallographic information embedded in complex materials systems [2].

Central to this theme is scanning electron nanodiffraction (SEND) or four-dimensional scanning transmission electron microscopy (4D-STEM), which generates position-resolved diffraction datasets that can be further analyzed to extract nanoscale properties of materials. SEND data has proven essential for several applications, such as strain analysis, orientation mapping, grain clustering, and phase identification [3-5]. However, such applications have so far been limited to Bragg diffraction of ideal crystalline materials. On the other hand, fluctuation electron microscopy (FEM), which was developed prior to the adoption of 4D-STEM, has been applied successfully in the study of disordered materials [6]. The principle of FEM is to extract an order parameter to understand the degree of medium-range order within amorphous materials by studying the intensity fluctuations within diffraction patterns. Recently, cepstrum analysis ($C_p = \mathcal{F}\{\log[I(\boldsymbol{k})]\}$) has been introduced for the study of both ordering and disordering in materials systems. This analytical technique has been applied to study nano- to atomic-scale lattice strain and crystalline defects [7, 8].

The principle of fluctuation cepstral STEM (FC-STEM) is to measure the fluctuations associated with interatomic distances and orientations. This is possible, because the quefrency peaks of the radial profile given by the exit wave power cepstrum can be directly related to the projected atomic potential in the thin sample limit. The FC-STEM analysis is performed as follows: (1) the recorded diffraction patterns are converted to cepstral patterns (Figure 1a), (2) a background-corrected radial profile is computed (Figure 1b), and (3) fluctuation analysis is performed on regions surrounding the selected quefrency peaks (Figure 1c and 1d). Each pixel in the resulting FC-STEM image reconstruction represents the intensity variation for the selected quefrency range. Using a cycled silicon anode harvested from a lithium-ion battery as a sample, we acquired SEND datasets within different regions of interest and performed the aforementioned FC-STEM analysis to deconvolute complex phase information embedded within the material. In this case, the first peak FC-STEM image provides bright contrast in the amorphous regions (Figure 1c), while the second and third peaks give the bright contrast for the crystalline regions (Figure 1d). A comparison of FC-STEM with other electron imaging and diffraction visualization methods (including dark-field, bright-field, EDS elemental composition, and FEM) is made in Figure 2. These examples demonstrate that FC-STEM analysis provides clear contrast separation for distinct phases comprising the complex, disordered material. The insights gained from the application of FC-STEM are especially useful for identifying heterogeneous nanoscale characteristics, which has broad impact on battery materials that are otherwise not accessible through conventional characterization methods [9].

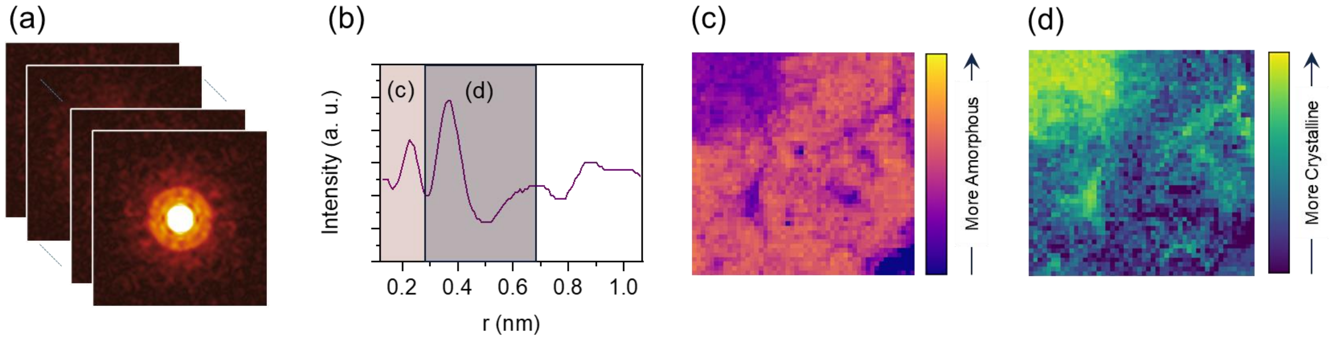

Figure 1. Procedure for FC-STEM analysis. (a) Starting cepstrum stack computed from SEND dataset with (b) corresponding radial cepstrum profile. Image reconstruction of (c) amorphous material and (d) crystalline material

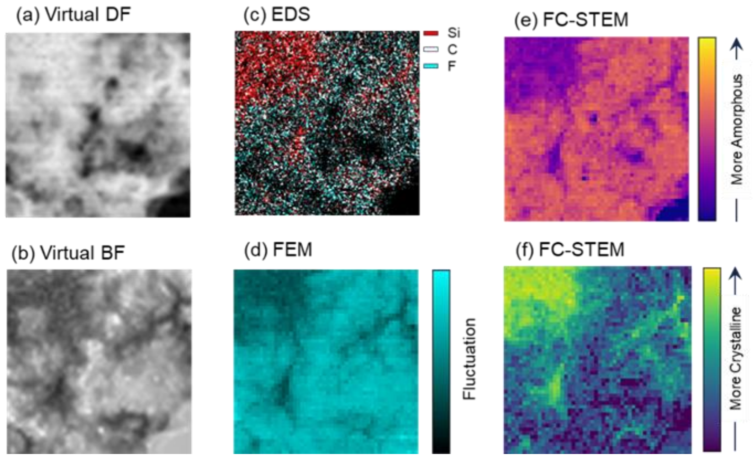

Figure 2. Comparison of different electron imaging visualizations of the same region. (a) Virtual dark field (DF), (b) Virtual bright field (BF), (c) energy-dispersive X-ray spectroscopy (EDS) compositional map, (d) FEM, and (e, f) FC-STEM of amorphous and crystalline regions.

References:

[1] S Pidaparthy et al., J. Electrochem. Soc. **168**(10) (2021), p. 100509.
[2] J-M Zuo et al., arXiv2110.02070 (2021).
[3] R Yuan, J Zhang, and J-M Zuo, Ultramicroscopy **207** (2019), p. 112837.
[4] K-H Kim et al., Micron **71** (2015), p. 39.
[5] C Ophus, Microsc. Microanal. **25**(3) (2019), p. 563.
[6] JM Gibson and MMJ Treacy, Phys. Rev. Lett. **78**(6) (1997), p. 1074.
[7] E Padgett et al., Ultramicroscopy **214** (2020), p. 112994.
[8] Y-T Shao et al., Ultramicroscopy **231** (2021), p. 113252.

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