A survey of methods for predicting electronic structure

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The electronic band structure of a solid crystal is central to understanding its conductivity, magnetic ordering, as well as its optical properties. At present, the dominant choice for determining electronic structure is density functional theory (DFT), which has achieved a high degree of success despite the simplicity of the independent-electron model on which the Kohn-Sham construction is built. Standard DFT calculations however fail to predict the correct electronic properties of materials when the interaction between electrons is strong; the Mott insulator NiO is a classic example.

While different flavors of theory have been applied to different materials, a comprehensive, systematic survey that compares the different computational approaches to electronic structure prediction is lacking. We fill this gap through this study by examining three systems – Ag, graphene and FeSe – with the same set of tools: (1) local density approximation (DFT-LDA), (2) generalized gradient approximation (DFT-GGA), (3) a hybrid approach that combines electron-density and orbital methods (HSE functional), and (4) the inclusion of long-range dispersion (DFT+D3). Calculations using (5) GW perturbation theory and (6) dynamical mean theory (DFT+DMFT) are then carried out to address the many-body problem for electrons. Silver, graphene and FeSe were handpicked to represent a diversity of materials classes: a transition metal, a 2D semimetal, and a prototype for iron-based high-T_c superconductors [1, 2, 3]. Finally, simulated bands and photoemission spectral function maps are compared with data from high-resolution angle-resolved photoemission spectroscopy (ARPES) experiments we carried out on CVD-grown graphene on silicon, and on the Ag(111) surface.

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