Coherent X-ray diffraction imaging (CDI) is a powerful technique for operando materials characterization at the nanoscale.\(^1\) Uniquely, in this technique, the image resolution is not determined by the resolution of the optics used in the experiment but by the maximum angle through which the x-rays are scattered by the sample. The phase retrieval process can be computationally expensive, and the current methods have inherent deficiencies. First, this method does not include any physical knowledge of the material or the phenomena being studied. Furthermore, the resulting image is limited by the signal-to-noise of the data; so while signal may exist at greater angle, hence higher resolution, it may be insufficient for the phase retrieval to return a reliable image at that resolution.

There is tremendous opportunity to improve both the resolution and knowledge acquired from CDI through the implementation of data analysis methods that incorporate the physics of the material being studied. We have proposed to replace the phase retrieval step in CDI with machine learning algorithms to do atomistic model fitting of the data based on a training database derived from large scale molecular dynamics (MD) and statics (MS) simulations; hence the title, Atomistically Informed Coherent Diffractive Imaging. The fitted image will be inherently physical since it is derived from the physical properties of the atoms making up the material. It could also be significantly higher resolution, since the fitted model is inherently atomic resolution and the highest angle, weakly scattered signal in the measured patterns, can be directly fit with a reliability that surpasses the ability of current phase retrieval.

The proposed upgrade to the Advanced Photon Source (APS-U) will improve the coherence of the x-ray beams by a factor of at least 100. As such, coherent x-ray diffraction data measured on modern beamlines will come at either 2-3 orders of magnitude faster, or have 2-3 orders of magnitude greater signal. This flood of data, faster or larger, will need a modern data analysis pipeline to analyze it and retrieve relevant features. The current CDI phase retrieval cannot stand up to the task of addressing such big data challenges. The goal of our proposed data analytics approach will work directly on the measured data, bypassing inefficient and often memory intensive phase retrieval step, and provide atomistic information quickly.

The scientific playgrounds in which the ideas are being explored are studies of materials deformations and corrosion. This presentation will introduce the concepts being explored in the AICDI project at Argonne National Laboratory and show some recent successes (and failures) in this work.

References