Recent Advances in the PANNA code

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Credits:

Emine Kucukbenli, Boston U (MA, USA)

Ruggero Lot, SISSA Trieste (I)

Franco Pellegrini, SISSA Trieste (I)

Yusuf Shaidu, Berkeley (CA, USA)

Mandana Safari, SISSA Trieste (I)

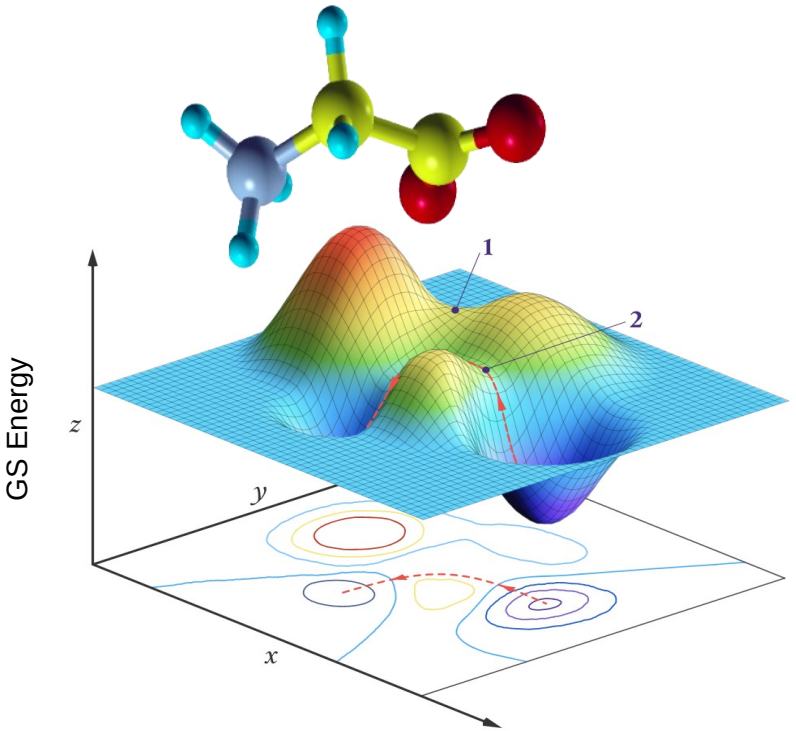
PANNA

Properties from Artificial Neural Network Architectures

https://gitlab.com/pannadevs/panna

Atomistic modelling of materials

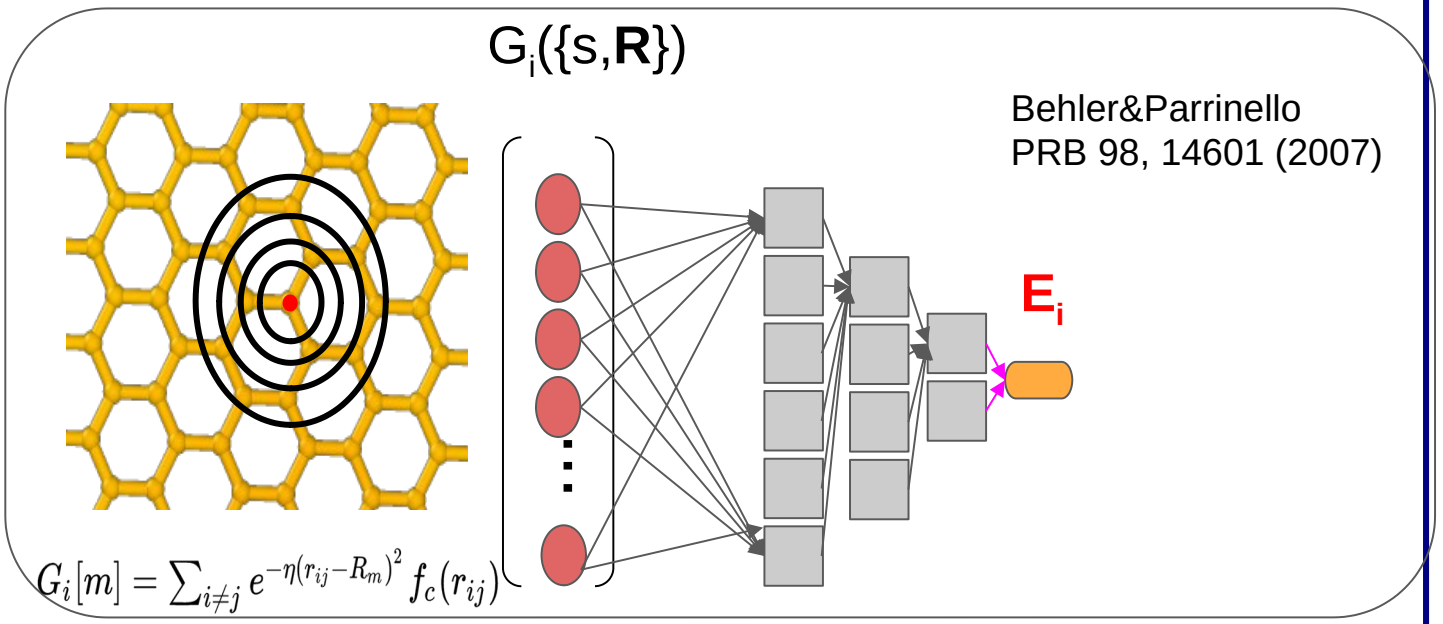
$E(\{\mathbf{R}\})$



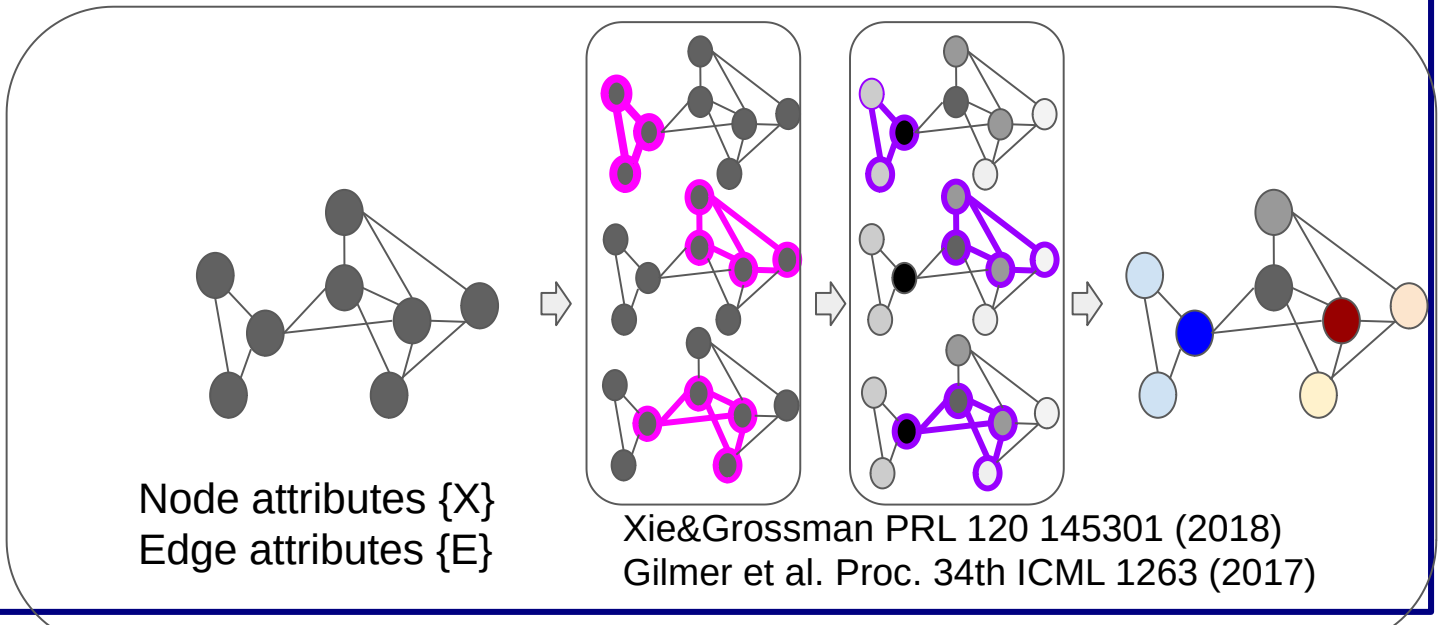
- Equilibrium geometry
- Energy differences, e.g. binding energy
- Forces, MD

Approximating Energy via NNs

Fully Connected
A2A NN



Graph NN



Replace the expensive DFT total energy calculations (or other accurate methods) with an interatomic potentials built to reproduce DFT data in a variety of environments

$$E(c) = \sum_{\alpha} \sum_{i \in \alpha} \varepsilon_{\alpha}(\mathbf{d}_i) + \text{long range contrib}$$

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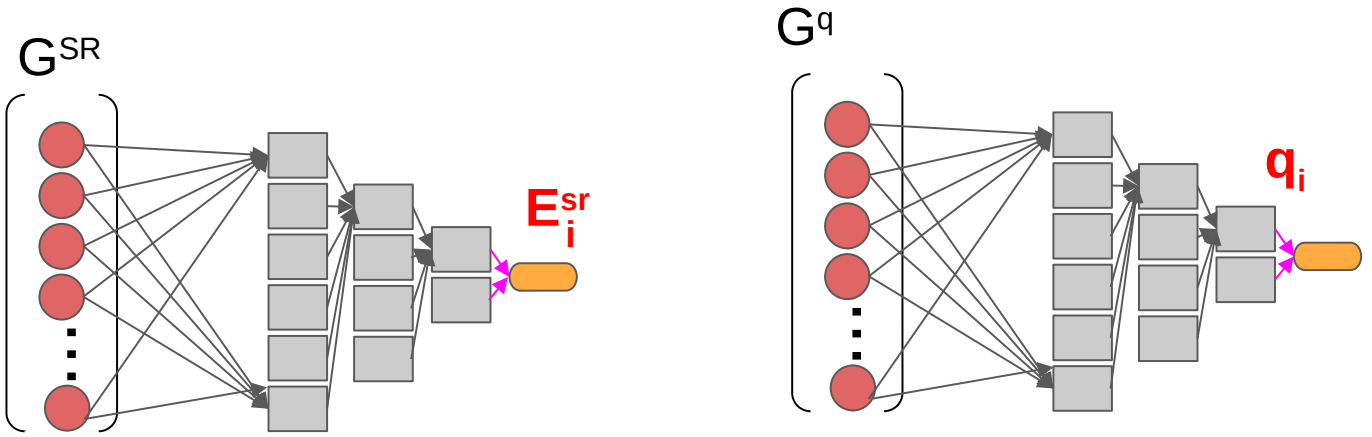
$$E(c) = \sum_{\alpha} \sum_{i \in \alpha} \varepsilon_{\alpha}(\mathbf{d}_i) + \text{ long range contrib}$$

Learning charges

$$E = \sum_i E_i^{sr} + \sum_{i,j} f_s \frac{q_i q_j}{r_{ij}}$$



Train Q-NN on DFT charge density decomposition
Calculate electrostatic contribution and subtarct from DFT energy
Train the SR-NN

Artrith, N., Morawietz, T. & Beheler J. (2011) PBR, 83(15)
Handley, C.M., & Popelier, P.L.A. (2009) J Chem Theory and Computation, 5(6),1474-1489.

✓ **Flexible functional form** for the non-ES part

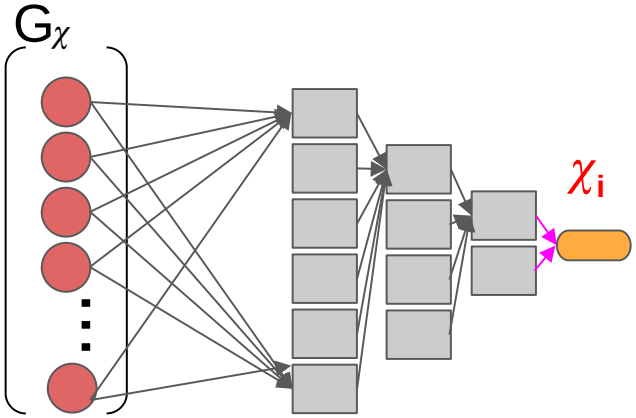
✗ **No non-local charge transfer**: Q locally determined

Learning charge equilibration

$$E(\{q_i\}) = \sum_{i=1}^{N} \left(E_i^0 + \chi_i q_i + \frac{1}{2} J_{ii} q_i^2 \right)$$

$$+ \frac{1}{2} \iint \frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} \, d\mathbf{r} \, d\mathbf{r}',$$

- Ei energy of the isolated atom i
- χ_i : electronegativity of atom i, depends on atomic env.
- J_{ii}: atomic hardness, depends on element type only.
- Charge density is sum of Gaussians normalized to q_i for each atom i, with a spread of α_i



$$\frac{\partial E}{\partial q_i} = 0; \quad \forall\, i = 1, \ldots, N$$

$$\left(\begin{array}{ccc|c} & A_{i,j} & & \begin{matrix}1\\ \vdots \\ 1\end{matrix} \\ \hline 1 & \ldots & 1 & 0 \end{array}\right) \begin{pmatrix} q_1 \\ \vdots \\ \frac{q_N}{\lambda} \end{pmatrix} = \begin{pmatrix} -\chi_1 \\ \vdots \\ \frac{-\chi_N}{q_{\text{tot}}} \end{pmatrix}$$

Ghasemi, S. A., Hofstetter, A., Saha, S., & Goedecker, S. (2015) PRB, 92(4).

✗ **Restricted functional form** for the non-ES part

✓ **Non-local charge transfer** can be captured

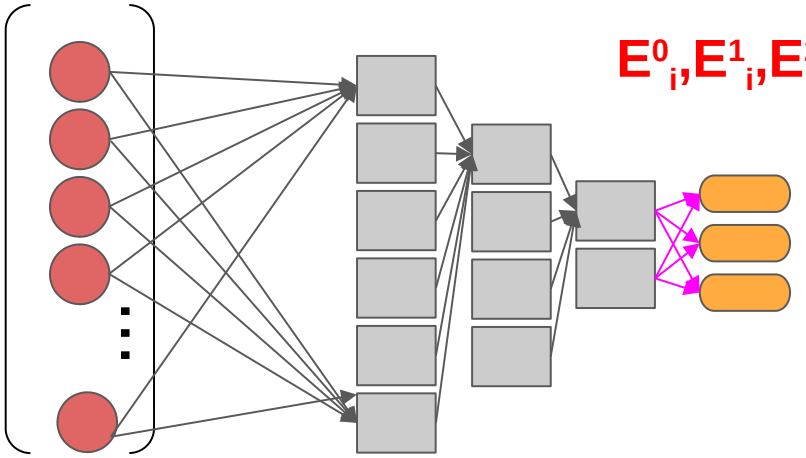
$$E(\{q_i\}) = \sum_{i=1}^{N}\left(E_i^0 + \chi_i q_i + \frac{1}{2}J_{ii}q_i^2\right) + \frac{1}{2}\iint \frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\,d\mathbf{r}\,d\mathbf{r}',$$

$$\Rightarrow \sum_i^N \left(E_i^0 + E_i^1 q_i + \frac{1}{2}E_i^2 q_i^2\right)$$

E^0_i, E^1_i, E^2_i Environment dependent Taylor expansion coefficients

$$\frac{\partial E}{\partial q_i} = 0; \quad \forall\, i = 1, \ldots, N$$

G



$$\left(\begin{array}{ccc|c} & A_{i,j} & & \begin{matrix}1\\ \vdots \\ 1\end{matrix} \\ \hline 1 & \ldots & 1 & 0 \end{array}\right)\left(\begin{matrix} q_1 \\ \vdots \\ q_N \\ \hline \lambda \end{matrix}\right) = \left(\begin{matrix} -E_1^1 \\ \vdots \\ -E_1^N \\ \hline q_{\text{tot}} \end{matrix}\right)$$

Shaidu, Kucukbenli et al.; in preparation.

$$E(\{q_i\}) = \sum_{i=1}^{N}\left(E_i^0 + \chi_i q_i + \frac{1}{2}J_{ii}q_i^2\right) + \frac{1}{2}\iint \frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\,d\mathbf{r}\,d\mathbf{r}',$$

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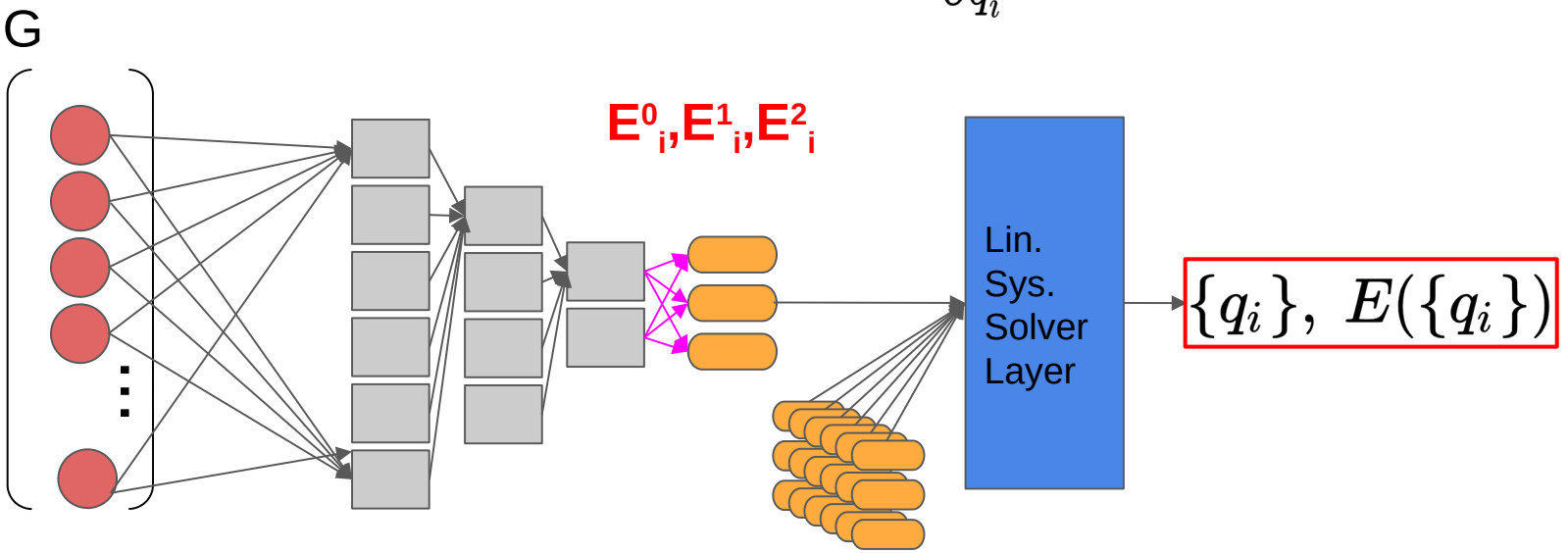


Shaidu, Kucukbenli et al.; in preparation.

✓ **Flexible functional form** for the non-ES part

✓ **Non-local charge transfer** can be captured

Environment-dependent CE at work:

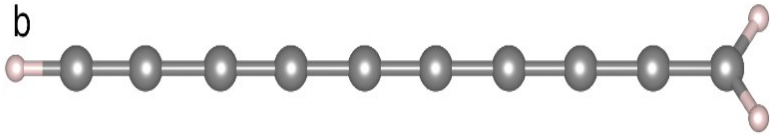
Data from:
Ko, Finkler, Goedecker and Behler *Nat Commun* 12, 398 (2021)

$C_{10}H_2$

a

$(C_{10}H_3)^+$

b



- Same # of electrons, different external potential
- For the left hand side, local descriptor stays the same (cutoff: 4.23A)
- **RMSE**
  Energy: 1.2 meV/atom
  Forces: 73 meV/A

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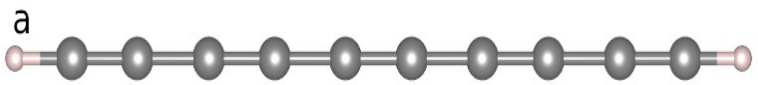
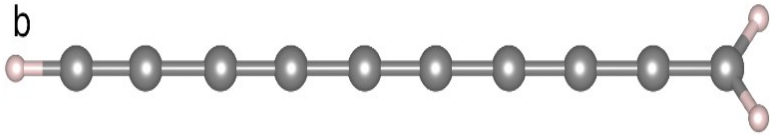


$(C_{10}H_3)^+$

b



- Same # of electrons, different external potential
- For the left hand side, local descriptor stays the same (cutoff: 4.23A)
- **RMSE**
  Energy: 1.2 meV/atom
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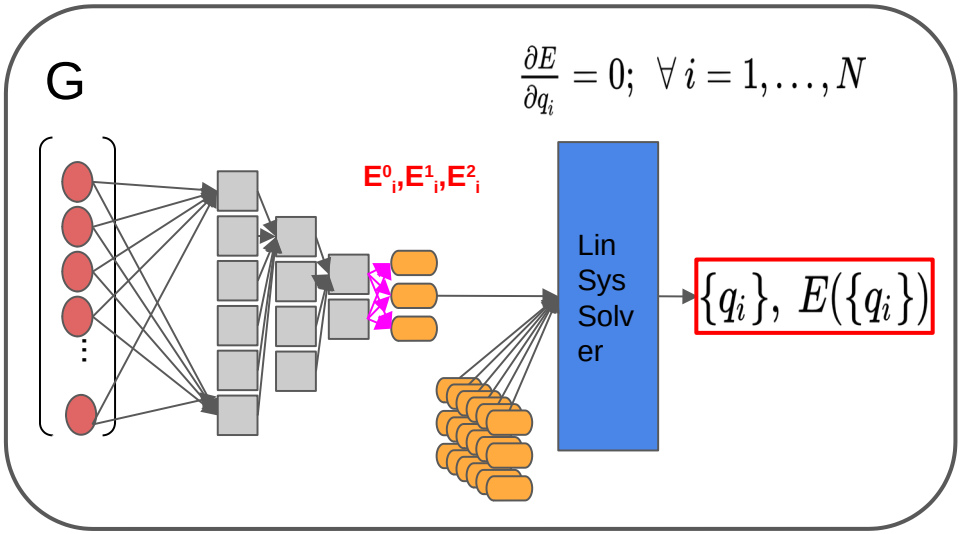
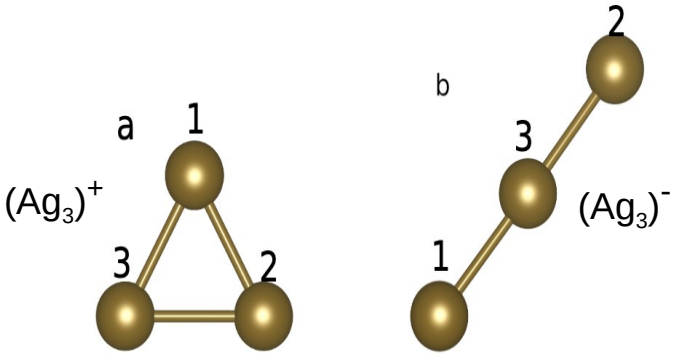
- Using DFT charges in the loss function is optional:



	charge (me)	energy (meV/atom)	force (meV/Å)	γ_q
w DFT charges	9.3	1.2	73	1.0
w\o DFT charges	77.4	1.2	73	0
Ko et al.	6.6	1.2	78	-
No LR treatment	-	1.6	122	-

Environment-dependent CE at work:

Data from:
Ko, Finkler, Goedecker and Behler *Nat Commun* 12, 398 (2021)



- Networks without charge information cannot distinguish the two manifolds. → learning task is not well defined

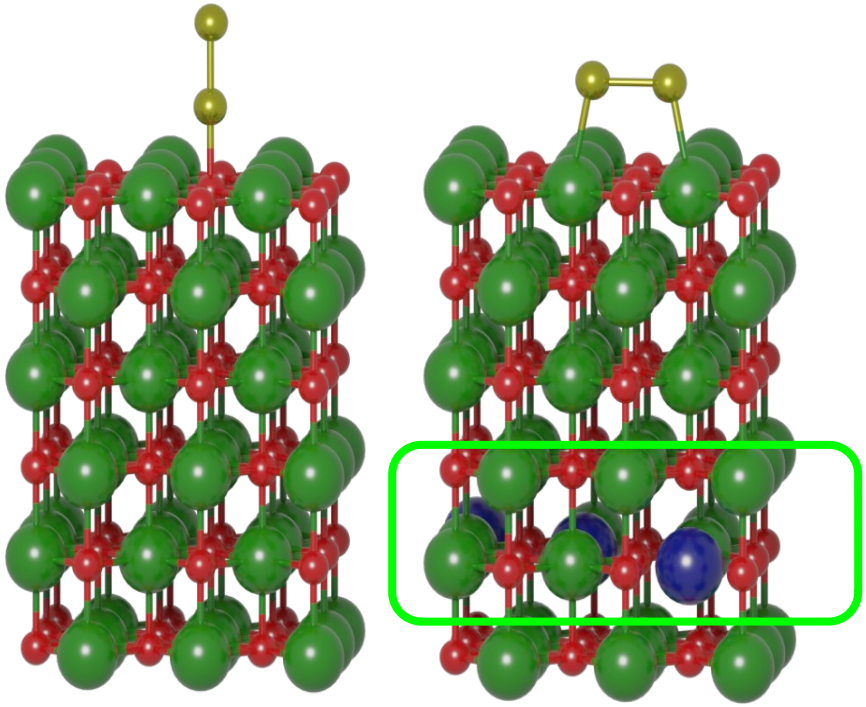
- Different # of electrons, same external potential → two different E(v) maps to learn.
- **RMSE**
  Energy: ~1 meV/atom
  Forces: 24 meV/A

	charge	energy	force	γ_q
	(me)	(meV/atom)	(meV/Å)	
w\o DFT charges	614.3	0.96	24	0.0
w DFT charges	2.1	1.15	24	1.0
Ko et al.	10.0	1.32	32	-
No LR treatment	-	311.40	1390	-

Environment-dependent CE at work:

Data from:
Ko, Finkler, Goedecker and Behler *Nat Commun* 12, 398 (2021)

Equilibrium position of Au_2 on MgO(001):



Al substitution in 5th layer, >10A

RMSE:

	charge (e)	energy (meV/atom)	force (meV/Å)	γ_q
w\o DFT charges	0.50	0.21	44	0.0
w DFT charges	0.07	0.27	68	1.0
Ko et al.	0.01	0.22	66	-
No LR treatment	-	2.31	133	-

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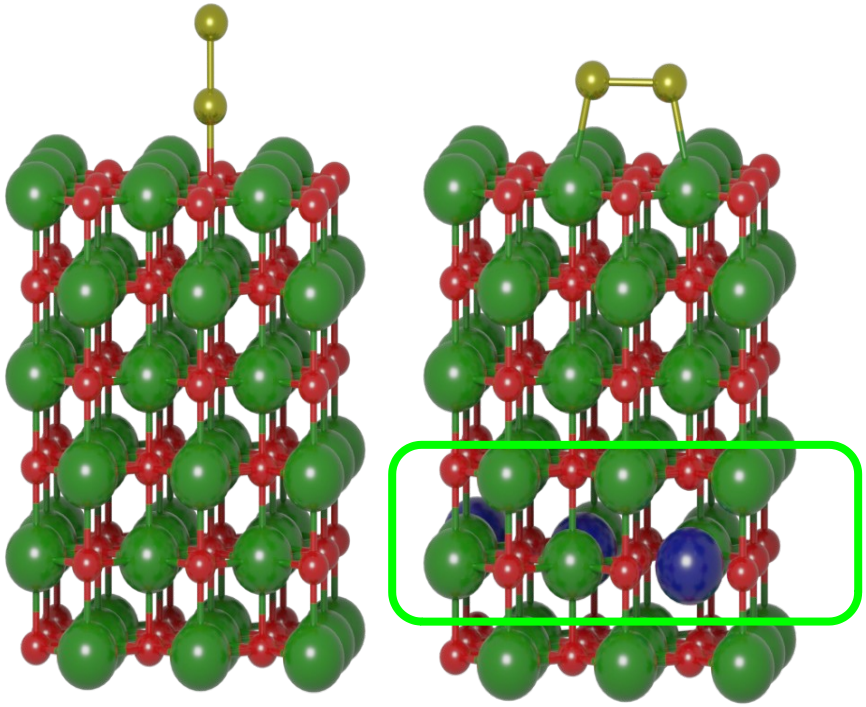


Al substitution in 5th layer, >10A

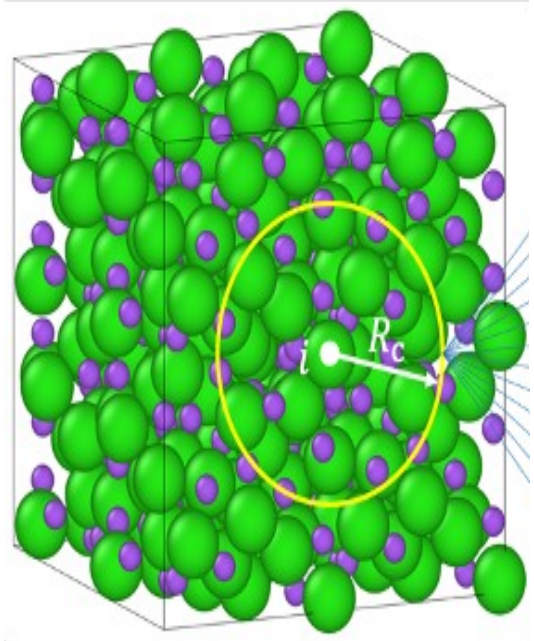
RMSE:

	charge (e)	energy (meV/atom)	force (meV/Å)	γ_q	ΔE (meV)
w\o DFT charges	0.50	0.21	44	0.0	-65
w DFT charges	0.07	0.27	68	1.0	-59
Ko et al.	0.01	0.22	66	-	-41
No LR treatment	-	2.31	133	-	400
DFT					-3

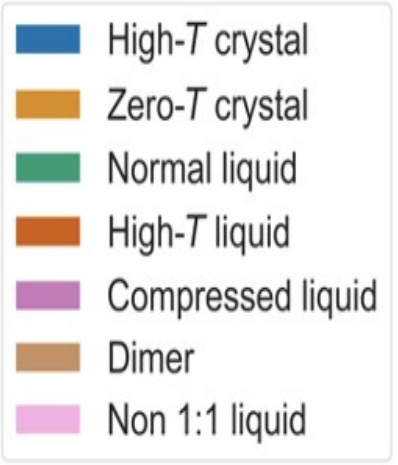
Environment-dependent CE at work

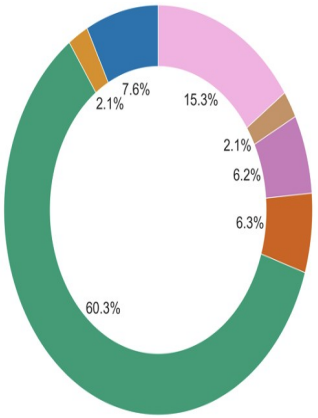
Data from:
Li, Kucukbenli et al. Cell Rep Phys Sci 2, 100359 (2021)

NaCl



$N \sim 112,000$





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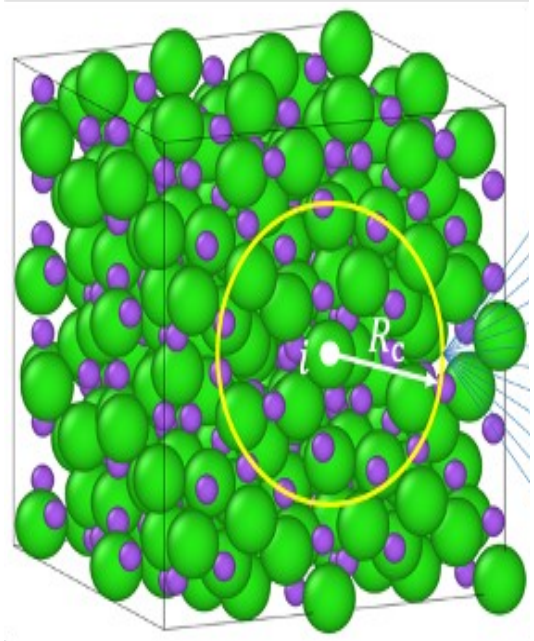

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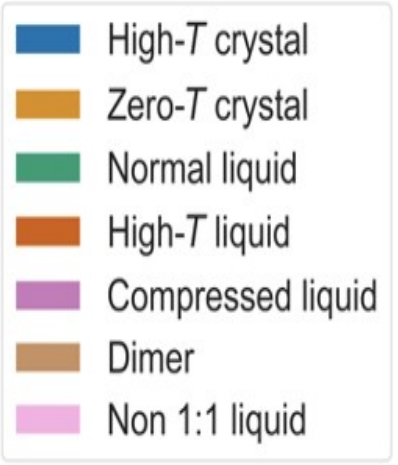
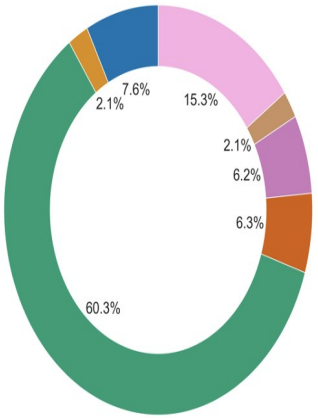




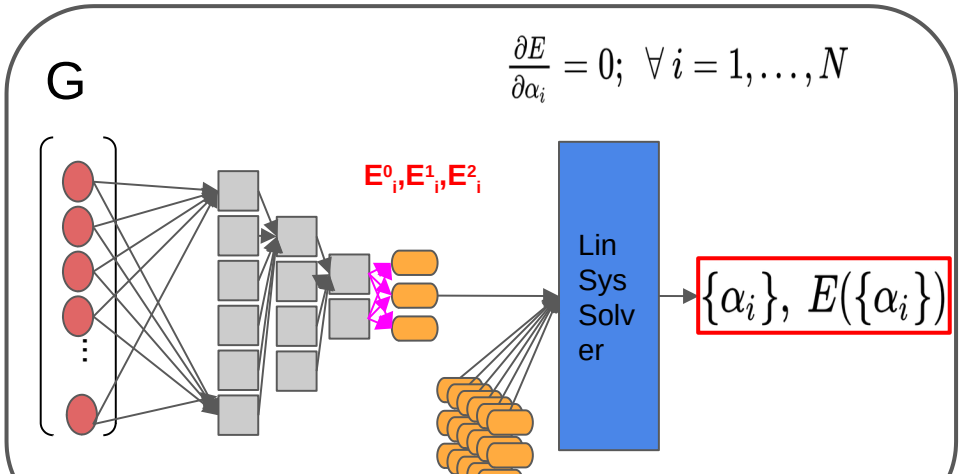
- Wide range of external potential and total electron variation
- **RMSE**
  Energy: ~1 meV/atom
  Forces: 30 meV/A

	energy (meV/atom)	force (meV/Å)	R_c (Å)
No LR treatment	1.35	41	8
Env. dept. CE	0.98	30	5.3

Taylor Expansion Formalism for vdW

$$E(\{\alpha_i\}) = \sum_i^N \left(E_i^0 + E_i^1 \alpha_i + E_i^2 \alpha_i^2\right) - \sum_{ij} \frac{\alpha_i \alpha_j}{r_{ij}^6}$$

- E_i^0 ,E_i^1 ,E_i^2 : Taylor expansion coefficients that are environment dependent.
- α's are proxy parameters for environment dependent polarization of atoms
- We solve the coupled equations again, but this time for polarization variables, and without any constraint on their sum.



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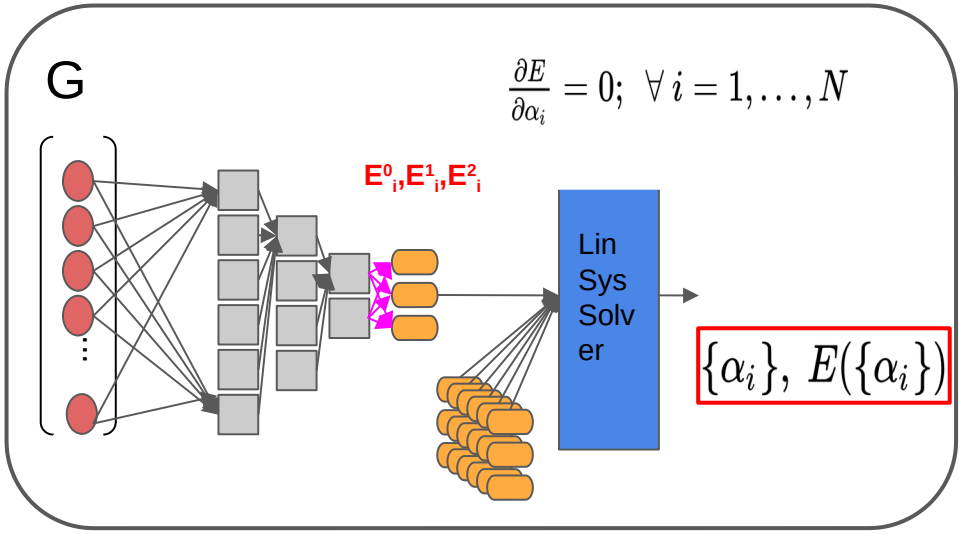
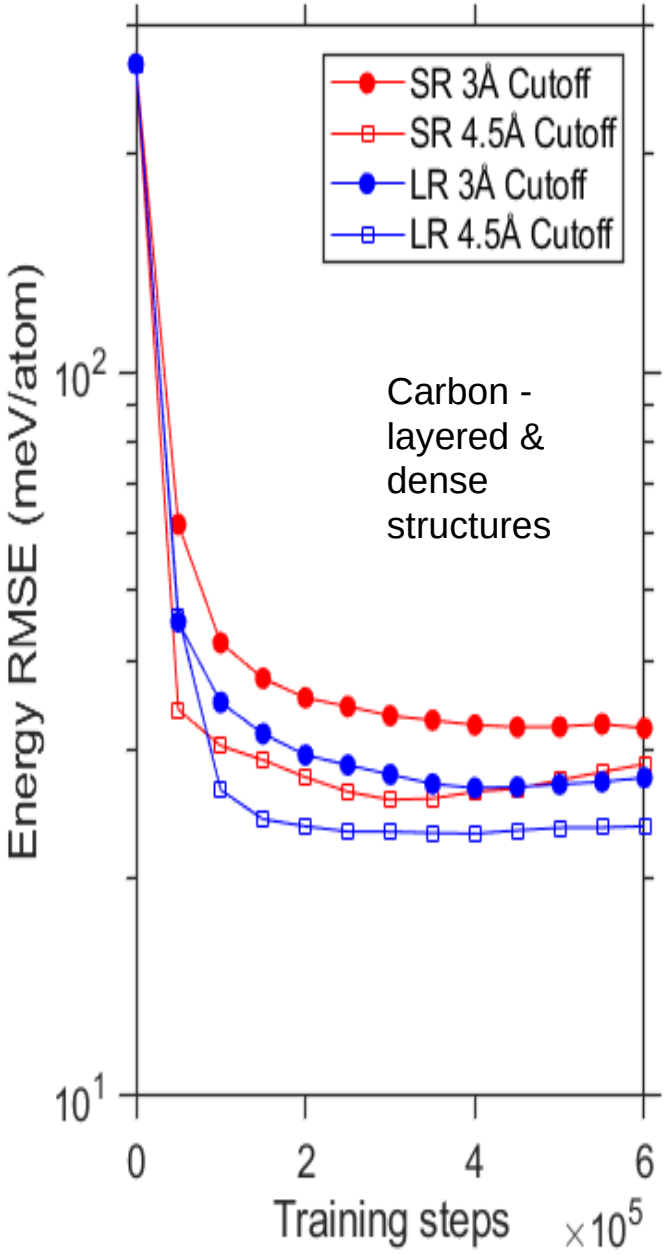
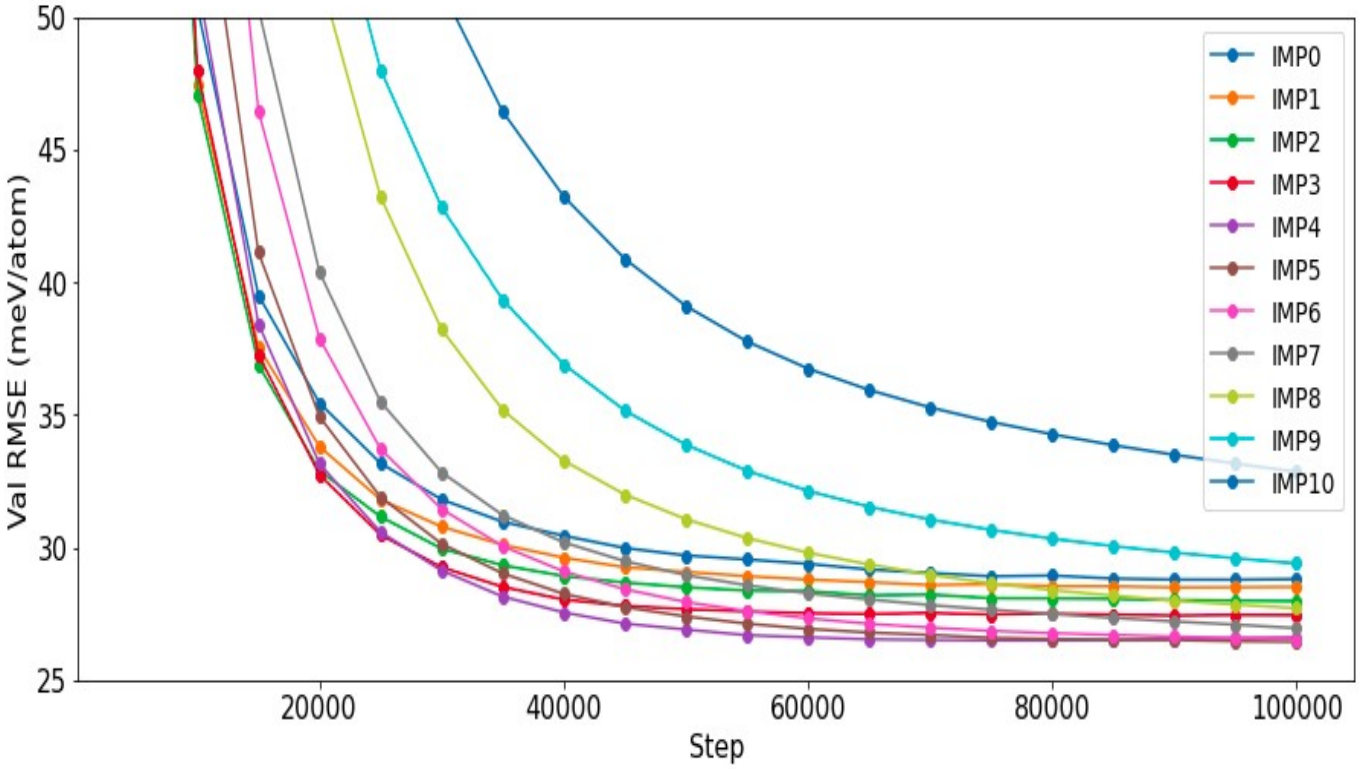


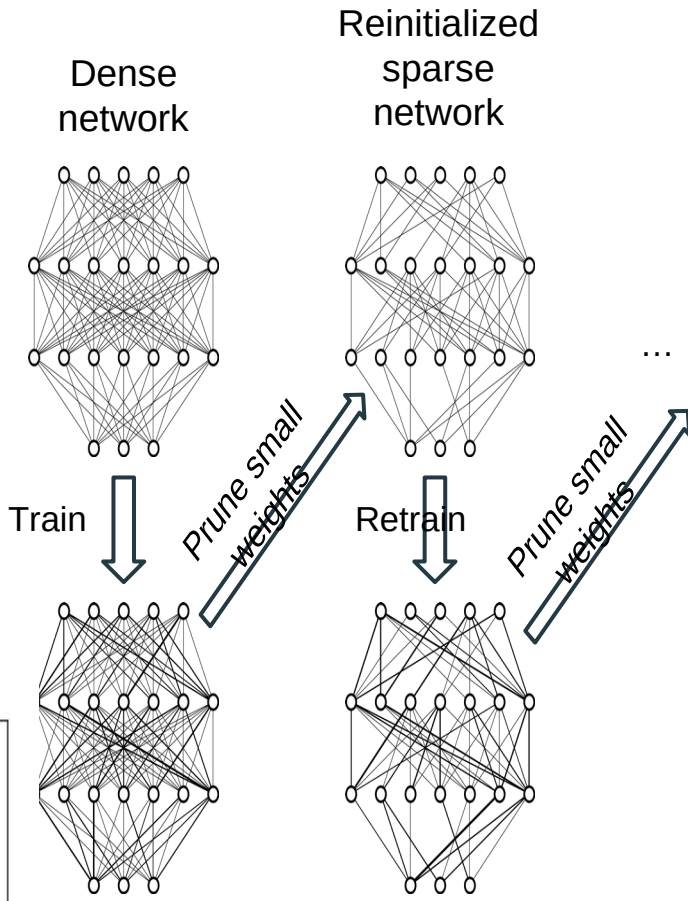


Shaidu, Kucukbenli et al. npj CompuMats 7 (2021)

Iterative Magnitude Pruning

1. Train a NN
2. Remove (prune) X% of the connections with the smallest absolute weights
3. Reset the remaining weights to the init. values and retrain
4. Go to step 2

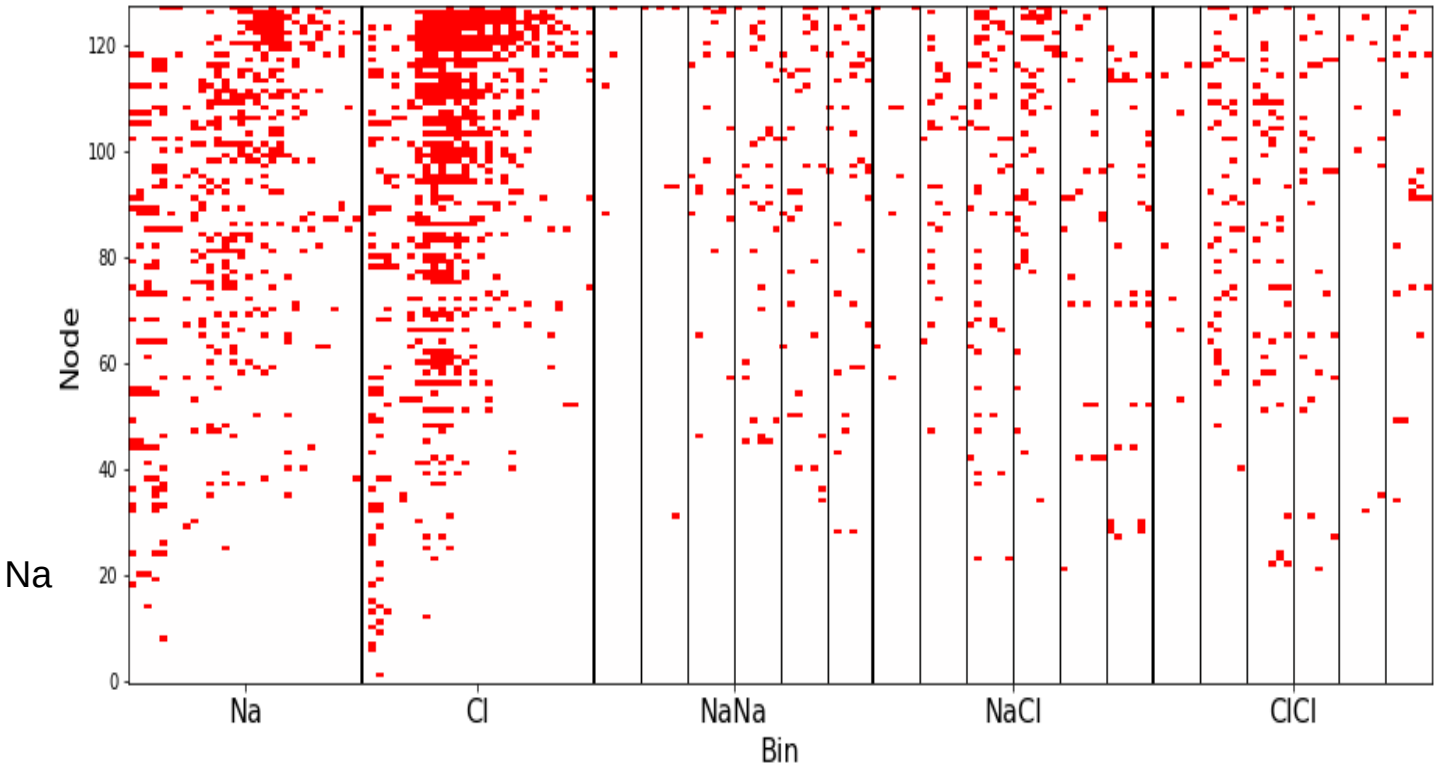


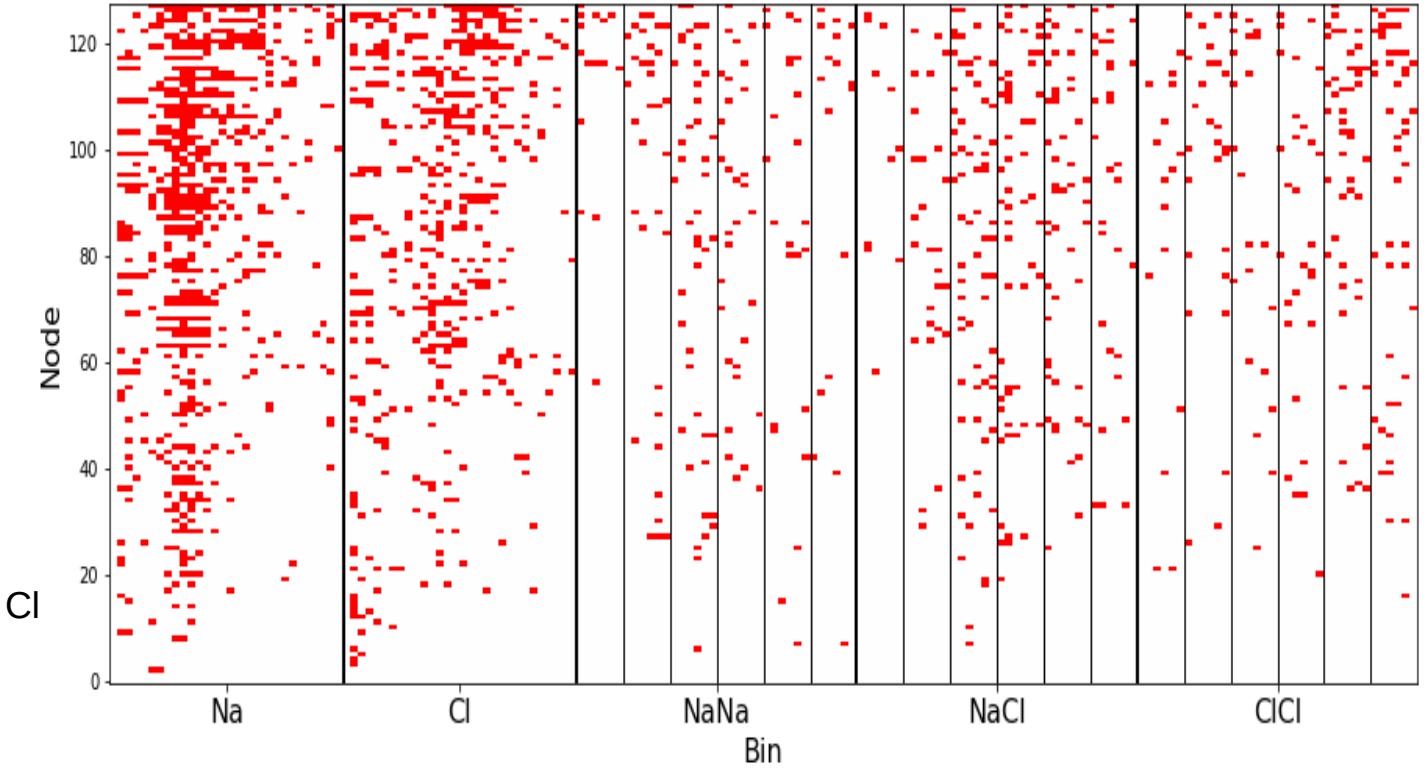


Often reduction to **<10%** size of the original network!

IMP for NaCl:

- Network is heavily informed by the radial part of the atomic environment descriptor.
- Connections to features describing the atomic environment far away is retained, but mostly for one atom.



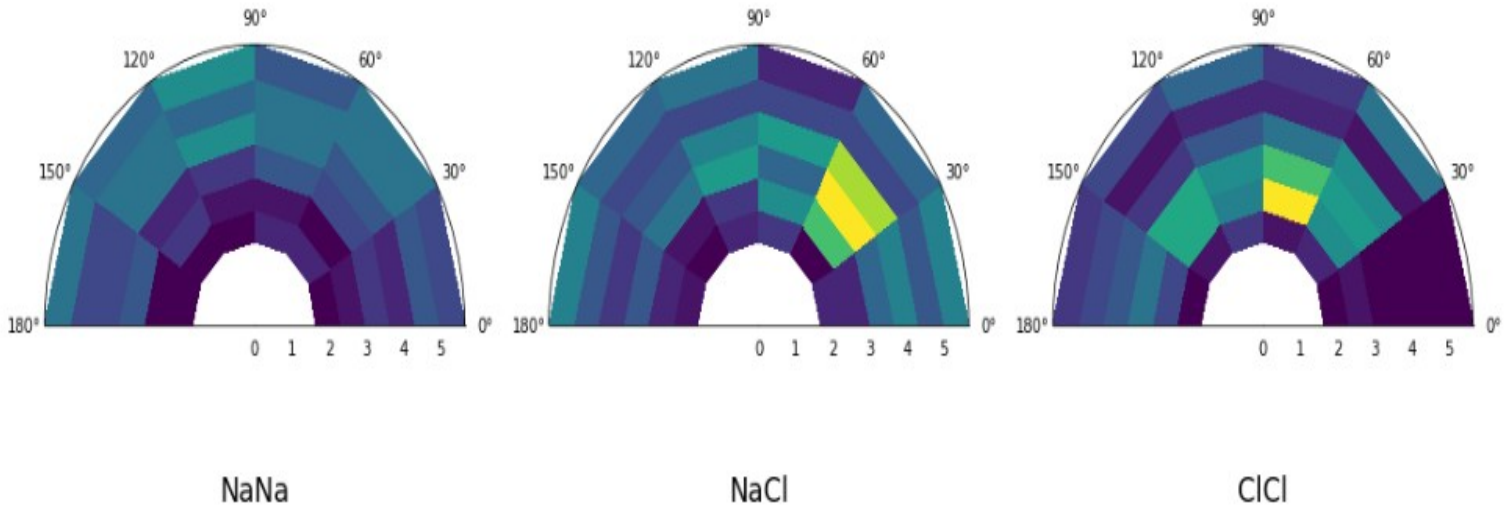


IMP for NaCl - remaining connections:

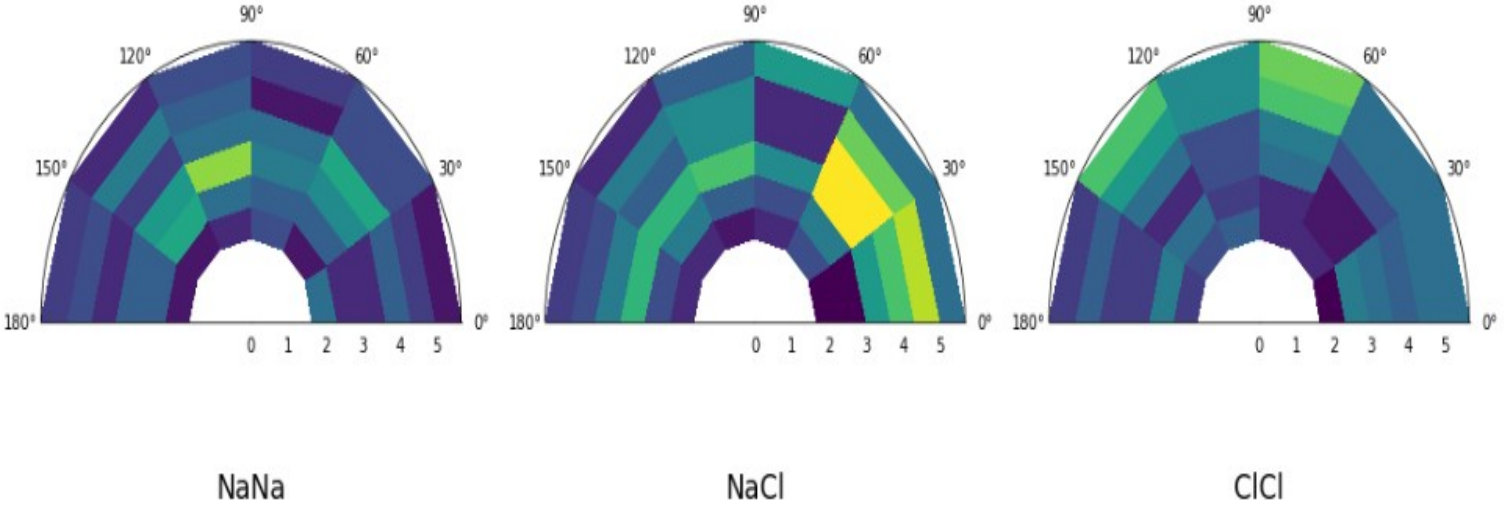
- Average connection remaining for the angular part:
- Significant angles are ~45 for NaNaCl andClClNa

  → corners of the NaCl cube.

Na



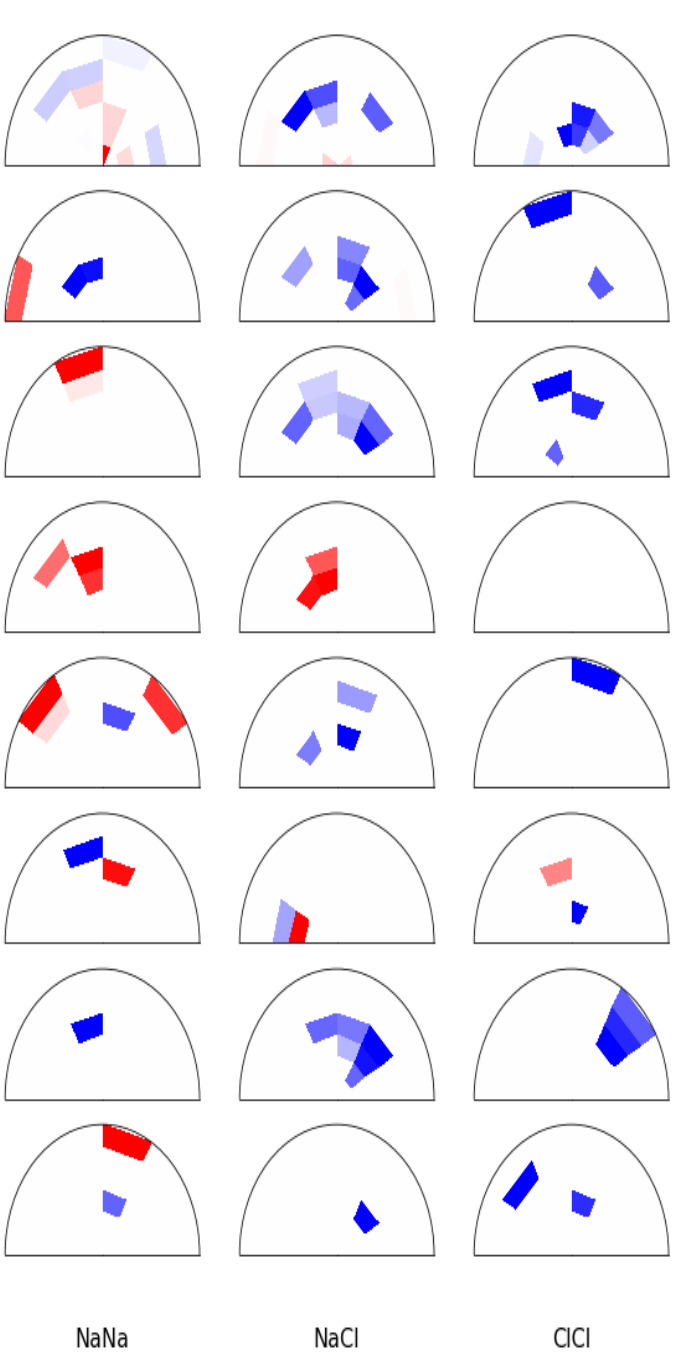
Cl

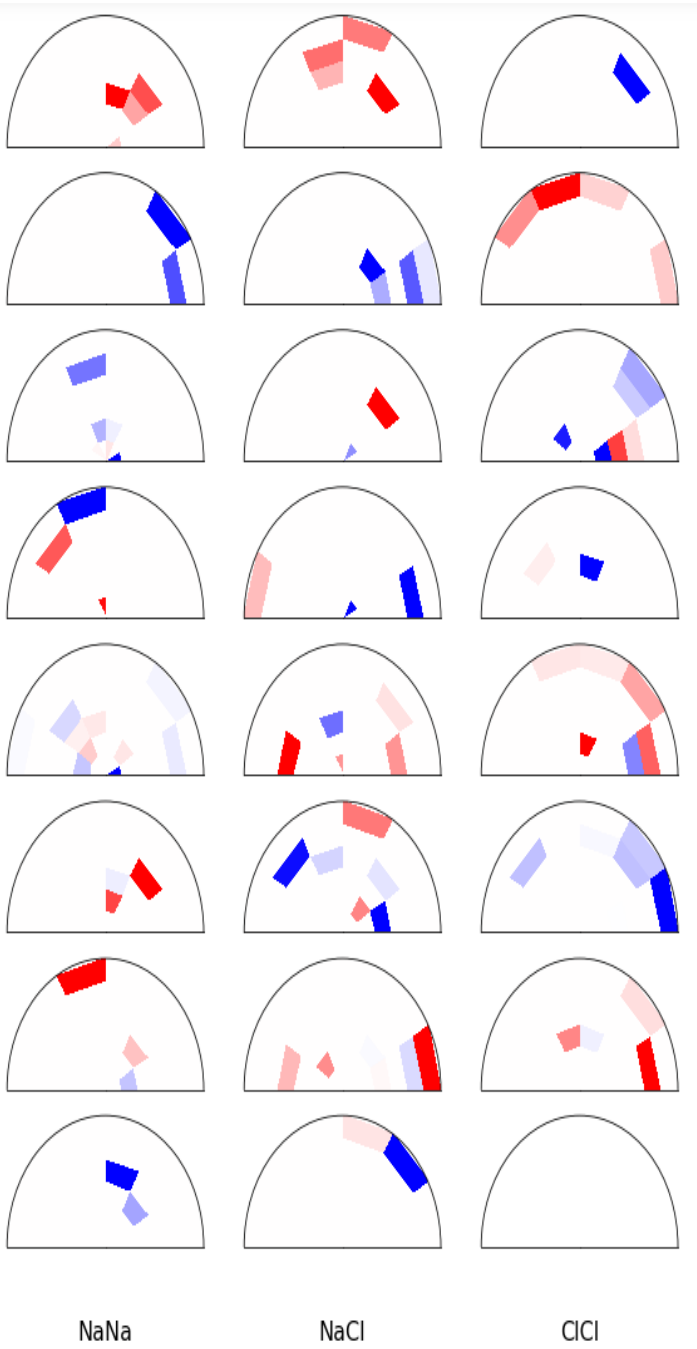


IMP for NaCl

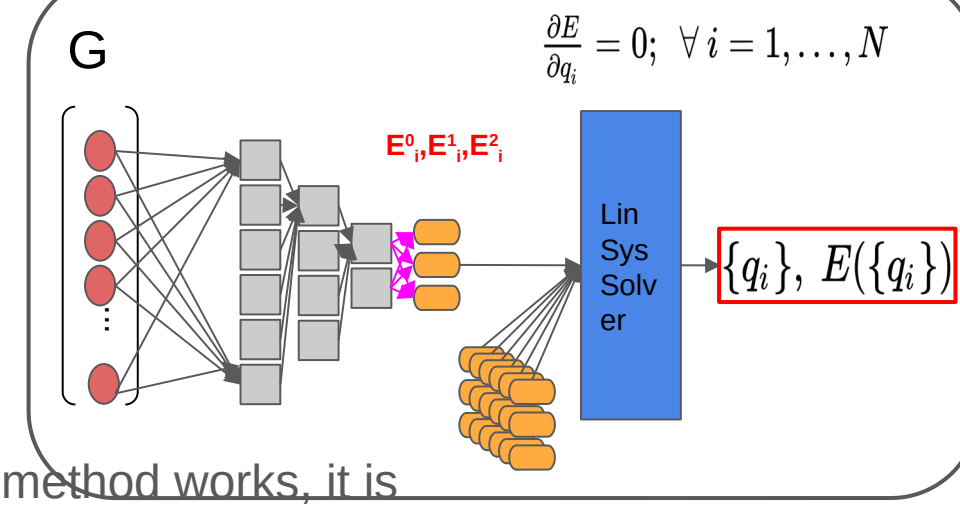
Masks multiplied by weights of single nodes (rows) divided by species (columns), for Na (left) and Cl (right):

- Nodes mostly work with local information in angles
- Long-range angle information up to cutoff is retained.

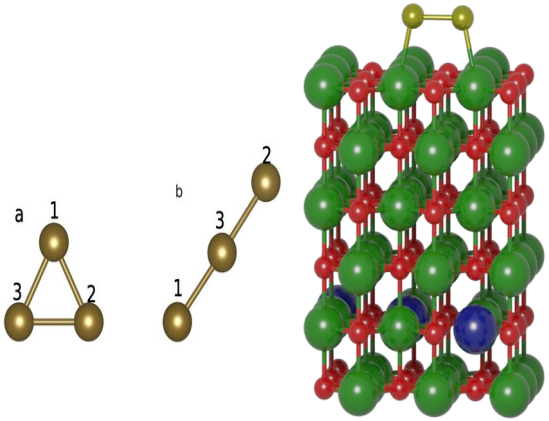




Take away:



- LR interaction via Taylor expansion: Environment-dependent Charge Equilibration method works, it is feasible and is particularly useful for:
  - Non-local charge transfer,
  - Multiple E(v) mapping,
  - Reducing R_{cut} significantly
- Taylor expansion method can be extended to vdW (what else? Can it be used as poor man's "*attention*"?)
- Pruning can reduce the network size & shed light into what's important for the learning task.
- All implemented in **PANNA**: https://gitlab.com/PANNAdevs/panna



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