

On-lattice interatomic models

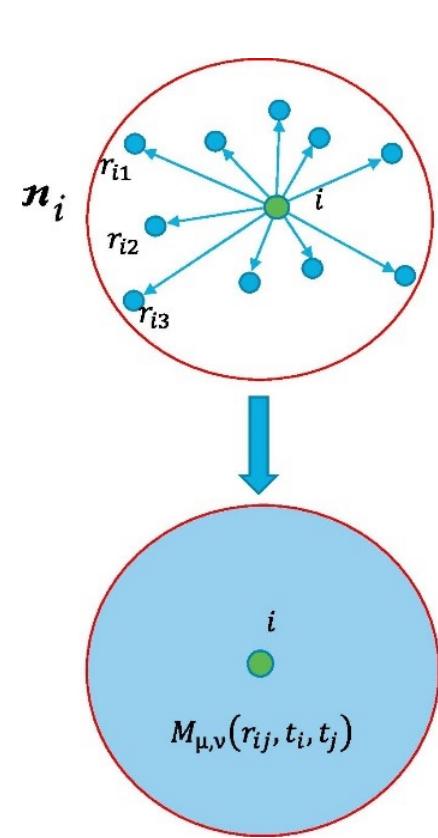
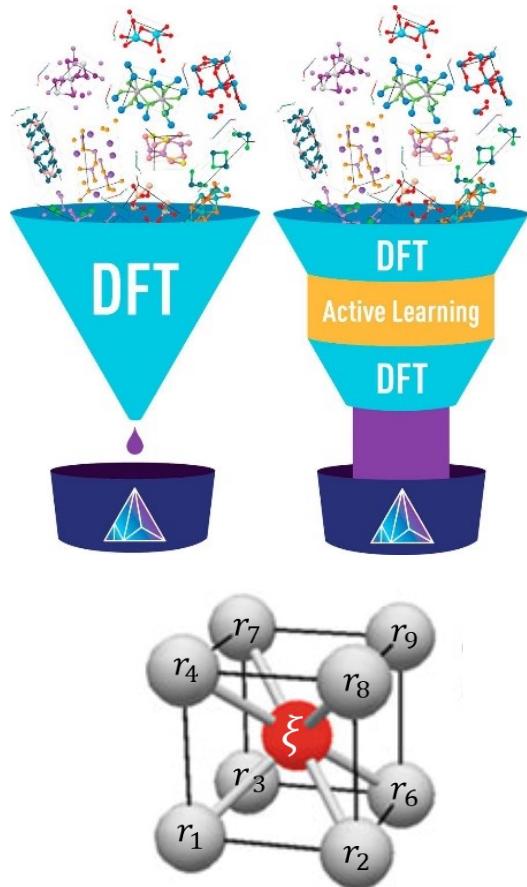
Speakers: Tatiana Kostiuchenko
Vadim Sotskov

Group leader: Alexander Shapeev

Skoltech Multiscale Modelling



MTP
Moment tensor potential



**Малоранговый машинно-обучаемый потенциал
LRP**
Low-rank interatomic potential

OFF -lattice

ON -lattice

Lecture content

1. The difference between **off-** and **on-lattice** models
2. Why on-lattice models are actual
3. Monte Carlo methods
4. Commonly used on-lattice models:
 - Cluster expansion method
 - Generalized perturbation method
 - Low-rank interatomic potential
5. Frontier problems solved by these methods
6. About Cython and LRP-code applications

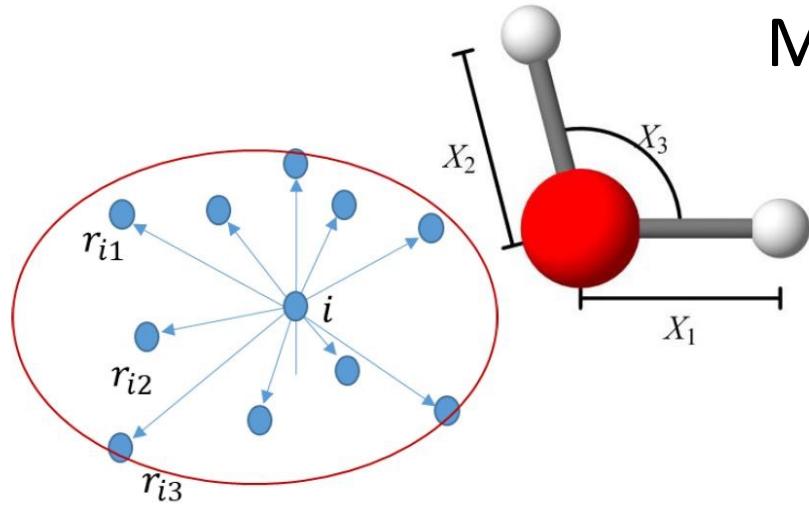
1. The difference between **off- and on-lattice** models

The difference between off- and on-lattice models



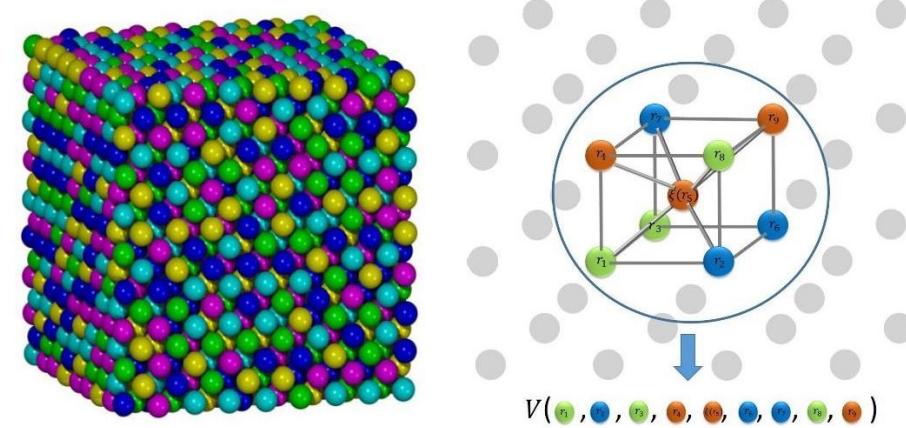
Can account for:
Stresses and forces

Magnetic moments
Many-body interactions



Can account for:
Local distortions

- Fixed crystalline lattice
Without the possibility of
- account for explicit stresses

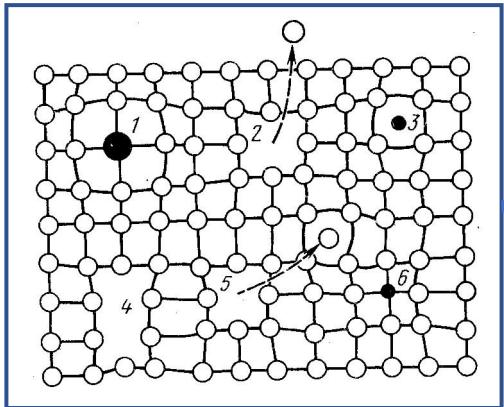


2. Why on-lattice models are actual

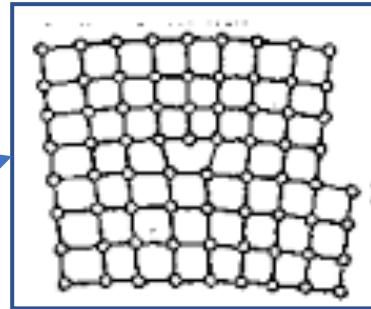
Why on-lattice models are actual

Multicomponent alloys

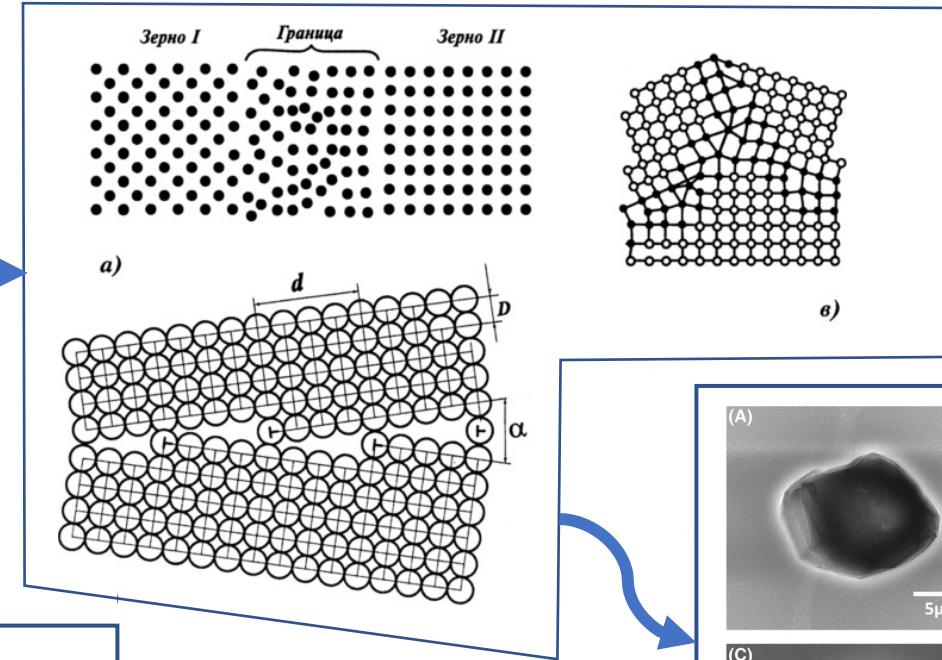
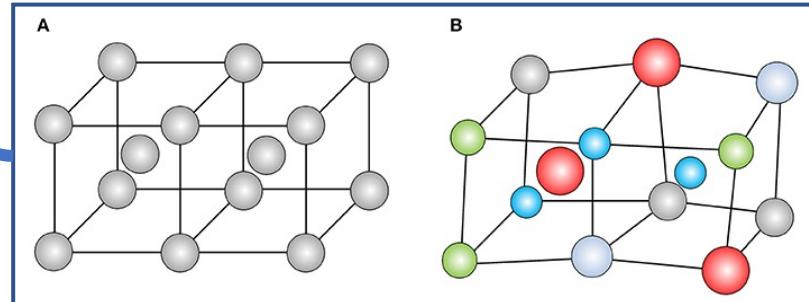
Slowing down the diffusion of point defects



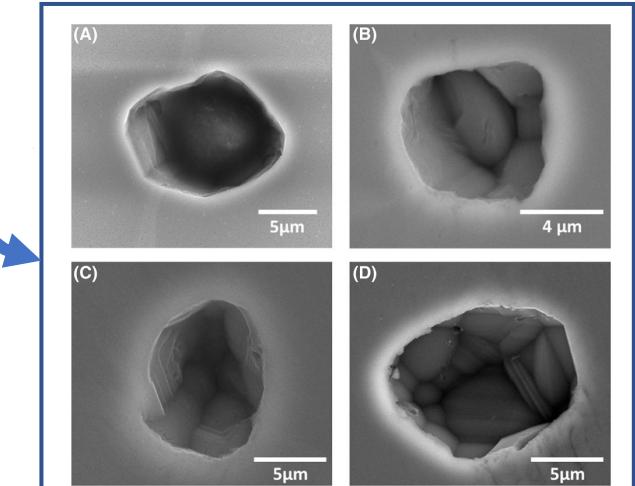
Dislocation movement



Crystal lattice distortion



Movement of grain boundaries



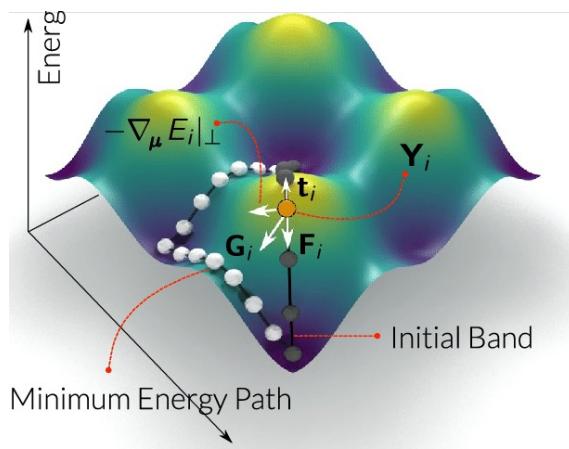
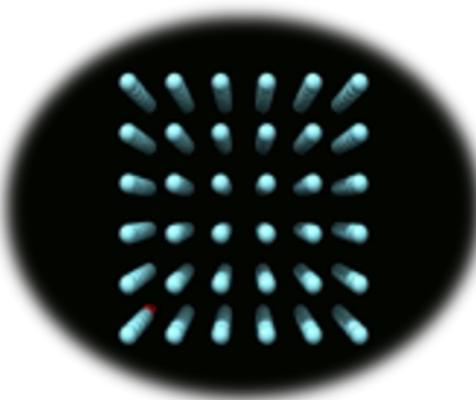
Formation of bulk defects

Why on-lattice models are actual

Multicomponent alloys

Molecular dynamics

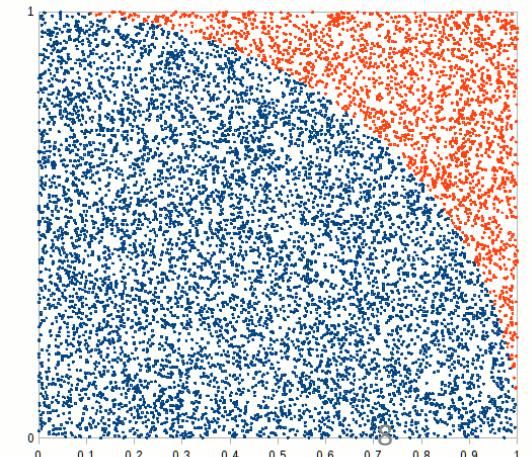
- relies on numerical solution of Newton's equations of motion
- reproduce the dynamics of a system



Monte-Carlo method

- relies on equilibrium statistical mechanics
- generates states according to appropriate Boltzmann distribution

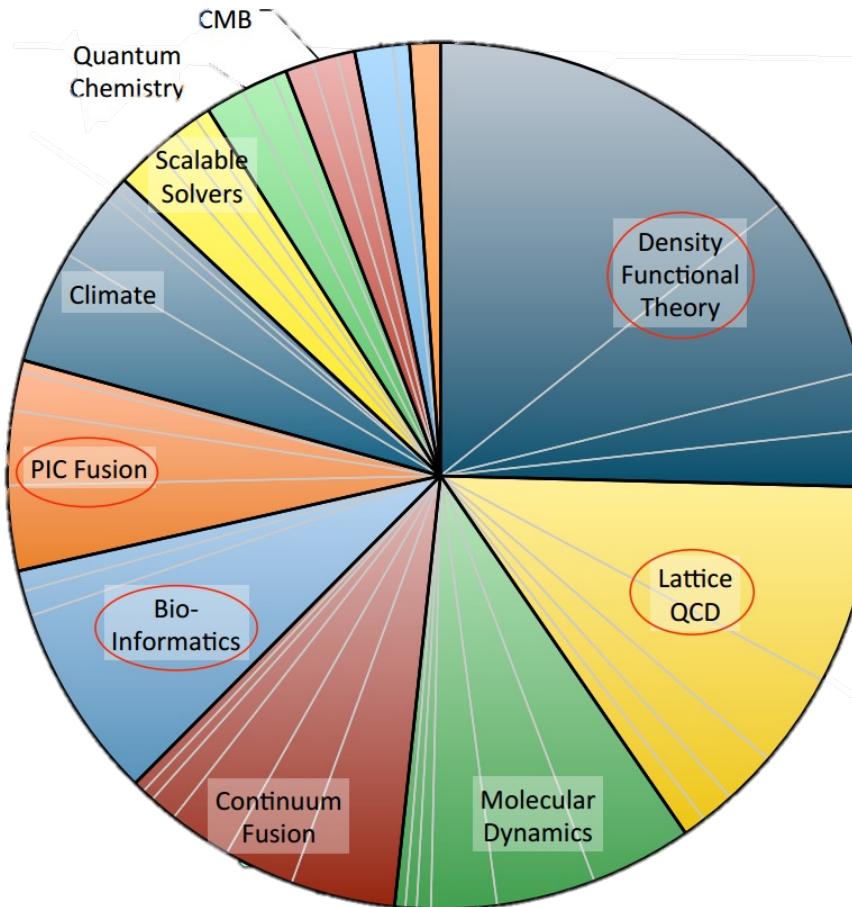
Ising model:
mathematical model of
Ferromagnetism
in statistical mechanics



Why on-lattice models are actual

Multicomponent alloys

Molecular dynamics (MD)



Monte Carlo method

3. Monte Carlo methods

Monte Carlo methods

The basics

A_i – the state of the system, a u_i – probability of being in this state

Mean value of thermodynamic function:

$$\bar{F} = \sum_{A_i} F(A_i) u_i, u_i \geq 0, \sum_i u_i = 1$$

Gibbs distribution:

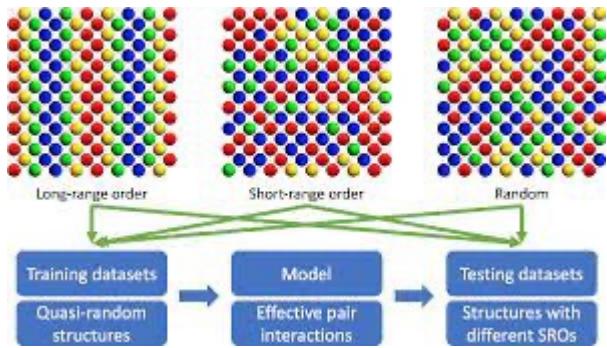
$$u_i = \frac{\exp(-\beta U_N(A_i))}{\sum_{A_j} \exp(-\beta U_N(A_j))},$$

$U_N(A_i)$ – energy of the system in state A_i

$$\beta = \frac{1}{k_B T}$$

Markov chain + Law of large numbers

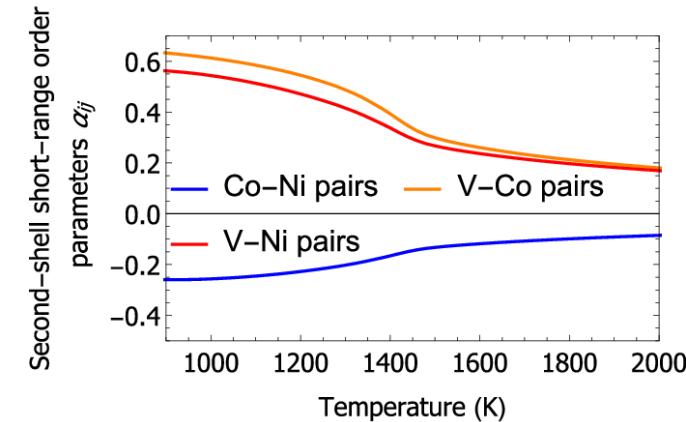
$$\bar{F} = \frac{1}{M} \sum_k^M F(A_{i_k}), A_{i_1}, \dots, A_{i_M} - \text{“events”}$$



Monte Carlo methods

Canonical Monte Carlo

1. We randomly **select the node** of crystalline lattice (with a probability $\frac{1}{N}$)
2. Randomly **select a neighbor** (neighboring node)
3. Calculate the **energy difference ΔE** before and after event.
The **probability** of the event is $p = \exp(-\beta\Delta E)$
4. Generate random number $\epsilon \in [0; 1]$ and **compare** with p .
If $p > \epsilon$, then the event is “accepted”
5. Calculate **thermodynamic functions** for new state, **updated statistics**



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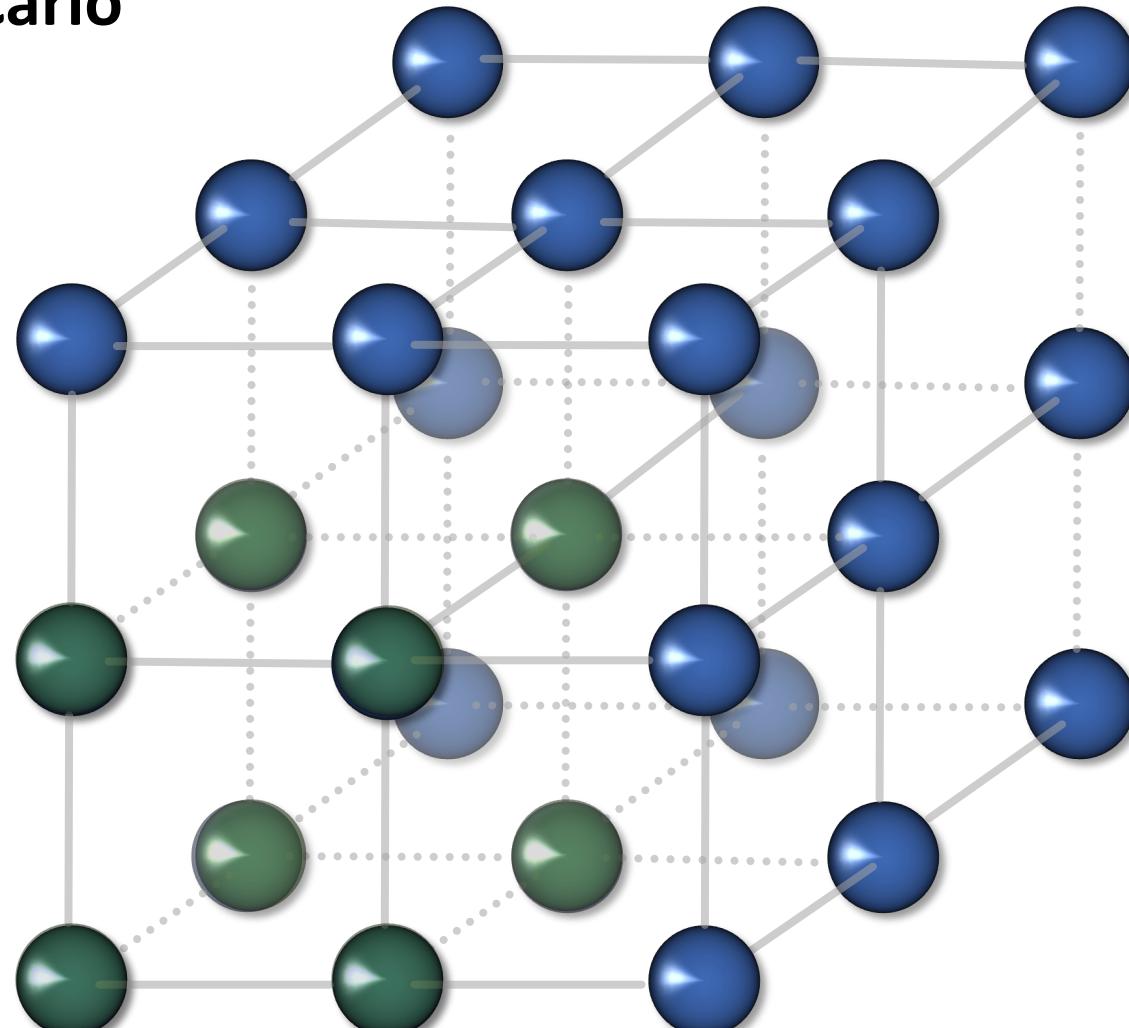
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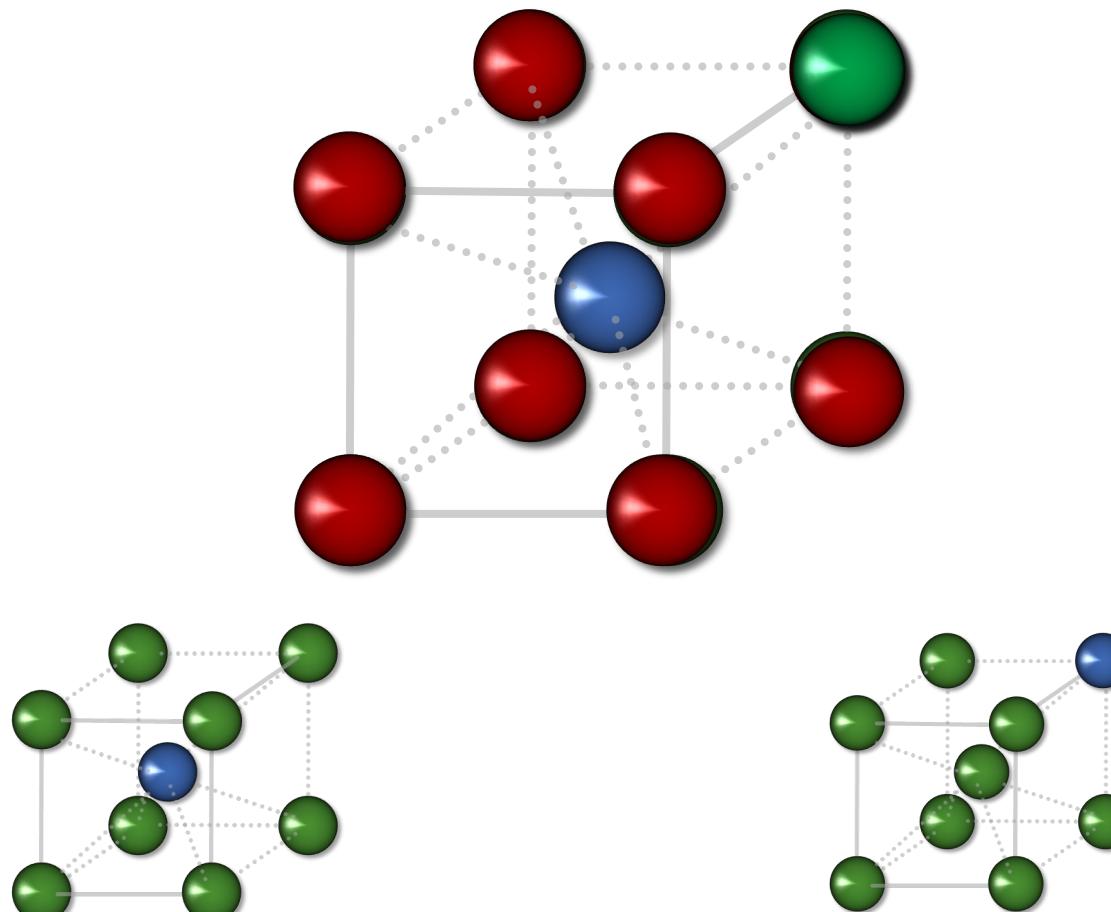
If $p > \epsilon$, then the event is “accepted”

IF NOT

5. Calculate **thermodynamic functions** for new state,
updated statistics

$$\Delta E = E_1 - E_2$$

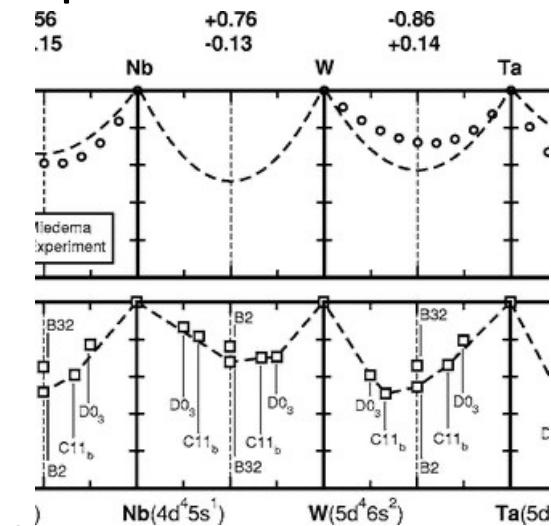
$$\rightarrow E_{current} += \Delta E \text{ or } 0$$



Monte Carlo methods

Grand Canonical (μVT) Monte Carlo

1. We randomly **select the node** of crystalline lattice (with a probability $\frac{1}{N}$)
2. Randomly **select another chemical element** (from the system) for replacement
3. Calculate the **energy difference ΔE** before and after event
The **probability** of the event is $p = \exp(\mu N - \beta \Delta E)$
4. Generate random **number $\epsilon \in [0; 1]$** and **compare** with p .
If $p > \epsilon$, then the event is “accepted”
5. Calculate **thermodynamic functions** for new state, **updated statistics**



macrocanonical ensemble = thermodynamic equilibrium (thermal and chemical)

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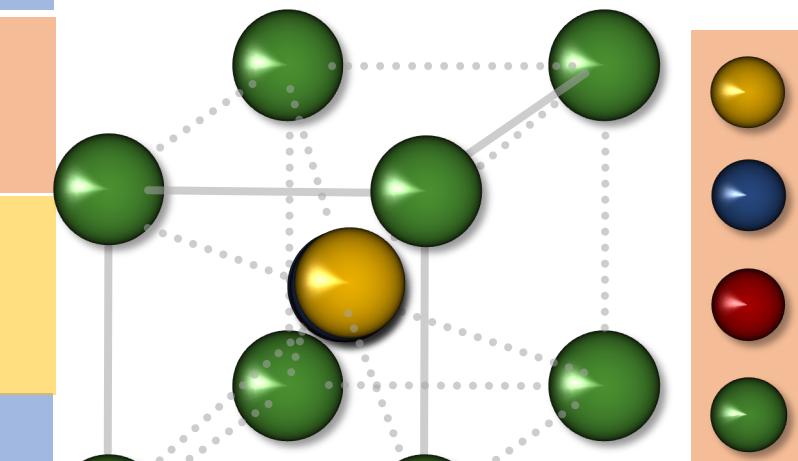
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5. Calculate **thermodynamic functions** for new state, **updated statistics**

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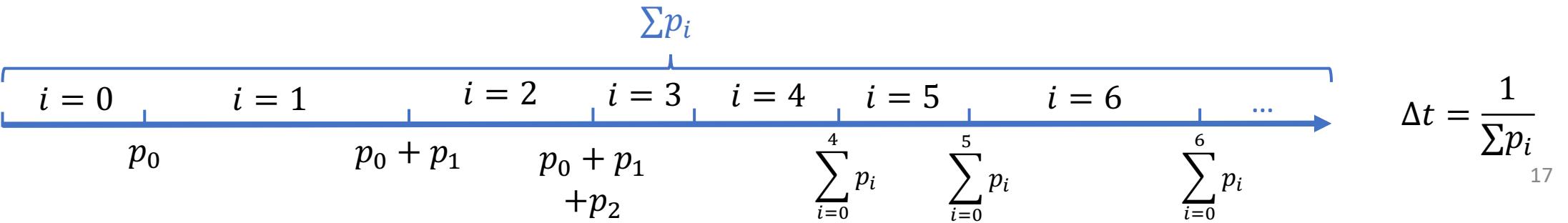
$$\Delta E = E_1 - E_2 \\ \rightarrow E_{current} += \Delta E \text{ or } 0$$



Monte Carlo methods

Kinetic Monte Carlo

1. $t = 0$, start with initial state k
 2. Make a list of states i that can be obtained from k with one event step
 3. Calculate the common rate as a $\sum p_i$, where p_i is a probability of k transfer to i
- Thus we get the sum of probabilities.
4. Generate random number $\xi \in [0; 1]$. Compare $\xi \cdot \sum p_i$ and detect which state i we come to with an event
 5. Calculate **thermodynamic functions** for new state, **updated statistics**, $t = t + \Delta t$



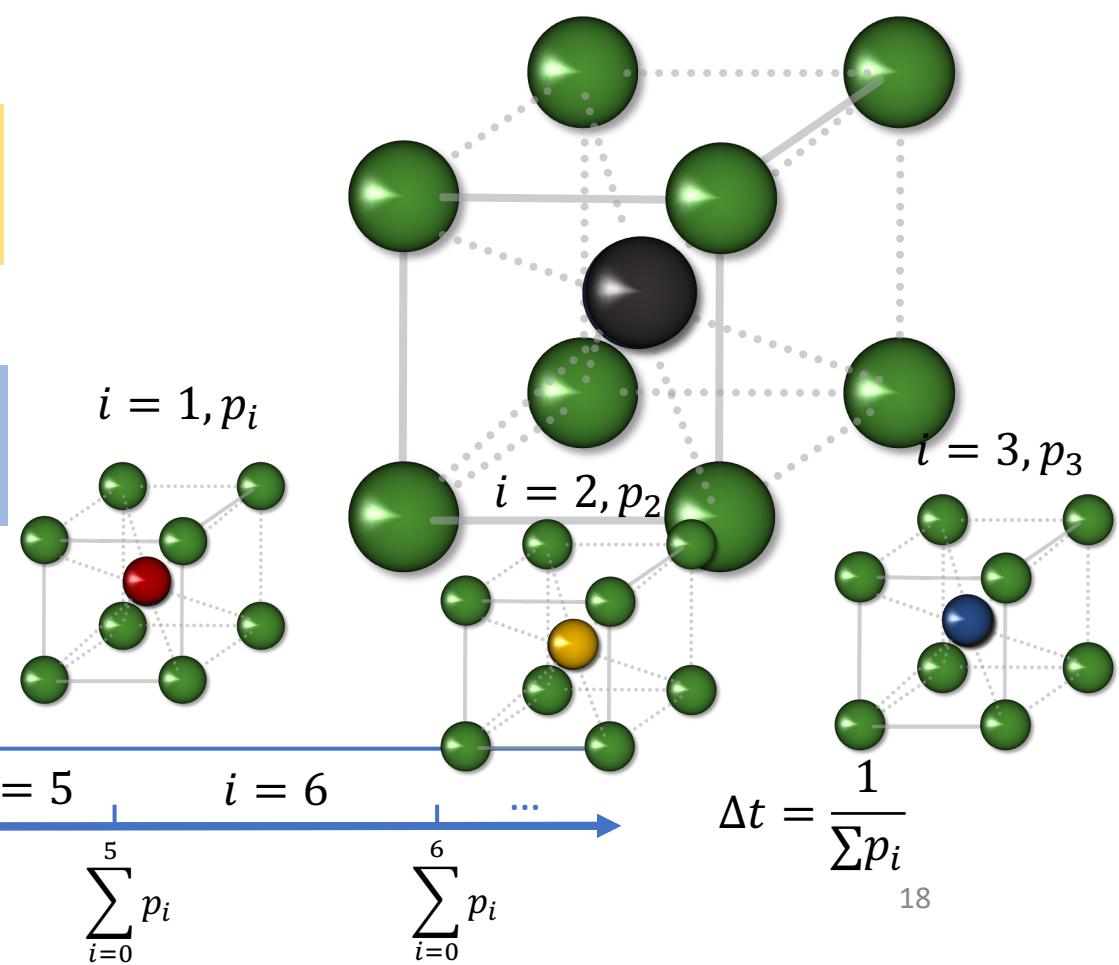
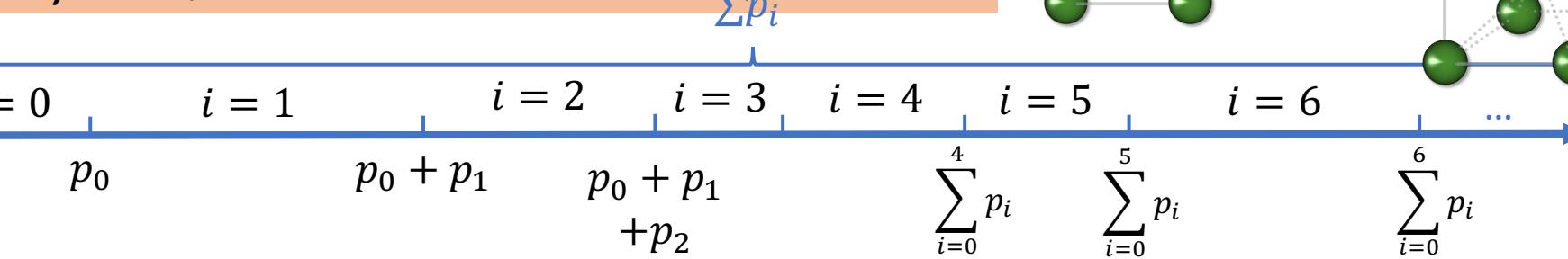
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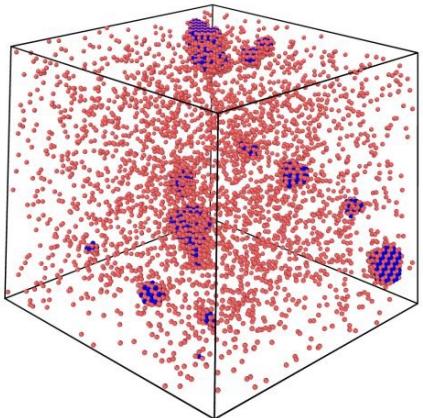
$$\Delta t = \frac{1}{\sum p_i}$$

Monte Carlo methods

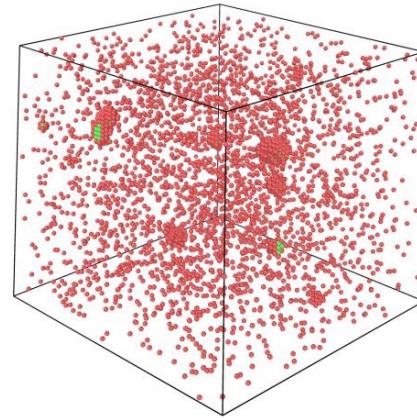
Kinetic Monte Carlo

Mechanism of Re precipitation in irradiated W-Re alloys from kinetic Monte Carlo simulations

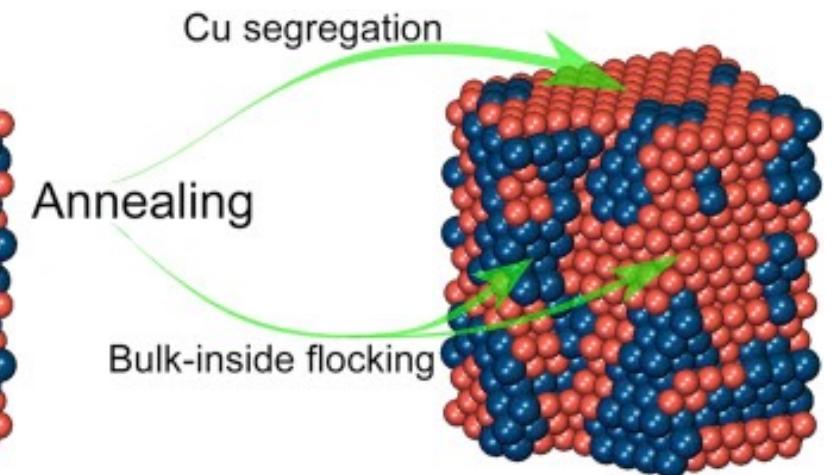
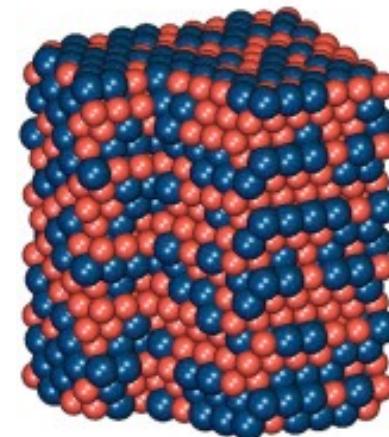
[doi: 10.1103/PhysRevB.96.094108](https://doi.org/10.1103/PhysRevB.96.094108)



(a) W-1.8at%Re alloy, 0.5 at% vacancy concentration.



(b) W-1.4at%Re alloy, 0.1 at% mixed-interstitials.



4. Commonly used on-lattice models

Commonly used on-lattice models

- Cluster expansion method
- Generalized perturbation method
- Low-rank interatomic potential

Commonly used on-lattice models

Cluster expansion method

Energy of the system with determined cfg

J_α – fit to first principal calculations

Empty cluster energy
cluster

$$E(\sigma) = J_{\text{null}} + \sum_i J_i \sigma_i + \sum_{ij} J_{ij} \sigma_i \sigma_j + \sum_{ijk} J_{ijk} \sigma_i \sigma_j \sigma_k + \dots$$

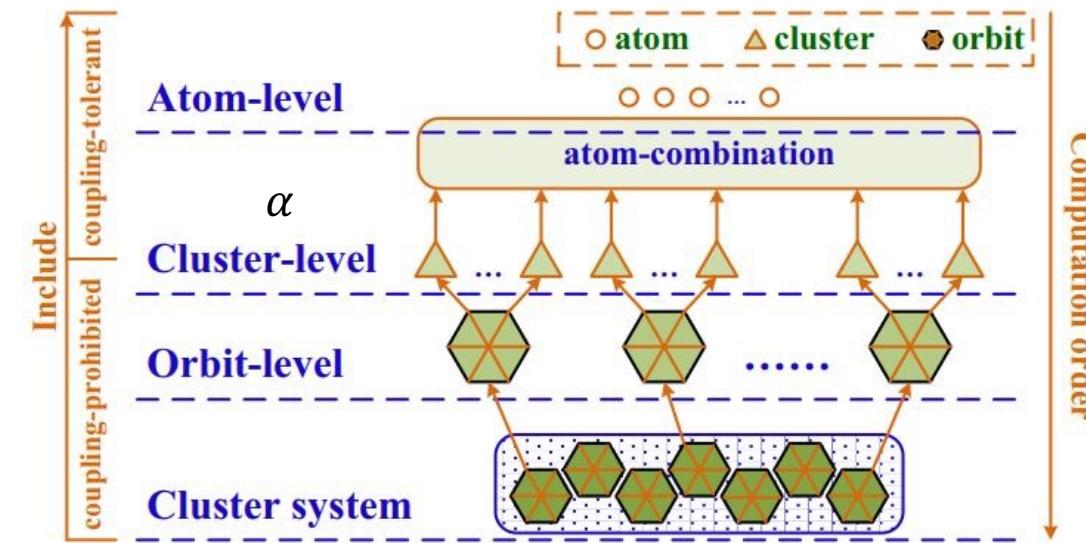
The further the accurate...

Point cluster	pair cluster	triplets cluster
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$$E(\sigma) = \sum_{\alpha} m_{\alpha} J_{\alpha} \langle \prod_{p \in \alpha} \sigma_p \rangle$$

Effective cluster interaction
Atoms positioning
site

Example for binary alloy



Commonly used on-lattice models

Effective interaction model

J_{ij} exchange-coupling parameter for the magnetic moments

$$H = \frac{1}{2} \sum_i \sum_j V_{ij} + \frac{1}{2} \sum_i \sum_j J_{ij} \overrightarrow{M}_i \overrightarrow{M}_j , \quad \overrightarrow{M}_i = (M_i, \phi, \theta)$$

chemical interactions between atoms

$\Delta E = \frac{1}{2} \sum_i \sum_j J_{ij} \cdot \Delta(\overrightarrow{M}_i \overrightarrow{M}_j)$ – fit for same chem cfg, but different magnetic states

Table 1

Fitted parameters (in meV) for the magnetic (J_{ij}) and chemical (V_{ij}) pair interactions in the EIM (Eq. (1)).

	1nn	2nn	3nn	4nn	5nn
J_{Fe-Fe}	-11.65800	-1.67630	0.51512	0.49840	0.22120
J_{Co-Co}	-7.60200	-6.58240	-5.16990	1.95370	0.13135
J_{Fe-Co}	-5.98790	-2.69460	-2.00000	-0.04000	0.01000
V_{Fe-Fe}	10.62000	-4.94300	-	-	-
V_{Co-Co}	10.62000	-4.94300	-	-	-
V_{Fe-Co}	-11.92500	4.10800	-	-	-

Commonly used on-lattice models

Generalized perturbation method

+Effective interaction model

- 1) Effective medium (homogeneous random alloy without any short-range order) which represents a random alloy configuration on average.

$$\widetilde{V_S^{(n)}} = \frac{1}{2^n} V_S^{(n)}$$

Concentration-variable effective interaction from GPM

- 2) Consider the energetics of the corresponding clusters embedded in this effective medium.

Configuration determination	$\xi_S^{(n)} = \frac{1}{n} \sum_{p \in S} \prod_{i=1,n} \sigma_{i,p}$	<div style="display: flex; justify-content: space-around; align-items: center;"> Geometry class Sites in cluster </div>
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$$E_{conf} = \sum_{n,S} V_S^{(n)} \xi_S^{(n)}$$

Effective
interaction

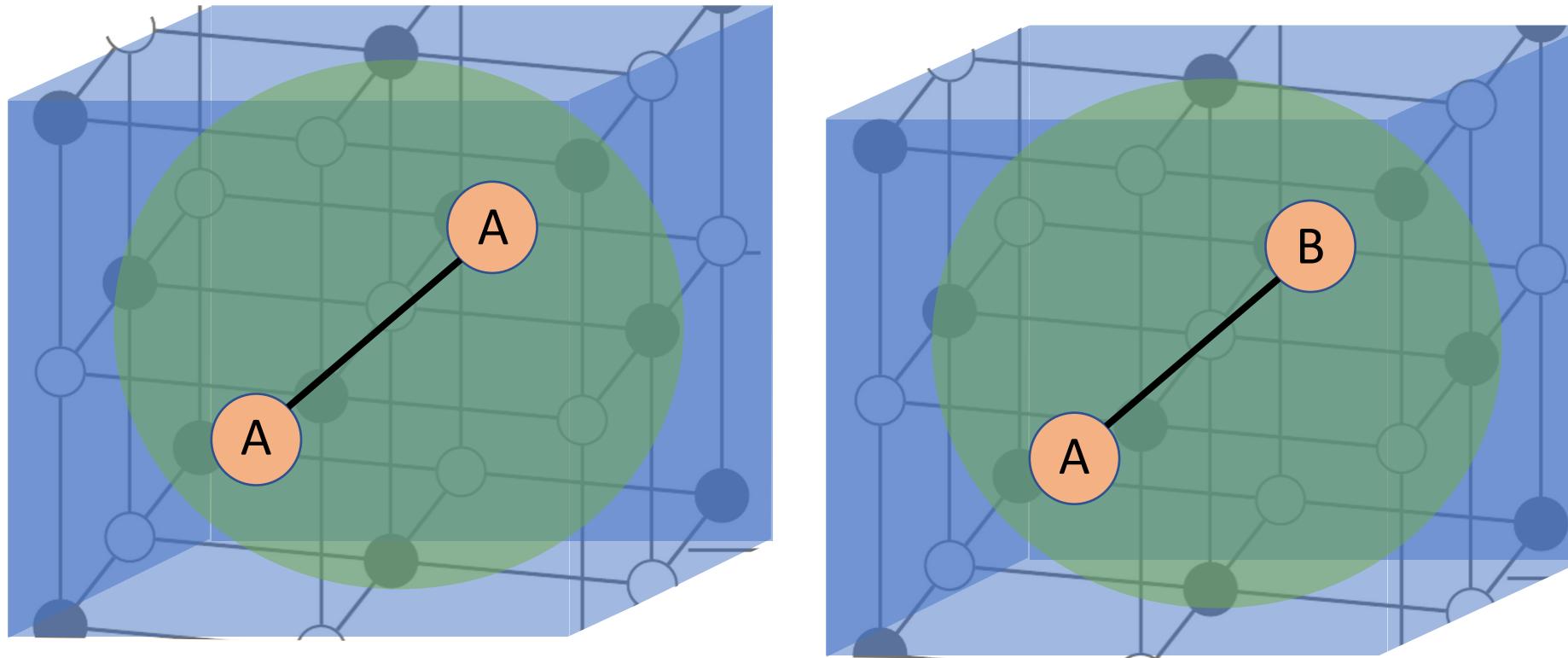
$$\xi_S^{(n)} = \langle \sigma \rangle^n$$

all the clusters with
an even (odd) number
of A atoms embedded

Commonly used on-lattice models

Generalized perturbation method

+Effective interaction model

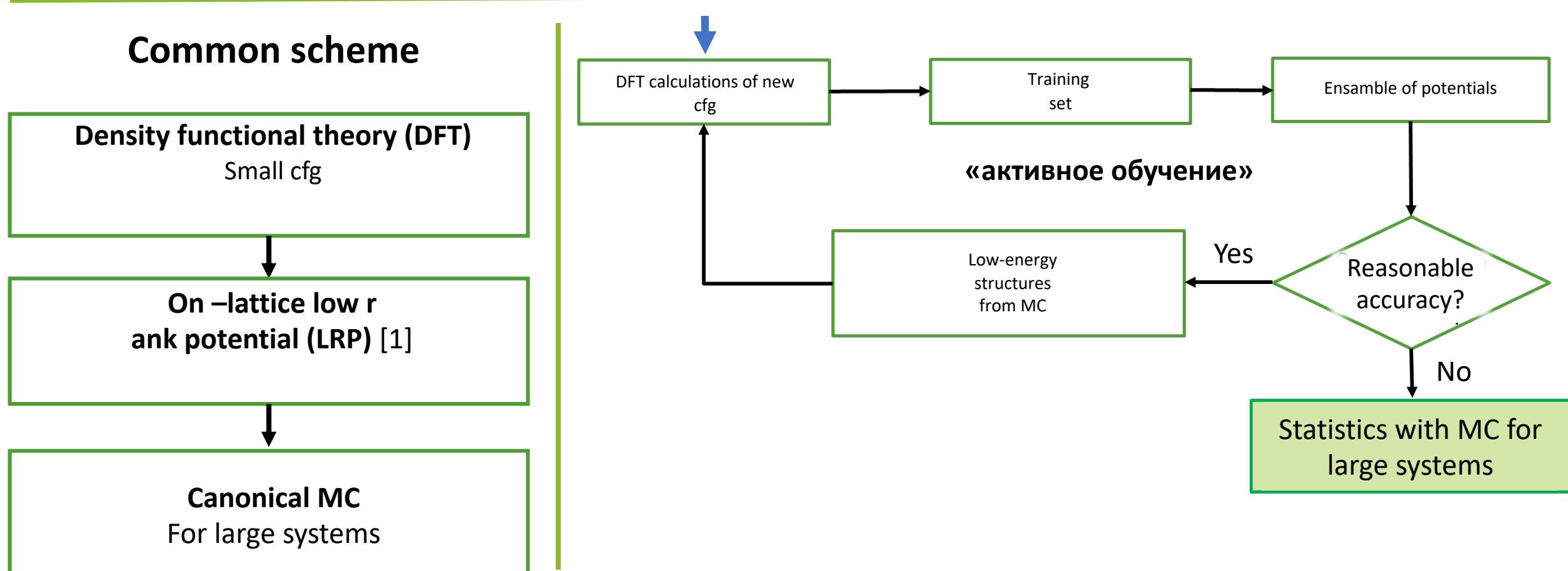


Effective medium = homogeneous random alloy

Commonly used on-lattice models

Low-rank interatomic potential

Methodology



Commonly used on-lattice models

Low-rank interatomic potential

Представление энергии атомной системы:

$$E(\sigma) = \sum_{\xi \in \Omega} V(\sigma(\xi + r_1), \dots, \sigma(\xi + r_9)) \quad (1)$$

химические типы ближайших
атомов-соседей

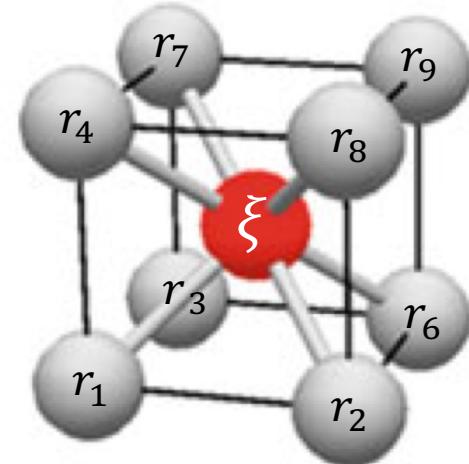
V — потенциал, тензор с 4^9 параметрами.

Разложение «тензорного поезда» [3] позволяет уменьшить число параметров до $4 \cdot 9 \cdot 5^2$ ($rank = 5$).
Задача регрессии:

$$\frac{1}{K} \sum_{k=1}^K |E(\sigma^{(k)}) - E^{\text{qm}}(\sigma^{(k)})|^2 \quad (2)$$

DFT

$$\sigma(x) = \left\{ \begin{array}{l} \text{Nb} \\ \text{Mo} \\ \text{Ta} \\ \text{W} \end{array} \right\}$$

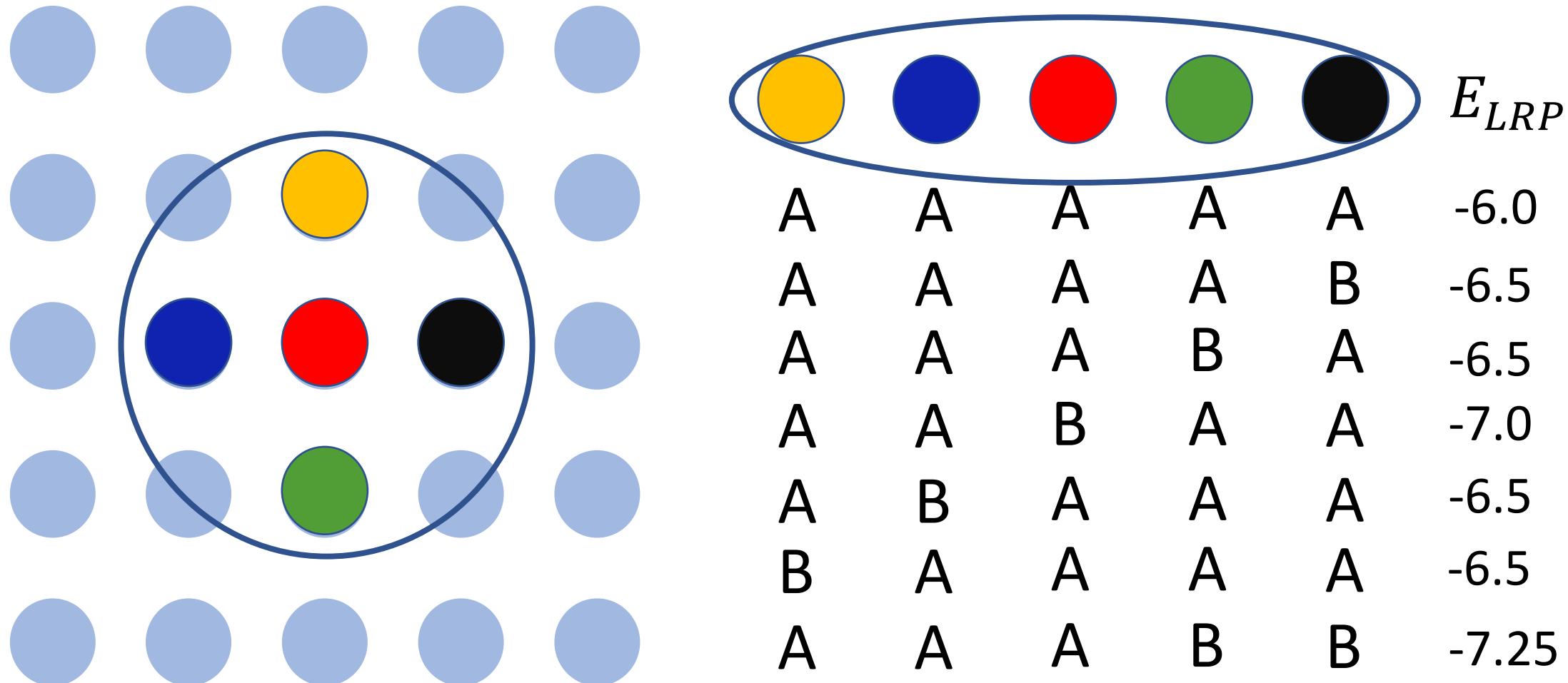


[1] Alexander Shapeev. Accurate representation of formation energies of crystalline alloys with many components. Computational Materials Science, 139:26-30, 2017 (The Low-Rank Interatomic Potential).

[3] Ivan V Oseledets. Tensor-train decomposition. SIAM Journal on Scientific Computing, 33(5):2295-2317, 2011.

Commonly used on-lattice models

Low-rank interatomic potential

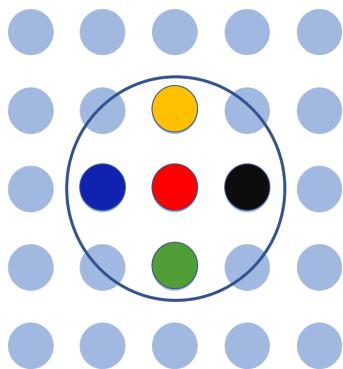


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Commonly used on-lattice models

Low-rank interatomic potential

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E _{LRP}					
A	A	A	A	A	-6.0
A	A	A	A	B	-6.5
A	A	A	B	A	-6.5
A	A	B	A	A	-7.0
A	B	A	A	A	-6.5
B	A	A	A	A	-6.5
A	A	A	B	B	-7.25

$$A_{ij} = \begin{array}{c} \text{[blue bar]} \\ \times \\ \text{[blue bar]} \\ \times \\ \text{[blue bar]} \end{array}$$

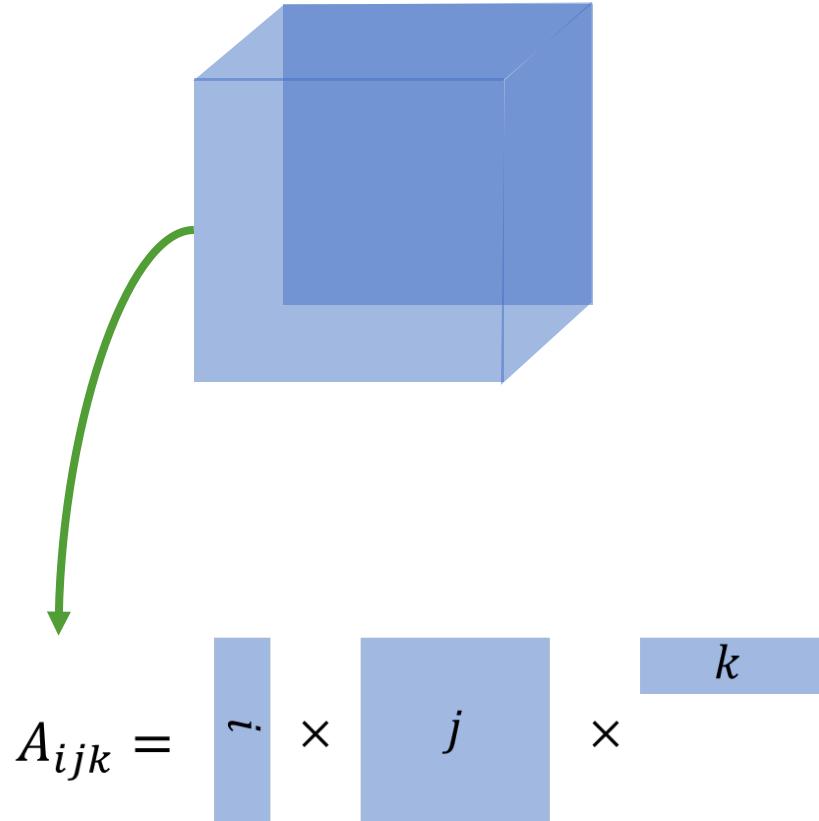
In BCC 9-dimensional tensor

$$\begin{array}{ccc} \text{[blue 3x3 grid]} & = & \text{[blue vertical bar]} \times \text{[blue horizontal bar]} \end{array}$$

$$A_{23} = \begin{array}{c} i=2 \\ \times \\ j=3 \end{array}$$

Commonly used on-lattice models

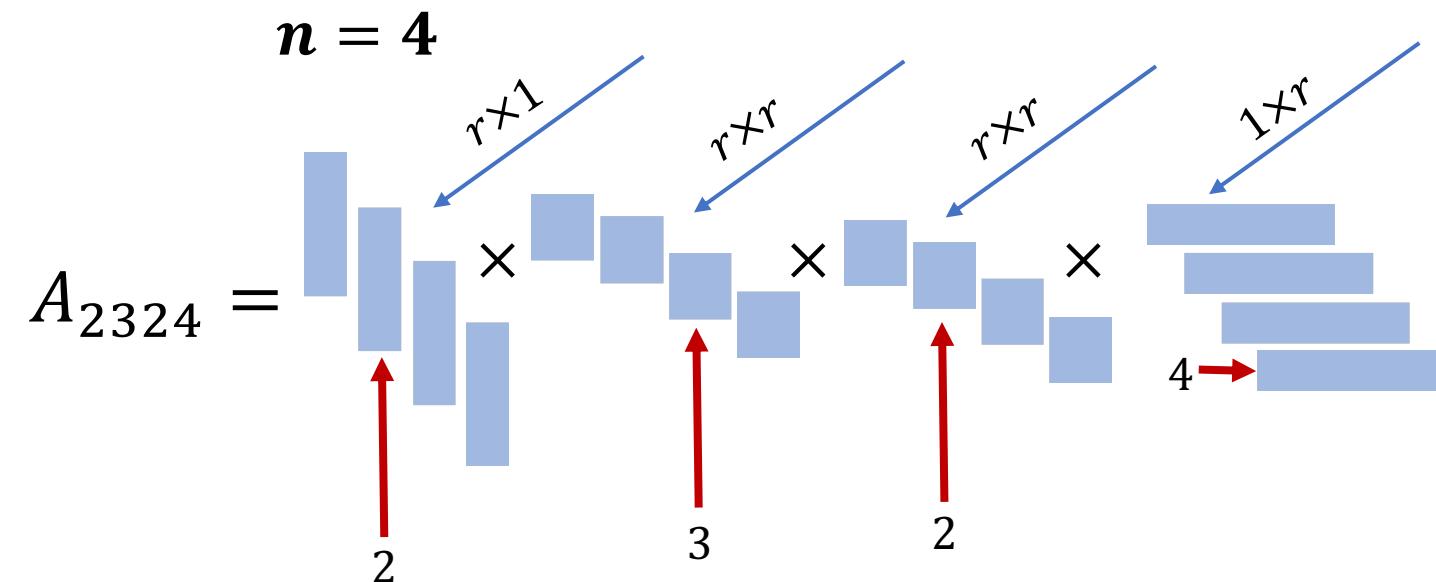
Low-rank interatomic potential



A diagram illustrating the decomposition of a 3D tensor into three 2D matrices. On the left, a large blue 3D cube represents the tensor A_{ijk} . A green arrow points from this cube down to a horizontal row of three blue rectangles. The first rectangle is labeled i , the second j , and the third k . Between the i and j rectangles is a multiplication sign (\times), and between the j and k rectangles is another multiplication sign (\times).

$$A_{ijk} = i \times j \times k$$

Tensor train (TT) nmr^2



A diagram illustrating the Tensor Train (TT) decomposition for $n=4$. The expression $A_{2324} =$ is followed by a sequence of four tensors connected by multiplication signs (\times). Above each tensor is a blue arrow pointing to its right, labeled $r \times 1$. Below each tensor is a red arrow pointing upwards, labeled with its rank: 2, 3, 2, and 4 respectively. The final rank 4 is also indicated by a red arrow pointing to the right of the last tensor.

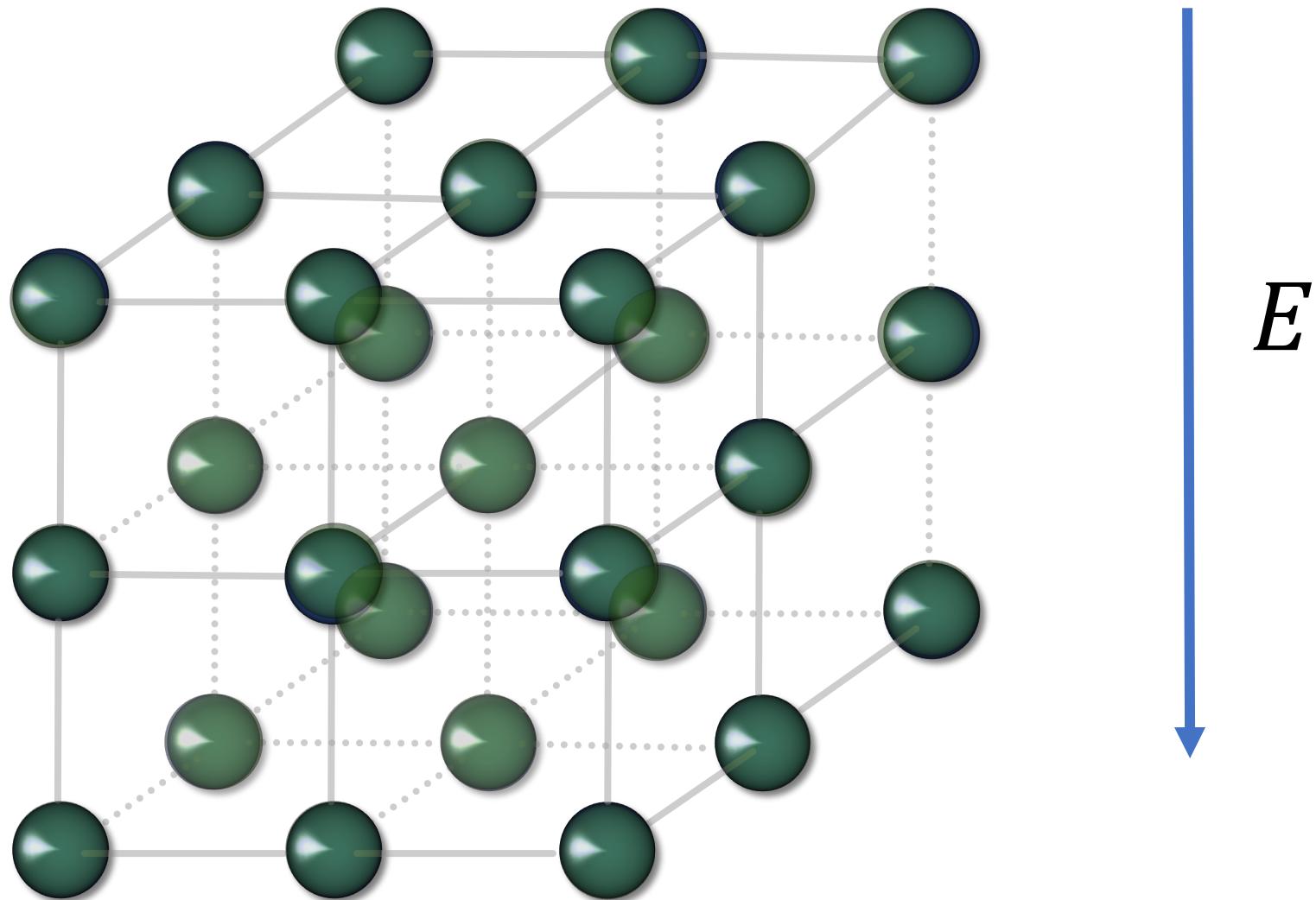
$$A_{2324} =$$

$n = 4$

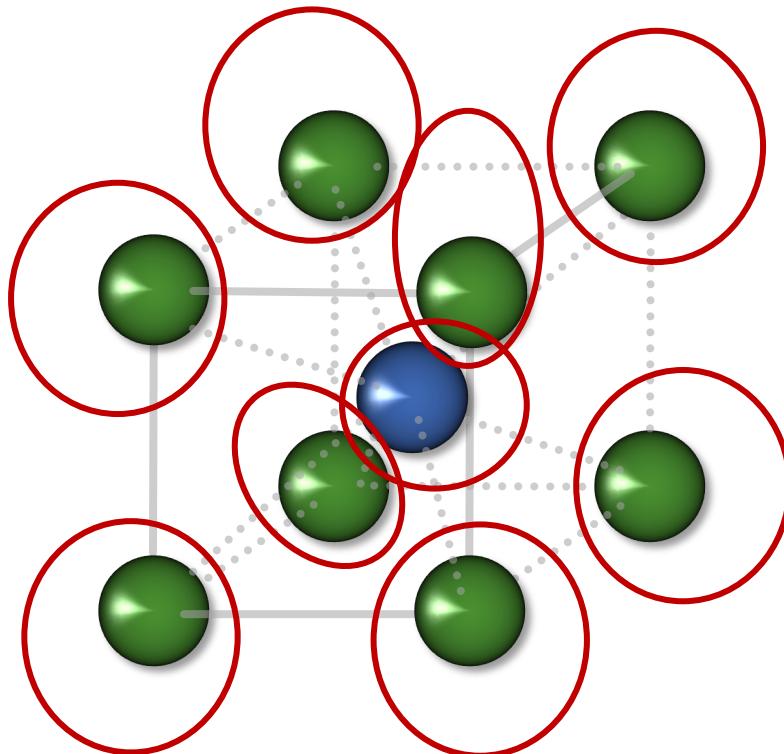
$r \times 1$ $r \times r$ $r \times r$ $1 \times r$

2 3 2 4

Accounting for local lattice distortions



Accounting for local lattice distortions

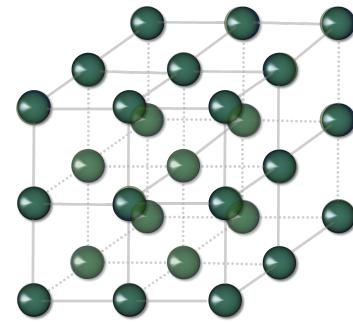


E_{local}

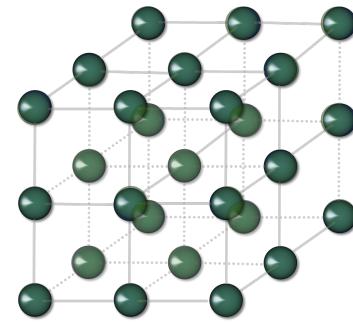
Accounting for local lattice distortions

UnRelaxed scenario

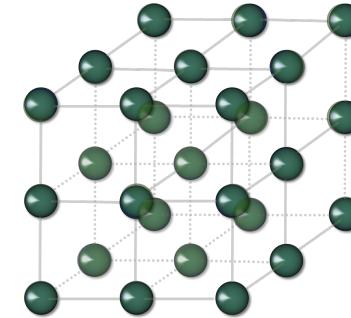
Learn from QM



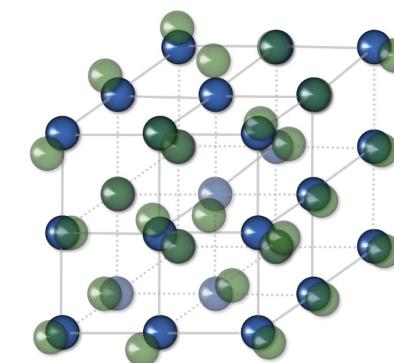
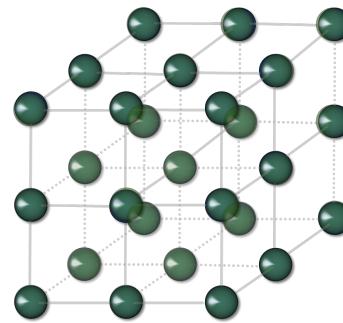
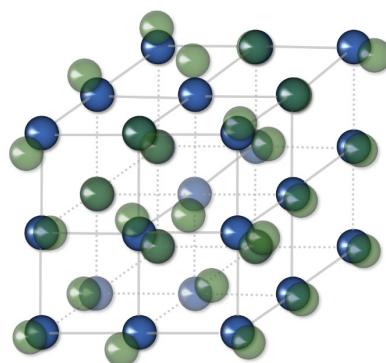
How it looks in
MC simulation



Predicts



Relaxed scenario

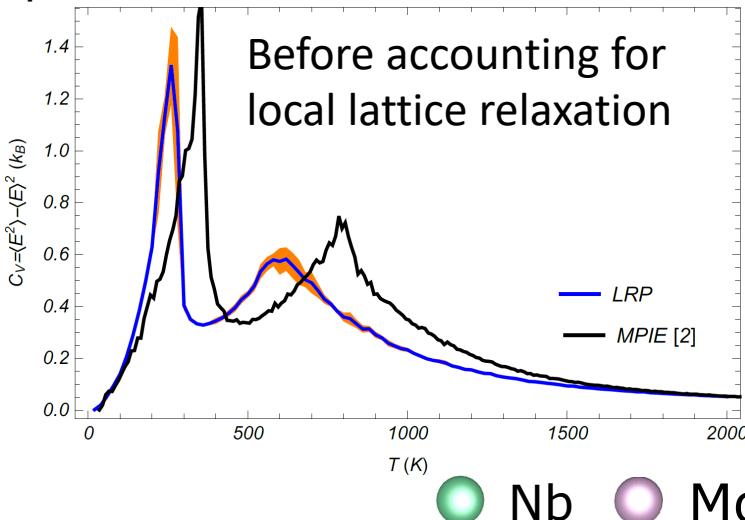


5. Frontier problems solved by these methods

Low-rank interatomic potential

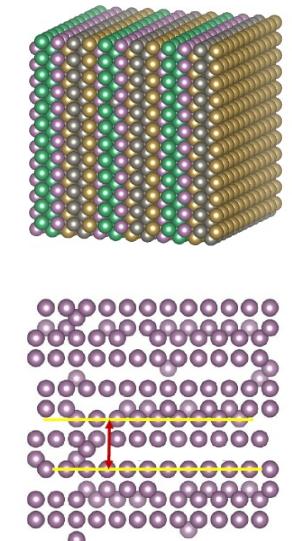
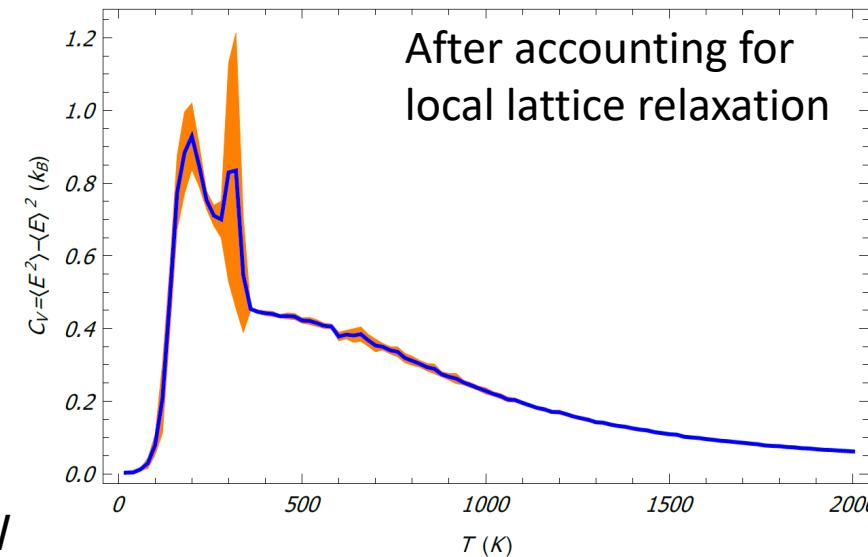
- A qualitatively new methodology has been developed and tested for the study of complex multicomponent alloys with low-rank machine learning potentials
- Investigation of the influence of local lattice distortions in the equiatomic system Nb-Mo-Ta-W

Impact of lattice relaxations on phase transitions in a high-entropy alloy studied by machine-learning potentials



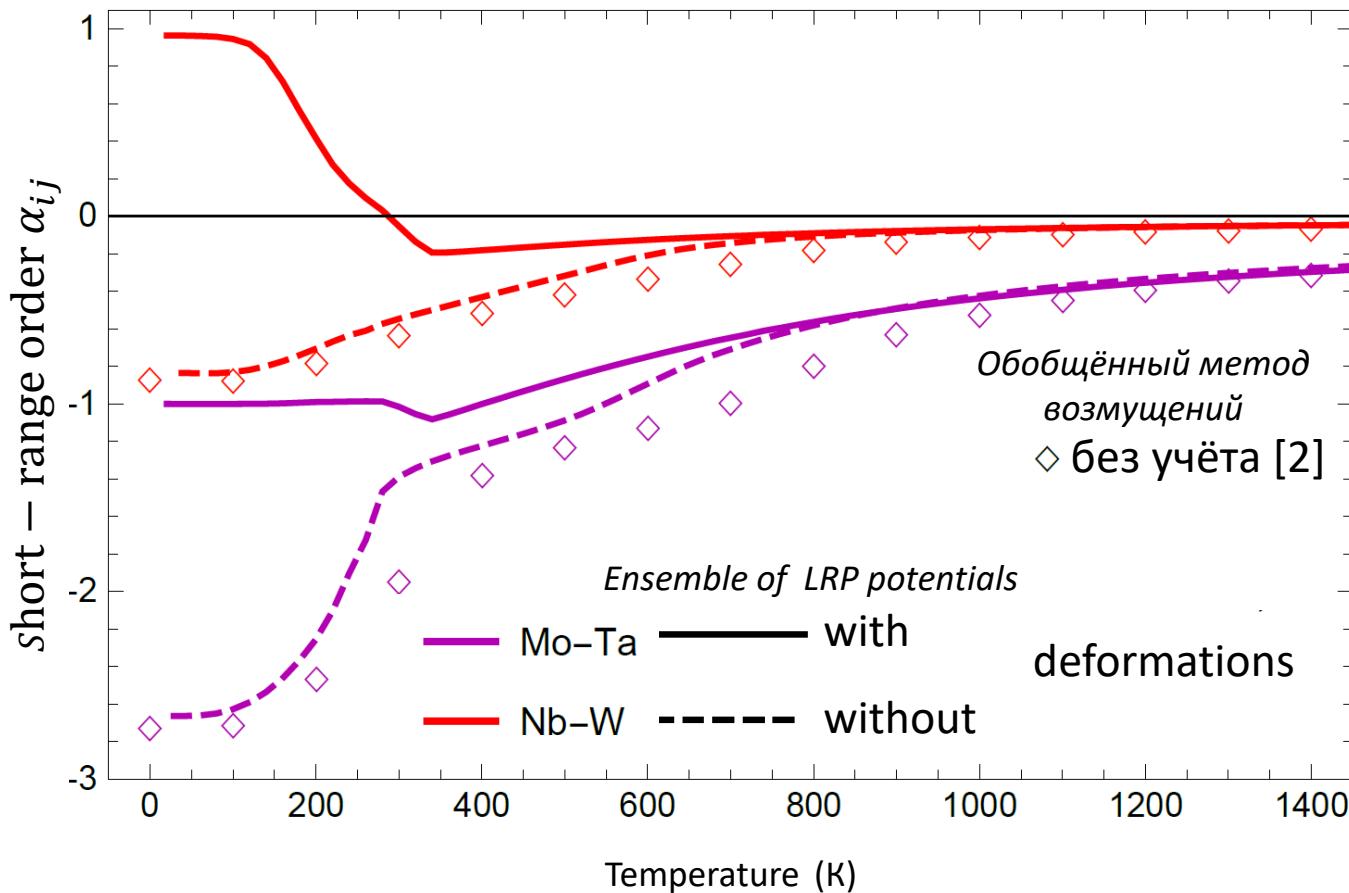
npj nature partner journals

Collaborators: Fritz Körmann
Jörg Neugebauer



Low-rank interatomic potential

With and without taking into account local deformations



Warren–Cowley short-range order parameters:

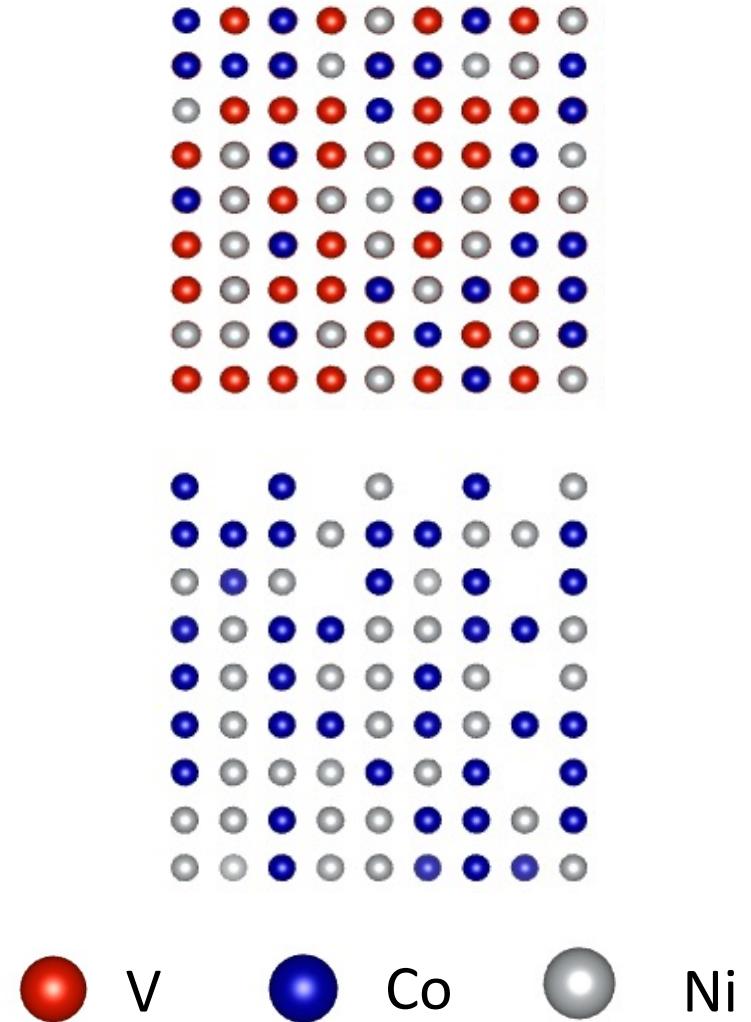
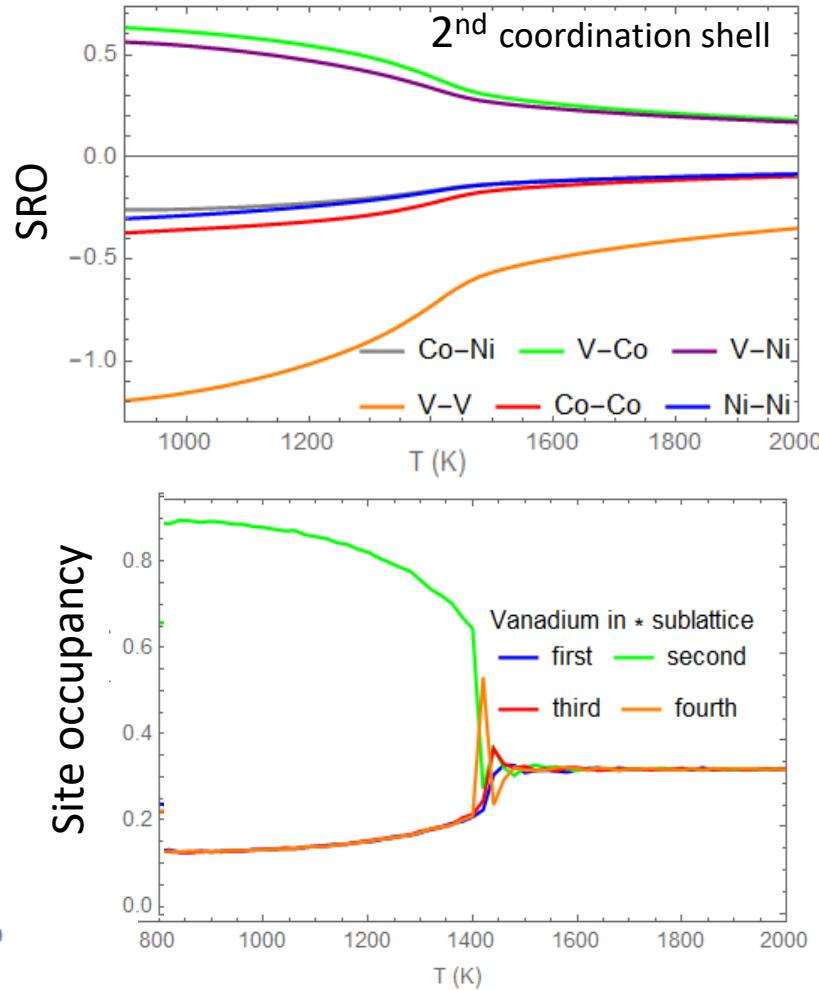
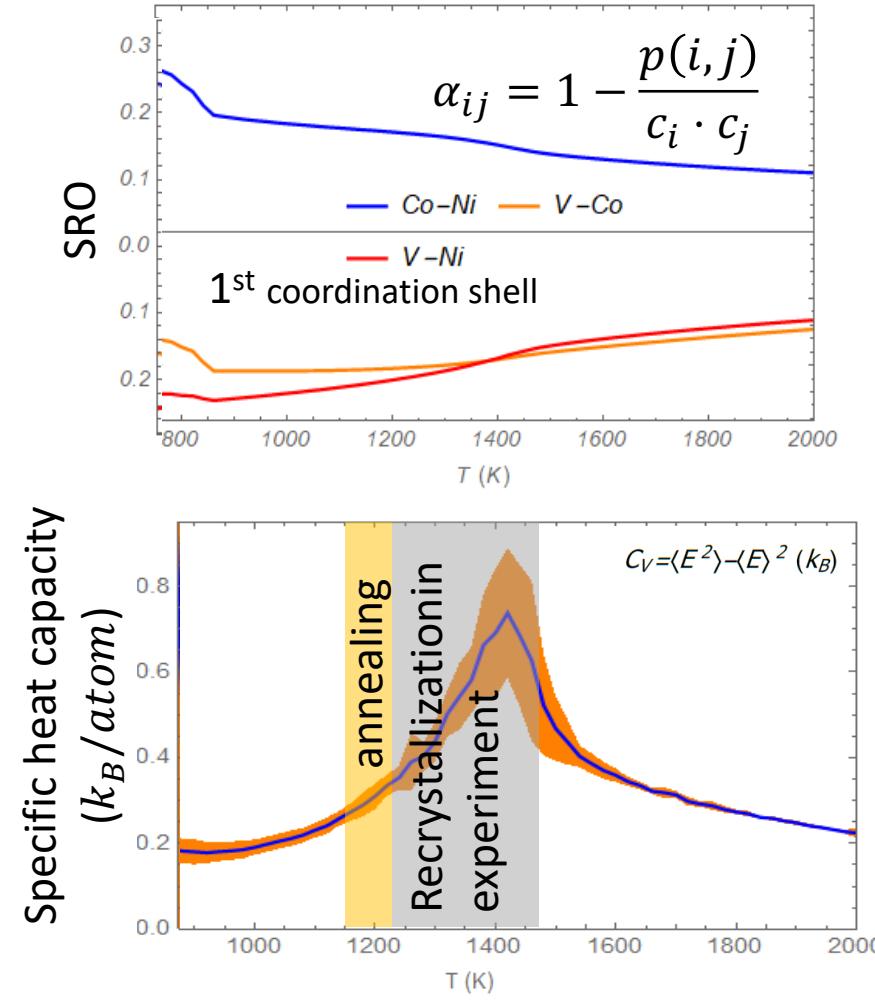
$$\alpha_{ij} = 1 - \frac{p(i,j)}{c_i \cdot c_j}$$

i, j – chemical elements

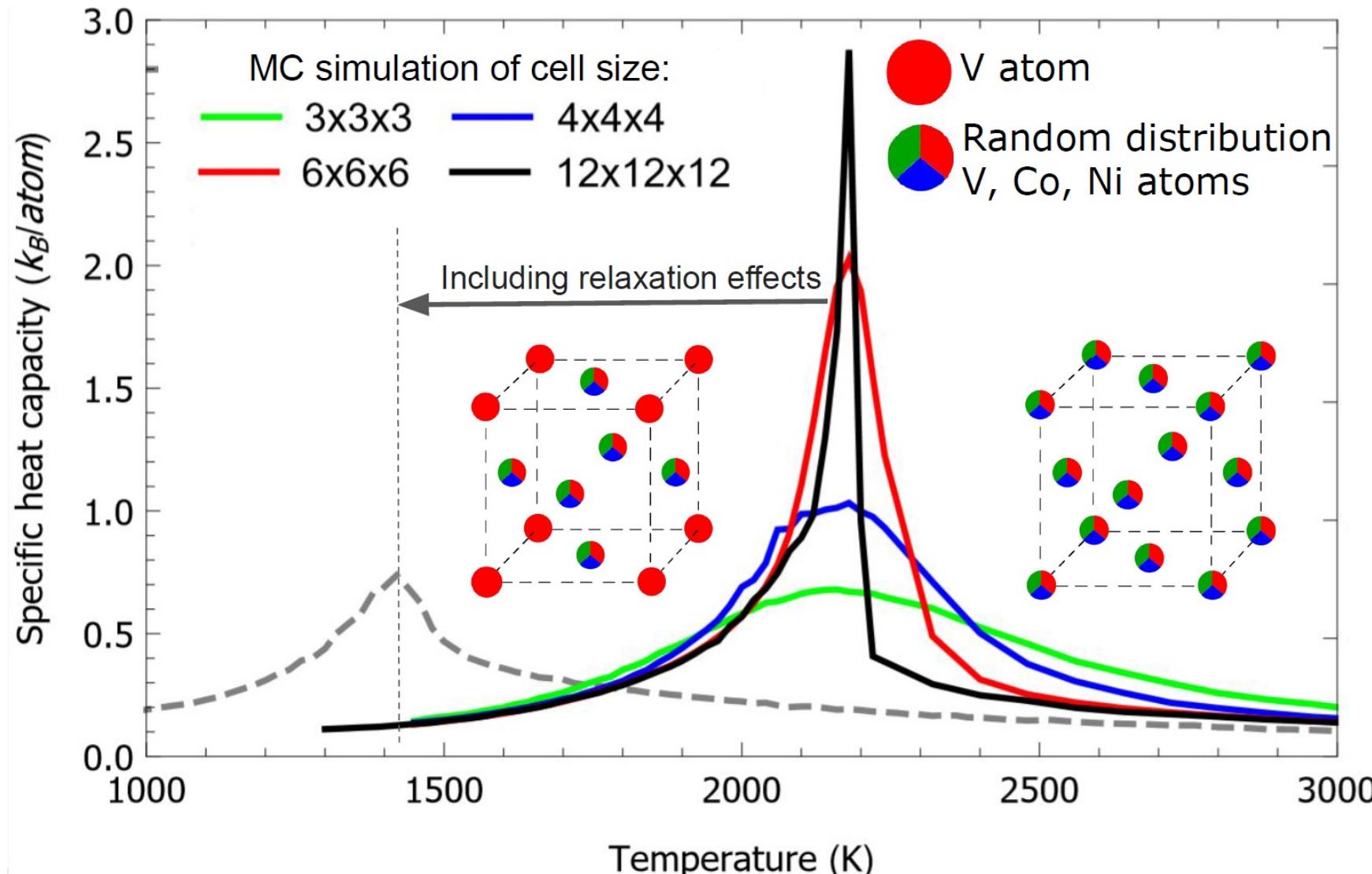
c_j, c_i – concentrations

$p(i,j)$ – probability to find as neighbors

Low-rank interatomic potential



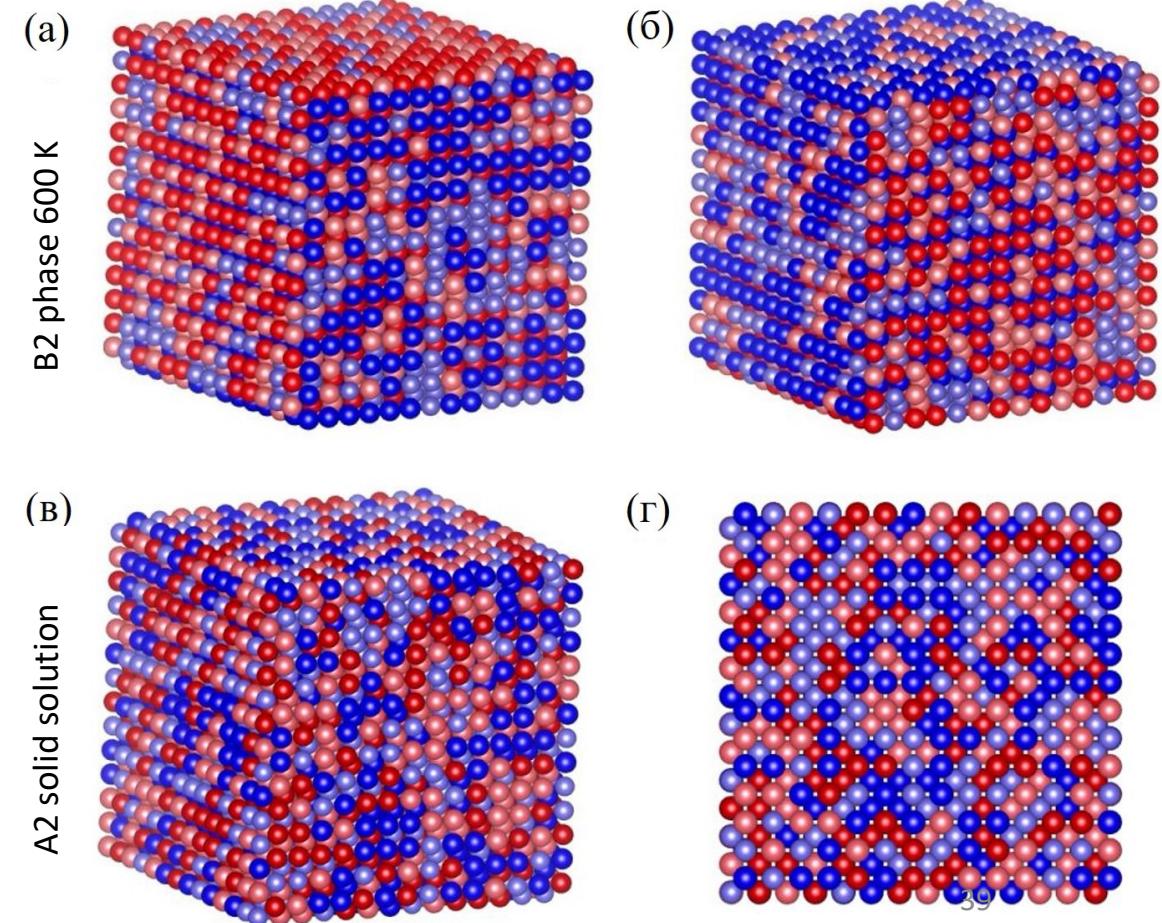
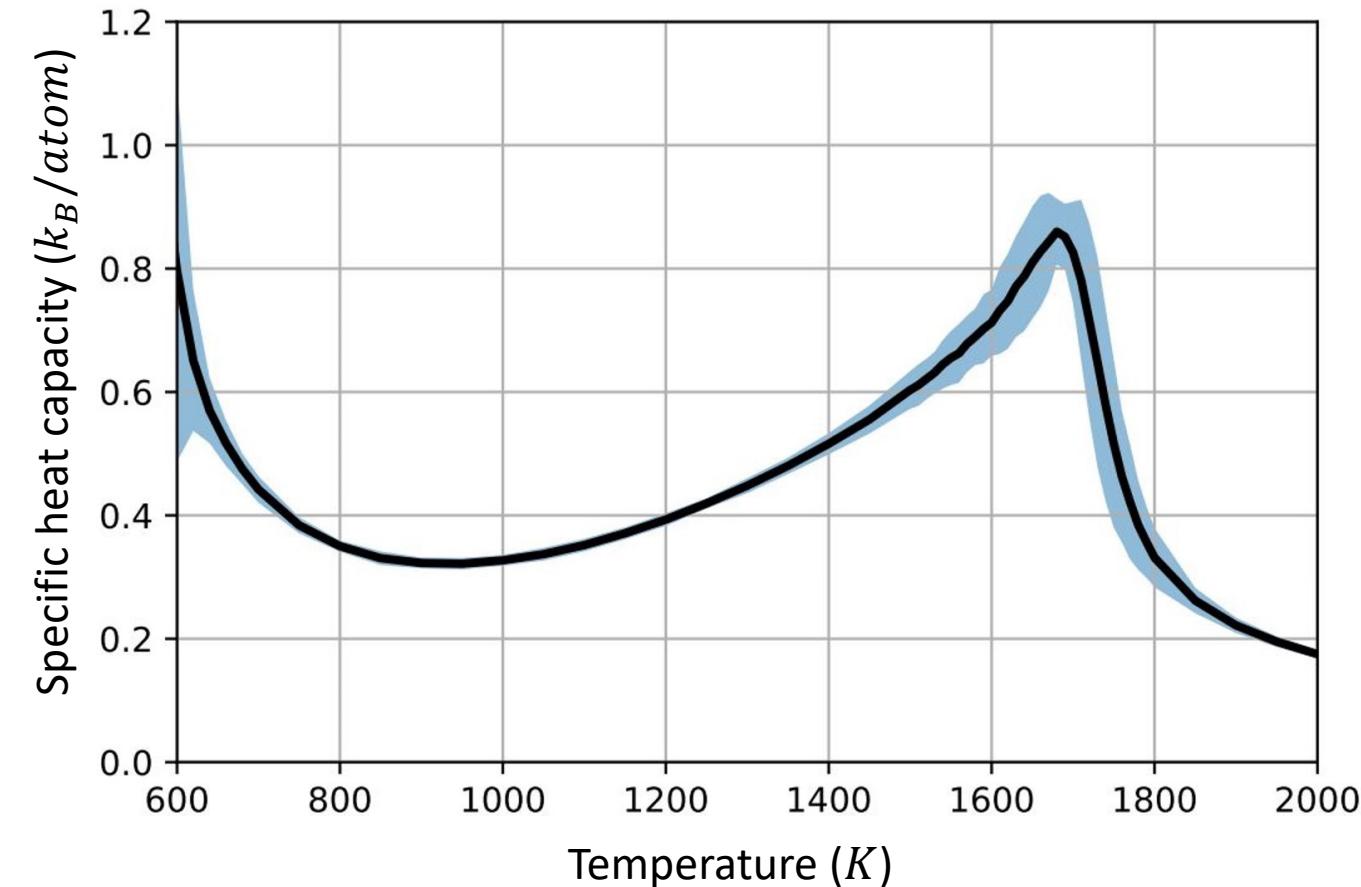
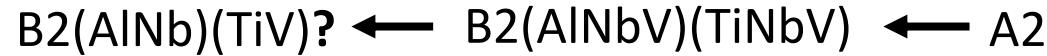
Low-rank interatomic potential



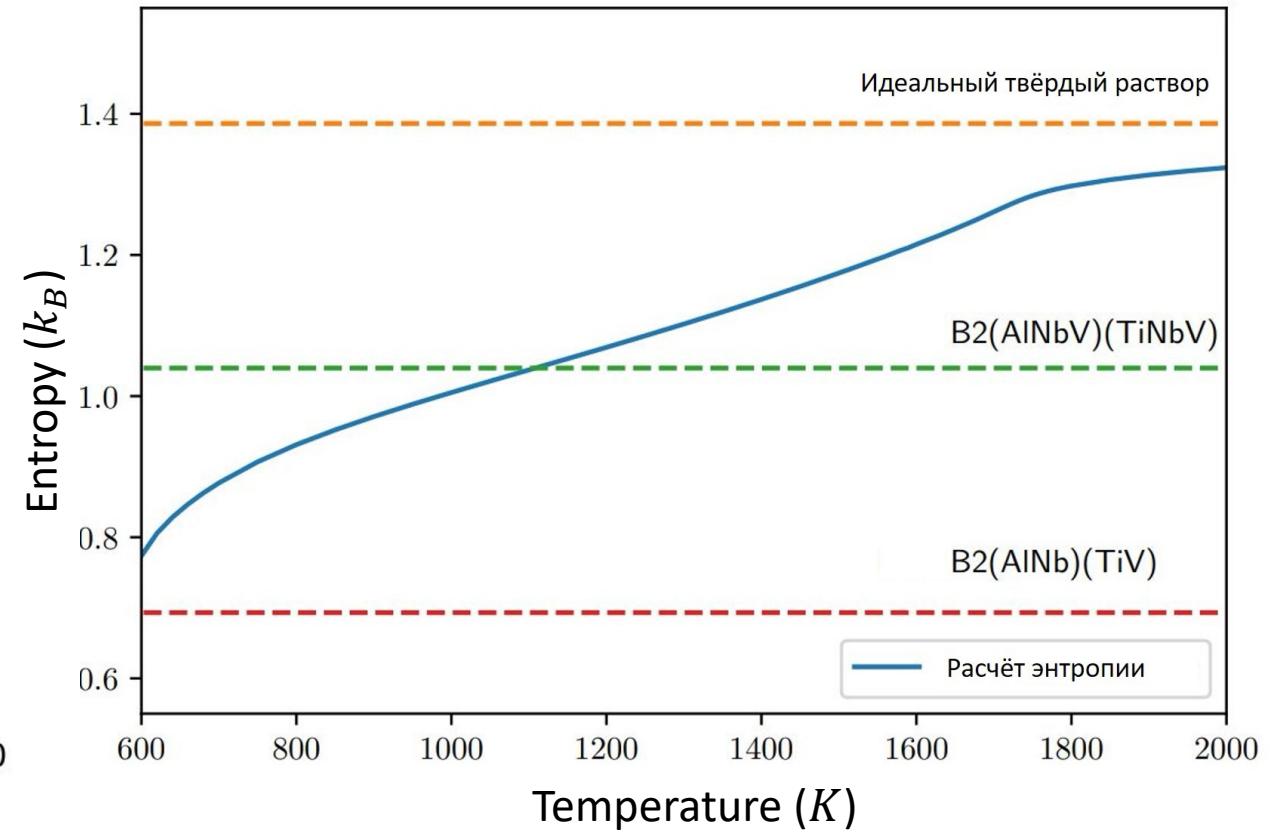
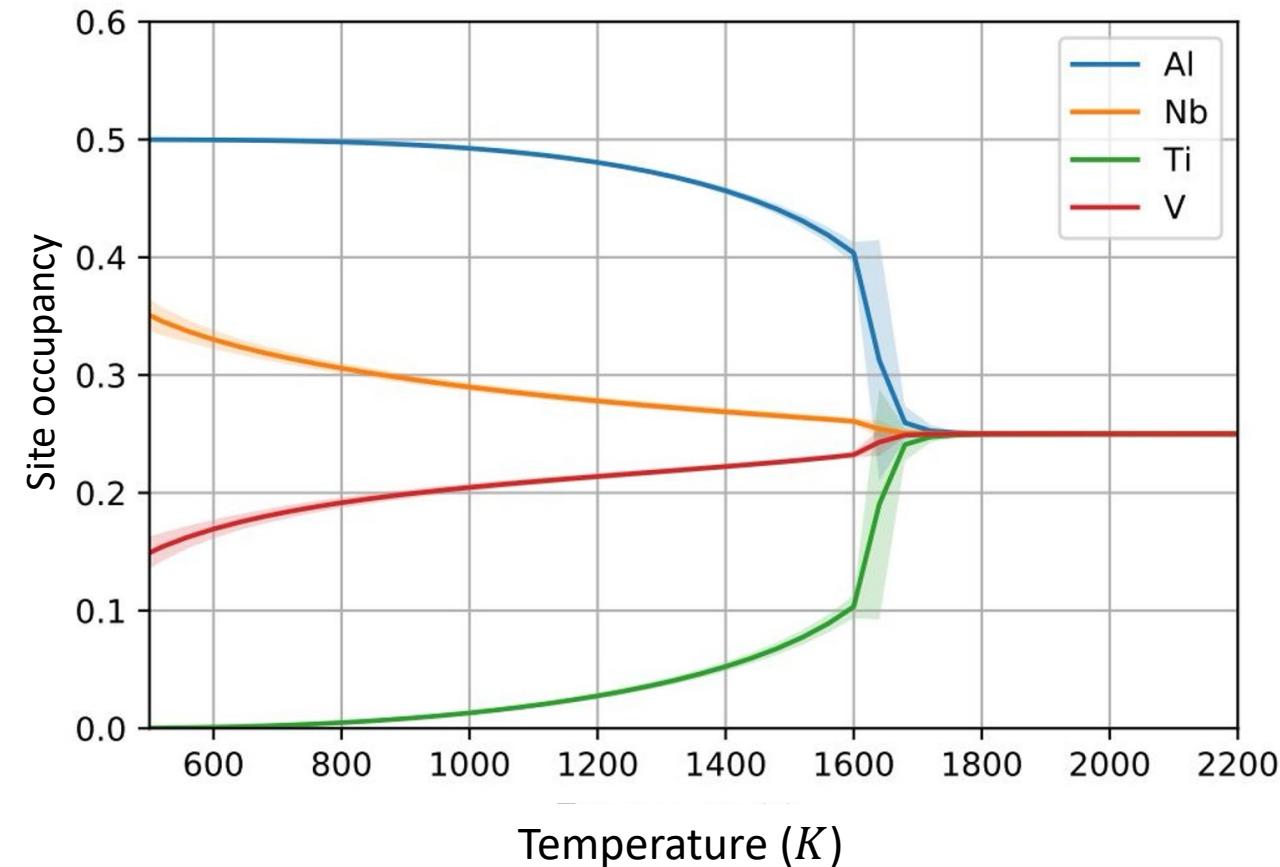
$$C_v = \frac{\langle E^2 \rangle - \langle E \rangle^2}{k_B T^2}$$

Low-rank interatomic potential

AlNbTiV and B2 phase formation

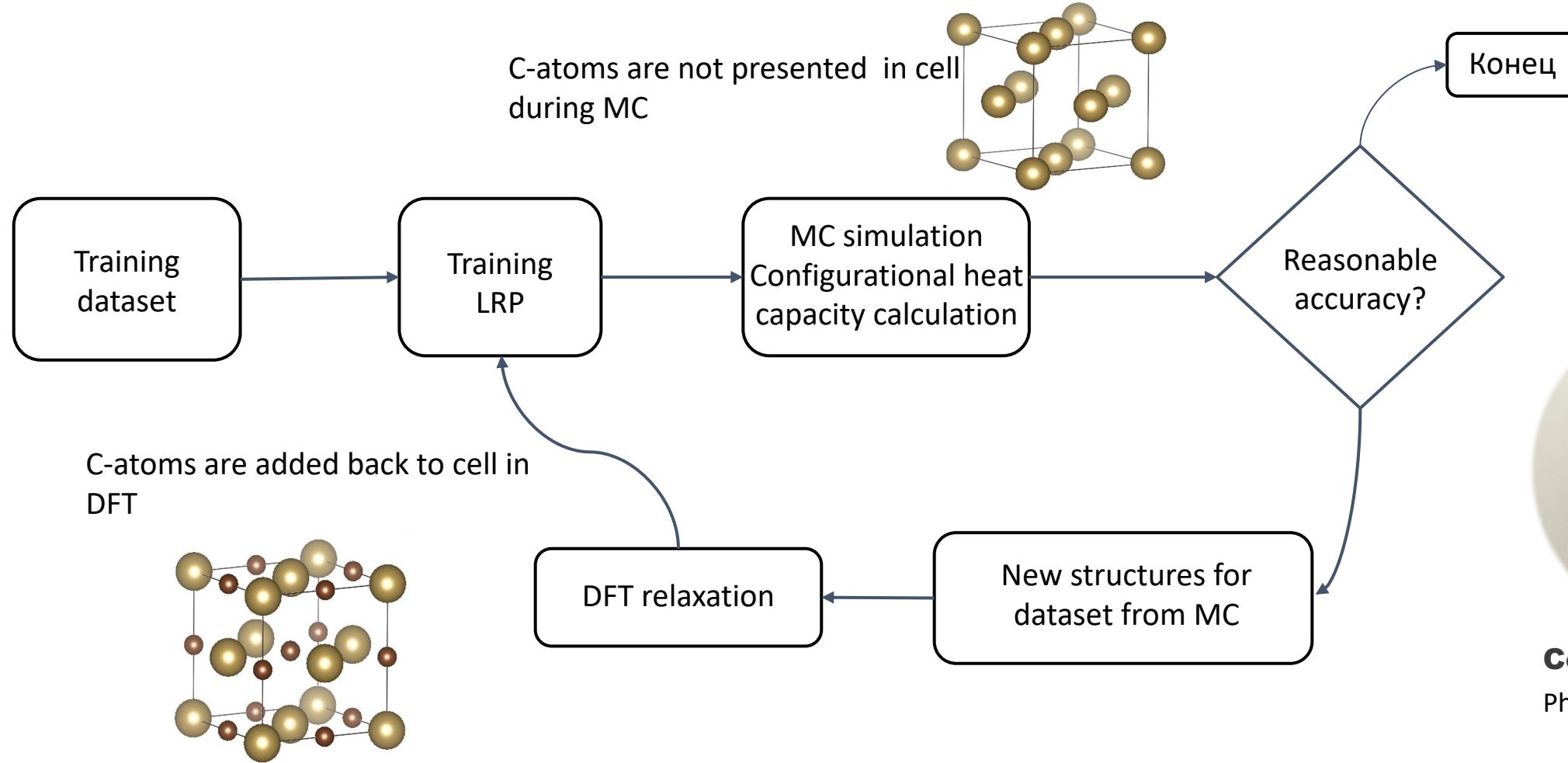


Low-rank interatomic potential



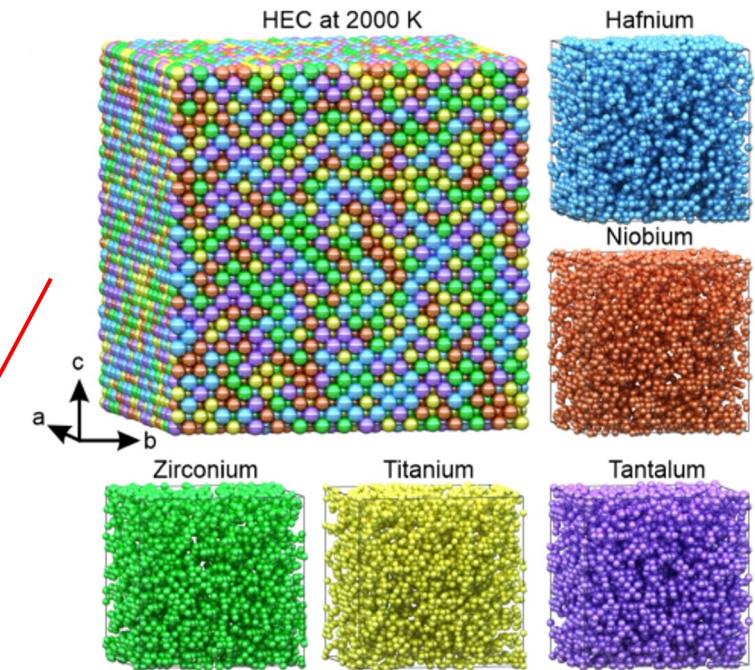
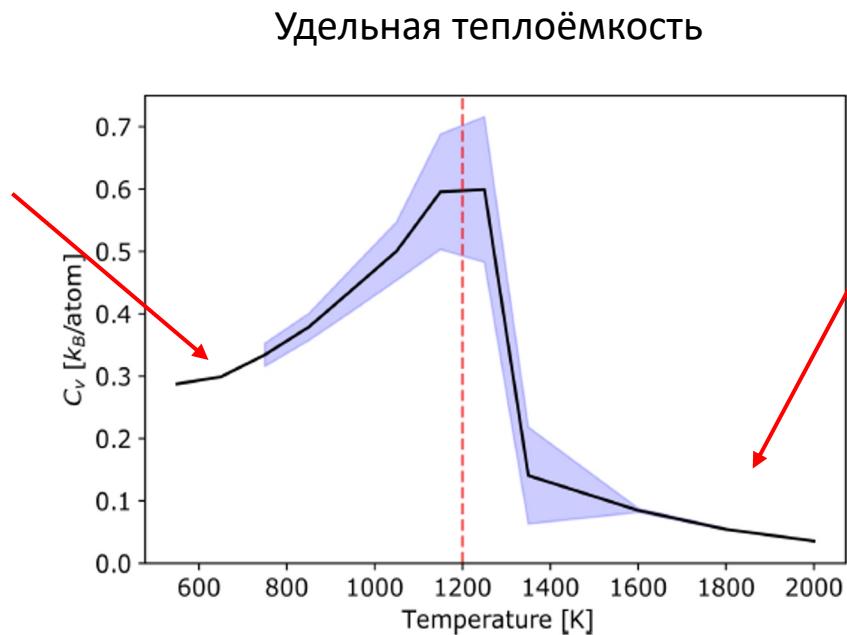
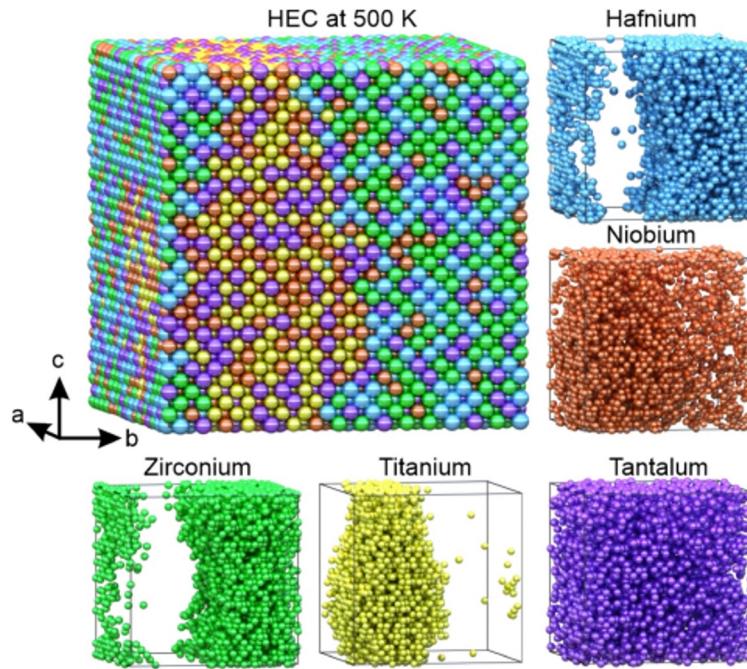
New structures search: HfTaTiNbZrC₅

Methodology – Canonical Monte Carlo (CMC) + Low-rank potential (LRP)



Сотсков Вадим
PhD student

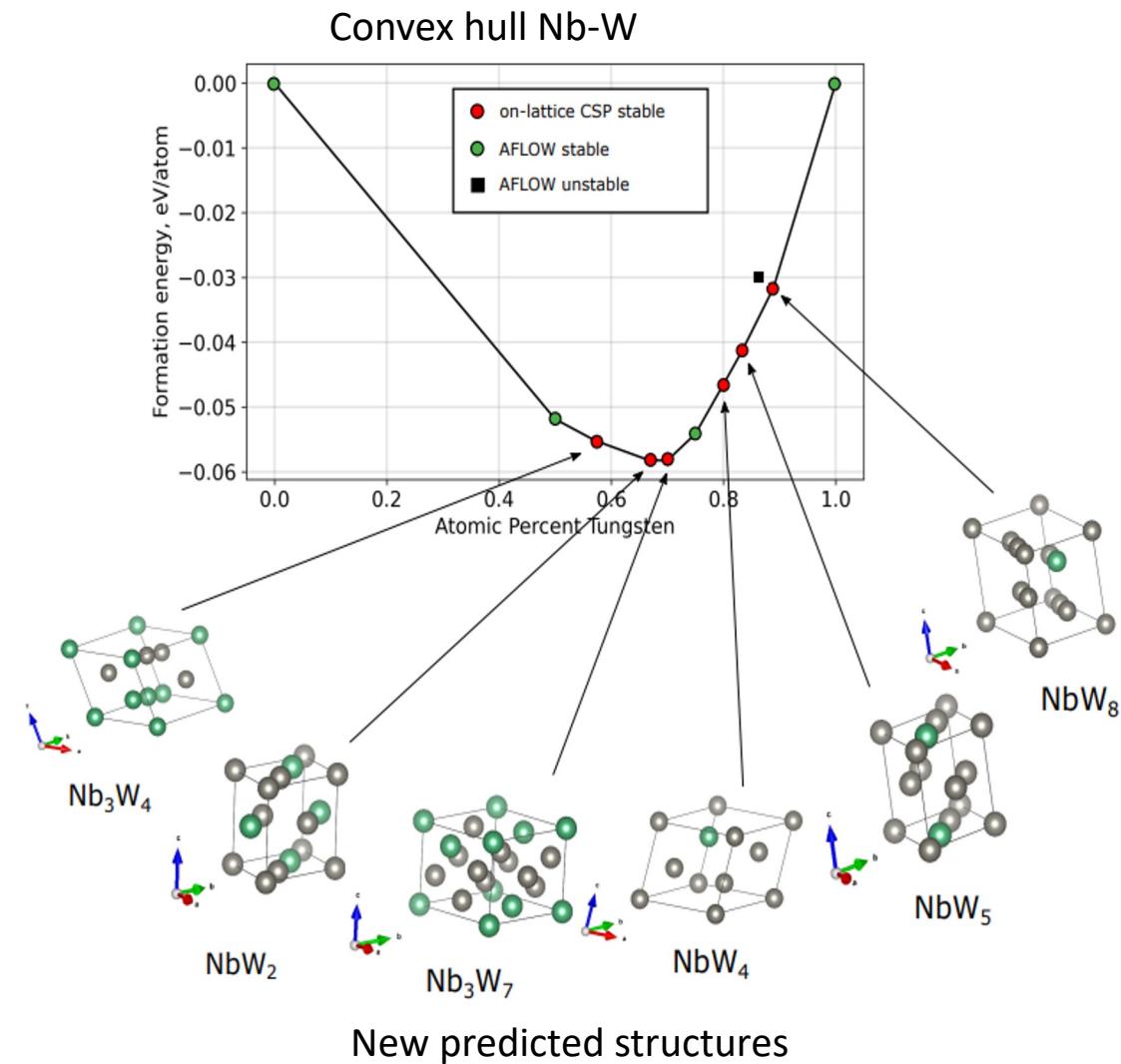
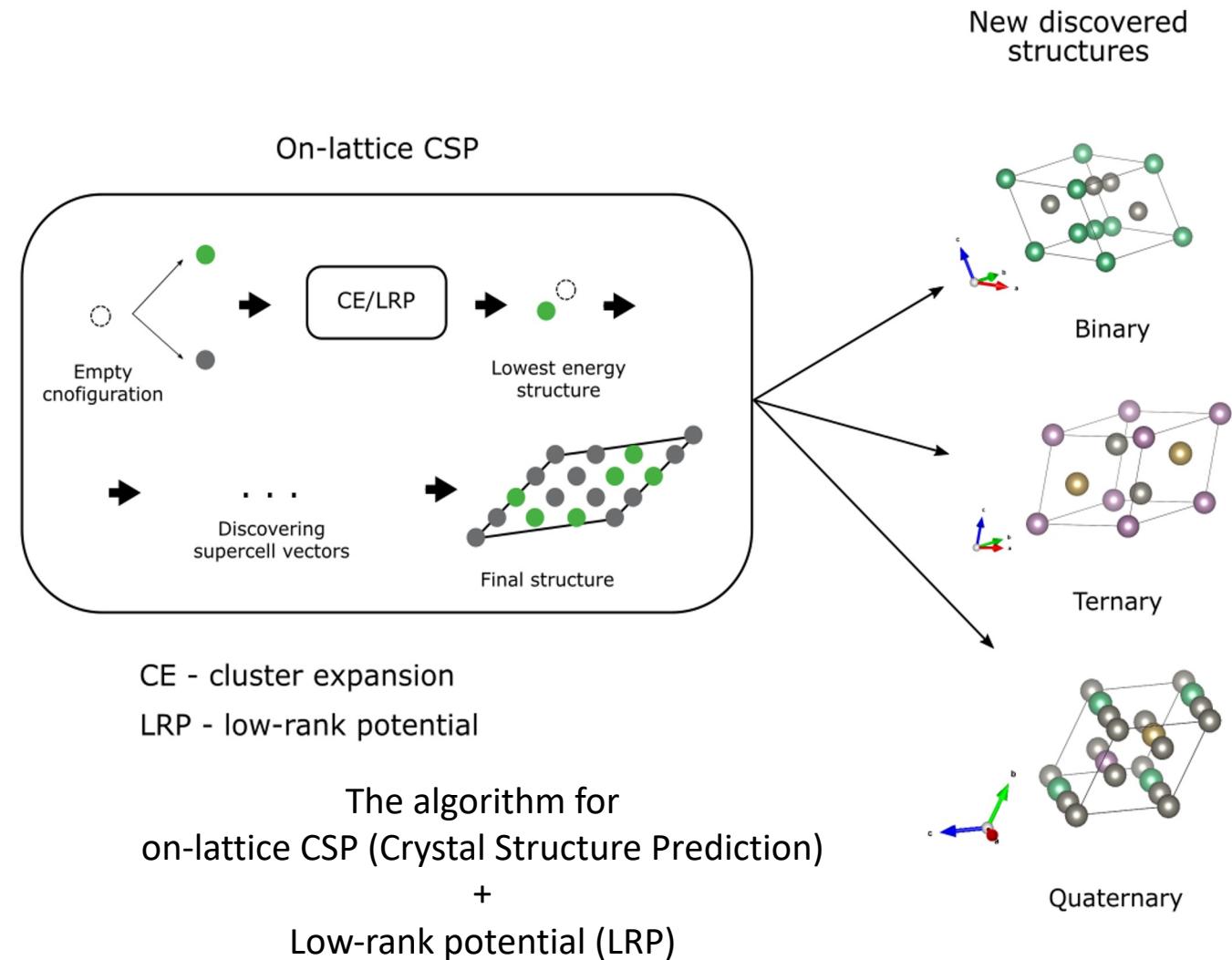
New structures search: SS stability HfTaTiNbZrC₅



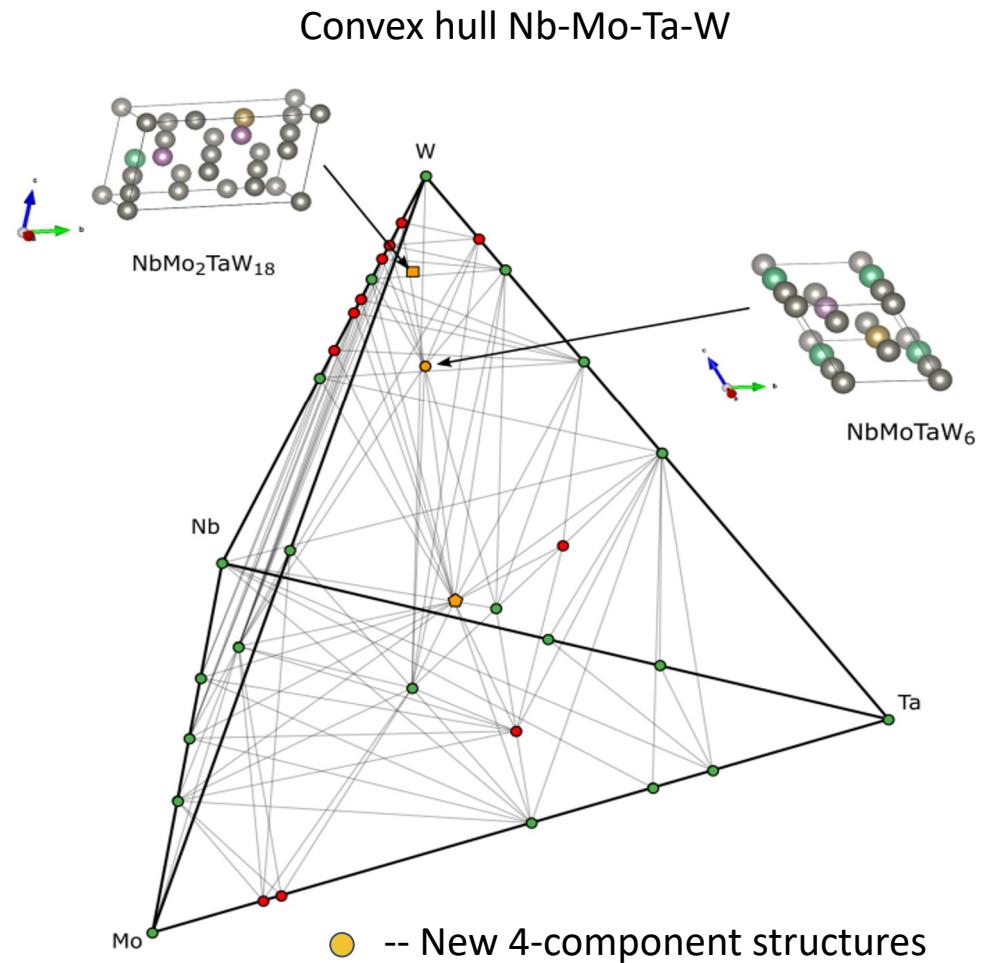
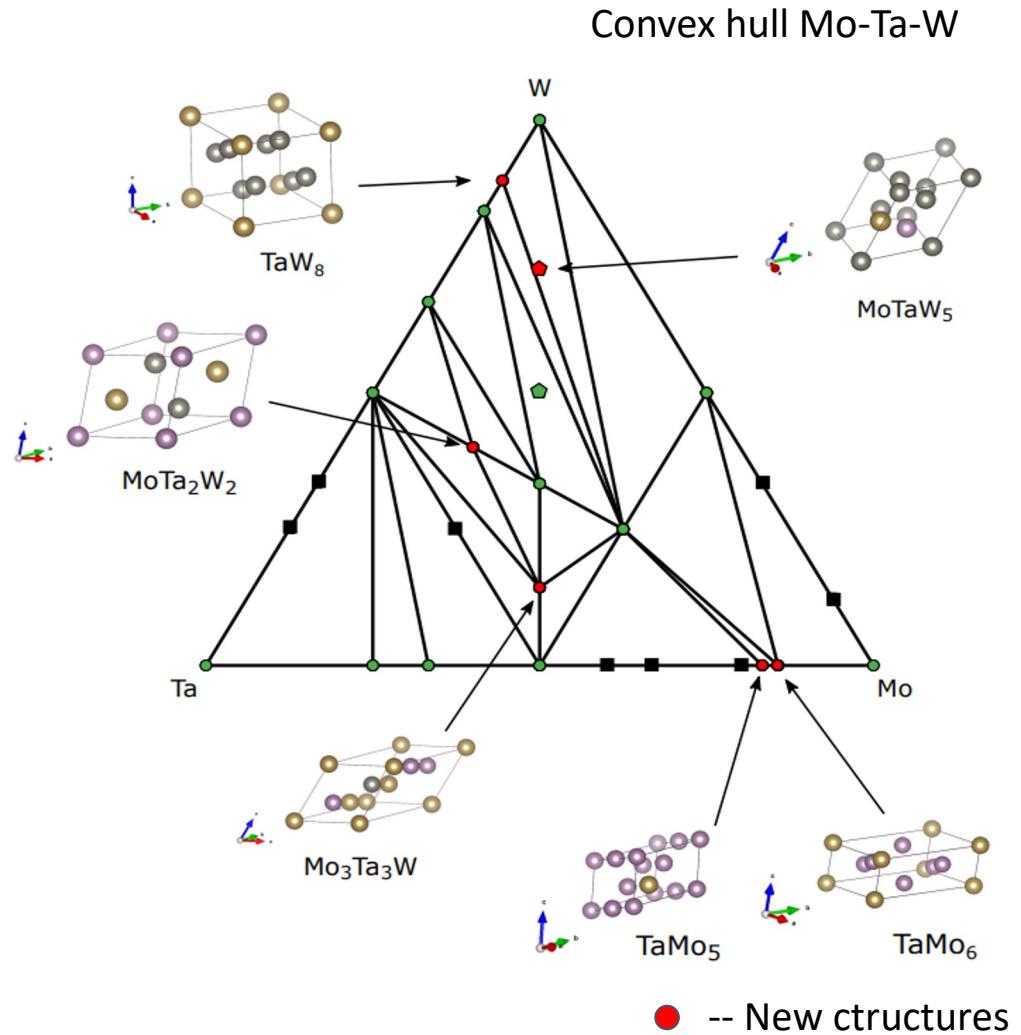
500 K
Two solid solutions coexist
Ti-Nb-Ta-C и Zr-Hf-Ta-C



New structures search : structure prediction



New structures search : structure prediction



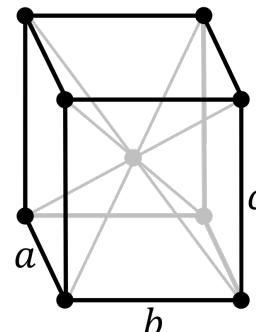
New structures search: Convex hull construction

Prediction of new stable high-entropy alloys from *ab initio* calculations with ML-potentials

1. Development of an algorithm for constructing convex hulls of stable structures for metal alloys with a given type of crystal lattice (bcc, fcc)

23 V Ванадий	
41 Nb Ниобий	42 Mo Молибден
73 Ta Тантал	74 W Вольфрам

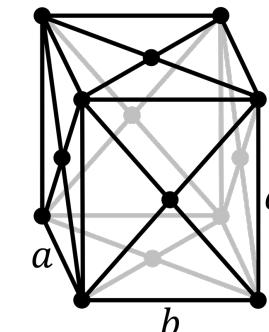
Refractory alloys
bcc



2. The search of new alloys

	29 Cu Медь
46 Pd Палладий	47 Ag Серебро
78 Pt Платина	79 Au Золото

Precious metals
fcc

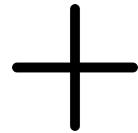


Zinkovich Victoria
Master student Sk

New structures search: Convex hull construction

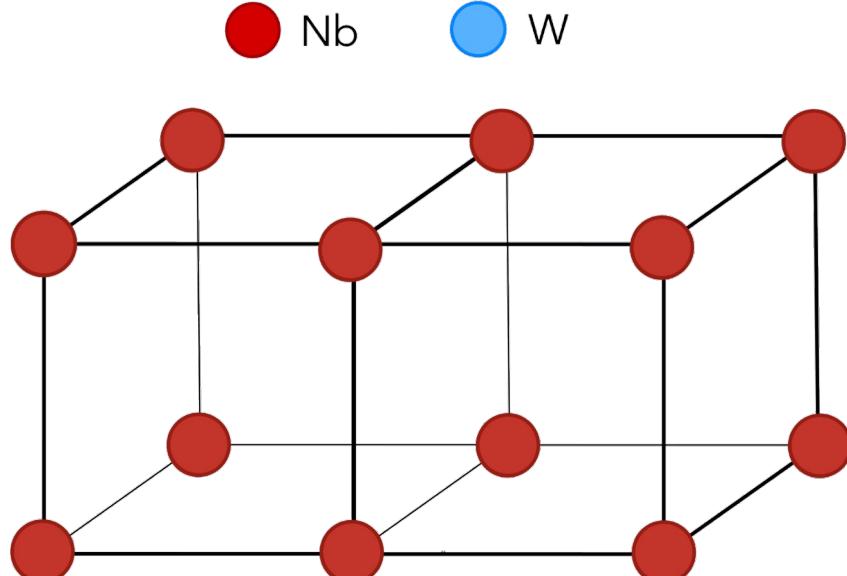
1. Brute-force method

for different types of atoms at the sites of a crystal lattice (coloring)



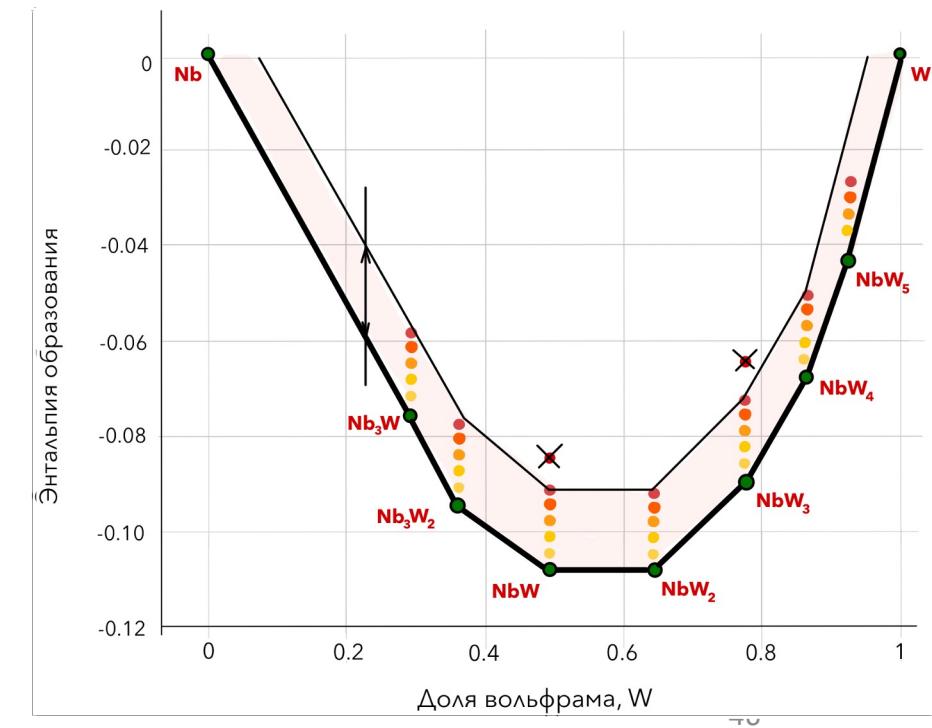
2. LRP

as an interatomic model for calculating the energy of structures



New HEA search

We start with structures with two atoms in a supercell and gradually increase the number of atoms

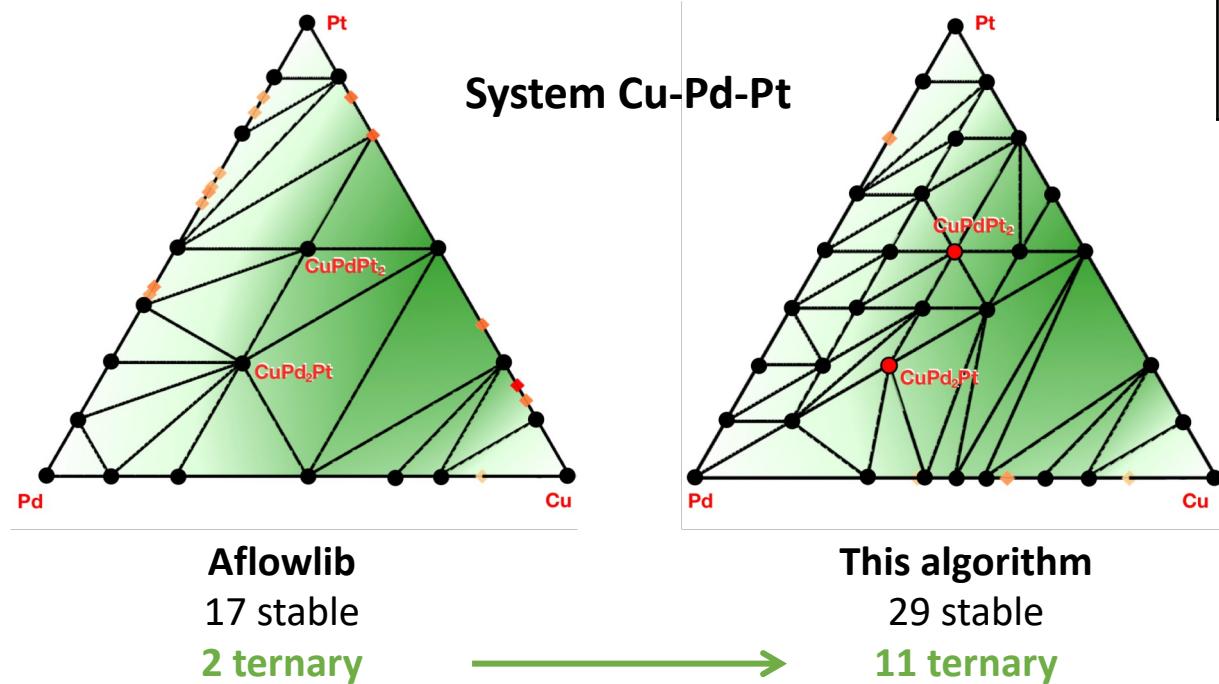


New structures search: Convex hull construction

1. The method was **tested** on six systems :

- Nb-W, Nb-Mo-W, V-Nb-Mo-Ta-W
- Cu-Pt, Cu-Pd-Pt, Cu-Pd-Ag-Pt-Au

	bcc	fcc
binary	62	19
ternary	41	59
quaternary	32	1
	$\sum = 135$	$\sum = 79$



2. As a result of the algorithm, **214 new alloys** were found that are not in the known databases (Aflow, NIST, OQMD)

6. LRP-code application

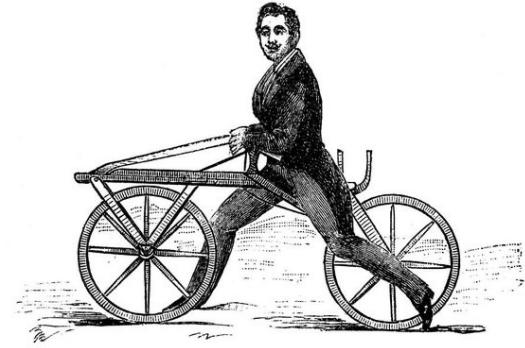


About Cython and LRP-code application

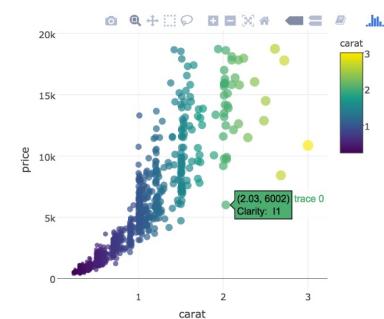
Cython is an **optimising static compiler** for both the **Python** programming language and the extended Cython programming language (based on **Pyrex**). It makes writing C extensions for Python as easy as Python itself.

Cython gives you the combined power of Python and C to let you

- write Python code that calls back and forth from and to C or C++ code natively at any point.
- easily tune readable Python code into plain C performance by adding static type declarations, also in Python syntax.
- use combined source code level debugging to find bugs in your Python, Cython and C code.
- interact efficiently with large data sets, e.g. using multi-dimensional NumPy arrays.
- quickly build your applications within the large, mature and widely used CPython ecosystem.
- integrate natively with existing code and data from legacy, low-level or high-performance libraries and applications.



```
In [5]: library(plotly)
set.seed(100)
d <- diamonds[sample(nrow(diamonds), 1000), ]
plot_ly(c = type = "scatter", mode = "markers",
        x = carat, y = price,
        color = -carat, size = -carat,
        text = -paste("Clarity:", clarity))
```



```
19. Sep 09:31 boot
21. Sep 15:50 dev
19. Sep 09:32 etc
21. Sep 15:52 home
7 30. Sep 2015 lib -> usr/lib
34 23. Jul 10:01 lib64 -> usr/lib
96 1. Aug 22:45 lost+found
396 30. Sep 2015 mnt
16 21. Sep 15:52 opt
8 21. Sep 08:15 private -> /home/encrypted
4096 12. Aug 15:37 proc
560 21. Sep 15:50 root
7 30. Sep 15:50 run
4096 30. Sep 2015 sbin -> usr/bin
6 21. Sep 2015 srv
300 21. Sep 15:51 sys
4096 12. Aug 15:45 tmp
4096 23. Jul 10:25 usr
4096 49
```