TITLE:
Orbital-density dependency driving a functional theory for spectral properties

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Energy functionals which depend explicitly on each individual orbital density, rather than the total charge density, appear naturally when applying self-interaction corrections to density-functional theory (e.g. in the case of the Perdew-Zunger or the Koopmans-compliant [1] functionals). In these cases the total energy is not invariant under unitary rotations of the orbitals, and functional minimization leads to orbital-dependent Hamiltonians.

Rather than a limitation, we argue that this is a powerful feature, and show how it is possible to interpret the orbital-dependency of densities and potentials as an effective and discretized frequency dependency, in close analogy to the quasi-particle approximation of frequency-dependent self-energies. Such framework is naturally amenable to describe electronic spectroscopies [2], and is free from e.g. the constraint of having derivative discontinuities in the exact formulation. We believ that a functional theory for the spectral density is therefore emerging, able to address at the same time total energies and spectral properties

[1] I. Dabo, A. Ferretti, N. Poilvert, Y. Li, N. Marzari, M. Cococcioni, Phys. Rev. B 82, 115121 (2010); G. Borghi, A. Ferretti, L. Nguyen, I. Dabo, N. Marzari, Phys. Rev. B 90, 075135 (2014).
[2] A. Ferretti, I. Dabo, M. Cococcioni, N. Marzari, Phys. Rev. B 89, 195134 (2014).