1 Supplementary

Code and data to replicate our experiments can be found at https://github.com/ppope/rho-learn.

1.1 DFT Relaxations

We relax structures using Quantum Espresso (`v6.7`) and the AiiDA relaxation workflow [3]. The `fast` relaxation protocol without magnetization. For complete documentation of relaxation parameters see the Supplementary of [3]. Note for pseudo-potentials this protocol uses the `efficiency` set of the SSSP library `1.2.1` [4]. We use the PBE exchange-correlation functional for all relaxations.

1.2 Hyperparameters for model training

We document all hyperparameters used for training the SCN models in the accompanying file `hyperparams.yml`. This set was modified from the offical SCN implementation in the OCP repo [1]. In particular a much smaller model was used than the state-of-the-art SCN results.

1.3 Initializing SCF runs in Quantum Espresso with learned densities

An SCF run may be initialized with a custom density, e.g. one generated from a machine-learning model, using the `startingpot` input parameter of Quantum Espresso (QE) [2]. Importantly, we use QE compiled with `HDF5` support, rather than machine-dependent `dat` binary files. To initialize an SCF run with a learned density, we follow the file format of the `HDF5` charge density files expected by QE. This format includes coefficients of the *reciprocal* charge densities and their associated Miller indices. Only data with associated reciprocal space vector G with $|G|^2 \leq$ `ecutrho` is written, where `ecutrho` is the energy cutoff for charge density in suitable units. We validate our charge-density read/write implementation is correct by checking that convergence of ground-truth densities are unaffected by reading/writing.

References

[1] Open Catalyst Project - `ocp` repository. URL `https://github.com/Open-Catalyst-Project/ocp`.

[2] Quantum Espresso - pw.x input description: startingpot. URL `https://www.quantum-espresso.org/Doc/INPUT_PW.html#idm899`.

[3] Sebastiaan P Huber, Emanuele Bosoni, Marnik Bercx, Jens Bröder, Augustin Degomme, Vladimir Dikan, Kristjan Eimre, Espen Flage-Larsen, Alberto Garcia, Luigi Genovese, et al. Common workflows for computing material properties using different quantum engines. *npj Computational Materials*, 7(1):136, 2021.

[4] Gianluca Prandini, Antimo Marrazzo, Ivano E Castelli, Nicolas Mounet, and Nicola Marzari. Precision and efficiency in solid-state pseudopotential calculations. *npj Computational Materials*, 4(1):72, 2018.