CHGCNN: Crystal Hypergraph Neural Networks

A Universal Framework for Material System Representations

Alex Heilman, Weiyi Gong, Qimin Yan

Department of Physics, Northeastern University

March 5, 2024

Overview

- Crystal graphs

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- Crystal graphs, MPNNs

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- Crystal graphs, MPNNs, limitations

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- Crystal hypergraphs

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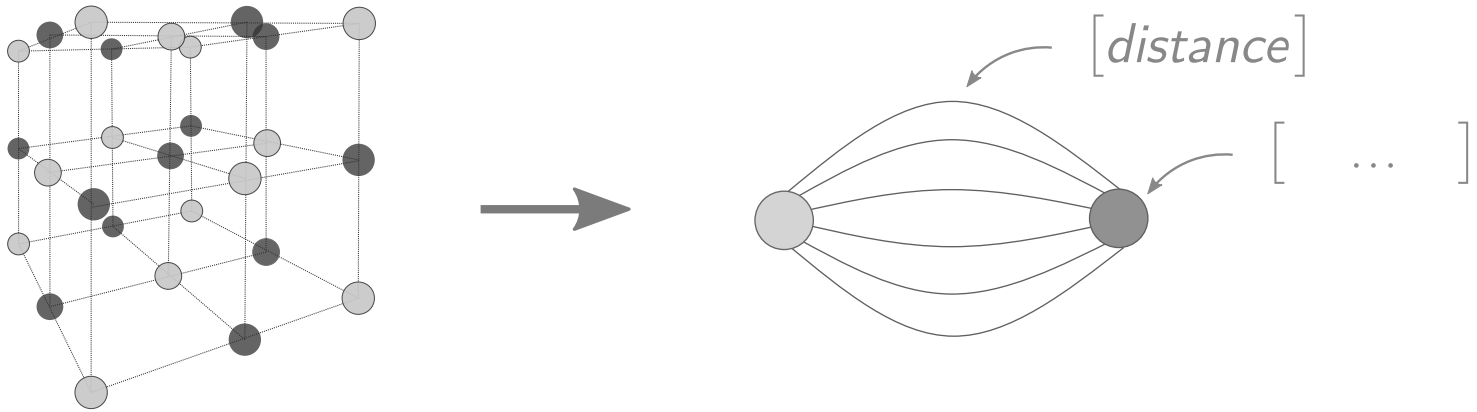
- Crystal hypergraphs, hyperedge types, MPNN extensions
- See them in action! Comparative testing

Crystal Graph Construction

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



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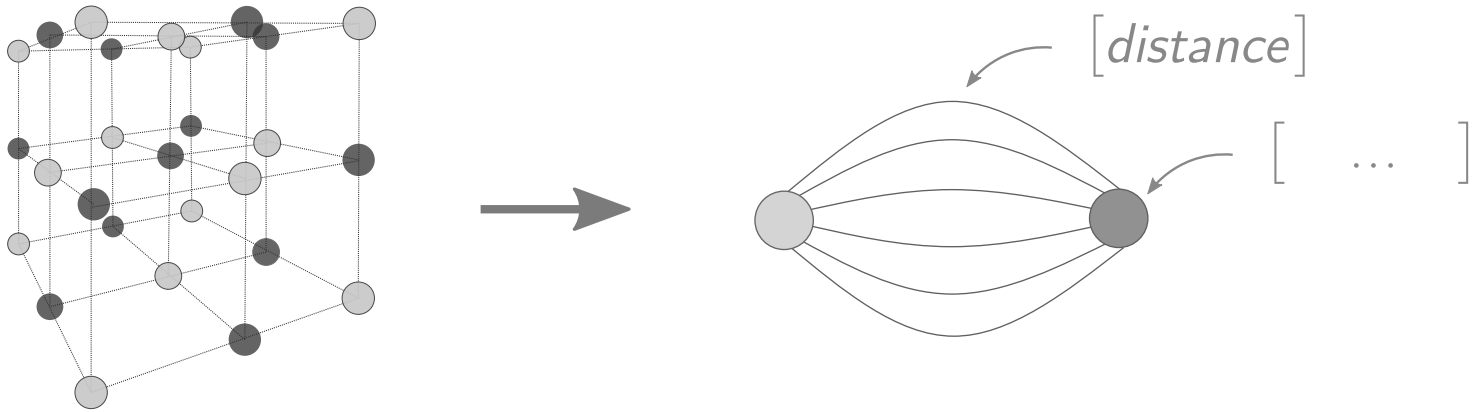

- Atoms are nodes
- Edges denote 'neighbors'; based on distance
- Feature vectors

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Message Passing on Graphs

MPNN framework[2]:

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MPNN framework[2]:

- Message m for each node i:

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

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We'll need to generalize this later...

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Graph Limitations

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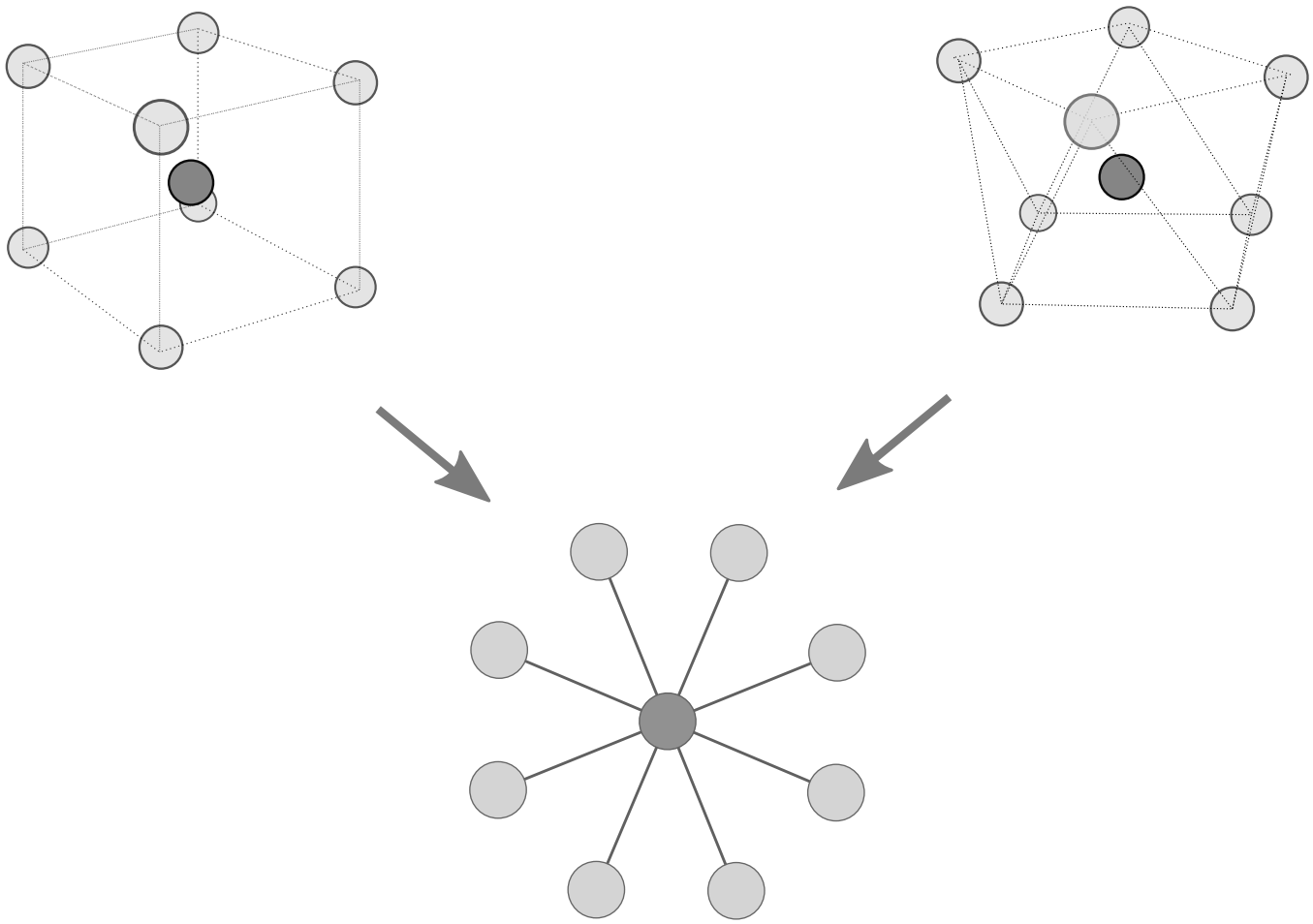

These two crystalline structures have same representations!

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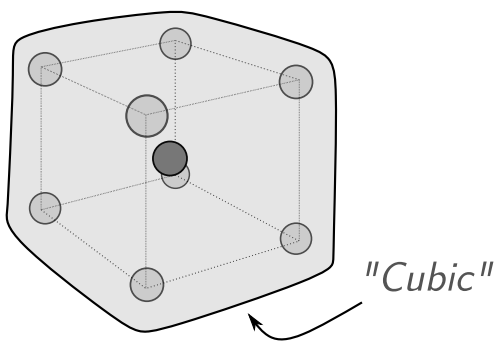
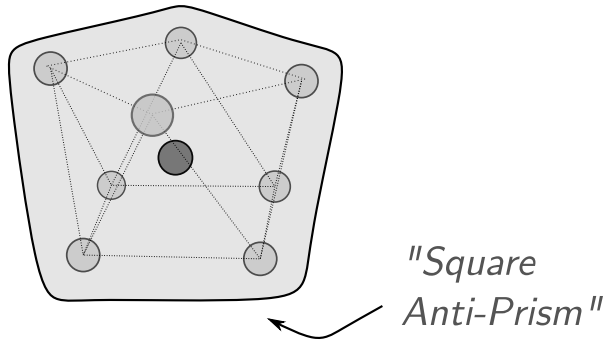
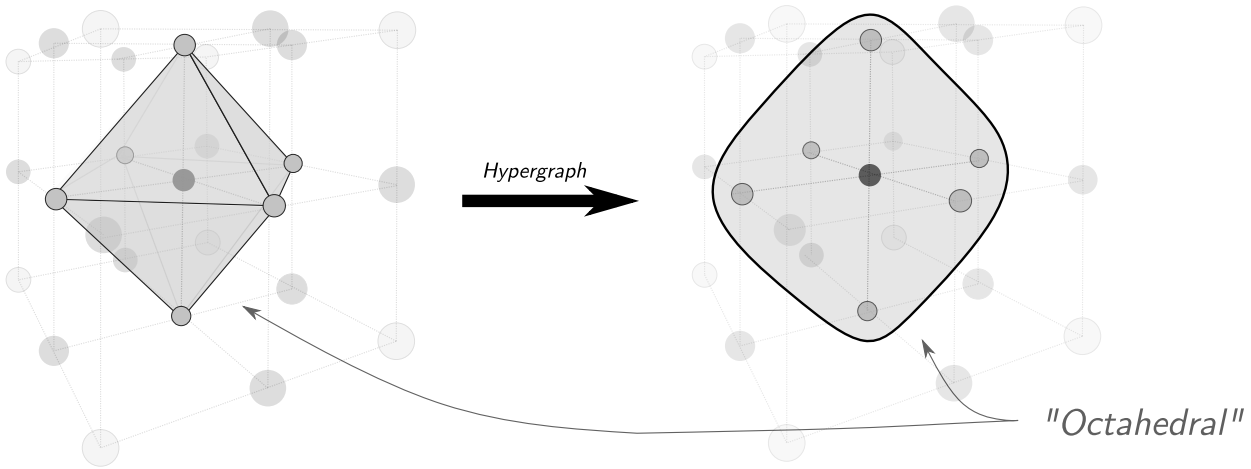




So we may define hyperedges for this local geometry.

Local Environments as Hyperedges

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



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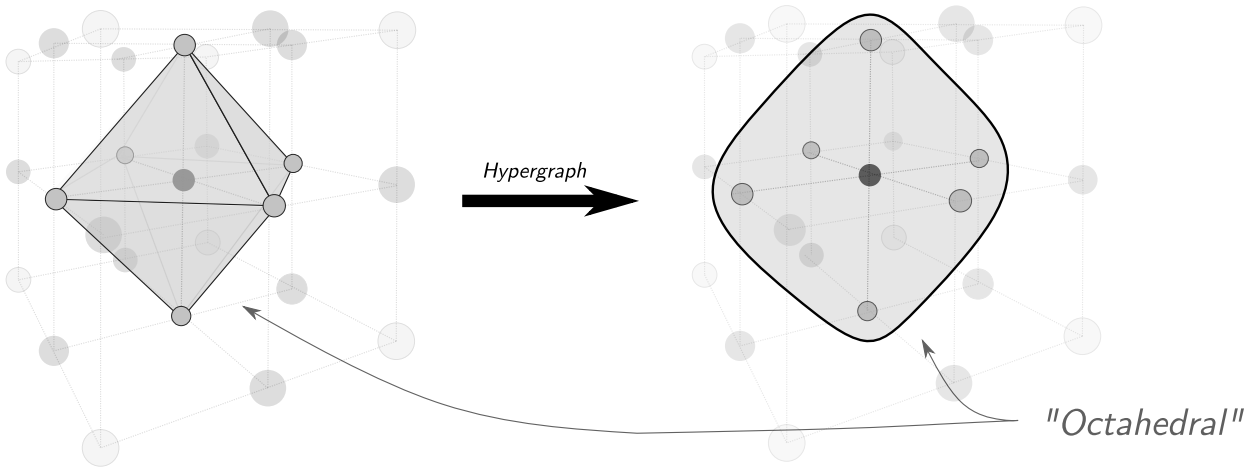

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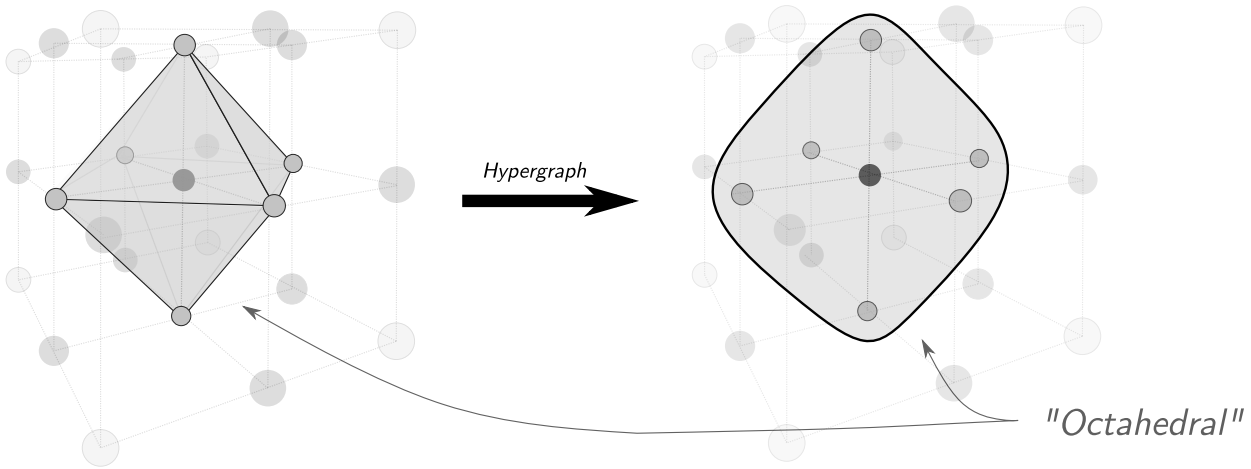

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• Geometry is encoded quantitatively with continuous symmetry measures or structure order parameters[3] as features

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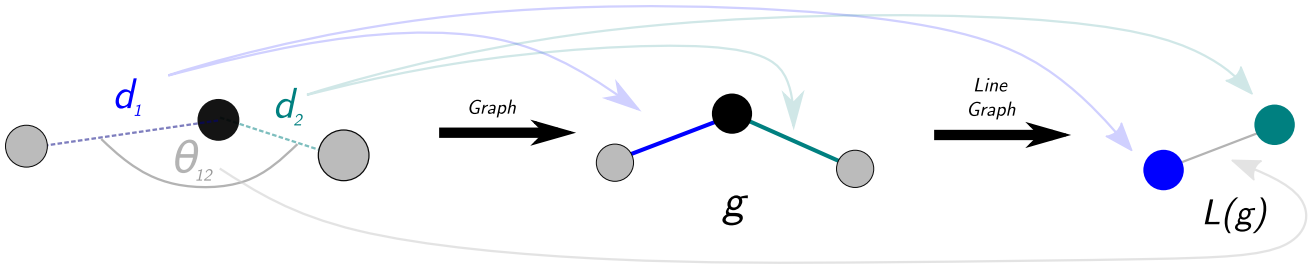
Another Example: Triplets as Hyperedges

Many modern models utilize bond angle information[4].



This is edge feature in a derived, auxiliary, 'line-graph'.

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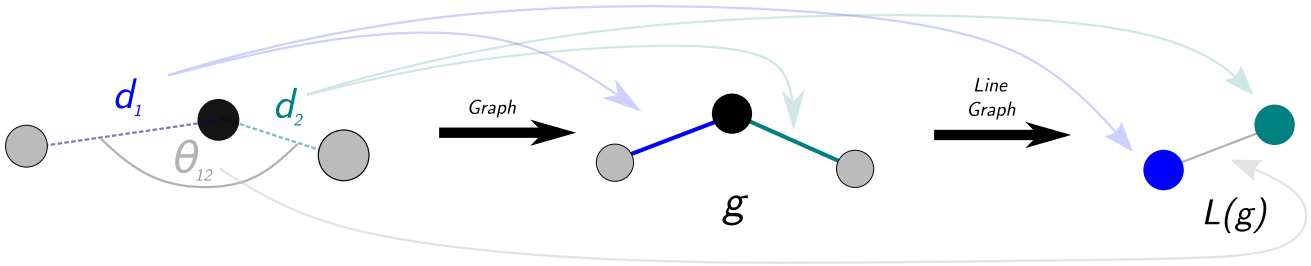

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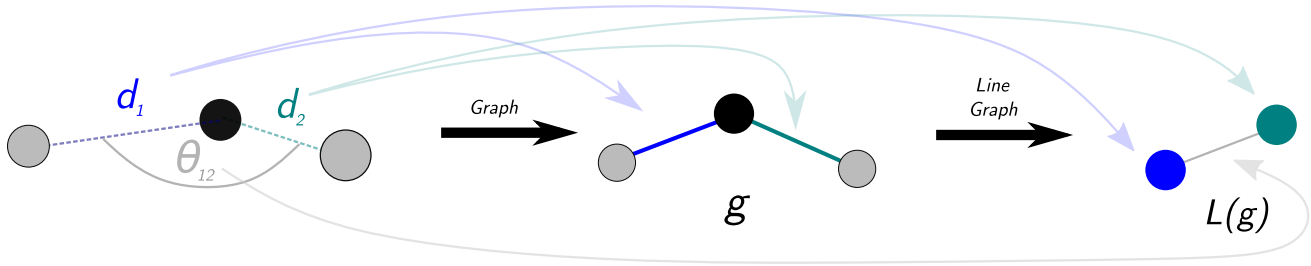

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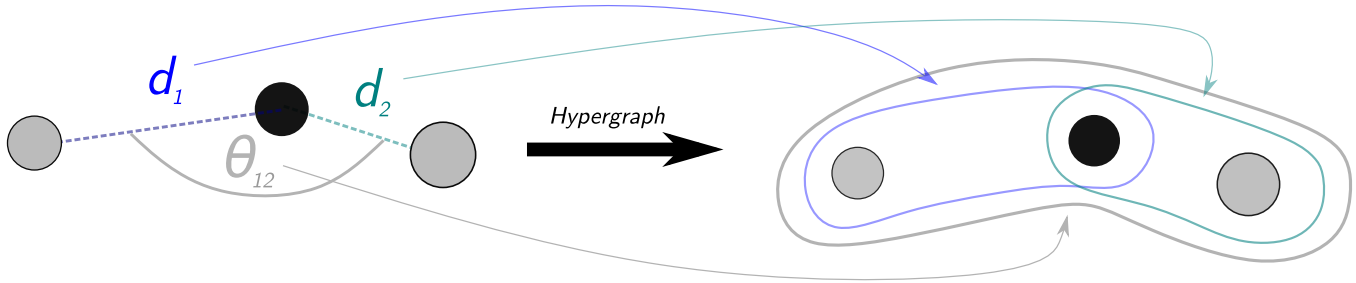

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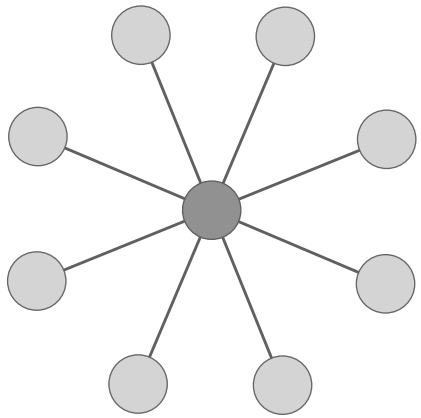

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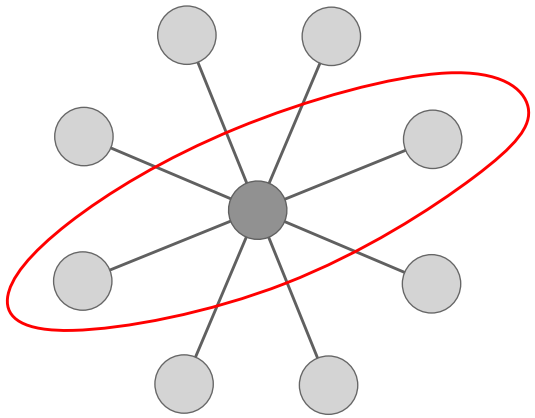

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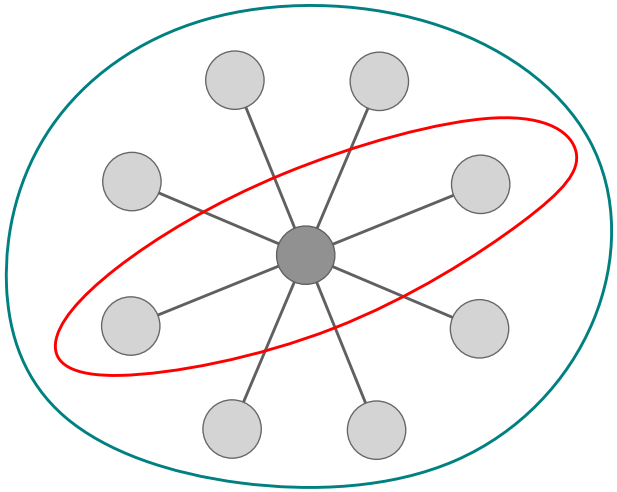

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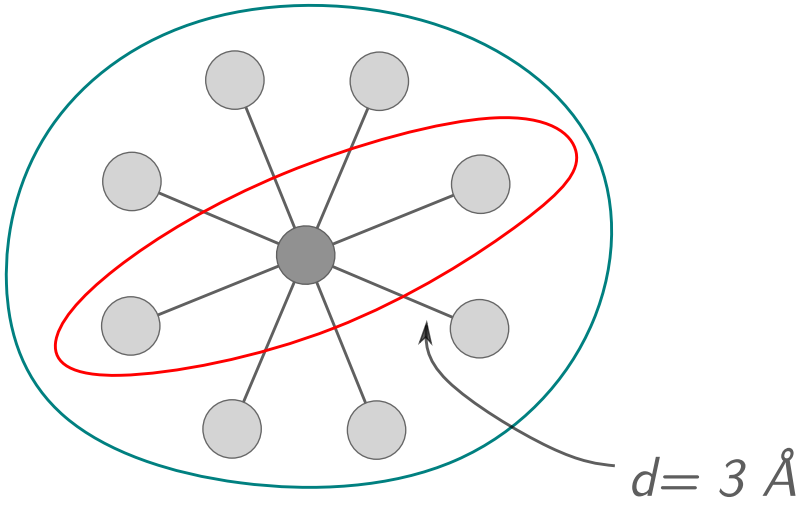

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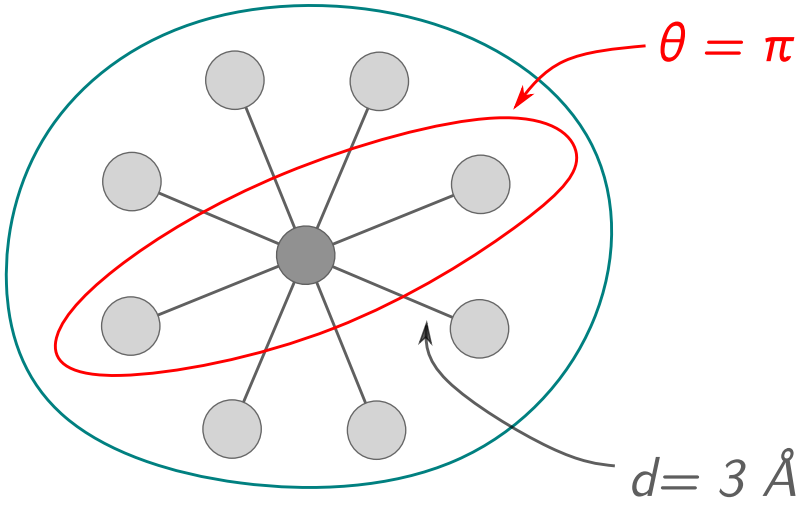

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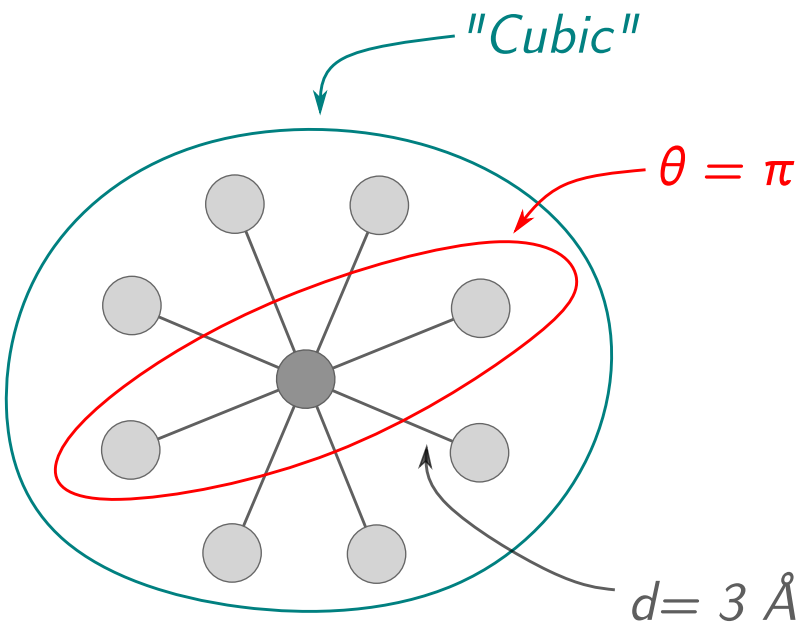

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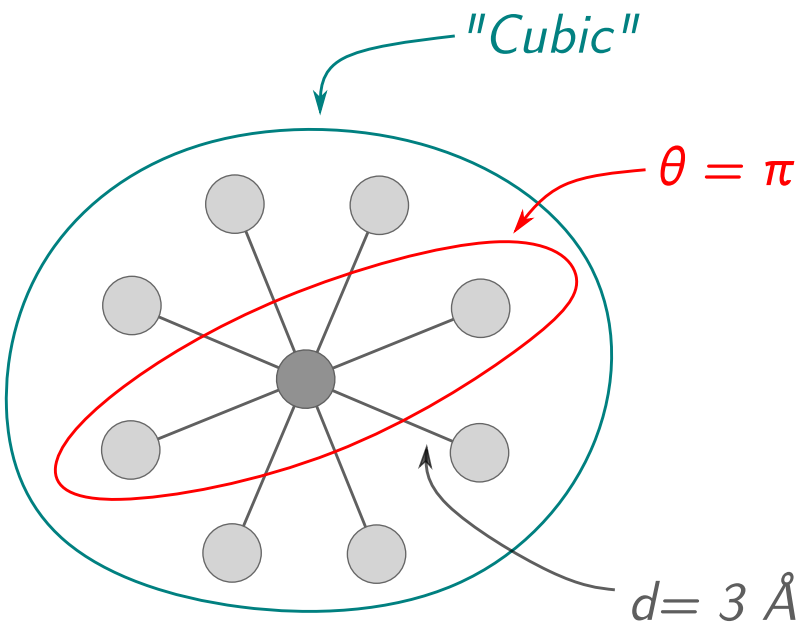

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But..... how do we update these representations?

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Three approaches are immediatley obvious.

Three Approaches to Hypergraph Convolution

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3. Hyperedge Aggregation: One message for each hyperedge

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3. $n_i' = n_i + f(n_i \oplus h_j \oplus \text{AGG}\{n_k \in h_j\}) \rightarrow$ scales as $\mathcal{O}(nm)$!

Approach (**3.**) seems a good balance of message quality and scaling behaviour

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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
Motif-only	0.387
Bond & Motif	**0.301**

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**
Bond & Motif	0.058

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

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

Looking Forward

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- Improve convolutional layers for hyperedges.

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- Improve convolutional layers for hyperedges.
- Apply to novel tasks related to geometry.

Thanks for your time!

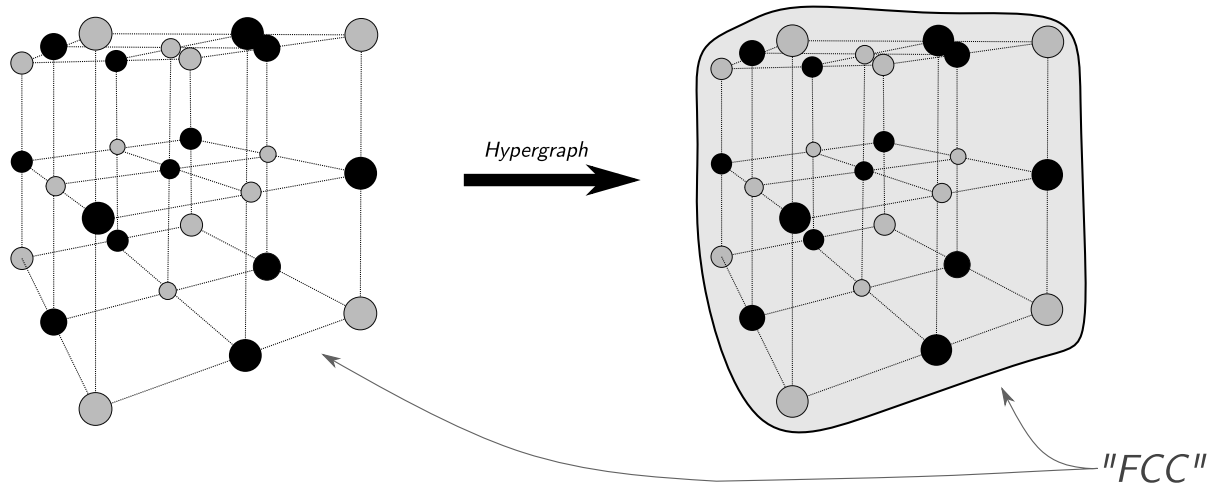
Hyperedge Index

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.

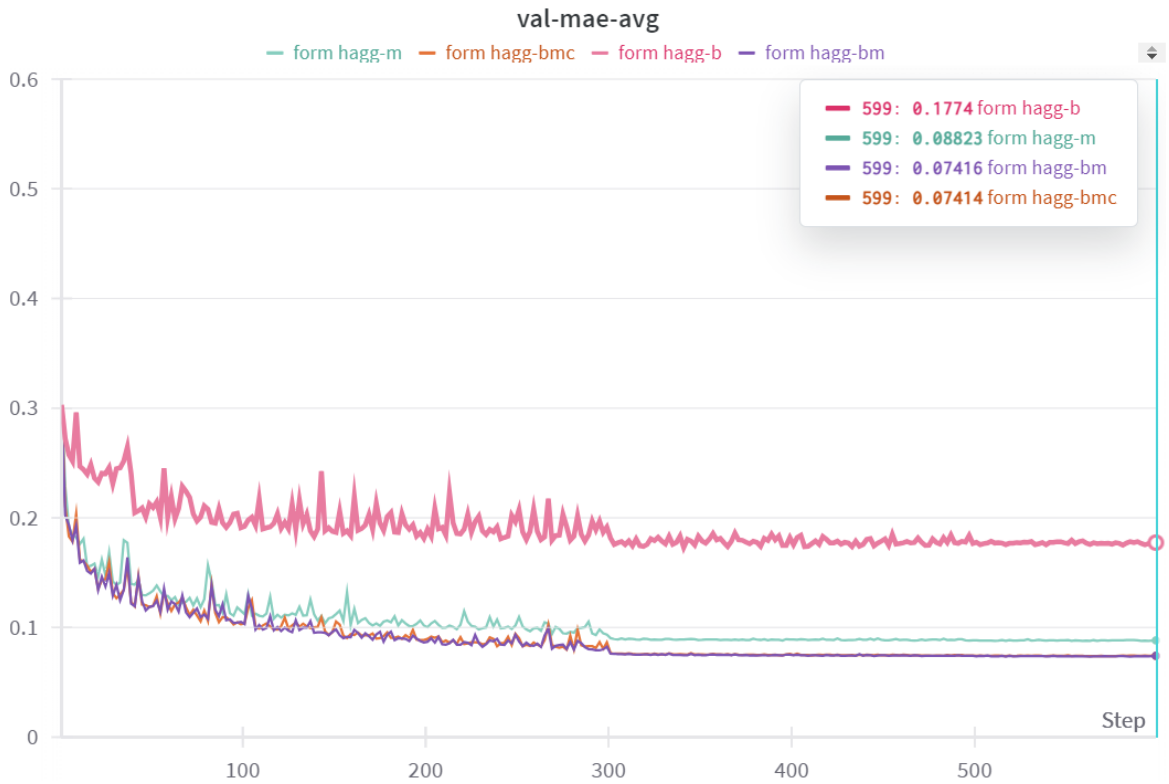


These 'unit' cell hyperedges allow for the explicit inclusion of global crystalline structure properties, i.e. point group information.

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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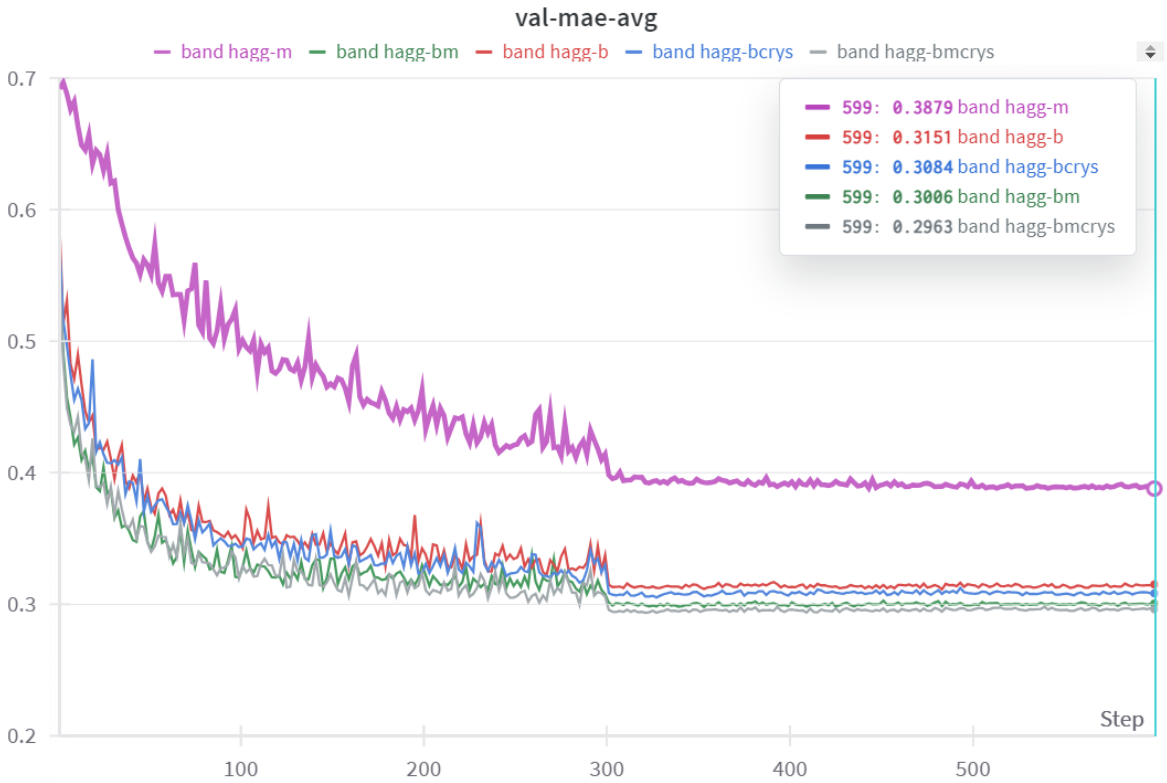
Below are results for band gap training, based on a dataset including 152,605 materials.



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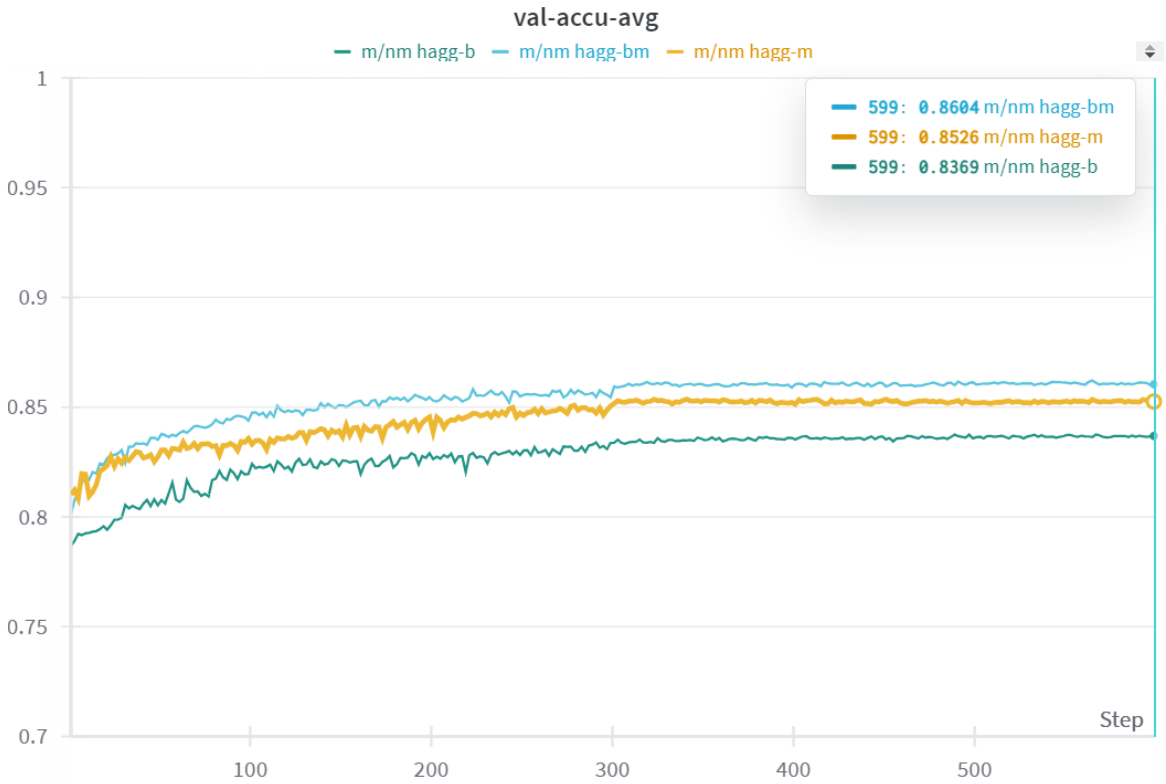

Model	Best Accuracy
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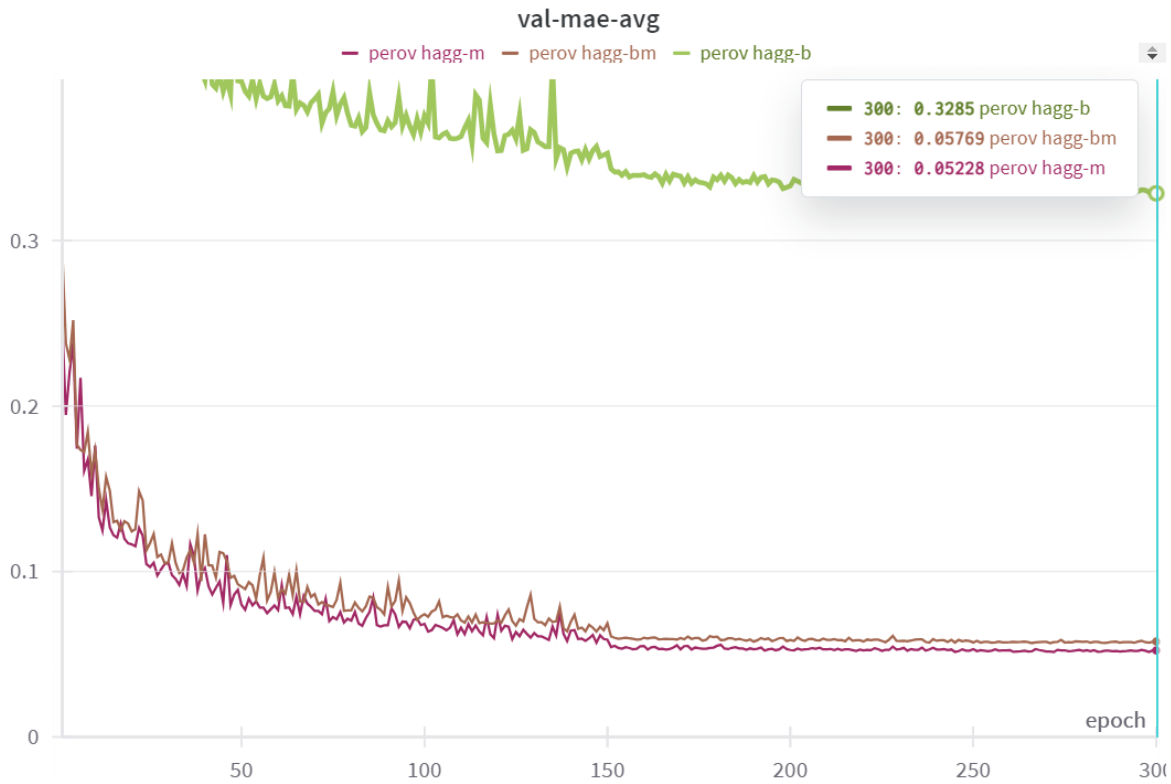
(MatBench) Perovskites

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



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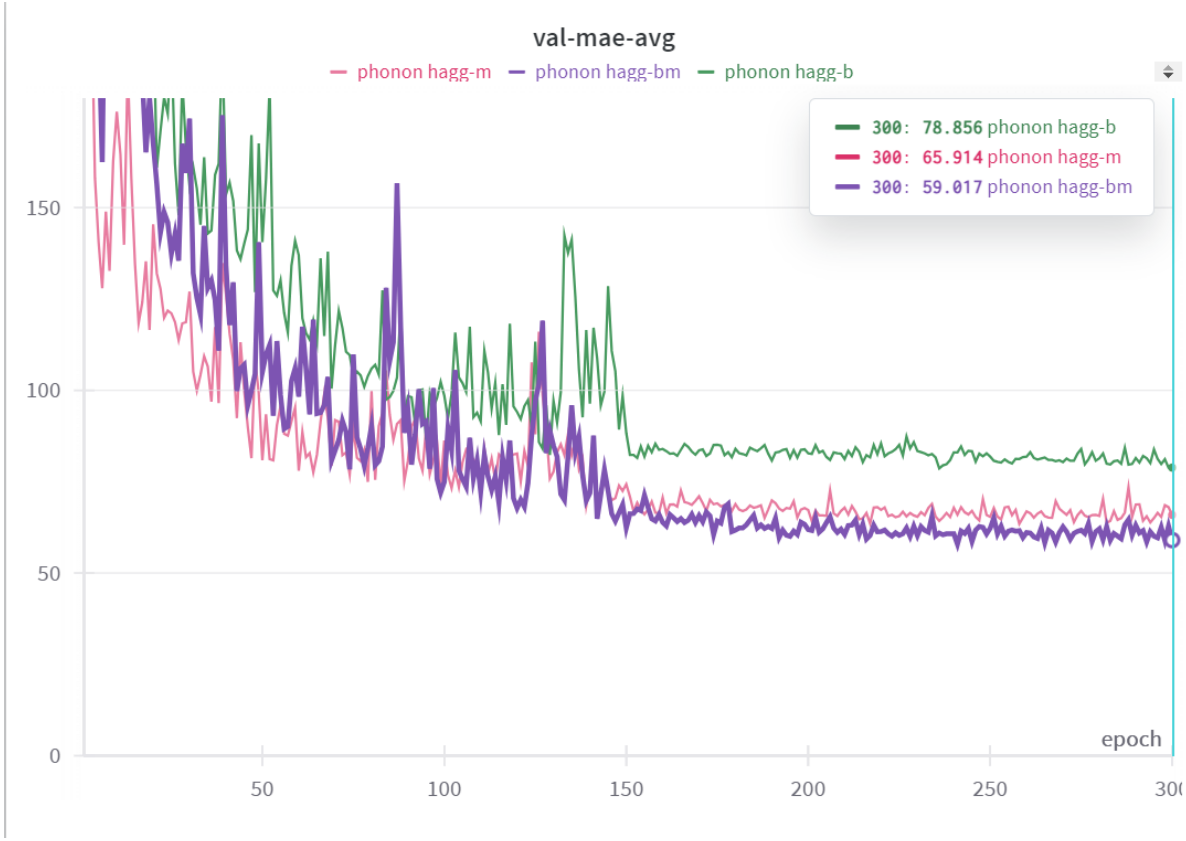
Below, we test on a dataset of 1,265 materials with the target being the highest calculated frequency optical phonon mode peak.



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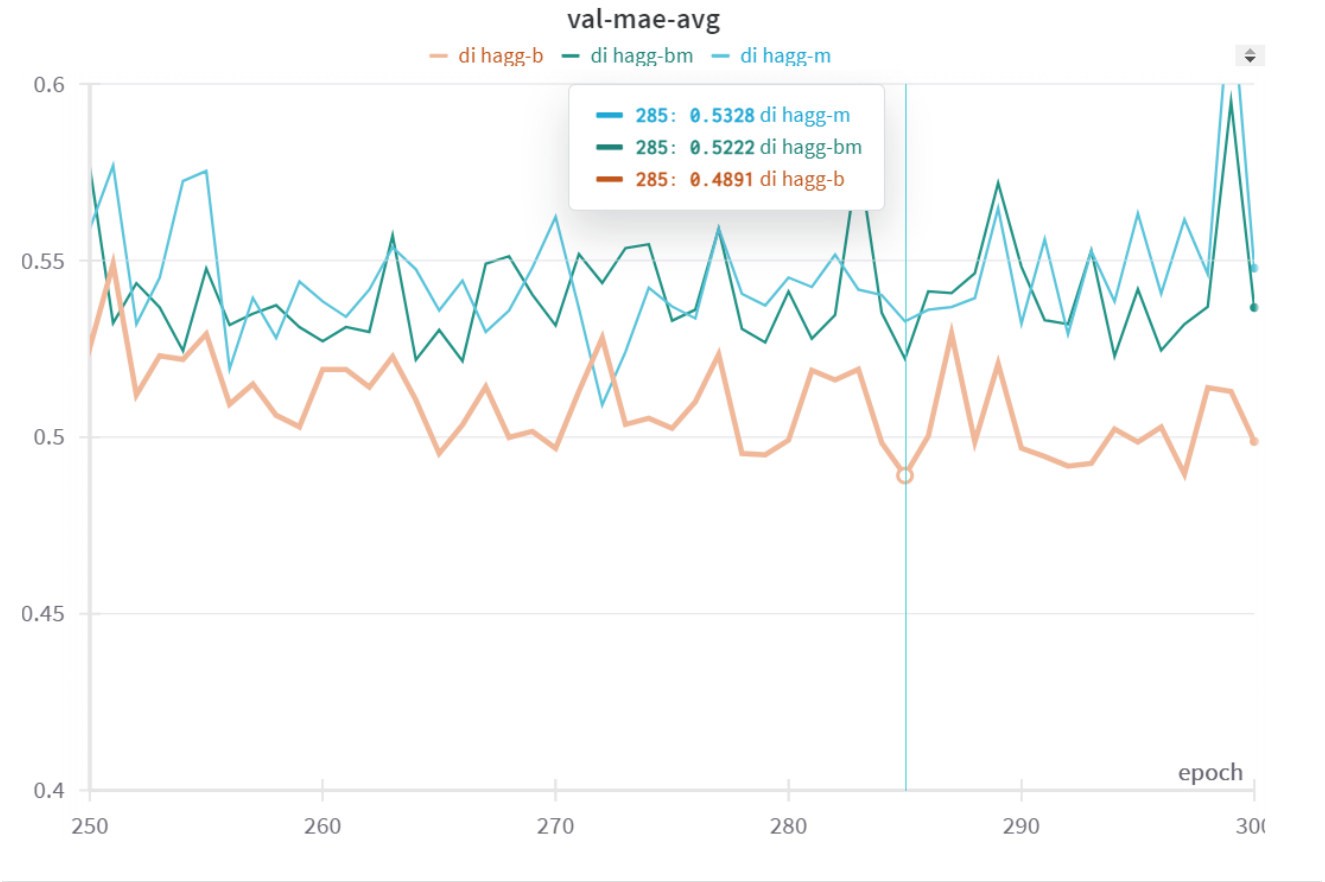
Bond & Motif	**59.0**

(MatBench) Dielectrics

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



Model	Best MAE
Bond-only	**.4891**
Motif-only	.5328
Bond & Motif	.5222