Electron energy loss spectroscopy: An atomic-resolution complement to photon-based spectroscopies – theory and applications

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Density functional theory (DFT) has been developed to the point that electronic, optical and magnetic properties of 3D and 2D materials, defects, interfaces, and complex nanostructures can be calculated with sufficient accuracy to compare with experimental data and play an equal-partner role in materials engineering. In recent years, however, aberration-corrected scanning-transmission-electron microscopes have acquired extraordinary spatial resolution in electron-energy-loss spectroscopy, which is an analog of x-ray and optical spectroscopies, but with atomic-scale resolution. Theory, however, has lagged behind in its ability to calculate what is being measured.

In this talk, we will describe the development of the theory and computer codes that combine DFT in the all-electron Projector Augment Wave (PAW) formulation with dynamical scattering theory. We track the evolution of the microscope's focused electron beam within the sample, i.e., we include diffraction and interference effects, and compute directly through matrix elements the excitation of either core or valence electrons. In combination with STEM data, we have a powerful tool to probe the electronic and magnetic properties of complex nanostructures that rivals photon-based spectroscopies. We will show applications to core-electron excitations in complex 3D oxides [1] and to valence-electron excitations in pristine monolayer graphene [2] and in impurities in graphene [3] and BN, demonstrating that the method provides robust signatures of distinct impurity configurations.

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