Linear and non-linear eigensolver-based techniques for quantum modeling and simulations: applications to carbon-based materials

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In a recent article [Phys. Rev. B 79, 115112, (2009)], the FEAST algorithm has been presented as a general purpose eigenvalue solver which combines accuracy, robustness, high-performance and (linear) parallel scalability. Here, FEAST will be presented beyond the "black-box" solver as a fundamental modeling framework for the electronic structure problem spanning the research fields of solid-state physics and quantum chemistry.

First, it will be shown that FEAST can naturally address the original numerical complexity of the electronic structure problem as formulated by Slater in 1937 [Phys. Rev. 51, 846-851, (1937)]. The non-linear eigenvalue problem of type $\mathbf{A}(\lambda)\mathbf{x} = \lambda\mathbf{B}\mathbf{x}$ arising from the muffin-tin decomposition of the real-space domain is first derived and then reformulated to be solved exactly within the FEAST framework. This new framework is presented as a fundamental and practical solution for performing both accurate and scalable electronic structure calculations, bypassing the various issues of using traditional approaches such as linearization and pseudopotential techniques.

Second, we present a modification of the FEAST algorithm that solves the full non-linear eigenvector problem of type $\mathbf{A}(\mathbf{x})\mathbf{x} = \lambda\mathbf{B}\mathbf{x}$ (i.e. Schrödinger/Poisson) as an efficient and robust alternative to traditional self-consistent field method (such as Newton-Broyden, Anderson mixing, direct inversion of the iterative subspace and mixed Pulay techniques). The resulting approach is potentially one order of magnitude faster than conventional iterative methods, and it converges to the correct solution regardless of the choice of initial guess.

Finally, we present and discuss FEAST-based time-domain propagation techniques for time dependent quantum problem. Efficient time-dependent simulations have become increasingly important for characterizing the electron dynamics under time dependent external perturbations such as electromagnetic fields, pulsed lasers, and particle scattering. Using innovative spectral decomposition and discretization of time-ordered evolution operators, we aim to address the traditional limitations in term of trade-off between robustness and performances in such calculations.

In order to illustrate the efficiency of the FEAST framework, numerical examples are provided for various molecules and carbon-based materials using our in-house all-electron finite element implementation for the DFT/Kohn-Sham/LDA problem.