# Surrogate models for the electron density and related scalar fields


Joseph W. Abbott[1], Sanggyu Chong[1], Alan Lewis[2], Michele Ceriotti[1]

[1]Lab-COSMO, EPFL, Switzerland, [2]Univeristy of York, UK


## Background

- ML surrogate models for DFT can bypass the Kohn-Sham equations with more favourable scaling
- Scalar fields derived from Kohn-Sham orbitals can be useful learning targets → accelerating DFT, probing electronic structure
- Equivariant learning of schemes built atop a modular software stack allow different scalar fields to be flexibly targeted at scale
- This can be applied to quantities such as the LDOS for STM imaging

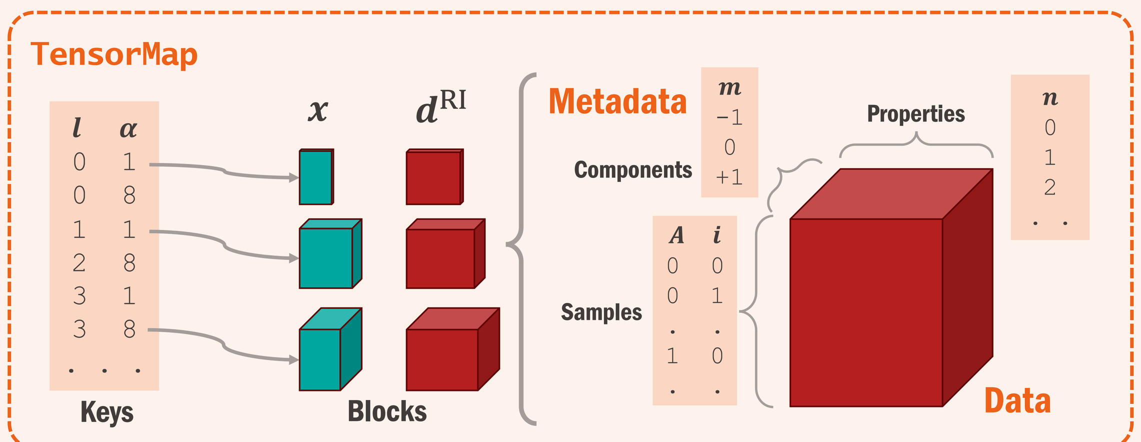
## Modular software ecosystem

### /luthaf/rascaline

- Evaluating structural representations
- *Work-in-progress*: Python-API for Clebsch-Gordan iterations. Later: learnable representations?

### /lab-cosmo/metatensor

- Sparse storage format for atomistic data
- *Lingua franca* for building end-to-end ML workflows
- Operations for manipulating data + metadata

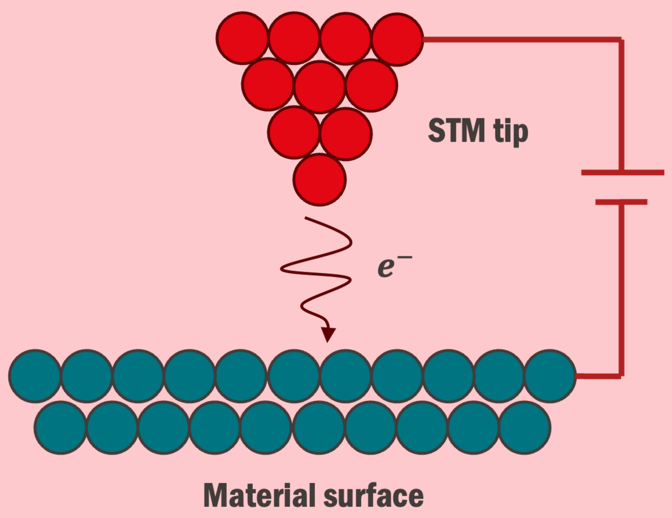



metatensor: data interchange format

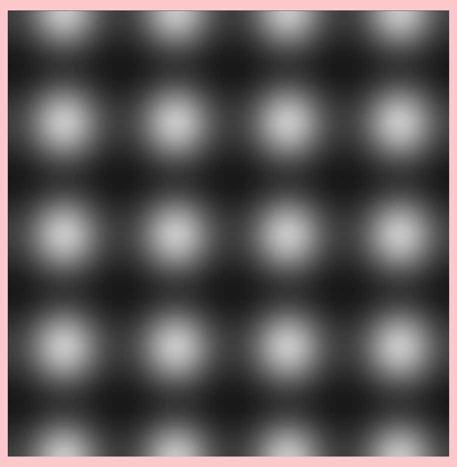
### /jwa7/rho_learn

- Custom metatensor/PyTorch modules for equivariant learning of scalar fields and tensors
- Integration with FHI-aims: calculators + parsers
- Gradient-based model training
  - Reduced memory-requirements, scalable
  - Models of arbitrary complexity (i.e. NNs)

## Application: ML-driven STM imaging




STM experiment setup. $e^-$ tunnel between tip and material surface



DFT reference 2D slice of the ILDOS of a Si (100) slab

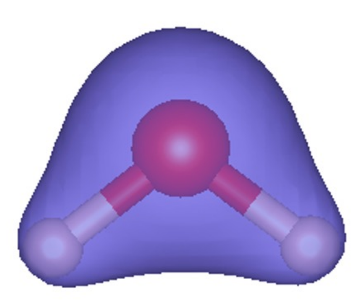
- Scanning tunnelling microscopy (STM) → experimental technique to probe the electronic structure of materials
- Surface STM images → 2D contour plots of the local density of states (LDOS) resolved at Fermi energy $\epsilon_F$
- Target scalar field: integrated LDOS with KS-orbital weighting:

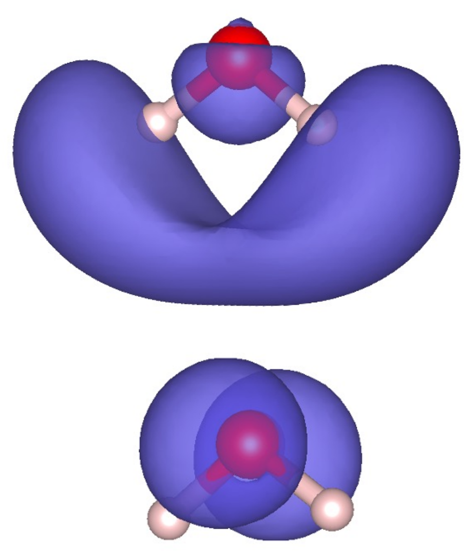
$$W(a, \epsilon, V) = \sum_{\epsilon'=\epsilon}^{\epsilon+V} g(\epsilon - \epsilon_a)$$

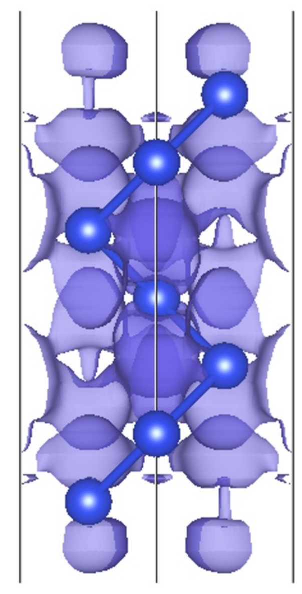
- Considerations: energy alignment and long-range effects

## Scalar fields $\rho(\boldsymbol{r})$ of interest

| $\rho(\boldsymbol{r}) \equiv$ electron density | $\rho(\boldsymbol{r}, a') \equiv$ KS-orbital density | $\rho(\boldsymbol{r}, \epsilon) \equiv$ LDOS |
|---|---|---|
| $W(a) = n_a^{\mathrm{occ}}$ | $W(a) = \delta_{aa'}$ | $W(a, \epsilon) = g(\epsilon - \epsilon_a)$ |






### FHI-aims extension for constructing scalar fields

$$\rho(\boldsymbol{r}) = \sum_{i,j \in \mathrm{AO}} \left[ \sum_{a \in \mathrm{KSO}} W(a)\, C_{ij}(a) \right] \phi_i(\boldsymbol{r})\, \phi_j(\boldsymbol{r})$$

- General expression for constructing a scalar field from KS-orbitals
- KS-orbital weights $W(a)$ dictate the specific scalar field

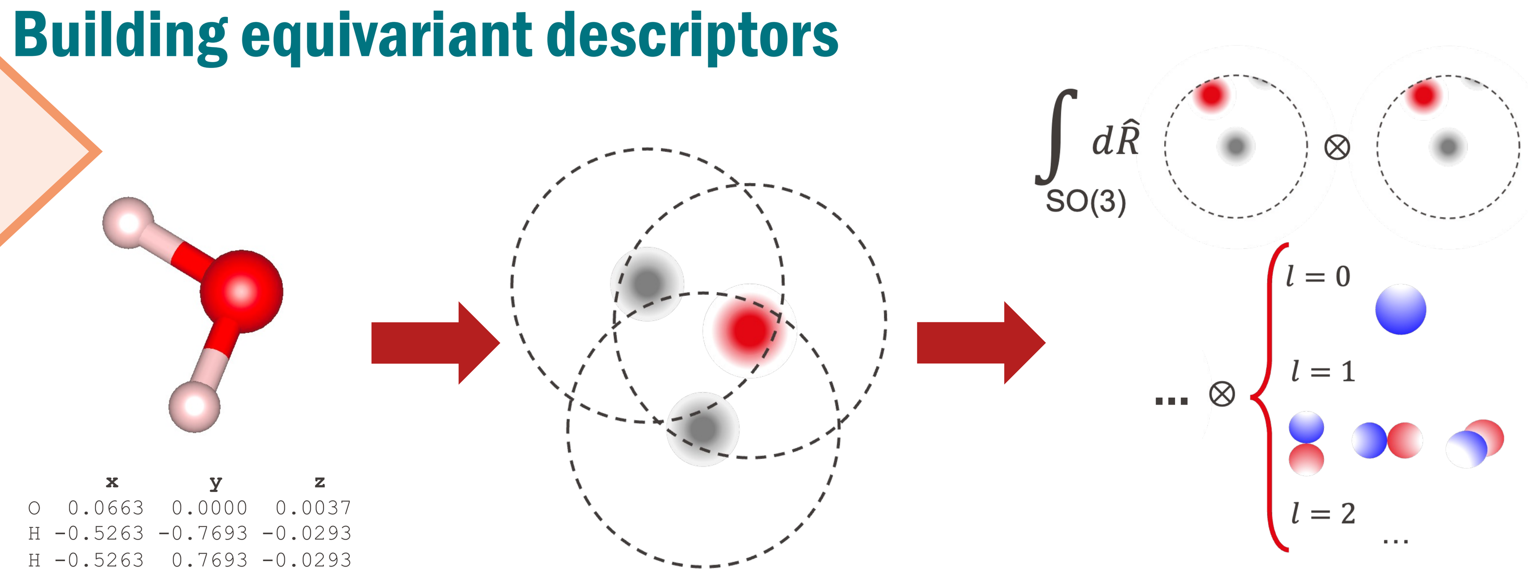
### Generating learning targets – RI fitting

$$\rho(\boldsymbol{r}) \approx \rho^{\mathrm{RI}}(\boldsymbol{r}) = \sum_b d_b^{\mathrm{RI}}\, \varphi_b(\boldsymbol{r})$$

- Real-space scalar field decomposed onto a fitted basis
- $\{d_b^{\mathrm{RI}}\}$ are the equivariant ML targets

## End-to-end workflow for $\rho$-learning

### Building equivariant descriptors




1. Nuclear coordinates & atom types
2. Local atom-centered environments
3. Rotational symmetrisation and/or Clebsch-Gordan iterations

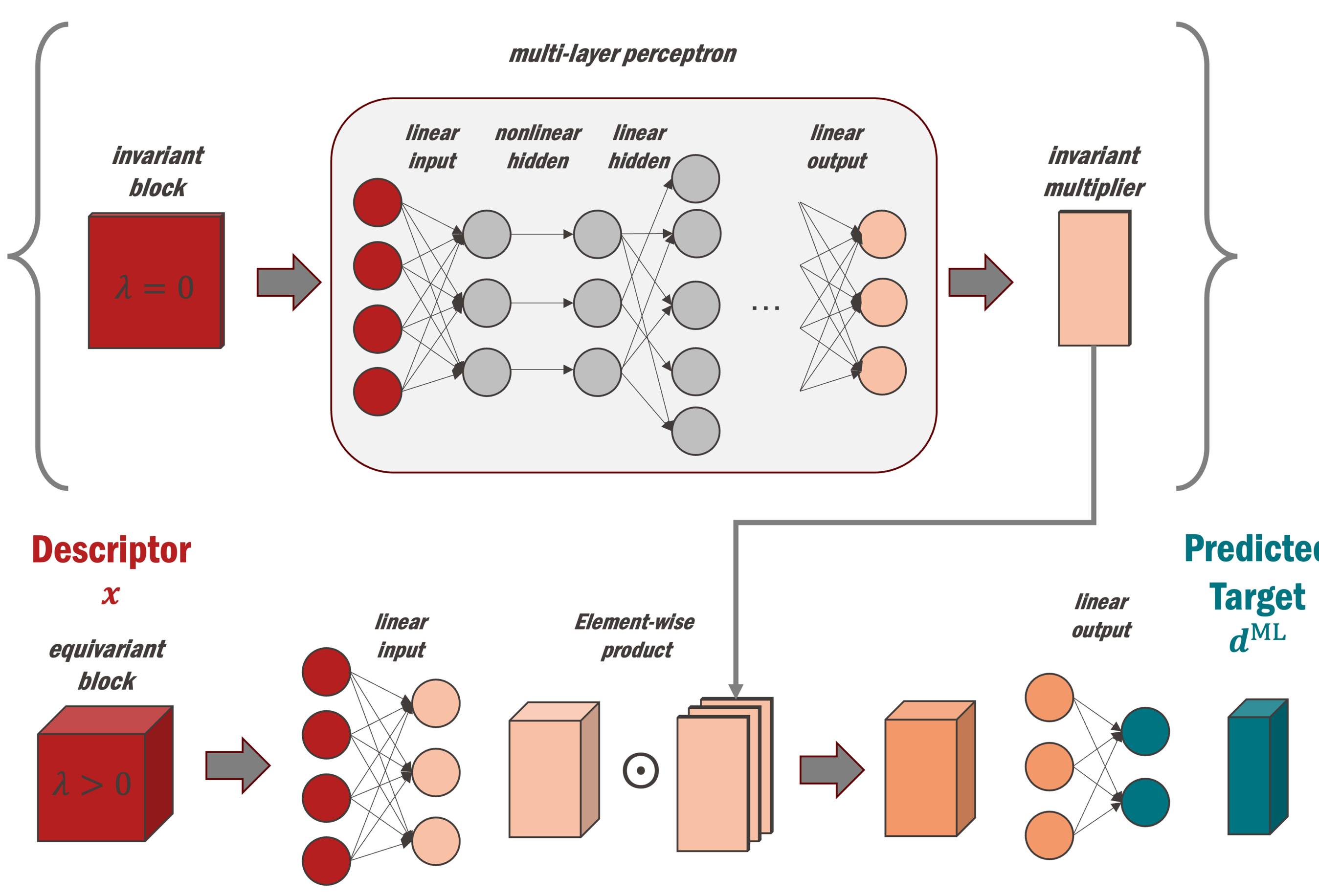
### Generating learning targets

FHI-aims — The ab initio materials simulation package

Converge SCF, define $W(a)$, run RI-fitting procedure

**rho_learn demo:**
→ end-to-end learning of the HOMO in gas phase water, integrated with FHI-aims

### Training a model




Incorporating nonlinearities into the model. Invariant blocks are passed through a multi-layer perceptron and used as a nonlinear multiplier for equivariant blocks.

- Descriptor and target decomposed in the angular basis → equivariant learning with model for each species and $l$-channel
- Non-orthogonal RI-basis → all $\{d_b^{\mathrm{RI}}\}$ are coupled → overlap matrix, $\hat{S}$, required for loss evaluation (memory intensive!)

$$\mathcal{L} = \Delta\tilde{d} \cdot \hat{S} \cdot \Delta\tilde{d}$$

- Use of nonlinearities can improve model performance

/jwa7/rho_learn

## Next Steps

1. LDOS-learning for STM image generation of Si surfaces and beyond
2. Make rho_learn fully torch-scriptable for shippable models
3. Further integrate rho_learn with FHI-aims for derived quantities + DFT acceleration
4. Unify ML-infrastructure for electronic structure surrogate models → different targets, different QC codes

## References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