Boosting Ab-initio Molecular Dynamics with Deep Learning

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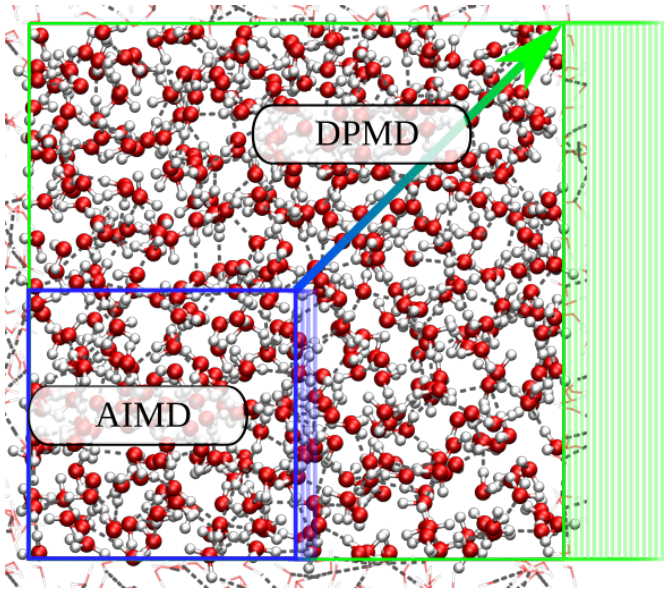
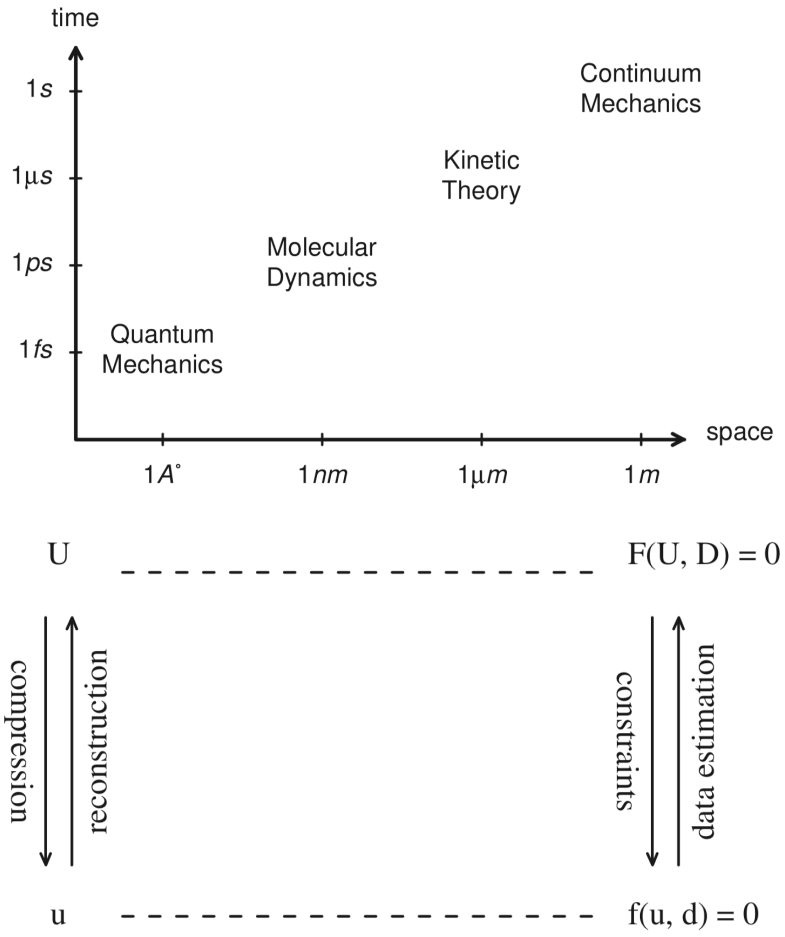
From AIMD to DPMD

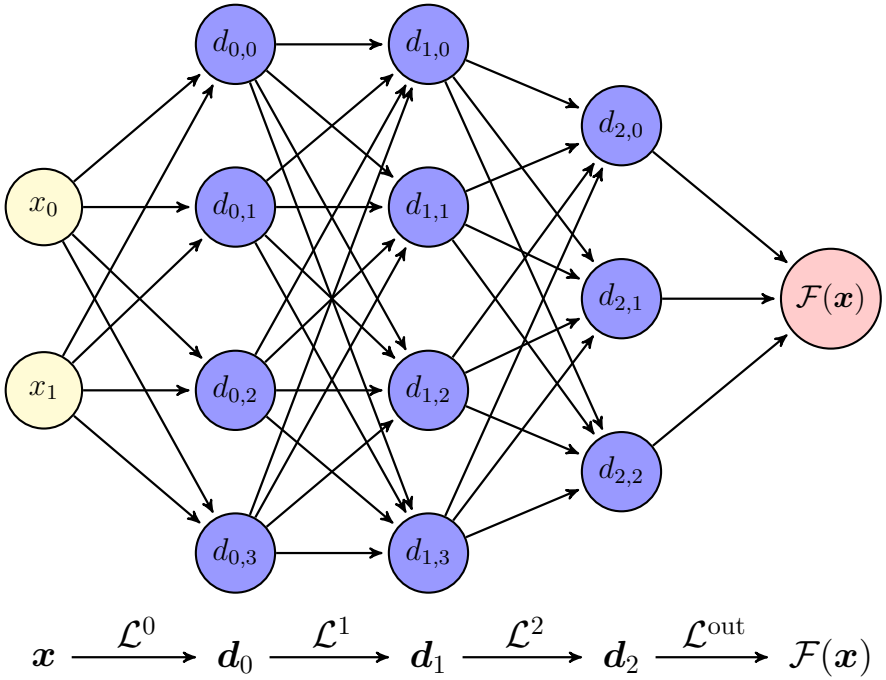
- *Ab-initio* molecular dynamics (AIMD) (Car and Parrinello, 1985) opened the way to simulations of materials without relying on empirical potentials;
- After 30+ years the main limit of AIMD is its computational cost that severely restricts accessible size and time scales;
- Boosting AIMD with Deep Potential molecular dynamics (DPMD):



Why deep learning could help



Multi-scale modeling problems



High-dimensional functions;
Composion of non-linearity.

Outline

- Modeling a potential energy surface (PES);
- Modeling the electronic structure information;
- A deep reactive PES model for water;
- Conclusion.

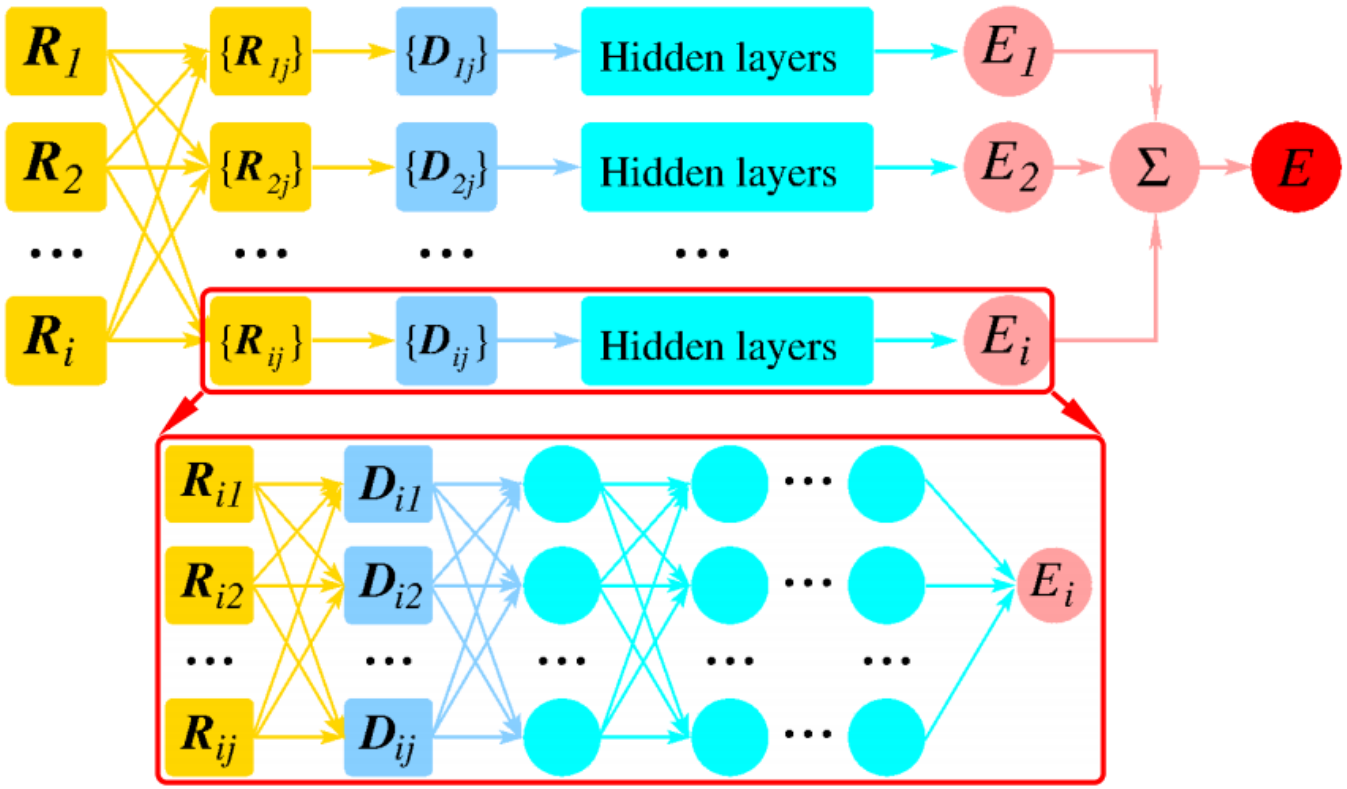
Requirement for a reliable PES model

$$E = E(\boldsymbol{r}_1, \boldsymbol{r}_2, \cdots, \boldsymbol{r}_i, \cdots, \boldsymbol{r}_N)$$

- accuracy (e.g. uniform);
- efficiency (e.g. linear scaling);
- physical constraint (e.g. extensivity, symmetry);
- no human intervention/ end-to-end.

Typical construction

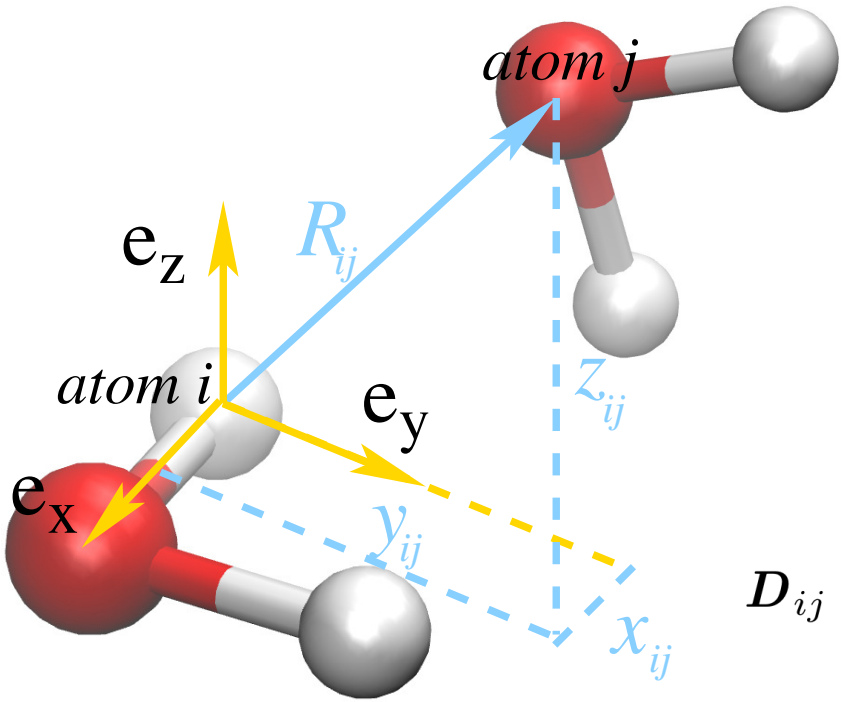
$$E = \sum_i E_i,\ E_i = E_{s(i)}(\boldsymbol{r}_i, \{\boldsymbol{r}_j\}_{j\in\mathcal{N}(i)}),\ \mathcal{N}(i) = \{j : r_{ij} = |\boldsymbol{r}_{ij}| \le r_c\}$$



$E_i(\boldsymbol{r}_i, \{\boldsymbol{r}_j\}_{j\in\mathcal{N}(i)})$ represented by DNNs with $symmetrized$ inputs.

Behler, J., Parrinello, M. (2007). Phys. Rev. Lett., 98(14), 146401.

Coordinates in a local frame



$$\boldsymbol{D}_{ij} = \{1/R_{ij}\}$$

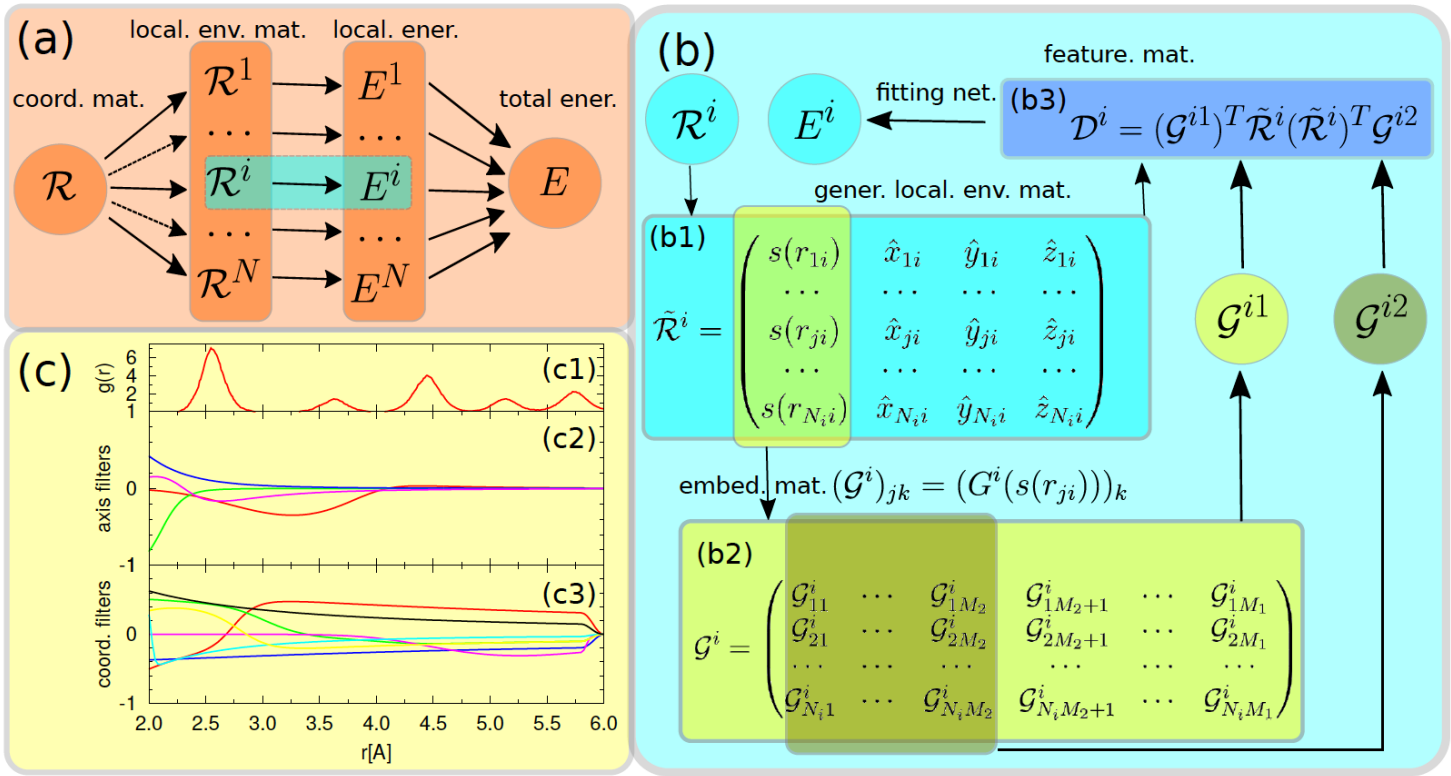
or

$$\boldsymbol{D}_{ij} = \{1/R_{ij}, x_{ij}/R_{ij}^2, y_{ij}/R_{ij}^2, z_{ij}/R_{ij}^2\}$$

J. Han, L. Zhang, R. Car, W. E, Comm. in Comp. Phys., 23, 629 (2018);

L. Zhang, J. Han, H. Wang, R. Car, W. E, Phys. Rev. Lett. 120, 143001 (2018);

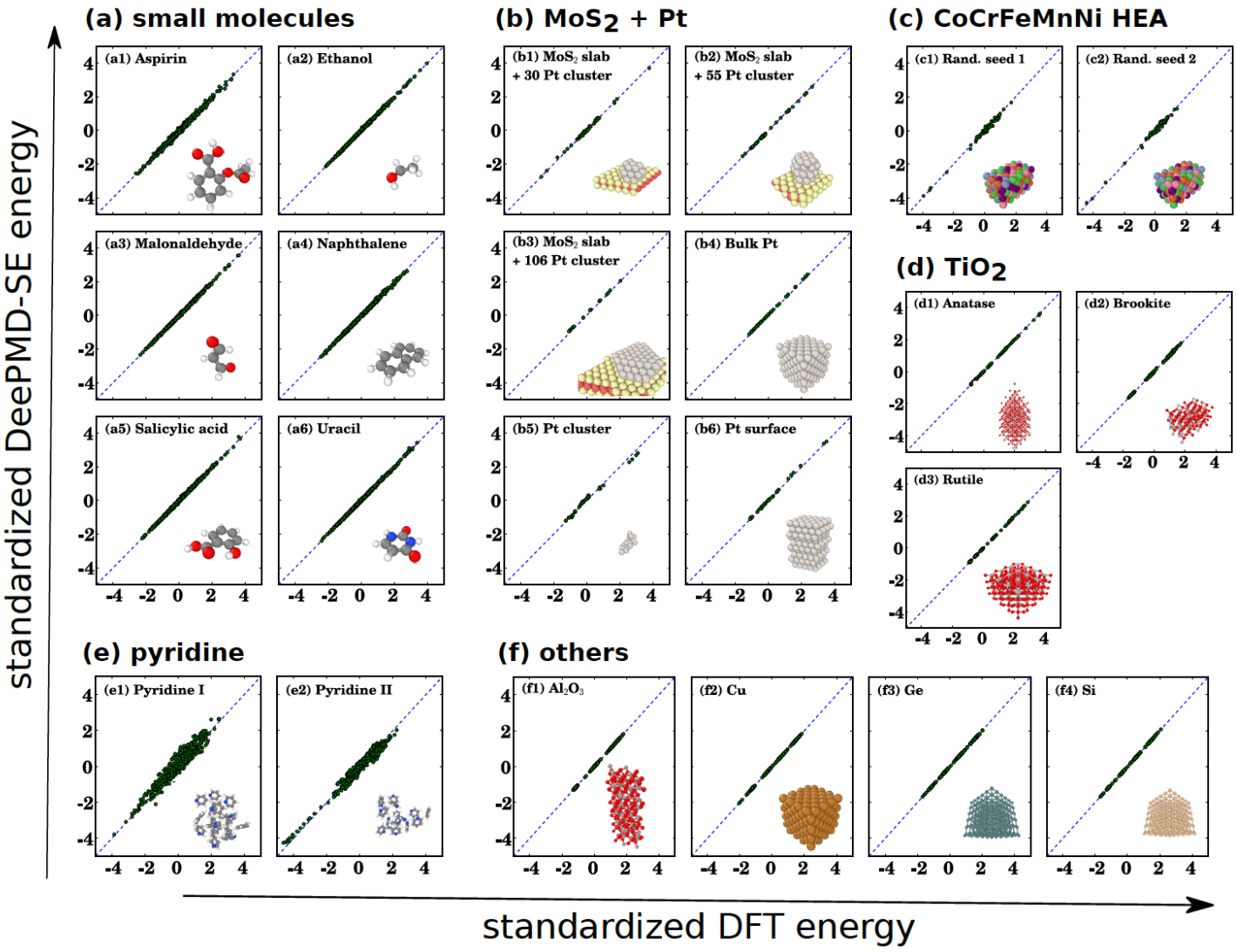
Adaptive and faithful descriptors



- Translation and Rotation: $(\mathcal{R}^i(\mathcal{R}^i)^T)$: $\Omega^i_{jk} = \boldsymbol{r}_{ji} \cdot \boldsymbol{r}_{ki}$,
- Permutation: $((\mathcal{G}^{i1})^T\mathcal{R}^i)$: $\sum_{j\in\mathcal{N}(i)} g(\boldsymbol{r}_{ji})\boldsymbol{r}_{ji}$,
- Finally, we propose: $\mathcal{D}^i = (\mathcal{G}^{i1})^T\mathcal{R}^i(\mathcal{R}^i)^T\mathcal{G}^{i2}$.

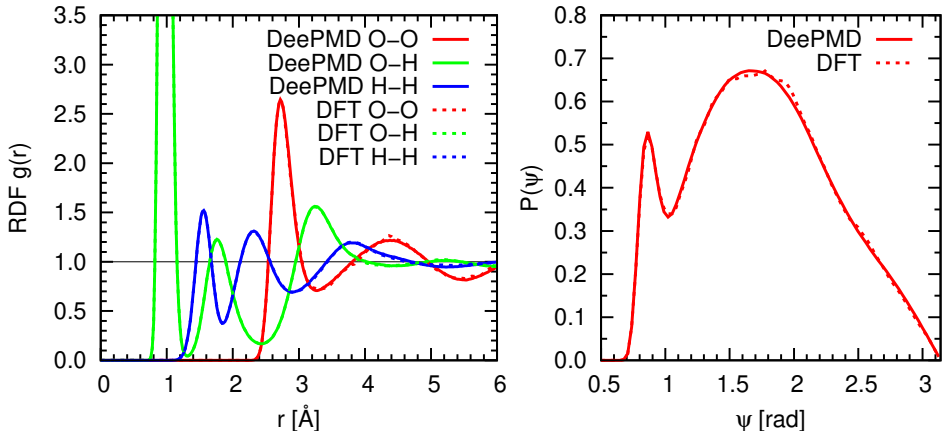
L. Zhang, J. Han, H. Wang, W. Saidi, R. Car, W. E, Advances in Neural Information Processing Systems 31, 4441 (2018).

Modeling various systems with the same principle

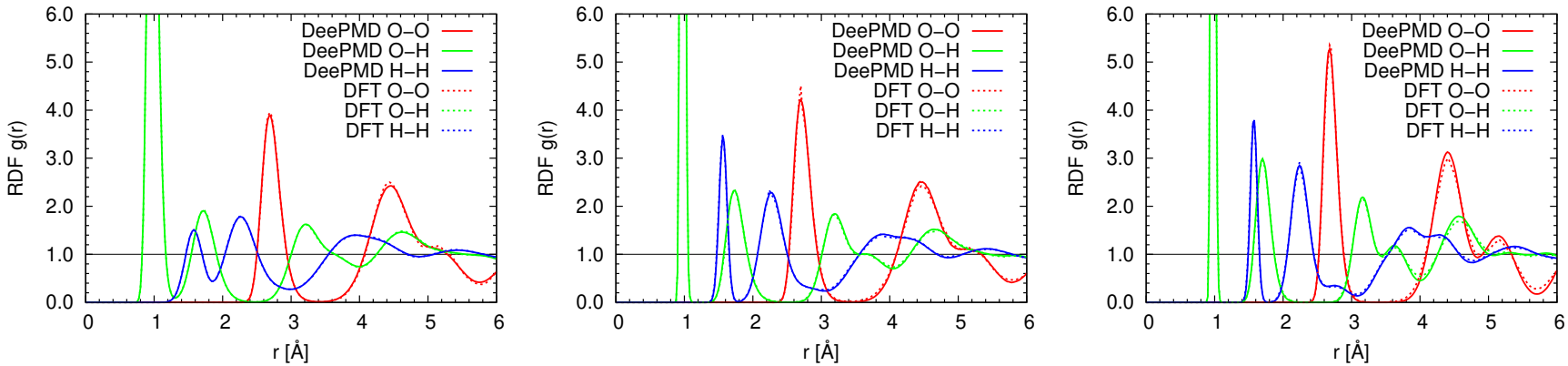


Water at different thermodynamic conditions

- The path integral water structures (ambient cond.)

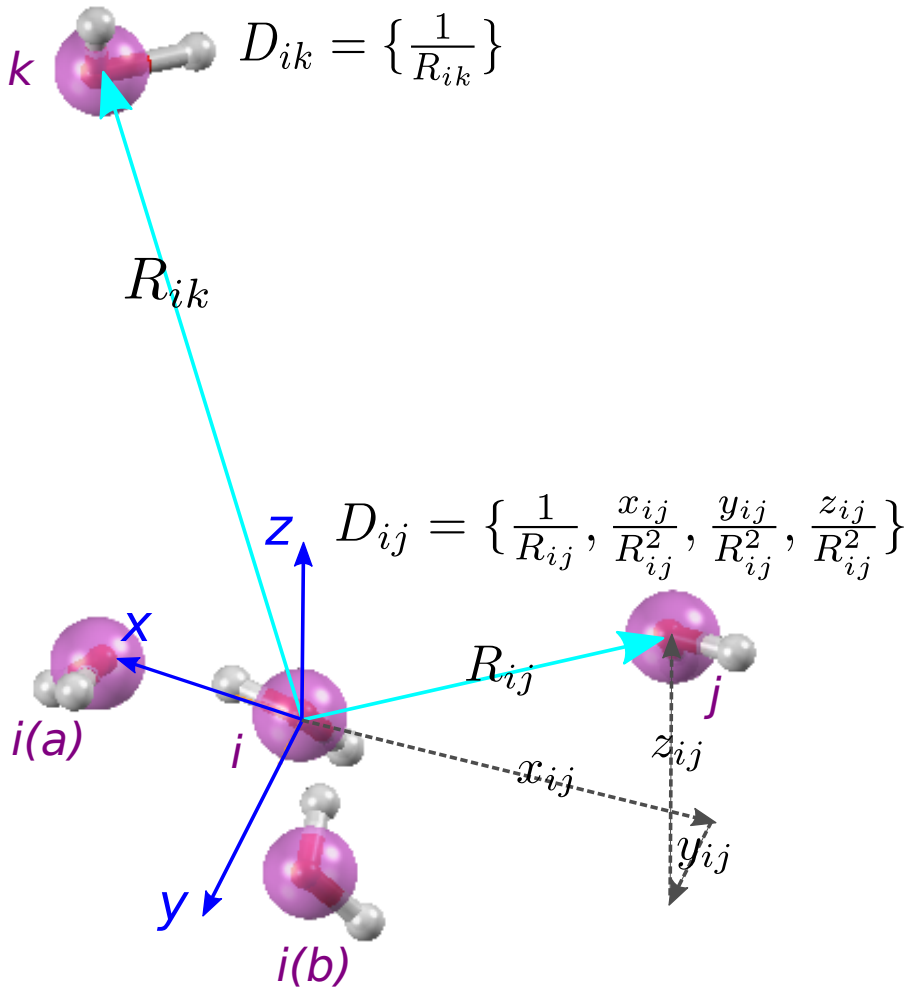


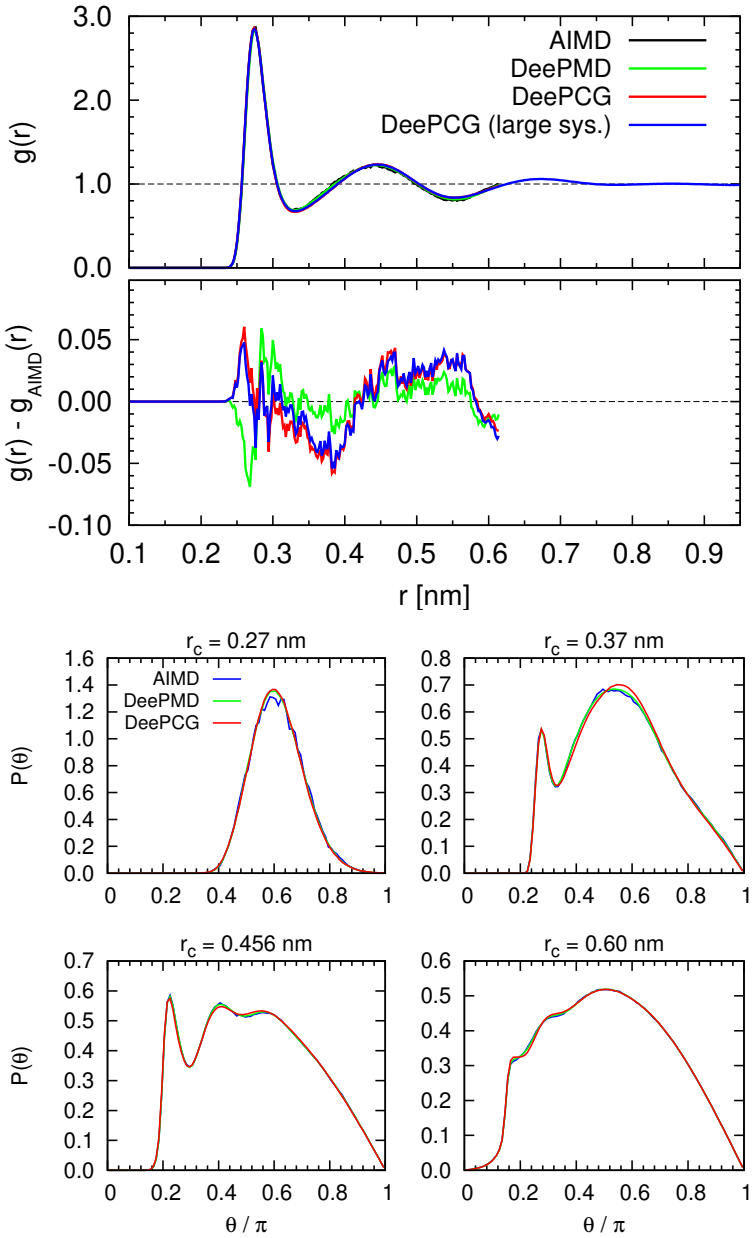
- Ice in different thermodynamic states



PI-ice, P=1.0 bar, T=273 K; ice P=1.0 bar,T=330 K; ice P=2.13 bar,T=238 K.

DeePCG for coarse-graining

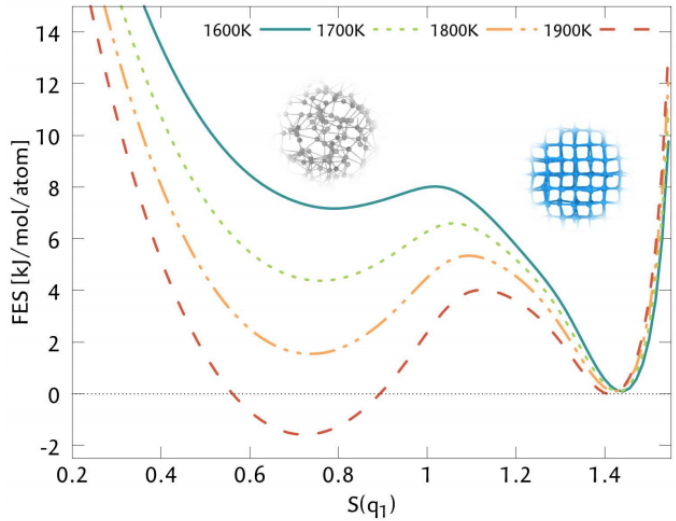


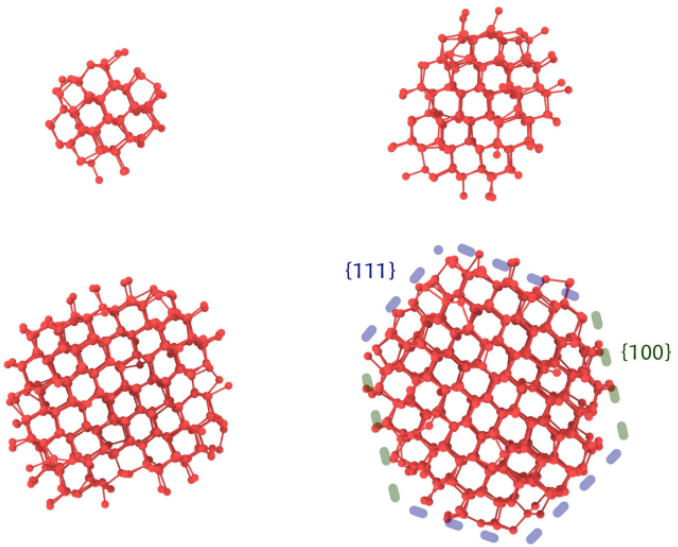


L. Zhang, J. Han, H. Wang, R. Car, W. E, J. Chem. Phys., 149, 034101 (2018)

Extention to other applications

Rare events:





L. Bonati and M. Parrinello, Phys. Rev. Lett. 121, 265701

Nonadiabatic excited-state dynamics:

THE JOURNAL OF PHYSICAL CHEMISTRY Letters
Letter
Cite This: J. Phys. Chem. Lett. 2018, 9, 6702−6708
pubs.acs.org/JPCL

Deep Learning for Nonadiabatic Excited-State Dynamics

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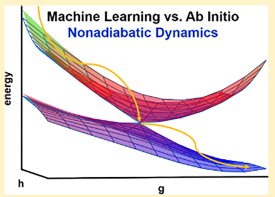
Supporting Information

ABSTRACT: In this work we show that deep learning (DL) can be used for exploring complex and highly nonlinear multistate potential energy surfaces of polyatomic molecules and related nonadiabatic dynamics. Our DL is based on deep neural networks (DNNs), which are used as accurate representations of the CASSCF ground- and excited-state potential energy surfaces (PESs) of CH_2NH. After geometries near conical intersection are included in the training set, the DNN models accurately reproduce excited-state topological structures; photoisomerization paths; and, importantly, conical intersections. We have also demonstrated that the results from nonadiabatic dynamics run with the DNN models are very close to those from the dynamics run with the pure ab initio method. The present work should encourage further studies of using machine learning methods to explore excited-state potential energy surfaces and nonadiabatic dynamics of polyatomic molecules.



Chen, Wen-Kai, et al. J. P. C. Lett. 9.23 (2018): 6702-6708.

Irradiation damage simulations:

Deep learning inter-atomic potential model for accurate irradiation damage simulations[a)]

Hao Wang,[1] Xun Guo,[1] Linfeng Zhang,[2] Han Wang,[3, b)] and Jianming Xue[4, c)]

[1)]*State Key Laboratory of Nuclear Physics and Technology, School of Physics, CAPT, HEDPS, and IFSA Collaborative Innovation Center of MoE College of Engineering, Peking University, Beijing 100871, P. R. China;*

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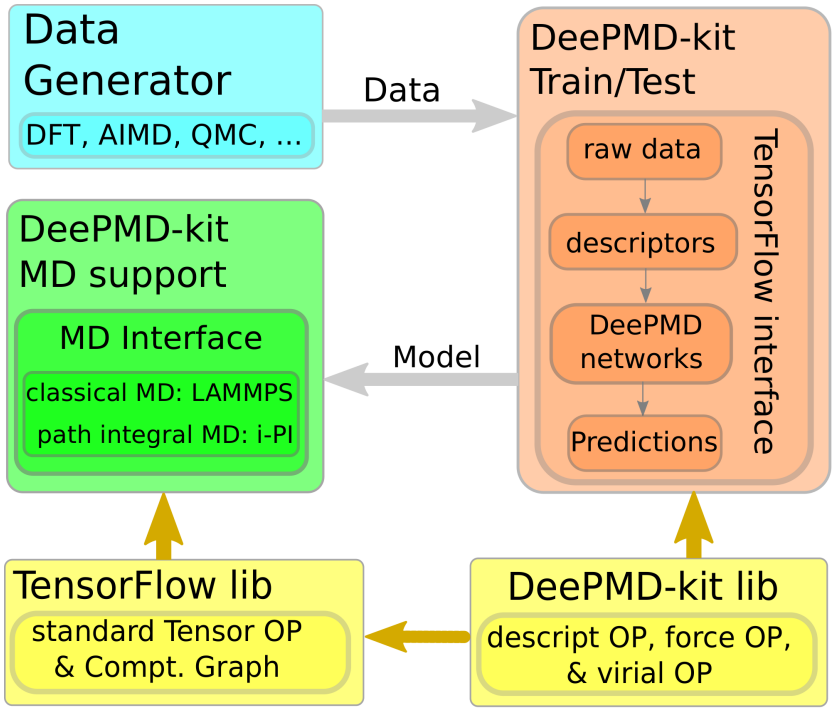
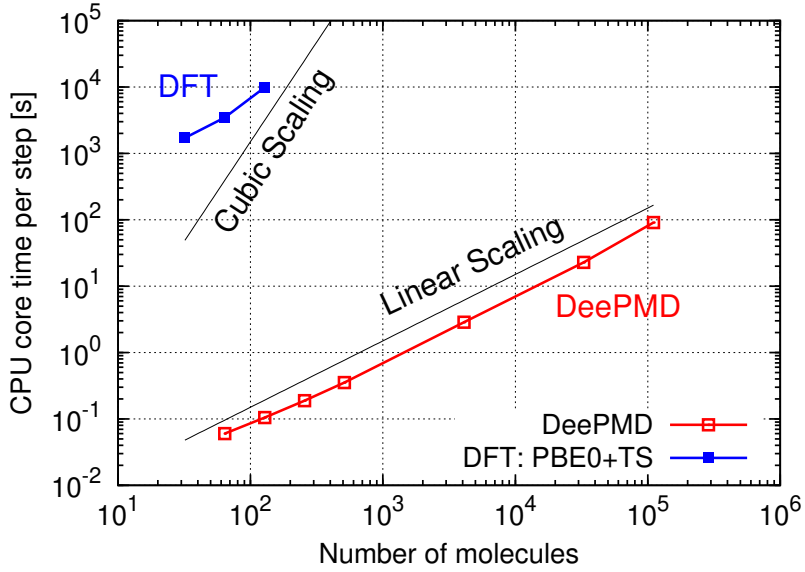
[4)]*State Key Laboratory of Nuclear Physics and Technology, School of Physics, CAPT, HEDPS, and IFSA Collaborative Innovation Center of MoE College of Engineering, Peking University, Beijing 100871, P. R. China.*

(Dated: 3 April 2019)

arxiv.org/abs/1904.00360

Open source software DeePMD-kit





- TensorFlow: efficient network operators
- LAMMPS, i-PI; MPI/GPU support.

Free download from https://github.com/deepmodeling/deepmd-kit

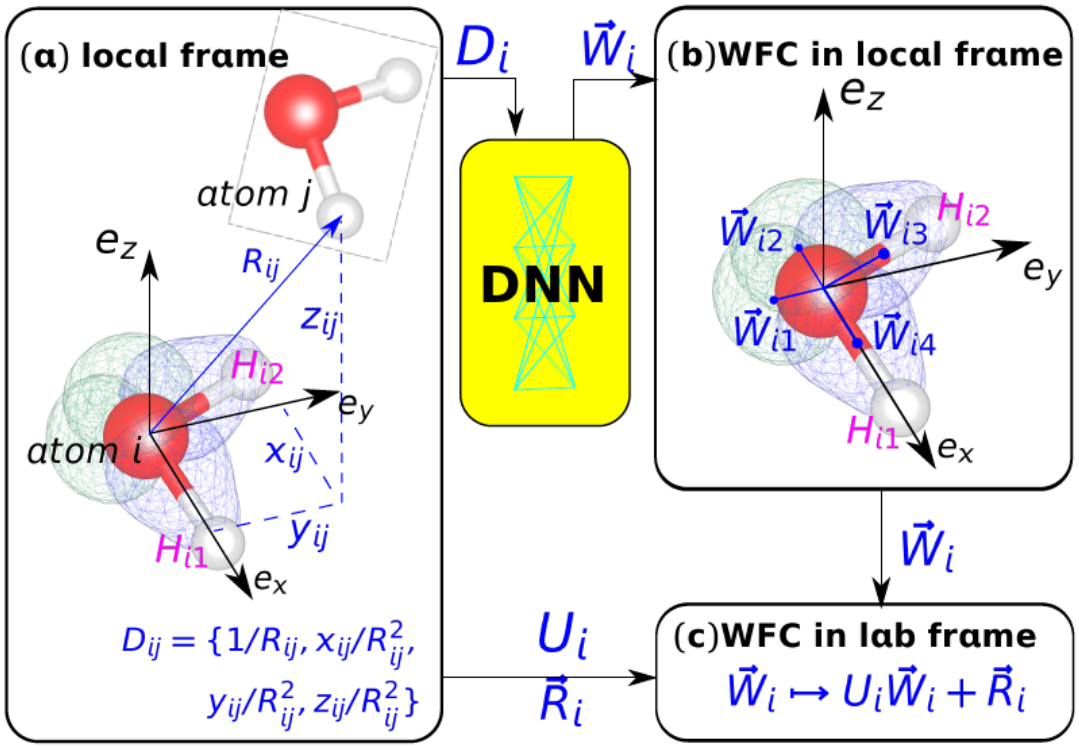
Comp.Phys.Comm., 0010-4655 (2018).

Modeling the electronic information

- Many observables depend also on electronic structure information that is accessible in AIMD simulations;
- Examples are the electronic charge density, the cell dipole, the atomic polarizability, etc.;
- In AIMD, these quantities derive consistently from the quantum mechanical ground state of the electrons within DFT.

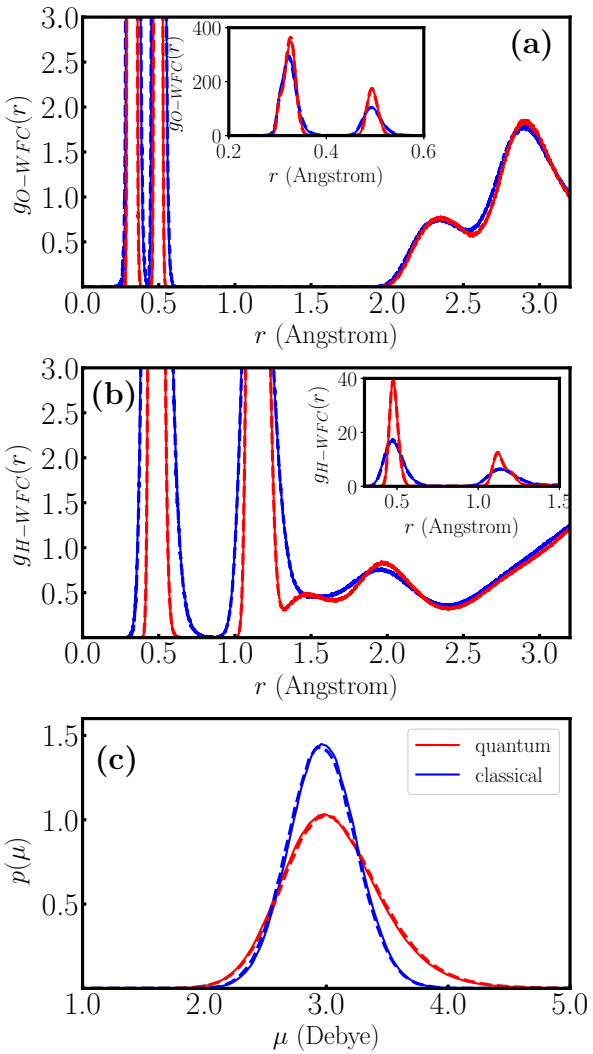
Modeling the maximally localized Wannier Functions

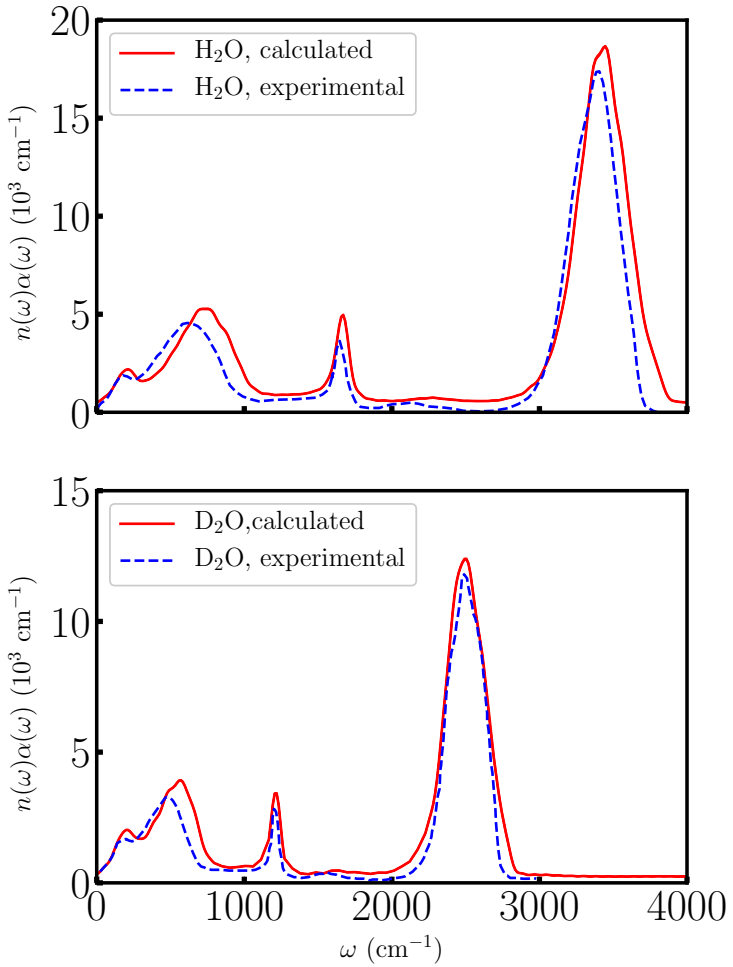
$$\{\vec{W}_n(\boldsymbol{r})\} = \min_{U_{\boldsymbol{k}}} \Omega[U_{\boldsymbol{k}}] = \min_{U_{\boldsymbol{k}}} \sum_n (\langle r^2 \rangle_n - \langle \bar{\boldsymbol{r}}^2 \rangle_n)$$



Nicola Marzari and David Vanderbilt. Phys. Rev. B 56.20 (1997): 12847.

Configuration distribution and IR spectrum



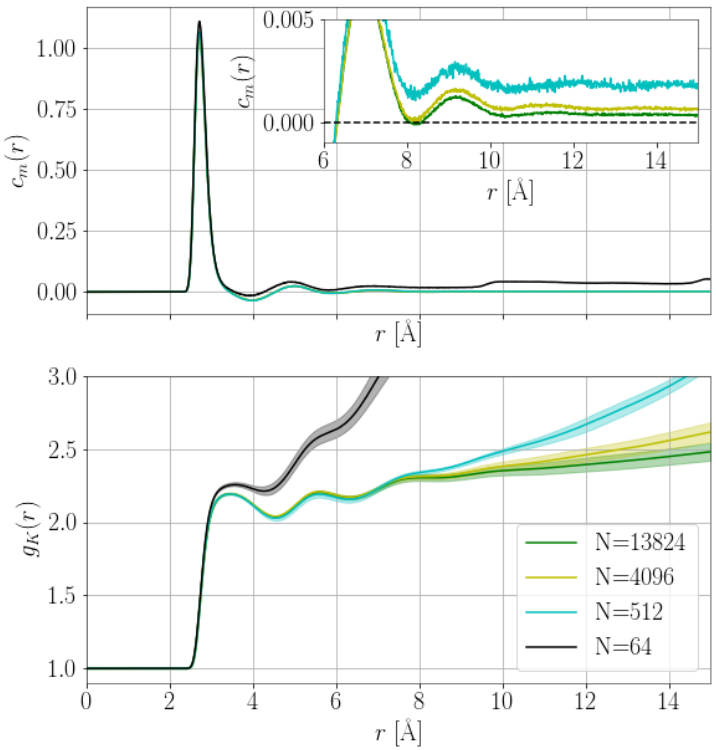


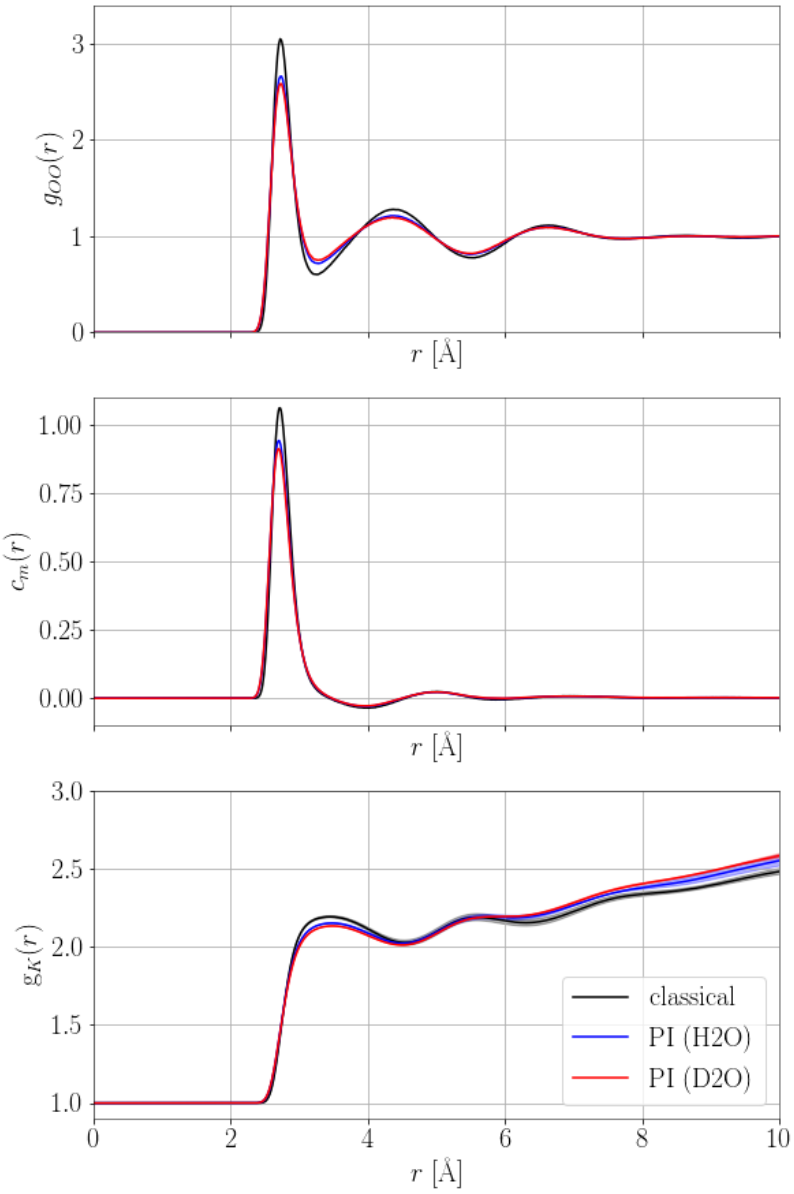
Left: Pair coorelation functions of ionic and MWLF center positions and disctribution of magnitude of molecular dipole.

Right: Infra-red spectrum calculated from 500 ps microcanonical DPMD simulation of 512 water molecules at $T = 300K$.

L. Zhang, et al., in preparation.

Static dielectric property

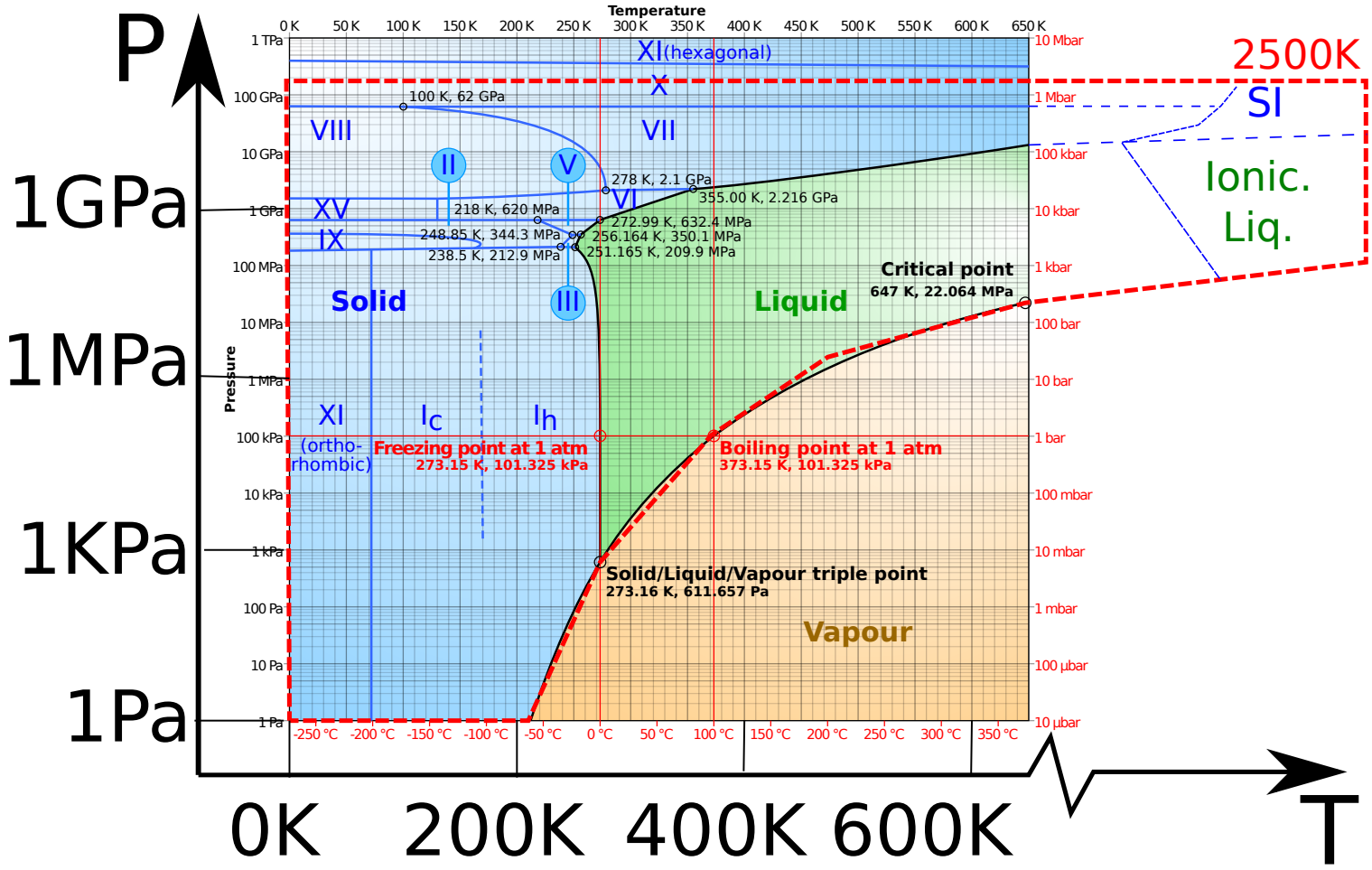




ϵ_{H_2O}: 78.57 (Exp.: 78.39)

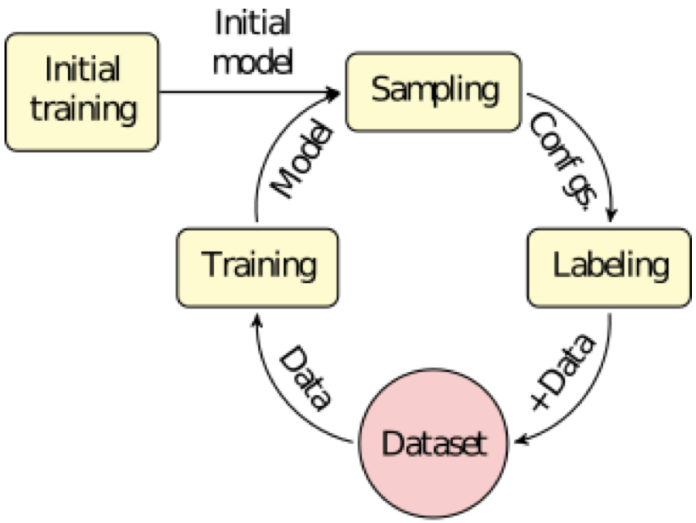
ϵ_{D_2O}: 77.56 (Exp.: 78.06)

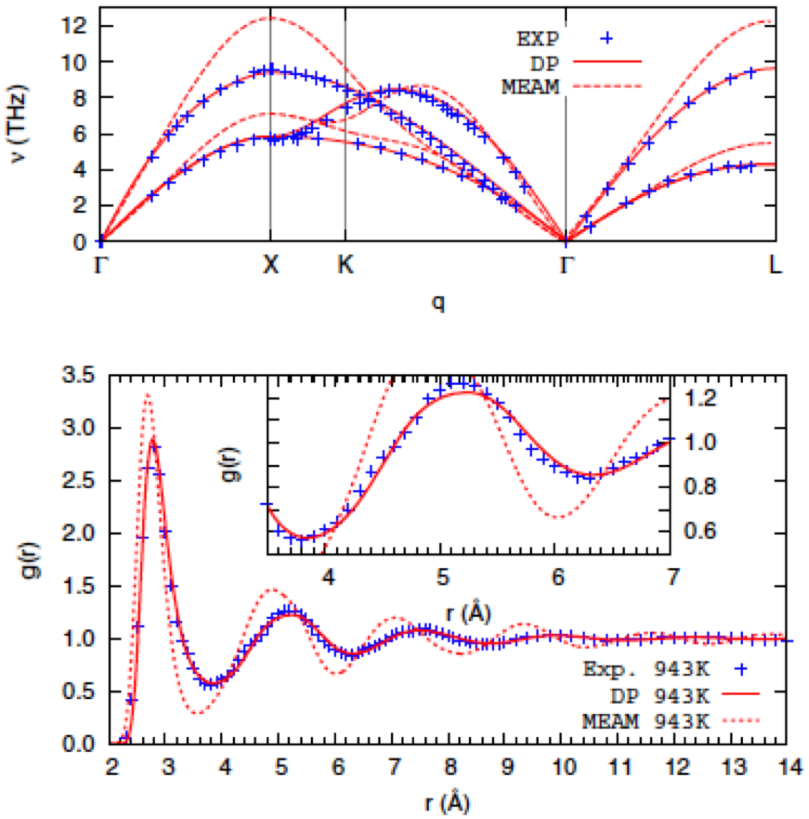
Can we model water on the whole phase diagram by extending AIMD with DPMD?



Experimental results were collected by Martin Chaplin, et al.
at http://www1.lsbu.ac.uk/water/water_phase_diagram.html

Optimal incremental learning: the DP-GEN scheme



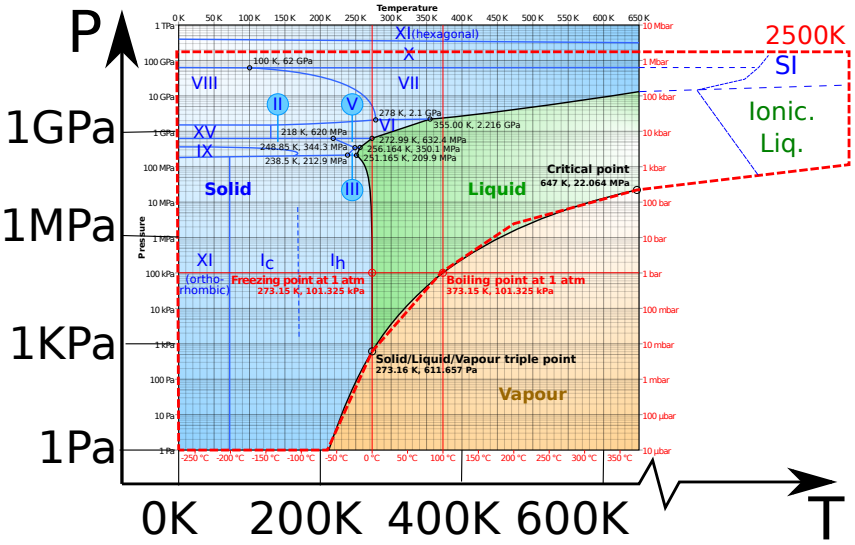


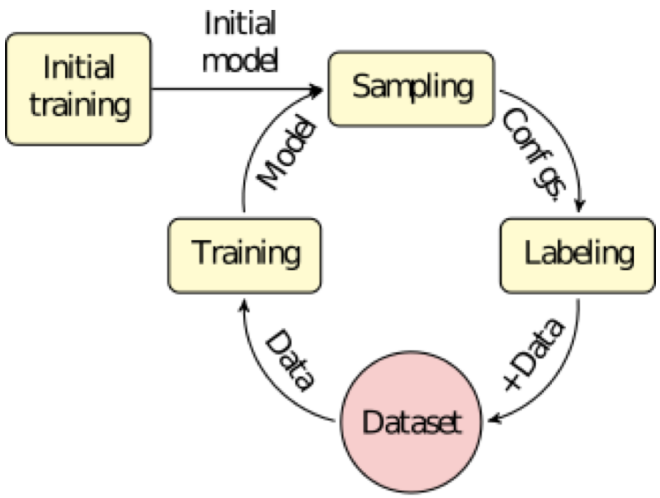
Labeling when model deviation is large: $\epsilon = \max_i \sqrt{\langle \| \boldsymbol{f}_i - \langle \boldsymbol{f}_i \rangle \|^2 \rangle}$.

See, e.g. principle of maximal disagreement in "Query by Committee" by Seung, Opper, Sompolinsky (1992).

L. Zhang, D. Lin, H. Wang, R. Car, W. E, Phys. Rev. Mat. 3, 023804 (2019)

DP-GEN for water





- **Reference model:** DFT at the classical SCAN level;
- **Starting configurations:** relaxed Ice I-XV at $T = 0$ K and equilibrated liquid at $T = 330$ K;
- **Range of thermodynamic conditions:** red dashed box;
- **number of MD snapshots:** DPMD exploration: 1.4 billion, DFT calculation: 32 thousand ($\sim$0.002% of the former).

  Typical AIMD trajectory: 100 thousand snapshots (50-100 *ps*).
- **number of DP-GEN iterations:** 100.

Thermodynamic integration (TI) for the phase diagram

Hamiltonian TI

$(H^{(ref)}, p, T) \mapsto (H, p, T)$

↓

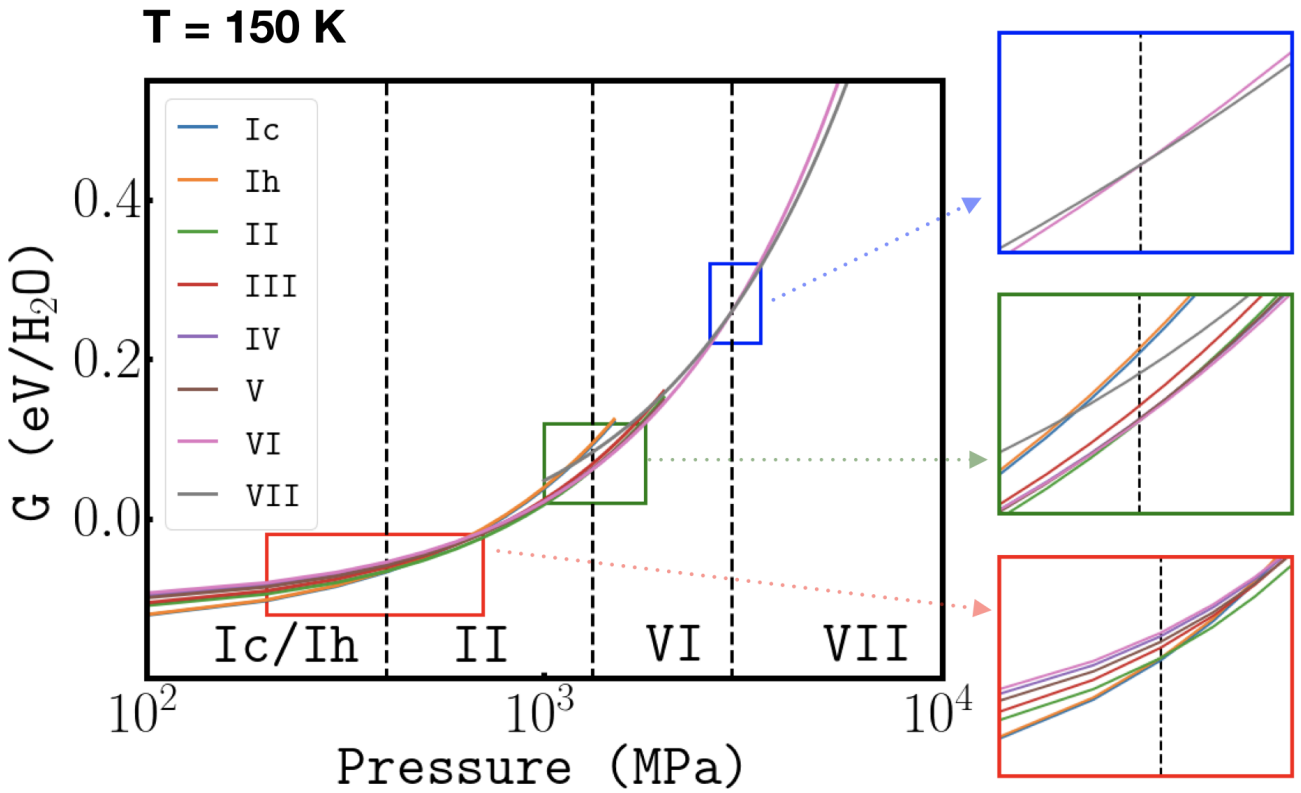
TI

$(H, p_1, T_1) \mapsto (H, p_2, T_2)$

↓

Clapeyron Eqn.

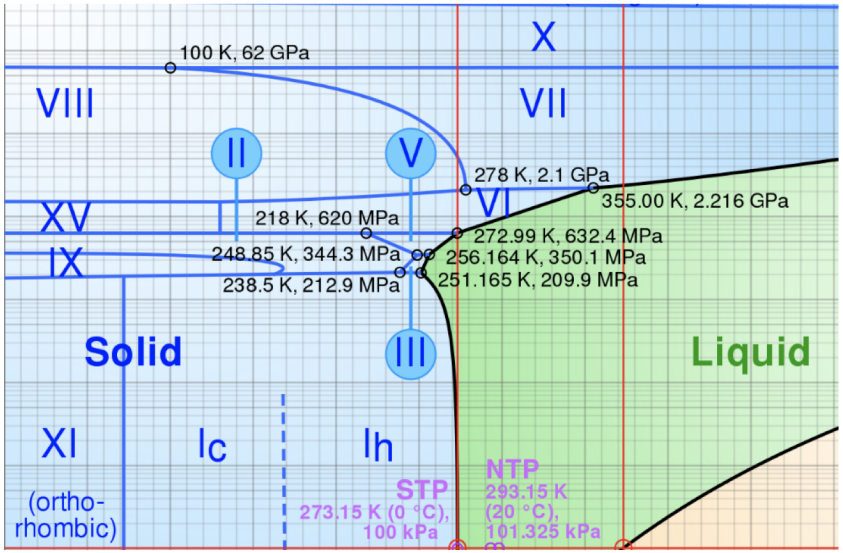
$$\frac{dp}{dT} = \frac{h_{II} - h_I}{T(v_{II} - v_I)}$$



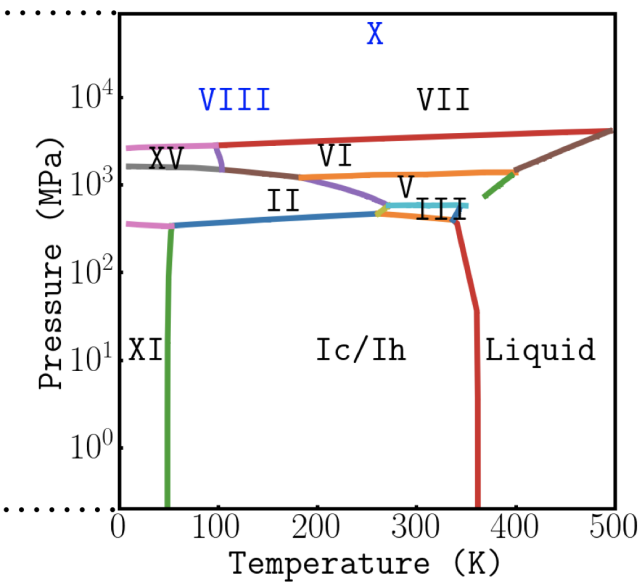
Special issues: size effect; proton disorder, etc.

Water phase diagram modeled by DP+SCAN

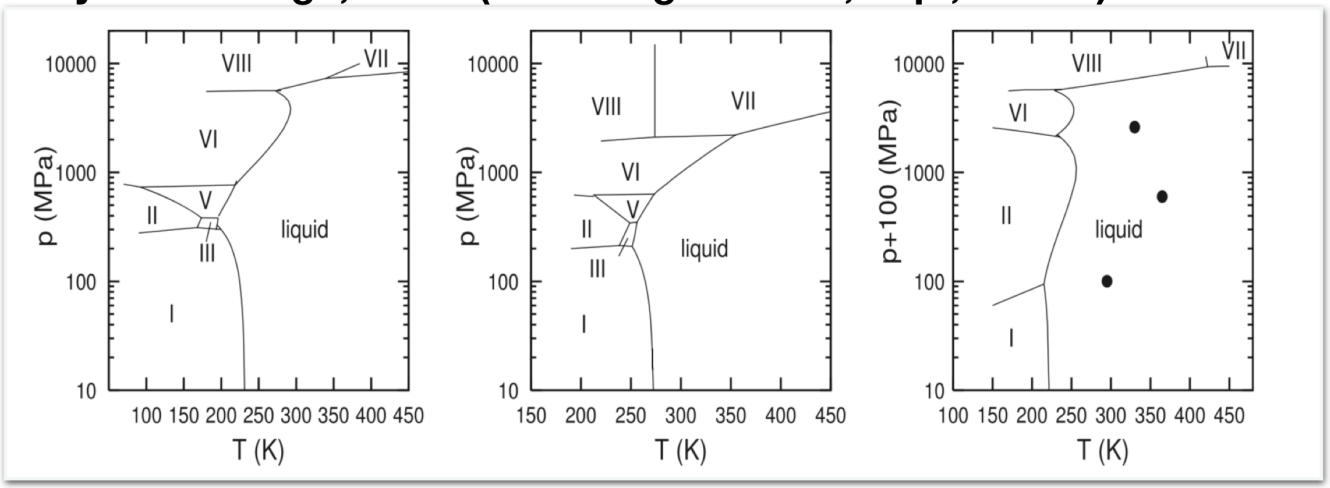
(a) Exp. collected by Martin Chaplin



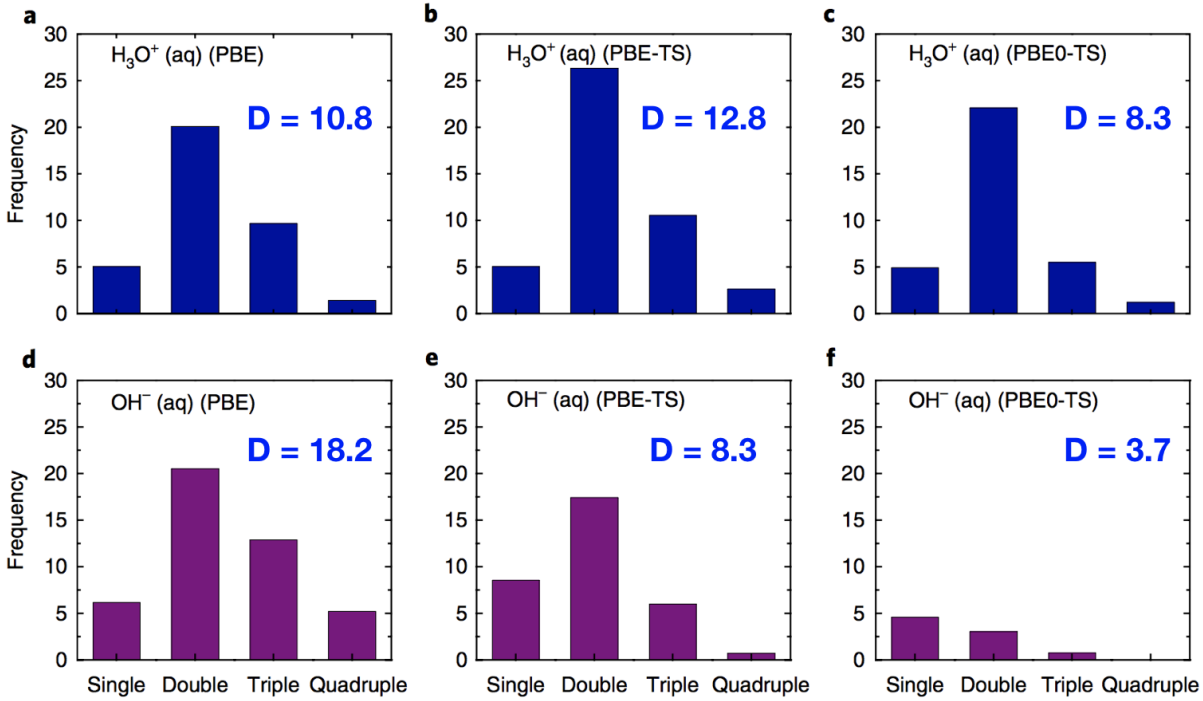
(b) current result



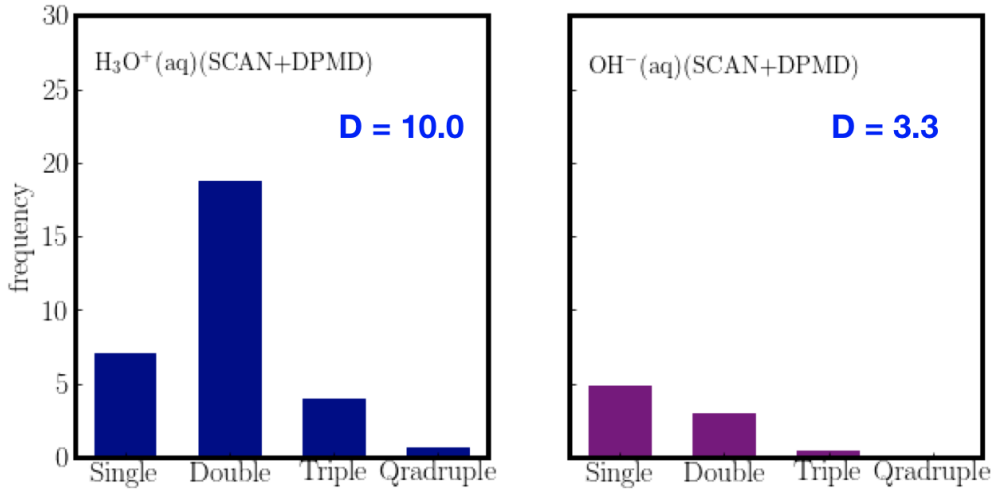
(c) Work by Carlos Vega, et al. (Left to right: TIP4P, Exp., SPC/E)



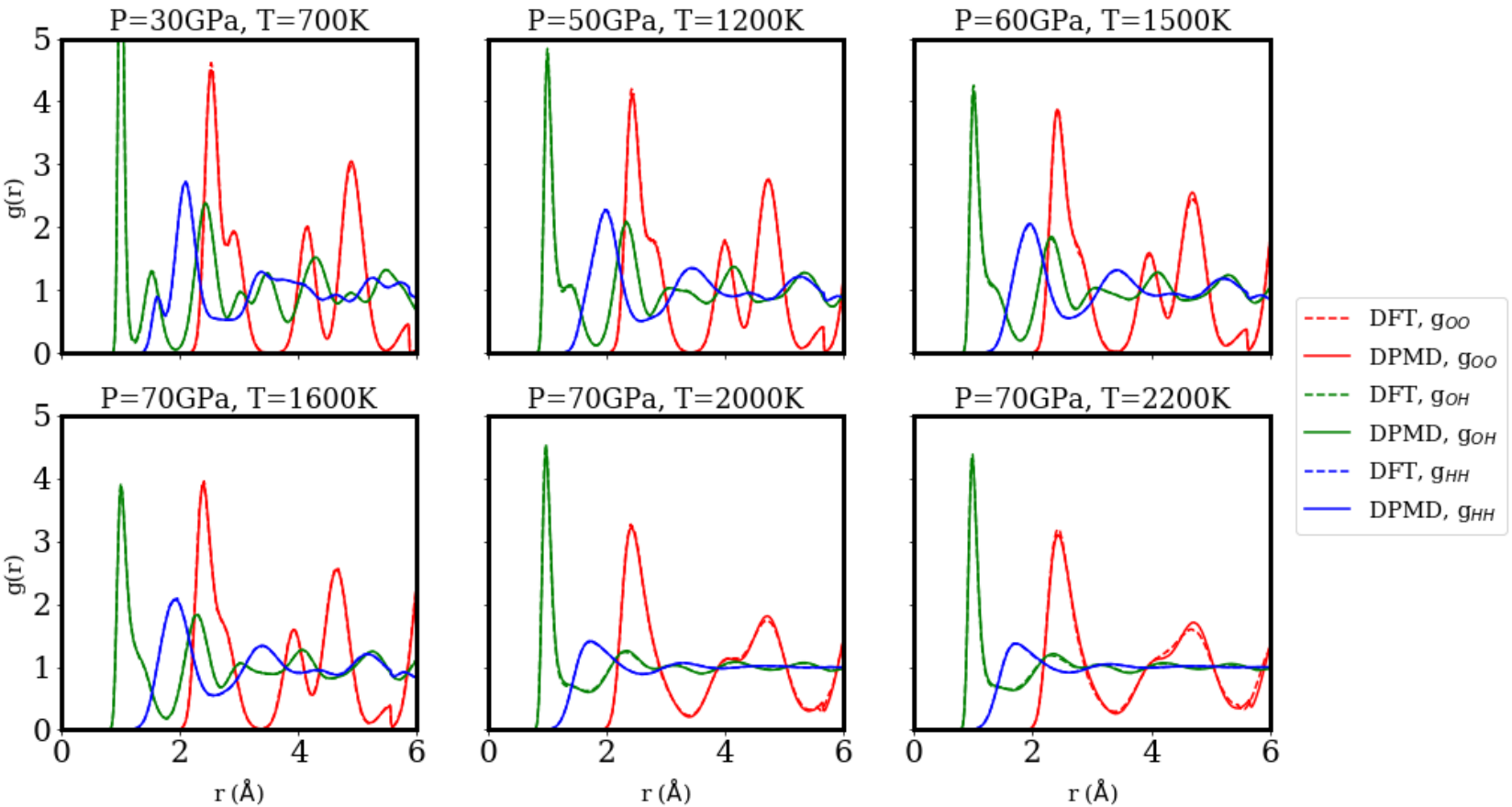
Proton transport at room temperature



(M. Chen, et al. "Hydroxide diffuses slower than hydronium in water because its solvated structure inhibits correlated proton transfer." *Nature chemistry* (2018): 1.)

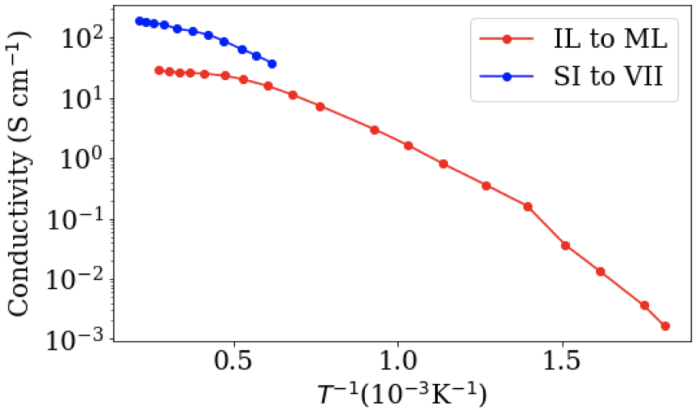


High-pressure phases modeled by DP+SCAN

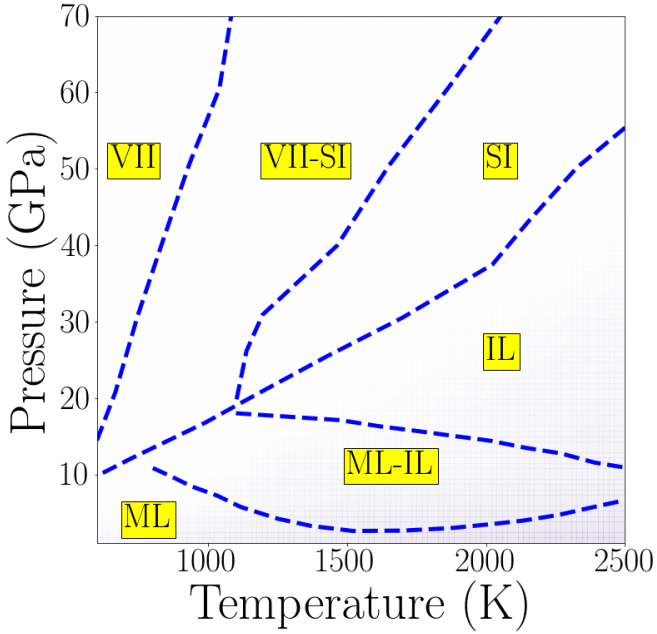


High-pressure phases modeled by DP+SCAN

Exp. by Marius, et. al

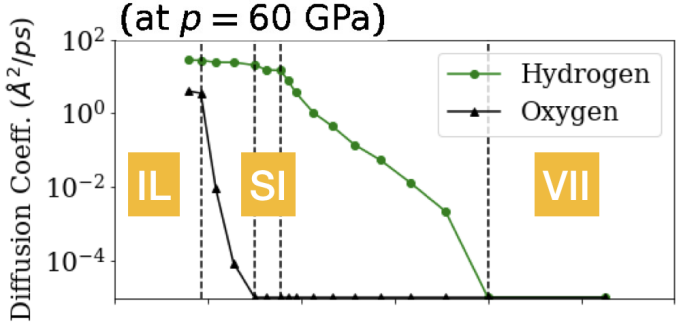


("Experimental evidence for superionic water ice using shock compression", Nature Physics 14, 297–302 (2018))

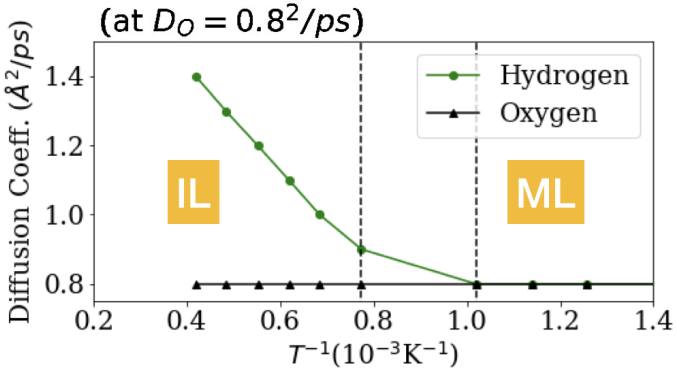


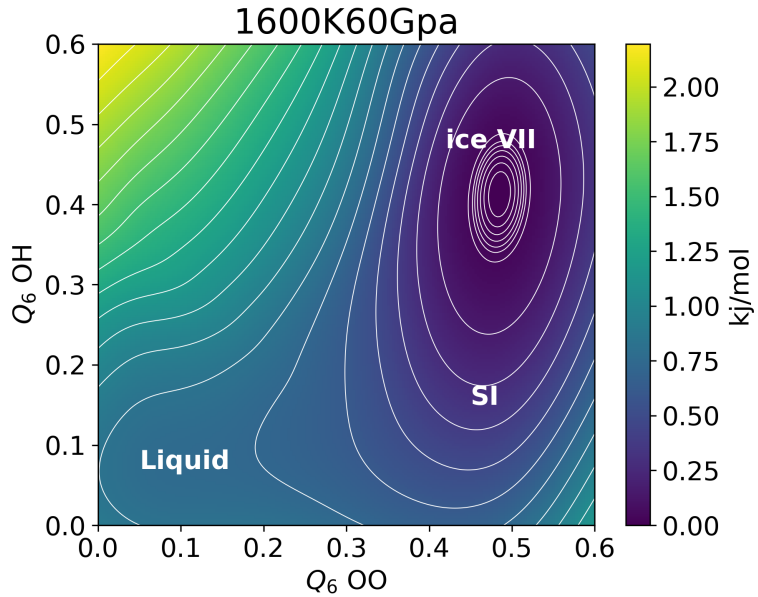
DPMD simulation:

(at $p = 60$ GPa)



(at $D_O = 0.8^2/ps$)





High-pressure phases modeled by DP+SCAN

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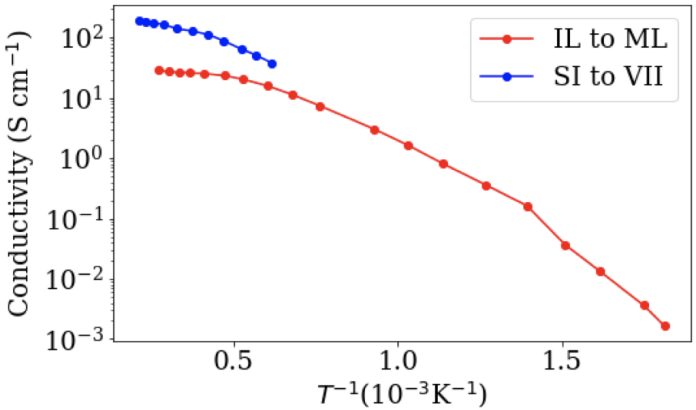
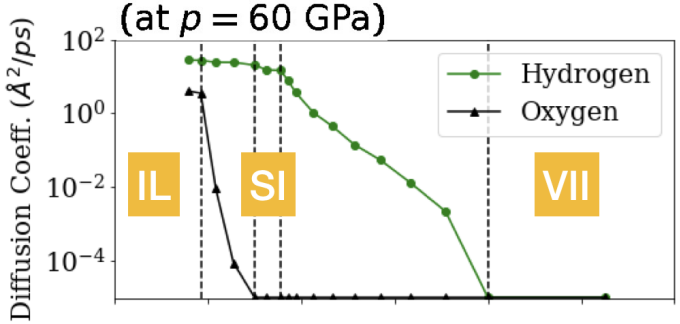


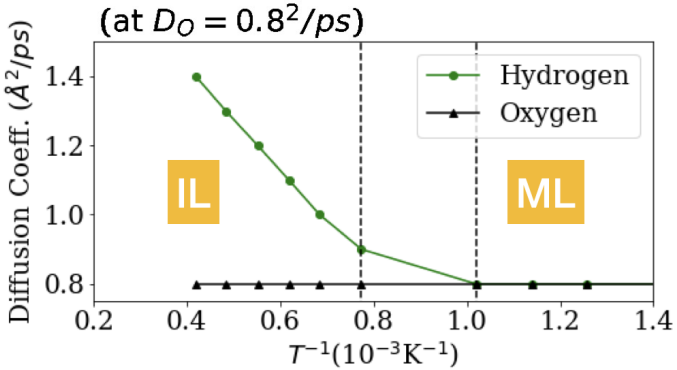
("Experimental evidence for superionic water ice using shock compression", Nature Physics 14, 297–302 (2018))

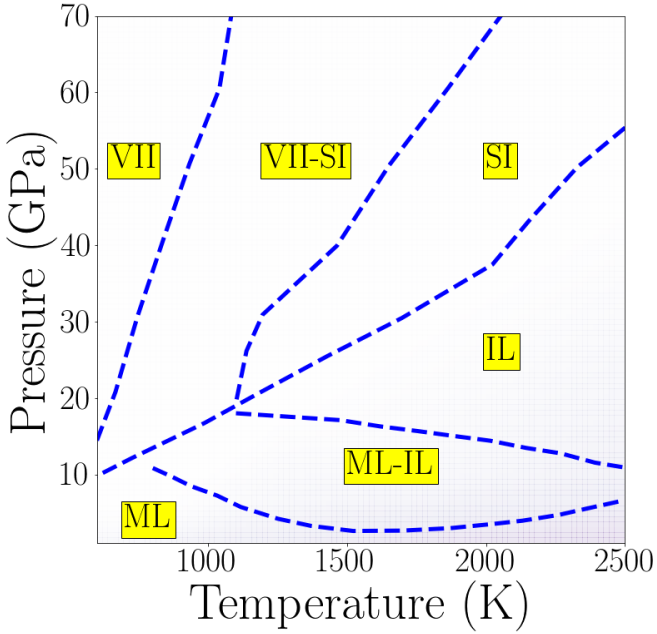
DPMD simulation:

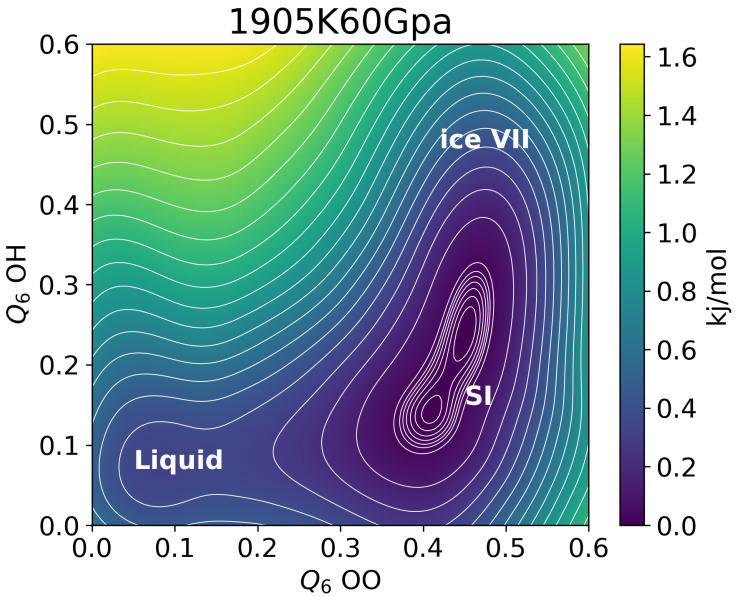
(at $p = 60$ GPa)



(at $D_O = 0.8^2/ps$)







High-pressure phases modeled by DP+SCAN

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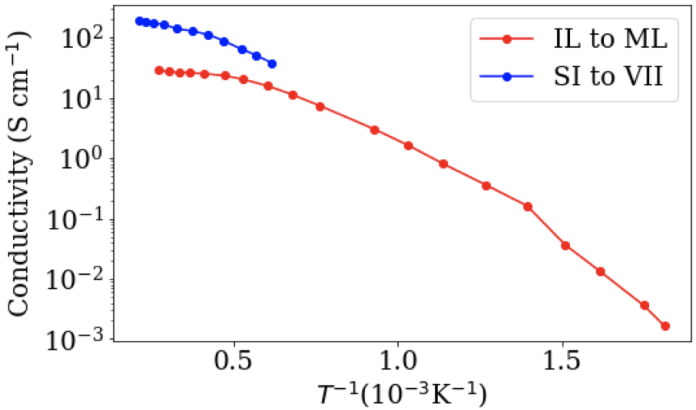
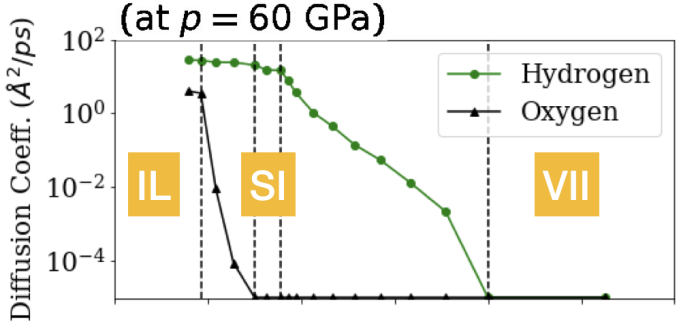


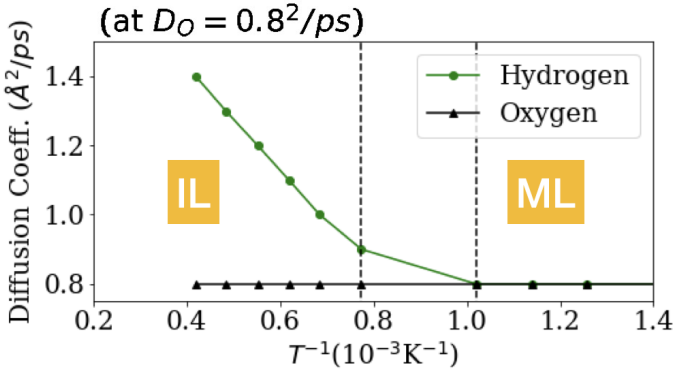
("Experimental evidence for superionic water ice using shock compression", Nature Physics 14, 297–302 (2018))

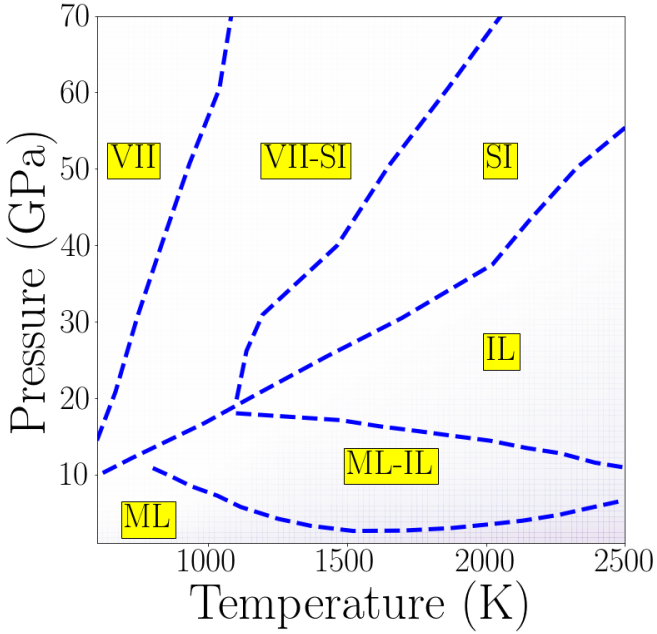
DPMD simulation:

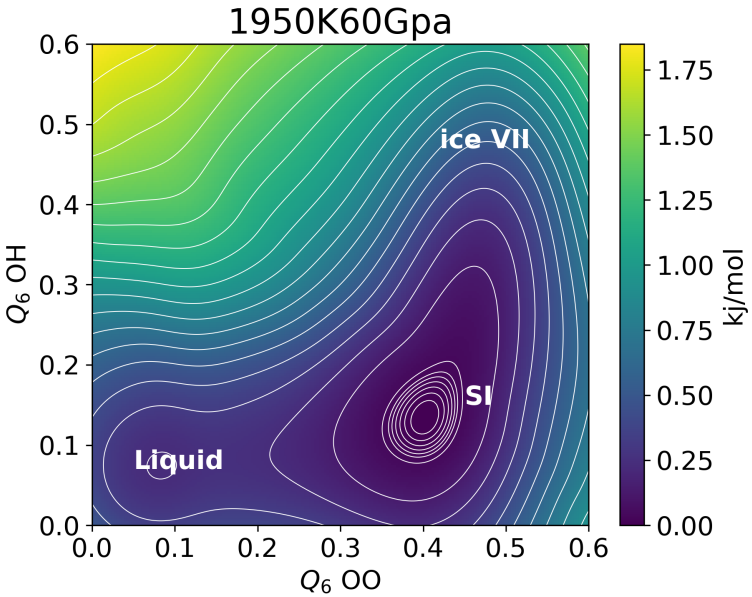
(at $p = 60$ GPa)



(at $D_O = 0.8^2/ps$)







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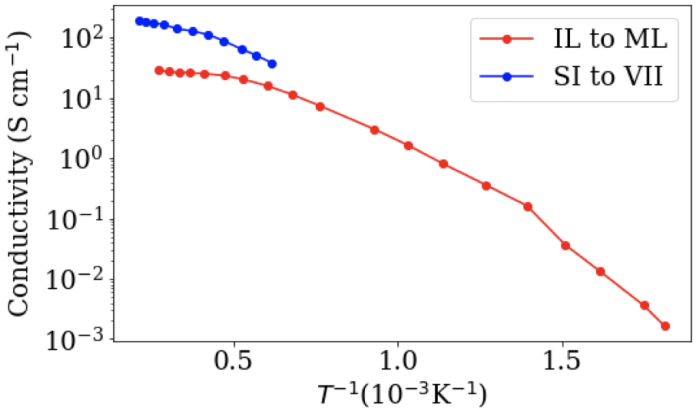

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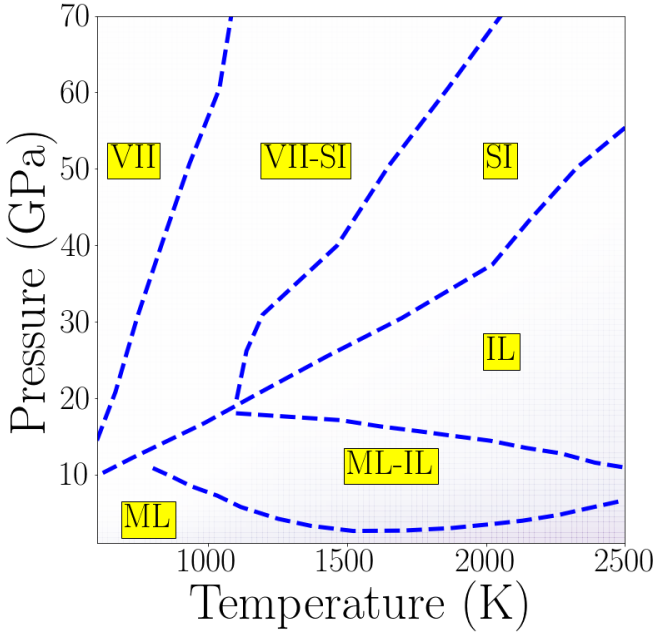

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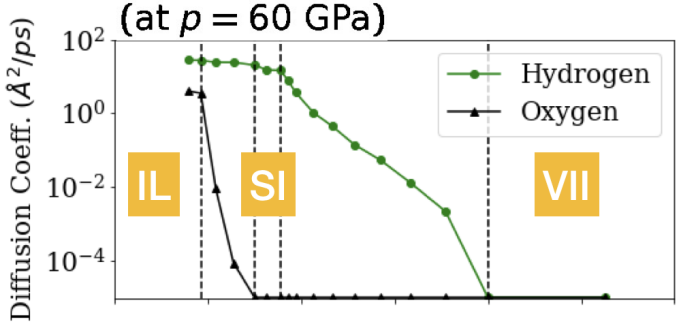
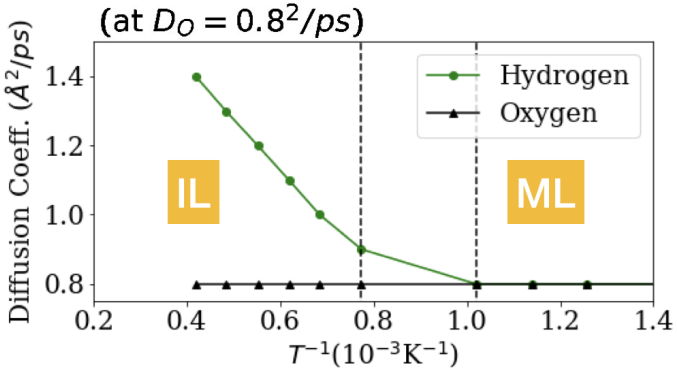
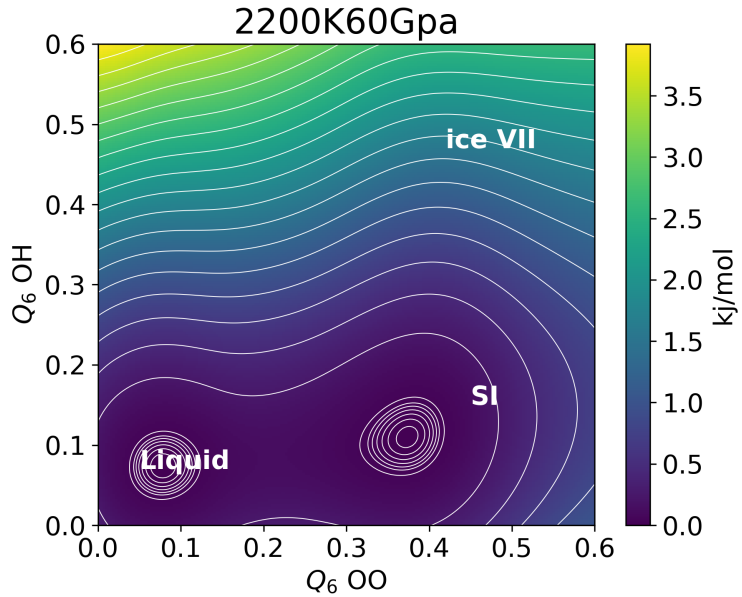


(at $D_O = 0.8^2/ps$)





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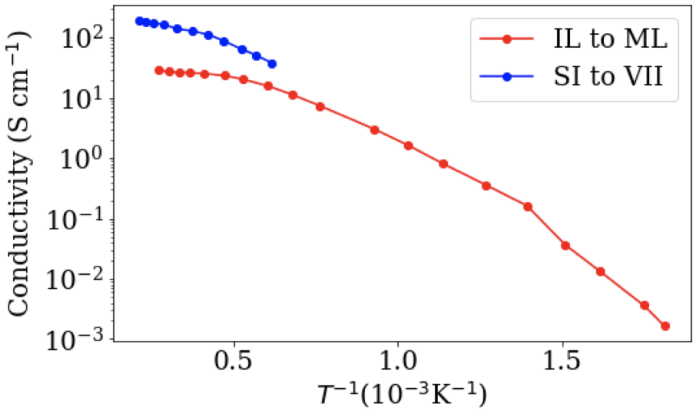

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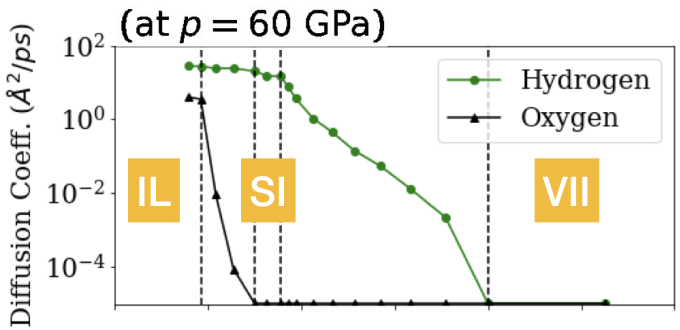

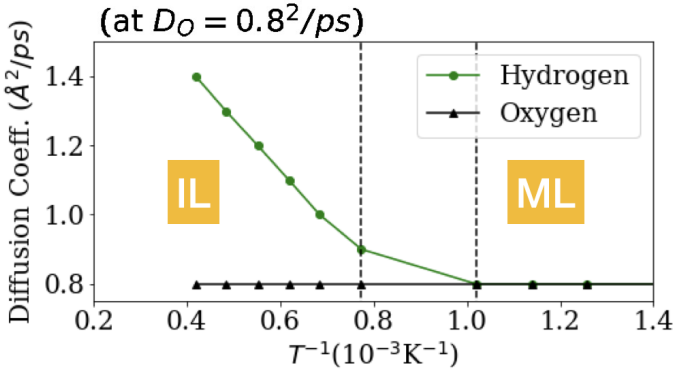

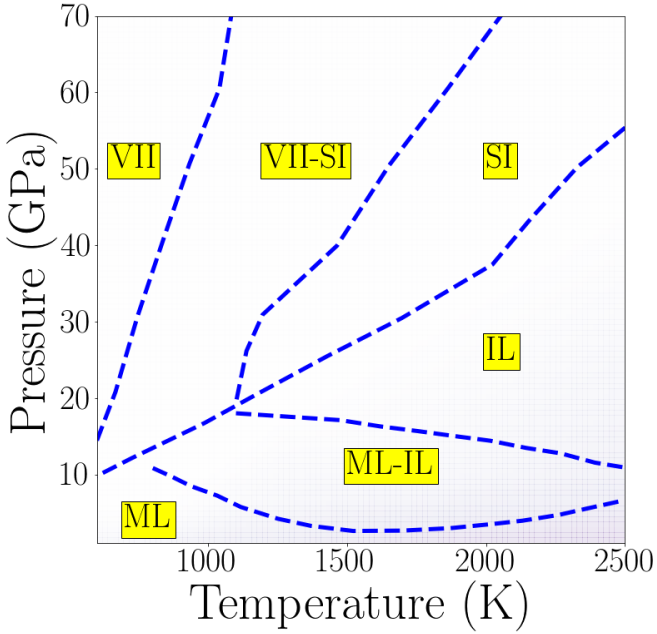

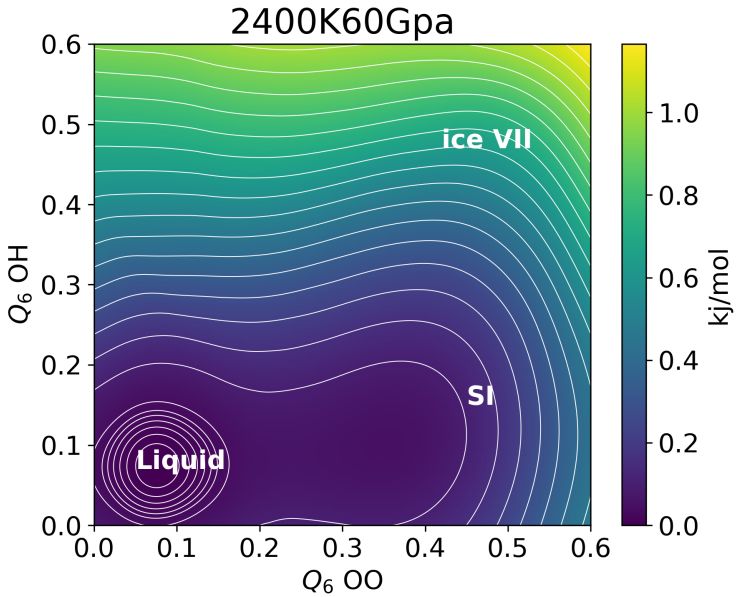

Conclusions

- Multiscale molecule modeling via deep learning: model and data;
- DPMD extends the AIMD accuracy to much larger size and time scales, opening the way to studies that would be otherwise impossible;
- Issues for further study: accuracy of the PES/data quality, long-range interaction...

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- Mohan Chen, Xinzheng Li, Yuzhi Zhang (Peking Univ.)
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