doi:10.1017/S1431927621000799
Microsc. Microanal. 27 (Suppl 1), 2021

Building an edge computing infrastructure for rapid multi-dimensional electron microscopy

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The development of aberration correction for scanning transmission electron microscopy (STEM) has made sub-ångström diameter electron probes routine at acceleration voltages even below 100kV [1, 2, 3]. Such probes can locate individual atomic columns in 3D materials and single atoms in layered materials with picometer-level precision, enabling microscopists to quantify complex phenomena such as ferroelectric polarization, symmetry breaking events in Kitaev materials, etc [4, 5, 6]. Along with aberration correction, the advent of high-speed direct electron detectors has made even more complex STEM measurements possible [7, 8]. These detectors allow microscopists to record the entire convergent beam electron diffraction (CBED) pattern at every scan position, resulting in a four-dimensional dataset, often referred to as 4D-STEM [9]. Some of the most ground-breaking recent electron microscopy results have used such datasets as the starting point for high precision strain measurements, differential phase-contrast microscopy, and ptychography [10, 11, 12, 13]. However, the major roadblock preventing widespread 4D-STEM adoption is computational, despite the highly detailed materials physics it illuminates. 4D-STEM generates gigabytes of data per second, and thus real-time observation-based feedback is impossible without rapid data processing [14]. Of the currently pursued approaches globally, machine learning-based routines have shown some of the most significant promises in speeding up the analysis pipeline to provide near real-time quantification of three-dimensional atomic structure from 4D-STEM data [15]. However, there is no system globally that combines neural networks with direct electron detectors in a microscopy system to deliver near-real-time materials physics.

Here in this work, we describe the components of a prototype system under development at ORNL that couples machine learning-based data analysis with electron microscopy to deliver rapid materials energetics from the microscope, with the setup's schematic demonstrated in **Fig. 1**. The electron beam transmitted through the sample encodes materials property information in its scattering behavior, captured with a pixelated detector at every scan position. For conventional electron microscopy, the scan points are chosen to satisfy Nyquist's sampling criterion, which means the scan step size is at most half the microscope resolution. In contrast to the process described before, our setup's initials scan positions are sparse to enable rapid data acquisition. The pixelated detector's data is then streamed to the edge computer, where the CBED images are converted to atomic structure descriptors using pre-trained neural networks. The neural networks are trained using simulated CBED patterns data on various material systems and at different beam conditions to prevent overfitting. These atomic descriptors are then streamed to a high-performance computer (HPC) to calculate the region's energetics being imaged in the microscope using first-principles density functional theory simulations. Using Gaussian process-based Bayesian Optimization (GP-BO), a map is subsequently generated from the calculated materials energetics of the imaged region [16]. However, since the initial scan sampling is sparse, the GP-BO estimates will have variable uncertainty in the area imaged by the microscope. The next set of scan positions are thus chosen to reduce this uncertainty, with these positions then fed back to the microscope acquisition computer, completing the feedback loop. The process is repeated till the desired uncertainty is reached over the entire field of view.

Microscopy, as practiced currently, is siloed. The acquisition, data-reconstruction, and simulations are performed on separate platforms by human operators. The system that we describe here aims to remove these bottlenecks and directly deliver materials physics rapidly and in an unsupervised fashion.

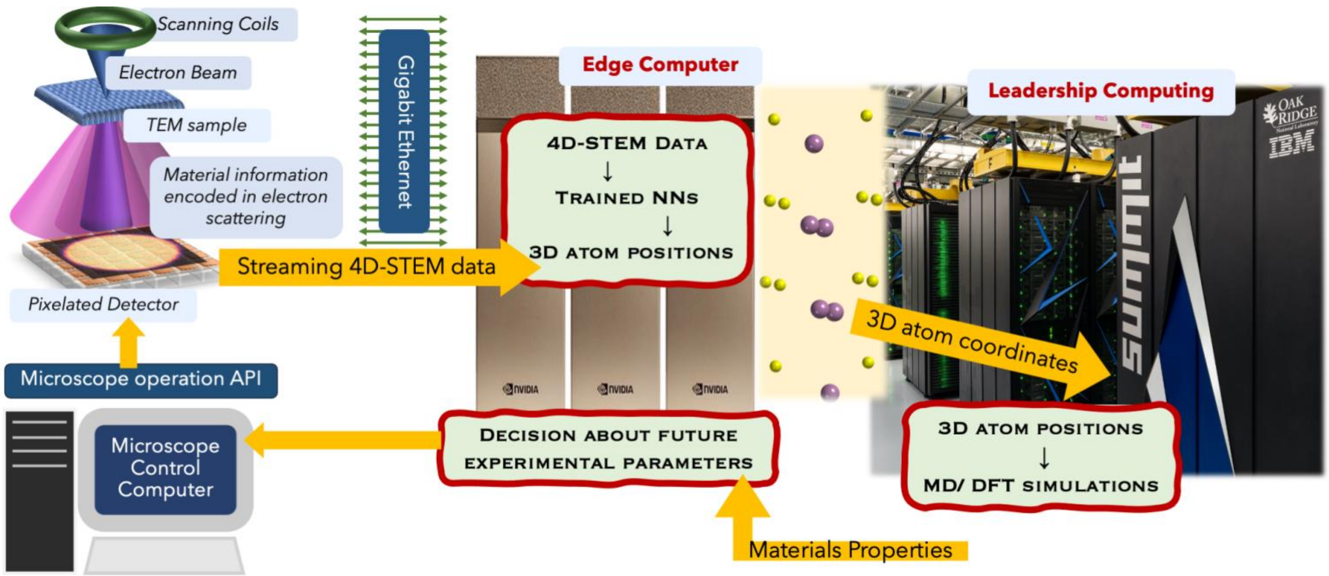

Figure 1. Schematic of an electron microscope connected to edge and leadership-class computing. The pixelated direct electron detector in the TEM is connected to an edge computer. During the course of a 4D-STEM experiment, data is streamed over gigabit ethernet from the detector. The edge system has GPU accelerated trained neural nets that rapidly convert the collected 4D-STEM data to atomic descriptors, which are streamed to the leadership class computer. Based on the atomic coordinates, energetics are calculated through first-principles simulations on the HPC. This is streamed back to the edge system, where based on gaussian process regressions, regions of further interest are located. These regions are the next set of 4D-STEM experiments and are streamed back to the microscope control computer.

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https://doi.org/10.1017/S1431927621000799 Published online by Cambridge University Press