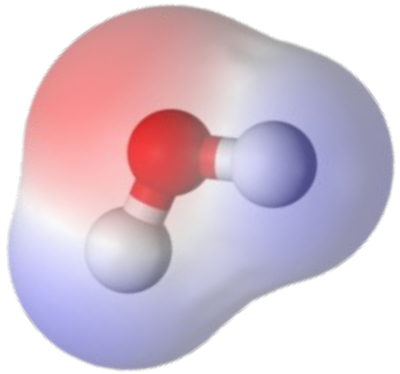
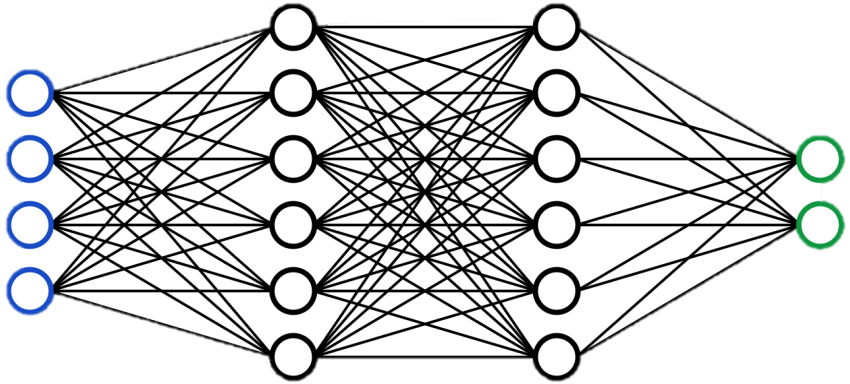
Tutorial Session: Deep Modeling with Deep Dipole



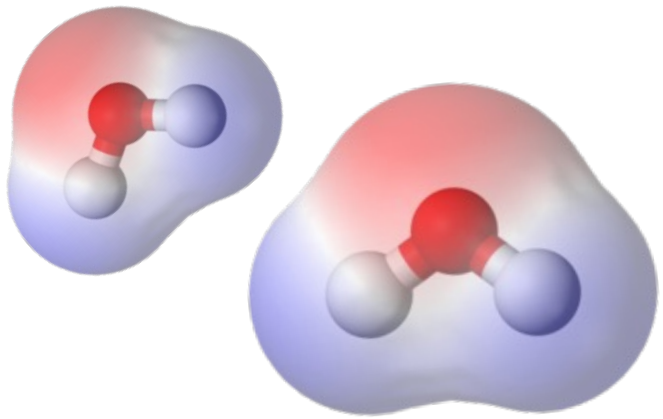


Instructor: Pinchen Xie

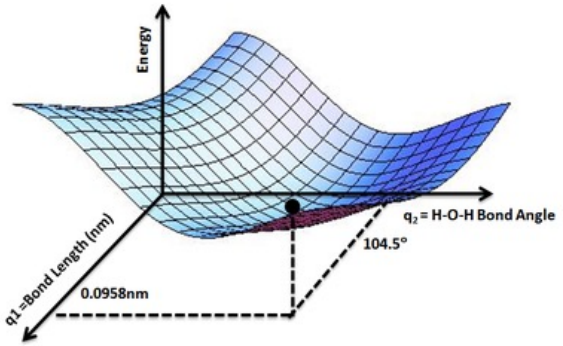
June, 2024

Slides at: github.com/CSIprinceton/workshop-june-2024/tree/main/hands-on-sessions/day-2-DW-DPLR/7-deep-wannier

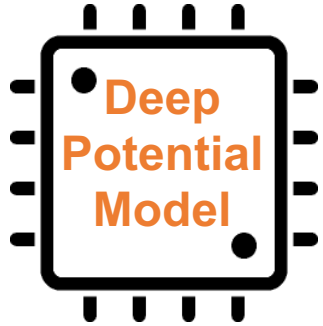
What does Deep Dipole do?

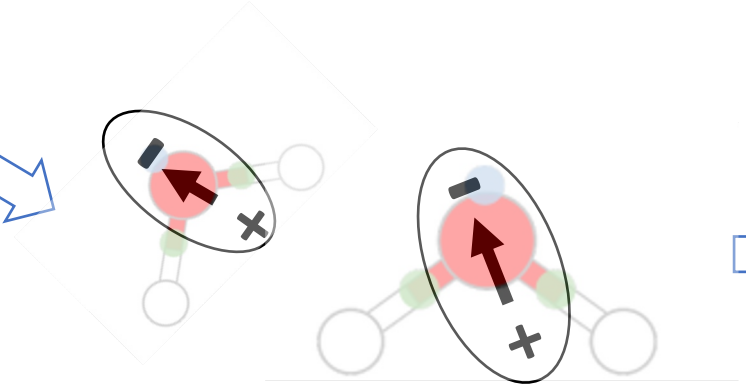


Electronic Structures

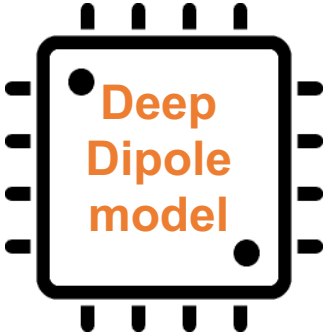


Potential Energy Surface





Atomic Dipole

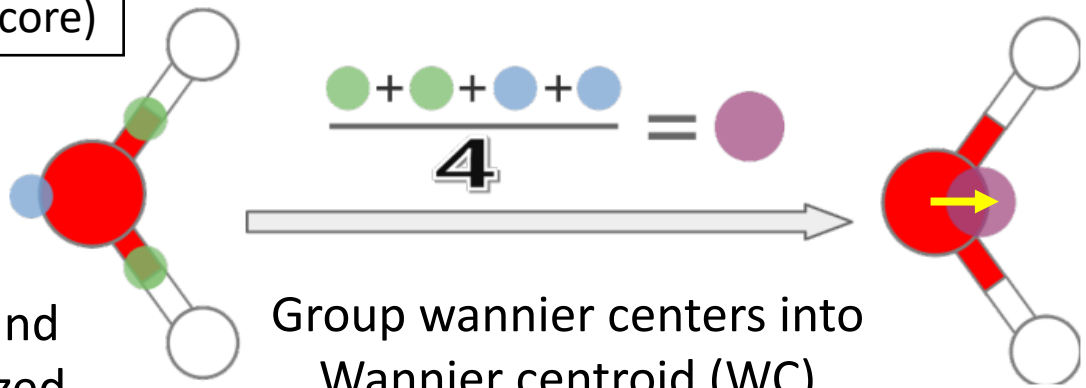


Largely depends on ionic positions and
electronic orbitals in <u>local</u> chemical environments

How to define an atomic dipole?

Example: the water molecule

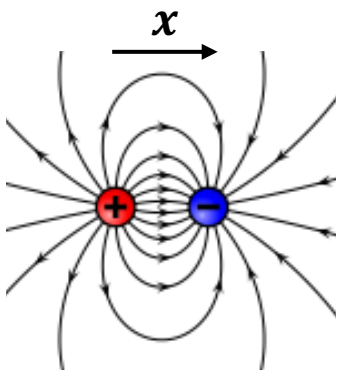
Valence Electron: 8
Wannier centers: 4
Hydrogen ion (+1e)
Oxygen ion (+6e, with frozen core)



Atomic coordinates $\{\boldsymbol{r}_i\}$ and assigned maximally localized wannier centers $\{\boldsymbol{W}_j\}$

Implemented in Wannier90

Group wannier centers into Wannier centroid (WC)



Classical picture

The atomic dipole: $q\boldsymbol{x}$

8e

Marzari, Nicola, et al. "Maximally localized Wannier functions: Theory and applications." Reviews of Modern Physics 84.4 (2012): 1419.
Zhang, Linfeng, et al. "A deep potential model with long-range electrostatic interactions." *The Journal of Chemical Physics* 156.12 (2022): 124107.

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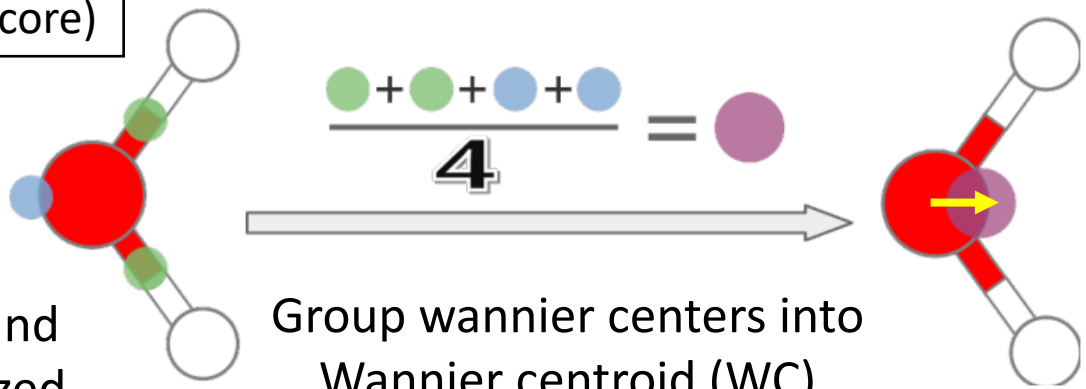

Group wannier centers into Wannier centroid (WC)

Atomic coordinates $\{\boldsymbol{r}_i\}$ and assigned maximally localized wannier centers $\{\boldsymbol{W}_j\}$

Implemented in Wannier90

Training label:
Position vector from WC to central atom (e.g. Oxygen)

You can also let the dipole moment to be the training label!

Marzari, Nicola, et al. "Maximally localized Wannier functions: Theory and applications." Reviews of Modern Physics 84.4 (2012): 1419.

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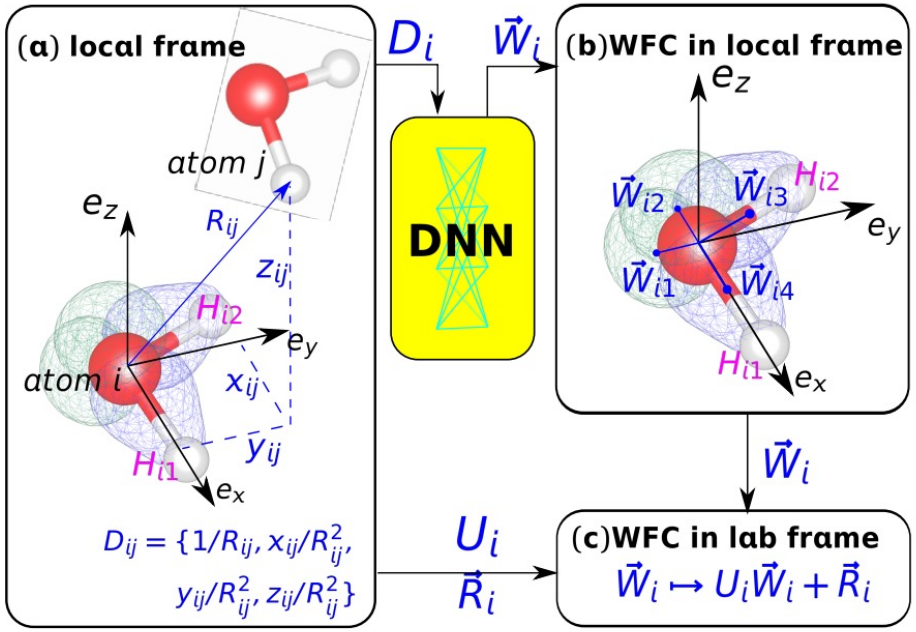
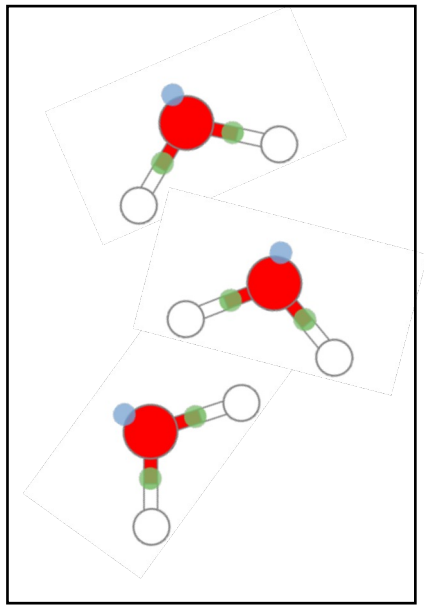
What is a Deep Dipole model?

The open box



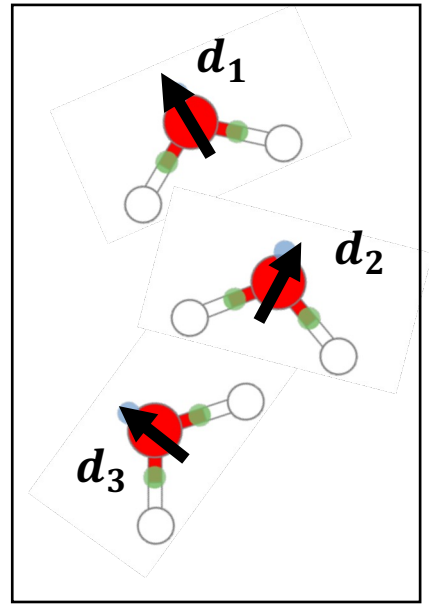
The black box



Atomic coordinates $\{\boldsymbol{r}_i\}$

Equivariant

Deep Dipole



Vectors associated with central atoms

Zhang, Linfeng, et al. "Deep neural network for Wannier function centers." *arXiv preprint arXiv:1906.11434* (2019).

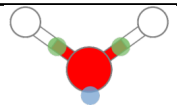
Zhang, Linfeng, et al. "Deep neural network for the dielectric response of insulators." Physical Review B 102.4 (2020): 041121.

hands-on-sessions/day-2-DW-DPLR/7-deep-wannier/1-wannier90-example/ice_mlwf_exercise

Hands-on Exercise 1 -- 30 minutes

See [Marzari, Nicola, et al. "Maximally localized Wannier functions: Theory and applications." Reviews of Modern Physics 84.4 (2012): 1419] for details

Goal: Compute maximally localized Wannier centers of given water configuration with Quantum-Espresso and Wannier90.



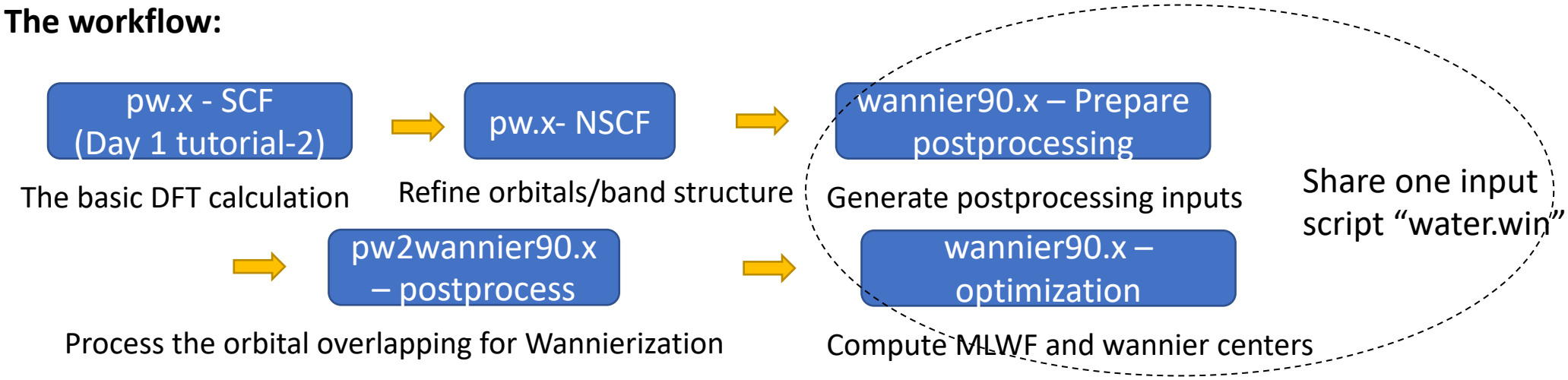
How:

(1) Read and execute “run.sh” (line by line so you can see the succession of outputs) ~ 10mins

(2) Meanwhile, get familiar with the inputs&outputs

(3) Visualize the final output “water_centres.xyz” with OVITO.

The workflow:



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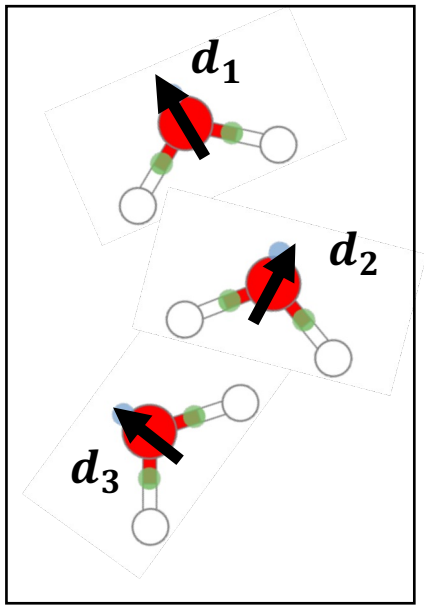
Basic setup of the wannier90 input

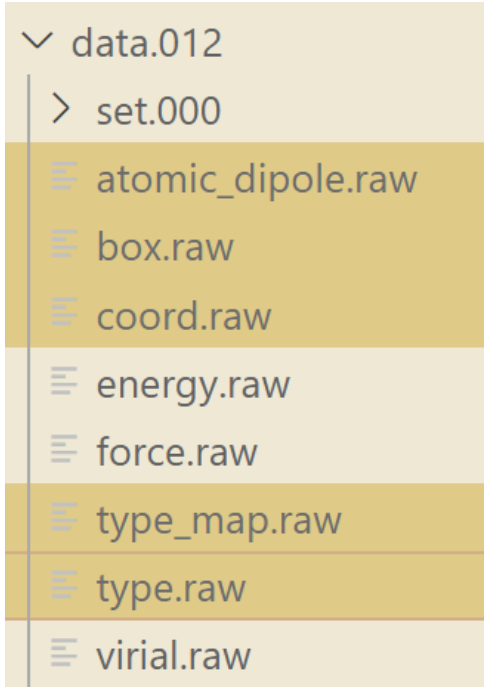
```
num_wann  = 64     ← Number of wannier centers = #Occupied orbitals
num_iter  = 100    ← Iterations for MLWF optimization. 100 is default. If not converging in 100 steps,
probably k-grid is too sparse.
write_xyz        = True

guiding_centres = .true. ← Provide an initial guess for the optimization problem
begin projections ← specify the atomic orbital as initial guess
  O: sp3
end projections
```

https://github.com/wannier-developers/wannier90/raw/v3.1.0/doc/compiled_docs/user_guide.pdf

Training Deep Dipole: Dataset



- Required data



Optional: dipole.raw

- Format

Number of central atoms

atomic_dipole.raw:(#frames, #sel_atom×3)

Concatenated in the same order as the central atoms appear in coord.raw

dipole.raw: (#frames, 3) <- the sum over all central atoms

atomic_dipole.npy&dipole.npy:

Same thing but in numpy format.

Training Deep Dipole: parameters

Difference between training Deep Potential & Deep Dipole

	fitting_net	loss
Energy model	"fitting_net": { "type": "ener", "neuron": [240, 240, 240], },	"loss": { "start_pref_e": 0.01, "limit_pref_e": 1, "start_pref_f": 100, "limit_pref_f": 1, "start_pref_v": 0, "limit_pref_v": 0, },
Dipole model	"fitting_net": { "type": "dipole", "sel_type": [0], "neuron": [100, 100, 100], },	"loss": { "type": "tensor", "pref": 1.0, "pref_atomic": 1.0, },

0.0 if there is no dipole.npy

Type of central atom

Hands-on Exercise 2 -- 10 minutes

Goal: Train a deep dipole with the dataset

Folder: hands-on-sessions/day-2-DW-DPLR/7-deep-wannier/2-train_dipole_model

How:

(1) Check “input.json”

(2) Before training, examine dataset and verify that the number of columns in “atomic_dipole.raw” is compatible with "sel_type” and the number of water molecules.

(3) Run “train.sh”. Check logs.

(4) Terminate the training due to the lack of time. We provide the trained model in

hands-on-sessions/day-2-DW-DPLR/7-deep-wannier/dipole_model

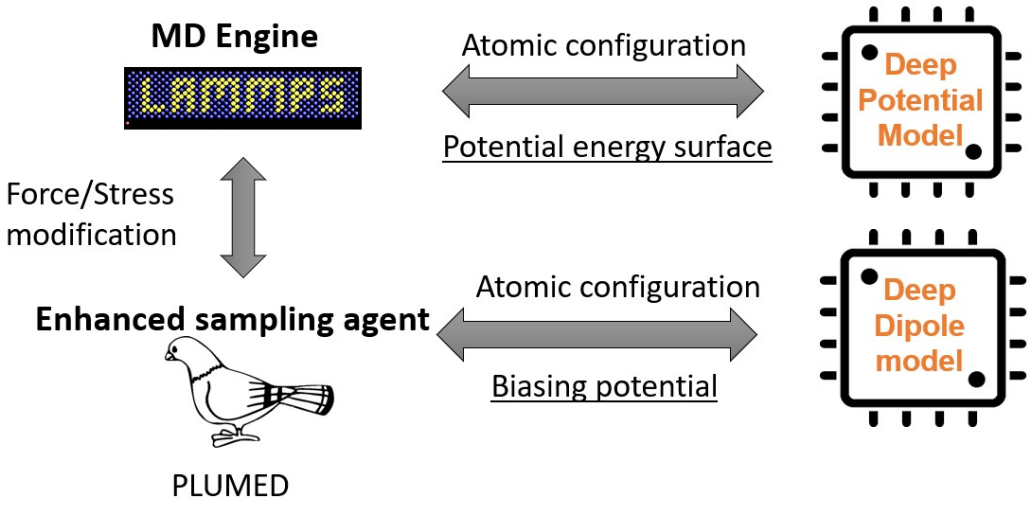
Application of Deep Dipole

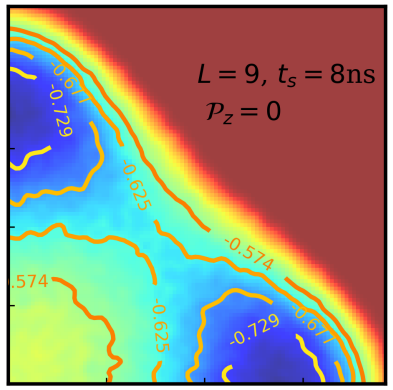
- **Postprocessing (Today)**

  After a regular DPMD simulation. Process the dumped trajectory with deep dipole model. You can compute dipole distribution / polarization/ dielectric response …..

- **Enhanced sampling**

  Pair Lammps up with PLUMED and use deep dipole model as a collective variable.





A cross section of the 3D free energy surface for the polarization of PbTiO3.

Reference:
arXiv preprint arXiv:2205.11839.
PLUMED plugin:
https://github.com/y1xiaoc/deepmd-plumed

Hands-on Exercise 3 -- 20 minutes

Goal:

(1) Run a DPMD for liquid water with provided Deep Potential model. Then analyzed the dumped trajectory with Deep Dipole model.

Folder: hands-on-sessions/day-2-DW-DPLR/7-deep-wannier/3-MD_exercise

How:

(1) Follow “run.sh”

(2) Compute and plot the distribution of model prediction. Example script:

hands-on-sessions/day-2-DW-DPLR/7-deep-wannier/dipole_distribution.py

(3) Experiment with “dipole_model.eval”!