A computational methodology for kinetic energy density dependent meta-GGA functionals in finite-element based DFT calculations with accelerated self-consistent field iterations

Srinibas Nandi,[1] and Phani Motamarri*[1]

[1]*Computational and Data Sciences, Indian Institute of Science Bangalore*
phanim@iisc.ac.in

First principles modelling methods have become a ubiquitous tool for studying various properties of materials in diverse applications, with Kohn-Sham density functional theory (DFT) being the frontrunner. While Kohn Sham DFT is theoretically exact, the specific form of exchange-correlation energy remains elusive and is approximated using different levels of theory with increasing complexity. Kinetic energy density dependent meta-GGA (τ-mGGA) functionals like SCAN that incorporate kinetic energy density alongside local electron density and gradient of electron density, offer improved accuracy but pose computational challenges. To this end, the regularized-restored SCAN (r^2SCAN) exchange-correlation functional represents a promising compromise between numerical stability and accuracy, meeting the most known constraints of density functional approximations. The current work introduces finite-element discretization and an efficient numerical implementation of τ-mGGA functionals in the open-source DFT-FE code[1] to conduct large-scale electronic structure calculations. Despite improved accuracy, the usage of τ-mGGA functionals is somewhat limited due to the numerical instability[2] and slow convergence nature of the self-consistent field (SCF) iterations associated with these functionals. To mitigate this, we propose a new preconditioned mixing scheme within the aforementioned finite-element framework to accelerate the convergence of SCF iterations when using τ-mGGA functional, leveraging a low-rank approximation of the dielectric matrix.[3] We assess the robustness and efficiency of aforementioned computational methods across a range of system sizes and material systems highlighting the applicability of the framework to generic boundary conditions.

References

1. Das, S.; Motamarri, P.; Subramanian, V.; Rogers D.M.; Gavini V. *Comput. Phys. Commun.*, **2022**, *280*, 108473
2. Lehtola, S.; Marques M.A.L. *J. Chem. Phys*, **2022**, *157*, 174144
3. Das S. ; Gavini V. *Phys. Rev. B*, **2023**, *107*, 125113