Numerical modeling of radiation effects in solids: principal features, limitations and perspectives

Pascale Geysermans
CNRS – ICMPE, 15, rue Georges Urbain
94407 Vitry-sur-Seine cedex, France

Pascale.Geysermans@glvt-cnrs.fr

What is Multiscale ?

microscopic constituents of matter = atoms (nanometers/femtoseconds)
---> they determine the behavior of the material at the macroscopic scale (centimeters/seconds)

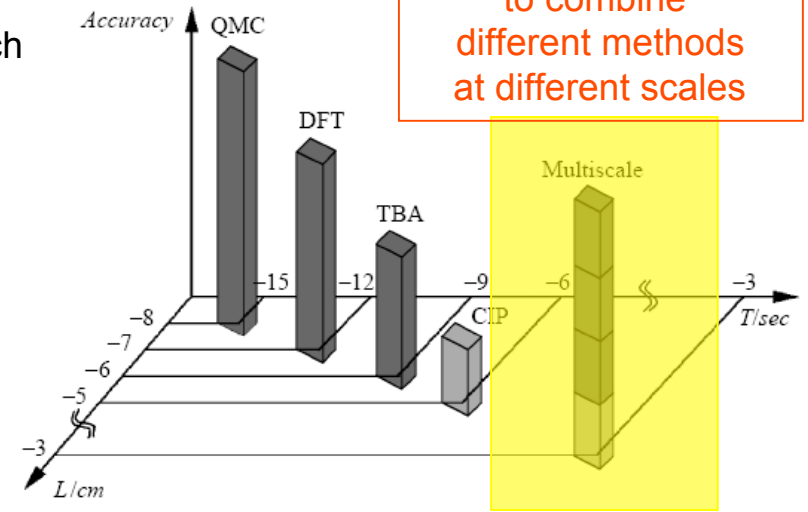
* Atomic scale (~10^{-9}m,10^{-15}-10^{-12}s), electrons, quantum-mechanical methods
  quantum Monte Carlo and quantun chemistry : ~ 10 - 100 atoms
  DFT : 100-1000 atoms : static properties

* Microscopic scale (~10^{-6}m,10^{-12}-10^{-9}s), atoms, classical interatomic potentials
  Molecular Dynamics, Atomic Monte Carlo

* Mesoscopic scale (~10^{-4}m,10^{-9}-10^{-6}s), lattice defects such as dislocations, grain boundaries, phenomenological theories
  Larger-scaled entities, Dislocation Dynamics

* Macroscopic scale (~10^{-2}m,>s), continuum medium
  Finite Elements (elastic continuum)

Multiscale approach : to combine different methods at different scales

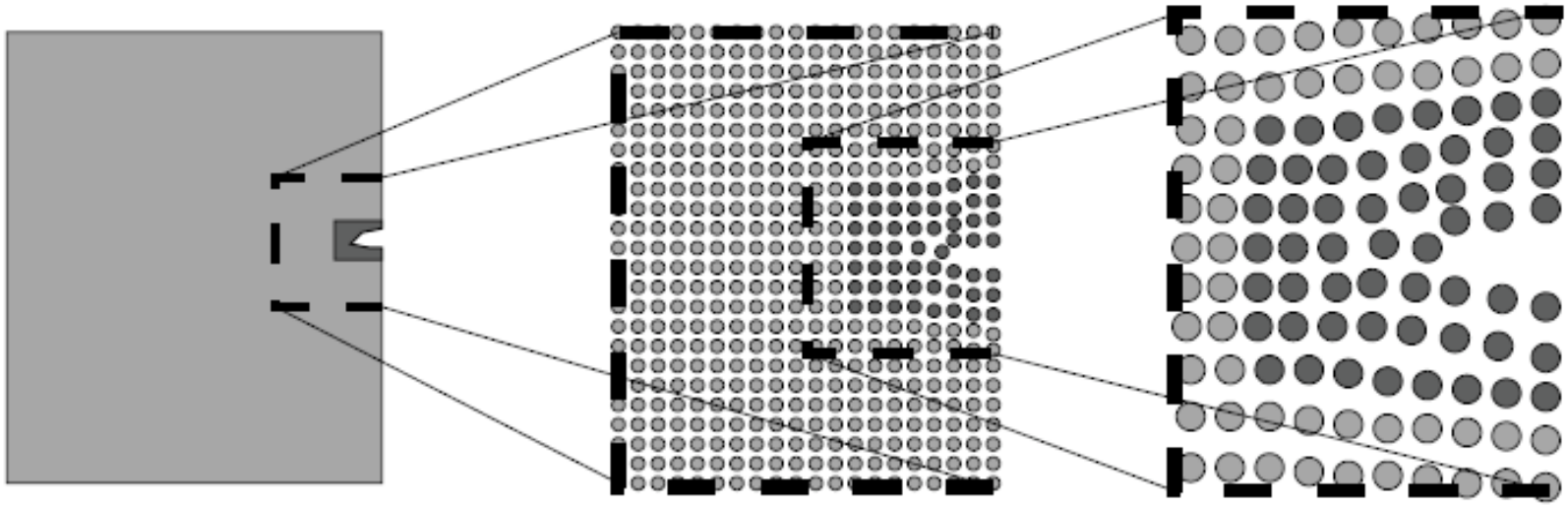


Kaxiras, *Handbook of Theoretical and Computational* ..., vol **X**, (2005) 1-33

sequential method : information obtained from more detailed scale calculations are input into the larger scale simulation

concurrent method : different scales are concurrently considered and communicate with a type of hand-shaking procedure, system partitioned into domains

Schematic view of geometrical decomposition in a concurrent multiscale simulationof a slab with small crack



macroscopic scale:
modeled as a continuum :
Finite elements

atomic scale:
atomistic simulations
using empirical potentials
molecular dynamics

microscopic scale:
forces derived from quantum
mechanical calculations

PLAN

Introduction to molecular dynamics and its limitations

Atomistic simulations of displacements cascades
* positive contributions
* limitations

Quantum mechanical calculations : DFT
* how to complete metals description provided by classical empirical potentials

Sequential multiscale modeling of the resistivity recovering experimental results
* coupling of ab initio results as input data in an event-based Monte Carlo simulation

Molecular dynamics : an avantageous description of dislocation core

Concurrent multiscale modelling of a dislocation without and with impurities within its core

Conclusions

Molecular Dynamics : Atomistic simulations

Time evolution of a set of N interacting classical particles

by integration of Newton's equation of motion (deterministic technique)

$$m_i \frac{d^2 \vec{r}_i}{dt^2} = \vec{F}_i$$

Classical approximation is valid when

$$\Lambda = \sqrt{\frac{2\pi\hbar^2}{mk_B T}} << a$$

(very light elements Li, Ar or
very low temperature < T_θ drop in the specific heat,
anomalous expansion coefficient)

→ defects in crystals, surfaces and interfaces, ...

Statistical mechanics method

set of configurations distributed according to some statistical ensemble

quantities are obtained as averages of the various instantaneous quantities

microscopic behavior ⟷ equilibrium thermodynamics
non-equilibrium processes

Forces ⟷ **Potential Models**

Simulation is realistic if it mimics the behavior of the real system

$$\vec{F}_i = -\nabla V(\vec{r}_1,\ldots\vec{r}_N)$$

gradient of a potential energy function depending on the positions of the particles

$$V = \sum_{i=1}^{N}\sum_{j>i}^{N} U(r_{ij}) \qquad \vec{F}_i = \sum_{j>i}^{N} \vec{F}_{ij}$$

Pair potential approximation :

no adequate description of many-body effects in metals

(E_c/T_m; E_v/E_c, C_{11}/C_{44})

$$V = \sum_{i=1}^{N}\sum_{j=i+1}^{N}\varphi(r_{ij}) + \sum_{i=1}^{N}\phi(\rho_i) \quad , \quad \text{with } \rho_i = \sum_j \psi(r_{ij})$$

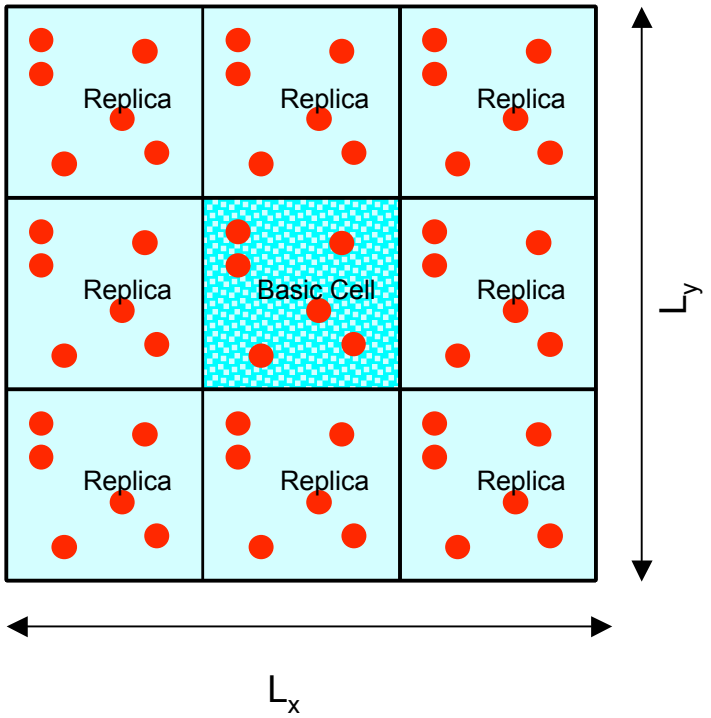
superposition of contributions from neighboring atoms

repulsive pairwise term of the Born-Mayer type

$$\phi(\rho_i) = -\sqrt{\sum_{j=1}^{N}\theta(r_{ij})}$$

Finnis-Sinclair scheme

Parameters fitted on experimental quantities :
cohesive energy, elastic constants, defect formation energy



Periodic Boundary Conditions

infinite system
no surface effect

minimum image criterion : at most one among all pairs will interact
short-range of the common potentials : interaction range < L/2

surfaces : PBC are removed --> slab

!! long range defect (dislocation)

Time / Size limitations of Molecular Dynamics

$N \approx 10^3 - 10^6$ atoms

$t \approx 10^{-12} - 10^{-7}$ s

Size limitation

Mean square displacements

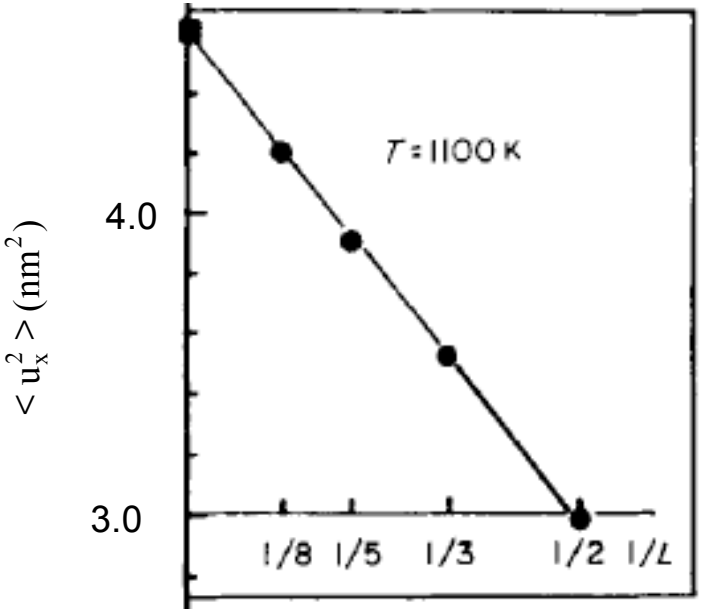
finite temperature T≠0K

≠ system sizes : 32<N<4000

harmonic approximation
classic statistical limit

$$\left\langle u_x^2 \right\rangle = \frac{kT}{m}\int_0^{\omega_{max}} \frac{g(\omega)}{\omega^2} d\omega$$

g(ω)is the frequency spectrum



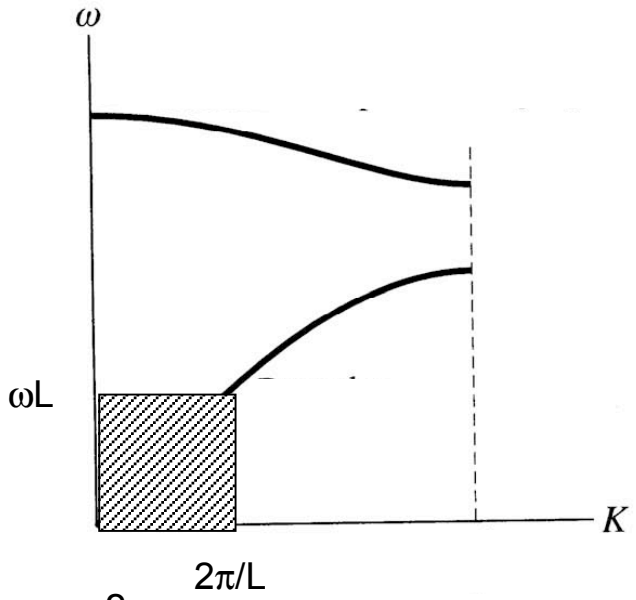
Cubic periodic system of size L

→ minimum k value of 2π/L, cut-off frequency ω_L (proportionnal to 2π/L)

$$\left\langle u_x^2 \right\rangle_L = \frac{kT}{m}\int_{\omega_L}^{\omega_{max}} \frac{g_L(\omega)}{\omega^2} d\omega$$

...

$$\left\langle u_x^2 \right\rangle_L = \left\langle u_x^2 \right\rangle_{L_0} + kT \propto \left(\frac{1}{L_0} - \frac{1}{L}\right)$$



4000 atoms ≡ equilibrium quantities but non-equilibrium processes ?

DISPLACEMENT CASCADES

Development of models of damage → understanding of elemental processes

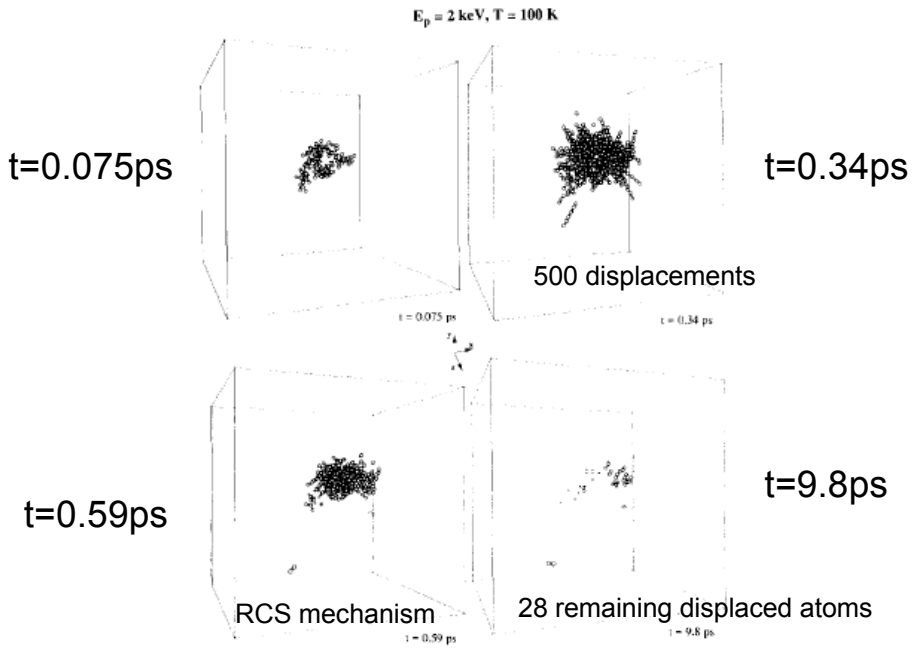
damage < elastic collision : energetic primary knock-on atoms

→ cascade of displacements ~ 10 ps, 10 nm

→ interstitials, vacancies

→ MD : well-adapted method

Formation and relaxation of a 2keV cascade in a lattice at T=100K



A.F. Calder and D.J. Bacon, J. Nucl. Mater., **207** (1993) 25

* primary knock-on atom (PKA) > 1keV
(kinetic energy transfered to one atom, along a defined direction ≡ simulate the elastic collision)
avoid any chanelling

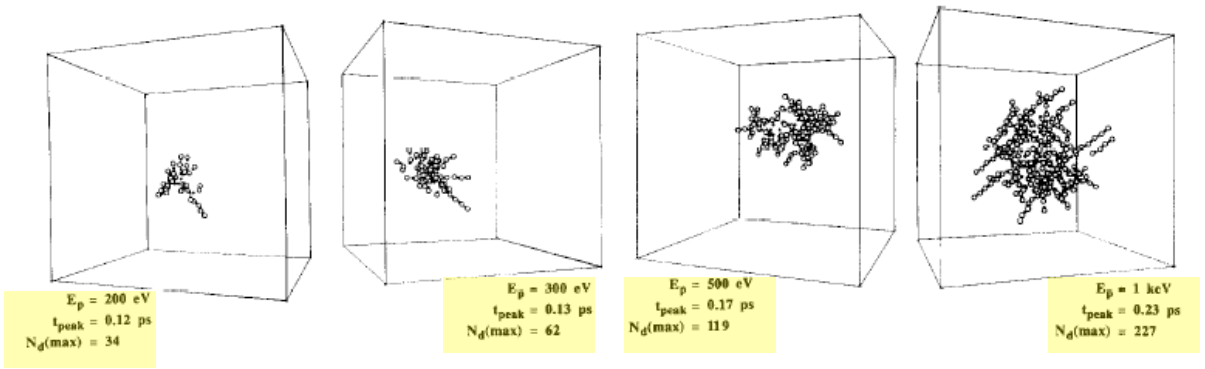
*high density of displaced atoms
in a spacially localized region > predictions by the binary collision approximation

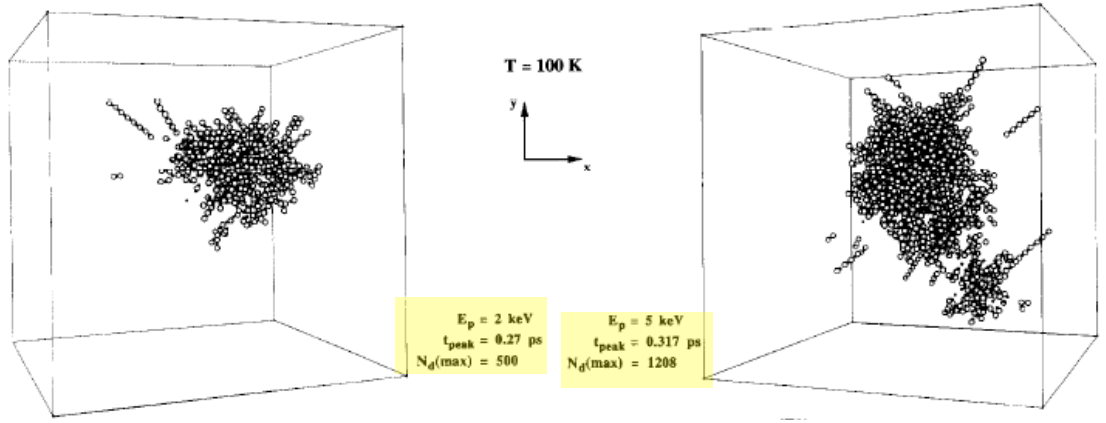
* most of them recombine with vacant sites during the cooling phase
* ejection and replacement collision sequences by interstitials (RCS)
---> vacancy-rich core and interstitial-rich periphery

spacially segregated defects -->
% of isolated or clustered residual defects

size of the displacement cascade increases with increasing PKA energy
---> mutiple well-defined subcascades

low density





high density

subcascades

after chanelling

→

increase of the residual defects
(decrease of replacements)

t_{peak} : time of maximum number displaced atoms depends on the energy PKA

→ dispersed results

Displacement energy threshold to introduce in BCA (energy required to displace an atom, T, PKA energy)

Evidence that the number of free (vacancy and interstitial) defects
that survive the thermal spike is much smaller than the BCA value
These values are included in chemical rate theory models of radiation effects

Evidence of chanelling

limited size of the system ---> interaction across the periodic boundary --> limited to "small PKA energy"

microcanonical statistical ensemble (NVE) --> increasing of the temperature
uniform algorithm are not adapted

no thermal conductivity by electrons is introduced (electron-phonon coupling)

lack of statistics : only a few successives cascades are realized (<10)

alloy (some recent works on Fe-Cr)

difficulty to precisely caracterize the nature and the position of defects

Stability of interstitials in α-Fe : magnetism

irradiation : production of atomic defects : self-interstitials, vacancies

migration energy unusually large : 0.30 eV (< 0.1 eV other metals)
discrepancies between experiments and classical potential

ab initio calculations

DFT : quantum mechanical atomistic calculations.
transforms the complex many-body problem
of interacting electrons and nuclei into a coupled set
of one-particle (Kohn-Sham) equations, which are
more manageable.

Parameter free calculations of all ground state
physical observables (charge and spin densities,
total energy and related quantities ...), no dynamics

different implementations :
plane-wave basis functions,
pseudopotentials for valence electrons
linearized methods for all-electron calculations, ...

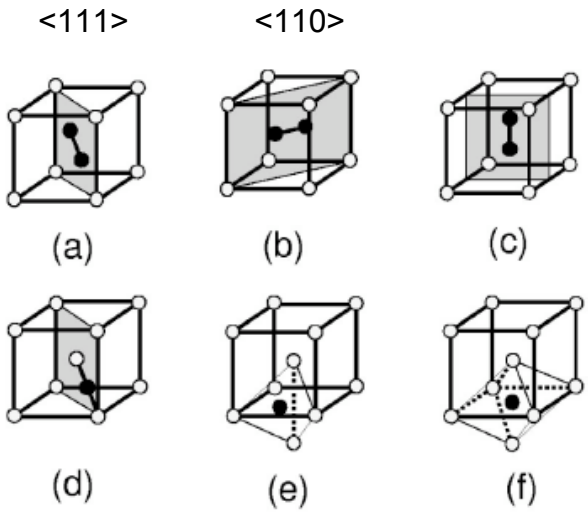
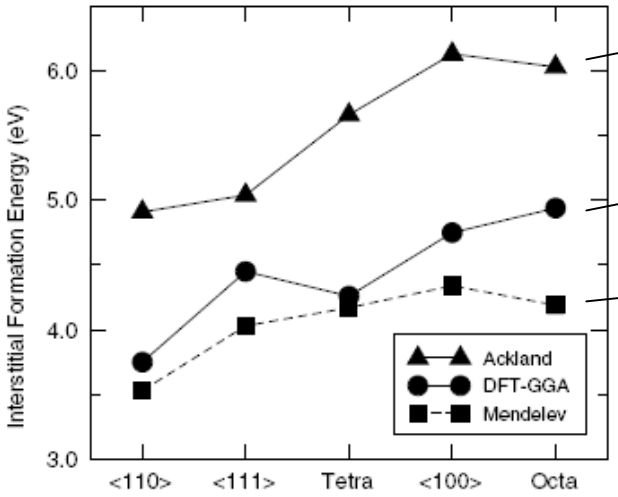


FIG. 1. Schematic pictures of interstitials studied here: (a) ⟨111⟩ dumbbell, (b) ⟨110⟩ dumbbell, (c) ⟨100⟩ dumbbell, (d) crowdion, (e) tetrahedral, and (f) octahedral.

First-principles molecular dynamics :
* electronic total energy & forces at each time step
* minimization of the electronic total energy
cpu consumming,
limited to rather small system sizes and short time sequences

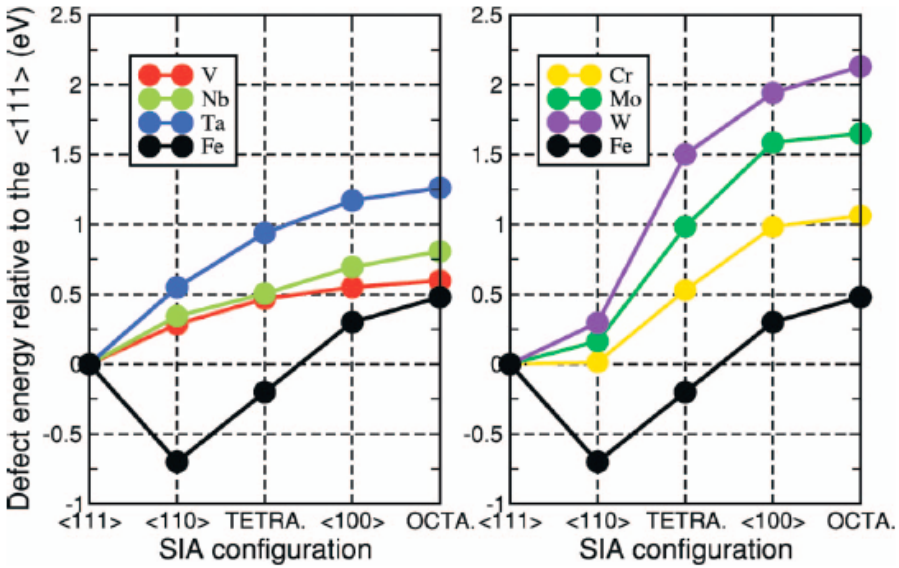


FS and MEAM widely used in MD simulations of displacement cascades

agreement with experiments (migration energy)

fit parameters to formations energies of <110>, <111>,<100>
significant discrepancies for configurations which where not include into the fit

C. Fu, F. Willaime, PRL, **92** (2004) 175503



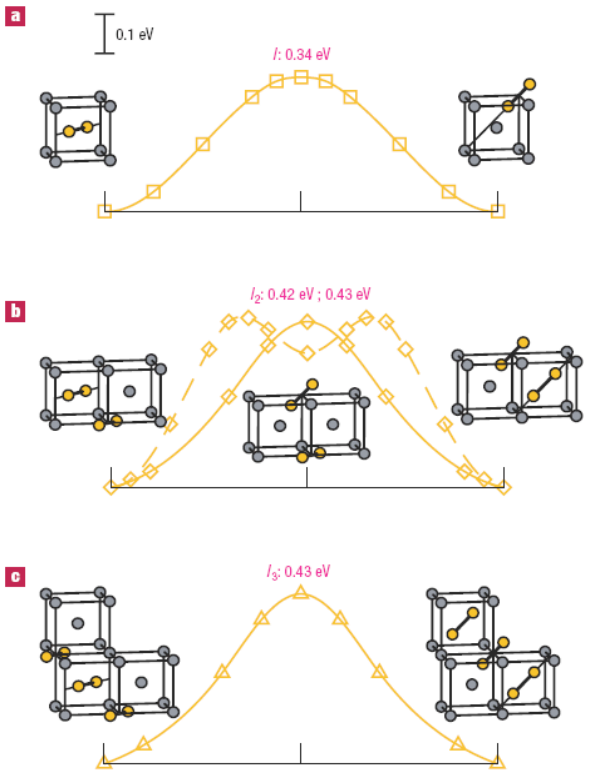
ab initio calculations of
local magnetic moments
local antiferromagnetic

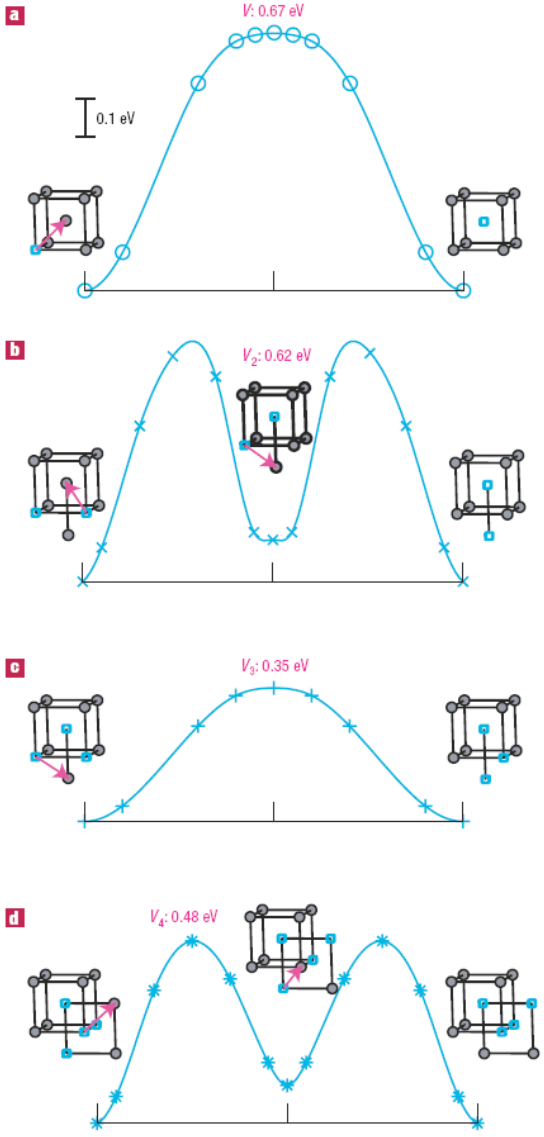
bulk value : 2.3 μ_B

$0.7 < \mu_B < -0.9\ \mu_B$

D.Nguyen-Manh, A.P.Horsfield and L. Dudarev, PRB **73** (2006) 020101

Migration energy of interstitial-type or vacany-type defects
I, I_2, I_3, V, V_2, V_3, V_4





Kinetic Monte Carlo simulations

---> multiscale moddeling approach of the resistivity recovery experiments

C. Fu, et al. , nature mater., **4** (2005) 68

Multiscale Modeling

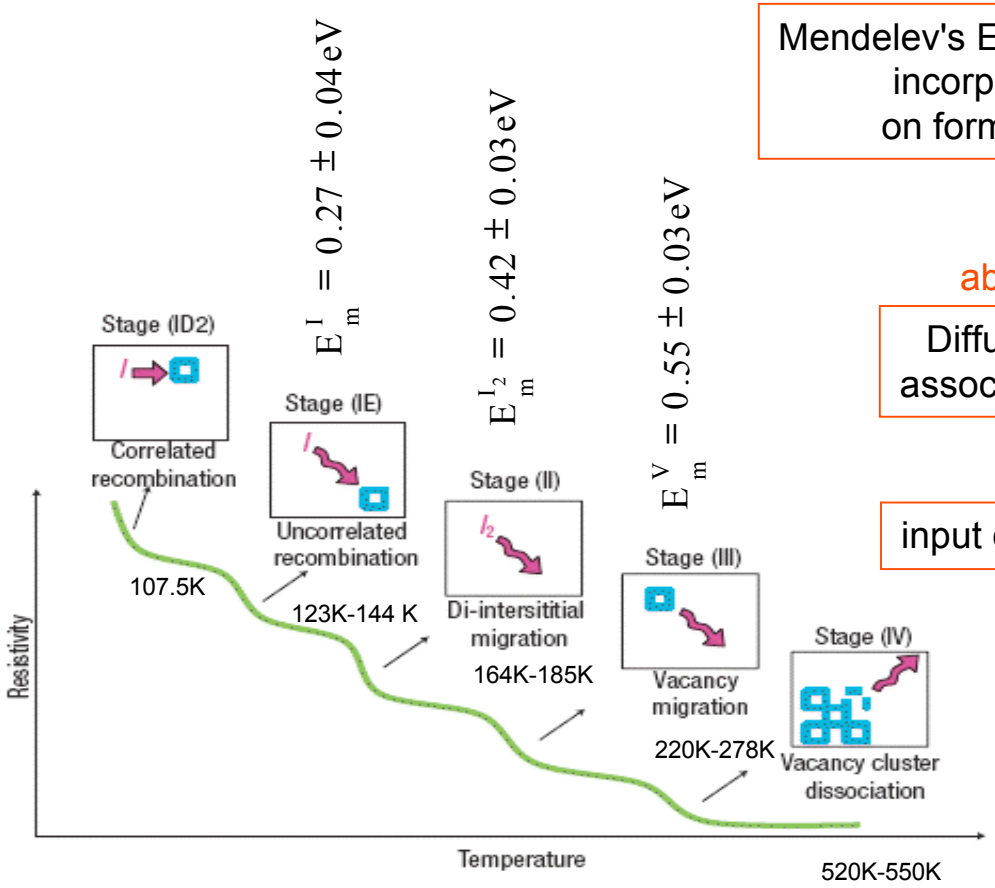
EAM potentials : difficulties to reproduce different stages because of bad predictions for activation energies
$E_m^{I2} < E_m^{I}$; E_m^{V} too large

Mendelev's EAM potential is better for stage IE : incorporating fitting of parameters on formation energies of interstitials

ab initio calculations

Diffusion mechanisms and associated activation energies

input data in KMC simulations



Kinetic Monte Carlo

different defects ≡ **OBJECTS** without any atomic details

the atomic transport is reduced to single diffusion events
corresponding to jumps of the mobile species

↓

continuous coordinates in the simulation cell
nature (number of self-interstitials, vacancy, impurity atoms, ...)
shape (cluster , loop)
mobility → diffusion and interaction together
dissociation rate if necessary → eventually annihilate or aggregate with other defects

Primary defects are introduced in the system rate corresponding to the dose rate

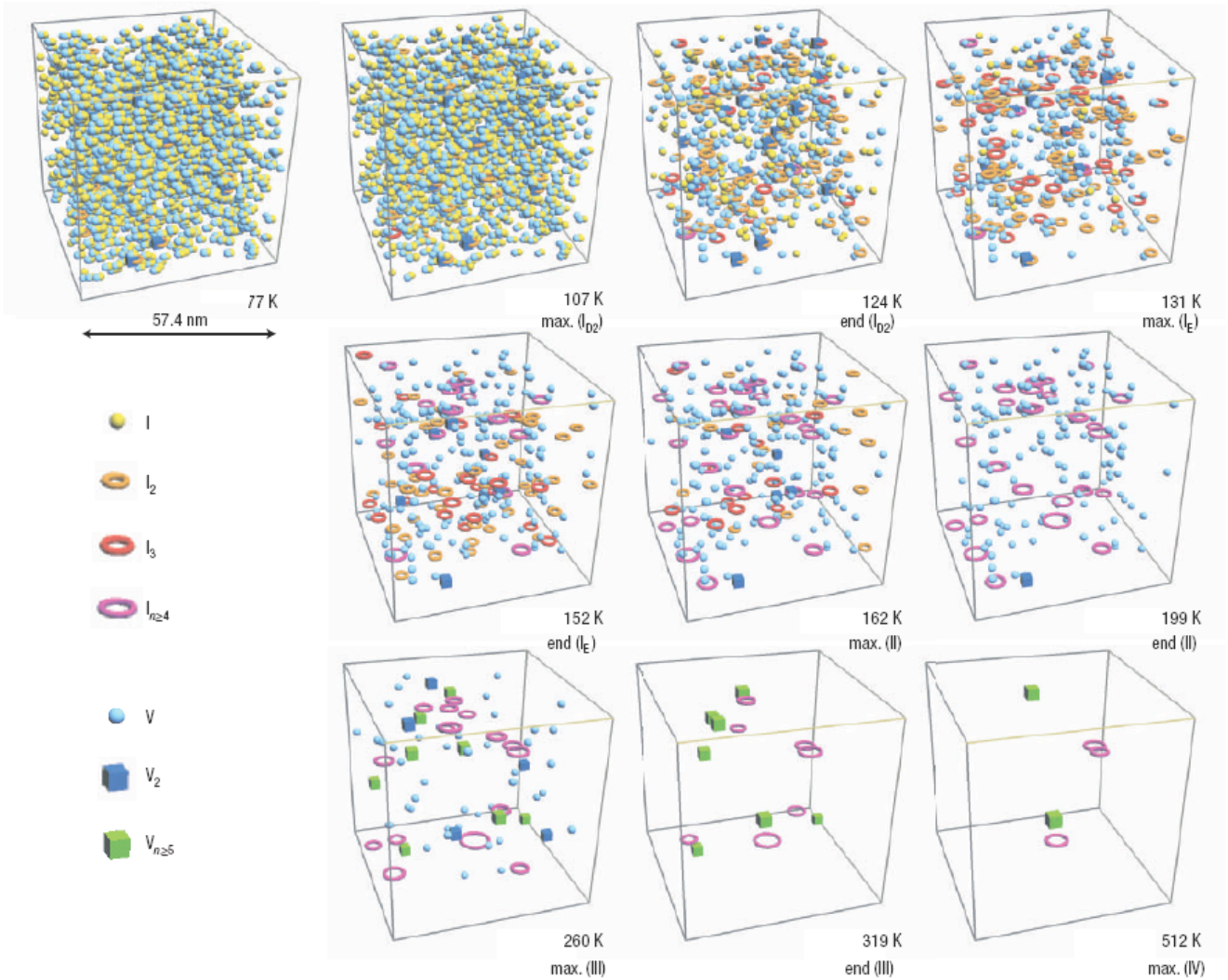
delay of all possible events between objects is computed to sample the probability
distribution of their occurence

initial Frankel pairs concentration : 60 % (< experiments)

Distance between Frenkel pairs : 4 a

d_r recombination for I-V = 3.3 a

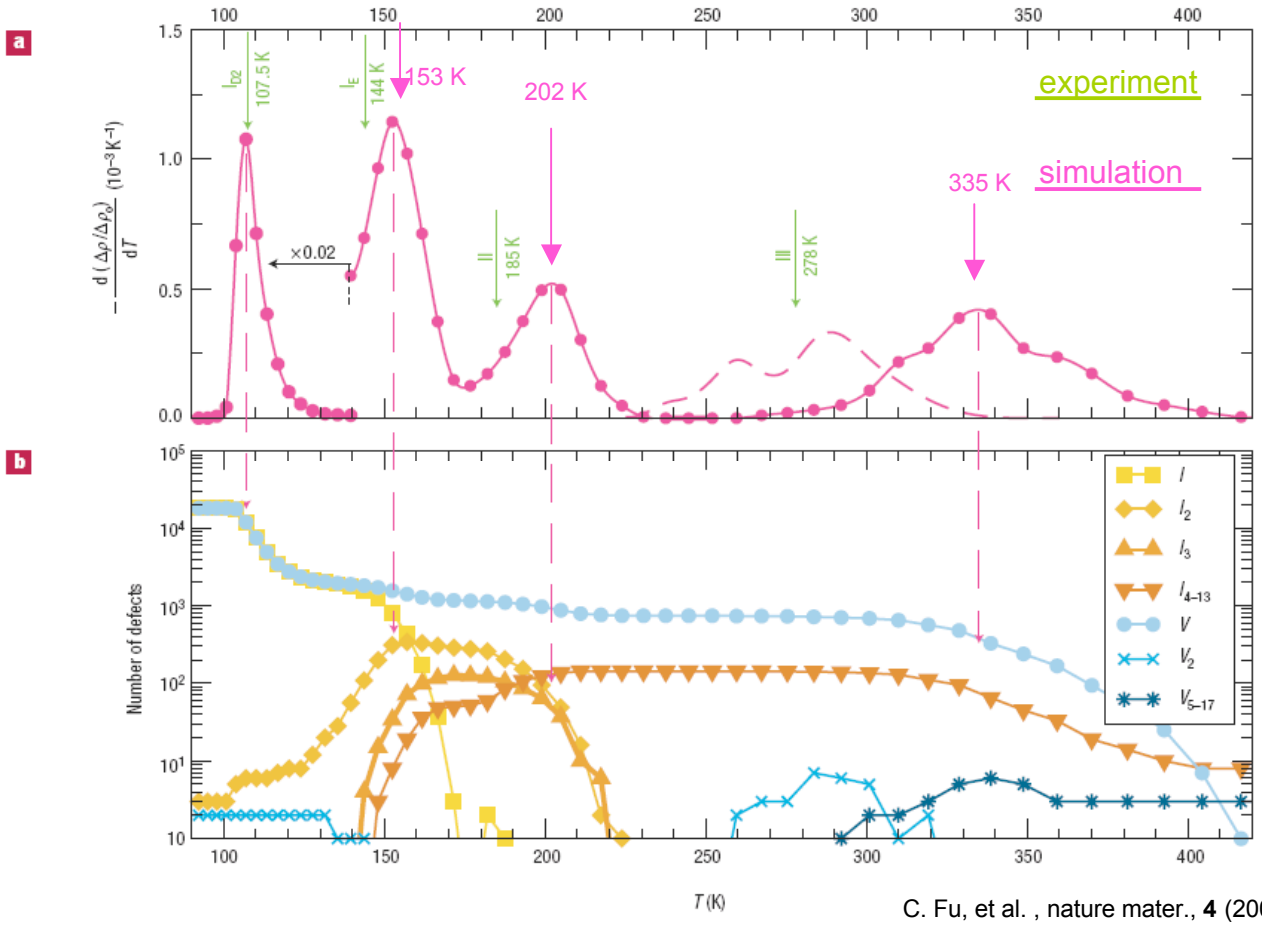
cell size : 300 - 600 nm



C. Fu, et al. , nature mater., **4** (2005) 68

resistivity is proportional to the remaining number of I and V (isolated + cluster)

→ simulated resistivity recovery plot



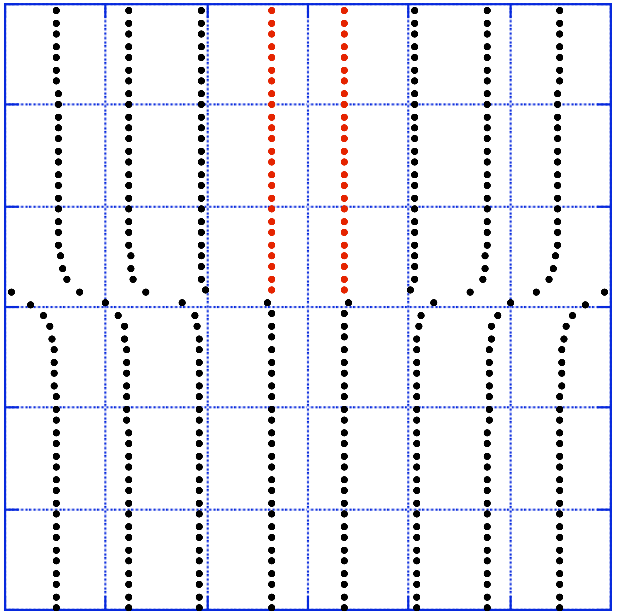
stage II :
migration of I_2 and I_3
various clustering and annihilation
only $I_{n\geq4}$

stage III:
$V_{2\text{-}4}$

C. Fu, et al. , nature mater., **4** (2005) 68

How Atomistic Simulations allows a complete description of the dislocation, in continuity with the elastic theory

Dislocation Core is part of the system such that when a dislocation is introduced in the crystal at position of minimal energy, according to elastic theory, atoms move if an energy minimization calculus is done

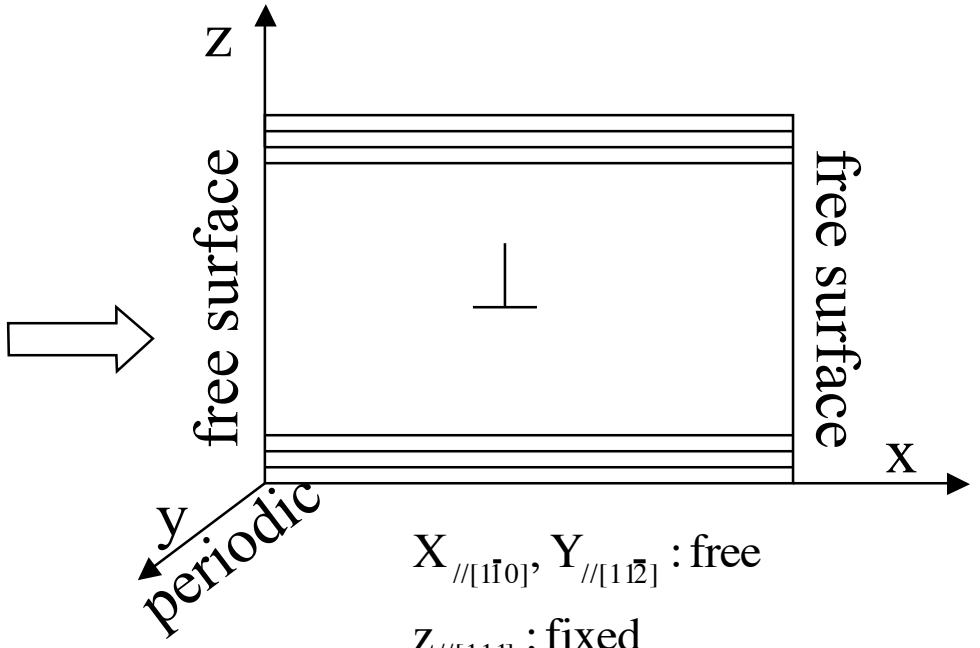


σ_{zz} ⟹

large tilt of [111] planes

edge dislocation --> elastic theory --> displacement field

$$\begin{cases} u_{x\,//[1\bar{1}0]} = \dfrac{b}{2\pi}\left[\text{Arc tan}\,\dfrac{z}{x} + \dfrac{1}{2(1-\nu)}\dfrac{xz}{x^2+z^2}\right] \\ u_{y\,//[11\bar{2}]} = 0 \\ u_{z//[111]} = \dfrac{b}{8\pi(1-\nu)}\left[((1-2\nu)\ln(x^2+z^2) + \dfrac{x^2-z^2}{x^2+z^2}\right] \end{cases}$$



A. Aslanidès et al., Comput.Mater.Sci.**10**(1998) 401

Scale changing

Elastic theory ⟺ Numerical simulation

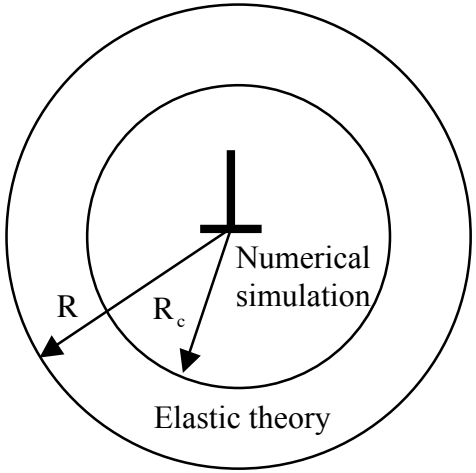
$$E^{tot}_{excess}(R_c,R) = E^{core}_{excess}(R_c) + E^{elastic}_{excess}(R_c,R)$$

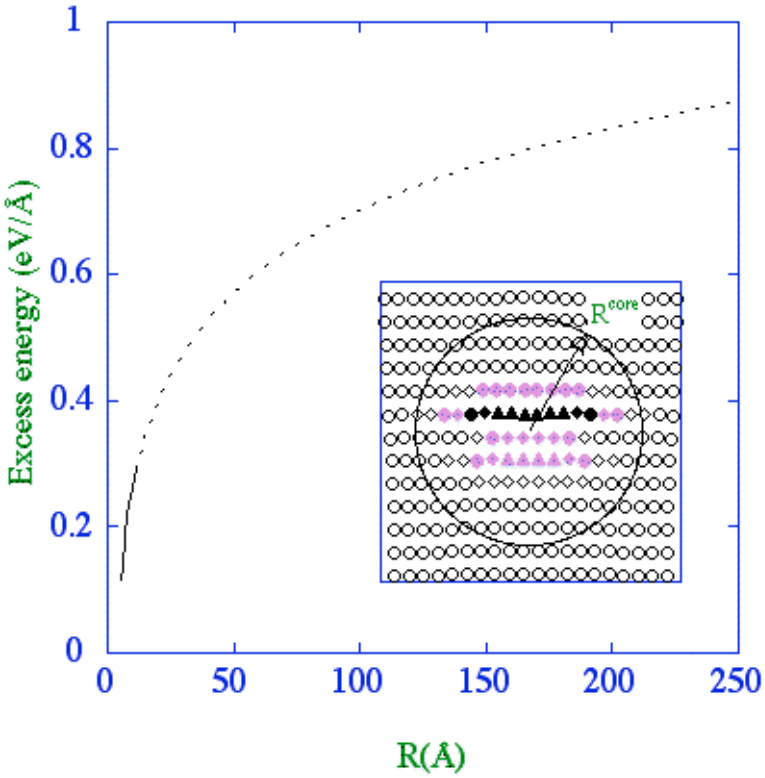
$$E^{tot}_{excess}(R_c,R) = E^{core}_{excess}(R_c) + \frac{\mu b^2}{4\pi(1-\nu)} \ln\left(\frac{R}{R_c}\right) L$$

$$\varepsilon = \frac{E^{tot}_{excess}(R_c,R)}{V}$$

$$\frac{d\varepsilon}{dR_c} = 0$$

$$\Rightarrow R_c = 11.4\text{Å}$$

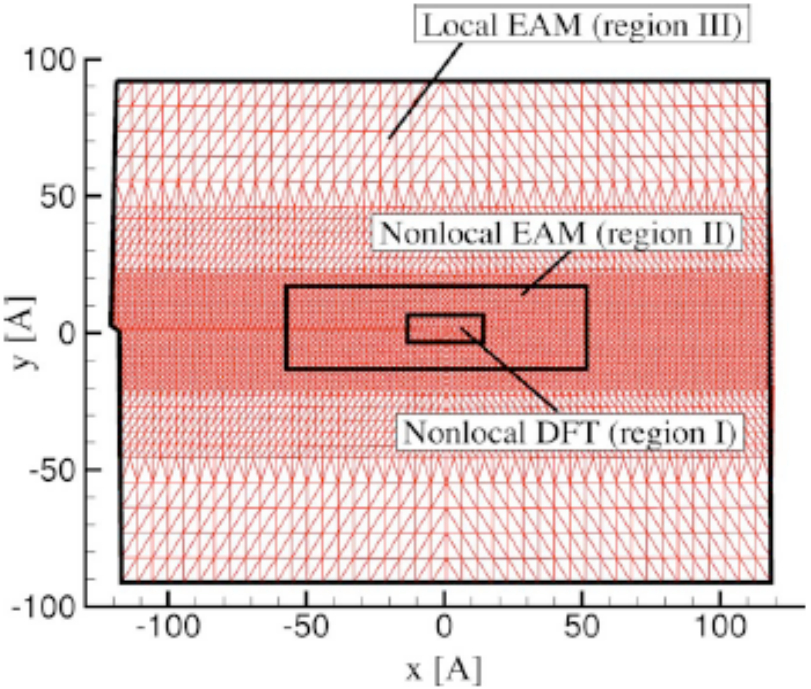




A. Aslanidès et al., Comput.Mater.Sci.**10**(1998) 401

Multiscale approach for metals

Lu, Tadmor, Kaxiras, PRB 73, 024108 (2006)



→ different atomic species within region I,
without developping reliable semi-empirical potential

$$E^{int}[I,II] = E_{EAM}[I+II] - E_{EAM}[I] - E_{EAM}[II]$$

(impurities) (impurities)

→ particularly useful in dealing with impurities

Total Energy

deformation gradients, finite elements

$$E_{tot} = E[I+II] + \sum_{j=1}^{N_{loc}} n_j E_j^{loc}(\{\mathbf{F}\})$$

$$E[I+II] = E_{DFT}[I] + E_{EAM}[II] + E^{int}[I,II]$$

$$E^{int}[I,II] = E_{EAM}[I+II] - E_{EAM}[I] - E_{EAM}[II]$$

$$E[I+II] = E_{DFT}[I] - E_{EAM}[I] + E_{EAM}[I+II]$$

Forces

$$-\mathbf{F}_i^{II} = \frac{\partial E_{tot}}{\partial \mathbf{q}_i^{II}} = \frac{\partial E_{EAM}[I+II]}{\partial \mathbf{q}_i^{II}} + \frac{\partial \sum_{j=1}^{N_{loc}} n_j E_j^{loc}(\{\mathbf{F}\})}{\partial \mathbf{q}_i^{II}}$$

$-\mathbf{F}_i^{III}$ id.

$-\mathbf{F}_i^{I}$: contributions from DFT atoms and the nearby EAM region II

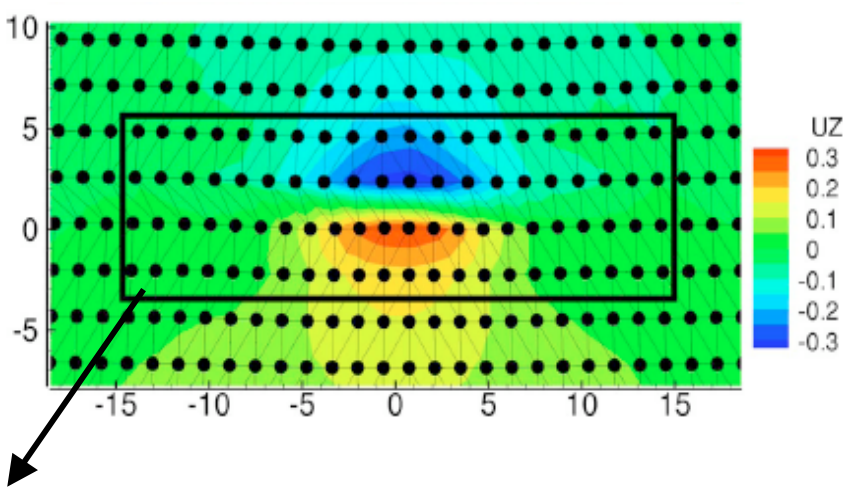
edge dislocation in Aluminum : core structure and H impurity

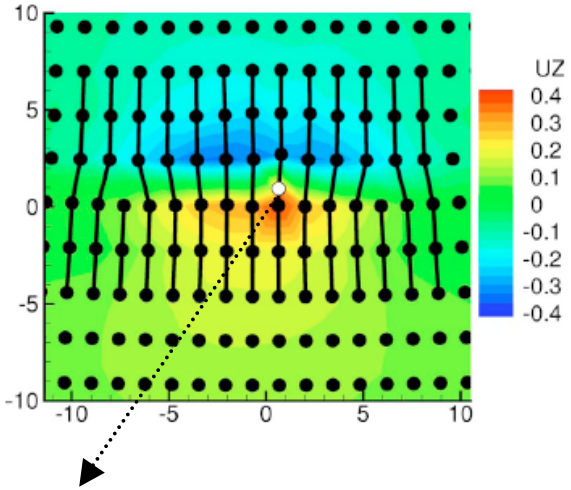
Lu, Tadmor, Kaxiras, PRB 73, 024108 (2006)

quantum mechanical calculation → impurities in the discolation core

long-range effects of the dislocation stress field

→ new insight in the problem of hydrogen embrittlement of metals





DFT I : 84 atoms (30 Å X 9 Å X 4.86 Å)

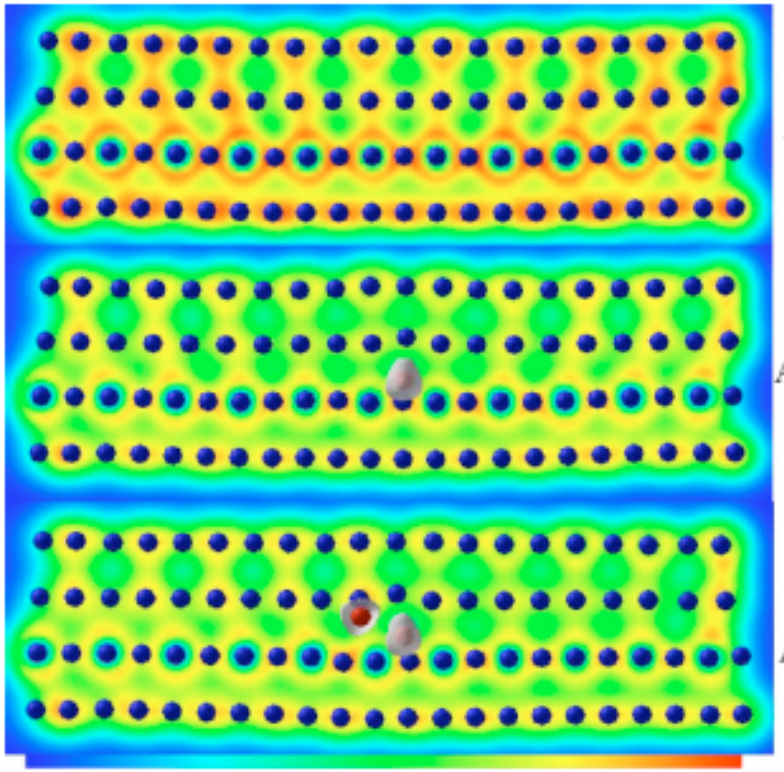
dissociation : 5.6 Å (exp : 5.5 Å, DM : 11-15 Å)

column of hydrogen impurities at the dislocation core

spreading of the core : 13 Å
(≡ H lower the stacking fault energy)

→ H-enhanced dislocation mobility

Electron density distribution at the dislocation core



strong bonding
more directionnal above the slipping plane
more spherical on the side of the two partials

charge accumulation on H atoms
negatively charged
covalent bonding across the slip plane is disrupted

ionic bonding between H and Al

→ wider dislocation core

Lu, Tadmor, Kaxiras, PRB 73, 024108 (2006)

Conclusions

some examples of the recent concepts in multiscale simulations methods

field in rapid progress, powerful techniques (DFT, MD)

qualitative but also quantitative modeling

sequential multiscale simulation : reliability of the phenomenological model
+ accuracy of the relevant parameters entering the model

concurrent multiscale simulation : more sophisticated and computationally demanding
more general applicability (no a priori physical quantities, no phenomenological models)
BUT the actual challenge are

* practical method
* ? partition of domains in the system
* problems related to the coupling between the different domains (error estimation)

this presentation is not exhaustive but illustrative of what multiscale modeling is !!