

QHFlow

: Accelerating DFT with Equivariant Flow Matching



Seongsu Kim¹, Nayoung Kim¹, Dongwoo Kim², and Sungsoo Ahn¹

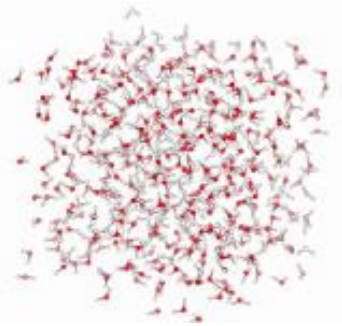
¹KAIST, ²POSTECH

Sep 2025 (Presented at NVIDIA BioNeMo Reading group)

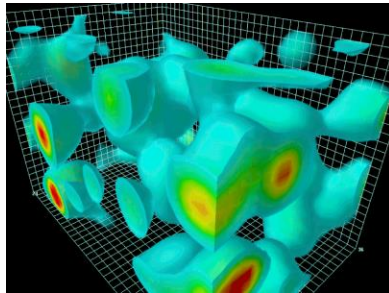
NeurIPS 2025 spotlight

Atomistic interaction modeling for materials

Atomistic Interaction Modeling



Molecular Dynamics



Wavefunction

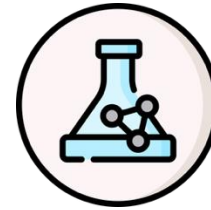
Example

- Classical Force Field
- Machine Learning Force Field (MLFF)
- Machine Learning Interatomic Potential (MLIP)
- Density Functional Theory (DFT)
- Wavefunction based method (QVMC, CCSD)

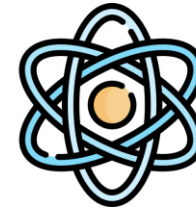
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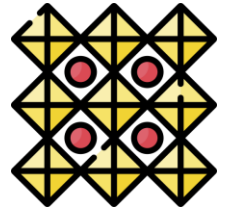
Material Understanding



Chemical Reaction

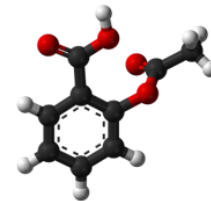


Quantum Effects



Solid State Effects

Material Discovery



Molecule



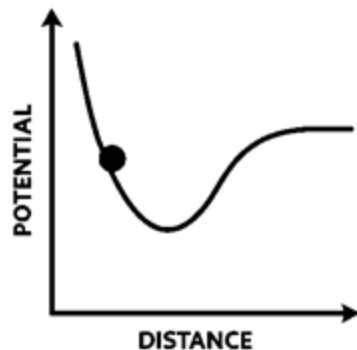
Drug



Battery cathode

Our interests: DFT

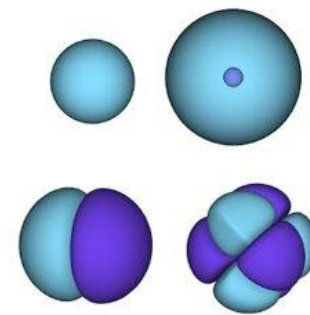
Classical Efficiency



 **Balanced**



Quantum Accuracy



Level of Theory

(classical) Interatomic potential

Electron density

Wavefunction

Speed

Fastest

Speed

Accuracy

Slowest

Accuracy

Low

Fidelity

Poor quantum interaction

High

Accurate quantum interaction

Examples

Classical Force Field

Density Functional Theory

QVMC, CCSD(S)

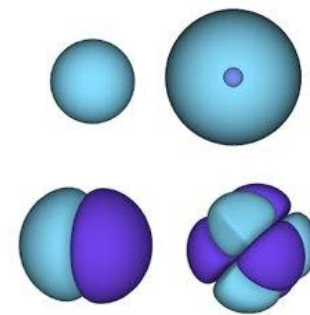
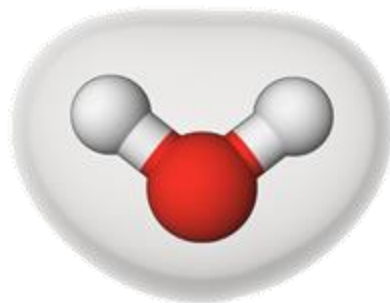
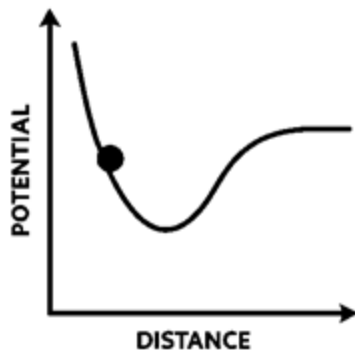
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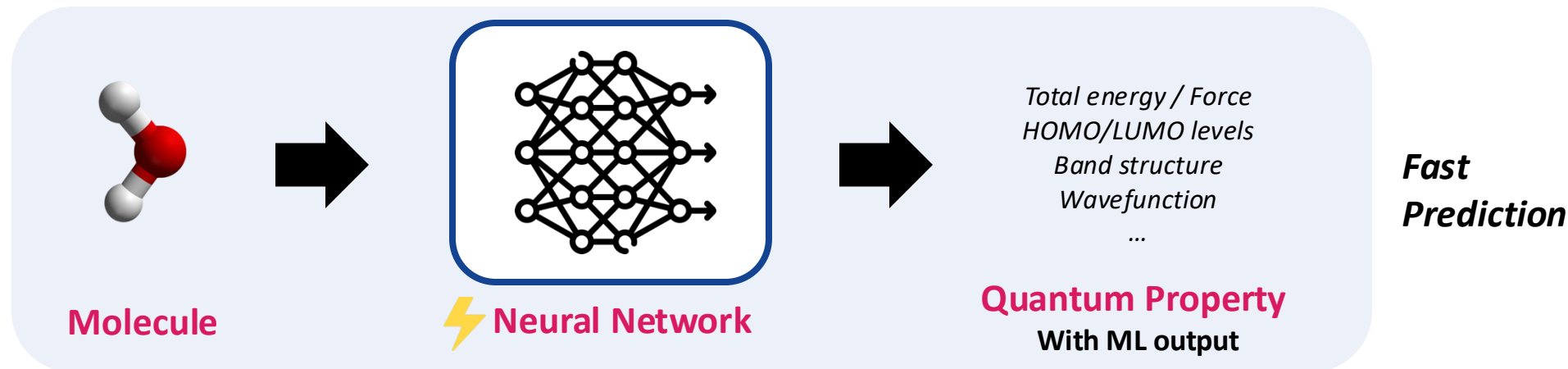
QVMC, CCSD(S)

DFT is a common standard in computational chemistry for both industry and academia

Our goal: ML-DFT

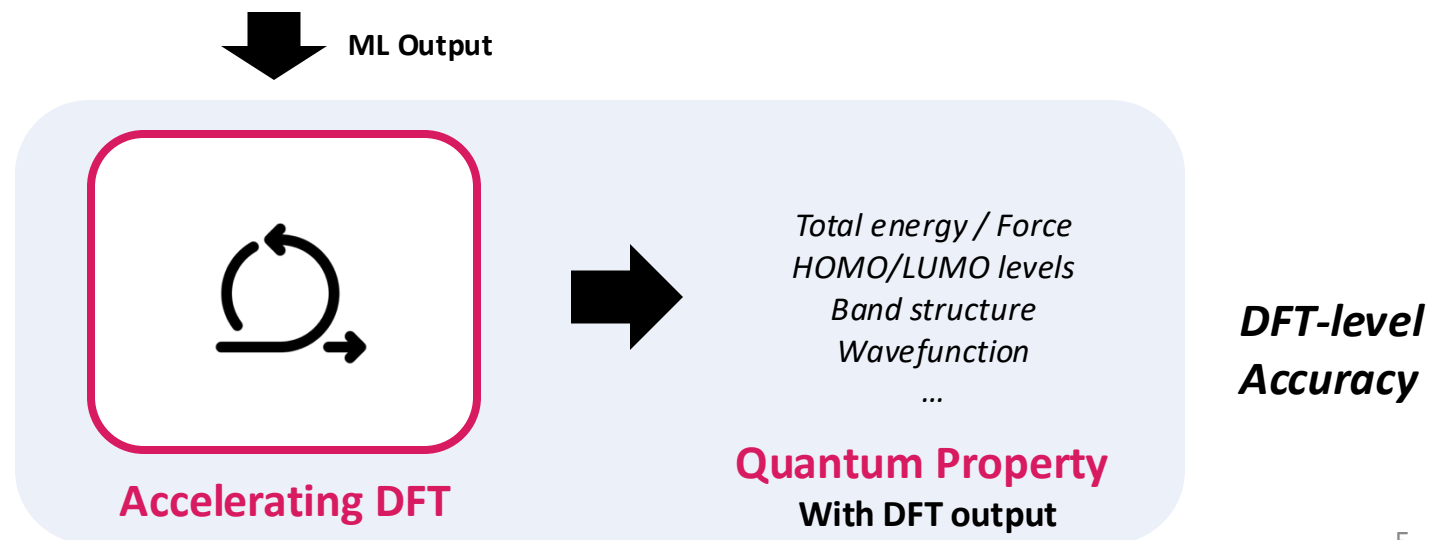
Goal 1: Fast Prediction

(One-shot approximation)

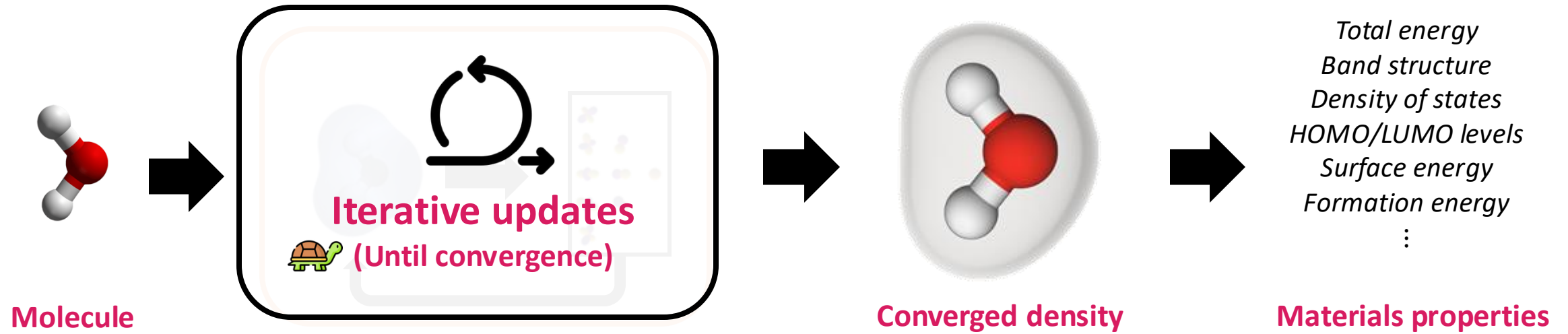


Goal 2: DFT Acceleration

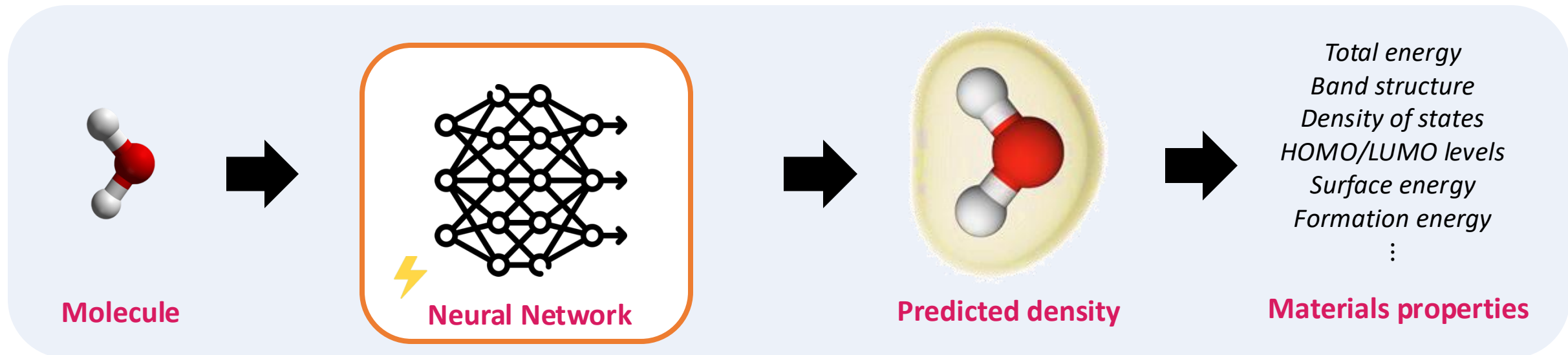
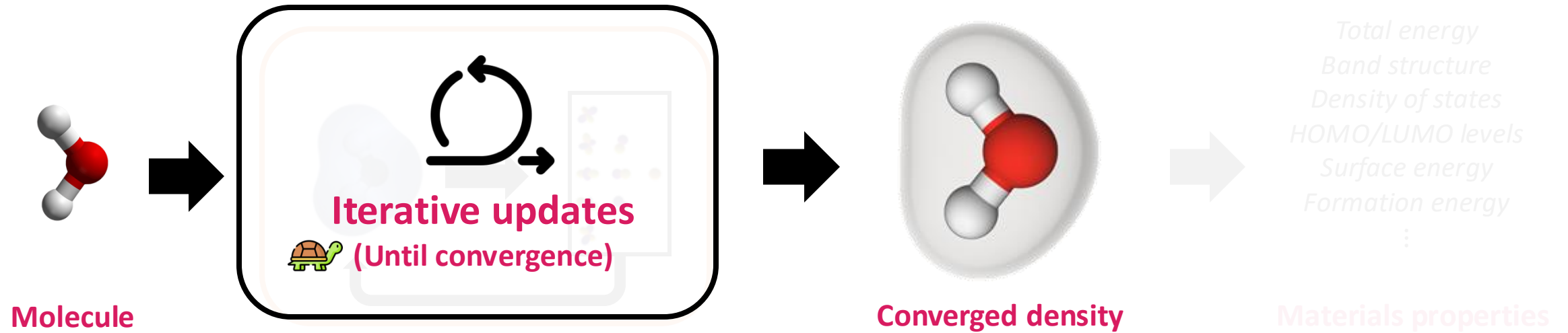
(With DFT calculation)



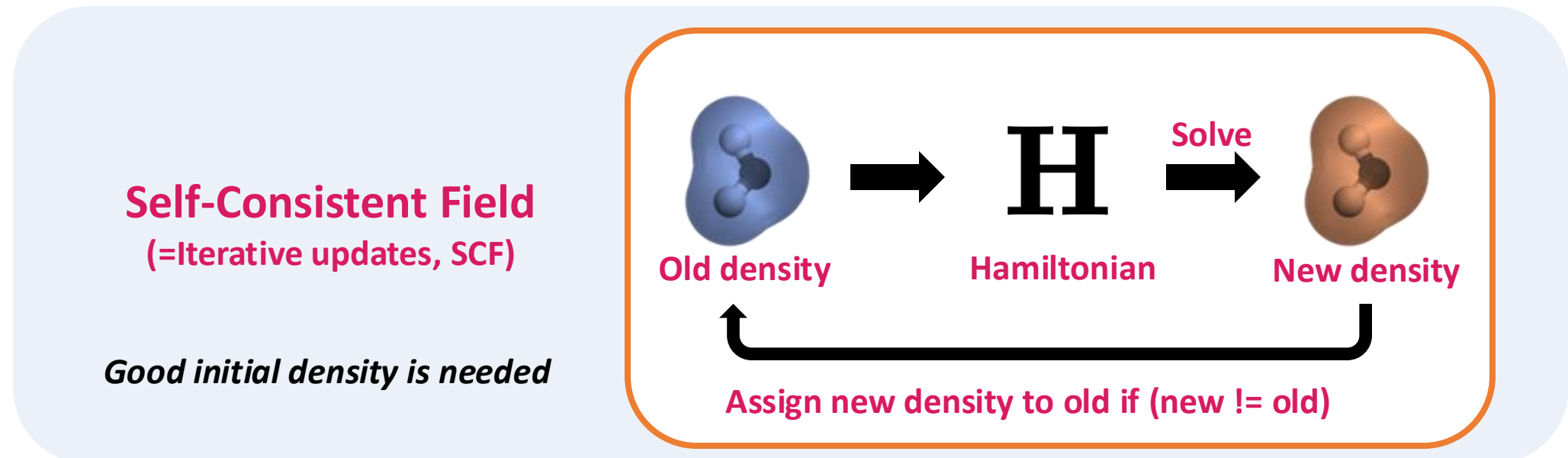
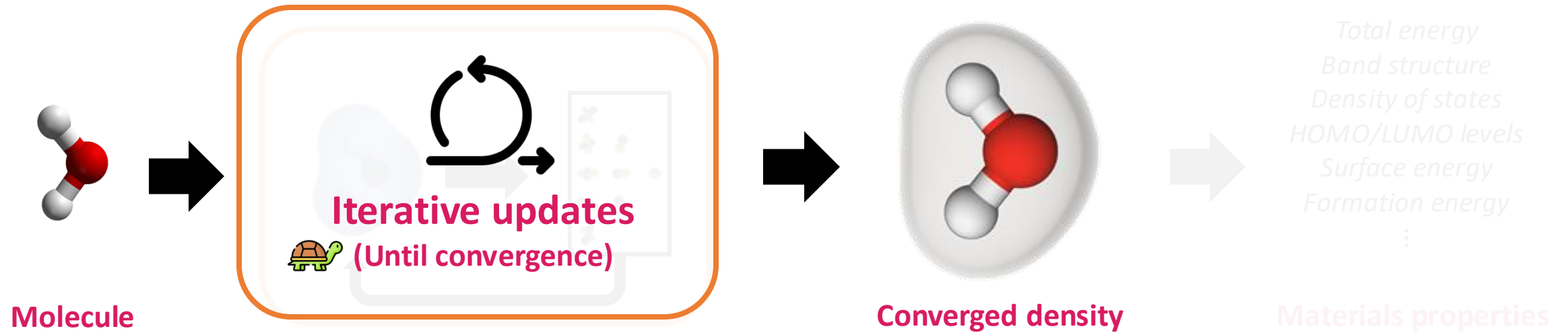
What is DFT?



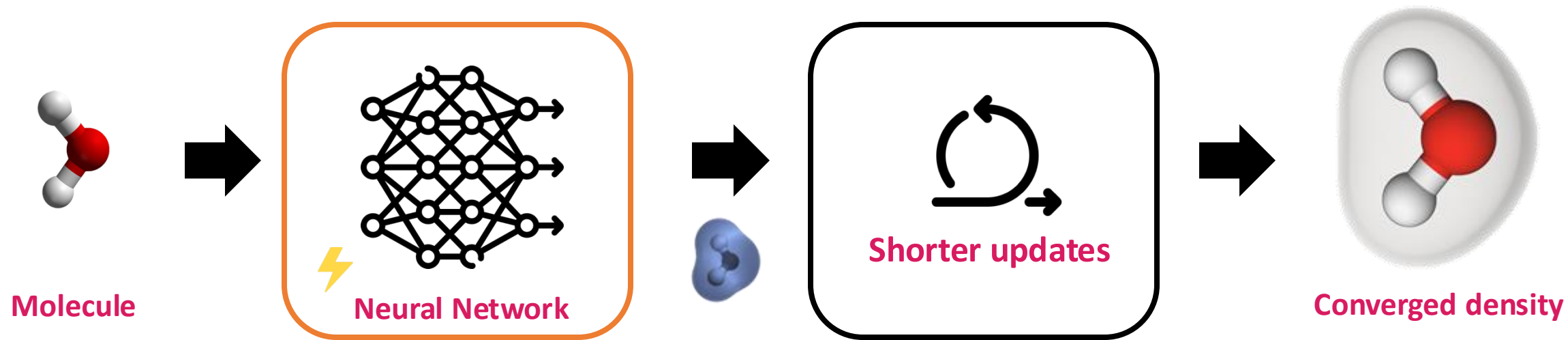
Our goal: (1) DFT Approximation



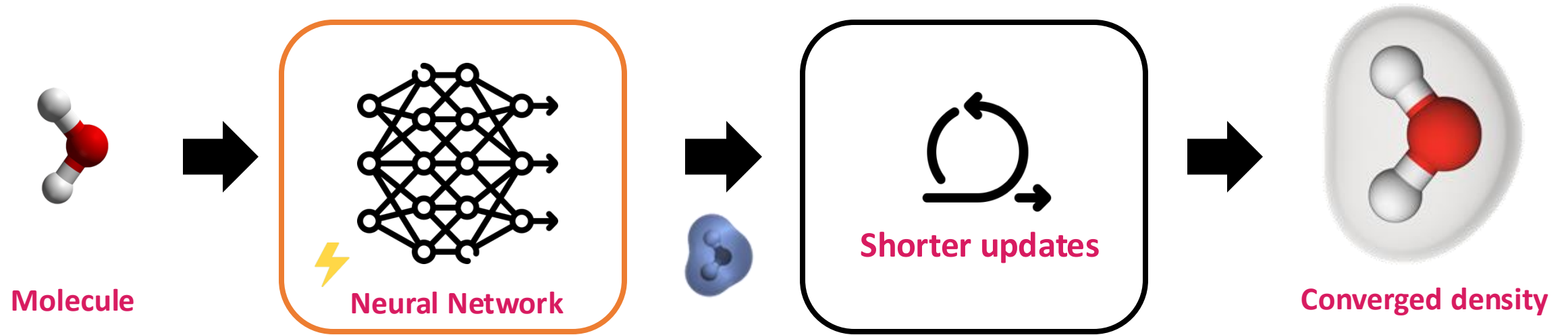
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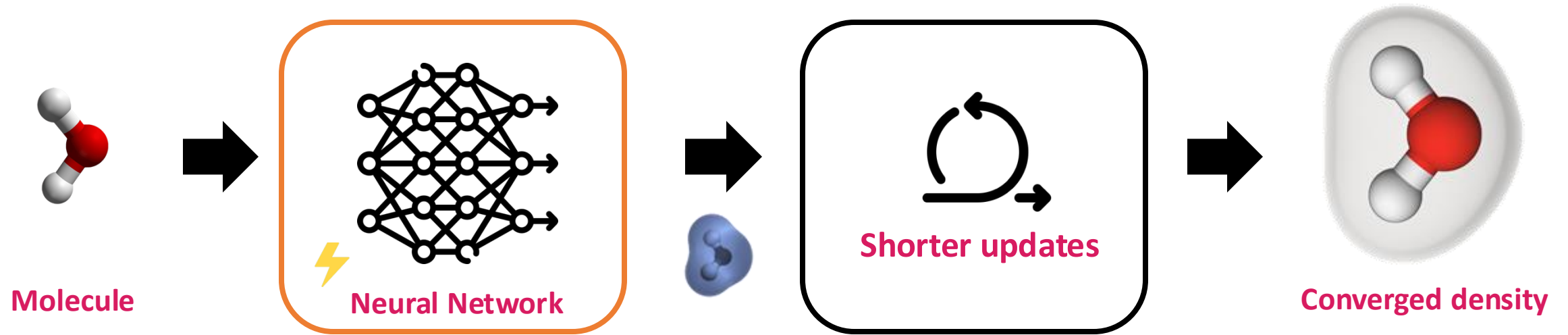


Our goal: (2) DFT Acceleration



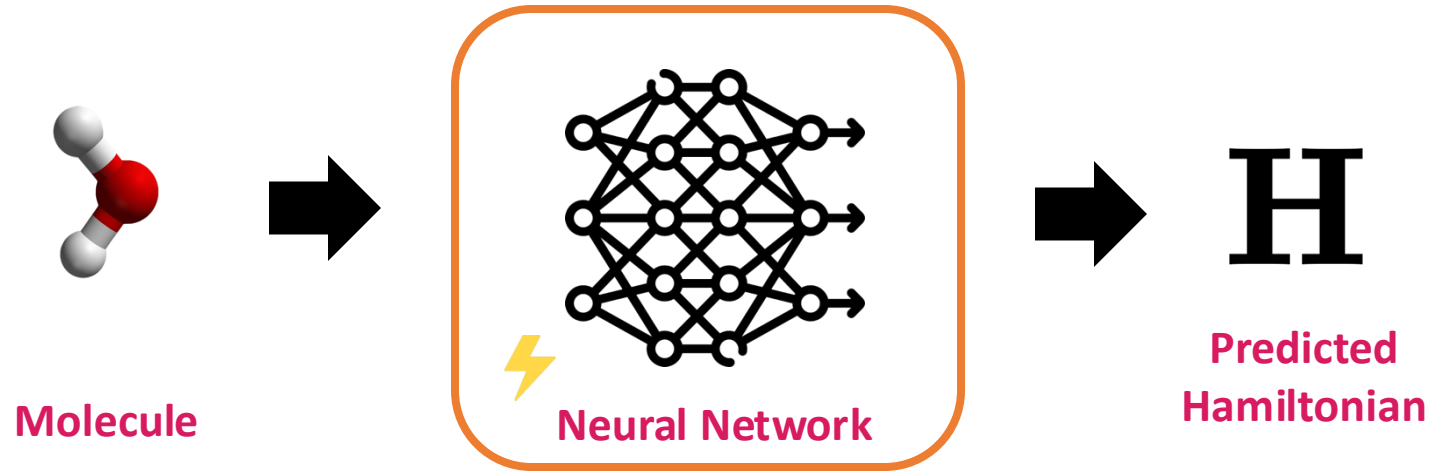
Spoiler: with ML acceleration..

Our goal: (2) DFT Acceleration



Spoiler: with ML acceleration..
-70% steps
-50% total time
(vs. conventional density initialization)

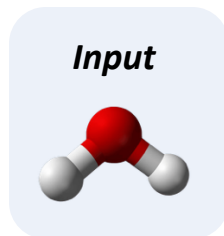
Our objective



Objective (as ML task):

Predict the Hamiltonian matrix from atomic geometry without SCF iterations

ML-DFT vs. MLFF / MLIP



Output format

Prediction Target

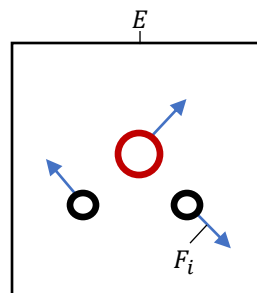
Available Properties

Correctability

Energy Error (MD17)

MLFF / MLIP

NequIP, Equiformer



Energy / Force

Energy, Force, Stress
(+ Scalar properties)

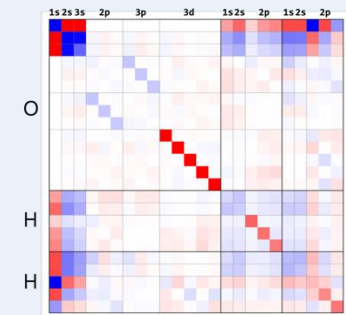
No

1

Equiformer=1

ML-DFT

QHNet, QHFlow



DFT Hamiltonian matrix

All DFT properties

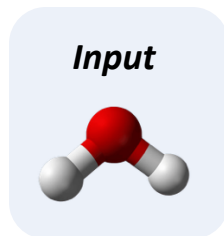
Energy, Force, Stress,
Electron density
Wavefunction, DOS, etc.

Yes

1/50

QHFlow / Equiformer

ML-DFT vs. MLFF / MLIP



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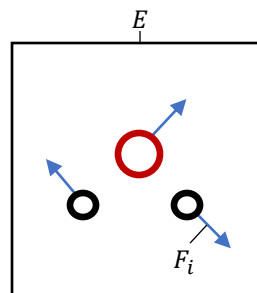
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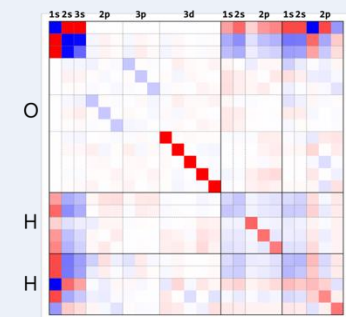
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QHFlow / Equiformer

Schrödinger Equation

The **Hamiltonian** of interest stems from the ***Schrödinger Equation***

$$\hat{H}\psi = E\psi$$

This equation is the **master equation of chemistry** (Quantum mechanics)

$\hat{H}$: *Hamiltonian operator* of the system

E : *Total energy* of the system

ψ : *Wavefunction*

By solving the equation, we can get all information about the material!

However, *directly solving it is extremely hard* - $O(N!)$ (Full Configuration Interaction)

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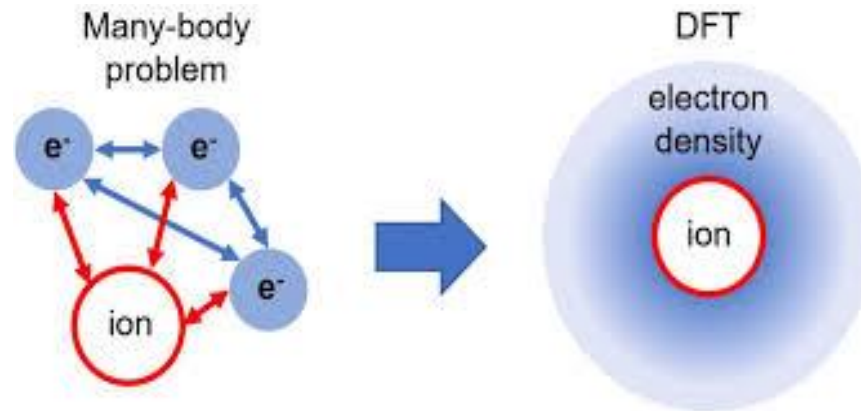
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Kohn-Sham (KS) Density Functional Theory

(Kohn-Sham) DFT is a practical approximation of the **Schrödinger Equation**

Main idea: Reformulate *many-body interaction* to *functional of the density* ρ



Schematic of the DFT assumption

Advantage: make the complex problem into *small independent problem*

$$\hat{H}\psi = E\psi$$

Original Schrödinger Equation

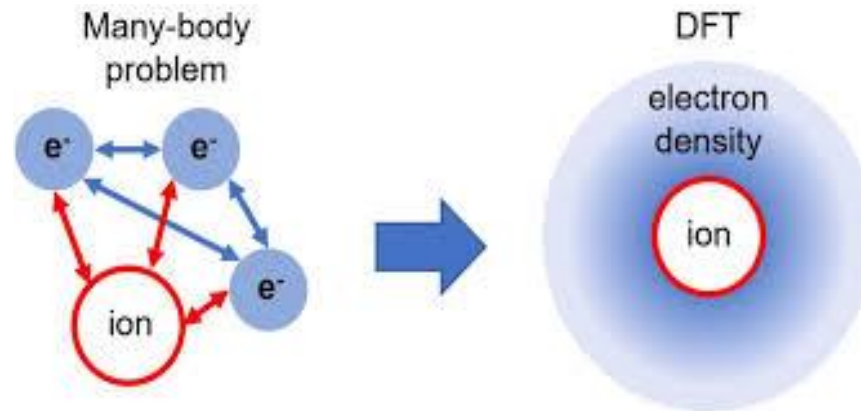
$$(\hat{H}_{\text{KS}}[\rho]\phi_i)(r) = \epsilon_i\phi_i(r), \quad \rho(r) = \sum_i |\phi_i(r)|^2$$

Kohn-Sham DFT formulation

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Kohn-Sham DFT formulation

DFT in matrix formulation

KS-DFT equation can be converted into **the matrix form**

Also known as the **Roothaan-Hall DFT (RH-DFT)**:

$$\mathbf{HC} = \mathbf{SC}\epsilon \quad \text{RH-equation}$$

$\mathbf{H}$: *Hamiltonian matrix (or called Fock matrix F)*

$\mathbf{C}$: *Density coefficient matrix*

$\mathbf{S}$: *Overlap matrix*

ϵ : *Diagonal orbital energy matrix*



With RH-DFT, we can handle the **density function** ρ as the coefficient matrix $\mathbf{C}$

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With RH-DFT, we can handle the **density function** ρ as the coefficient matrix **C**

DFT in matrix formation

$$\mathbf{H}\mathbf{C} = \mathbf{S}\mathbf{C}\epsilon$$

$\mathbf{S}$ is fixed when the system is given, easy to calculate (it depends on orbital basis set)

$\mathbf{C}$, ϵ can be obtained when $\mathbf{H}$, $\mathbf{S}$ are known

If we know the $\mathbf{H}$, then we can get electron density from obtained $\mathbf{C}$

(Note) $\mathbf{C}$ is not uniquely determined due to gauge freedom, so not a good target for supervised learning

From $\mathbf{H}$, $\mathbf{C}$, and ϵ , we can calculate the *HOMO/LUMO, Energy, Force*, etc..

We introduce **QHFlow**,

a **Hamiltonian prediction** framework

high-order **equivariant flow matching** with

invariant tensor-expansion priors

QHFlow significantly improves DFT (SCF) performance

- Reduces the **Hamiltonian error UP TO 73%**
- Reduces the **SCF iteration steps UP TO 68%**
- Reduces the **total DFT (SCF) time UP TO 54%**

High-order Equivariant Flow Matching for Density Functional Theory Hamiltonian Prediction

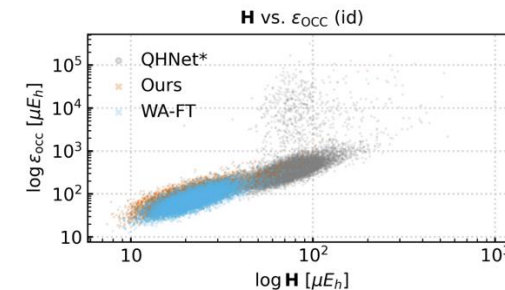
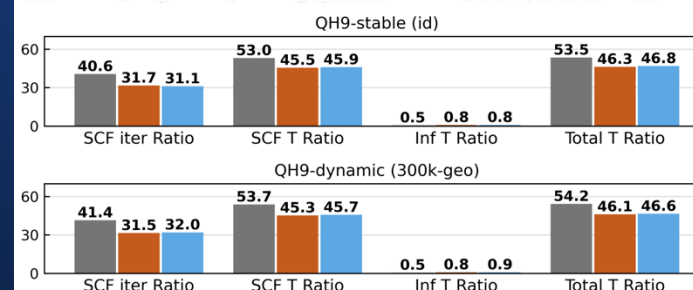
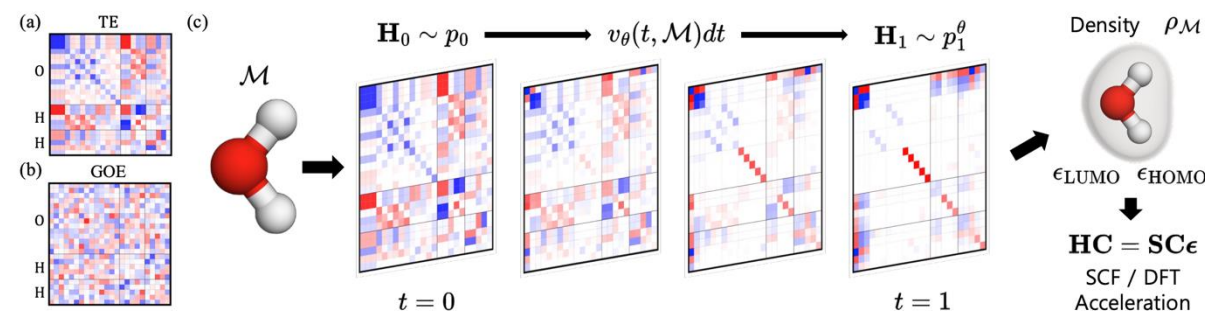
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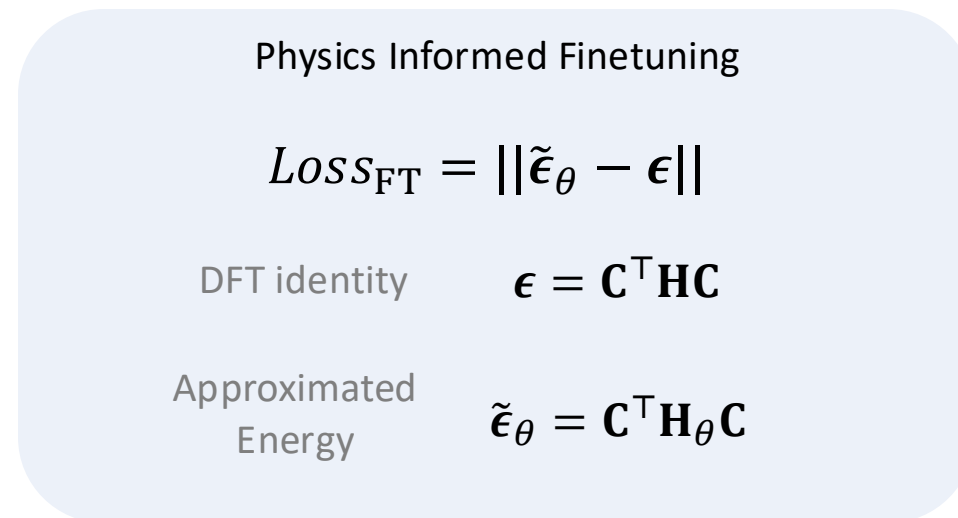
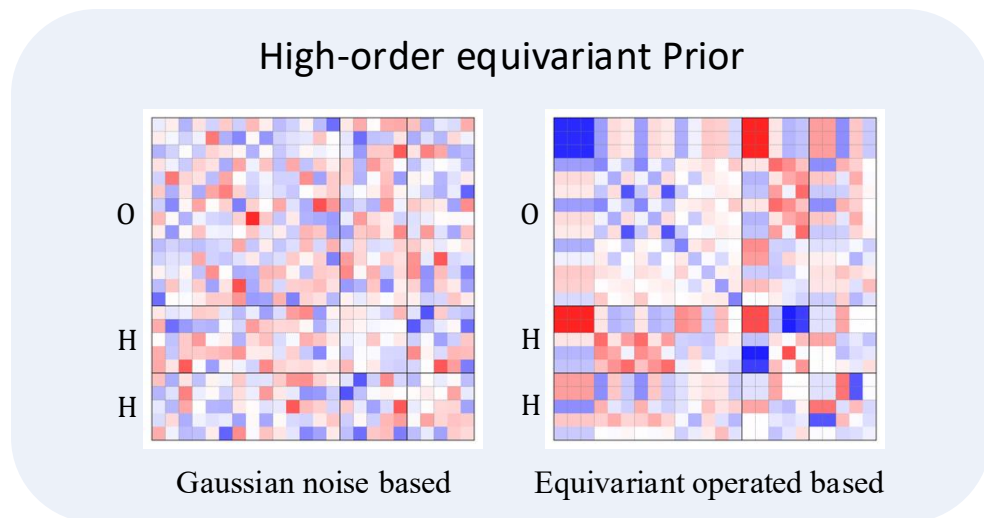
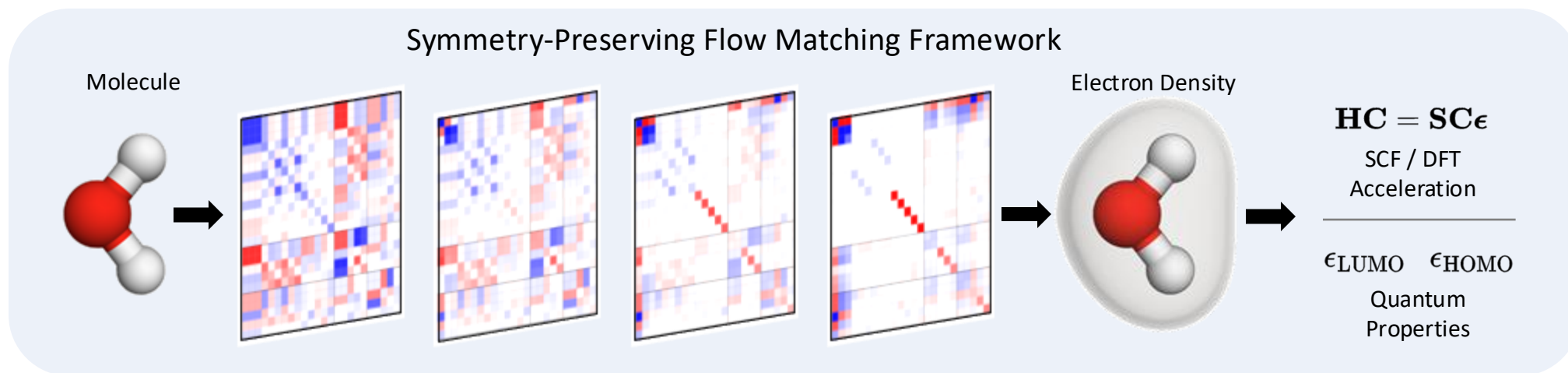
{seongsu.kim, nayoungkim, sungsoo.ahn}@kaist.ac.kr

dongwoo.kim@postech.ac.kr

<https://github.com/seongsukim-ml/QHFlow>



Overview of QHFlow

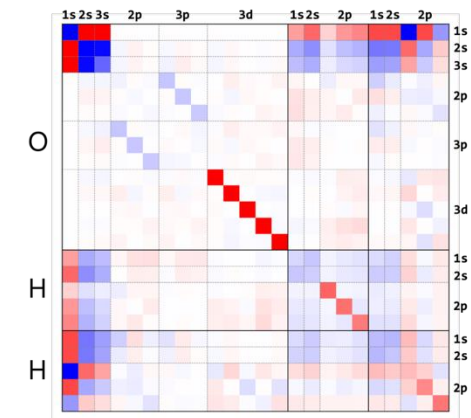
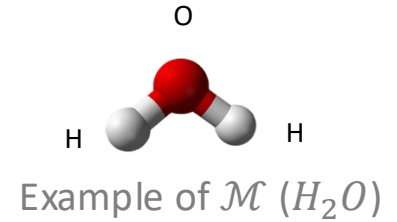


Problem setting

Objective: predict the Hamiltonian matrix $\mathbf{H}$

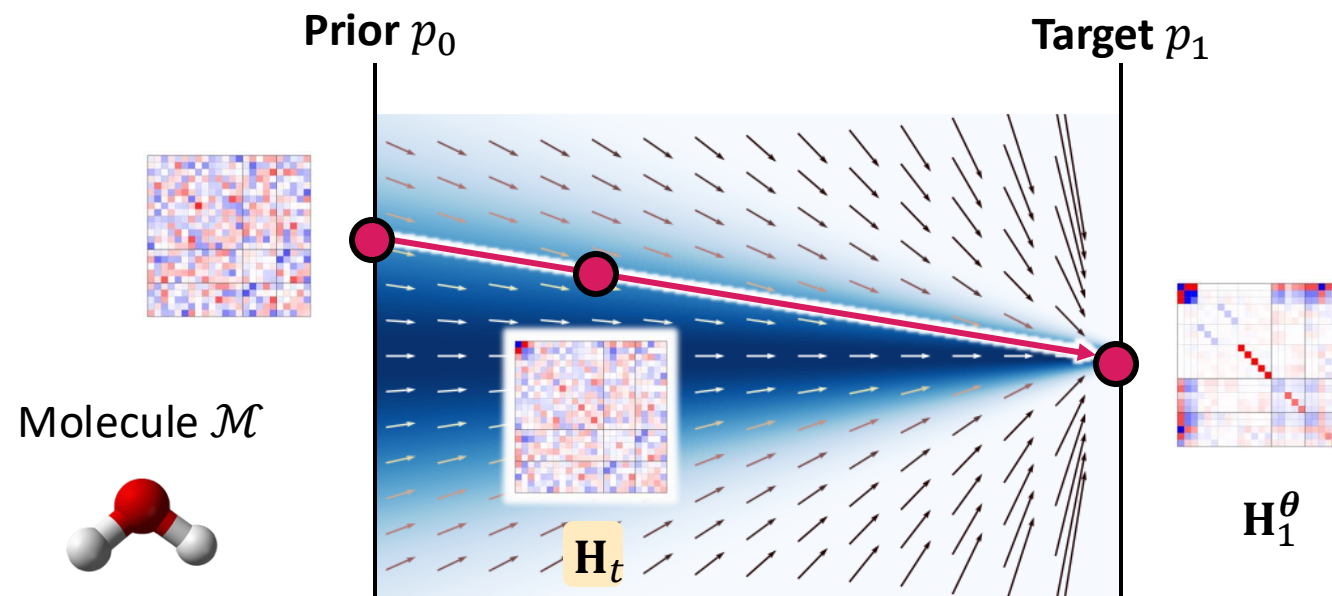
Input: (1) Molecular geometry $\mathcal{M}$ (atomic numbers Z , atomic positions X)
(2) Conventional Hamiltonian initial guess matrix $\mathbf{H}_{\text{init}}$

$$\mathbf{H}_{\text{pred}} = f_{\theta}(\mathcal{M}, \mathbf{H}_{\text{init}}) + \mathbf{H}_{\text{init}}$$



Example of Hamiltonian matrix $\mathbf{H}$ (H_2O)

Our method: Molecule conditioned flow matching



Insight: Learning the end point prediction $\mathbf{H}_1^\theta$ from middle point $\mathbf{H}_t$

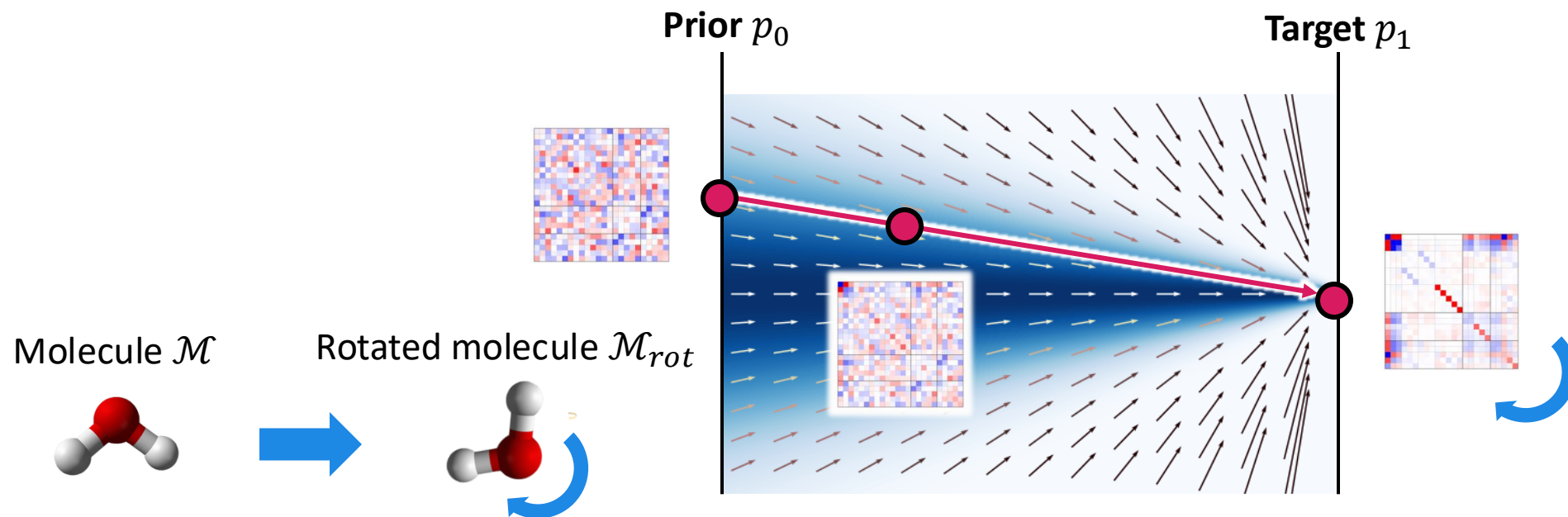
Vector field:

$$v_t^\theta = \frac{\mathbf{H}_1^\theta - \mathbf{H}_t}{1 - t}$$

Objective function:

$$\mathcal{L}_{\text{CFM}} = \mathbb{E}_{(\mathbf{H}, \mathcal{M}) \sim \mathcal{A}, t \sim \mathcal{U}(0,1), \mathbf{H}_t \sim p_t(\cdot | \mathbf{H})} \left[\frac{1}{(1 - t)^2} \left\| \mathbf{H}_1^\theta(\mathbf{H}_t, \mathcal{M}) - \mathbf{H}_{1, \mathcal{M}} \right\|_2^2 \right]$$

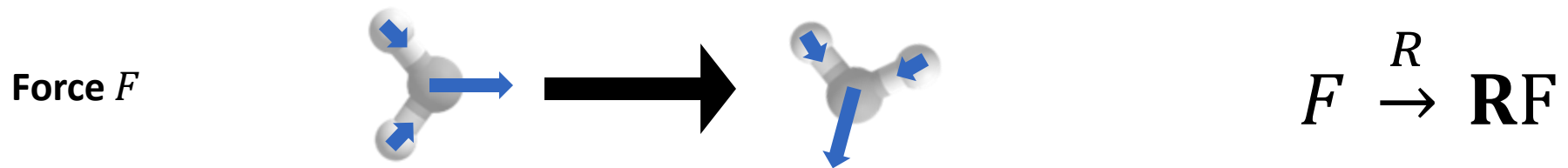
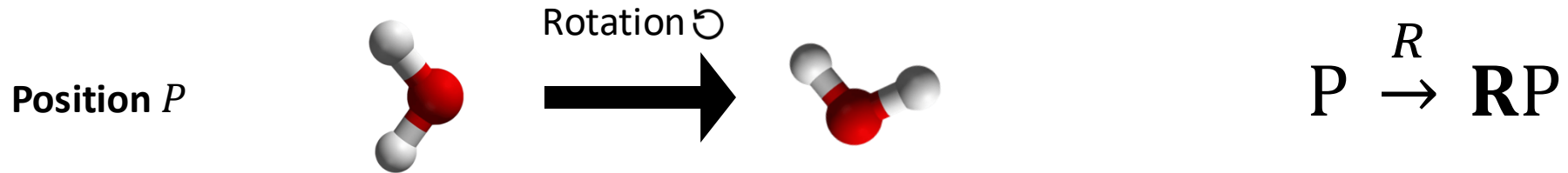
Our method: Equivariant flow matching



Preserving ***symmetry*** with flow matching → Equivariant flow matching

Equivariant property of Hamiltonian (Symmetry)

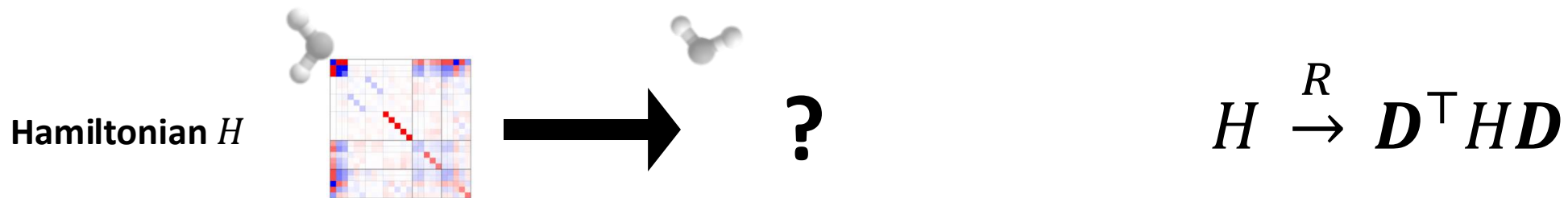
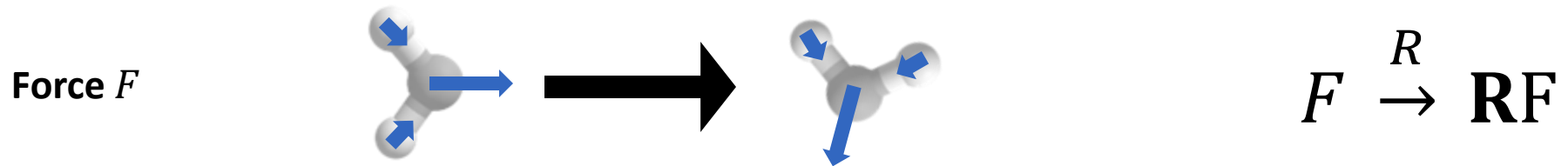
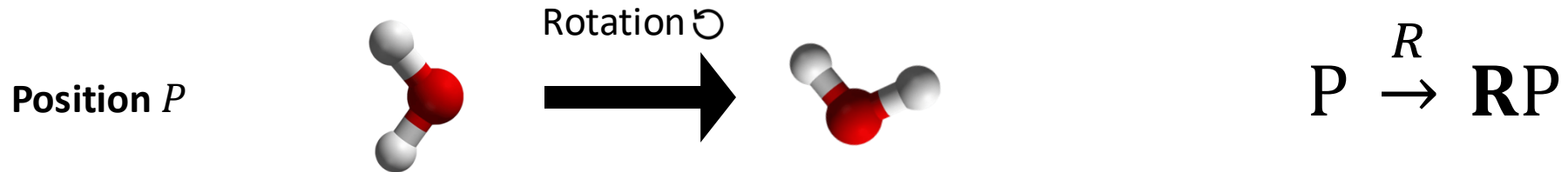
Just like **forces**, **Hamiltonian matrix** rotates along with the system



$D(R)$ plays a role of high-order rotation

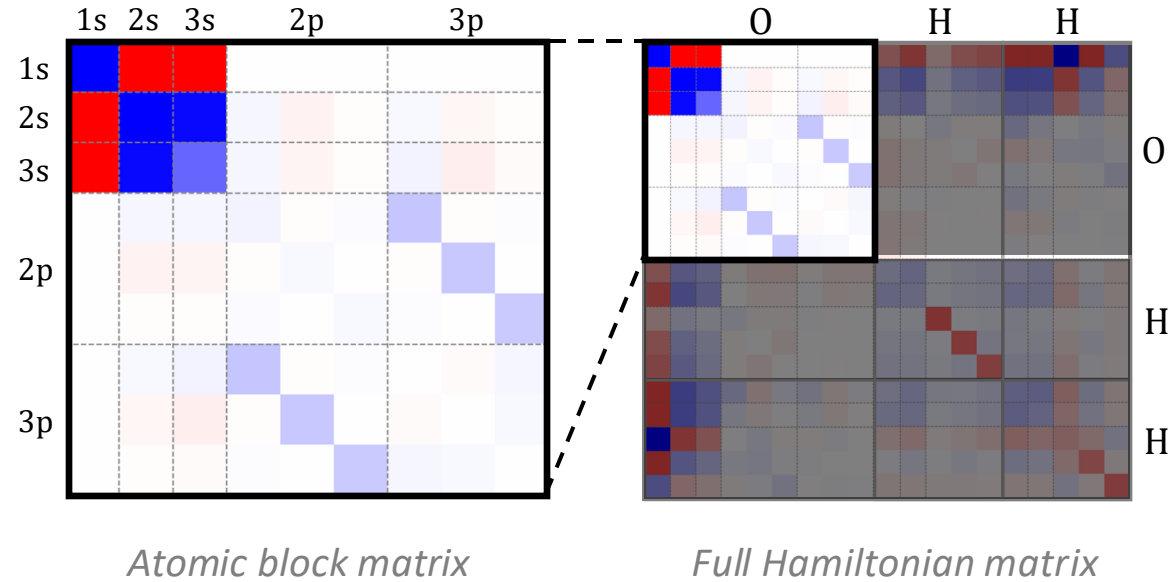
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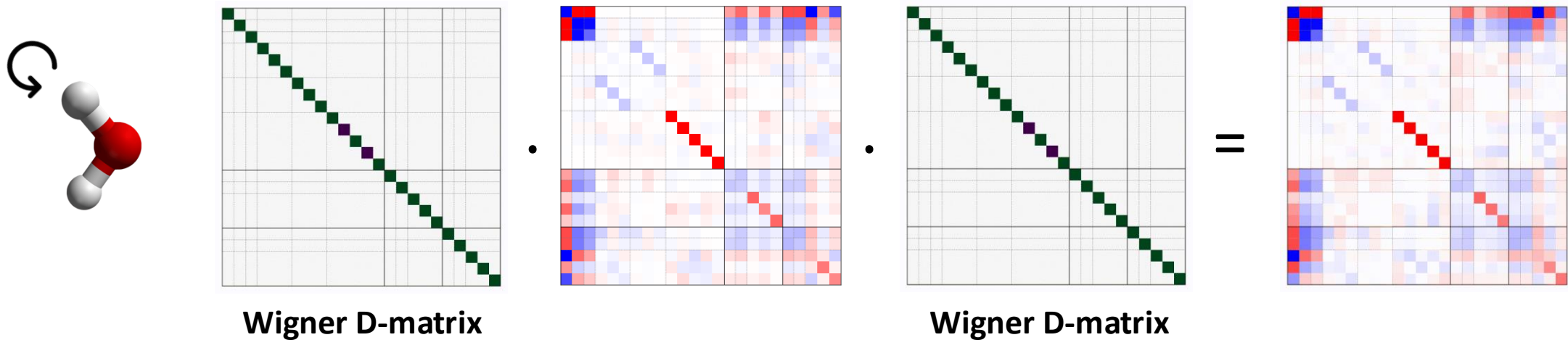
Equivariance of Hamiltonian



Hamiltonian matrix has **high-order equivariant structure**

Each block matrix needs a special type of rotation matrix!
Wigner D-matrix

Equivariance of Hamiltonian



$$\mathbf{D}^T \mathbf{H} \mathbf{D} = \mathbf{H}_{\text{rot}}$$

Applying a **special rotation matrix** for rotation of the Hamiltonian matrix
(Wigner D-matrix)

Equivariant flow matching for Hamiltonian

Rotated Hamiltonian has the same probability for any Hamiltonian, that is

$$p_t(\mathbf{R} \cdot \mathbf{H}) = p_t(\mathbf{H})$$

Following *Song et al*, this property can be satisfied by

(1) **Rotation invariant prior distribution ($t = 0$)**

$$p_0(\mathbf{R} \cdot \mathbf{H}) = p_0(\mathbf{H})$$

(2) **Rotation equivariant vector field**

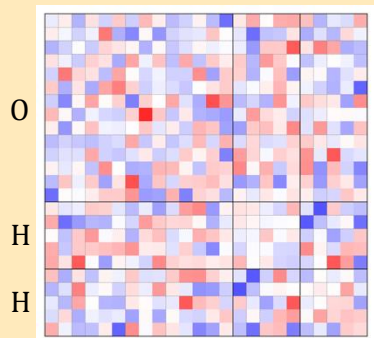
$$\mathbf{v}_t(\mathbf{R} \cdot \mathbf{H} | \mathbf{R}\mathcal{M}) = \mathbf{R} \cdot \mathbf{v}_t(\mathbf{H} | \mathcal{M})$$

Our method: Invariant prior design

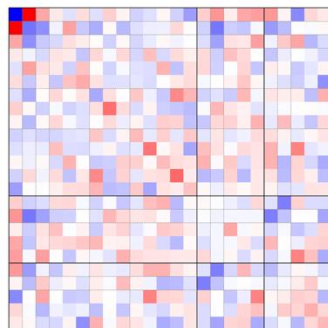
Two types of prior satisfying the invariance:

$$p_0(\mathbf{R} \cdot \mathbf{H} | \mathcal{RM}) = p_0(\mathbf{H} | \mathcal{M})$$

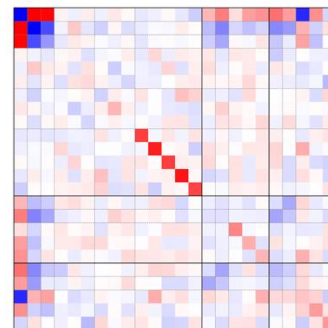
GOE
Gaussian Orthogonal Ensemble



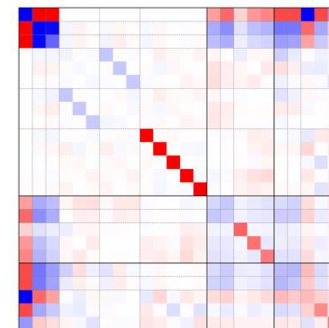
$t = 0$



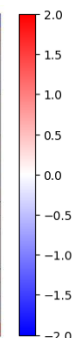
$t = 1/3$



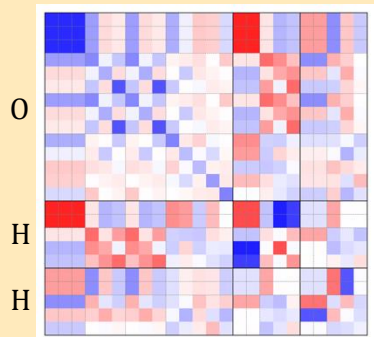
$t = 2/3$



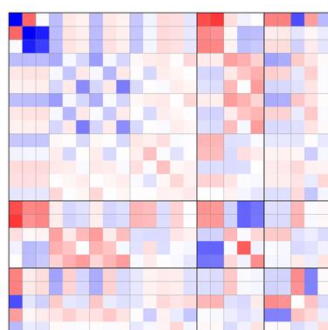
$t = 1$



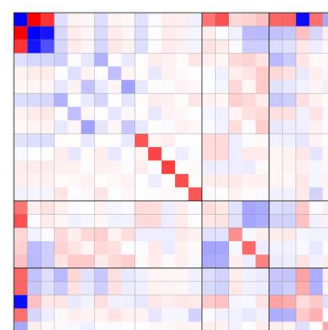
TE
Tensor Expansion



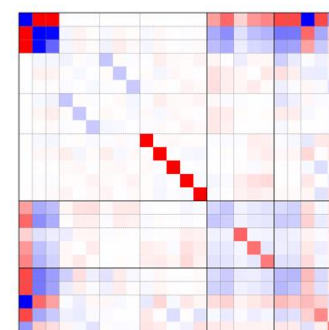
$t = 0$



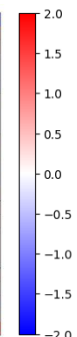
$t = 1/3$



$t = 2/3$

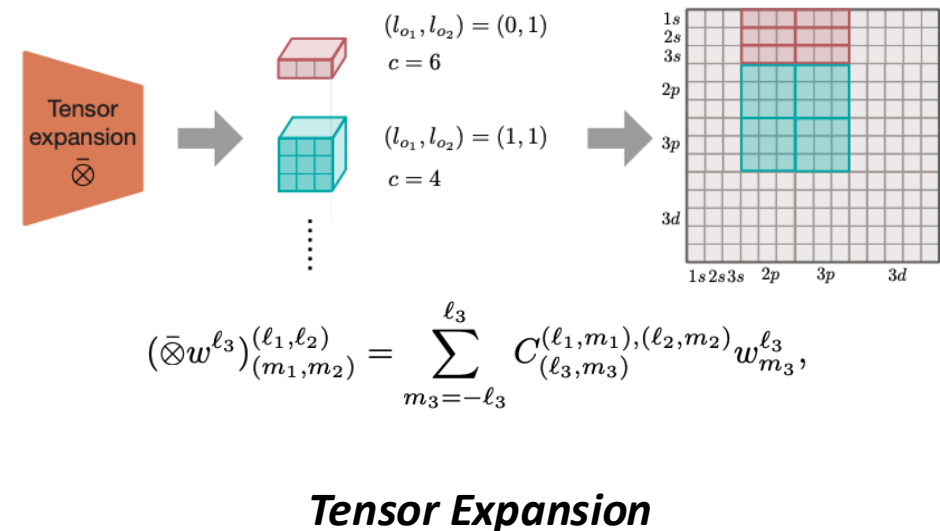
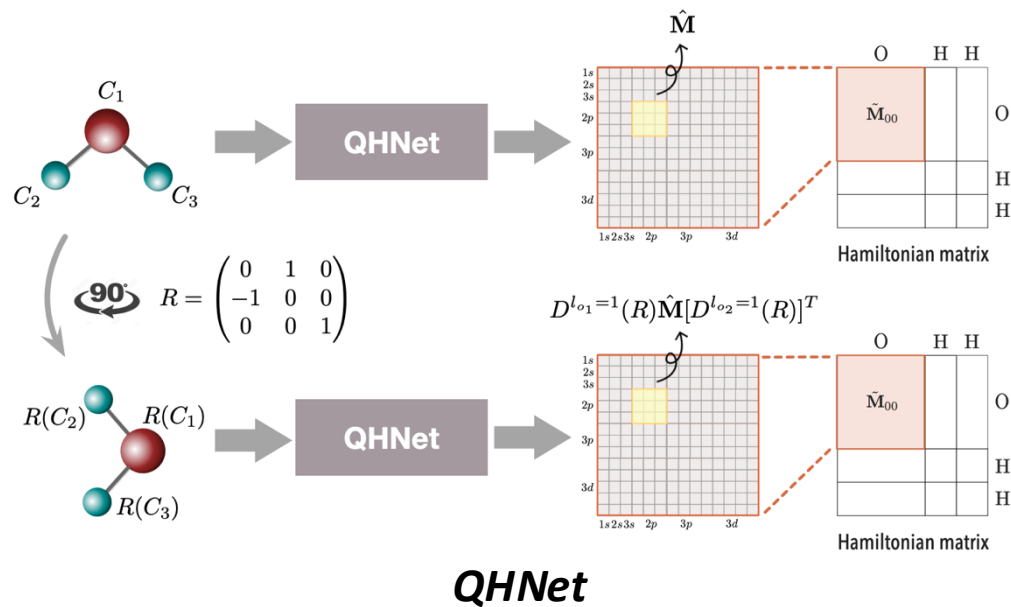


$t = 1$



Equivariant vector field

Using rotation equivariant Hamiltonian prediction architecture (QHNet, Yu et al, 2023)



Modified the architecture to introduce two additional inputs:

- t : **Time conditioning** for flow matching
- $\mathbf{H}_t$: the **current Hamiltonian** matrix

Our method: Physics-informed finetuning

- **Approximated orbital energies $\tilde{\epsilon}$** (from *Li et al*)

$$\tilde{\epsilon}_{\theta} = \mathbf{C}^{\top} \mathbf{H}_{\theta} \mathbf{C}$$

- **Ground-truth orbital energies ϵ**

$$\epsilon = \mathbf{C}^{\top} \mathbf{H} \mathbf{C}$$

- Our finetuning aligns these two values

$$Loss_{FT} = ||\tilde{\epsilon}_{\theta} - \epsilon||$$

$$Total Loss = Loss_{flow} + Loss_{FT}$$

$$\mathbf{H} \mathbf{C} = \mathbf{S} \mathbf{C} \epsilon$$

RH-DFT equation

$$\mathbf{C}^{\top} \mathbf{S} \mathbf{C} = \mathbf{I}$$

Identity property

$$\mathbf{C}^{\top} \mathbf{H} \mathbf{C} = \epsilon$$

Orbital energy

Experiment

DFT Approximation – MD₁₇

- Molecular dynamics trajectory for four small molecules
 - Same chemical formula Z with *different atomic positions* X

$$HC = SC\epsilon$$

H : Hamiltonian MAE

ϵ_{occ} : occupied energy MAE

S_c : Similarity score of the coefficients

Model	Water (3 atoms)			Ethanol (9 atoms)			Malonaldehyde (9 atoms)			Uracil (12 atoms)		
	$H \downarrow$ [μE_h]	$\epsilon_{occ} \downarrow$ [μE_h]	$S_c \uparrow$ [%]	$H \downarrow$ [μE_h]	$\epsilon_{occ} \downarrow$ [μE_h]	$S_c \uparrow$ [%]	$H \downarrow$ [μE_h]	$\epsilon_{occ} \downarrow$ [μE_h]	$S_c \uparrow$ [%]	$H \downarrow$ [μE_h]	$\epsilon_{occ} \downarrow$ [μE_h]	$S_c \uparrow$ [%]
(2019) SchNOrb	165.4	279.3	100.00	187.4	334.4	100.00	191.1	400.6	99.00	227.8	1760.	90.00
(NIPS'21) PhiSNet	15.67	85.53	100.00	<u>20.09</u>	102.04	99.81	21.31	100.6	99.89	<u>18.65</u>	143.36	99.86
(ICML'23) QHNet*	<u>11.70</u>	<u>26.06</u>	100.00	27.99	99.33	<u>99.99</u>	29.60	100.16	<u>99.92</u>	26.80	127.93	<u>99.87</u>
(ICML'25) SPHNet	23.18	182.29	100.00	21.02	<u>82.30</u>	100.00	<u>20.67</u>	<u>95.77</u>	99.99	19.36	<u>118.21</u>	99.99
Ours	4.93	19.29	100.00	5.33	29.03	100.00	3.80	22.68	99.99	3.68	30.54	99.99

- Our model shows **71% error reduction** on Hamiltonian
- Our model shows lower error on **occupied energy** ϵ_{occ} and higher **coefficient similarity score** S_c
 - Higher physical fidelity

DFT Approximation – QH9

- The **general molecule dataset**, which consists of various chemical formulas

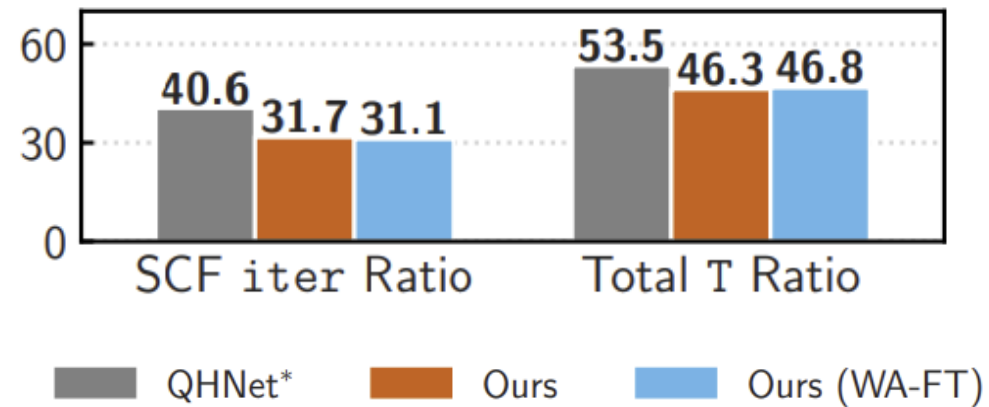
Dataset	Model	<i>H Error</i>	<i>Energy related error</i>				
		$H \downarrow [\mu E_h]$	$\epsilon_{occ} \downarrow [\mu E_h]$	$\mathcal{S}_c \uparrow [\%]$	$\epsilon_{LUMO} \downarrow [\mu E_h]$	$\epsilon_{HOMO} \downarrow [\mu E_h]$	$\epsilon_{\Delta} \downarrow [\mu E_h]$
(ICML'23) QHNet (ICLR'25) WANet (ICML'25) SPHNet	QHNet*	77.72	963.45	94.80	18257.34	1546.27	17822.62
	WANet	80.00	833.62	96.86	-	-	-
	SPHNet	45.48	334.28	97.75	-	-	-
	Ours	22.95	119.67	99.51	437.96	179.48	553.87
	Ours (WA-FT)	23.85	101.92	99.56	187.48	92.22	206.15
	w/ Finetuning						
QH9-stable (id)	QHNet*	69.69	884.97	93.01	25848.83	1045.99	25370.10
	SPHNet	43.33	186.40	98.16	-	-	-
	Ours	20.01	84.54	99.04	321.20	130.74	395.83
	Ours (WA-FT)	20.55	72.64	99.16	171.24	77.96	179.57
QH9-stable (ood)	QHNet*	88.36	1170.50	93.65	23269.41	2040.06	22407.96
	WANet	74.74	416.57	99.68	-	-	-
	SPHNet	52.18	100.88	99.12	-	-	-
	Ours	25.94	103.11	99.59	425.18	175.18	547.33
QH9-dynamic (300k-geo)	QHNet*	121.39	5554.36	86.02	53505.09	4352.76	50424.86
	SPHNet	108.19	1724.10	91.49	-	-	-
	Ours	45.91	442.56	98.65	1344.68	479.71	1605.03
	Ours (WA-FT)	46.60	424.75	98.74	912.10	403.51	1047.88

H : Hamiltonian MAE
 ϵ_{occ} : occupied energy MAE
 $\mathcal{S}_c$: Similarity score of the coefficients
 ϵ_{LUMO} : LUMO energy MAE
 ϵ_{HOMO} : HOMO energy MAE
 ϵ_{Δ} : LUMO-HOMO energy diff. MAE

- Our model show a **53% error reduction** on average
- With the **finetuning (WA-FT)**, additional **improvement in Energy** with a trade-off in **Hamiltonian error**

DFT Acceleration – QH₉

Initialization of the SCF process



- Reduces the **SCF steps** calculations by **about 69% (= 1-0.31)**
- Reduces the **total time** of SCF convergence by **about 54% (= 1-0.46)** (Inference time included)

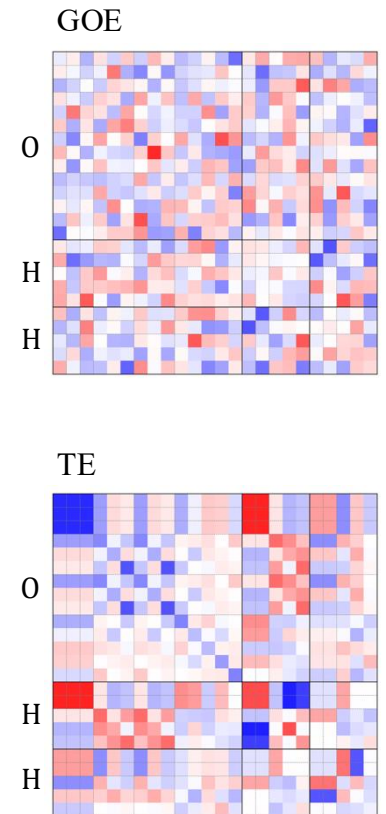
Here, 100% is the conventional initialization (*MinAO*), and a lower value implies more efficiency

We tested on the first 300 samples from each test split

Ablation – Prior distribution

- Influence of the prior distribution on QH9

Data	Prior	$H \downarrow [\mu E_h]$	$\epsilon_{\text{occ}} \downarrow [\mu E_h]$	$\mathcal{S}_c \uparrow [\%]$
id	GOE	25.93	154.65	99.39
	TE	22.95	119.61	99.51
ood	GOE	20.41	87.32	98.95
	TE	20.01	84.54	99.04
geo	GOE	29.39	122.14	99.49
	TE	25.94	103.11	99.59
mol	GOE	46.78	419.68	98.65
	TE	45.91	442.56	98.65



- **TE** prior consistently yields lower errors than **GOE**
- Highlights importance of designing appropriate priors for Hamiltonian prediction

Thank you!

Open to talk!

Interested in *AI for science, computational biology, and material design!*

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