Critical reassessment of recent developments in functional theory: From Hartree-Fock to *i*-DMFT

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A recent scientific endeavour has put forth a variation of one-particle reduced density matrix functional theory (DMFT) based on an entropic extension of the Hartree-Fock functional. This *i*-DMFT would reach the accuracy of sophisticated configuration interaction methods while its computational cost, however, would be still comparable to Hartree-Fock. These supposedly promising prospects call for some reassessment and a further specification of the role of universality and duality in functional theory in general: First, we conclusively reveal that *i*-DMFT assigns to every molecule \mathcal{M} its own functional $\mathcal{F}_{\mathcal{M}}$ and thus violates fundamental principles of functional theory. Despite abandoning universality almost entirely, *i*-DMFT still does not reach chemical accuracy. Our numerical calculations also show that the expectation values for various one-particle observables are not correctly reproduced. All these insights suggest a two-fold change of paradigm: (i) the Hartree-Fock functional might be the wrong reference for the development of functional approximations and (ii) the quest in chemistry for accurate descriptions of energy surfaces would result in a simple *molecular geometry* functional theory rather than in a DMFT or DFT.

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