Designing computationally efficient Laplacian-based kinetic energy density functionals

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In recent years interest has grown in the use of the orbital-free kinetic energy density (KED) in density functional theory, either for "deorbitalizing" metaGGA functionals or to remove orbitals entirely from the Kohn-Sham formulation. The Perdew-Constantin (PC) KE metaGGA [1] and later variants demonstrate the effectiveness of the use of the Laplacian of the density to model the transition between the slowly varying electron gas and single-orbital limits in such functionals. Unfortunately, the use of the Laplacian in this context creates unphysically spiky Pauli potentials that are difficult to converge and lead to inherently noisy results. To ameliorate this problem, we construct and test a smoothness measure based on the variational description of Poisson's equation, applied to the Laplacian-generated terms in the potential. Optimization of this measure is used to develop dramatically smoother models of the electron localization transition.

We discuss new smoothed kinetic energy density functionals derived from PC and the recent RPP [2] designed to remove the dependence on kinetic energy of the r2SCAN metaGGA. Applying these to standard test sets of molecules and solids yield performance in predicting structural properties that is on par or better than the parent functional, and, for VASP calculations for solids, does so with consistently less wall time. However, the number of self-consistent steps needed for convergence can be significantly longer for Laplacian=based potentials than for the original orbital-dependent potentials, particularly for molecular dynamics calculations, indicating the existence of instabilities that remain to be resolved.

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[1] J. P. Perdew and L. A. Constantin, Phys. Rev. B 75, 155109 (2007).
[2] A Kaplan and J Perdew Phys. Rev. Materials 6, 083803 (2022)