Modelling of radiation damage in tungsten including He production

C.S. Becquart[1], C. Domain[2]

A. De Backer[1]

M.F. Barthe[3]

M. Hou[4], C. Ortiz[5]

[1] Unité Matériaux Et Techniques, UMET, UMR 8207, Villeneuve d'Ascq, France

[2] EDF R&D, MMC Centre des Renardières, Moret sur Loing, France

[3] CEMHTI, UPR 3079 , Orléans, France

[4] Physique des Solides Irradiés et des Nanostructures CP234, Université Libre de Bruxelles, Bd du Triomphe, B-1050 Brussels, Belgium

[5] Laboratorio Nacional de Fusión por Confinamiento Magnético, CIEMAT, E-28040 Madrid, Spain

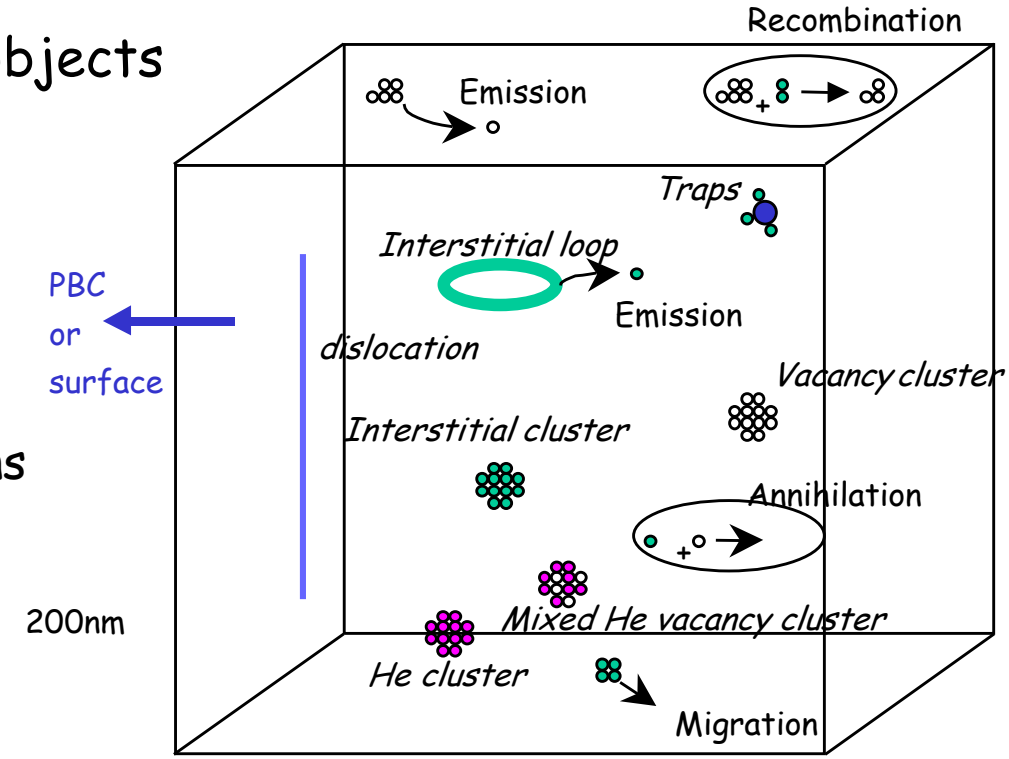
EUROPEAN FUSION TECHNOLOGY PROGRAMME: CEA Contrat V 3542.001

PROGRAMME INTERDISCIPLINAIRE ENERGIE DU CNRS 2006-2009

Object Kinetic Monte Carlo: LAKIMOCA

Microstructure = objects defined by:

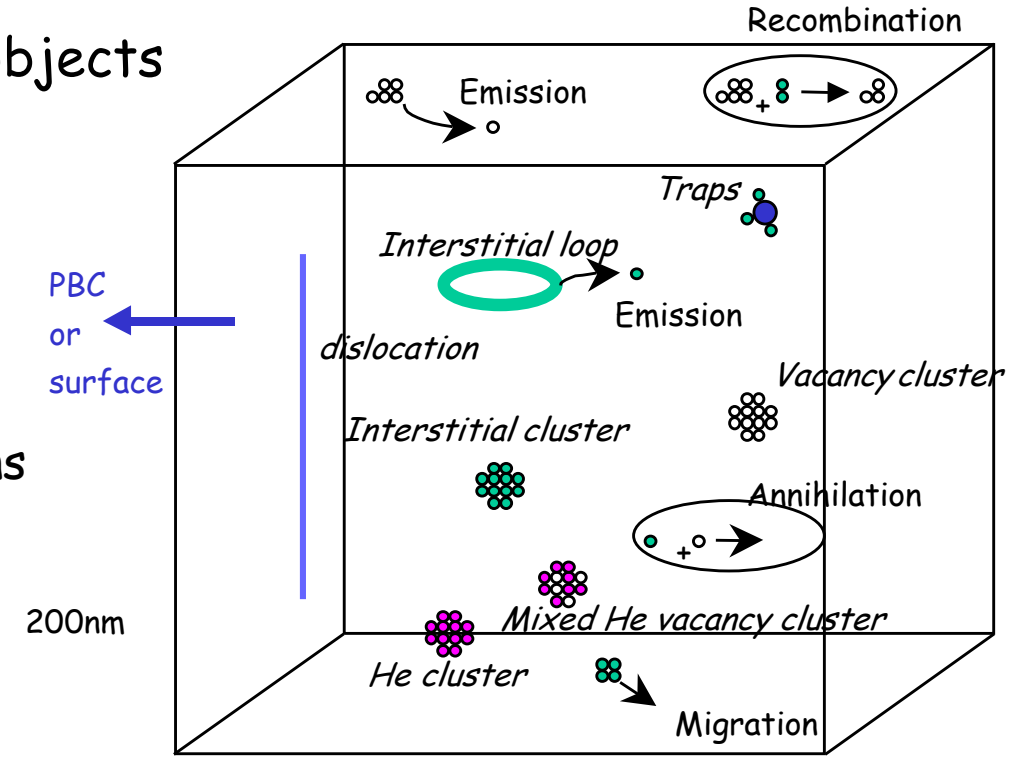
- type
- centre-of-mass position
- reaction radius
- possible reactions



Object Kinetic Monte Carlo: LAKIMOCA

Microstructure = objects defined by:

- type
- centre-of-mass position
- reaction radius
- possible reactions



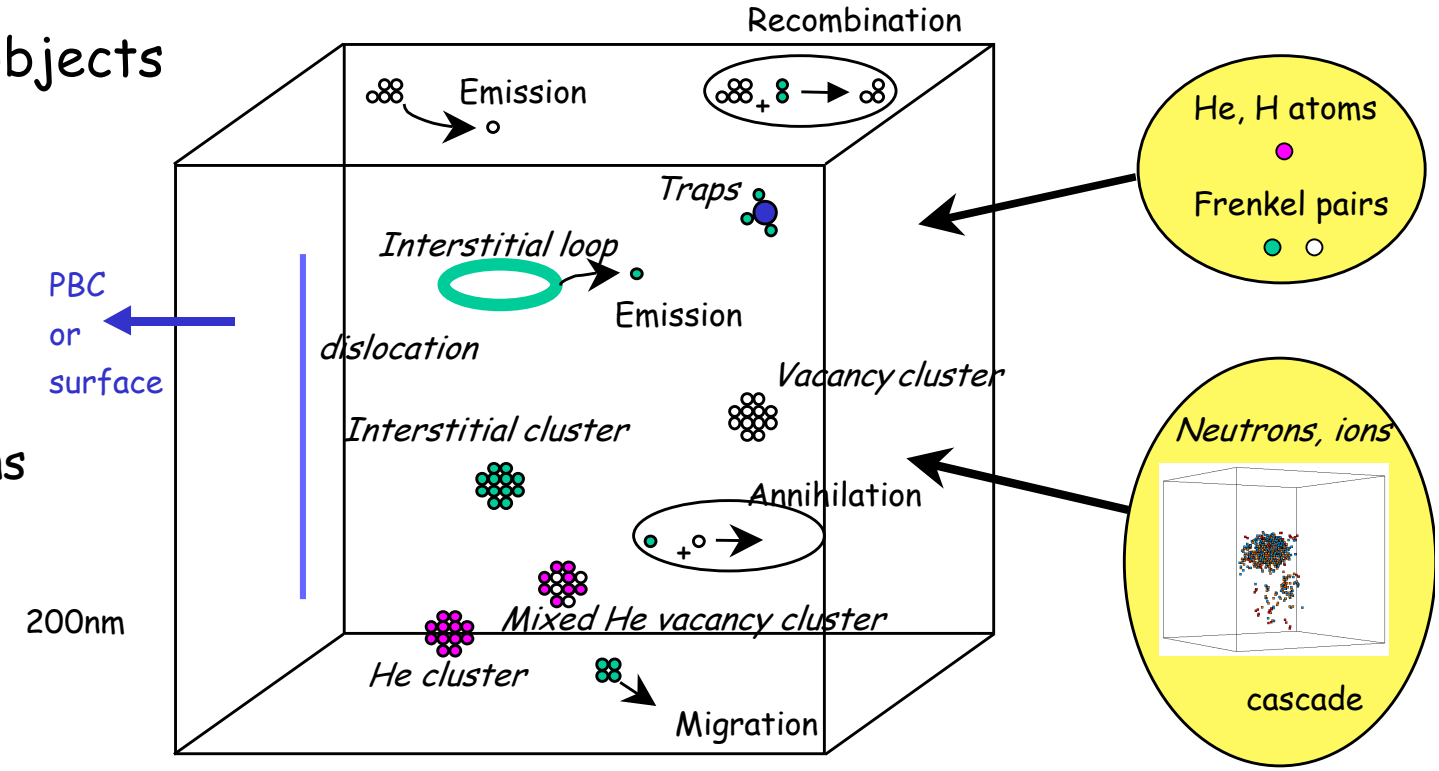
Properties of He (and H), interactions with vacancy and vacancy clusters, with SIAs and impurities (binding and migration energies): ab initio calculations.

===> Use He desorption experiments to check.

Object Kinetic Monte Carlo: LAKIMOCA

Microstructure = objects defined by:

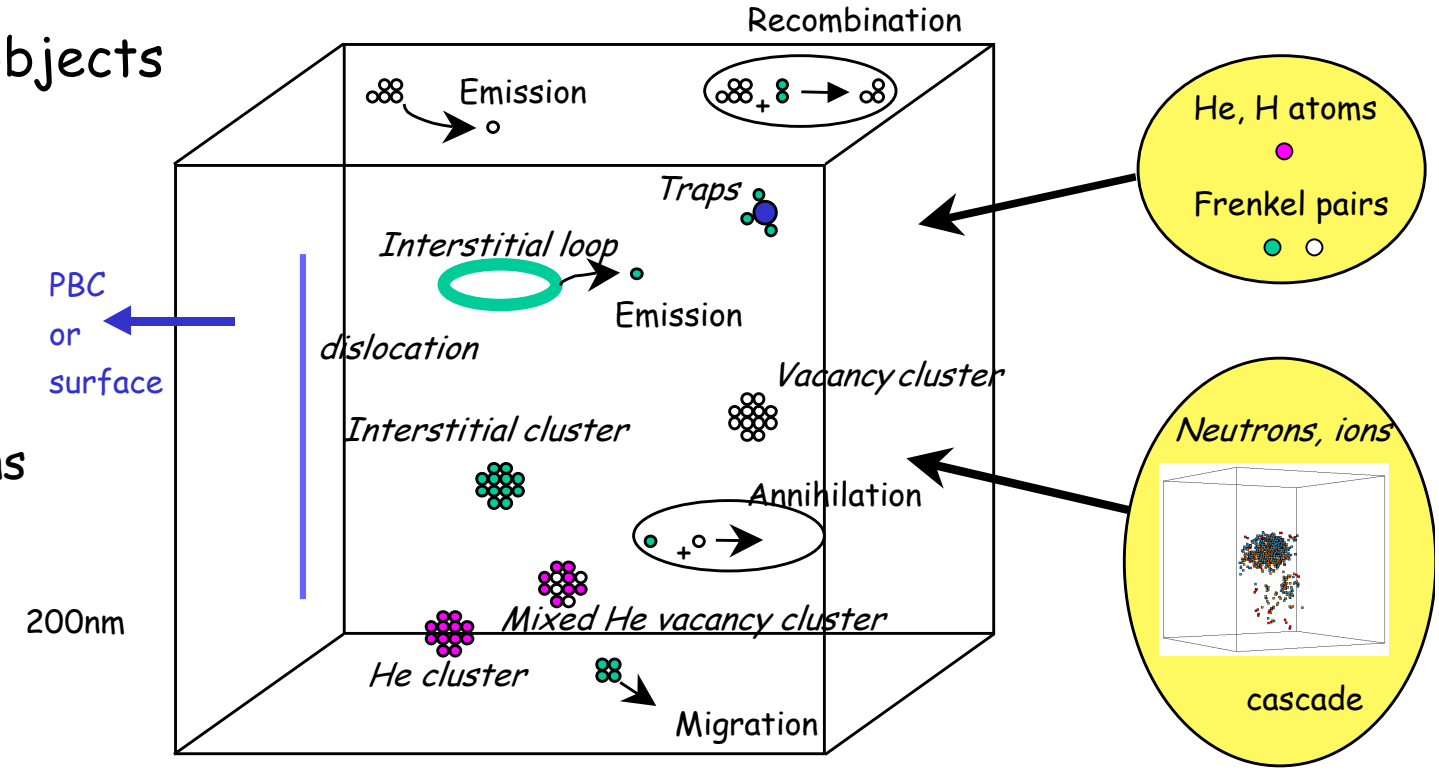
- type
- centre-of-mass position
- reaction radius
- possible reactions



Object Kinetic Monte Carlo: LAKIMOCA

Microstructure = objects defined by:

- type
- centre-of-mass position
- reaction radius
- possible reactions



External events: He implantation, damage…

==> BCA code Marlowe adjusted on exp. results

ab initio method

- DFT
- PAW
- VASP (Vienna Ab initio Simulation Package)
- Exchange and correlation: *GGA* (PW91)
- All atomic positions relaxed
- Constant volume calculations
- Supercells with PBC.
- Cut off W: 350 eV.
- 54 atoms + 5x5x5 kpoints, 128 atoms + 3x3x3 kpoints, 250 atoms + 3x3x3 kpoints

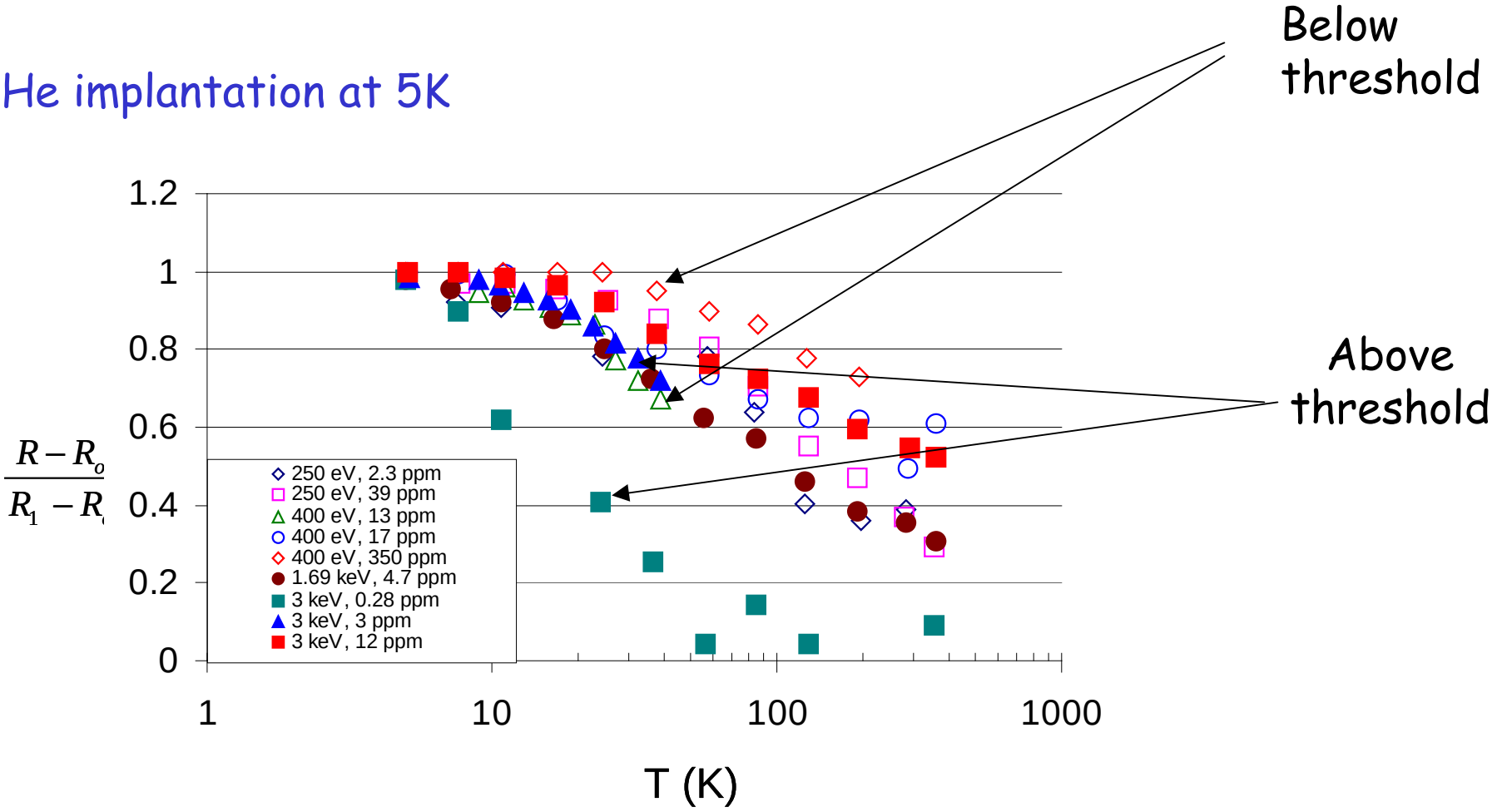
Formation, binding and migration energies

Ab initio BO MD: NVT ensemble, time step 1 fs, cutoff 350 eV and 1 kpt

G. Kresse and J. Hafner, Phys. Rev. B **47**, 558 (1993); ibid. 49, 14 251 (1994)
G. Kresse and J. Furthmüller, Comput. Mat. Sci. **6**, 15 (1996)
G. Kresse and J. Furthmüller, Phys. Rev. B **55**, 11 169 (1996)

He desorption experiments

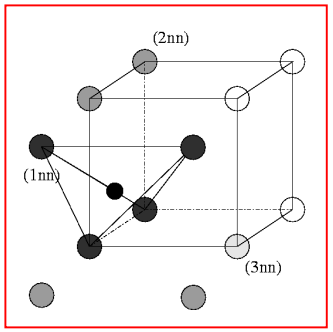
He implantation at 5K



A.S. Soltan, R. Vassen and P. Jung, J. Appl. Phys. **70**(2) (1991) 793

He

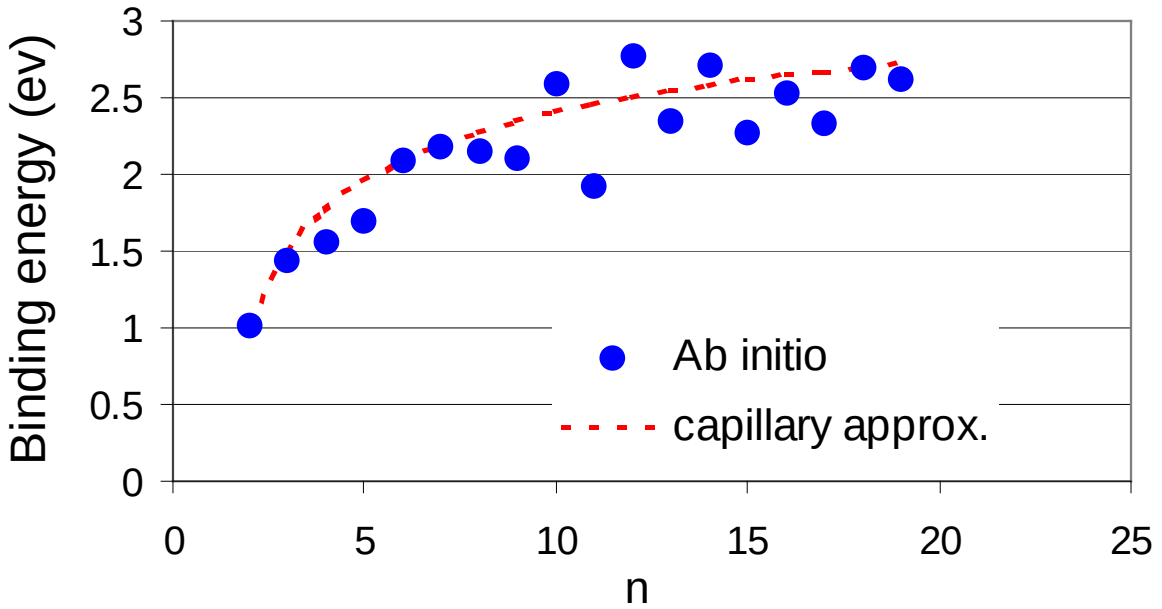
	Ab initio	Exp. (eV)	Empirical potentials (eV)
Octahedral	6.38		7.83 [HENR04] **5.47** [WILS72]
Tetrahedral	**6.16**		5.71 [WILS72]
Substitutional	4.70		
Migration energy E_{He}^{mig}	0.06	0.28 (^{3}He [AMA84]) 0.24-0.32 (^{4}He [WAG79])	0.29 [HENR04] 0.24 [WILS72]



E_{mig}(He) very low: discrepancy with exp. results. Consistent with tendency to form clusters [BEC06], agreement with Soltan [SOLT91]

J. Amano and D. Seidman, J. Appl. Phys. **56** (1984) 983
A. Wagner and D.N.Seidman, Phys. Rev. Lett. **42** (1979) 515
C.S. Becquart & C. Domain, Phys. Rev. Lett. **97** (2006) 196402.
A.S. Soltan, R. Vassen and P. Jung, J. Appl. Phys. **70**(2) (1991) 793

He clusters



(total number of He in the resulting cluster)

$$E^b(n \to (n-1)+1) = E^{for}(n-1) + E^{for}(1) - E^{for\cdot}(n)$$

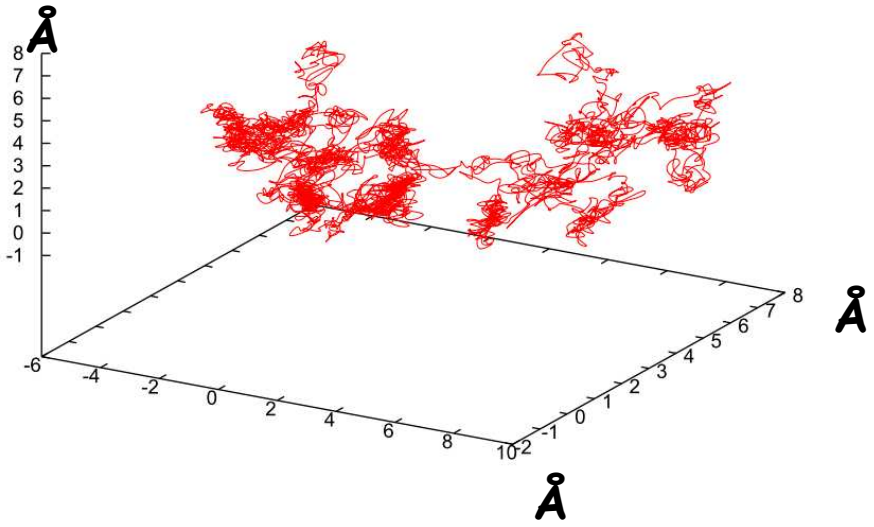
$$= \left[\left(E^b(2) - E^{for}(1)\right)\left((n)^{2/3} - (n-1)^{2/3}\right)\right] / \left(2^{2/3} - 1\right) + E^{for}(1)$$

He clusters

Small clusters are mobile

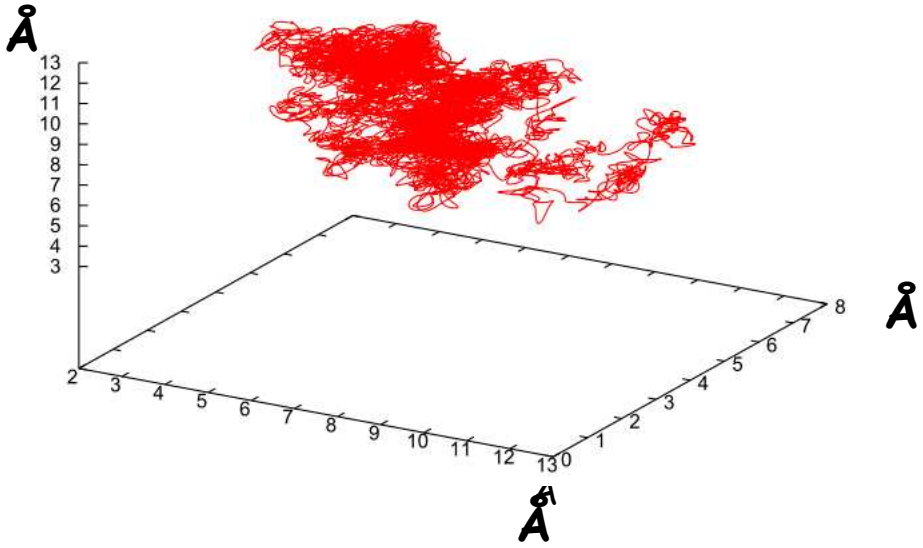
Ab initio MD at 1000 K, center of mass (CM) trajectories

44 ps



5He

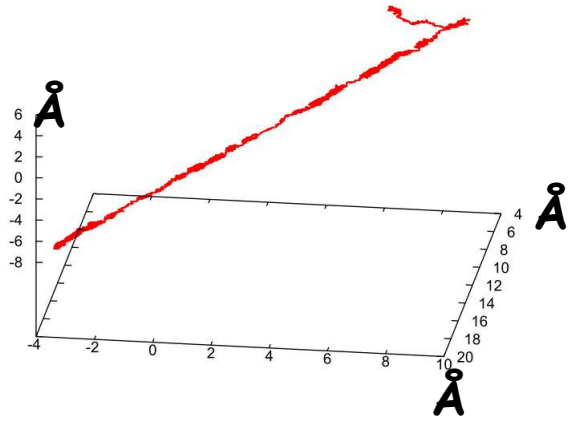
82 ps



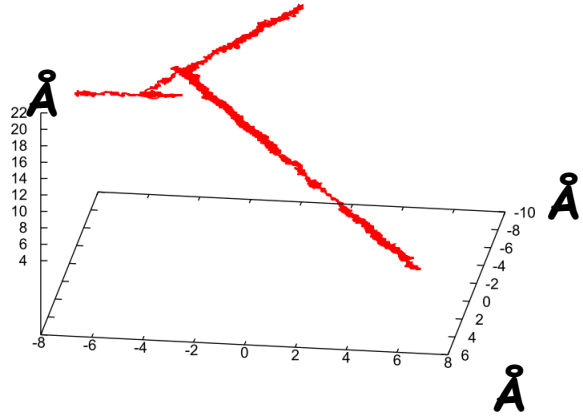
10He

2He

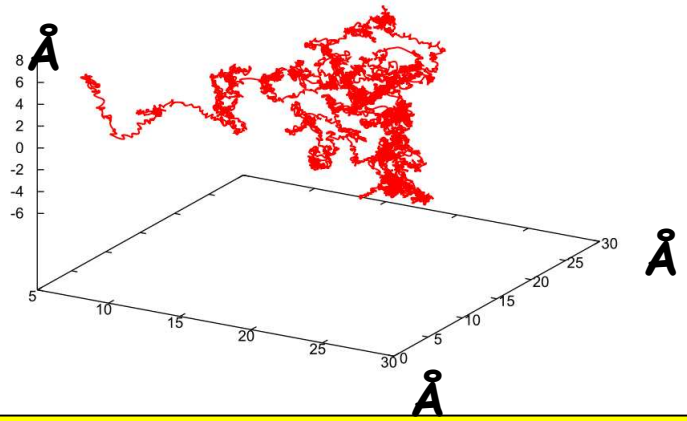
150 K : 103 ps



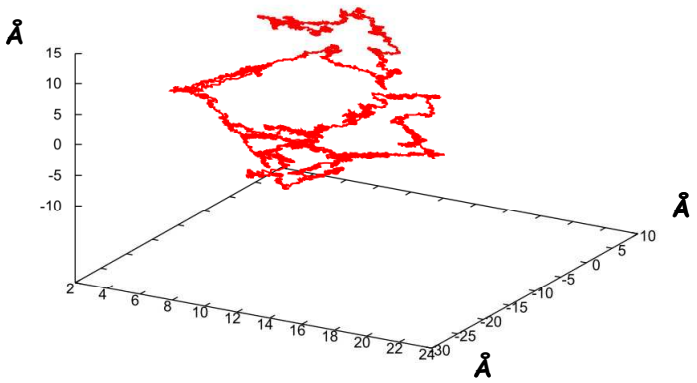
300 K : 105 ps

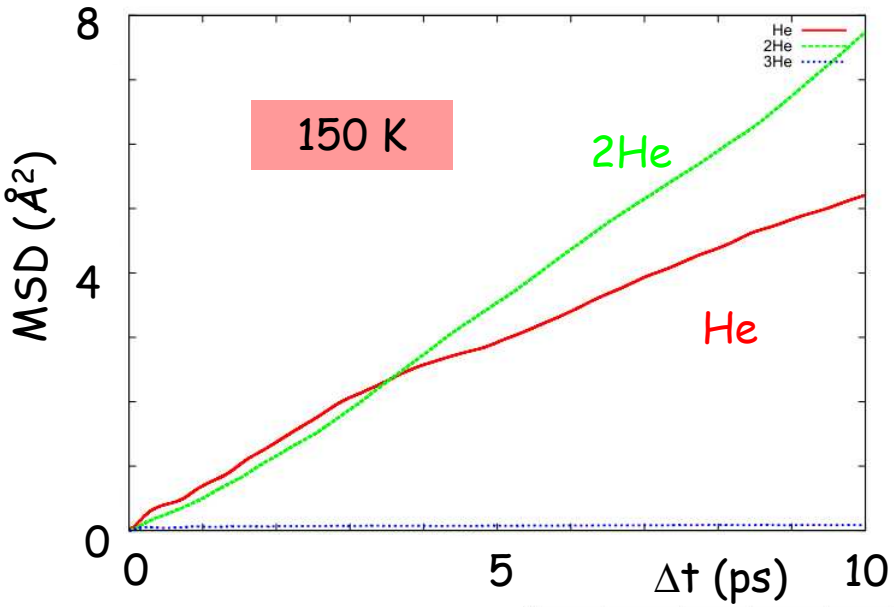


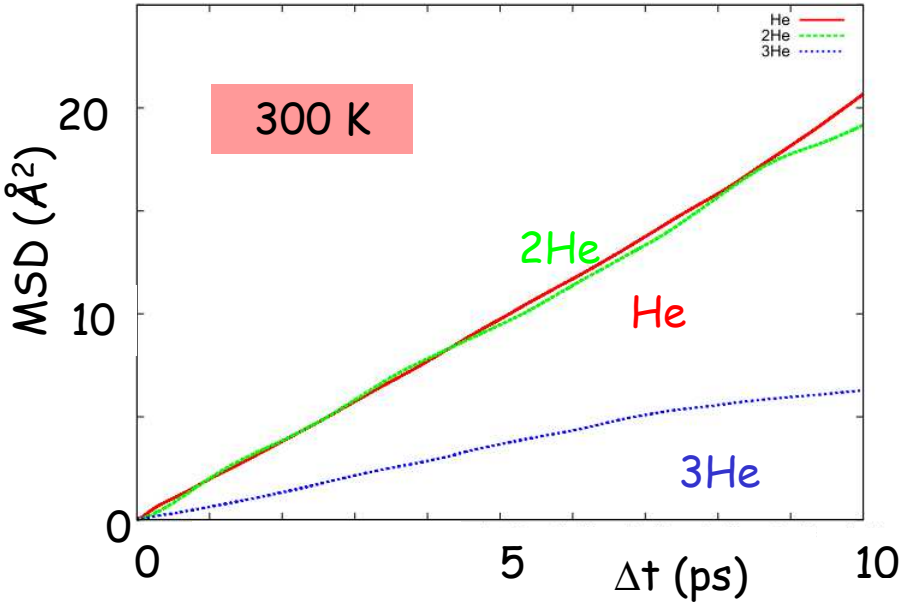
1000 K : 103 ps

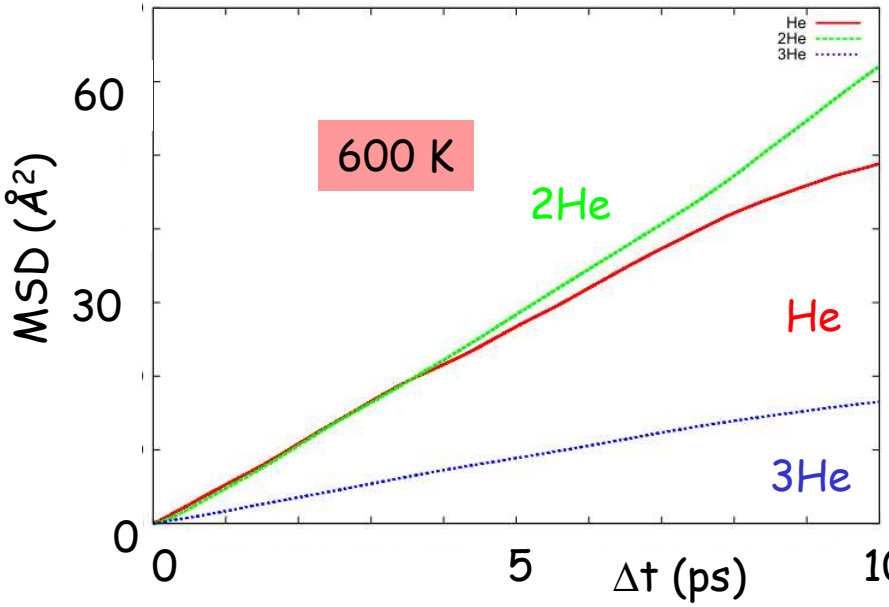


600 K : 107 ps









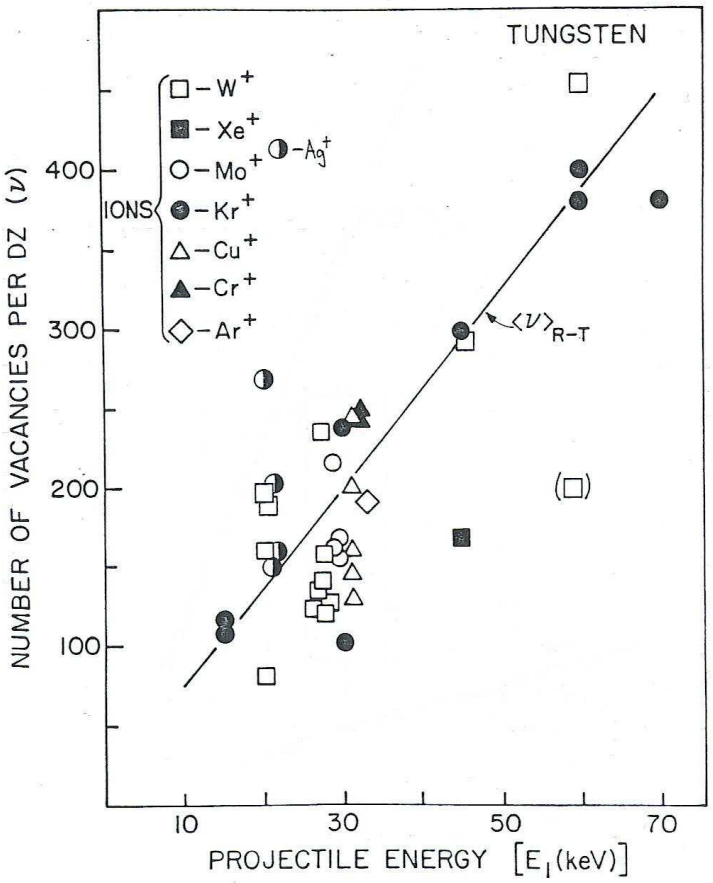
Center of Mass MSD

Emig He ~ 0.04 eV

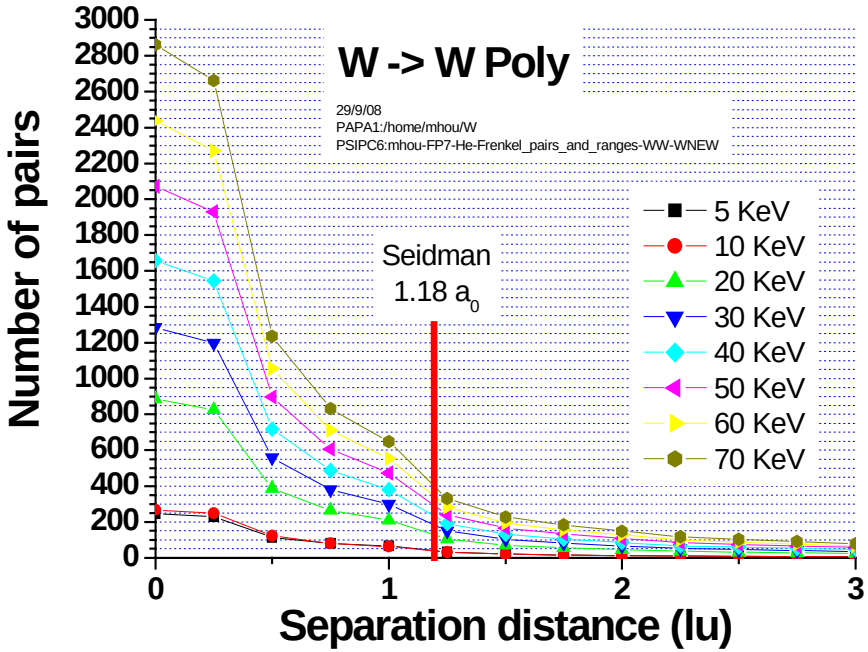
Emig 2He (1D) ~ 0.027 eV

Emig 3He ~ 0.07 eV

External events: MARLOWE parameterized on exp. results



D. Pramanik and D. N. Seidman, Jour. Appl. Phys. **54** (1983) 6352 ; D. N. Seidman, R. S. Averback, and R. Benedek, Phys. Stat. Solidi (b) **144** (1987) 85.



M. Hou, C. J. Ortiz, C.S. Becquart, C. Domain, U. Sarkar, A. Debacker, J. Nucl. Mater. 403 (2010) 89.

Check parameterization: OKMC simulations of He desorption

Two steps:

-implantation in 399×400×1001 l.u. box

Exp. rate = $10^{15} s^{-1} m^{-2}$ ==> 16 He per s

PBC in two directions: thin foil 317.3 nm thick.

Surface perpendicular to ⟨001⟩ direction

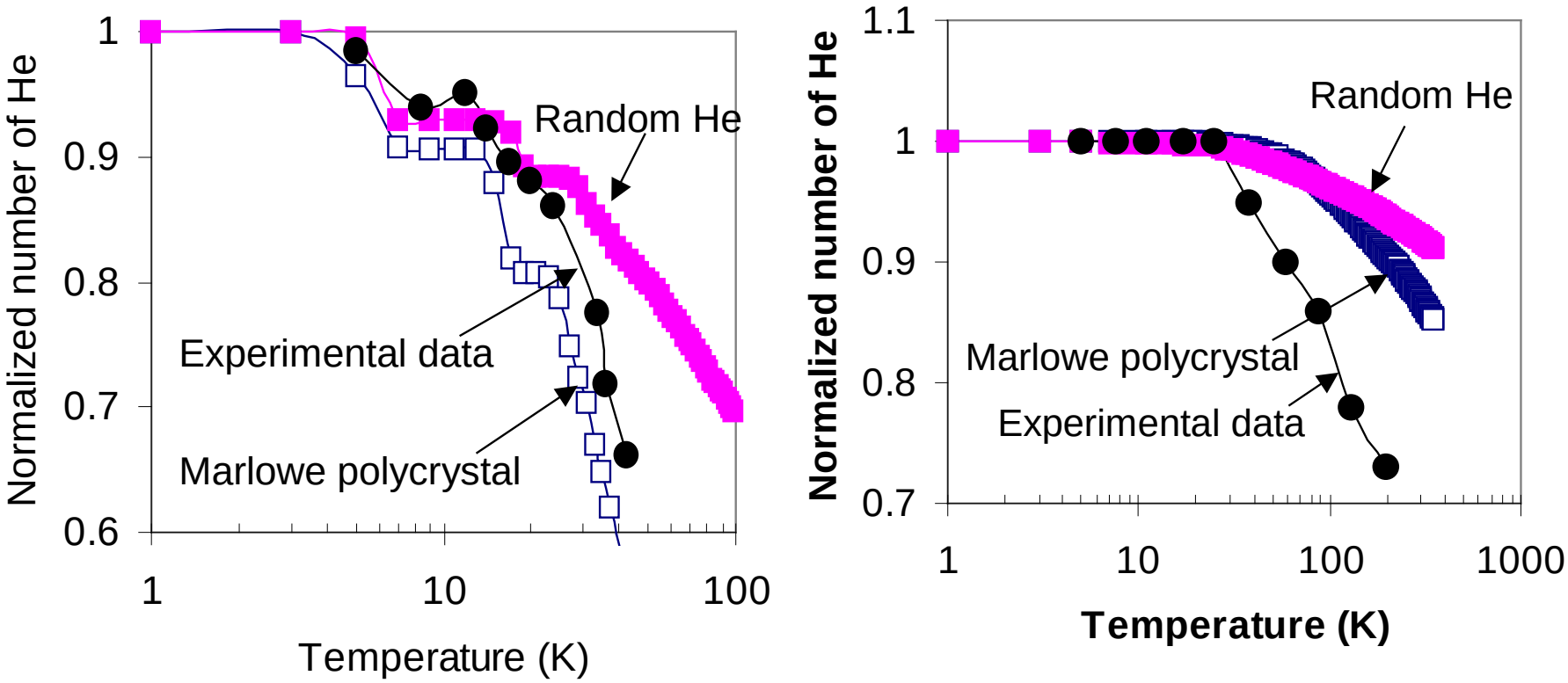
-isochronal annealing

T increase of 2K every 60 s

Monitor the total number of defects (not objects) in the box

C.S. Becquart, C. Domain, U. Sarkar, A. DeBacker, M. Hou, accepted in J. Nucl. Mater. 403 (2010) 75.

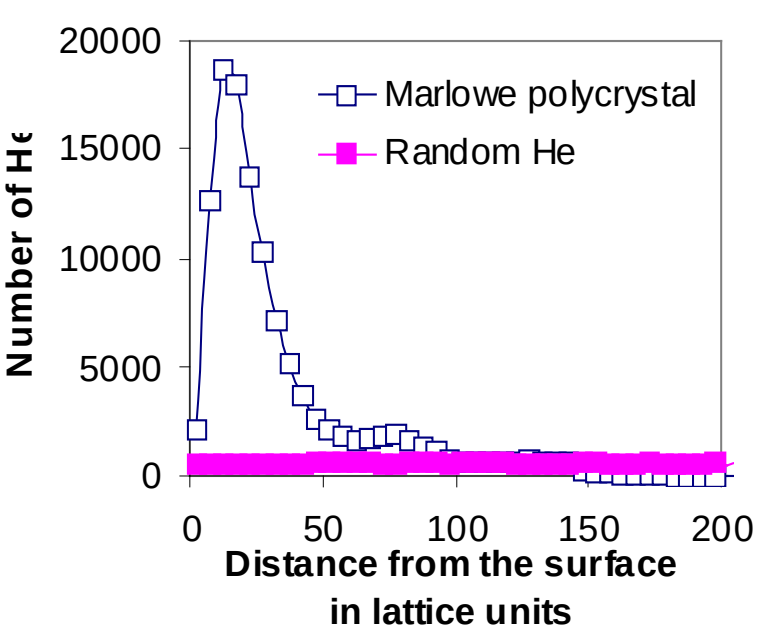
OKMC simulations of He desorption implanted «under threshold»

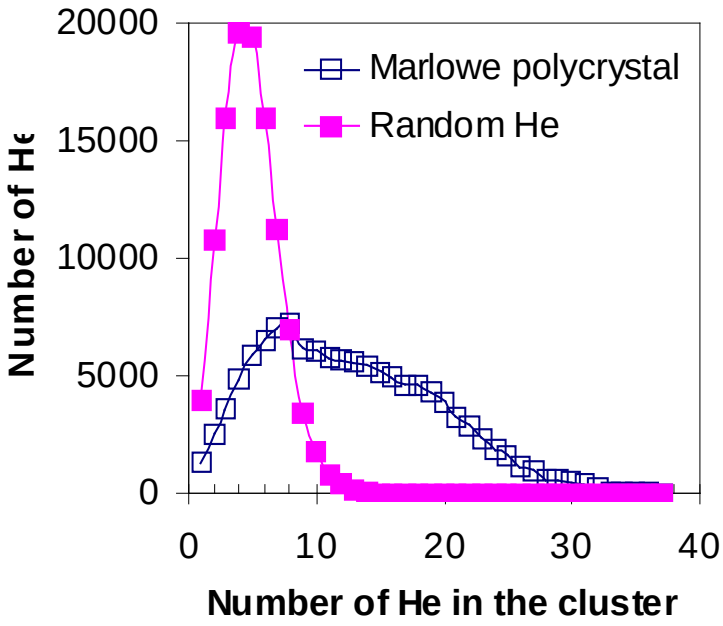


Exp: 400 eV He, 13 ppm He and 350 ppm He

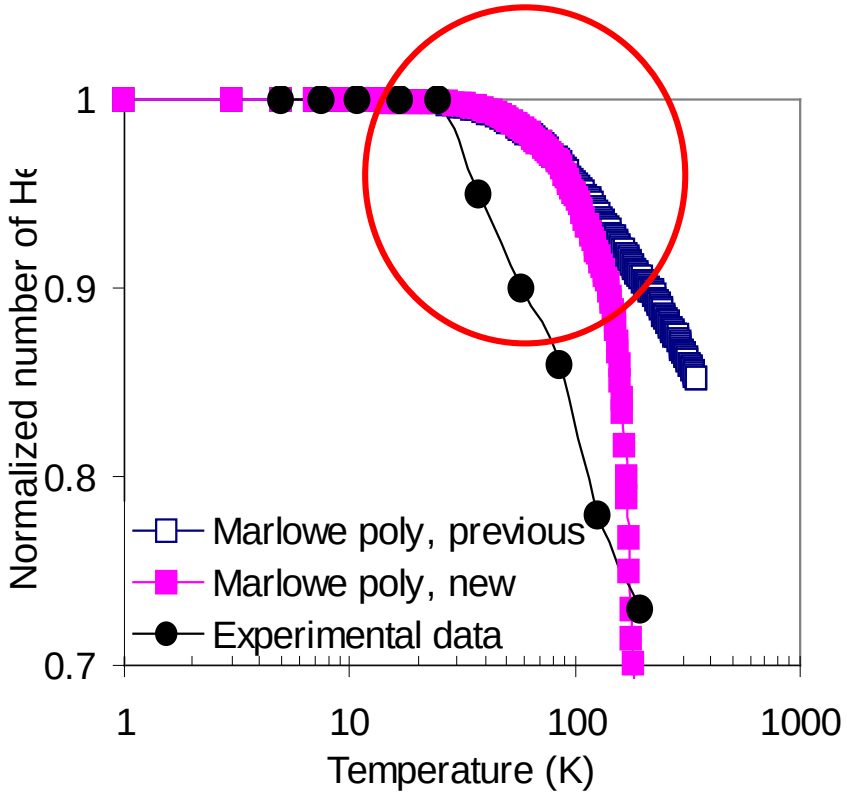
- Relevance of He distribution

Mean spatial distribution of He at the end of implantation





Exp: 350 ppm 400 eV He

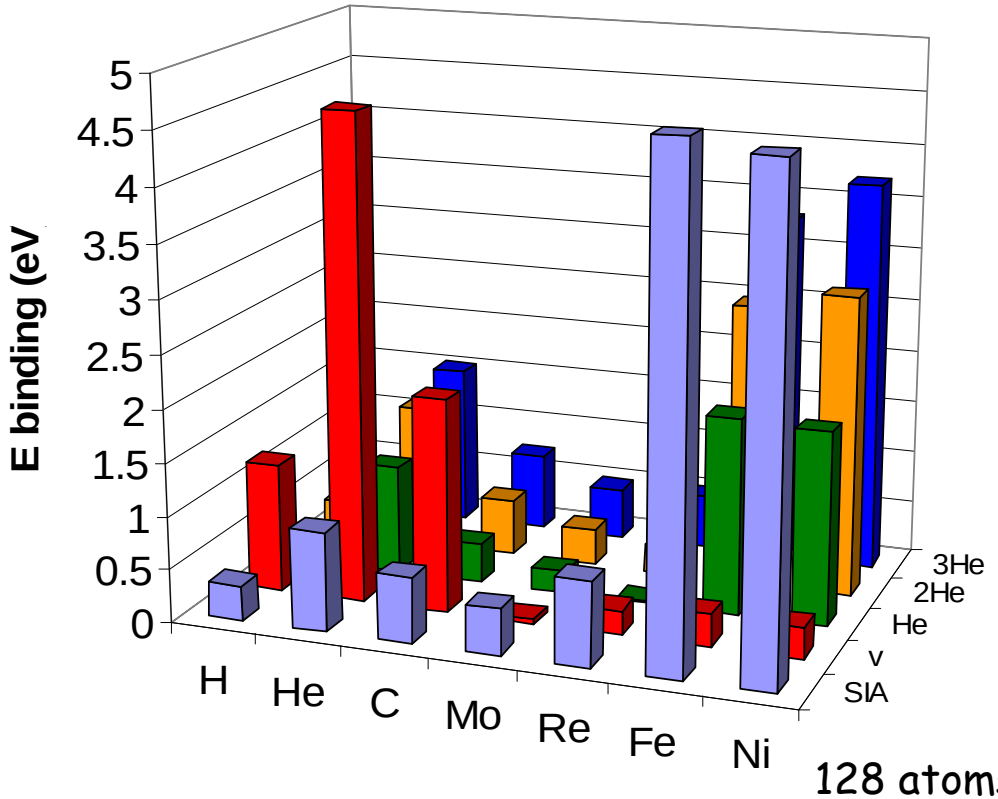


Migration of small He clusters…

Role of impurities…

Exp: 350 ppm 400 eV He

Role of impurities (ab initio)

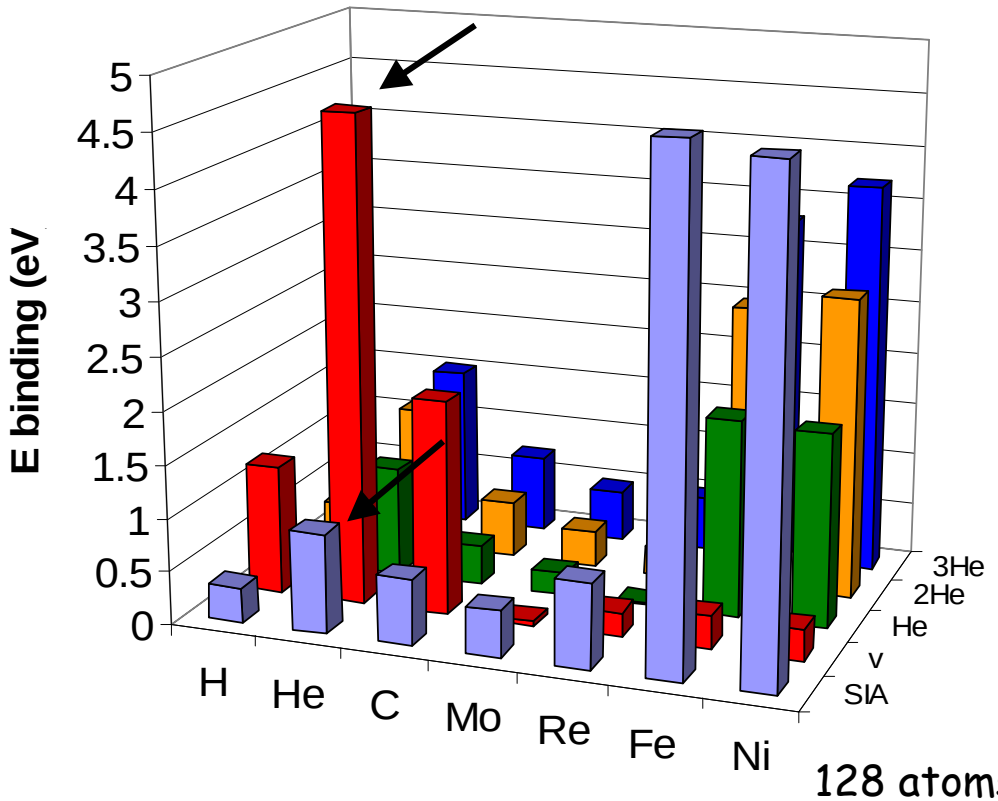


128 atoms 3x3x3 kpoints

Most impurities trap He and small He clusters

===> potential bubble nucleation centers

Role of impurities (ab initio)



128 atoms 3x3x3 kpoints

Most impurities trap He and small He clusters

===> potential bubble nucleation centers

Vacancy and voids

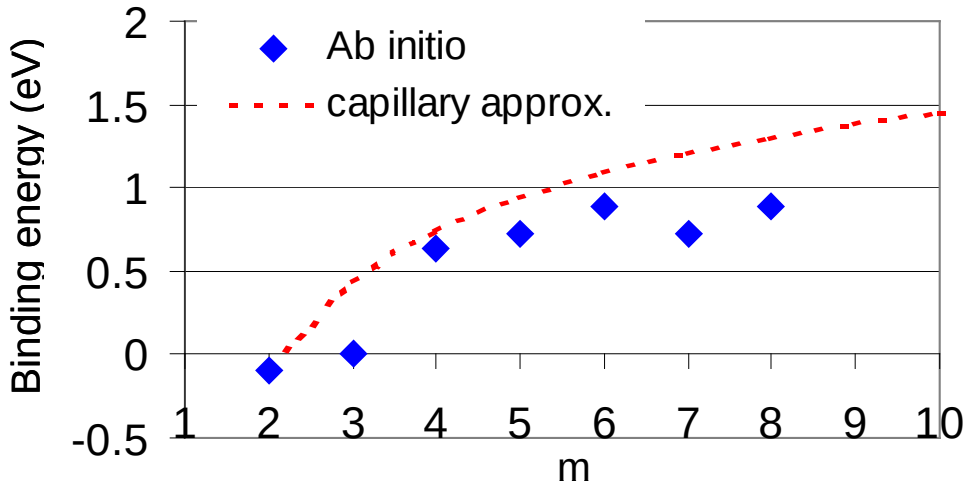
VASP: divacancy unstable; not predicted by empirical potentials; agreement (now) between DFT calculations

Exp: vacancy loops [BUSW70; HAUS72; RAU70] (TEM) and voids (FIM) [SEID78] formed under heavy particle irradiation.

BUT

-2v stabilised by C and O [VASP] or H [Kato, ICFRM-15]

-Cascade condition: all vacancies in narrow volume

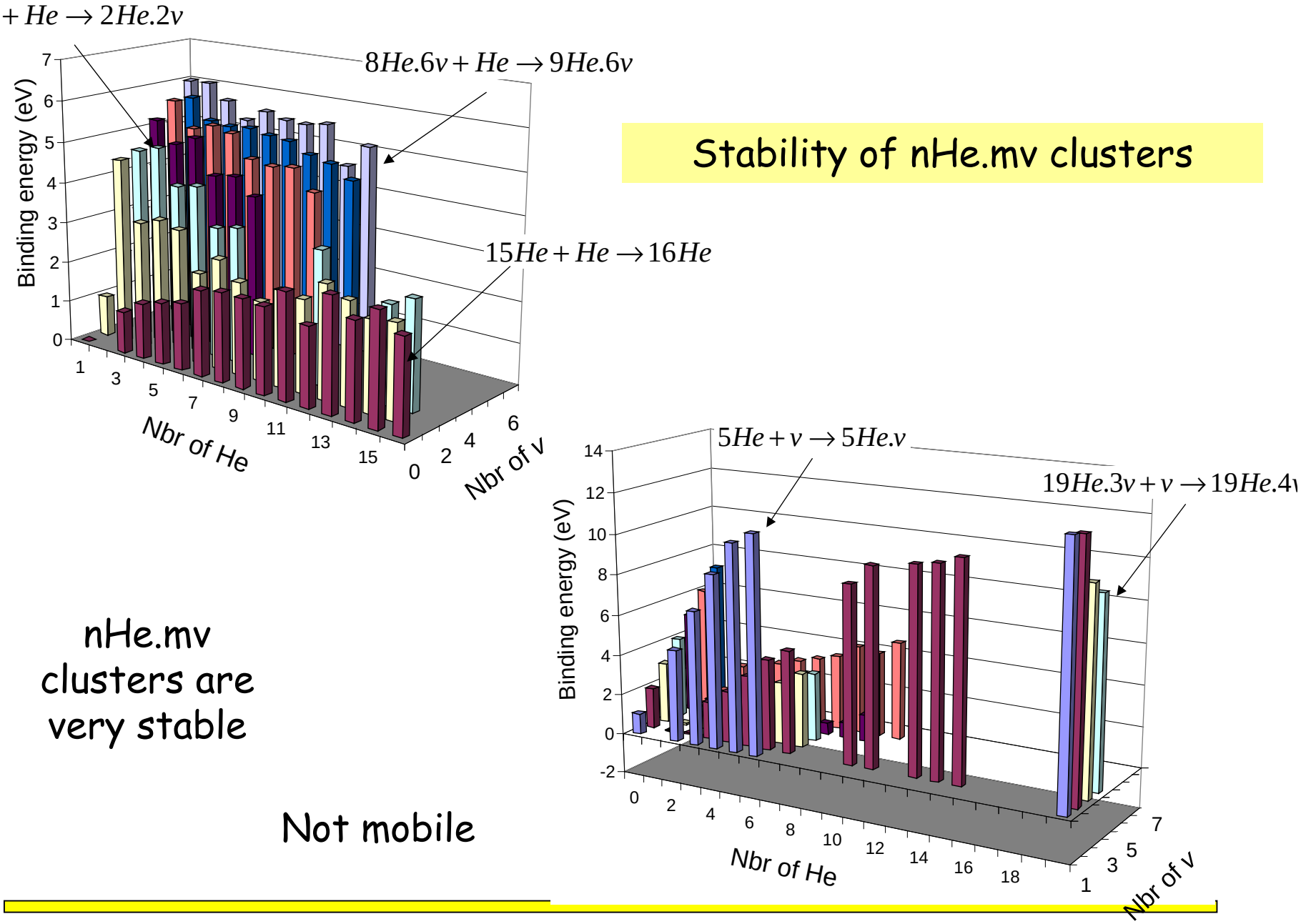


(total number of vacancies in the resulting cluster)

J.T. Buswell, Phil. Mag. **22** (1970) 391 and 787; F. Häussermann, Phil. Mag. **25** (1972) 561 and 583 ; R.C. Rau, Phil. Mag. **18** (1970) 1079.

D. N. Seidman, Surf. Science **70** (1978) 532.

Stability of nHe.mv clusters

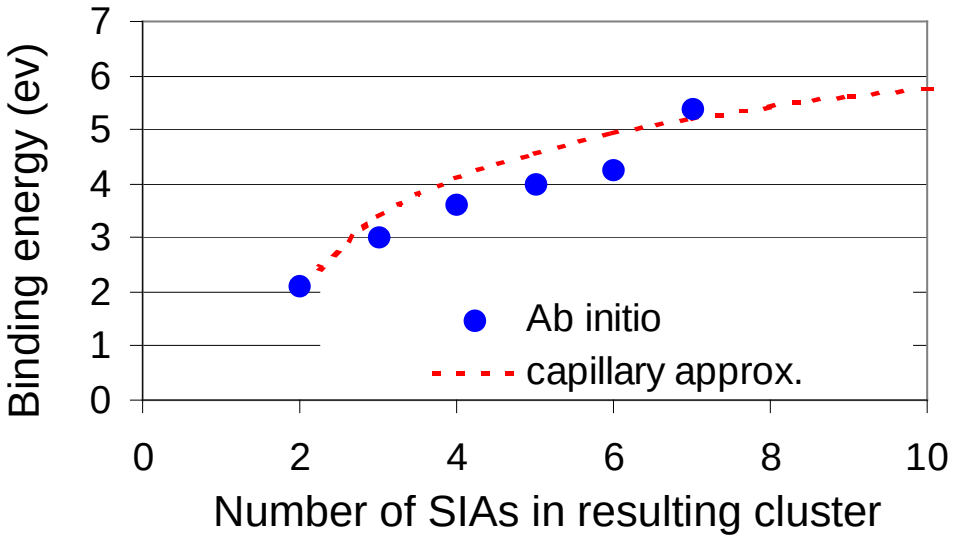


nHe.mv clusters are very stable

Not mobile

SIA and SIA clusters

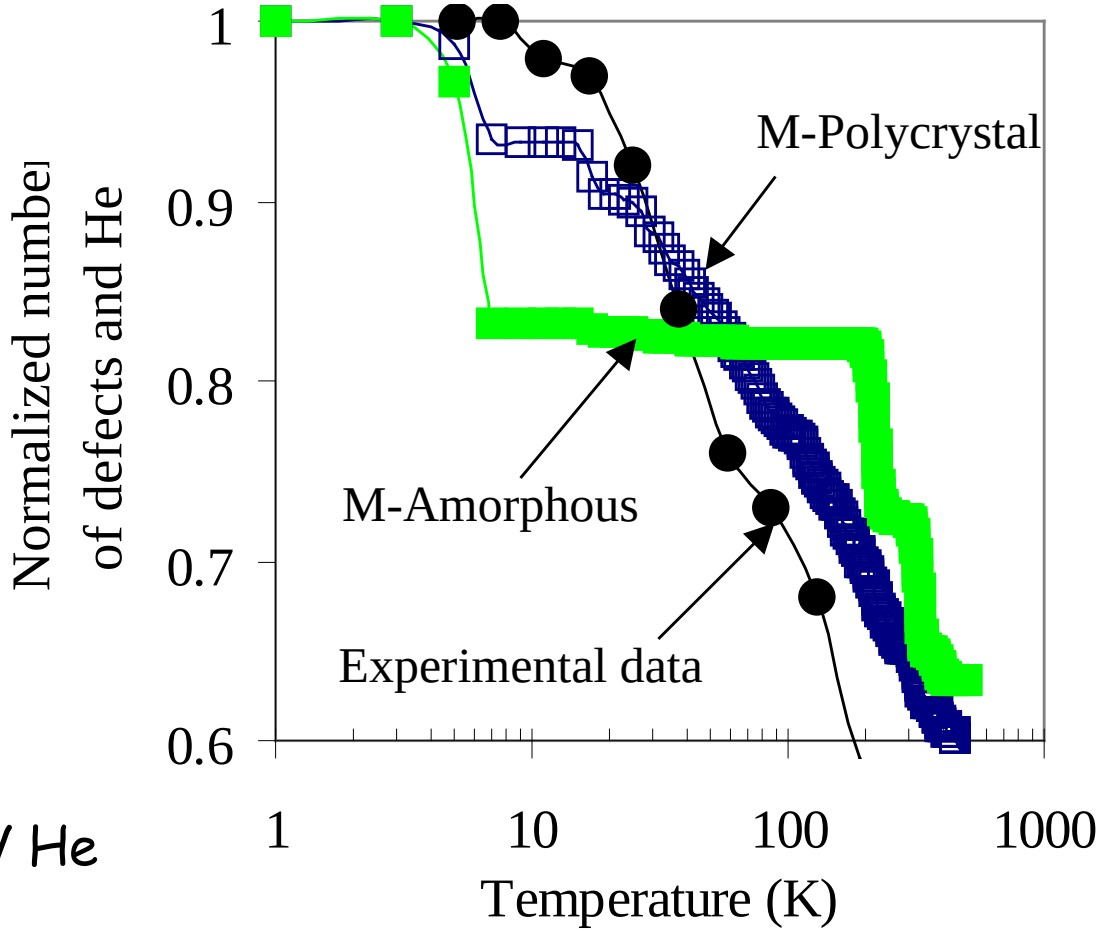
SIA very mobile: 0.013 eV ; 1D motion with Erot = 0.38 eV [DERL07]



SIA clusters very stable and also very mobile [DUDA08]

P.M. Derlet, D. Nguyen-Manh and S.L. Dudarev, Phys. Rev. B **76** (2007) 054107-1

S. L. Dudarev, C.R. Physique **9** (2008) 409.



Experiment better reproduced when W is considered as a polycrystal

12 ppm 3 keV He

- Relevance of the choice of matrix structure on He and primary damage distribution and on longer term evolution.

Influence of He atoms in the void formation of 800 keV ^{3}He implanted in tungsten

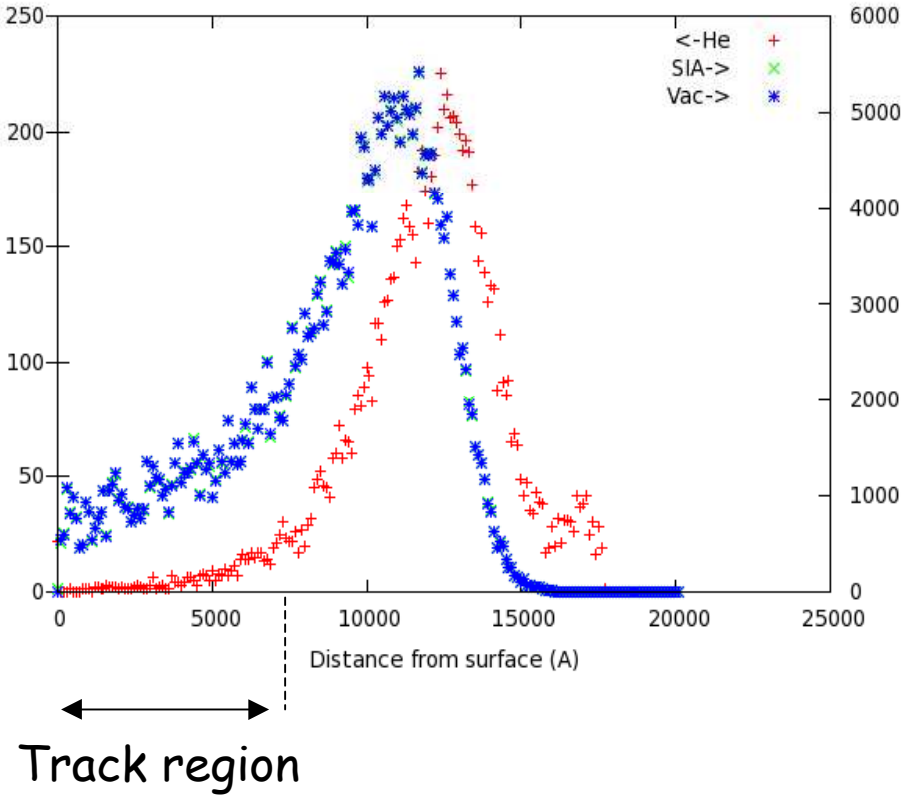
Exp. results of P.E. Lhuilier PhD thesis CEMHTI Orléans: the higher the fluence, the smaller the voids **in the track region**

1/ Implantation stage done at 300 K with experimental flux and fluences (10^{14}, 10^{15}, 10^{16} and 5×10^{16} ions per cm^2).

2/ Isochronal annealing stage : ΔT = 10 K every 3600 s up to 900 K.

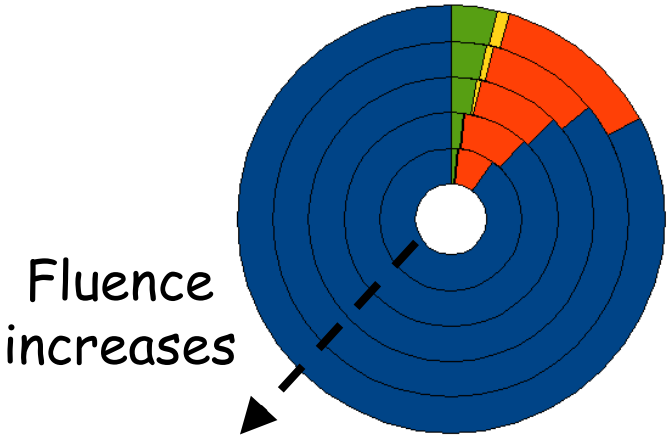
.

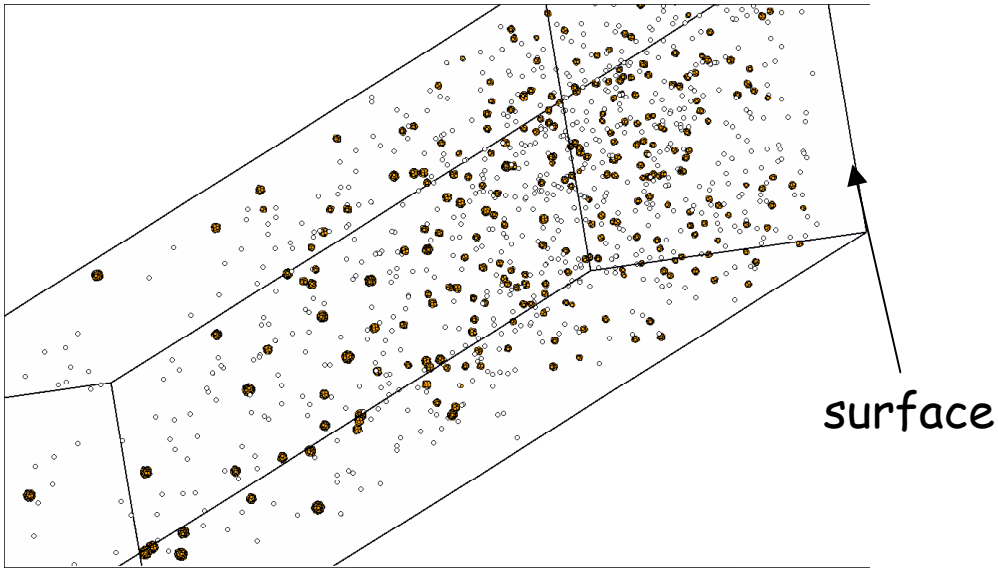
a 800 keV ^{3}He atom produces on average 33 FP (Marlowe)



Box size: 195×203×2208 a_0 = 64×64×700 nm; track region: 3.75 % of the He atoms and 25.4 % of the point defects: 221 FP per He atom

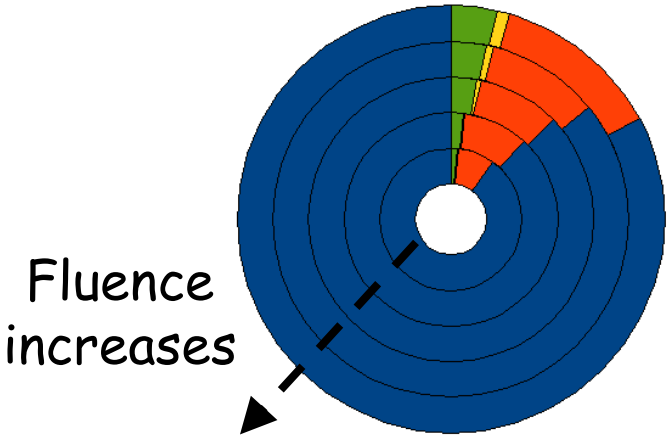
RT implantation ==> SIA and nHe clusters are mobile: annihilation, desorption ...

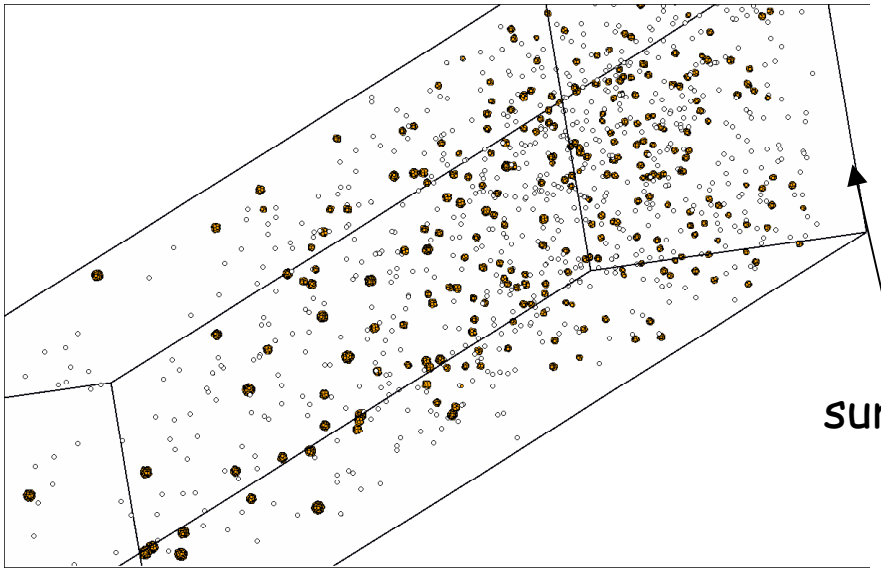




Defect population after implantation stage: v, mv, nHe.pSIA, He.v, nHe.v

RT implantation ==> SIA and nHe clusters are mobile: annihilation, desorption ...



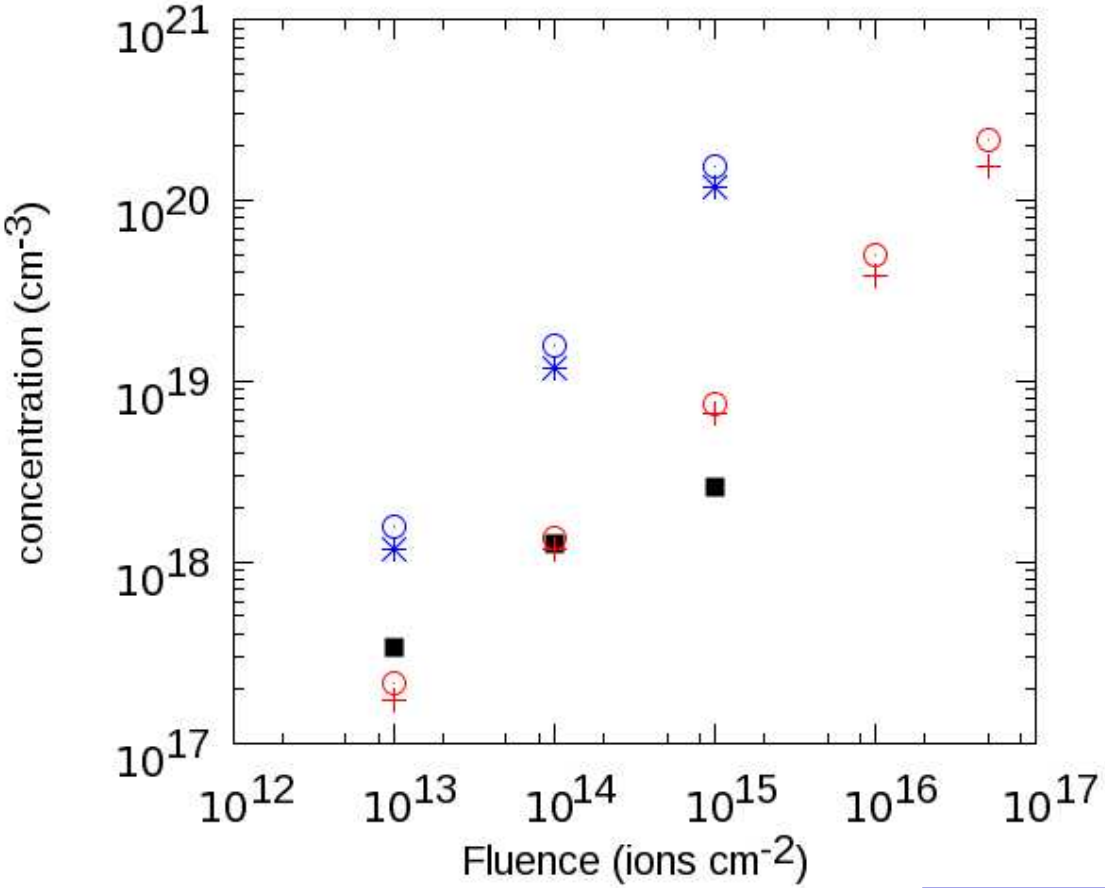


v mv m>1 1He.1v nHe.mv m>1

non mobile

Defect population after implantation stage: v, mv, nHe.pSIA, He.v, nHe.v

End of implantation sequence

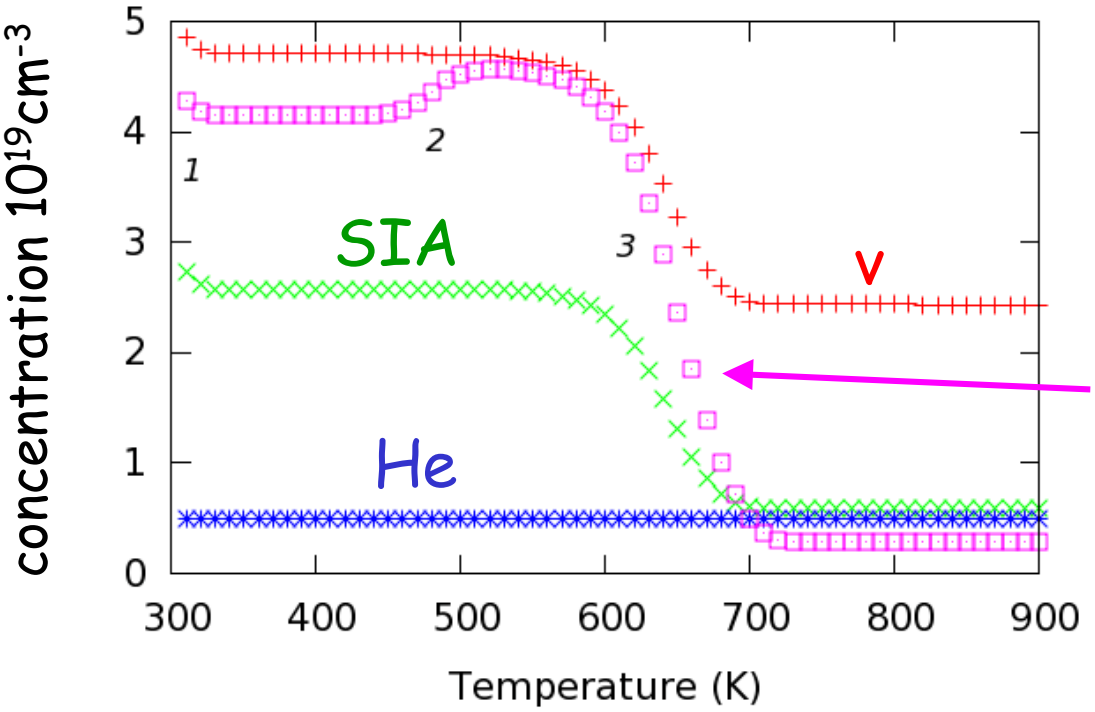


Experimental data ■ (PAS)
SRIM ⊙
Marlowe ✳
Okmc, *v* +
Okmc, total number of *v* ⊙

Good agreement with exp. data

During the isochronal annealing, v start moving, small mv clusters emit

10^{16} ion/cm^{-2}

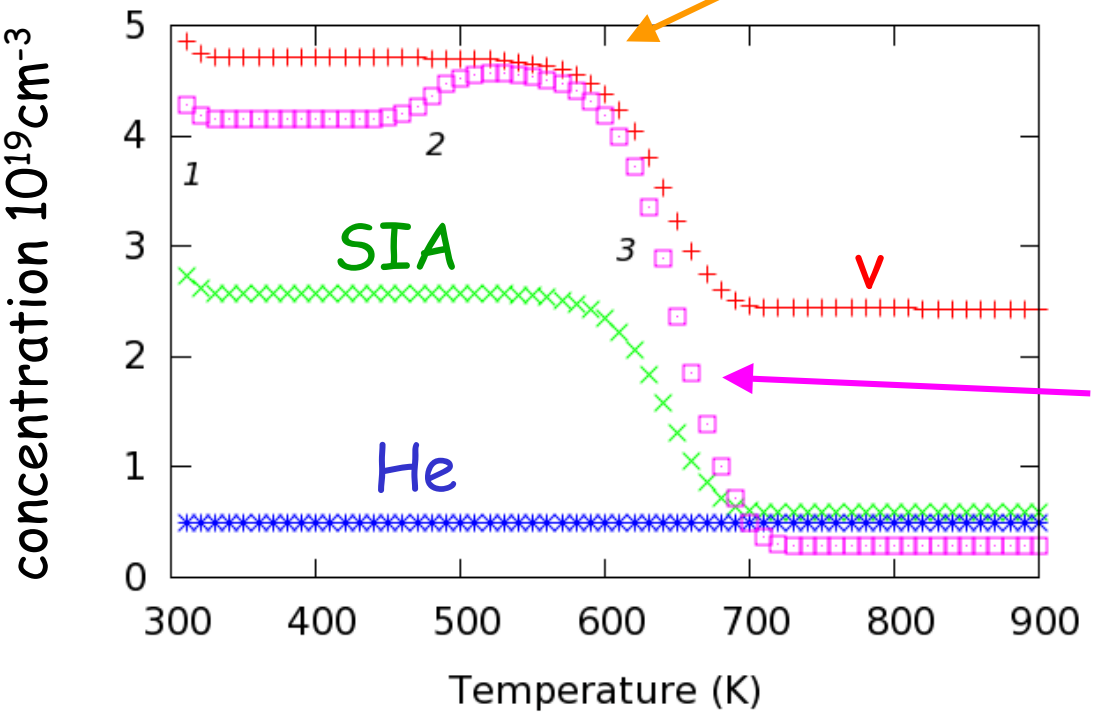


v clusters (mv, nHe.mv)

During the isochronal annealing, v start moving, small mv clusters emit

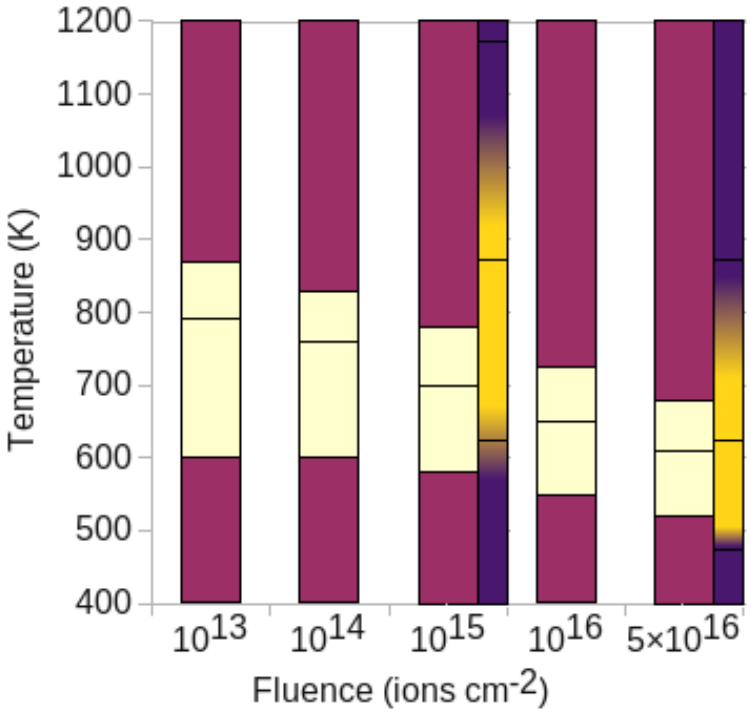
Clustering phase

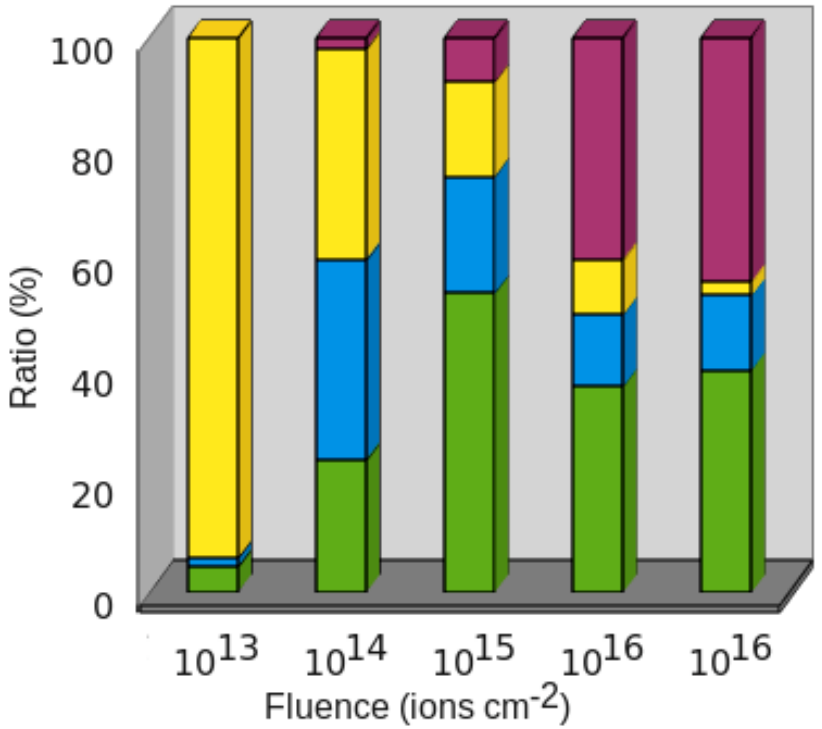
10^{16} ion/cm^{-2}



v clusters (mv, nHe.mv)

Clustering T range depends on fluence. Amount of desorption, annihilation and clustering also

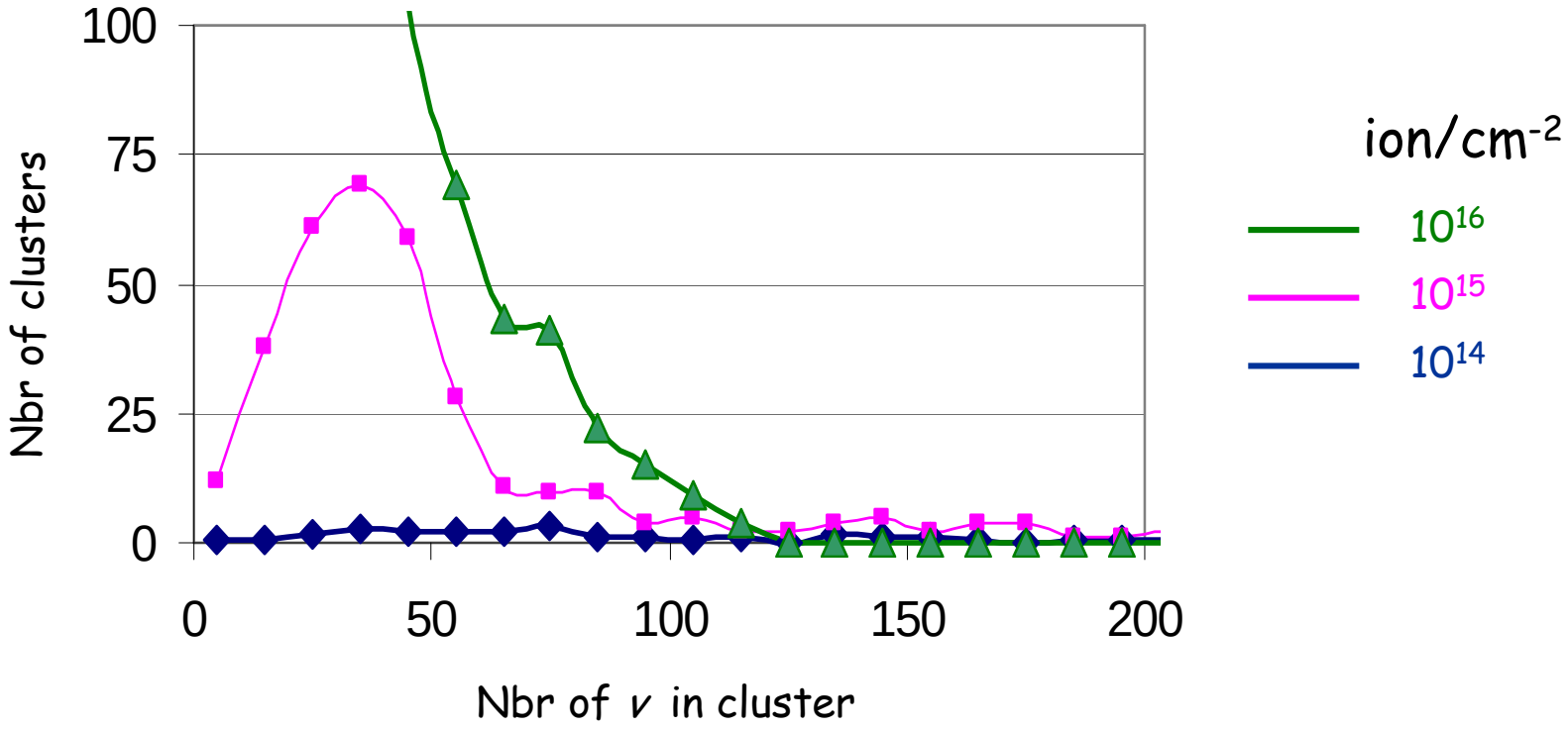




At 900K

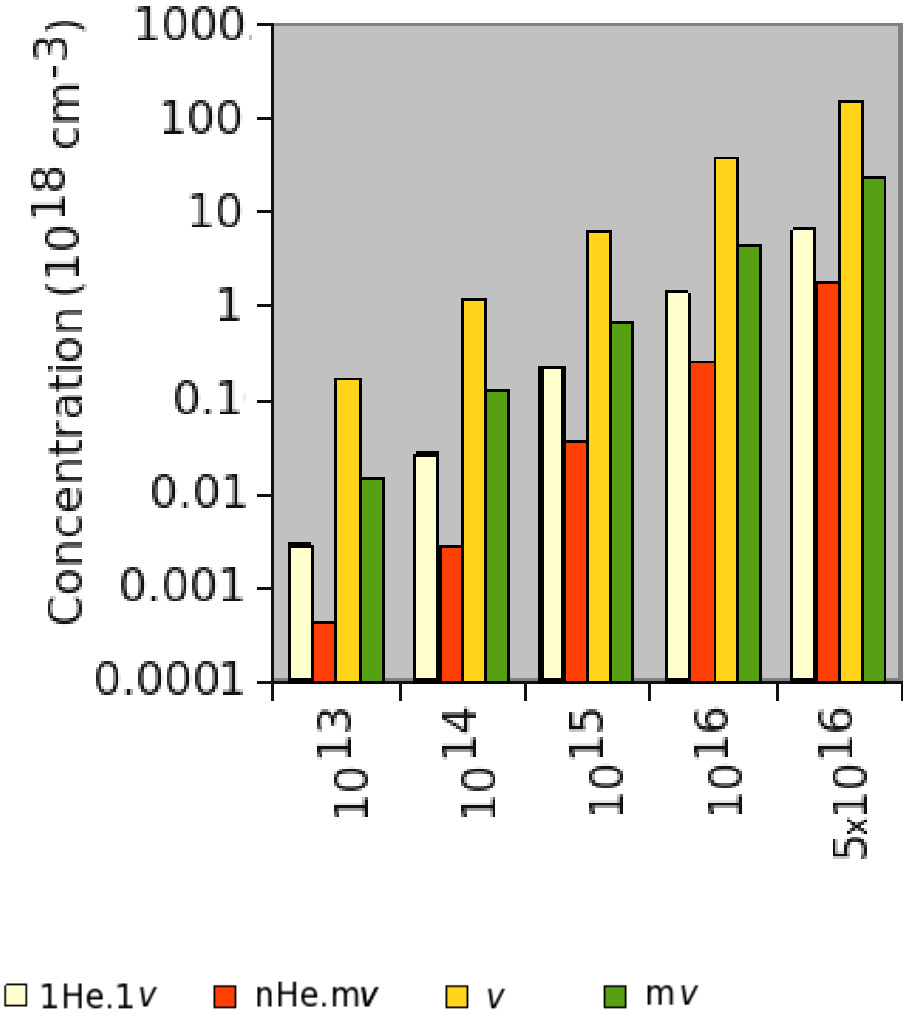
Desorption
Annihilation
Mixed clustering
Pure clustering

Vacancy cluster distribution at 900 K

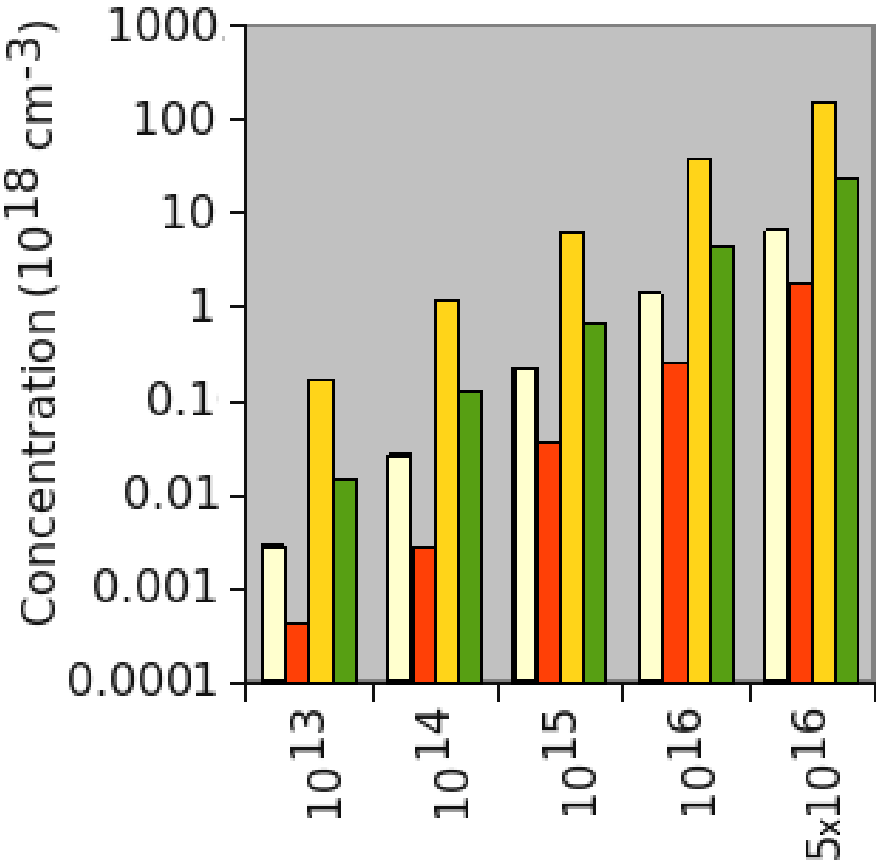


The higher the fluence, the smaller the v clusters as observed experimentally

Cluster distribution at the end of the implantation

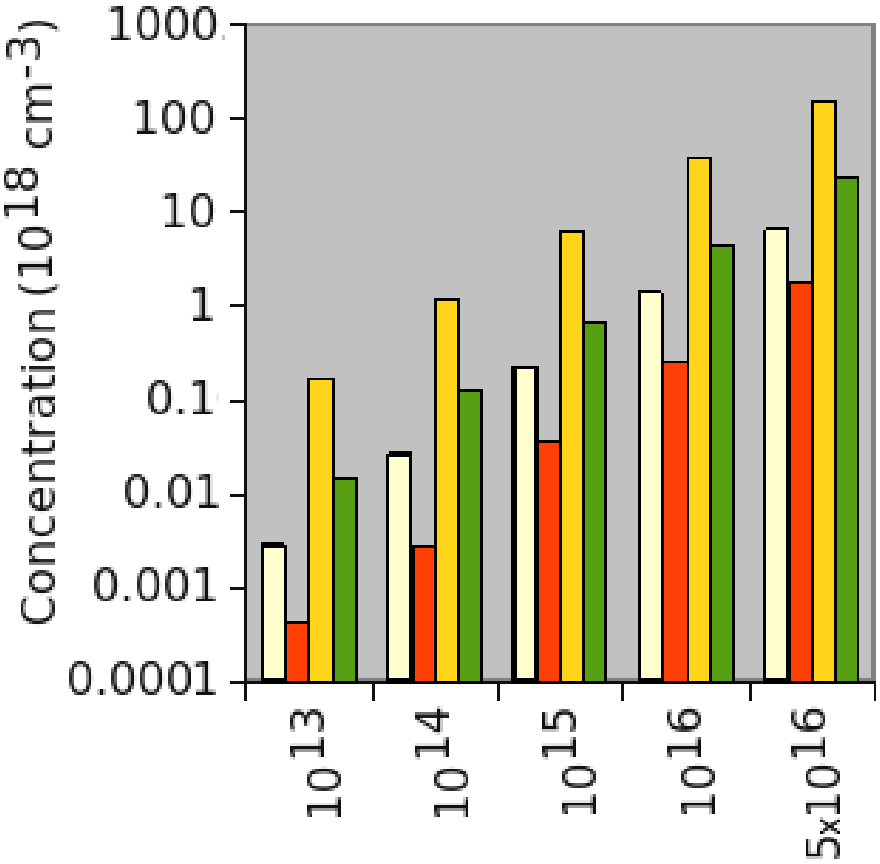


Cluster distribution at the end of the implantation



Free vacancies

Cluster distribution at the end of the implantation

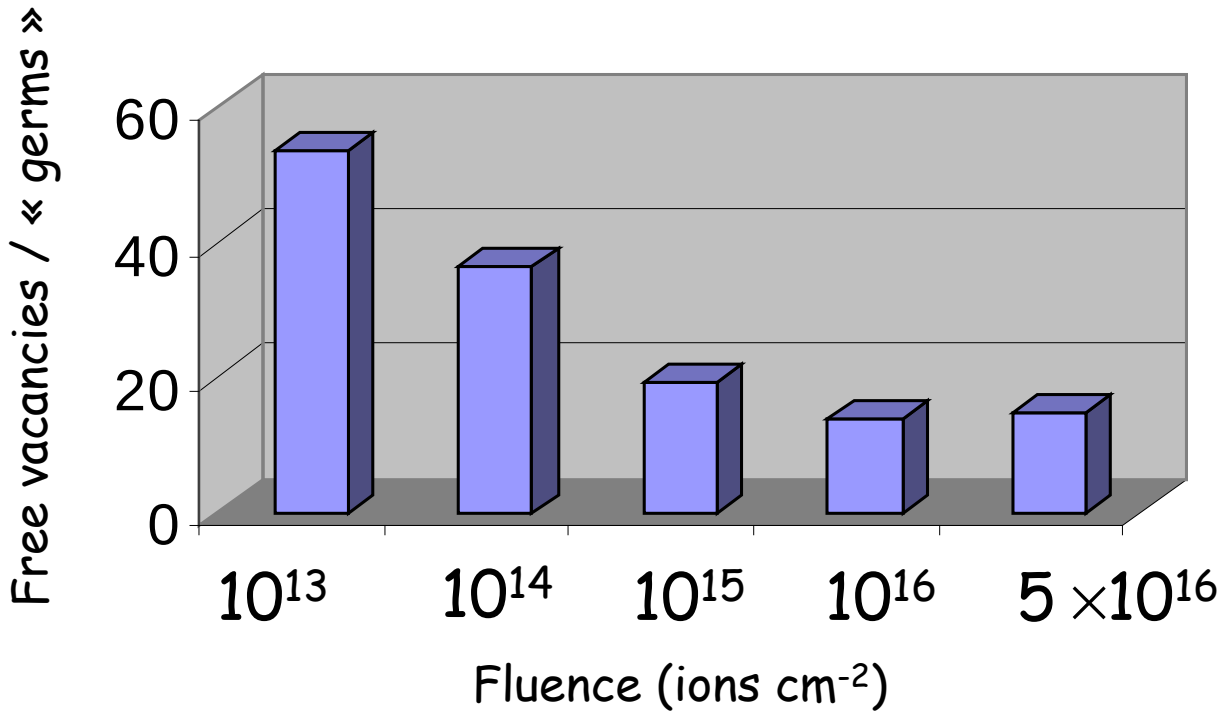


Germs

nHe.pSIA 1He.1*v* nHe.m*v* *v* m*v*

Free vacancies

At high fluence, it is easier to form clusters because the vacancies have less distance to cover. Furthermore, there are more germs and less desorption.



« germs » = non mobile clusters = nv.mHe + nSIA.mHe

Without He, not the same trends anymore

Conclusions

==> Ab initio calculations:

- He is very mobile
- small He clusters are mobile, 2He clusters perform 1D motion at low T.
- He and He clusters bind to almost everything

- di-vacancy is unstable
- vacancies stabilise He clusters

==> OKMC model parameterized on ab initio data and exp.:

- relevance of implantation model
- need more information on mobility of He clusters; trap mutation

OKMC model can help interpret experimental results and vice versa