GRACE universal interatomic potential for materials discovery and design

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ICAMS
INTERDISCIPLINARY CENTRE FOR ADVANCED MATERIALS SIMULATION

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- Matteo Rinaldi
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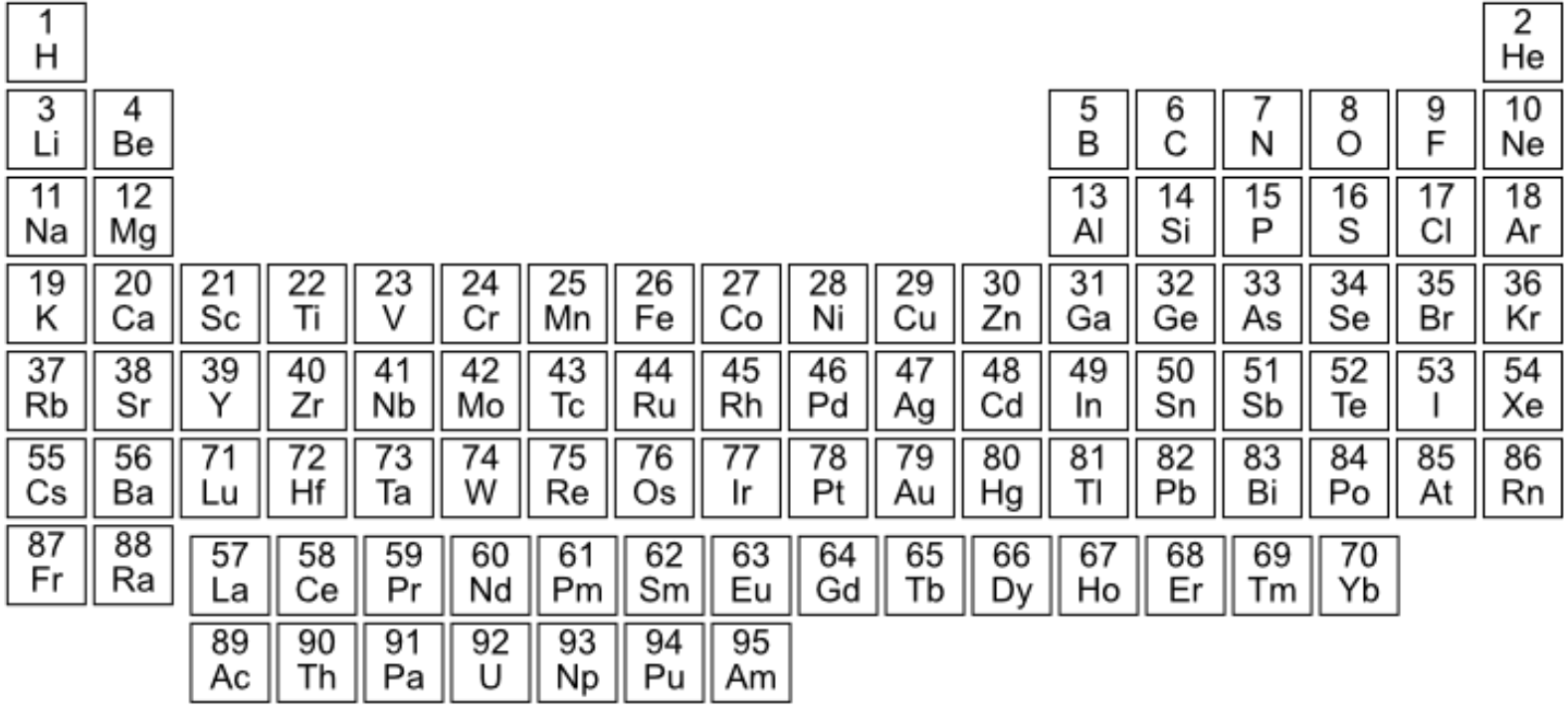

1 H
2 He
3 Li
4 Be
5 B
6 C
7 N
8 O
9 F
10 Ne
11 Na
12 Mg
13 Al
14 Si
15 P
16 S
17 Cl
18 Ar
19 K
20 Ca
21 Sc
22 Ti
23 V
24 Cr
25 Mn
26 Fe
27 Co
28 Ni
29 Cu
30 Zn
31 Ga
32 Ge
33 As
34 Se
35 Br
36 Kr
37 Rb
38 Sr
39 Y
40 Zr
41 Nb
42 Mo
43 Tc
44 Ru
45 Rh
46 Pd
47 Ag
48 Cd
49 In
50 Sn
51 Sb
52 Te
53 I
54 Xe
55 Cs
56 Ba
71 Lu
72 Hf
73 Ta
74 W
75 Re
76 Os
77 Ir
78 Pt
79 Au
80 Hg
81 Tl
82 Pb
83 Bi
84 Po
85 At
86 Rn
87 Fr
88 Ra
57 La
58 Ce
59 Pr
60 Nd
61 Pm
62 Sm
63 Eu
64 Gd
65 Tb
66 Dy
67 Ho
68 Er
69 Tm
70 Yb
89 Ac
90 Th
91 Pa
92 U
93 Np
94 Pu
95 Am

100 years of Quantum Mechanics

- Hamiltonian operator

$$\hat{H} = -\sum_i \frac{\hbar^2}{2m}\nabla_i^2 - \sum_I \frac{\hbar^2}{2M_I}\nabla_I^2 - \sum_{iI} \frac{Z_I e^2}{|\boldsymbol{R}_I - \boldsymbol{x}_i|} + \sum_{ij}^{i\neq j} \frac{1}{2}\frac{e^2}{|\boldsymbol{x}_i - \boldsymbol{x}_j|} + \sum_{IJ}^{I\neq J} \frac{1}{2}\frac{Z_I Z_J e^2}{|\boldsymbol{R}_I - \boldsymbol{R}_J|}$$

kinetic energy of electrons	kinetic energy of nuclei	electron-nucleus interaction	electron-electron interaction	nucleus-nucleus interaction

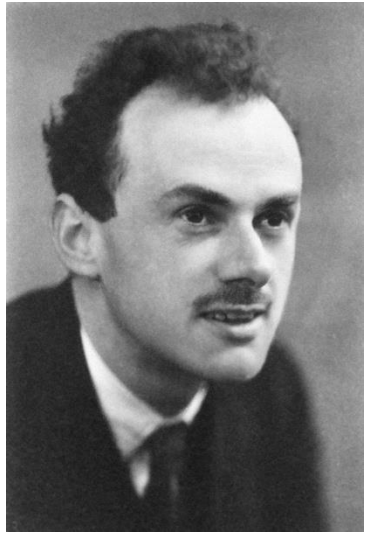
- Schrödinger equation

$$E\Psi = \hat{H}\Psi$$

Erwin Schrödinger

➔ **Precise prediction of atomic interactions, at high computational cost**

Direction from nearly 100 years ago



Paul Dirac

The mathematical theory of a large part of physics and the whole of chemistry are thus completely known, and the difficulty is only that the exact application of these laws leads to equations much too complicated to be soluble.

It therefore becomes desirable that approximate practical methods of applying quantum mechanics should be developed, which can lead to an explanation of the main features of complex atomic systems without too much computation.

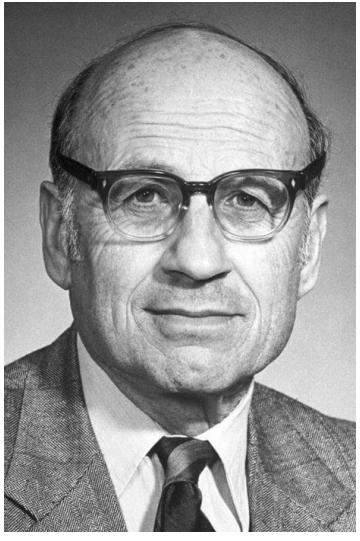
P.A.M. Dirac, Proc. R. Soc. Lond. (1929)

Density functional theory

- From many electrons to one effective electron

$$\left(-\frac{\hbar^2}{2m}\nabla^2 + V_{ion}(\boldsymbol{x}) + V^H(\boldsymbol{x}) + V^{XC}(\boldsymbol{x})\right)\psi_n(\boldsymbol{x}) = \varepsilon_n\psi_n(\boldsymbol{x})$$

- kinetic energy of electron
- interaction with nuclei
- Hartree potential: electrostatic interaction between electrons
- Exchange-Correlation potential: approximation required



Walter Kohn

- The most cited concept in the physical sciences
- Widely used software (VASP, …)

- **Nearly universally applicable across the periodic table**

➔ **Accurate prediction of atomic interaction, moderate computational cost**

Density functional theory

- From many electrons to one effective electron

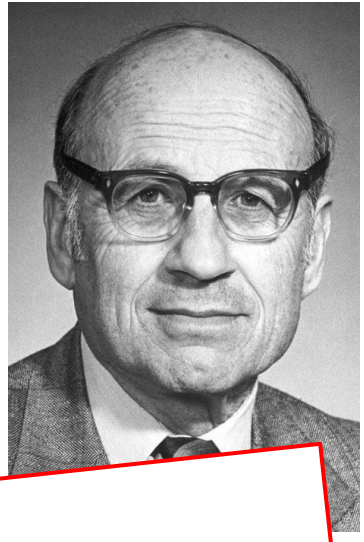
$$\left(-\frac{\hbar^2}{2m}\nabla^2 + V_{ion}(\boldsymbol{x}) + V^H(\boldsymbol{x}) + V^{XC}(\boldsymbol{x})\right)\psi_n(\boldsymbol{x}) = \varepsilon_n\psi_n(\boldsymbol{x})$$

kinetic energy of electron

interaction with nuclei

Hartree potential: electrostatic interaction between electrons

Exchange-Correlation potential: approximation required



- The most cited concept in th
- Wi

- **Still about 1 million times too slow**
- **Need much faster models of the atomic interaction**

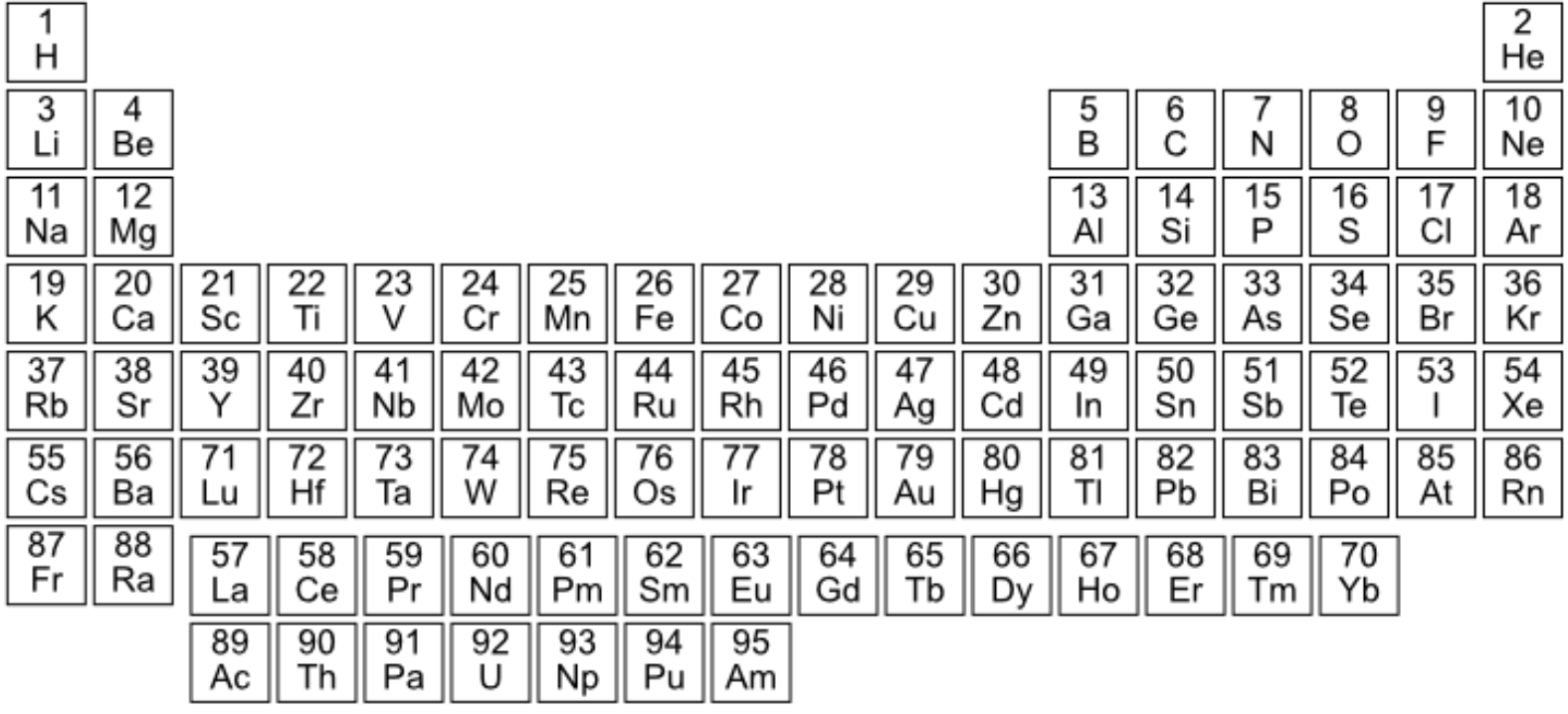
applicable across the periodic table

Accurate prediction of atomic interaction, moderate computational cost

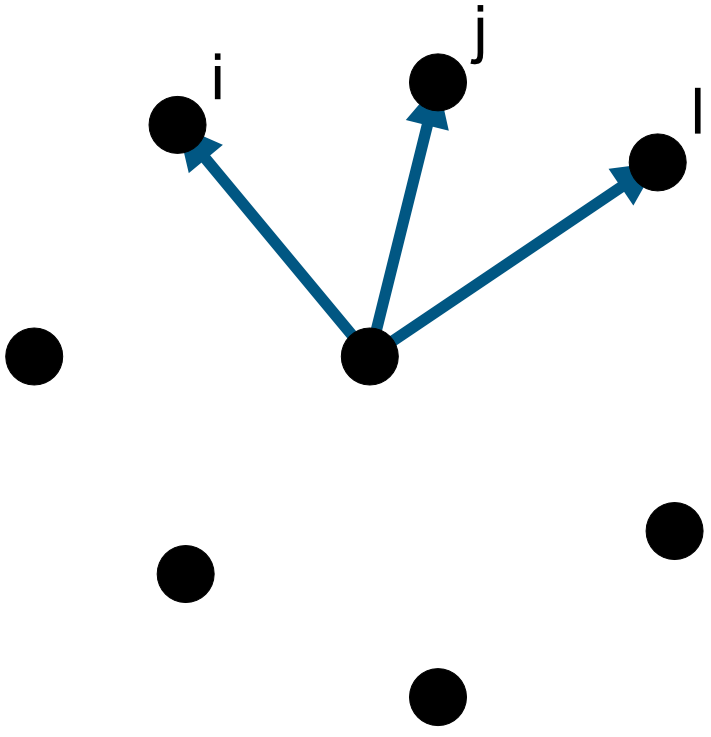
Outline

1. **Challenges**
2. **Atomic Cluster Expansion (ACE)**
3. **Graph Atomic Cluster Expansion (GRACE)**
4. **Efficient evaluation**
5. **Foundational interatomic potential**
6. **Conclusion**

1. Challenge: geometric and chemical complexity



Challenge: geometric complexity



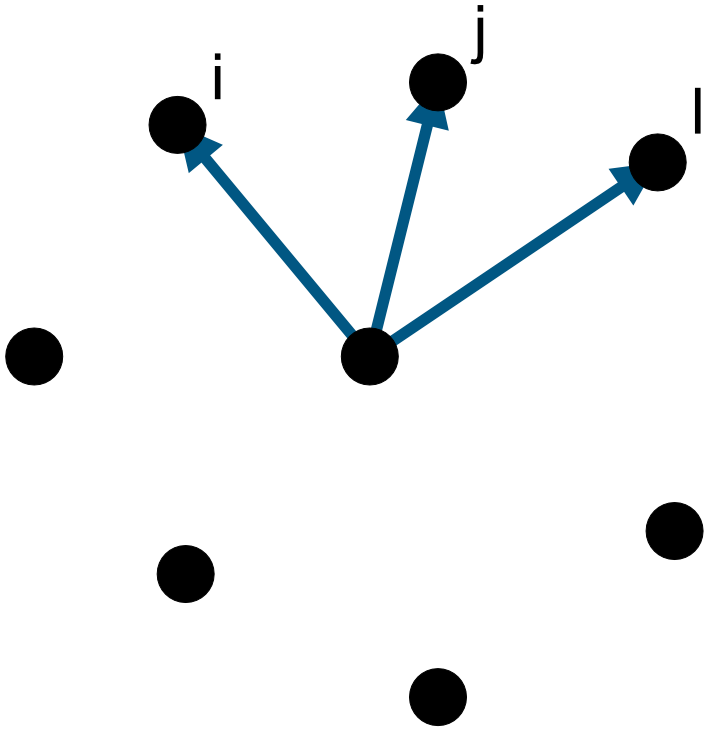
- **Challenge:** complexity

N^K

number of neighbors

body order

Challenge: geometric complexity



- **Challenge:** complexity

N^K

number of neighbors

body order

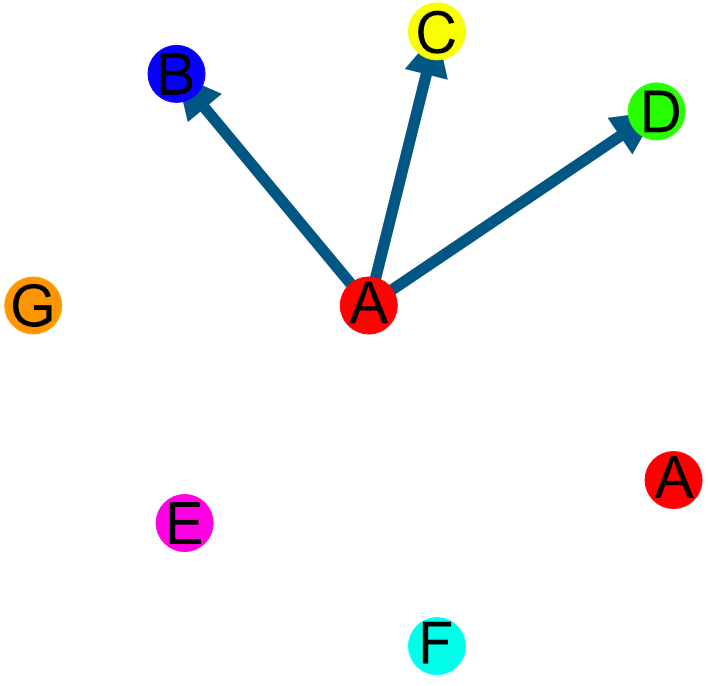
- **Solution:** product representation

$$\sum_{ijl}^{N} x_i y_j z_l = (\sum_{i}^{N} x_i)(\sum_{i}^{N} y_i)(\sum_{i}^{N} z_i)$$

N^3 operations $\qquad$ $3N$ operations

➔ **Linear scaling with number of neighbors**

Challenge: chemical complexity

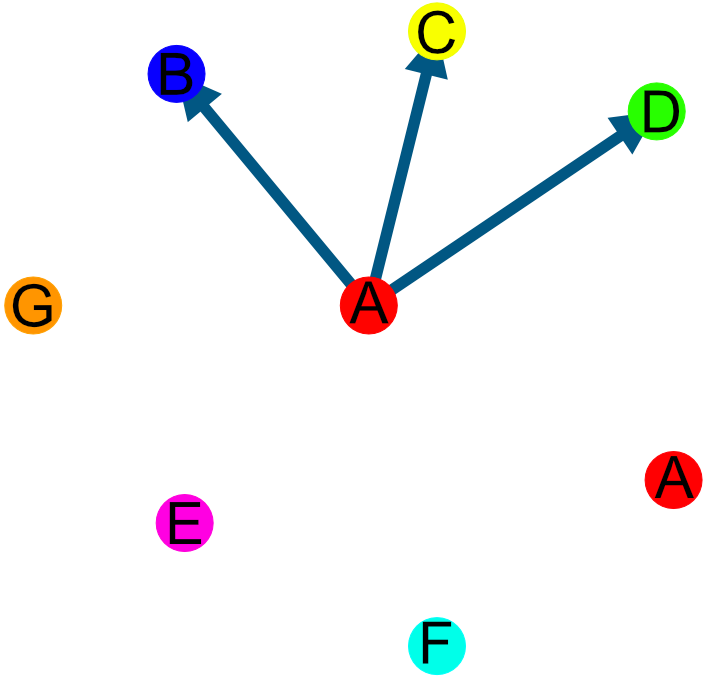


- **Challenge:** complexity for ~100 chemical elements

V_{ABCD}

$100^4 = 10^8$ entries

Challenge: chemical complexity



- **Challenge:** complexity for ~100 chemical elements

$$V_{ABCD}$$

$100^4 = 10^8$ entries

- **Solution:** tensor decomposition

$$V_{ABCD} = \sum_{k=1}^{K} \lambda_k c_A^{(k)} c_B^{(k)} c_C^{(k)} c_D^{(k)}$$

$c_A^{(k)}$

$K \times 100$ entries

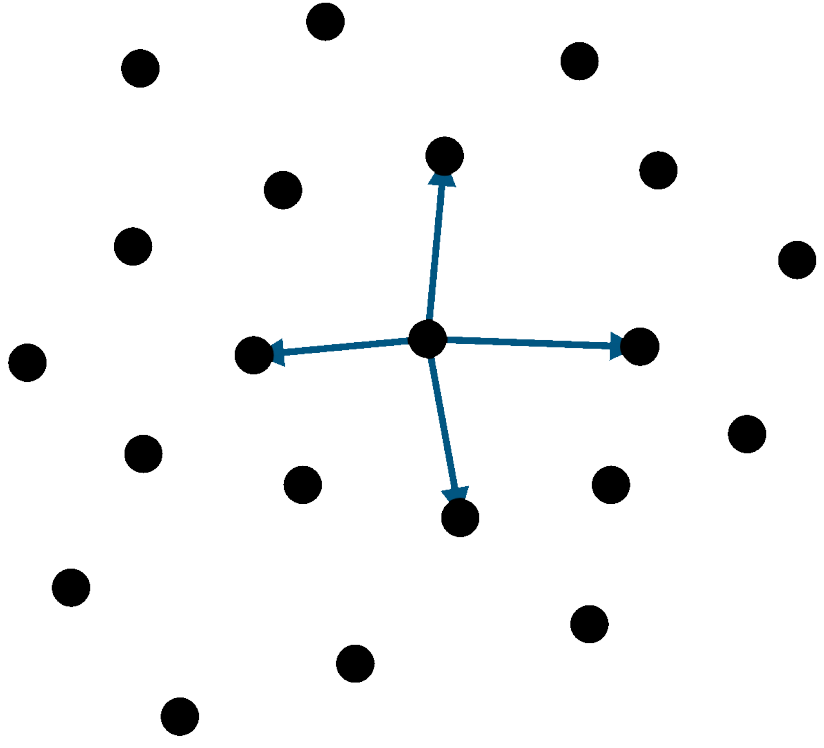
➔ **Nearly independent of number of chemical elements**

2. Atomic Cluster Expansion

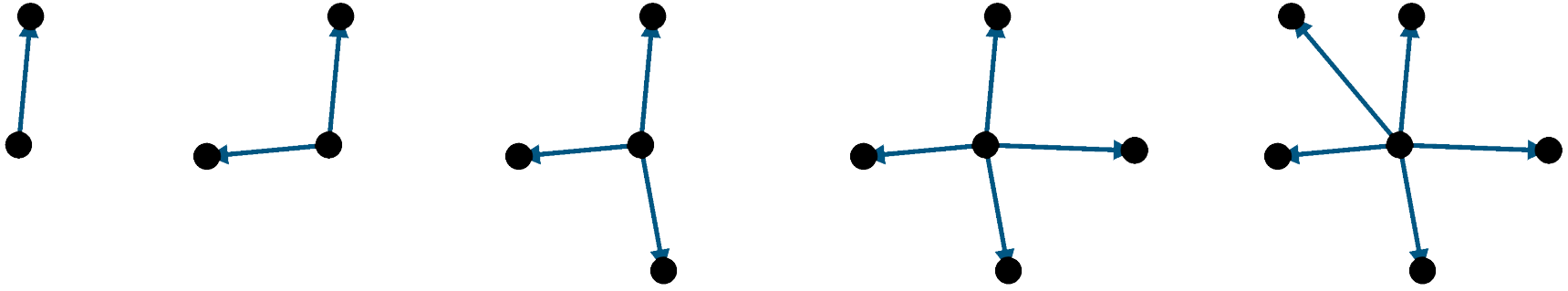
Learning atomic energies

Atomic energies

$$E = \sum_i E_i$$



Star graphs

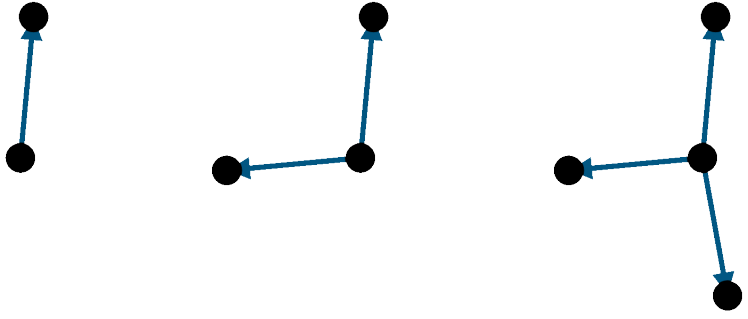


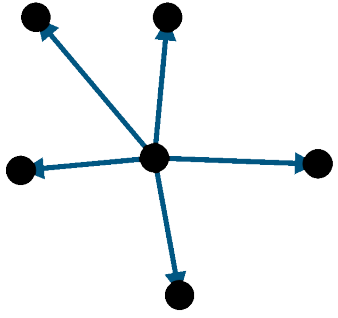
node = atom

edge = bond

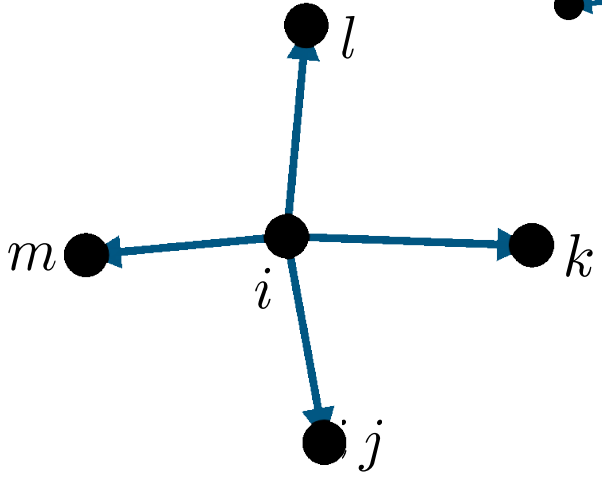
root node = atom for which energy is computed

Atomic cluster expansion





- Basis functions on graphs



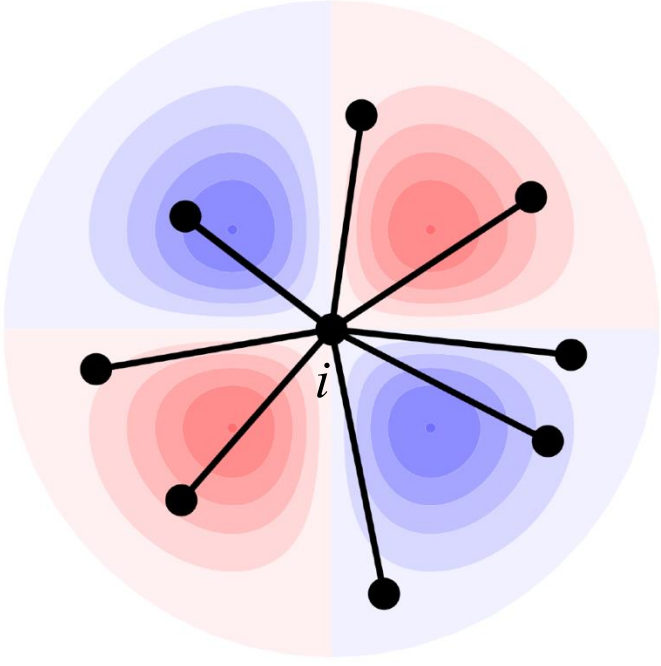
$$\Phi_{\alpha \mathbf{v}} = \phi_{i v_1}(\boldsymbol{r}_{ji}) \phi_{i v_2}(\boldsymbol{r}_{ki}) \phi_{i v_3}(\boldsymbol{r}_{li}) \phi_{i v_4}(\boldsymbol{r}_{mi})$$

Atomic cluster expansion

- **Basis functions are complete**

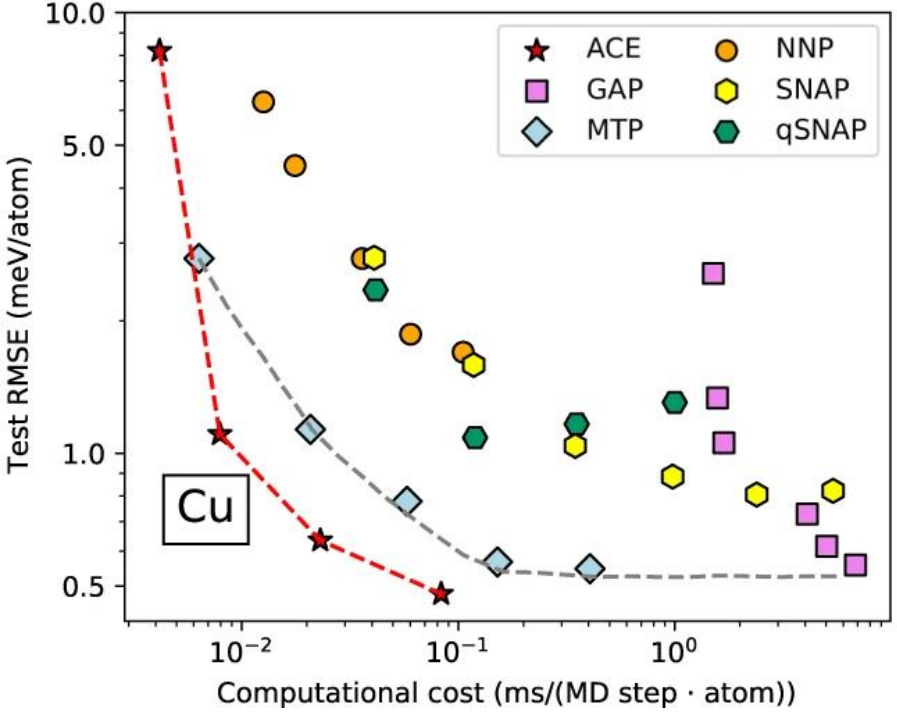
$$E = \sum_{\alpha \mathbf{v}} J_{\alpha \mathbf{v}} \Phi_{\alpha \mathbf{v}}$$

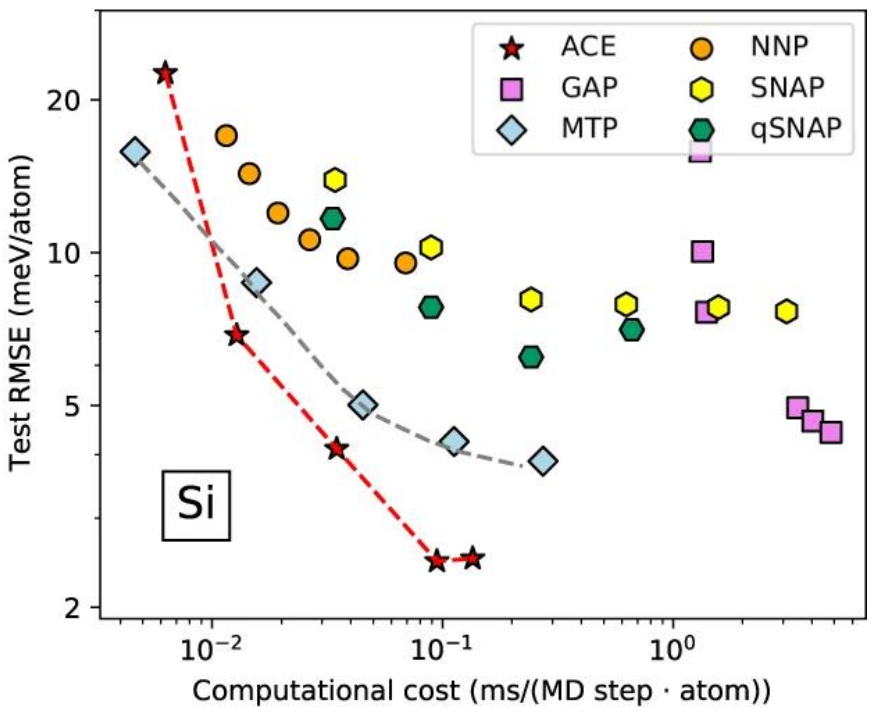
expansion coefficients



Drautz, PRB 99 (2019) 014104

Efficiency



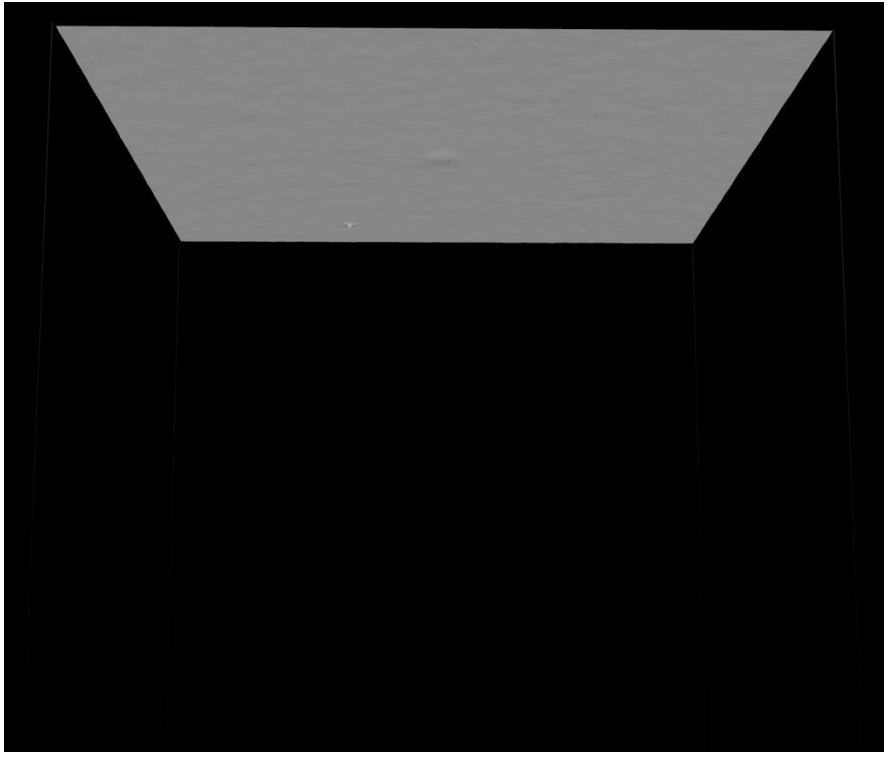


Reference: Zuo, et al, J. Phys. Chem. A 124, 731 (2020)

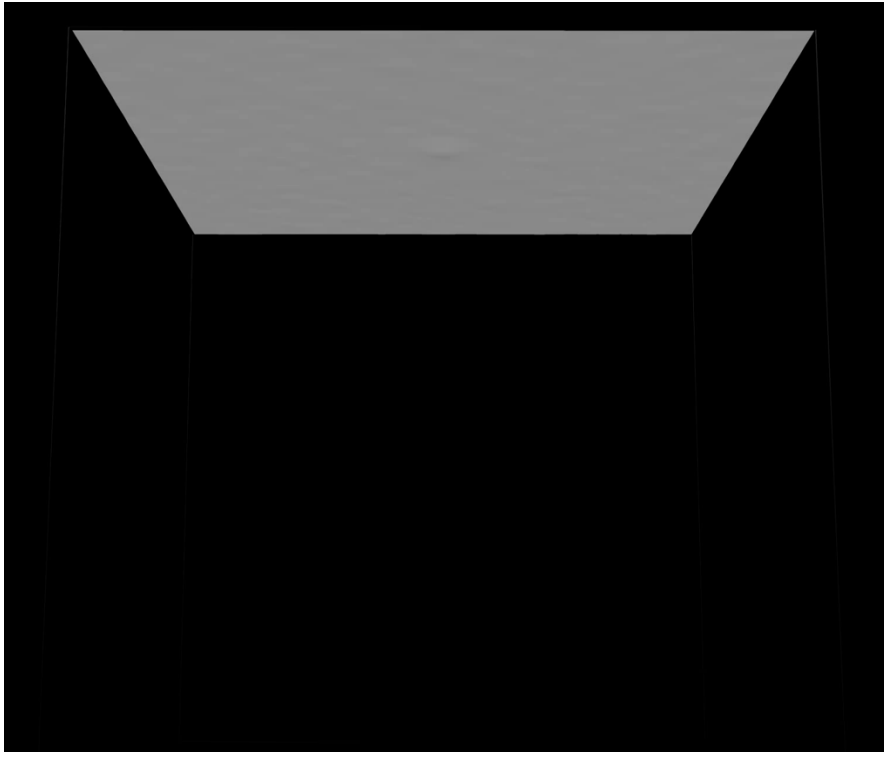
Y. Lysogorskiy, et al, npj Comput. Mater. (2021)

Example: nanoindentation

Copper



Tungsten

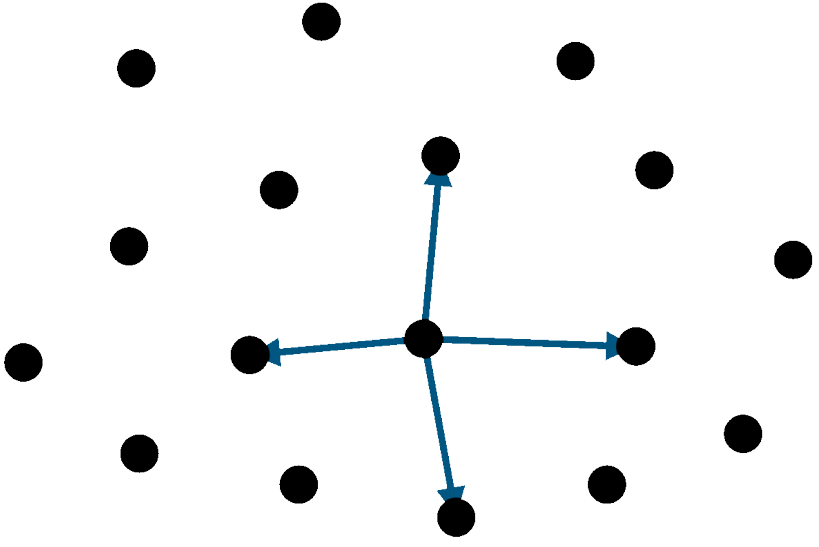


Immel, Sutmann (2025)

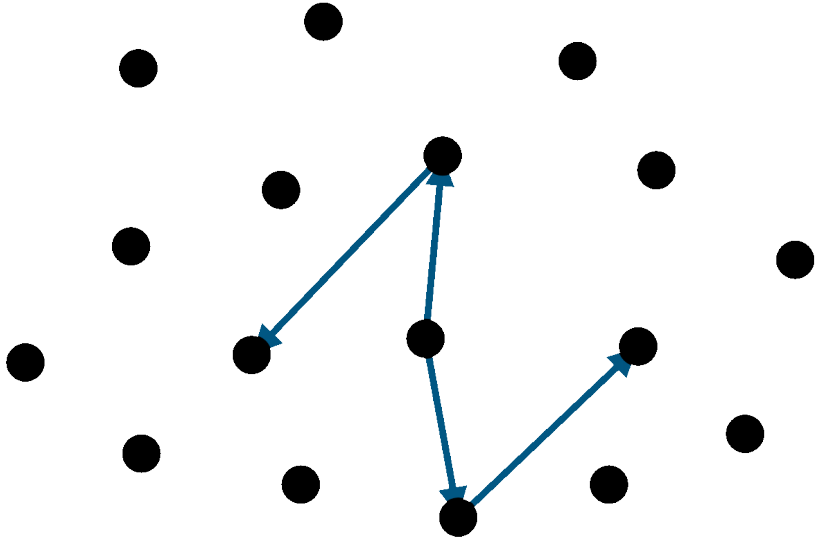
~ several millions of atoms for several millions of time steps

3. Graph Atomic Cluster Expansion

Further possible graphs

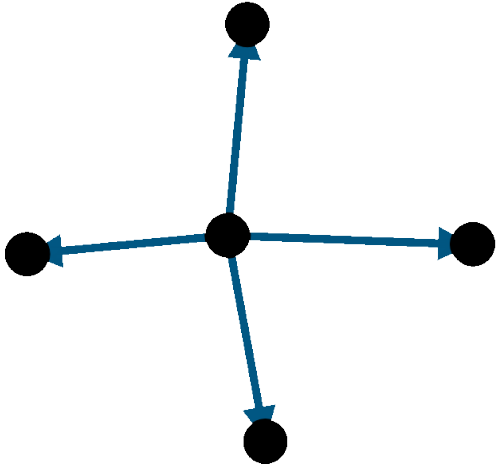


- Connecting to 4 neighboring atoms

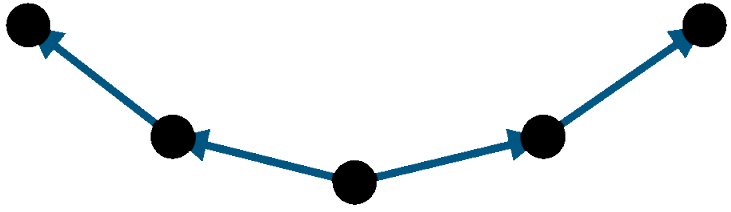


- Alternative to the same atoms

Graph topology



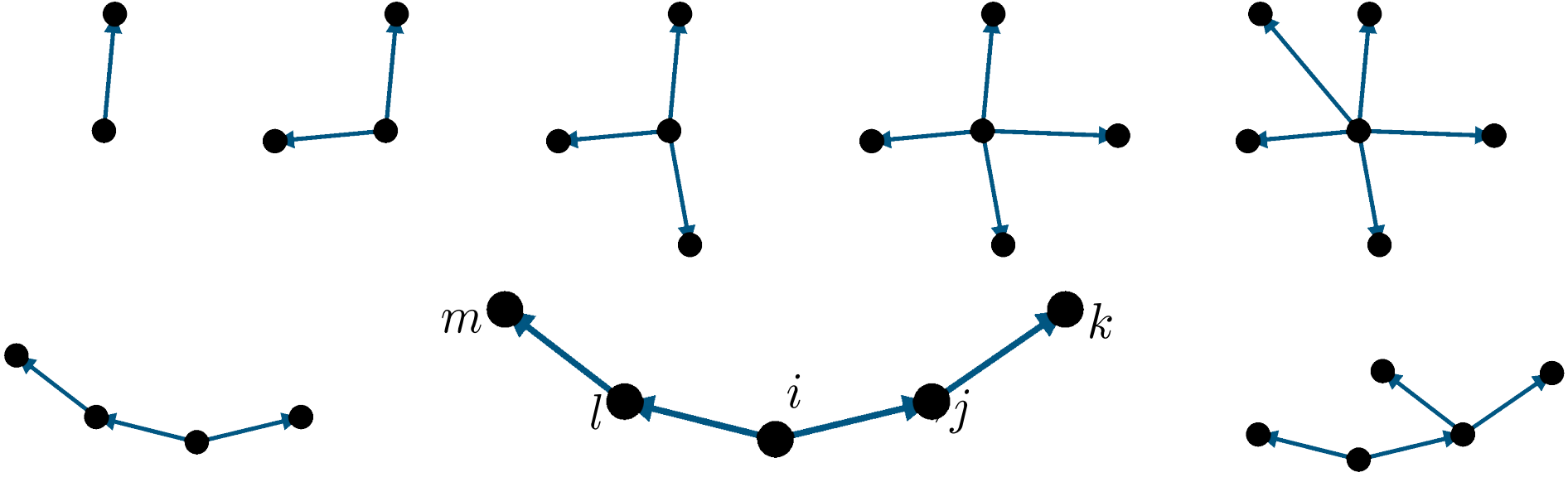
1 layer star graph



2 layers tree graph

Graph atomic cluster expansion (GRACE)

- Incorporate tree graphs in addition to star graphs



$$\Phi_{\alpha \mathbf{v}} = \phi_{i v_1}(\boldsymbol{r}_{ji}) \phi_{j v_2}(\boldsymbol{r}_{kj}) \phi_{i v_3}(\boldsymbol{r}_{li}) \phi_{l v_4}(\boldsymbol{r}_{ml})$$

Graph atomic cluster expansion (GRACE)

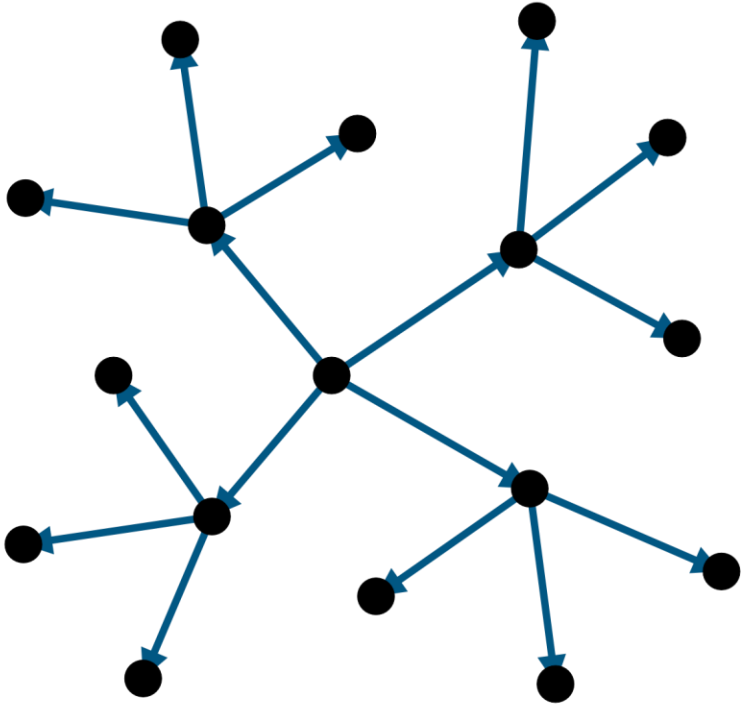
Multi-atom basis function

$$\int \Phi^*_{\alpha\mathbf{v}}(\boldsymbol{\sigma})\Phi_{\beta\mathbf{v}'}(\boldsymbol{\sigma})\, d\boldsymbol{\sigma} = \delta_{\alpha\beta}\delta_{\mathbf{v}\mathbf{v}'}$$

orthonormal

$$\sum_{\alpha\mathbf{v}} \Phi^*_{\alpha\mathbf{v}}(\boldsymbol{\sigma})\Phi_{\alpha\mathbf{v}}(\boldsymbol{\sigma}') = \delta(\boldsymbol{\sigma} - \boldsymbol{\sigma}')$$

complete



Cluster expansion

$$E = \sum_{\alpha\mathbf{v}} J_{\alpha\mathbf{v}}\Phi_{\alpha\mathbf{v}}$$

expansion coefficients

Bochkarev, Lysogorskiy, Drautz, Phys. Rev. X 14 (2024) 021036

Graph atomic cluster expansion (GRACE)

Multi-atom basis function

$$\int \Phi^*_{\alpha\mathbf{v}}(\boldsymbol{\sigma})\Phi_{\beta\mathbf{v}'}(\boldsymbol{\sigma})\, d\boldsymbol{\sigma} = \delta_{\alpha\beta}\delta_{\mathbf{v}\mathbf{v}'}$$

orthonormal

$$\sum_{\alpha\mathbf{v}} \Phi^*_{\alpha\mathbf{v}}(\boldsymbol{\sigma})\Phi_{\alpha\mathbf{v}}(\boldsymbol{\sigma}') = \delta(\boldsymbol{\sigma}-\boldsymbol{\sigma}')$$

complete

$\mathrm{CPU\ time} \propto N^{|\boldsymbol{v}|}$ — leading order; number of neighbors

$\mathrm{CPU\ time} \propto N$ — fast ✓; high body-order ✓

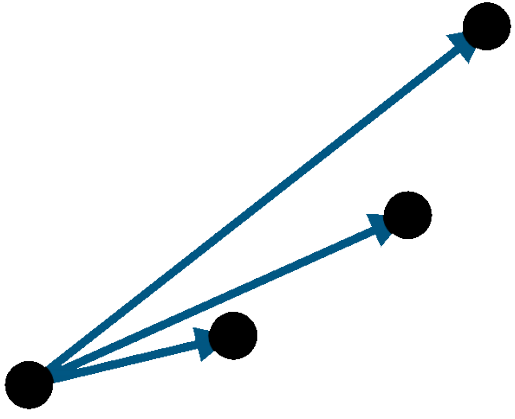
Cluster expansion

$$E = \sum_{\alpha\mathbf{v}} J_{\alpha\mathbf{v}}\Phi_{\alpha\mathbf{v}}$$

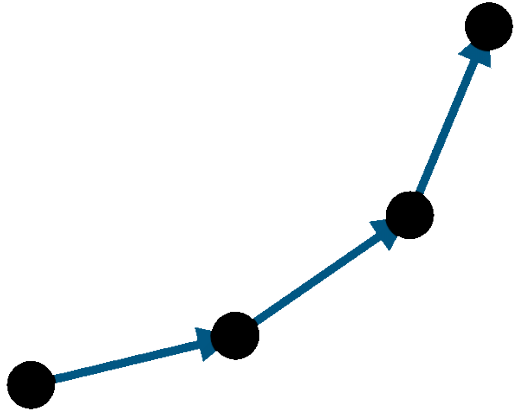
$J_{\alpha\mathbf{v}}$: expansion coefficients

Bochkarev, Lysogorskiy, Drautz, Phys. Rev. X 14 (2024) 021036

Completeness and sensitivity



Star (direct/local)



Tree (indirect/semilocal)

Competing graphs:

- Different sensitivity
- Different chemistry

Incorporating several topologies for sensitivity and chemical interpretation

Interactions from Quantum Mechanics

Schrödinger equation:

$$(\hat{H} - E)|\psi\rangle = 0$$

Green's function:

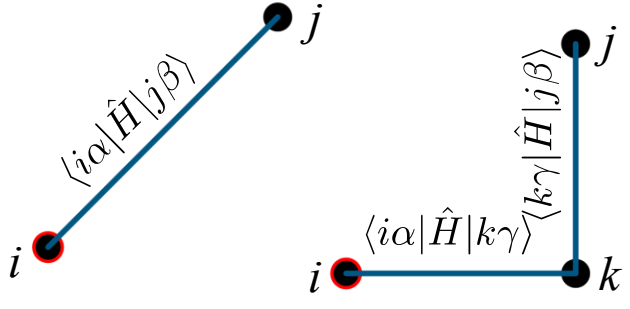
$$\hat{G}(E) = (E - \hat{H})^{-1}$$

$$\hat{G}(E) \approx a_0(E) + a_1(E)\hat{H} + a_2(E)\hat{H}^2 + a_3(E)\hat{H}^3 + \ldots$$

Matrix elements with atom-centred basis:

$\langle i\alpha|\hat{H}|j\beta\rangle$ Typically short-ranged

$$\langle i\alpha|\hat{H}^2|j\beta\rangle = \sum_{k\gamma}\langle i\alpha|\hat{H}|k\gamma\rangle\langle k\gamma|\hat{H}|j\beta\rangle$$



Interactions from Quantum Mechanics

Schrödinger equation:

$$(\hat{H} - E)|\psi\rangle = 0$$

Green's function:

$$\hat{G}(E) = (E - \hat{H})^{-1}$$

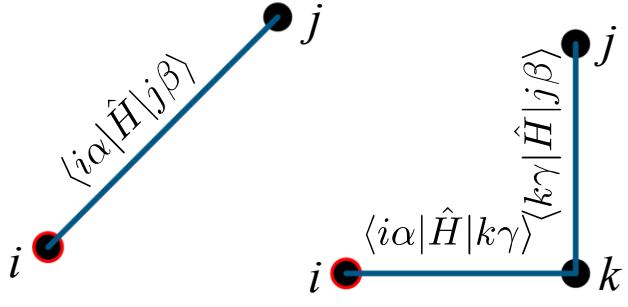
$$= a_0(E)\mathbb{1} + a_1(E)\hat{H} + a_2(E)\hat{H}^2 + a_3(E)\hat{H}^3 + \ldots$$

Direct and indirect/semilocal interactions in quantum mechanics

Matrix elements with atom-centred basis:

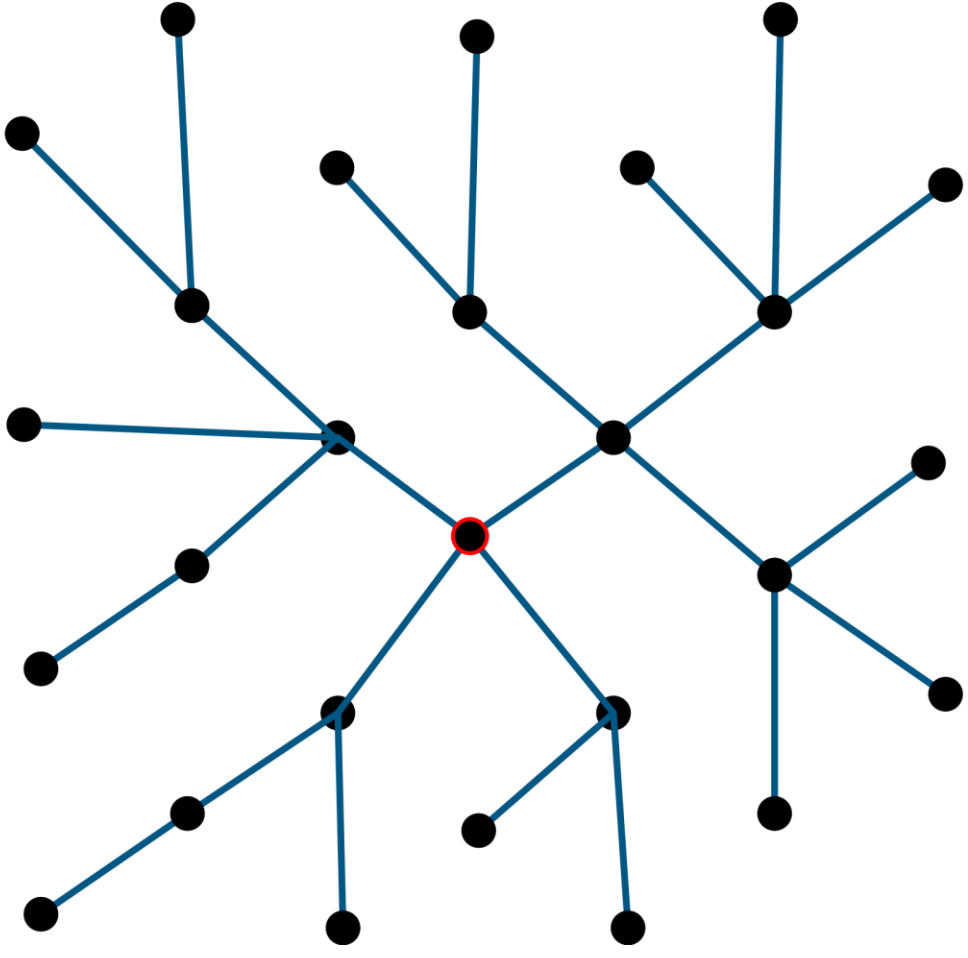
$\langle i\alpha|\hat{H}|j\beta\rangle$ Typically short-ranged

$$\langle i\alpha|\hat{H}^2|j\beta\rangle = \sum_{k\gamma}\langle i\alpha|\hat{H}|k\gamma\rangle\langle k\gamma|\hat{H}|j\beta\rangle$$



4. Efficient evaluation

Efficient evaluation of GRACE

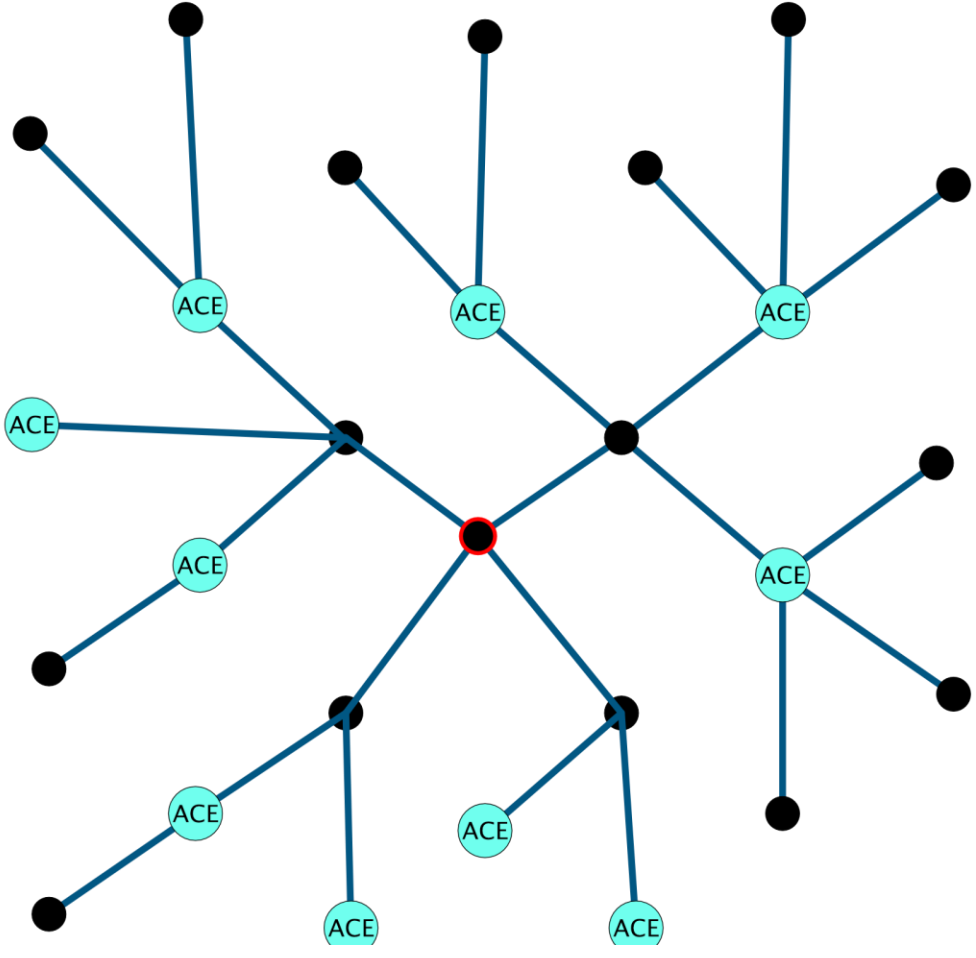


ACE is a complete expansion

➔ Transform local atomic environment to ACE and reduce reach of graph by one layer

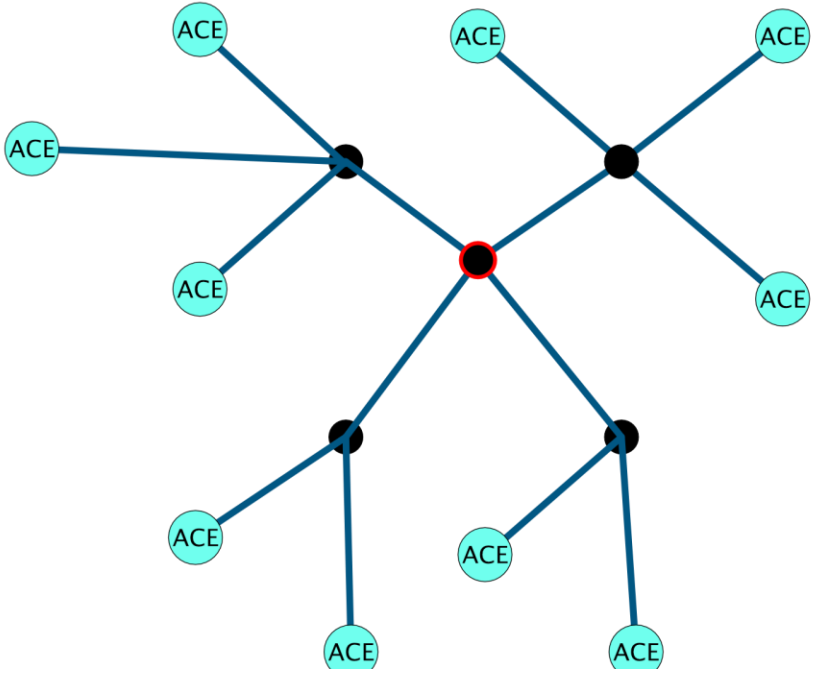
➔ Repeat until graph is reduced

Recursive evaluation



local ACE on layer 2

Recursive evaluation

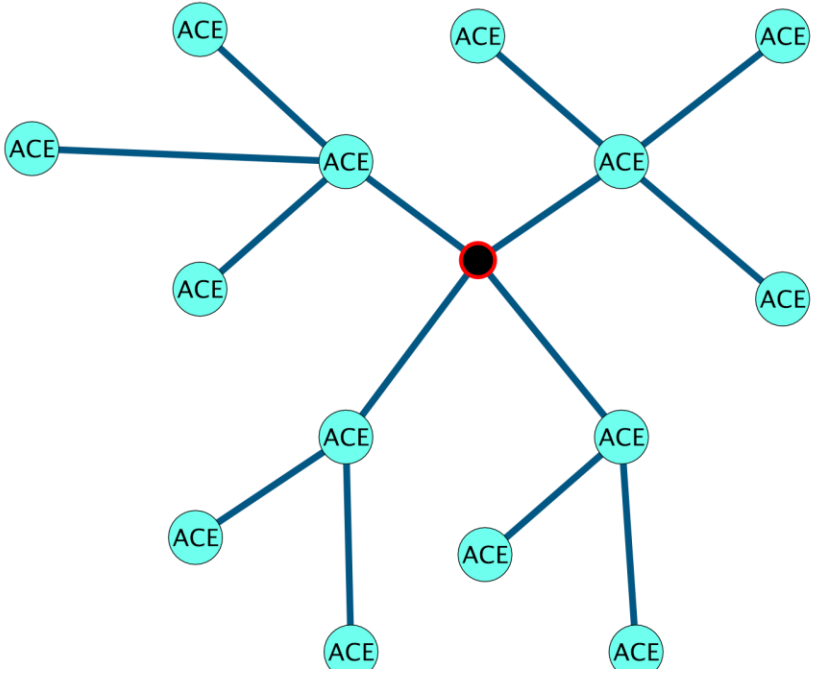


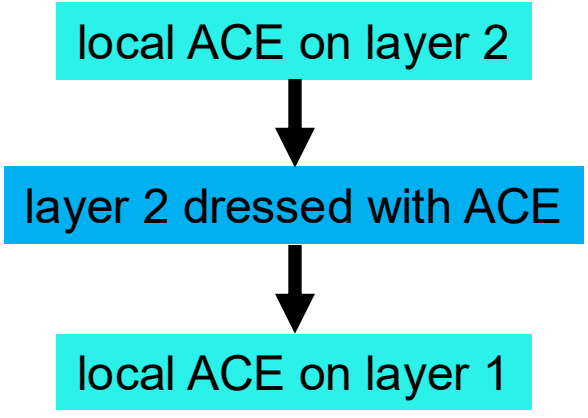
local ACE on layer 2

↓

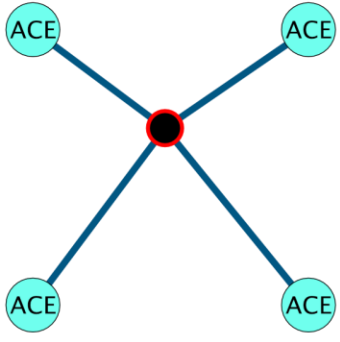
layer 2 dressed with ACE

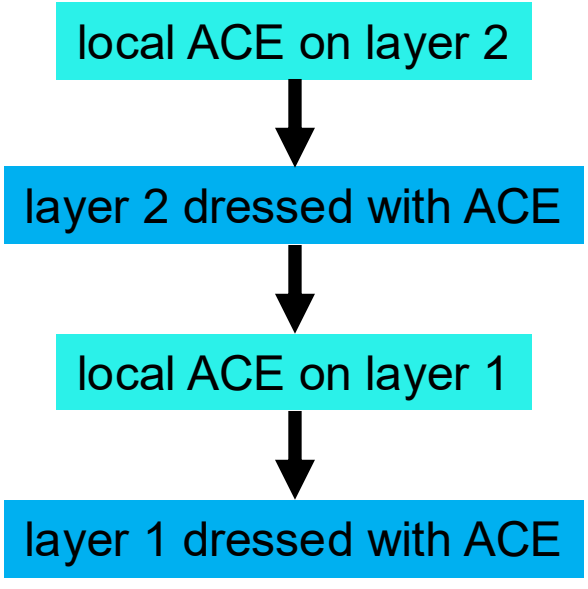
Recursive evaluation



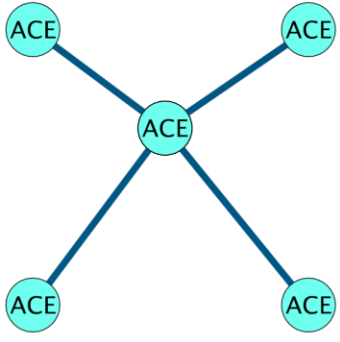


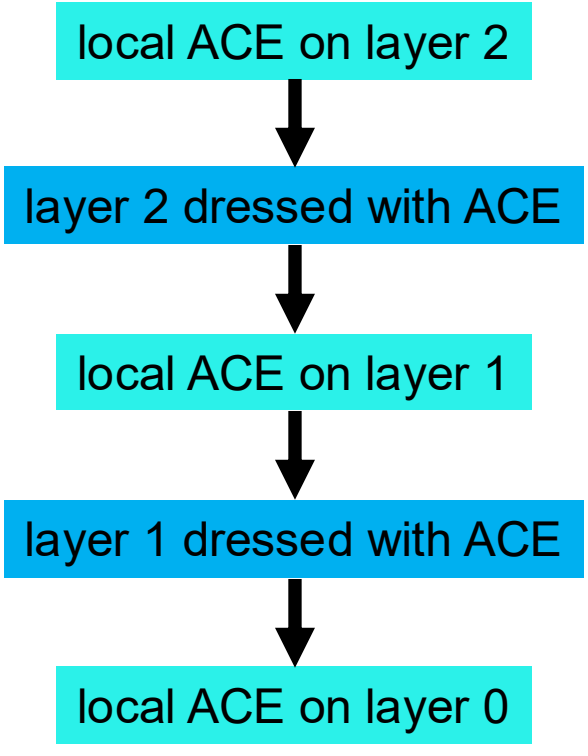
Recursive evaluation



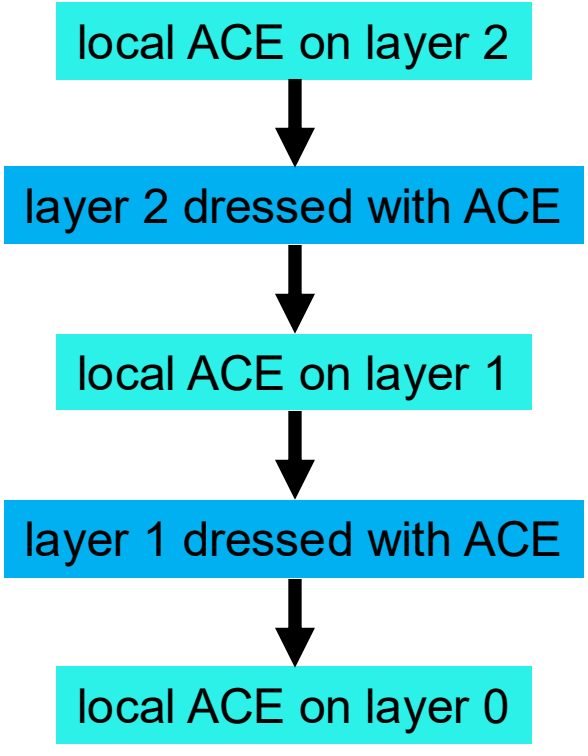


Recursive evaluation





Recursive evaluation

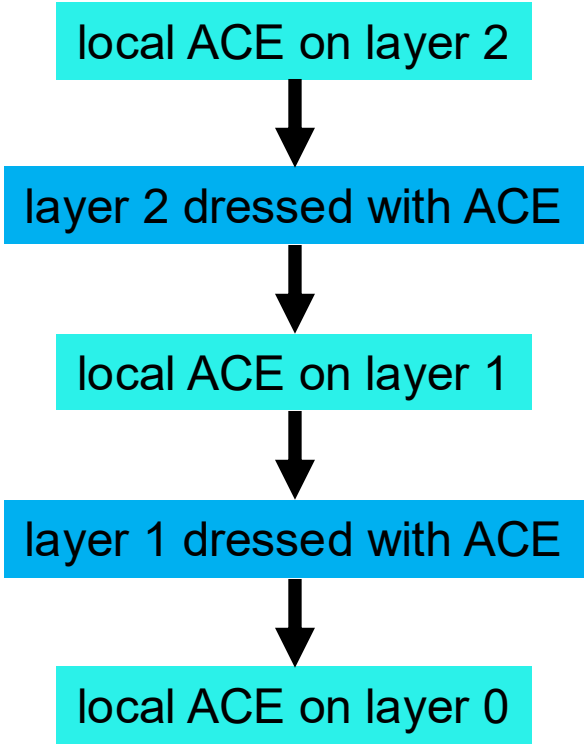


Recursive evaluation

- **Recursive evaluation incorporates all subgraphs**

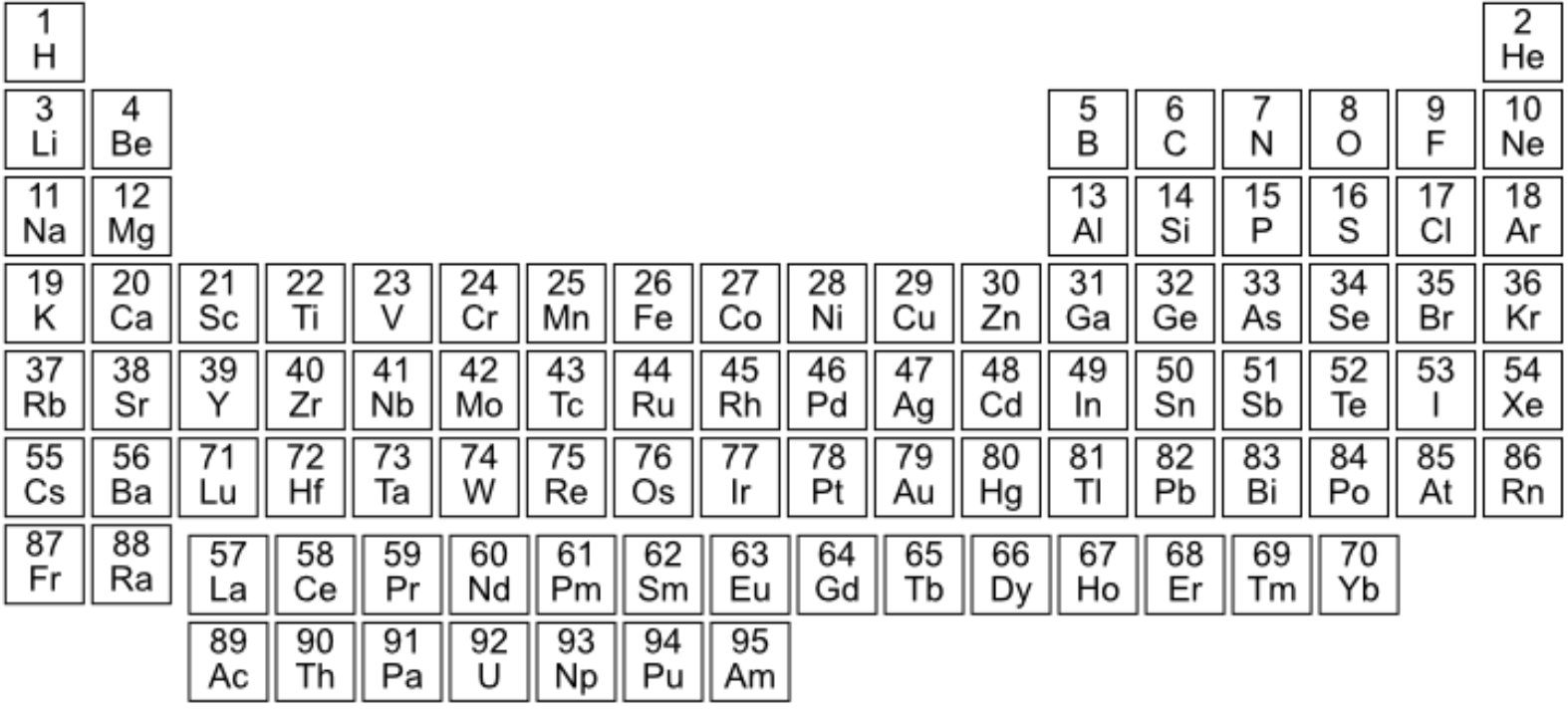
- **Recursive evaluation may be understood as message passing**

➔ Link to message passing (MACE, NequIP, …) and fast multipole method



Batzner et al, Nat. Comm. (2022), Batatia et al, NeurIPS (2022), Multi ACE, Batatia et al, Nat. Mach. Intel. (2025)

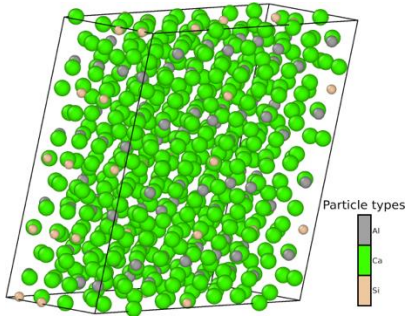
5. Foundational GRACE interatomic potential



GRACE foundation model

- Train on (nearly) all elements of the periodic table

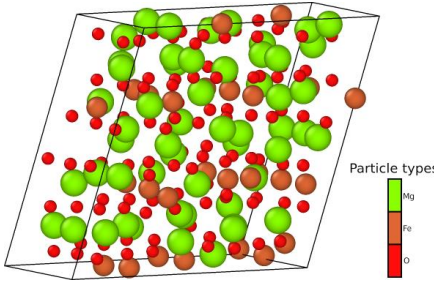
- Alexandria database (https://alexandria.icams.rub.de/)
- Materials project (https://next-gen.materialsproject.org/)
- OMAT24 dataset (about 118 million structures)



➔ Tremendous improvements over the past 1-2 years

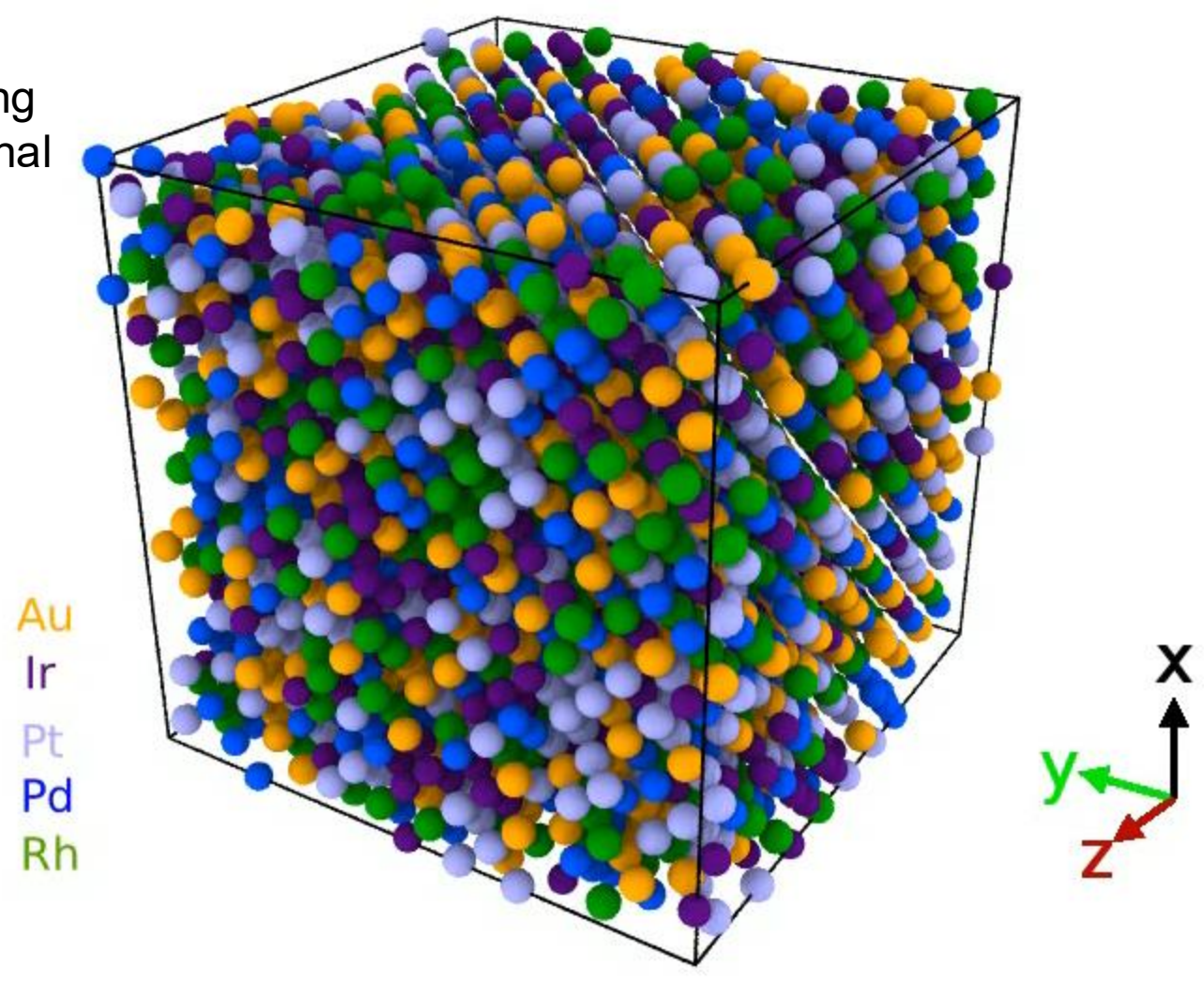
How can this work at all?

- Properties of chemical elements are strongly correlated
- Fewer than 10 dimensions required for all elements



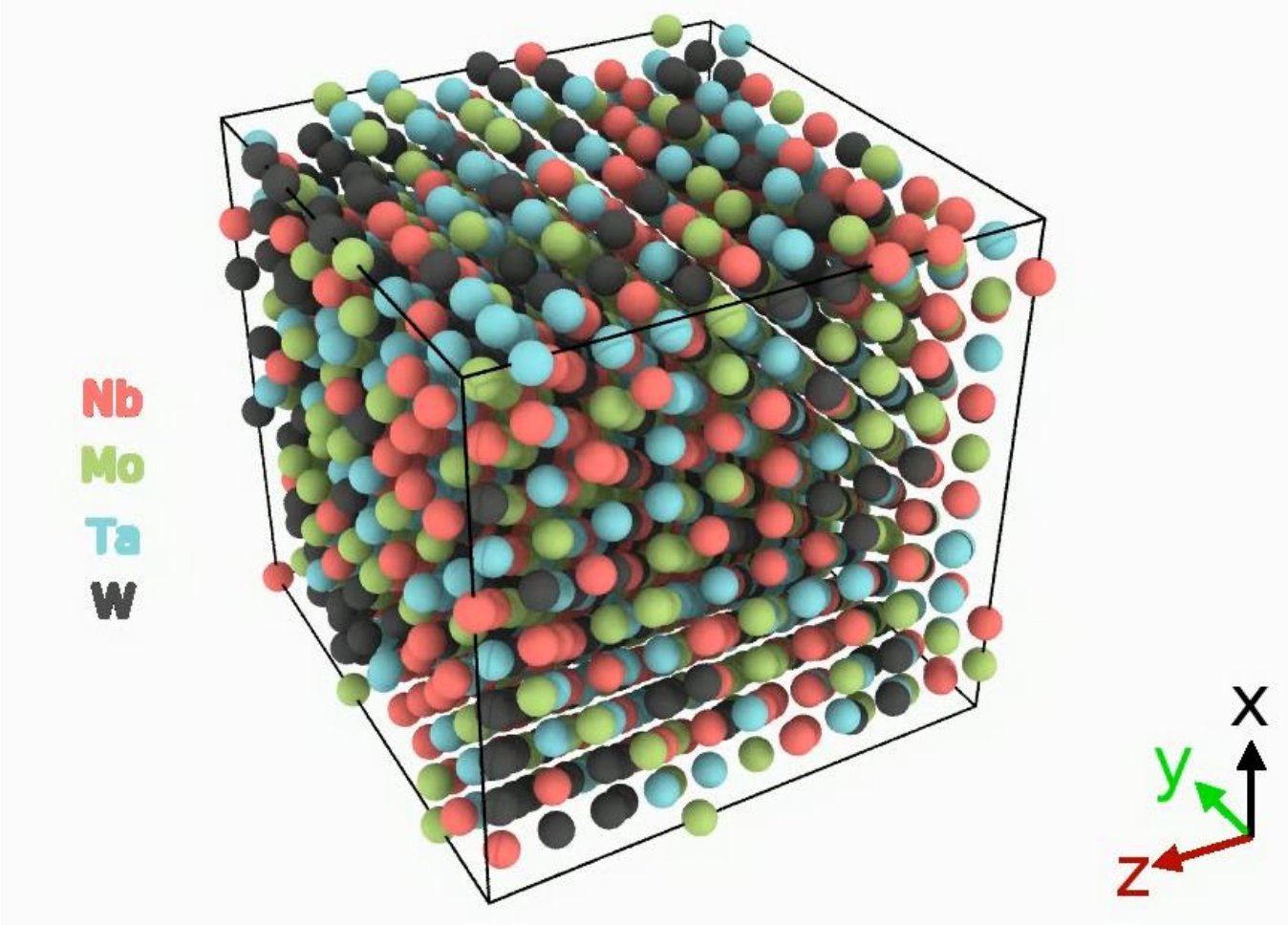
Phase separation in AuIrPtPdRh

Part of CRC 1625
Atomic-scale understanding and design of multifunctional compositionally complex solid solution surfaces



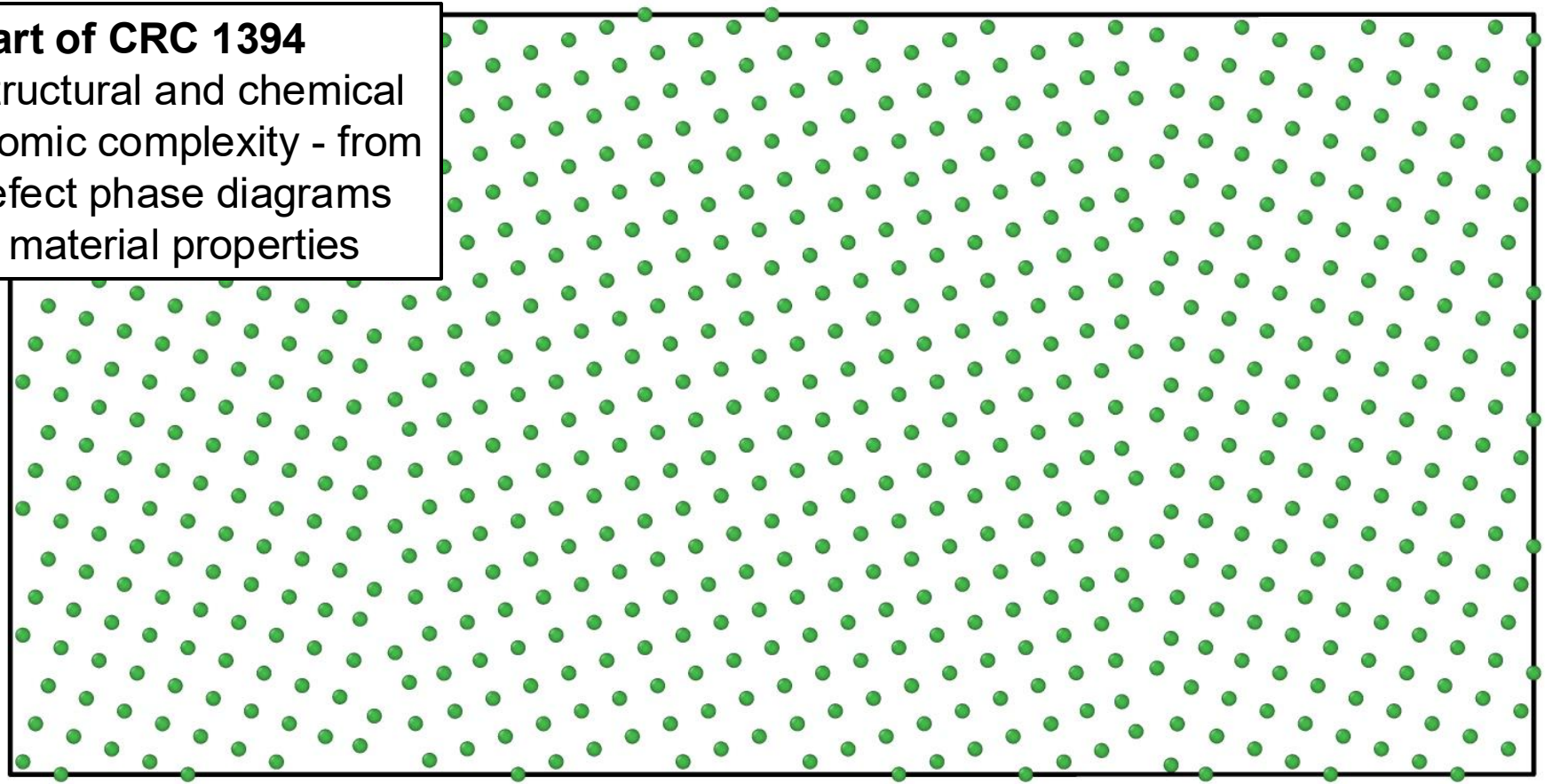
ICAMS

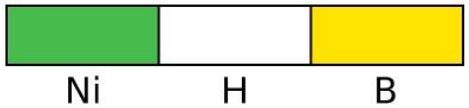
Example: NbMoTaW



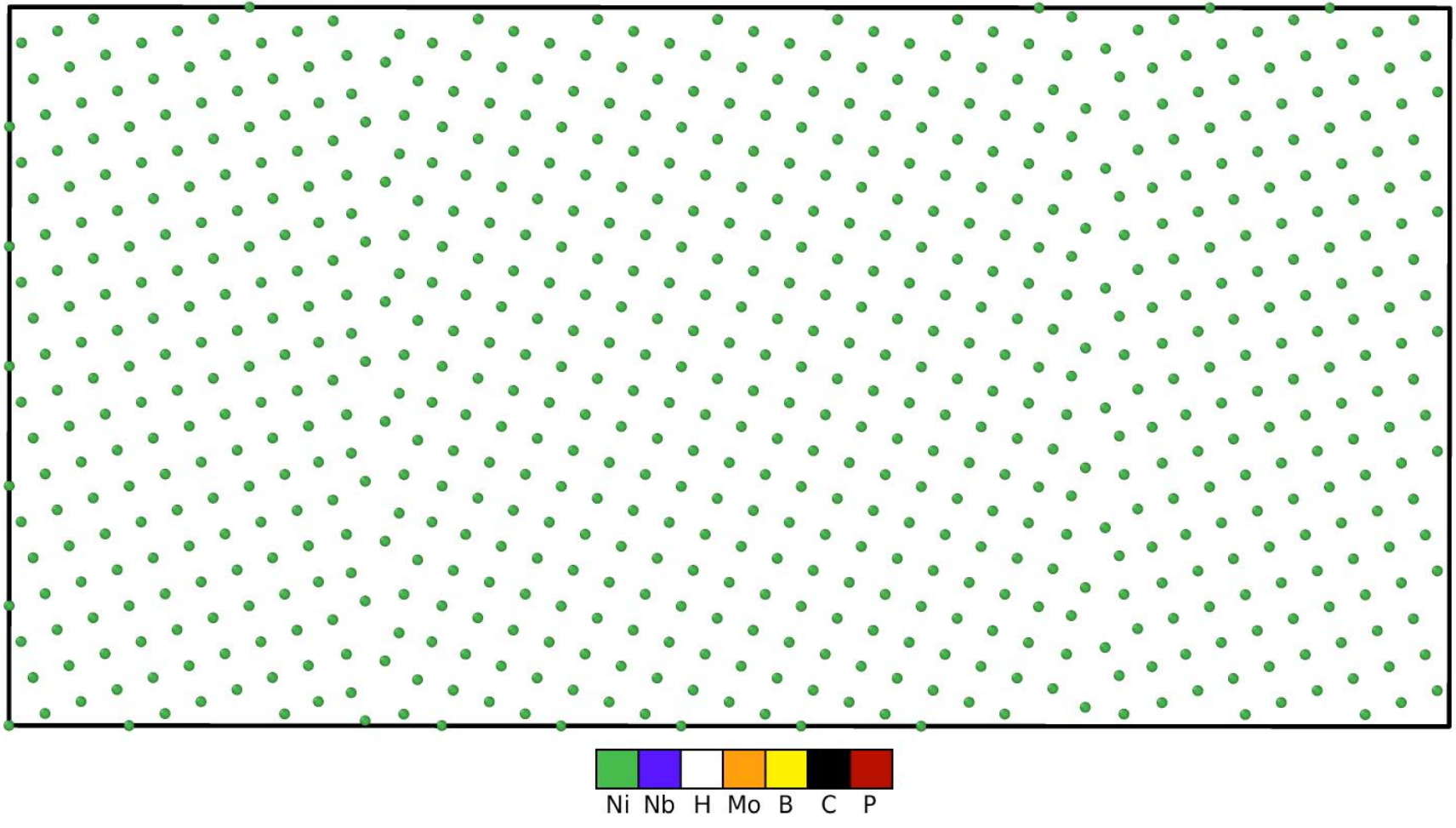
Grain boundary segregation in nickel

Part of CRC 1394
Structural and chemical atomic complexity - from defect phase diagrams to material properties

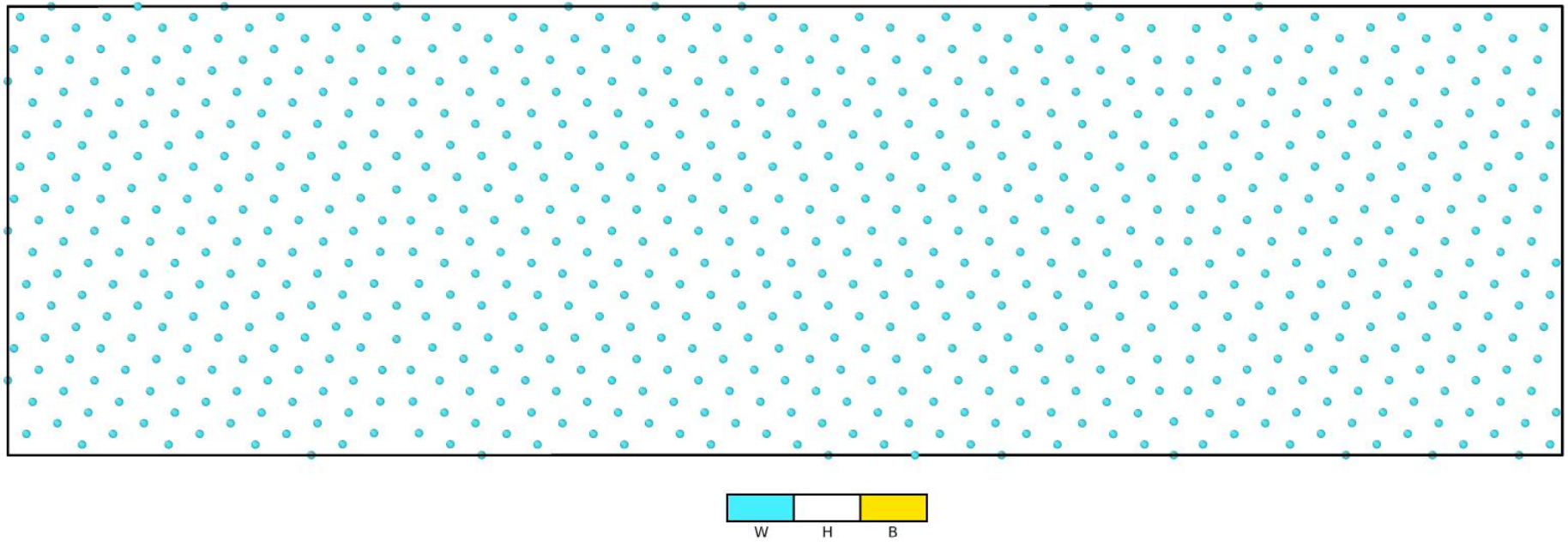




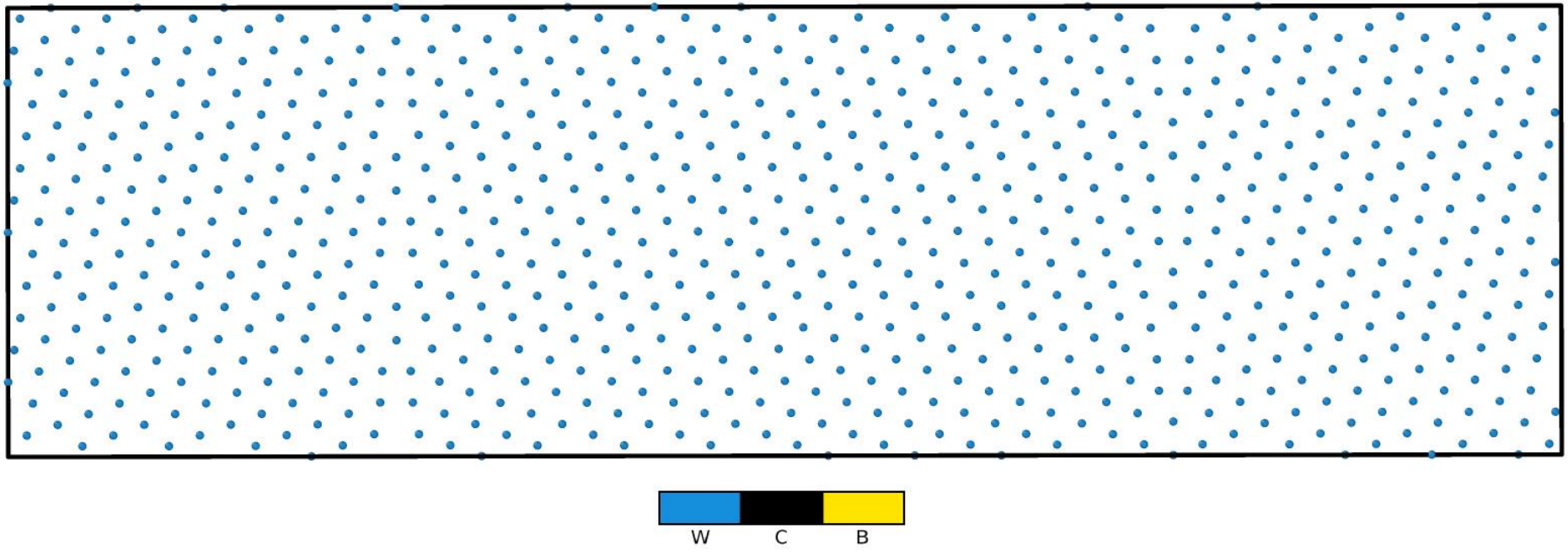
Grain boundary segregation in nickel



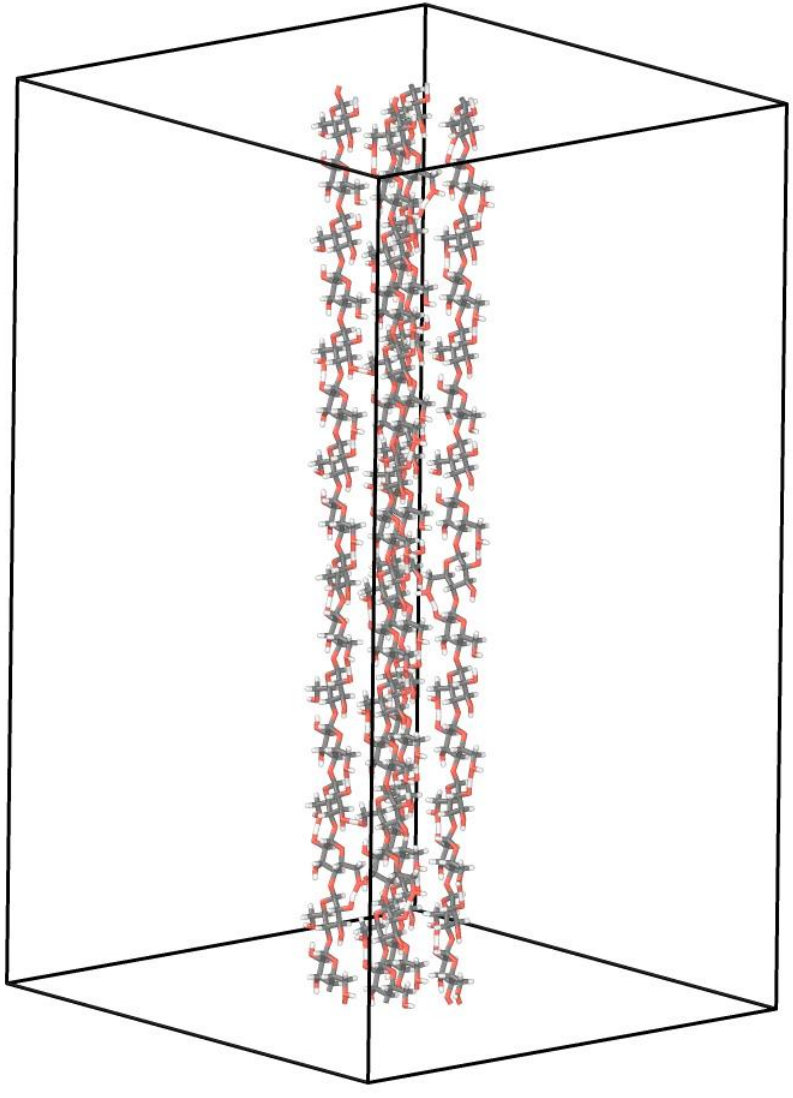
Grain boundary segregation in tungsten



Grain boundary segregation in tungsten



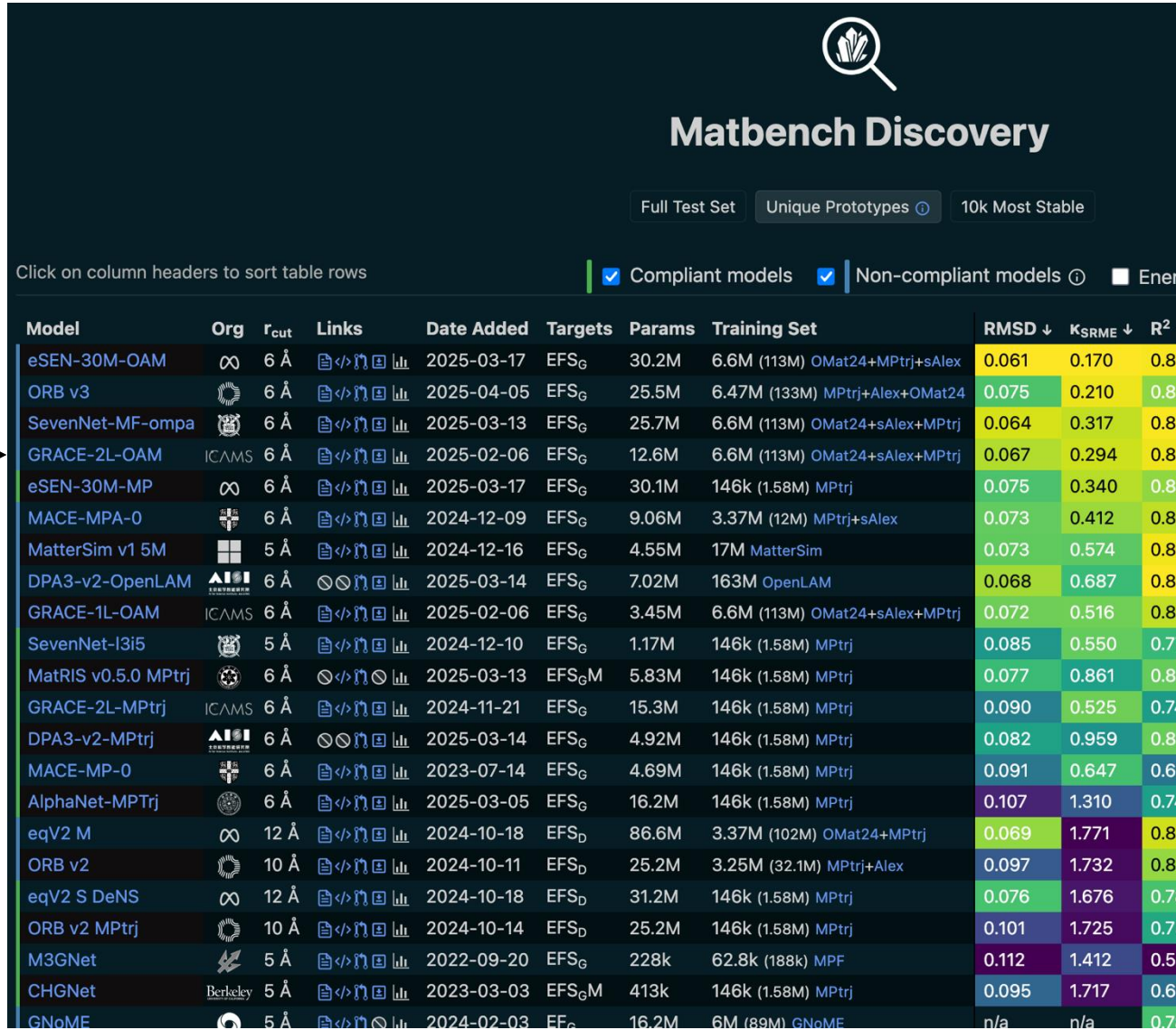
Burning cellulose



Matbench discovery leaderboard

- **GRACE training less than 1000 GPU h**
- **GRACE about 10x faster than some competing models**

Snapshot 27 May 2025



Model	Org	r_{cut}	Links	Date Added	Targets	Params	Training Set	RMSD ↓	κ_{SRME} ↓	R^2
eSEN-30M-OAM		6 Å		2025-03-17	EFS_G	30.2M	6.6M (113M) OMat24+MPtrj+sAlex	0.061	0.170	0.8
ORB v3		6 Å		2025-04-05	EFS_G	25.5M	6.47M (133M) MPtrj+Alex+OMat24	0.075	0.210	0.8
SevenNet-MF-ompa		6 Å		2025-03-13	EFS_G	25.7M	6.6M (113M) OMat24+sAlex+MPtrj	0.064	0.317	0.8
GRACE-2L-OAM	ICAMS	6 Å		2025-02-06	EFS_G	12.6M	6.6M (113M) OMat24+sAlex+MPtrj	0.067	0.294	0.8
eSEN-30M-MP		6 Å		2025-03-17	EFS_G	30.1M	146k (1.58M) MPtrj	0.075	0.340	0.8
MACE-MPA-0		6 Å		2024-12-09	EFS_G	9.06M	3.37M (12M) MPtrj+sAlex	0.073	0.412	0.8
MatterSim v1 5M		5 Å		2024-12-16	EFS_G	4.55M	17M MatterSim	0.073	0.574	0.8
DPA3-v2-OpenLAM		6 Å		2025-03-14	EFS_G	7.02M	163M OpenLAM	0.068	0.687	0.8
GRACE-1L-OAM	ICAMS	6 Å		2025-02-06	EFS_G	3.45M	6.6M (113M) OMat24+sAlex+MPtrj	0.072	0.516	0.8
SevenNet-l3i5		5 Å		2024-12-10	EFS_G	1.17M	146k (1.58M) MPtrj	0.085	0.550	0.7
MatRIS v0.5.0 MPtrj		6 Å		2025-03-13	EFS_GM	5.83M	146k (1.58M) MPtrj	0.077	0.861	0.8
GRACE-2L-MPtrj	ICAMS	6 Å		2024-11-21	EFS_G	15.3M	146k (1.58M) MPtrj	0.090	0.525	0.7
DPA3-v2-MPtrj		6 Å		2025-03-14	EFS_G	4.92M	146k (1.58M) MPtrj	0.082	0.959	0.8
MACE-MP-0		6 Å		2023-07-14	EFS_G	4.69M	146k (1.58M) MPtrj	0.091	0.647	0.6
AlphaNet-MPTrj		6 Å		2025-03-05	EFS_G	16.2M	146k (1.58M) MPtrj	0.107	1.310	0.7
eqV2 M		12 Å		2024-10-18	EFS_D	86.6M	3.37M (102M) OMat24+MPtrj	0.069	1.771	0.8
ORB v2		10 Å		2024-10-11	EFS_D	25.2M	3.25M (32.1M) MPtrj+Alex	0.097	1.732	0.8
eqV2 S DeNS		12 Å		2024-10-18	EFS_D	31.2M	146k (1.58M) MPtrj	0.076	1.676	0.7
ORB v2 MPtrj		10 Å		2024-10-14	EFS_D	25.2M	146k (1.58M) MPtrj	0.101	1.725	0.7
M3GNet		5 Å		2022-09-20	EFS_G	228k	62.8k (188k) MPF	0.112	1.412	0.5
CHGNet	Berkeley	5 Å		2023-03-03	EFS_GM	413k	146k (1.58M) MPtrj	0.095	1.717	0.6
GNoME		5 Å		2024-02-03	EF_G	16.2M	6M (89M) GNoME	n/a	n/a	0.7

https://matbench-discovery.materialsproject.org/

Conclusions

- Foundational GRACE interatomic potential
- Fast and accurate
- Exhaustive databases available, but still somewhat limited

- Models, code, tutorials: https://gracemaker.readthedocs.io