Development of a charge-self-consistent DFT+DMFT method and its application to crystal-field splitting

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Strongly correlated electron systems exhibit various phenomena that are difficult to describe within conventional density functional theory (DFT). Combining DFT with dynamical mean-field theory (DMFT) provides a powerful framework for treating local dynamical correlations [1]. In one-shot DFT+DMFT calculations, however, correlation-induced changes in the charge density are not fed back into the DFT calculation. Charge self-consistent (CSC) DFT+DMFT overcomes this limitation by updating the charge density using the interacting Green's function and iterating the DFT and DMFT calculations self-consistently.

In this work, we developed a Wannier-based CSC DFT+DMFT framework including spin-orbit coupling (SOC) using Quantum ESPRESSO (QE), Wannier90, and DCore, with a particular focus on f-electron materials. The Wannier representation provides a localized orbital basis that facilitates model analysis and the inclusion of SOC, making it well suited for studying crystal-field splitting and orbital character in correlated f-electron systems.

For the benchmark, we applied our framework to Ce_2O_3 without SOC using the Hubbard-I impurity solver and compared the results with those obtained using solid_dmft [2], an existing CSC DFT+DMFT implementation based on QE, Wannier90, and TRIQS. The convergence behavior and band structure show good agreement, demonstrating the consistency of the two implementations (Fig. 1). Based on this validation, we applied the framework to γ-Ce and CeB_6 to investigate the crystal-field splitting within the $J = 5/2$ multiplet [3][4]. In both systems, the inclusion of charge self-consistency modifies the relative energies of the crystal-field states and improves the agreement of the crystal-field levels with experiment. These results suggest that the correlation-induced redistribution of the charge density is important for accurately describing the crystal-field level structure of Ce-based 4f-electron materials.

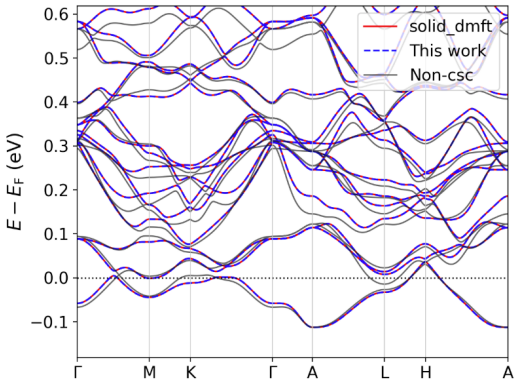


Fig. 1. Wannier band structure of Ce_2O_3

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