The density is not enough

Nicola Marzari[1]

[1]Theory and Simulation of Materials, EPFL, Lausanne, Switzerland

Materials' simulations based on density-functional theory (DFT) have become an extremely powerful and widely used tool for scientific discovery and technological advancement. Still, in the current approximations, they remain an imperfect tool for predicting materials' properties, with open and urgent challenges in the quest towards qualitative and quantitative accuracy.

Several of these challenges stem from the remnants of self-interaction in the electronic-structure framework, leading to qualitative failures in describing some of the fundamental processes involved e.g. in energy applications - from charge-transfer excitations to photoemission spectra to the structure and reactivity of transition-metal complexes.

I'll discuss these challenges in realistic case studies, and present a brief overview of some of our suggestions for possible solutions - including constrained DFT, DFT + onsite and intersite Hubbard terms, and Koopmans' compliant energy functionals. In particular, I'll highlight how Koopmans' compliant functionals point to a beyond-DFT formulation where both total energies and spectroscopic properties can be accounted for. Such framework will be illustrated with applications to real systems and with simplified models that can be solved exactly.

Work done in collaboration with I. Dabo (Penn State), A. Ferretti (U. of Modena), Matteo Cococcioni (EPFL), G. Borghi (EPFL), Linh Nguyen (EPFL), D. D. O' Regan (EFPL), H. J. Kulik (MIT), C. H. Park (SNU), Patrick Sit (CUHK).