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Building Atomic and Plasmonic Devices via Electron Beams: from Desired Structures to Desired Properties

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The evolution of the aberration correction in Scanning Transmission Electron Microscopy (STEM) has enabled multiple advances including high-resolution structural imaging, pm-level position detection of atomic columns, and ~3-5 mV resolution electron energy loss spectroscopy of the chemical and physical functionalities. These developments have further enabled the precise manipulation of the atomic structure of materials, including removal and formation of atomic planes in 3D materials, controlled atomic motion, and even direct atomic assembly of homo- and heteroatomic artificial molecules.

These advances naturally pose a question whether the atomically-precise structures with desired structures or functionalities can be created via electron beam manipulation [1]. Addressing this challenge in a systematic way requires a solution of several closely related tasks, ranging from engineering (beam and spectroscopy controls) to image analysis of the microscope data streams towards semantic segmentation, the discovery of the action and effect mechanisms for beam manipulation, and finally learning structure-property relationships in atomic systems.

Recently, the introduction of the programmable interfaces by several microscope manufacturers have enabled deploying Python codes as a part of the data acquisition, analysis, and control loop of electron microscope, allowing for implementation of automated experiment workflows. In this presentation, I will discuss several recent advances in STEM-EELS automated experiments as applied to quantum and nanoplasmonic systems.

As a first example, we explore the relationship between local structures and their plasmonic properties. For a static data set, we demonstrate the use of the variational encoder-decoder and dual autoencoders with common latent space to build local structure-property relationships. These allow to predict the possible plasmonic responses for a given materials system, and establish what configuration is likely to produce a desired plasmonic response. However, this approach suffers from out of distribution shift inevitable for correlative methods, and allows for only limited extrapolation for different microscope settings even for the same material. We further develop active learning methods that allow to learn these structure-property relationship in real time, and demonstrate its applicability for STEM-EELS of edge plasmons in 2D materials [2].

Secondly, we develop the instrumental framework for direct electron beam manipulation of materials, exploring the limits of sculpting 3D materials and the associated evolution of the plasmonic responses. The pathways towards the design of plasmonic structures with the desired functionalities are discussed.

Finally, we discuss the opportunities enabled by the electron beam in STEM towards the atomic and nanoscale manipulation, including atomic motion, atomic assembly, and introduction of novel structural orders [3-5]. Harnessing these phenomena towards applications requires real-time analysis of the data streams from the electron microscopes, achieved here *via* the ensemble learning iterative training (ELIT) approach for deep convolutional neural networks (DCNNs). With ELIT DCNN [4], the data streams can be converted to atomic coordinates and identities in real time, enabling robust atom identification and targeted intervention.

Further opportunities enabled by the application of active reinforcement learning in electron beam manipulation are discussed.

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