Regularization of the reduced density laplacian to improve de-orbitalization

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In the current setting of applications of density functional theory (DFT) to ab initio simulations of very large systems there is a profound tension between complexity of the exchange-correlation (XC) approximation and computational cost. Explicit dependence on the Kohn-Sham [1] orbitals is introduced in meta-GGA XC functionals to gain accuracy. The cost is both computational (time) and conceptual, since the generalized KS procedure typically is the only feasible one. De-orbitalization [2] attempts to retain meta-GGA accuracy by replacing the KS kinetic energy density τ_{KS} with a function of the density, its dimensionless gradient s, and dimensionless Laplacian q. The key quantity in this transformation is the α chemical region indicator, which goes from $\alpha(s, \tau_{KS})$ to $\alpha(s, q)$.

Recently, we have focused on issues arising from de-orbitalization of two versions of the Tao-Mo XC called *rreg* TM
[3] and *v2-sreg*TM [4, 5]. They use both α and an another indicator z' and z_{rev} respectively. We found that the original de-orbitalization strategy does not work well for such two-indicator functionals. Numerical exploration showed that the abrupt removal (Heaviside step function) of regions in which $q < 0$ significantly improves error comparisons on molecular and solid test sets. However, this cutoff modifies the behavior of α relative to its orbital-dependent parent by introducing singularities and other odd disparities.

To address this challenge, we introduce a novel variable, $q_s = q\sigma(q)$ in which $\sigma(q)$ is a sigmoid function. In de- orbitalization, q_s, replaces q. The key advantage of q_s is that it eliminates effectively all $q < 0$ regions smoothly, hence retains continuous, smooth behavior for α.

We validate the de-orbitalization of our selected metaGGA functionals using standard molecular and solid test sets. procedure. Use of q_s instead of q thus extends de-orbitalization reasonably effectively to two-indicator XC functionals.

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