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 to the von Weizsacker or single electron-pair 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 Laplacian-dependent 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. However, the number of self-consistent steps needed for convergence depends upon choice of optimizer and material and can be significantly longer for smoothed than for non-smoothed Laplacian-based potentials, indicating the existence of undiagnosed secondary issues.

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[2] A Kaplan and J Perdew Phys. Rev. Materials 6, 083803 (2022)