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# **Ab initio-trained neural-network driven kinetic Monte Carlo simulations of microstructure evolution of iron alloys**

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# Outline

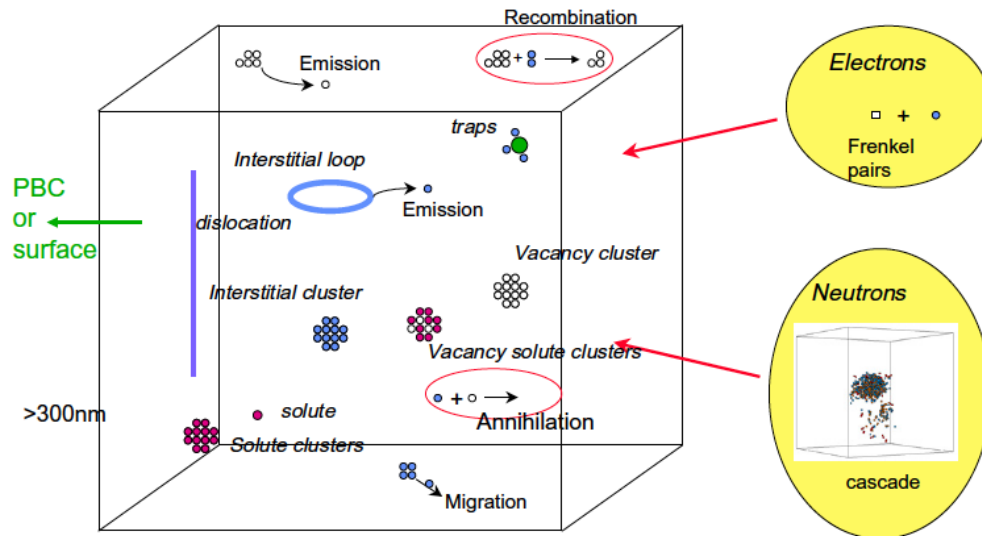
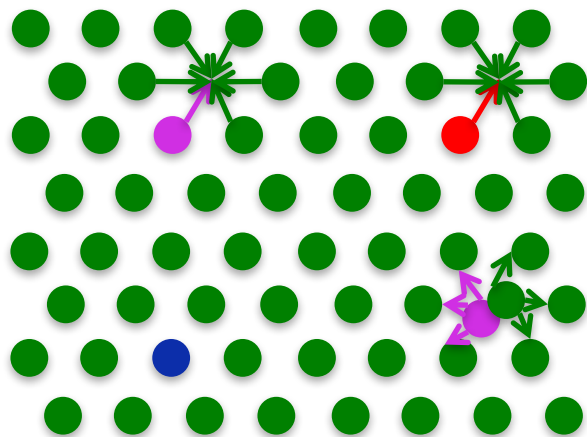
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- Kinetic Monte Carlo method
- DFT-ANN-based prediction of migration barriers
  - Thermal aging of dilute FeCu alloys
- DFT-ANN prediction of cohesive energy and migration barriers
  - Rigid-lattice potentials (phase diagrams)
  - Thermal aging of Fe-20%Cr alloys
  - Lattice-free potentials
- Conclusions

# Kinetic Monte Carlo method

Courtesy of C. Domain (EDF).

- System evolution driven by defect jumps.



- One transition is randomly selected according to the transition rates.

$$\Gamma_j = \Gamma_0 \exp\left(-\frac{E_j^{\text{mig}}}{k_B T}\right)$$

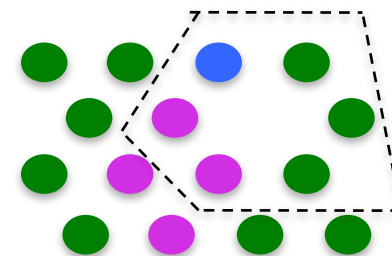
- One transition is randomly selected according to the transition rate distribution

- Simulation time is advanced with the residence-time algorithm.

$$\Delta t = \frac{1}{\sum_j P_j}$$

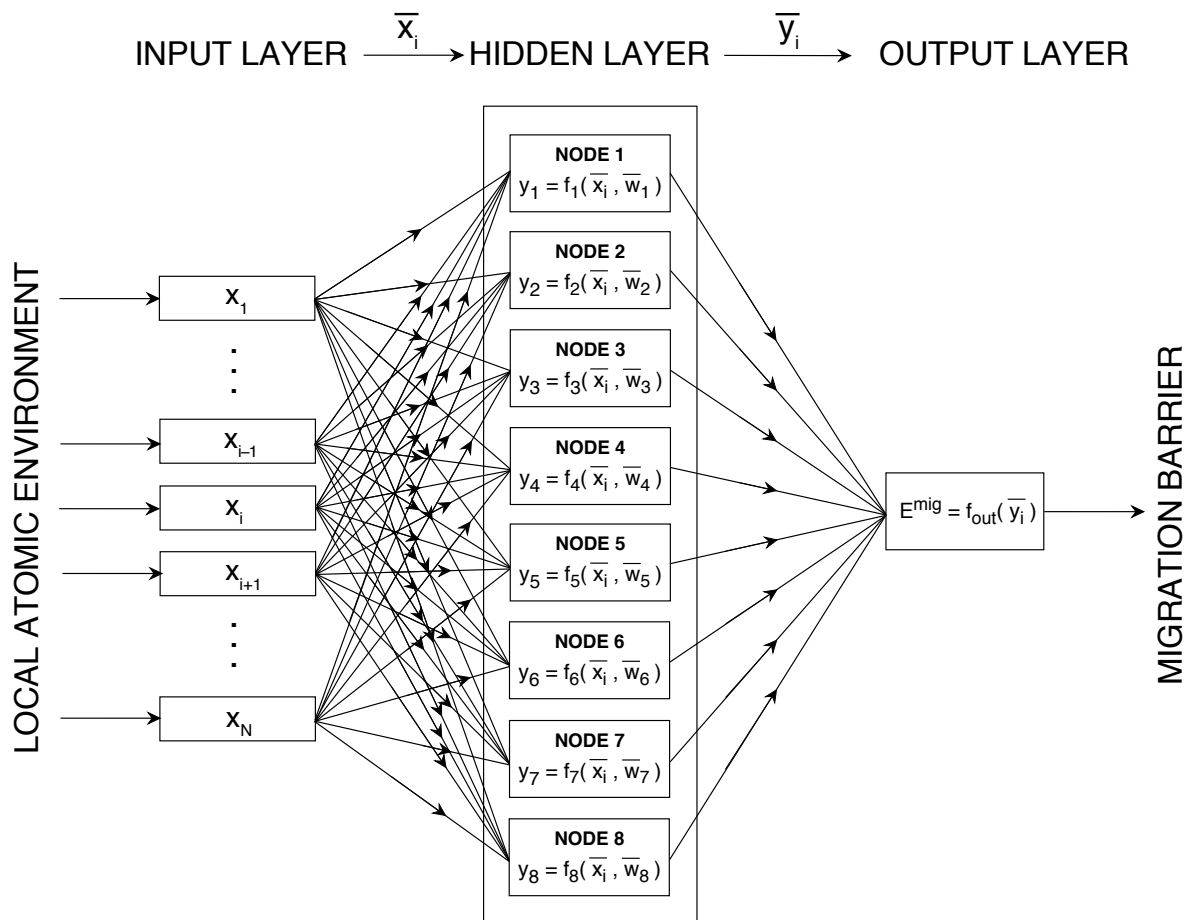
## PROBLEM

- Transition rates depend on the composition of the local chemical environment → impossible to calculate them all.



- Difficult to achieve a fully reliable physical description of the system with traditional cohesive models or saddle-point energy models.

# Artificial neural network approach



- Mathematical structures to learn **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 related to an **unknown local chemical composition**, by fitting a set of parameters in each node of the hidden layer on a **pool of known training cases**.

- The selection of training cases must be **representative of all possible chemical compositions**.

- ANNs provide accurate predictions in a very computationally efficient way, and can be applied *on the fly* to atomistic simulations.

# Scope of the work

- Improve the **prediction accuracy of atomic migration rates** in kinetic Monte Carlo (KMC) simulations of microstructure evolution.
- Migration barriers ( $E^{\text{mig}}$ ) are highly **non-linear functions of the local chemical composition**. First-principles methods provide accurate calculations of  $E^{\text{mig}}$ , 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.

# Interatomic potentials vs ANN

## Interatomic potentials (IP)

- Fitted on experimental/*ab initio* data.
- Highly customizable to desired specific properties.
- Computationally cheap.
- System-specific.
- Fitting is a time-consuming and non-trivial/non-linear task.
- Difficult to be accurate on many alloy properties at once.

N. Castin *et al.*, *J. Chem. Phys.* **132**, 074507 (2010)

N. Castin *et al.*, *J. Chem. Phys.* **135**, 064502 (2011)

N. Castin *et al.*, *J. Nucl. Mater.* **429**, 315 (2012)

## Density Functional Theory (DFT)

- Provides detailed description of alloy thermodynamic and kinetic properties.
- Can be applied to complex multicomponent alloys with little complexity addition.
- Computationally expensive (for large amount of configurations).

**EAM → DFT**

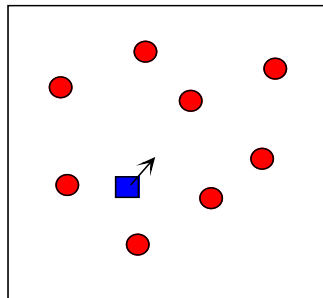
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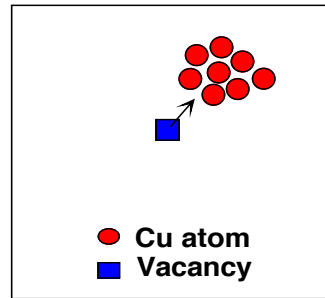
# Simulation of FeCu thermal aging

- 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:



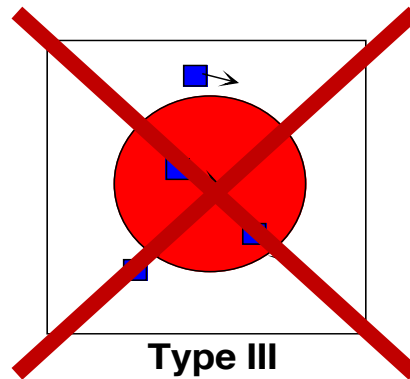
Type I

**DILUTE ALLOY**  
**(1000 cases)**



Type II

**SMALL CLUSTERS**  
**(1000 cases)**



Type III

**LARGE CLUSTERS**  
**(> 80 Cu)**

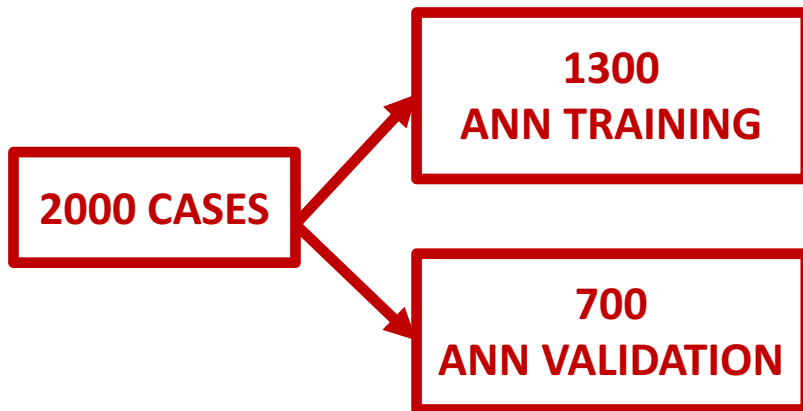
Type-III cases are **not included** because of DFT cell size limitations (250 atoms).



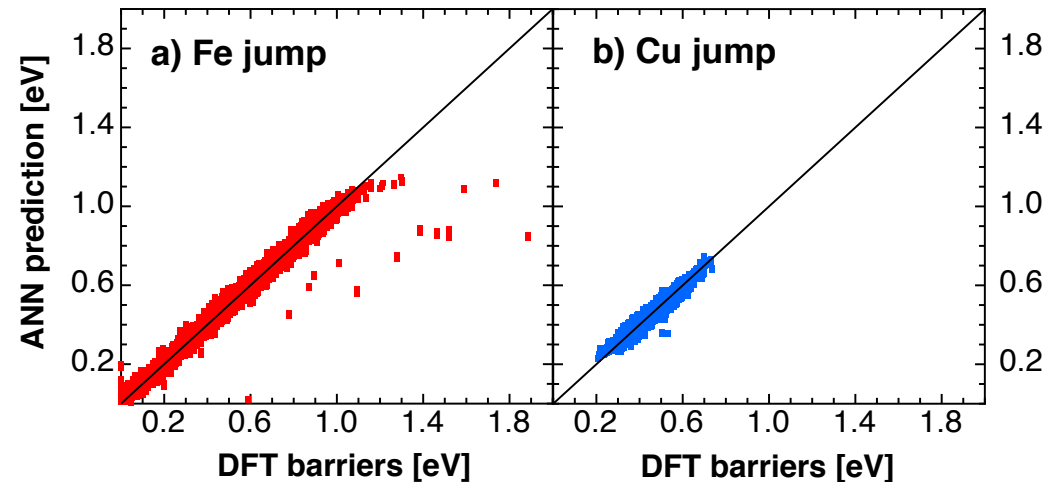
# DFT database

- **Computational cost** ( $\approx 5000$  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 migration barriers)as well as to perform a clever careful choice of the samples in the database.

10 million core-hours



**ANN PREDICTION PERFORMANCE**  
(average error: 0.023 eV)

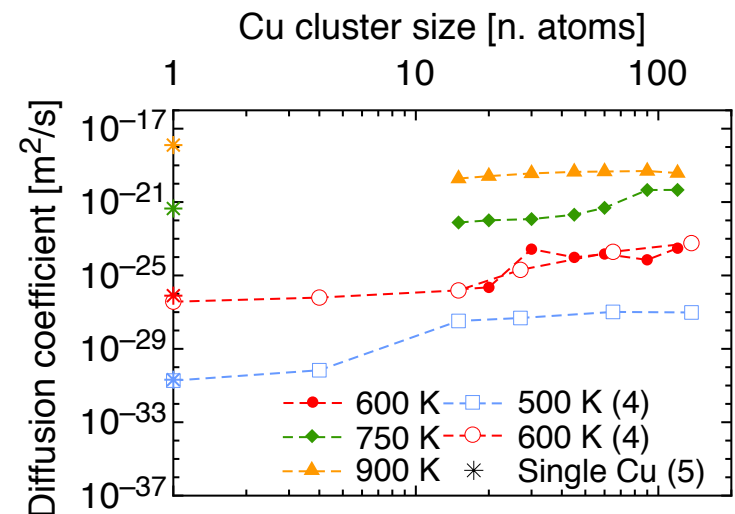


# Hybrid KMC simulations of thermal aging

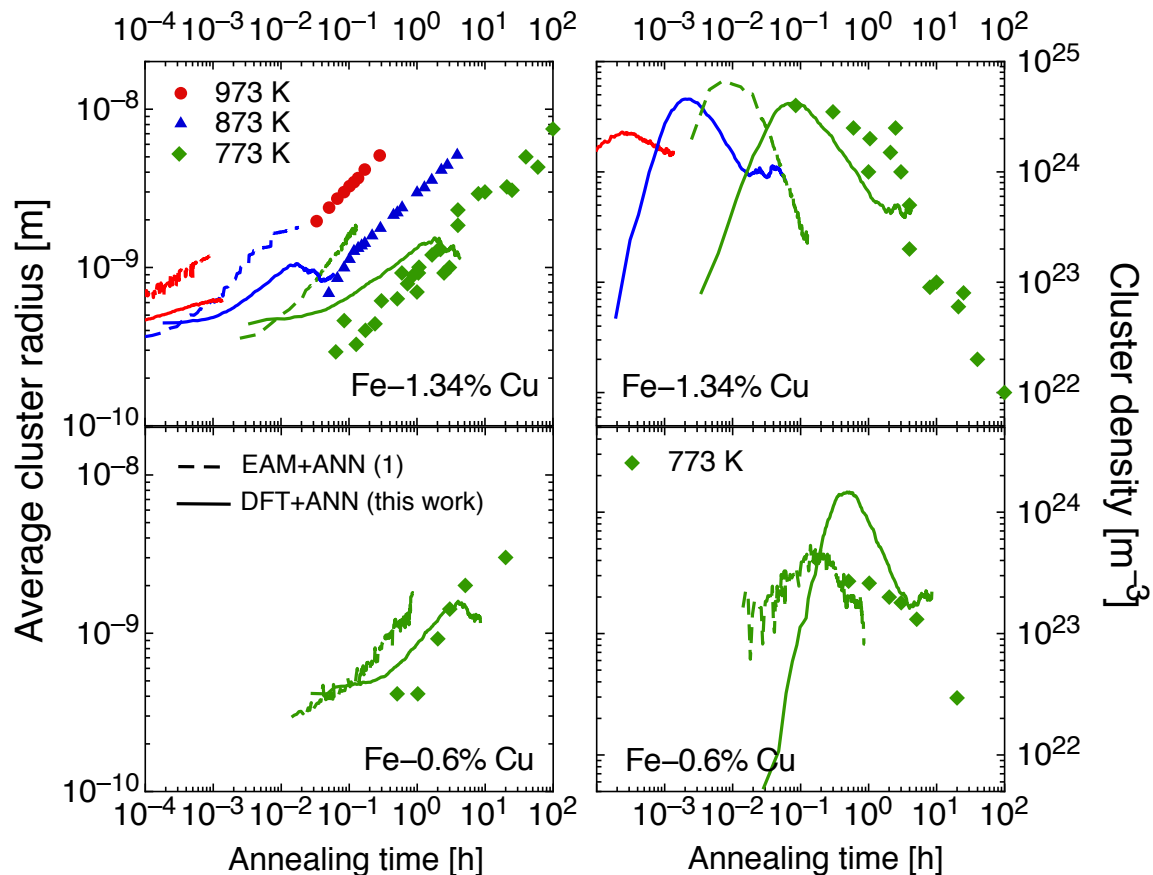
- **Hybrid atomistic-object KMC** approach<sup>(1)</sup> (Cu clusters above 15 atoms are treated as objects).
- **Object part** is parameterized by AKMC simulations driven by the same neural network.
- Simulation **time is rescaled** to match the equilibrium vacancy concentration, but it is **not fitted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

## Cu-CLUSTER DIFFUSION COEFFICIENTS



# Thermal aging results



Excellent timescale matching!

**P1** - Excessive cluster density in 0.6%Cu alloy → **underestimation of Cu solubility** (due to wrong DFT prediction of solution energy).

**P2** - Late coarsening phase suddenly stops → this stage is not correctly reproduced due to the **lack of type-III cases** in the training database.



**SUCCESS!**  
DFT properties fully transferred to KMC



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

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# DFT-ANN potentials

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

**TOTAL ENERGY**

$$E_{\text{tot}} = \sum_x \sum_{a_x}^{N_{a_x}} E_x^{\text{ANN}} \rho(a_x)$$

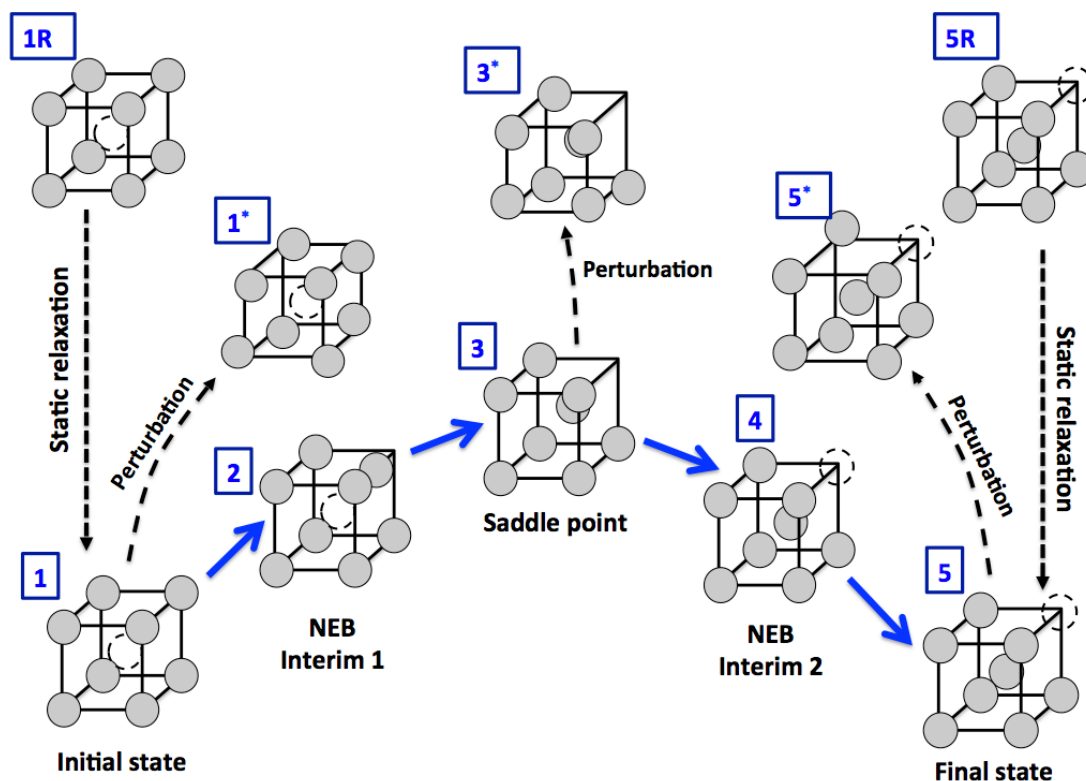
$x = \text{Fe, Cr, Cu}$   
 $a_x = \text{atom of species } x$

- Each  $E_x^{\text{ANN}}$  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.

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

# DFT database sets

50 million core-hours



FeCu  $\rightarrow$  2000 vac. jumps  
FeCr  $\rightarrow$  2000 vac. + 5600 int. jumps

## DFT datasets

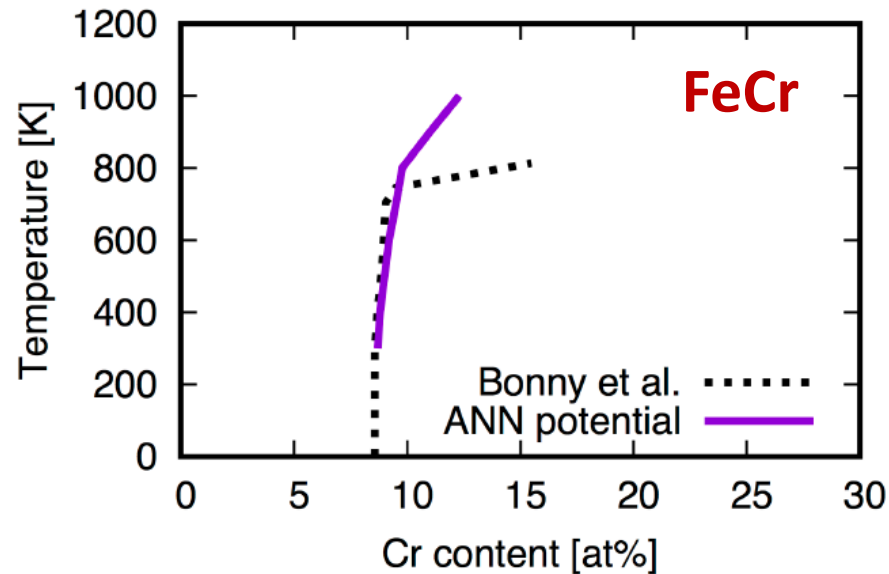
- I. Energy of **fully relaxed** equilibrium configurations (1, 5).
- II. **Saddle-point** energies of selected migration events (3).
- III. Energies of several **intermediate relaxation steps** (2, 4, from 1R to 1, from 5R to 5).
- IV. Energies of **randomly perturbed** configurations (1\*, 3\*, 5\*).

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

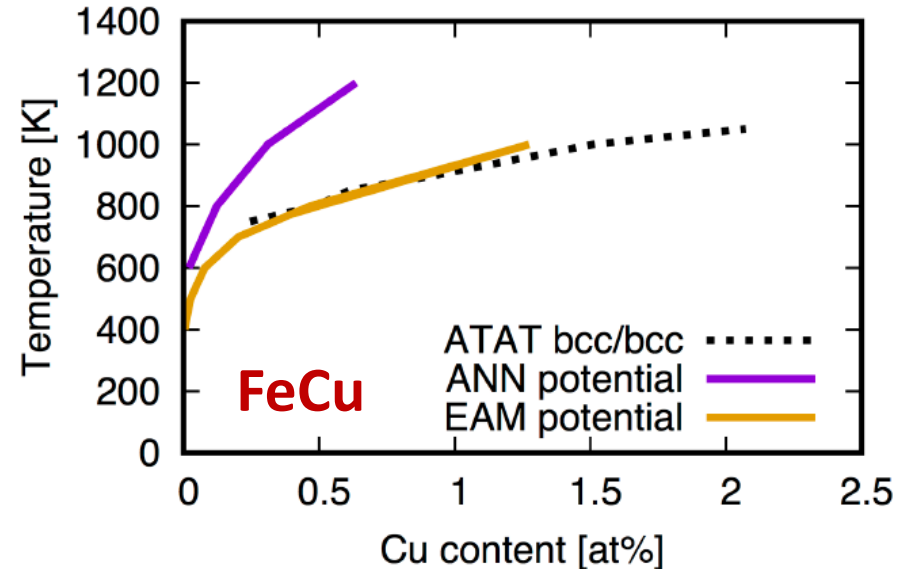
# Rigid-lattice potentials

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

## PREDICTED PHASE DIAGRAMS (DATASET I)



Satisfactory! (missing  $\sigma$  phase)



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**:

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

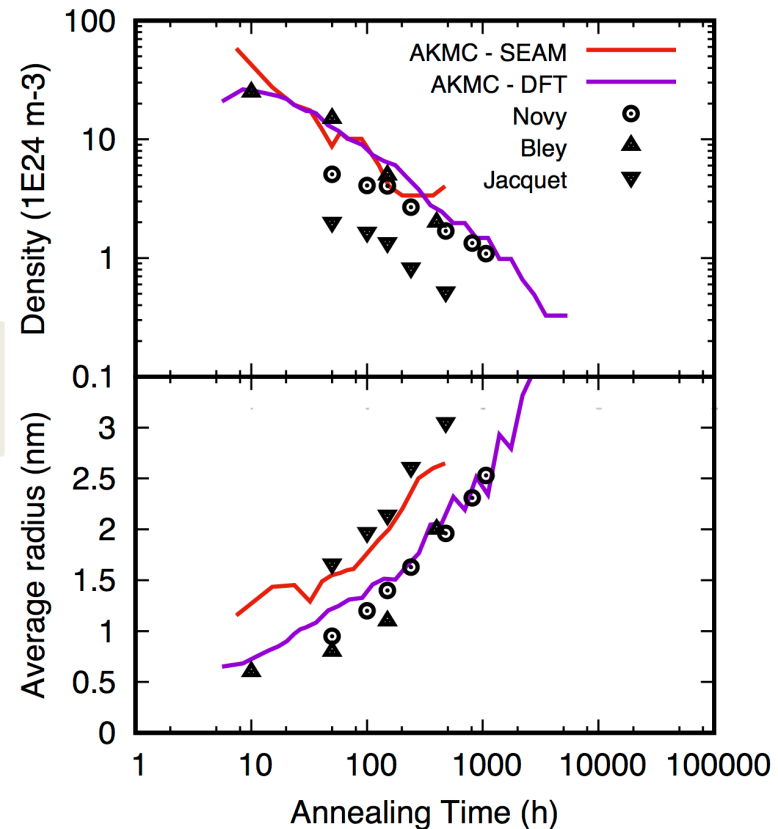
ANN 1 (green) and ANN 2 (blue) are indicated above the terms.

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

Saddle-point energy (set II)

Results analyzed **with the eyes of atom probe**:

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

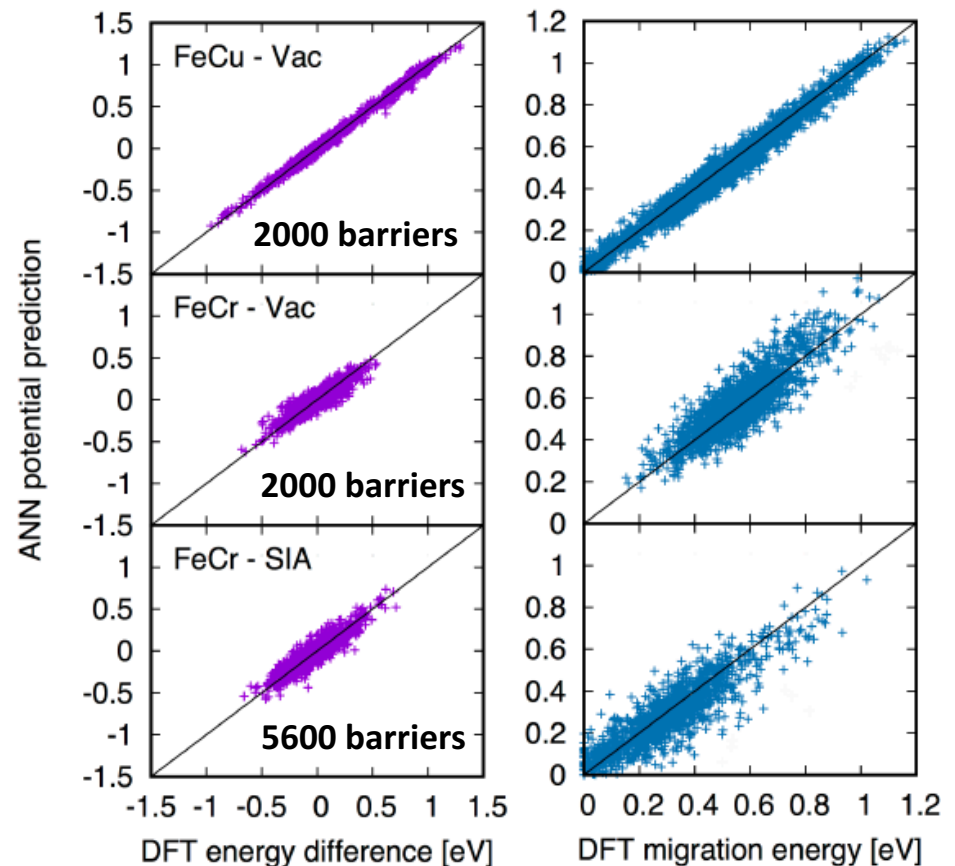




# Lattice-free potentials

- 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

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- 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.

**THANKS FOR YOUR ATTENTION!**

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