What is wrong with DFT ?

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Electronic-structure modeling has become a very powerful tool to understand, predict, or design the properties of complex materials and devices. It is also an imperfect tool, with many 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 e.g. mixed-valence complexes, electron-transfer excitations, and even single-particle energies. I'll discuss these effects in realistic case studies, and suggest possible solutions based on constrained DFT, on extended Hubbard functionals, or on imposing a generalized Koopmans' condition. I'll also highlight how the calculation of magnetic properties (NMR/EPR) or the use of wavefunction techniques can provide stringent validation criteria for novel developments.