

LLMs for DFT simulation script

Inmaculada Yong Moncusí Laso^{1,2}

Dorye Esteras², Andrei Tomut², Stephan Roche³, Jose H. Garcia²

¹ *Universidad Autonoma de Barcelona, Barcelona, Spain*

² *Catalan Institute of Nanoscience and Nanotechnology, Barcelona, Spain*

³ *Institució Catalana de Recerca i Estudis Avançat, Barcelona, Spain*

imoncusi@icn2.cat

Abstract

Recent advances in Large Language Models (LLMs) have enabled code generation and scientific workflow assistance, opening new opportunities in computational materials science [1]. Density Functional Theory (DFT) remains a key method for predicting electronic and magnetic properties, but requires careful expert configuration of parameters, particularly in SIESTA, where convergence and numerical settings strongly affect accuracy and reproducibility [2,3]. In this work, we evaluate the capability of LLMs to generate functional SIESTA scripts across representative material classes. A benchmarking methodology is developed to assess correctness and physical validity, supported by a knowledge graph encoding domain constraints and workflow dependencies. An iterative refinement process with feedback improves agreement with reference approaches. However, variability in parameter selection across iterations remains a key limitation, highlighting the need for more robust and standardised convergence strategies.

References

- [1] Brown, T. et al., Language Models are Few-Shot Learners, NeurIPS (2020).
- [2] Martin, R. M., Electronic Structure: Basic Theory and Practical Methods, Cambridge University Press (2004).
- [3] Soler, J. M. et al., Physical Review B, 61 (2000) 13639.
- [4] Yang, K. et al., Physical Review B, 104 (2021) 224429.
- [5] Shi, H. et al., Physical Review B, 92 (2015) 205118.

Figures

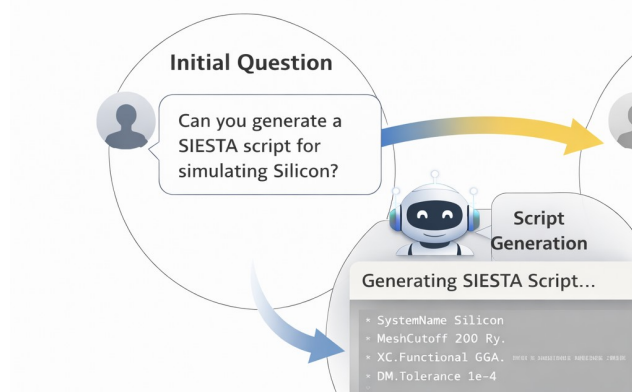


Figure 1: Generation process of SIESTA scripts