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SMINS-4 Structural Materials for Innovative Nuclear Systems

Manchester, UK

July 11th-14th, 2015

Ab initio-trained neural-network driven Monte Carlo simulations of microstructure evolution in irradiated iron alloys

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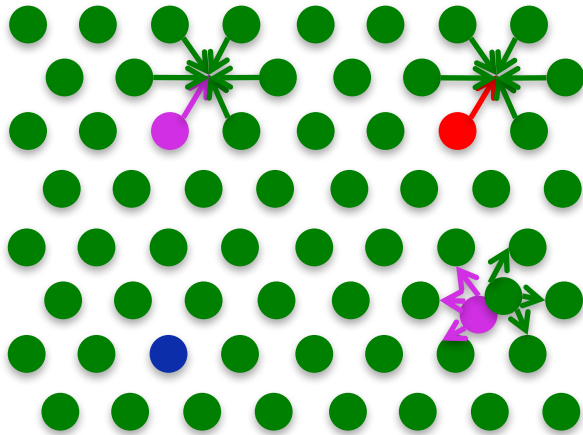


Outline

- Introduction: Transition rates in KMC simulations
- Neural networks
- Thermal ageing of FeCu alloys
- Advanced modelling of FeCu and FeCr alloys
- Conclusions

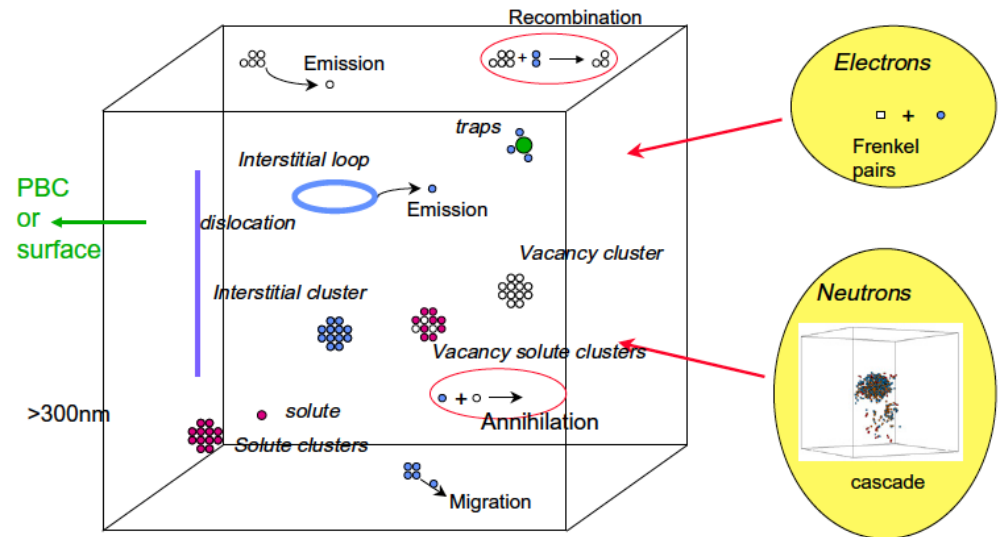
Kinetic Monte Carlo

ATOMISTIC KMC



Atomic transition rates

OBJECT KMC



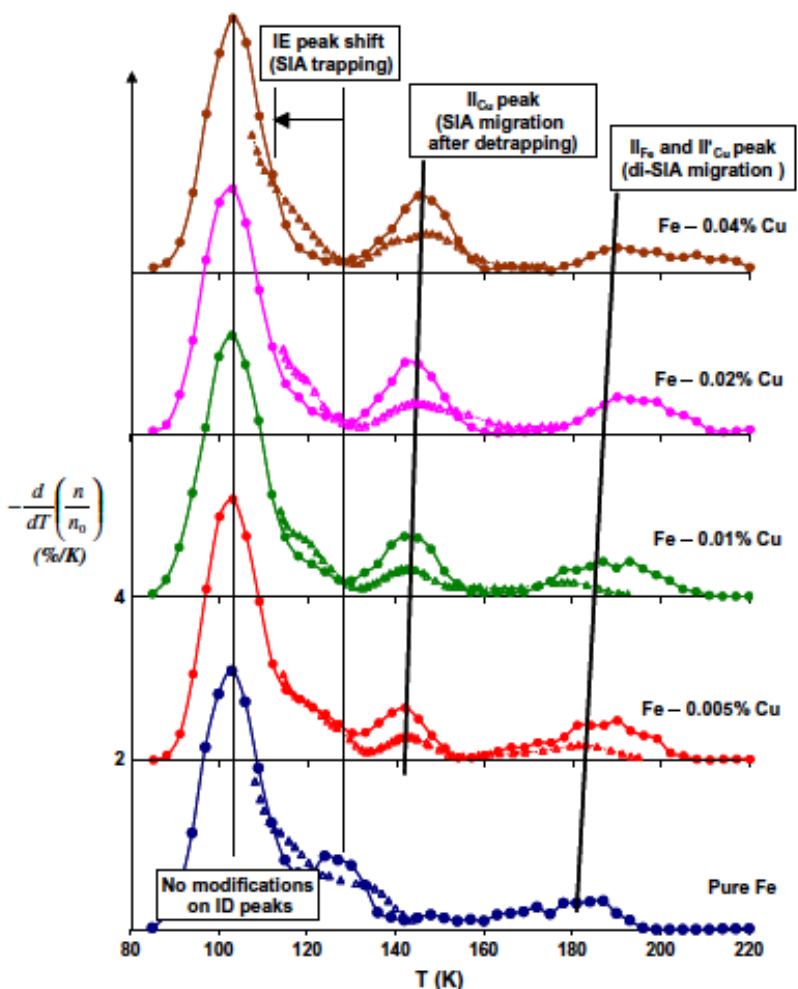
Object event rates, obtained by AKMC

- Microstructure evolution driven by point-defect migration.
- Migration barriers depend on local atomic environment.
- Too many combinations!
- Poor thermodynamic and kinetic descriptions with inaccurate predictions.

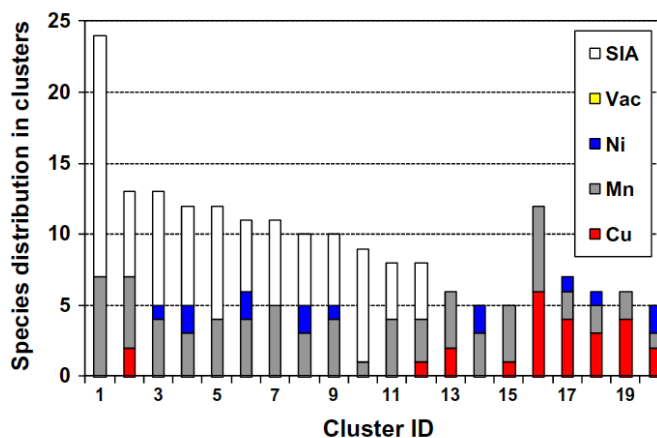
KMC simulations

- [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., submitted to Phys. Status Solidi A (2016).
- [4] M. K. Miller et al., J. Nucl. Mater. **437**, 107-115 (2013).

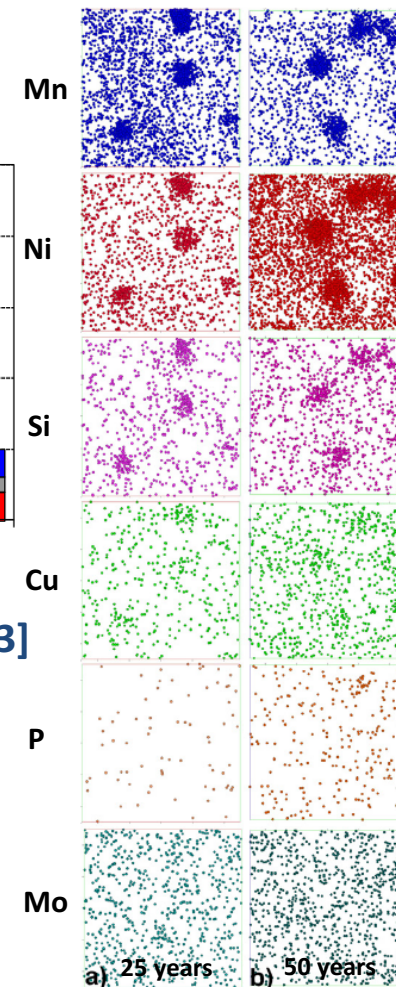
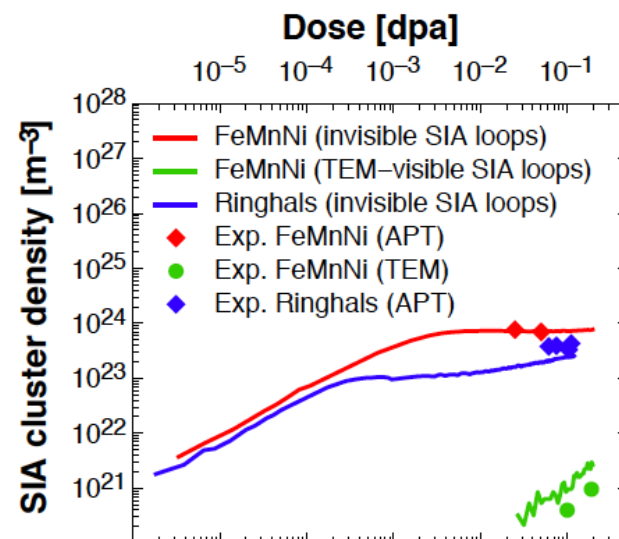
Resistivity recovery simul. in Fe alloys [1]



Composition of solute clusters [2]

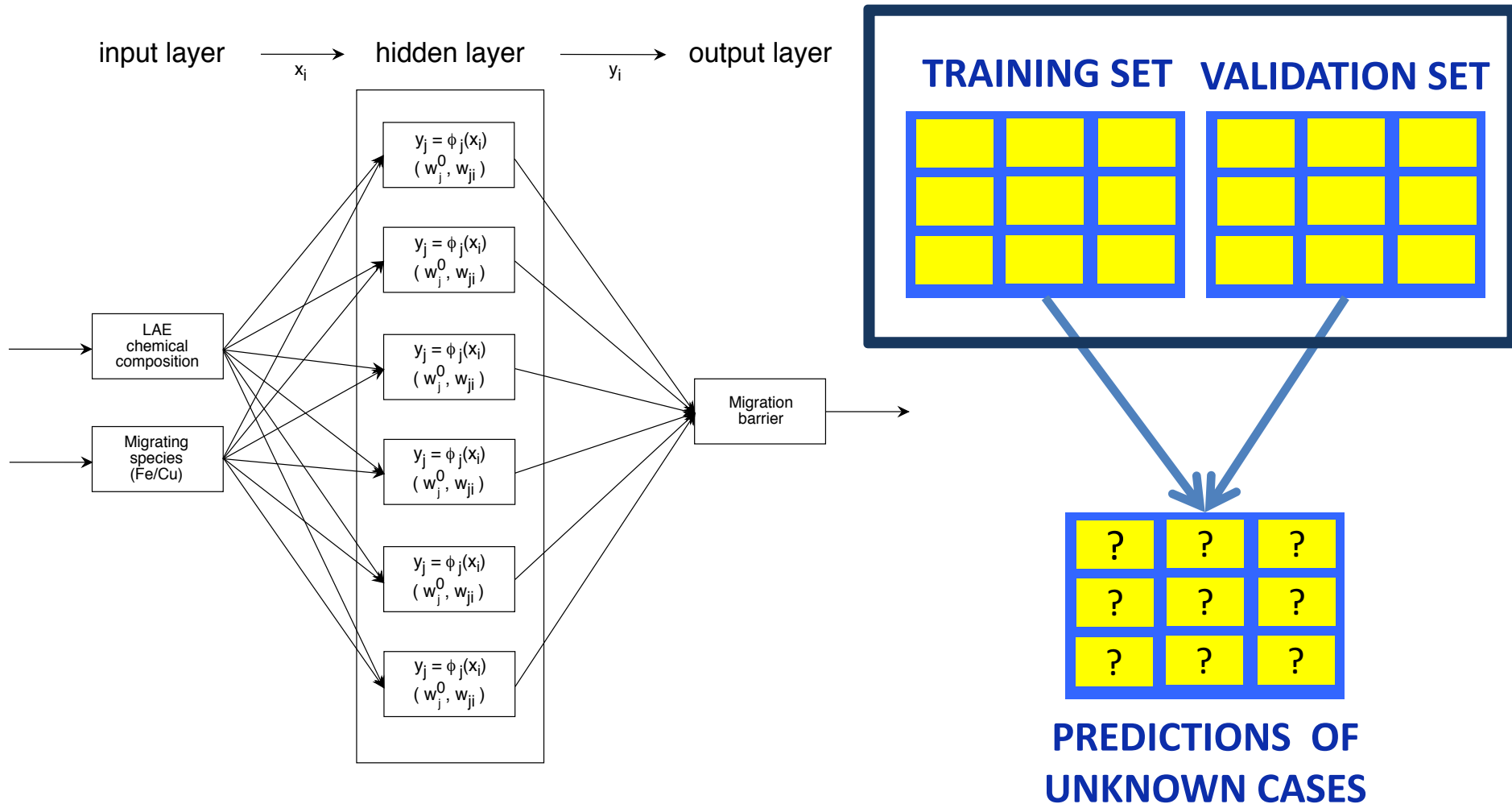


SIA cluster density in RPV steels [3]



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

Artificial neural networks



EAM-ANN simulations of FeCu thermal aging

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

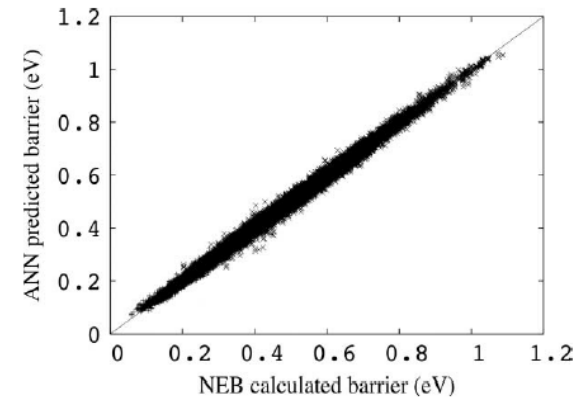
- Hybrid AKMC/OKMC simulations of Cu precipitation in α -Fe (**thermal ageing**).
- Clusters $N \geq 15$ considered as objects.

10000 barriers!

• 3 stages

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

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

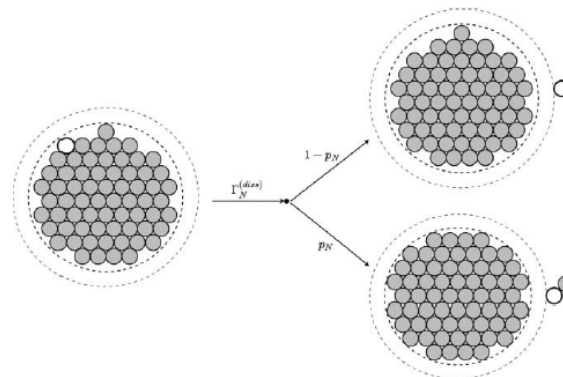
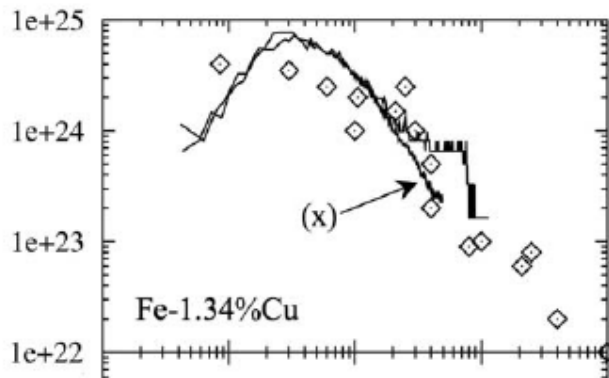
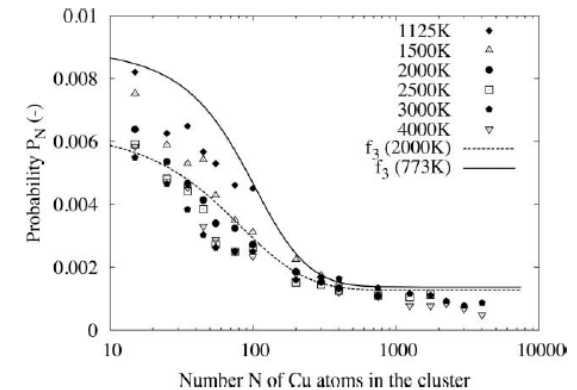


$\ln(D_N(T))$

$\ln(t_N(T))$

$p_N(T)$

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



Interatomic potential vs DFT

Interatomic potentials

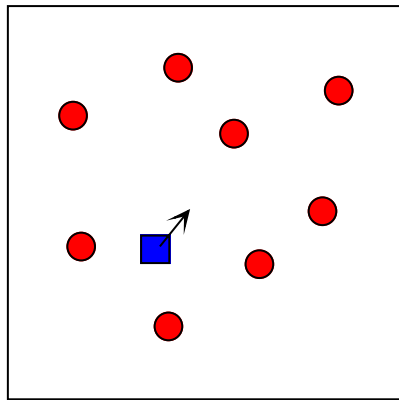
- Fitted on experimental/*ab initio* data.
- Highly customizable to desired specific properties.
- Computationally cheap.
- System-specific.
- Fitting is a time-consuming and non-trivial/non-linear task.
- Difficult to be accurate on many alloy properties at once.

Density Functional Theory

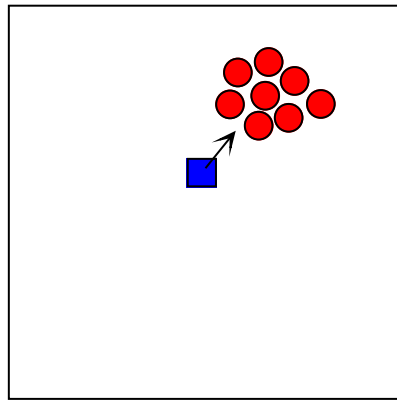
- Provides detailed description of alloy thermodynamic and kinetic properties (no need of compromises).
- Can be applied to complex multicomponent alloys with little complexity addition.
- Computationally expensive (for large amount of configurations).
- Limited simulation-cell size and amount of computations.

Neural network training

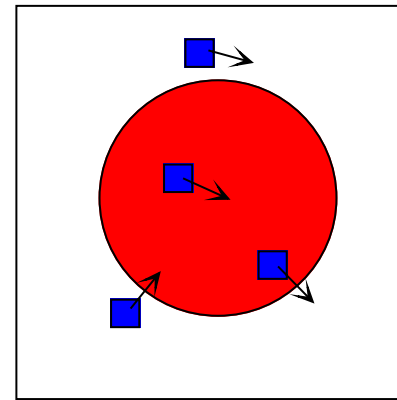
- Test case: thermal aging of FeCu alloys (only 1 vacancy).
- 2000 configurations (training + validation).
- Maximize variety of selected atomic environments.



Type I



Type II



Type III

● Cu atom
■ Vacancy

3 millions core-hours!

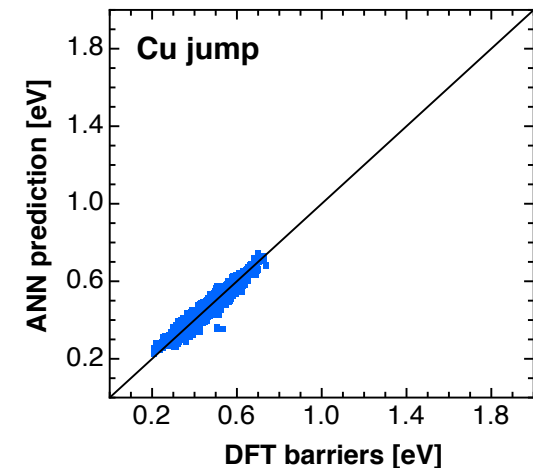
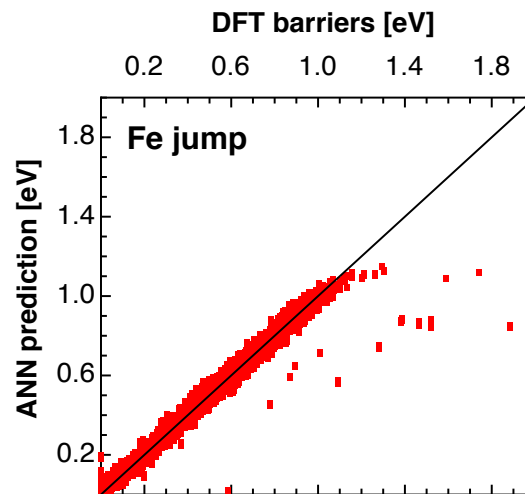
DFT settings

PAW-PBE, 300 eV, 3^3 k-points

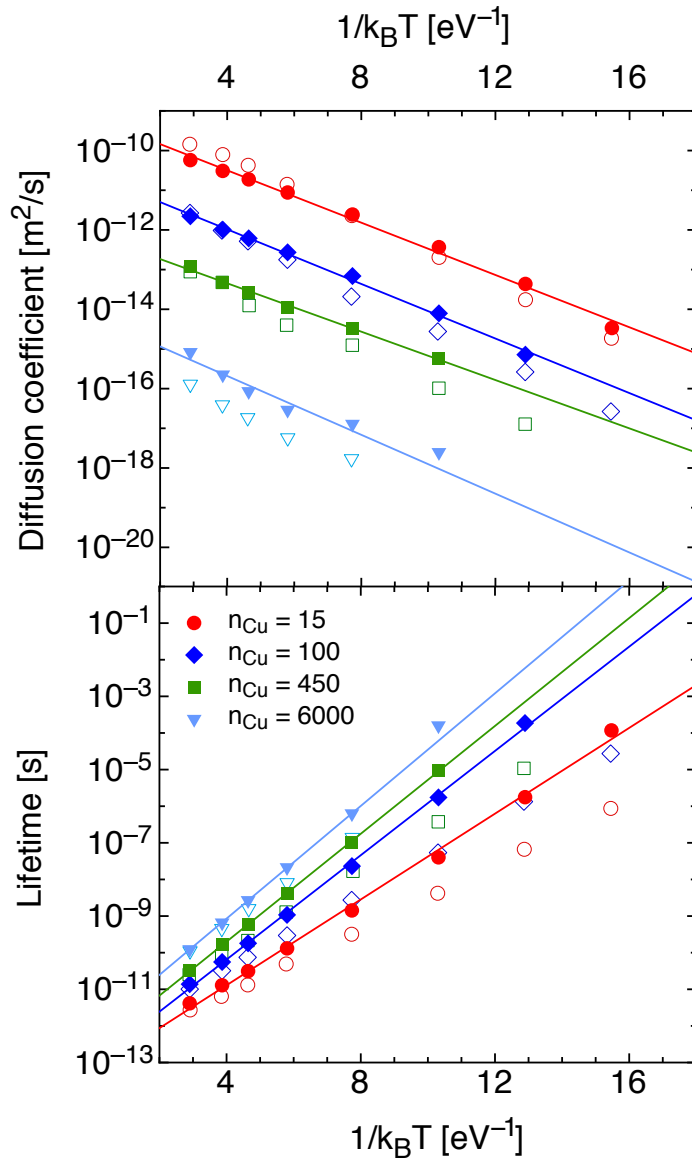
Full-kp relaxations of end states

NEB relaxations with 2^3 kp grid

Full-kp energy calculation of saddle point



Cu cluster stability & mobility



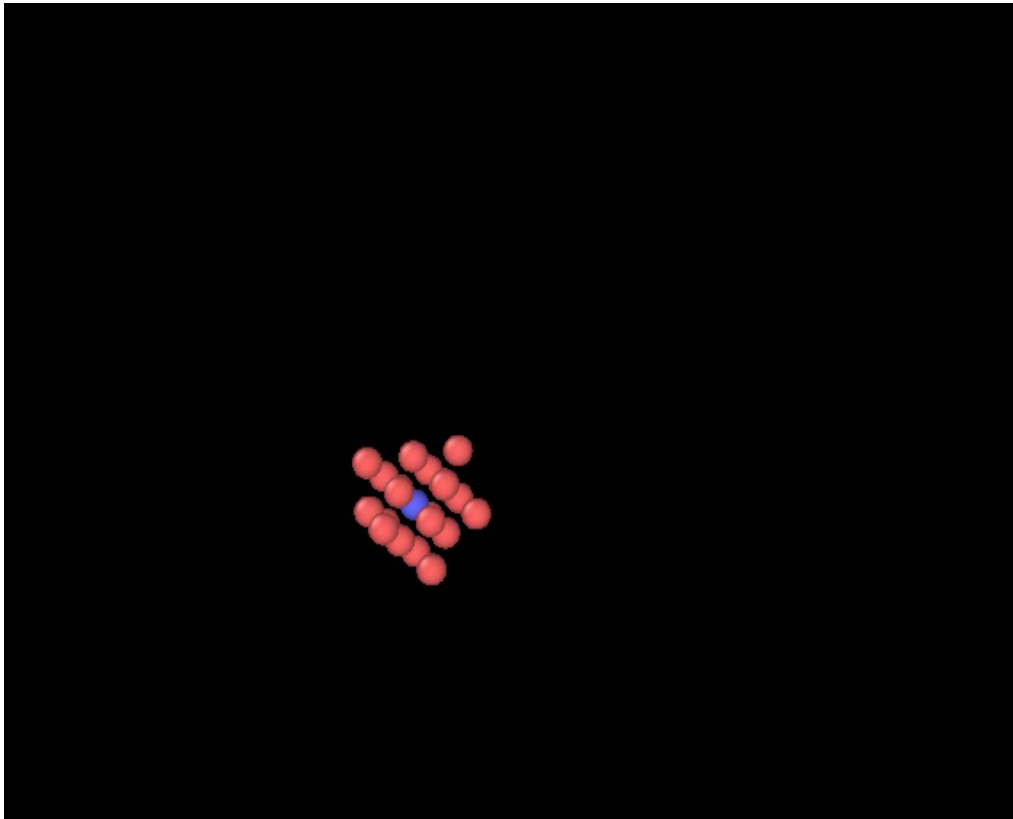
Cu Cluster properties are now different!

- 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.

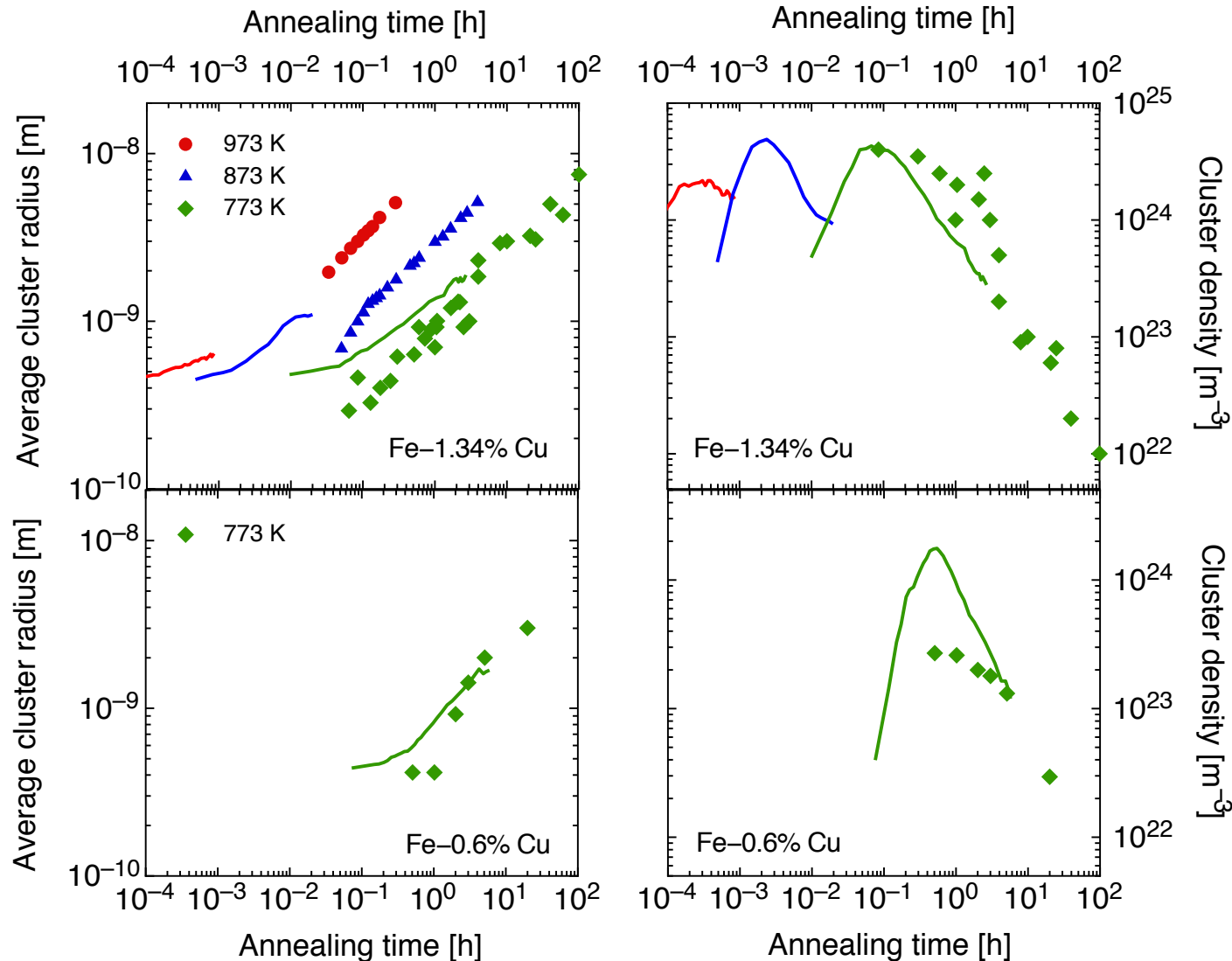
Cu cluster dissociation

- 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 (*).



$$\overline{n_{\text{Cu}}}$$

Thermal ageing of FeCu alloys



- General satisfactory agreement.
- Improved time-scale match with respect to IAP-based study.
- Overestimation of cluster density in dilute alloy.
- Incorrect DFT prediction of solubility limit (0.76 eV vs 0.5 eV).

FeCu test case - conclusions

- It is nowadays possible to obtain large DFT database and apply advanced regression schemes.
- Improved thermal ageing evolution (well-described time scales).
- Thermodynamic description is not accurate because of inexact DFT prediction. 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.

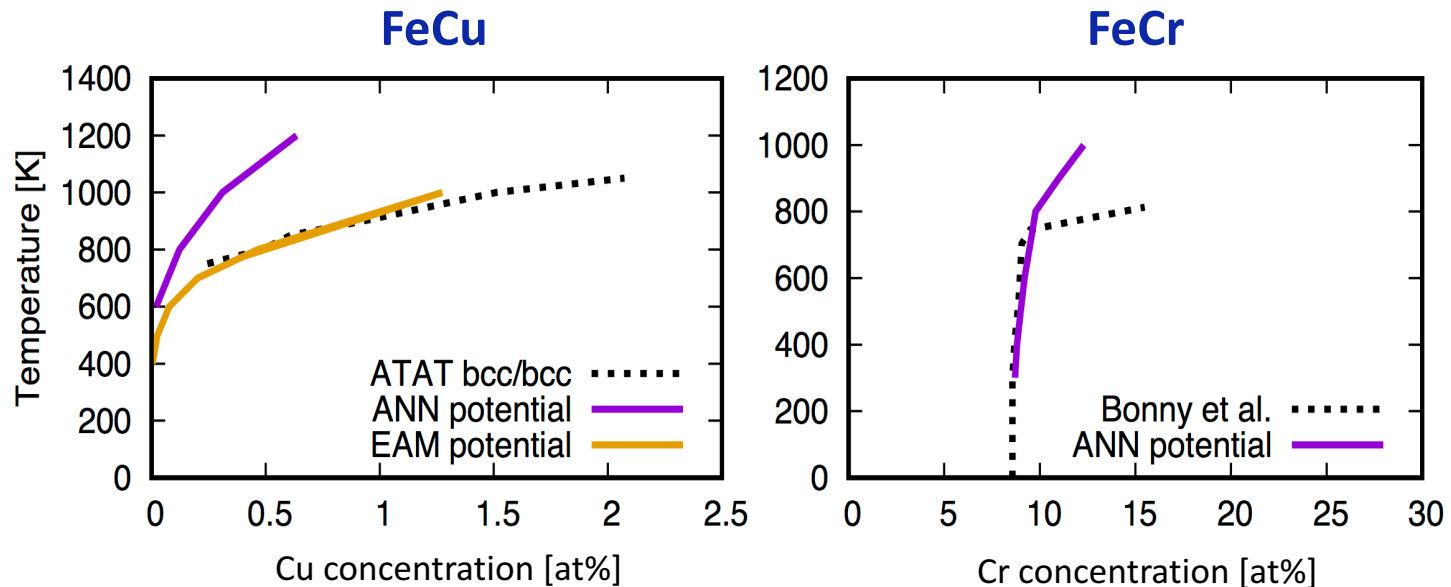
(1) Rigid-lattice FeCu and FeCr potentials

TOTAL ENERGY: $E_{\text{tot}}^{\text{ANN}}$

$r(a)$ 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)



Underestimated Cu solubility

Very satisfactory agreement

(2) Thermal ageing of FeCr alloys

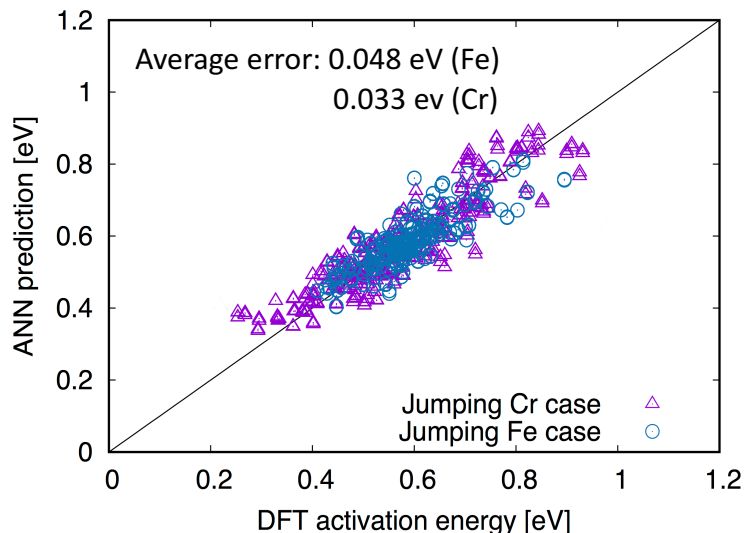
VACANCY MIGRATION
BARRIER:

$$E_{ij}^{\text{mig}}$$

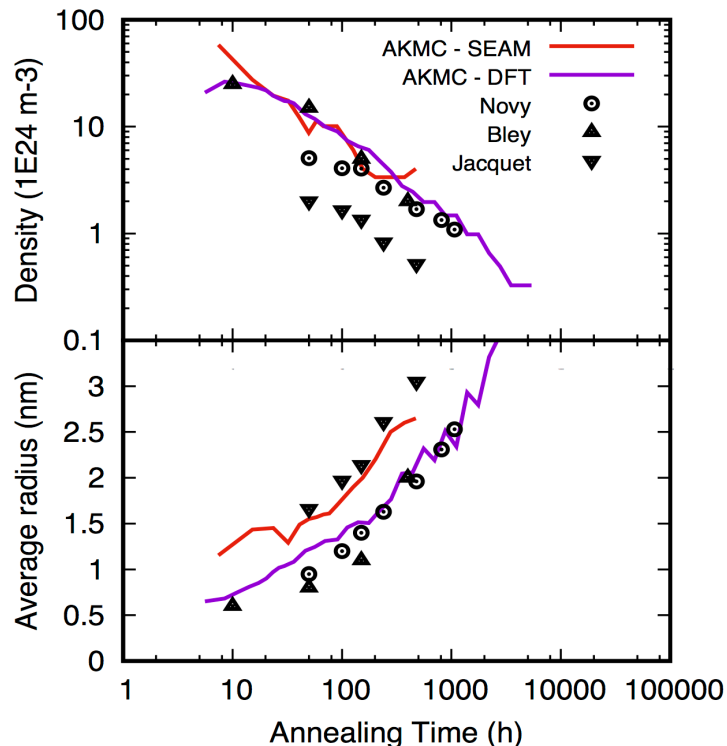
Provided by the rigid-lattice potential

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

Accuracy of migration energy prediction



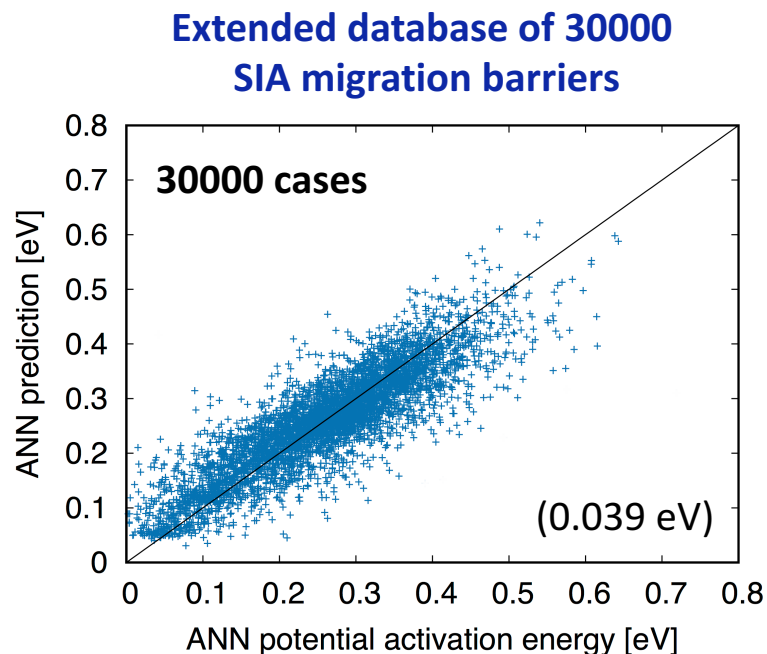
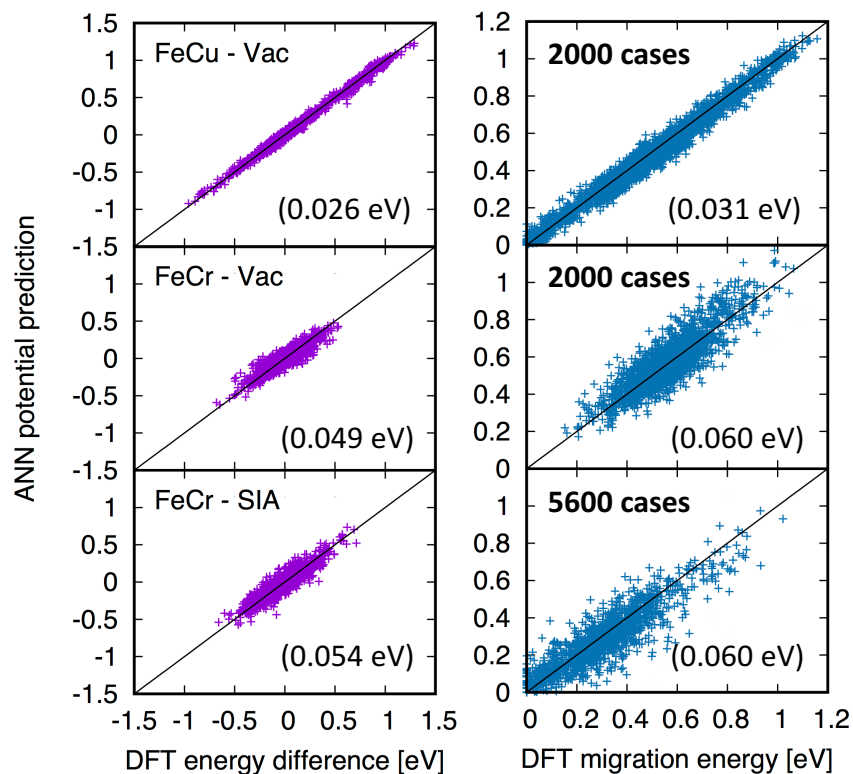
Thermal ageing in Fe-20%Cr alloy



(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 ($\Delta E + E_{\text{sad}}$)



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 electron-irradiated FeCr alloys and FeMnNi alloys is foreseen, with the final goal of approaching real RPV steel compositions.

THANKS FOR YOUR ATTENTION!