Finite-element methods for noncollinear magnetism and spin-orbit coupling in real-space pseudopotential density functional theory

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We present an efficient finite-element approach[1] for large-scale real-space pseudopotentials density functional theory (DFT) calculations incorporating noncollinear magnetism and spin-orbit coupling effects. The approach, implemented within the open-source DFT-FE[2] computational framework, fills a significant gap in real-space DFT calculations using finite element basis sets, which offer several advantages over traditional DFT basis sets. In particular, we first derive the finite-element (FE) discretized governing equations involving two-component spinors, leveraging a local reformulation of DFT electrostatics. To evaluate the widely used GGA exchange-correlation potentials in these governing equations under the locally collinear approximation, we devise a numerical strategy that avoids the computation of ill-defined gradients of magnetization direction near zero magnetization. Additionally, we utilize an efficient self-consistent field iteration approach based on Chebyshev filtered subspace iteration procedure exploiting the sparsity of local and non-local parts of FE discretized Hamiltonian to solve the underlying nonlinear eigenvalue problem based on a two-grid strategy. Further, we propose using a generalized functional within the framework of noncollinear magnetism and spin-orbit coupling with a stationary point at the minima of the Kohn-Sham DFT energy functional to develop a unified framework for computing atomic forces and periodic unit-cell stresses. Validation against plane-wave implementations shows excellent agreement in ground-state energetics, vertical ionization potentials, magnetic anisotropy energies, band structures, and spin textures. The proposed method achieves significant speed ups for semi-periodic and periodic systems with 5000-7000 electrons in terms of minimum wall times compared to widely used plane-wave implementations on CPUs in addition to exhibiting notable computational advantage on GPUs for material systems with as many as 20,000 electrons. Furthermore, extensions of the afforementioned computational framework within the projector augmented wave formalism[3] to further reduce the computational cost will also be discussed, highlighting the potential applications to studying TMD bilayers with low twist angles incorporating spin-orbit coupling effects. The proposed approach offers a fast, scalable, and systematically convergent framework for large-scale DFT calculations accounting for noncollinear magnetism and spin-orbit coupling, enabling more complex material simulations and extending the range of ab-initio studies.

References

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