All-electron Electronic Structure Pathway to Challenges in Molecules, Materials and "Energy"

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This talk describes recent advances of all-electron electronic structure methods as implemented in the FHI-aims code, covering semilocal and hybrid DFT for periodic and non-periodic systems on equal footing, as well as many-body perturbation theory such as the random-phase approximation and GW. Our approach builds on numerically tabulated atom-centered basis sets,[1] which facilitate very high numerical accuracy,[2] but that can also be employed for routine production simulations up to very large, complex solid or molecular systems (1,000s of atoms). Key methodological components that will be discussed include: The massively parallel eigenvalue solver library ELPA; a new electronic structure infrastructure "ELSI" that will bundle approaches to solve or circumvent the DFT eigenvalue problem towards even larger systems; and an accurate, localized resolution of identity strategy for the two-electron Coulomb operator that facilitates all-electron $O(N)$ hybrid DFT calculations up to 1,000 atoms,[3,4] as well as many-body theory beyond DFT. Finally, we show how these developments enable us to make accurate predictions for complex, nanostructured materials, particularly electronically tunable, crystalline organic-inorganic hybrid materials of interest for photovoltaic or optoelectronic applications.

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[4] A. Ihrig *et al.*, New J. Phys. **17**, 093020 (2015).