Using Quantum Monte Carlo to Illuminate the Properties of 2D Materials: A CrI3 Case Study

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In recent years, 2D materials have garnered a wealth of interest because of their unique low-dimensional physics, exotic magnetism, and the relative ease with which their properties can be tuned via doping, crinkling, strain, and stacking. The first magnetic 2D material to be discovered, monolayer (ML) CrI_3, is particularly fascinating due to its ground state ferromagnetism, which can be employed to design spintronic materials. Yet, because monolayer materials are notoriously difficult to probe experimentally, much remains unresolved about their properties. In this talk, I will discuss how Diffusion Monte Carlo (DMC) was used to accurately predict the atomic magnetic moments, lattice parameters, and geometry of ML CrI_3, as an example 2D material. Notably, we find that the atomic magnetic moments in CrI_3 are 3.62 μB per chromium and -0.145 μB per iodine, which are both larger than previously anticipated, supporting a potentially large ligand superexchange-dominated magnetic anisotropy. Our DMC-predicted lattice constant (a_0) of 6.87 Å is also very close to the experimental a_0 = 6.84 Å, demonstrating the predictive power of DMC for geometry and magnetism in 2D materials.