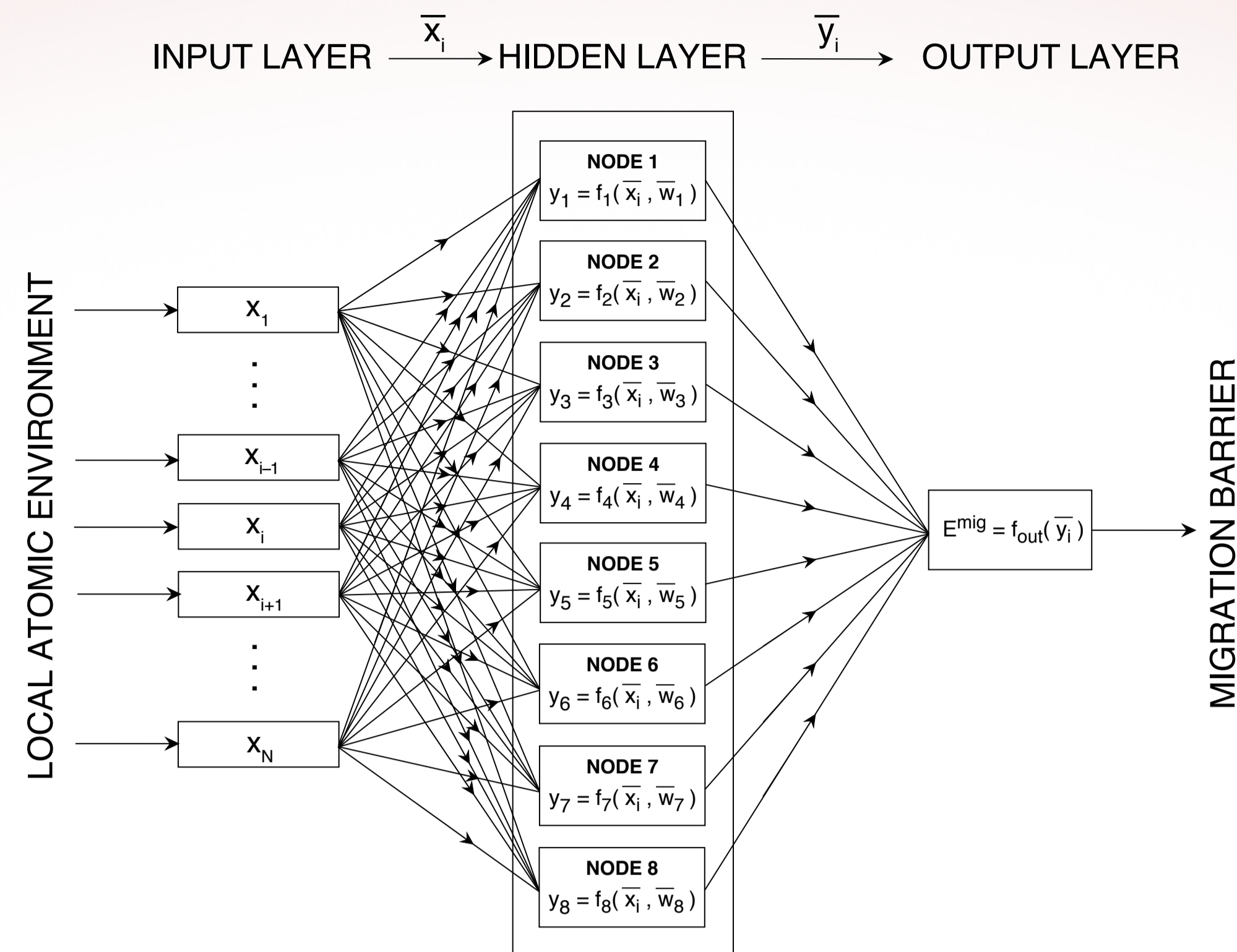


SCOPE OF THE WORK

- Improve the **prediction accuracy of atomic migration rates** in kinetic Monte Carlo (KMC) simulations of microstructure evolution.
- Migration barriers (E^{mig}) are highly **non-linear functions of the local chemical composition**. First-principles methods provide accurate calculations of E^{mig} , although at a very high computational cost.
- The amount of **reliable first-principles physical information** transferred to KMC models can be maximized by training **artificial neural networks (ANN)** on **Density Functional Theory (DFT)** databases of migration barriers.
- The proposed DFT-ANN approach has two advantages:
 - ✓ The **physical description** of the system can be considerably **enriched** with respect to traditional cohesive models.
 - ✓ As opposed to ANNs trained over interatomic potentials, DFT potentials are available for (**almost**) **any alloys**. Therefore, increasing the chemical complexity of the system can be done with little additional effort.

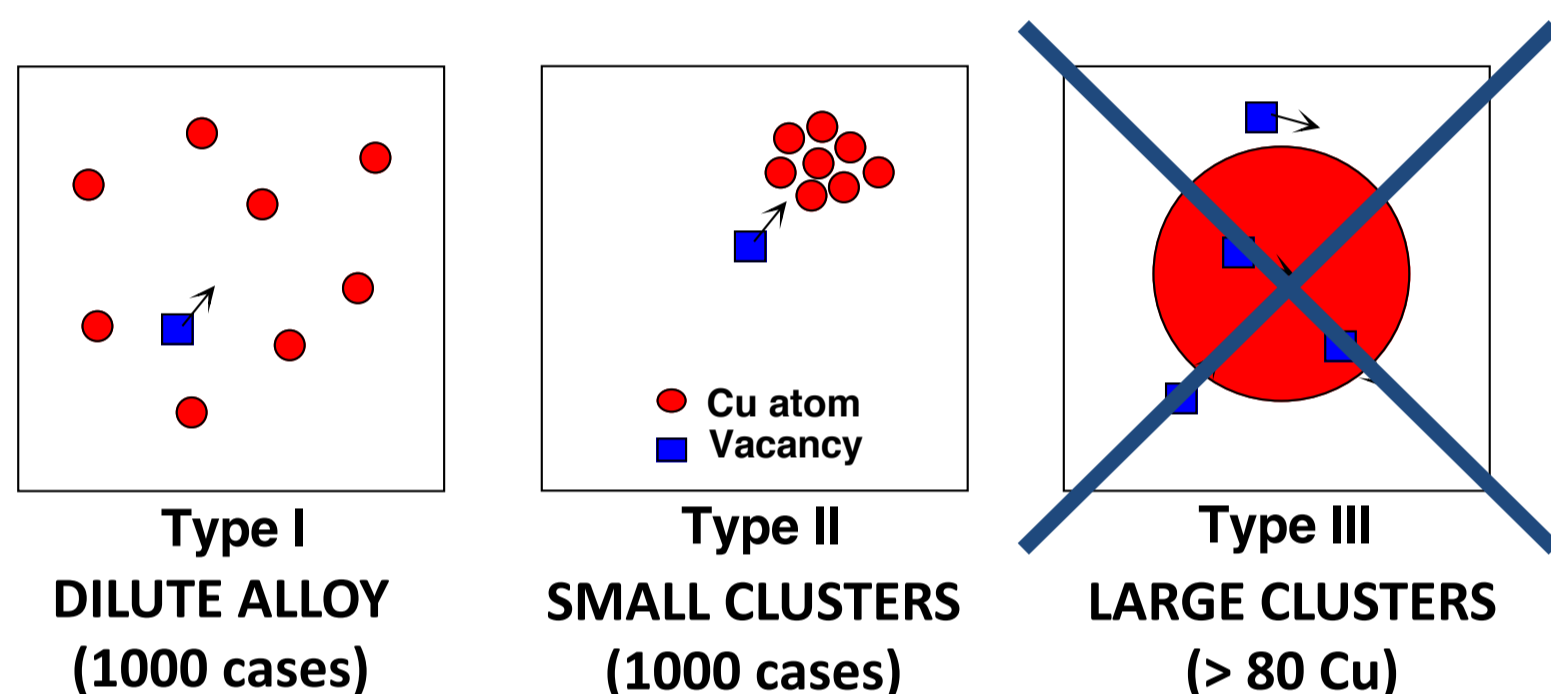
ARTIFICIAL NEURAL NETWORKS



- Mathematical structures aimed at **learning complex relationships** between given input and output variables, for which usually no simple expressions can be formulated.
- Here, ANNs are used as **regression tools** to provide a value of the migration barrier (output) related to an **unknown local chemical composition** (input). This is achieved by fitting a set of parameters in each node of the hidden layer (see figure) on a **pool of known training cases**. The larger the pool, the more precise the prediction.
- The selection of training cases must be **representative of all possible chemical compositions** that can be encountered during the simulation.
- Once trained, the ANNs provide accurate predictions in a very computationally efficient way, and can be applied *on the fly* to atomistic simulations.
- Previously, ANNs based on EAM interatomic potentials have been successfully applied to predict KMC migration barriers⁽¹⁾. This work attempts for the first time to **replace the EAM pool with DFT**.

A) DFT-ANN MIGRATION ENERGY PREDICTION⁽²⁾

- A database of **2000 migration barriers** is obtained by means of DFT-NEB calculations with the Vienna *ab initio* Simulation Package (VASP).
- The database **maximizes** the variety of the local atomic environments (LAE) around the hopping vacancy and the distribution of migration energy values.
- The samples cases are selected within **two out of three LAE types** occurring during Cu precipitation:

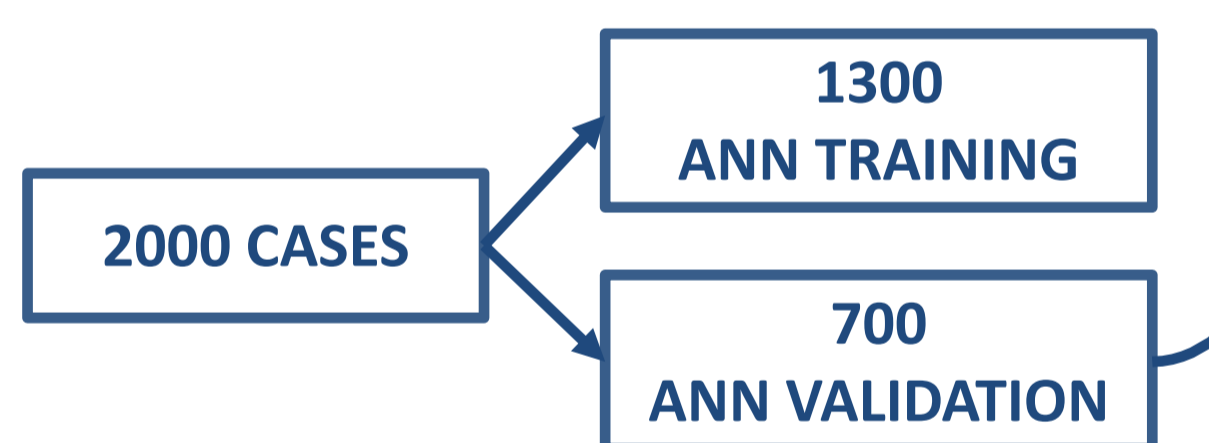
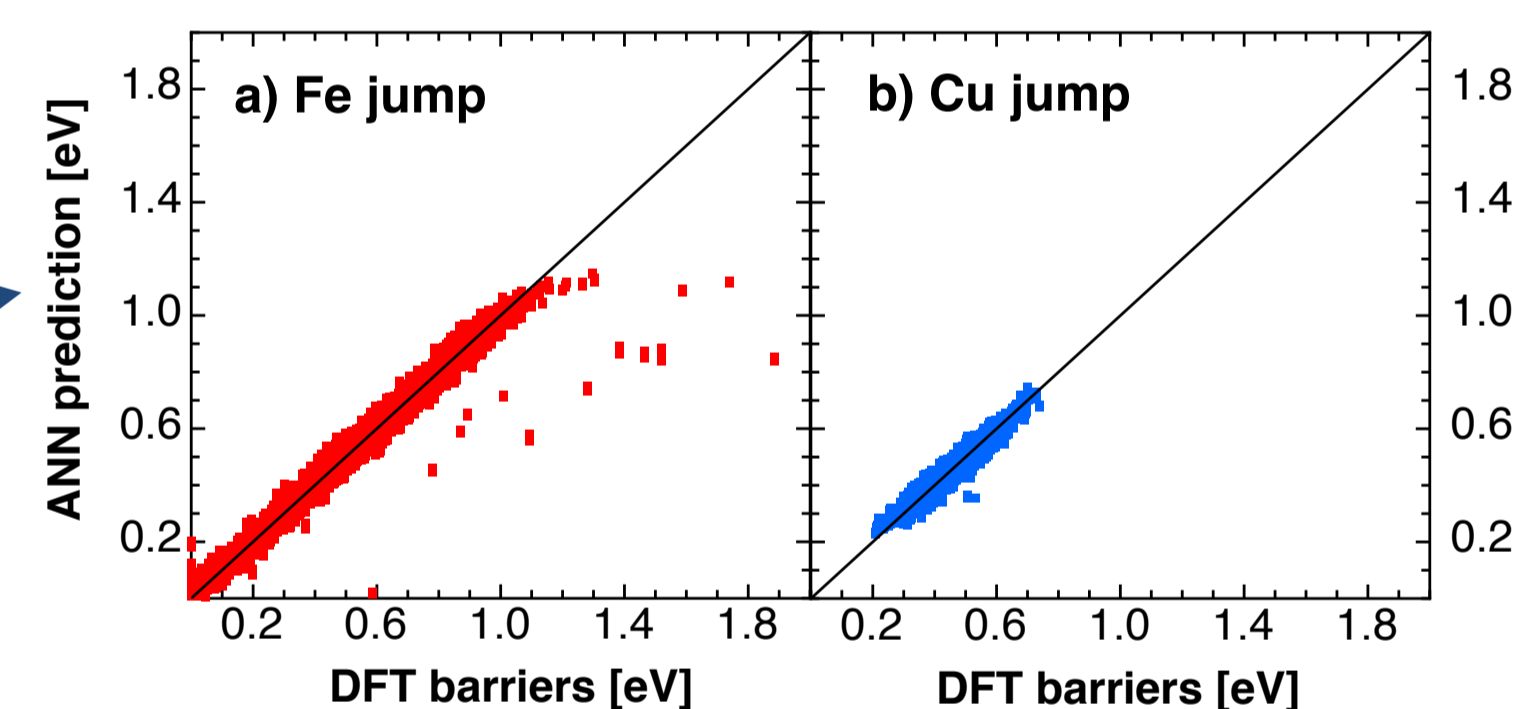


10 million core-hours

Type-III cases are **not included** because of DFT cell size limitations (250 atoms).

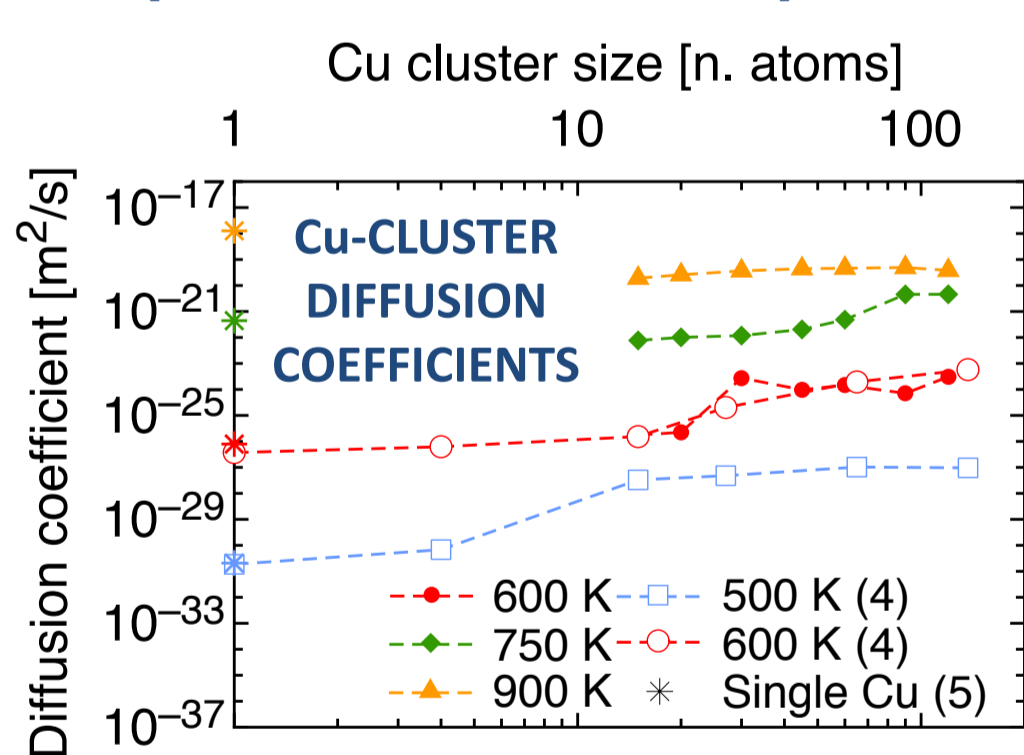
- **Computational cost** (≈ 5000 core-hours/NEB) compels to find optimum between:
 - ✓ NEB accuracy (PAW-PBE, $3 \times 3 \times 3$ kpoints, 300 eV cutoff)
 - ✓ DFT cell size (250 atoms)
 - ✓ Amount of sample cases (2000 barriers)

ANN PREDICTION PERFORMANCE
(average error: 0.023 eV)



THERMAL AGING OF FeCu ALLOYS (HYBRID KMC)

- **Hybrid atomistic-object KMC** approach⁽¹⁾ (Cu clusters above 15 atoms are treated as objects).
- **Object part** is parameterized by AKMC simulations driven by the same neural network.
- Simulation **time is rescaled** to match the equilibrium vacancy concentration, but it is **not fitted on experiments**.
- **Improved prediction** of precipitation kinetics with respect to previous EAM-ANN work⁽¹⁾:
 - ✓ Very satisfactory agreement with exp. timescale
 - ✓ Well reproduced evolution of the 1.34%Cu alloy

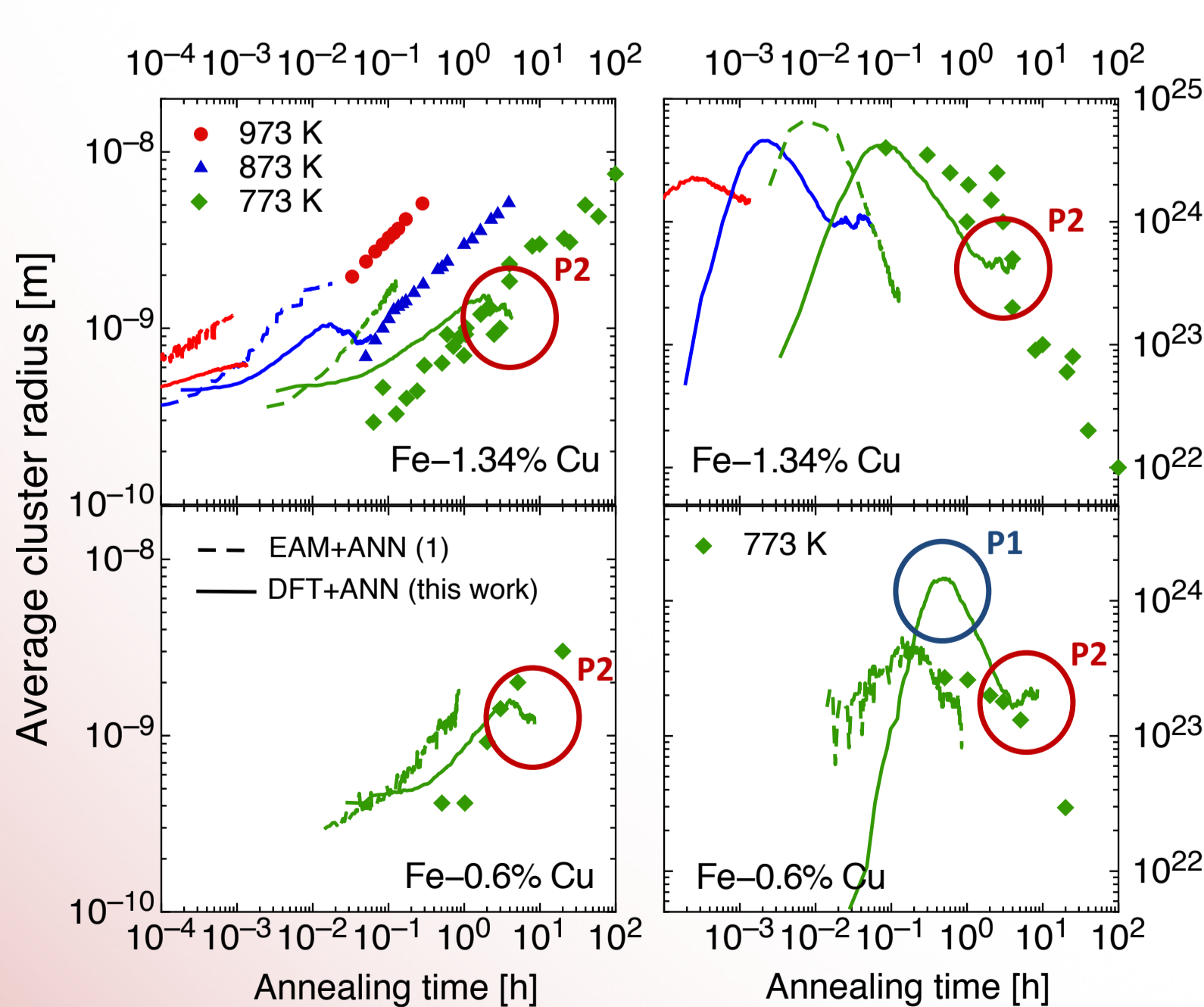


Increasing diffusion coefficient with cluster size shows that precipitation is mainly **driven by coarsening of medium and large clusters**.

SUCCESS!
DFT properties fully transferred to KMC

Model limitations:
P1: NO CONTROL OVER THERMODYNAMICS
P2: SMALL DFT CELL SIZE

- P1** - Excessive cluster density in 0.6%Cu alloy \rightarrow **underestimation of Cu solubility** (due to wrong DFT prediction of solution energy).
- P2** - Late coarsening phase suddenly stops \rightarrow this stage is not correctly reproduced due to the **lack of type-III cases** in the training database.



B) DFT-ANN COHESIVE MODELS⁽³⁾ (POTENTIALS)

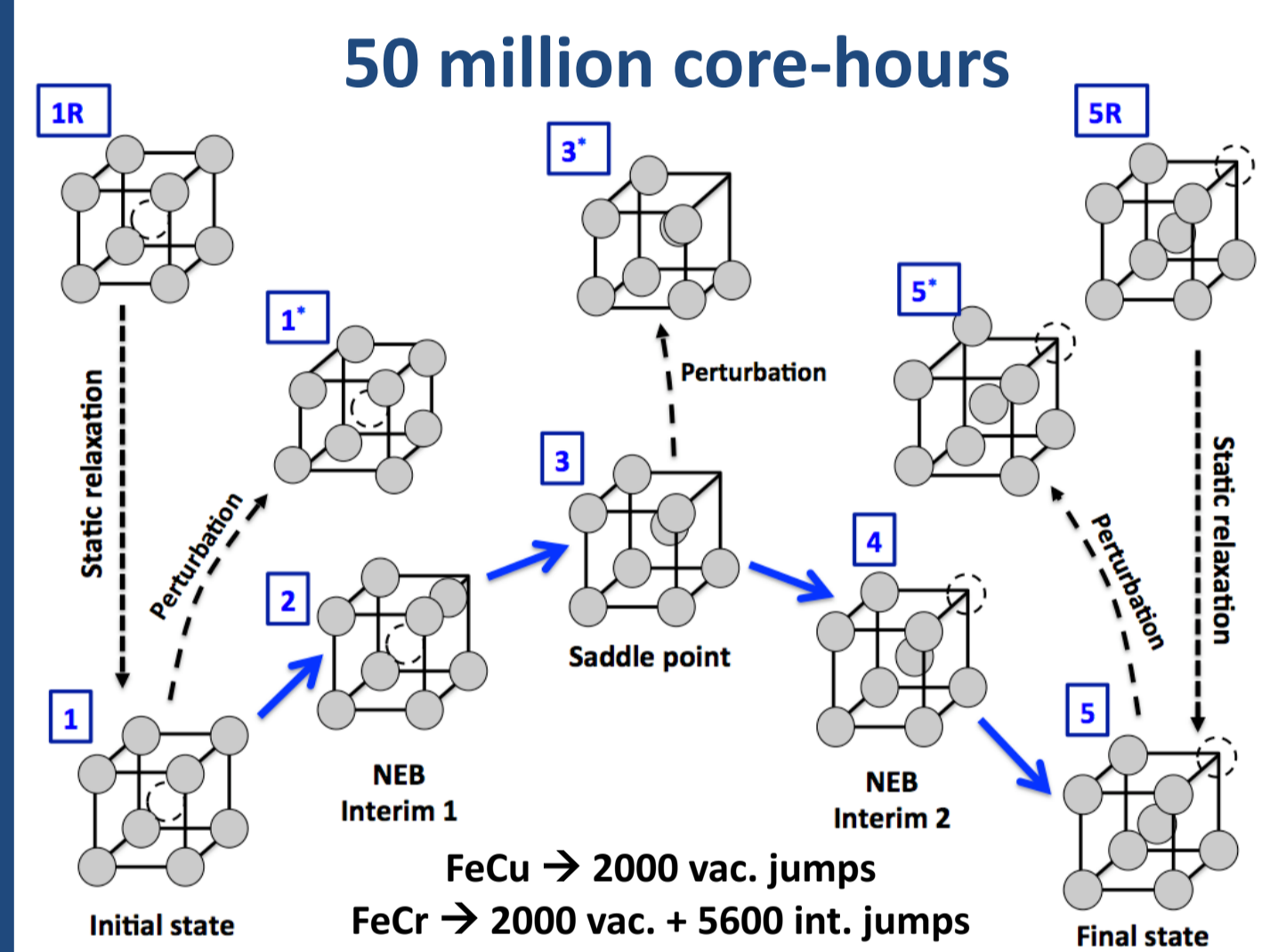
- Objective: exploit DFT database **more extensively**, to predict not only migration barriers but also the **total energy** associated to a given atomic configuration.

$$\text{TOTAL ENERGY} \quad E_{\text{tot}} = \sum_x \sum_{a_x} E_x^{\text{ANN}} \rho(a_x) \quad x = \text{Fe, Cr, Cu} \quad a_x = \text{atom of species } x$$

- Each E_x^{ANN} provides an **estimate of the energy assigned to each atom** of species x , and is calculated by a dedicated neural network.

- $\rho(a_x)$ is the **atomic density** at the position of atom a_x , written as a series of spherical harmonics.

$$\rho(a_x)(r, \theta, \phi) = \sum_{n=1}^{\infty} \sum_{l=0}^n \sum_{m=-l}^l C_{nlm}^{(a)} R_n(r) Y_{lm}(\theta, \phi)$$



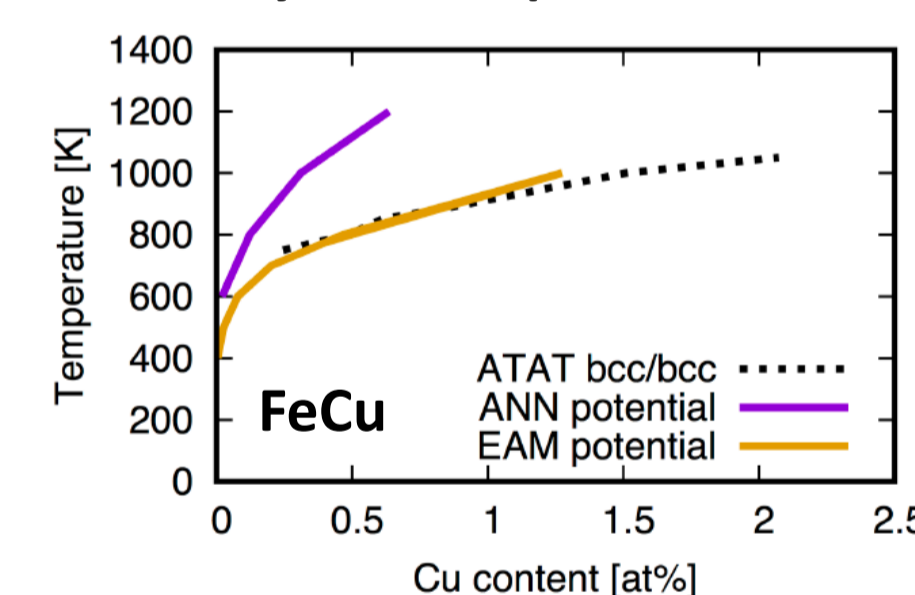
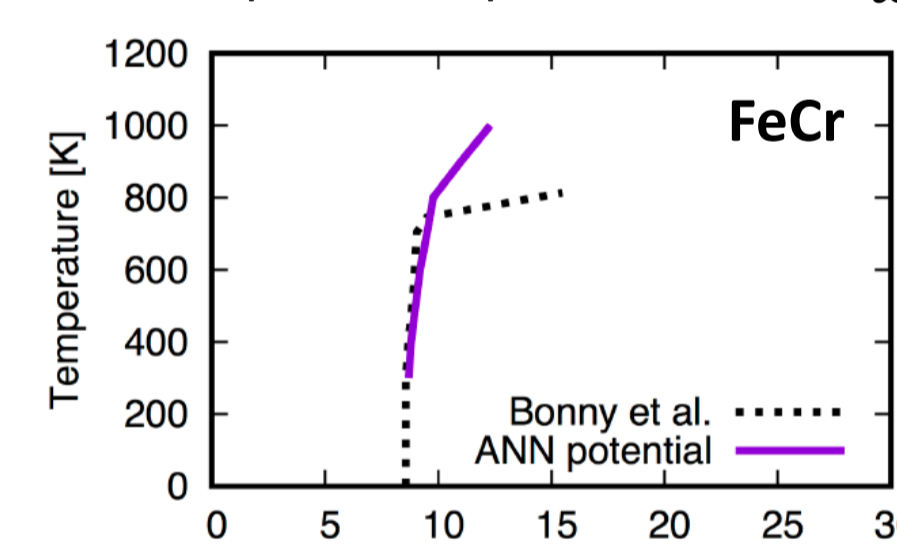
DFT datasets

- Energy of **fully relaxed equilibrium configurations** (1, 5).
- Saddle-point** energies of selected migration events (3).
- Energies of several **intermediate relaxation steps** (2, 4, from 1R to 1, from 5R to 5).
- Energies of **randomly perturbed configurations** (1*, 3*, 5*).

Thanks to set IV, the neural networks learn that the energy should increase when departing from equilibrium states.

RIGID-LATTICE POTENTIALS

- Metropolis MC prediction of E_{tot} at **thermodynamic equilibrium**, based on ANN trained on dataset I.



PREDICTED PHASE DIAGRAMS

- FeCr \rightarrow satisfactory
- FeCu \rightarrow confirmed underestimated solubility
- Now we have a mean to control the thermodynamic properties of the system!

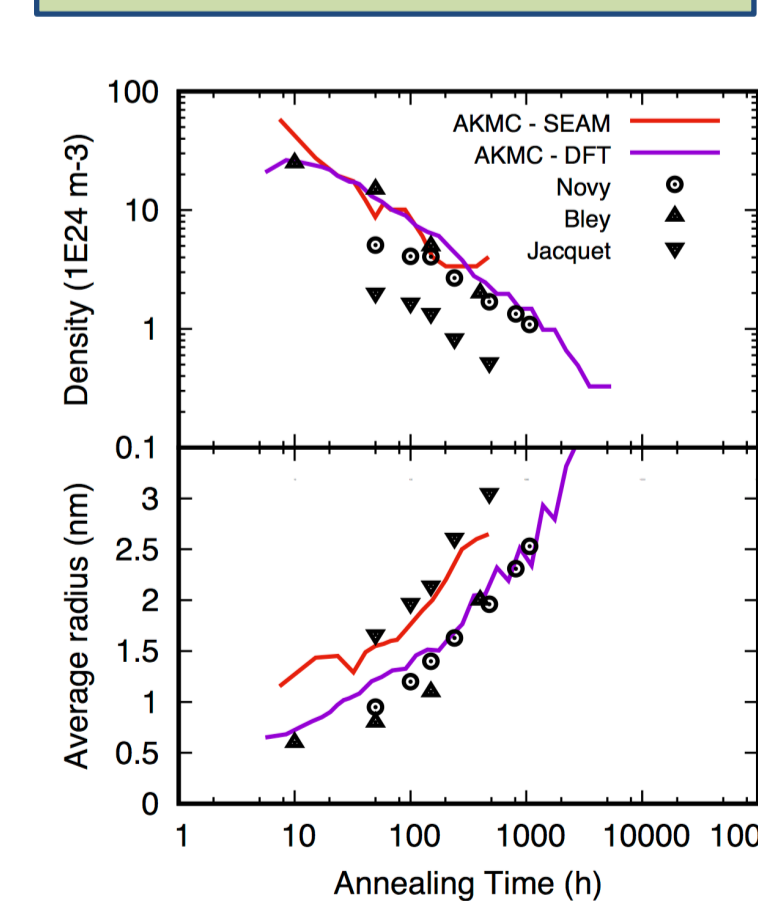
Fe-20%Cr THERMAL AGING

- AKMC simulations (FeCr + 1vac) at 500 °C.
- Migration barriers obtained with **two separate neural networks**:

$$E^{\text{mig}} = \frac{1}{2} \Delta E_{\text{tot}}^{\text{ANN}} + E_{\text{act}}^{\text{ANN}}$$

End-state energy difference from rigid-lattice potentials (set I)

Saddle-point energy (set II)



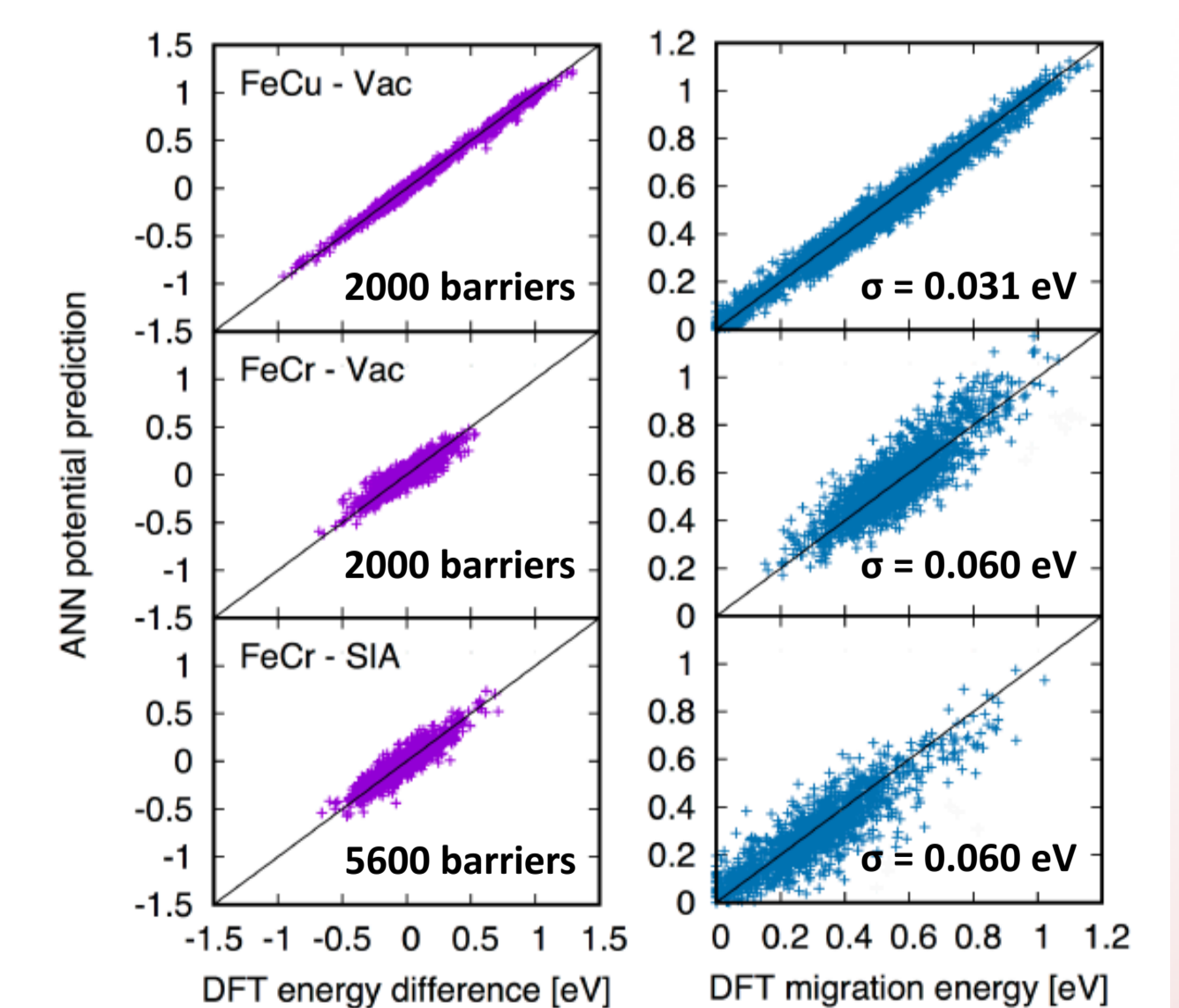
Very good agreement with experiments!

Results analyzed with the eyes of atom probe:

- ✓ 60% of atoms are **randomly discarded**.
- ✓ **Random distortions** in atomic coordinates due to the measurement.

LATTICE-FREE POTENTIALS

- All sets (I to IV) are used. No explicit force fitting, but **excellent force prediction** thanks to dataset IV.
- Application: **re-calculation of migration barriers**. Very good comparison with DFT barriers!



Trustworthy tools to extend the DFT database with **more sample cases and larger simulation cells!** (e.g. 30000 SIA jumps in FeCr).

CONCLUSIONS

- This new DFT-ANN approach can ensure an accurate physical description of the multifold properties of an alloy in KMC and other mesoscale simulations, maximizing the amount of information transferred from first-principles calculations. It is therefore a very promising support to microstructure evolution simulations of alloys in a wide range of applications and conditions.
- The computational challenges due to the heavy requirement of DFT calculations can be addressed by combining several ANNs to create accurate rigid-lattice and lattice-free potentials. The splitting between equilibrium and saddle-point ANNs allows for the choice of the most appropriate methods to target thermodynamic and kinetic properties separately, methods that can possibly differ from one another.
- The following planned application is modeling the microstructure evolution of FeMnNi alloys, representative of reactor pressure vessel steels.

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