Machine Learning Applied to Materials Science

Hypergraphs, Equivariant Networks, and Tensors

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June 5, 2025

Overview

- Crystal graphs, MPNNs, and hypergraphs

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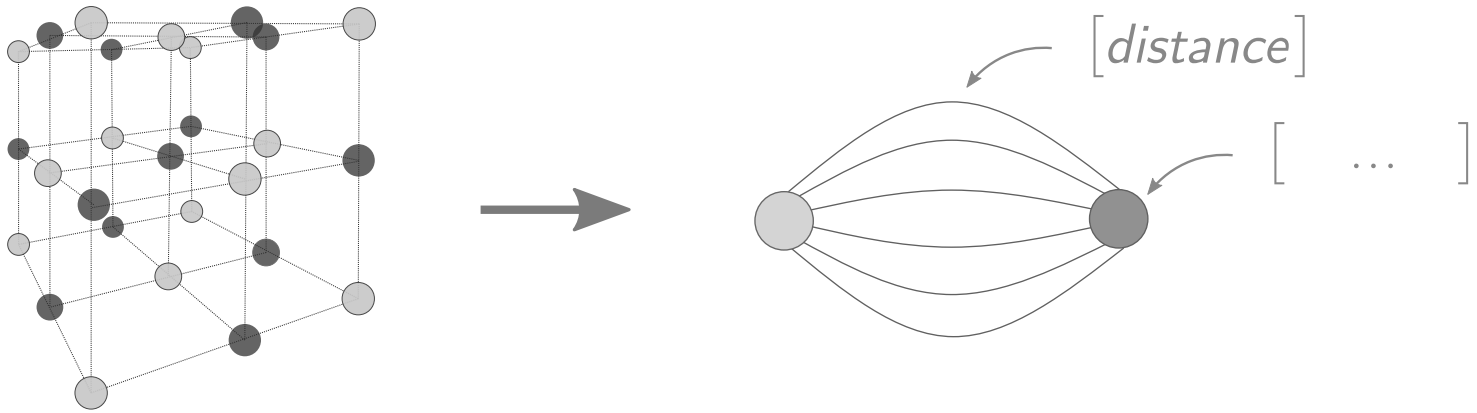
- Equivariant Networks and Tensors

Crystal Graph Construction

Usually represent crystalline systems as graphs (nodes and edges)[1]:



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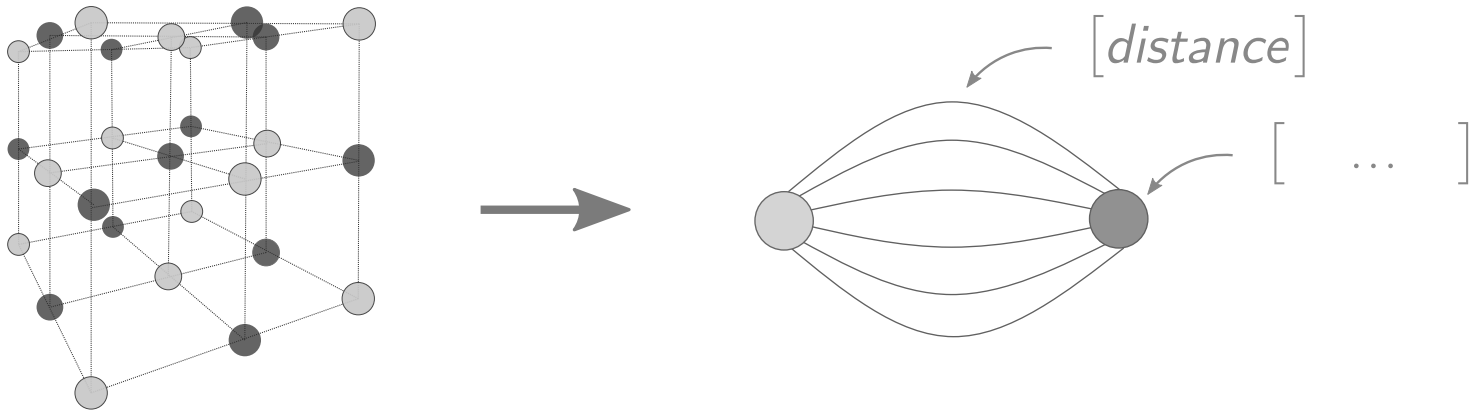

- Atoms are nodes
- Edges denote 'neighbors'; based on distance
- Feature vectors

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Message Passing on Graphs

MPNN framework[2]:

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MPNN framework[2]:

- Message m for each node i:

$$m_i^{t+1} = \sum_{n_j \in \mathcal{N}(i)} M_t(n_i^t, e_{ij}, n_j^t)$$

- Update function U for layer t:

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We'll need to generalize this later...

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Graph Limitations

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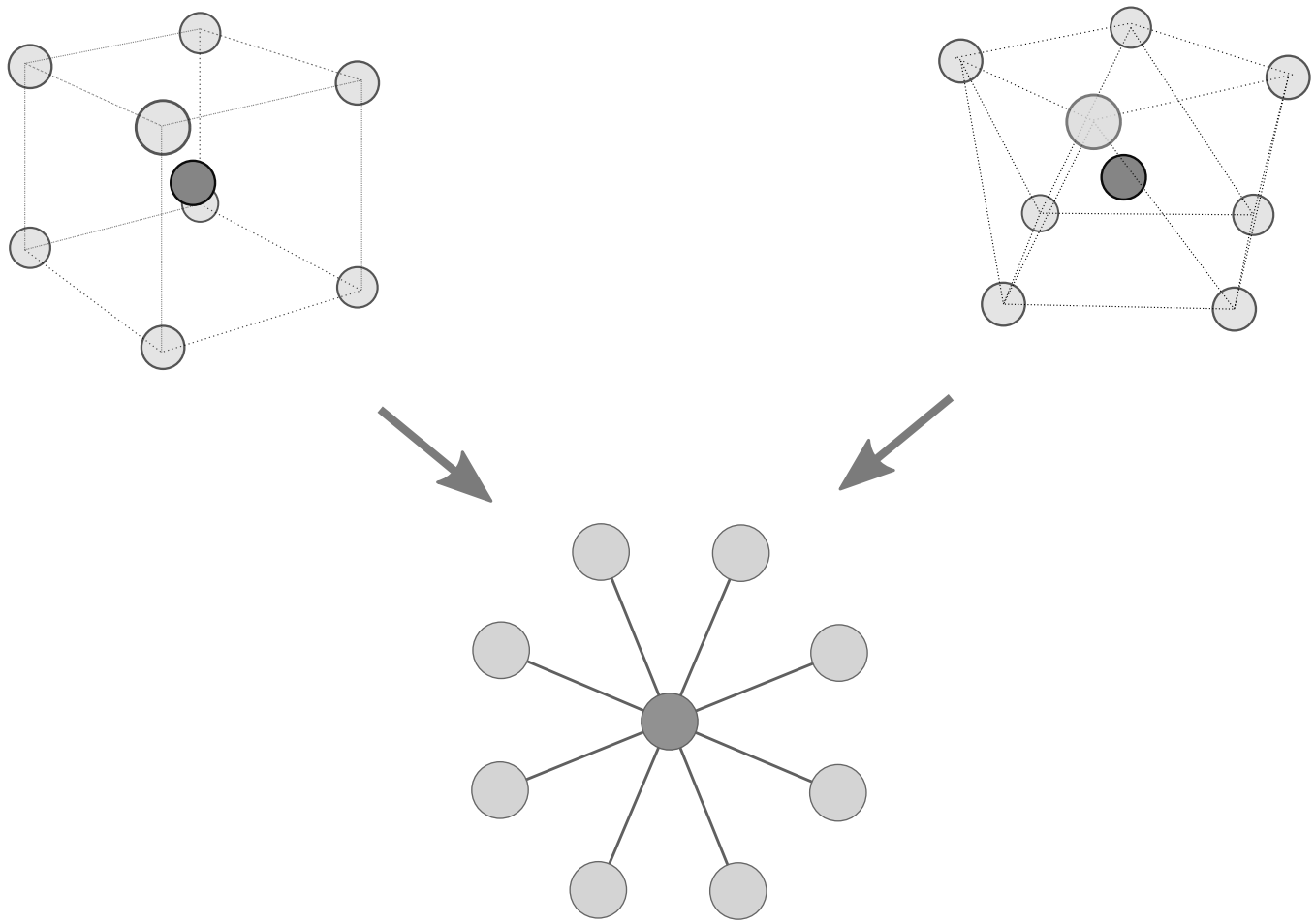

These two crystalline structures have same representations!

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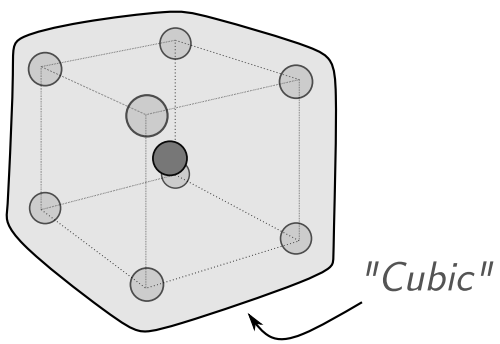
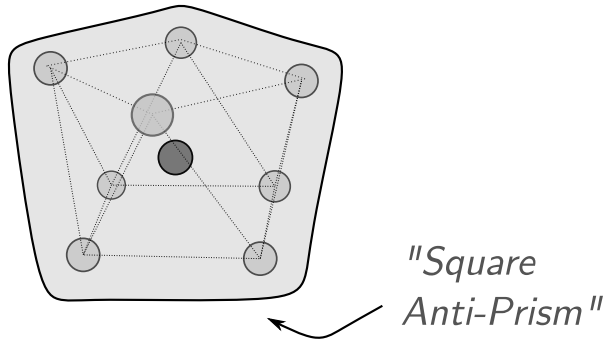
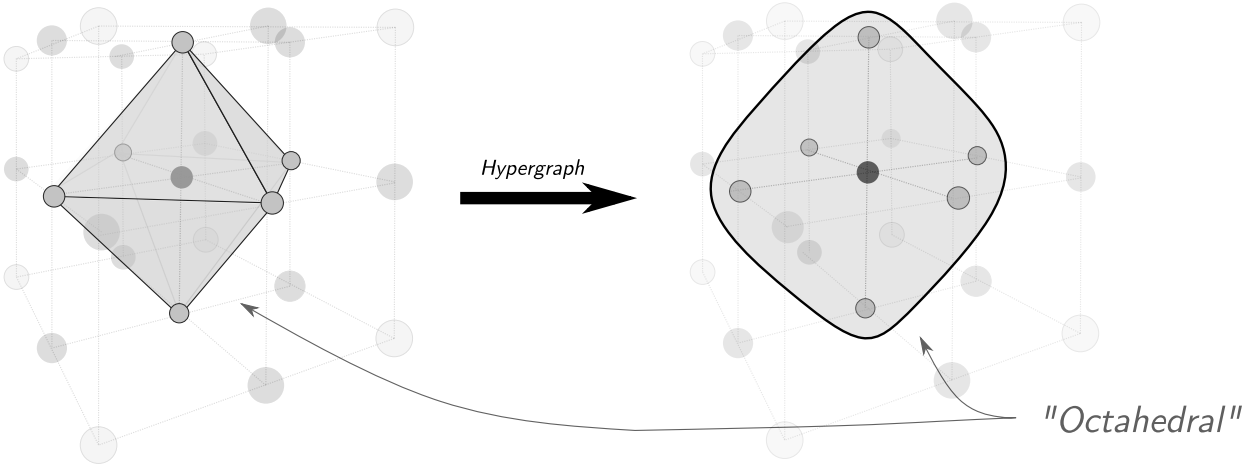




So we may define hyperedges for this local geometry.

Local Environments as Hyperedges

Construct a hyperedge for each atom's entire local environment:



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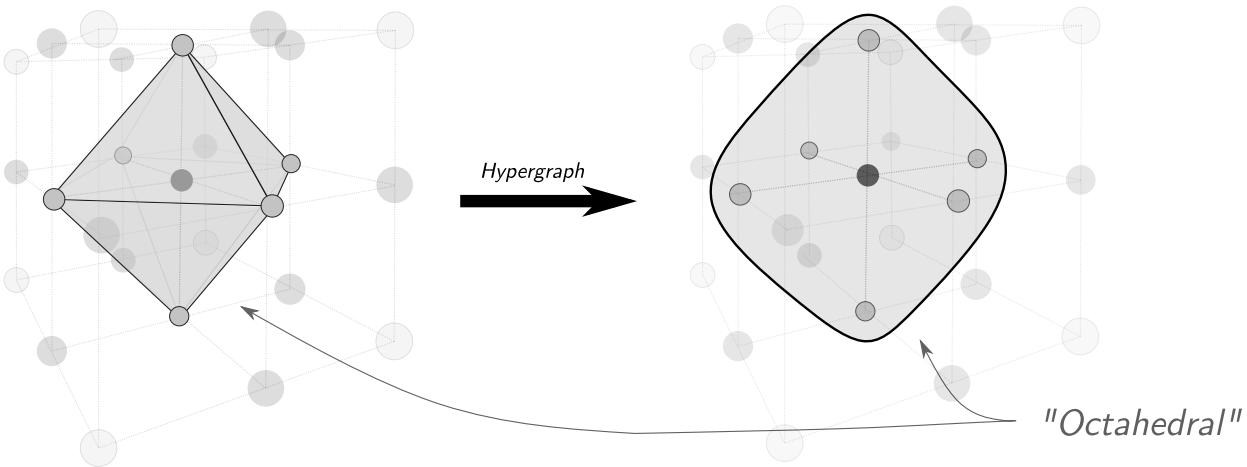

These contain the entire first shell of neighbors.

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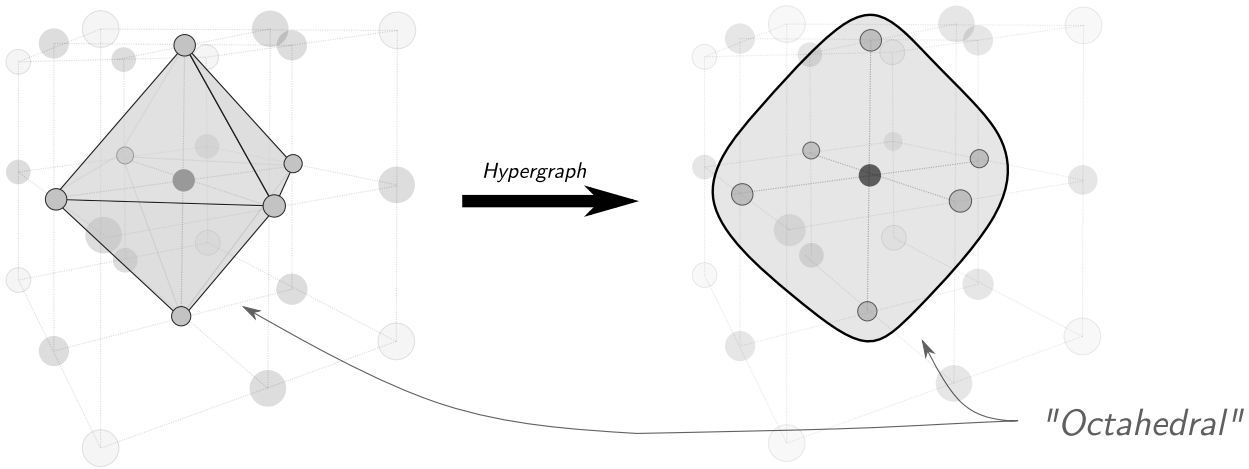

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- Geometry is encoded quantitatively with continuous symmetry measures or structure order parameters[3] as features

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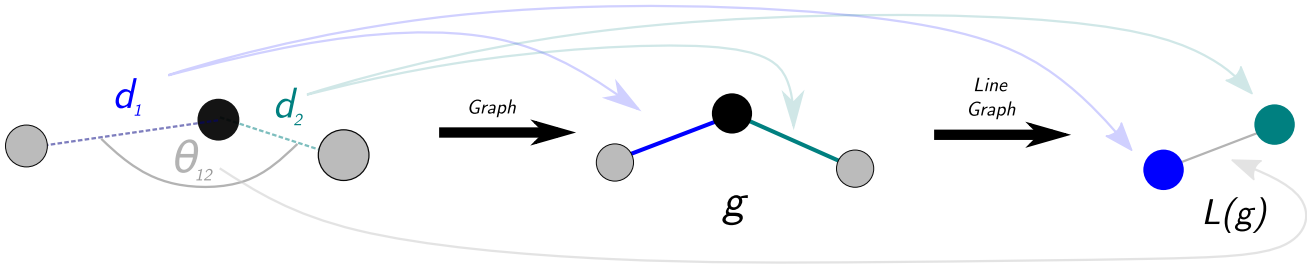
Another Example: Triplets as Hyperedges

Many modern models utilize bond angle information[4].



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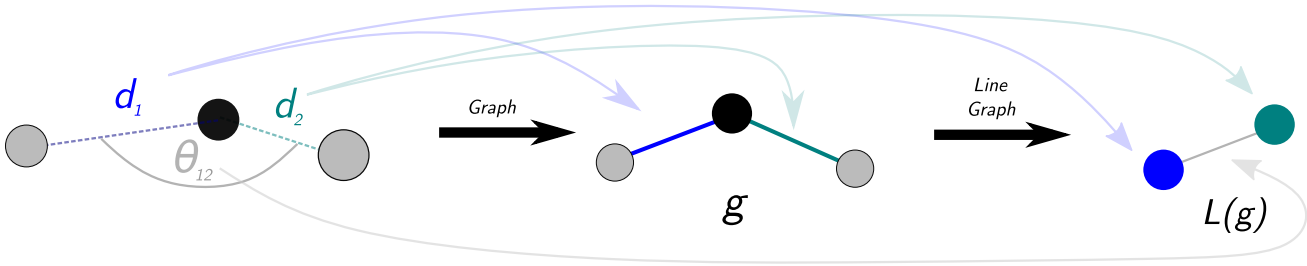

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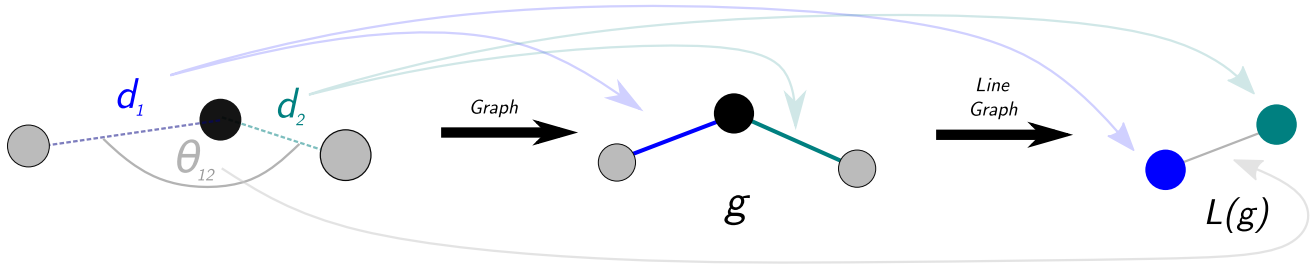

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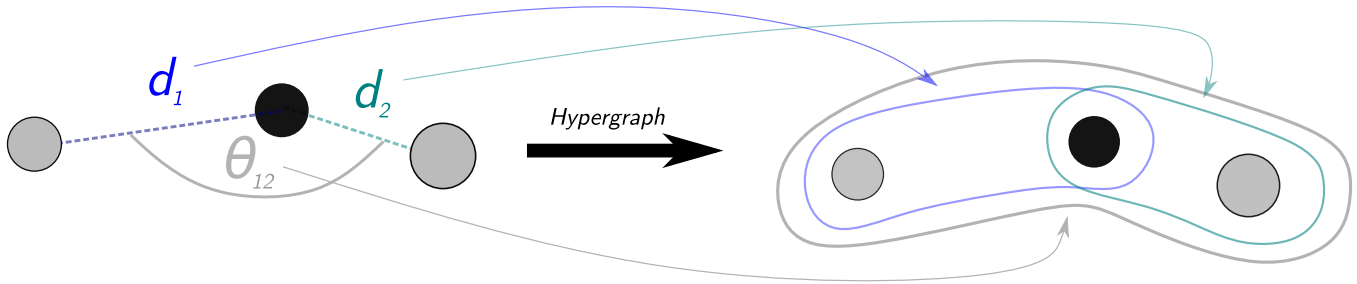

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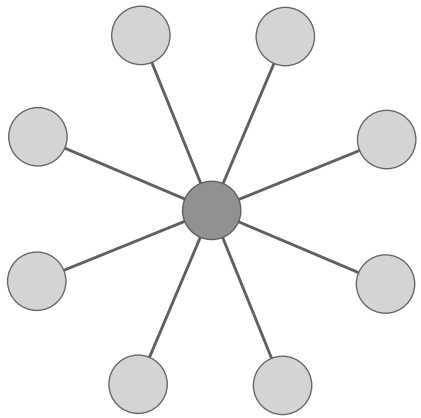

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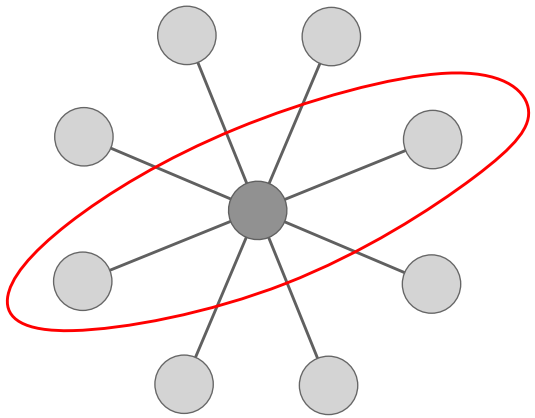

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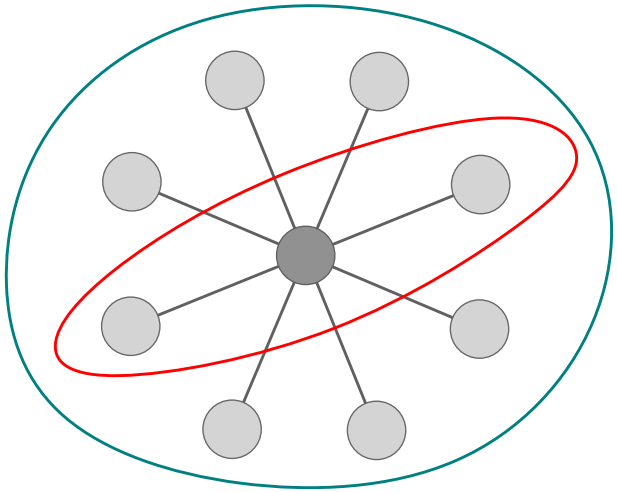

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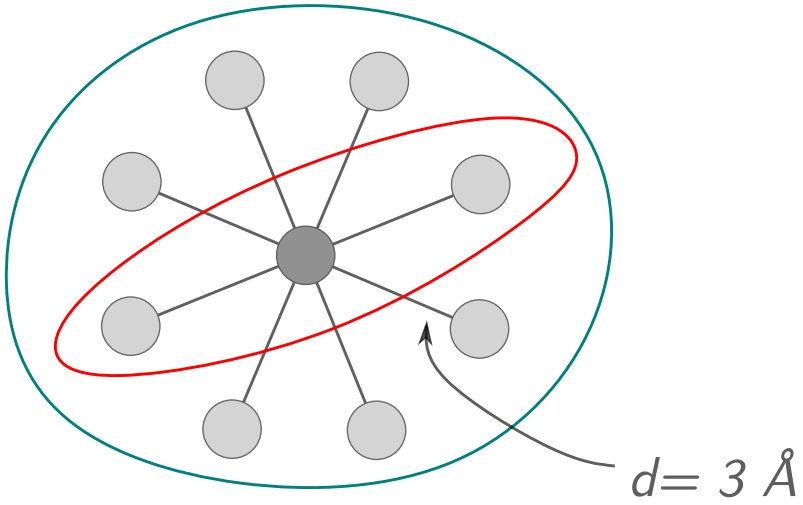

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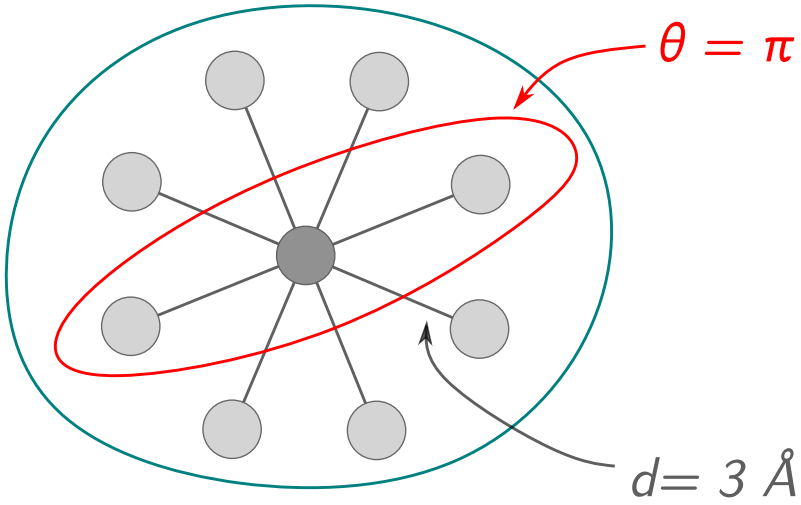

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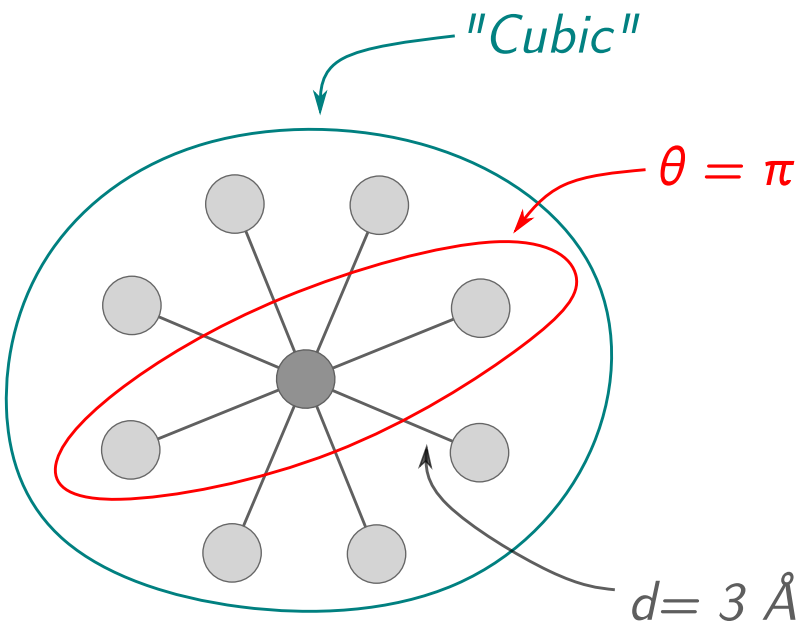

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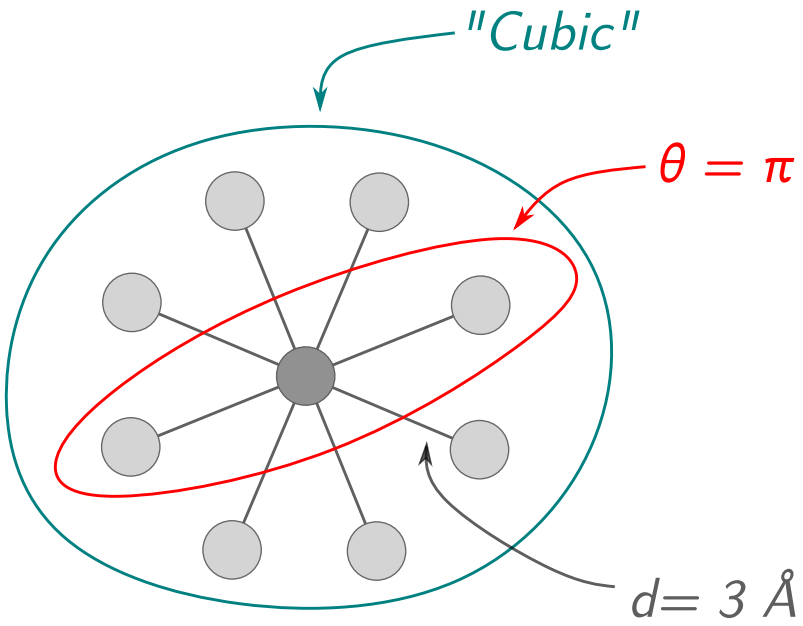

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But..... how do we update these representations?

Extending Message Passing to Hypergraphs

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$$m_i^{t+1} = \sum_{h_j \ni n_i} M_t(n_i^t, \underbrace{h_j^t, \{n_w^t | n_w \in h_j\}}_{e_{ij}, n_j})$$

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- Acts as testbed for what features/order correlations are most important for task.

Hyperparameters and Model Architecture

For each convolutional structure, testing was done for a model with 3 convolutional layers, an initial learning rate of 0.01, hidden node features of dimension 64, and a hidden output layer of dimension 128 (Similar to CGCNN's architecture). The loss function utilized is MSE.

Datasets are split 80% for training and 20% for validation tests.

Materials Projet Datasets

Below are results for various regression and classifications tasks for 152,605 materials.

Formation Energy

Model	Best MAE (eV/Atom)
Bond-only	0.177
Motif-only	0.088
Bond & Motif	**0.074**

Band Gap

Model	Best MAE (eV)
Bond-only	0.315
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Metal/Non-metal

Model	Best Accuracy
Bond-only	.837
Motif-only	.853
Bond & Motif	**.860**

MatBench Datasets

Below, we present results on validation sets for various MatBench target sets.

Model	Phonons (1,265) Best MAE (cm^{-1})	Refractive Indices (4,764) Best MAE
Bond-only	78.9	**.4891**
Motif-only	65.9	.5328
Bond & Motif	**59.0**	.5222

Model	Perovskites (18,829) Best MAE (eV/Atom)
Bond-only	0.329
Motif-only	**0.052**
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Overview of Experimental Results

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• In the case of Perovskites (a harder set of formation energy targets), this geometrical information seems even more impactful. Though, it also may just help more for the smaller dataset available for training.

• Phonons (also heavily dependent on geometrical information) seem to also benefit greatly from motif-level features

Conclusion

- Graphs are limited in their expression of higher-order (above pair-wise) features of crystal structures

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- Crystal hypergraphs give us a natural way to encode different types of features in one representation

Equivariant Functions

An equivariant function $f : X \to Y$ is one that satisfies the following equality:

$$f\left(D_X(g) \cdot x\right) = D_Y(g) \cdot f(x)$$

Equivariant Convolution

Enforcing equivariance often restricts our choice of function; while also guaranteeing a natural condition for certain tasks.

Hence, equivariance conditions may often restrict our trainable parameter space (requiring less data in training) while providing similar expressibility of the model (giving similar, or sometimes better performance).

Equivariant Convolution cont.

Example: Convolutional Networks and Translation
Traditional convolutional neural networks (CNNs) are actually equivariant under the group action of translation (in 2D for the usual input of images).

Special Orthogonal Group in 3D

The special orthogonal group of 3 dimensions, $SO(3)$, is the group describing 3 dimensional rotations.
It has irreducible representations indexed by a rotational order ℓ and a harmonic order m, termed the spherical harmonics Y_m^ℓ.
Note that products of spherical harmonics can be decomposed in terms of another linear superposition of spherical harmonics via Clebsch-Gordon Coefficients $C^{\ell_f m_f}_{\ell_1 m_1 \ell_2 m_2}$

$$Y_{\ell_1}^{m_1}(\Omega) Y_{\ell_2}^{m_2}(\Omega) = \sum_{\ell_3, m_3} \sqrt{\frac{(2\ell_1+1)(2\ell_2+1)}{4\pi(2\ell_3+1)}} C^{\ell_3 m_3}_{\ell_1 m_1 \ell_2 m_2} C^{\ell_3 0}_{\ell_1 0 \ell_2 0} Y_{\ell_3 m_3}(\Omega)$$

Tensor Field Networks/SO(3)

NEquiP utilizes a convolutional structure that is equivariant under the $SO(3)$ group, following that introduced in the Tensor Field Networks paper

Layer-to-layer convolution defined as:

$$\mathcal{L}^{(l_o)}_{acm_o} = \sum_{m_f, m_i} C^{l_o m_o}_{l_f m_f, l_i m_i} \sum_{b \in S} F^{l_f l_i}_{cm_f}(\vec{r}_{ab}) V^{l_i}_{bcm_i}$$

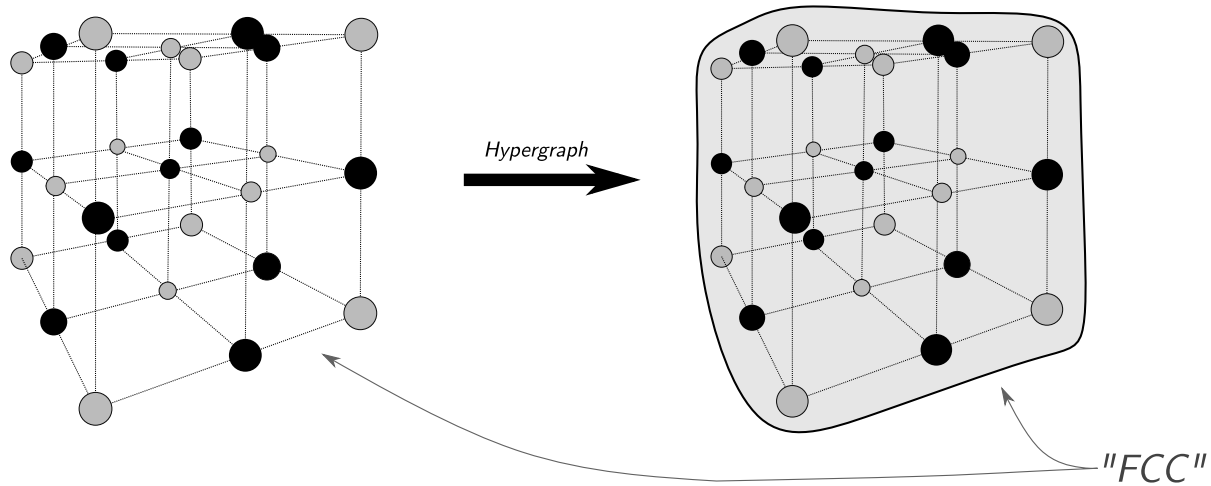
Hyperedge Index

Computationally, we define a *hyperedge index* of dimension 2 (as in previous hypergraph works):

[[node-index, ...],
[hyperedge-index, ...]]

Cell Hyperedges

Another order of hyperedge we may consider is that describing the entire unit cell of some crystalline structure.

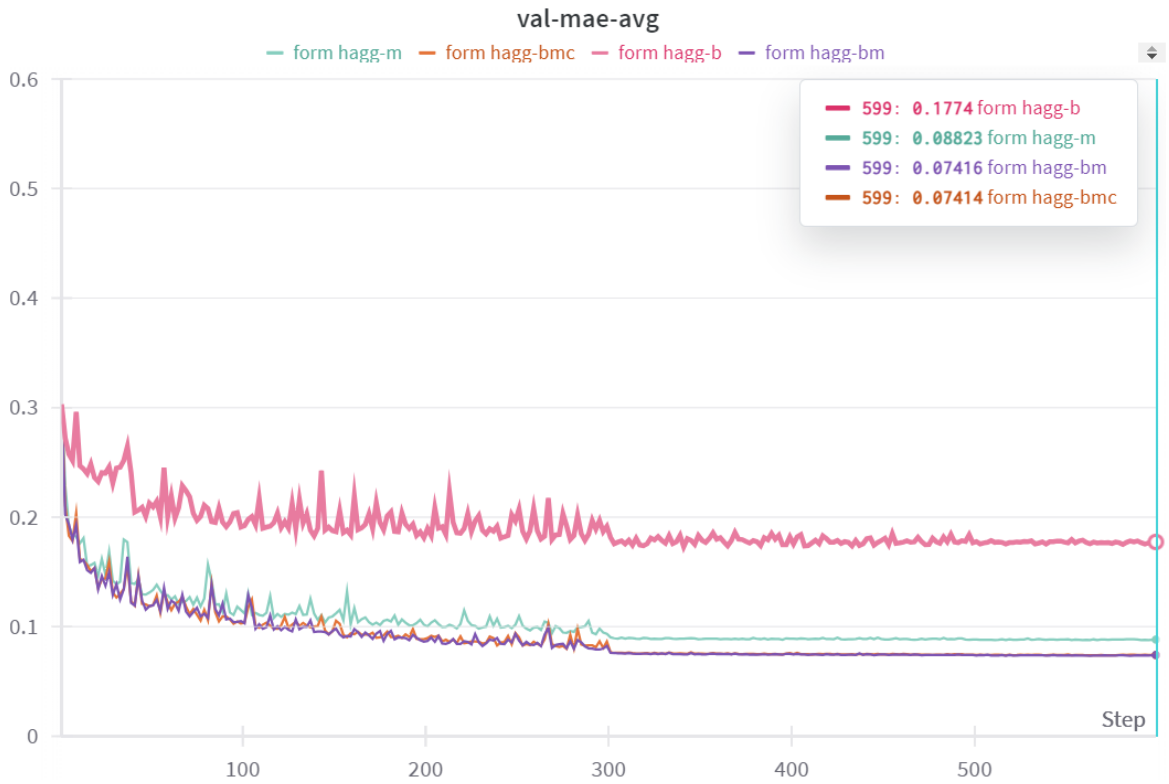


These 'unit' cell hyperedges allow for the explicit inclusion of global crystalline structure properties, i.e. point group information.

This unit cell feature also may be learned through convolution and potentially used as a dynamic state vector.

(Materials-Project) Formation Energy

Below, validation MAE is shown through training for formation energy. The total dataset includes 152,605 materials.



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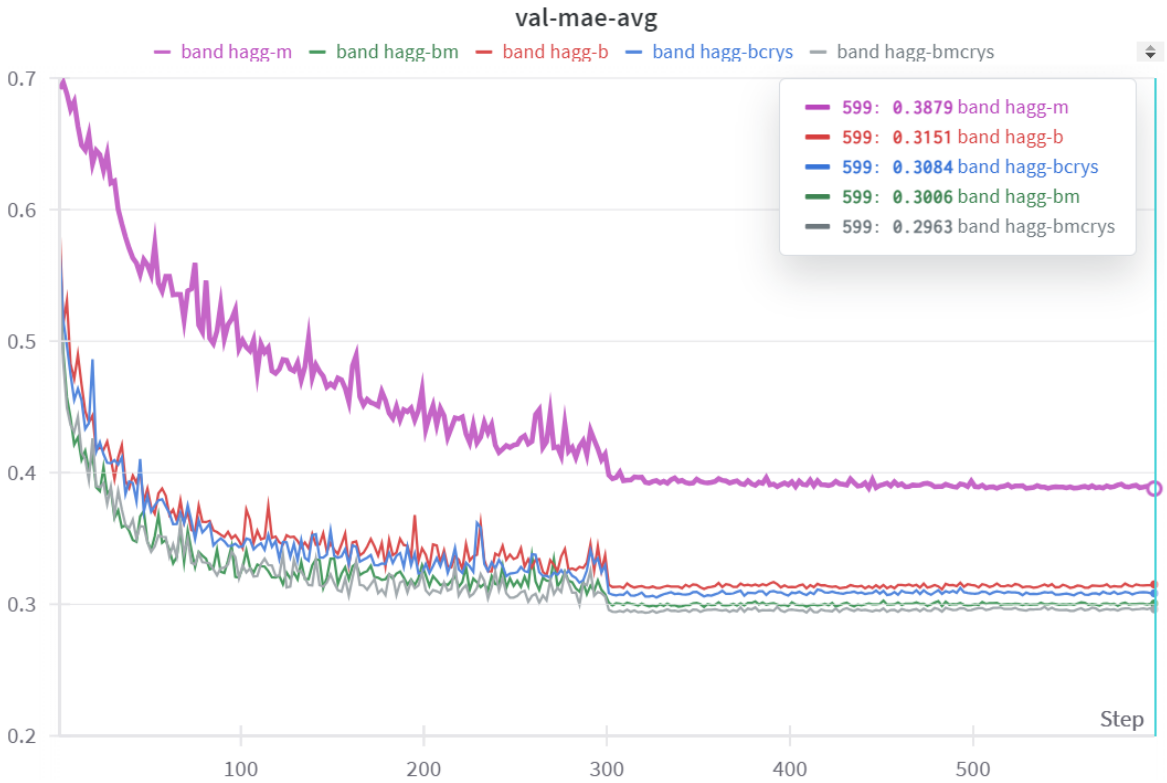

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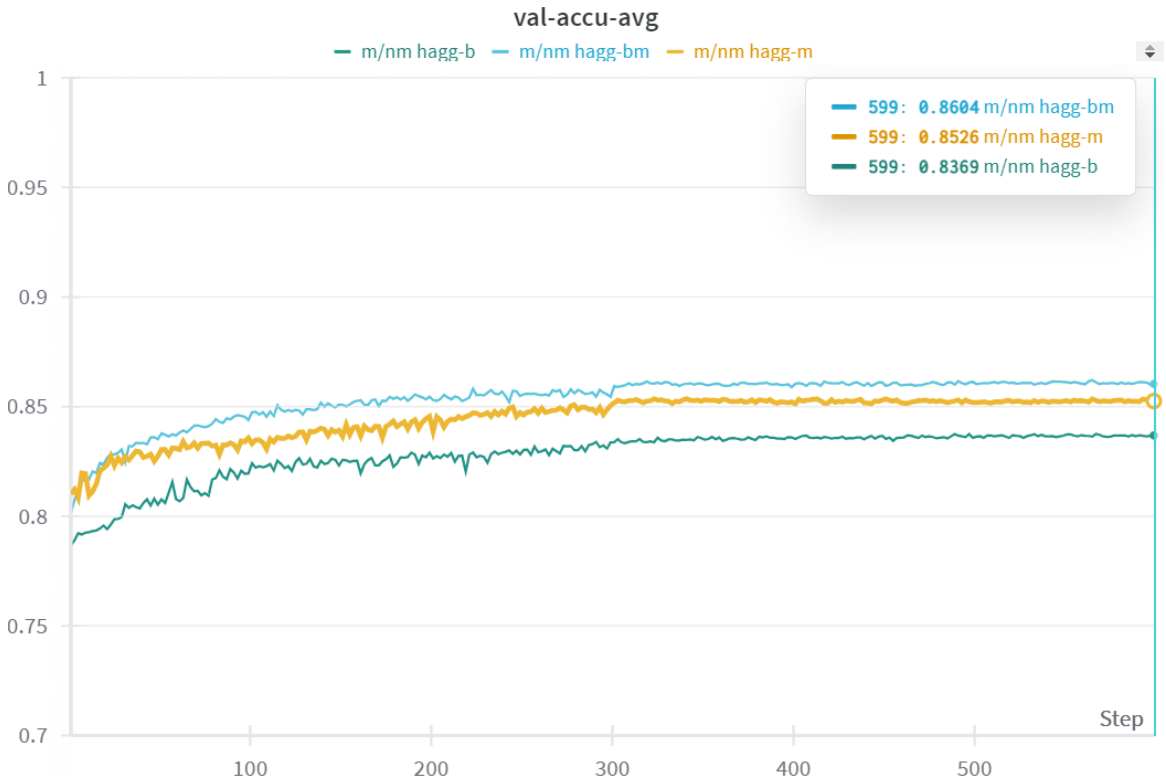
(Materials-Project) Metal/Non-metal

Below, we test metal/non-metal classification for 152,605 materials.



Model	Best Accuracy
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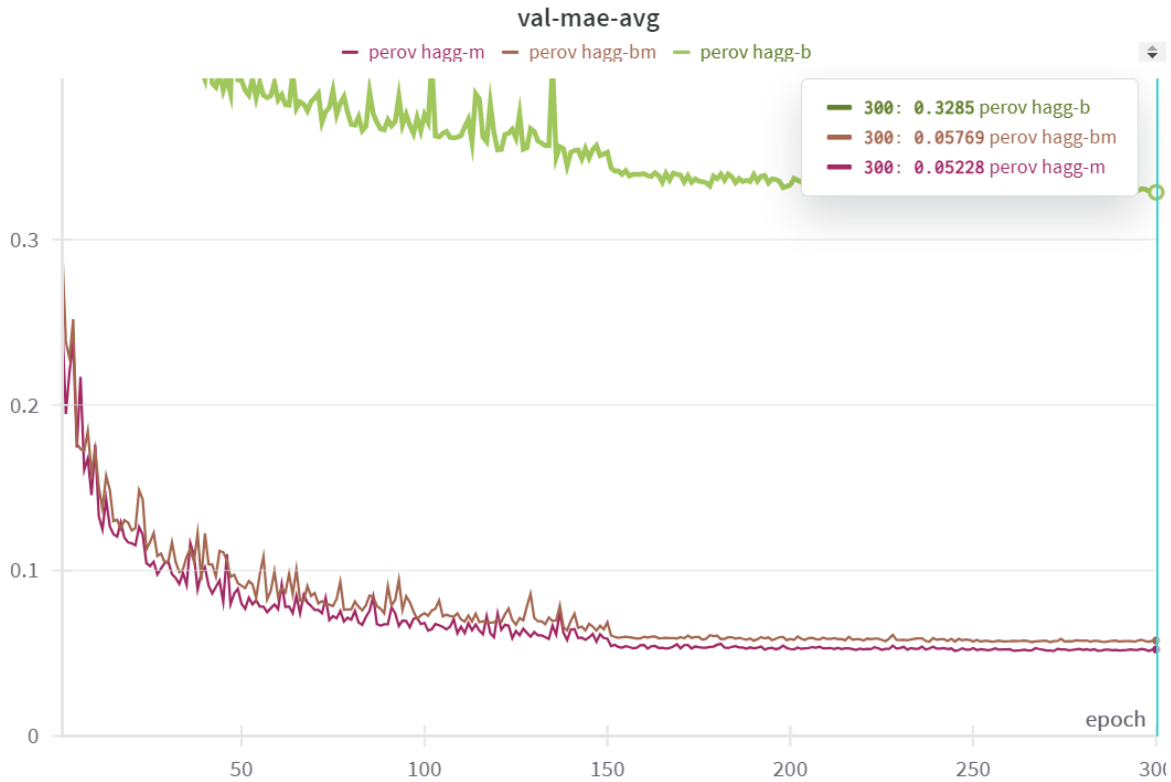
(MatBench) Perovskites

Below, we test on 18,928 calculated formation energies for Perovskites.



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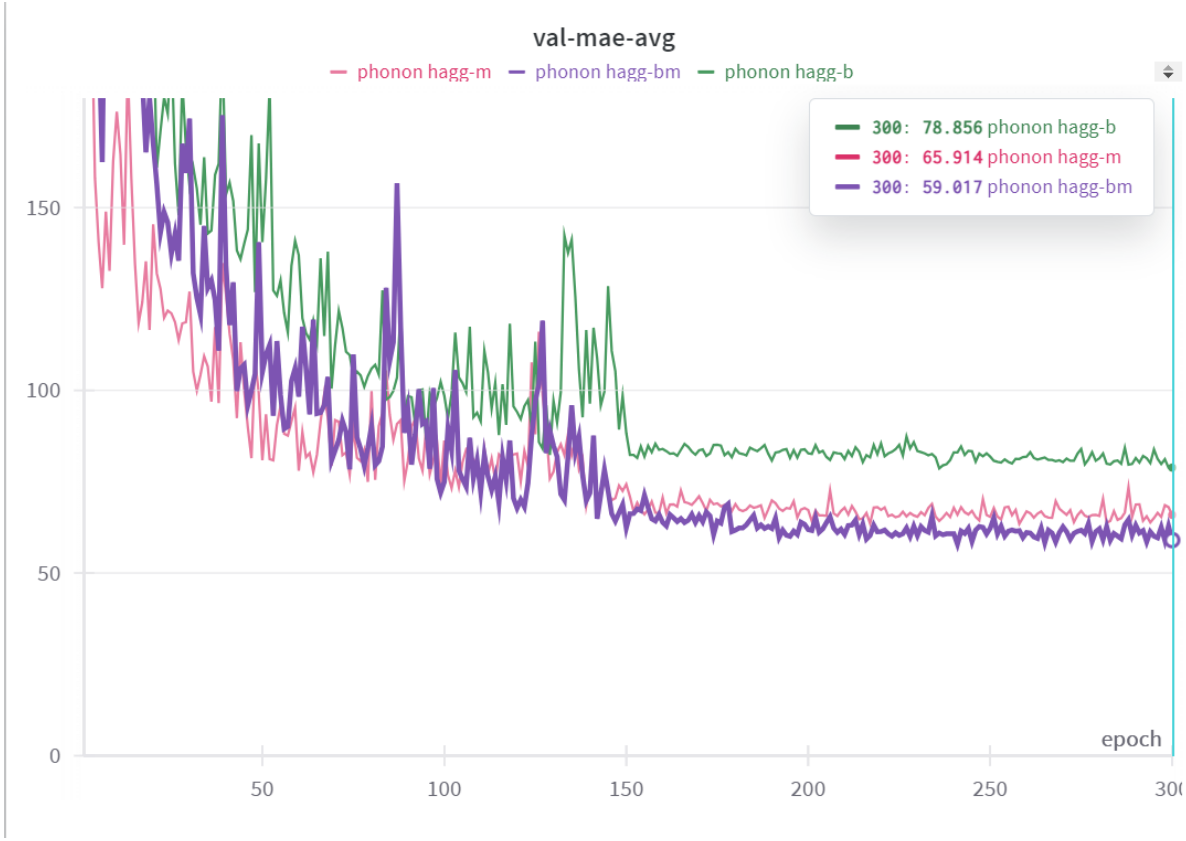
(MatBench) Phonons

Below, we test on a dataset of 1,265 materials with the target being the highest calculated frequency optical phonon mode peak.



Model	Best MAE (cm^{-1})
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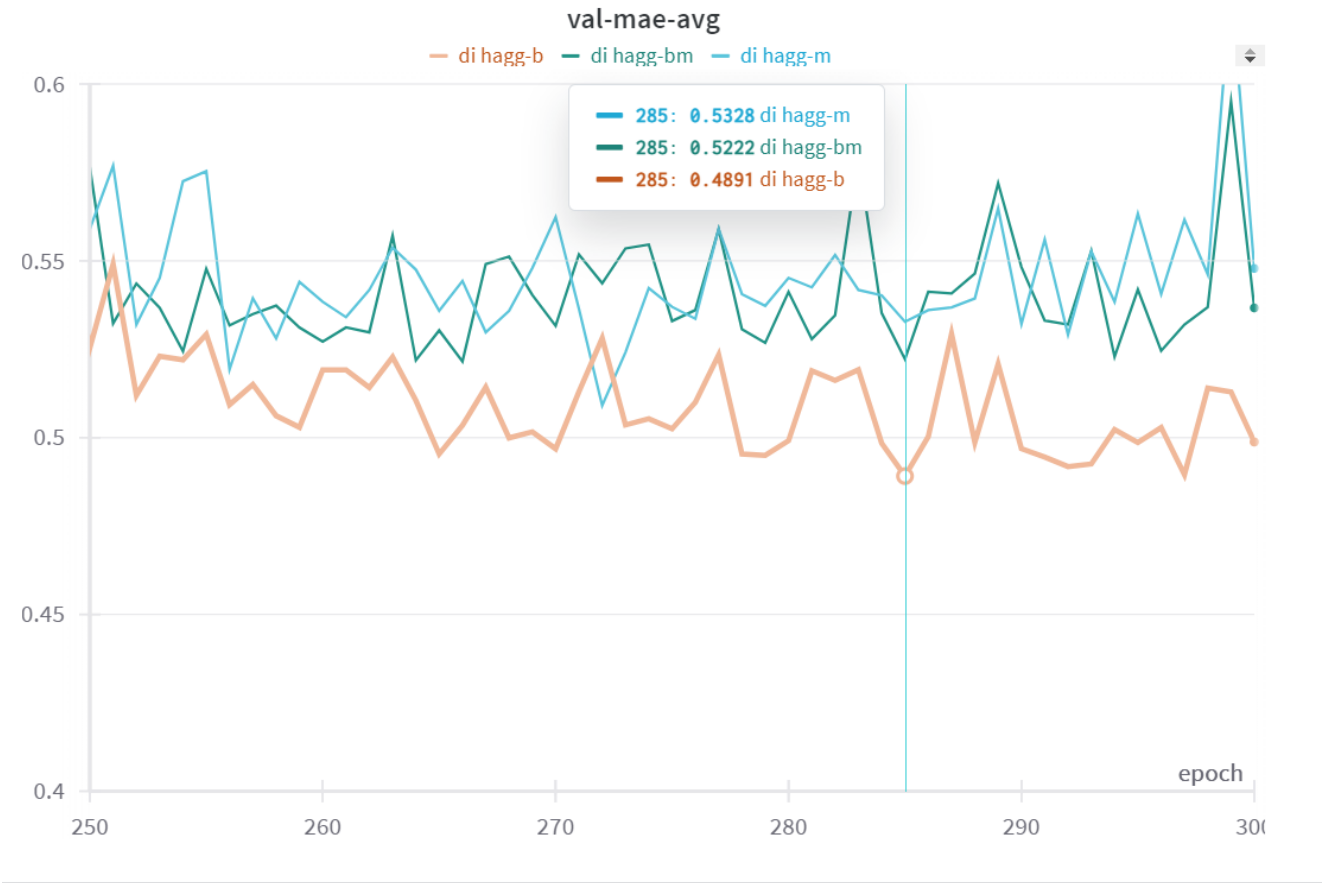
(MatBench) Dielectrics

Below, we test on the calculated refractive index of 4,764 materials



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