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Métodos de simulación *ab initio* y aplicaciones a materiales bajo irradiación

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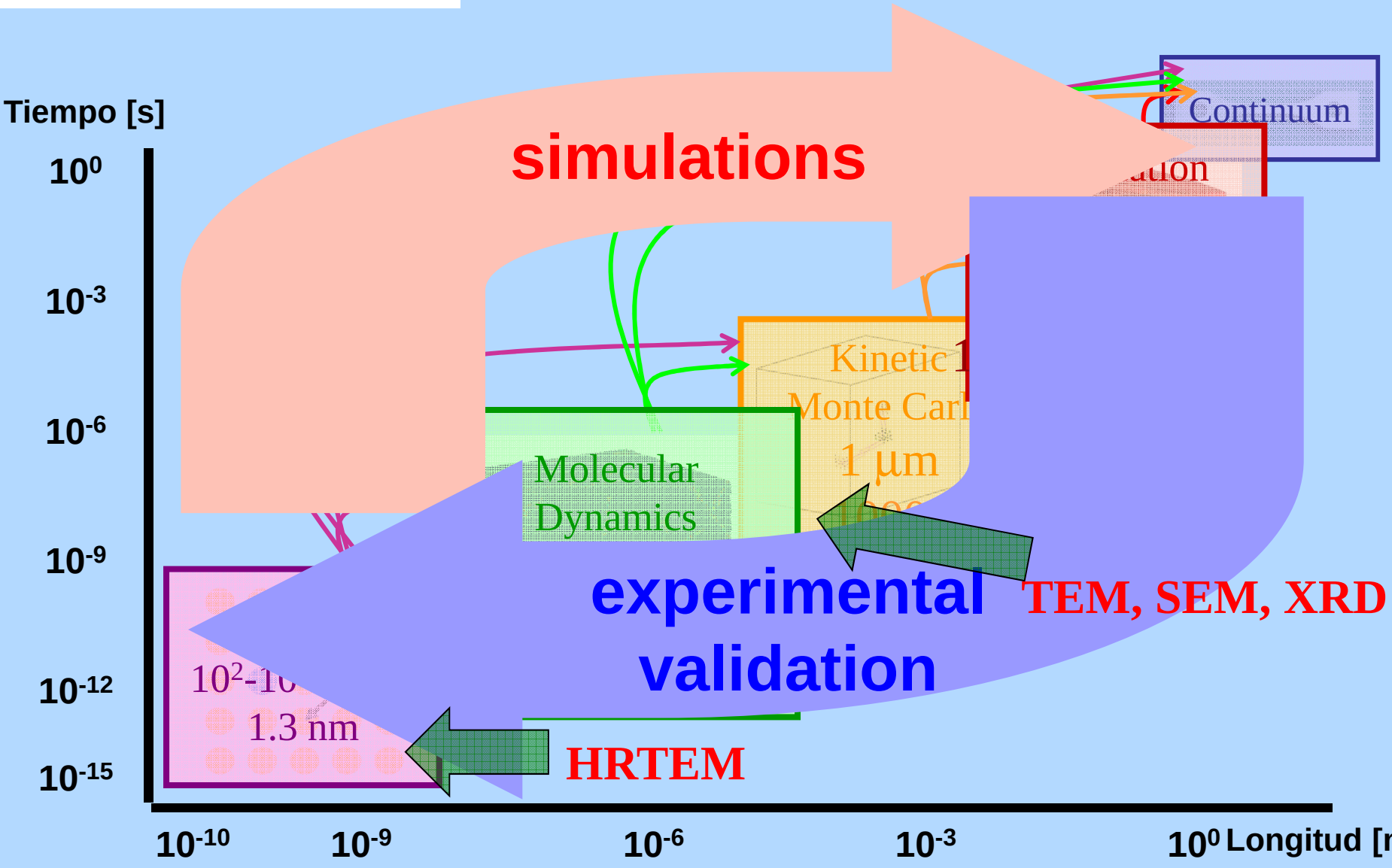
MULTISCALE MODELLING PARADIGM



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Solving Schrödinger's equation in solids

Form of potential

(non-)selfconsistent
"Muffin-tin" MT
atomic sphere approximation (ASA)
Full potential : FP
pseudopotential (PP)

Relativistic treatment of the electrons

non relativistic
semi-relativistic
fully-relativistic

exchange and correlation potential

Hartree-Fock (+correlations)
Density functional theory (DFT)
Local density approximation (**LDA**)
Generalized gradient approximation (**GGA**)
Beyond LDA: e.g. **LDA+U**

$$\left[-\frac{1}{2}\nabla^2 + V(r)\right]\varphi_i^k = \varepsilon_i^k \varphi_i^k$$

Schrödinger – equation
(Kohn-Sham equation)

Representation of solid

non periodic
(cluster)
periodic
(unit cell)

Basis functions

plane waves : PW
augmented plane waves : APW
atomic oribtals. e.g. Slater (STO), Gaussians (GTO), LMTO, numerical basis

Treatment of spin

Non-spinpolarized
Spin polarized
(with certain magnetic order)

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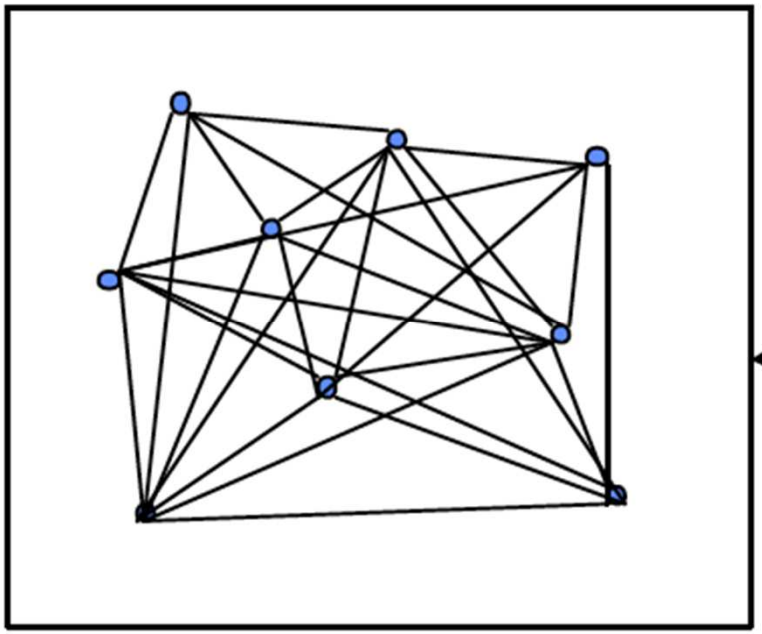
How does DFT work

$$\sum_{i<j}^{N} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}$$

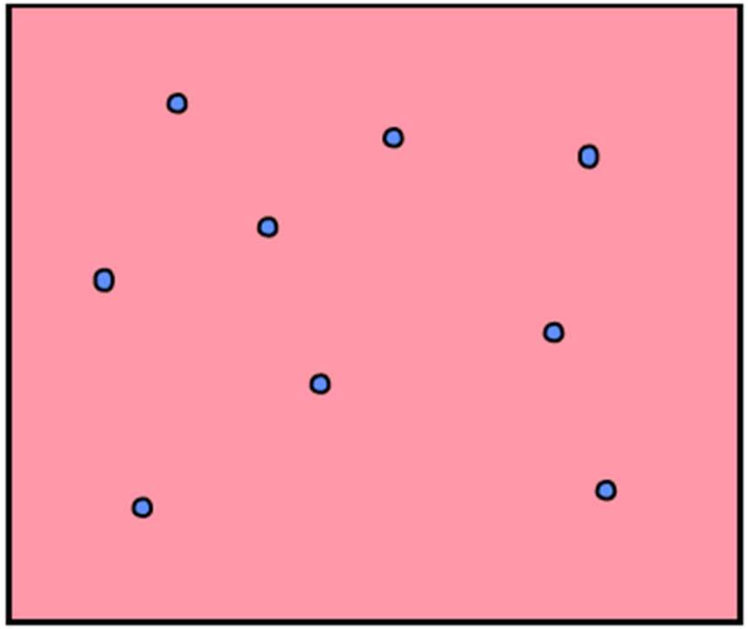
$$\Psi(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N)$$

$$v_H(\mathbf{r}) + v_{xc}(\mathbf{r})$$

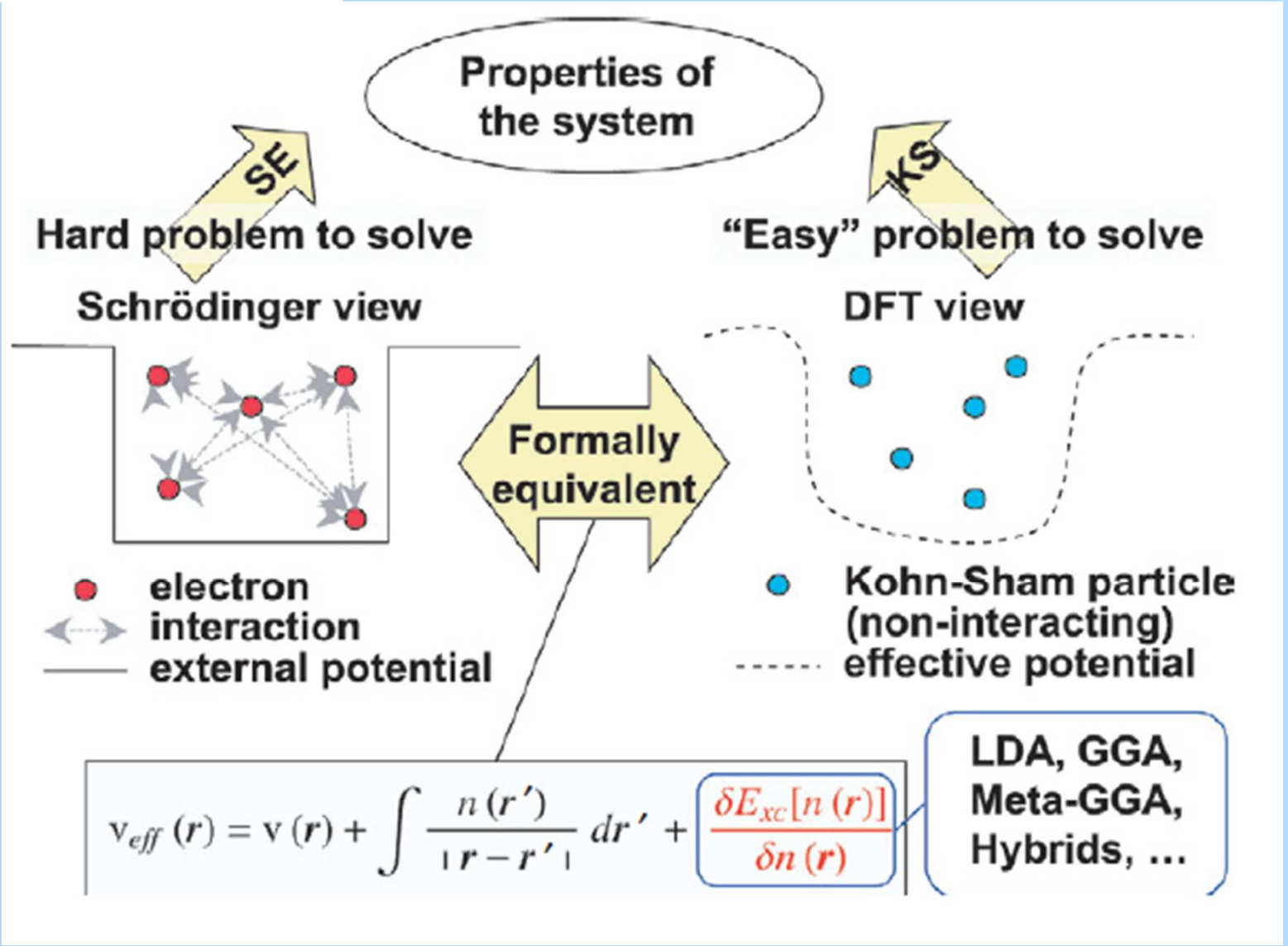
$$\phi_i(\mathbf{r})$$



E, ρ



DFT AND FUNCTIONALS



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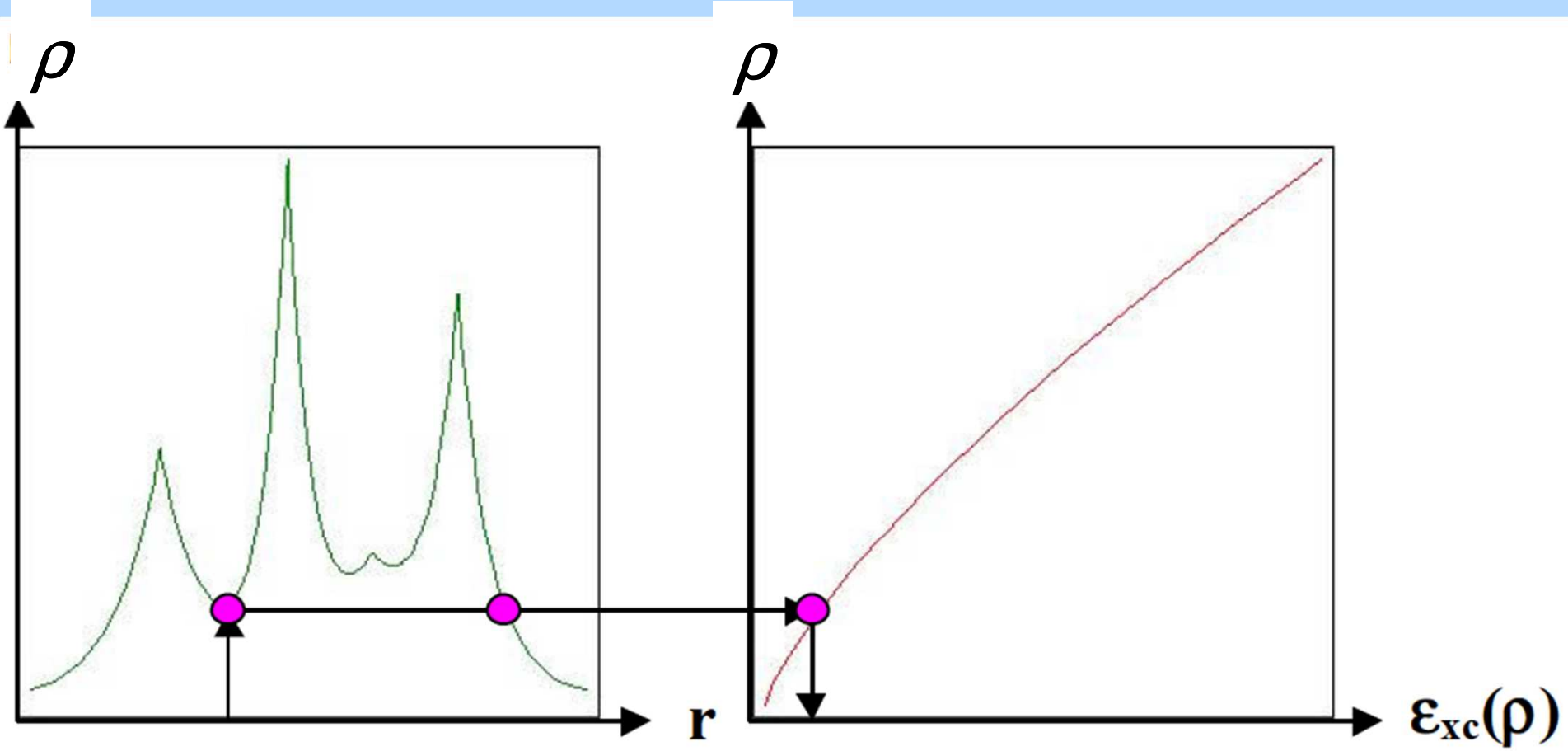
LDA approximation

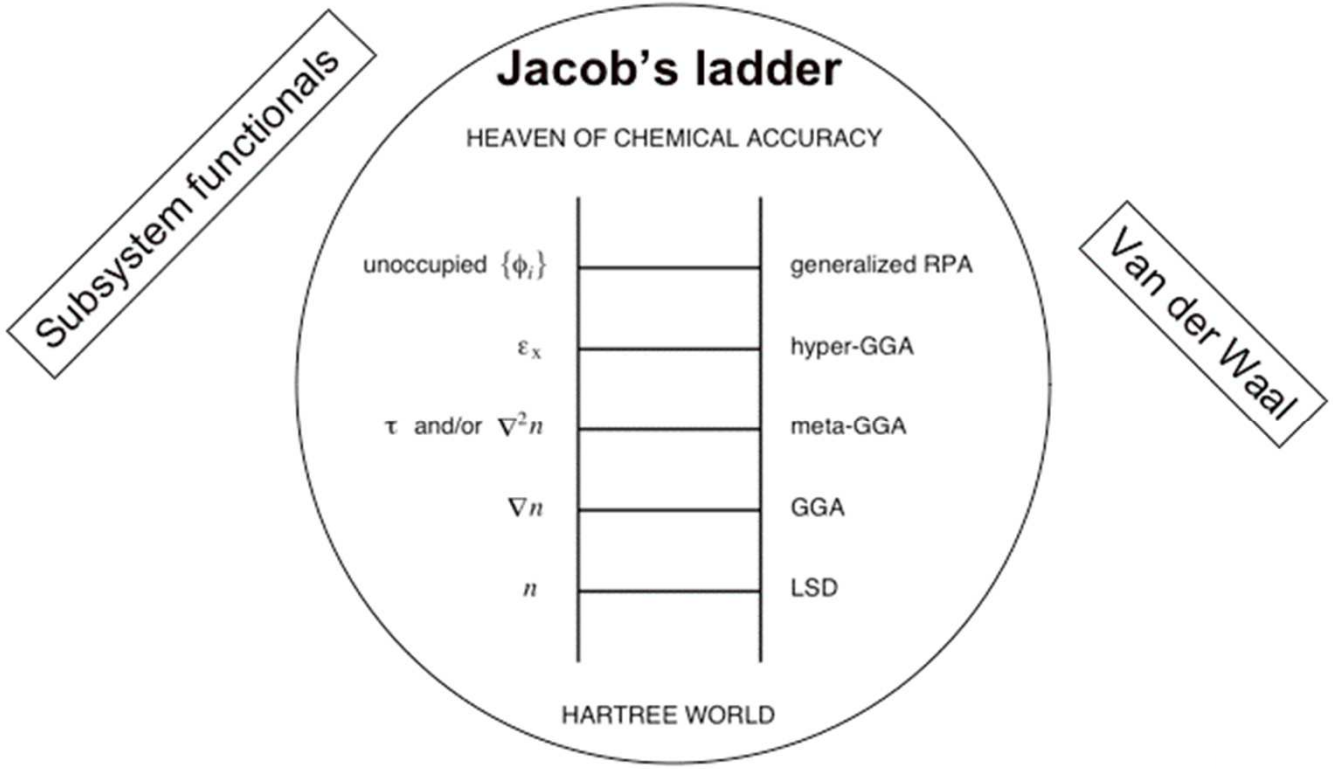


Exchange energy density of the inhomogeneous system

Exchange energy density of the homogeneous electron gas

Accuracy limiting factor

Hierarchy of functionals



From J.P. Perdew et. al. JCP 123, 062201 (2005).

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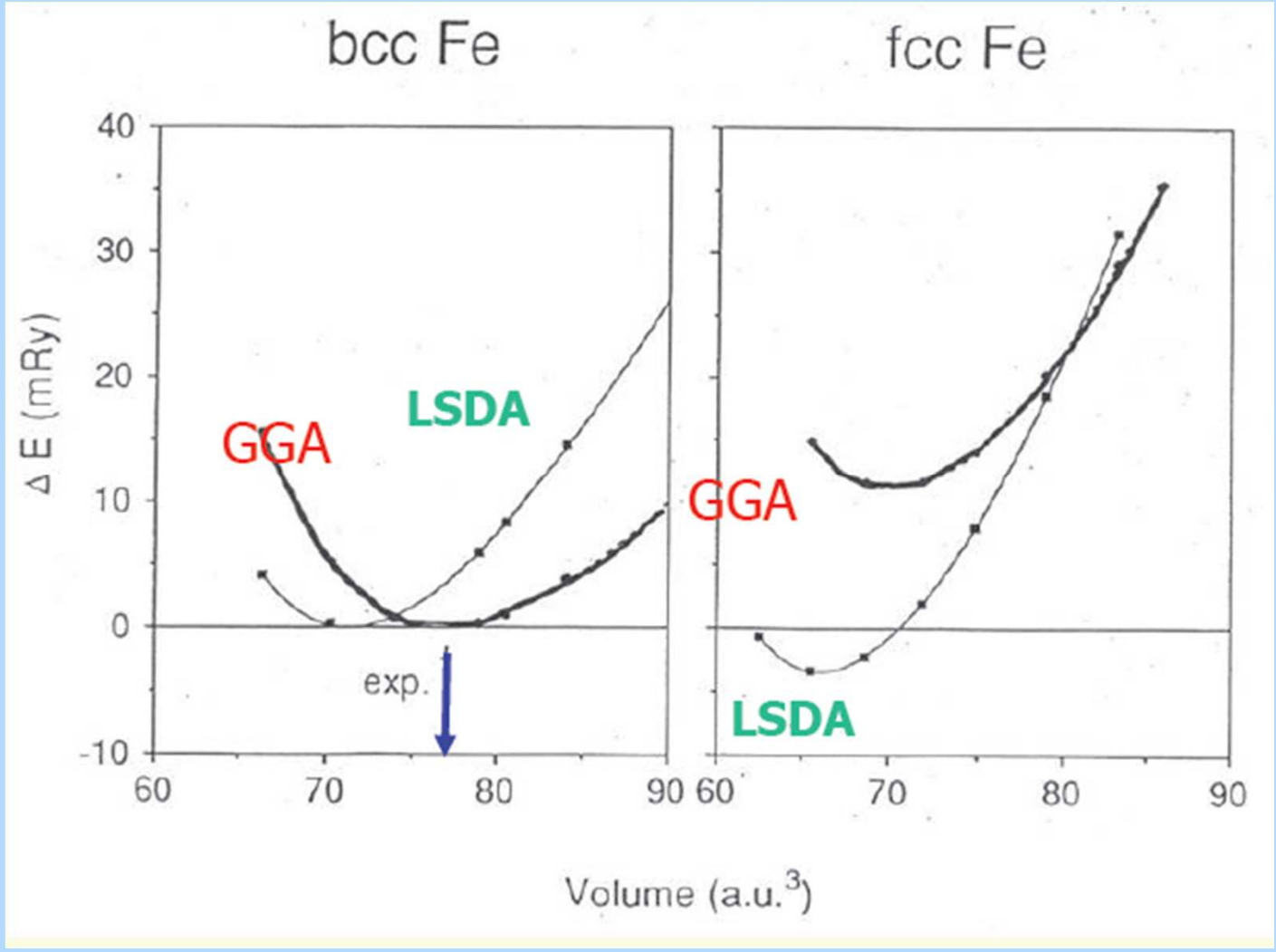
DFT ground state of iron



LSDA

NM
fcc
in contrast to experiment

GGA

FM
bcc
Correct lattice constant

Experiment

FM
bcc

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Electronic structure determination

THREE BASIC METHODS:

- Plane waves and grids → solution of Schr. and Poisson eqs.
  simplicity and speed of FFT
  smooth pseudopotentials

- Localized atomic(-like) orbitals → atomic-like states
  good scaling with size

- Atomic sphere methods → atomic-like features near nucleus
  smooth functions between atoms
  most demanding

Augmented: APW, KKR

Linearized: LAPW, LMTO

→ most precise full-potential

APW+lo e.g.TM 3d

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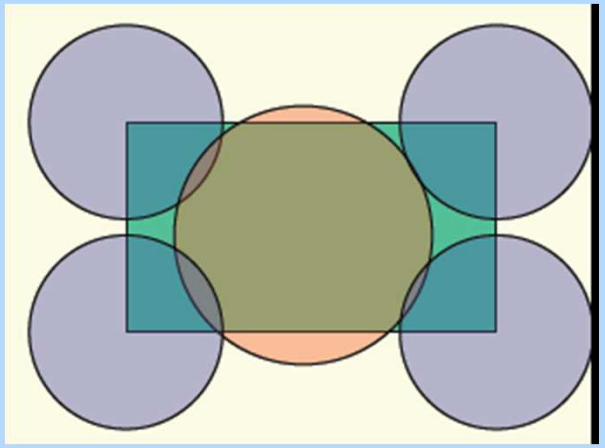
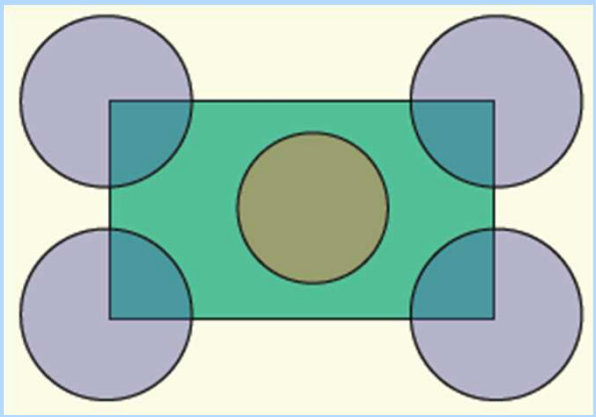
Shape approximations to "real" potentials

1. Atomic sphere approximation (ASA)
   - overlapping spheres "fill" all volume
   - potential spherically symmetric



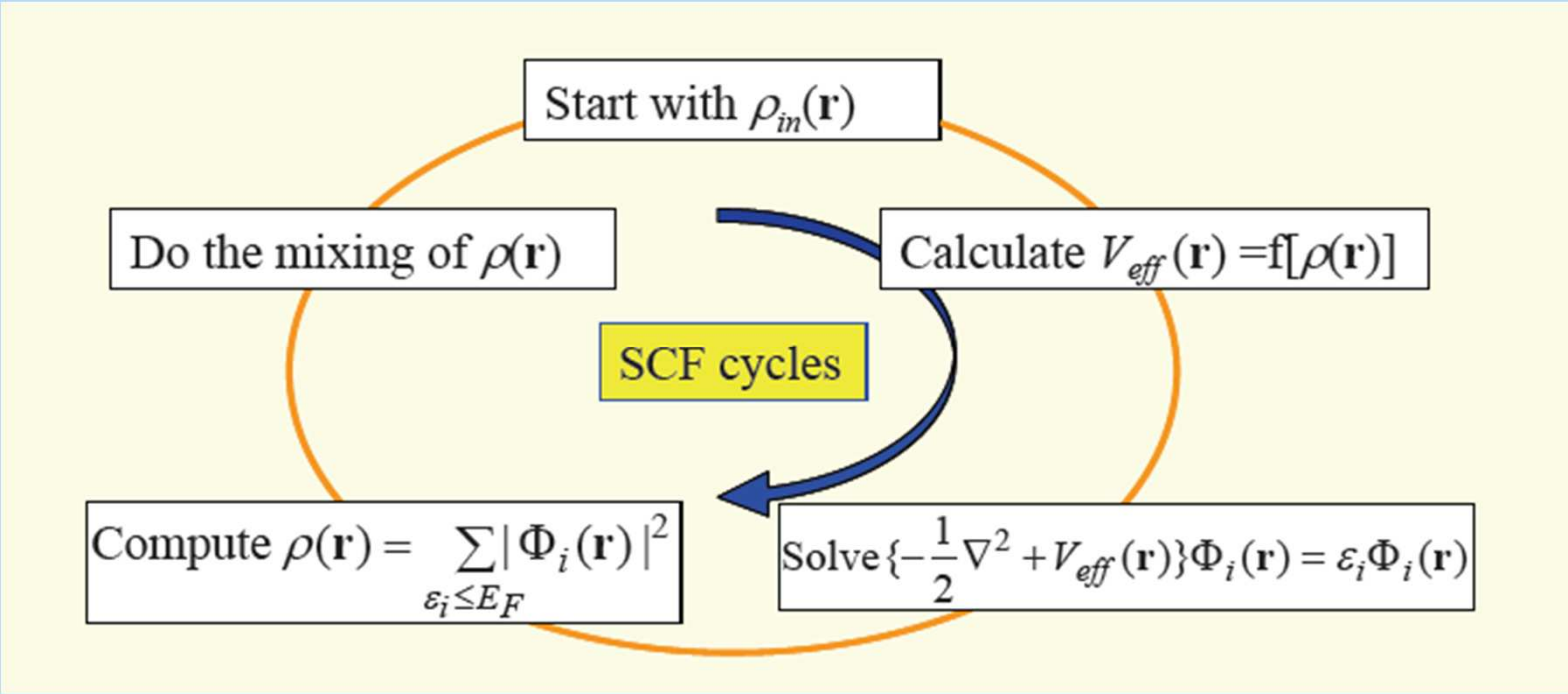
2. "muffin-tin" approximation (MTA)
   - non-overlapping spheres with spherically symmetric potential +
   - interstitial region with V=const.



3. "full"-potential
   - no shape approximations to V

Self-consistent field (SCF) calculations

- In order to solve $H\Psi=E\Psi$, we need to know the potential V
- For $V(r)$ we need the electron density $\rho(r)$
- The density $\rho(r)$ can be obtained from $\Psi(r)^*\Psi(r)$
- $\Psi(r)$ is unknown before $H\Psi=E\Psi$ is solved



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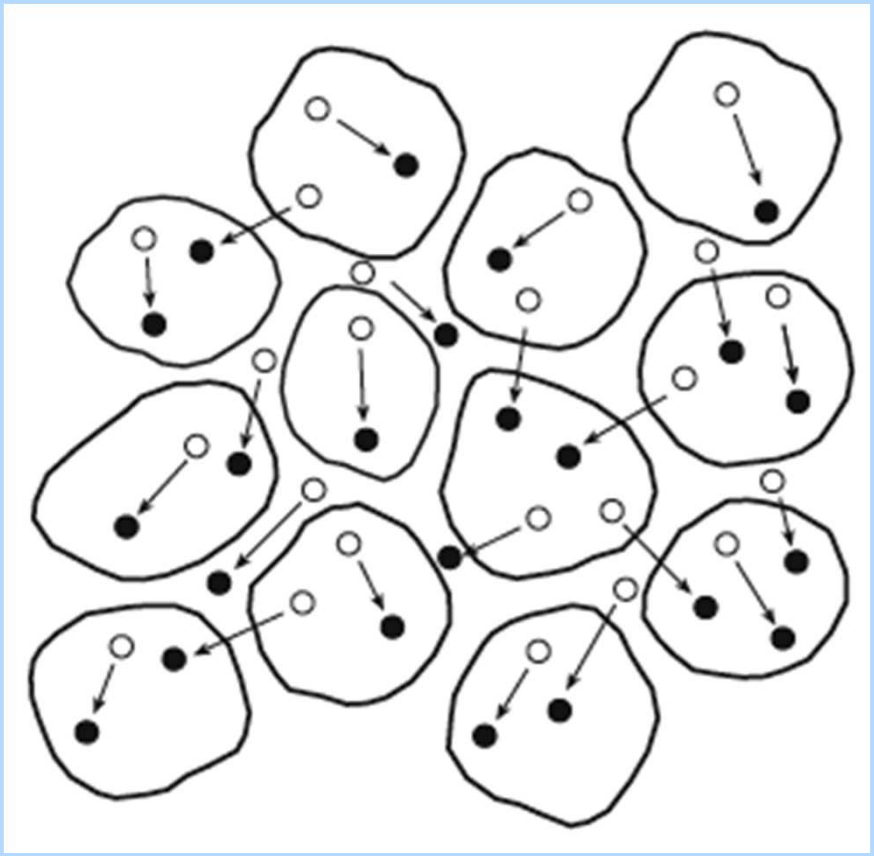
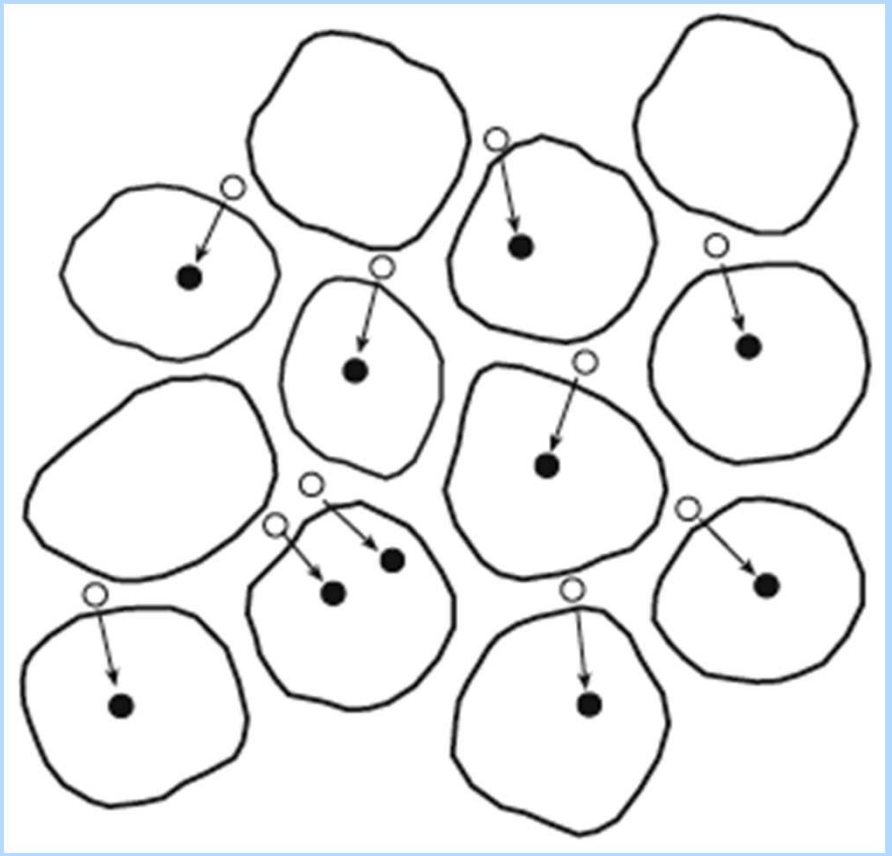
Irradiated nanocrystalline solids

Ovidko 2005





High irradiation dose

Low irradiation dose

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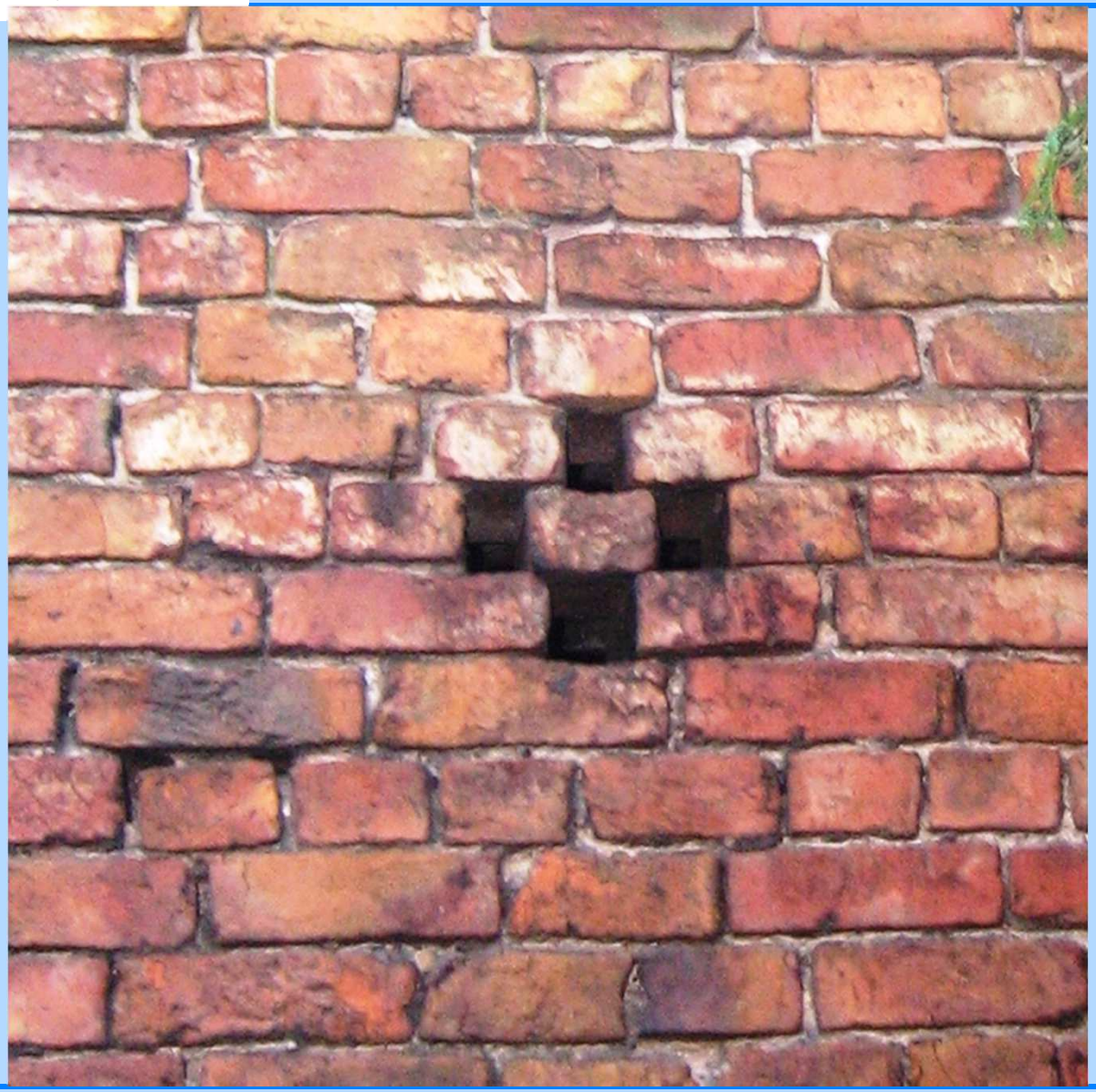
Tetravacancy



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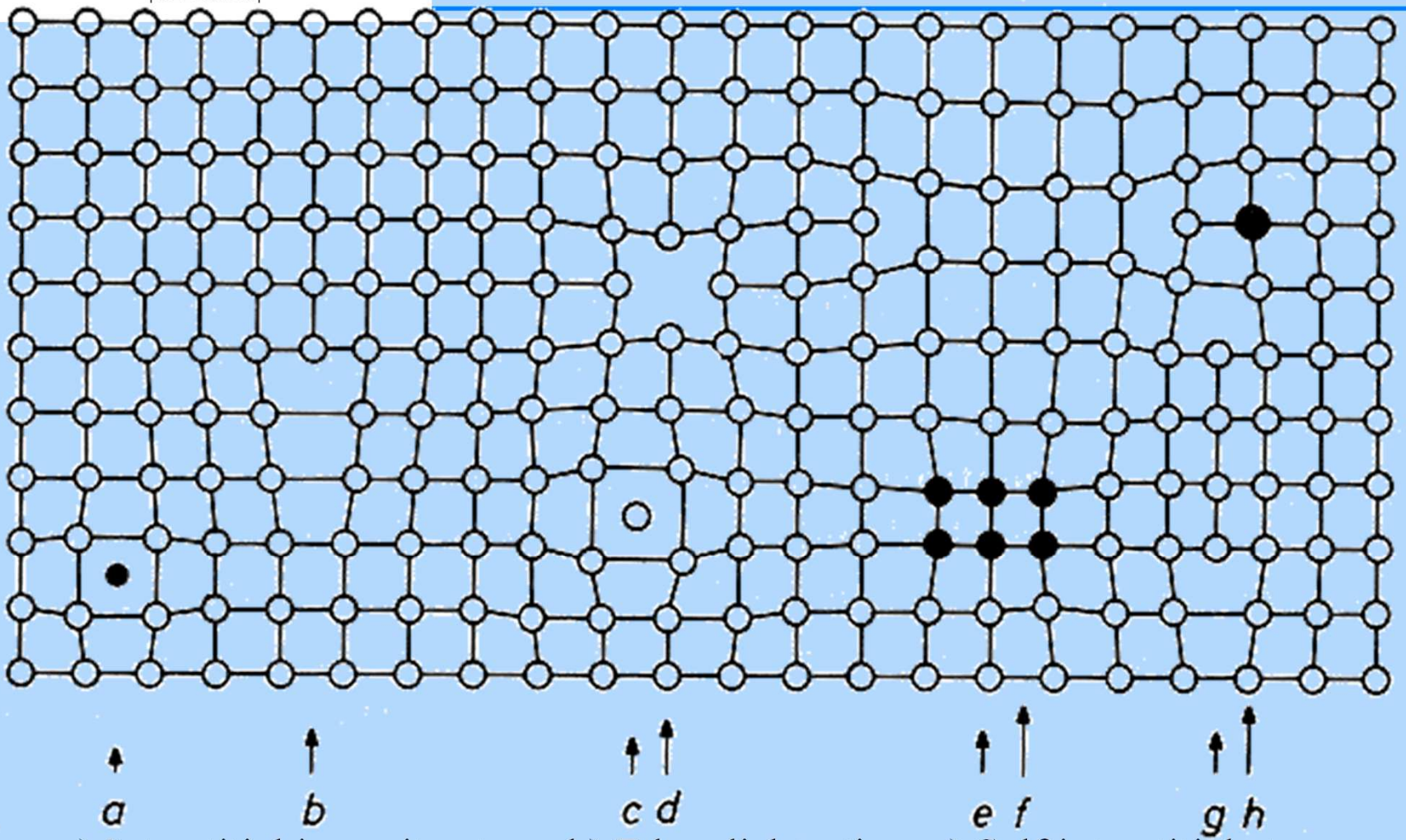
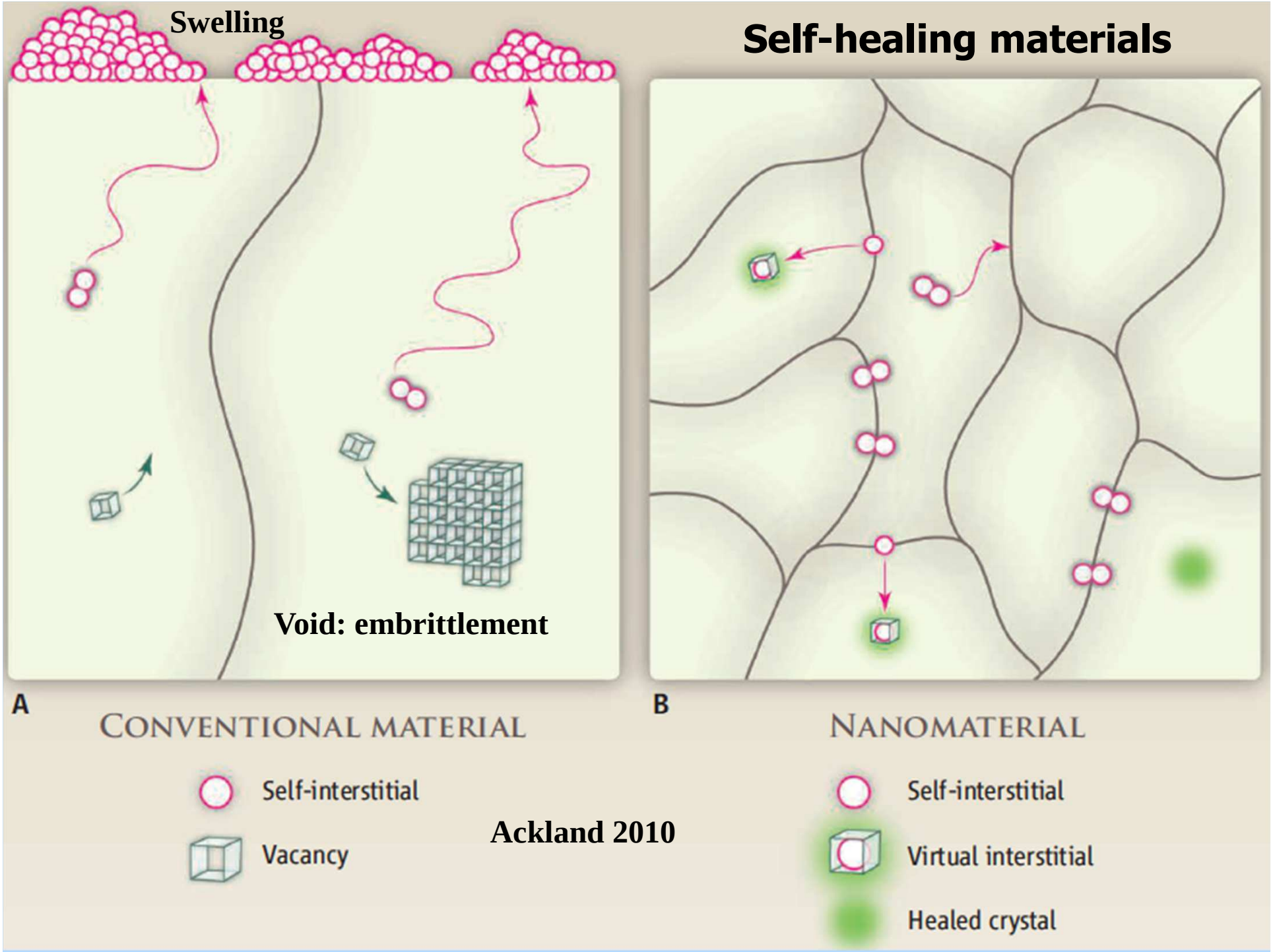
Defects in crystals

H. Föll



a) Interstitial impurity atom, b) Edge dislocation, c) Self interstitial atom, d) Vacancy, e) Precipitate of impurity atoms, f) Vacancy type dislocation loop, g) Interstitial type dislocation loop, h) Substitutional impurity atom


Swelling
Self-healing materials
Void: embrittlement
A
CONVENTIONAL MATERIAL
B
NANOMATERIAL
Self-interstitial
Vacancy
Ackland 2010
Self-interstitial
Virtual interstitial
Healed crystal

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Irradiated W microstructural evolution

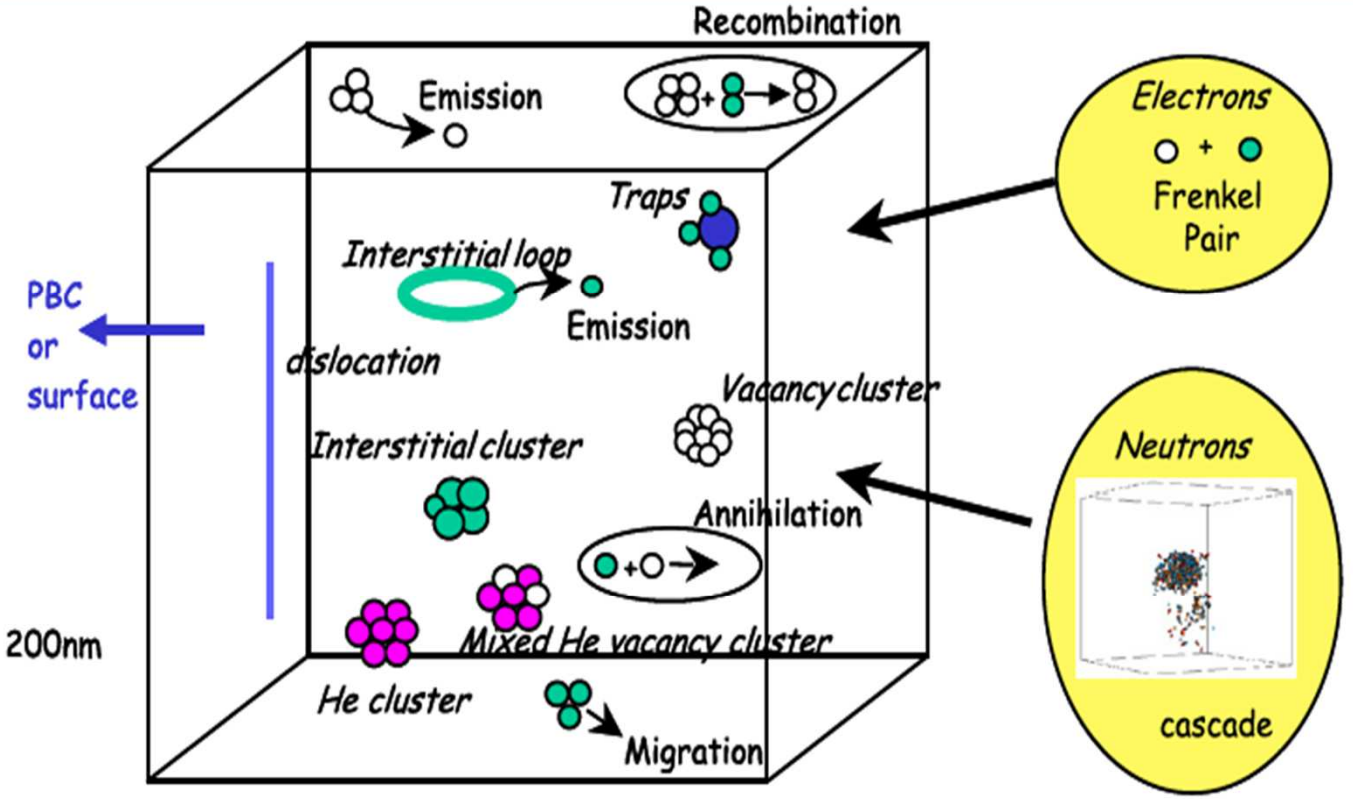

Fig. 1. Summary of the different events: migration, recombination, emission of single entities or trapping as well as electron or neutron irradiations, taking place in an object KMC simulation using the LAKIMOCA code. The white spheres are the vacancies, the green ones, the SIAs; the pink ones represent the helium atoms.

Ab initio data parameterise an OKMC model: analyse evolution of defect population during He desorption from W

Becquart 2010

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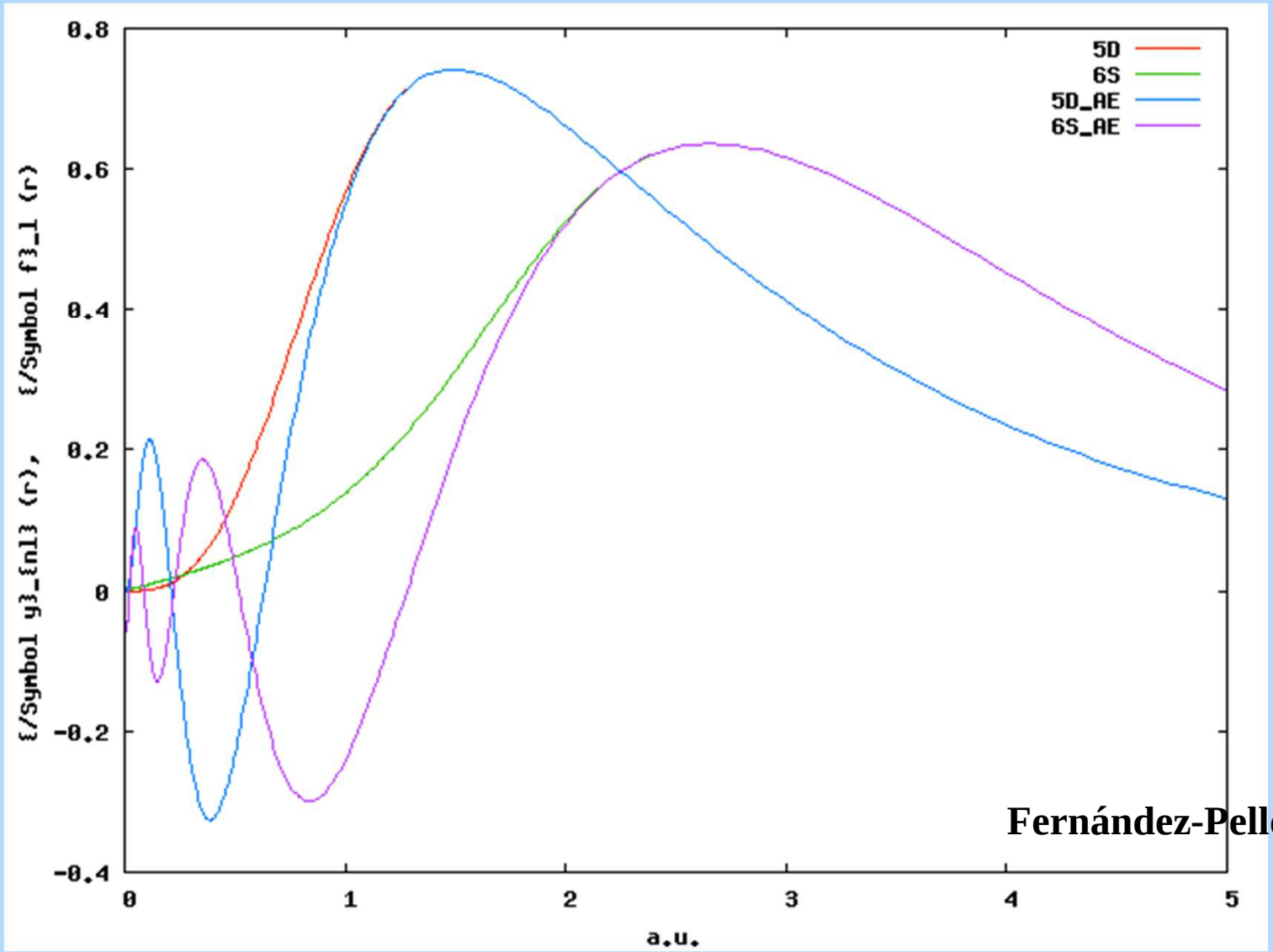
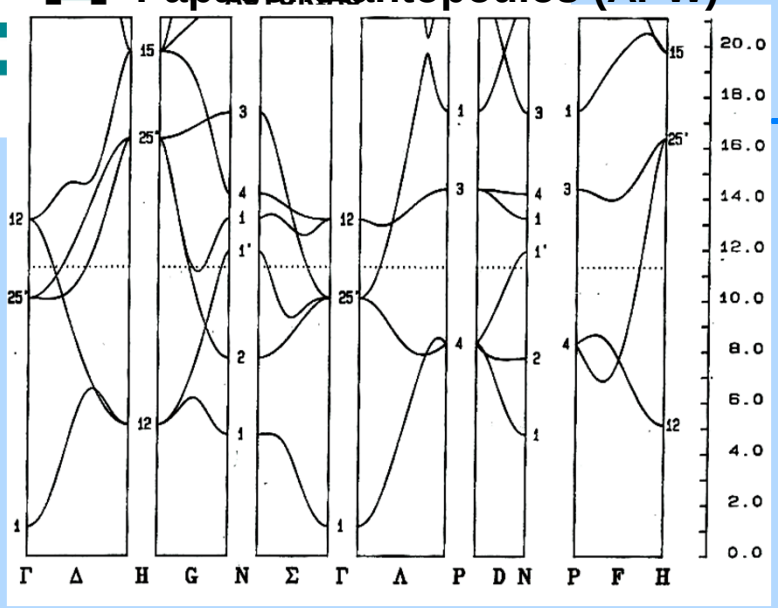
W Pseudopotential, QE, $5d^4 6s^2$, PBE

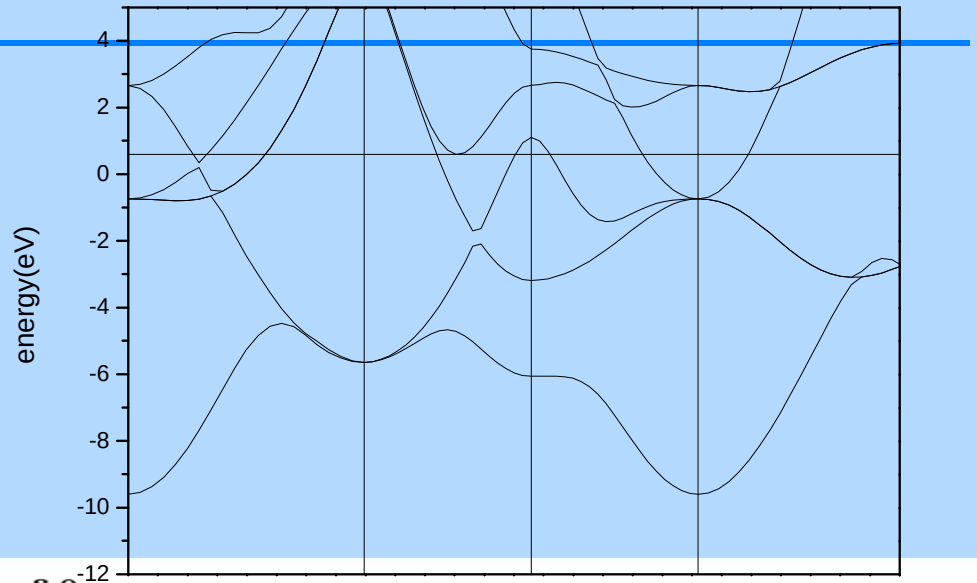


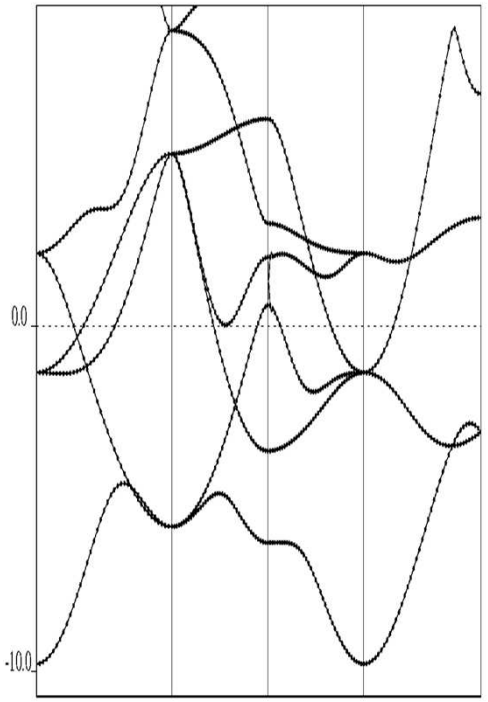
Fernández-Pello 2012

Papaconstantopoulos (APW)

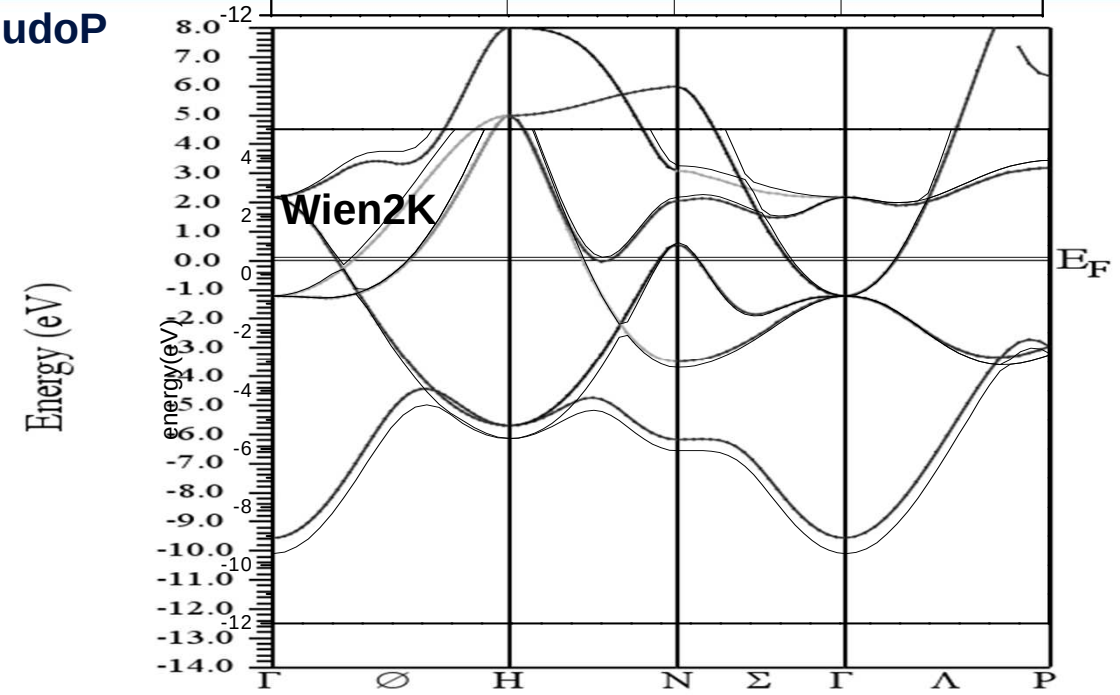


Fireball-GGA rc(s)=4.5 rc(p)=4.2 rc(d)=4.7
Lattice parameter=3.165 A





QE-PseudoP



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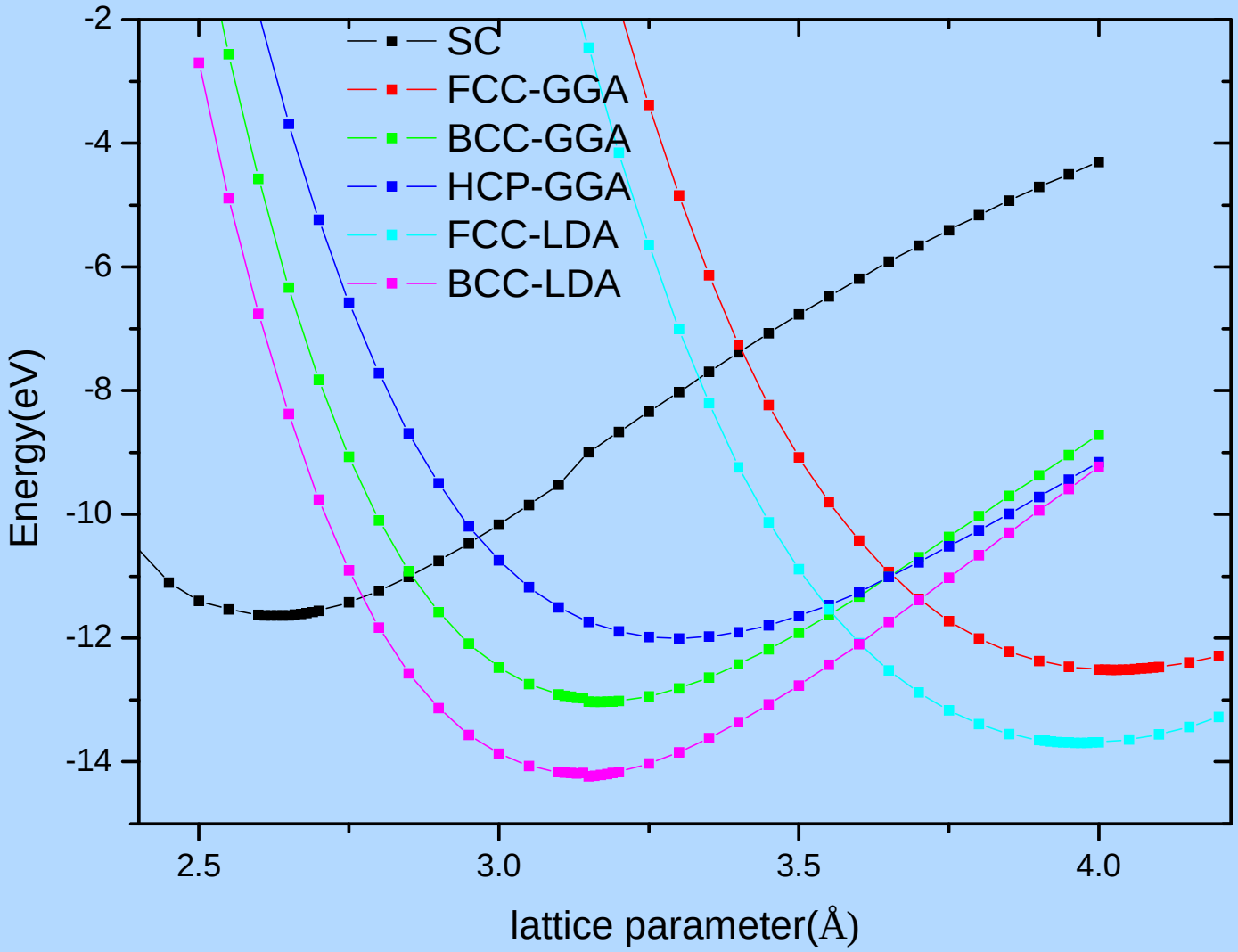
W bulk, E vs. V curves



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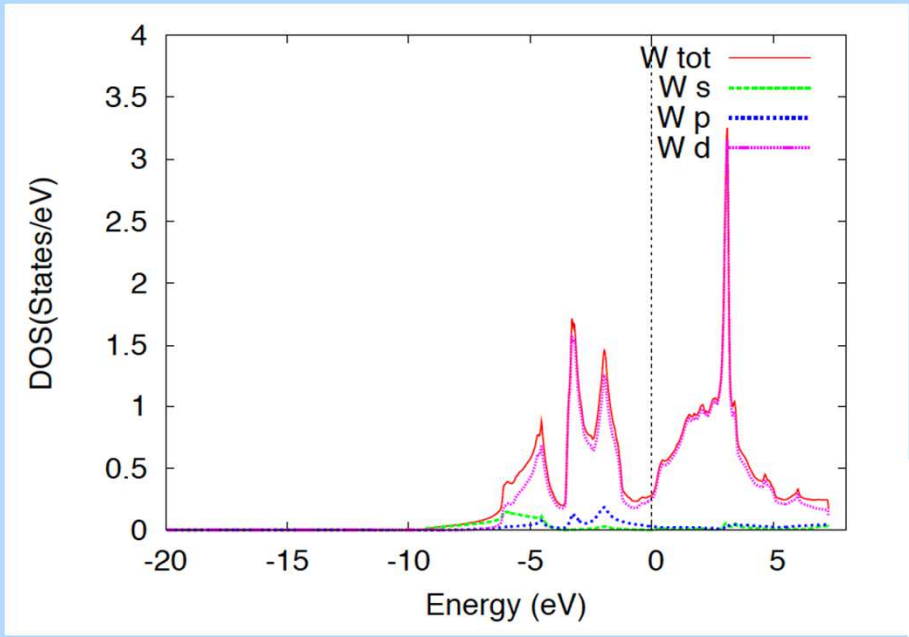
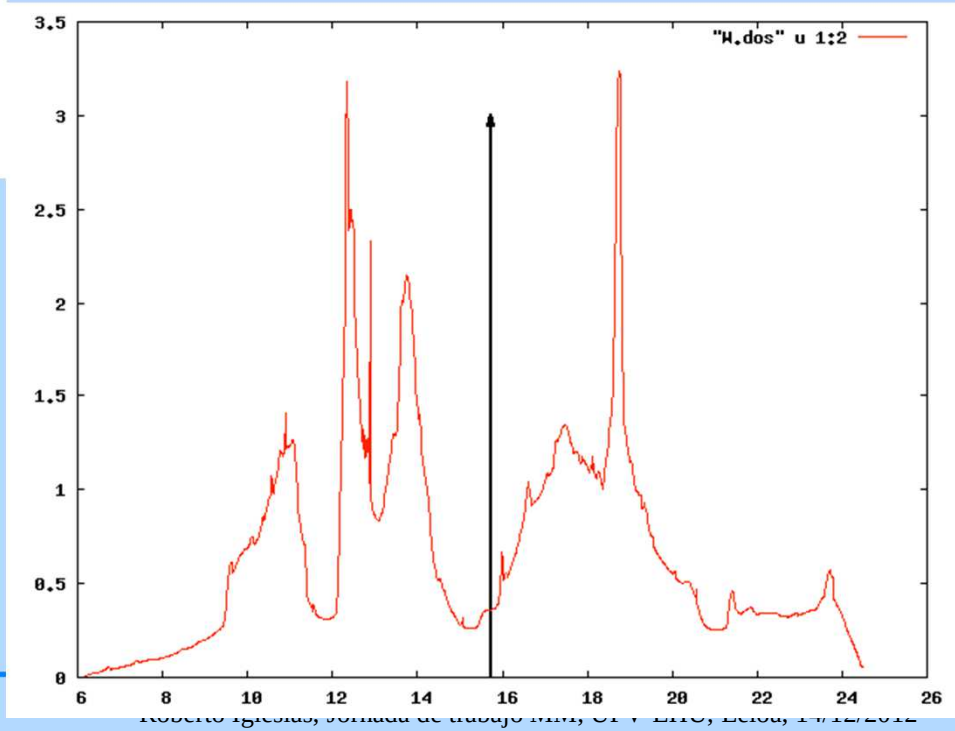
W DOS



Wien2k

QE-PseudoP



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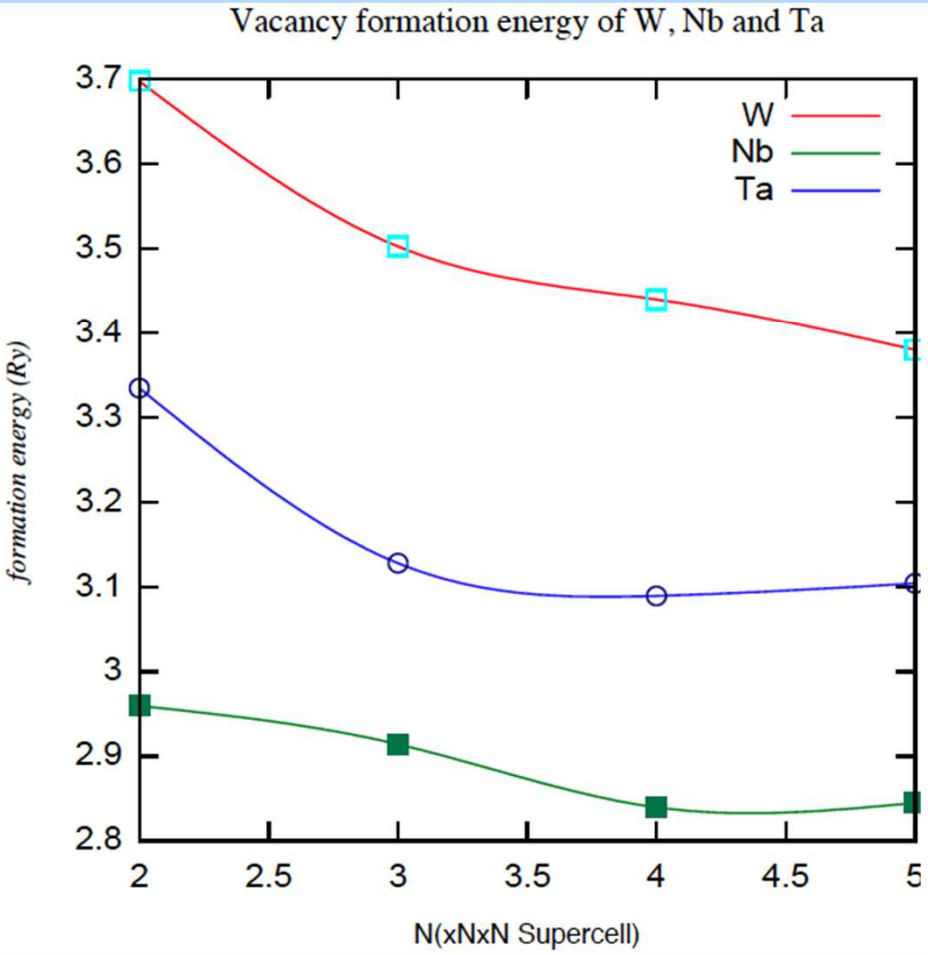
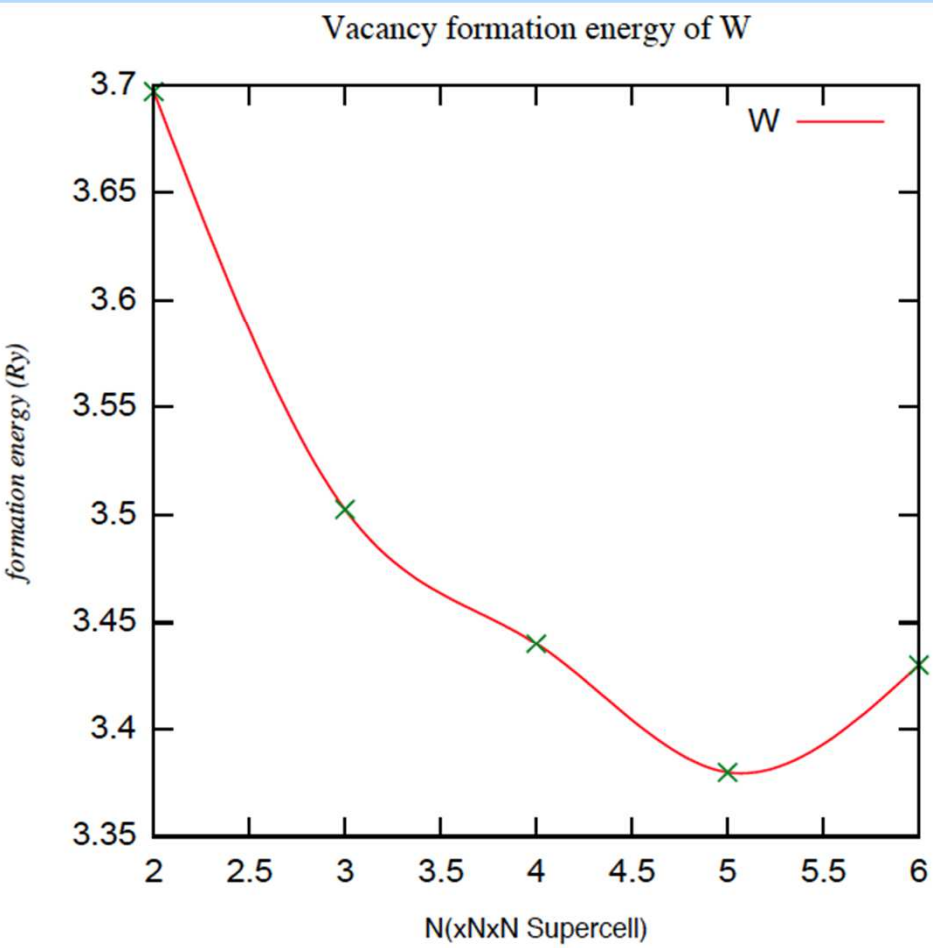
MonoV formation energies





Fernández-Pello 2012

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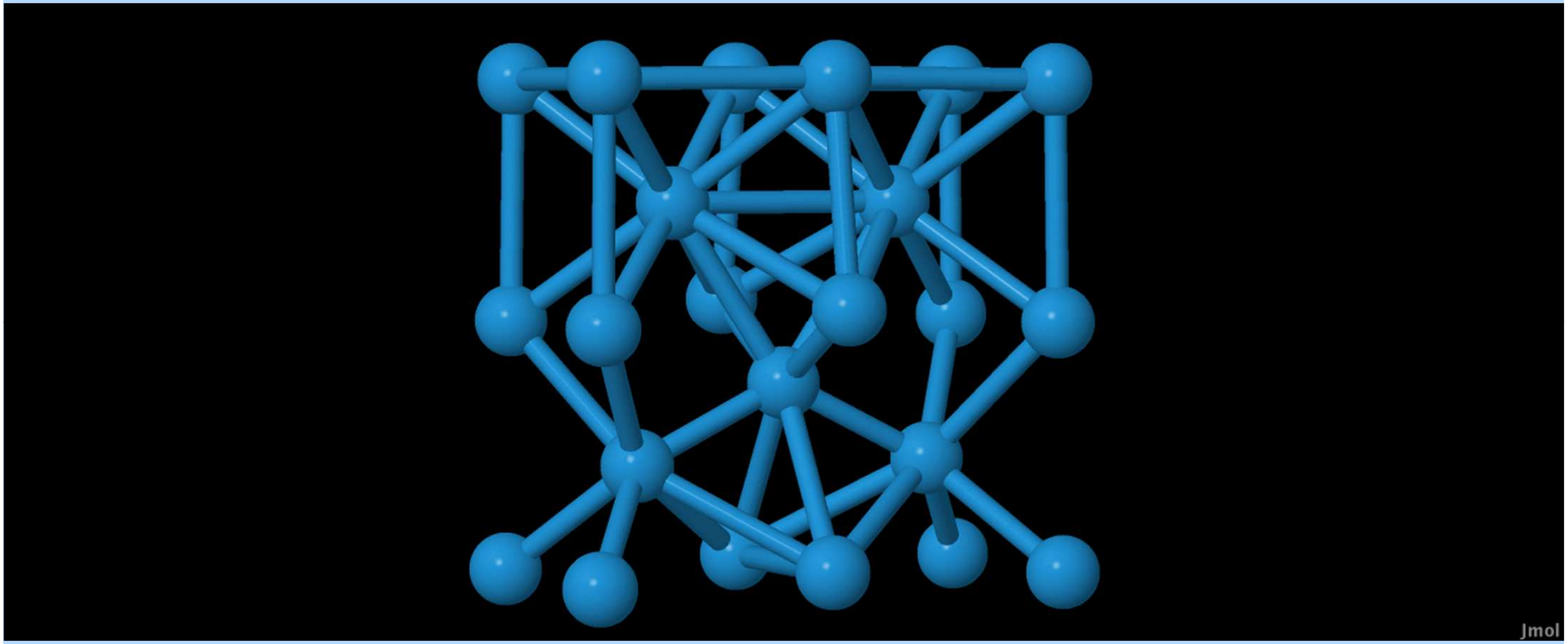
Tetra to octa migration pathways in W



González 2012

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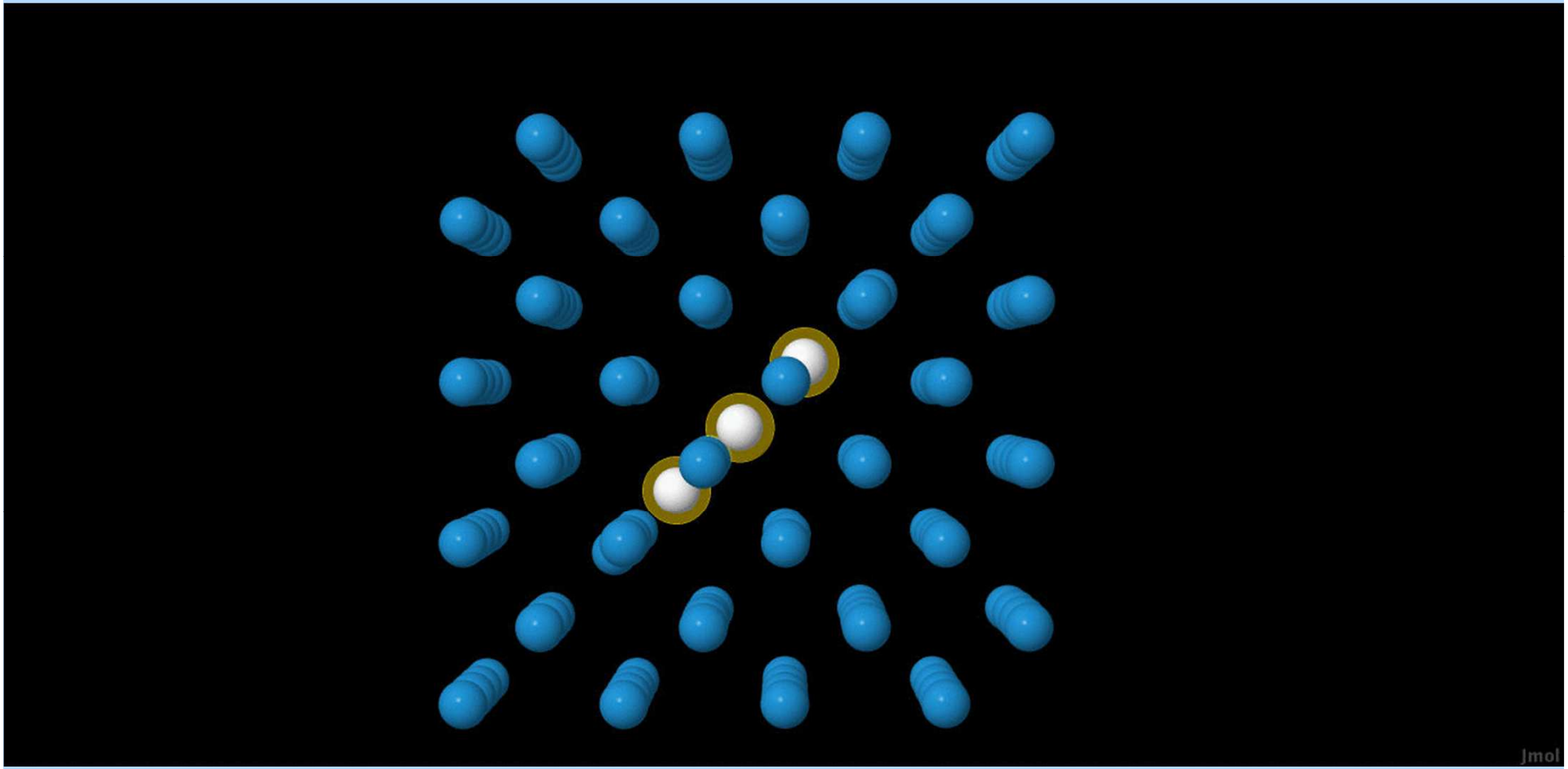
Migración de una *dumbbell* [111] (W)



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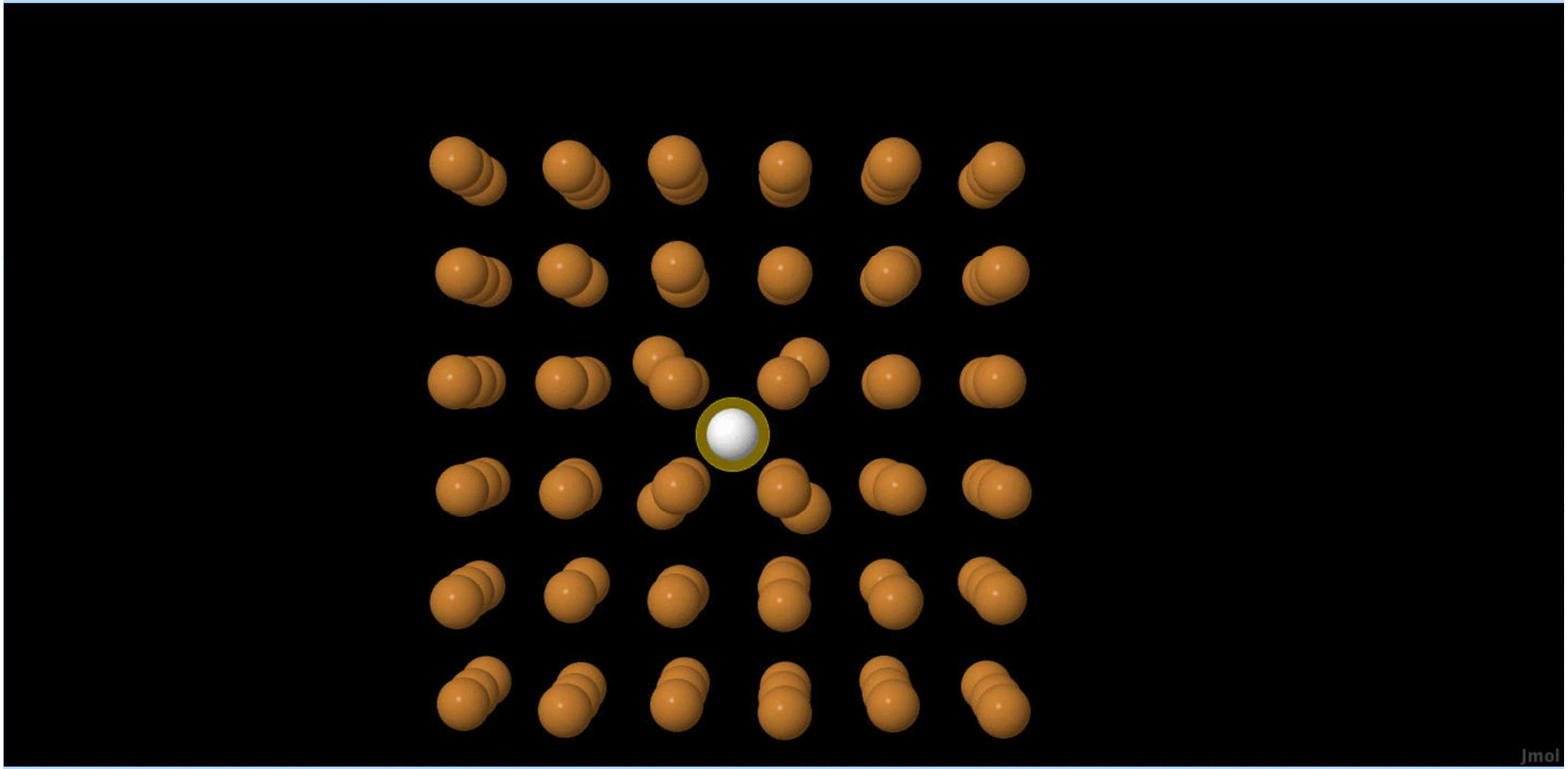
Migración desde hueco tetra a octa (Cu)



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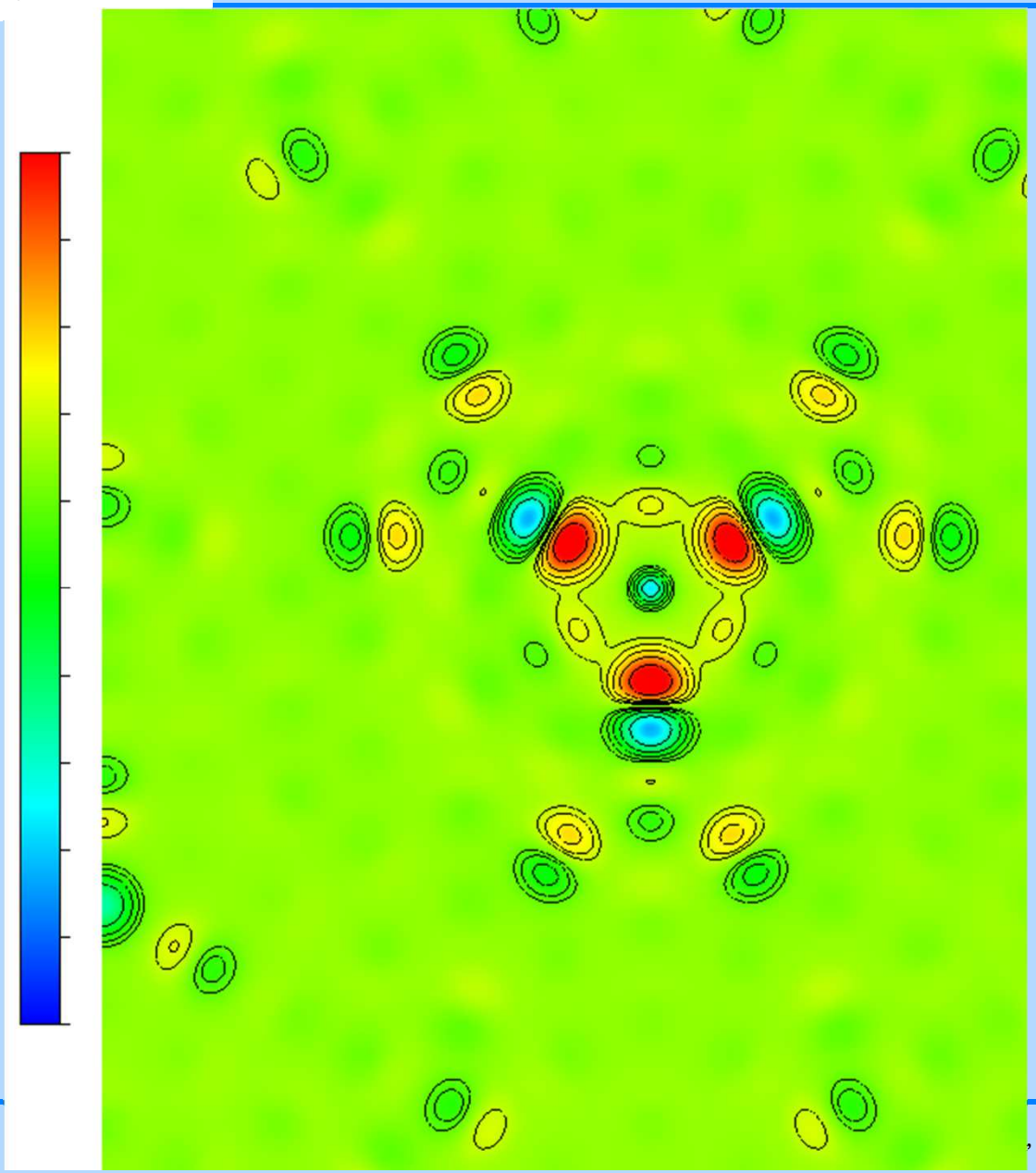
PbHe tetra charge density



Fraile 2012

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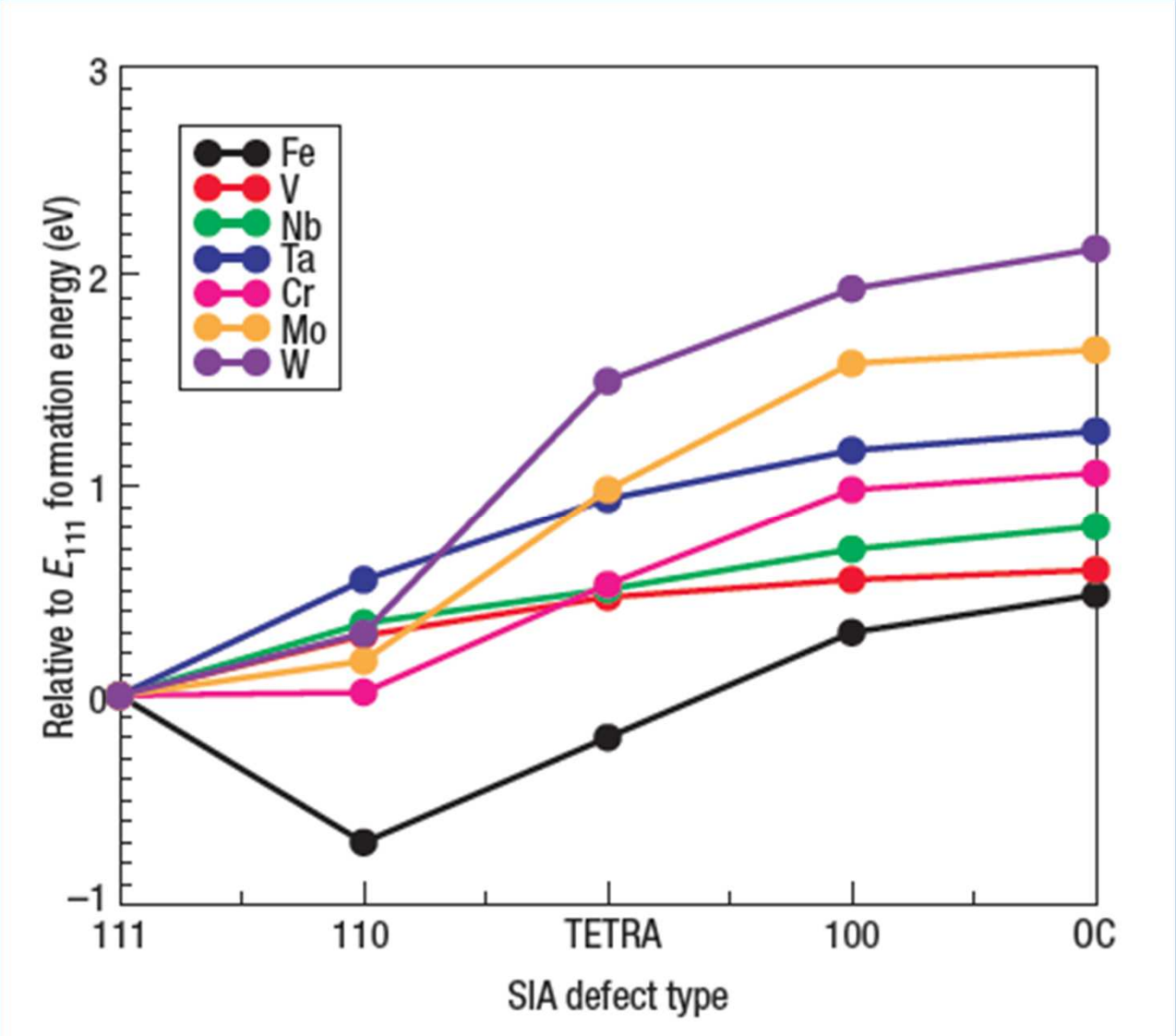
SIA E_f in bcc metals by DFT: role of magnetism



Nguyen-Manh 2006

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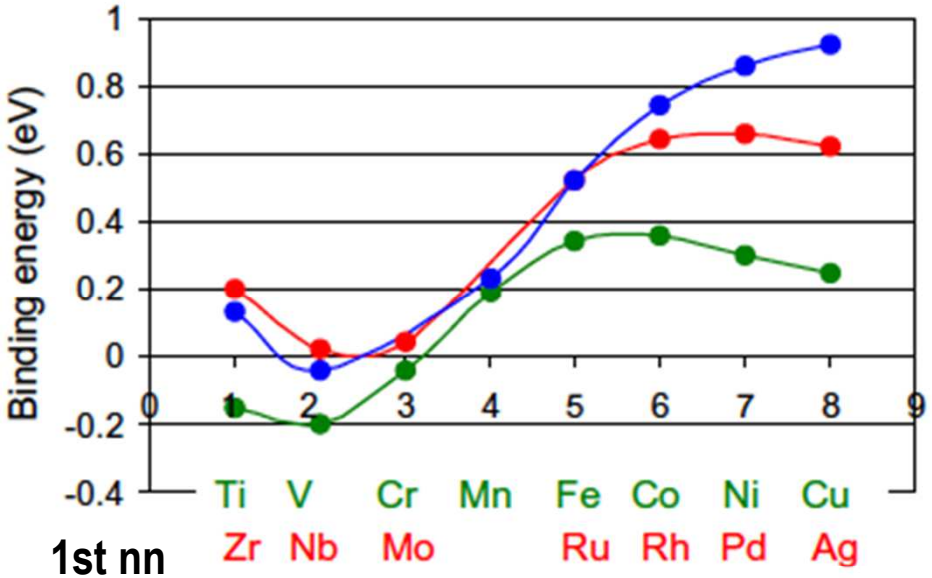
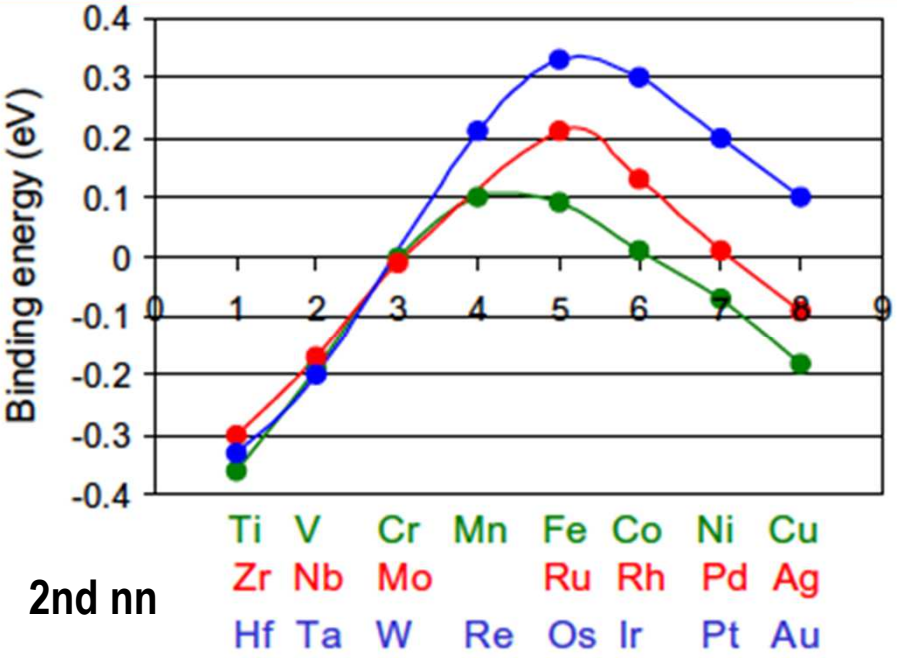
Solute PD interactions in W

Becquart 2012

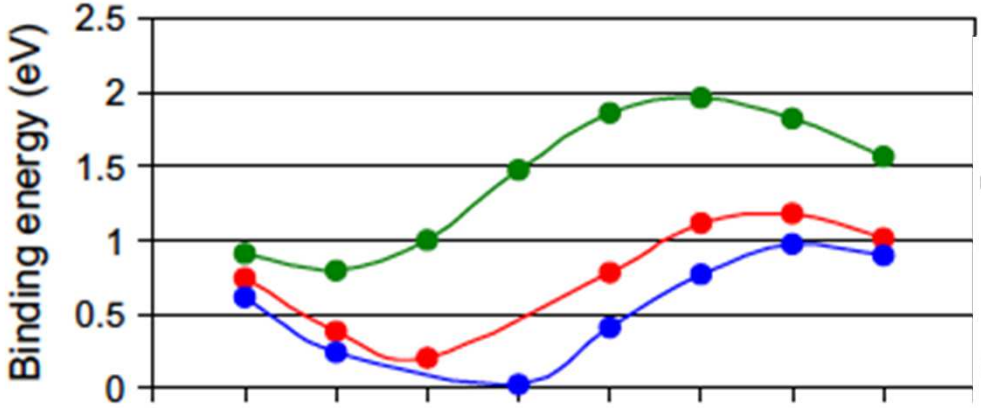
Solute ats-V



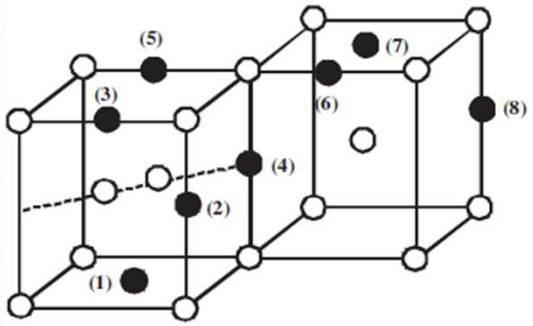
1st nn



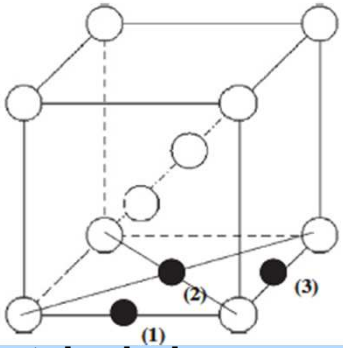
2nd nn



Subs. TM ats-He



8 octahedral around <110> dumbbell



3 octahedral around <111> crowdion

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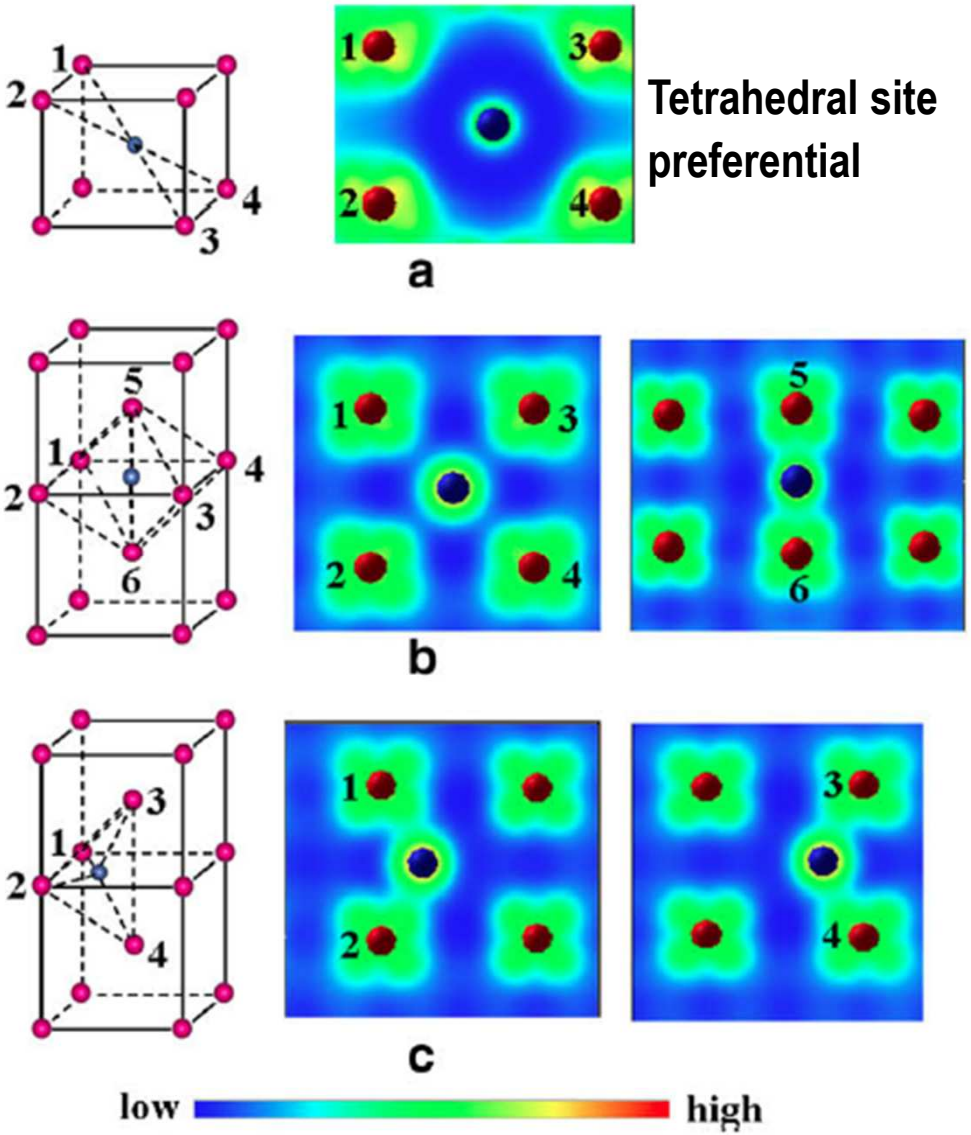
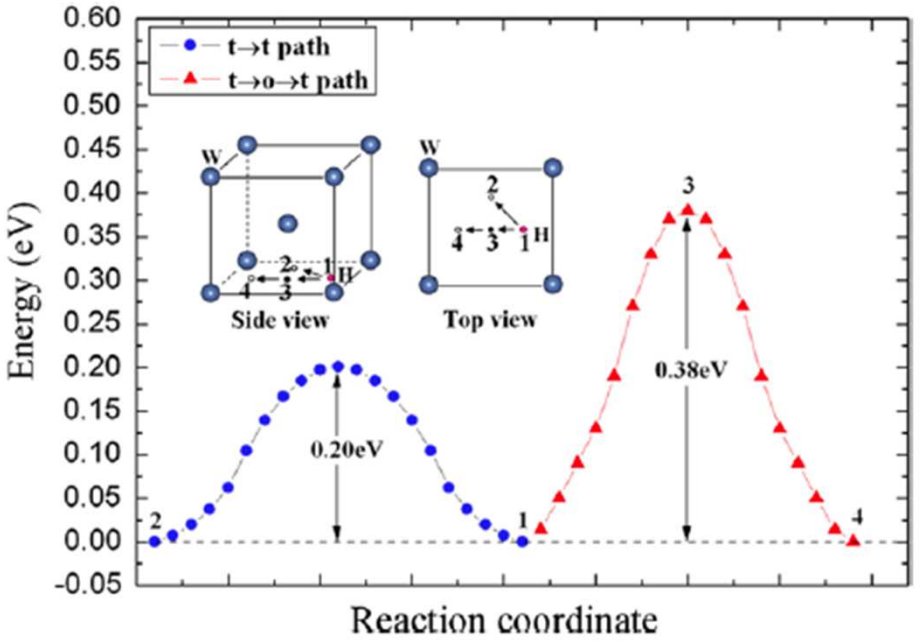
H stability and diffusion in W



Tetrahedral site preferential



Diffusion energy profile and paths

Liu 2009

Charge density maps

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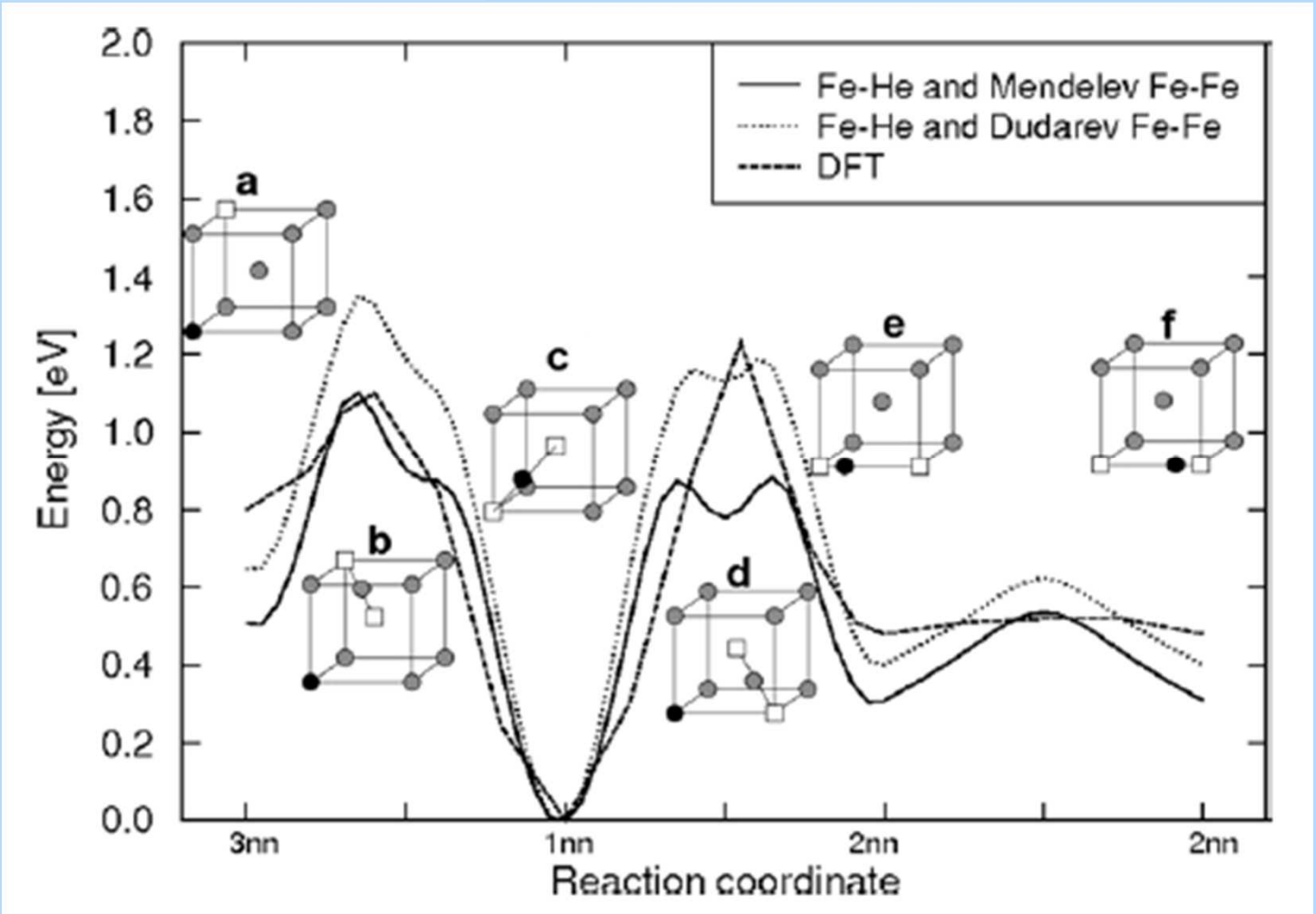
Energy landscapes



Juslin 2008

HeV_2 complexes in Fe: MD with a new Fe-He pair potential

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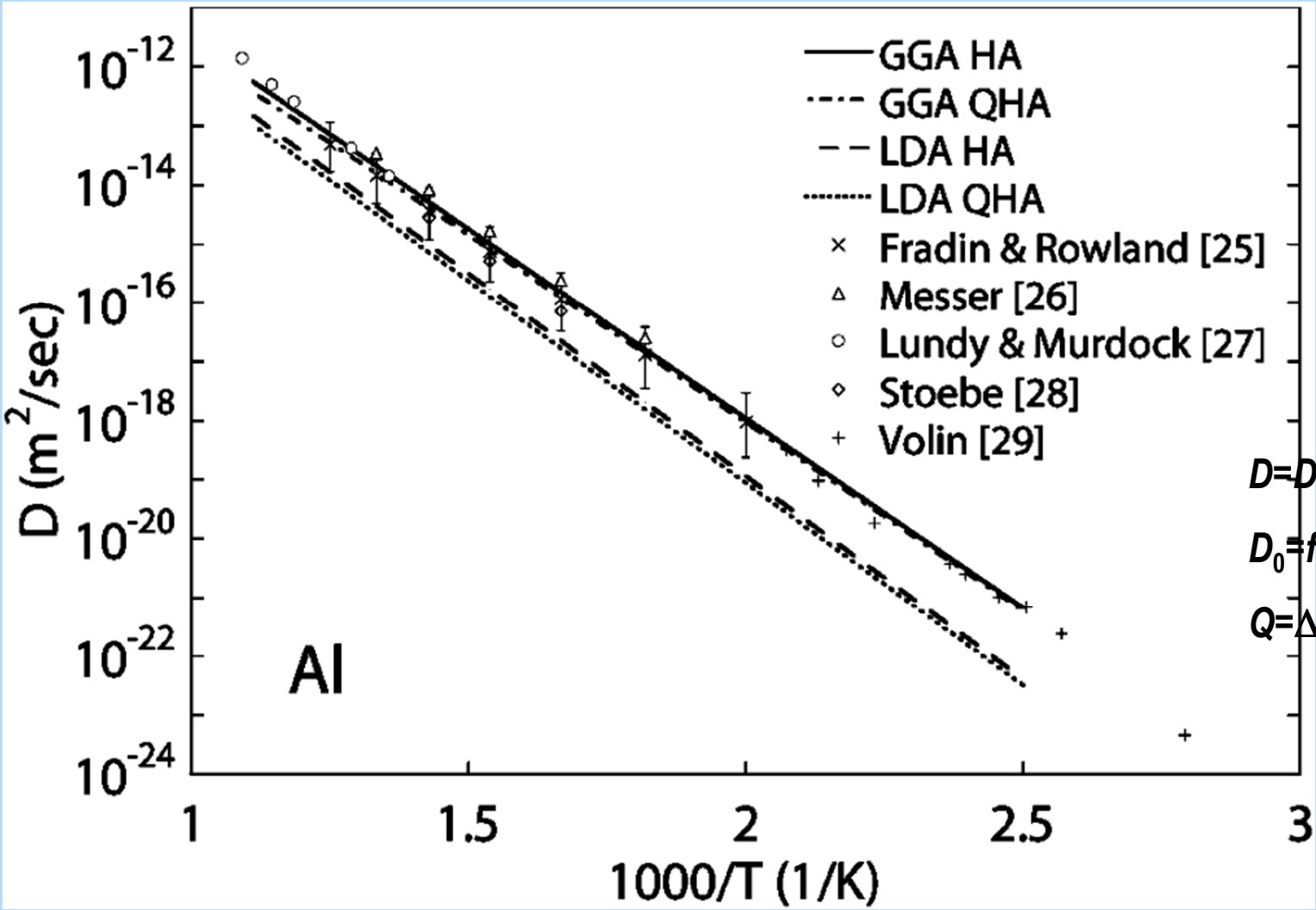
Self-diffusion coefficients



$D=D_0\exp(-Q/k_BT)$

$D_0=fa_0^2\nu^*\exp(\Delta S_f^{vib}/k_B)$

$Q=\Delta H_f+\Delta H_m$

Mantina 2008

Arrhenius plot for Al: activation energy and diffusion prefactor using VASP

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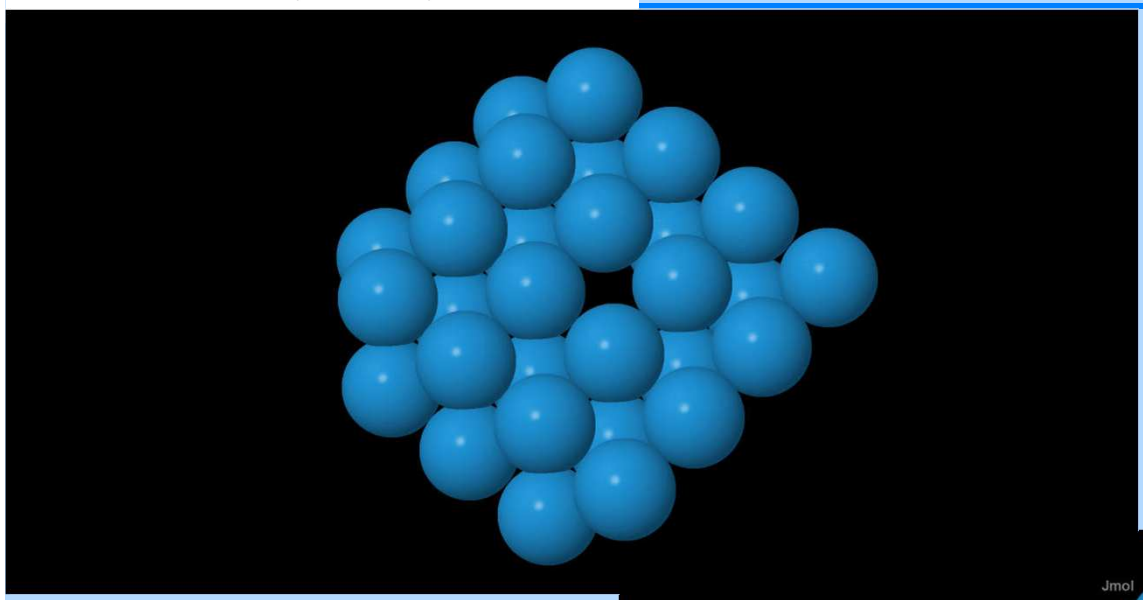
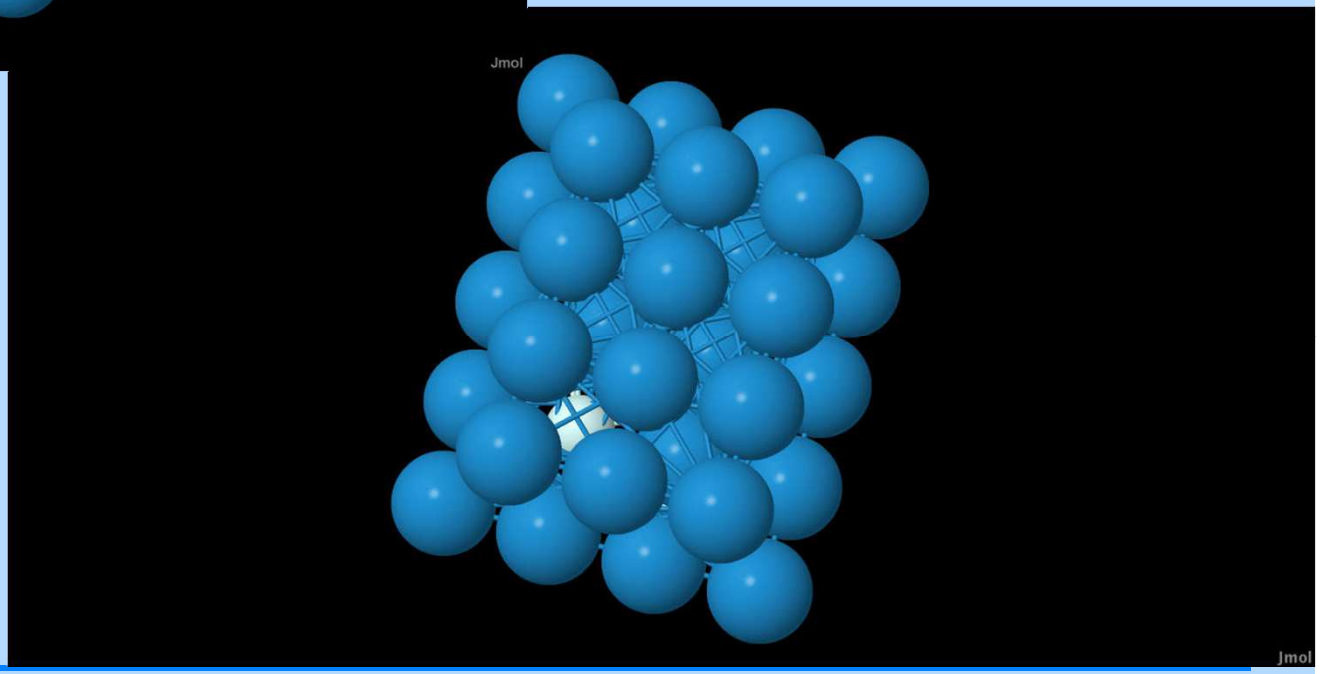
Surfaces with vacancies and defects





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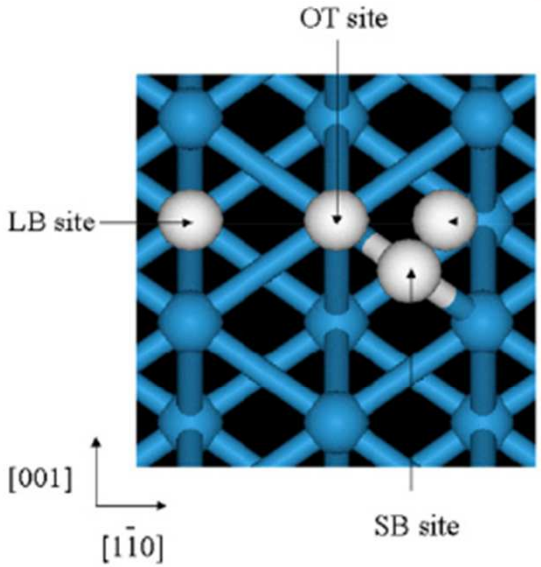
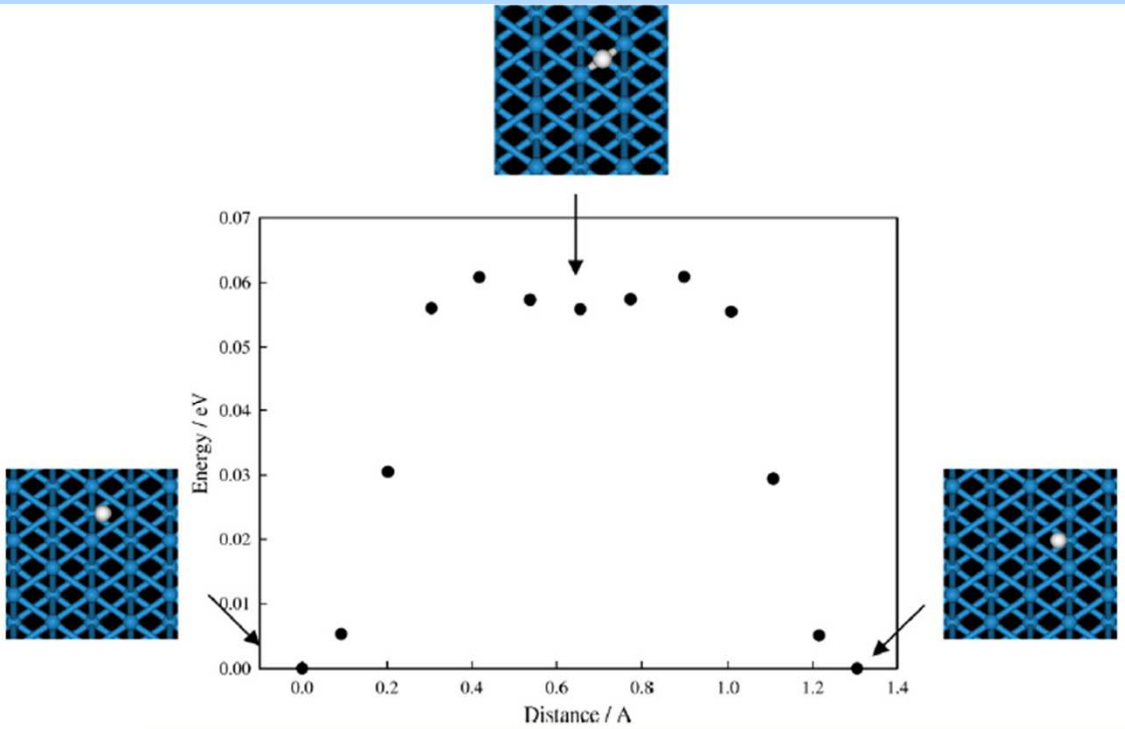
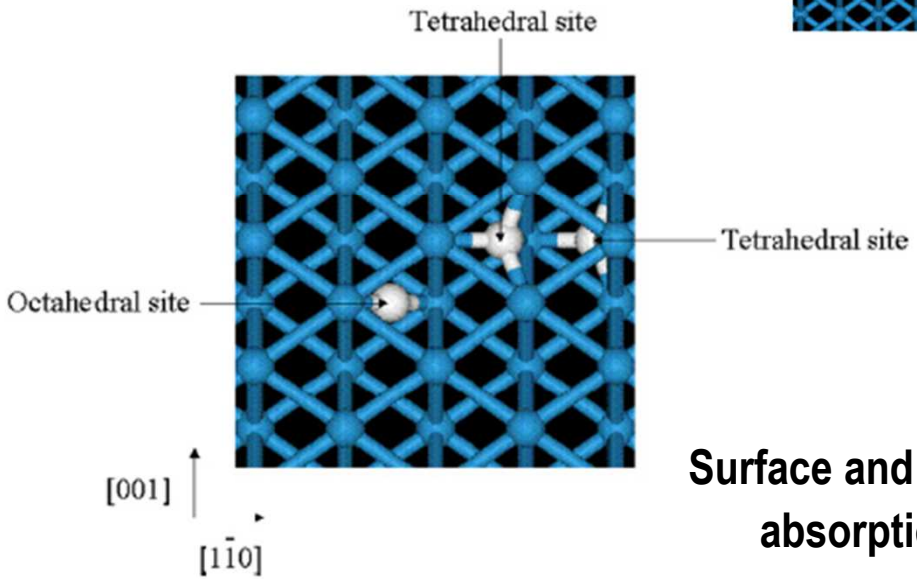
H on W (110) surface







H diffusion potential energy curve on W (110) (1x1)-H surface

Nojima 2007

Surface and subsurface absorption sites

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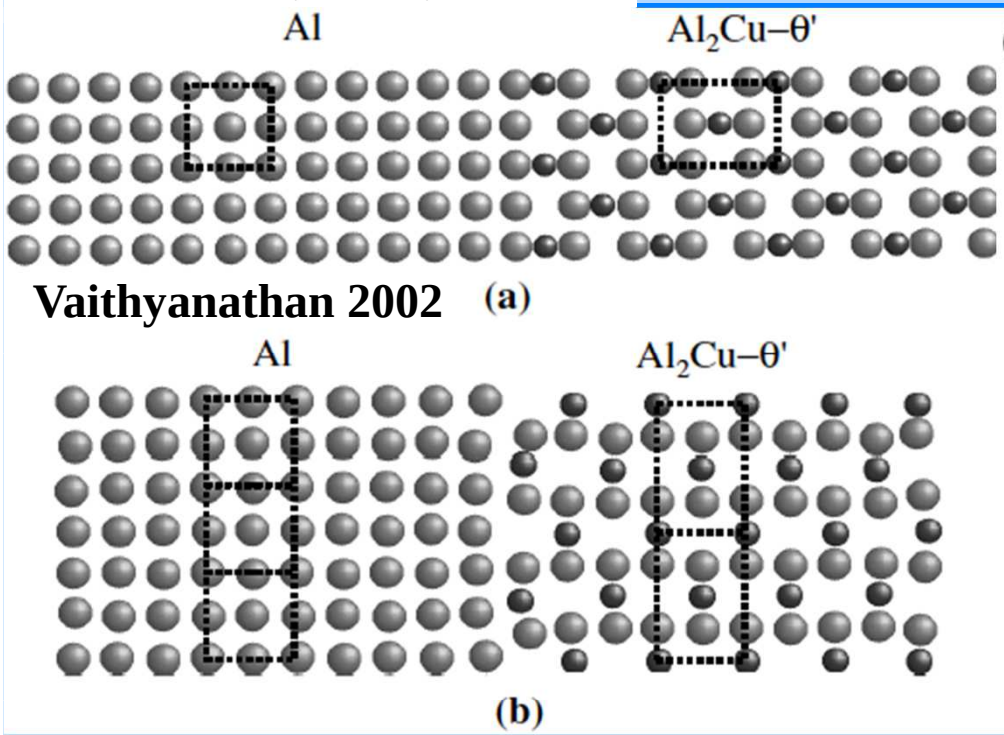
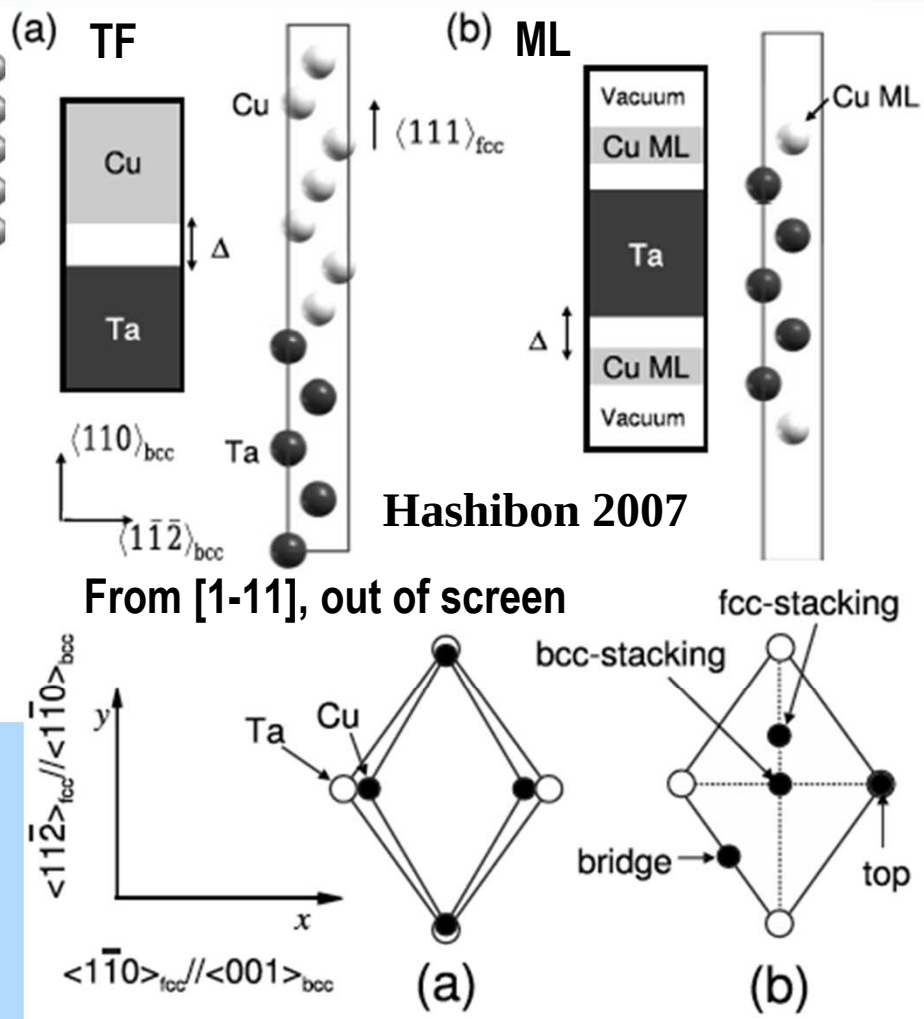
IFs with DFT



a) Coherent (100) IF

b) Semicoherent (001) IF



Al_2Cu solid solution/Thin Cu film on Ta/ β''-Mg_5Si_6/α-Al

Incommensurate: one on top of each other

Commensurate cells: strained Cu on unstrained Ta

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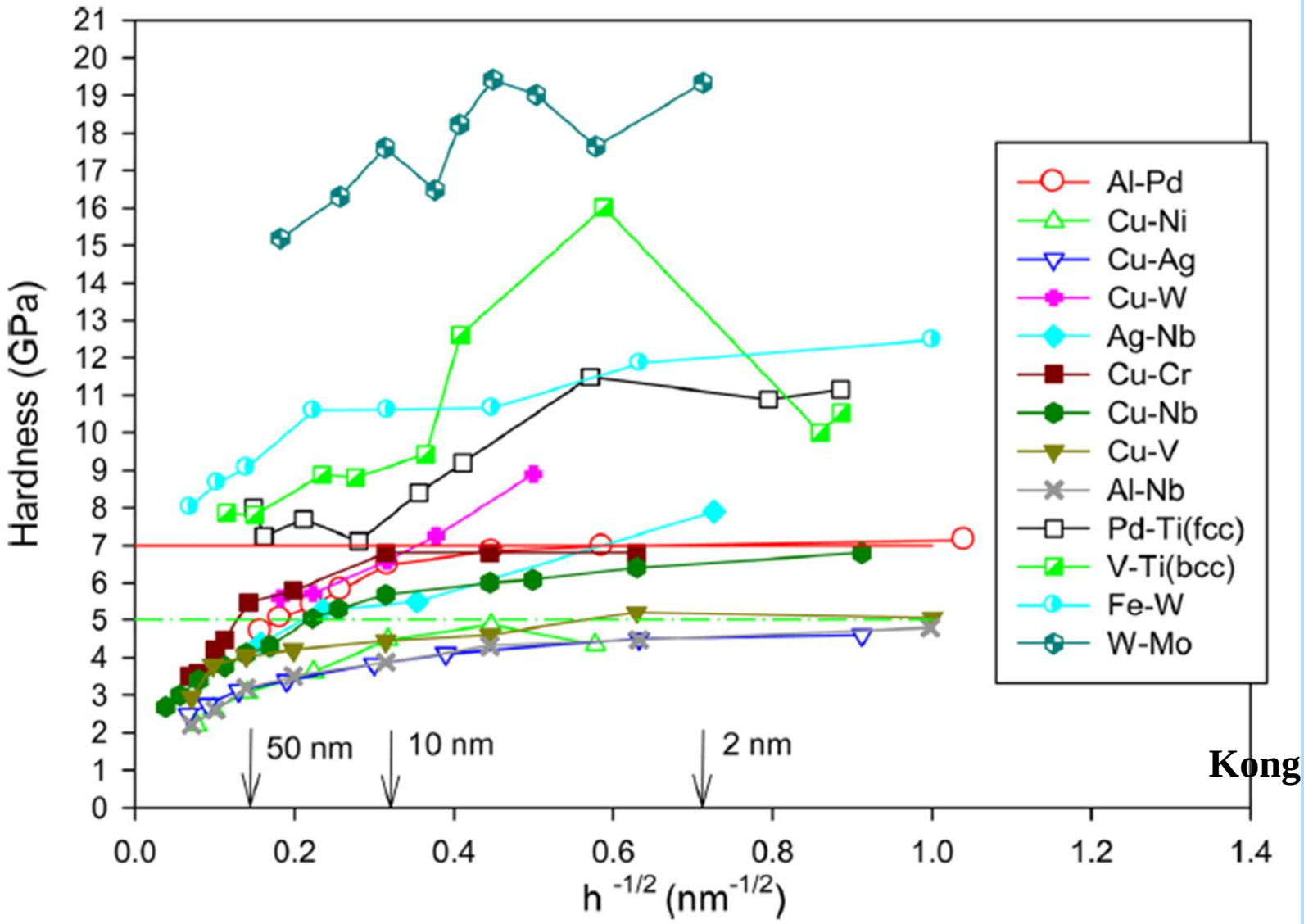
Strengthening in MLs with $\Delta H_f < 0$



Kong 2011

Symbols: open, fcc/fcc; solid, fcc/bcc; half-filled, bcc/bcc

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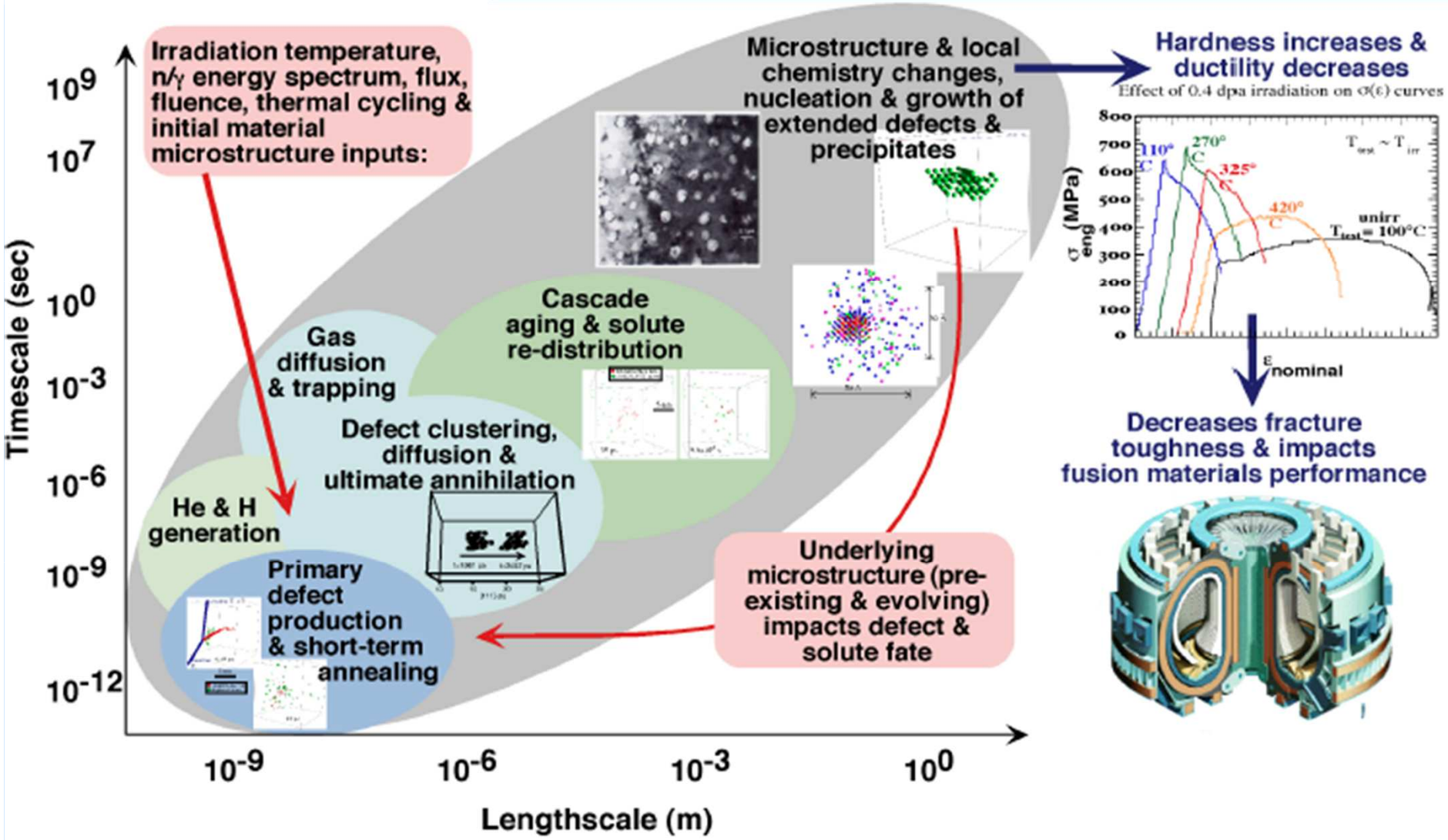
Procesos de daño por radiación



Cortesía de B. Wirth

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? Darío Sergio Ángeles Roberto César