Electronic correlations in solids via finite-temperature Green's functions

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Imaginary-time/frequency Green's function formalism offers a viable route to modeling the electronic structure of solids competitive with the traditional wavefunction based approach. Its fundamental advantage stems from simultaneous inclusion of the effects of both electronic correlations and finite temperature, the latter attainable without an explicit reference to the excited states. A temperature-dependent Green's function, if evaluate self-consistently, provides a direct access to the thermodynamic properties, and its continuation to real frequencies yields one-electron spectra. Another valuable advantage consists in a particular suitability of Green's functions for embedding techniques based on a multilevel self-energy approximation.

Self-consistent Green's function method with the second-order perturbative self-energy (GF2), unlike its wavefunction counterpart MP2, is known to perform qualitatively correct in a variety of cases including disappearance of the band gap and emergence of strong correlations. In this work we explore GF2 applied to realistic crystalline systems described by full quantum-chemical Hamiltonians. We also investigate routes to a higher-level non-perturbative treatment of electronic correlations in solids where simple perturbative schemes are insufficient.