Automating Local Orbital Selection in the LAPW+lo Framework

Automating Input Files for Density Functional Theory using Linearized Augemented Plane Waves and Local Orbitals

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In Short

- LAPW+lo methods offer benchmark quality precision.
- Selecting which atomic local orbitals (LOs) to use in a calculation is not always straightforward.
- Aim to better understand precision vs. computational expense trade-off in LOs.
- HQ dataset is needed to answer this question.
- End goal: recommend LO set for a user-specified precision.

Density functional theory (DFT) is the most widely used method in modern computational condensed-matter physics and chemistry as reflected by the existence of dozens of implementations [1]. The numerical precision of density-functional-theory (DFT) calculations depends on a variety of computational parameters, one of the most critical being the basis-set size. The basis set in DFT depends on the implementation. Linearized-augmented plane wave implementations with local-orbitals (LAPW+lo) methods have recently been shown to achieve micro-Hartree precision [1]. The method requires the user to specify local orbitals to be used in the calculation's basis set which are active in atomically centered spherical regions, called muffin tins. The precision of the method therefore relies on the user's ability to specify a good set of local orbitals. Specifying too many will slow down the calculation, while specifying too few could sacrifice precision.

This project seeks to better understand the balance between precision and computational time depending on the local orbitals used in an LAPW+lo calculation. To do this, we will use a set of 71 elemental solids chosen to represent the periodic table, of various crystal structures. These structures have been used in previous reproducibility and precision of DFT studies [2, 3]. A specific LAPW+lo implementation will also be used, namely, `exciting` which is a full-potential all-electron package implementing DFT and methodology beyond [1]. A major benefit of using an all-electron code is that it can explore the physics of core electrons and not just valence electrons. The code implements a family of linearized augmented planewave (LAPW) methods including the LAPW+lo framework. With `exciting` a user is allowed to control physical parameters such as the muffin-tin radius, the cutoff energy and the number of local orbitals. The dual basis set of the LAPW+lo framework is demonstrated in Figure 1, with an diagram containing two different elements, a and b, in a unit-cell. The dual basis set in the LAPW+lo can be mathematically represented in the MT regions (e.g., $\mathbf{r} \in MT_a$) and the interstitial region I as:

$$\phi_{\mathbf{G}+\mathbf{k}}(\mathbf{r}) = \begin{cases} \sum_\nu A^\alpha_{\mathbf{G}+\mathbf{k},\nu} u_\nu(r_a) Y_{lm}(\mathbf{r_a}) & \mathbf{r} \in \mathrm{MT}_a \\ \frac{1}{\sqrt{\Omega}} e^{i(\mathbf{G}+\mathbf{k})r} & \mathbf{r} \in I. \end{cases}$$

The different muffin-tin volumes in each atom can be made to touch but no larger, i.e., they are non-overlapping. The larger the muffin-tin volume, the smaller the interstitial region between atoms. A smaller interstitial region, generally needs less planewaves than a larger intersittial region to achieve the same precision. This trade-off, however, is dependent on the local orbitals used in the calculation.

The project will use local orbital sets for 71 elemental solids that have been converged by hand to a total energy precision of 10E-4 eV/atom in Ref. [2]. This local orbital set, however, is computationally expensive. This is why, the project uses an algorithm to remove local orbitals one by one from the set and measure the effect on the calculation's precision. The end result is therefore varying degrees of calculation precision (and computational time) and local orbital set sizes.

The end goal of the project is to recommend a local orbital set for a given material and user-specified precision, e.g., TiO_2 with a total energy per atom precision of 10 meV/atom. To realize this goal, a statistical learning model, or recommender system, will need to be built that can predict with some likelihood the precision a given local orbital set will offer. In order to train such a recommender system, a high-quality benchmark dataset is needed, which is the goal of this project.

WWW

`https://sol.physik.hu-berlin.de/`

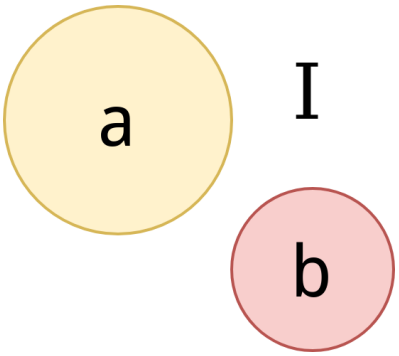

Figure 1: *The LAPW+lo framework can make use of a dual basis. Atomic-like basis functions are active in a spherically symmetric atomic-centered volume called the muffin-tin. The muffin tins are denoted for two different elements, a and b. The interstitial region uses planewaves.*

More Information

[1] Gulans, Andris, Anton Kozhevnikov, and Claudia Draxl. "Microhartree precision in density functional theory calculations." *Phys. Review B* bf97, (2018). doi:10.1103/PhysRevB.97.161105

[2] Lejaeghere, Kurt, Gustav Bihlmayer, Torbjörn Björkman, Peter Blaha, Stefan Blügel, Volker Blum, Damien Caliste et al. *Science* **351**, 154104 (2016). doi:10.1126/science.aad3000

[3] Carbogno, C., Thygesen, K.S., Bieniek, B. et al. *npj Comput Mater* **8**, 69 (2022). doi:10.1038/s41524-022-00744-4

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