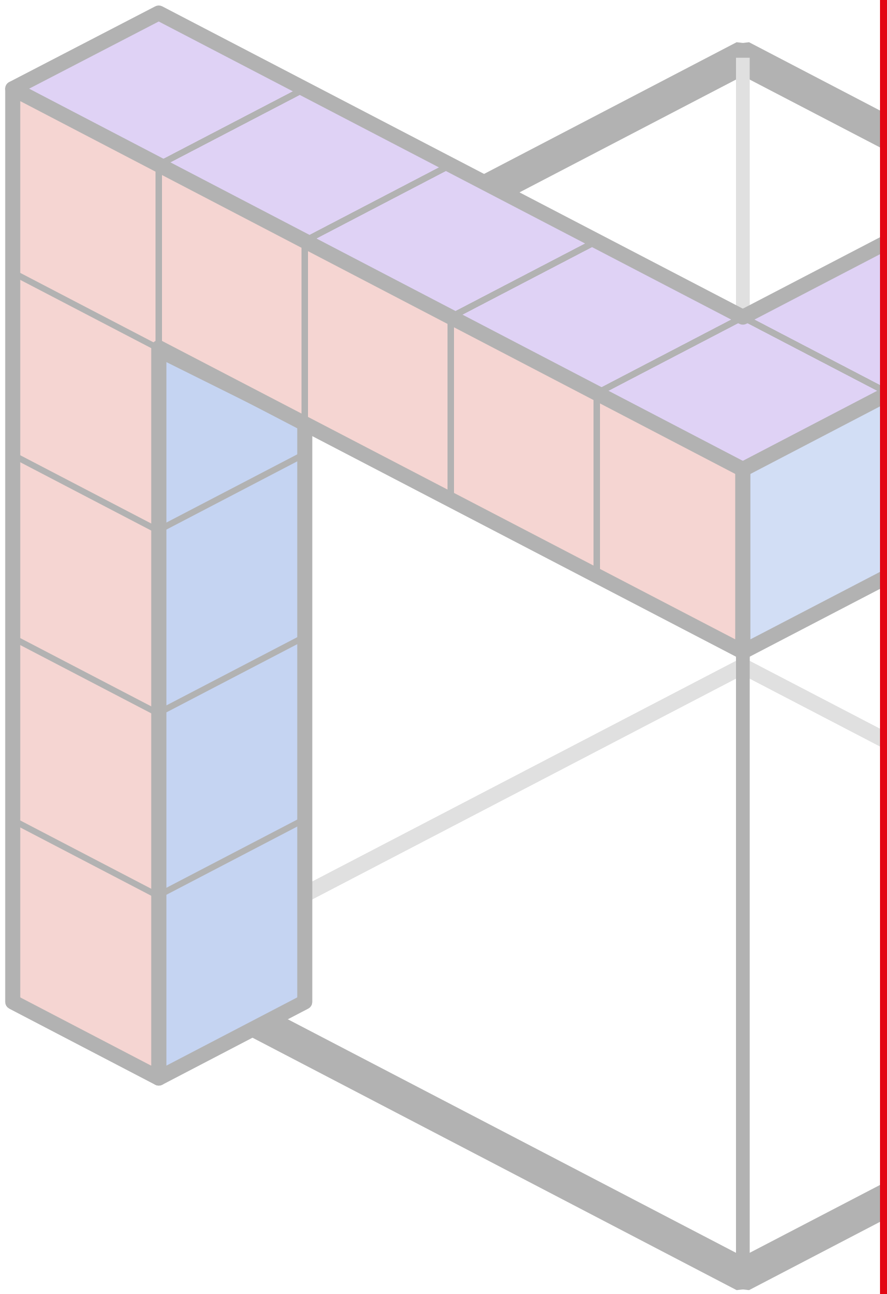
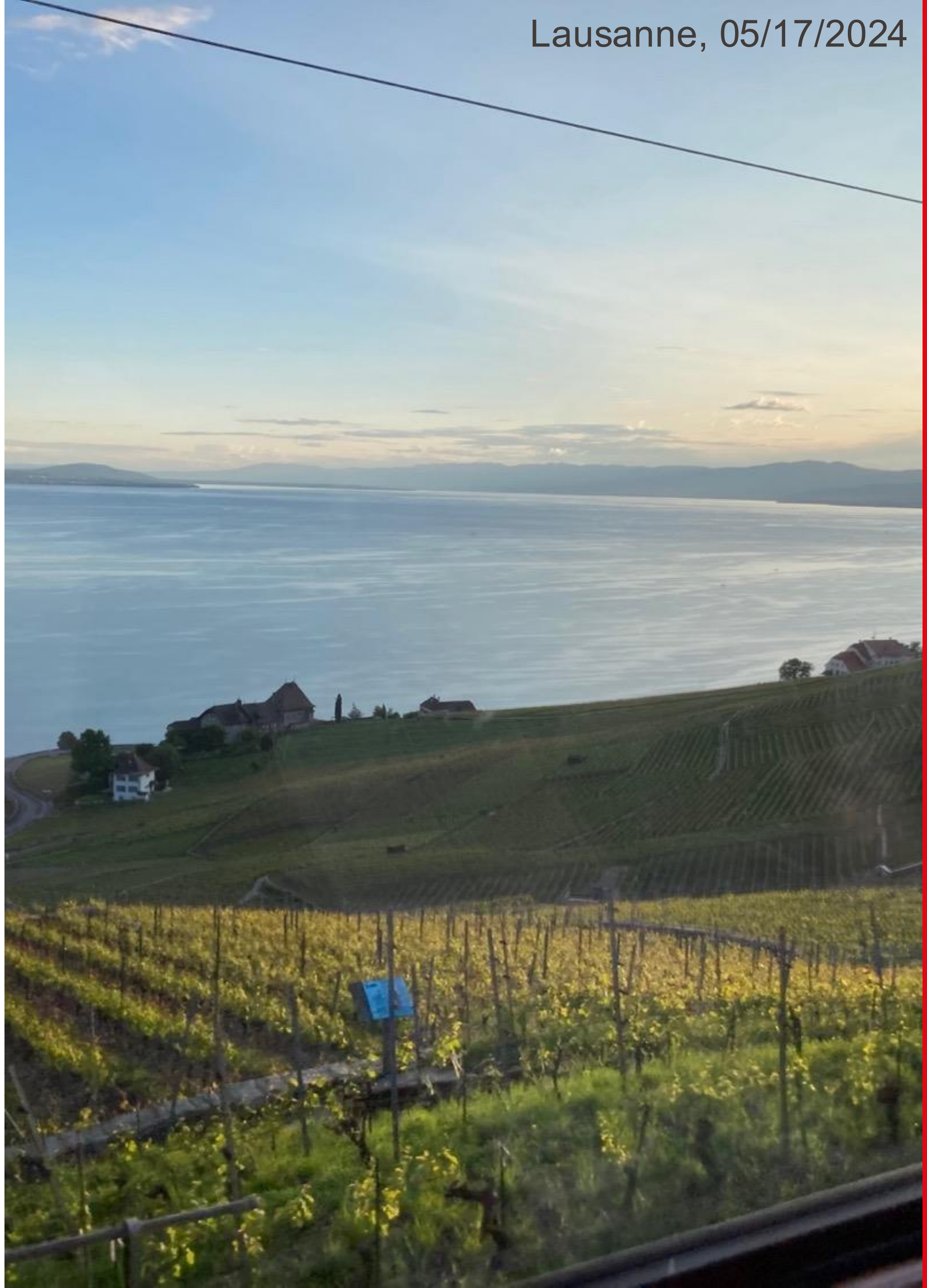


Scientific Applications of the `metatensor` Ecosystem

Flash talks by multiple speakers


Lausanne, 05/17/2024

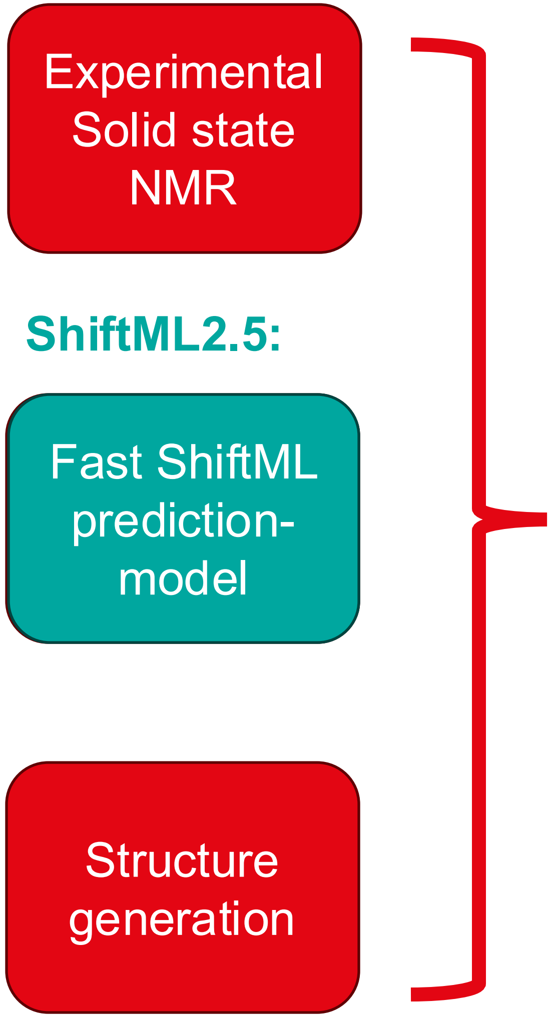
ShiftML 2.5: Fast shielding predictions for organic solids

Matthias Kellner

COSMO
COMPUTATIONAL SCIENCE AND MODELING

LRM
Laboratory of Magnetic Resonance

NMR crystallography

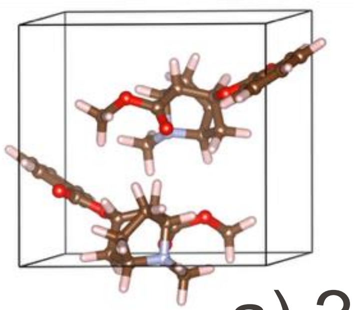


Experimental Solid state NMR

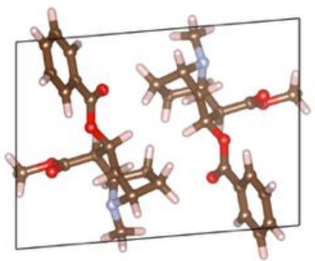
ShiftML2.5:

Fast ShiftML prediction-model

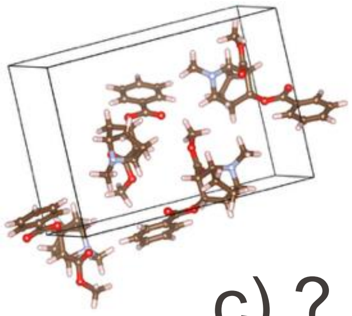
Structure generation



a) ?



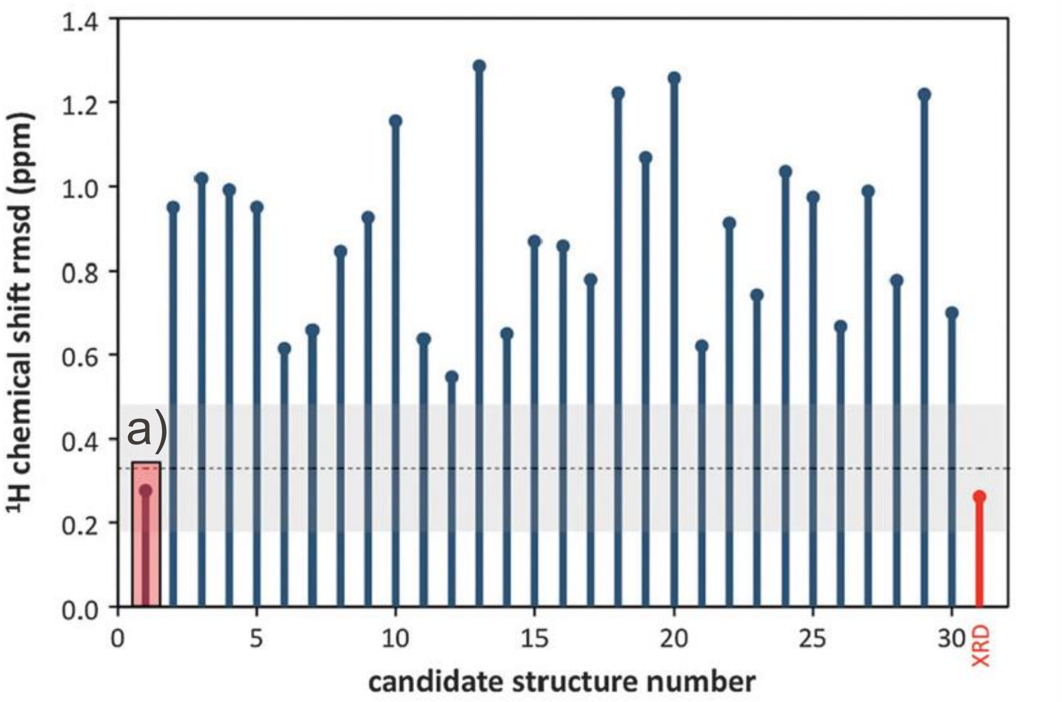
b) ?



c) ?

[1] Baias et al. doi: 10.1039/c3cp41095a

Compare ab initio shielding calculations (GIPAW) with Experimentally measured ones and choose candidate that agrees most.

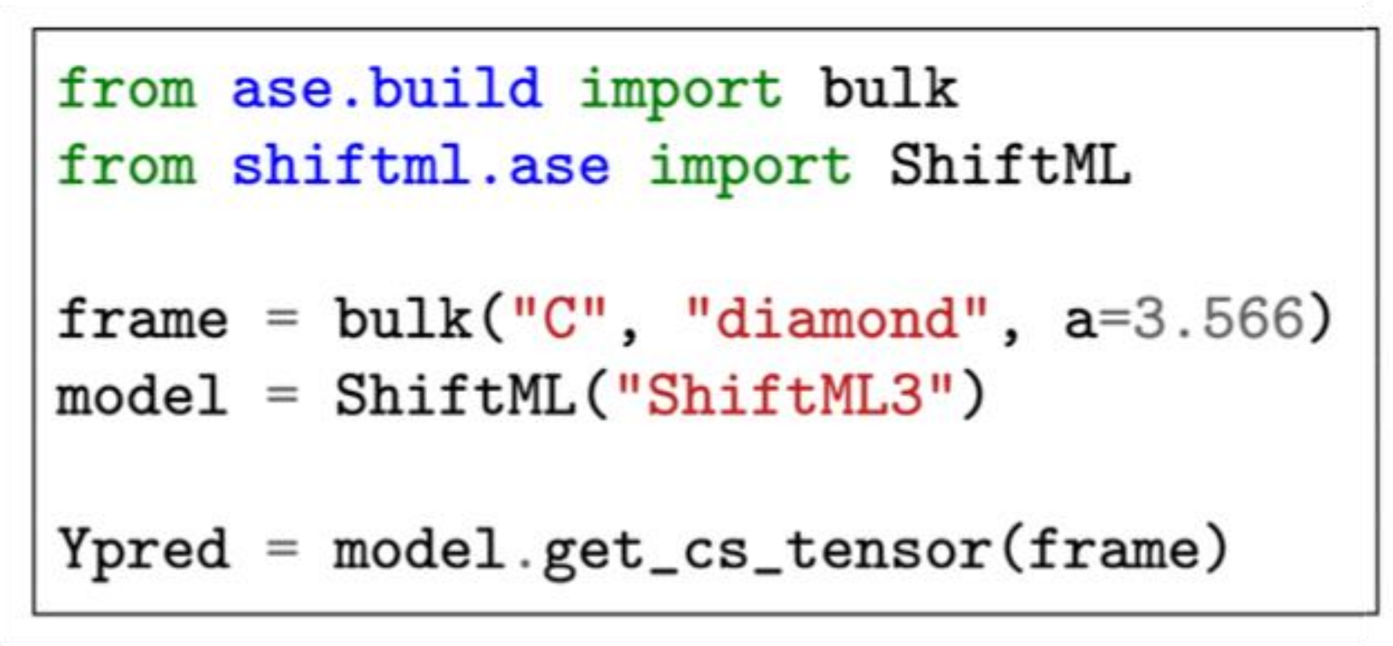


Advantages over XRD crystallography?

- Works with powdered samples
- And amorphous drug formulations

How does metatensor help?

1) ML-Models for tensorial properties (`metatrain`)
2) Deploy models (`metatomic`)

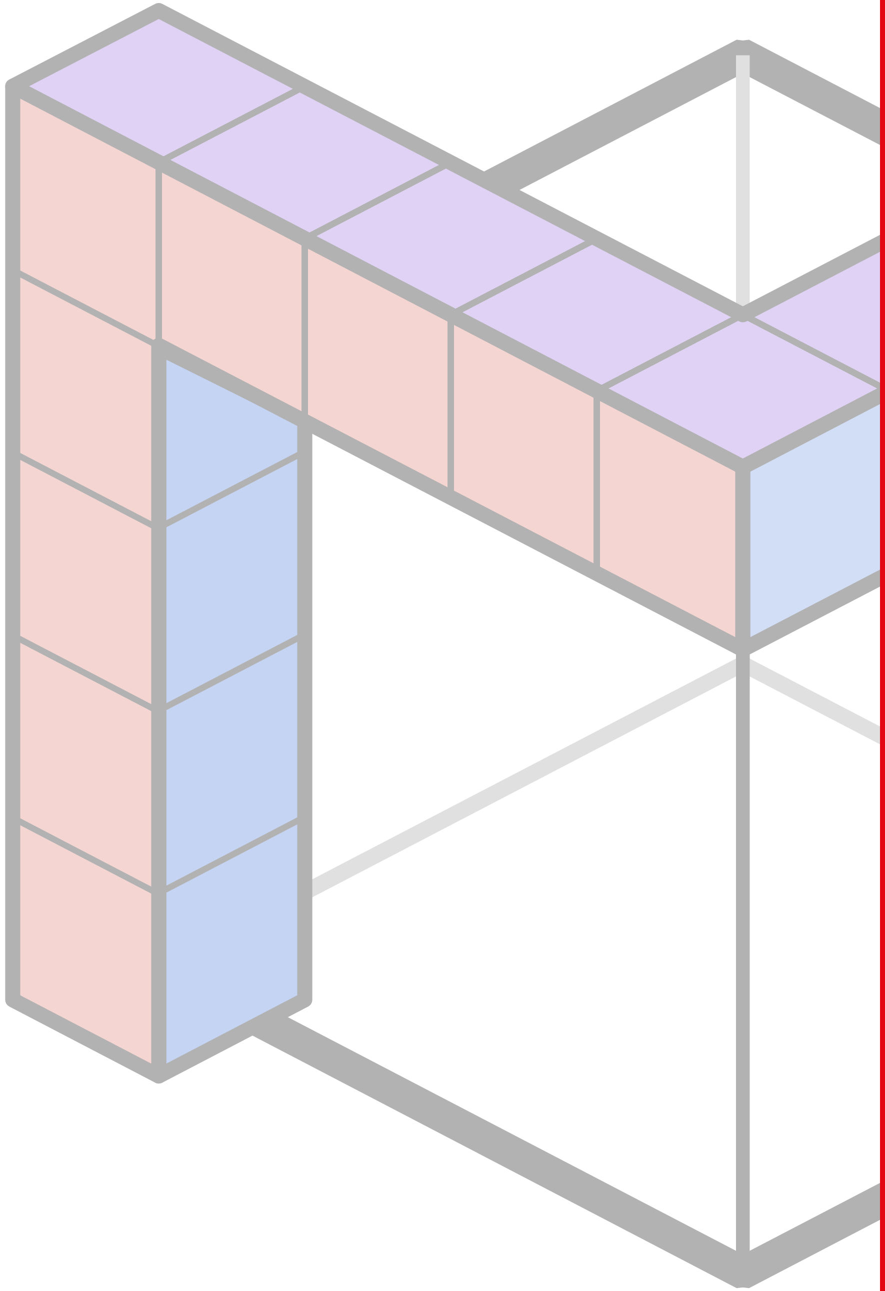


```python
from ase.build import bulk
from shiftml.ase import ShiftML

frame = bulk("C", "diamond", a=3.566)
model = ShiftML("ShiftML3")

Ypred = model.get_cs_tensor(frame)
```

Deploy model and make it installable on various operating systems and architectures!

PyPI release soon:

`lab-cosmo/ShiftML`

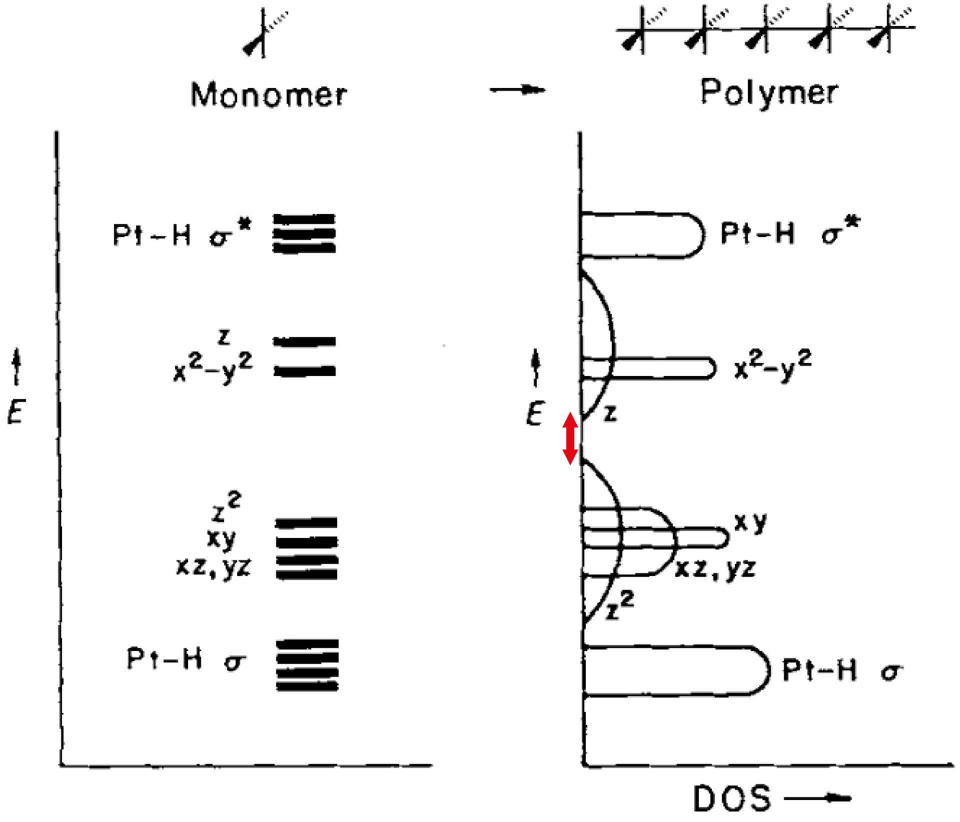


Machine Learning the Electronic Density of States

Wei Bin How

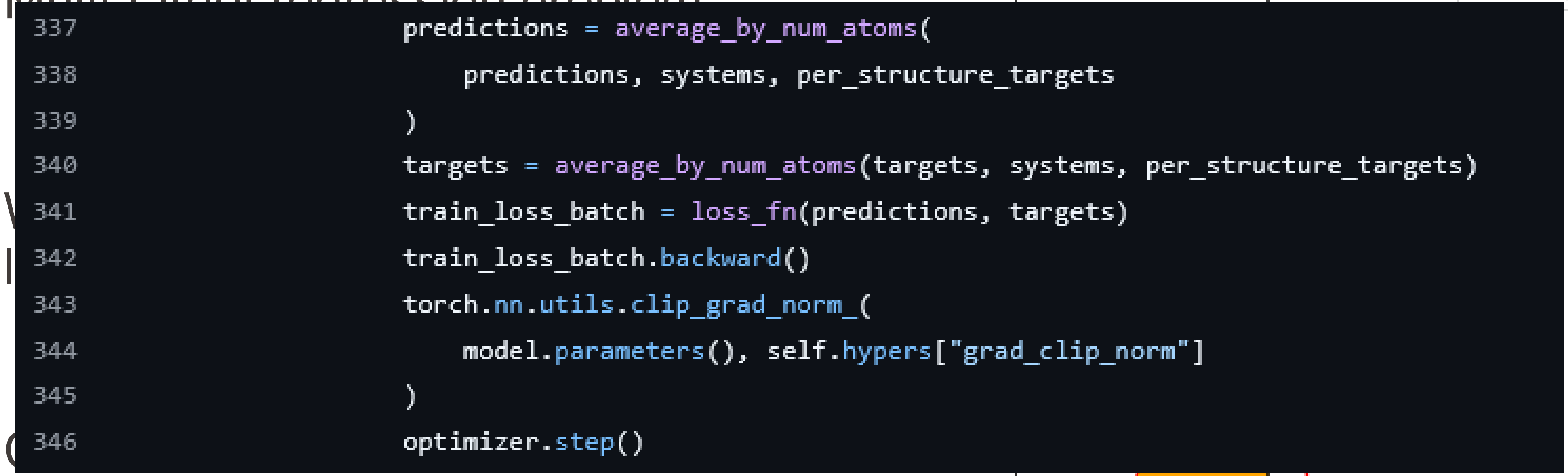
Density of States

- Bulk analogue to energy levels in discrete molecules
- Useful for predicting and explaining the optoelectronic properties of materials
  - Bandgaps – Optical absorption and conductivity
  - Electronic heat capacity



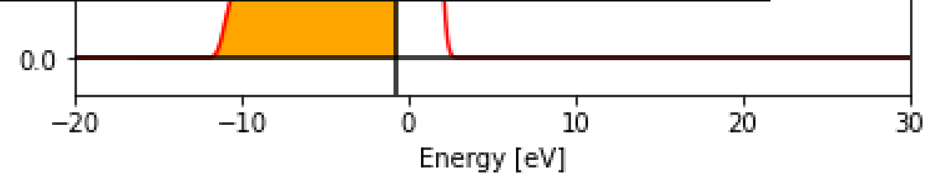
Density of States

- Multi-target regression problem

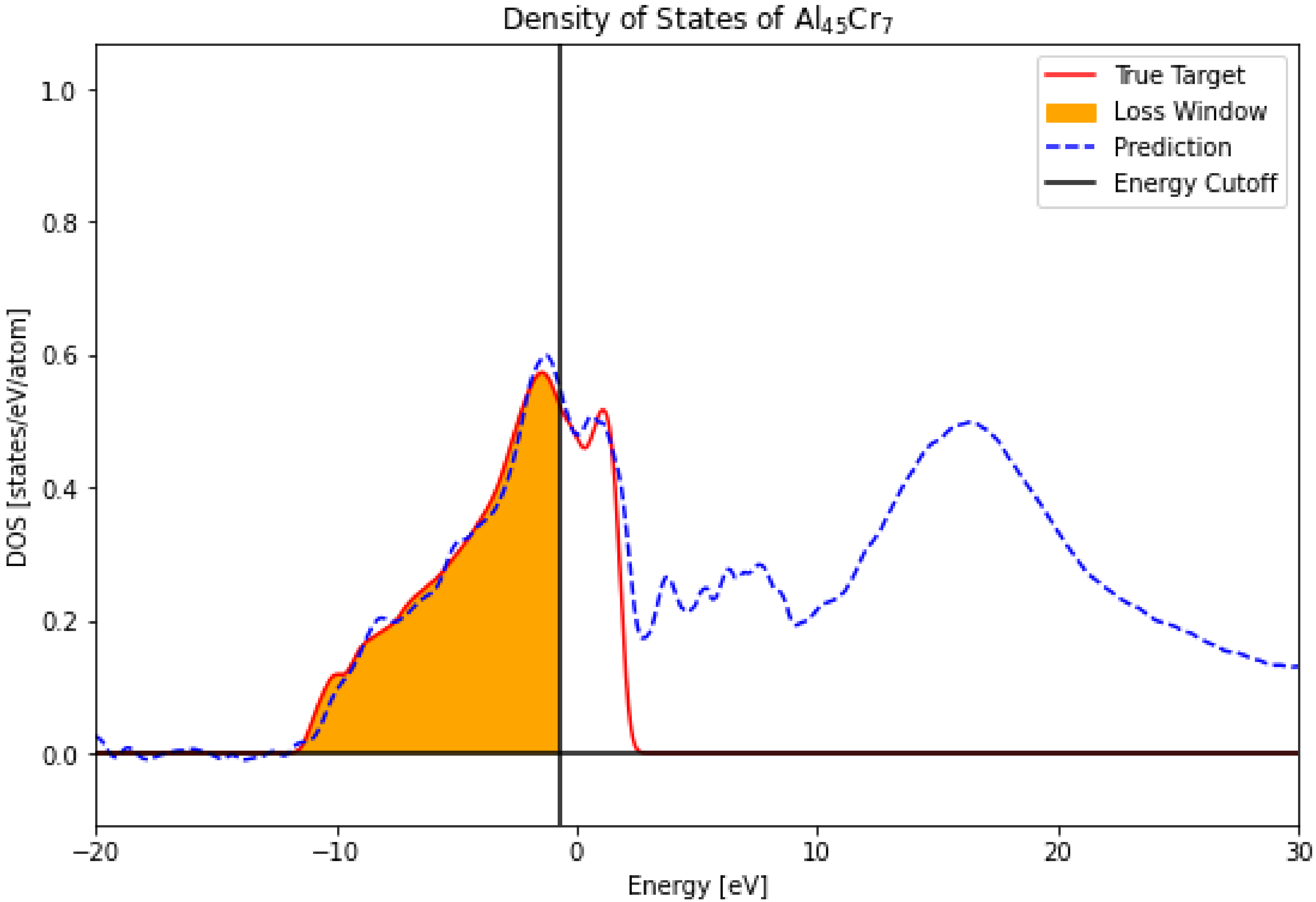


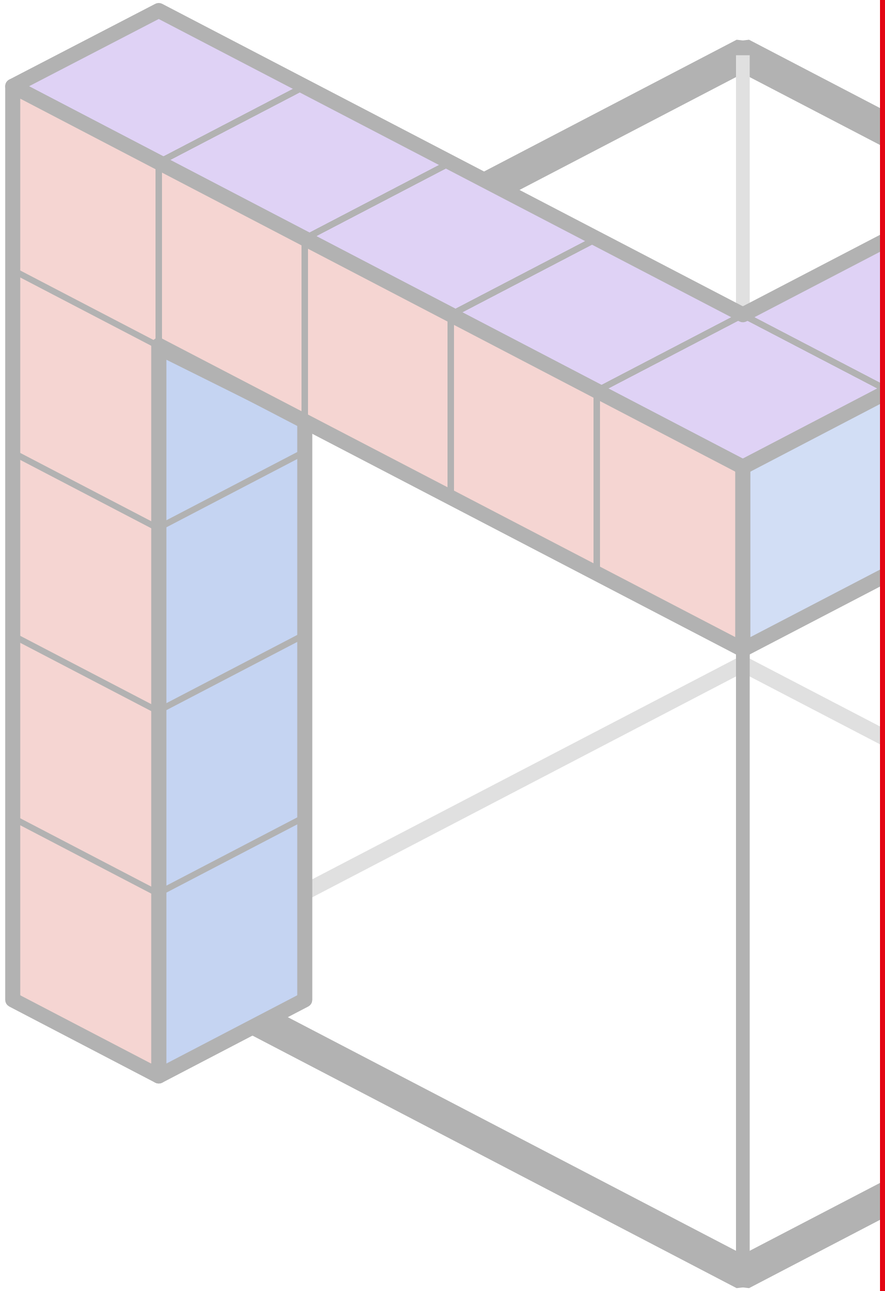
```
                    predictions = average_by_num_atoms(
                        predictions, systems, per_structure_targets
                    )
                    targets = average_by_num_atoms(targets, systems, per_structure_targets)
                    train_loss_batch = loss_fn(predictions, targets)
                    train_loss_batch.backward()
                    torch.nn.utils.clip_grad_norm_(
                        model.parameters(), self.hypers["grad_clip_norm"]
                    )
                    optimizer.step()
```

- V
  I
- C
  - Customizable evaluation window
  - Overcomes finite eigenvalue issue
  - Crucial for highly diverse datasets



Density of States





The λ-MCoV model
(Maximally Coupled Vector)

Michelangelo Domina

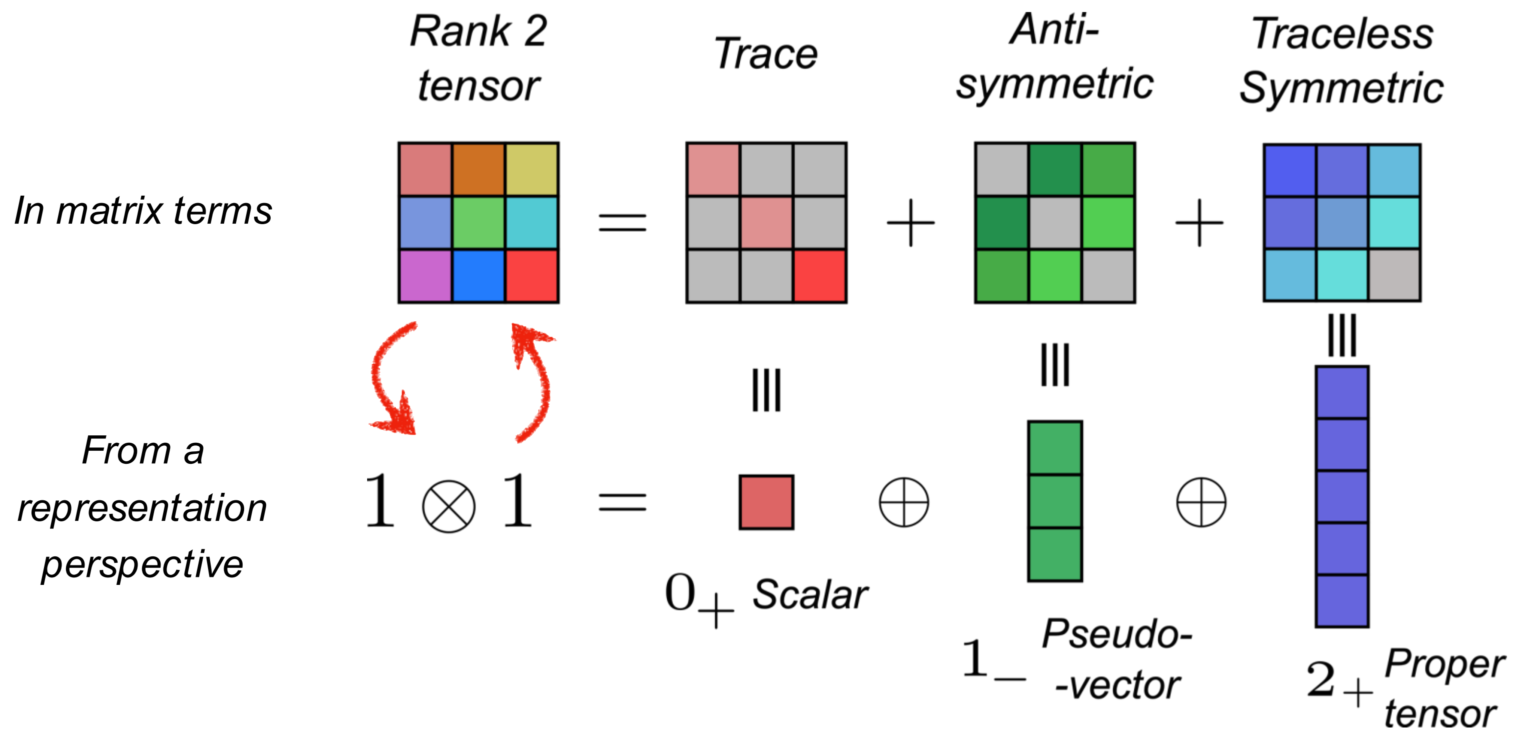
Together with:

Filippo
Bigi

Paolo
Pegolo

Our targets: irreducible tensors

A rank 2 (Cartesian) example



These are our targets

Physically relevant rank 2 tensors:

- *Moment of inertia*
- *Stress tensor*
- *Polarizability tensor*
- *Susceptibility tensor*
- *Conductivity tensors*
- *Hessian Matrix*
- …

The Architecture

A scalar-based equivariant model

arXiv > physics > arXiv:2505.05404

Physics > Chemical Physics

[Submitted on 8 May 2025]

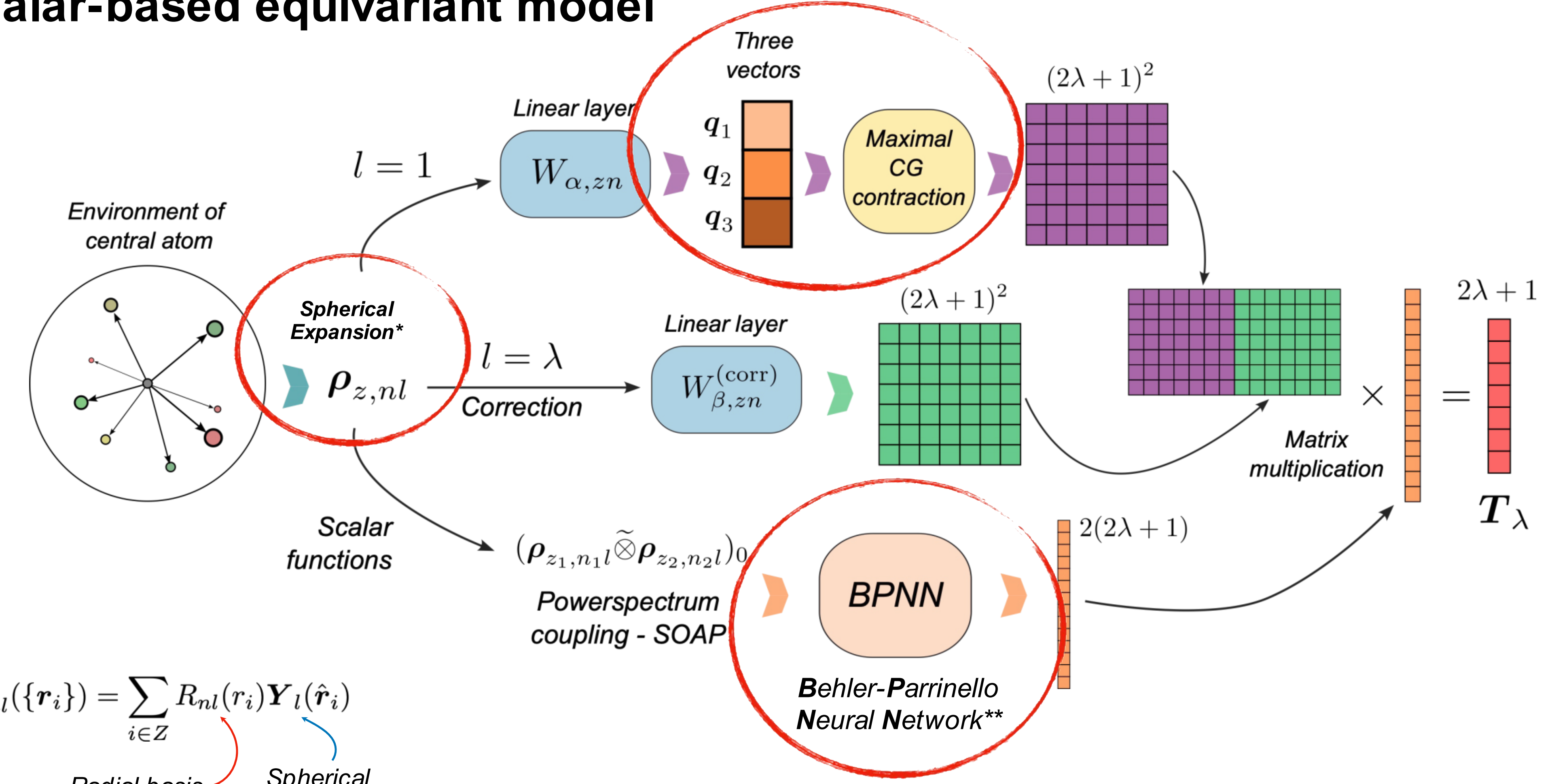
Representing spherical tensors with scalar-based machine-learning models

Michelangelo Domina, Filippo Bigi, Paolo Pegolo, Michele Ceriotti

If curious, read the pre-print!

Core of the architecture

Based on our mathematical results



$$^{*}\,\boldsymbol{\rho}_{Z,nl}(\{\boldsymbol{r}_i\}) = \sum_{i\in Z} R_{nl}(r_i)\boldsymbol{Y}_l(\hat{\boldsymbol{r}}_i)$$

Radial basis

Spherical harmonics

Phys. Rev. Lett. **98, 146401 (2007)

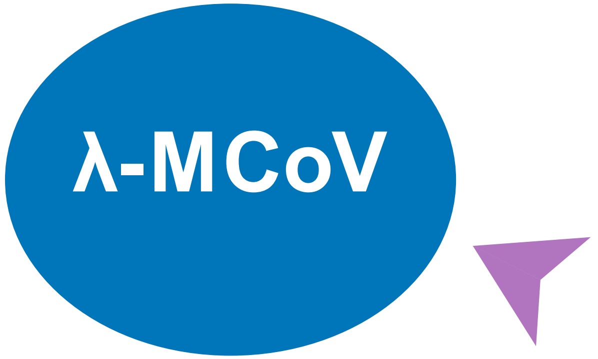
MICHELANGELO DOMINA

METATENSOR ECOSYSTEM WORKSHOP

λ-MCoV

*All the scalar functions are modeled by a **Behler-Parrinello NN**, with **SOAP** powerspectrum as features*

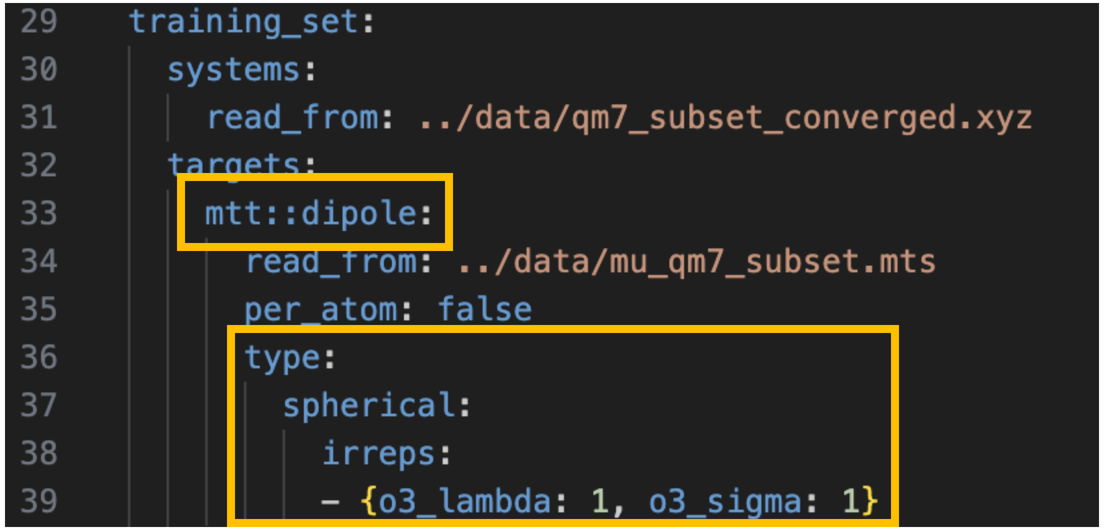
*All the features **are spherical expansion-based** with almost no coupling (only **maximal**) involved*



*Seamless inclusion of **non-linearity***

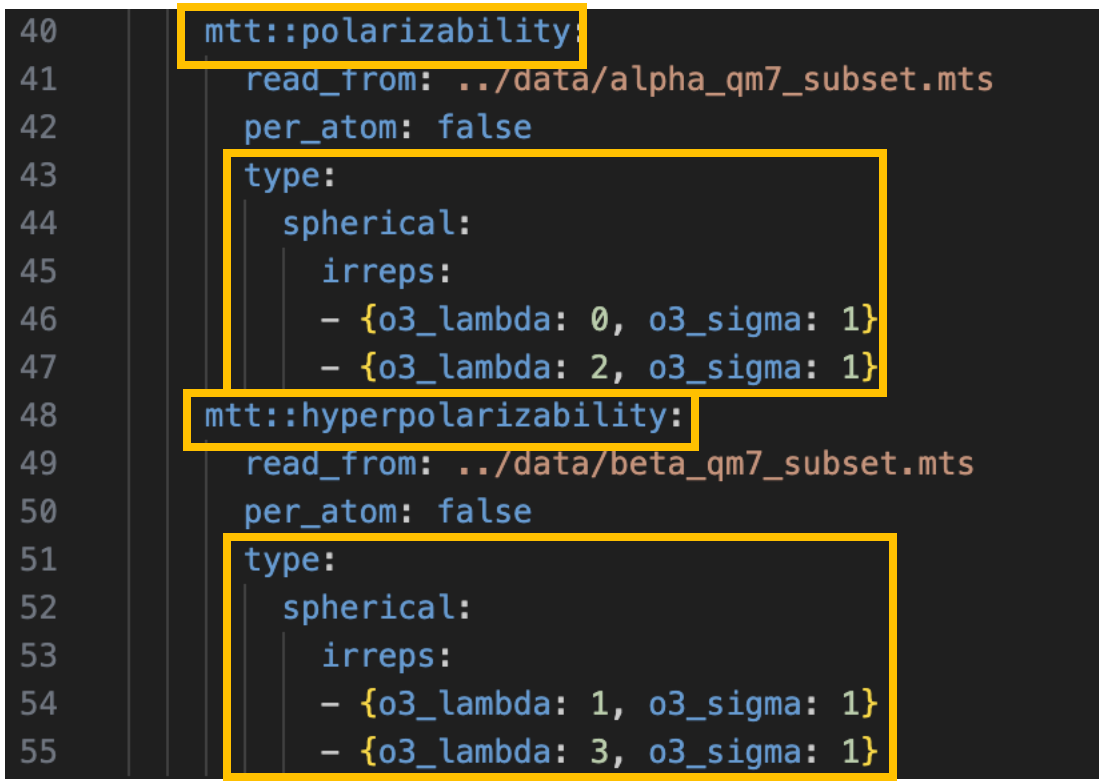
***Minimal architecture** for the scalar functions*

Use the model with metatrain



```yaml
training_set:
  systems:
    read_from: ../data/qm7_subset_converged.xyz
  targets:
    mtt::dipole:
      read_from: ../data/mu_qm7_subset.mts
      per_atom: false
      type:
        spherical:
          irreps:
          - {o3_lambda: 1, o3_sigma: 1}
```

options.yaml



```yaml
    mtt::polarizability:
      read_from: ../data/alpha_qm7_subset.mts
      per_atom: false
      type:
        spherical:
          irreps:
          - {o3_lambda: 0, o3_sigma: 1}
          - {o3_lambda: 2, o3_sigma: 1}
    mtt::hyperpolarizability:
      read_from: ../data/beta_qm7_subset.mts
      per_atom: false
      type:
        spherical:
          irreps:
          - {o3_lambda: 1, o3_sigma: 1}
          - {o3_lambda: 3, o3_sigma: 1}
```

Simultaneous *irreducible targets (presented in TensorMaps):*

- *Dipole*
- *Polarizability*
- *Hyper-polarizability*

Train and evaluate

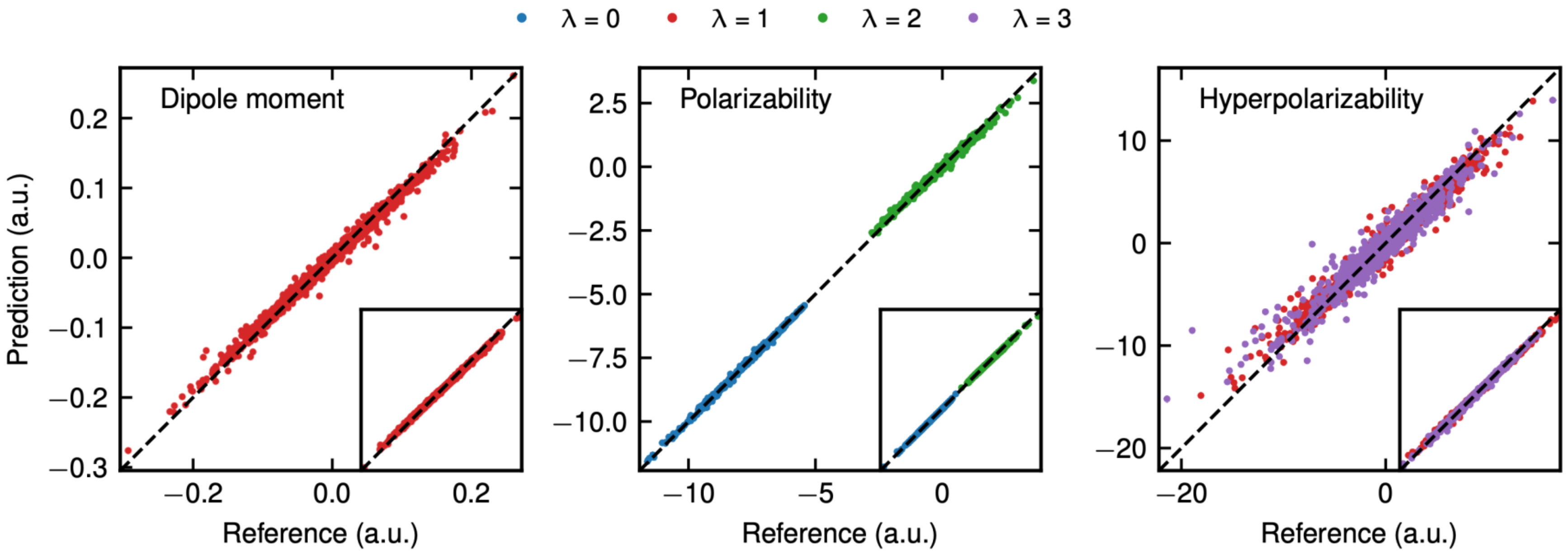
As easy as running

```
% mtt train options.yaml
```

```
% mtt export model.ckpt −o model.pt
```

```
% mtt eval model.pt eval.yaml −b 128 −e extensions −o outputs.mts
```

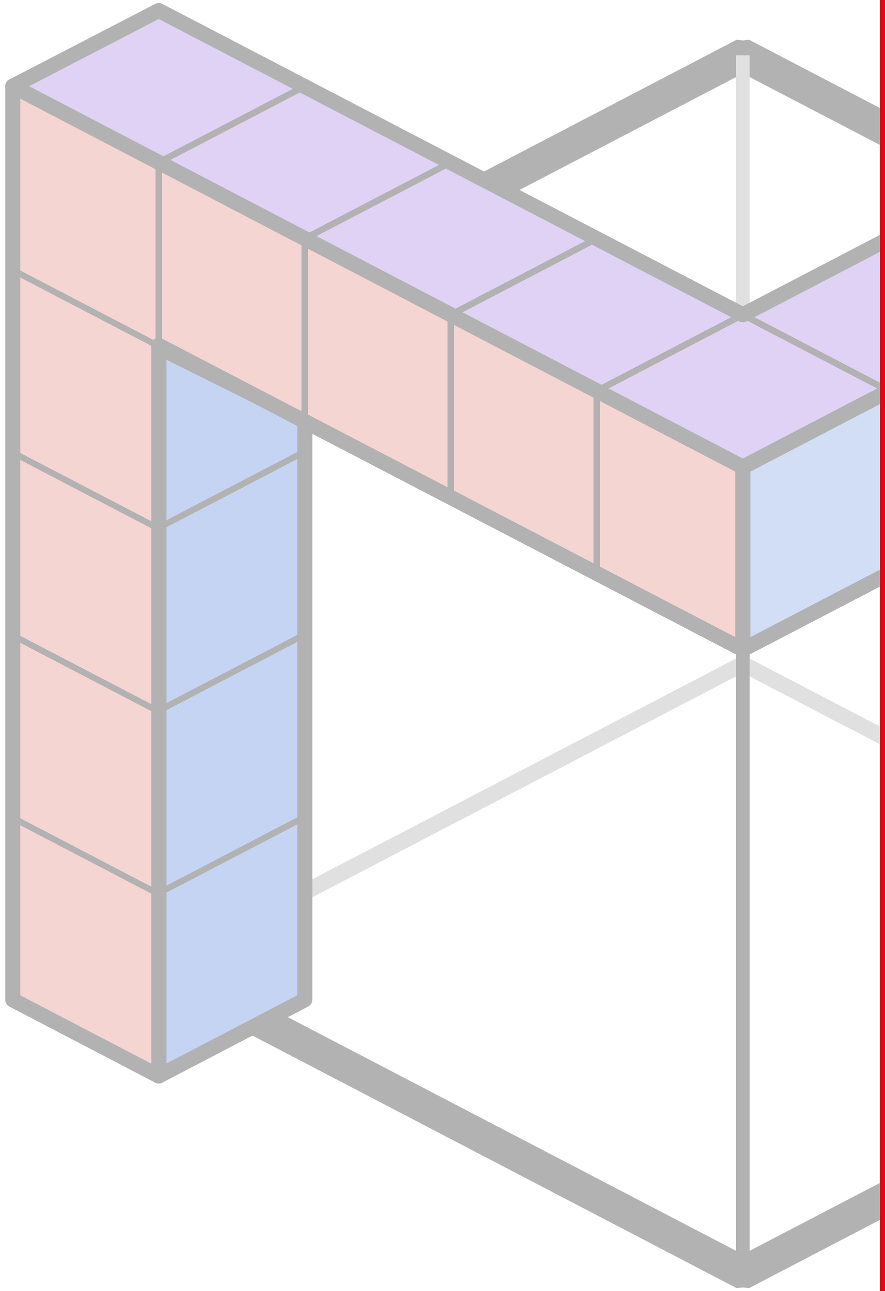
***Train** the model* ▶ ***Export** the model* ▶ ***Evaluate** the model*



*Evaluated on data calculated from the **QM7** dataset*

- *J. Am. Chem. Soc.* ***131****, 9732 (2009)*
- *Phys. Rev. Lett.* ***108****, 059301 (2012)*

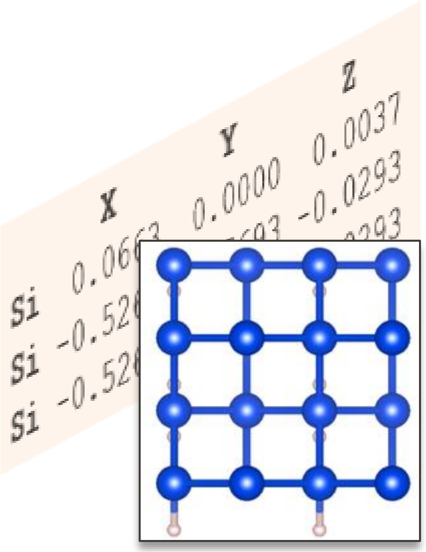
Materials Cloud https://doi.org/10.24435/materialscloud:zq-a6



Learning electronic structure targets with `metatensor`

Joseph W. Abbott

Surrogate models for electronic structure



first principles quantum models

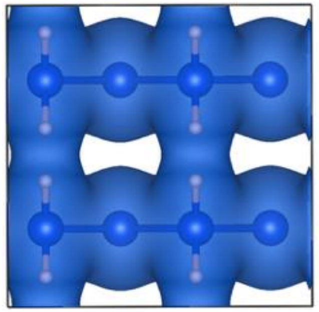
i.e. Kohn-Sham DFT

$$HC = SC\epsilon$$

$$\psi(\boldsymbol{r}) = \sum_i c_i \phi_i(\boldsymbol{r})$$

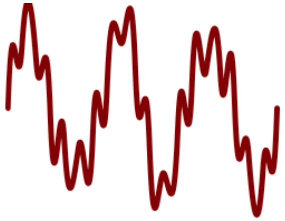
KS-orbitals

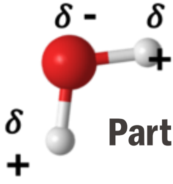
Hamiltonian, H



Electronic density, ρ

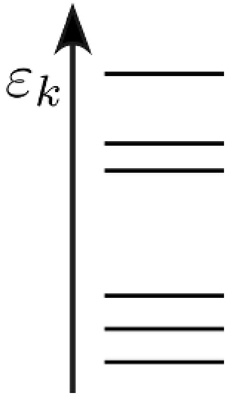
Potential energy surface, V





Partial charges

Dipole moment

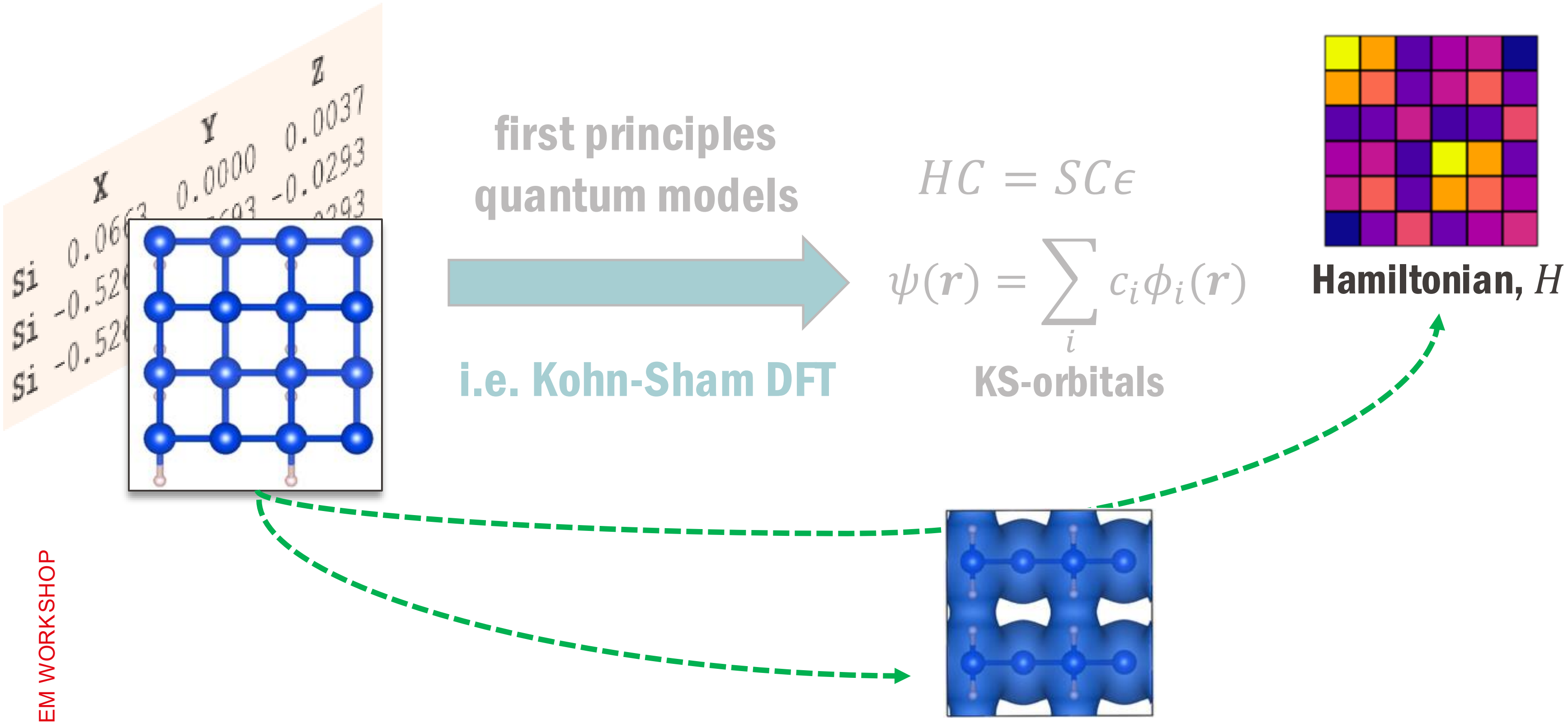


MO energies / bands

Electronic structure methods:

- **Calculate** electronic structure to **understand** structure-property relationships
- Problem: system-size scaling, expensive for large systems

Surrogate models for electronic structure



first principles quantum models

i.e. Kohn-Sham DFT

$HC = SC\epsilon$

$\psi(\mathbf{r}) = \sum_i c_i \phi_i(\mathbf{r})$

KS-orbitals

Hamiltonian, H

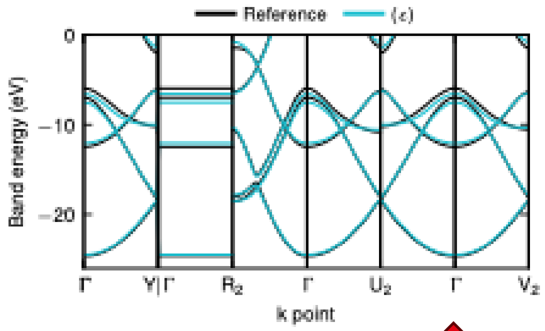
Electronic density, ρ

Why?

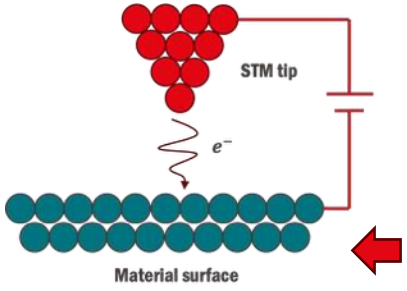
→ Speed up SCF

→ 1-step access to electronic properties

→ Directly probe electronic structure



Band diagrams via periodic Hamiltonians

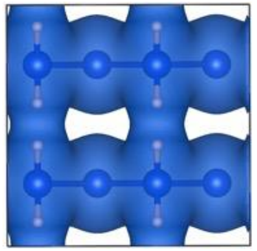


Predicting surface electronic structure in slab systems

Data-driven surrogate models:

- Trained on electronic structure data
- **Predicts** targets at a fraction of the cost

Predict electronic structure targets *on a basis*



Electronic density, ρ

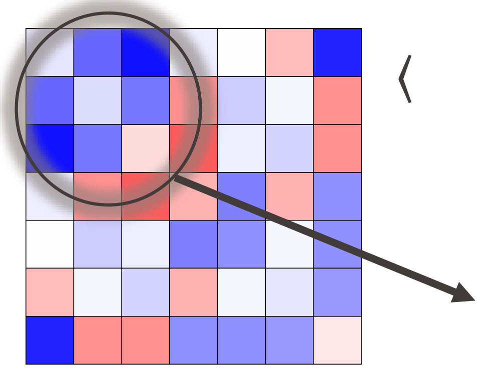
$$\rho(\boldsymbol{r}) \approx \sum_{inlm} d_{inlm}\varphi_{inlm}(\boldsymbol{r})$$

→ The learning targets are coefficients of atom-centered (ρ) or pairs of atom-centered ($\widehat{H}$) basis functions

per-atom

Target coefficients

per-pair



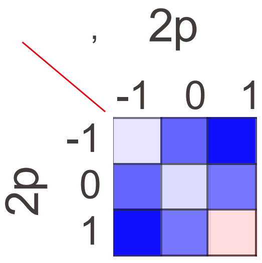
Hamiltonian,

$$\langle \qquad ' \; ' \quad '\rangle$$

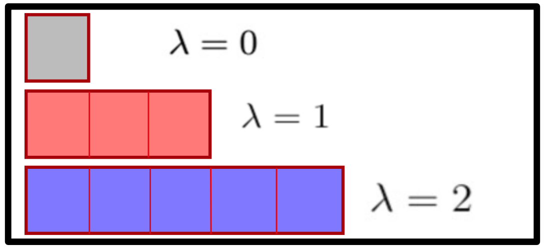
“uncoupled”

$$| \quad \rangle \otimes |' \quad '\rangle = \bigoplus_{\lambda\mu} | \quad \rangle \langle \quad ; ' \quad ' | \quad \rangle$$

“coupled”



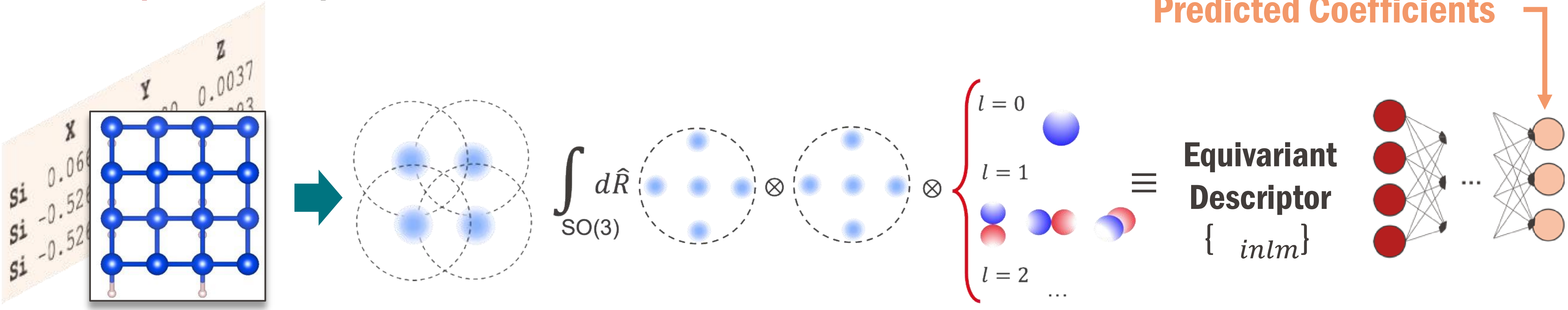
couple →



Irreducible spherical components

Predict electronic structure targets *on a basis*

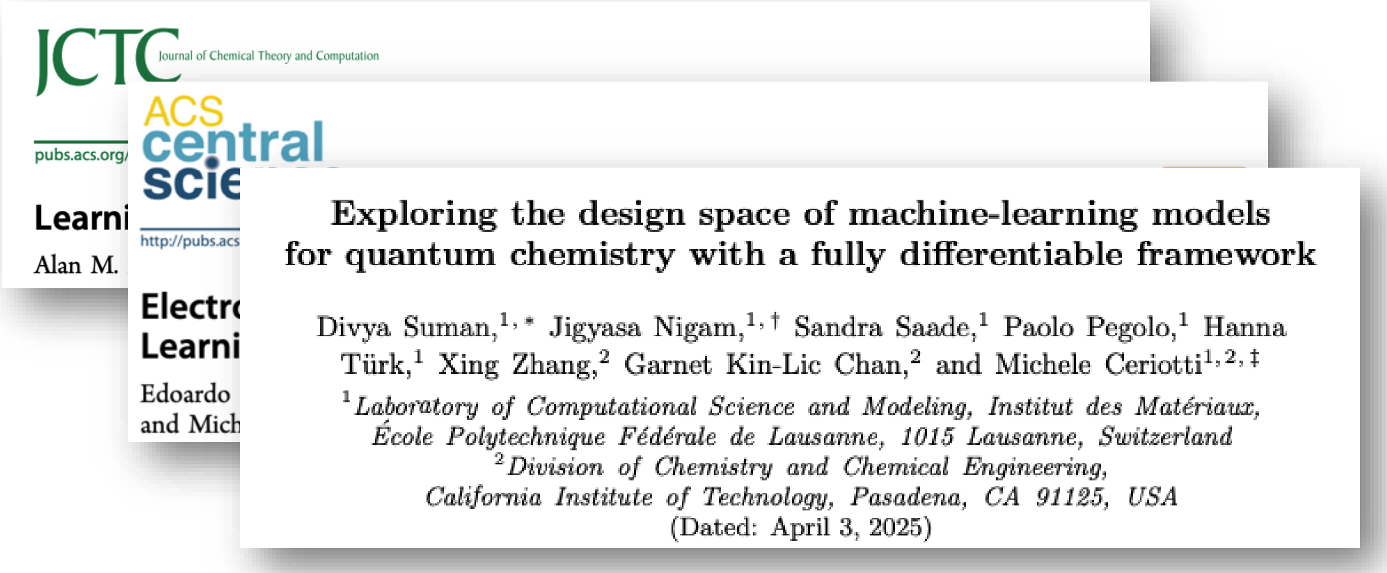
1. Descriptor-based equivariant NNs



Predicted Coefficients

Equivariant Descriptor $\{\ inlm\}$

Permutation + translation invariance, rotation equivariance, i.e. -SOAP



JCTC Journal of Chemical Theory and Computation

pubs.acs.org/

Learni

Alan M.

ACS central scie

http://pubs.acs

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and Mich

Exploring the design space of machine-learning models for quantum chemistry with a fully differentiable framework

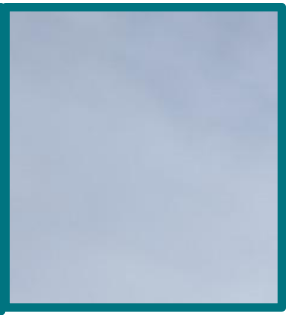
Divya Suman,[1,*] Jigyasa Nigam,[1,†] Sandra Saade,[1] Paolo Pegolo,[1] Hanna Türk,[1] Xing Zhang,[2] Garnet Kin-Lic Chan,[2] and Michele Ceriotti[1,2,‡]

[1] *Laboratory of Computational Science and Modeling, Institut des Matériaux, École Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland*

[2] *Division of Chemistry and Chemical Engineering, California Institute of Technology, Pasadena, CA 91125, USA*

(Dated: April 3, 2025)

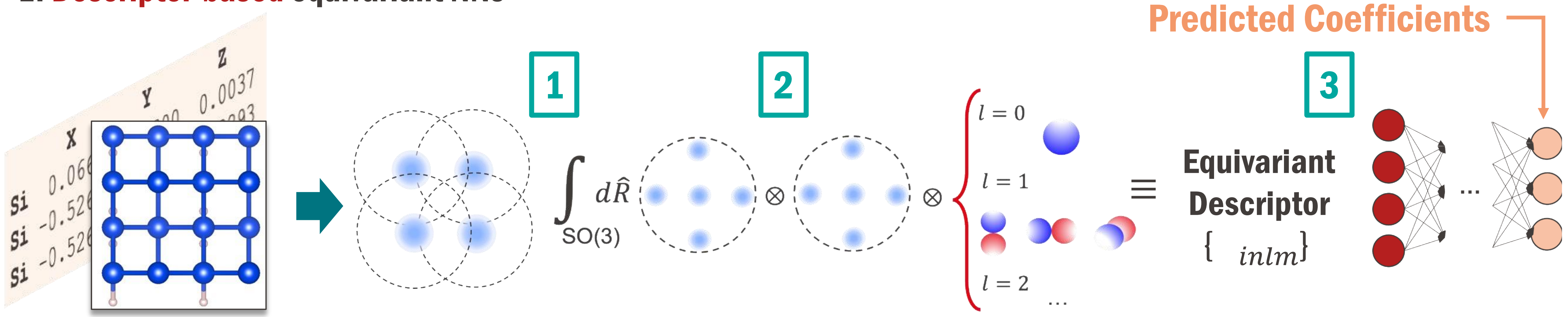
Divya



Jigyasa

Predict electronic structure targets *on a basis*

1. **Descriptor-based** equivariant NNs



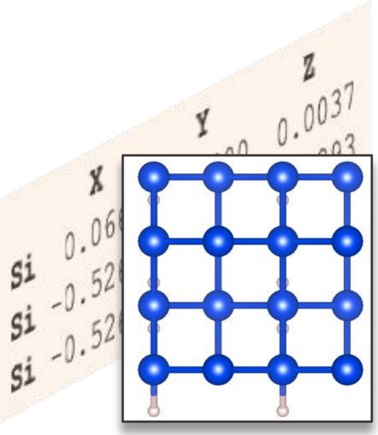
Predicted Coefficients

Permutation + translation invariance, rotation **equivariance**, i.e.   -SOAP

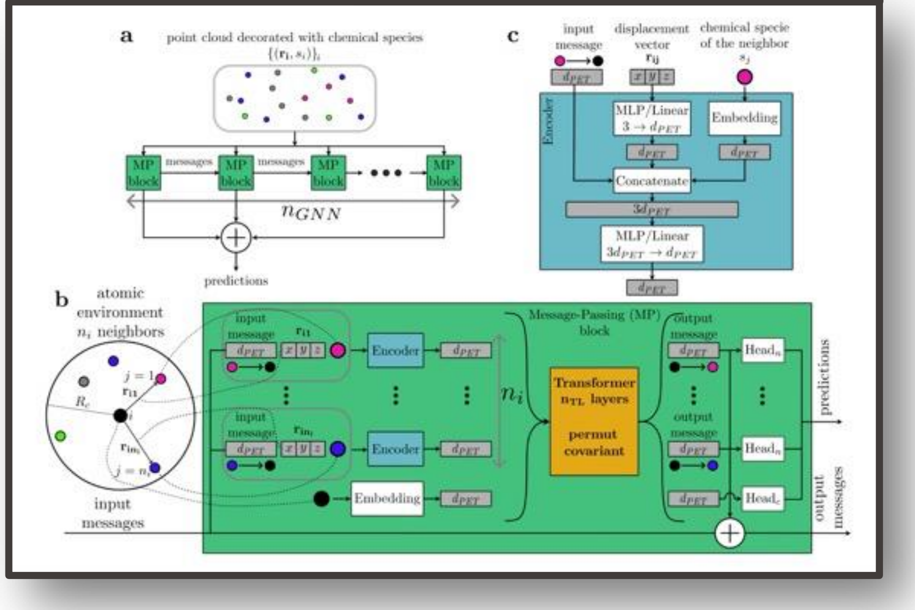
```
1. featomic.SphericalExpansion{ByPair}

2. featomic.clebsch_gordan.EquivariantPowerSpectrum{ByPair}

3. metatensor.torch.learn.nn.EquivariantLinear
```

Predict electronic structure targets *on a basis*

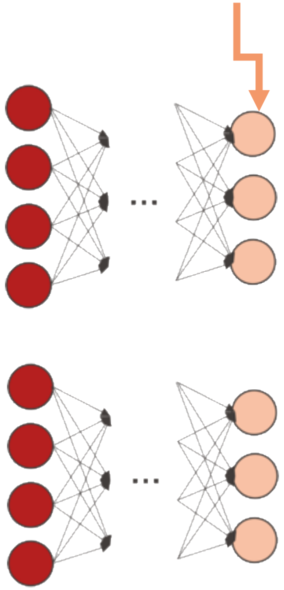
2. **Transformer-based** GNNs



Point-Edge Transformer (PET)



Predicted Coefficients (, ͡)



Paolo

à Rotation **equivariance** learned via data augmentation

à Different heads for multi-target prediction

Coming soon: PET for electronic structure targets on a basis!

Cool resources to check out…

Build your own equivariant models

metatensor-learn how-to guides

featomic how-to guides

→ <u>See also</u>: Guillaume's tutorial session on metatensor + metatomic

*Coming (*very*) soon* tutorials for building equivariant descriptors!

See the building blocks in action

The Atomistic Cookbook

→ Equivariant model for the polarizability

→ Periodic Hamiltonian learning

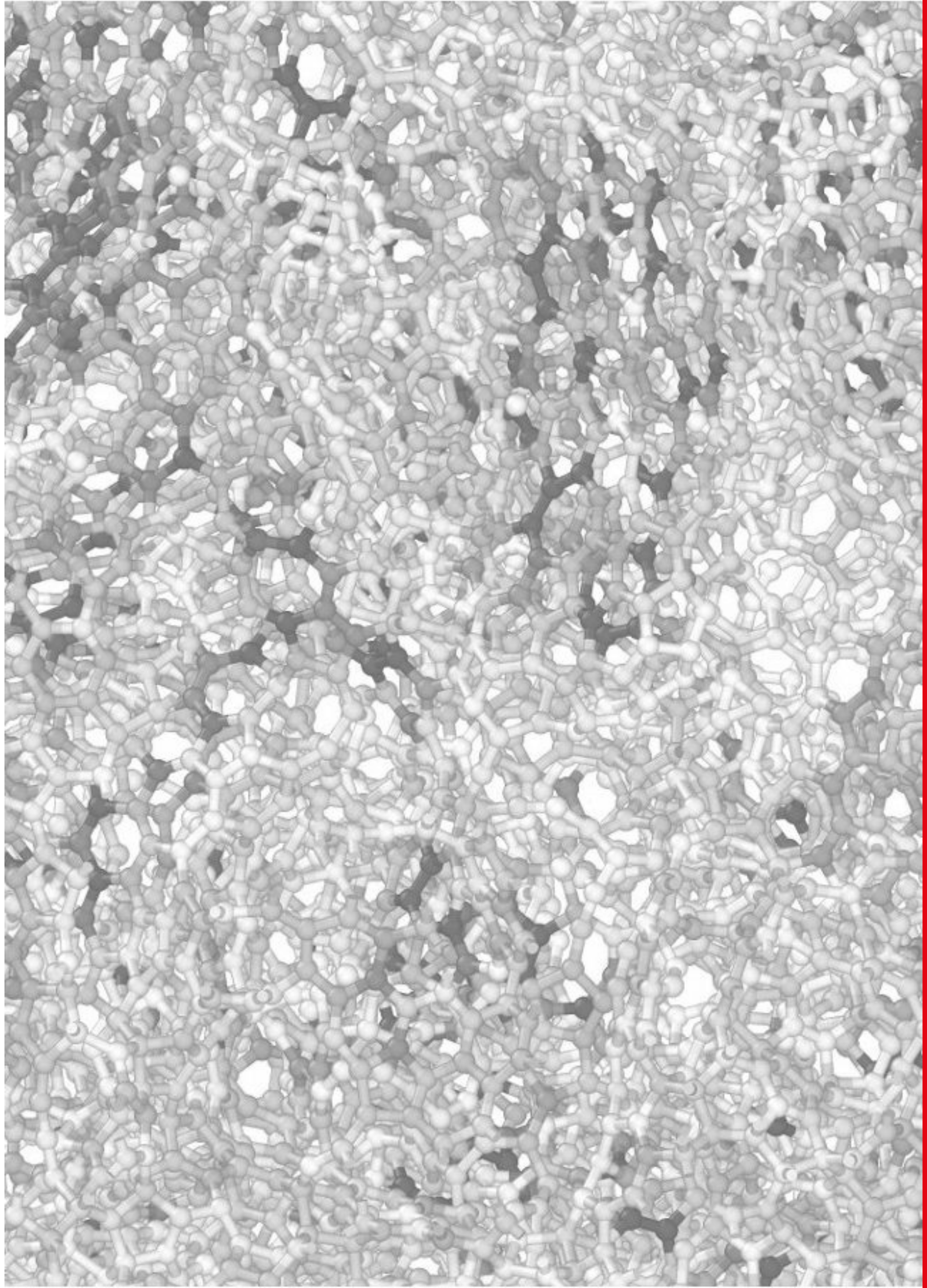
→ Hamiltonian learning with indirect targets

Divya

Hanna

Jigyasa

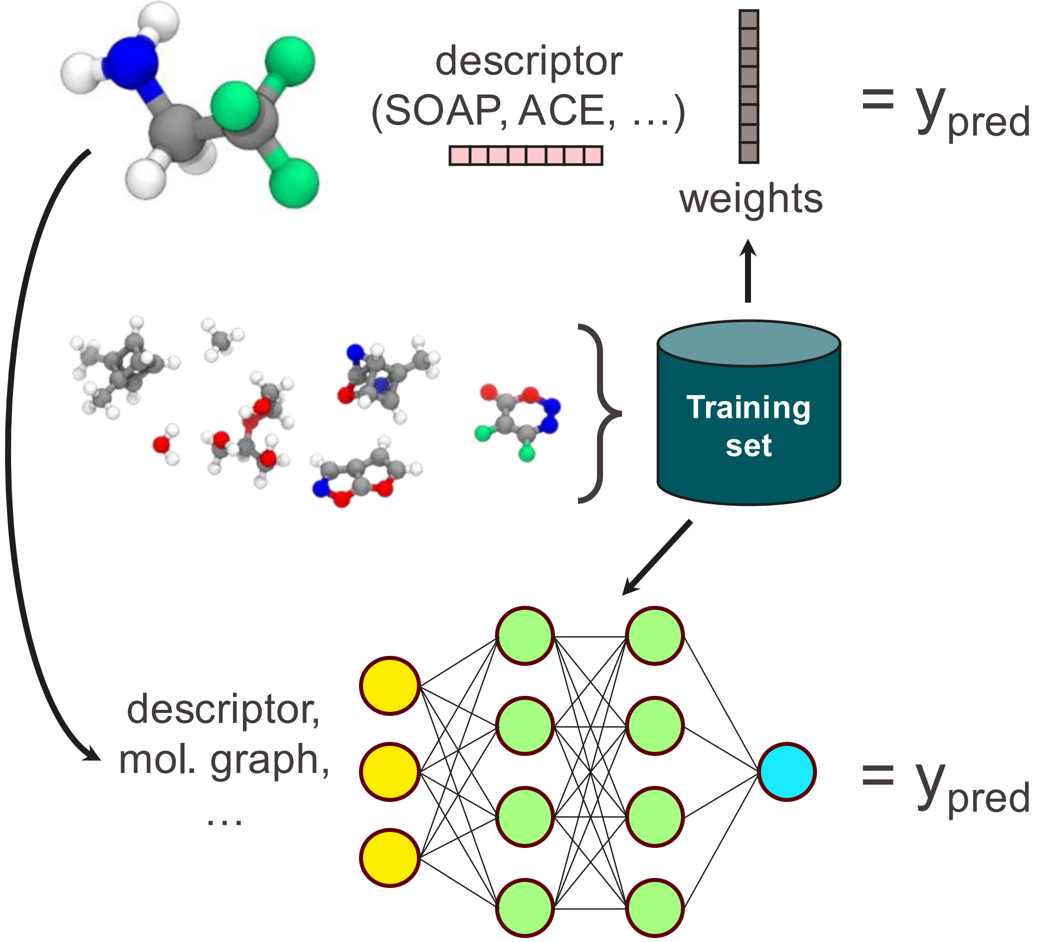
Paolo

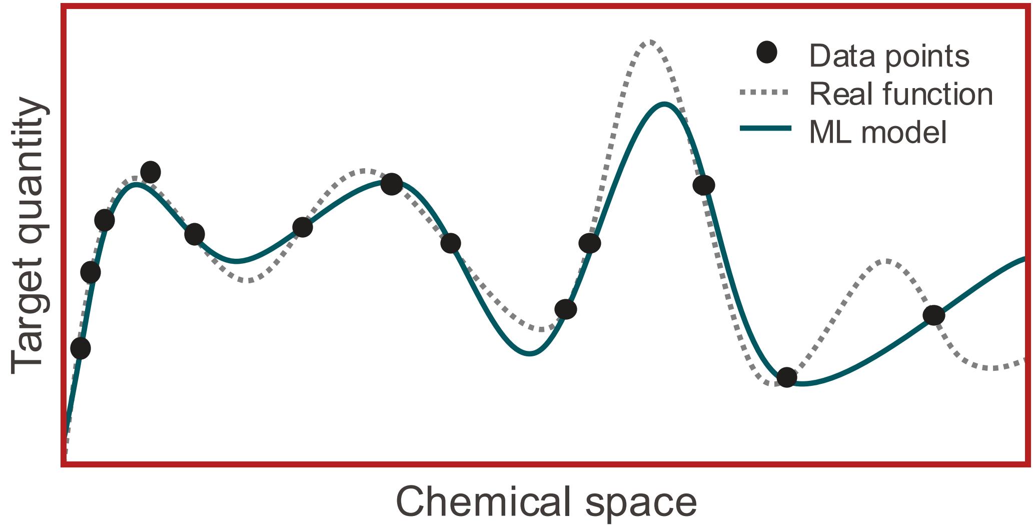


Uncertainty Quantification with Prediction Rigidities in `metatrain`

Sanggyu “Raymond” Chong

ML Models are Intrinsically Statistical





Can we quantify the **trustworthiness** or **robustness** of model predictions?

Prediction Rigidities (PR)

- Quantifies the **robustness** of model predictions at different “levels”

$$\mathcal{L}\left(\mathbf{w},\lambda,\epsilon_\star\right) \approx \mathcal{L}(\mathbf{w}_o) + \underbrace{\frac{1}{2}(\mathbf{w}-\mathbf{w}_o)^\top \mathbf{H}_o(\mathbf{w}-\mathbf{w}_o)}_{\text{second-order expansion around } \mathrm{w}_\mathrm{o}} + \underbrace{\lambda(\epsilon_\star - \tilde{y}(\mathbf{x}_\star,\mathbf{w}))}_{\text{Lagrange multiplier}}$$

$$\mathbf{H}_o = \frac{\partial^2 \mathcal{L}}{\partial \mathbf{w}\, \partial \mathbf{w}^\top}\bigg|_{\mathbf{w}_o} \approx \sum_i \frac{\partial \tilde{y}_i}{\partial \mathbf{w}} \frac{\partial^2 \ell_i}{\partial \tilde{y}_i^2} \frac{\partial \tilde{y}_i}{\partial \mathbf{w}^\top}$$

$$\mathcal{L}_c(\epsilon_\star) \approx \mathcal{L}(\mathbf{w}_o) + \frac{1}{2} \frac{(\boxed{\epsilon_\star} - \tilde{y}(\mathbf{x}_\star,\mathbf{w}_o))^2}{\frac{\partial \tilde{y}_\star}{\partial \mathbf{w}}\Big|_{\mathbf{w}_o}^\top \mathbf{H}_o^{-1} \frac{\partial \tilde{y}_\star}{\partial \mathbf{w}}\Big|_{\mathbf{w}_o}}$$

$\epsilon_\star$ can target:

- **global, local, component-wise** predictions
- cheap & effective UQ!

$$R_\star \equiv \frac{\partial^2 \mathcal{L}_c(\epsilon_\star)}{\partial \epsilon_\star^2}\bigg|_{\epsilon_\star = \tilde{y}(\mathbf{x}_\star,\mathbf{w}_o)} = \left(\frac{\partial \tilde{y}_\star}{\partial \mathbf{w}}\bigg|_{\mathbf{w}_o}^\top \mathbf{H}_o^{-1} \frac{\partial \tilde{y}_\star}{\partial \mathbf{w}}\bigg|_{\mathbf{w}_o} \right)^{-1} = \frac{1}{\mathbf{f}_\star^\top (\mathbf{F}^\top \mathbf{F})'^{-1} \mathbf{f}_\star}$$

NN model: **last-layer features**

LLPR UQ in metatrain

- Simple wrapper provided:

```
from metatrain.utils import LLPRUncertaintyModel
llpr_model = LLPRUncertaintyModel(model)
```

- Compute covariance → compute inv_covariance → calibrate

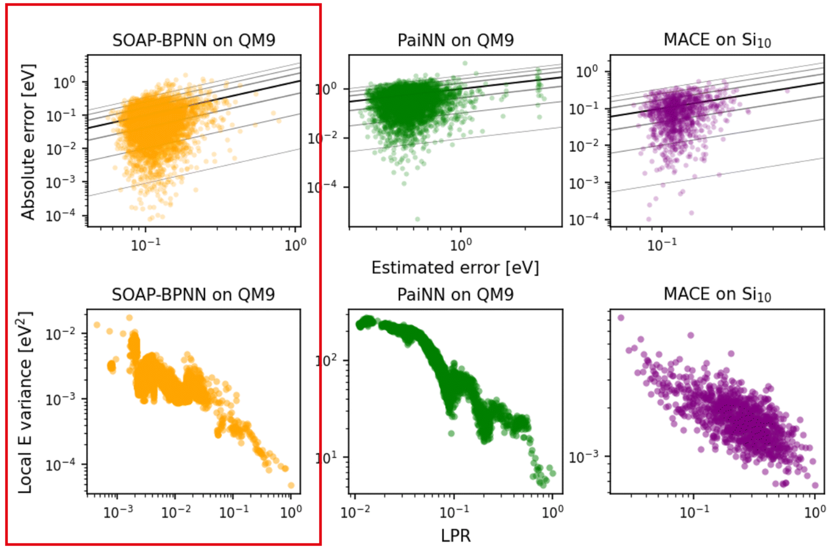
```
llpr_model.compute_covariance(train_loader)
llpr_model.compute_inv_covariance()
llpr_model.calibrate(calib_loader)
llpr_model.generate_ensemble(orig_weights, n_members)
```

- Users interact with:

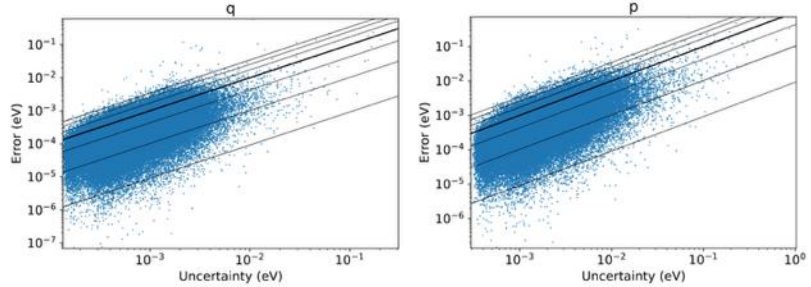
```
outputs['mtt::aux::{target}_uncertainty’]
outputs['mtt::aux::{target}_ensemble’]
```

*You will interact with some of this in **tutorial B**!*

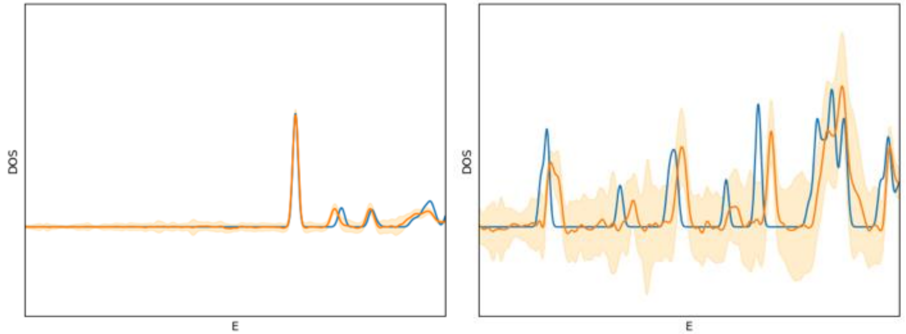
Hall of LLPR UQ



LPR



FlashMD

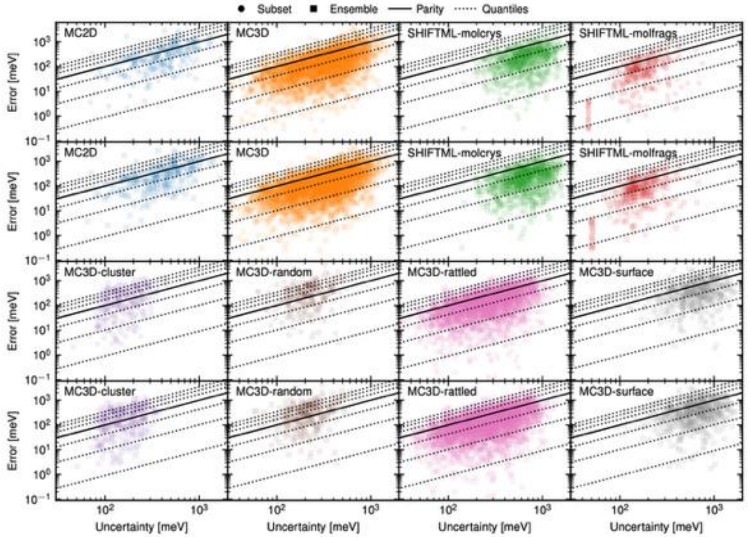


ML-DOS

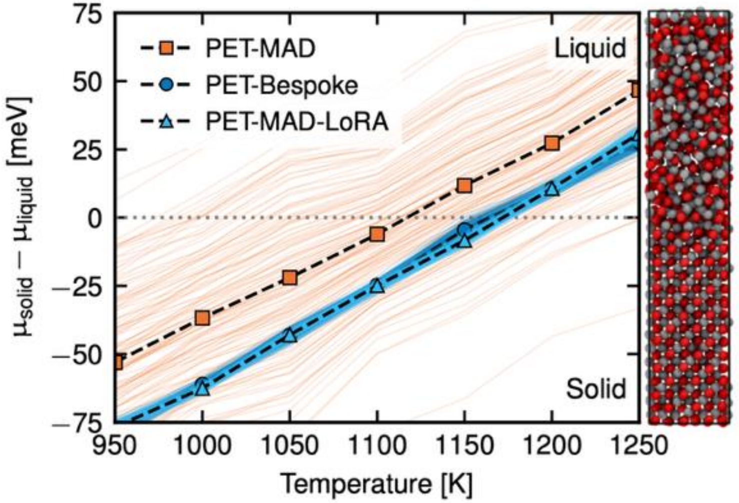
MLIPs

PET-MAD

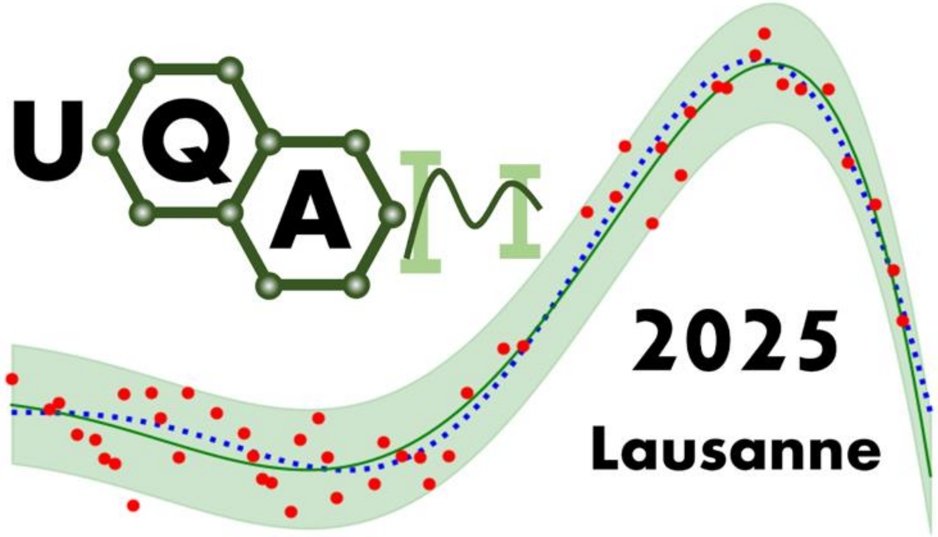
A UNIVERSAL INTERATOMIC POTENTIAL



LLPR ensemble



Workshop on Uncertainty Quantification in Atomistic Modeling



“From uncertainty-aware DFT to ML”

November 25-28, 2025

CECAM Headquarters, Lausanne, CH

Picture courtesy of Psi-k

cecam
Centre Européen de Calcul Atomique et Moléculaire

DAEMON

ROYAL SOCIETY OF CHEMISTRY