Local hybrid exchange-correlation functionals based on the static dielectric function

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Kohn-Sham density functional theory (KS-DFT) provides an appealing cost/performance ratio, but its accuracy crucially depends on the choice of the (approximate) exchange-correlation (xc) functional. A new class of flexible, accurate, and efficient functionals - local hybrid functionals (local hybrids) – emerged during the past 16 years [1]. Local hybrids replace the constant exact-exchange (EXX) admixture of traditional (global) hybrids by a real-space-dependent one, governed by a so-called local mixing function (LMF). The latter is a key ingredient in constructing local hybrids, and a variety of different LMFs are known [1].

In this work we construct the LMF as an inverse of the static dielectric function, ε_s^{-1} ("d-LMF"), following the concept of screened Coulomb interactions of electrons in semiconductors [2-4]. We build ε_s as a parameterized function of the local ionization potential (LIP), defined in refs. [5,6]. Our ansatz for the LIP is more flexible than the original one ($\frac{1}{8}\left|\frac{\nabla\rho}{\rho}\right|^2$, where ρ is the electron density) and employs, in addition to the gradient of the density, also its Laplacian. The theoretical foundation of such d-LMFs is solidly grounded in the idea of screening exact exchange locally and they thus provide a more satisfactory justification than some other proposals in the literature, while being sufficiently flexible to suggest practical usefulness. They furthermore justify the use of the reduced density gradient in some previously suggested LMFs. Other crucial ingredients of local hybrids are the semi-local exchange-energy density (which is mixed with the EXX one) and the semi-local correlation energy. PBE exchange [7] and B95 correlation [8] turned out to be a particularly successful combination in conjunction with the d-LMF. Our preliminary validation studies of the resulting local hybrids are promising: the functionals yield good results simultaneously for atomization energies, reaction barriers, total atomic energies, and some other properties. In this work we also analyze d-LMFs in the context of known exact constraints.

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