DEEP LEARNING FOR MOLECULAR DATA

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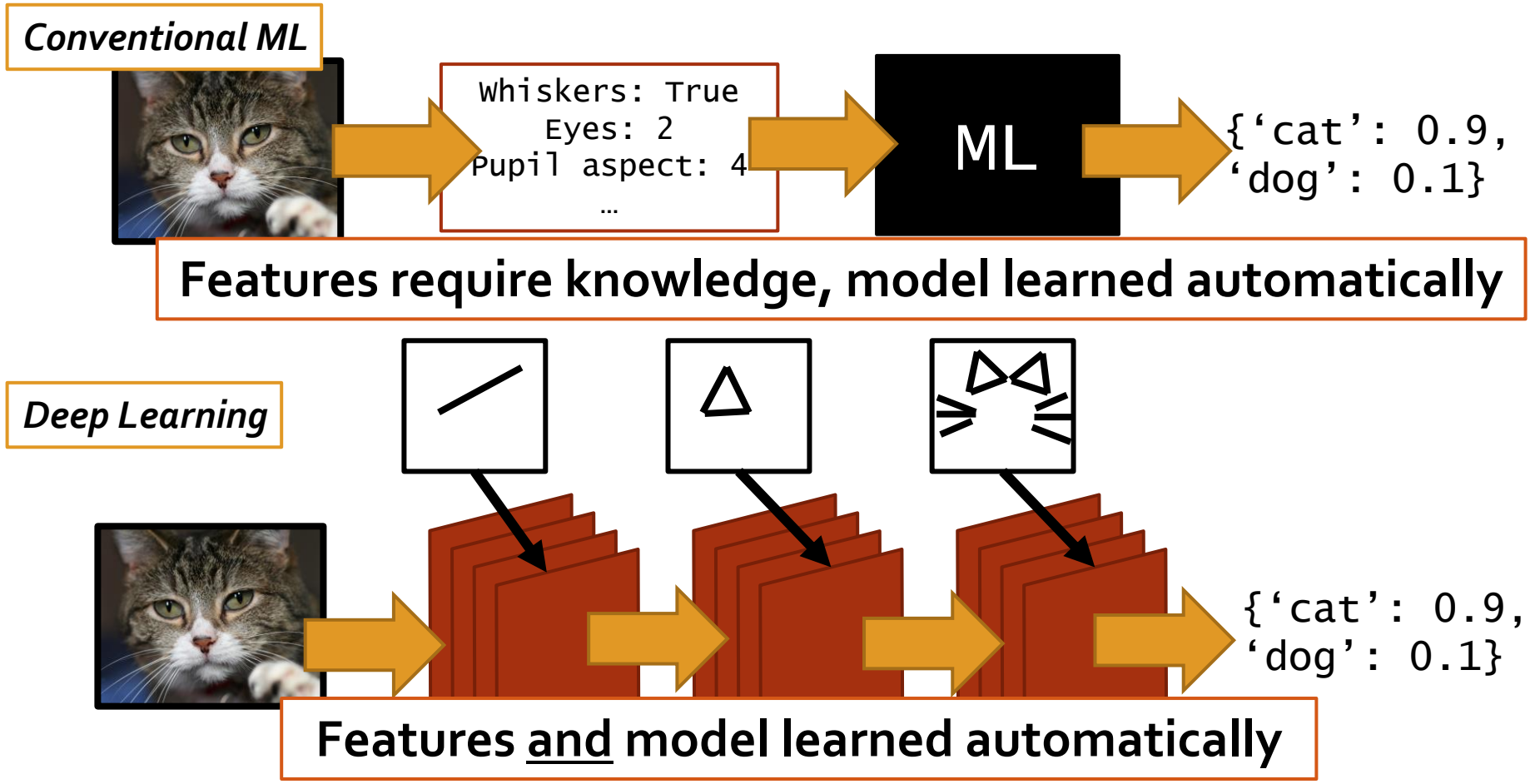
What is different about deep learning?



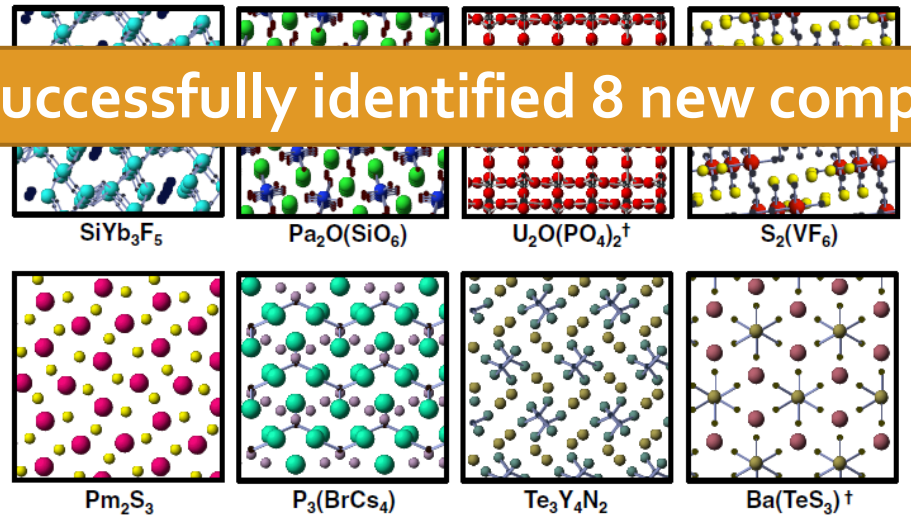
FIRST, WHAT CONVINCED ME DEEP LEARNING WAS WORTHWHILE

Predicting Crystalline Materials

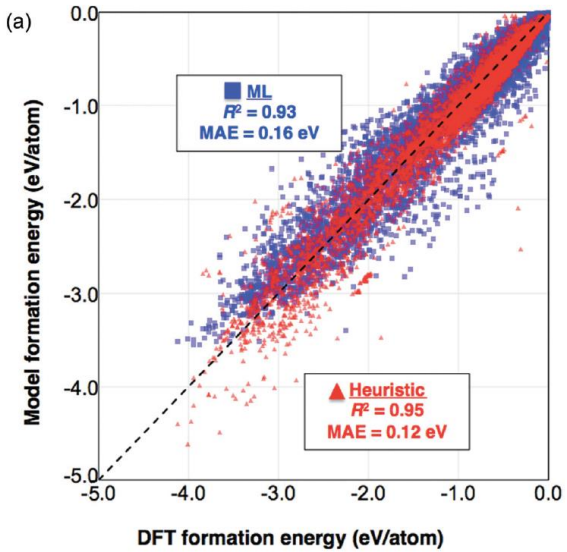
Goal: Identify new crystalline materials

Model: Given, composition predict ΔH_f

Initial attempt: 2014



Successfully identified 8 new compounds!



Error still ~10% typical values

Can we do even better with deep neural networks?

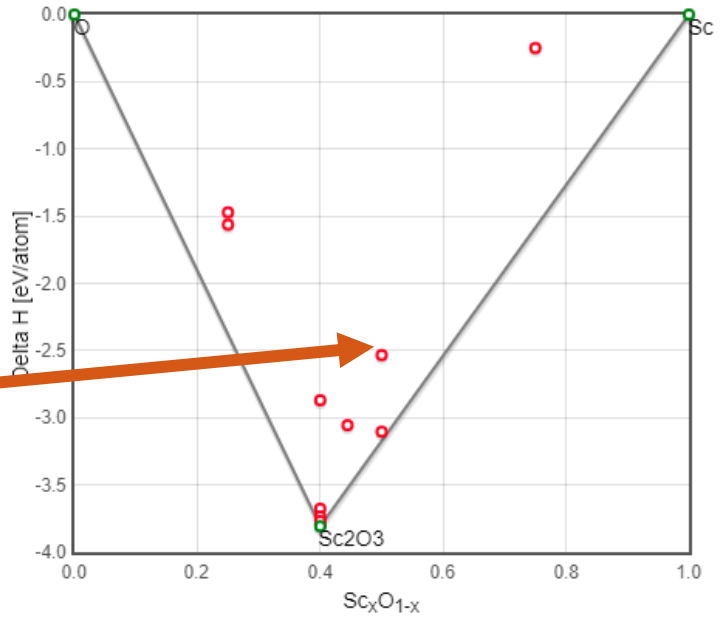
Ref: Meredig et al. PRB. (2014), 094104. doi: 10.1103/PhysRevB.89.094104

Training Data

Data Source: Open Quantum Materials Database
~470k DFT Calculations

Why OQMD?

- ✔ Open ✔ Large
- ✔ Contains unstable materials



Training Set:

Subset: Lowest energy structure

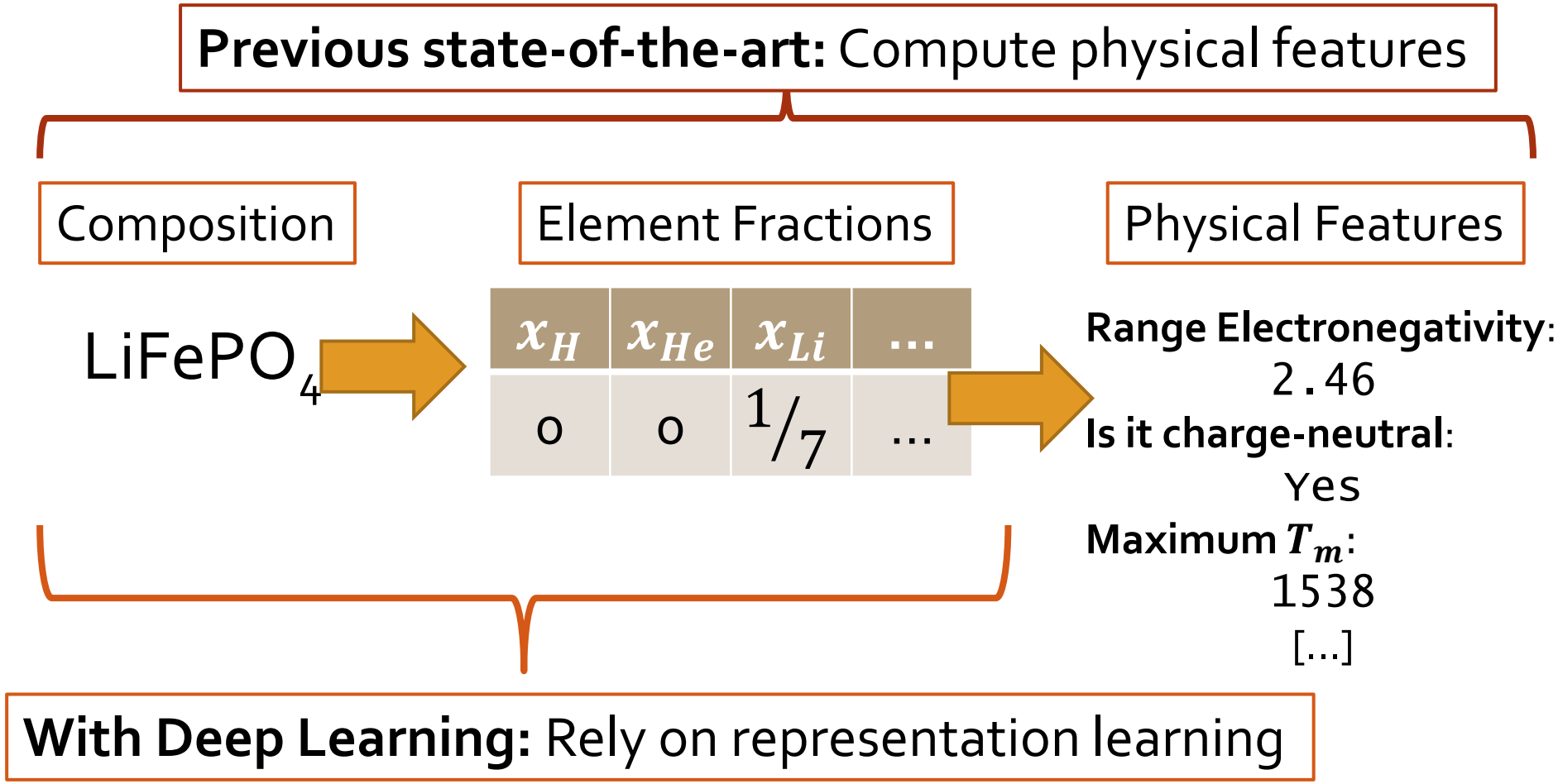
Size: 230336

Input: Composition

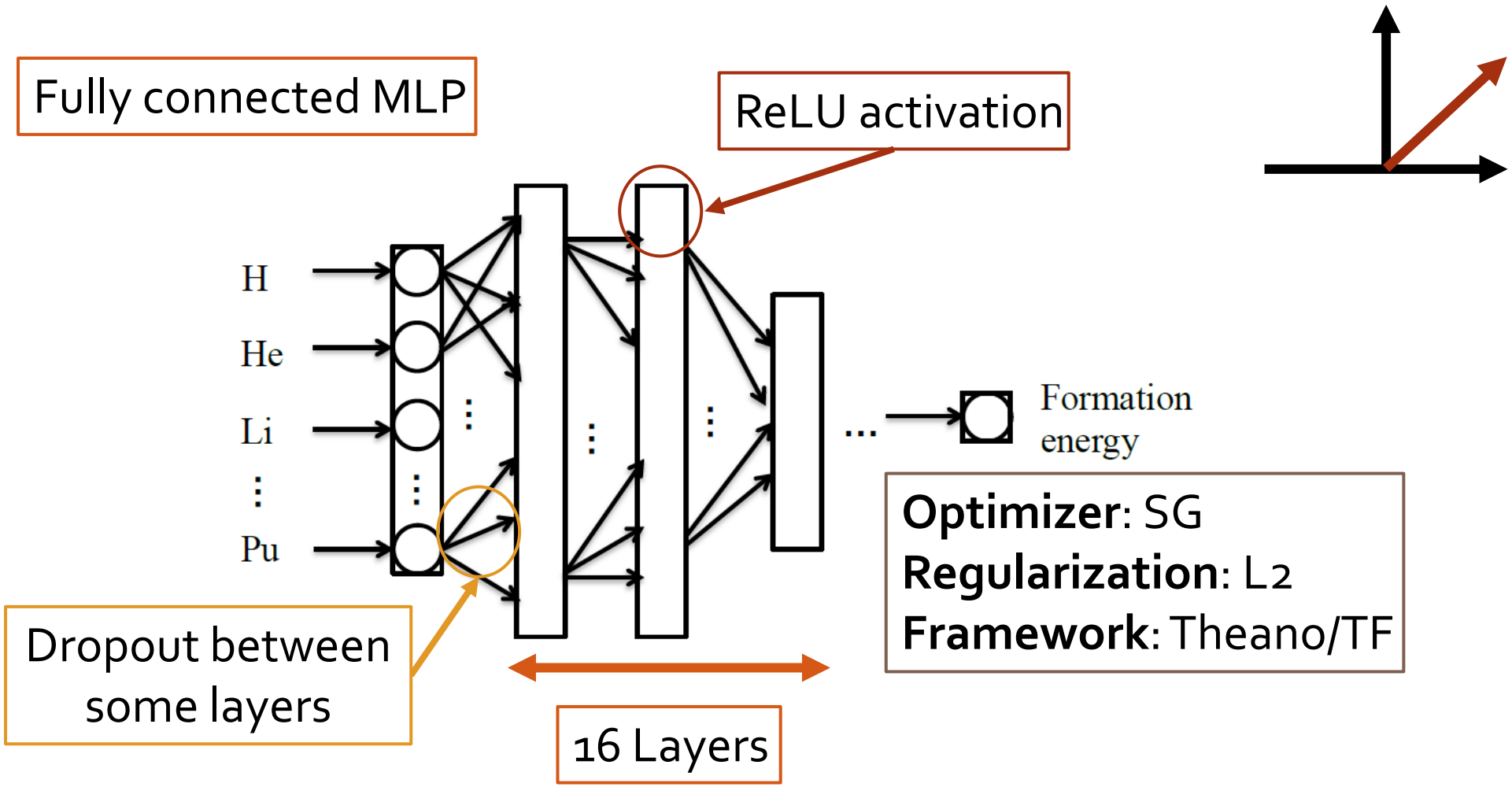
Output: Formation Enthalpy ΔH_f

Ref: Jha et al., Sci. Rep. (2020) 17593

Input Features

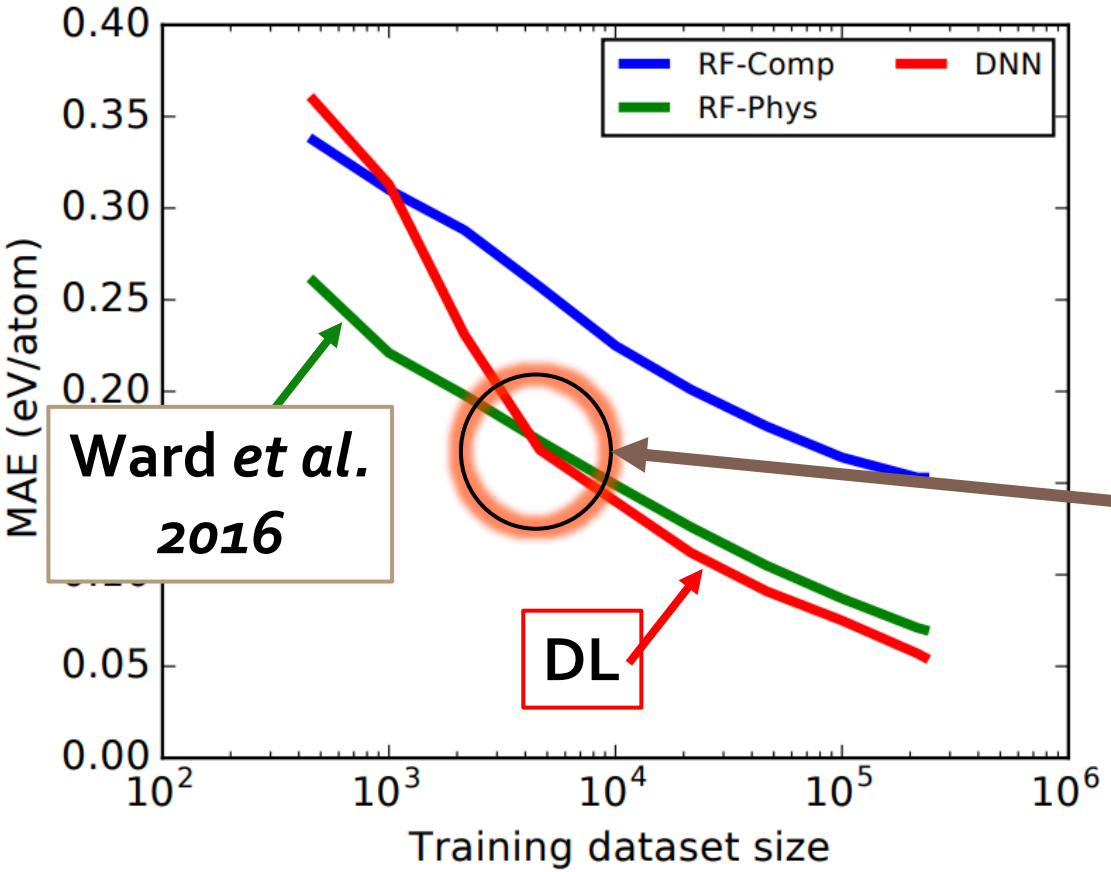


ElemNet Architecture: Nothing Fancy



Ref: Jha et al., Sci. Rep. (2020) 17593

Better than conventional learning?



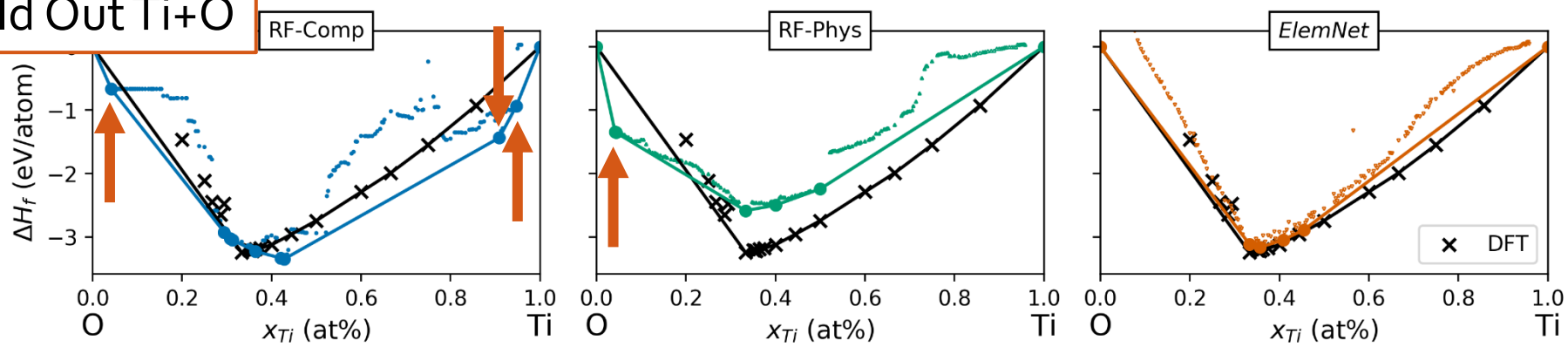
(ML) Meredig 2014: 0.092 eV/atom
(ML) Ward 2016: 0.070 eV/atom
(DL) Jha 2017: 0.055 eV/atom

Beats state-of-the-art with
no input from materials scientist,
and only 4000 data points!

Ref: Jha et al., Sci. Rep. (2020) 17593

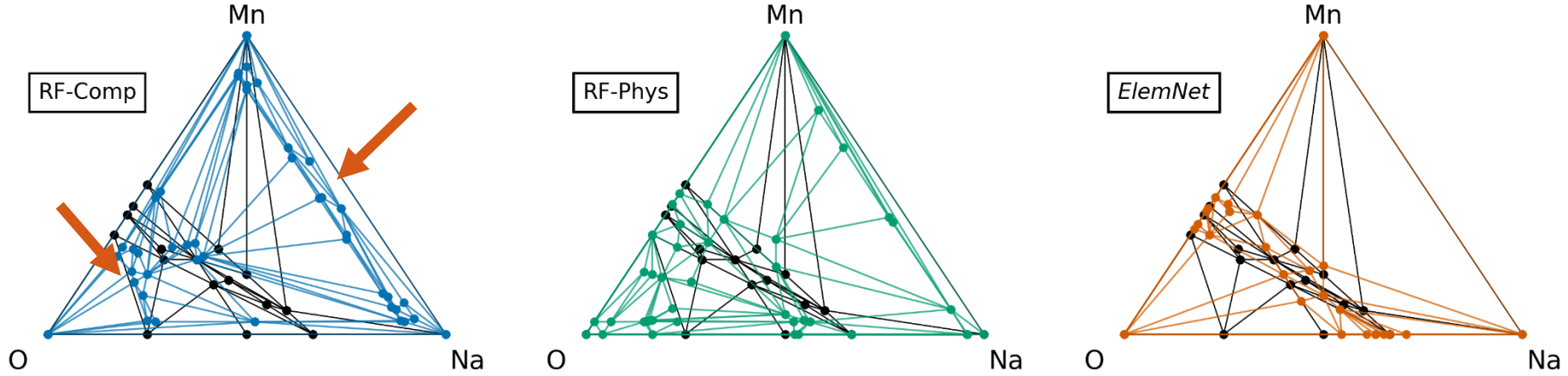
Can DL interpolate between elements?

Hold Out Ti+O



Deep Learning Yields Fewer Spurious Predictions

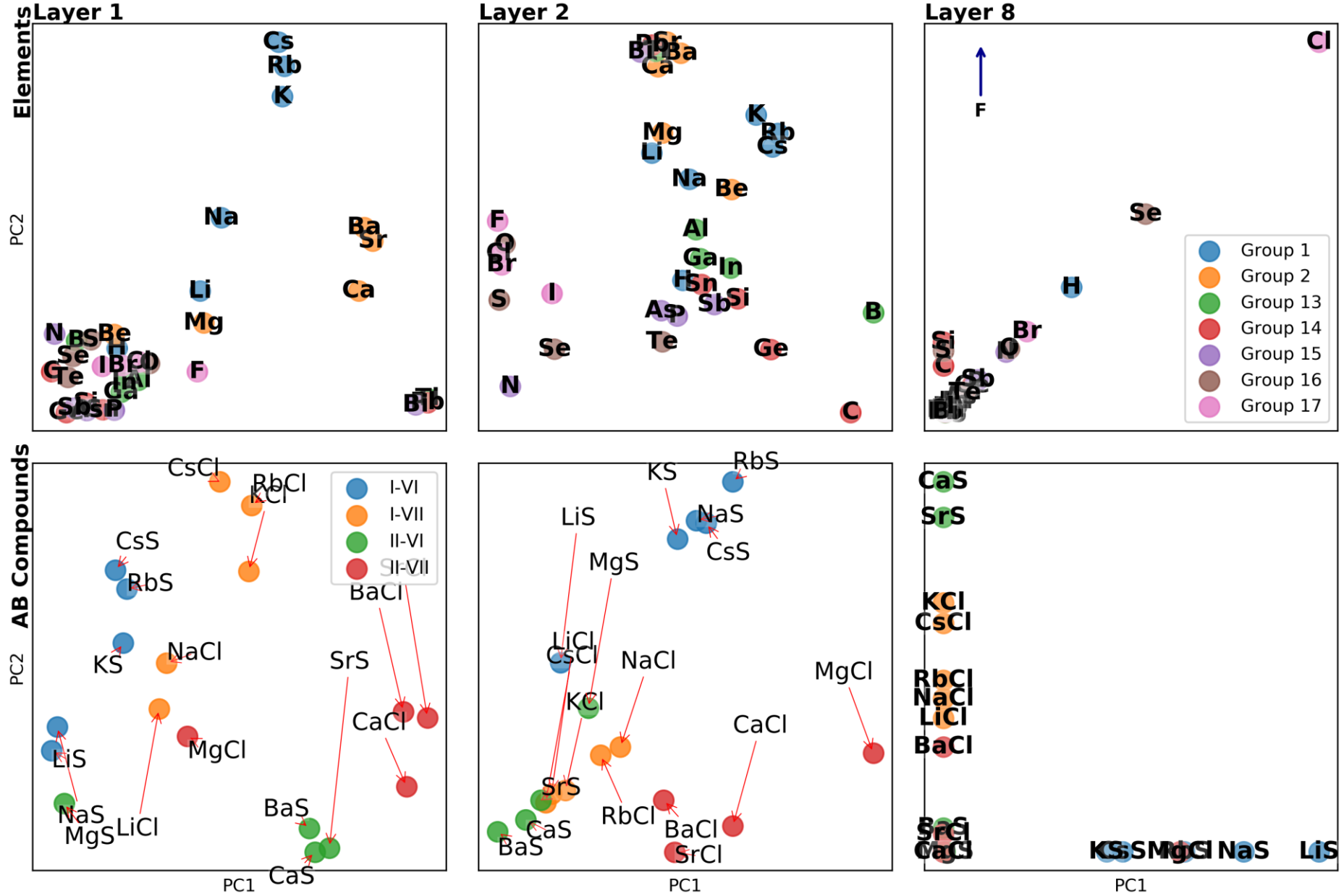
Hold Out Na-Fe-Mn-O



How is this possible? Model hasn't seen Ti-O bonds

Ref: Jha et al., Sci. Rep. (2020) 17593

How is it working so well?



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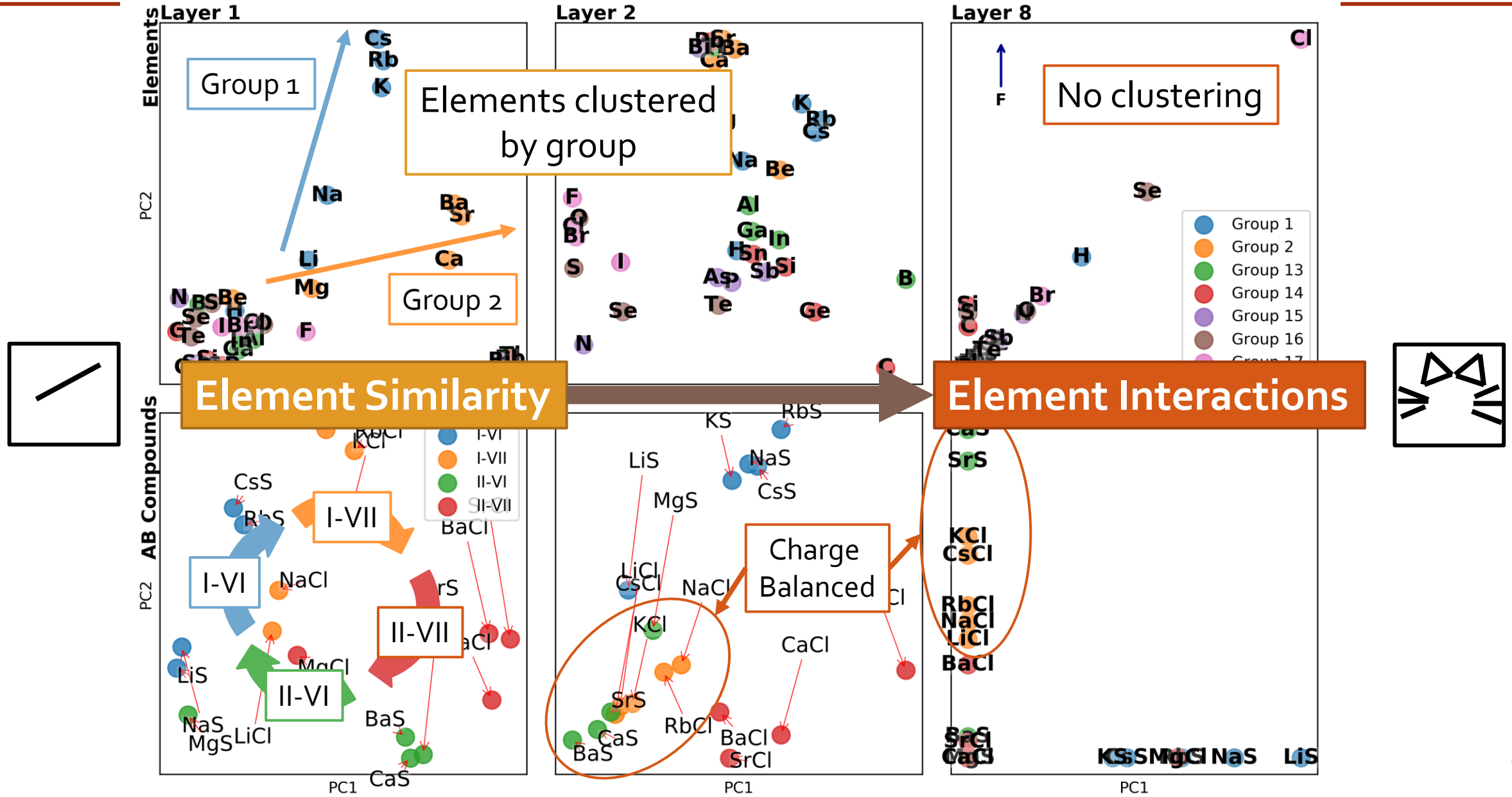
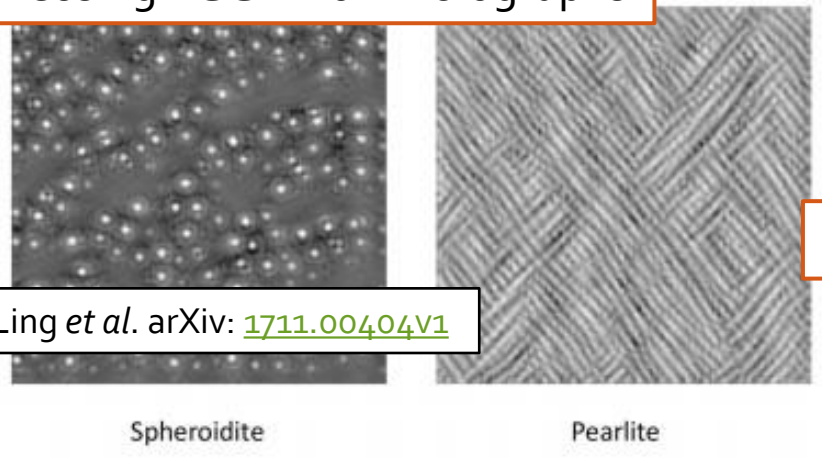


Big Opportunity: Transfer Learning

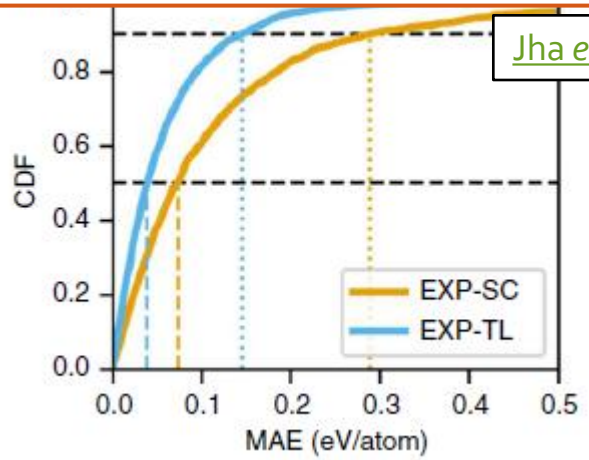
Features learned on one dataset applicable to others

Reusing VGG16 for micrographs



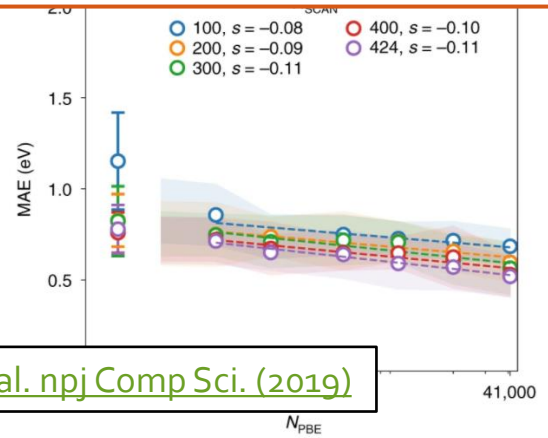
Ling *et al*. arXiv: 1711.00404v1

Reusing ElemNet for experimental data



Jha *et al*. Nat Comm. (2020)

Learning from multiple fidelities



Chen et al. npj Comp Sci. (2019)

Deep Learning is not Cure-All

Table 3: Summary of performances(test subset): conventional methods versus graph-based methods. Graph-based models outperform conventional methods on 11/17 datasets.

Category	Dataset	Metric	Best performances - conventional methods	Best performances - graph-based methods
Quantum Mechanics	QM7	MAE	KRR(CM): 10.22	**DTNN: 8.75**
	QM7b	MAE	KRR(CM): 1.05	**DTNN: 1.77***
			Multitask: 0.0150	**MPNN: 0.0143**
			Multitask(CM): 4.35	**DTNN: 2.35**
		E	XGBoost: 0.99	**MPNN: 0.58**
		E	XGBoost: 1.74	**MPNN: 1.15**
		E	XGBoost: 0.799	**GC: 0.655**
		PRC	Logreg: 0.129	**GC: 0.136**
Biophysics	MUV	AUC-PRC	**Multitask: 0.184**	Weave: 0.109
	HIV	AUC-ROC	**KernelSVM: 0.792**	GC: 0.763
	BACE	AUC-ROC	**RF: 0.867**	Weave: 0.806
	PDBbind(full)	RMSE	**RF(grid): 1.25**	GC: 1.44
Physiology	BBBP	AUC-ROC	**KernelSVM: 0.729**	GC: 0.690
	Tox21	AUC-ROC	KernelSVM: 0.822	**GC: 0.829**
	ToxCast	AUC-ROC	Multitask: 0.702	**Weave: 0.742**
	SIDER	AUC-ROC	**RF: 0.684**	GC: 0.638
	ClinTox	AUC-ROC	Bypass: 0.827	**Weave: 0.832**

* As discussed in section 4.4, DTNN outperforms KRR(CM) on 14/16 tasks in QM7b while the mean-MAE is skewed due to different magnitudes of labels.

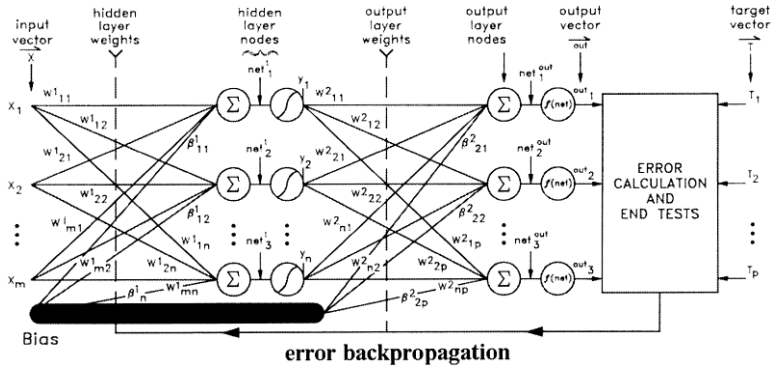
For benchmark problems of learning from molecular data, conventional ML better for **6/17** cases

Ref: Wu *et al.* Chem Sci. (2017) 10.1039/C7SC02664A

MANY APPROACHES FOR DEEP LEARNING WITH MOLECULAR DATA

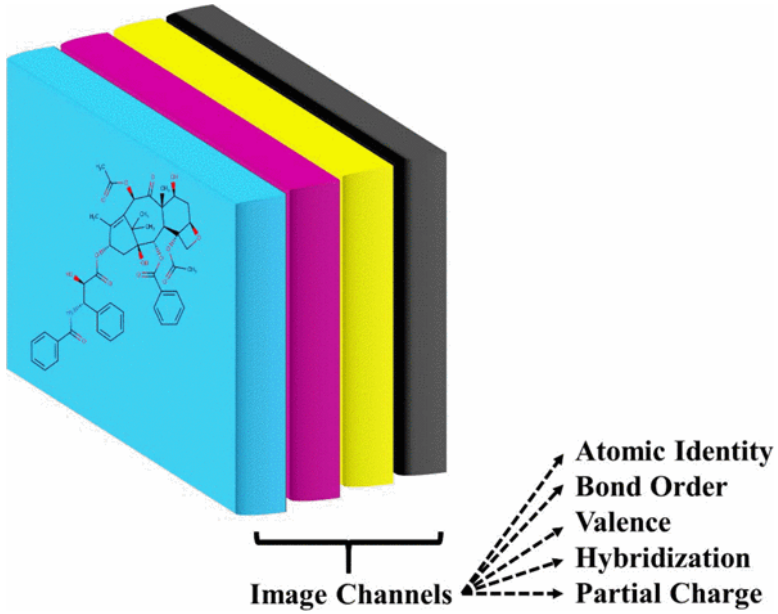
How do we do deep learning with molecules?

As a normal supervised learner



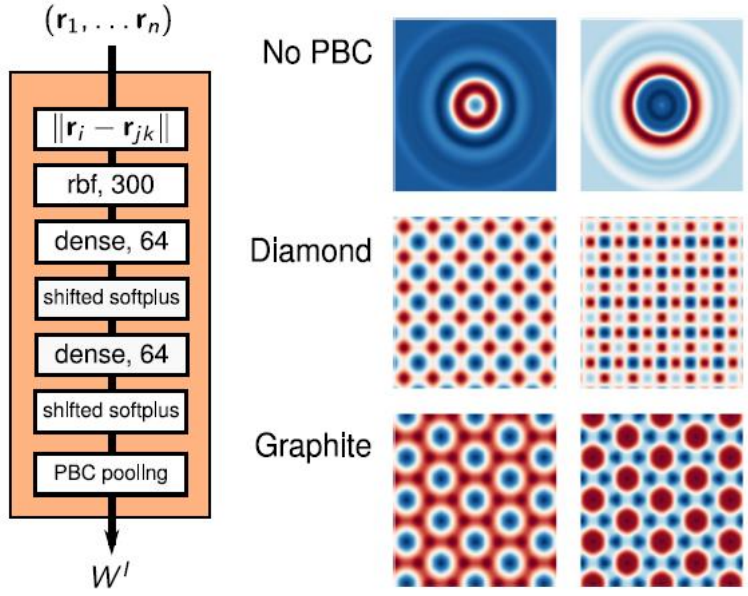
Ex: Ulmer et al. CTPS. (1998), 311.

Repurposing Image Models



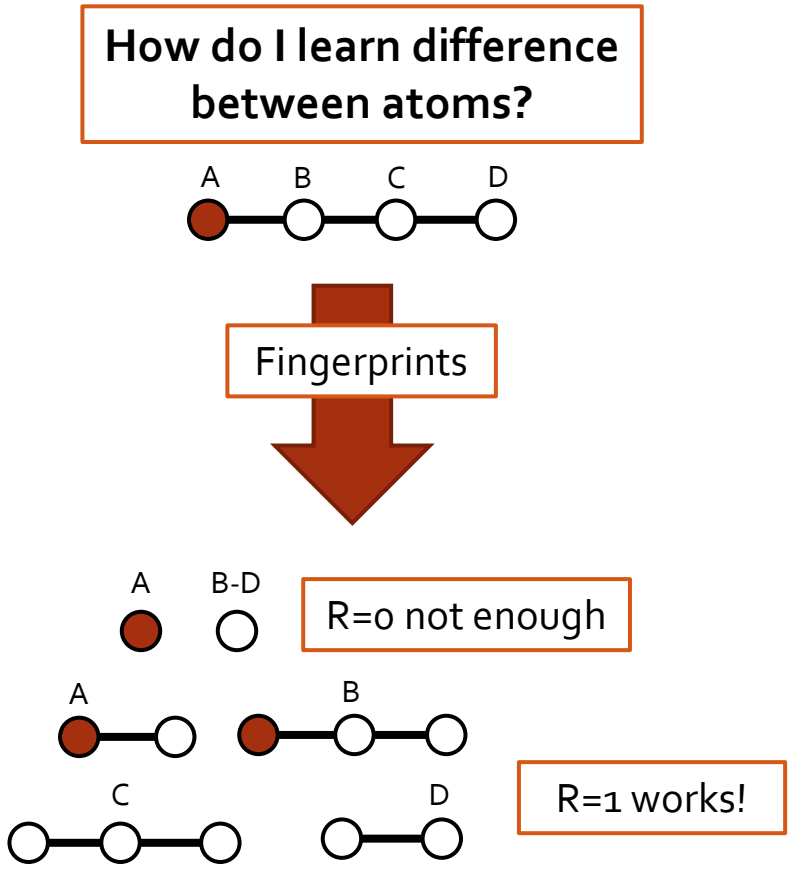
Ex: Goh et al. IEEE WACV. (2018)

Message-Passing Networks



Ex: Schütt et al. JCP, (2018)

Our focus: Message-Passing Networks

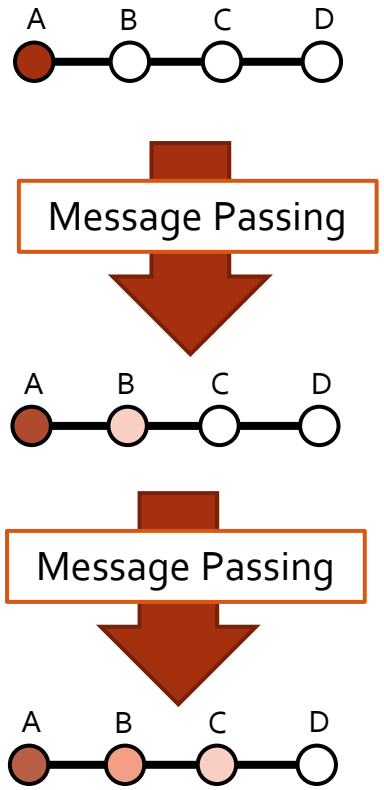


Problems:

1. Combinatorics: Many features!
2. No similarity between groups (is A:B > B:C)

Our focus: Message-Passing Networks

How do I learn difference between atoms?



Atomic fingerprints are learned by "convolution"

Message Passing is Two Steps

Step 1: Message generation

$$m(B) = f_M(\text{B}, \text{A}, \text{—}) + f_M(\text{B}, \text{C}, \text{—}) =$$

Step 2: Update

$$B^{\{n+1\}} = f_U(\text{B}, \text{m}) =$$

Solution: Message passing gives continuous features

~~1. Combinatorics: Many features!~~ Fixed feature count

~~2. No similarity between groups (is A:B > B:C)~~ Distances

New Challenge: Learning update and message functions

Formalizing Message Passing Neural Networks

A generalized form of neural networks for graph data, introduced by Gilmer et al. (Google)

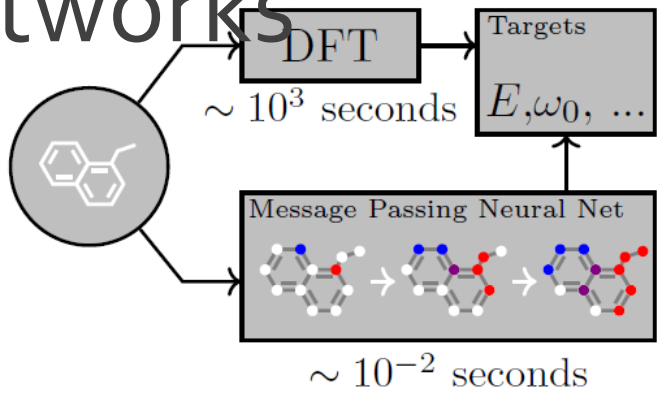


Figure 1. A Message Passing Neural Network predicts quantum properties of an organic molecule by modeling a computationally expensive DFT calculation.

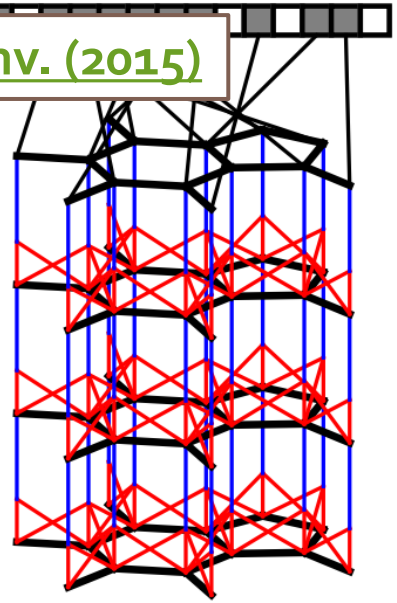
Existing strategies mostly variants of

- $m_v^{t+1} = \sum_{w \in N(v)} \boldsymbol{M_t}(h_v^t, h_w^t, e_{vw})$ 1. **Gather** messages from neighboring nodes
- $h_v^{t+1} = \boldsymbol{U_t}(h_v^t, m_v^{t+1})$ 2. **Update** node state given messages
- $\hat{y} = \boldsymbol{R}(\{h_v^T | v \in G\})$ 3. **Readout** graph properties given node states

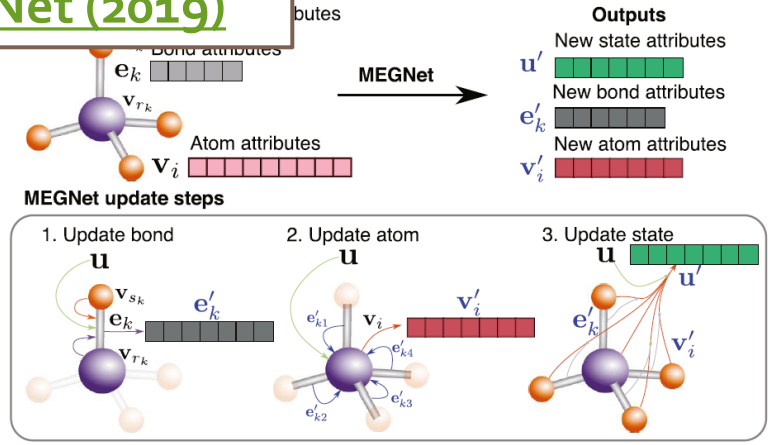
Ref: Gilmer et al. (2017). arXiv:1704.01212v2

Many variations of MPNNs

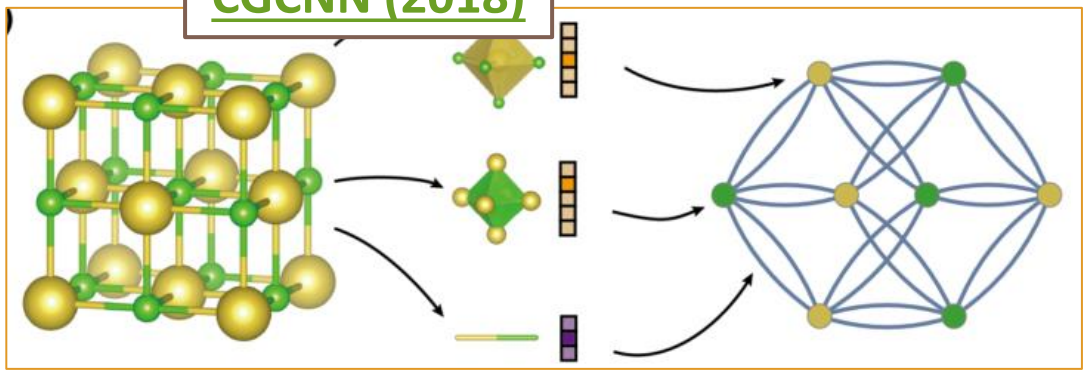
Graph Conv. (2015)



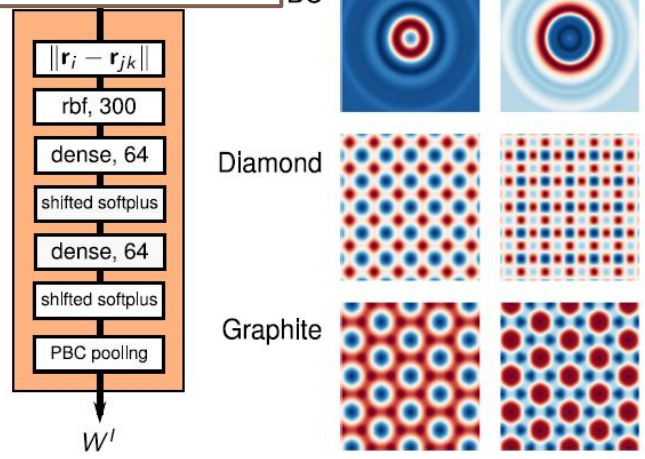
MEGNet (2019)



CGCNN (2018)

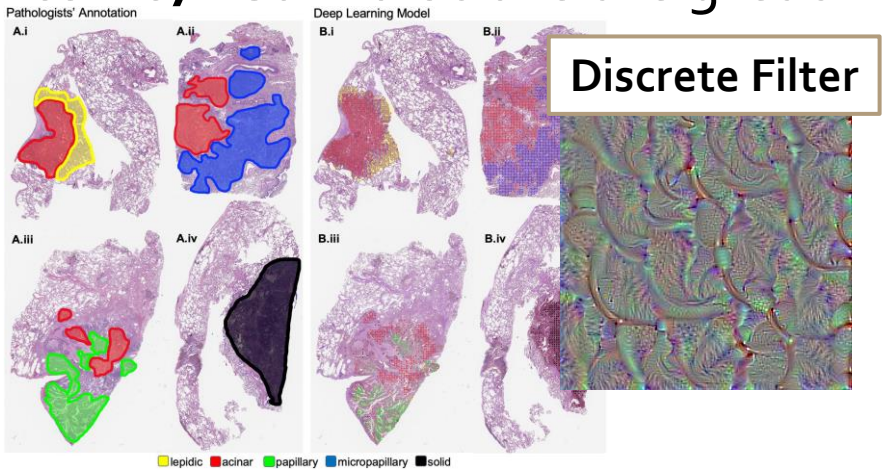


SchNet (2018)



SchNet: Continuous Convolutions

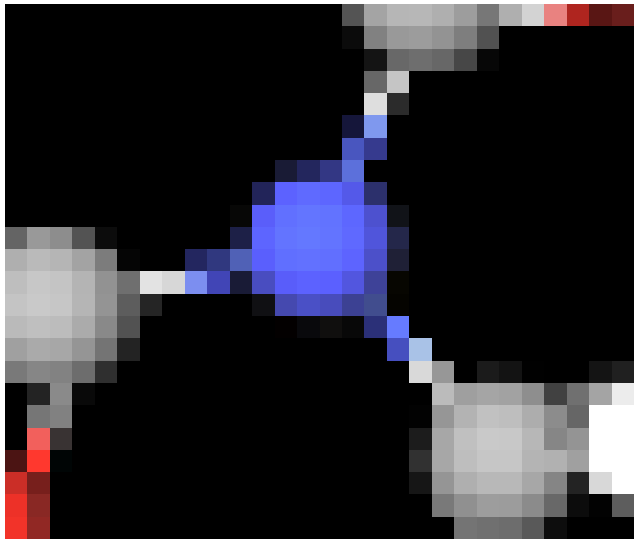
Opportunity: Convolutions are great!



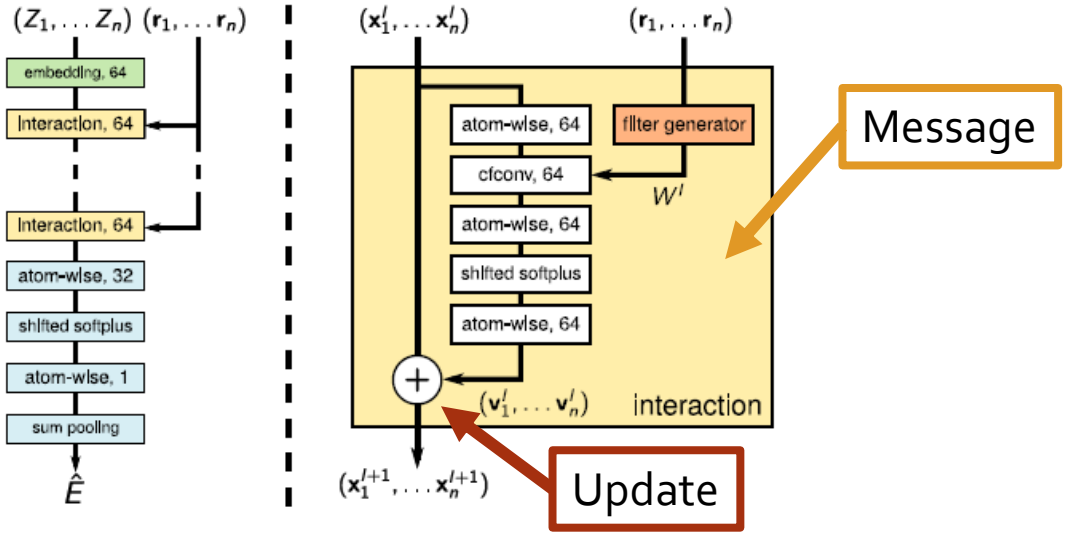
Wei *et al.* Sci Rep. (2019), 3358

Discrete Filter

Problem: Atoms are not pixel

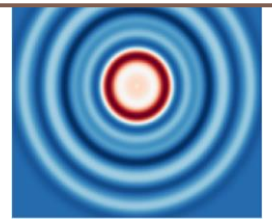


Key Innovation: "Continuous Convolutions"



$$\text{cfconv}(x_i) = \sum_{j}^{n_{atoms}} x_j \circ W(r_j - r_i)$$

Continuous Filter



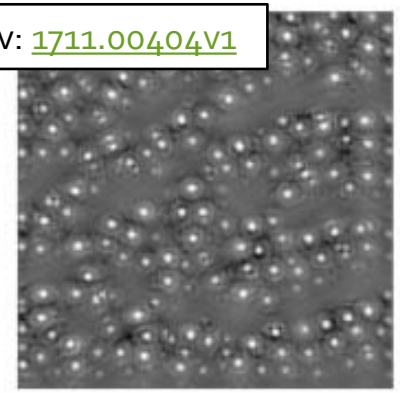
Ref: Schütt et al. JCP, (2018)

Take-home Points

Deep Learning has many advantages…

- Learning from sparse features
- High model complexity -> High accuracy
- Transfer learning

Ling *et al.* arXiv: 1711.00404v1

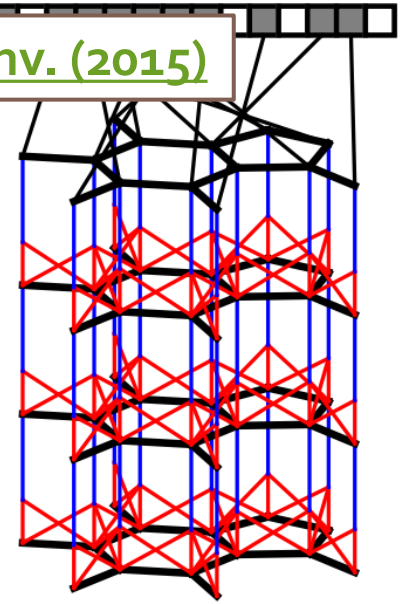


Spheroidite

Major method for molecules: Message Passing Networks

- Automatically learning fingerprints
- Combination of "message," "update" and "readout"
- Many variations in the literature

Graph Conv. (2015)



Practical Exercises: Learn how to implement these in TensorFlow