Correlation in 2D Materials: Magnetism, Proximity Effects, and Reactivity Through the Lens of Quantum Monte Carlo

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2D materials are an exciting new class of materials that hold great promise for a wide range of engineering applications because of their exceptional tunability. Indeed, these materials' band gaps, magnetic and topological properties, and conductivity can be dramatically altered just by stretching or stacking them. Despite their promise, however, our modern understanding of 2D materials is partially obscured by the fact that most have been computationally modeled using Density Functional Theory (DFT), a theory that cannot readily describe electron correlation and is known to yield widely varying results for such central quantities as band gaps and molecular binding energies depending upon the exchange-correlation functional employed.

In this presentation, I will describe my group's recent efforts to model an array of 2D materials using fully correlated quantum Monte Carlo (QMC) techniques and the unanticipated physics these efforts have revealed. I will begin by demonstrating how stochastic methods can advance beyond mean field techniques to properly resolve the geometry, spin density, magnetic moment of monolayer CrI_3, the first 2D magnetic material discovered. This modeling reconciles features observed through scattering experiments but that could not previously be reproduced using less accurate modeling. Subsequently, I will turn to showing how correlated sampling techniques can be used to model the catalytic reactions and the catalytic properties of these materials. Altogether, this work paints a more complete picture of the electronic structure and properties of this increasingly important class of materials, while also demonstrating the utility of quantum Monte Carlo methods for doing so.