Tutorial Session: Deep Modeling with Deep Dipole

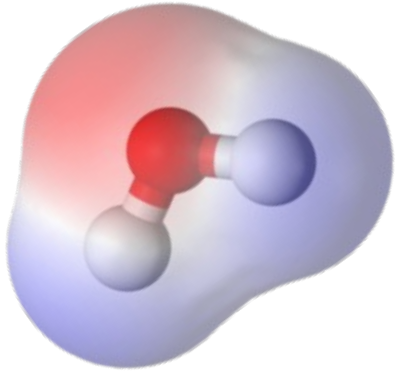
Preparation

- Clone or pull the latest Git Repo

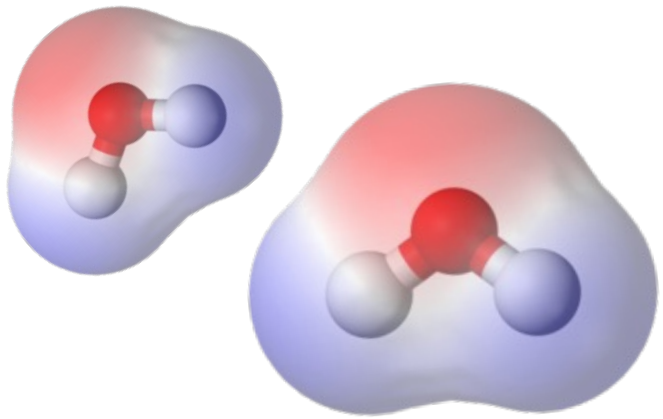
git clone https://github.com/CSIprinceton/workshop-june-2024.git

- Slides at:

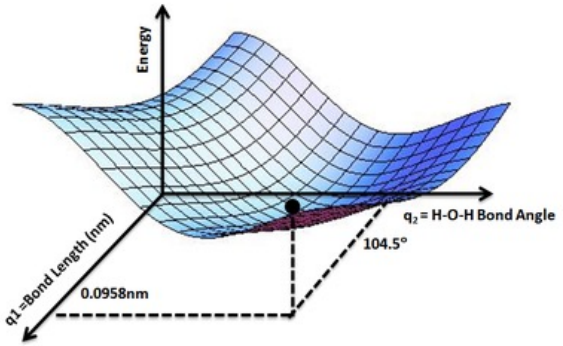
github.com/CSIprinceton/workshop-june-2024/tree/main/hands-on-sessions/day-2-DW-DPLR/7-deep-wannier



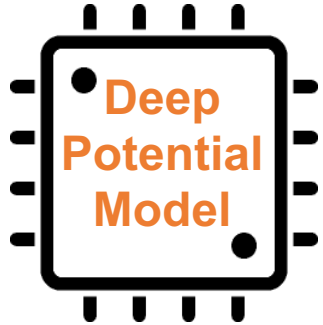
What does Deep Dipole do?

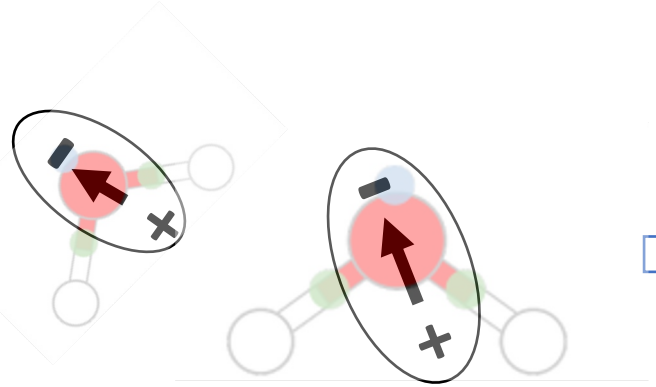


Electronic Structures

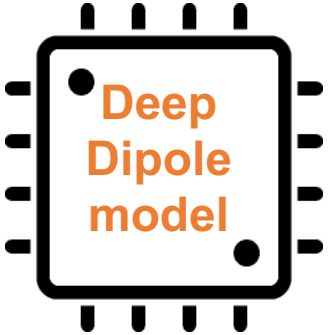


Potential Energy Surface





Atomic Dipole

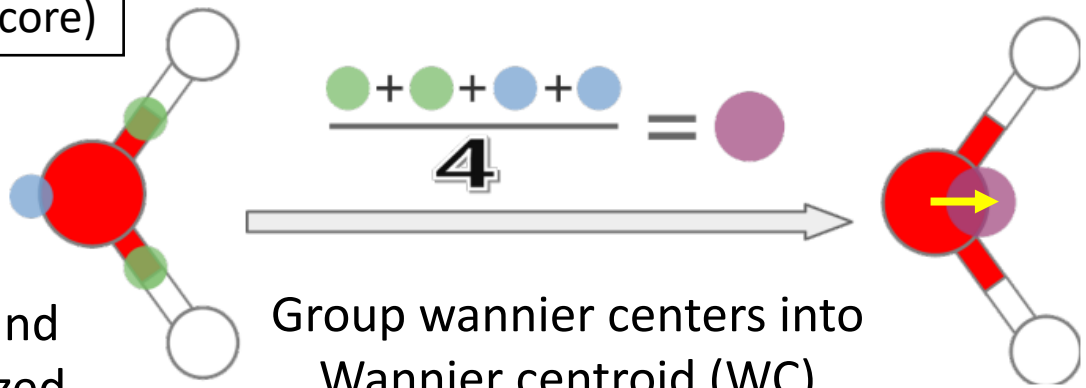


Largely depends on ionic positions and
electronic orbitals in <u>local</u> chemical environments

How to define an atomic dipole?

Example: the water molecule

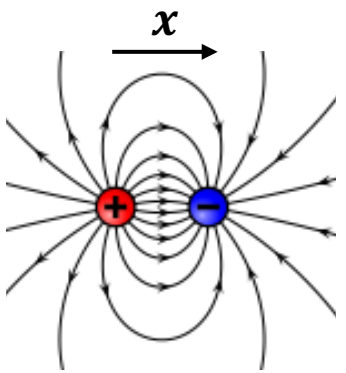
Valence Electron: 8
Wannier centers: 4
Hydrogen ion (+1e)
Oxygen ion (+6e, with frozen core)



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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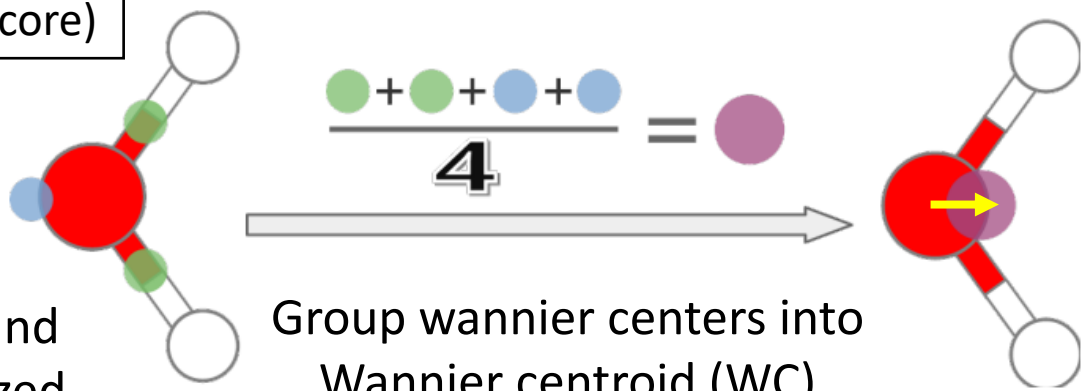


Group wannier centers into Wannier centroid (WC)

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

Implemented in Wannier90

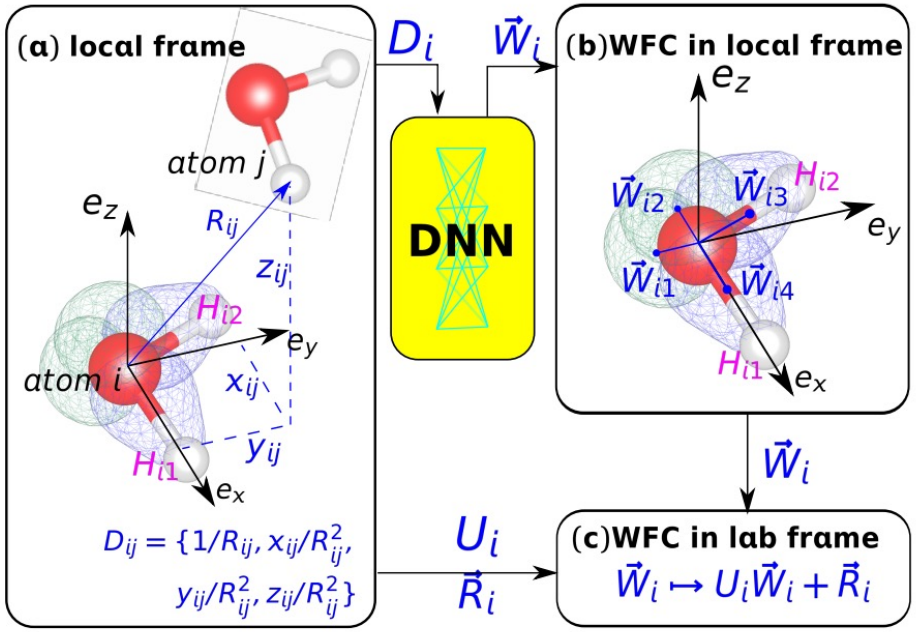
Training label:
Position vector from central atom to WC
(e.g. Oxygen)

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

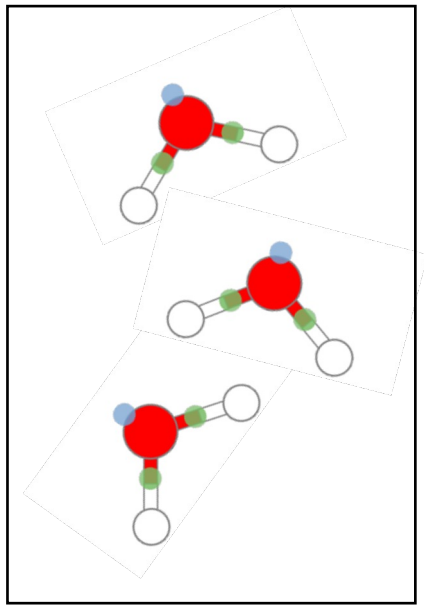
Marzari, Nicola, et al. "Maximally localized Wannier functions: Theory and applications." Reviews of Modern Physics 84.4 (2012): 1419.
Zhang, Linfeng, et al. "A deep potential model with long-range electrostatic interactions." *The Journal of Chemical Physics* 156.12 (2022): 124107.

What is Deep Dipole model?

The open box

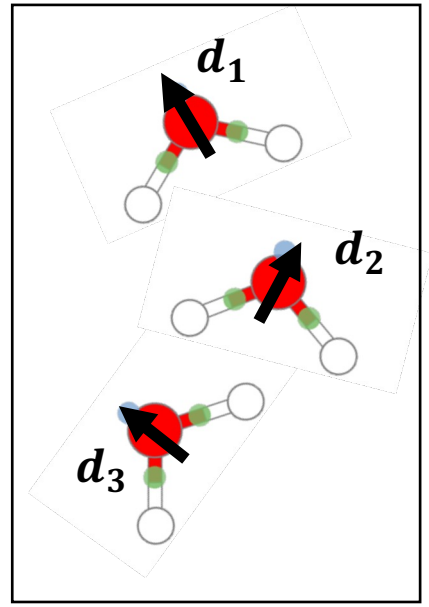


The black box



Equivariant

Deep Dipole



Atomic coordinates $\{r_i\}$

Vectors associated with central atoms

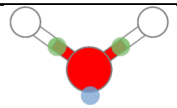
Zhang, Linfeng, et al. "Deep neural network for Wannier function centers." *arXiv preprint arXiv:1906.11434* (2019).
Zhang, Linfeng, et al. "Deep neural network for the dielectric response of insulators." Physical Review B 102.4 (2020): 041121.

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:

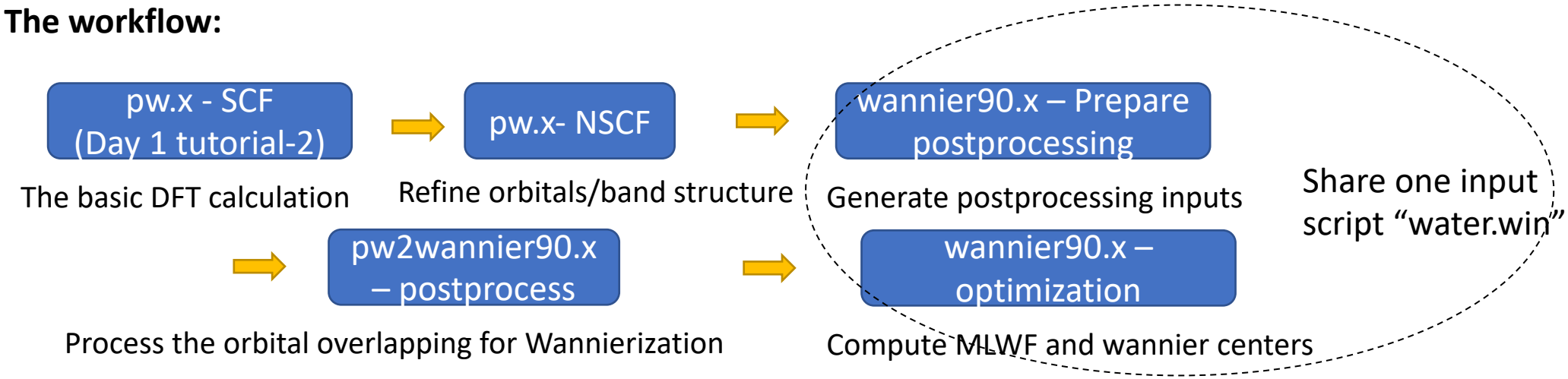
("sbatch run_della.sh" if you are using della)

(1) Read and execute "run_azure.sh" (line by line so you can see the succession of outputs) ~ 15mins

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

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

The workflow:



Hands-on Exercise 1 -- 30 minutes

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

How:

(“sbatch run_della.sh” if you are using della)

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

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

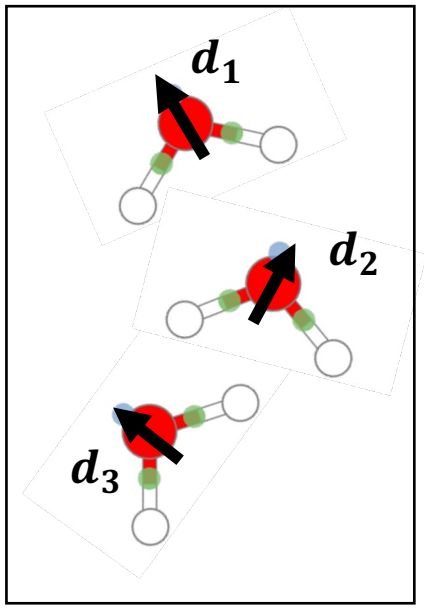
(3) Visualize the final output “water_centres.xyz” with OVITO.

Basic setup of the wannier90 input

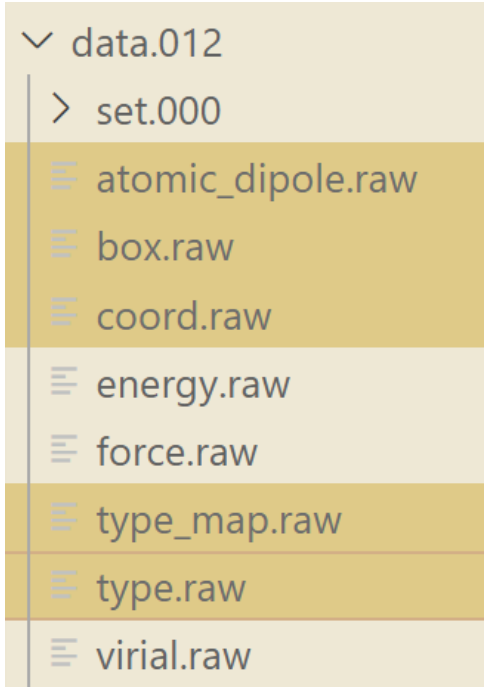
```
num_wann  = 64     ← Number of wannier centers = #Occupied orbitals
num_iter  = 100    ← Iterations for MLWF optimization. 100 is default. If not converging in 100 steps,
probably k-grid is too sparse.
write_xyz        = True

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

Number of central atoms

atomic_dipole.raw: The vector associated to central atom: (#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. (“sbatch train.sh” if you are using della)

(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

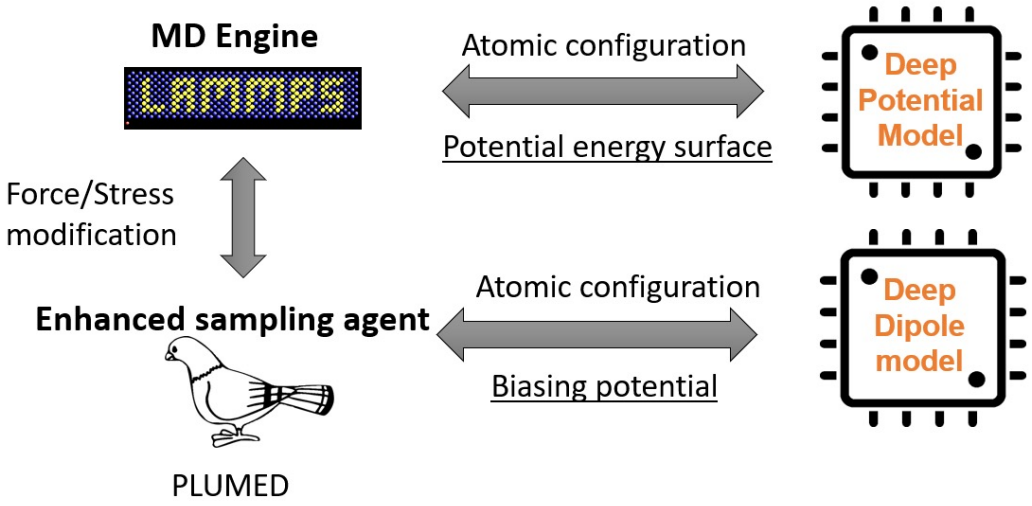
Application of Deep Dipole

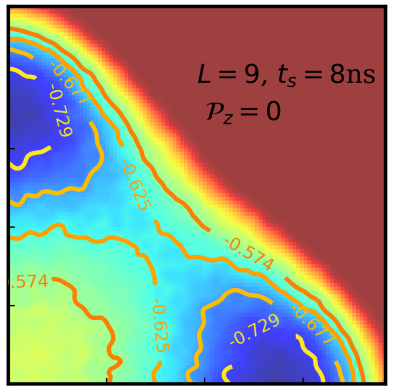
- **Postprocessing (Today)**

  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: 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 "3-MD_exercise/run.sh" ("sbatch run.sh" if you are using della)

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

7-deep-wannier/wannier_distribution.py

(2, Virtual Machine) Check the manuscript 7-deep-wannier/wannier_distribution.py

(3) See the results at 7-deep-wannier/Wannier_Centroid_distribution.py