Practical Introduction to Neural Network Potentials Day 3:

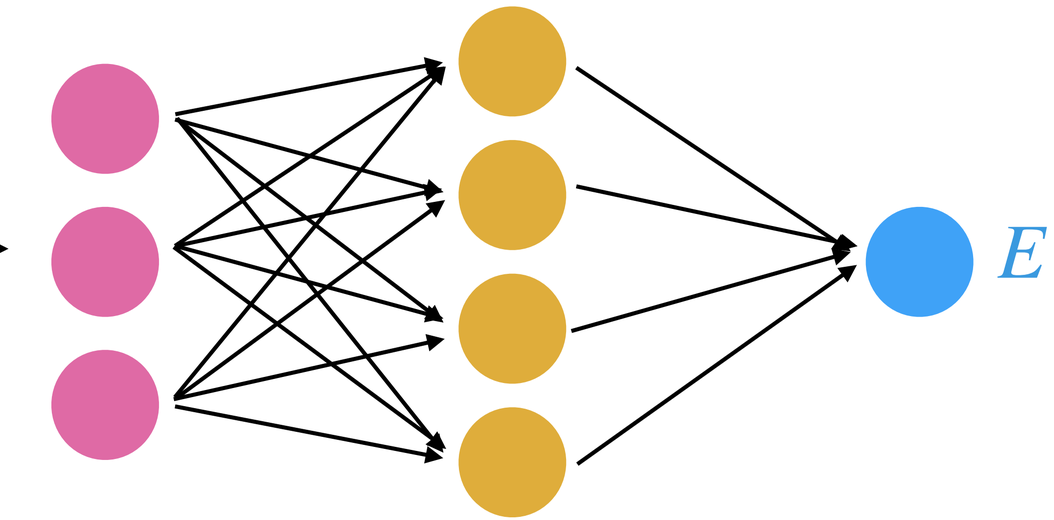
General neural network potentials

Meeting #2 Review: Modeling energies with a neural network

```
5

Cl          0.558980060028    -0.000003409116    -0.000010838753
C          -1.233013253888     0.000007518693     0.000023805641
H          -1.571271543066    -0.697791943059     0.759266356426
H          -1.571271410823     1.006427246976     0.224727628339
H          -1.571303371347    -0.308606540429    -0.983901358501
```

Raw xyz coordinates



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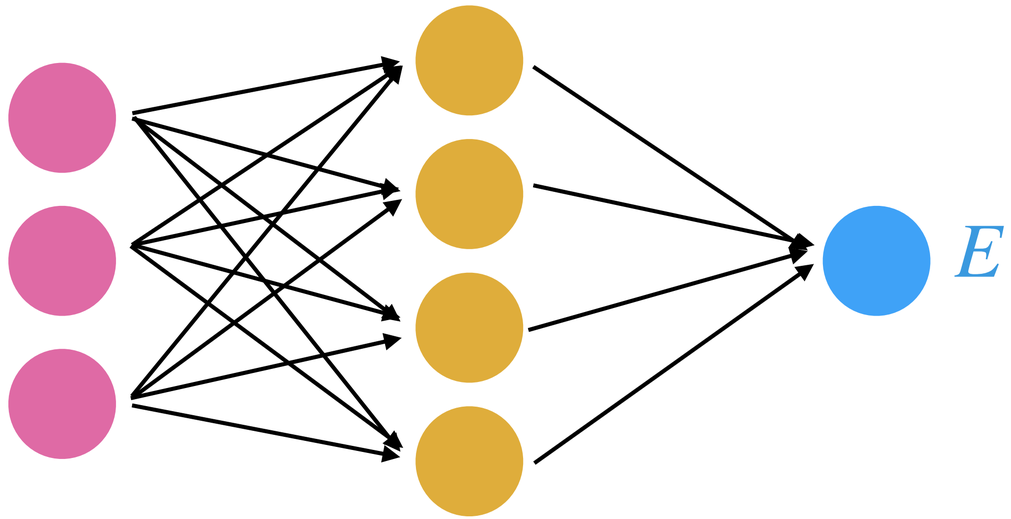

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What limitations does this model have?

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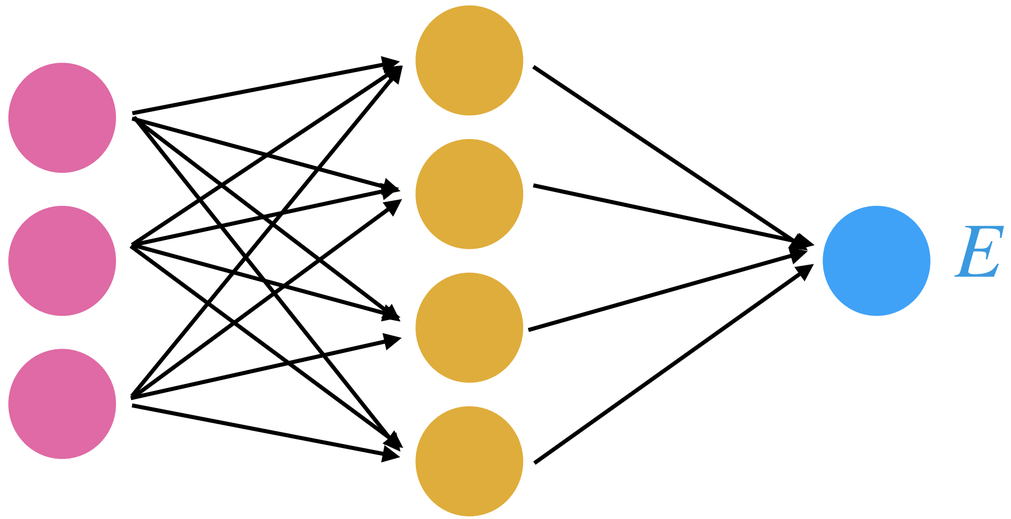

How did this model improve on our first model?

What limitations does this model have?

How could we improve this model?

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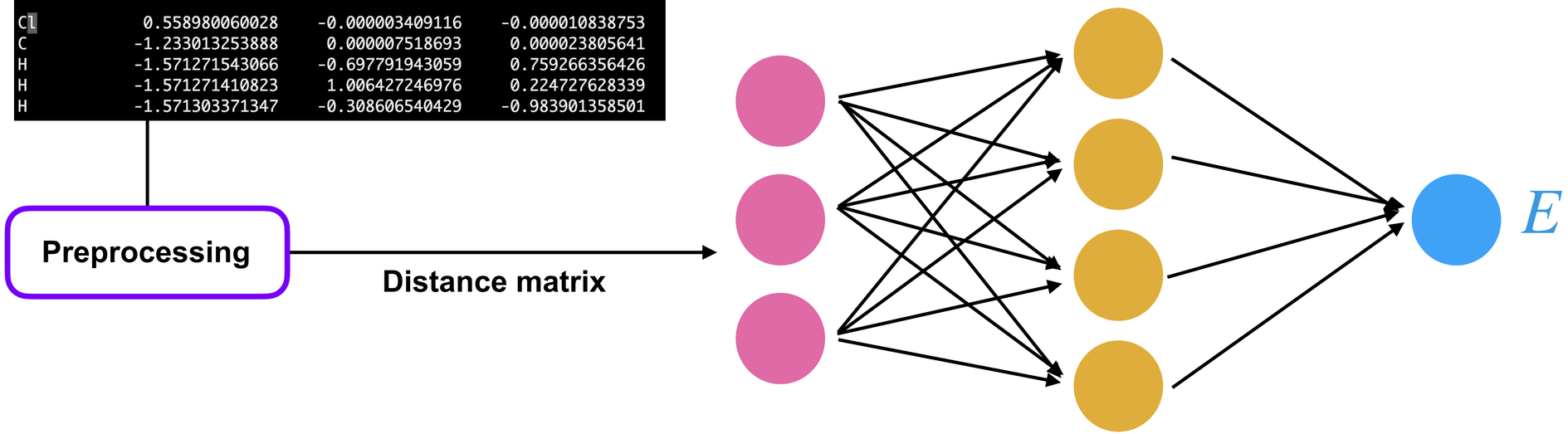

Roughly what was the error in this model?

Literature context for today's session:

Atom-centered symmetry functions for constructing high-dimensional neural network potentials

Cite as: J. Chem. Phys. **134**, 074106 (2011); https://doi.org/10.1063/1.3553717
Submitted: 08 December 2010 • Accepted: 21 January 2011 • Published Online: 16 February 2011

Jörg Behler

Chemical Science

EDGE ARTICLE

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Cite this: *Chem. Sci.*, 2017, **8**, 3192

ANI-1: an extensible neural network potential with DFT accuracy at force field computational cost†

J. S. Smith,[a] O. Isayev*[b] and A. E. Roitberg*[a]

Full • Submitted: 15 December 2019 • Accepted: 28 January 2020 • Published Online: 19 February 2020

Approaches for machine learning intermolecular interaction energies and application to energy components from symmetry adapted perturbation theory

J. Chem. Phys. **152**, 074103 (2020); https://doi.org/10.1063/1.5142636

Derek P. Metcalf[1], Alexios Koutsoukas[2], Steven A. Spronk[2], Brian L. Claus[2], Deborah A. Loughney[2], Stephen R. Johnson[2], Daniel L. Cheney[2], *and* C. David Sherrill[1,a)]

npj | Computational Materials

www.nature.com/npjcompumats

ARTICLE OPEN

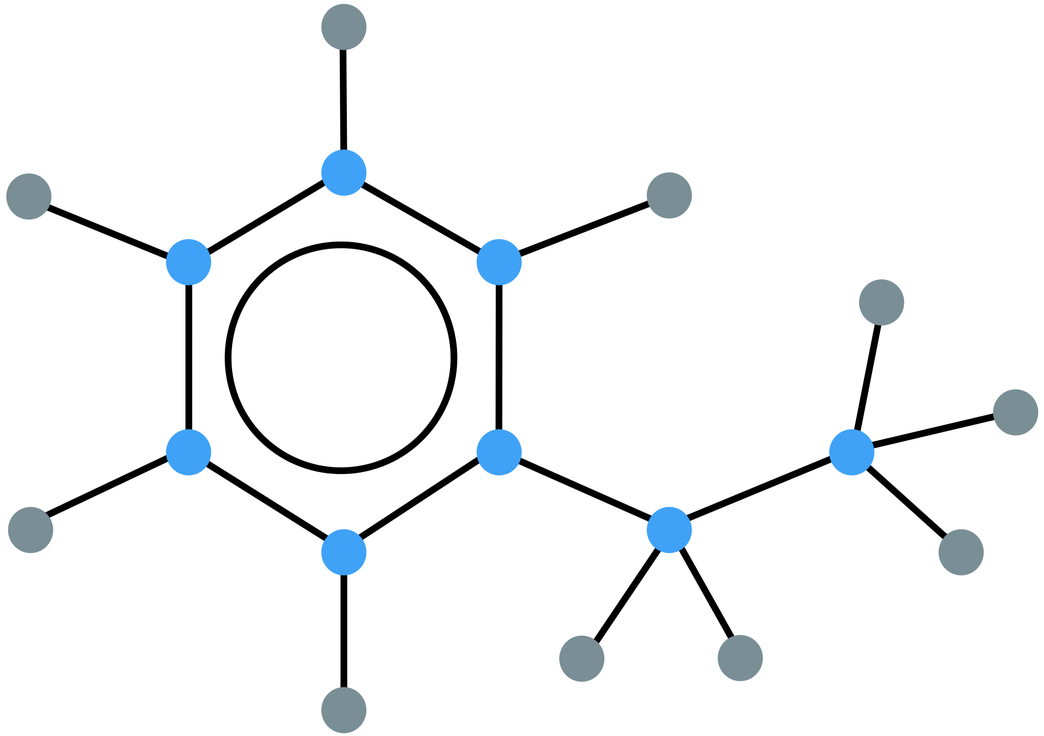
Check for updates

Inverse design of two-dimensional materials with invertible neural networks

Victor Fung[1✉], Jiaxin Zhang[2✉], Guoxiang Hu[3], P. Ganesh[1] and Bobby G. Sumpter[1]

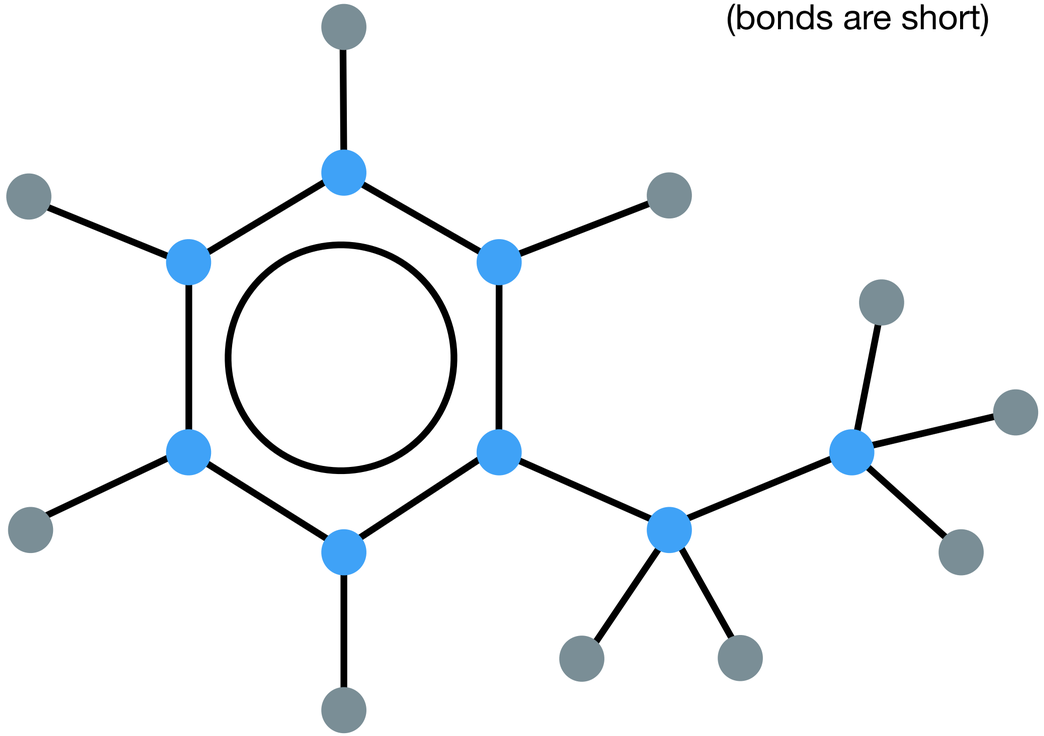
Force fields and energy locality

$$E^{FF} = \sum^{bonded} E_{ij}^{bond} + E_{ijk}^{angle} + E_{ijkl}^{torsion} + \sum^{non-bonded} E_{ij}^{elst} + E_{ij}^{vdW}$$



Force fields and energy locality

$$E^{FF} = \sum^{bonded} \underbrace{E_{ij}^{bond} + E_{ijk}^{angle} + E_{ijkl}^{torsion}}_{\text{Local by construction (bonds are short)}} + \sum^{non-bonded} \underbrace{E_{ij}^{elst} + E_{ij}^{vdW}}_{\text{Local by physical form (} E_{ij}^{elst} \text{ decays like } r_{ij}^{-1}, \; E_{ij}^{vdW} \text{ decays like } r_{ij}^{-6} \text{)}}$$



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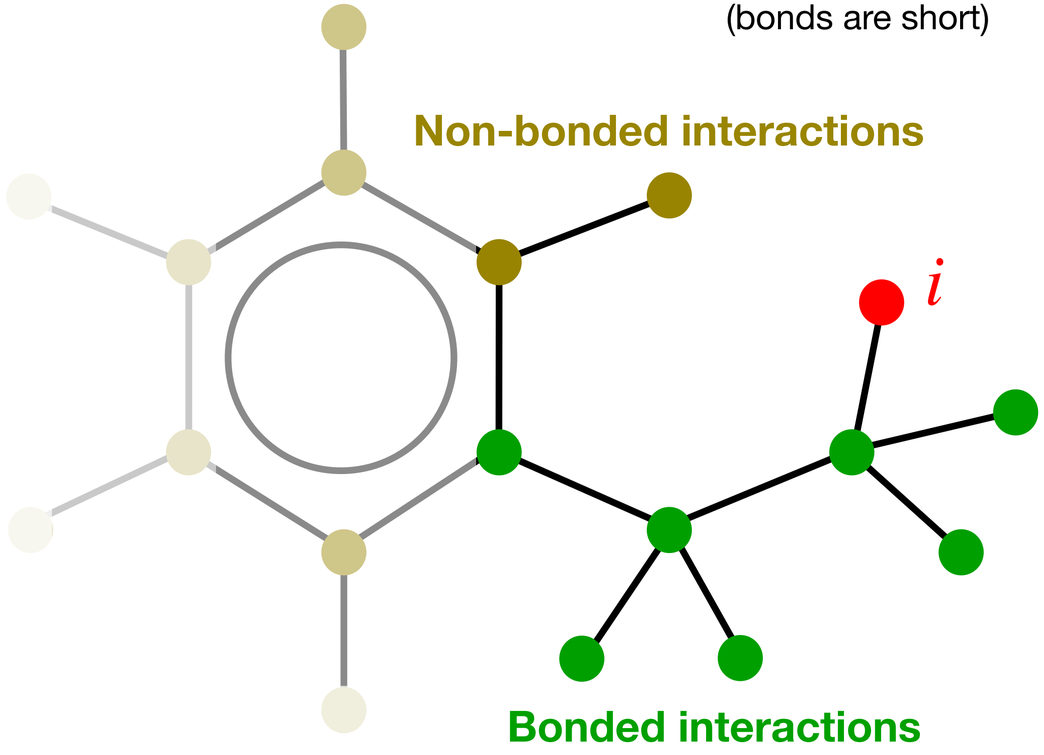
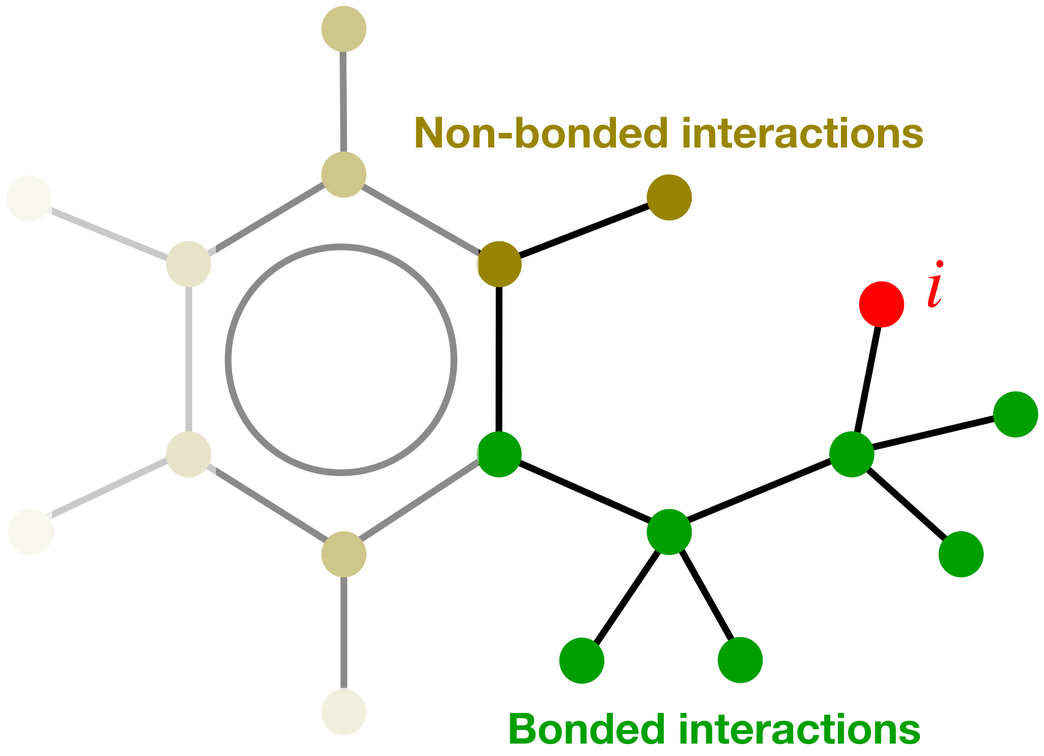


This is separable; it may be expressed as a sum of atomic contributions:

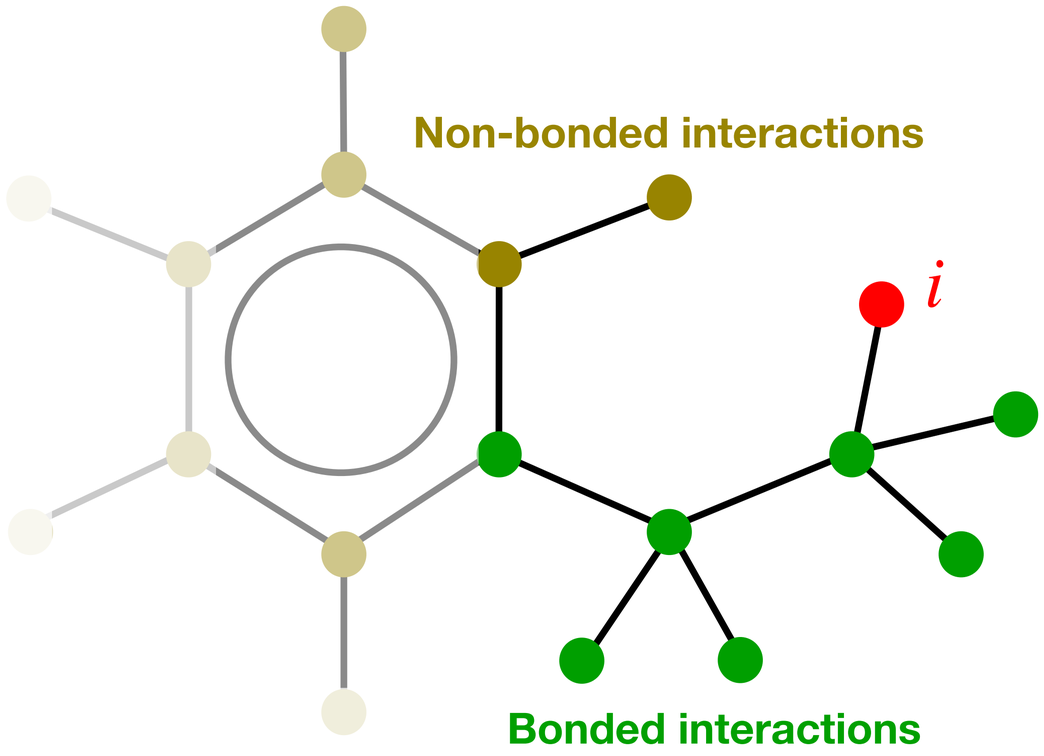
$$E^{FF} = \sum_{i}^{N} E_i^{FF}$$

Force fields and energy locality



What are some weaknesses of the force field energy function?

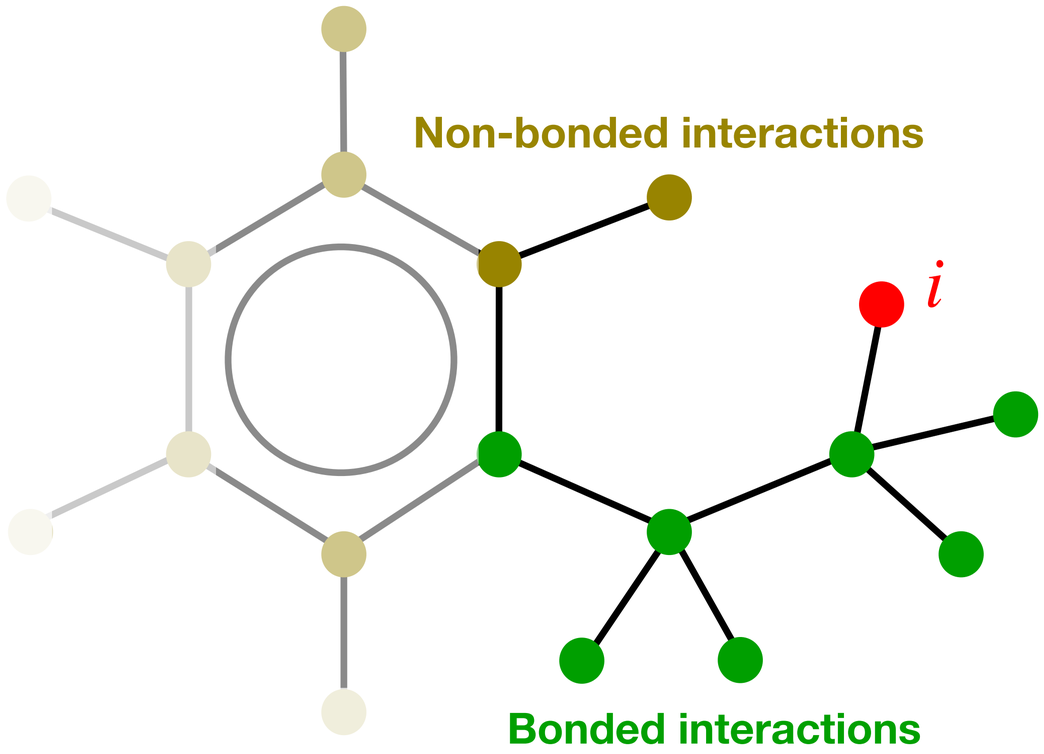
Force fields and energy locality



What are some weaknesses of the force field energy function?

- No reactions
- Requires atom-typing
- Bonds / angles aren't really springs
- Van der Waals aren't really Lennard-Jones
- Electrostatics isn't really just charge-charge Coulomb's Law

Force fields and energy locality



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$\Longrightarrow$ Force fields lack *flexibility* to express a complicated function*

* This point is an oversimplification — force fields can generally express very complicated functions but are still too constrained to have QM-like accuracy on arbitrary systems

“Fixing” force fields

1. Remove bonds
2. Describe atoms only by their element

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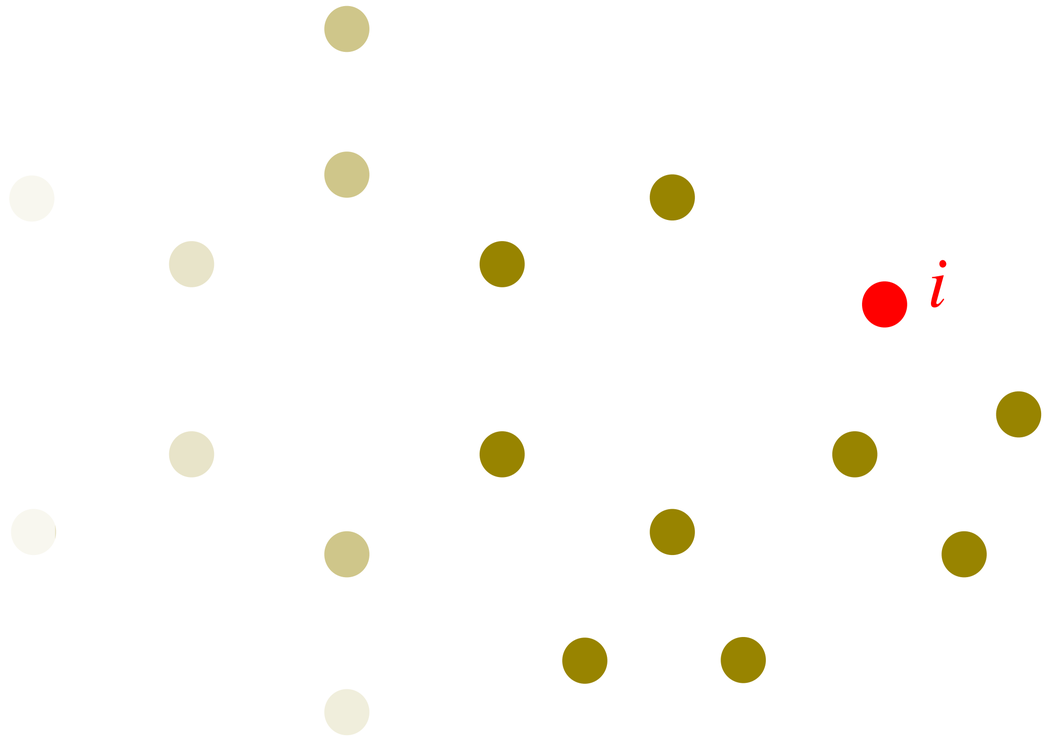
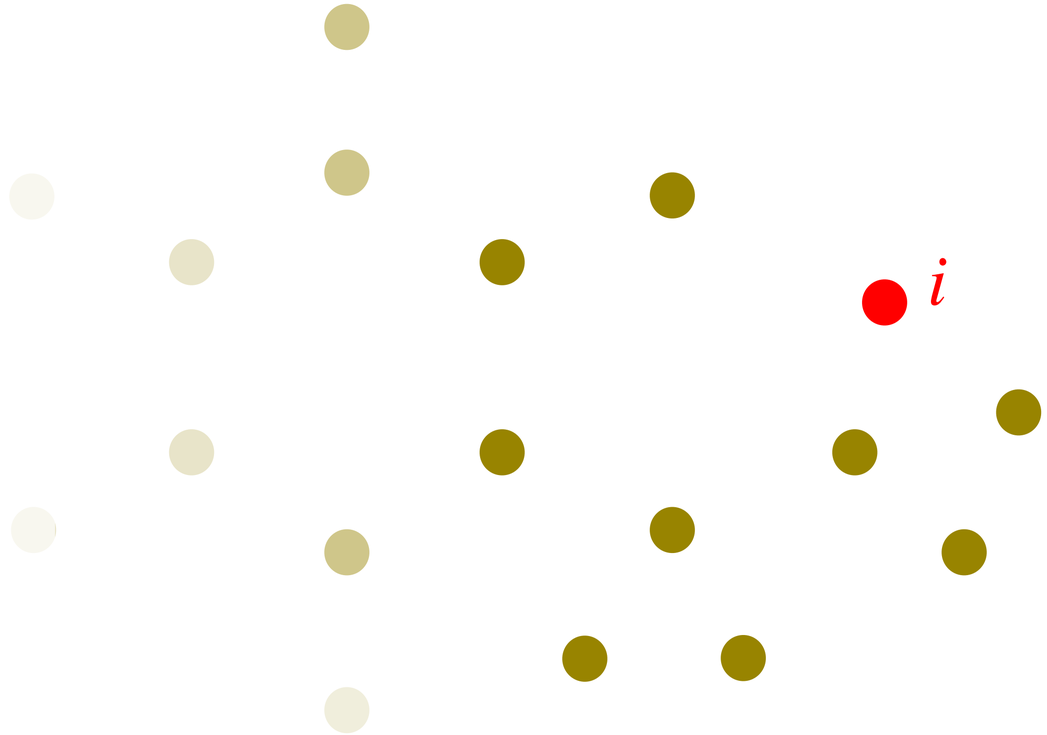


“Fixing” force fields

1. Remove bonds
2. Describe atoms only by their element
3. **Describe atomic *environments*, not just 2- 3- and 4-body interatomic distances**



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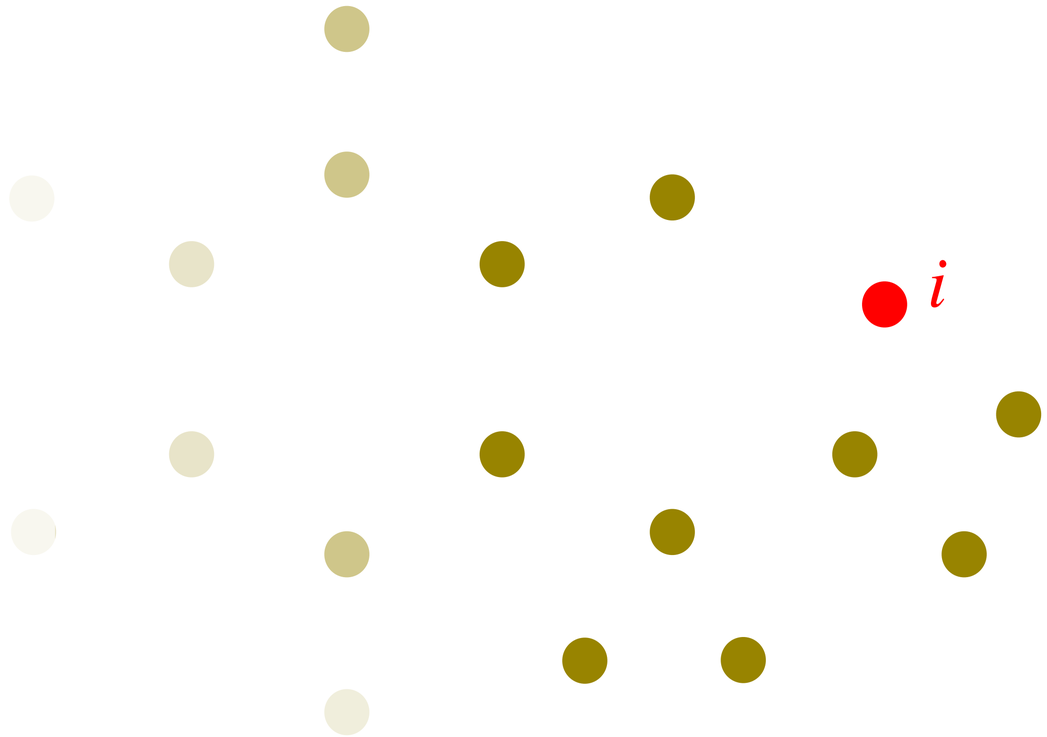
Atomic environment vectors (AEVs)



$$E = \sum_{i}^{N} E_i$$

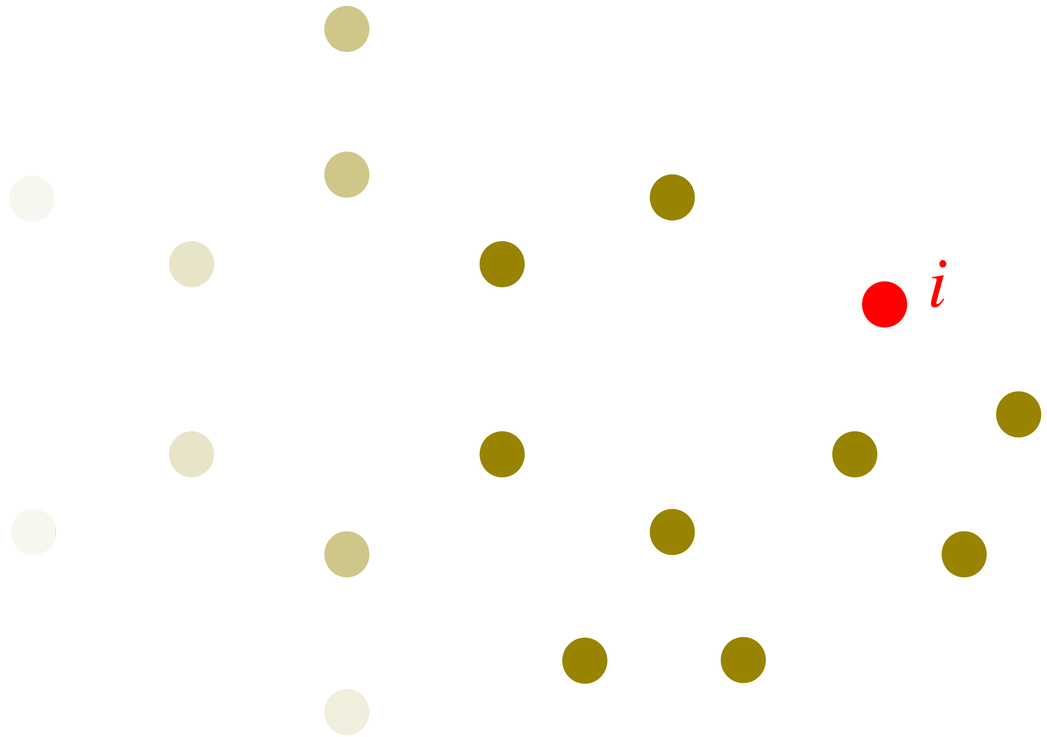
$$E_i = NN(\overrightarrow{x_i}); \quad \overrightarrow{x_i} \in \mathbb{R}^{n_f}$$

AEV of atom i,
A vector of numbers intended to describe its local environment.

Must be the same length n_f for all atoms

Atomic environment vectors (AEVs)

How about this one?



$$\overrightarrow{x_i} = \{Z_j r_{ij} \forall r_{ij} < r_c\}$$

With atom j atomic number Z_j

and cutoff distance r_c

Atomic environment vectors (AEVs)

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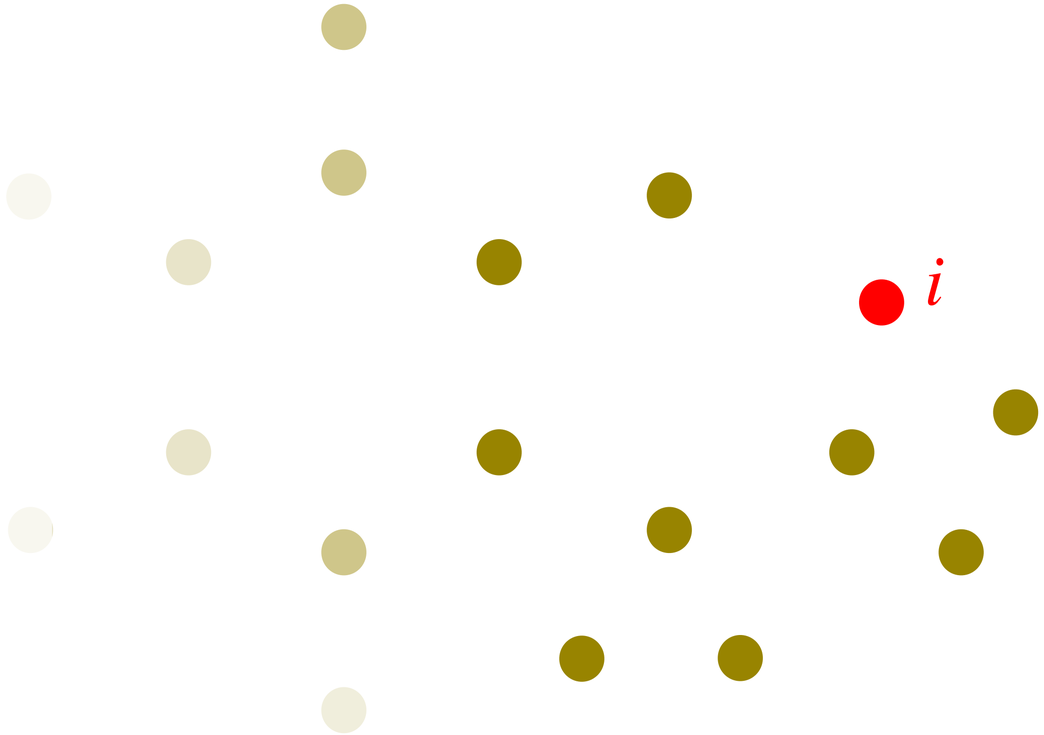


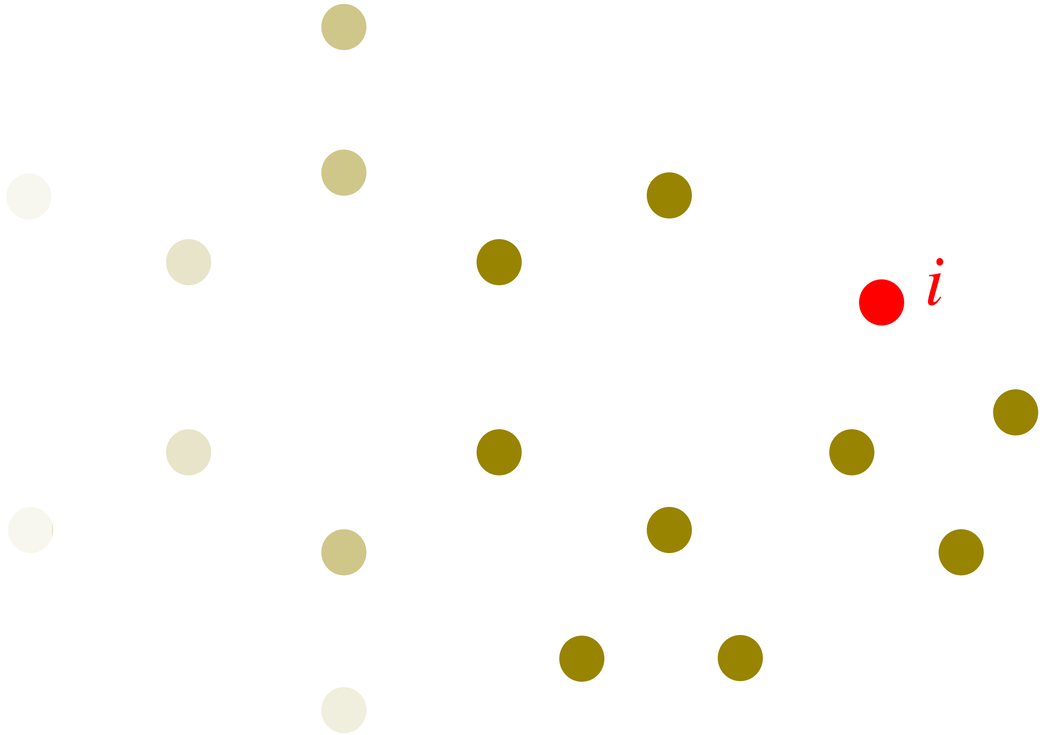
Variable length — number of atoms within a cutoff may be different between atoms

Atomic environment vectors (AEVs)

“Symmetry Functions”

Let’s try the same information, but without dependence on # j:

$$\overrightarrow{x_i} = \{ \sum_j e^{-\eta_k (r_{ij} - R_k)^2} \}_k$$



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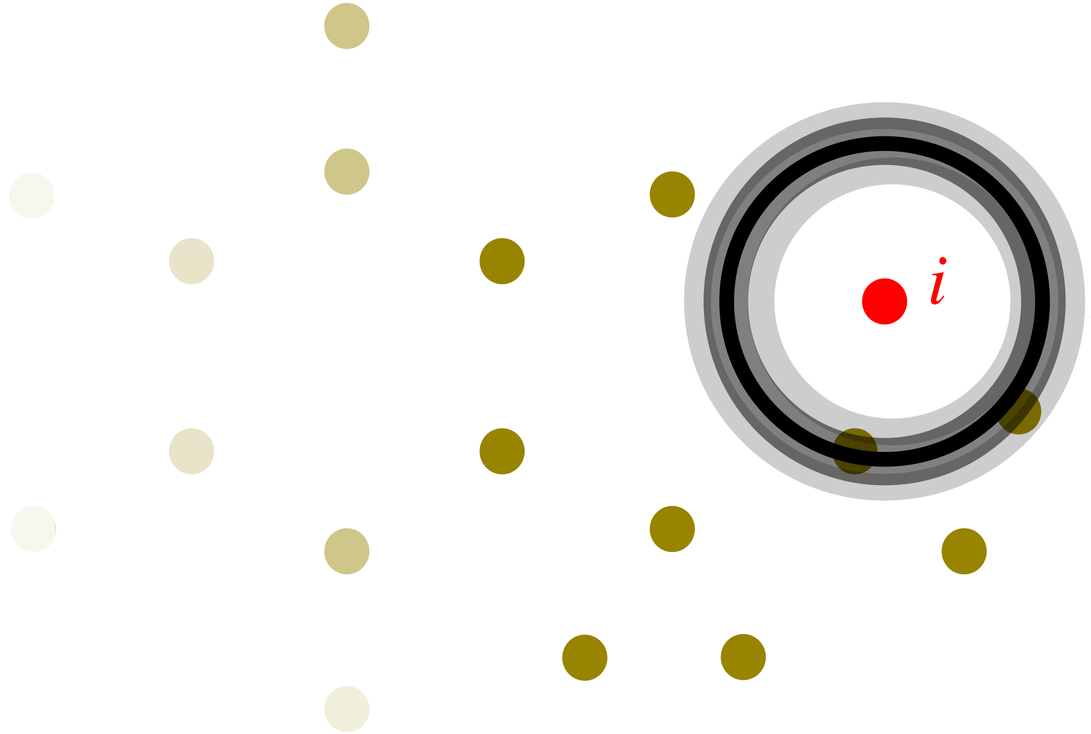

Each element of this set is a Gaussian placed at an offset of R_k from atom i. We choose a fixed number of elements k.

$k = 0$

$\eta_0 = 1.5$

$R_0 = 1.0$

Atomic environment vectors (AEVs)

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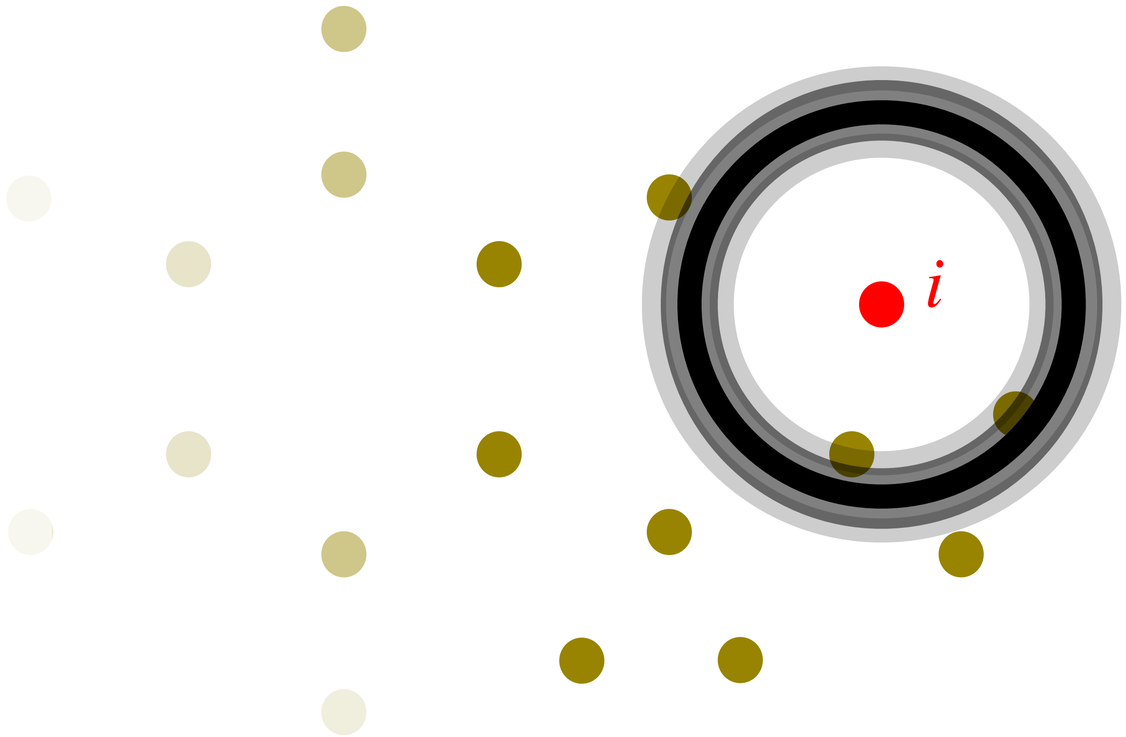

Each element of this set is a Gaussian placed at an offset of R_k from atom i. We choose a fixed number of elements k.

$k = 1$

$\eta_1 = 0.75$

$R_0 = 1.25$

Atomic environment vectors (AEVs)

"Symmetry Functions"

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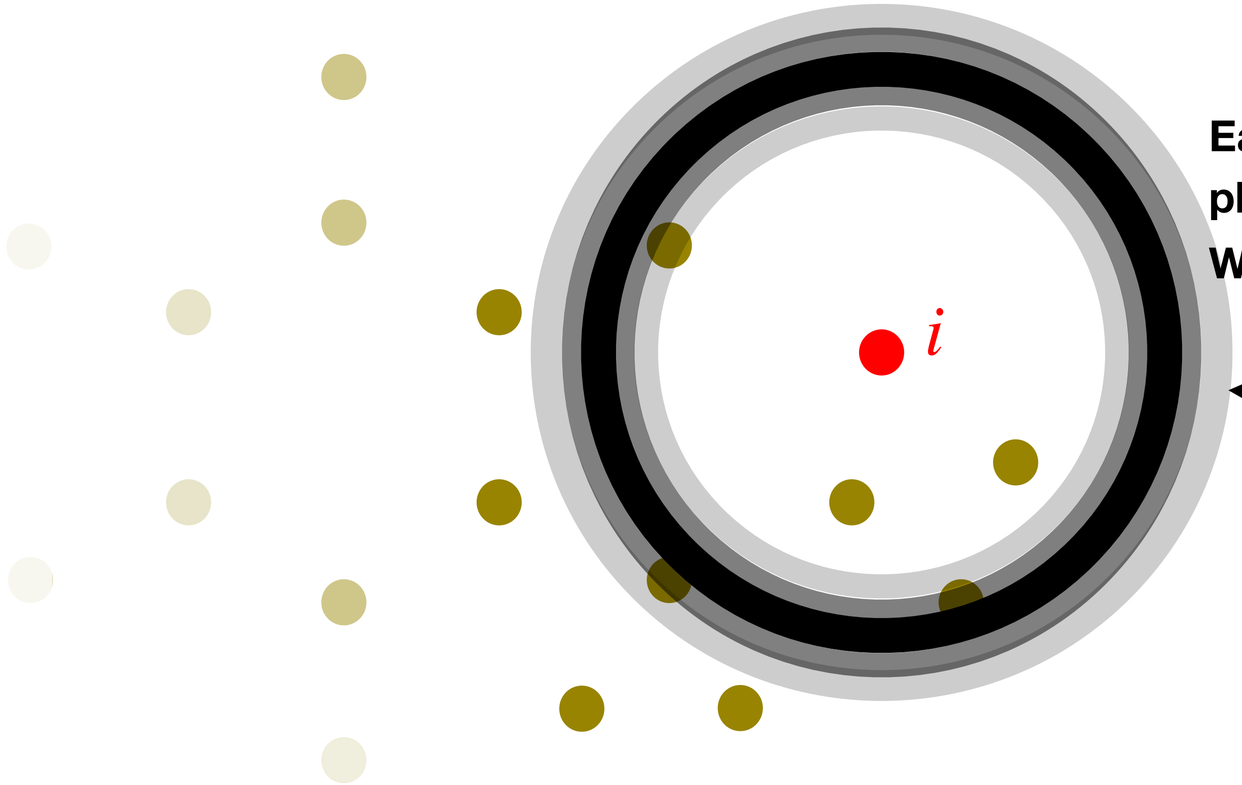
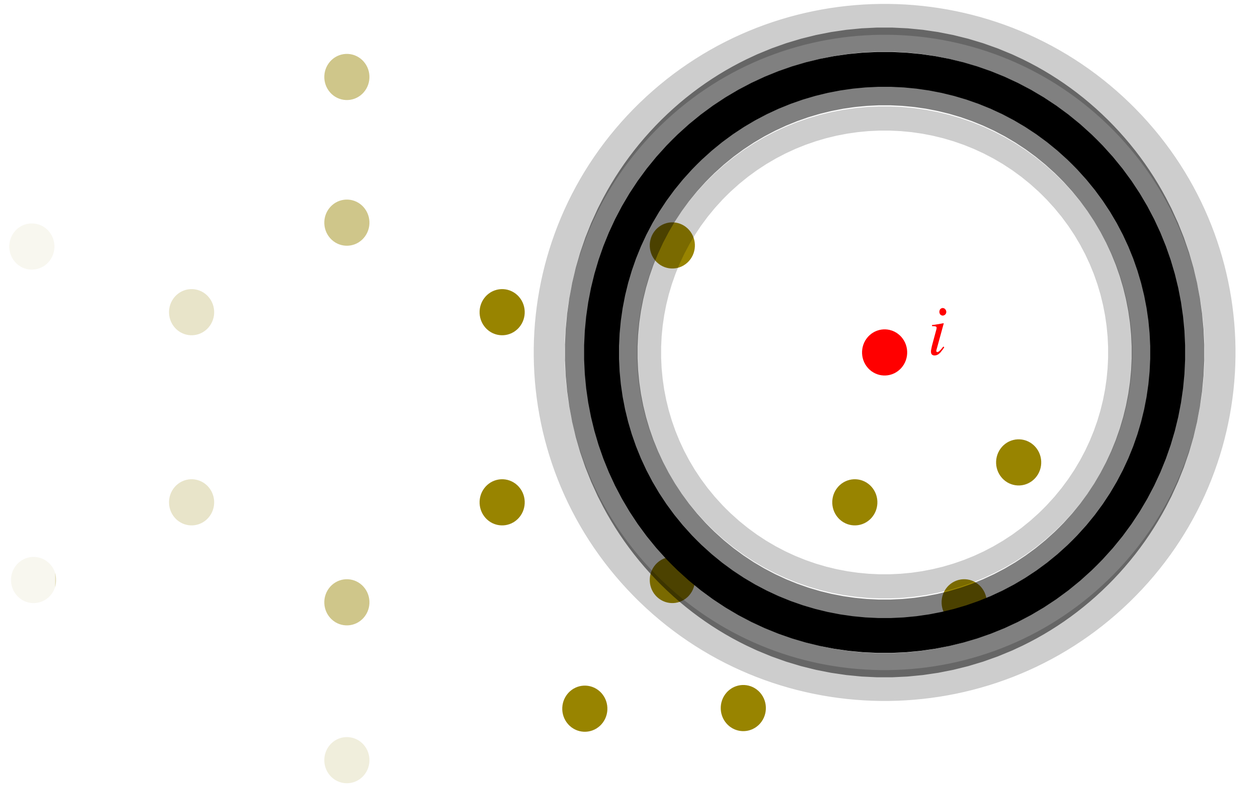


Each element of this set is a Gaussian placed at an offset of R_k from atom i. We choose a fixed number of elements k.

$$k = n_f$$
$$\eta_{n_f} = 0.5$$
$$R_{n_f} = 4.0$$

Atomic environment vectors (AEVs)



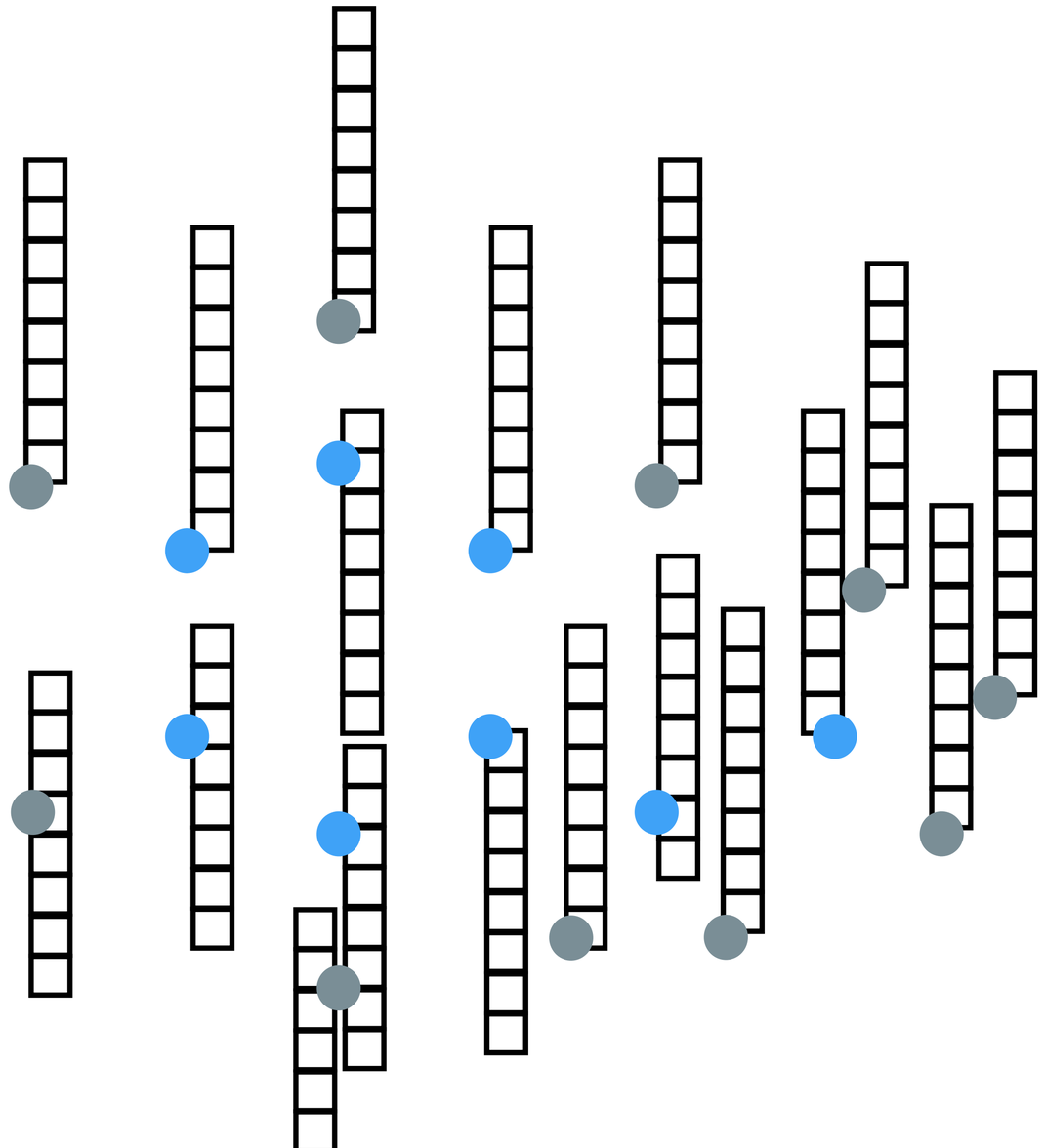
“Symmetry Functions”

$$\overrightarrow{x_i} = \{ \sum_j e^{-\eta_k (r_{ij} - R_k)^2} \}_k$$

Notice this does not distinguish between nearby elements.

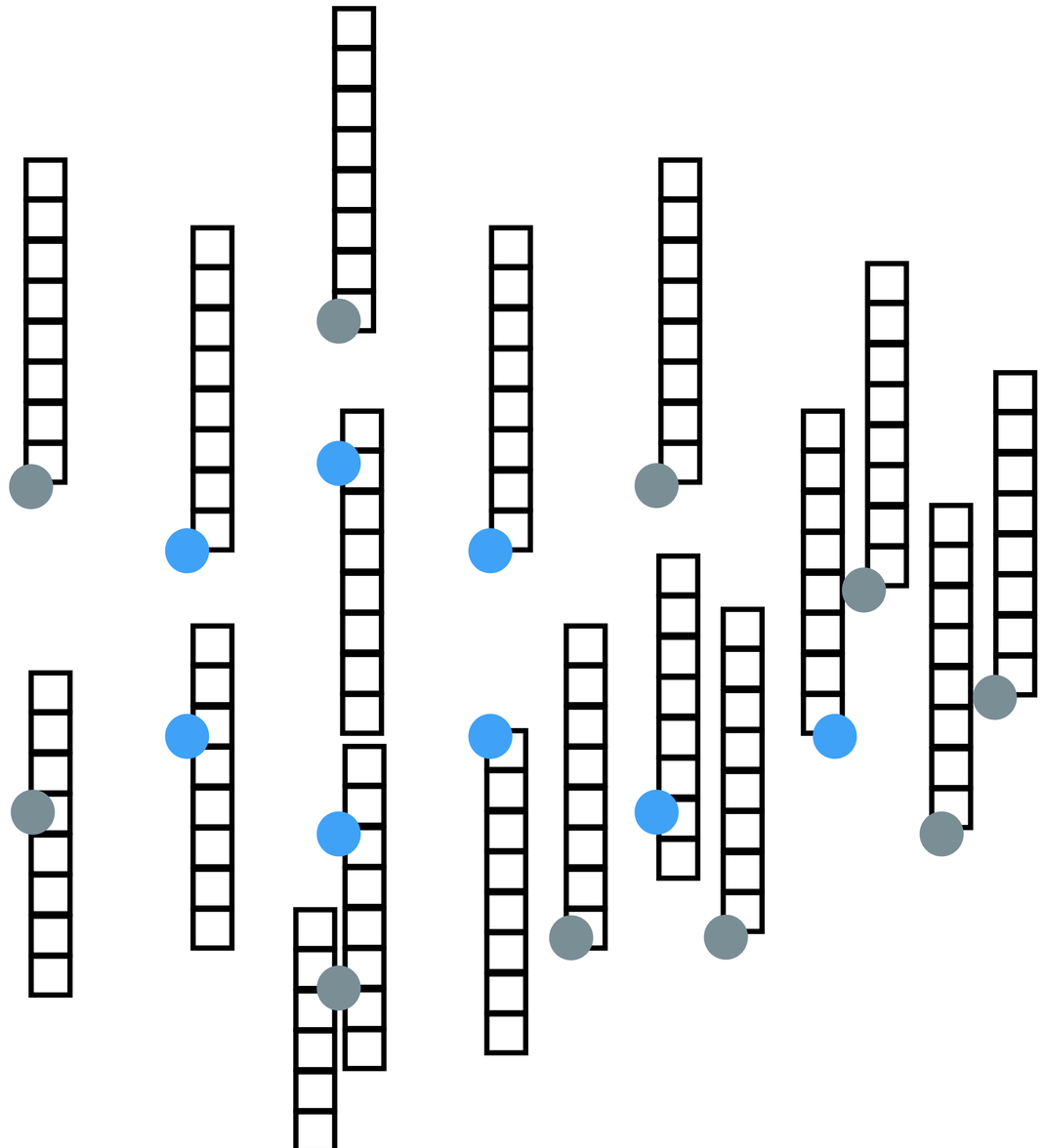
To fix this, we simply make n_{elem} vectors of this form, where each vector addresses a single element (H, C, etc.)

Atomic environment vectors (AEVs)



$$\overrightarrow{x_i} = \{ \sum_j e^{-\eta_k (r_{ij} - R_k)^2} \}_k = \begin{matrix} k = \\ \\ 1 \\ 2 \\ 3 \\ \\ \dots \\ \\ \\ n_f \times n_{elem} \end{matrix}$$

Atomic environment vectors (AEVs) : Exercise 3.1 and 3.2



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Let's see how $\overrightarrow{x_i}$ is implemented in Python

Behler-Parrinello Neural Networks (BPNN)

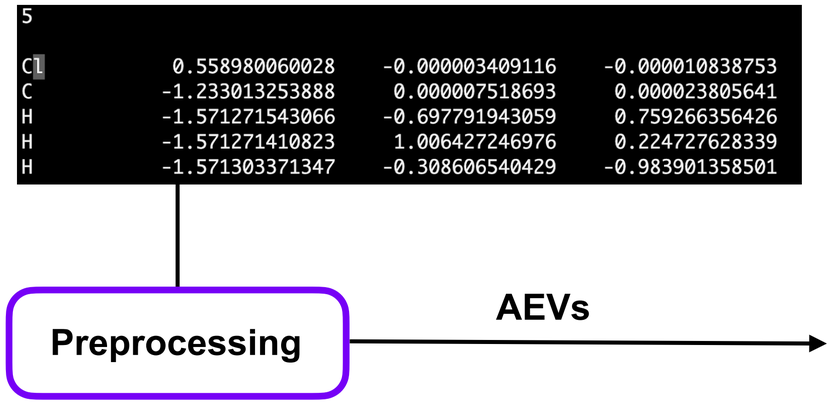
BPNN — a set of element-specific atomic-additive neural networks that use symmetry functions as input:

$$E^{NN} = \sum_{elem} \sum_{i}^{N_{elem}} NN_{elem}(\overrightarrow{x_i}),$$

$$\overrightarrow{x_i} = f(\mathcal{N}(i)),$$

for some simple function f and the neighborhood of atom i given by $\mathcal{N}(i)$.

Behler-Parrinello Neural Networks (BPNN)



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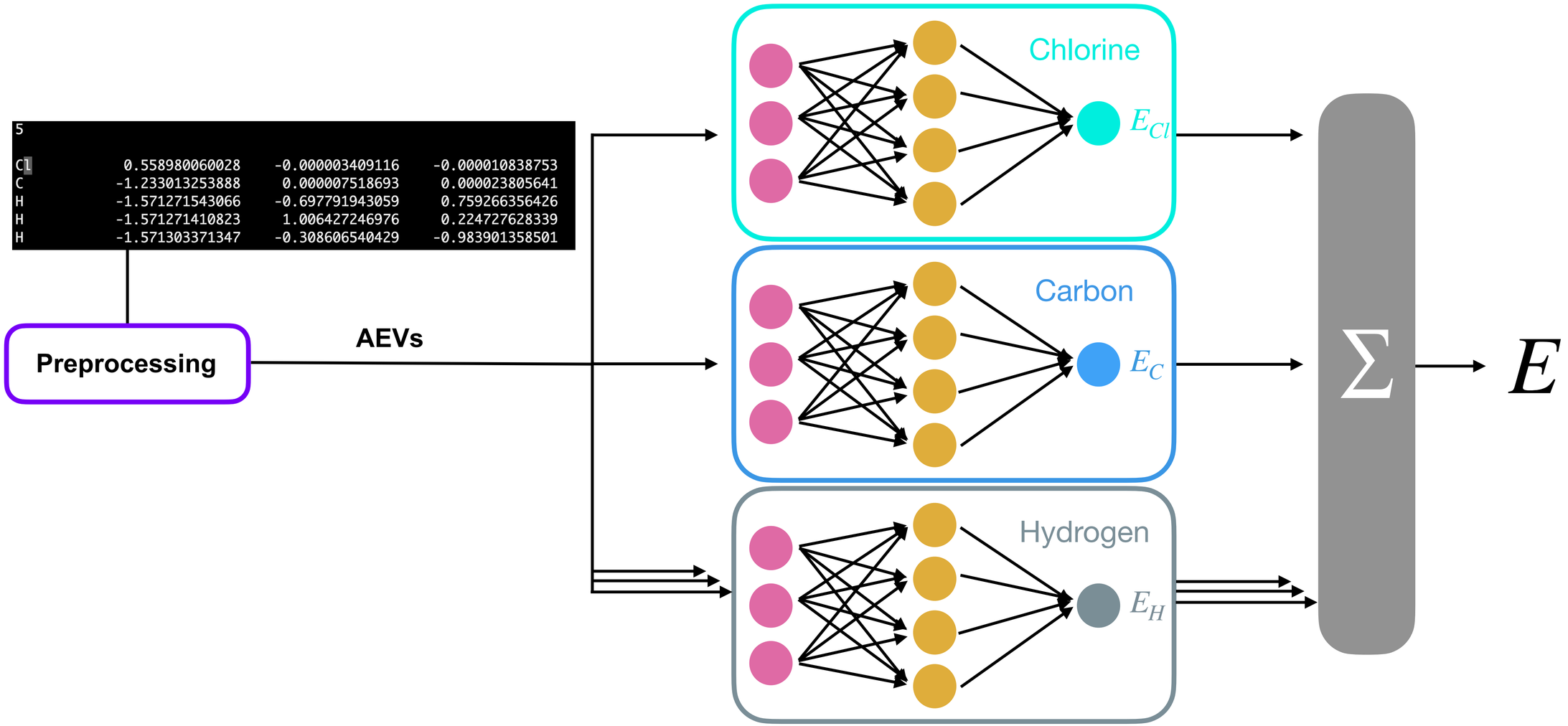

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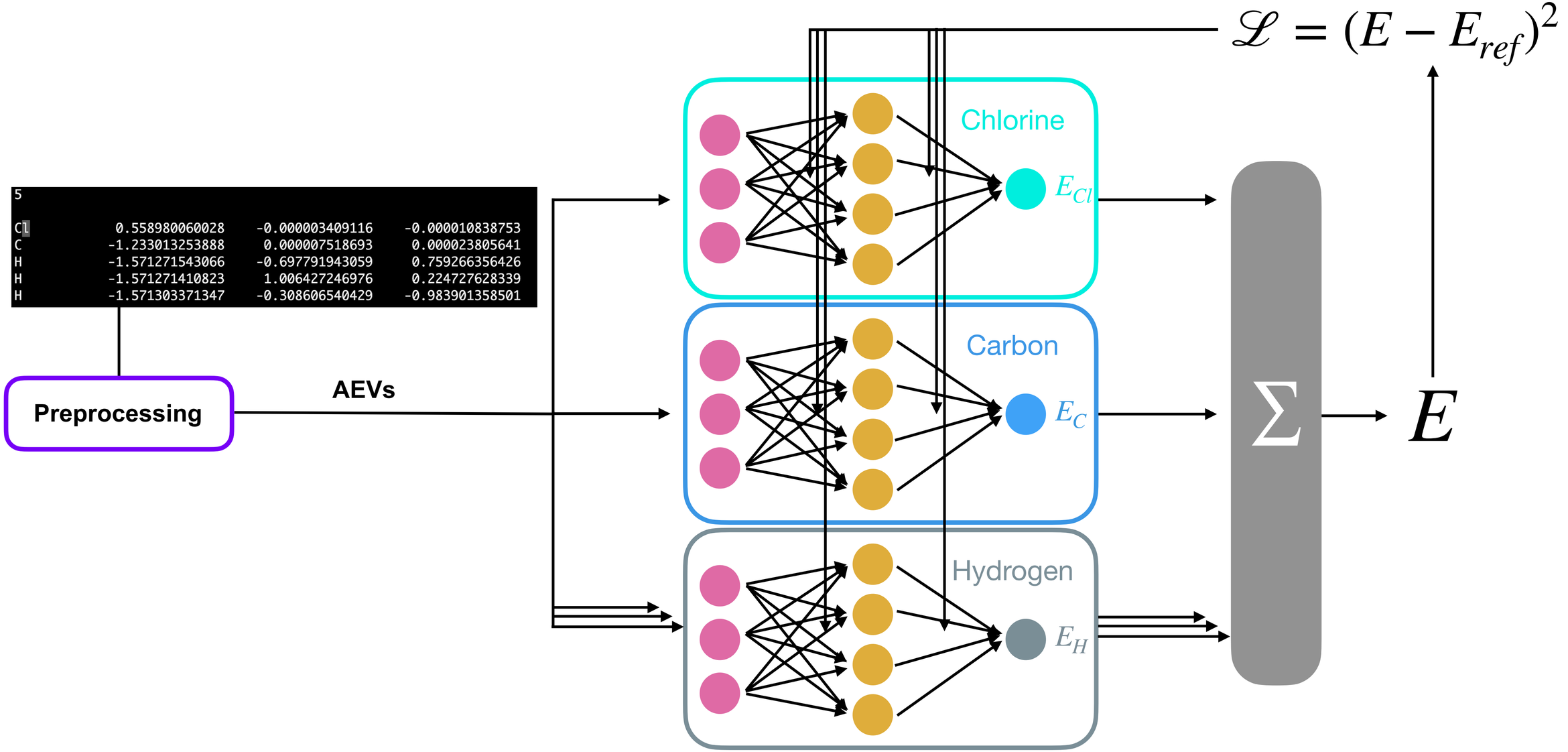
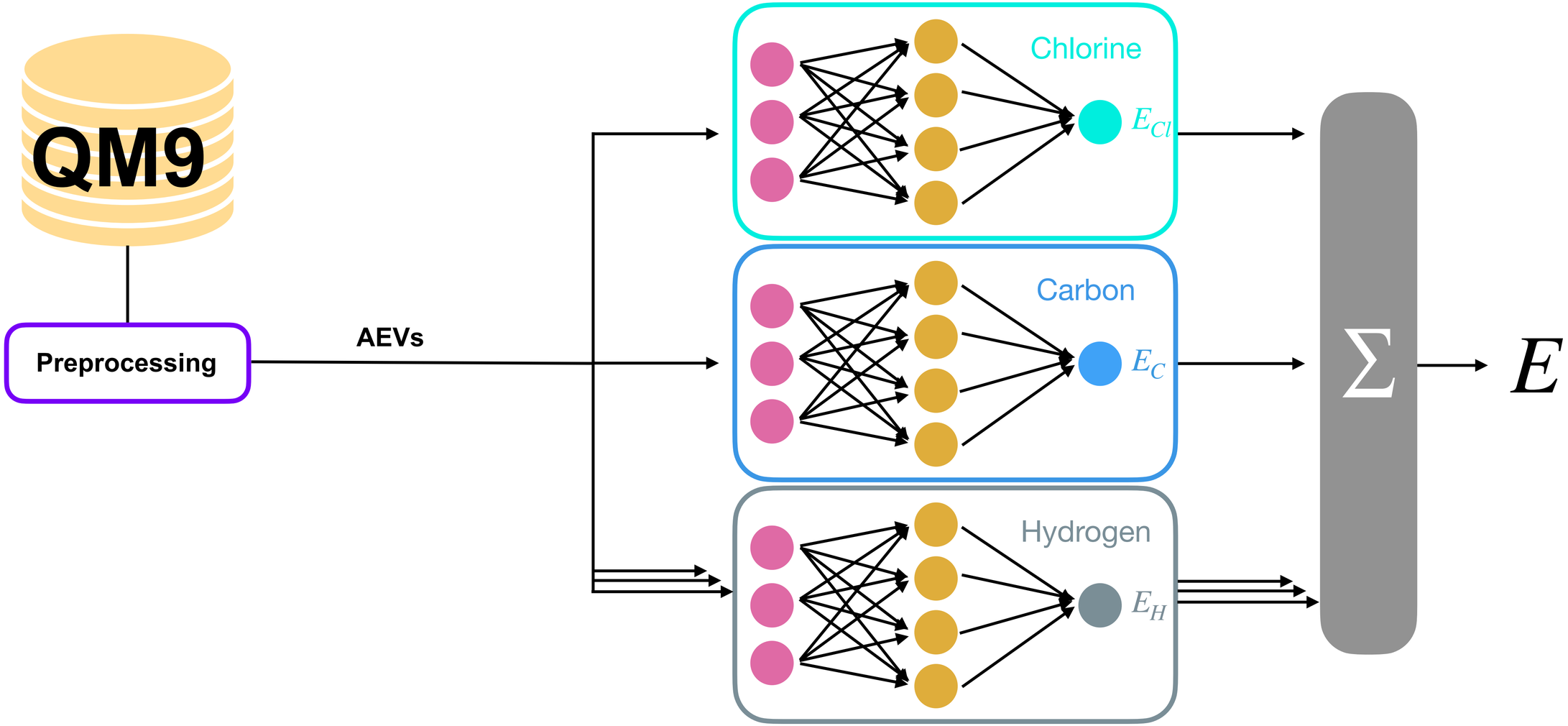


Behler-Parrinello Neural Networks (BPNN): Exercise 3.3



Reading Homework - links on GitHub

1.

Atom-centered symmetry functions for constructing high-dimensional neural network potentials

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Submitted: 08 December 2010 • Accepted: 21 January 2011 • Published Online: 16 February 2011

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Check out the *angular* symmetry functions, which incorporate triples of atoms to get angular resolution. Implement them yourselves in the existing code and see how they impact errors.

2.

Neural Message Passing for Quantum Chemistry

Justin Gilmer [1] **Samuel S. Schoenholz** [1] **Patrick F. Riley** [2] **Oriol Vinyals** [3] **George E. Dahl** [1]

Read this classic paper and introduction to message passing (/graph) neural networks in chemistry. The next two meetings will expand on this topic.