2944
doi:10.1017/S1431927622011035
Microsc. Microanal. 28 (Suppl 1), 2022

Exploring Local Physics and Structural Behaviors with Automated Experiment In 4D-STEM

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Four dimensional (4D) scanning transmission electron microscopy (STEM) has recently enabled discovery of symmetry-breaking distortions, internal electric and magnetic fields in complex materials, and atomic resolution imaging of light elements. However, 4D-STEM experiments can produce considerably large volumes of data, which tend to be analyzed only in post processing, therefore choice in where these data sets are acquired is critical. In addition, compared to ordinary high angle annular dark field (HAADF)-STEM imaging, the per-pixel acquisition time in 4D-STEM experiments tends to be roughly three orders of magnitude longer, which may impart a significant amount of electron dose to the specimen. To address these issues, we present a deep kernel learning [1,2] (DKL) workflow applied to the 4D-STEM methodology in the form of an automated experiment that chooses measurement locations on-the-fly guided by physics-based functions.

Choosing where to acquire 4D-STEM and other analytical measurements has traditionally been a task of the experienced microscopist. While experience plays a large and important role, a degree of bias – perhaps even unconscious – is naturally present during traditional experiments. In 4D-STEM, since the data volumes are typically too large to analyze on-the-fly, it is especially critical that the 4D-STEM measurement location is chosen carefully. As an alternative, the DKL workflow allows active learning of structure-property relationships. **Figure 1** shows the DKL workflow as it pertains to 4D-STEM methods. In short, the sample structure is obtained in the fully acquired (and fast) dark field image and is correlated with a diffraction pattern measurement. Hence a relationship is established between a local image patch and the scalarized diffraction pattern. Here, scalarized simply means reducing a diffraction pattern to a single scalar value by imparting intended physics, e.g., calculation of the center of mass shifts. DKL continues to learn and refine these relationships on-the-fly and adapts the subsequent measurement locations within the space defined by the dark field image

Consideration of **Figure 2** shows that different physical properties of a specimen may be used to guide automated experiments in 4D STEM [3]. Specifically, relative charge distribution analysis is correlated to local structure in Figure 2a, while differences in lattice spacing (i.e., strain) are correlated to local structure in Figure 2b. These two different modes of 4D STEM are each used to guide the autonomous searching of physics. The “acquisition” shown in blue combines maximizing the prediction and minimizing uncertainty, and effectively is a map in which the most intense pixel is the next measurement. In Figure 2a, the relative strongest CoM shift was learned to exist near and around the Silicon dopant atoms in graphene, which intuitively makes physical sense given the higher Z element having a stronger effect on the beam. In Figure 2b, it is seen that in vacuum, the model deemed there to be highest degree of uncertainty (no diffraction), and at vacuum-graphene interfaces, the model deemed there to be the largest difference in lattice spacing. This work lays the foundation for future experiments in 4D STEM where expensive data acquisition can now be specifically targeted for finding physics of interest as well as unexpected behaviors [4].

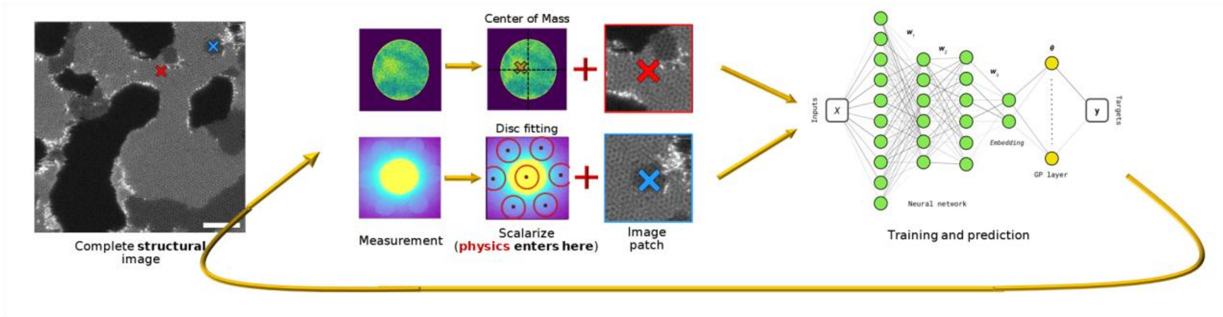

Figure 1. DKL workflow for 4D STEM automated experiments. A full structural dark field STEM image is acquired, then for a small number of random points in the image space diffraction patterns are acquired, and scalarized according to the intended physics. These scalar quantities along with the local image patches from where they were measured are sent as pairs into the DKL model, such that a structure-property relationship is derived. The next measurement location is guided by where the model predicts that the supplied physical criteria is maximized, and the model actively learns and with each new measurement and image patch.

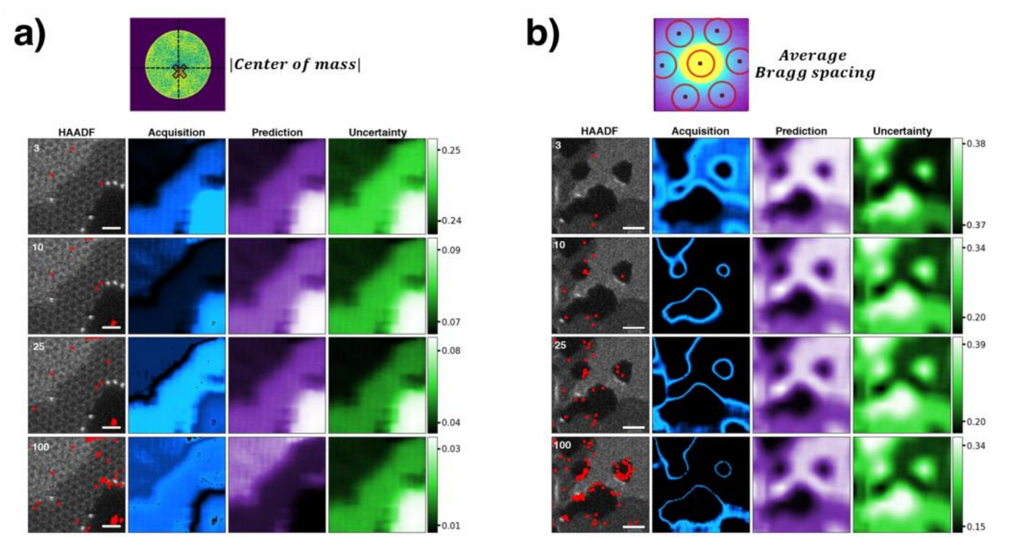

Figure 2. Two different schemes for 4D STEM automated experiment with DKL applied to twisted bilayer graphene. Relative electric field strengths are detected by computing the center of mass shifts of the beam, as is done in a), whereas relative lattice changes are measured by computing the average Bragg spacing in b).

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[4] This effort was based upon work supported by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES), Materials Sciences and Engineering Division (K.M.R., M.O.,

O.D., S.V.K.). Work was partially supported (MZ) and conducted using resources supported by Oak Ridge National Laboratory's Center for Nanophase Materials Sciences (CNMS), which is a U.S. DOE Office of Science User Facility.

https://doi.org/10.1017/S1431927622011035 Published online by Cambridge University Press