

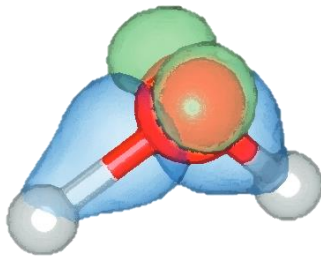
Tutorial Session: Deep Dipole

SPEAKER: PINCHEN XIE

PACM, PRINCETON

JUNE 8, 2022

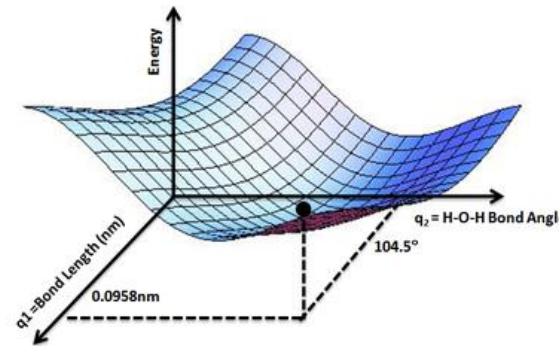
Beyond potential energy surface



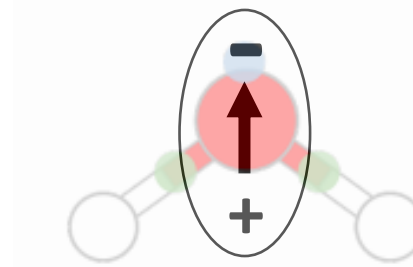
First-Principles theory (e.g., DFT/MBPT/...)

Atom

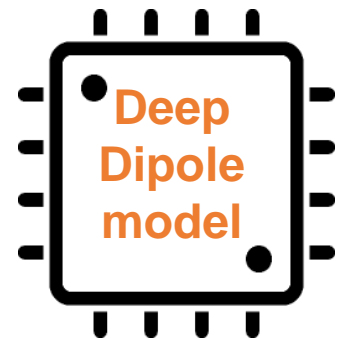
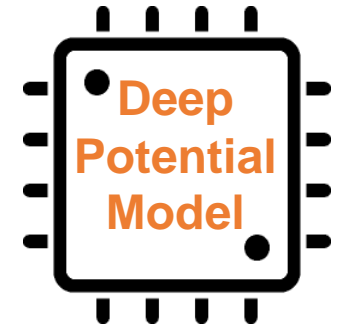
Electron



Potential Energy Surface

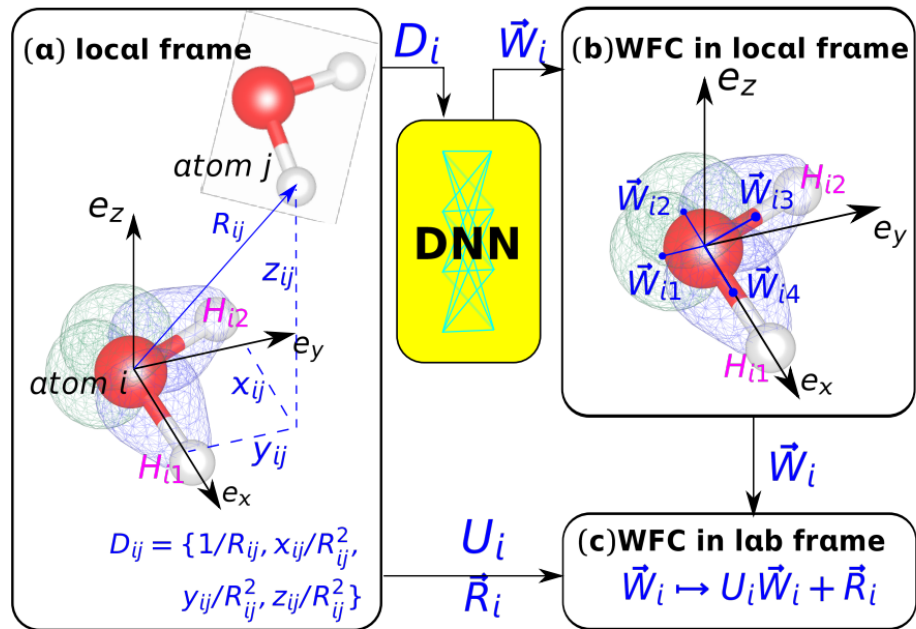


Atomic/Molecular Dipole

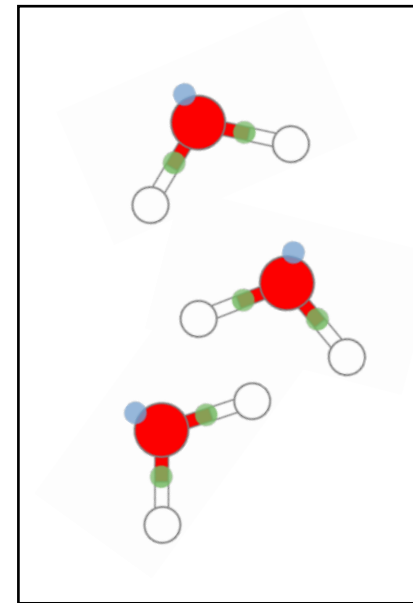


A very short introduction to Deep Dipole

The open box

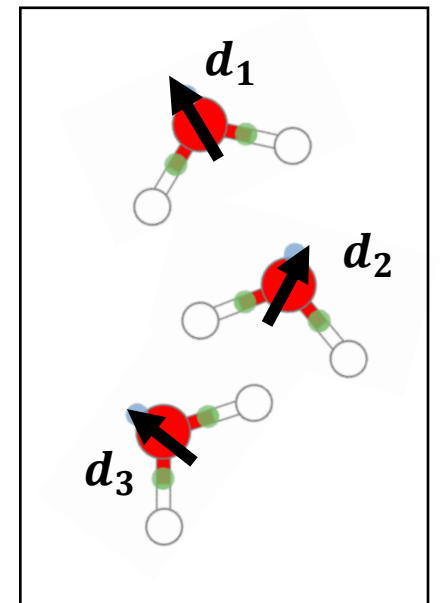


The black box



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

Deep Dipole



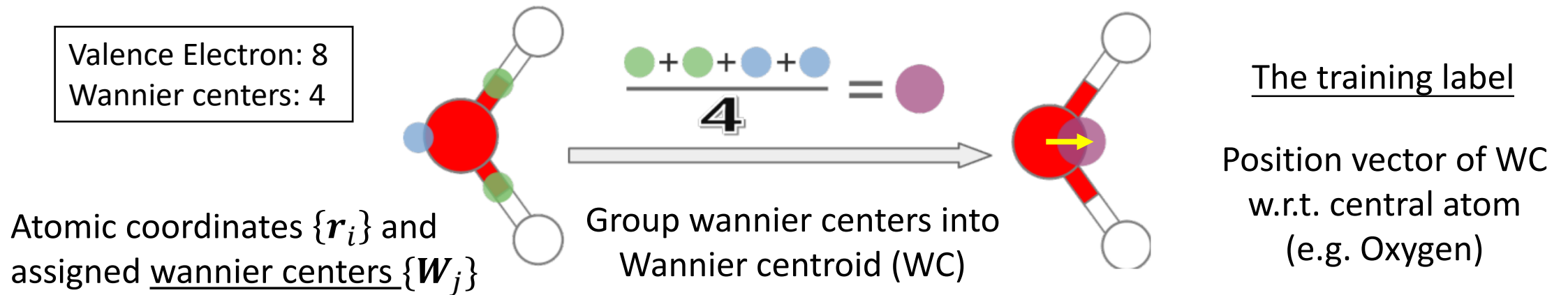
The dipole moment $\{\mathbf{d}_j\}$

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.

A very short introduction to Deep Dipole

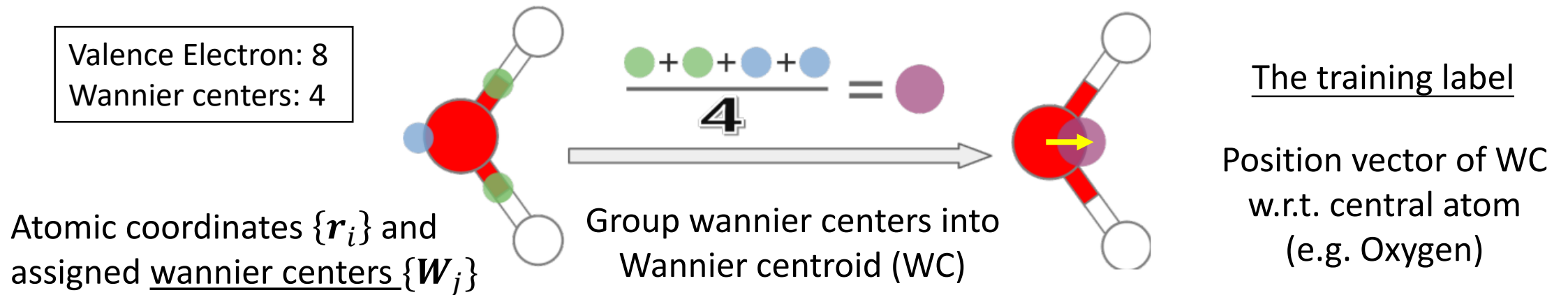
Example: the water molecule



A very short introduction to Deep Dipole

Example: the water molecule

Issue: wannier centers not unique (U(1) Gauge)



A very short introduction to Deep Dipole

Implemented in Wannier90

Maximally localized Wannier functions (MLWF)

The General wannier function:

$$|\mathbf{R}n\rangle = \frac{V}{(2\pi)^3} \int_{\text{BZ}} d\mathbf{k} e^{-i\mathbf{k}\cdot\mathbf{R}} \sum_{m=1}^J U_{mn}^{(\mathbf{k})} |\psi_{m\mathbf{k}}\rangle$$

Bloch orbitals

Arbitrary Unitary Rotation

The MLWF:

$$|\mathbf{R}n\rangle = \frac{V}{(2\pi)^3} \int_{\text{BZ}} d\mathbf{k} e^{-i\mathbf{k}\cdot\mathbf{R}} \sum_{m=1}^J U_{mn}^{(\mathbf{k})} |\psi_{m\mathbf{k}}\rangle$$

The optimal rotation minimizing the total spreading of wannier functions

Total spreading

$$\Omega = \sum_n [\langle \mathbf{0}n | r^2 | \mathbf{0}n \rangle - \langle \mathbf{0}n | \mathbf{r} | \mathbf{0}n \rangle^2]$$

$$= \sum_n [\langle r^2 \rangle_n - \bar{\mathbf{r}}_n^2]$$

wannier centers \mathbf{W}_n

Hands-on Exercise 1 -- 30 minutes

Goal:

- (1) Compute maximally localized wannier centers of given water configuration with Quantum-Espresso and Wannier90

Folder: hands-on-sessions/day-2/4-deep-wannier/wannier90 example/ice mlwf exercise

How:

- (1) Read and execute “run.sh” (better 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

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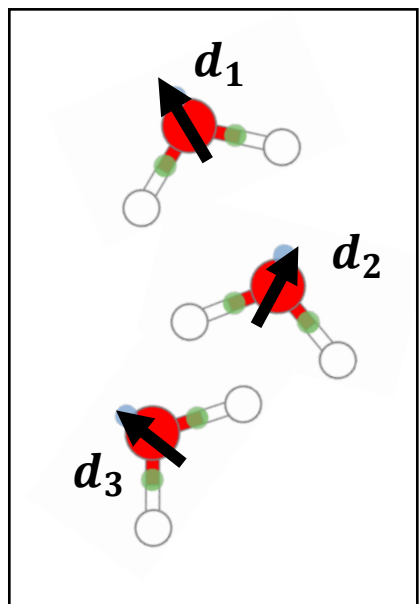
Basic setup of the wannier90 input

```
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
  0: sp3
end projections
```


Training Deep Dipole: Dataset

- Required data



```
data.012
└─> set.000
    atomic_dipole.raw
    box.raw
    coord.raw
    energy.raw
    force.raw
    type_map.raw
    type.raw
    virial.raw
```

Optional: dipole.raw

- Format

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

Reference atoms for dipole moments,
e.g. oxygen for H2O

Vector concatenated in the
same order as its reference atom
appearing in coord.raw

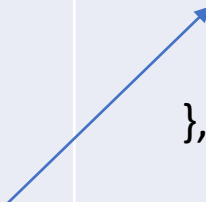

dipole.raw: (#frames, 3) <- the sum of atomic
dipole

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	<pre>"fitting_net": { "type": "ener", "neuron": [240, 240, 240], }</pre>	<pre>"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, }</pre>
Dipole model	<pre>"fitting_net": { "type": "dipole", "sel_type": [0], "neuron": [100, 100, 100], }</pre> 	<pre>"loss": { "type": "tensor", "pref": 1.0,  0.0 if there is no dipole.npy "pref_atomic": 1.0, }</pre>

Types of atom that carries a dipole.

Hands-on Exercise 2 -- 10 minutes

Goal:

(1) Train a deep dipole with the dataset hands-on-sessions/day-2/4-deep-wannier/data

Folder: hands-on-sessions/day-2/4-deep-wannier/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-2/4-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 / Electric field-driven MD

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

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-2/4-deep-wannier/MD exercise

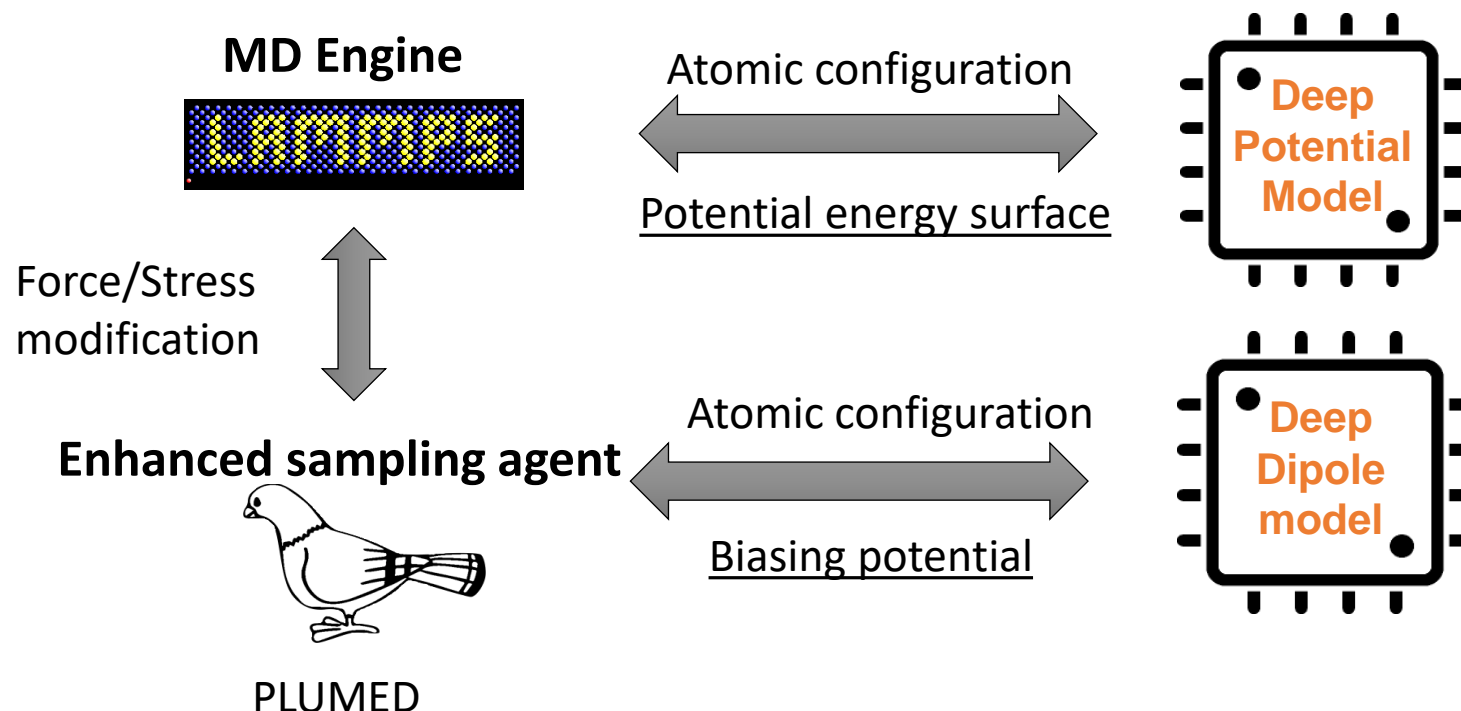
How:

- (1) Follow “run.sh”
- (2) Compute and plot the molecule dipole moments distribution with Python. Example script:

hands-on-sessions/day-2/4-deep-wannier/dipole distribution.py

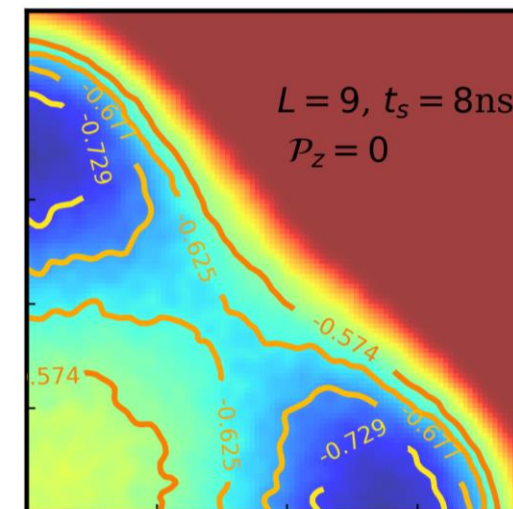
- (3) Experiment with “`dipole_model.eval`”!

Enhanced sampling



Global polarization from Deep Dipole

$$\mathcal{G}(T = 600K, \mathcal{P})$$



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

Reference: Xie, P., Chen, Y. E, W. and Car, R., 2022. Ab initio multi-scale modeling of ferroelectrics: The case of PbTiO₃. arXiv preprint arXiv:2205.11839.

PLUMED plugin: <https://github.com/y1xiaoc/deepmd-plumed> (not MPI-ready yet)