Accurate prediction of electronic and optical excitations in 3d and 2d materials from density functional theory

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Abstract

Density functional theory (DFT) has become the workhorse of first principles computational materials science. Nonetheless, it has struggled, often even qualitatively, in the description of electron and optical spectroscopy. Specifically, research has been fraught for decades with difficult questions as to the extent to which spectroscopic conclusions can be drawn from DFT even in principle, followed by serious concerns as to the reliability of typical approximation in DFT (including time-dependent DFT), especially for the solid state.

Here, a novel approach to overcoming these difficulties, involving Wannier-localization based optimal tuning of a screened range-separated hybrid functional, is presented. It is shown that quantitative accuracy for a wide range of semiconductors and insulators is achieved without any empiricism. Moreover, it is shown that an extension of the method achieves the same for 2d semiconductors and insulators. This opens the door to many DFT-based true predictions of electronic and optical properties, to high-throughput calculations, and to a systematic choice of the starting point for many-body perturbation theory calculations.