

solid_dmft: gray-boxing ab initio DFT+DMFT utilizing TRIQS

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Summary

Strongly correlated systems are a class of materials whose electronic structure is heavily influenced by the effect of electron-electron interactions. In these systems the attempt of an effective single-particle description often fails, and one has to explicitly take into account coulombic and exchange effects. Although density functional theory (DFT) plus dynamical mean-field theory (DMFT) has proven successful in describing strongly correlated electron systems for over two decades, it has only been very recently that ready-to-use software packages began to become available, with most scientific research carried out by self-written codes developed and used in research groups. Given the complexity of the method, there is also the question of whether users should implement the formalism themselves for each problem or whether ready-to-use black-box software, as is the case with many DFT software packages, is beneficial to the community.

The goal of solid_dmft is to find a middle ground, i.e. a *gray-box* tool as a ready-to-use implementation. This means that while the code contains all the functionality needed for many standard DMFT calculations, it is highly modular, based on open source and community-developed software, and therefore can be easily adapted to specific applications and needs. Hence, this project is targeted towards researchers aiming to apply DMFT methods on top of their DFT simulations to describe the physics of strongly correlated electron systems. While our approach allows one to fully perform these computations using standardized input flags to control the calculation without further need for elaborate coding, the final user can easily extend the implemented behavior by modifying the corresponding modules in the code.

The package is MPI-parallelized and written in Python 3, utilizing the publicly available TRIQS software library ([Parcollet et al., 2015](#)), and its available applications. The philosophy of the package is to increase reproducibility of DFT+DMFT calculations, provide clearer convergence metrics, and allow to run calculations for a large variety of systems without adapting the code manually, i.e. on this level similar to widely available DFT simulation packages.

Design Principles

The idea is to provide the full functionality of a full a DFT+DMFT calculation by merging the state-of-the-art implementations provided by the TRIQS library and its applications. This allows to easily run ab initio calculations for strongly correlated materials, as well as implement and test new features of TRIQS or benchmark new solvers against existing ones. solid_dmft manages the calls of the necessary routines to run the DFT calculations, to create the downfolded Hamiltonian, solve the resulting Hubbard-like Hamiltonian via DMFT, postprocess the data to calculate physically meaningful observables, and allow for charge-corrected feedback via charge self-consistency. The full DFT+DMFT cycle is presented in [Figure 1](#).

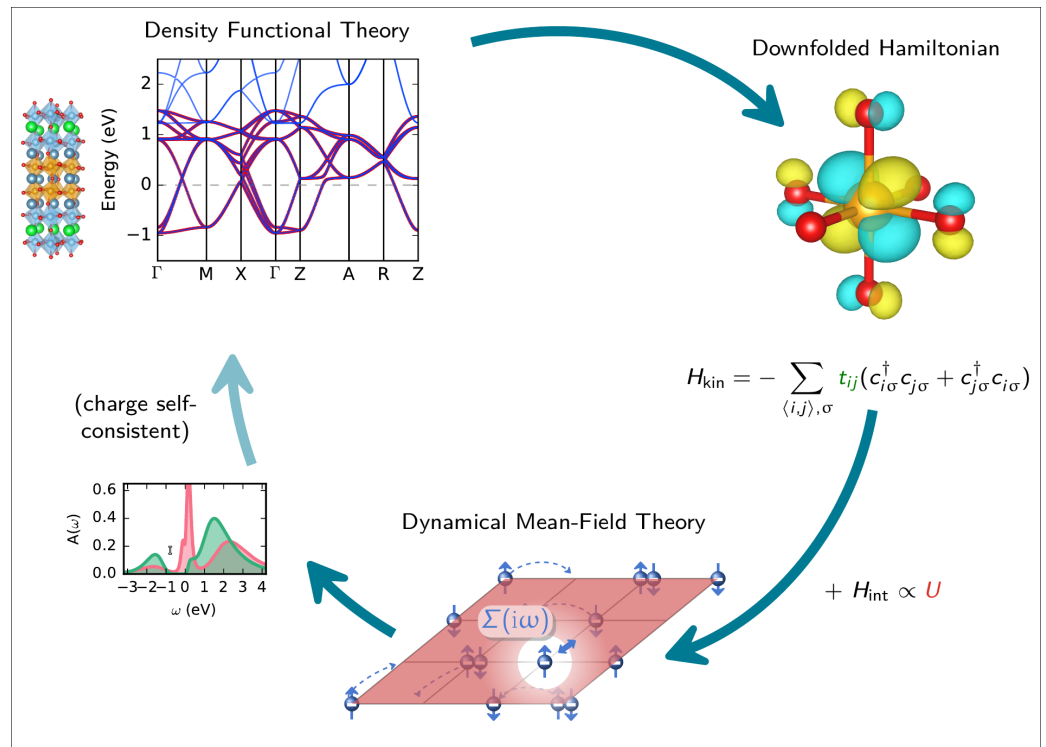


Figure 1: Full CSC DFT+DMFT cycle. Starting from a DFT calculation (top left), a downfolded Hamiltonian and projector functions are created via optimized projections on a local basis set (top right). By adding a specified interaction Hamiltonian H_{int} , a full interacting electron problem is created, to be solved via the DMFT equations (bottom) in TRIQS. After convergence in DMFT is reached, physical observables are calculated (bottom left). For fully CSC calculations a charge density correction is added to DFT and the cycle is restarted.

43 The code is designed to run on top of a DFT calculation or model system providing a low-energy
44 (downfolded) description of the periodic solid system. The DFT calculations can be performed
45 with any code that is compatible with TRIQS/DFTTools (Aichhorn et al., 2016). The input for
46 the DMFT calculation can be either provided directly as a Hamiltonian in reciprocal \mathbf{k} -space
47 in a localized basis set, or in terms of the overlap between the localized basis set and the
48 Kohn-Sham wavefunctions (so-called projector functions), and their respective eigenvalues.
49 The DFT output is converted by TRIQS/DFTTools into an HDF archive in a standardized
50 structure to be read or called by solid_dmft.

51 The code is designed to be modular in the same philosophy as the TRIQS software package,
52 relying on TRIQS functionalities to perform basic operations. Therefore, we split each part of
53 the simulation into separate stand-alone functions, to limit statefulness to a minimum and allow
54 for an easy extension to include new features. The modularity of the program also allows to
55 run, for example, the DMFT loop only via a call of a single function with well-defined input and
56 output, i.e. without running solid_dmft as a monolithic code. This ensures that the code can
57 be used in other projects. An abstracted solver class implements the various impurity solvers
58 available in TRIQS. Even though different solvers operate differently, solid_dmft allows to
59 seamlessly switch impurity solvers, with a simple input flag and adjusting the solver parameters.
60 A fully charge self-consistent (CSC) interface is implemented for Quantum ESPRESSO and the
61 Vienna ab-initio simulation package (VASP). solid_dmft allows also to perform inhomogenous
62 DMFT calculations, i.e. the treatment of multiple correlated and uncorrelated shells (impurity
63 problems) while converging the full lattice self-energy. After self-consistency is reached, either
64 via full CSC or just within the DMFT cycle, postprocessing scripts are available to perform
65 analytic continuation of imaginary Green's functions, and to calculate spectral functions.

As of now, `solid_dmft` has been successfully used in various peer-reviewed research studies (Beck & Georges, 2022; Hampel et al., 2019, 2020, 2021; Merkel & Ederer, 2021; Zhang et al., 2022), and provides stable releases matching the releases of the TRIQS library. We provide a full documentation including several tutorials and a reference manual. Examples and benchmark calculations can be found in the tutorials section of the documentation. Furthermore, we utilize an extensive CI workflow on GitHub to test every pull request and commit.

Statement of need

As mentioned, the number of ready-to-use DFT+DMFT codes is small, and all codes have been developed rather recently. Crucially, most of these codes adopt a black-box approach, where the complexity of the DMFT part is abstracted away from the final user (as in EDMFT (Haule et al., 2010), Amulet (Poteryaev & others, n.d.) or the DMFT implementation in Abinit (Romero et al., 2020)) and while such a strategy can reduce the numbers of free parameters to tune, it also inevitably hinders the flexibility of the approach. Other software packages like DFTwDMFT (Singh et al., 2021) and DCORE (Shinaoka et al., 2021) follow a very similar strategy as `solid_dmft`. Most notably, `solid_dmft` provides a flagship implementation of the TRIQS functionality to perform DFT+DMFT calculation and is easily extended. The benefits of this approach are twofold: on one hand, developers of TRIQS applications are able to benchmark their applications in a well tested framework. On the other hand, users can benefit from a standardized input-output structure compatible with the rest of the TRIQS ecosystem, allowing for easy reproducibility of results.

Given the complexity of the method, there is also the question of whether users should implement the formalism themselves for each problem or whether ready-to-use black-box software, as is the case with many DFT software packages, is beneficial to the community.

`solid_dmft` is developed in the spirit of a community code and supports external contributions that advance the capabilities of the program.

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References

- Aichhorn, M., Pourovskii, L., Seth, P., Vildosola, V., Zingl, M., Peil, O., Deng, X., Mravlje, J., Kraberger, G. J., Martins, C., Ferrero, M., & Parcollet, O. (2016). TRIQS/DFTTools: A TRIQS application for ab initio calculations of correlated materials. *Computer Physics Communications*, 204, 200–208. <https://doi.org/doi:10.1016/j.cpc.2016.03.014>
- Beck, H., S., & Georges, A. (2022). Charge self-consistent electronic structure calculations with dynamical mean-field theory using quantum ESPRESSO, wannier 90 and TRIQS. *Journal of Physics: Condensed Matter*, 34, 235601. <https://doi.org/10.1088/1361-648x/ac5d1c>
- Hampel, A., Beck, S., & Ederer, C. (2020). Effect of charge self-consistency in DFT + DMFT calculations for complex transition metal oxides. *Phys. Rev. Research*, 2, 033088. <https://doi.org/10.1103/PhysRevResearch.2.033088>
- Hampel, A., Lee-Hand, J., Georges, A., & Dreyer, C. E. (2021). Correlation-induced octahedral rotations in SrMoO₃. *Phys. Rev. B*, 104, 035102. <https://doi.org/10.1103/PhysRevB.104.035102>

- 109 Hampel, A., Liu, P., Franchini, C., & Ederer, C. (2019). Energetics of the coupled elec-
110 tronic-structural transition in the rare-earth nickelates. *Npj Quantum Materials*, 4, 5.
111 <https://doi.org/10.1038/s41535-019-0145-4>
- 112 Haule, K., Yee, C.-H., & Kim, K. (2010). Dynamical mean-field theory within the full-potential
113 methods: Electronic structure of CeIrIn₅, CeCoIn₅, and CeRhIn₅. *Phys. Rev. B*, 81,
114 195107. <https://doi.org/10.1103/PhysRevB.81.195107>
- 115 Merkel, M. E., & Ederer, C. (2021). Charge disproportionation and Hund's insulating behavior
116 in a five-orbital Hubbard model applicable to d^4 perovskites. *Physical Review B*, 104,
117 165135. <https://doi.org/10.1103/PhysRevB.104.165135>
- 118 Parcollet, O., Ferrero, M., Ayrat, T., Hafermann, H., Krivenko, I., Messio, L., & Seth, P.
119 (2015). TRIQS: A toolbox for research on interacting quantum systems. *Computer Physics*
120 *Communications*, 196(Supplement C), 398–415. [https://doi.org/https://doi.org/10.1016/](https://doi.org/10.1016/j.cpc.2015.04.023)
121 [j.cpc.2015.04.023](https://doi.org/10.1016/j.cpc.2015.04.023)
- 122 Poteryaev, A., & others. (n.d.). <http://amulet-code.org>.
- 123 Romero, A. H., Allan, D. C., Amadon, B., Antonius, G., Applencourt, T., Baguet, L., Bieder,
124 J., Bottin, F., Bouchet, J., Bousquet, E., Bruneval, F., Brunin, G., Caliste, D., Côté,
125 M., Denier, J., Dreyer, C., Ghosez, P., Giantomassi, M., Gillet, Y., ... Gonze, X. (2020).
126 ABINIT: Overview and focus on selected capabilities. *The Journal of Chemical Physics*,
127 152(12), 124102. <https://doi.org/10.1063/1.5144261>
- 128 Shinaoka, H., Otsuki, J., Kawamura, M., Takemori, N., & Yoshimi, K. (2021). DCore:
129 Integrated DMFT software for correlated electrons. *SciPost Phys.*, 10, 117. [https://doi.](https://doi.org/10.21468/SciPostPhys.10.5.117)
130 [org/10.21468/SciPostPhys.10.5.117](https://doi.org/10.21468/SciPostPhys.10.5.117)
- 131 Singh, V., Herath, U., Wah, B., Liao, X., Romero, A. H., & Park, H. (2021). DMFTwDFT: An
132 open-source code combining dynamical mean field theory with various density functional
133 theory packages. *Computer Physics Communications*, 261, 107778. [https://doi.org/https:](https://doi.org/10.1016/j.cpc.2020.107778)
134 [//doi.org/10.1016/j.cpc.2020.107778](https://doi.org/10.1016/j.cpc.2020.107778)
- 135 Zhang, R., Merkel, M. E., Beck, S., & Ederer, C. (2022). *Training biases in machine*
136 *learning for the analytic continuation of quantum many-body Green's functions*. arXiv.
137 <https://doi.org/10.48550/arXiv.2206.07493>