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Autonomous Electron Microscopy Enabling Physics Discovery: Applications in Plasmonics of 2D Systems

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The scanning transmission electron microscope (STEM) allows access to a broad range of physics derived from a multitude of different signal modalities across different length scales. Advances in aberration correction, electron source monochromation, and detector technologies have enabled, for example, measurements of pm-level deviations in atomic columns, optical and vibrational spectroscopies at the nano- and atomic scales, and even studying beam sensitive specimens. Until now, the decision on where to acquire these advanced measurements has been in the hands of the microscope operator, inevitably introducing a level of bias in the experiment. While the experience of the microscopist is not in question, the sample space is generally vast and may contain unexpected and unexplored phenomena; therefore, in most cases, the microscopist will find what they sought out to find, and only rarely will they stumble upon an unforeseen event. A scheme that intelligently searches the sample space requires active learning, especially for unseen specimens.

In this work, we demonstrate autonomous experiments in the STEM using a deep kernel learning [1] (DKL) workflow, targeting the discovery of specific physical phenomena *via* electron energy loss spectroscopic (EELS) signals. Physics is embedded into this by introducing a scalarizer function that operates on the EEL spectrum to extract the intended physics, for example, by finding the maximum peak intensity in a spectral region, or by calculating peak ratios. Here, we use the high angle annular dark field (HAADF)-STEM image to access the specimen structure, in which local image patches are created at every pixel, representing the local geometry. Next, several EEL spectra are randomly collected from the image space defined by the HAADF to initialize the DKL model, and these spectra are reduced to a single value defined by the scalarizer function's physics criteria, then correlated to the local image patches from where they were acquired. Hence, a structure-property relationship is immediately formed after only a handful of measurements. Finally, the so-called acquisition function guides where each subsequent measurement will be taken – which continues to be appended to the DKL model such that learning occurs with each new measurement.

The workflow is experimentally demonstrated [2] on several experimental systems using a NION monochromated aberration corrected STEM (MACSTEM) that allows Python-based control of hardware. In **Figure 1**, the DKL workflow is applied to a several-layer specimen of $MnPS_3$, a 2D van der Waal material that undergoes beam-induced transformations [3] and therefore its natural state is beam sensitive, which also lends well as a motivation in using this workflow. Two separate scalarizers are demonstrated here to show the difference in the actively learned structure-property relationships and therefore the acquisition pathway. In choosing to search for the maximum of the peak ratio between P_1 and P_2, the acquisition pathway is shown to be highly concentrated near the top edge of the flake, seen in Figure 1C. Meanwhile, if the maximum of P_2 is instead used, it is clear that the sampled points all reside within the flake as in Figure 1D. Physically, P_2 corresponds to the bulk plasmon, and exactly should be maximum while within the bulk of the material. An increased ratio of P_1 to P_2 on the other hand indicates the presence of an edge mode (and absence of the bulk mode) – in other words, this mode exists outside the flake but strongly localized to the edge. The presence of such an edge mode in $MnPS_3$ was previously not known until the DKL workflow was used in this example, which greatly motivates the use of autonomous experiments.

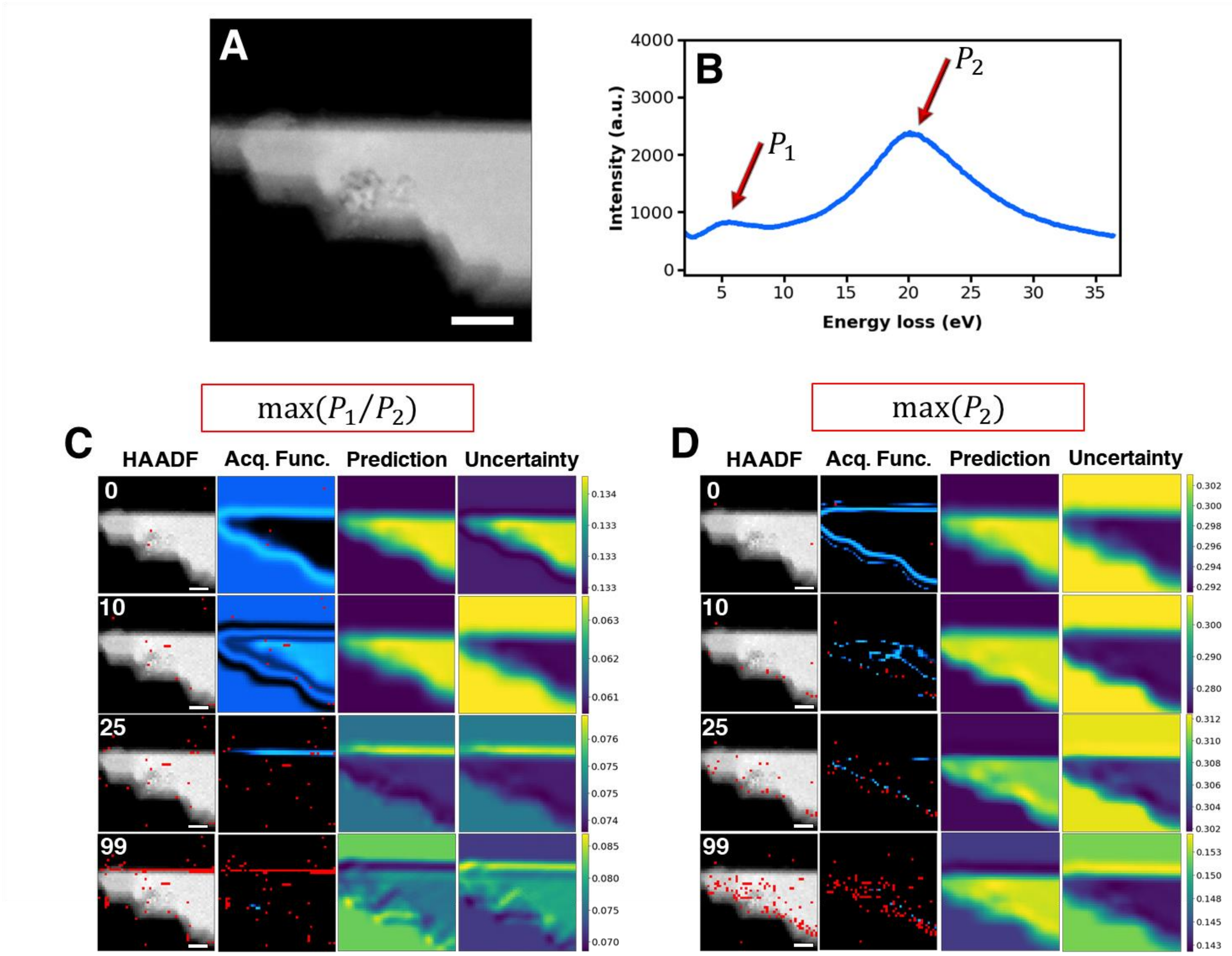

Figure 1. Autonomous experiments with DKL on a several-layer $MnPS_3$, a 2D van der Waal material, using two different scalarizer functions. HAADF-STEM image is shown in (A), where the EEL spectrum in (B) serves to illustrate how the scalarizer is chosen, shown with two prominent peaks, P_1 and P_2. The effect of different choice in physics-based scalarizers is compared in (C) and (D), where the maximum of the peak ratio between P_1 and P_2 is used to guide the automated experiment in (C), and the maximum of P_2 is used to guide that in (D).

While the active learning approach with DKL was demonstrated here for one material system, it is completely generalizable to any material system. In fact, any material that can be placed inside the microscope can be used with this workflow, since DKL actively learns relationships on-the-fly, i.e., no model pre-training is required. The DKL workflow also can be extended beyond spectral measurements

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like EELS and EDS to practically any type of higher dimensional analytical measurements, including 4D-STEM. Additionally, these principles can also be applied to other probe-based imaging techniques such as scanning probe microscopy. Combining the correlative predictions from DKL with instrument automation has proven to be successful in discovering physics in initial experiments and with more adoption, we hope DKL can be the harbinger of discovering many more phenomena [4].

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