

Koopmans spectral functionals: Bridging density-functional and many-body perturbation theory.

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Accurate first-principles predictions of spectral properties of materials are key to understanding, discovering and designing materials for countless applications - from electronics to energy harvesting and photonics. Koopmans spectral functionals provide a novel and functional approach - as opposed to diagrammatic techniques - to simultaneously and accurately describe ground state properties and charged excitations - i.e., the electronic band structure - of atoms, molecules, nanostructures and periodic crystals. This is achieved by augmenting standard density functionals with simple, yet physically motivated orbital-density-dependent corrections based on the three constraints of linearization, localization, and screening, to enforce the correct description of charged excitations.

After introducing the general framework and its implementation, I will discuss an extension of the theory to fully and consistently incorporate the spin degree of freedom. In this formulation, state-dependent potentials emerge that can be interpreted as a local counterpart to the electronic self-energy, albeit already accounting for spin-dependent interaction and screening effects beyond the random phase approximation typical of standard diagrammatic approaches (GW). I will present theoretical evidence supporting this view and numerical simulations demonstrating excellent agreement with experiments and state-of-the-art Green's function methods in predicting ionization potentials of a large set of molecules (the GW100 test set), as well as photoemission and absorption spectra of organic donors and acceptors, and band gaps and band structures of semiconducting and insulating crystalline materials. Being this a functional framework, the straightforward advantages are that forces and other derivatives are also readily accessible, and that the numerical parameters are those typical of density functional calculations.