Density-Functional Theory: Time to Move on?

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Density-functional theory can be a very powerful tool for scientific discovery and technological advancement. Still, it remains an imperfect tool, with open and urgent challenges in our quest towards qualitative and quantitative accuracy, and in our ability to perform quantum simulations under realistic conditions.

Several of these challenges stem from the remnants of self-interaction in our electronic-structure framework, leading to qualitative failures in describing some of the fundamental processes involved in energy applications - from charge-transfer excitations to photoemission spectra, to the structure and reactivity of transition-metal complexes. Ill discuss these challenges in realistic case studies, and our suggestions for possible solutions - including constrained DFT, DFT + onsite and intersite Hubbard terms, and Koopmans' compliant energy functionals. In particular, I'll discuss how self-interaction corrected functionals lead naturally to a beyond-DFT formulation where both total energies and spectroscopic properties can be accounted for.

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