Final Workshop, June 2019
Miraflores de la Sierra(Spain)

NANOFEATURE EVOLUTION MODELS FOR IRRADIATION EFFECTS IN RPV AND INTERNALS

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Moret sur Loing

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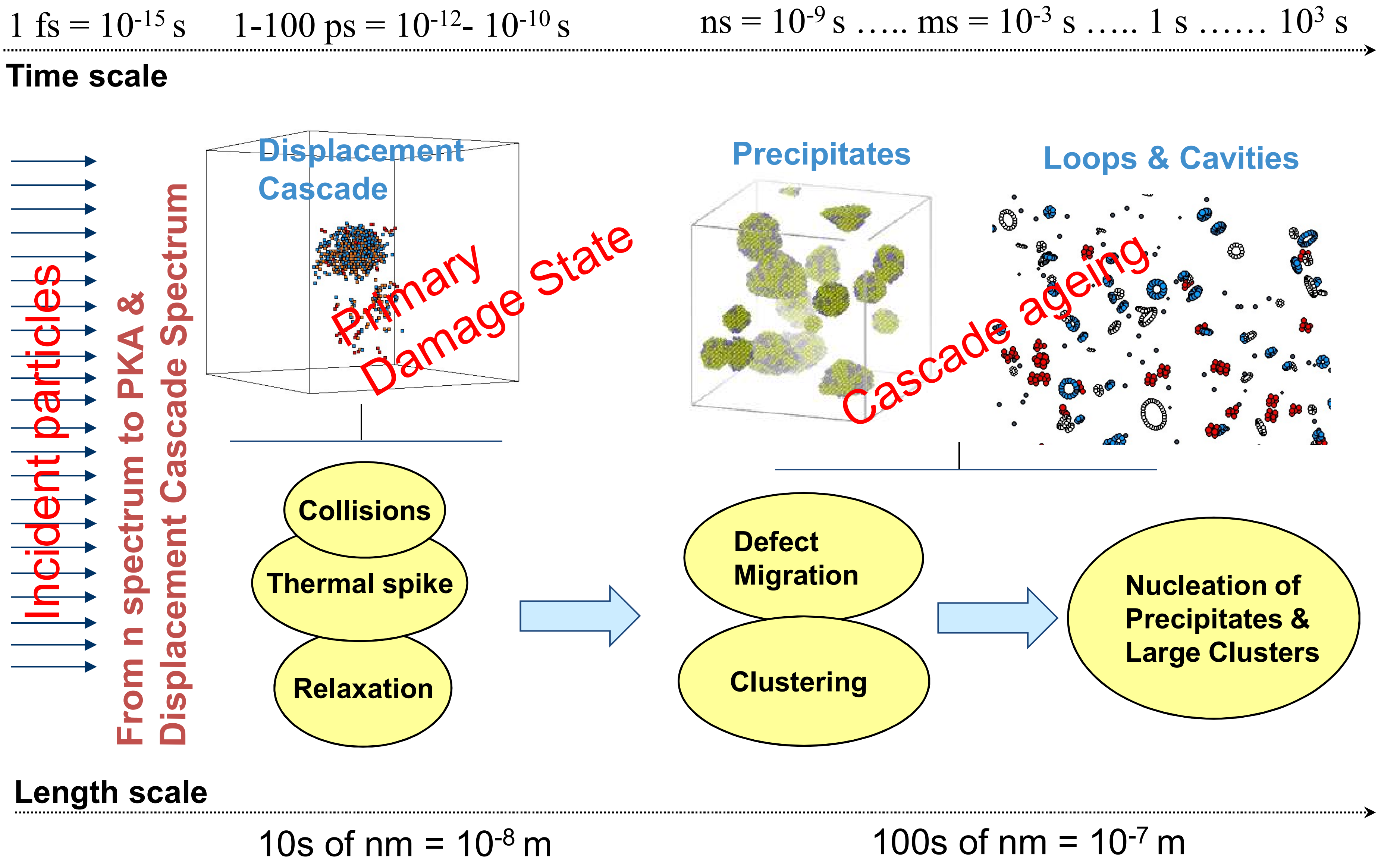
Outline

- ❑ Introduction
- ❑ Physics basis – finding mechanisms
- ❑ Multiscale models of nanofeature evolution in RPV steels
  - Predictive multi-scale modeling of FeCu and FeCr
  - Neutron irradiation and solute cluster growth in RPV steels
- ❑ Modeling ion beam conditions – the role of injected interstitials in austenitic alloys

Irradiation effects are inherently a multiscale problem



1 fs = 10^{-15} s 1-100 ps = 10^{-12}- 10^{-10} s ns = 10^{-9} s ….. ms = 10^{-3} s ….. 1 s …… 10^{3} s

Time scale

Incident particles

From n spectrum to PKA & Displacement Cascade Spectrum

Displacement Cascade

Primary Damage State

Precipitates

Loops & Cavities

Cascade ageing

Collisions

Thermal spike

Relaxation

Defect Migration

Clustering

Nucleation of Precipitates & Large Clusters

Length scale

10s of nm = 10^{-8} m

100s of nm = 10^{-7} m

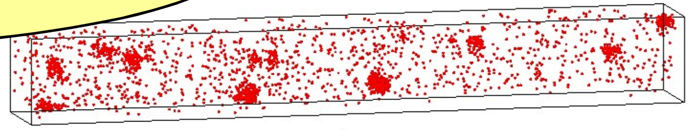
Irradiation effects are inherently a multiscale problem

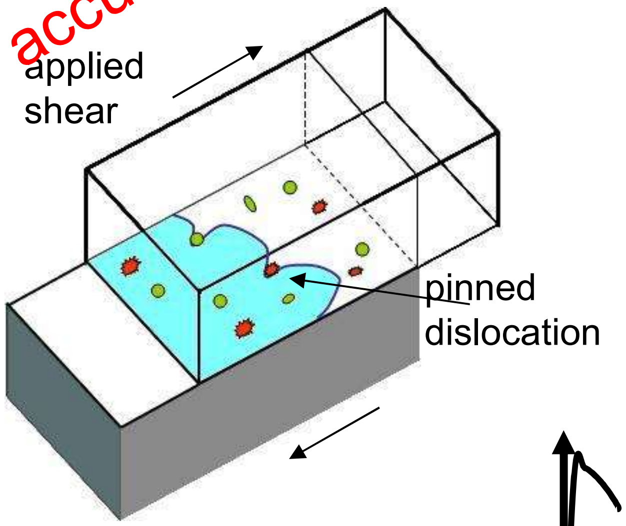
$\mu s = 10^{-3}$ s.. Years = 10^{7} - 10^{9} s

Time scale

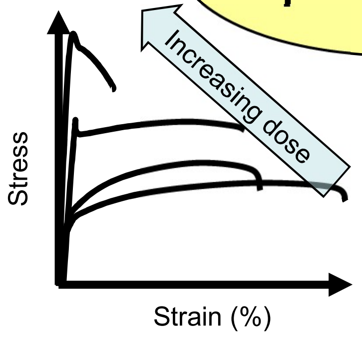
Cascade accumulation

Growing Concentration of Radiation Induced Defects while the Irradiation Proceeds



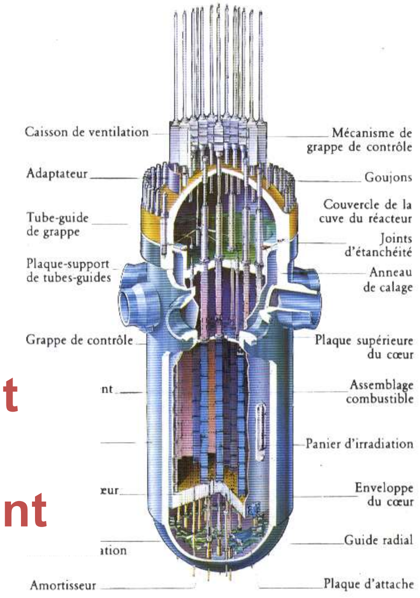


Mechanical Property Changes



Dislo/Defect Interaction
Yield Strength Increase
Loss of ductility

Component lifetime management

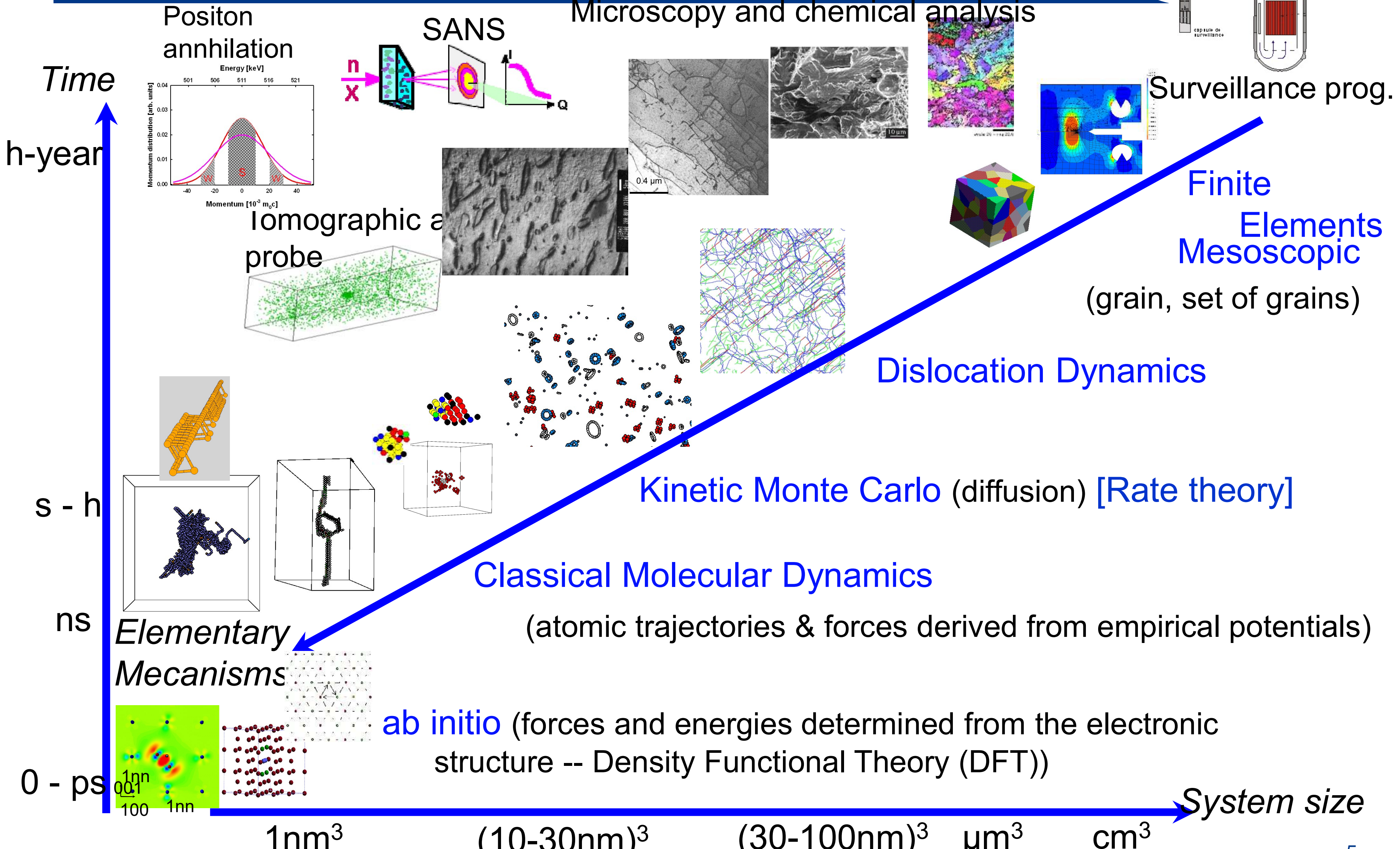


Length scale

10s of $\mu m = 10^{-5}$ m

cm = 10^{-2} m

Simulation tools



Positon annhilation

SANS

Microscopy and chemical analysis

Surveillance prog.

Time

h-year

Tomographic a
probe

Finite Elements

Mesoscopic

(grain, set of grains)

Dislocation Dynamics

Kinetic Monte Carlo (diffusion) [Rate theory]

s - h

Classical Molecular Dynamics

(atomic trajectories & forces derived from empirical potentials)

ns

Elementary Mecanisms

ab initio (forces and energies determined from the electronic structure -- Density Functional Theory (DFT))

0 - ps

System size

$1nm^3$ $(10\text{-}30nm)^3$ $(30\text{-}100nm)^3$ μm^3 cm^3

Relevant phenomena and appropriate computational methods for microstructure evolution

Phenomena

single displacement cascade

multiple cascades, cascade overlap

defect and solute migration and clustering

void swelling, hardening, embrittlement, creep, stress corrosion cracking, ...

microstructure evolution

collisional phase

quenching

annealing phase

defect/solute diffusion

mechanical property changes

10^{-14} s

10^{-11} s

10^{-8} s

10^{1} s

10^{4} s

> 10^{6} s

Methods

molecular dynamics

kinetic Monte Carlo

finite element

ab initio

MD dislocation dynamics

reaction rate theory, phase field 3D dislocation dynamics

10^{-9} m

10^{-7} m

10^{-6} m

> 10^{-3} m

Microstructure evolution modelling

Solute – defect properties
Binding energies
Migration energies
Transport coefficient

Primary damage
Cascade database

AKMC | Hybrid AKMC-OKMC | OKMC | Cluster Dynamics

Cohesive models:
Pair interaction
Cluster expansion
Concentration dependant pair model (CDP)
Neural network

Parameterisation:
DFT & MD results
Effective concentration dependant model
Treatment of all solutes

1D/3D modelling
Ion irradiation modelling

THE PHYSICS BASIS – FINDING MECHANISMS

Based on Deliverable D5.2 of the SOTERIA project

Multiscale modeling: the physics basis

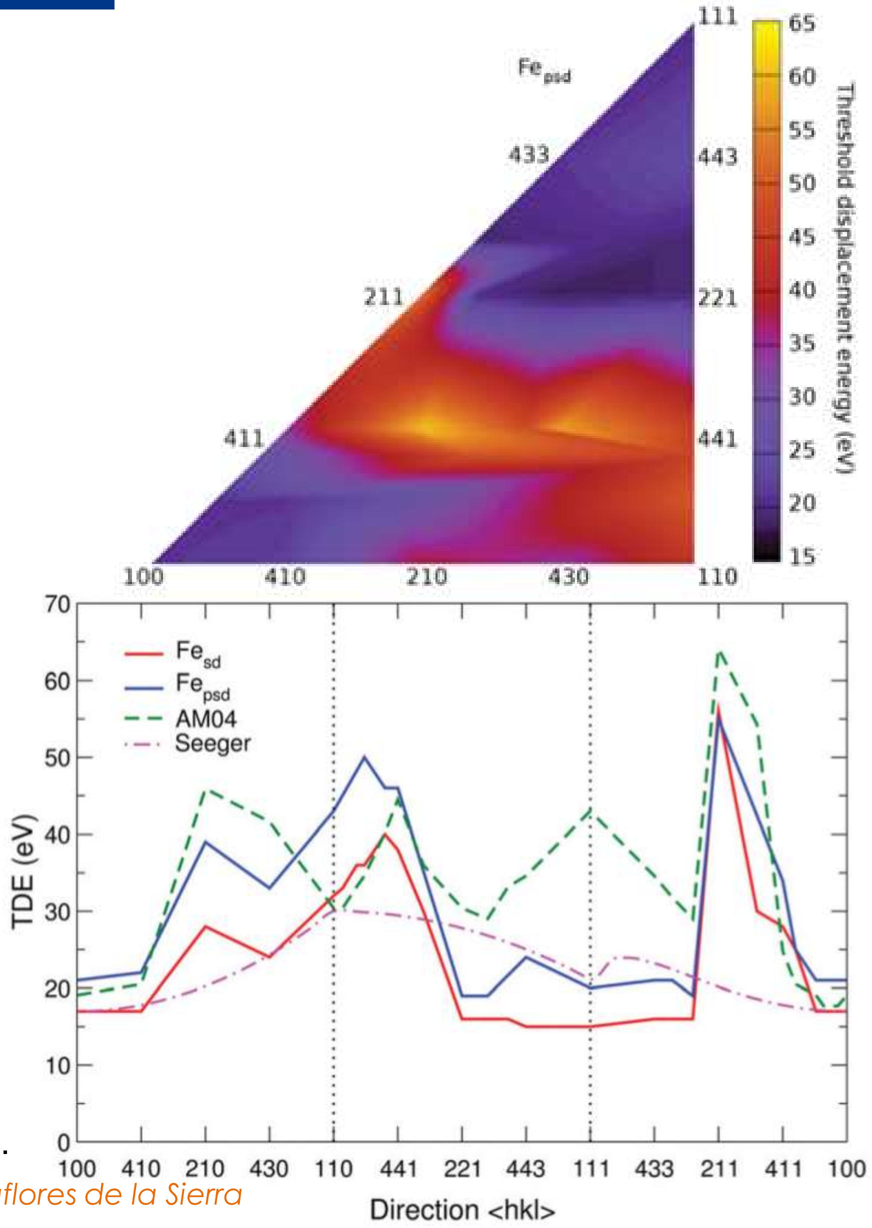
- ❑ The models and methods are only as good as their input
- ❑ Important to use best possible physics basis
- ❑ Large effort in SOTERIA to develop a mechanistic understanding of nanofeature evolution in RPV and internals

❑ First part based on the following SOTERIA publications:

- P. Olsson, C.S. Becquart, C. Domain, Mater. Res. Lett. **4** (2016) 219-225.
- L. Messina, M. Nastar, N. Sandberg, P. Olsson, Phys. Rev. B **93** (2016) 184302.
- L. Messina, N. Castin, C. Domain, P. Olsson, Phys. Rev. B **95** (2017) 064112.
- N. Castin, L. Messina, C. Domain, R. C. Pasianot, P. Olsson, Phys. Rev. B **95** (2017) 214117.
- M. Posselt, D. Murali, M. Schiwarth, Comp. Mater. Sci. **127** (2017) 284-294.
- C. Domain, C.S. Becquart, J. Nucl. Mater. **499** (2018) 582-594.
- C.S. Becquart, R.N. Happy, P. Olsson, C. Domain, J. Nucl. Mater. **500** (2018) 92.
- N. Castin, M.I. Pascuet, L. Messina, C. Domain, P. Olsson, R.C. Pasianot, L. Malerba, Comp. Mater. Sci **148** (2018) 116.

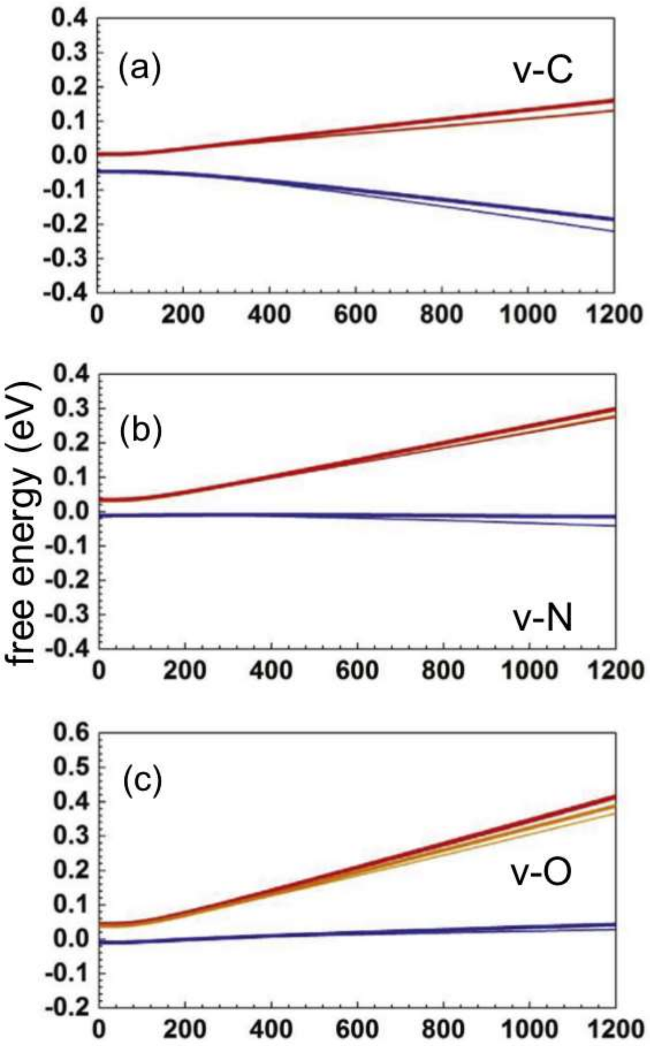
1) Threshold displacement energies

- Ab initio MD used to determine TDE in bcc Fe
- Some effect on average value (of reactor relevance)
  32 eV vs 40 eV
- Anisotropy different than canonical/historical models
- How to run AIMD simulations very important (approximation levels)
- AIMD results quite important for near-threshold conditions (NRT and KP should be modified for low energies)



P. Olsson, C.S. Becquart, C. Domain, Mater. Res. Lett. **4** (2016) 219-225.

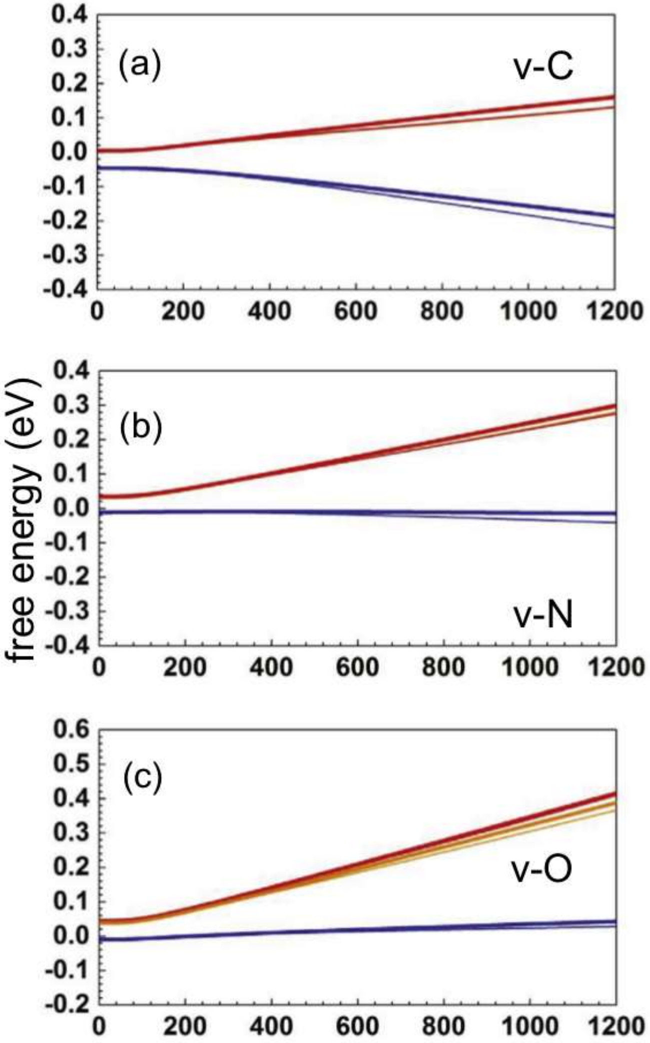
2) Free energy calculations in bcc Fe

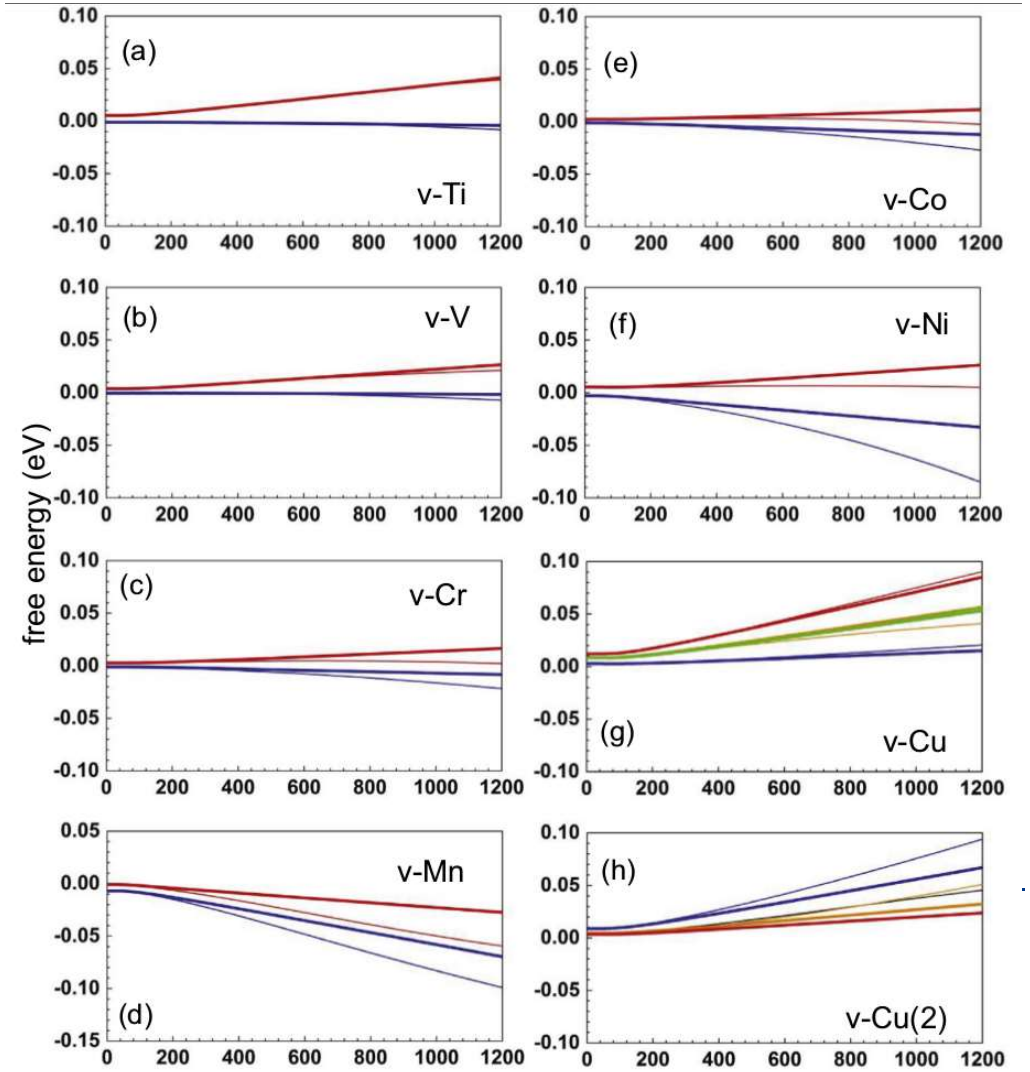


- ☐ Operation conditions are far from the DFT 0K conditions
- ☐ Free energy effects can be important **G = H – TS**
  - Phonons, electrons, magnons, anharmonicity, ...
- ☐ Vibrational free energy effects for small vacancy-solute clusters in bcc Fe
- ☐ Effect of modeling paradigm
- ☐ Method range of validity
- ☐ Order of magnitude (0.1 eV) can be important at operation conditions!
  - Not yet implemented in higher scale models

M. Posselt, D. Murali, M. Schiwarth, Comp. Mater. Sci. **127** (2017) 284-294.

2) Free energy calculations in bcc Fe





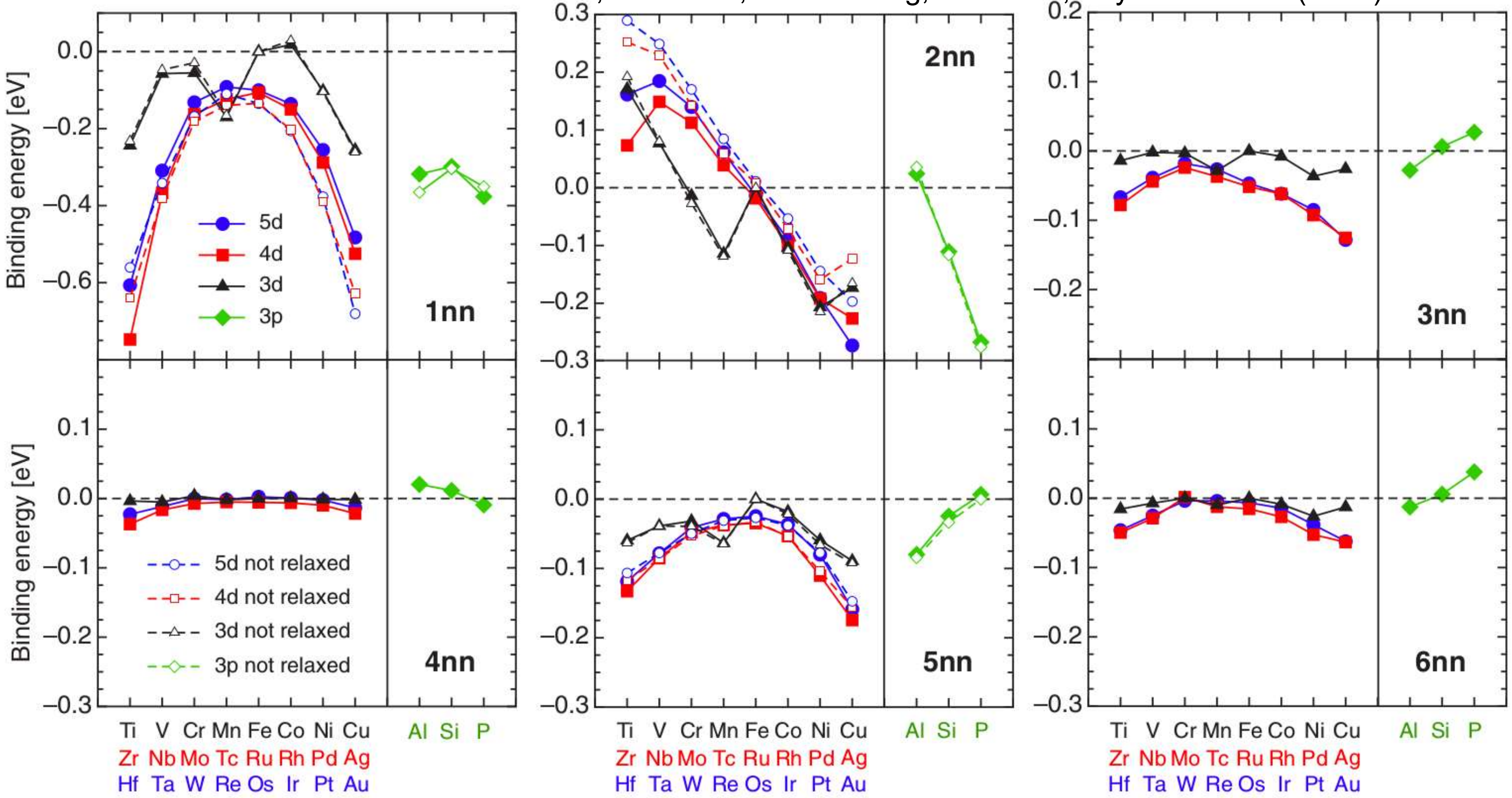
M. Posselt, D. Murali, M. Schiwarth, Comp. Mater. Sci. **127** (2017) 284-294.

3) Solute – vacancy **interactions** and kinetics in bcc Fe

- DFT database of solute-vacancy interactions
- Binding energies follow clear trends
- 3d-solutes affected by magnetism, 4d- and 5d mostly size effects

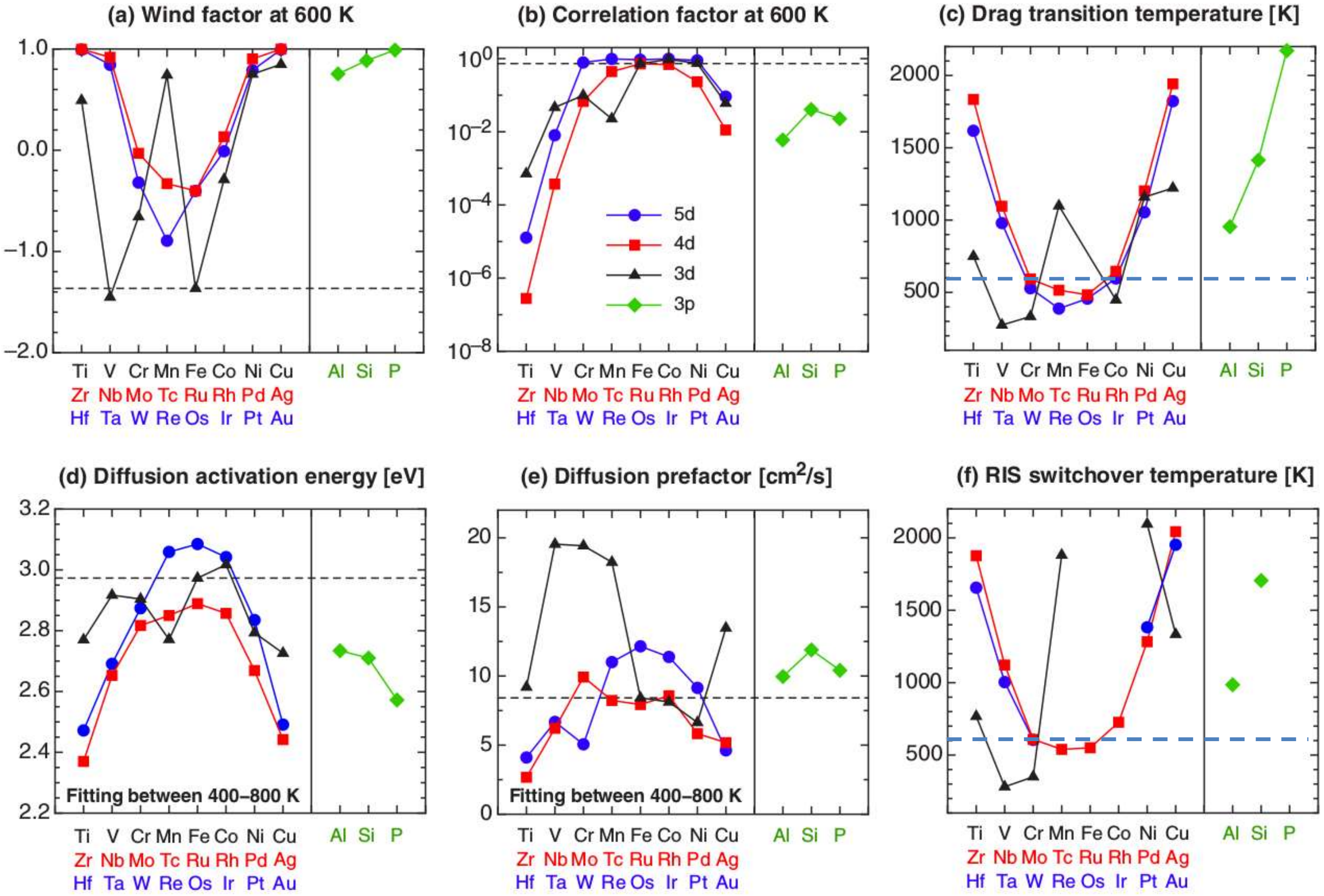
L. Messina, M. Nastar, N. Sandberg, P. Olsson, Phys. Rev. B **93** (2016) 184302.

VASP
PAW-PBE
250 atoms
bcc Fe
3^3 k-points
300 eV



3) Solute – vacancy interactions and **kinetics** in bcc Fe

- ☐ Self-consistent mean field theory coupling
- ☐ Solute drag by vacancies a general phenomenon (not limited to 1nn binding solutes!)
- ☐ Vacancy driven RIS enrichment for most RPV solutes
  - Exceptions Cr, V



(a) Wind factor at 600 K

(b) Correlation factor at 600 K

(c) Drag transition temperature [K]

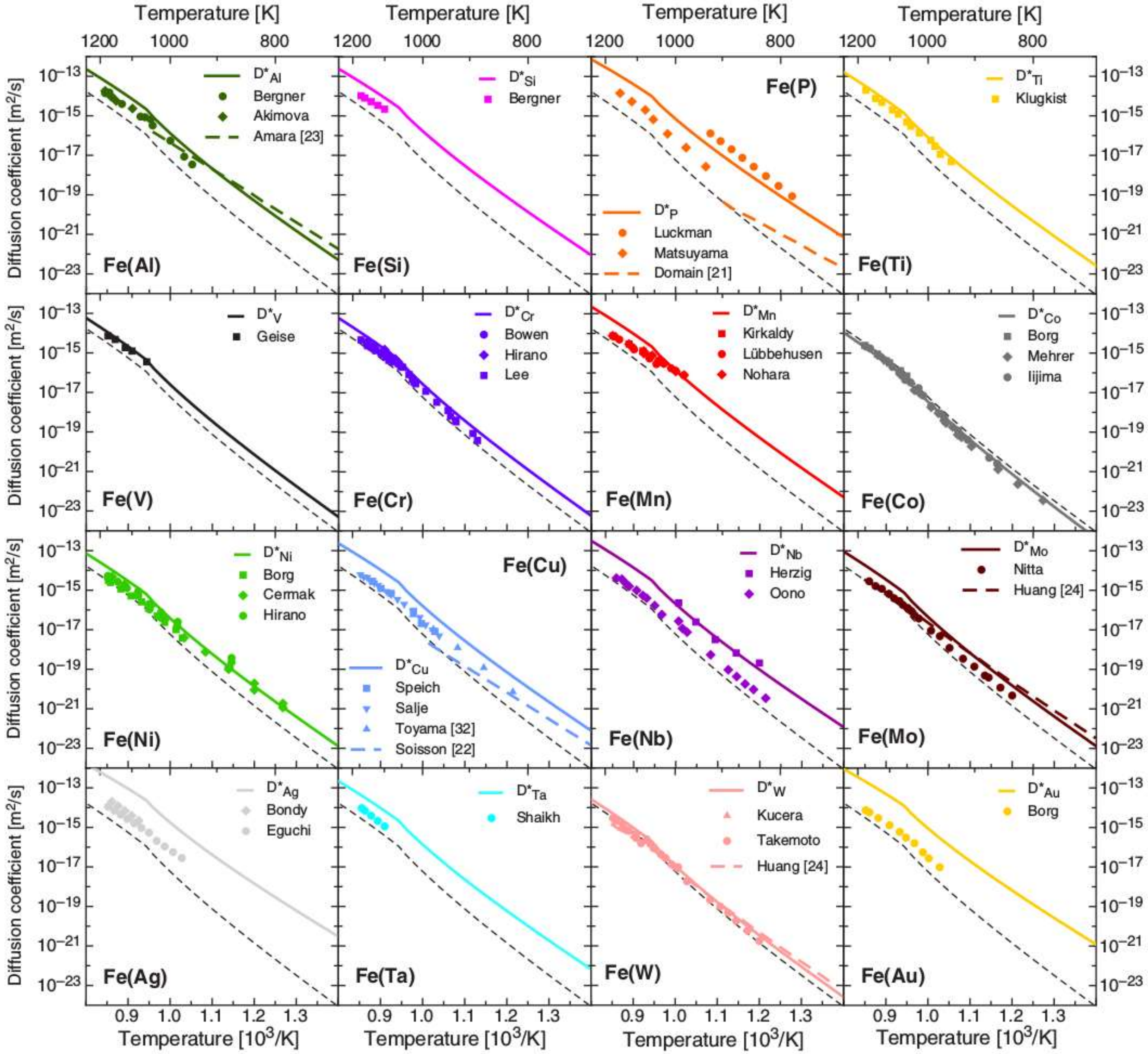
(d) Diffusion activation energy [eV]

(e) Diffusion prefactor [cm^2/s]

(f) RIS switchover temperature [K]

3) Solute – vacancy interactions and **kinetics** in bcc Fe

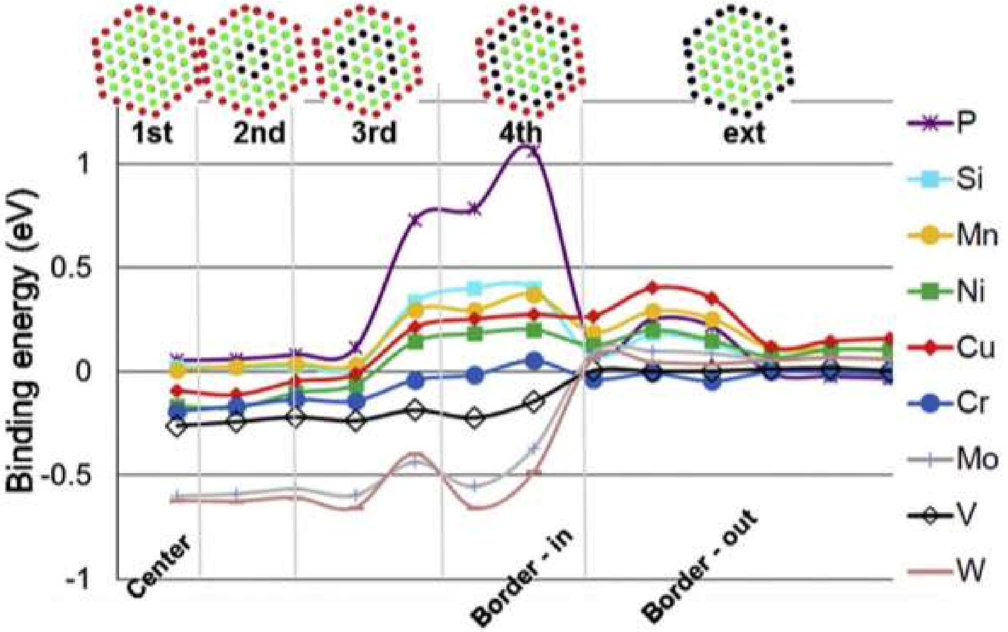
- Prediction of solute diffusion coefficients in good agreement with experiments
- Divergencies well understood
- Showcase of how to get low-T diffusion data

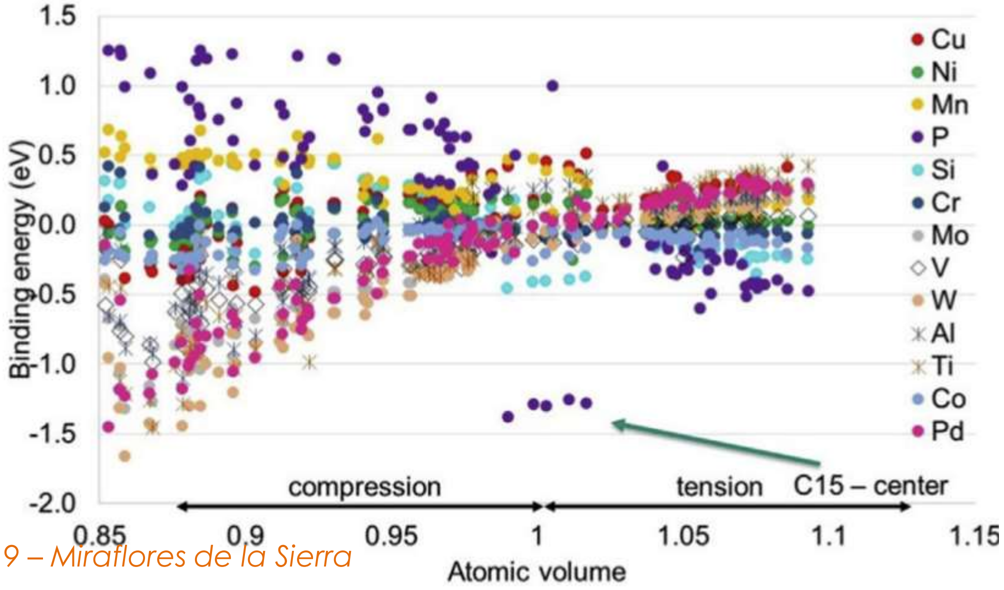


4) Solute – defect cluster interactions

- ❑ Large scale DFT calculations (1500 atoms)
- ❑ Provides new insight on how small SIA clusters interact with solutes
- ❑ Given attraction strength – small defects will "always" be trapped by solutes
- ❑ Some surprises!
  - Attracting/binding very general phenomenon
  - Size effects dominating (but also magn + local coordination)
  - Cr non-interacting (contradicting earlier work)

C. Domain, C.S. Becquart, J. Nucl. Mater. **499** (2018) 582-594.
C.S. Becquart, R.N. Happy, P. Olsson, C. Domain, J. Nucl. Mater. **500** (2018) 92.





MULTISCALE MODELS OF NANOFEATURE EVOLUTION IN RPV STEELS

Based on Deliverable D5.6 of the SOTERIA project

Multiscale modeling: microstructure evolution

- ❑ Microstructure evolution modelling using complementary strategies
- ❑ Comparison with large experimental data set
- ❑ Large effort in SOTERIA to develop a tools for mechanistic modelling of nanofeature evolution in RPV and internals

❑ Second part based on the following SOTERIA publications:

- De Backer et al. J. Phys.: Condens. Matter **30** (2018) 405701
- B. Pannier, PhD, Univ Lille, 2017
- G. Adjanor, 2 papers to be published in Phys Rev E. (arXiv:1808.10362, arXiv:1808.10715)
- L. Messina, N. Castin, C. Domain, P. Olsson, Phys. Rev. B **95** (2017) 064112.
- N. Castin, L. Messina, C. Domain, R. C. Pasianot, P. Olsson, Phys. Rev. B **95** (2017) 214117.
- N. Castin, M.I. Pascuet, L. Messina, C. Domain, P. Olsson, R.C. Pasianot, L. Malerba, Comp. Mater. Sci **148** (2018) 116.
- G. Bonny, C. Domain, N. Castin[1], P. Olsson[3], L. Malerba, Computational Materials Science **161** (2019) 309–320
- M. *Chiapetto*, L. *Messina*, C. S. Becquart, P. Olsson, L. Malerba, Nuclear Instruments and Methods in Physics Research Section B, **393** (2017) 105-109
- Castin et al., OKMC FeCuNiMnSiPCr submitted

Microstructure evolution modelling

Solute – defect properties
Binding energies
Migration energies
Transport coefficient

Primary damage
Cascade database

AKMC Hybrid AKMC-OKMC OKMC Cluster Dynamics

Cohesive models (rigid lattice):
Pair interaction
Cluster expansion
Concentration dependant pair model (CDP)
Neural network

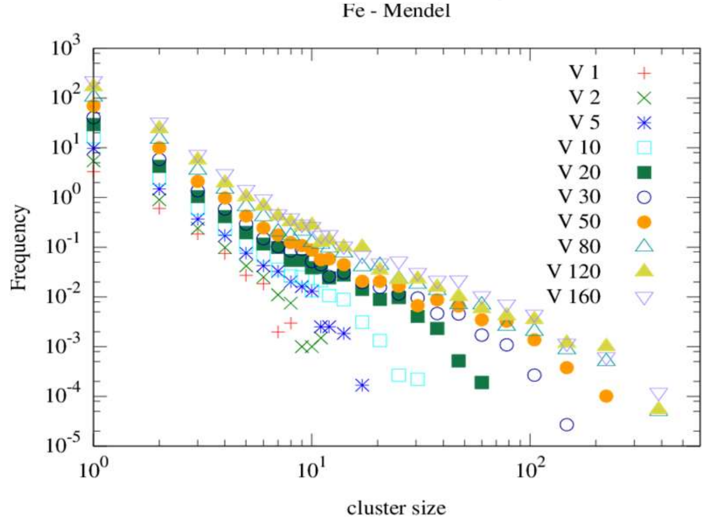
Parameterisation:
DFT & MD results
Effective concentration dependant model
Treatment of all solutes

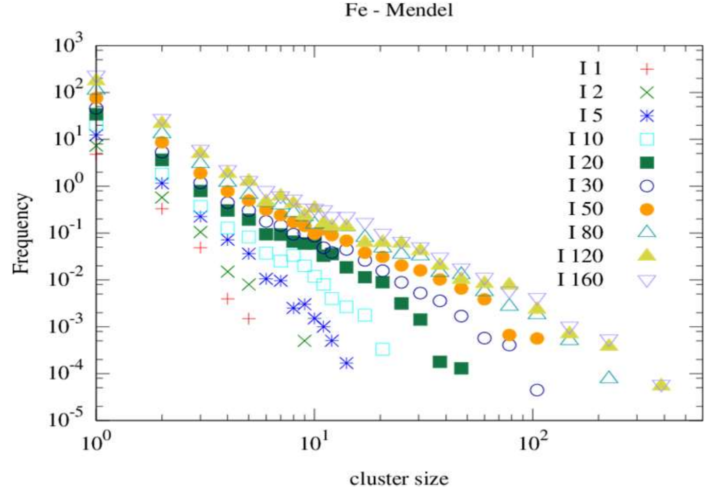
1D/3D modelling
Ion irradiation modelling

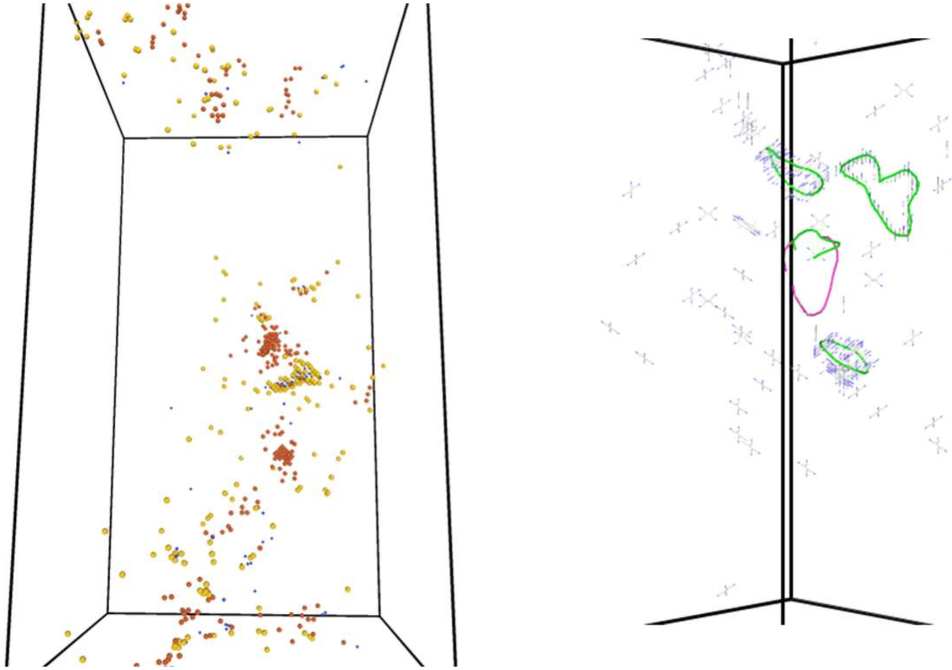
EAM potential FeCuNiMn → Metropolis MC (stability)

Primary damage:
new cascade database

- ☐ Thousands of displacement cascades







Formation of loops in the cascades

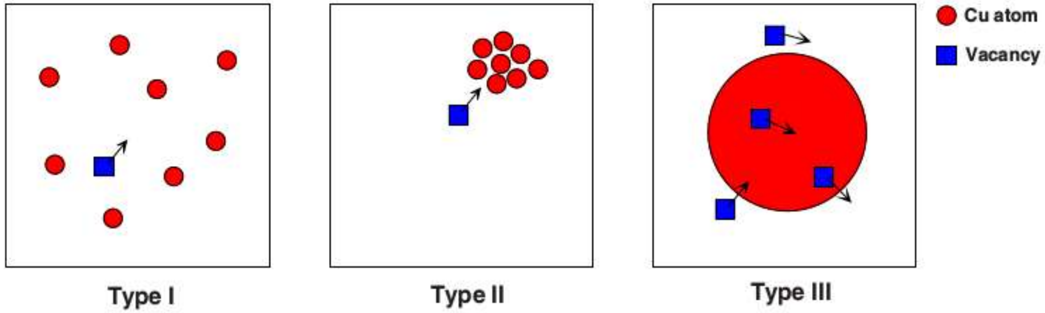
→ Quantification of the size distribution

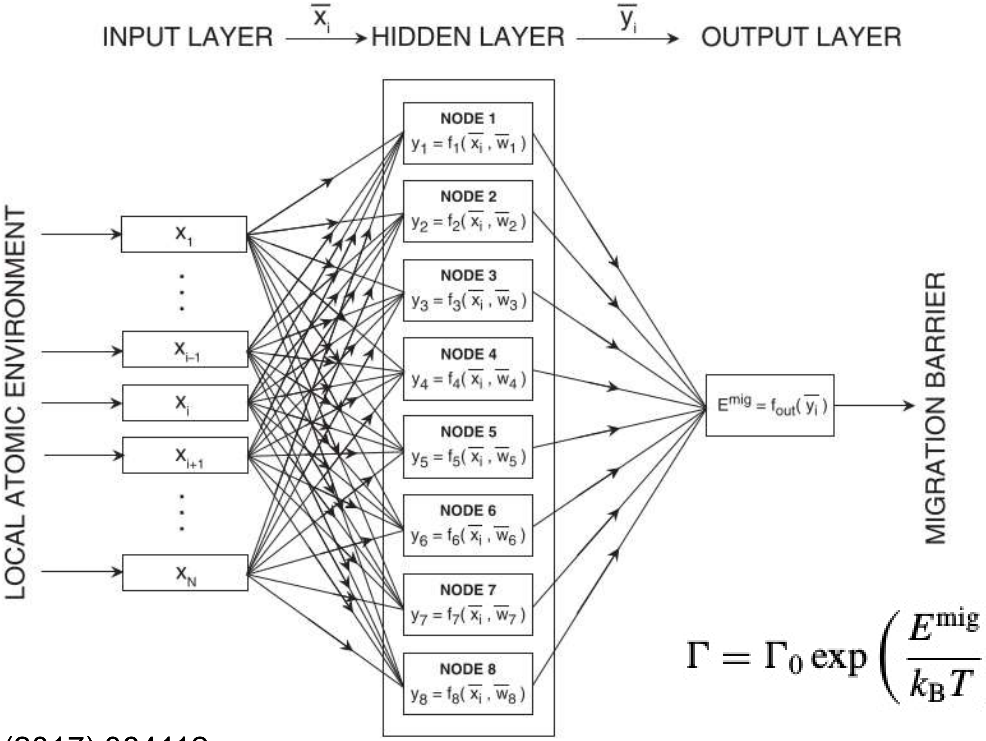
→ New unique cascade database in the SOTERIA plateform

De Backer et al. J. Phys.: Condens. Matter **30** (2018) 405701

Machine learning nanoscale evolution tools

- ❑ Artificial neural networks have recently begun to be applied to nanoscale evolution
- ❑ In SOTERIA, ANN’s were trained exclusively on DFT data (migration barriers) to transfer the physics basis fully to the nanoscale evolution
  - Examples:
    - FeCu + vacancies
    - FeCr + vacancies, SIAs
- ❑ A few thousand configurations required for each case (NEB’s)
- ❑ The ANN drives a hybrid AKMC/OKMC model



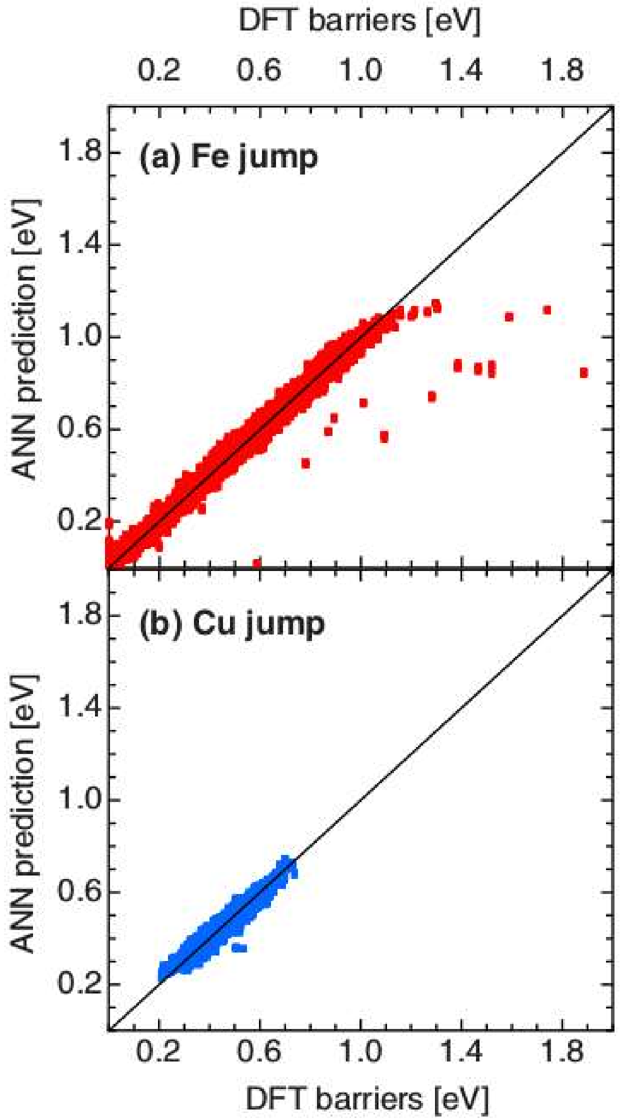


$$\Gamma = \Gamma_0 \exp\left(\frac{E^{\mathrm{mig}}}{k_{\mathrm{B}} T}\right)$$

L. Messina, N. Castin, C. Domain, P. Olsson, Phys. Rev. B **95** (2017) 064112.
N. Castin, L. Messina, C. Domain, R. C. Pasianot, P. Olsson, Phys. Rev. B **95** (2017) 214117.
N. Castin, M.I. Pascuet, L. Messina, C. Domain, P. Olsson, R.C. Pasianot, L. Malerba, Comp. Mater. Sci **148** (2018) 116.

Machine learning nanoscale evolution tools

- ☐ Successful training, good correlation factors
- ☐ The KMC simulations evolve according to the AKMC method, but when clusters grow beyond a threshold size, they become objects
- ☐ The OKMC part is first parameterized using AKMC
- ☐ Rapid KMC code framework
- ☐ Nanoscale evolution in FeCu and FeCr investigated



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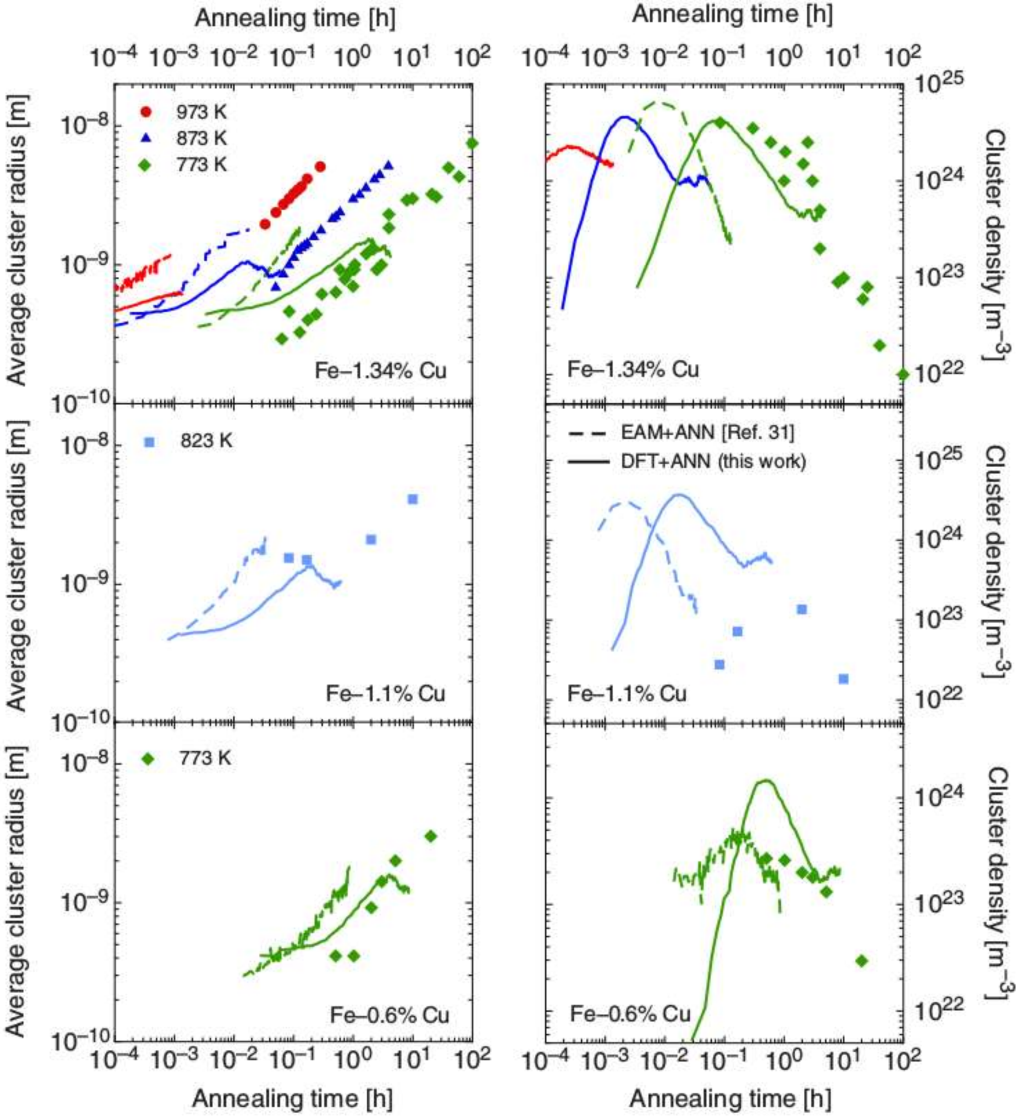
N. Castin, L. Messina, C. Domain, R. C. Pasianot, P. Olsson, Phys. Rev. B **95** (2017) 214117.
N. Castin, M.I. Pascuet, L. Messina, C. Domain, P. Olsson, R.C. Pasianot, L. Malerba, Comp. Mater. Sci **148** (2018) 116.

Machine learning nanoscale evolution tools

- ❑ Thermal ageing of FeCu
- ❑ No parameter adjustment – fully ab initio → KMC
- ❑ Very good agreement for 1.34% Cu
- ❑ Overestimated Cu-cluster density for 0.6% Cu → DFT solubility limit known issue for FeCu
- ❑ DFT physics fully transmitted to the KMC



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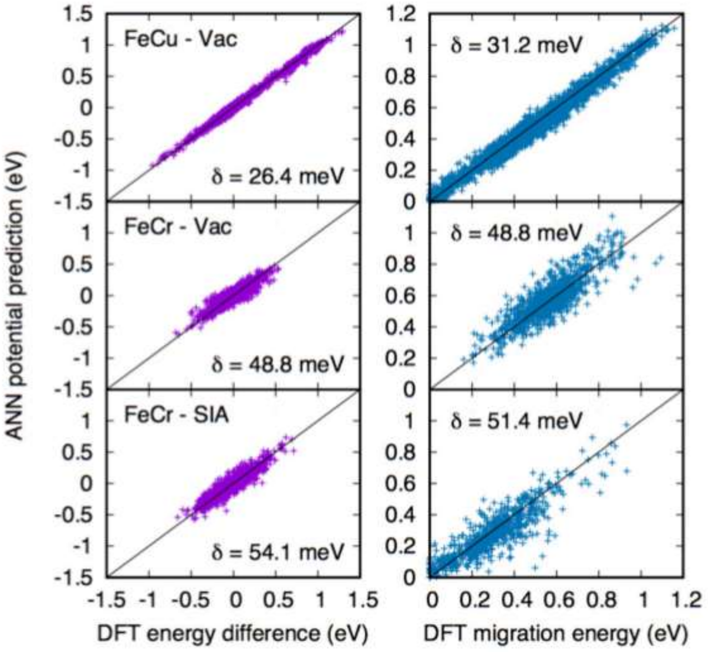
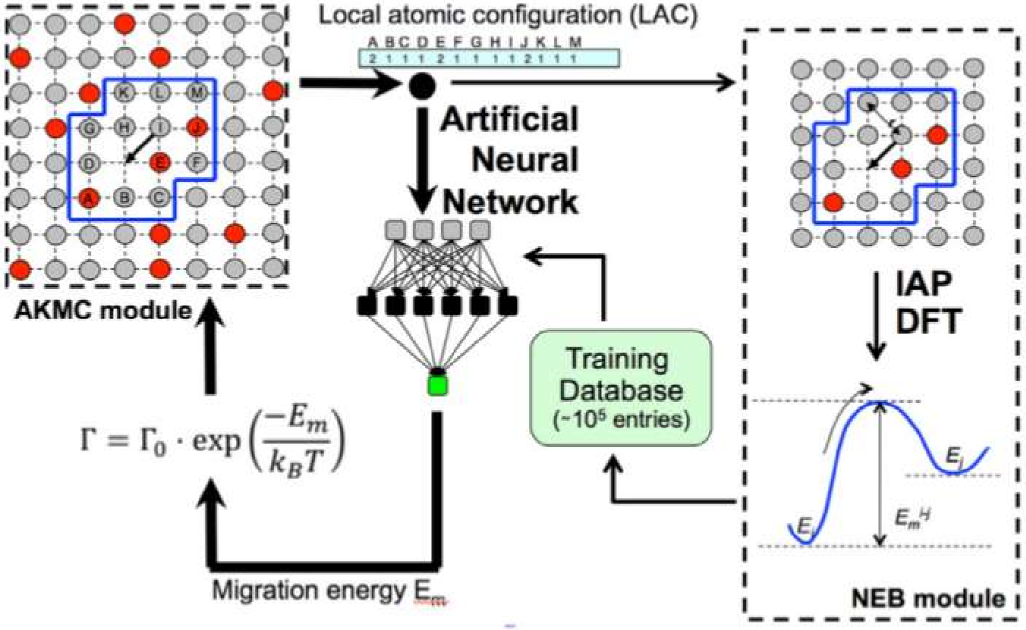
N. Castin, L. Messina, C. Domain, R. C. Pasianot, P. Olsson, Phys. Rev. B **95** (2017) 214117.
N. Castin, M.I. Pascuet, L. Messina, C. Domain, P. Olsson, R.C. Pasianot, L. Malerba, Comp. Mater. Sci **148** (2018) 116.

Machine learning nanoscale evolution tools

- ☐ Improved KMC/MD motor by fitting ANN potentials

- ☐ ANN pot trained first





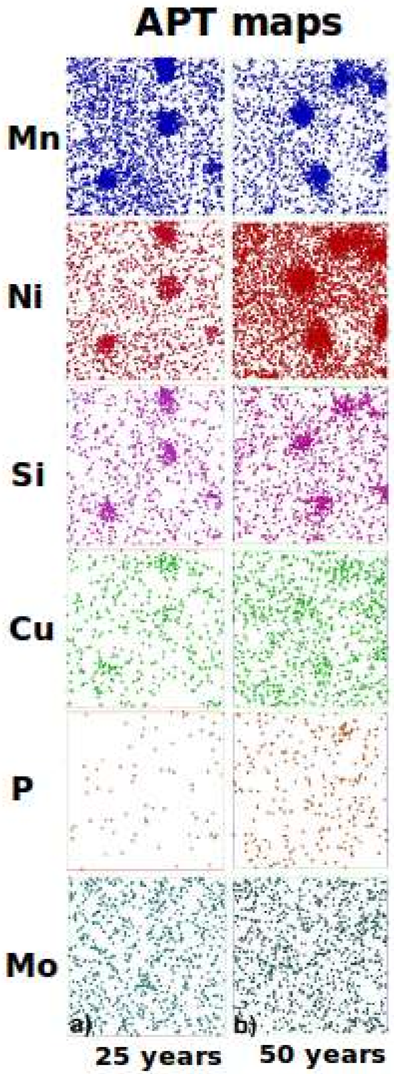
- Many more barriers can be used by training KMC motor on ANN potential predictions

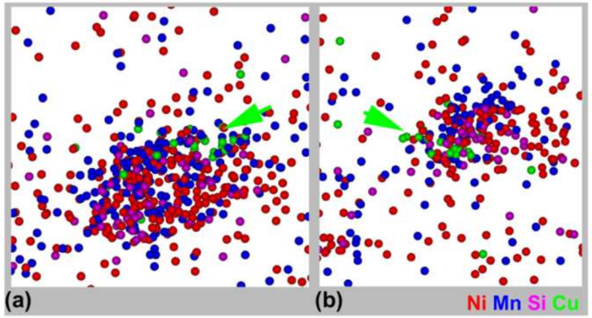
Conclusions

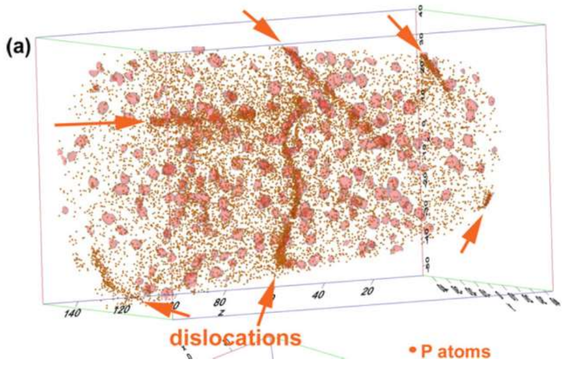
- ❑ Machine learning can be used to transfer the physics basis directly through the KMC scale
- ❑ Same power of analysis of mechanisms but no control over parameters to adjust and perform sensitivity studies
- ❑ Very computationally demanding
- ❑ DFT-ANN-KMC simulations are predictive – and in very good agreement with experiments

Modeling neutron irradiation in RPV steels

- ❑ **Issue:** Observed growth of solute clusters (Ni,Mn,...)
- ❑ Late-blooming effect or not?
- ❑ Mechanism for cluster growth?
- ❑ Object KMC model developed; Applied to model alloy and to RPV steel (Ringhals weld)





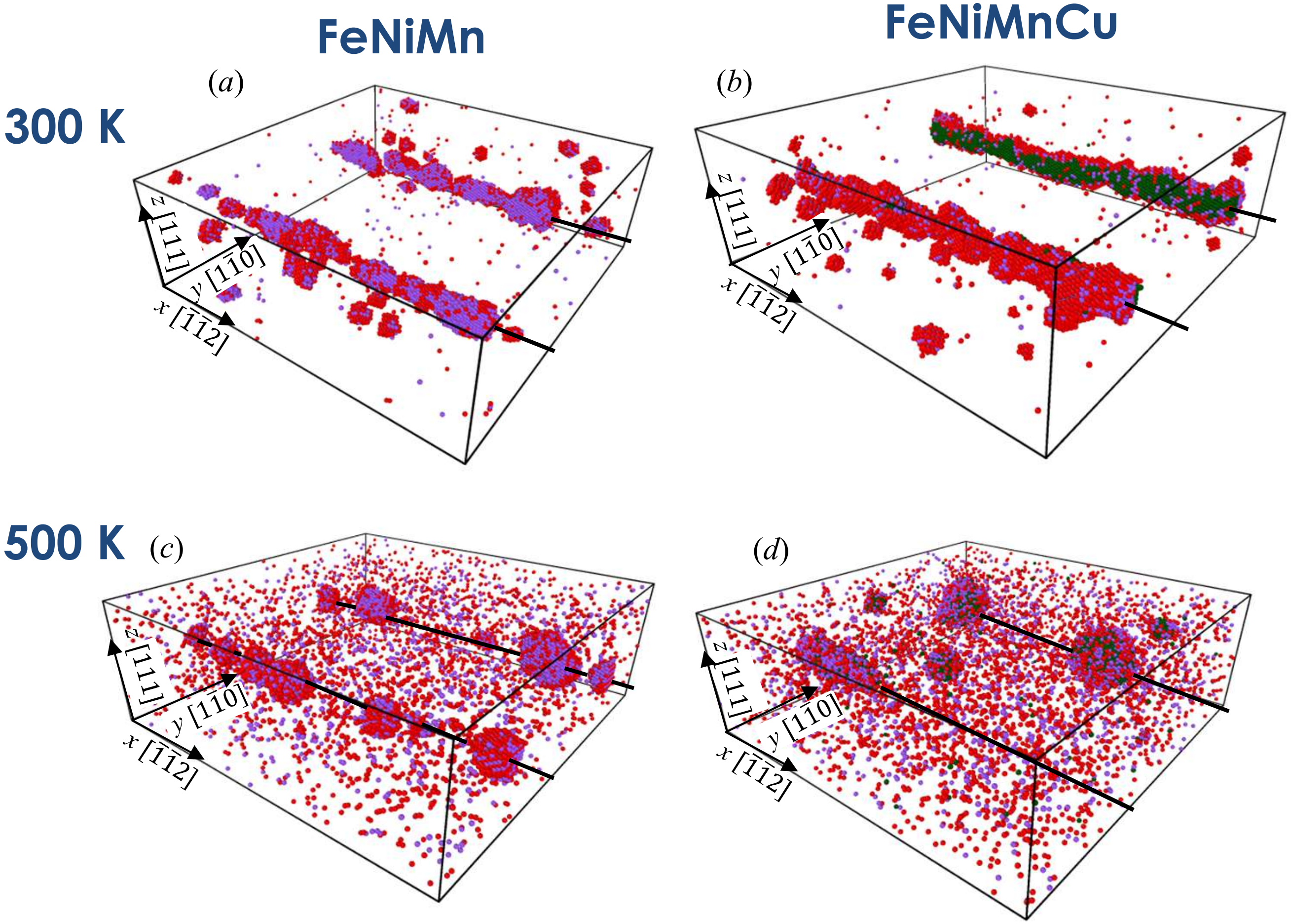


P. Efsing *et al.*, J. ASTM Int. **4** (2004).
Miller et al., *Journal of Nuclear Materials* **437** (2013).
US-NRC, Regulatory Guide 1.99 (1975).

Application of Metropolis MC

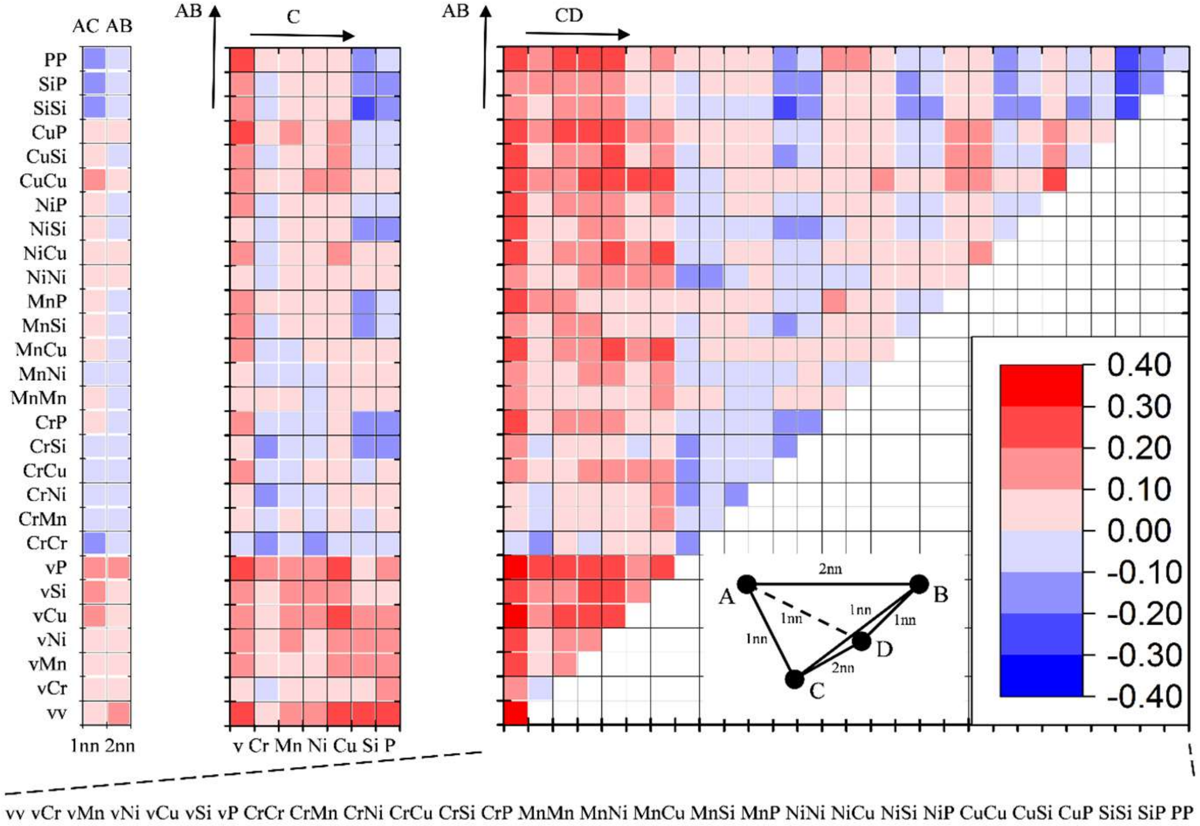
Study Ni, Mn, Cu, tendency to segregation at dislocations

Segregation of Ni, Mn and Cu at dislocations is energetically favoured in addition to being the consequence of solute transport towards sinks



M.I. Pascuet et al. Journal of Nuclear Materials 494 (2017) 311

Impressive examples of large scale DFT calculations for RPV steels use for cohesive model construction

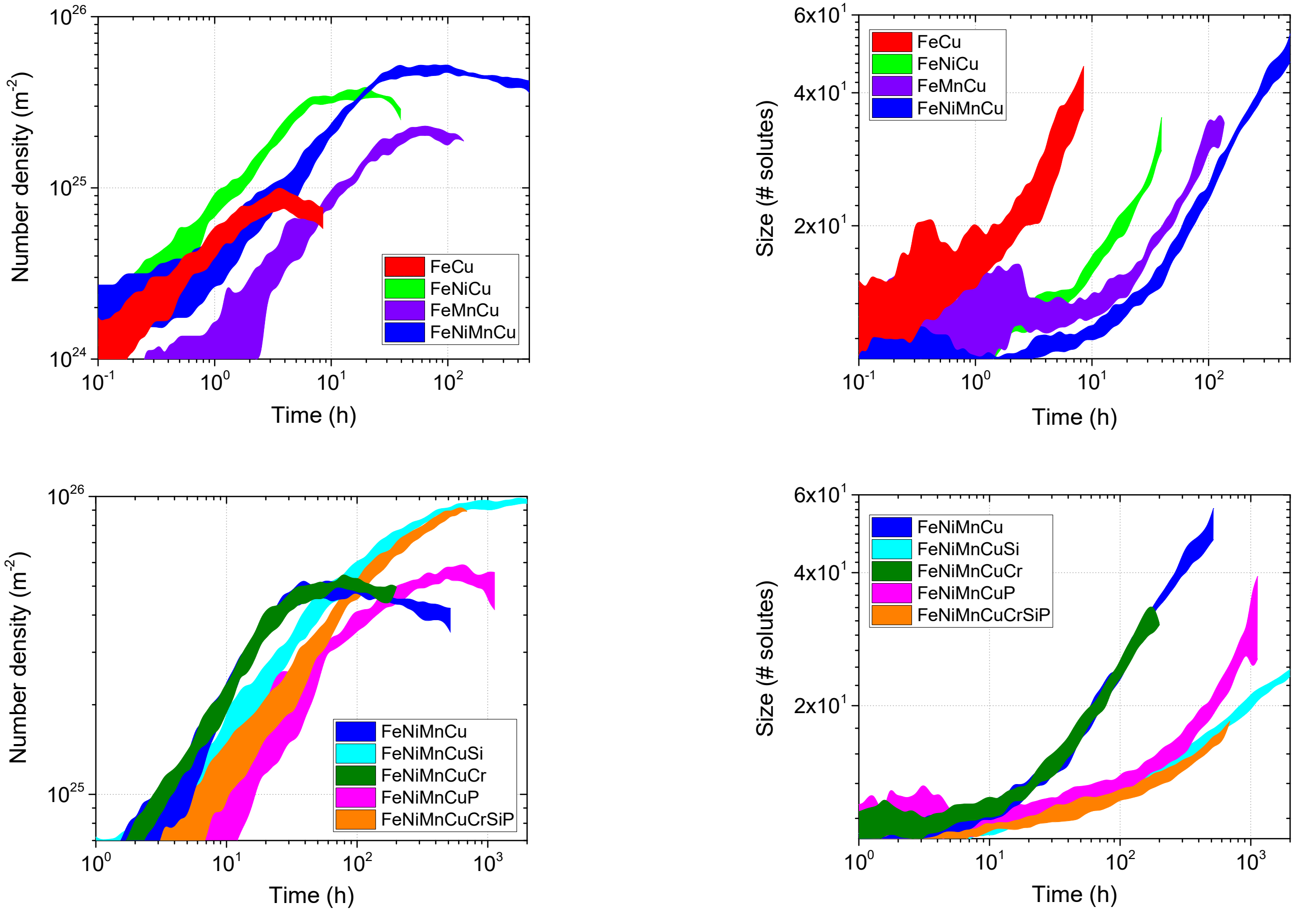


DFT database subset example

> 700 data points of binding energies up to triplets and quadruplets of solutes and vacancies

This study provides key data for the development of models that describe microstructural evolution in irradiated RPV steels

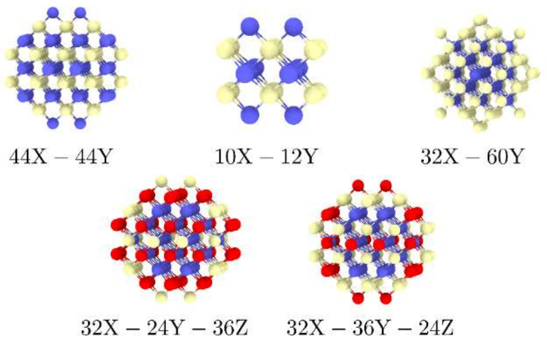
Cluster Expansion FeCrNiMnSiP

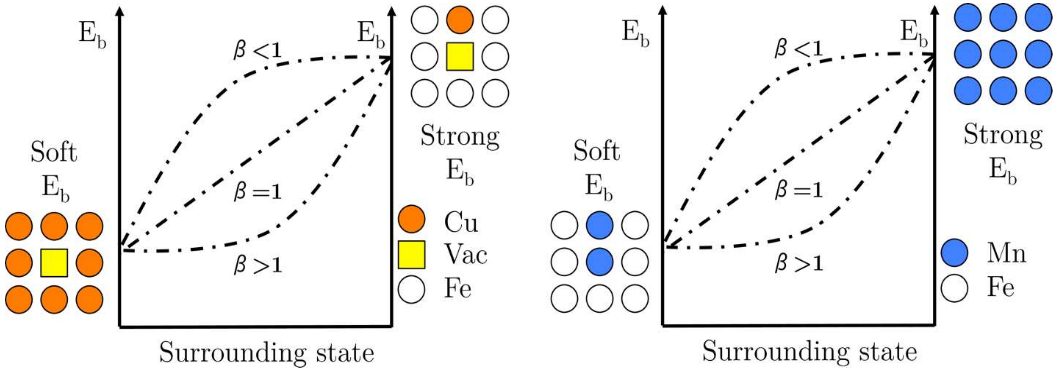


G. Bonny, C. Domain, N. Castin[1], P. Olsson[3], L. Malerba, *Computational Materials Science 161 (2019) 309–320*

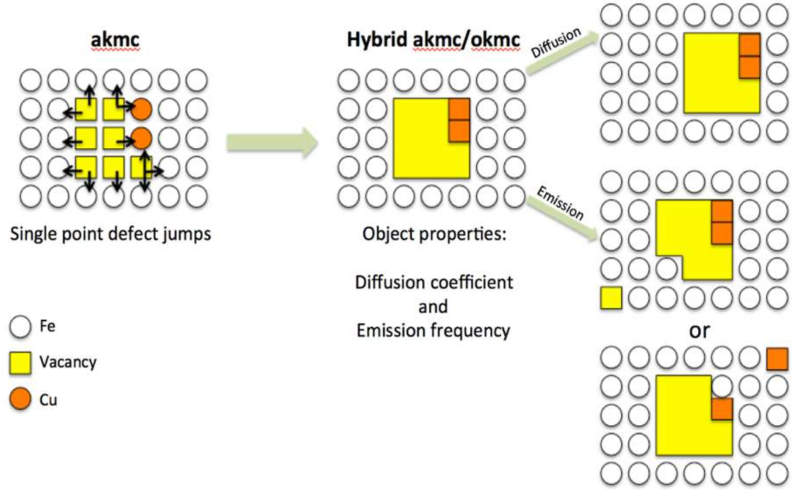
MICROSTRUCTURE EVOLUTION OF Fe-CuMnNiSi by AKMC BASED ON DFT

Development of an hybrid AKMC – OKMC model for complex alloys
Development of a new parameterization: Concentration Dependent Pair cohesive model adjusted on large database DFT calculations to describe energies of both pair interaction and Solute-vacancy clusters



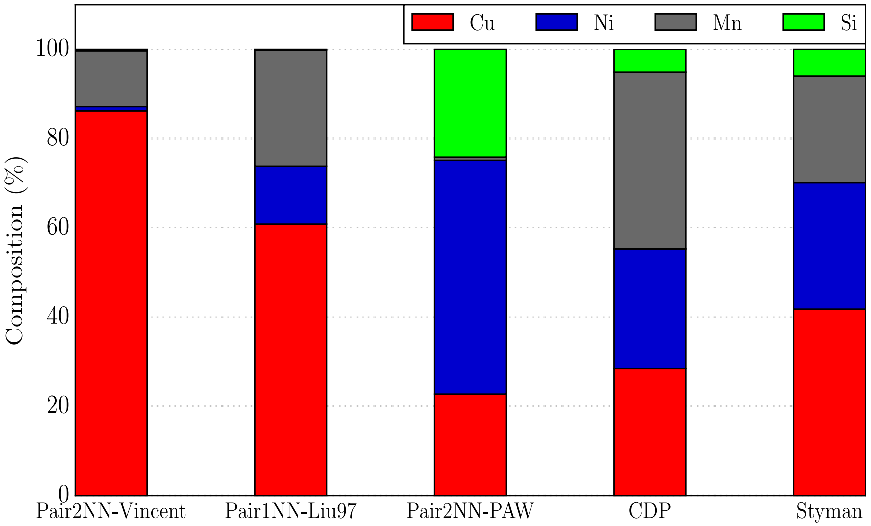


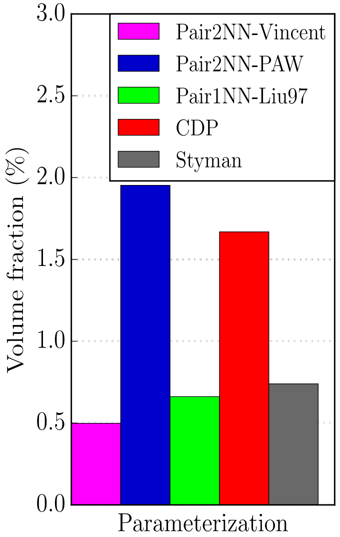
Hybrid AKMC – OKMC model



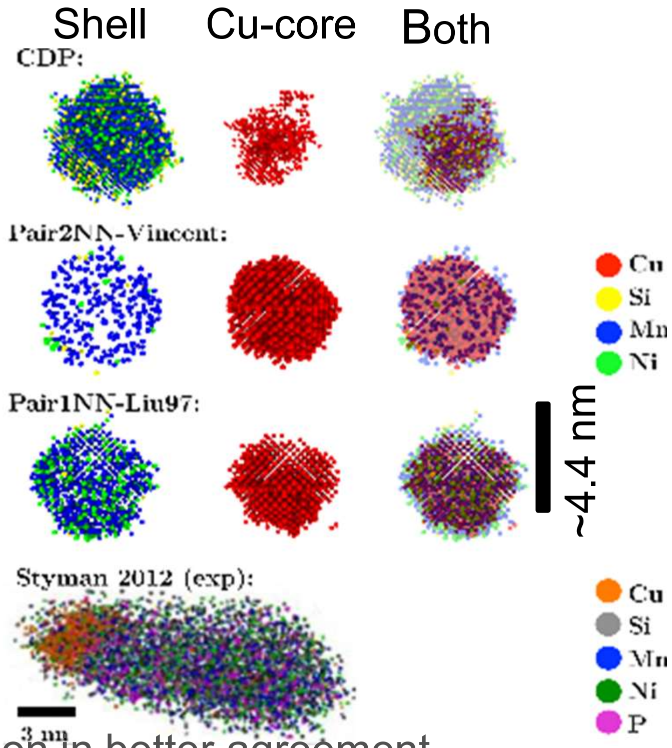
B. Pannier, PhD, 2017

MICROSTRUCTURE EVOLUTION OF Fe-CuMNNiSi by AKMC BASED ON DFT



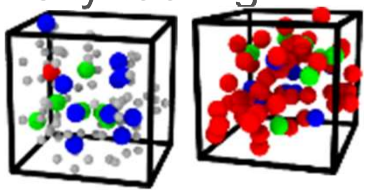


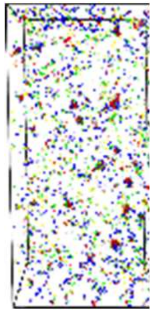
(90000 h) [1]



Thermal ageing: formation of solute rich cluster with composition in better agreement with experiments.

Microstructure under irradiation: better composition (cluster density too high and size too small compared to TAP experiments)



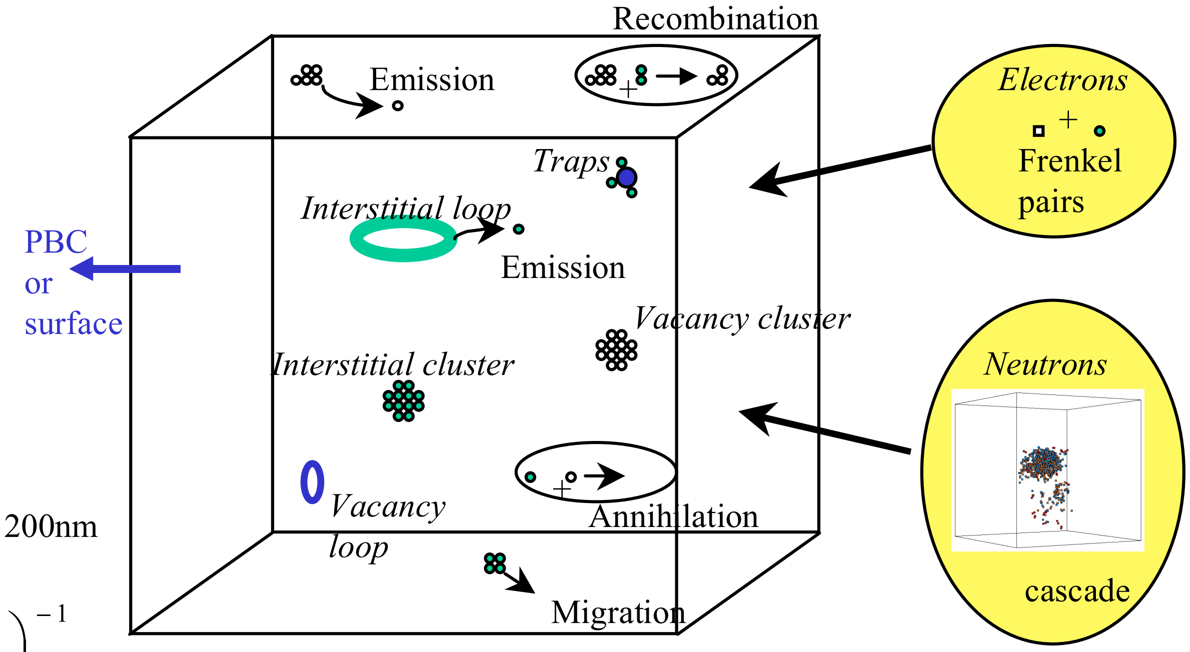


[1]: Styman et al., Progress in Nuclear Energy, 57, 2012

B. Pannier, PhD, 2017

Object Kinetic Monte Carlo

- Each object defined by:
  - type
  - centre-of-mass position
  - reaction radius
  - possible reactions



$\Gamma_i = \Gamma_i^0 \exp(-E_a / kT)$

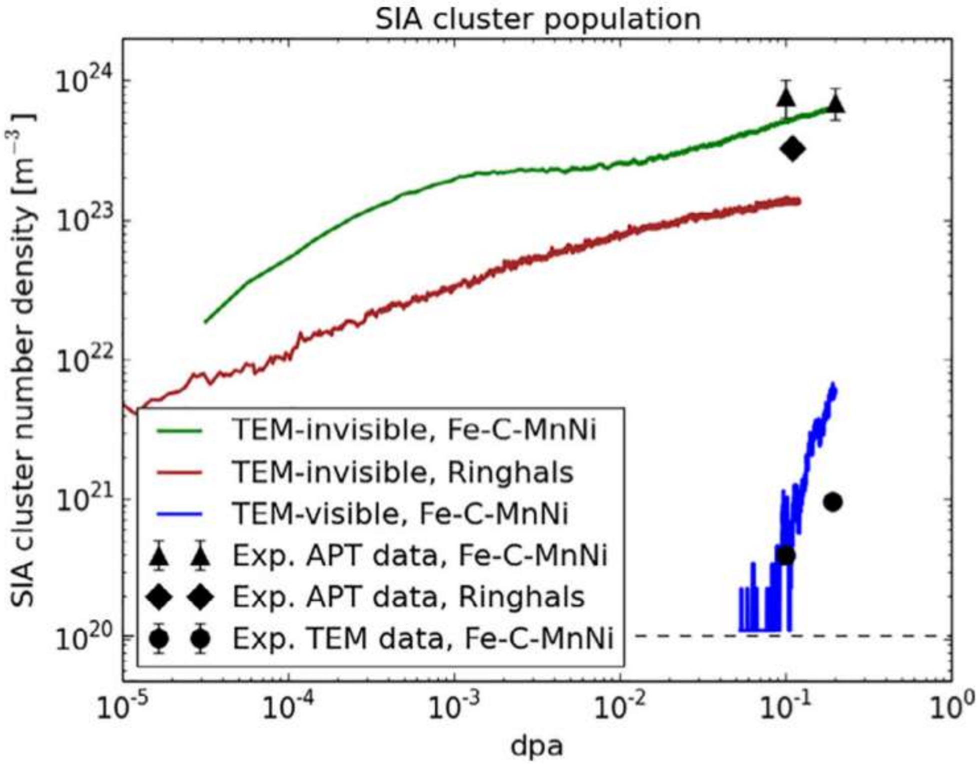
$$\text{Time step} = \left(\sum_{\substack{internal \\ events}} \Gamma_i + \sum_{\substack{external \\ events}} \Gamma_i \right)^{-1}$$

- Advantages:
  - Flexibility
  - Computing efficiency
  - Spatial distribution
- Drawbacks:
  - Large number of physical parameters
  - No atomic configurations

Modeling neutron irradiation in RPV steels

- **Issue:** Growth of solute clusters (Ni,Mn,...) observed
- Object KMC model developed
- Applied to model alloy and to RPV steel (Ringhals weld)
- Main ideas differentiating the alloy from the metal:
  - Grey alloy approach
  - SIA cluster diffusivity reduction due to solute interaction (from DFT)
  - 1D/3D motion depending on cluster size

$$\frac{D_n^{FeMnNi}}{D_n^{Fe}} = exp\left[-\frac{9C(E^b_{Mn}x_{Mn}+E^b_{Ni}x_{Ni})}{k_BT}\right]$$

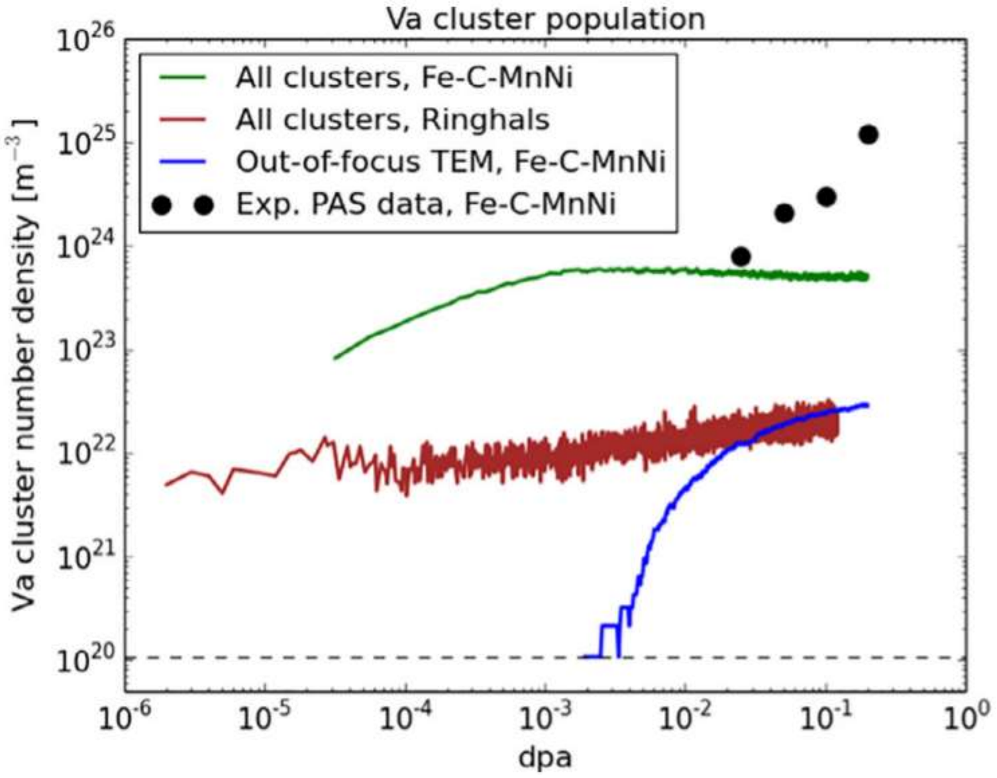


M. Chiapetto, L. Messina, C.S. Becquart, P. Olsson, L. Malerba, Nucl. Instr. Meth. Phys. Res. B **393** (2017) 105-109.

Modeling neutron irradiation in RPV steels

- Both SIA and vacancy cluster evolution is well represented by the model
- Importance of considering the experimental resolution!
- Dose rate effect:
  - clear predominance of single defects and smaller clusters at low dose rates

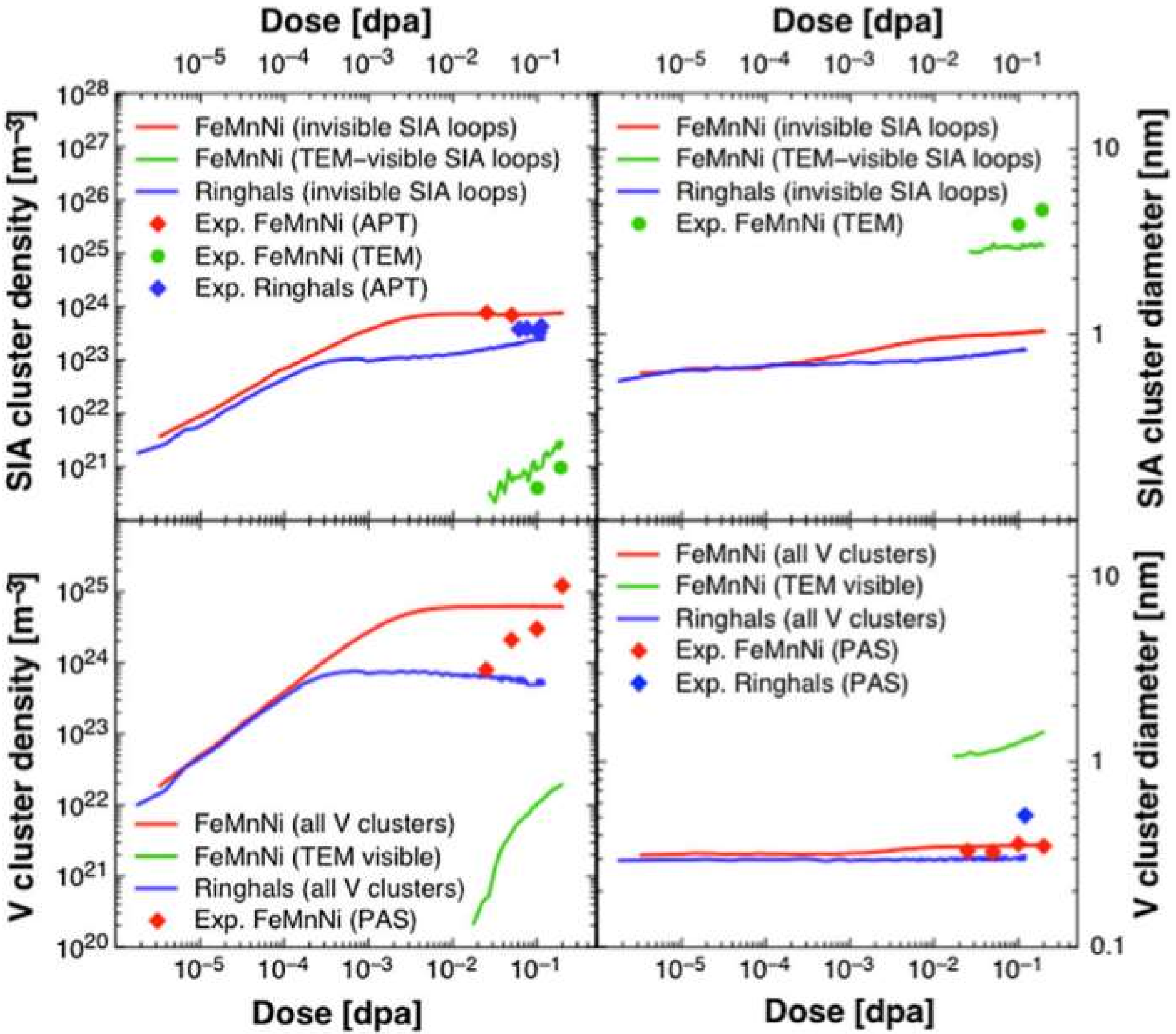
Property	Fe-C-MnNi [28]	Ringhals welds [9]
Composition [at.%]	1.2% Mn, 0.7% Ni	1.37% Mn, 1.58% Ni
Temperature	290 °C	284 °C
Neutron flux	$9.5 \cdot 10^{13}$ n/cm²s	$1.5 \cdot 10^{11}$ n/cm²s
Dpa flux	$1.4 \cdot 10^{-7}$ dpa/s	$2.7 \cdot 10^{-10}$ dpa/s [29]
Max dpa dose	0.2 dpa	0.12 dpa [29]
Carbon in matrix	134 at. ppm	100 at. ppm [30]
Dislocation density	$7 \cdot 10^{13}$ m^{-2}	NA
Average grain size	88 µm	NA



M. Chiapetto, L. Messina, C.S. Becquart, P. Olsson, L. Malerba, Nucl. Instr. Meth. Phys. Res. B **393** (2017) 105-109.

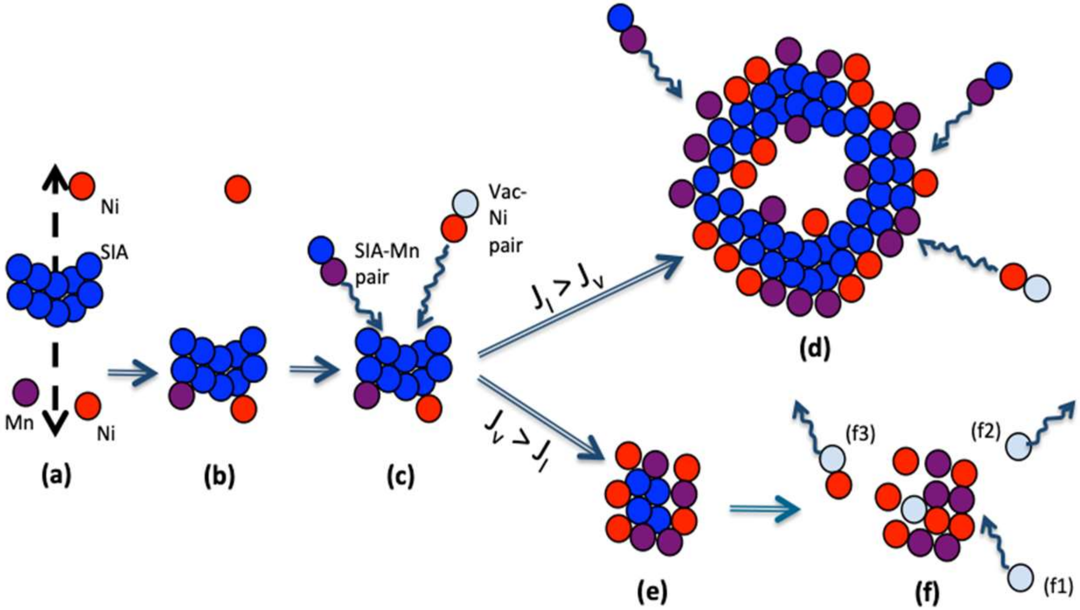
Modeling neutron irradiation in RPV steels

- ❑ Further model refinement:
  - RPV dislocation density
  - Role of dislocation bias
  - Vacancy cluster parameters refined using AKMC



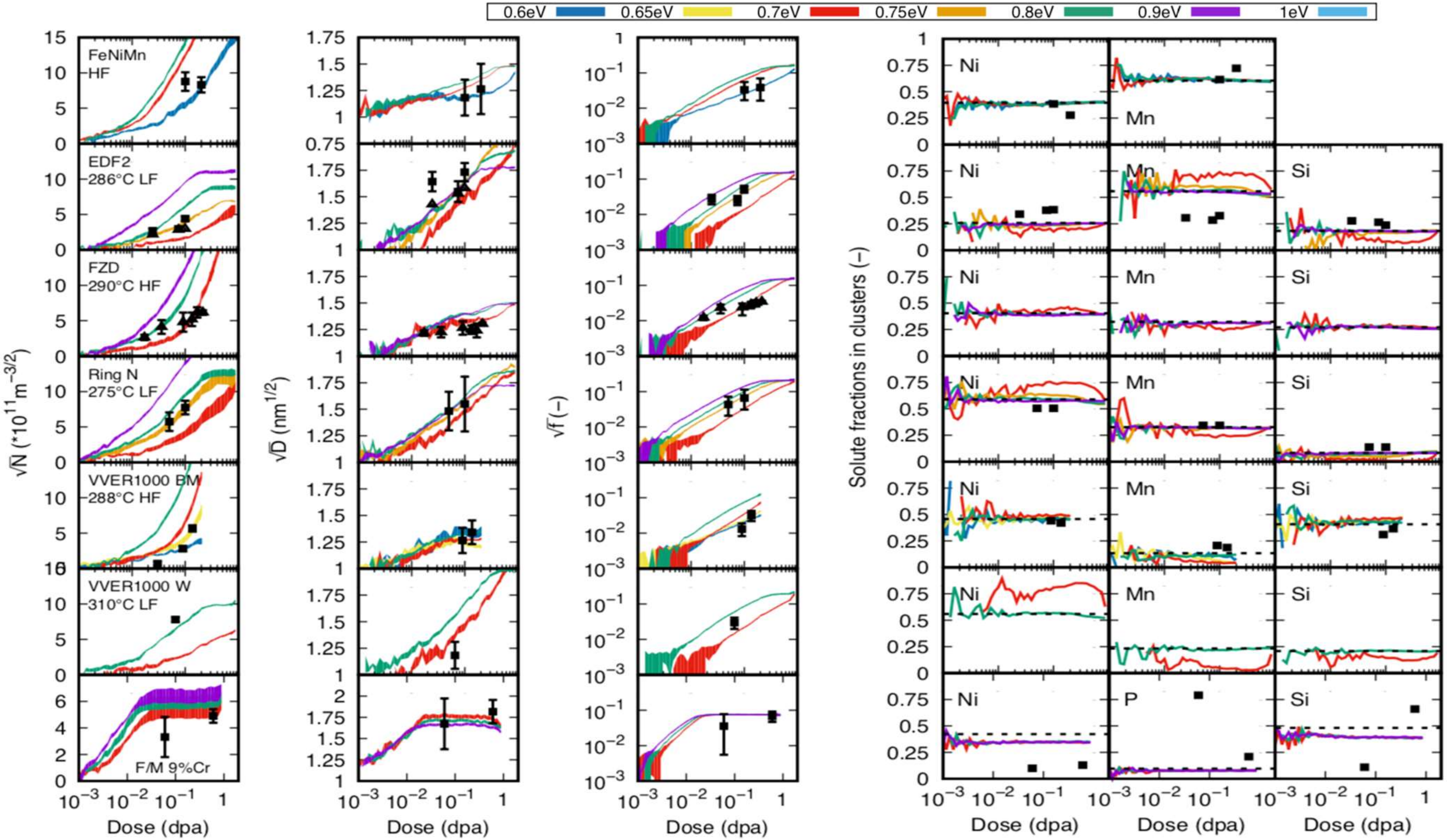
L. Messina, M. Chiapetto, P. Olsson, C.S. Becquart, L. Malerba, Phys. Stat. Solidi A **213** (2017) 2974

Microstructure evolution FeCuNiMnSiP by OKMC – explicite solute treatment



N. Castin et al, submitted

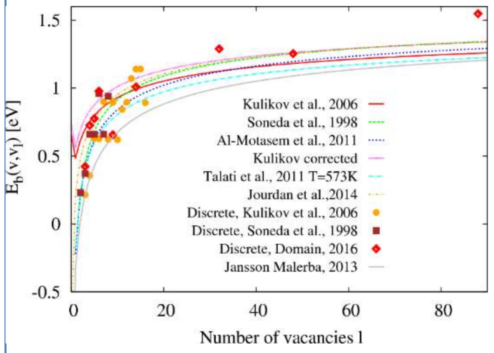
Microstructure evolution FeCuNiMnSiP by OKMC – explicite solute treatment

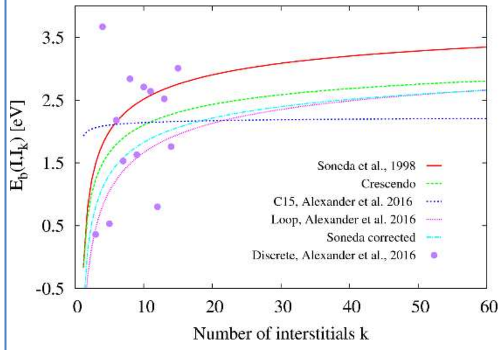


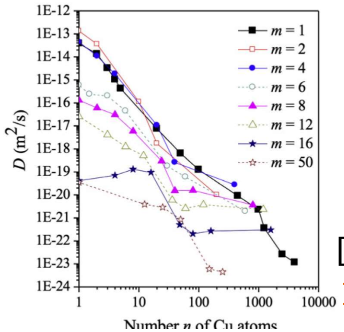
N. Castin et al, submitted

Cluster dynamics – irradiation Fe-0.1%Cu – 0.1 dpa

DFT, MD, AKMC results

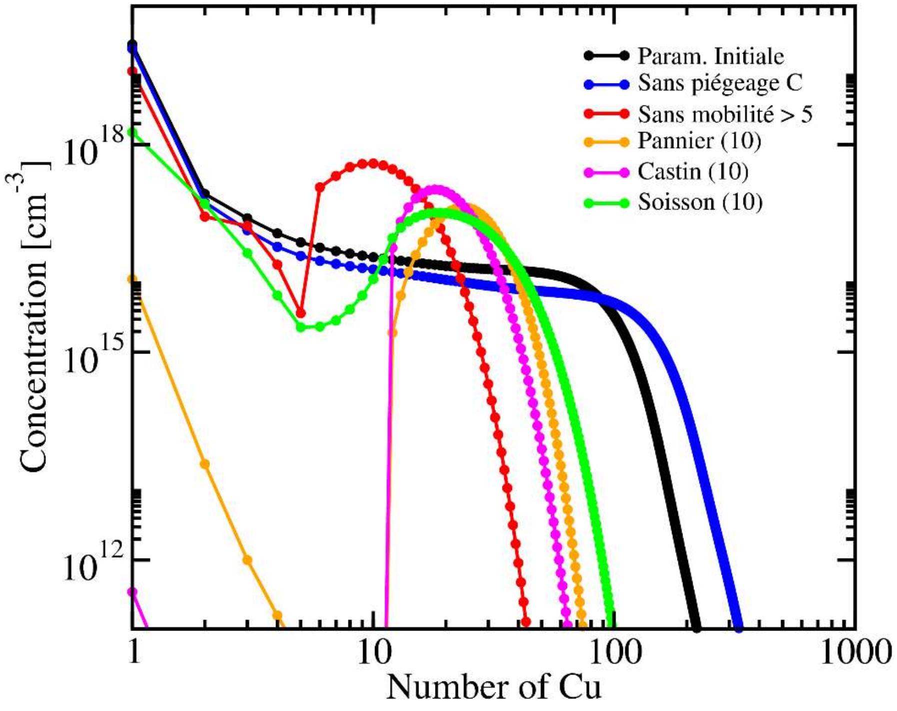






[Castin 2012]

CRESCENDO (co-dev CEA-EDF)



In progress:

- Treat multi-component alloys
- 1D/3D + trapping

Modeling neutron irradiation in RPV steels

❑ Conclusions:

- In RPV steels, the mechanism proposed for the observed growth of small solute clusters (Ni, Mn, Si, ...) has been heterogeneous nucleation on defect clusters

- That mechanism is here strengthened – the SIA cluster density perfectly matches the APT-seen solute cluster density

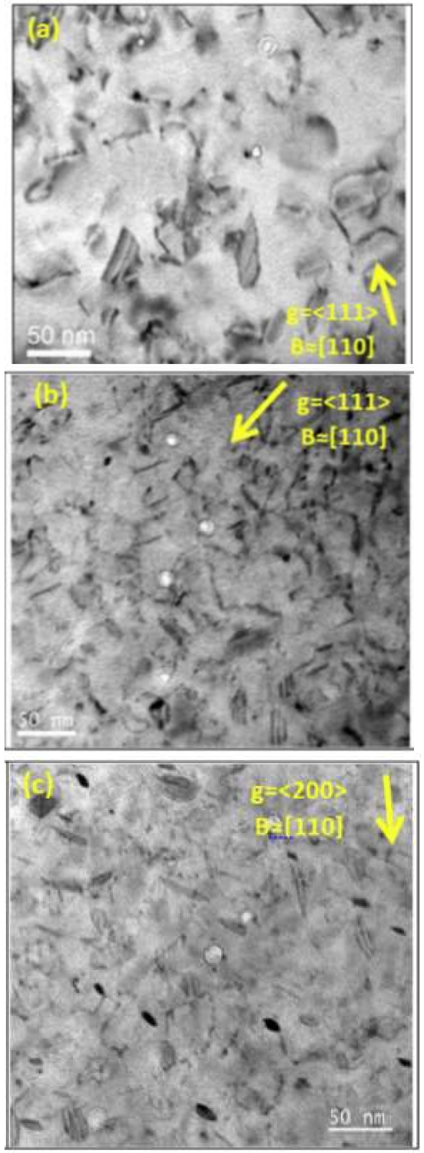
- Ab initio data and mean-field kinetics (many studies) support the mechanism

MODELING ION BEAM IRRADIATION IN AUSTENITIC STEELS

The role of injected interstitials

Self-ion irradiation of austenitic steel

- ❑ Ion beams often used as surrogates for neutron irradiation
  - Many issues with that! (flux, spatial distribution, injected SIAs, surface, ...)
- ❑ Injected SIAs have been shown to play a role (Lee JNM 1979)
- ❑ Cluster dynamics model developed here to study the issue in 304L steel
- ❑ CRESCENDO code



304L
450°C

5 dpa

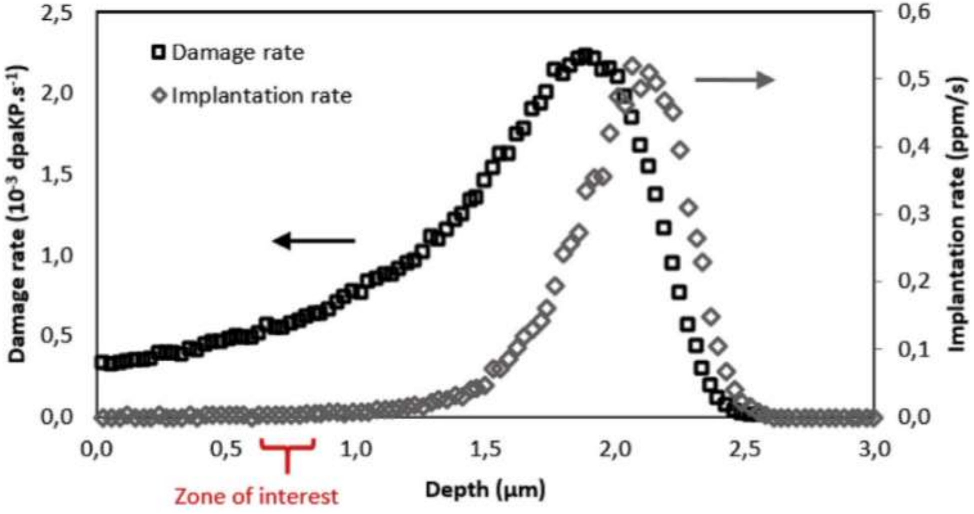
40 dpa

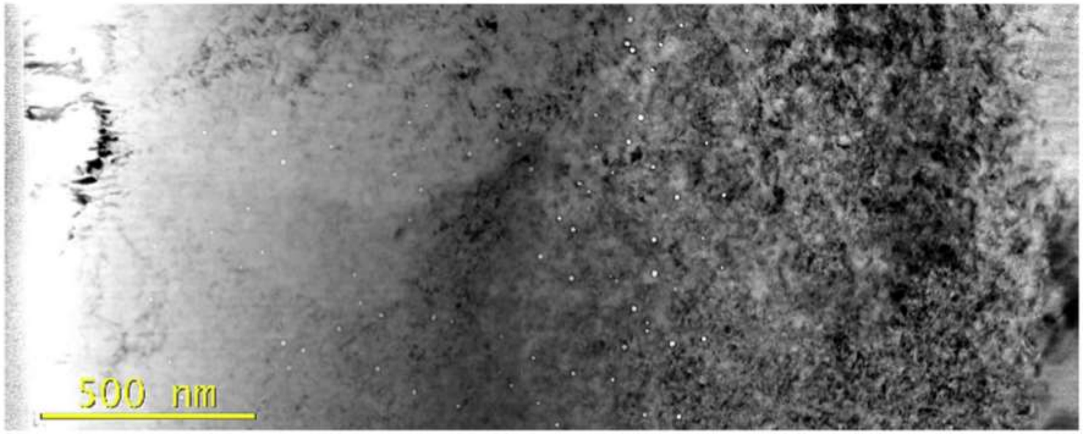
100 dpa

B. Michaut, T. Jourdan, J. Malaplate, A. Renault-Laborne, F. Sefta, B. Decamps, J. Nucl. Mater. **496** (2017) 166-176

Self-ion irradiation of austenitic steel

- SRIM (and similar simulations) show how the damage and implantation fluxes vary with depth
- Any bias between SIA/vac can have important consequences
- Self-ion irradiation implants SIAs
- Spatial resolution (depth) introduced in CD

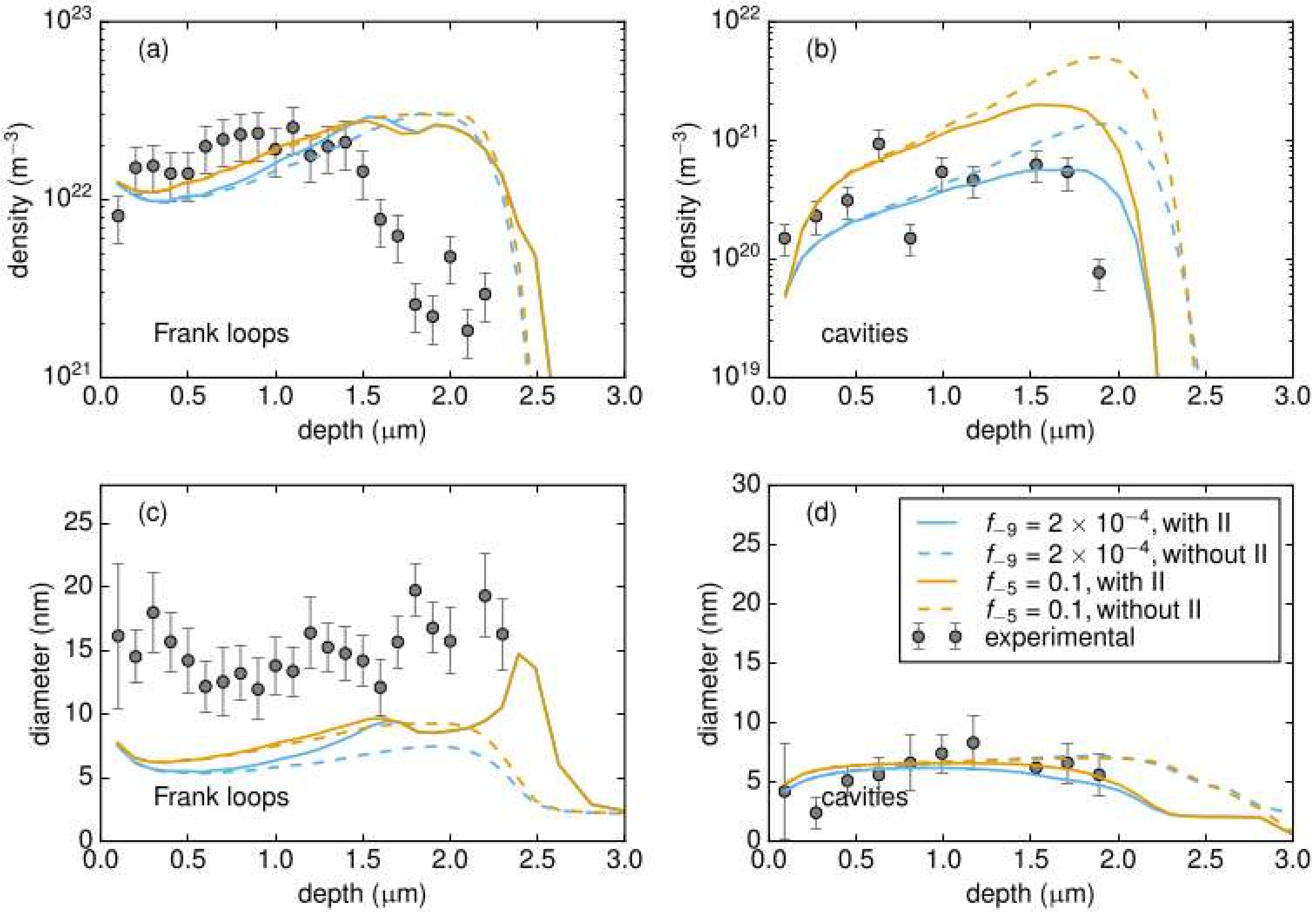




B. Michaut, T. Jourdan, J. Malaplate, A. Renault-Laborne, F. Sefta, B. Decamps, J. Nucl. Mater. **496** (2017) 166-176

Self-ion irradiation of austenitic steel

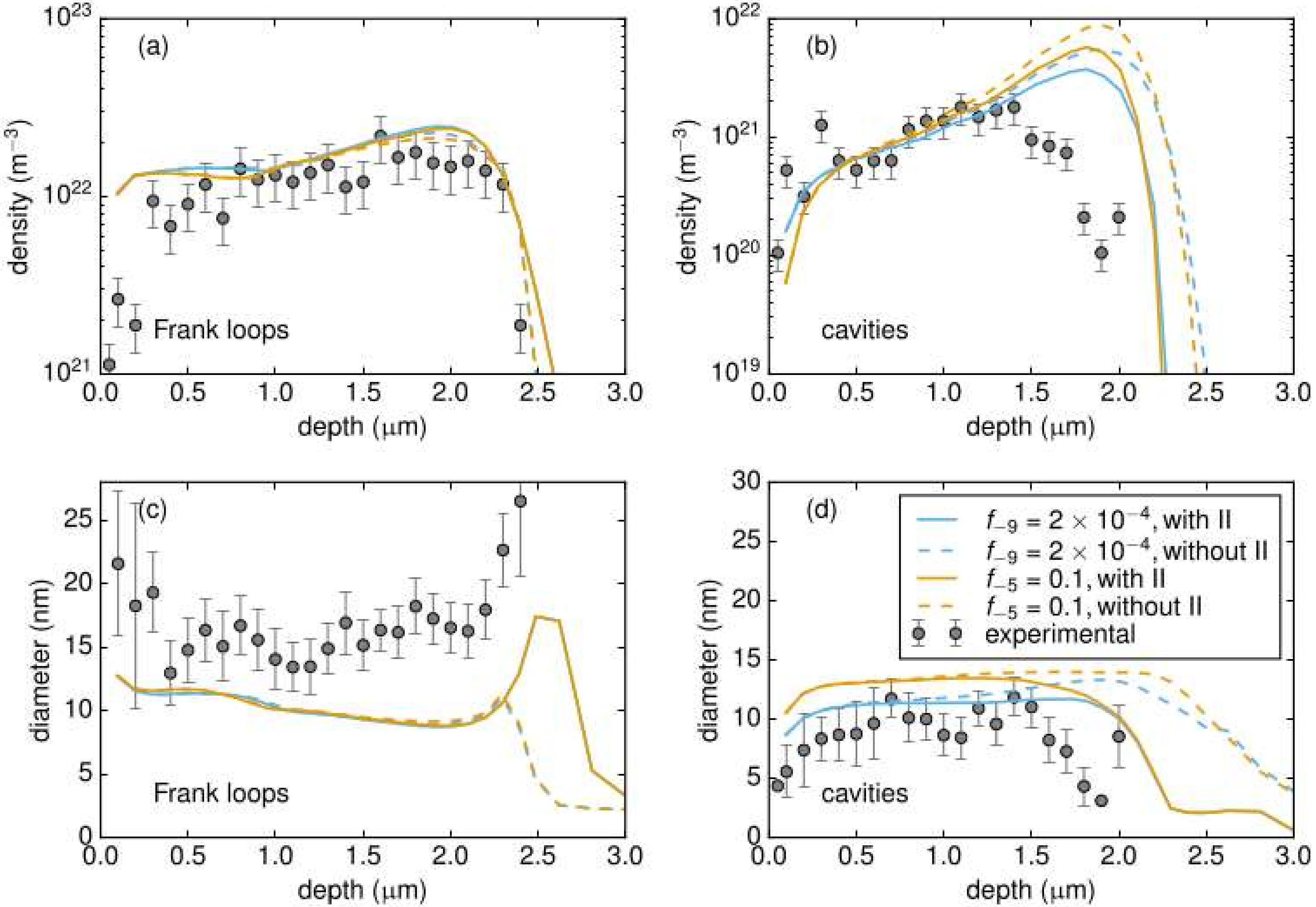
- 5 dpa results:
  - Two source terms (blue, orange)
  - Injected SIAs (II) (solid vs dashed)
  - Small depth effect of injected SIAs



B. Michaut, T. Jourdan, J. Malaplate, A. Renault-Laborne, F. Sefta, B. Decamps, J. Nucl. Mater. **496** (2017) 166-176

Self-ion irradiation of austenitic steel

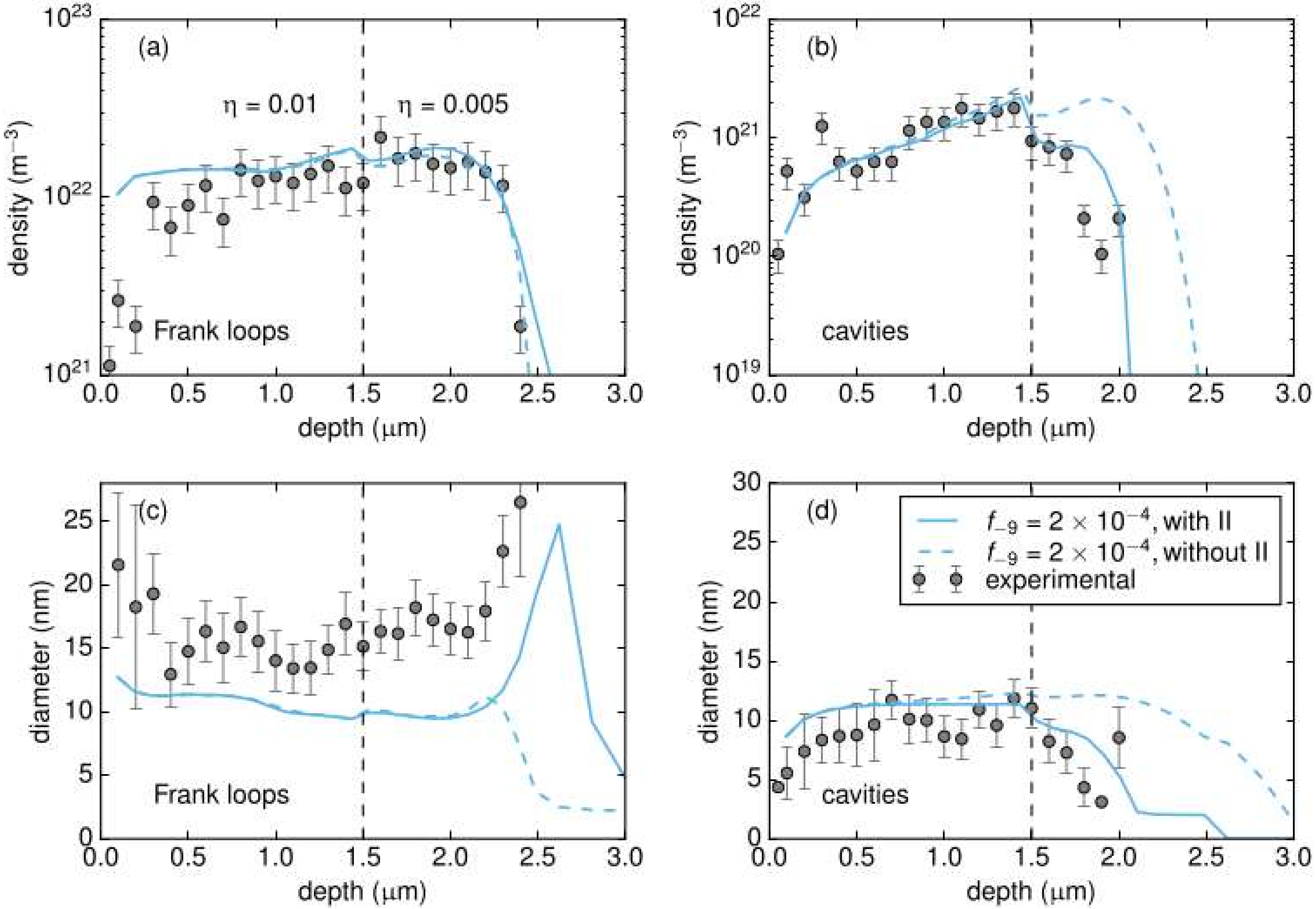
❑ 40 dpa results:
  - Two source terms (blue, orange)
  - Injected SIAs (II) (solid vs dashed)
  - Clear depth effect of injected SIAs for cluster sizes



B. Michaut, T. Jourdan, J. Malaplate, A. Renault-Laborne, F. Sefta, B. Decamps, J. Nucl. Mater. **496** (2017) 166-176

Self-ion irradiation of austenitic steel

- 40 dpa results:
  - Fraction of freely migrating SIAs proposed to vary with depth
  - First model: two zones
  - Rough model, needs refinement, but captures the main effect

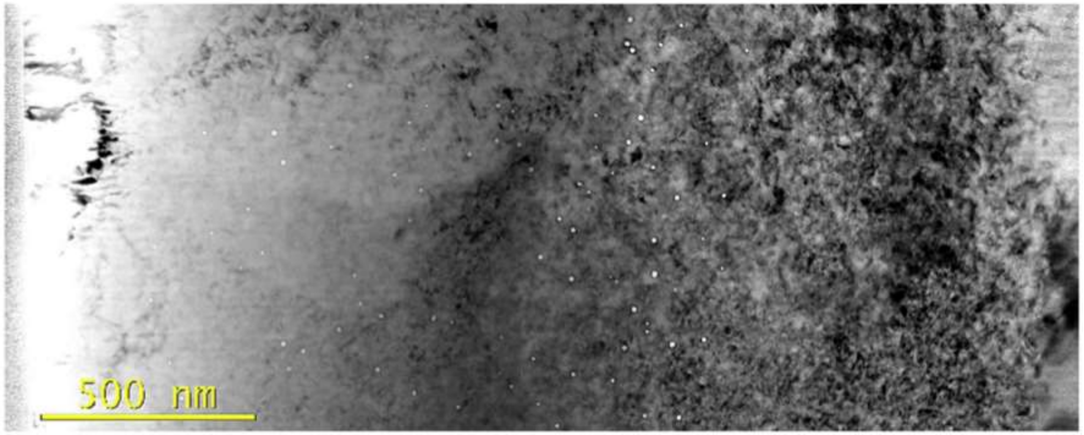


B. Michaut, T. Jourdan, J. Malaplate, A. Renault-Laborne, F. Sefta, B. Decamps, J. Nucl. Mater. **496** (2017) 166-176

Self-ion irradiation of austenitic steel

❑ Conclusions:

- Effect of injected SIAs (normally neglected) is important to consider
- Depth variation of freely migrating SIAs has significant effect
- More refined models needed
- Spatial resolution crucial considering strong depth dependence of damage



General conclusions

- ❑ A huge effort has been expended in the SOTERIA project to model the observed nanofeature evolution seen in RPV and internals under irradaition
- ❑ Advanced models and methods for the physics basis developed
- ❑ Mechanisms proposed from these results
- ❑ Larger-scale models (KMC, RT) have been developed to implement the mechanisms and investigate if they do explain the observations
- ❑ Microstructure modelling of FeCuNiMnSiP close to RPV
- ❑ Many success stories!

The SOTERIA Consortium

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ET DE SÛRETÉ NUCLÉAIRE

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