Deep learning for spectroscopy

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CSC Spring School in Quantum Chemistry 2024
19 April 2024

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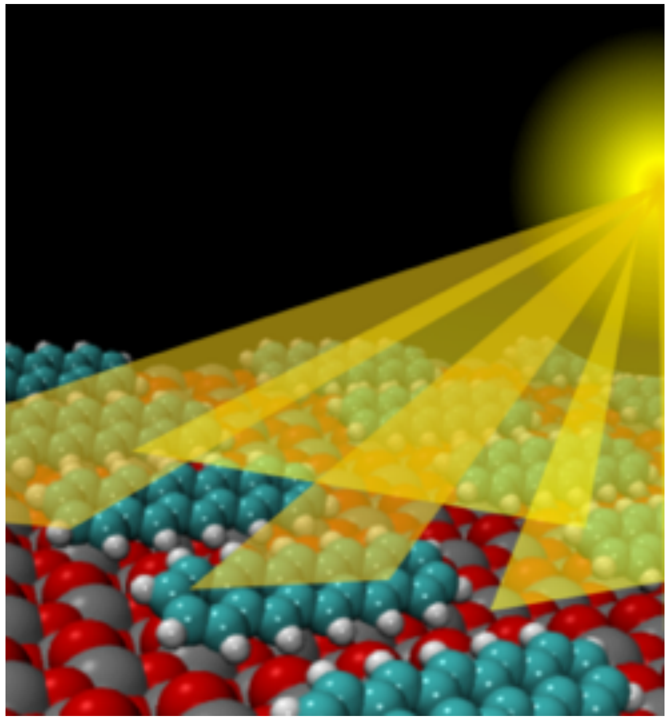
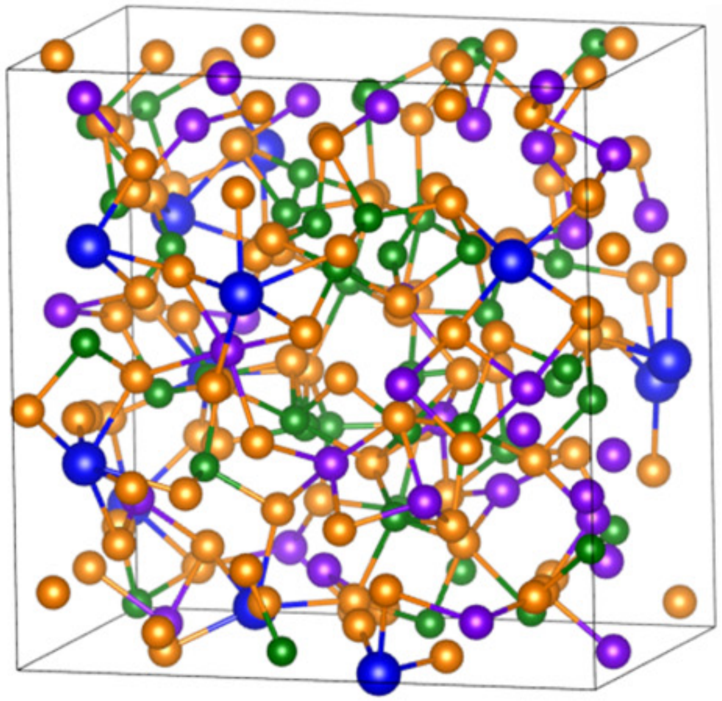
Optimising functional materials

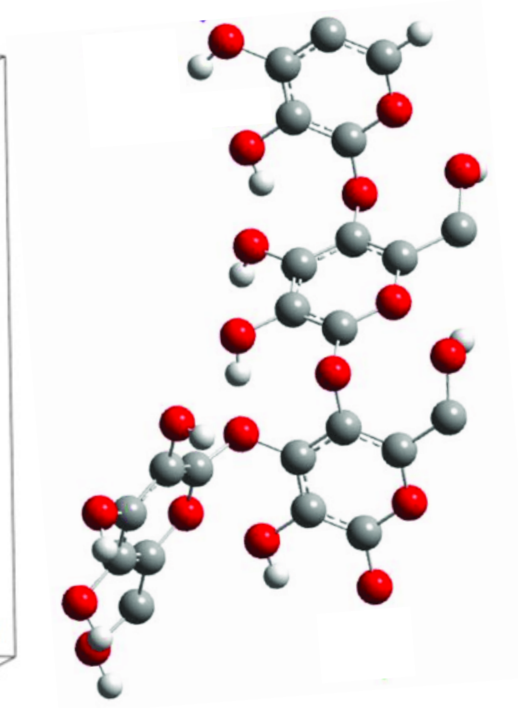
organic optoelectronics



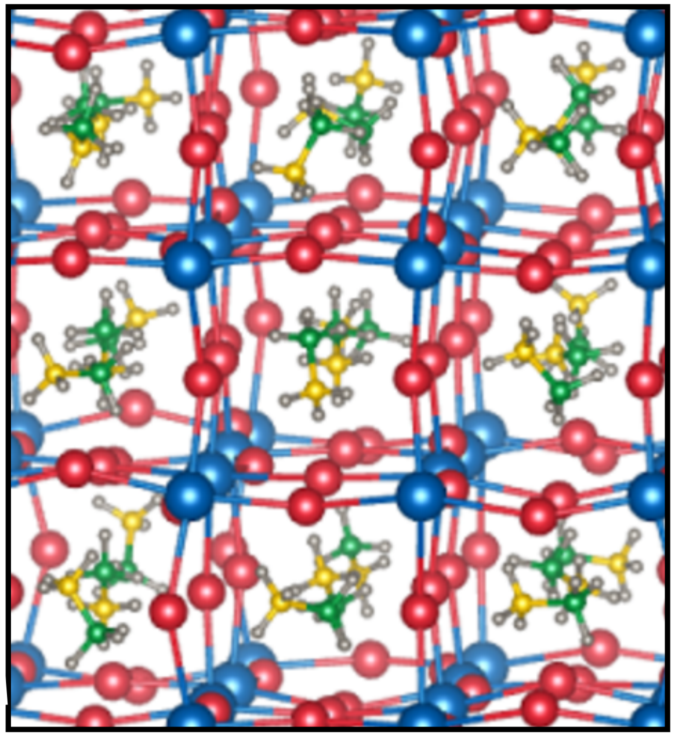
phase change memory



biofuels



organic/inorganic solar cells



Computational materials science science seeks to refine materials and optimize technologies

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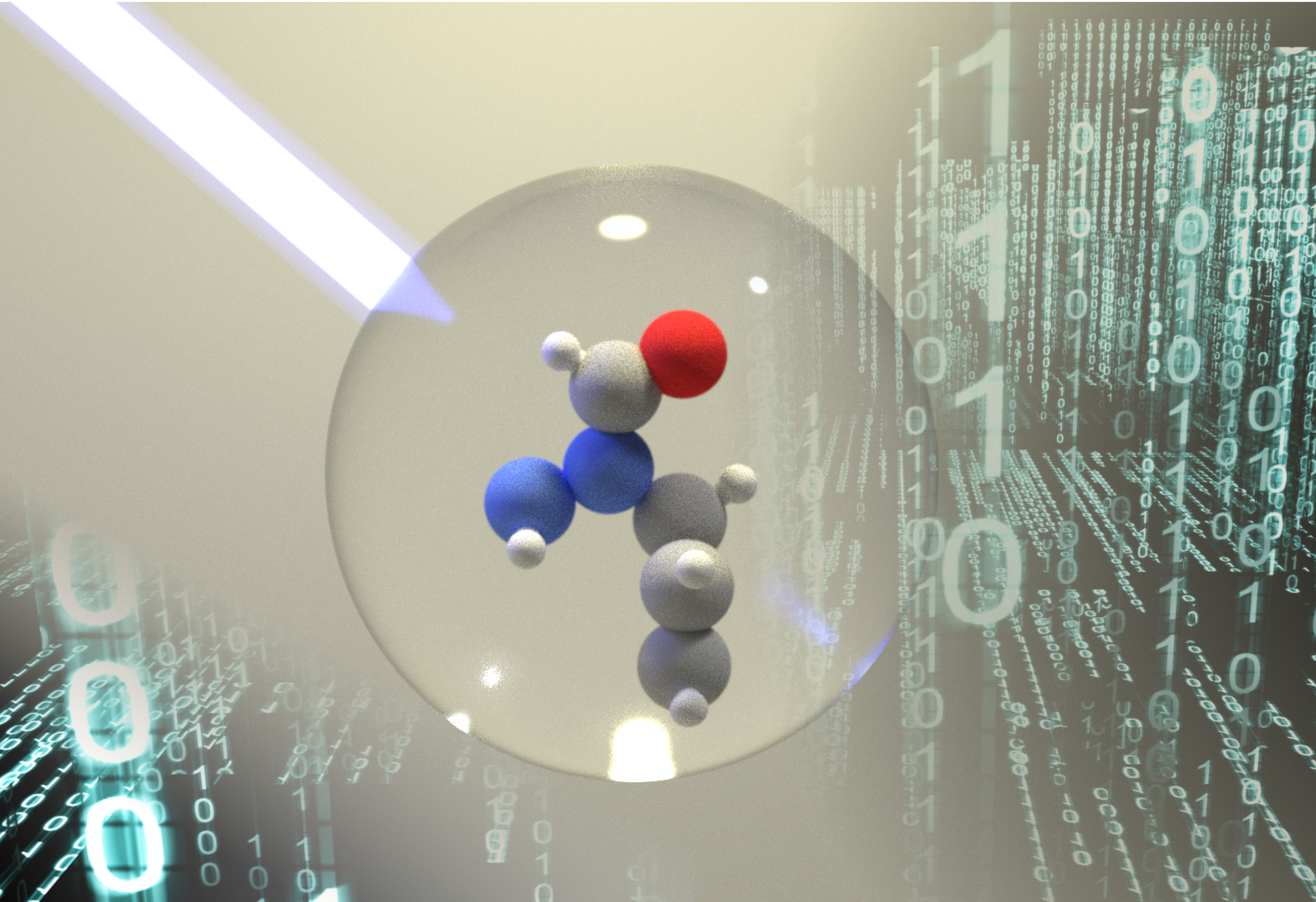
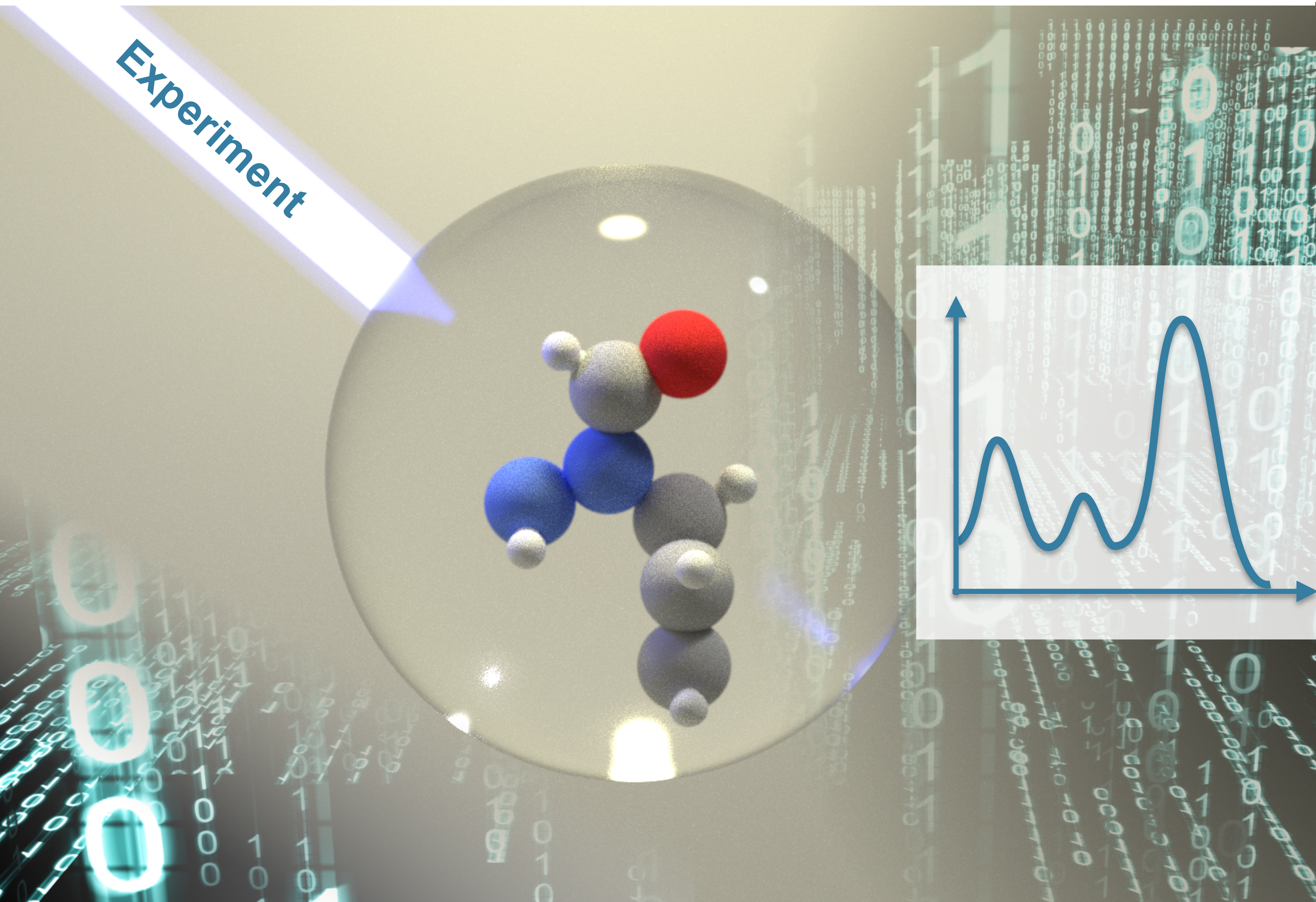
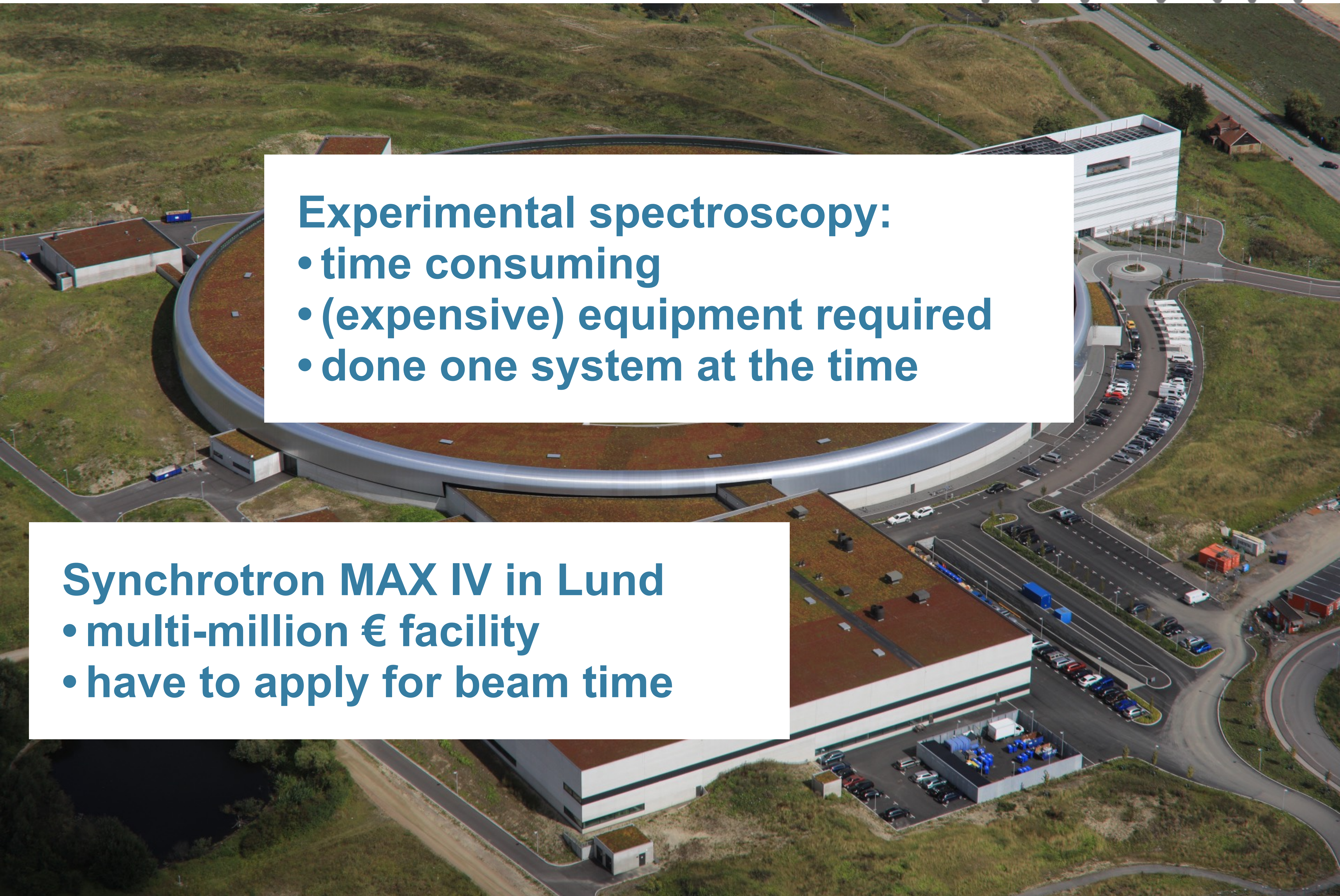
Key information from spectroscopy



Experimental spectroscopy



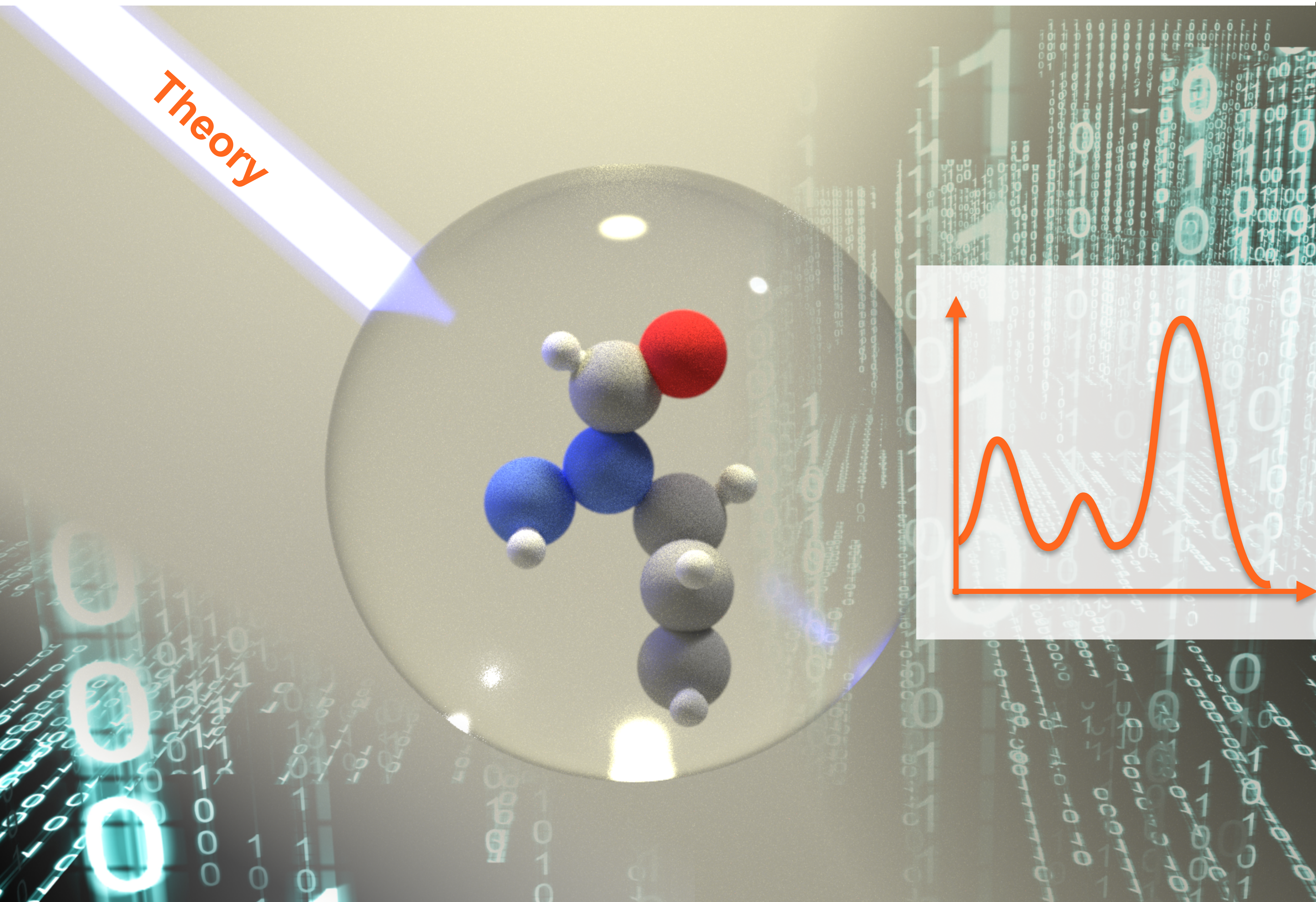
Experimental spectra



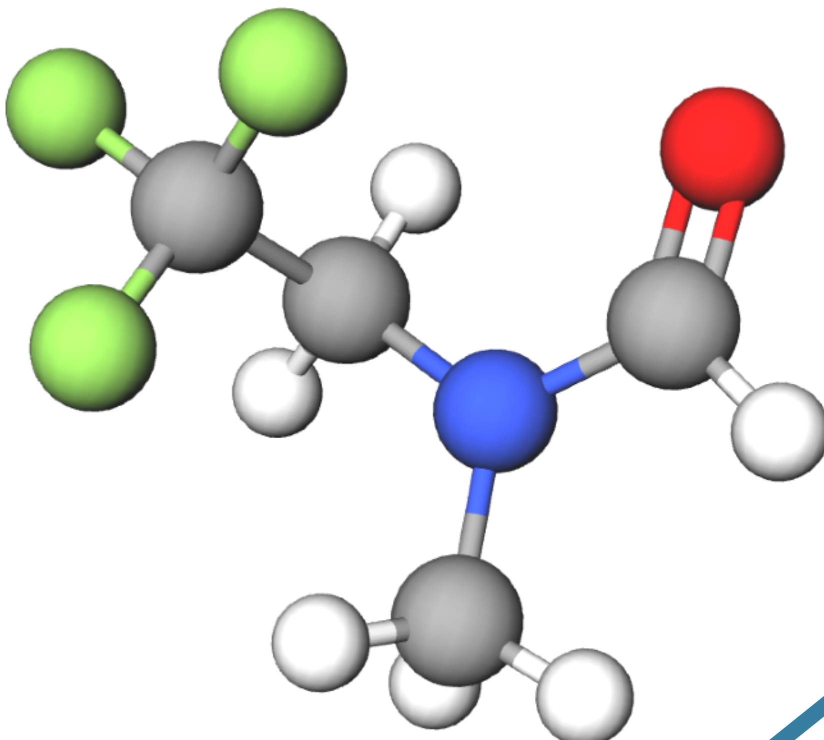
Experimental spectroscopy:
- time consuming
- (expensive) equipment required
- done one system at the time

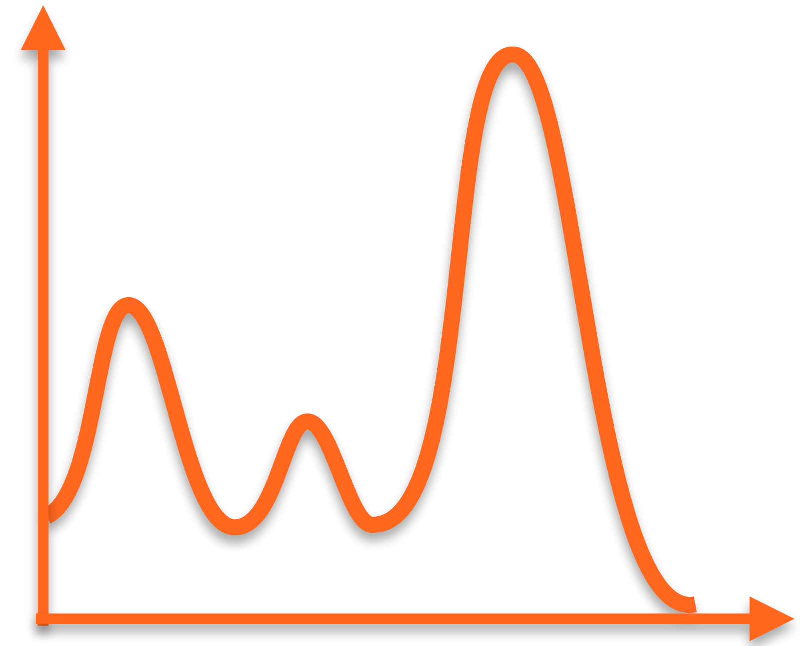
Synchrotron MAX IV in Lund
- multi-million € facility
- have to apply for beam time

Computational spectroscopy



Theoretical spectra





- GW
- BSE
- TDDFT
- etc.

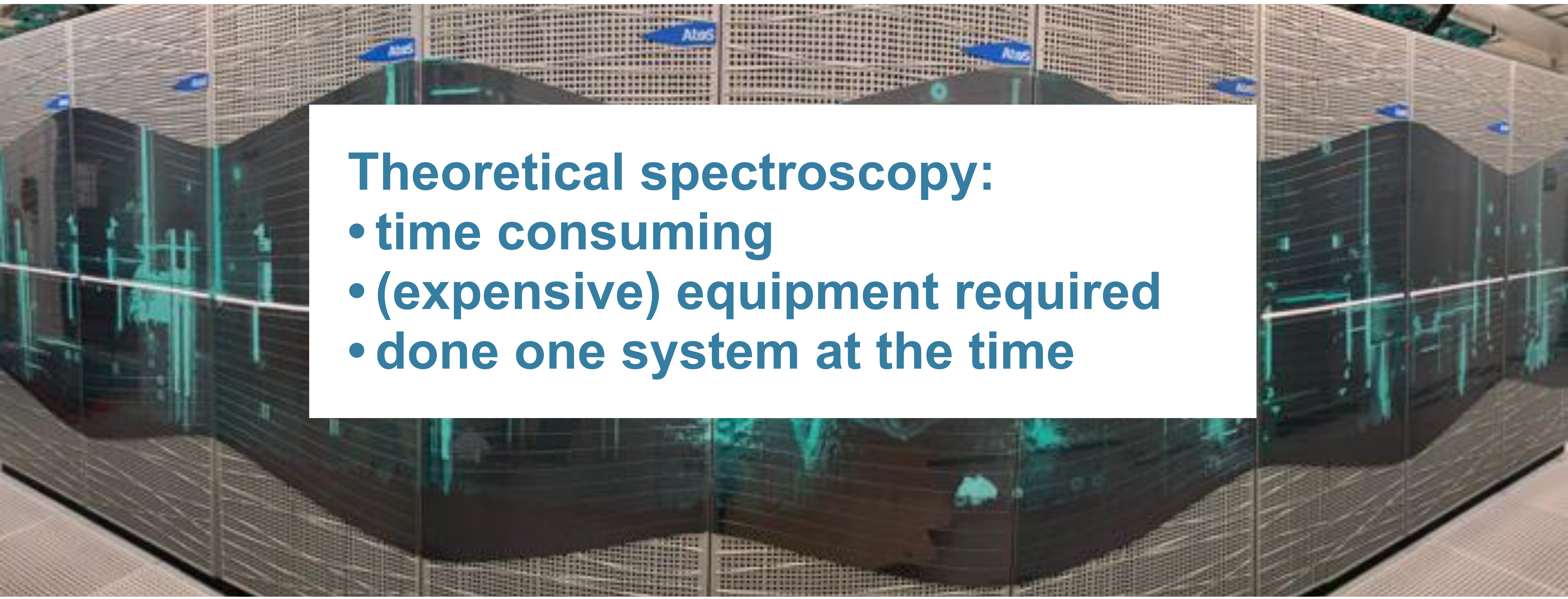
Quantum Mechanics:

- Schrödinger equation

$$H\Psi = E\Psi$$

Theoretical spectra

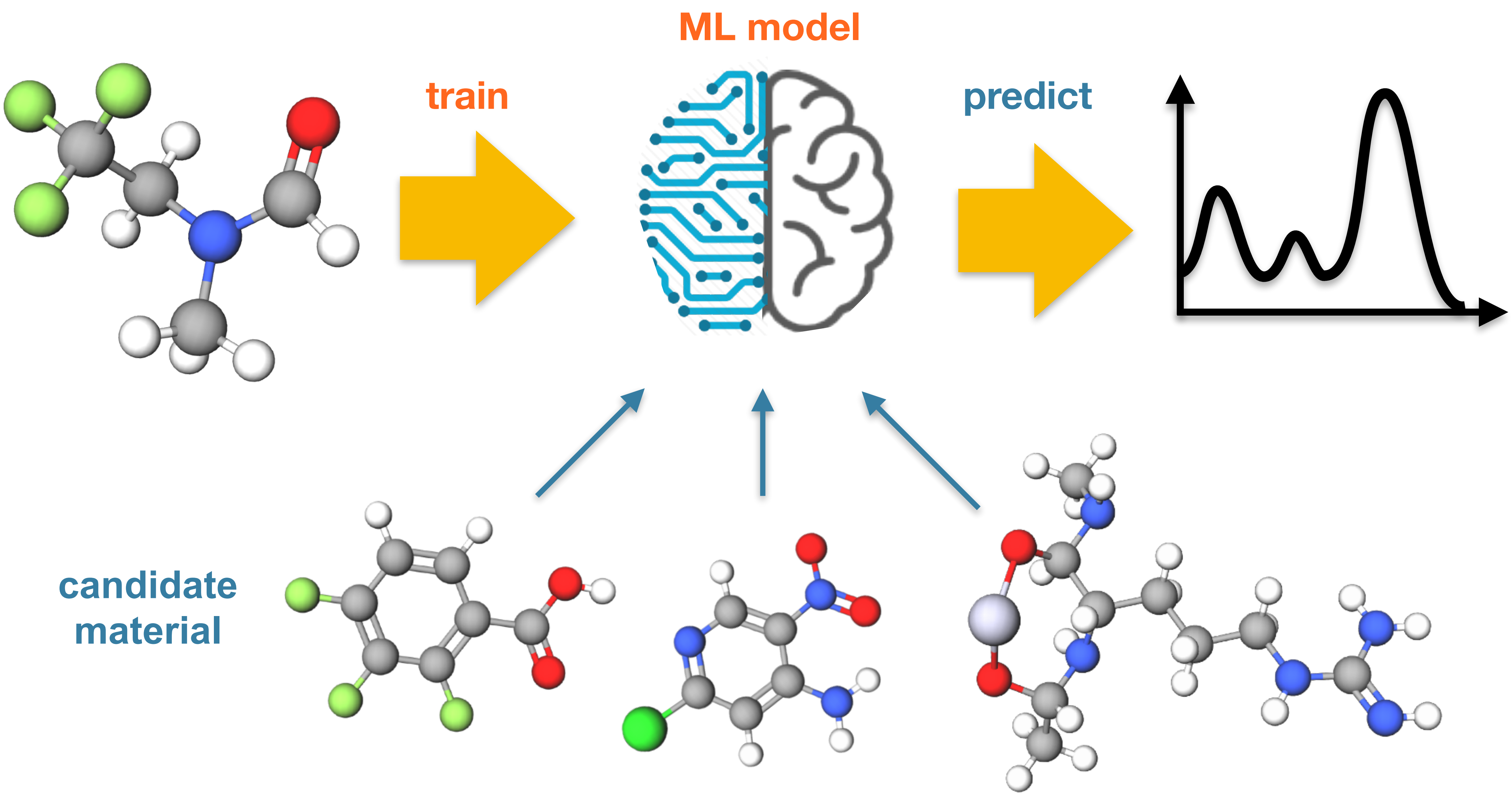
Quantum mechanical simulations are run on supercomputers.



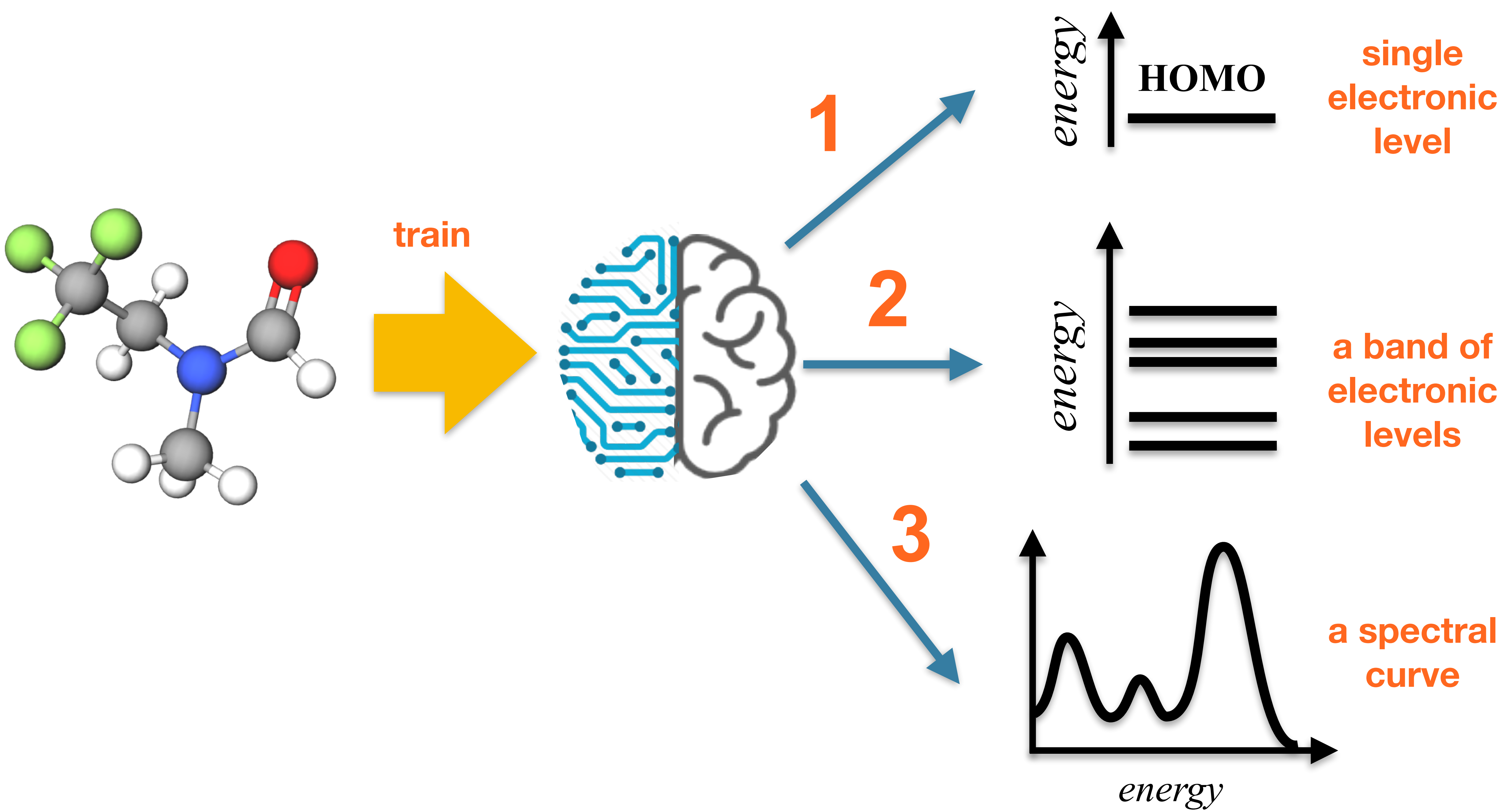
Theoretical spectroscopy:
- time consuming
- (expensive) equipment required
- done one system at the time

Mahti supercomputer at CSC

Machine learning to replace computation



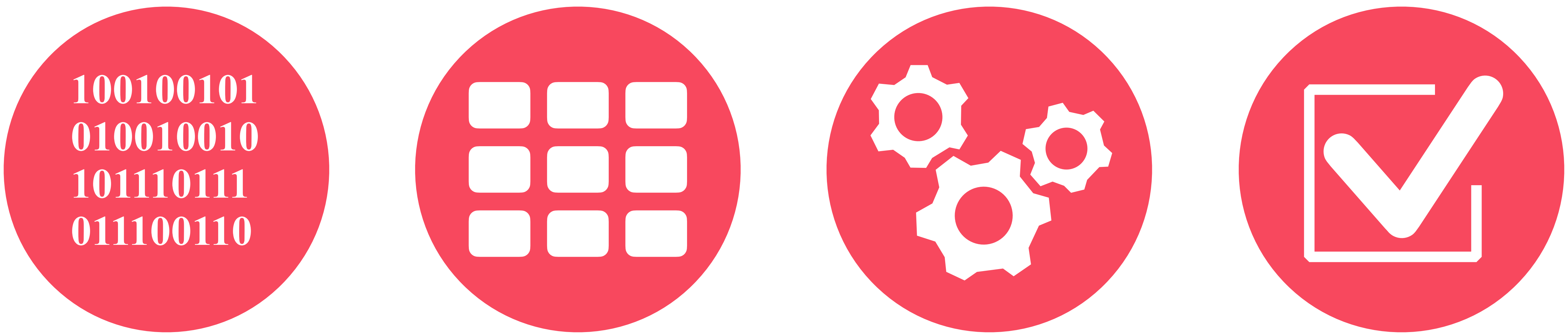
Machine learning objectives



Machine learning workflow

Dataset

ML Method

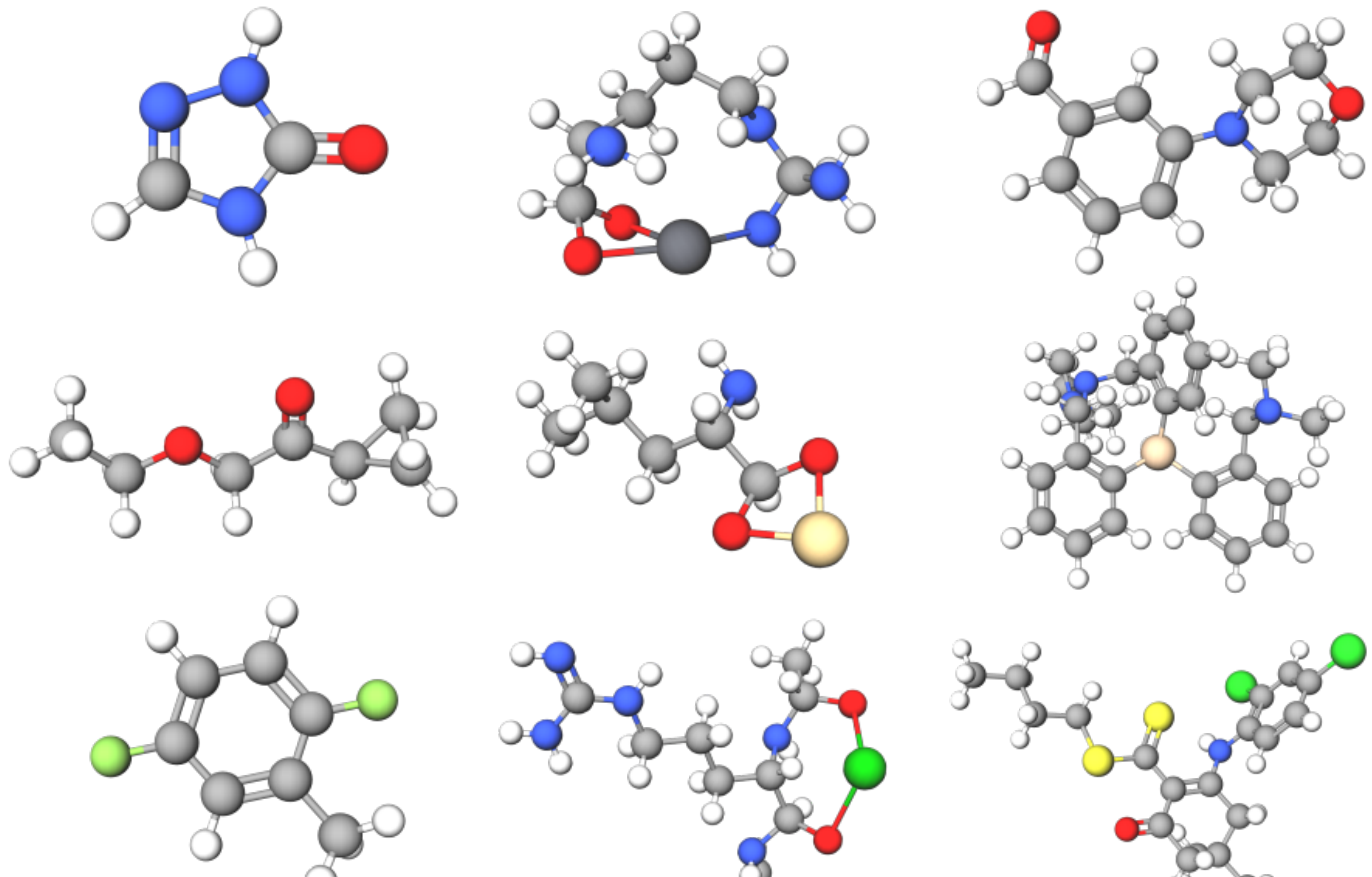


Representation

Quality Control

Dataset of molecular orbitals

QM9 AA OE

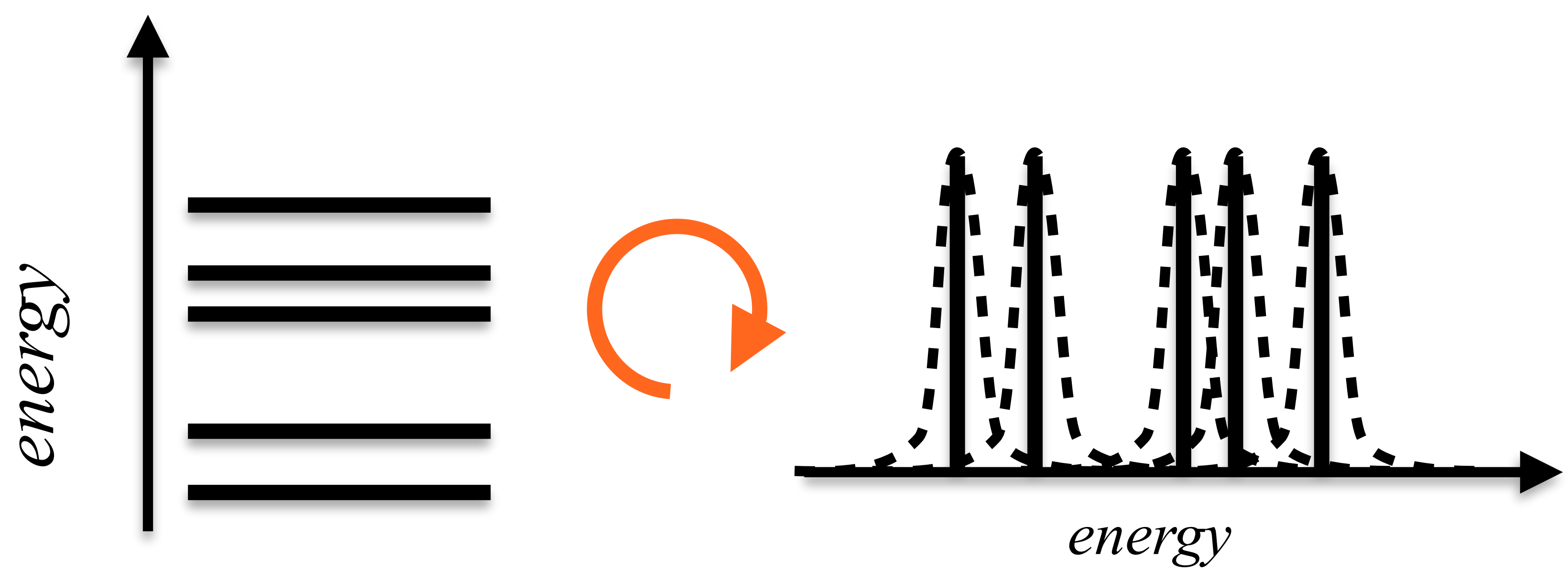


QM9	AA	OE
133,814 HOMOs	44,004 HOMOs	64,710 HOMOs

A. Stuke, et.al., J. Chem. Phys, 150, 204121 (2019)

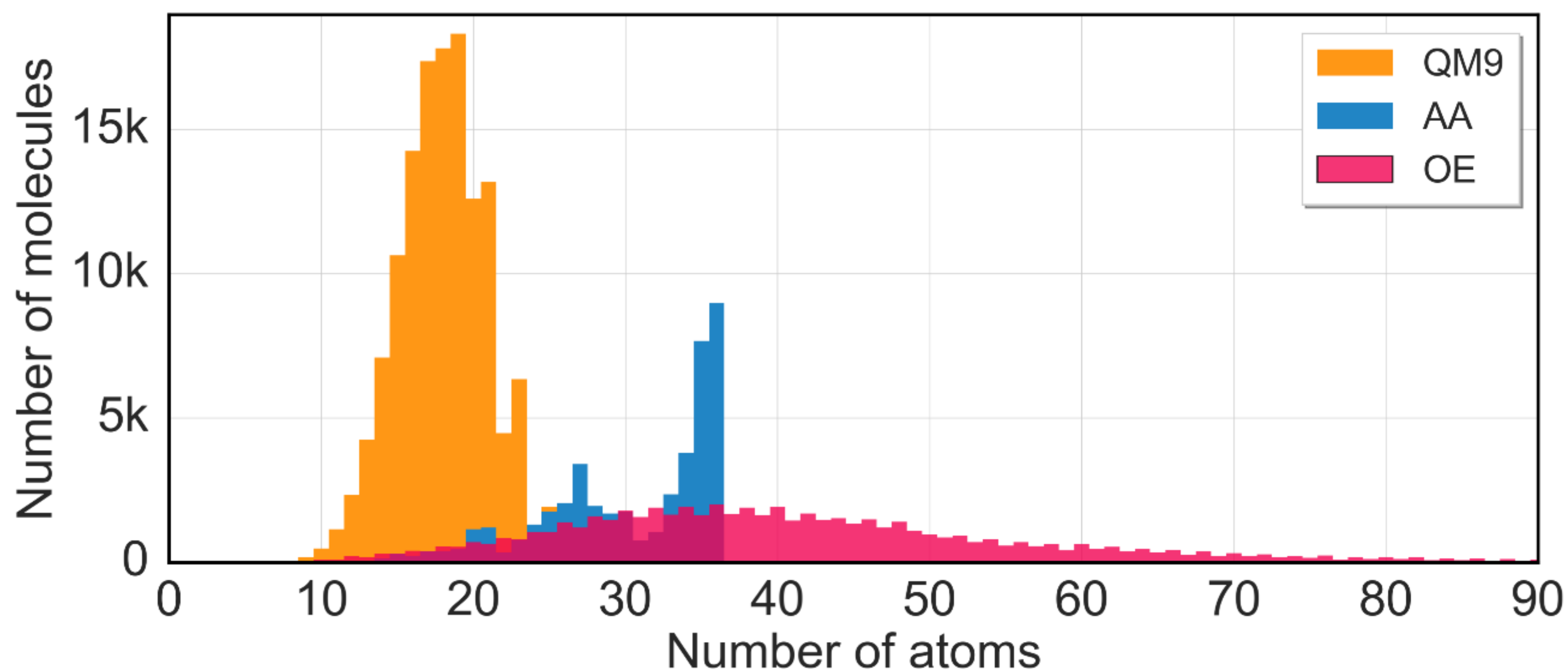
Computational details

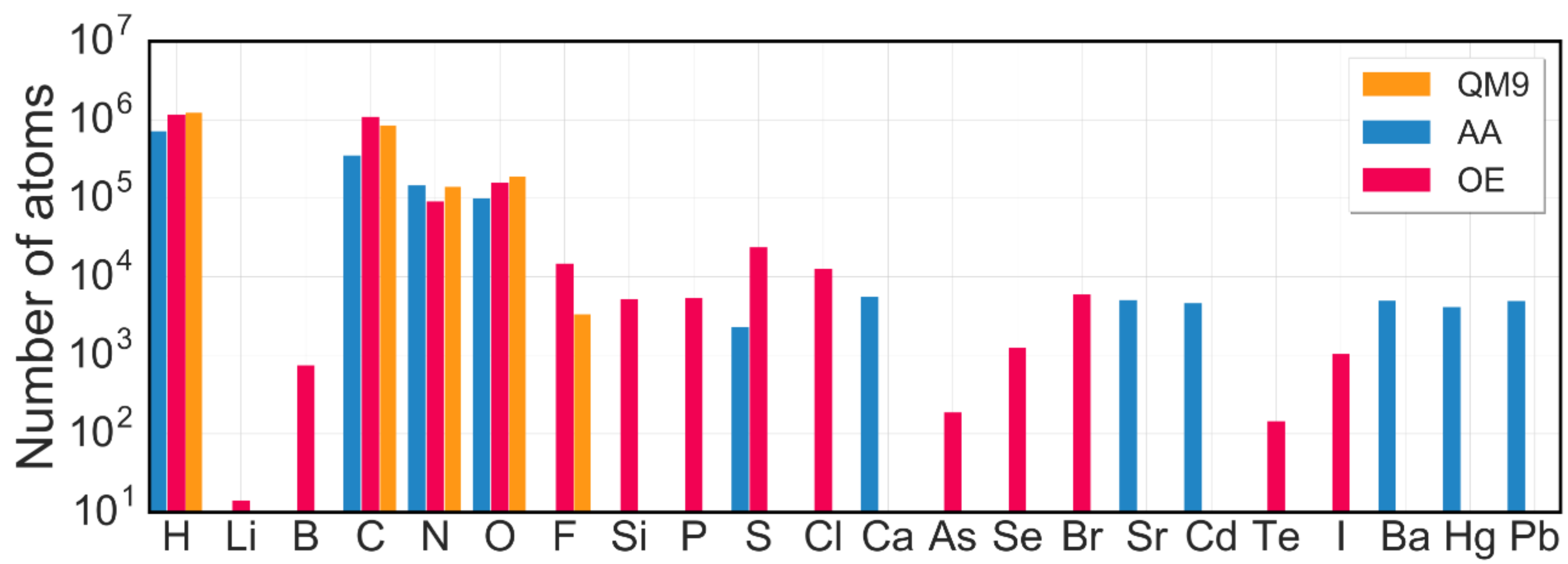
- FHI-aims for all calculations
- structure relaxed with PBE+vdW
- excitation energies: PBE Kohn-Sham eigenvalues
- spectra: Gaussian broadened eigenvalues



A. Stuke, et.al., J. Chem. Phys, 150, 204121 (2019)

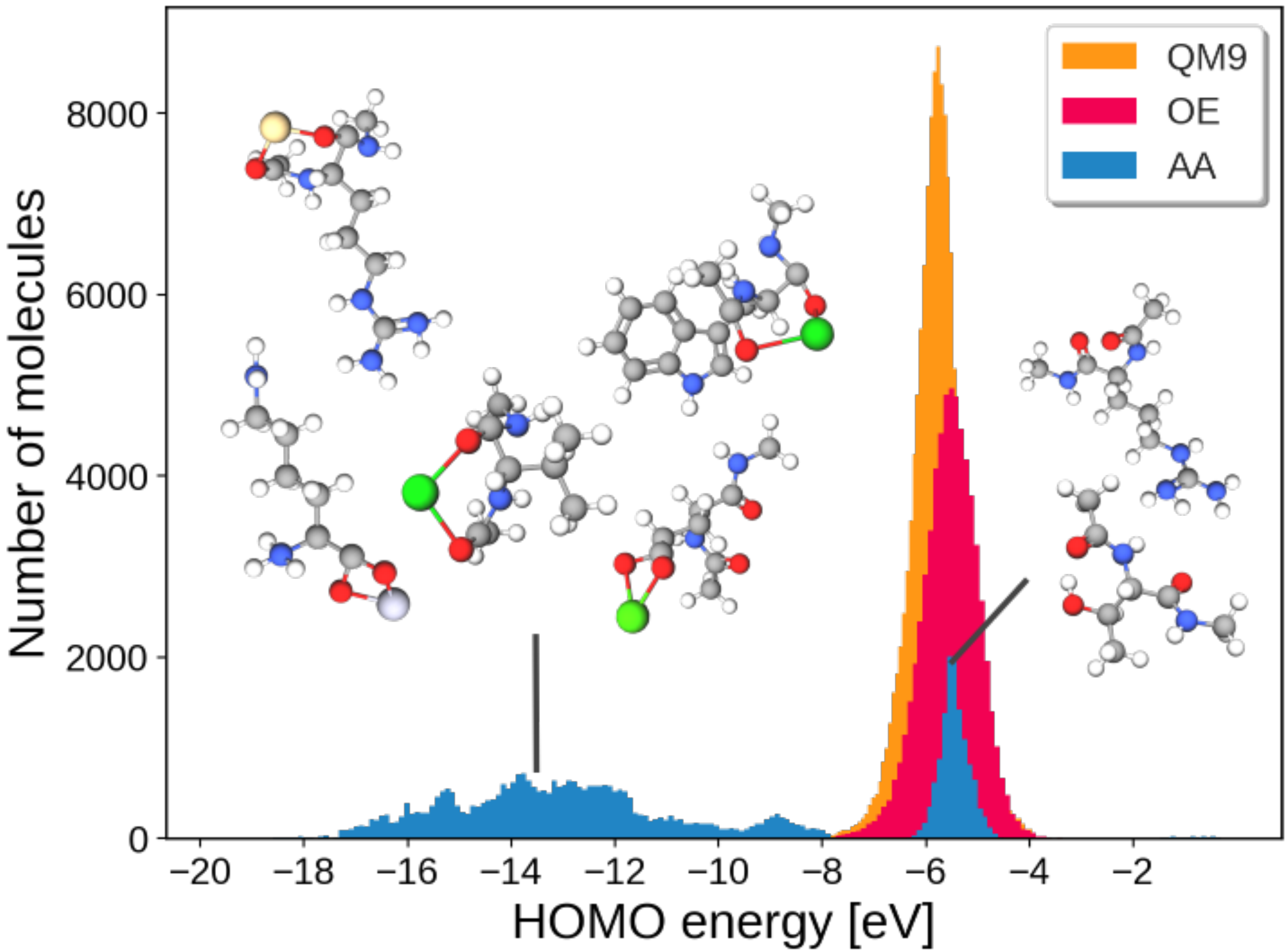
Dataset diversity



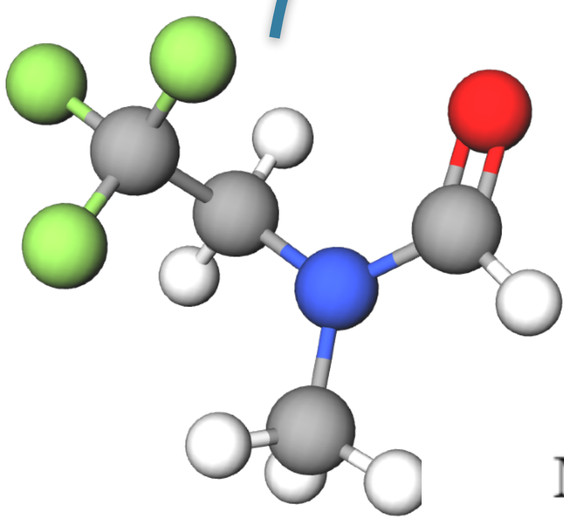


A. Stuke, et.al., J. Chem. Phys, 150, 204121 (2019)

HOMO distribution

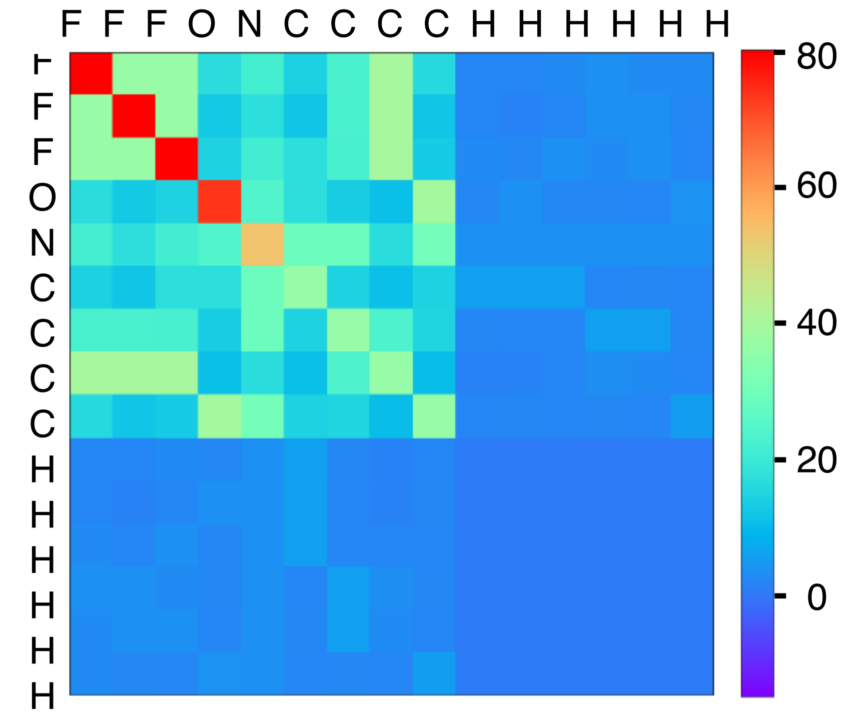


Molecular representations



"Coulomb" matrix

$$\mathbf{M}_{IJ} = \begin{cases} 0.5 Z_I^{2.4} & \text{for } I = J \\ \dfrac{Z_I Z_J}{|\mathbf{R}_I - \mathbf{R}_J|} & \text{for } I \neq J \end{cases}$$

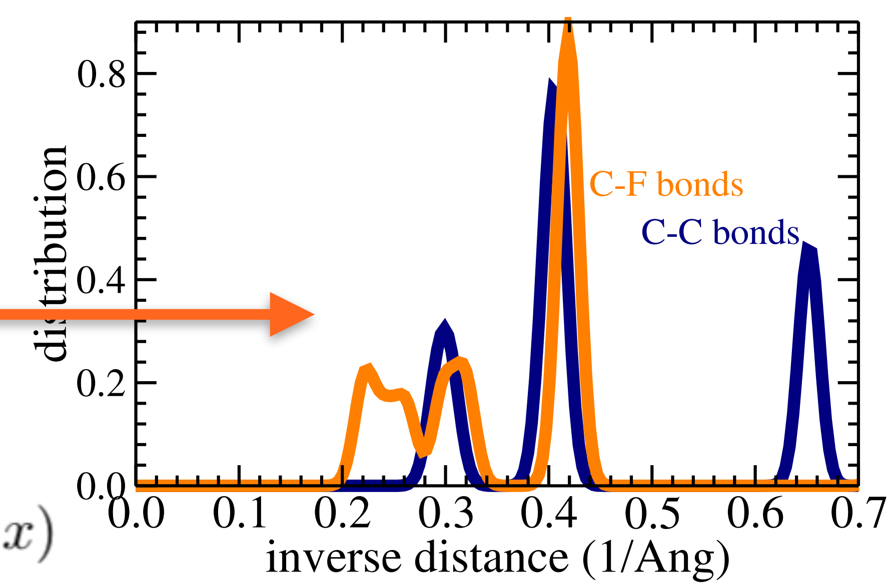


Many-body tensor

$$\mathrm{MBTR}_1^{Z_1}(x) = \sum_{l}^{|Z_1|} w_1^l d_1^l(x)$$

$$\mathrm{MBTR}_2^{Z_1,Z_2}(x) = \sum_{l}^{|Z_1|} \sum_{m}^{|Z_2|} w_2^{l,m} d_2^{l,m}(x)$$

$$\mathrm{MBTR}_3^{Z_1,Z_2,Z_3}(x) = \sum_{l}^{|Z_1|} \sum_{m}^{|Z_2|} \sum_{n}^{|Z_3|} w_3^{l,m,n} d_3^{l,m,n}(x)$$



A. Stuke, et.al., J. Chem. Phys, 150, 204121 (2019)

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Kernel Ridge Regression for HOMO

KRR - simple and powerful kernel-based regression tool

$$y^{ML}(R) = \sum_{i=1}^{N} \alpha_i K(R, R_i) \qquad K(R, R_i) = e^{-\frac{d(R,R_i)^2}{2\sigma^2}}$$

error^2 regularization

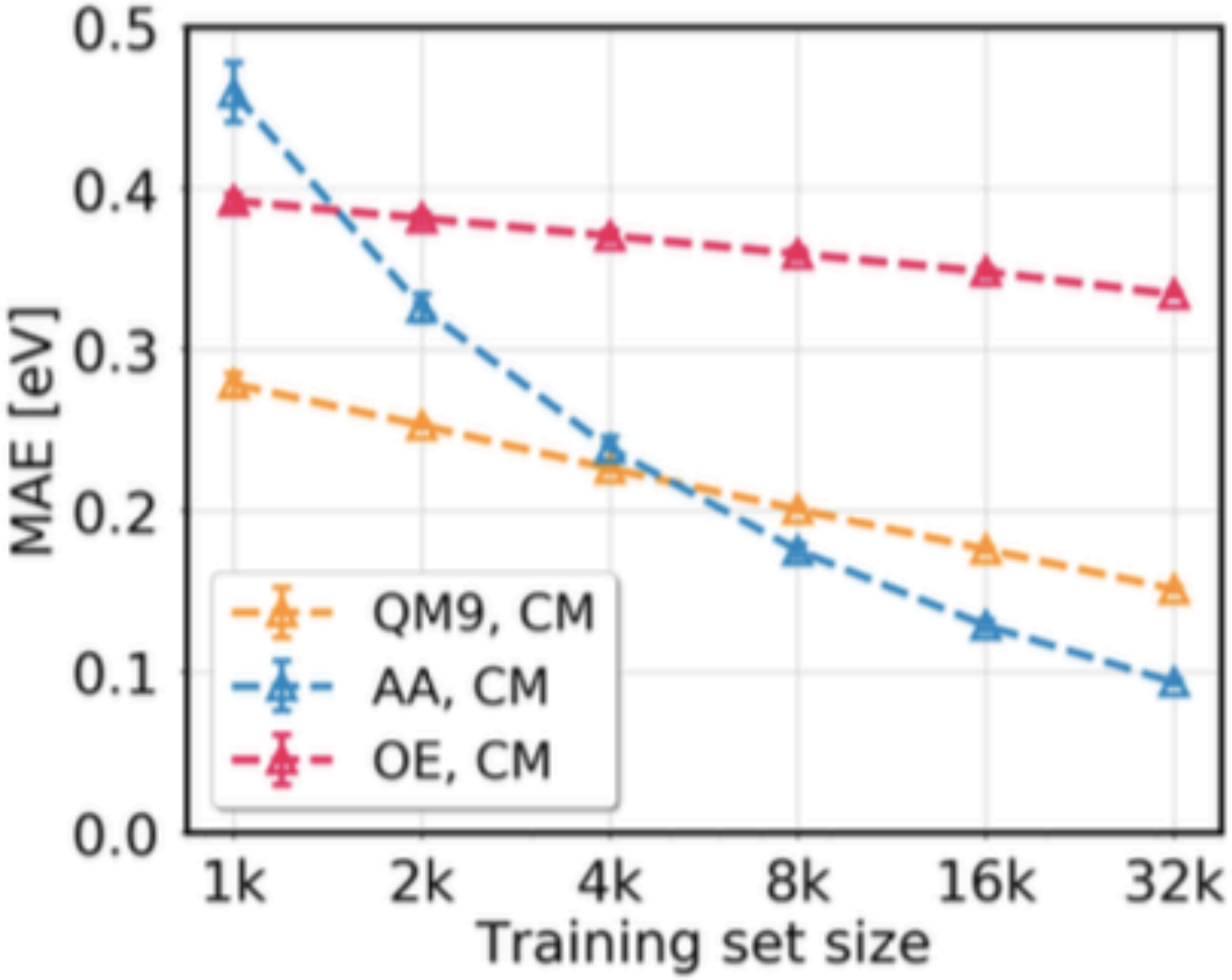
$$\arg\min_{\alpha} \sum_i [y^{ML}(\mathbf{R_i}) - y^{ref}(\mathbf{R_i})]^2 + \gamma \sum_i \alpha_i^2$$

$$\boldsymbol{\alpha} = (\mathbf{K} + \gamma \mathbf{I})^{-1} \mathbf{y^{ref}}$$

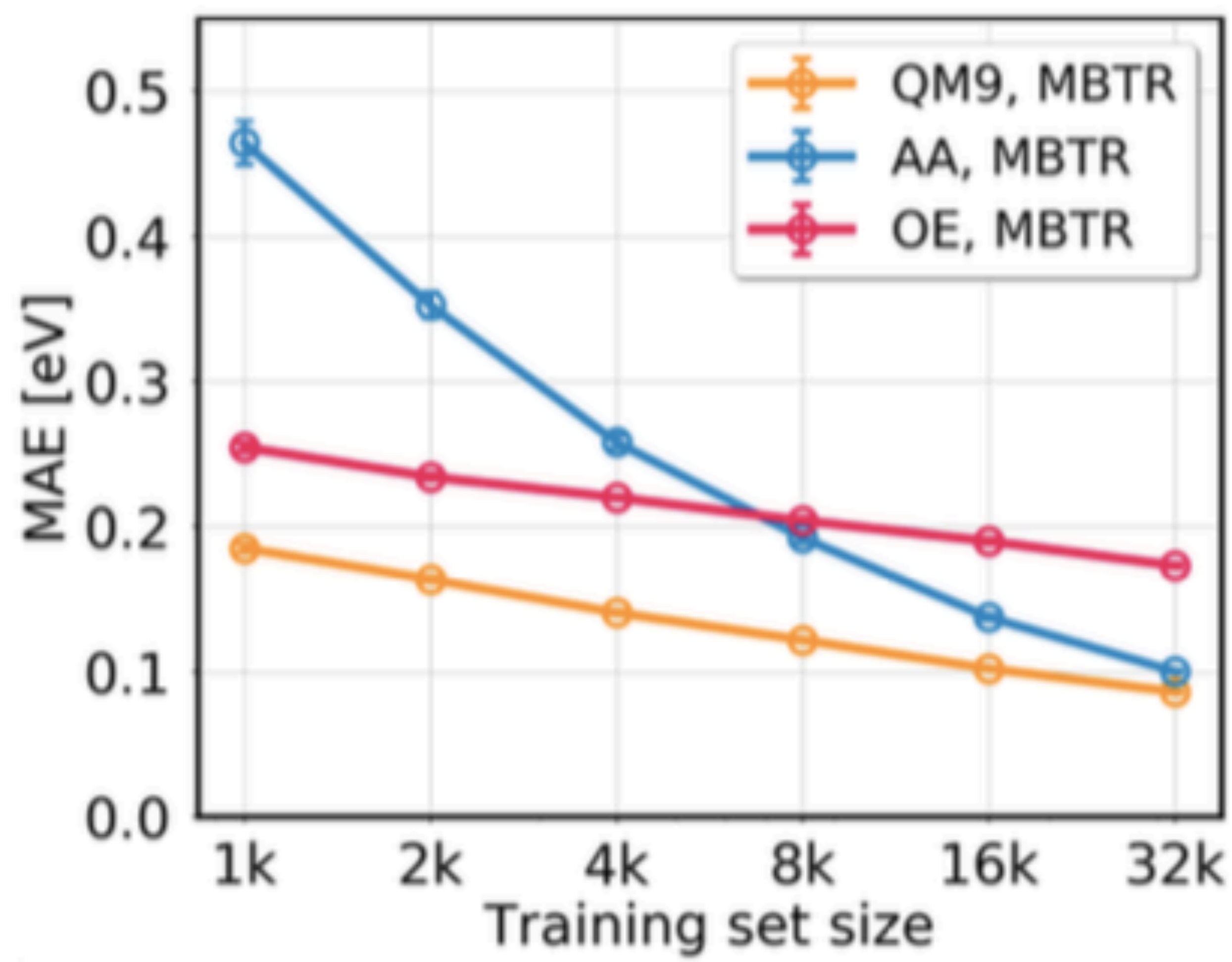
A. Stuke, et.al., J. Chem. Phys, 150, 204121 (2019)

Predictive power of ML models

"Coulomb" matrix



Many-body tensor



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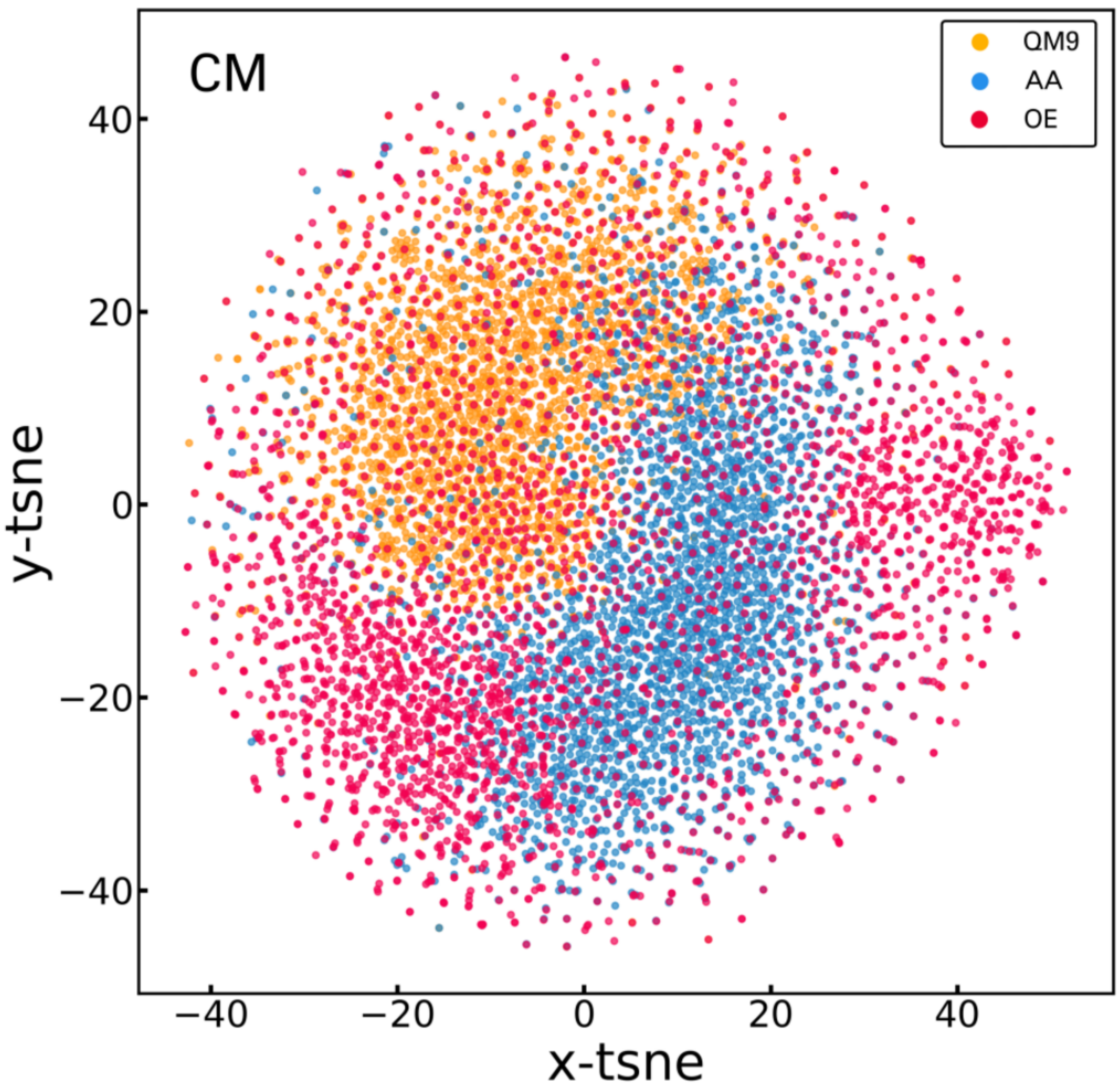
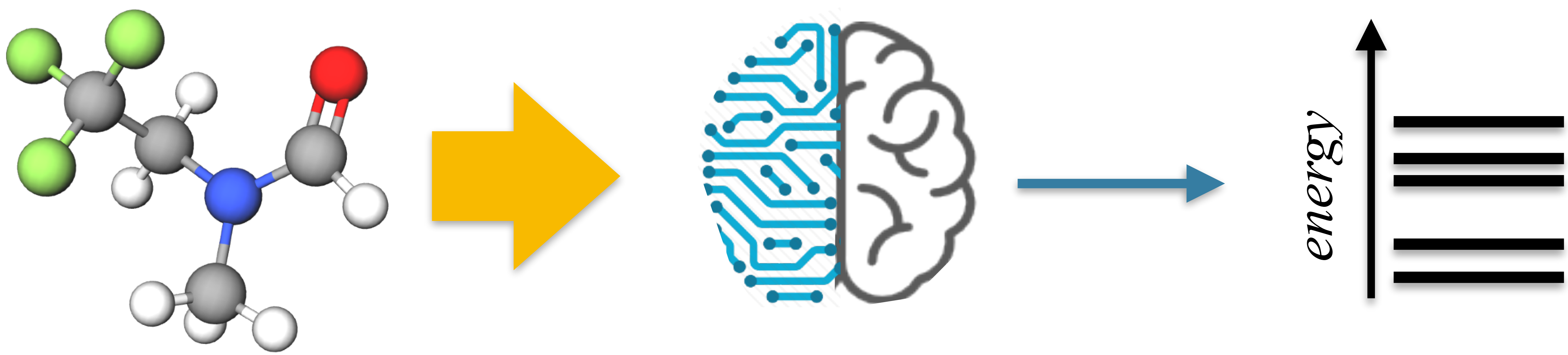
A. Stuke, et.al., J. Chem. Phys, 150, 204121 (2019)

Chemical diversity of datasets

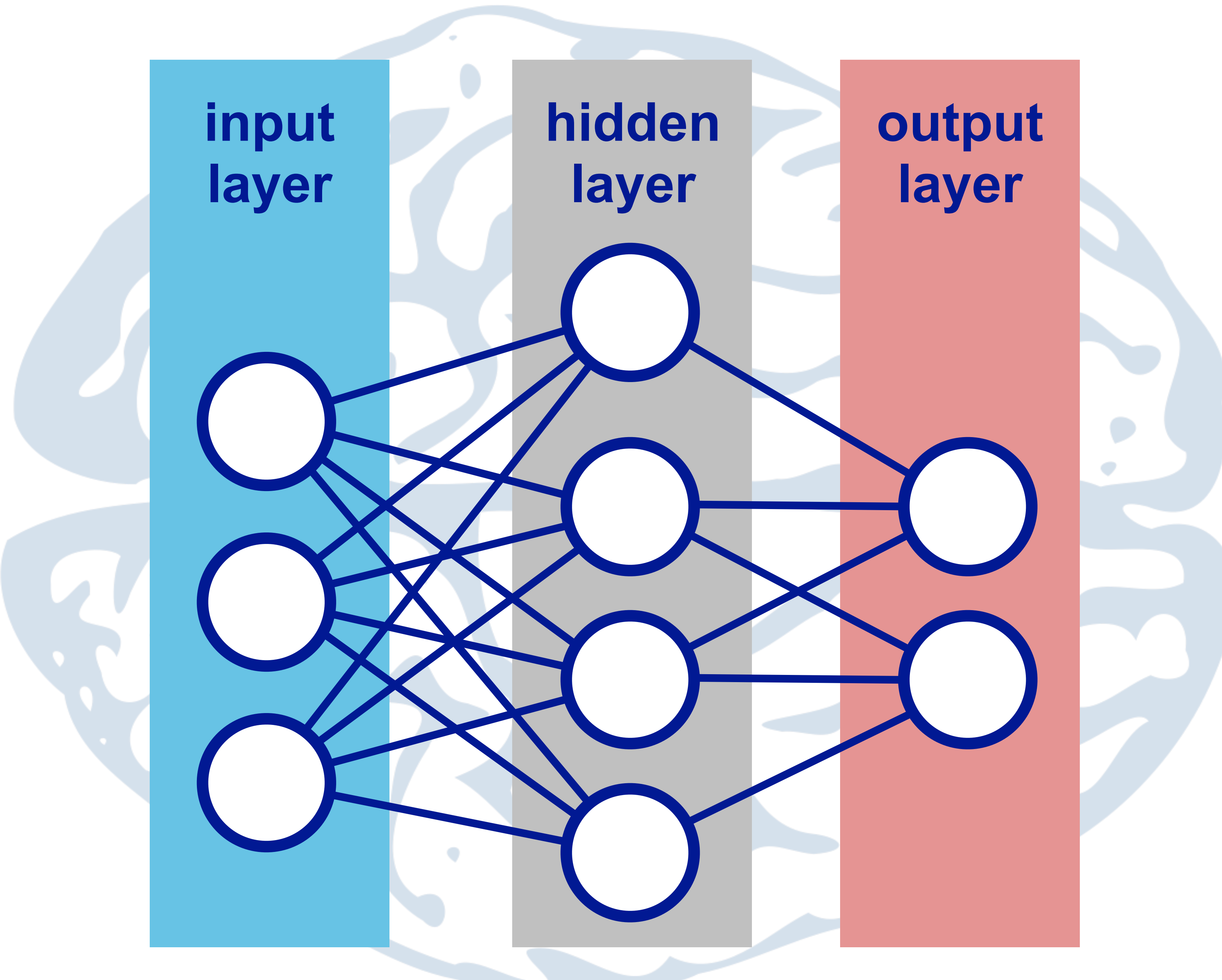


A. Stuke, et.al., J. Chem. Phys, 150, 204121 (2019)

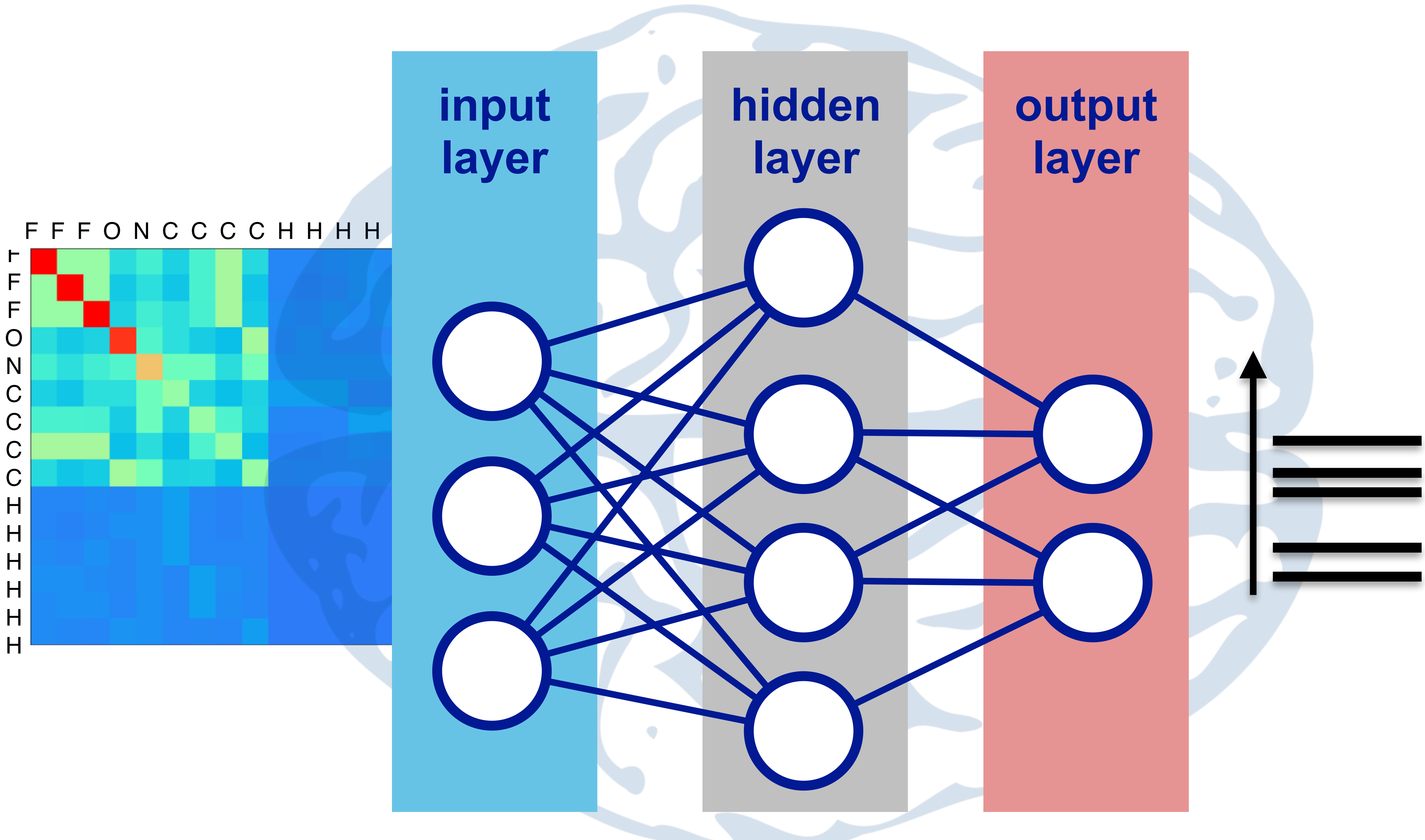
Learning a band of electronic states



Deep learning: artificial neural networks

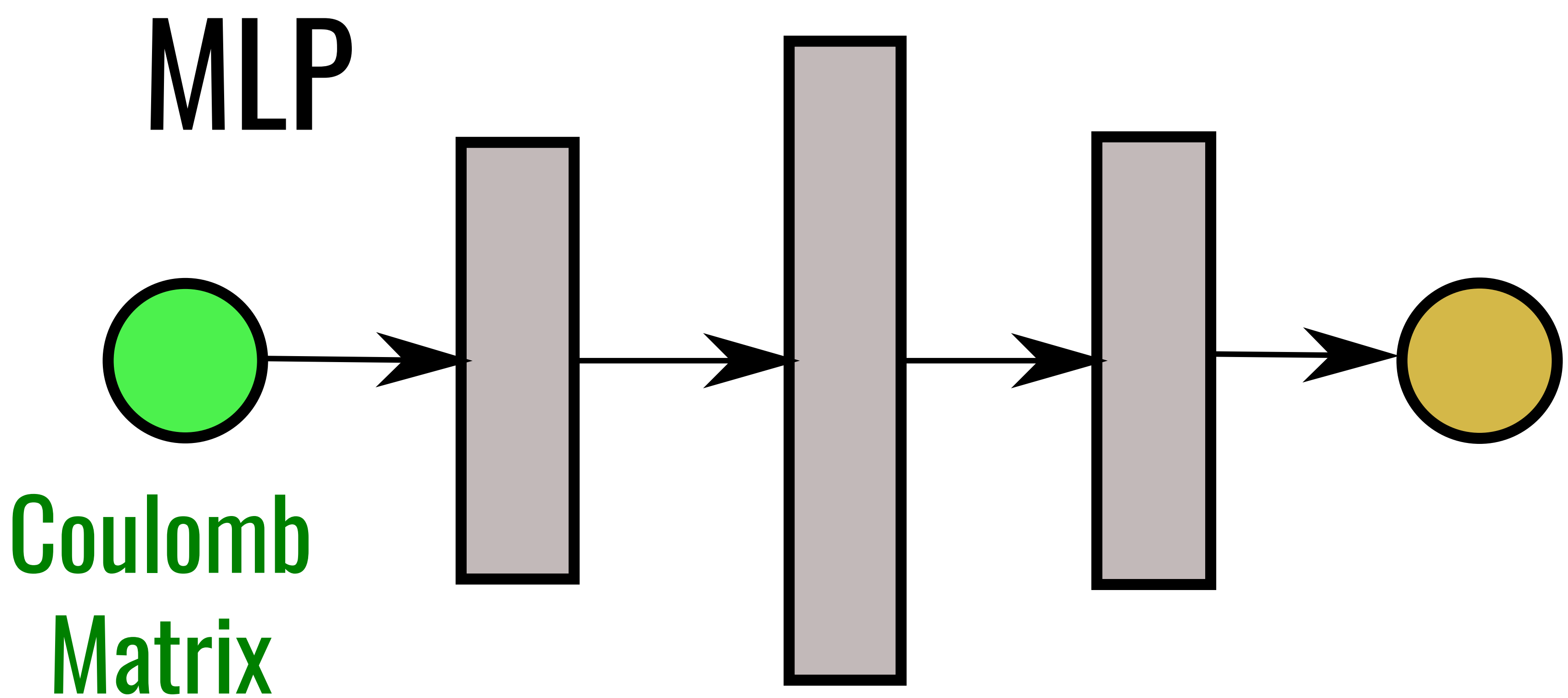


Deep learning: artificial neural networks



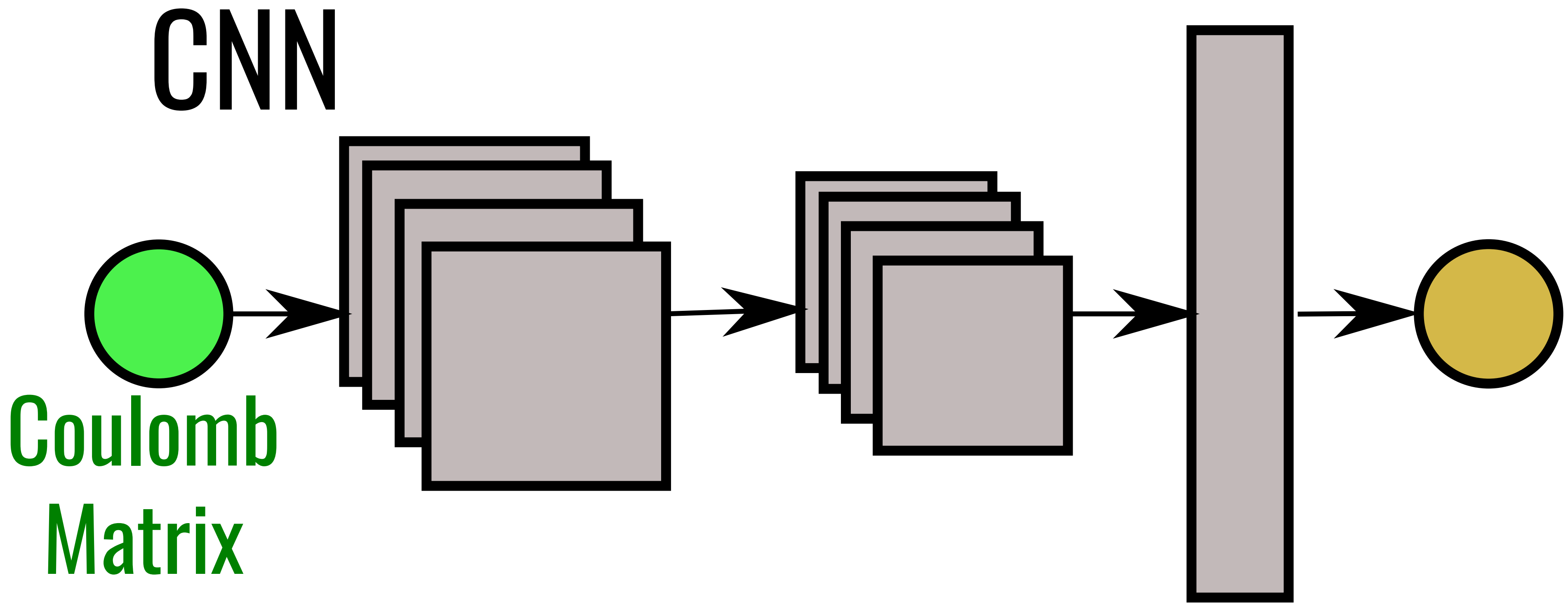
Ghosh et.al., Adv. Sci. 6, 1970053 (2019)

Multi layer perceptron (MLP) design



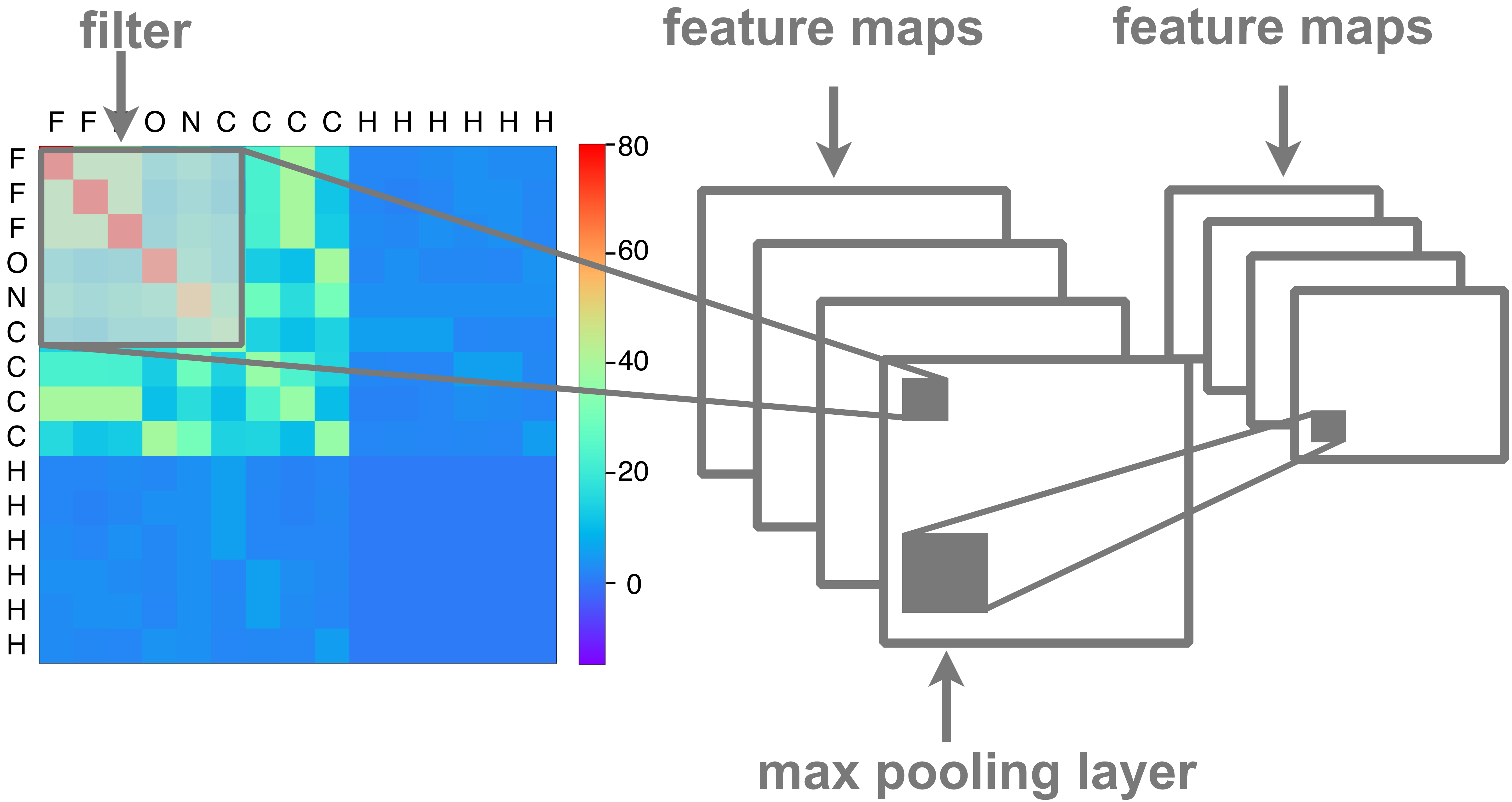
Ghosh et.al., Adv. Sci. 6, 1970053 (2019)

Convolutional neural network

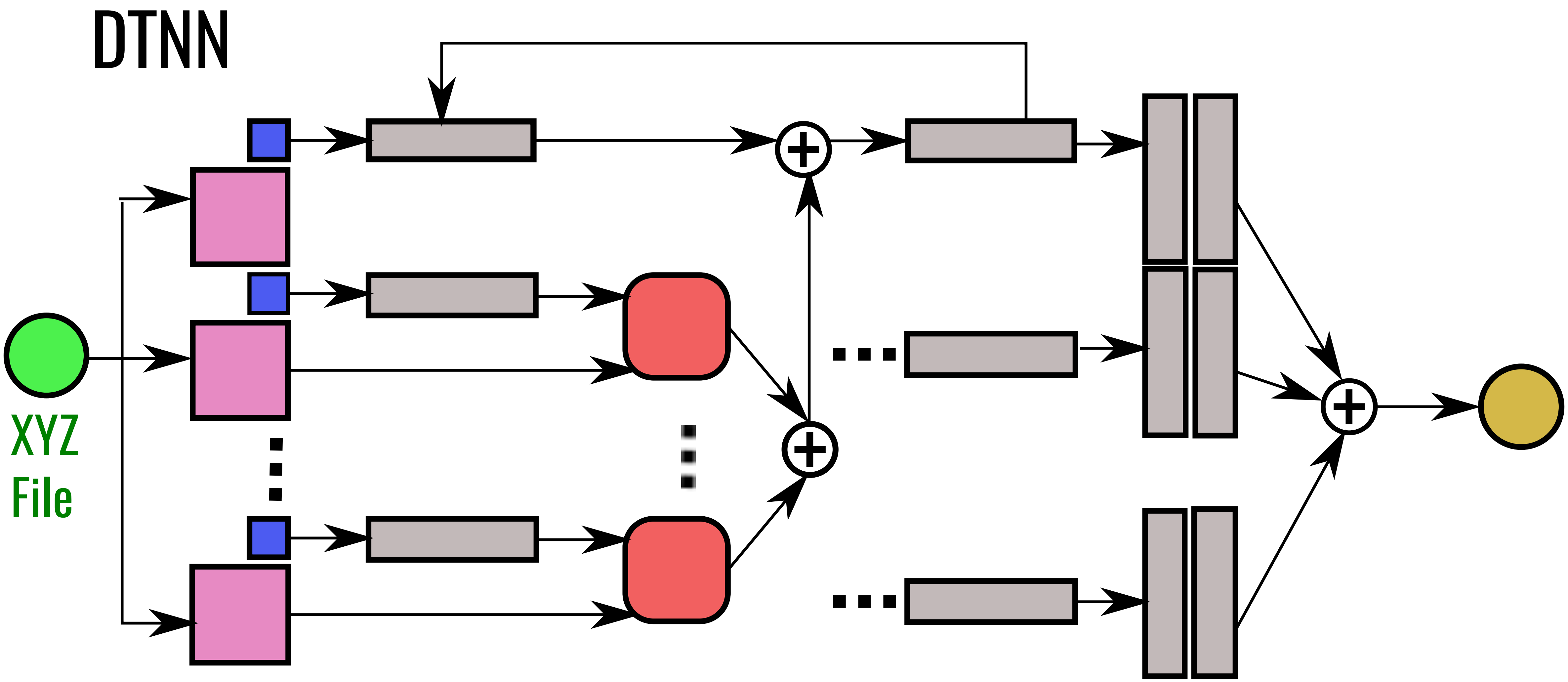


Ghosh et.al., Adv. Sci. 6, 1970053 (2019)

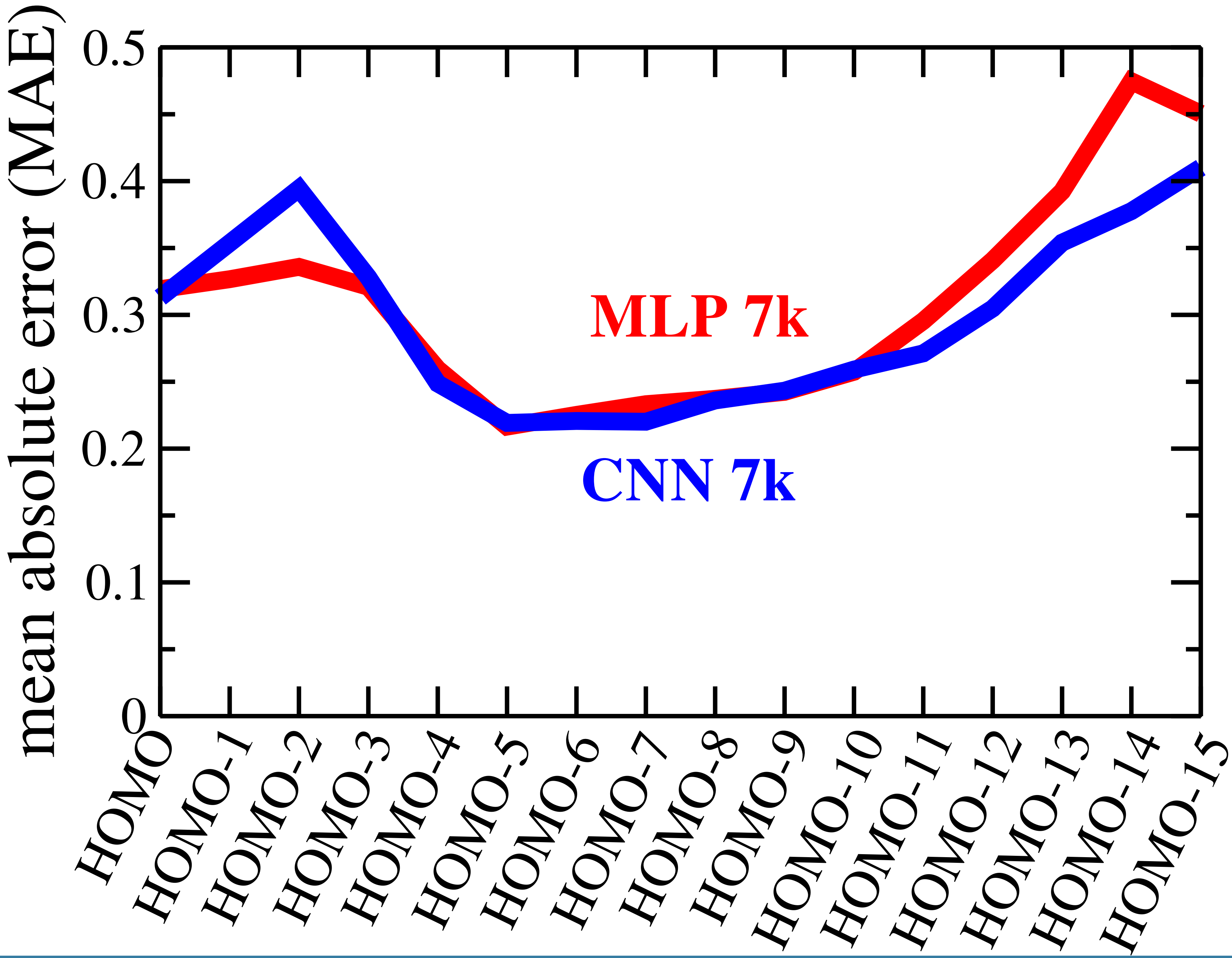
Convolutional neural network



Deep tensor neural network

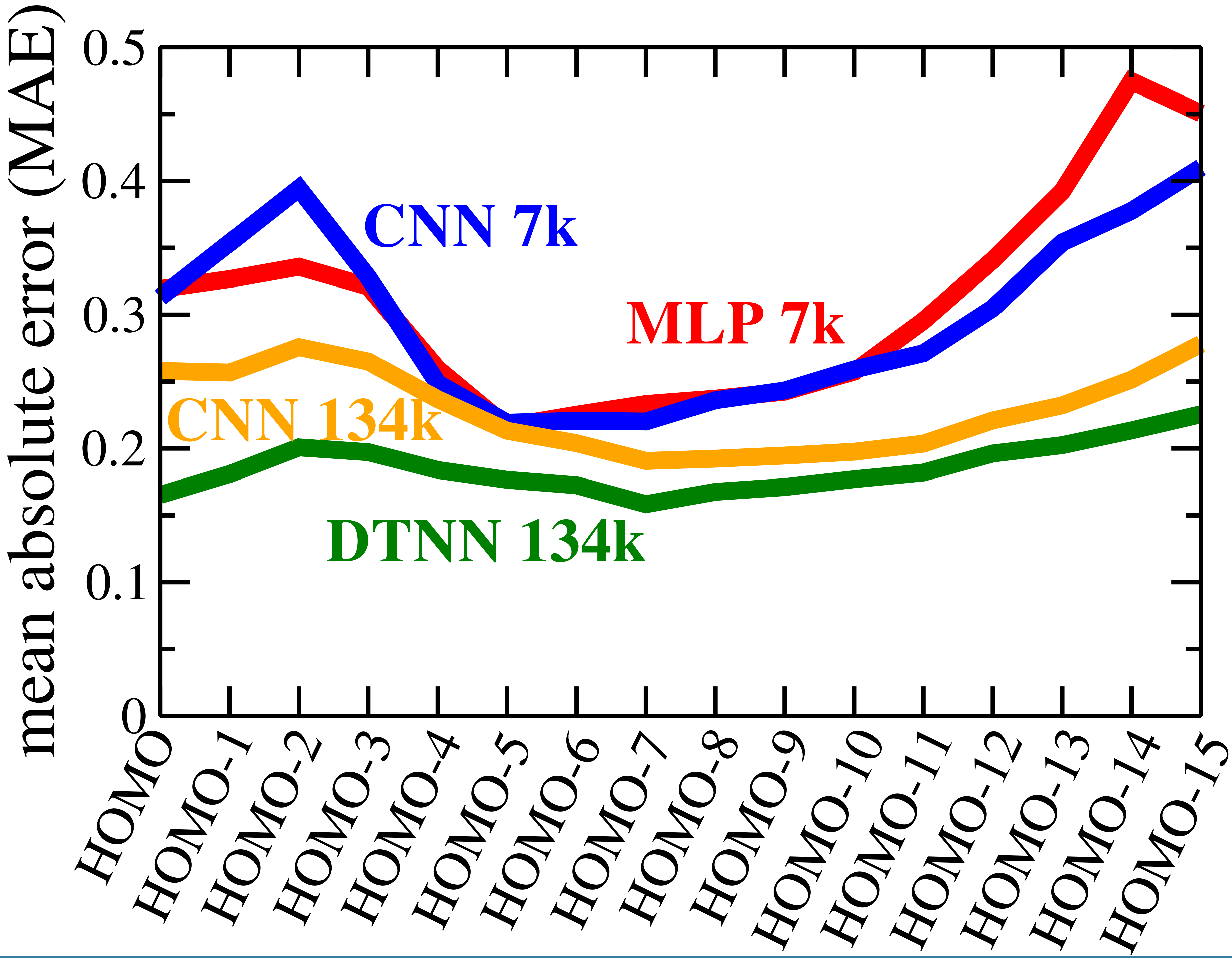


ANN performance comparison



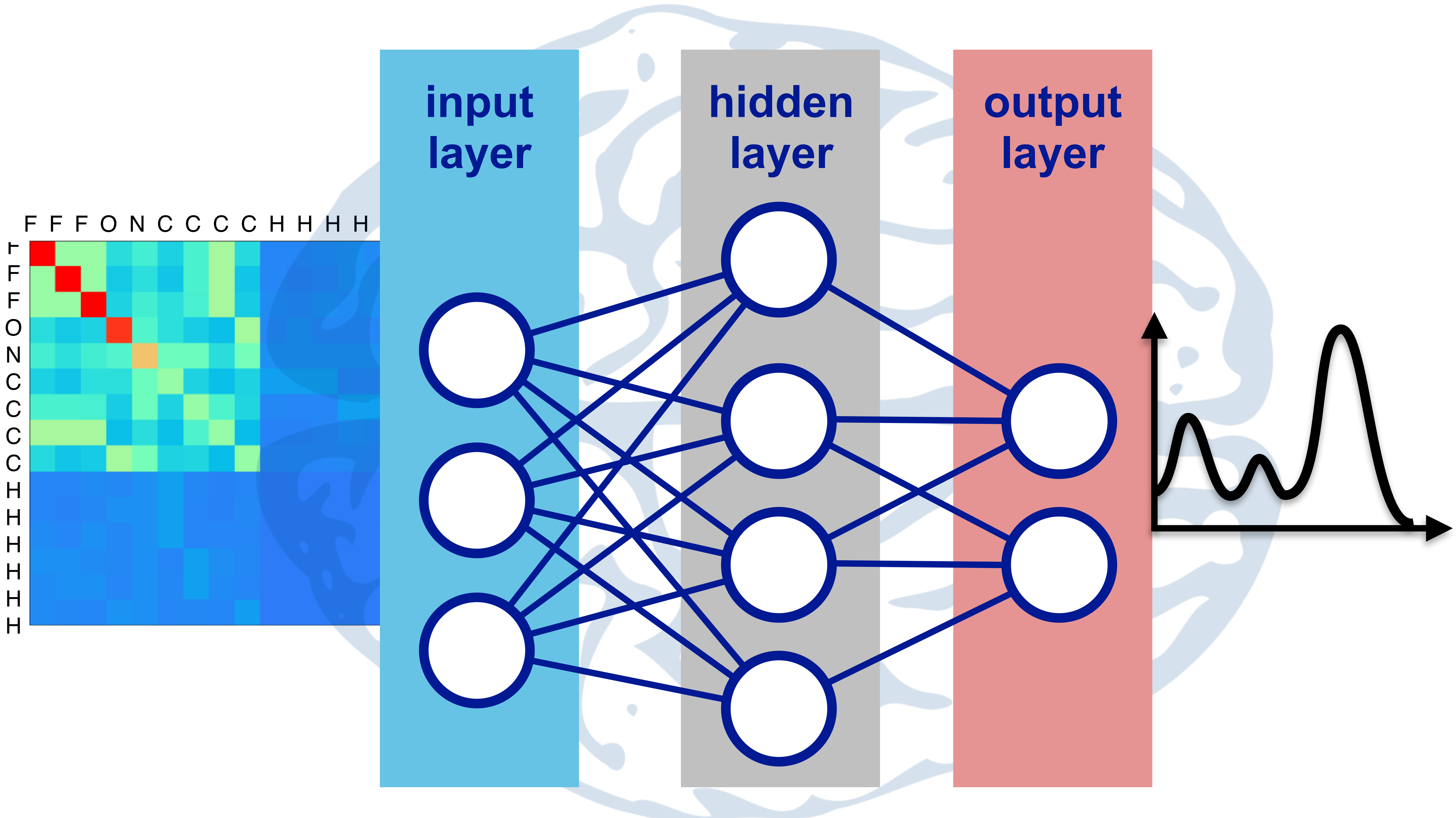
Ghosh et.al., Adv. Sci. 6, 1970053 (2019)

ANN performance comparison



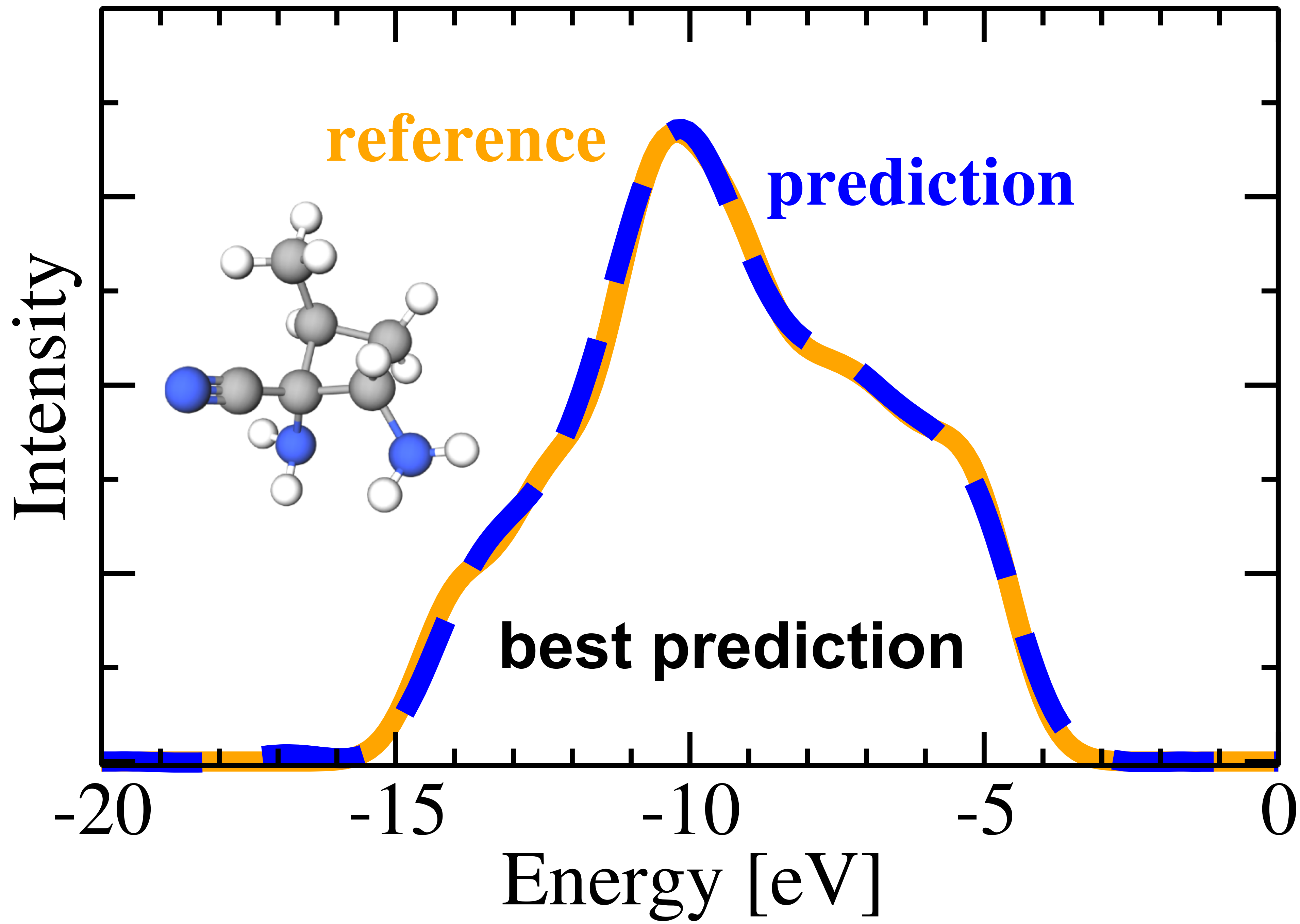
Ghosh et.al., Adv. Sci. 6, 1970053 (2019)

Deep learning: artificial neural networks



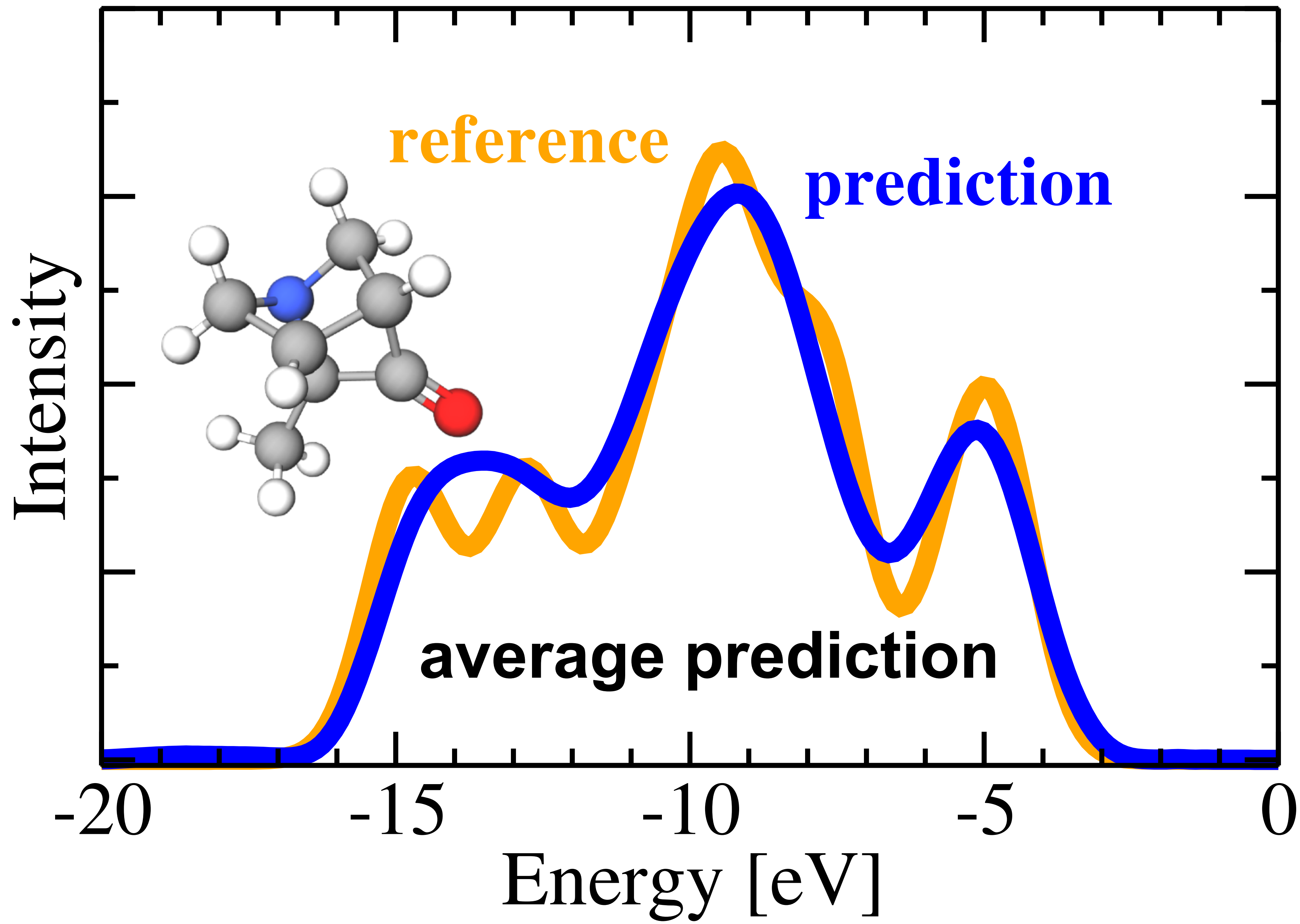
Ghosh et.al., Adv. Sci. 6, 1970053 (2019)

CNN predictions for spectra



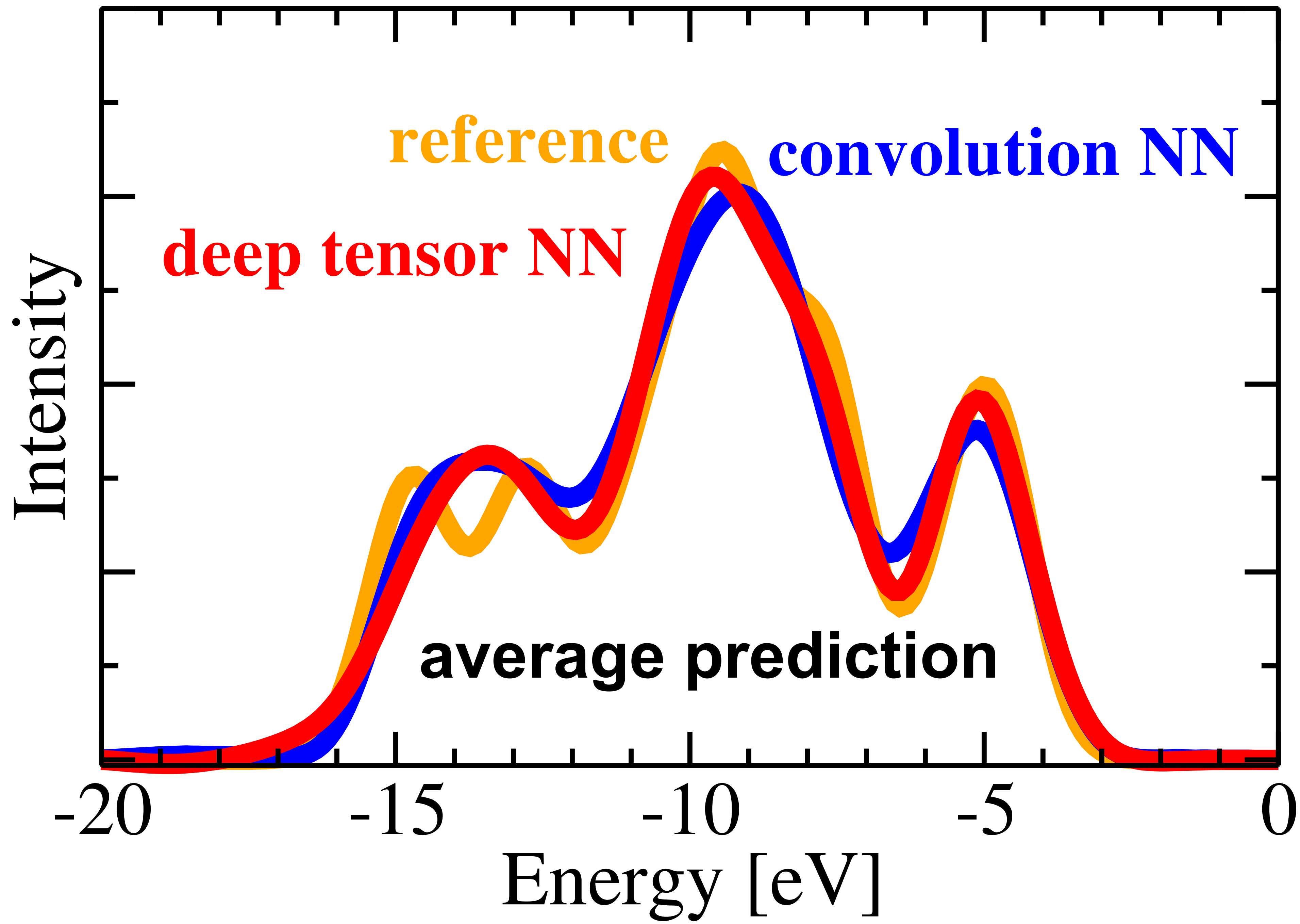
Ghosh et.al., Adv. Sci. 6, 1970053 (2019)

CNN predictions for spectra



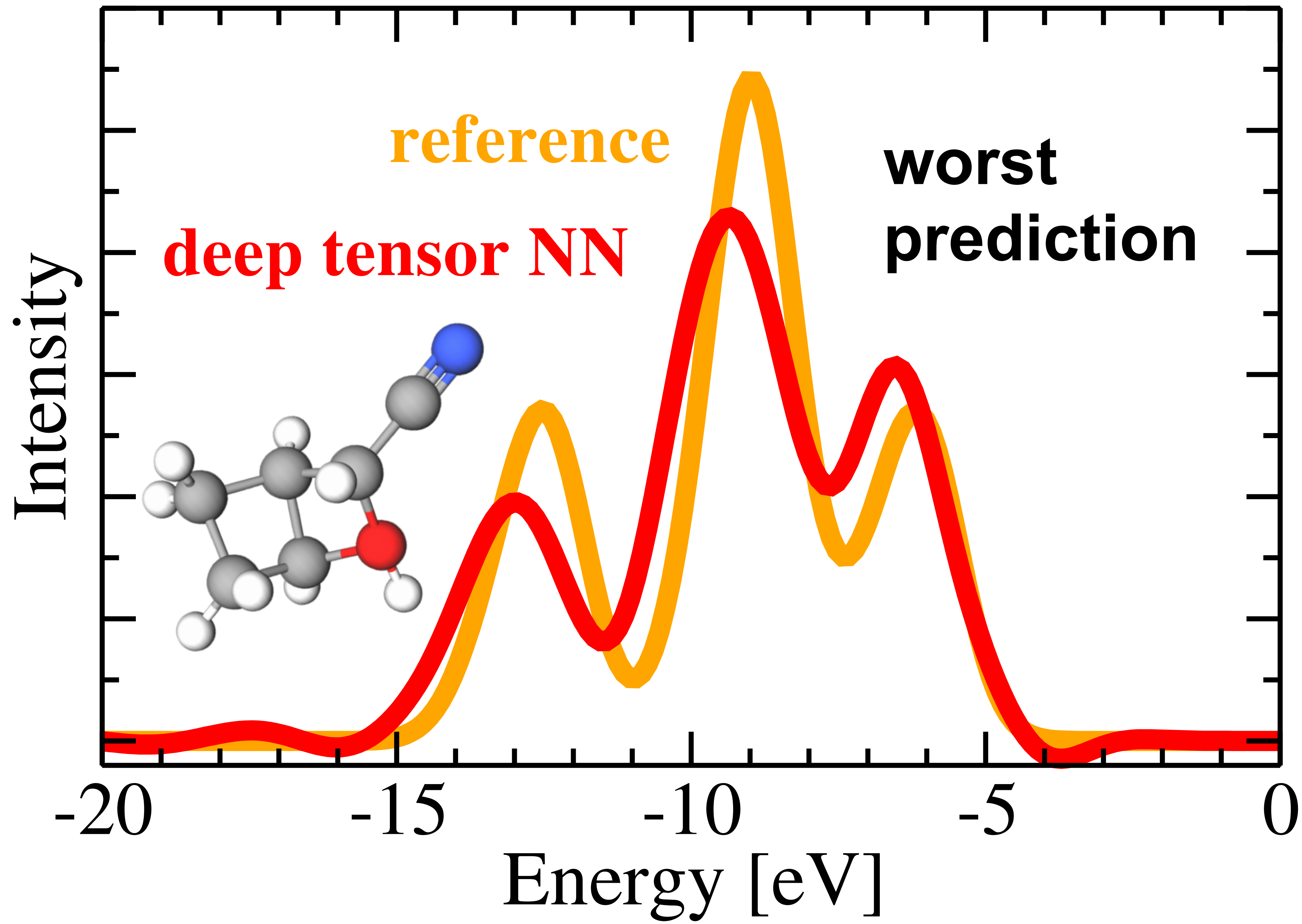
Ghosh et.al., Adv. Sci. 6, 1970053 (2019)

DTNN and CNN in comparison



Ghosh et.al., Adv. Sci. 6, 1970053 (2019)

DTNN worst case



Ghosh et.al., Adv. Sci. 6, 1970053 (2019)

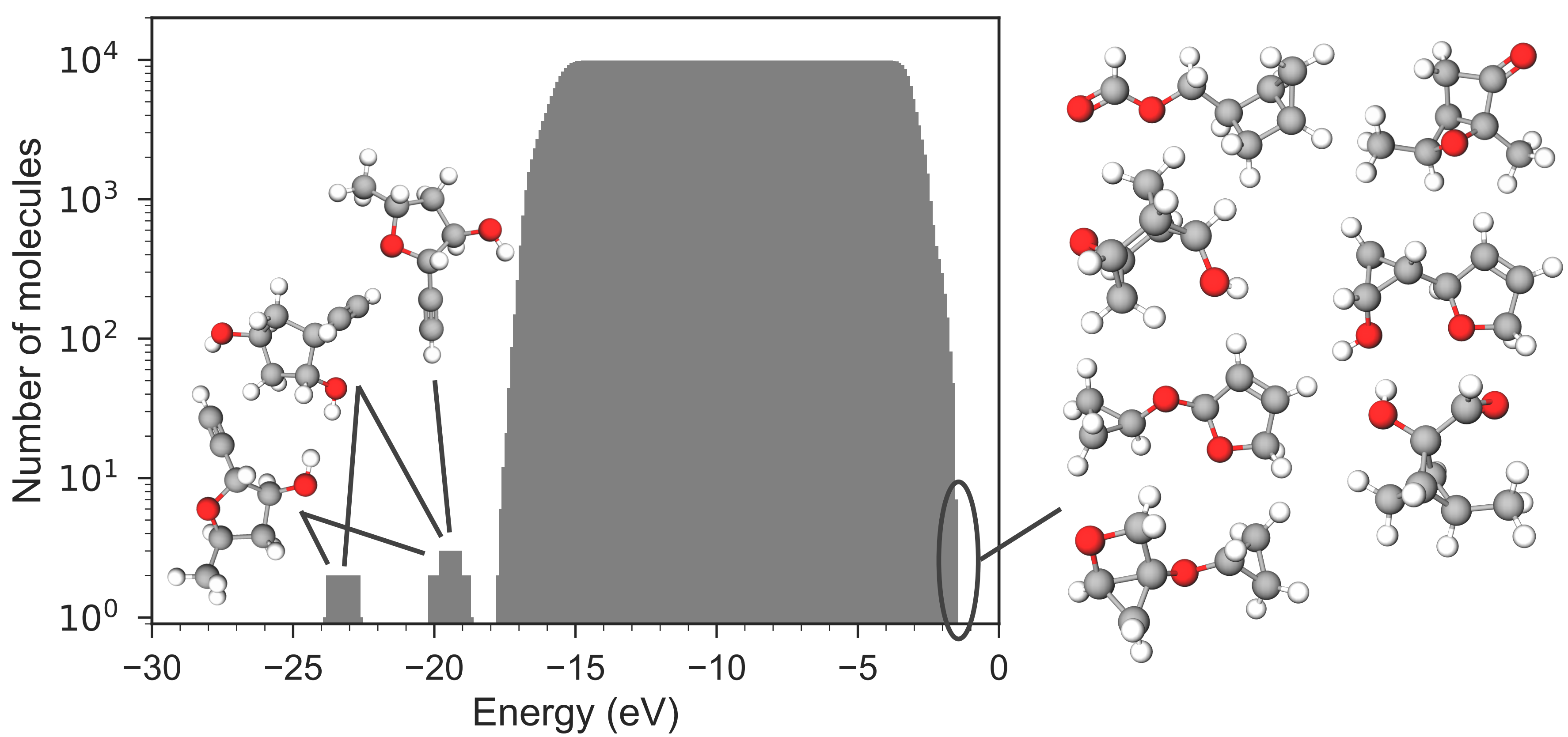
Neural network summary

NN type	MAE peak position	error in spectrum
convolutional	0.231 eV	3.9 %
deep tensor	0.186 eV	2.9 %

Ghosh et.al., Adv. Sci. 6, 1970053 (2019)

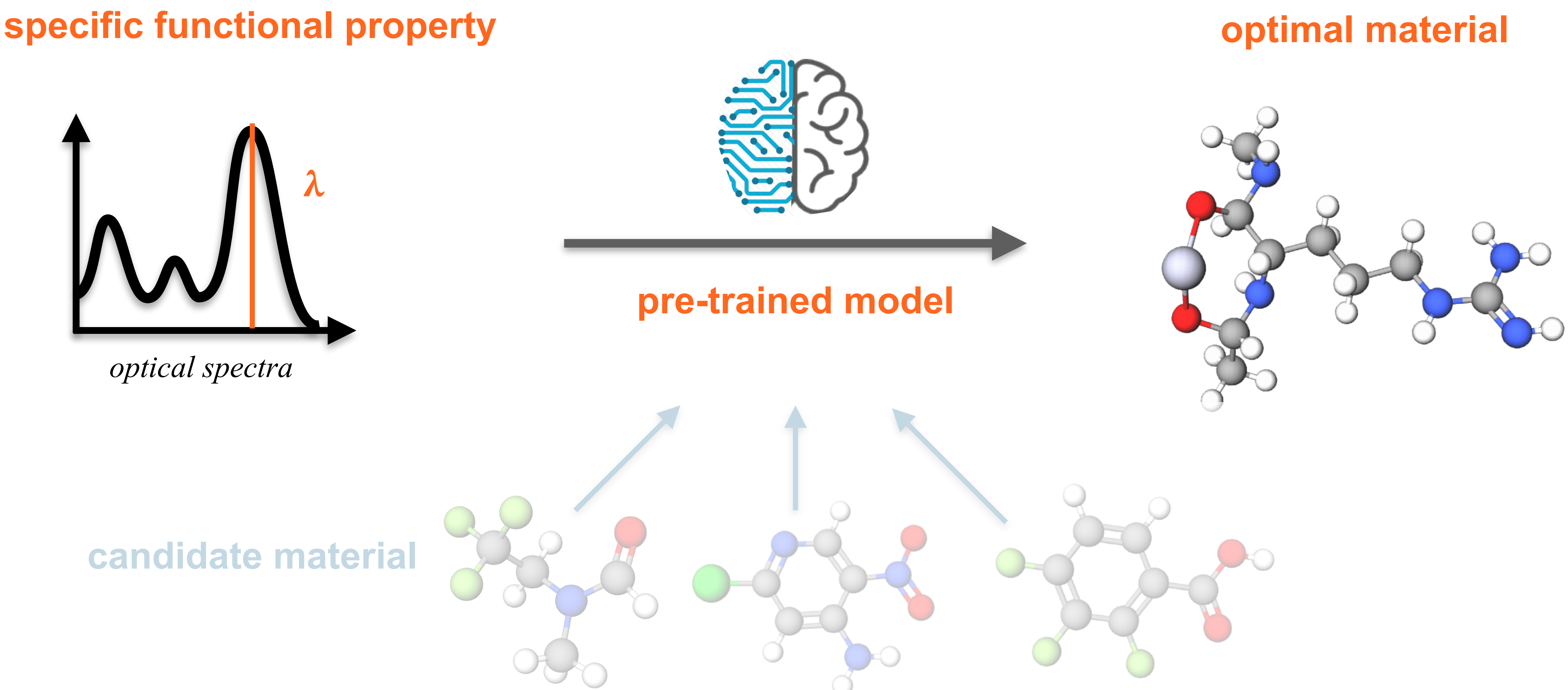
Test application

10k dataset diastereomers of $C_7H_{10}O_2$ → **instant spectral scan**

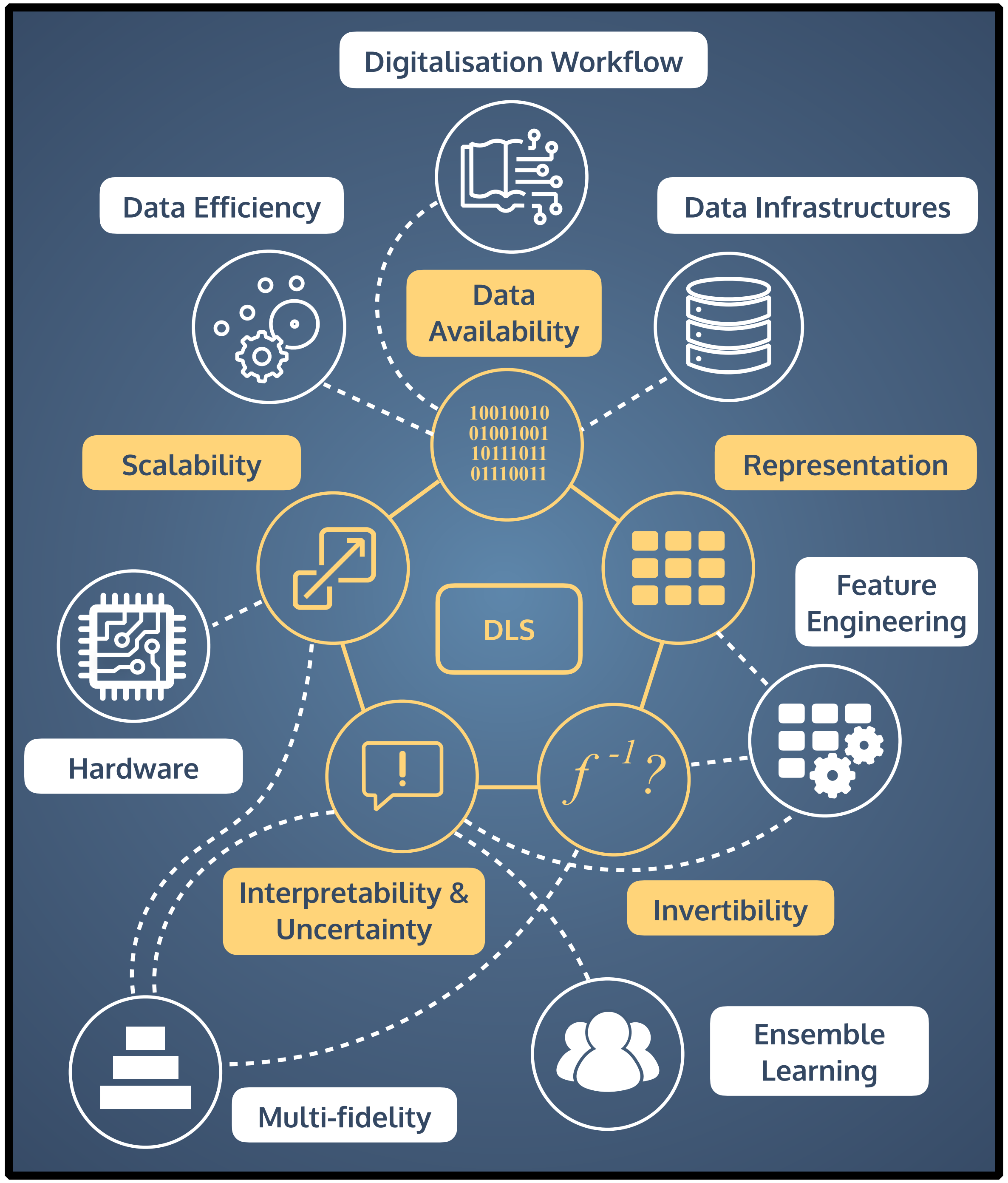


R. Ramakrischnan, P. O. Dral, M. Rupp, O. A. von Lilienfeld, JCTC 11, 2087 (2015)

Pre-screening applications



Challenges in deep learning spectroscopy

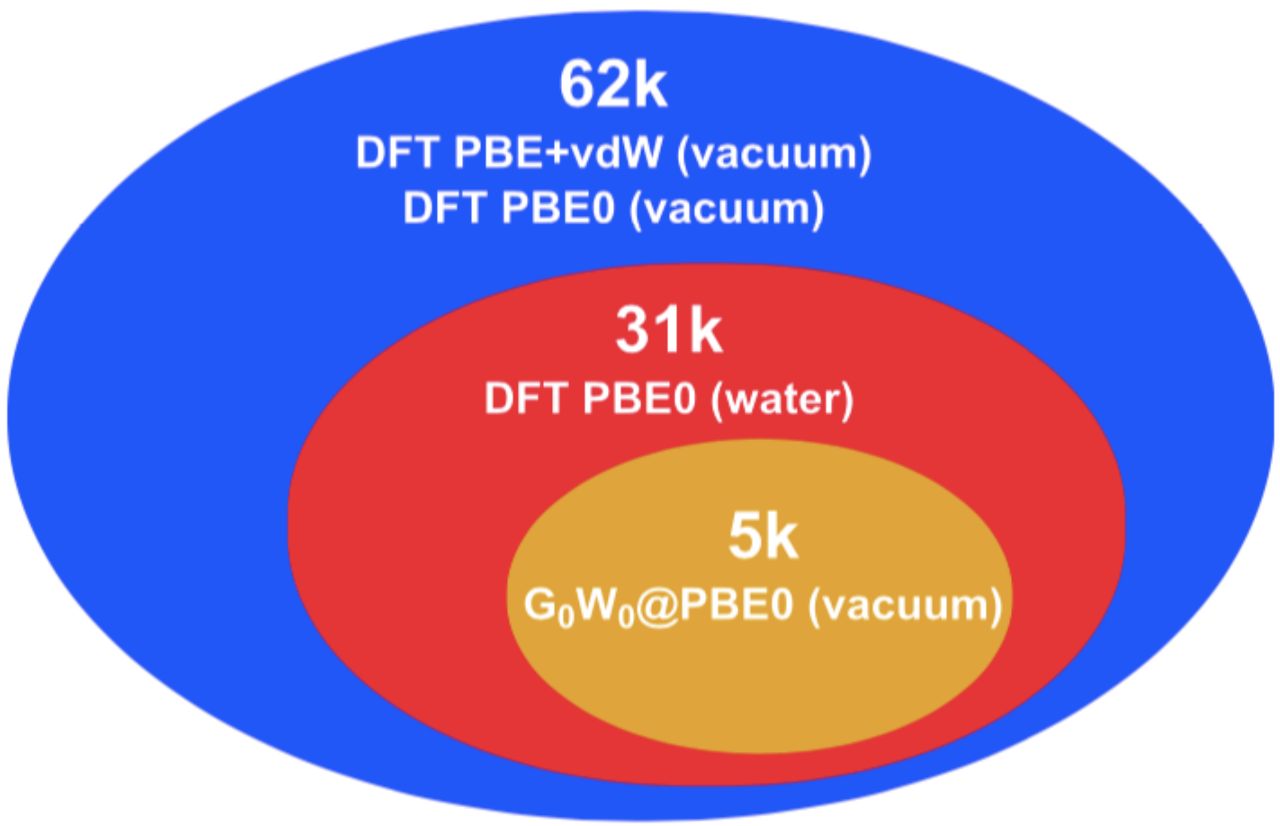


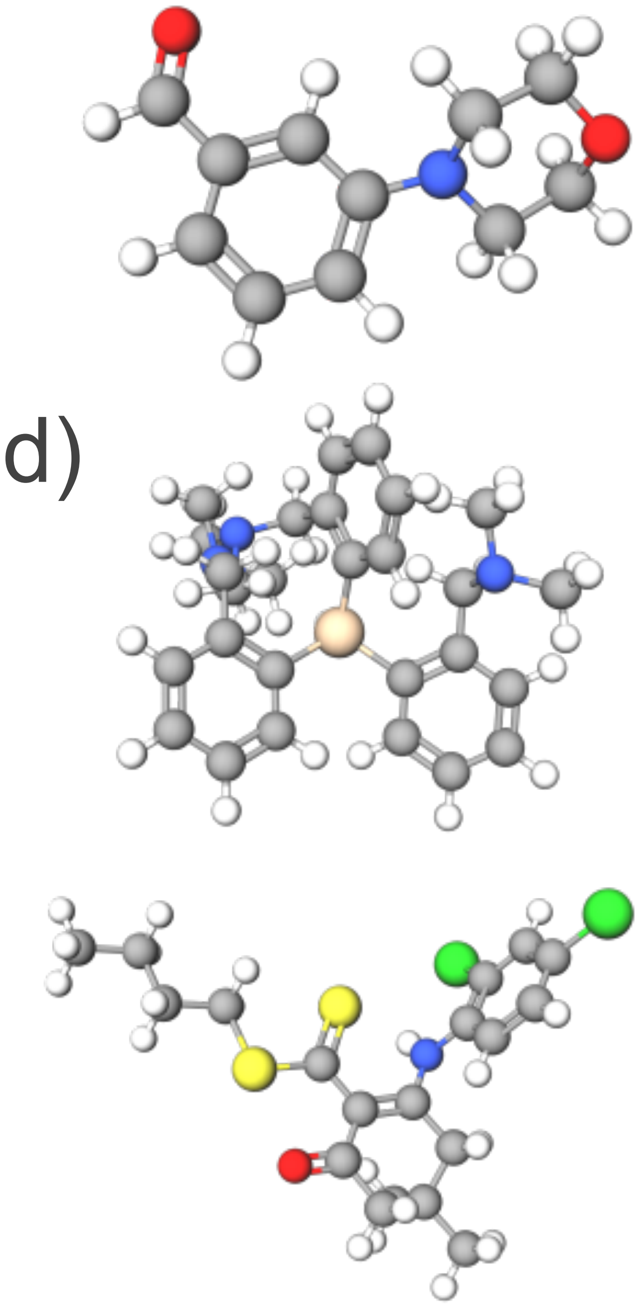
- forward mapping is easy, backward is difficult (invertibility)
- there is too little data (data availability)
- dataset curation is difficult (data digitalization)
- cannot extract physical insight from ML models (interpretability)
- what about noise and uncertainty?

H. Kulik et al., Electron. Struct., doi: 10.1088/2516-1075/ac572f (2022)

Multi-fidelity datasets

- for optoelectronic applications (conjugated)
- extracted from organic crystals
- energies and eigenvalues





OE62
HOMOs

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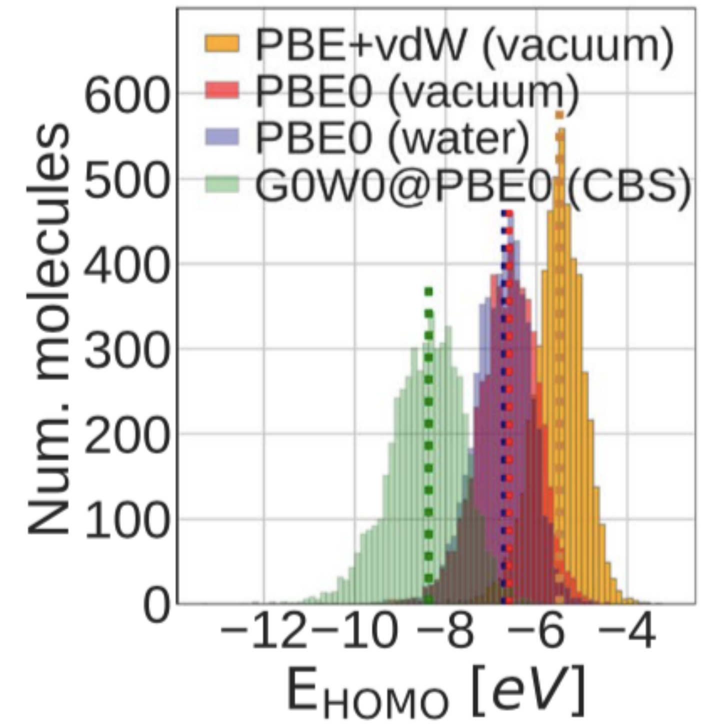
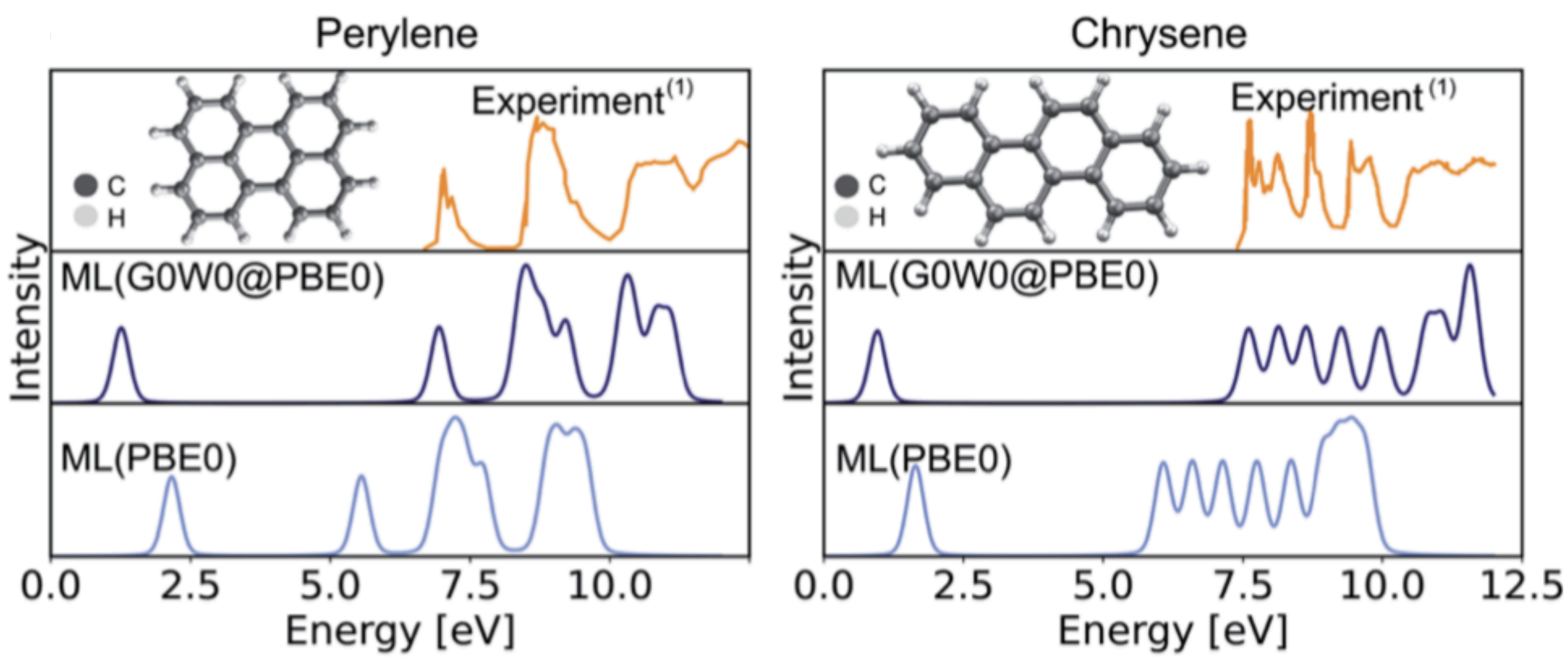
A. Stuke, et al., Sci. Data 7, 241722 (2020)

Multi-fidelity learning



- Δ-learning: learning the difference between PBE0 and G0W0
- ML spectra for MD trajectories



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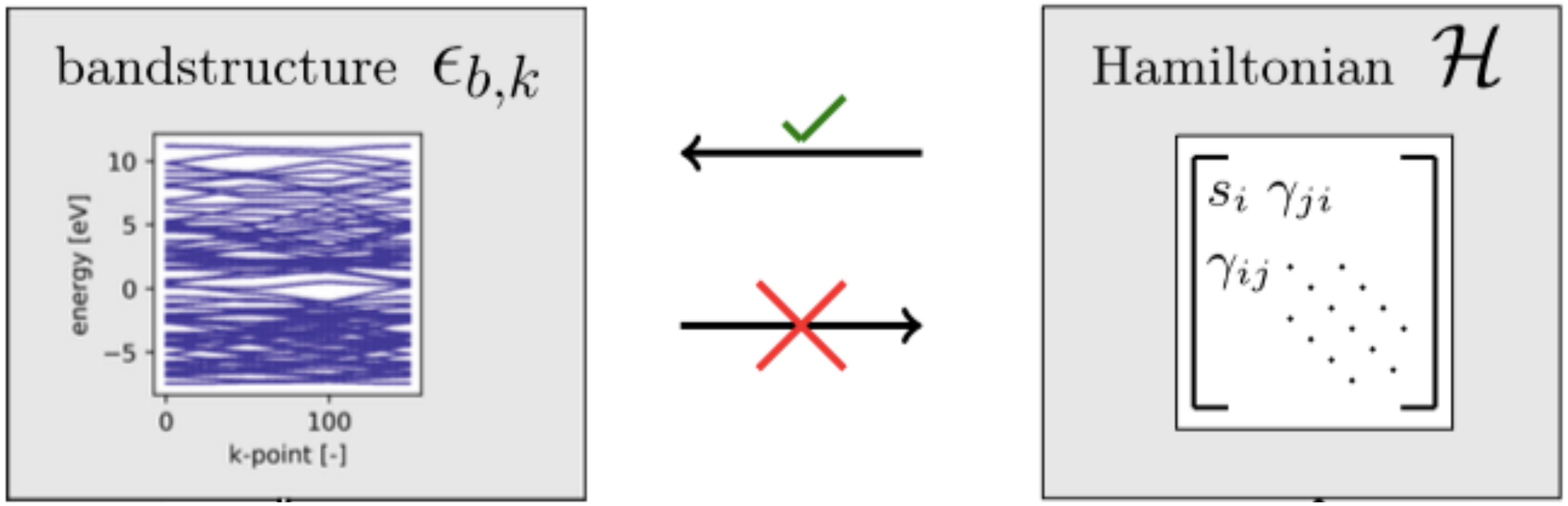
J. Westermayr and R. Maurer, Chem. Sci. 12, 10755 (2021)
J. Westermayr and P. Marquetand, Chem. Rev. 121, 9873 (2021)

Mapping bands to Hamiltonian

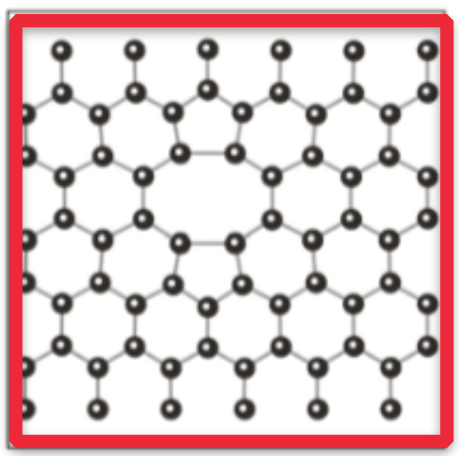


- tight-binding parameterisation for graphene defects
- varied H matrix elements to match reference bands
- variable nearest-neighbour cutoff (up to 10)

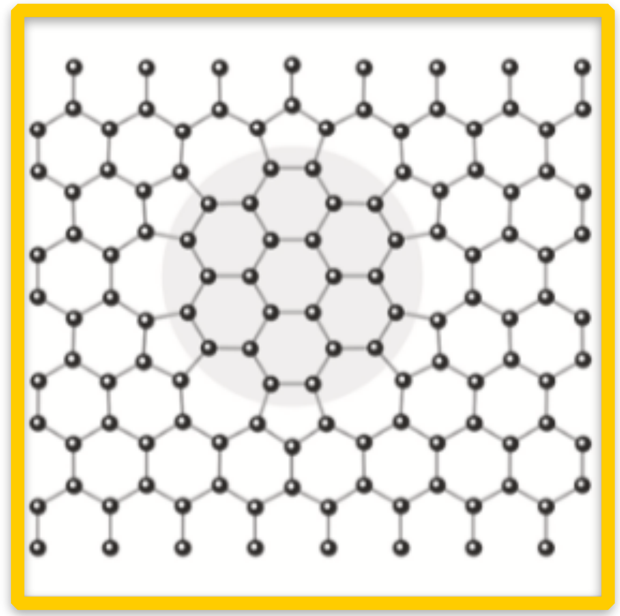
C. Schattauer, et al., npj Comput. Matter 8, 1 (2022)

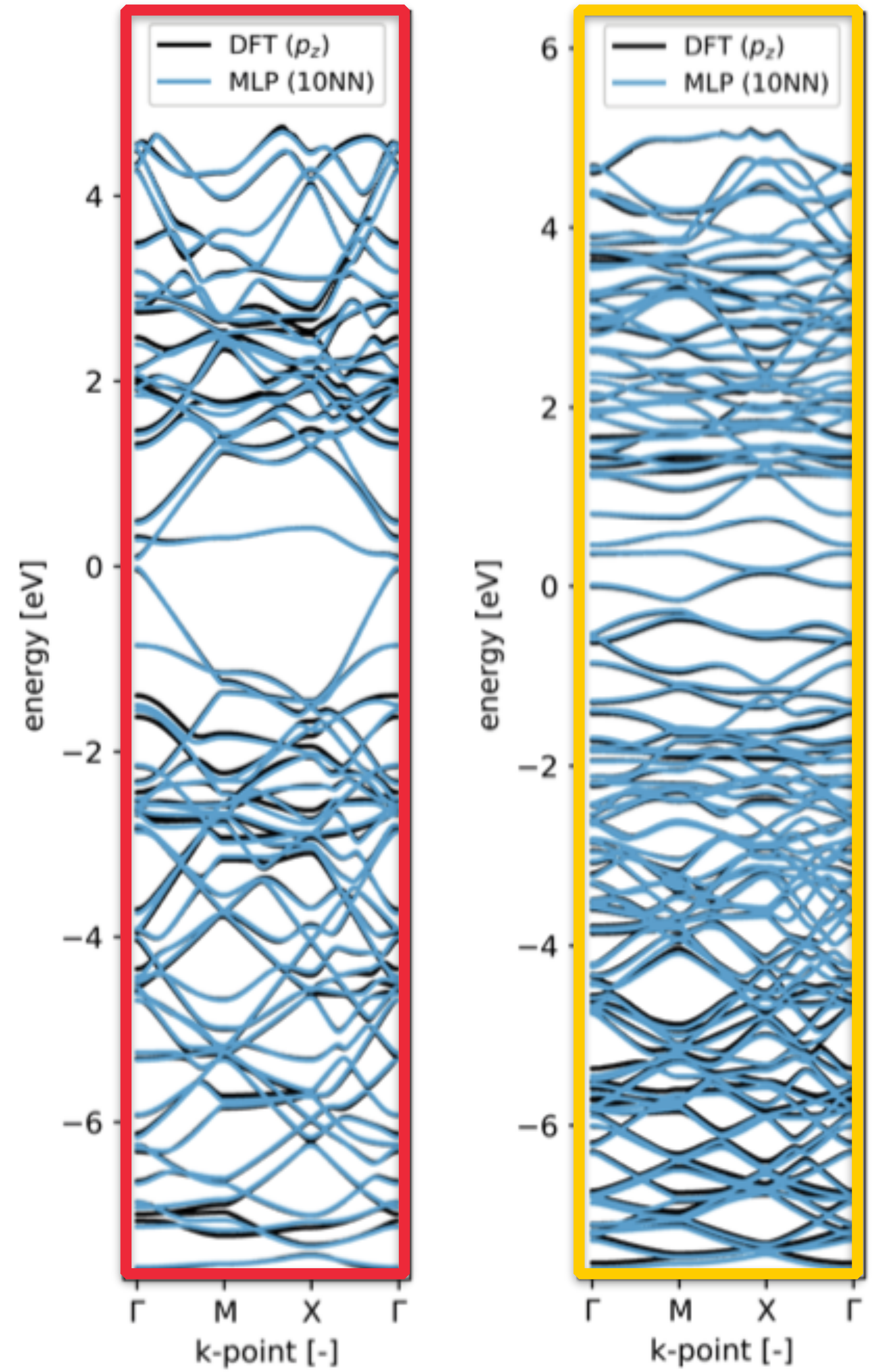
Designer Hamiltonian

vacancy defect



flower defect



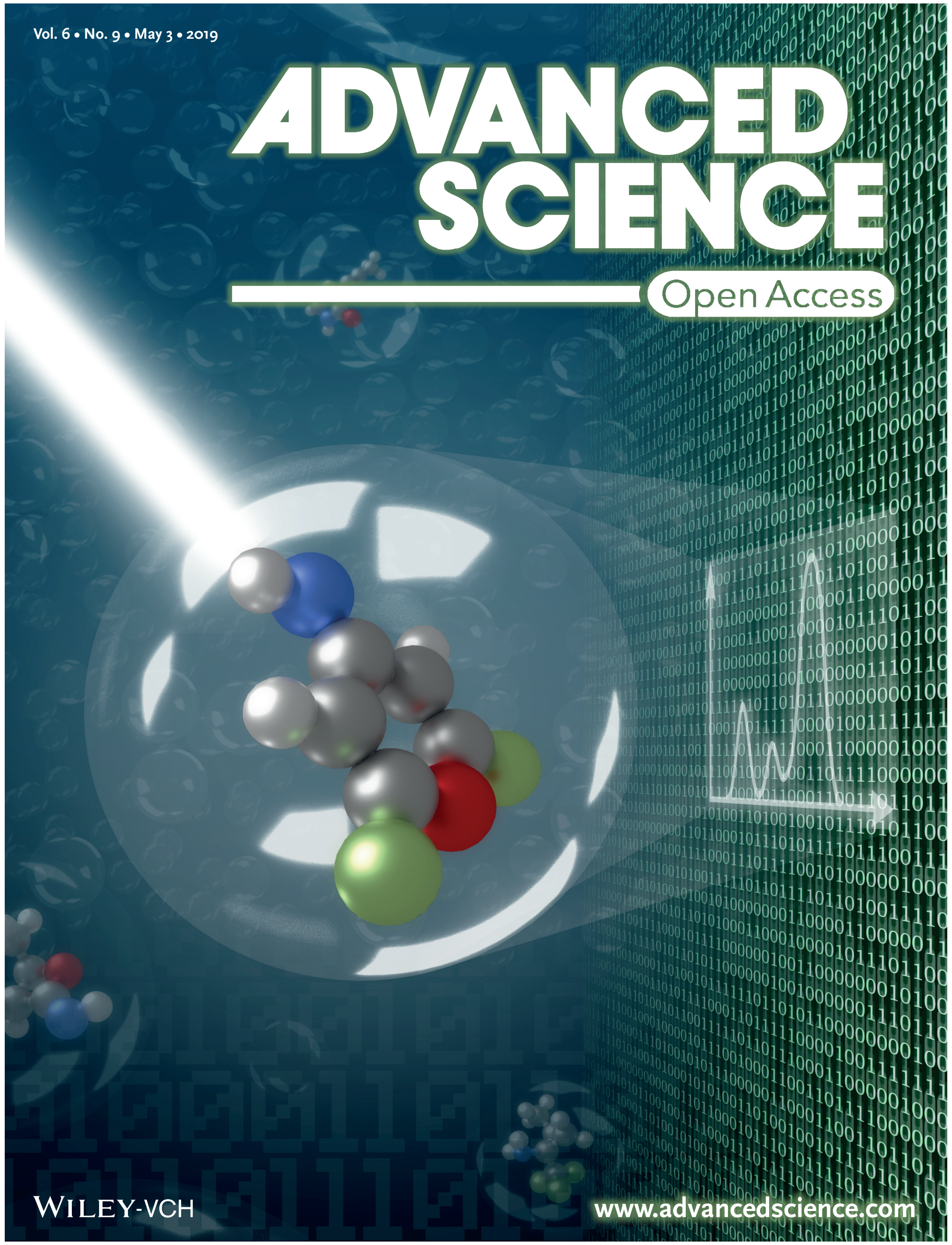


- successfully identified the H
- fast calculation of derived properties
- generalisable to 3D

C. Schattauer, et al., npj Comput. Matter 8, 1 (2022)

Summary

- deep learning can be used to learn spectroscopy!
- **community effort** needed: dataset quality, standards, digitalisation
- **method development** needed: invertibility and uncertainty



Acknowledgements

FCAI Finnish Center for Artificial Intelligence

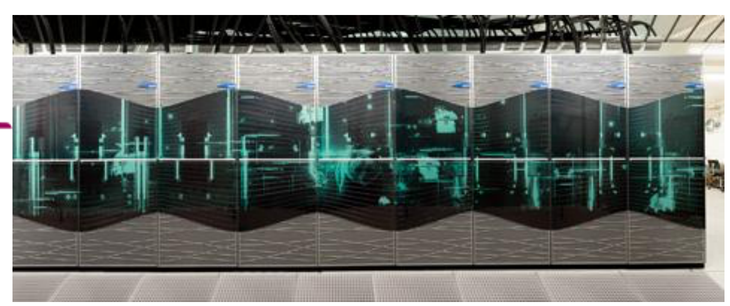
Finnish Center for AI (FCAI) highlight E
AI-driven design of materials

ACADEMY OF FINLAND

AI in Physical Sciences and Engineering
(AIPSE) project 2018-2021

Mahti

CSC



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