

Accurate, Scalable All-Electron Theory Across the Periodic Table: Organics, Inorganics, Hybrids

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This talk summarizes recent progress towards accurate, scalable all-electron theory of materials and molecules on equal footing in a framework based on numerically tabulated, atom-centered orbital basis functions, the FHI-aims code [1]. The primary production method is density-functional theory (DFT, semilocal and hybrid) as well as many-body perturbation theory such as *GW*, the random-phase approximation, the Bethe-Salpeter Equation, and others. Seamless scalability up to thousands of atoms (non-periodic and periodic) is guaranteed by the open-source "ELSI" infrastructure [2], a general-purpose interface for massively parallel (generalized) Kohn-Sham solutions using traditional $O(N^3)$ or better-scaling solvers, now used by several different electronic structure codes. We also touch upon recent work on benchmarking different approaches to spin-orbit coupling [3] and on a GPU implementation of key algorithms for (currently) semilocal density functional theory with significant speed-ups over multicore execution on the CPU part of a node. A key strength of the code is its ability to carry out hybrid DFT calculations [4] including spin-orbit coupling for periodic system sizes above thousand atoms without any significant approximations to the potential shape or basis set accuracy. This is particularly relevant in recent work on large, crystalline layered organic-inorganic hybrid perovskites, an emerging class of semiconductors that are tunable by leveraging the arsenal of synthetic organic chemistry and now subject to attention by a very large community. We demonstrate this tunability for several examples of lead-based [5] and lead-free [6] organic-inorganic hybrids, showing how computational predictions can facilitate effective candidate material selection for experimental synthesis [7].

The methodological work reported in this talk would not be possible without the continued support and contributions from the very large community of developers and users of the FHI-aims code, as well as the community around the ELSI infrastructure. The work on perovskites leans heavily on close collaborations with leading experimental colleagues, particularly the group of David Mitzi (Duke University).

References

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