BRITS-16 MPIPKS
12-15/9 2016
Dresden, Germany

Ab initio-trained neural-network driven Monte Carlo simulations in iron alloys

Pär Olsson[1], Luca Messina[1,2], Nicolas Castin[3], Christophe Domain[4], Roberto C. Pasianot[5]

[1] KTH Royal Institute of Technology, Stockholm, Sweden
[2]DEN-Service de Recherche de Métallurgie Physique, CEA, Université Paris-Saclay, France
[3] SCK•CEN, Nuclear Material Science Institute, Mol, Belgium
[4] EDF-R&D, Matériaux et Mécanique des Composants, Moret-sur-Loing, France
[5] Consejo Nacional de Investigaciones Científicas y técnicas (CONICET), Buenos Aires, Argentina

Financial support from:
- **Vattenfall AB**
- **Göran Gustafsson Stiftelse**
- **SOTERIA project**

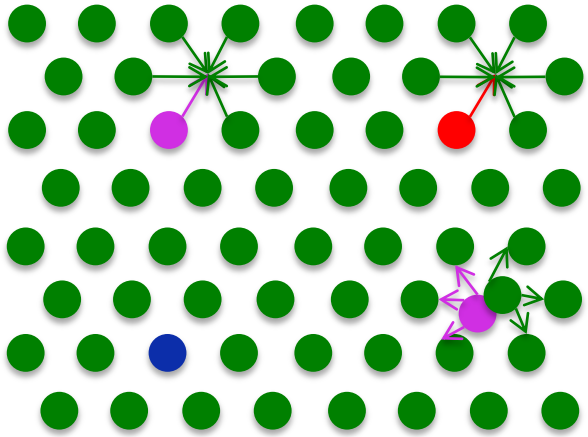
polsson@kth.se
messina@kth.se

Outline

- Introduction and motivation
- Neural networks
- Thermal ageing of FeCu alloys
- Advanced modelling of FeCu and FeCr alloys
- Conclusions

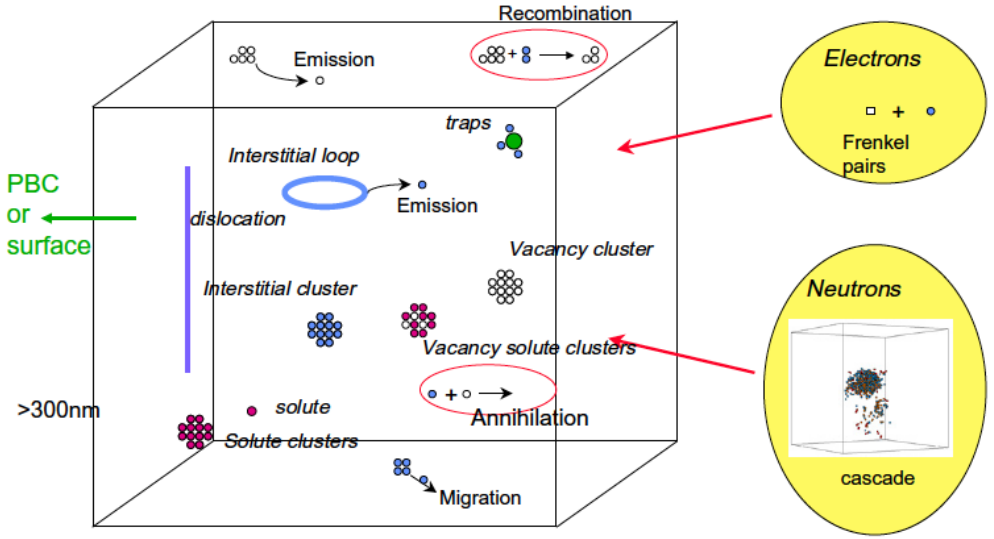
Simulating microstructure evolution

ATOMISTIC KMC



Atomic transition rates

OBJECT KMC



Object event rates, obtained by AKMC

- Microstructure evolution driven by **point-defect migration**
- Migration barriers depend on **local atomic environment** (LAE)
- KMC samples full trajectory of the studied system if all rates are included and transitions are rare events

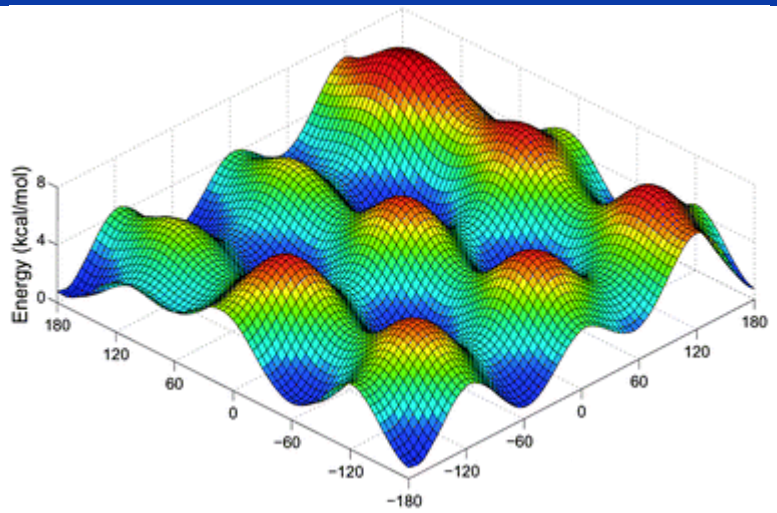
$$\Gamma = \Gamma_0 \exp\left(\frac{E^{\mathrm{mig}}}{k_{\mathrm{B}}T}\right)$$

$$\Delta t = -\frac{\ln(c)}{R}$$

Kinetic Monte Carlo

- The potential energy surface should drive the evolution
- How do we accurately represent a complicated **alloy** PES?
- Canonical idea is to simplify it according to some local model

$$E_a = E^{mig} = E^{mig}(\text{LAE})$$

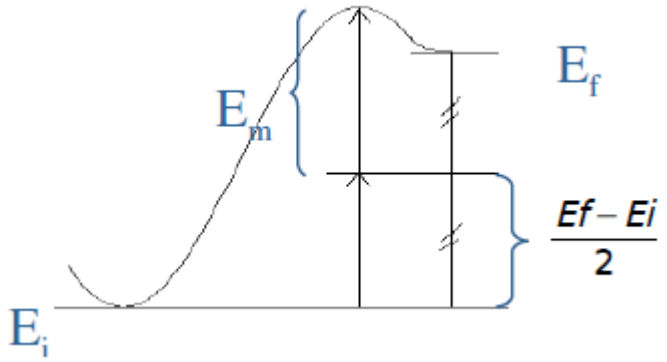


- Calculating all possible LAE on the fly can be expensive

- Models for barrier calculation, e.g:

FISE:

$$E_a = E_m + \frac{E_f - E_i}{2}$$



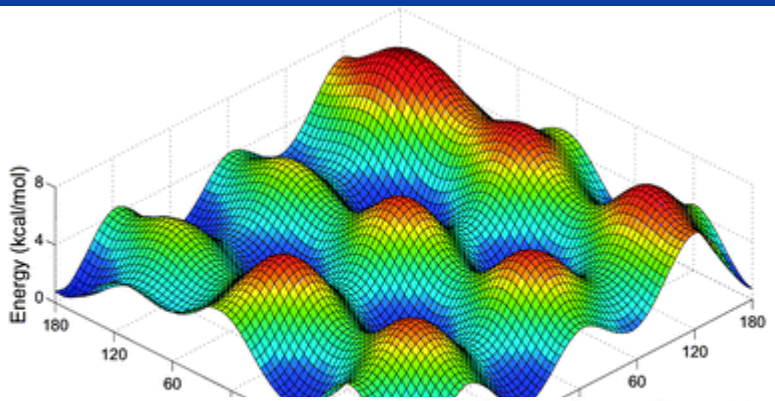
Cut bonds:

$$E_A = E(col) - E(ini) = e_A^{col} - \underbrace{\sum V_{Aj} - \sum V_{iV}}_{\text{liaisons coupées}}$$

Kinetic Monte Carlo

- The potential energy surface should drive the evolution
- How do we accurately represent a complicated **alloy** PES?
- Canonical idea is to simplify it according to some local model

$E_a = E^{mig} = E^{mig}(LAE)$

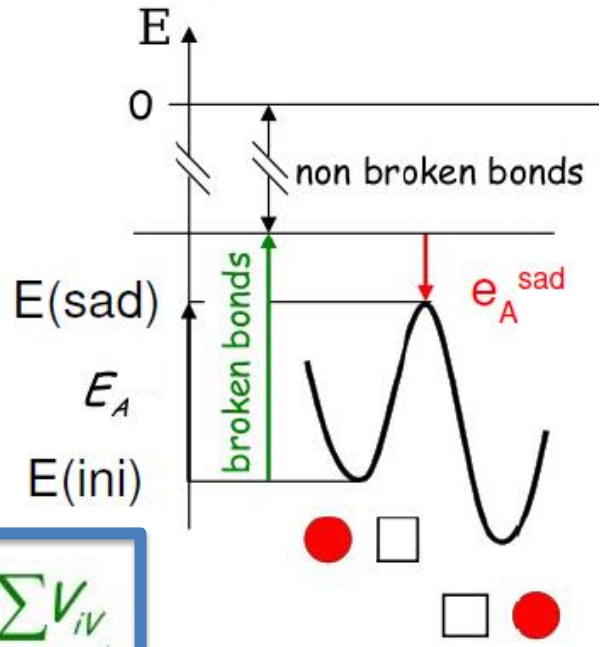


- Calculating all possible LAE on the fly can be expensive

- Models for barrier calculation, e.g:

FISE: $$E_a = E_m + \frac{E_f - E_i}{2}$$

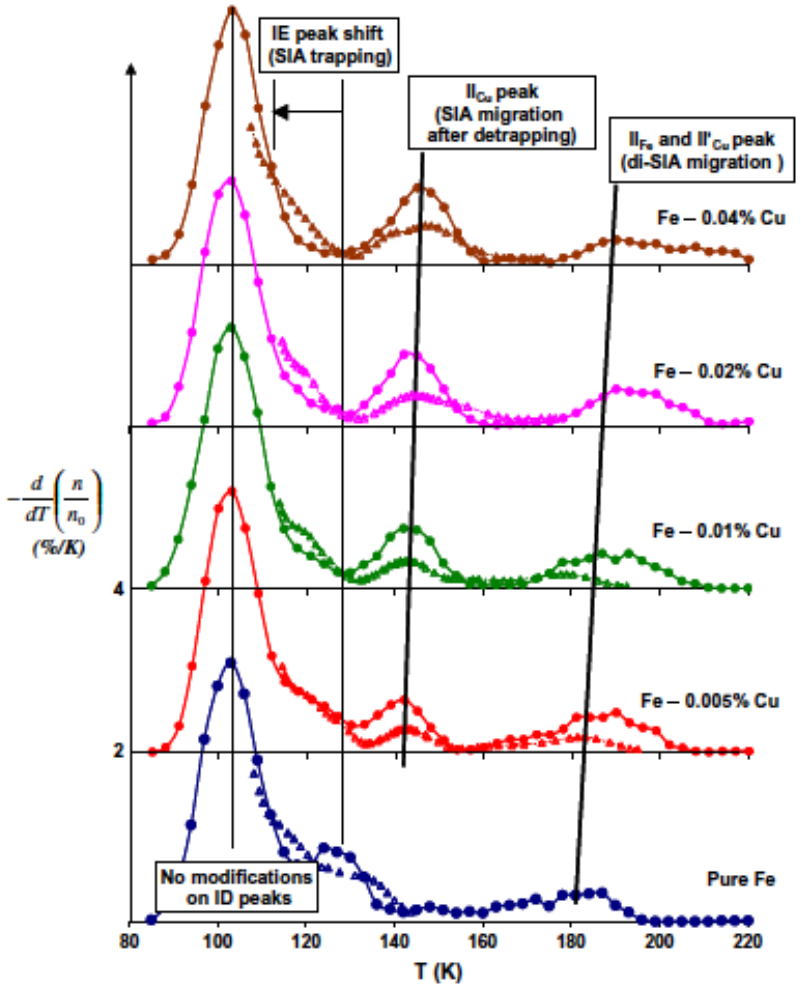
Cut bonds: $$E_A = E(col) - E(ini) = e_A^{col} - \underbrace{\sum V_{Aj} - \sum V_{iV}}_{\text{liaisons coupées}}$$



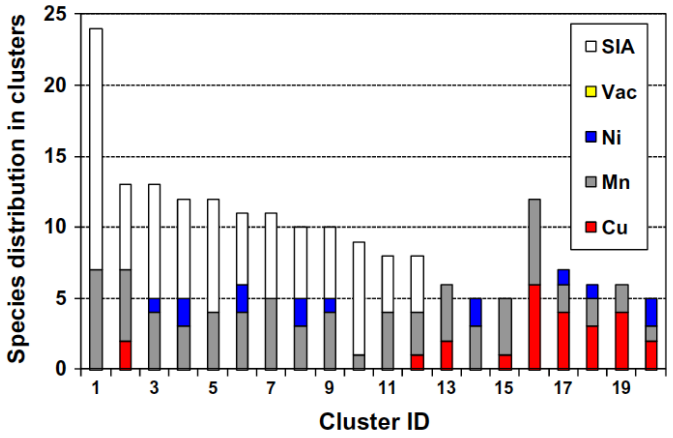
Some KMC examples

[1] R. Ngayam-Happy et al., J. Nucl. Mater. **407**, 16-28 (2010).
[2] R. Ngayam-Happy et al., J. Nucl. Mater. **426**, 198-207 (2012).
[3] L. Messina et al., Phys. Status Solidi A (Accepted 2016).
[4] M. K. Miller et al., J. Nucl. Mater. **437**, 107-115 (2013).

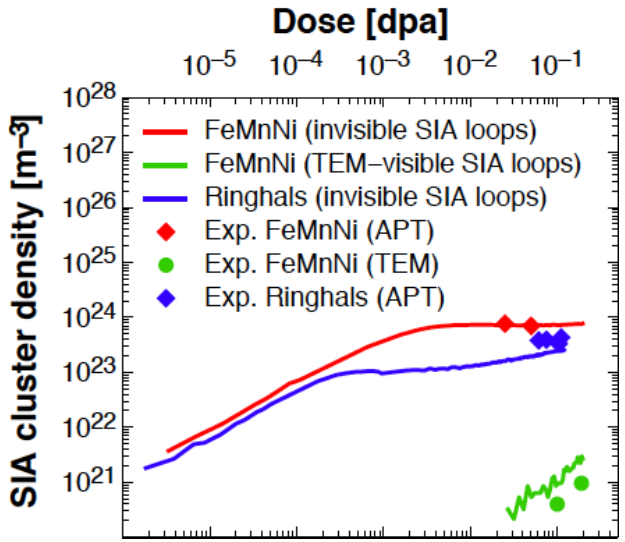
Resistivity recovery simul. in Fe alloys [1]

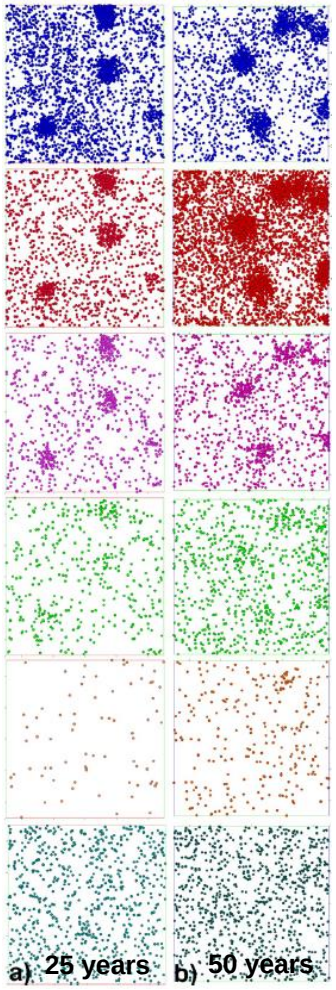


Composition of solute clusters [2]



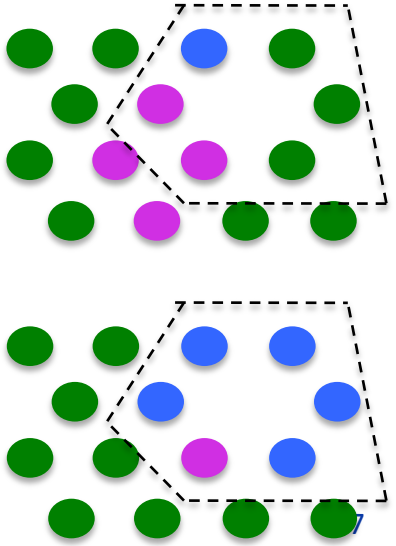
SIA cluster density in RPV steels [3]





APT maps of solute clusters in RPV surveillance samples [4]

Local Atomic Environment

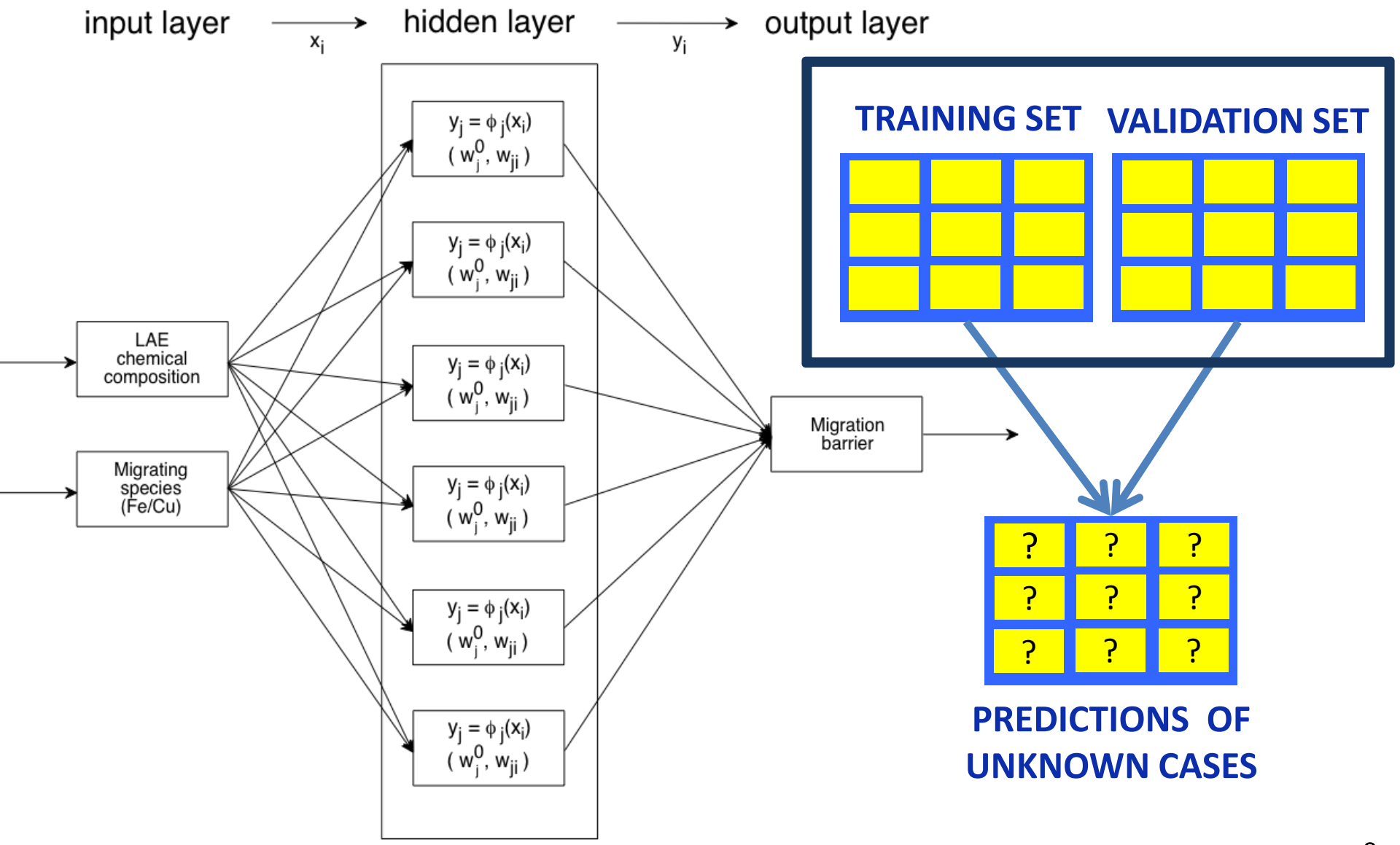


- How about supersaturation or concentrated cases?
- The LAE goes through a combinatorical explosion that is difficult to properly account for
- Castin implemented artificial neural network (ANN) to train barrier prediction on an alloy PES using EAM potentials

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

- Thousands of barriers used for training the ANN
- All the quirks of the EAM affect the ANN training and predictive power, and thus the KMC
- We here instead use massive amounts of DFT calculations as engine to train an ANN

Artificial neural networks

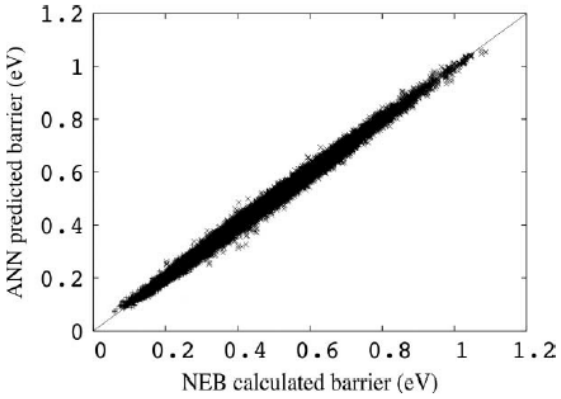


EAM-ANN simulations of FeCu thermal aging

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

10000 barriers

- Hybdrid AKMC/OKMC simulations of Cu precipitation in α-Fe (**thermal ageing**).
- Clusters N ≥ 15 considered as objects.

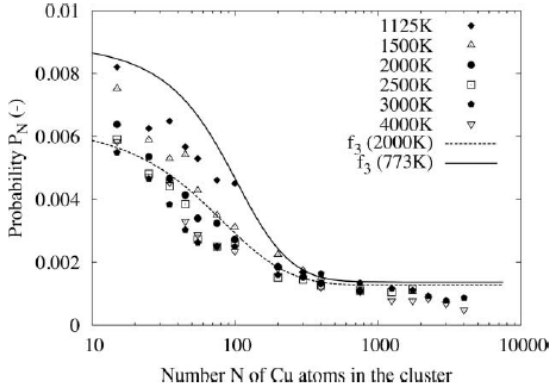


- <u>3 stages</u>

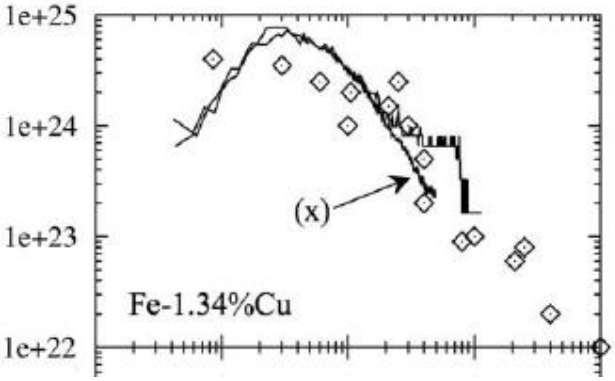
a) Barriers predicted with artificial neural network, based on 10^4 NEB calculations with EAM interatomic potential.

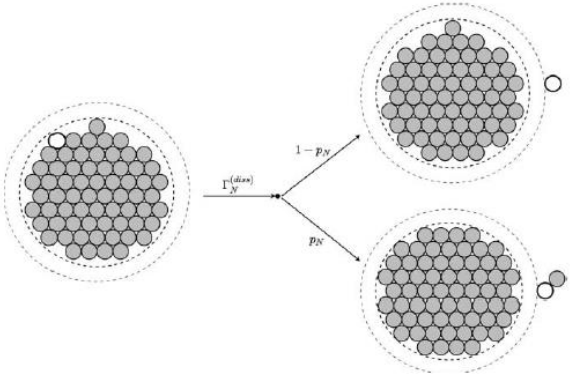
b) Calculation of stability and mobility of N ≥ 15 clusters. Clusters can emit the vacancy, or a V-Cu pair.

$$\ln(D_N(T)) = f_1(N, 1/k_BT) \qquad \ln(\tau_N(T)) = f_2(N, 1/k_BT) \qquad p_N(T) = f_3(N, 1/k_BT)$$



c) AKMC/OKMC simulations of thermal ageing. **Coalescence of mobile precipitates** plays an important role.





Interatomic potential vs DFT

Interatomic potentials

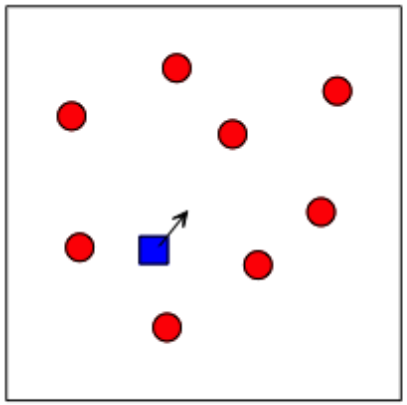
- Fitted on experimental/*ab initio* data.
- Highly costumizable to desired specific properties.
- Computationally cheap.
- System-specific.
- Fitting is a time-consuming and non-trivial/non-linear task.
- Questionable accuracy outside of fitting range

Density Functional Theory

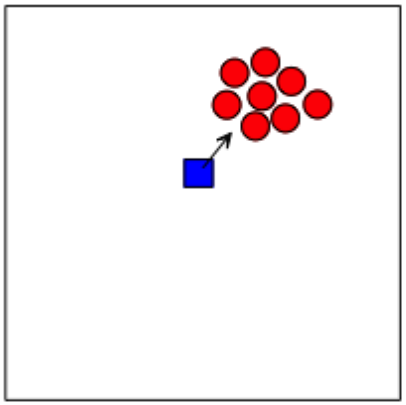
- Provides detailed description of alloy thermodynamic and kinetic properties (no need of compromises).
- Can be applied to complex multicomponent alloys with little complexity addition.
- Computationally expensive (for large amount of configurations).
- Limited simulation-cell size and amount of computations.

Neural network training

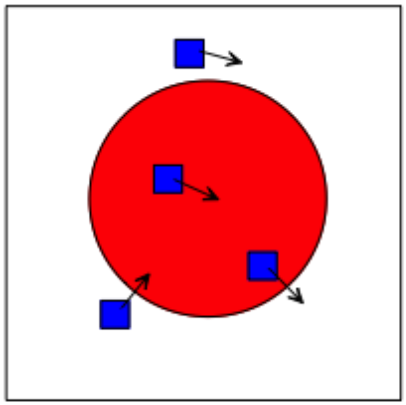
- **Test case**: thermal aging of FeCu alloys (only 1 vacancy).
- 2000 configurations (training + validation).
- Maximize variety of selected atomic environments.



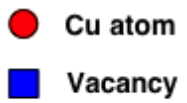
Type I



Type II

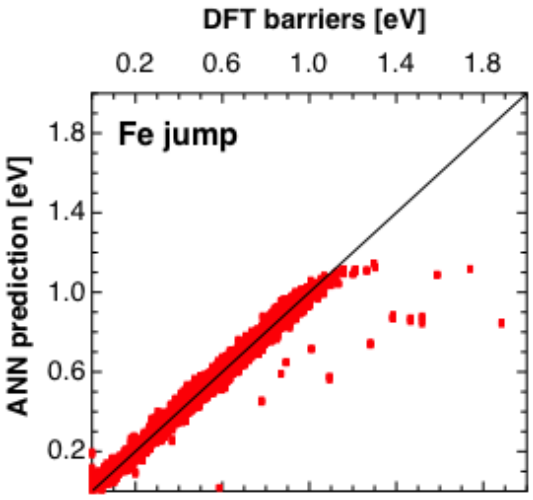


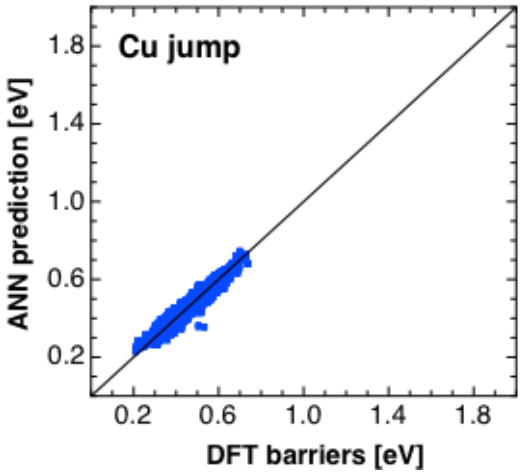
Type III



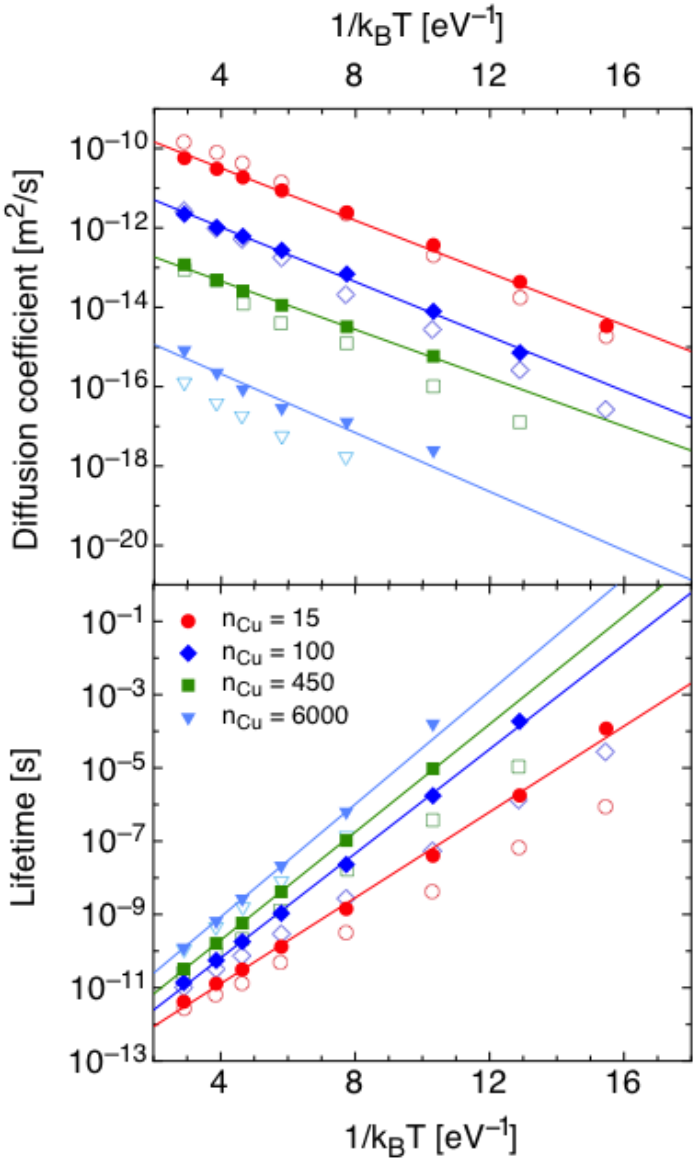
9 million core-hours!

DFT settings
PAW-PBE, 300 eV, 3^3 k-points
250-atom supercells
Full-kp relaxations of end states
CI-NEB relaxations with 2^3 kp grid
Full-kp energy calculation of saddle point





Cu cluster stability & mobility



Cu Cluster properties are now different!

- Much larger stability, higher dissociation energy

- Explained by low vacancy formation energy in BCC copper (0.85 eV in Cu vs 2.18 eV in Fe).

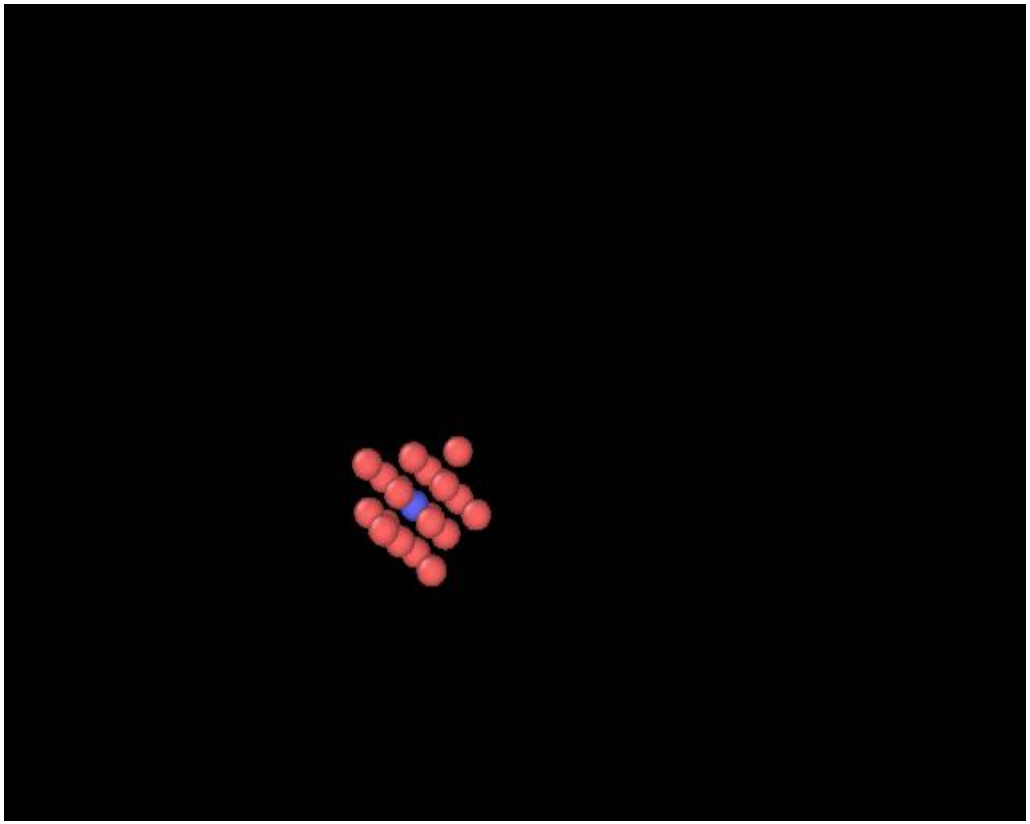
- Lower activation energy

- Much longer, temperature-dependent mean free paths.

Precipitation driven by coalescence of medium-sized clusters.

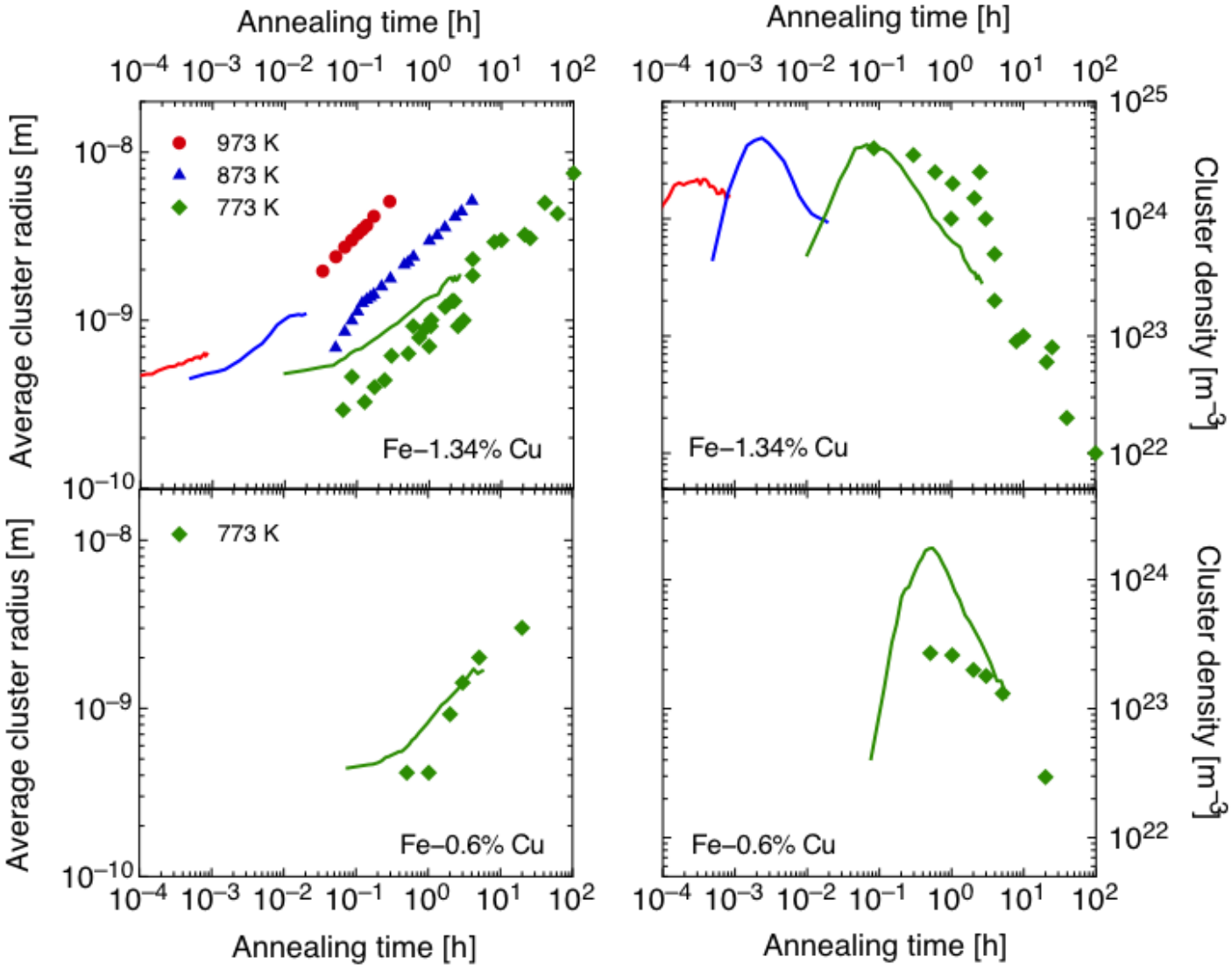
Cu cluster dissociation

- Frequent loss of Cu atoms before vacancy emission.
- Independent of cluster size and temperature until 1100 K.
- Related to vacancy-copper correlation (drag) active below 1100 K [(*)].



* L. Messina et al., *Phys. Rev. B* **90** (2014).

Thermal ageing of FeCu alloys



- General satisfactory agreement.
- Improved time-scale match with respect to EAM-based study.
- Overestimation of cluster density in dilute alloy.
- DFT-PAW prediction of solubility limit an issue (E_{subs}= 0.76 eV vs 0.5 eV).

L. Messina, N. Castin, C. Domain, P. Olsson, Submitted to J. Chem. Phys. (2016).

FeCu test case - conclusions

- We can generate large DFT databases and apply advanced regression schemes.

- Improved *prediction* of thermal ageing evolution (well-described time scales).

- Thermodynamic description is not accurate because of DFT issue for Cu in Fe -> Need to perform ANN regression on equilibrium energies and migration barriers separately.

- Technical limitations due to computational costs (box size and amount of calculations).

- **Next**: FeCr alloys and ANN-based cohesive models.

Advanced modeling of FeCu and FeCr

Objective: train two ANN's to perform thermodynamic and kinetic modeling separately.

THERMODYNAMIC ANN

Predicts total energy (cohesive model) of a given atomic configuration, by interpolation on a database of DFT-computed energies.

KINETIC ANN

Predicting the saddle-point energy of a given defect migration event, by interpolation on a database of DFT-computed migration barriers OR by using the TD cohesive model.

APPLICATIONS

1 **RIGID-LATTICE FeCu AND FeCr POTENTIAL**
- Phase diagram calculation by means of Metropolis Monte Carlo.
- Interpolation of DFT energies neglecting atomic forces.

2 **AKMC THERMAL AGEING OF FeCr ALLOYS**
- Vacancy migration barriers calculated by two separate ANN's using the rigid-lattice potential and DFT-computed saddle-point energies.

3 **LATTICE-FREE FeCu AND FeCr POTENTIALS**
- Atomic forces are included indirectly.
- Saddle-point energies are obtained and compared with the DFT values.
- Allows to extend the set of DFT migration barriers.

(1) Rigid-lattice FeCu and FeCr potentials

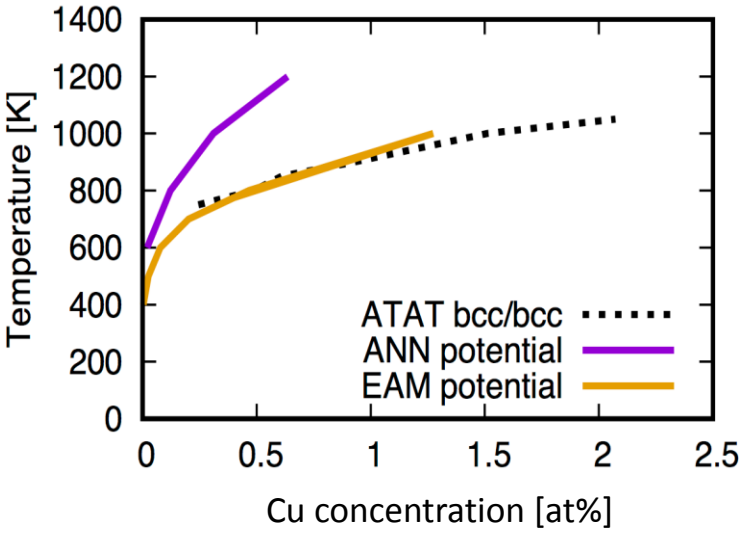
TOTAL ENERGY: $$E_{\text{tot}}^{\text{ANN}} = \sum_{a=1}^{N_{\text{Fe}}} E_{\text{Fe}}^{ANN}\rho(a) + \sum_{a=1}^{N_{\text{Cu}}} E_{\text{Cu}}^{ANN}\rho(a) + \sum_{a=1}^{N_{\text{Cr}}} E_{\text{Cr}}^{ANN}\rho(a)$$

$\rho(a)$ Local atomic density around atom a (describes univoquely the local atomic environment).

E_X^{ANN} Atomic energy functions (estimation of average energy assigned to each atom of species *x*). Outputs of the neural network trained on DFT energies of 4000 atomic configurations.

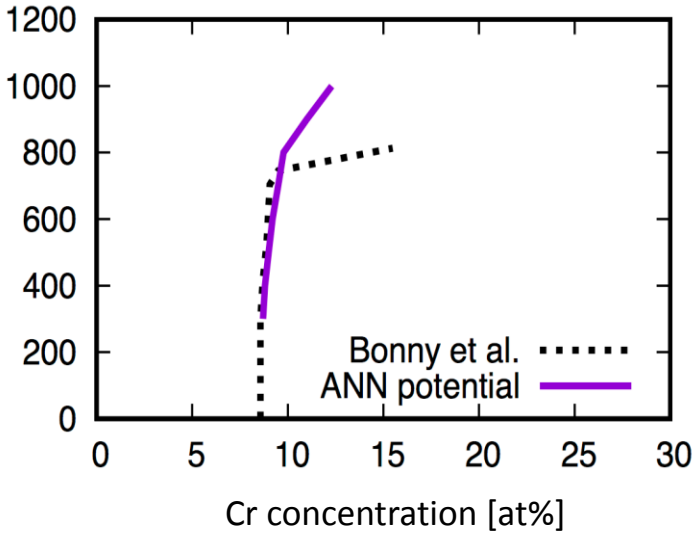
PREDICTED PHASE DIAGRAMS (MMC)

FeCu



Underestimated Cu solubility

FeCr



Very satisfactory agreement

(2) Thermal ageing of FeCr alloys

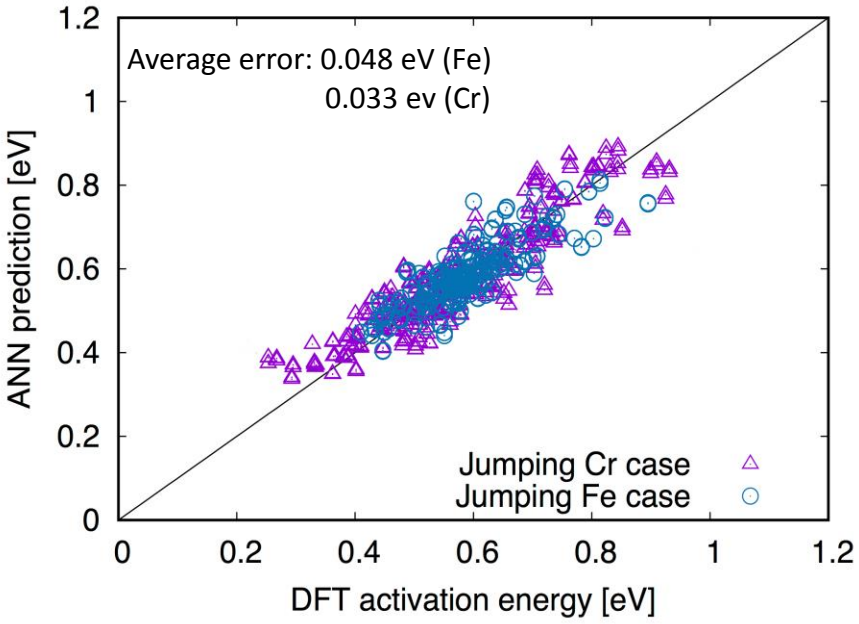
VACANCY MIGRATION BARRIER:

$$E_{ij}^{\mathrm{mig}} = \frac{1}{2}\left(E_{j,\mathrm{tot}}^{\mathrm{ANN}} - E_{i,\mathrm{tot}}^{\mathrm{ANN}}\right) + E_{\mathrm{saddle}}^{\mathrm{ANN}}$$

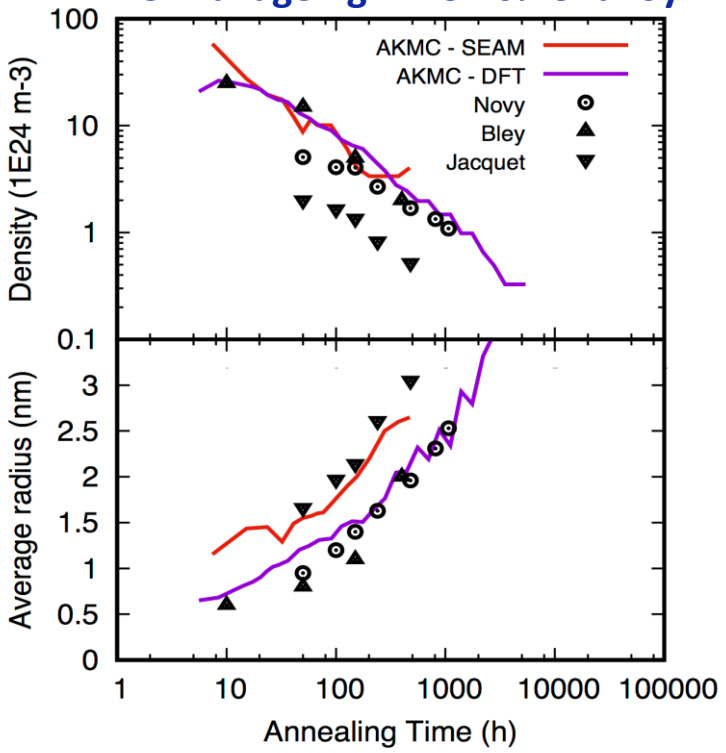
Provided by the rigid-lattice potential

Given by neural network trained on DFT-NEB calculations of 2000 vacancy migration events

Accuracy of migration energy prediction



Thermal ageing in Fe-20%Cr alloy



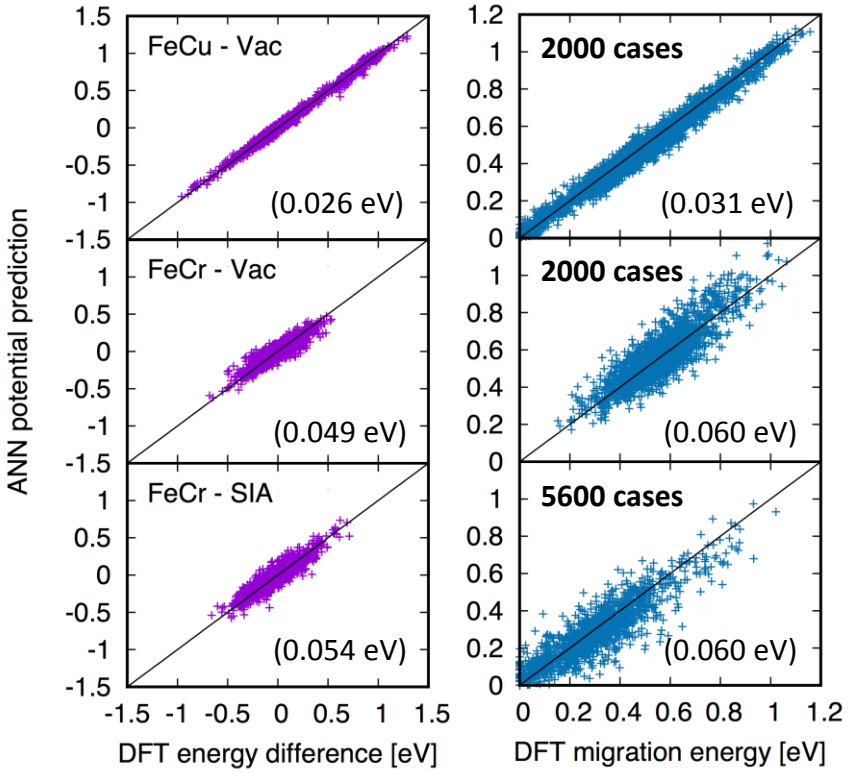
AKMC-SEAM: N. Castin, L. Malerba, J. Chem. Phys. **132,** 074507 (2010).

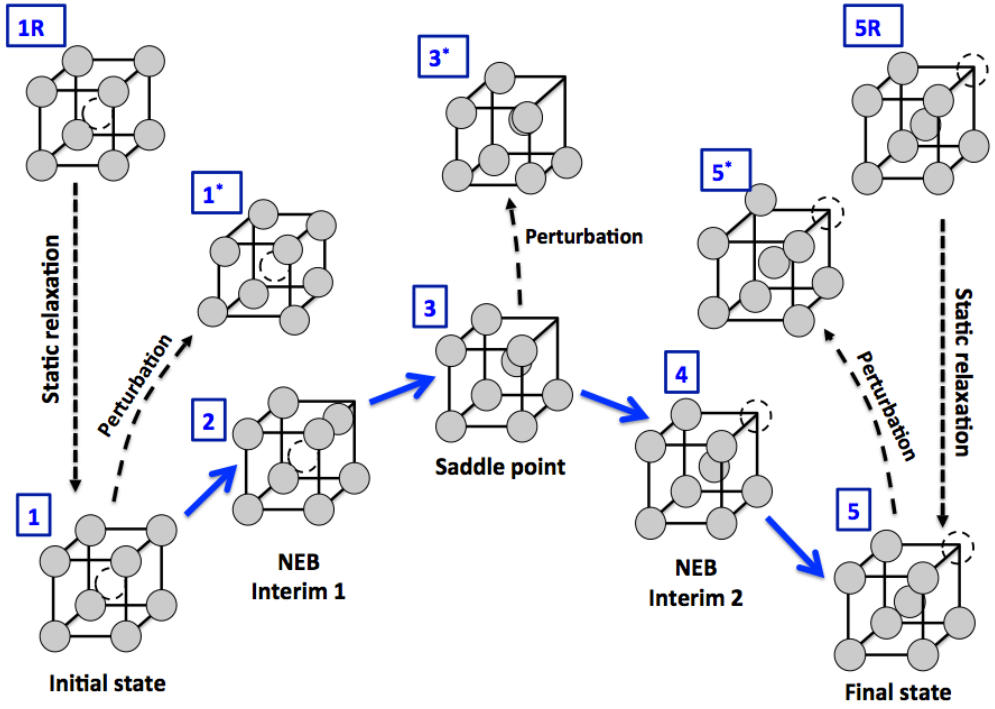
AKMC-DFT: N. Castin, L. Messina, C. Domain, R.C. Pasianot, P. Olsson, Submitted to J. Chem. Phys. (2016).

(3) Lattice-free FeCu and FeCr potentials

- Objective: introduce atomic displacements and forces in order to mimick the DFT saddle-point energies of defect migration and extend the computational capability of DFT.
- SIA migration requires much larger training database due to increased complexity.
- Forces introduced indirectly by training ANN on > 10^4 DFT-calculated energies of atomic configurations, where atoms were slightly displaced from their equilibrium positions.

Recalculated migration barriers ($\Delta E + E_{sad}$)

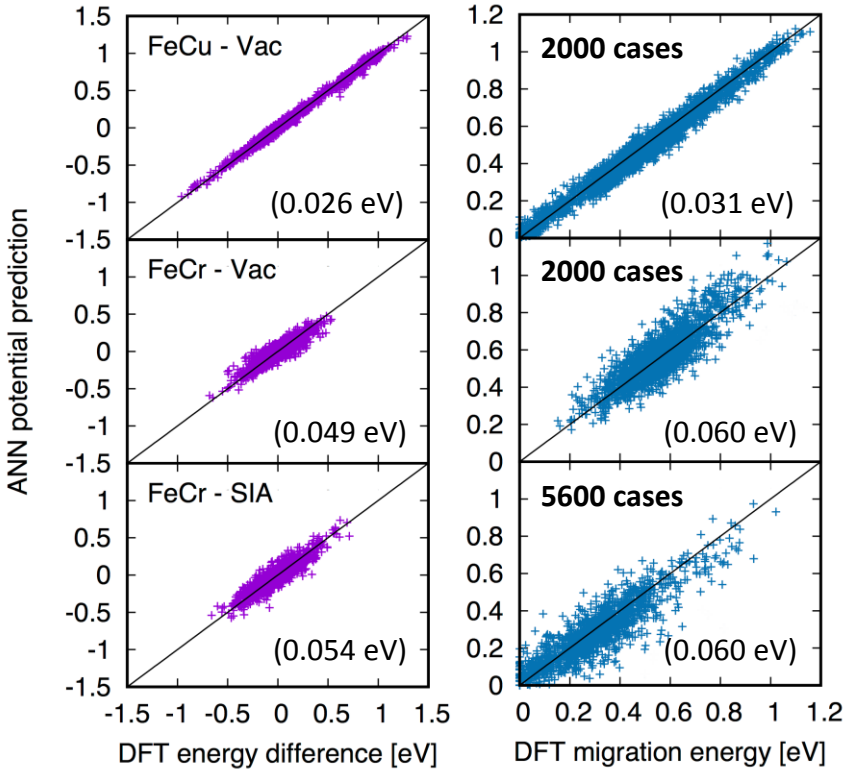




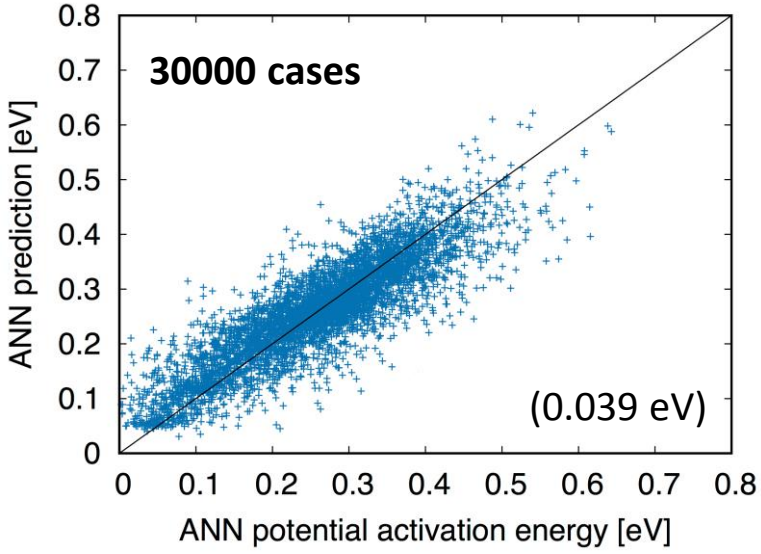
(3) Lattice-free FeCu and FeCr potentials

- Objective: introduce atomic displacements and forces in order to mimick the DFT saddle-point energies of defect migration and extend the computational capability of DFT.
- SIA migration requires much larger training database due to increased complexity.
- Forces introduced indirectly by training ANN on > 10^4 DFT-calculated energies of atomic configurations, where atoms were slightly displaced from their equilibrium positions.

Recalculated migration barriers ($\Delta E + E_{sad}$)



Extended database of 30000 SIA migration barriers



Conclusions & Perspectives

- First-of-a-kind parameterization of KMC simulations with fully DFT-based advanced regression scheme, applied to FeCu and FeCr alloys.
- Neural networks are widely employed to ensure full transferability of DFT properties to the KMC simulation.
- Portability to different and more complex systems with little additional complexity.
- Limitations due to the high computational cost of massive amounts of DFT calculations are overcome by developing ANN cohesive models mimicking DFT ("DFT potentials").
- Preliminary applications to FeCu and FeCr alloys shows satisfactory agreement with thermal ageing experiments.
- Future application to irradiated FeCr alloys and FeMnNi alloys is foreseen, with the final goal of approaching real steel compositions.

THANKS FOR YOUR ATTENTION!