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Introduction to the kinetic Monte Carlo in radiation damage physics

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Introduction to the kinetic Monte Carlo method in radiation damage physics

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- Metropolis MC for equilibrium systems
- MMC: order-disorder phase transformations
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Tutorial papers

A. F. Voter, Introduction to the Kinetic Monte Carlo Method, in Radiation Effects in Solids, Springer, 2005.

P. Kratzer, Monte Carlo and kinetic Monte Carlo methods, "Multiscale Simulation Methods in Molecular Sciences", Juelich, 2009.

M. Allen, D. Tildesley, Computer simulation of liquids, Oxford, 1990.

Metropolis MC for equilibrium system

THE JOURNAL OF CHEMICAL PHYSICS VOLUME 21, NUMBER 6 JUNE, 1953

Equation of State Calculations by Fast Computing Machines

NICHOLAS METROPOLIS, ARIANNA W. ROSENBLUTH, MARSHALL N. ROSENBLUTH, AND AUGUSTA H. TELLER,
Los Alamos Scientific Laboratory, Los Alamos, New Mexico

AND

EDWARD TELLER,* *Department of Physics, University of Chicago, Chicago, Illinois*

(Received March 6, 1953)

A general method, suitable for fast computing machines, for investigating such properties as equations of state for substances consisting of interacting individual molecules is described. The method consists of a modified Monte Carlo integration over configuration space. Results for the two-dimensional rigid-sphere system have been obtained on the Los Alamos MANIAC and are presented here. These results are compared to the free volume equation of state and to a four-term virial coefficient expansion.

I. INTRODUCTION

THE purpose of this paper is to describe a general method, suitable for fast electronic computing machines, of calculating the properties of any substance which may be considered as composed of interacting individual molecules. Classical statistics is assumed, only two-body forces are considered, and the potential field of a molecule is assumed spherically symmetric. These are the usual assumptions made in theories of

II. THE GENERAL METHOD FOR AN ARBITRARY POTENTIAL BETWEEN THE PARTICLES

In order to reduce the problem to a feasible size for numerical work, we can, of course, consider only a finite number of particles. This number N may be as high as several hundred. Our system consists of a square† containing N particles. In order to minimize the surface effects we suppose the complete substance to be periodic, consisting of many such squares, each square containing N particles in the same configuration. Thus we

Metropolis MC for equilibrium system

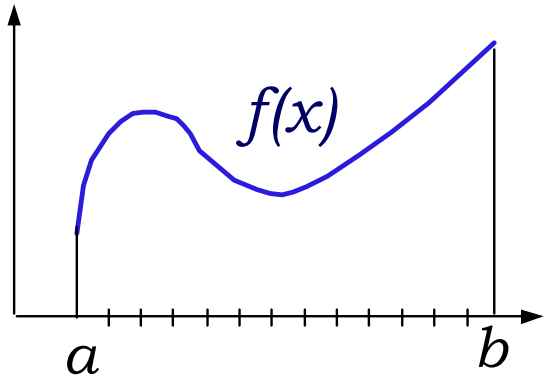
Evolution of statistical-mechanics integrals

$$\langle Q \rangle = \int dr_1 ... dr_N Q(R) e^{-U(R)/kT} \Big/ \int dr_1 ... dr_N e^{-U(R)/kT}$$

Configurational integral: $$Z = \int dr_1 ... dr_N e^{-U(R)/kT}$$

Integration

$$I = \int_a^b f(x)dx$$

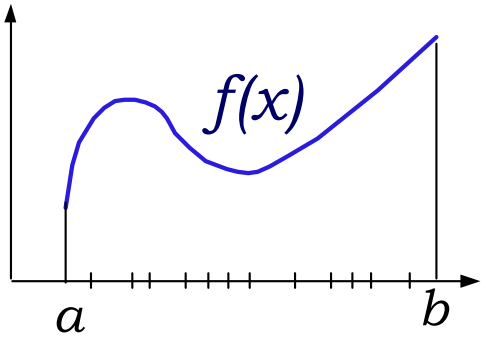


Regular integration

$$I \to \frac{b-a}{N}\sum_{i=1}^{N} f(x_i), \qquad x_i = a + \delta(i-0.5)$$

MC integration

$$I \to \frac{b-a}{N}\sum_{j=1}^{N} f(x_j), \qquad x_j = a + (b-a)\cdot\xi_j$$



Integration errors

$$\Delta I_{reg} \sim \delta^2 \sim \frac{1}{n^2} \qquad \Delta I_{MC} \sim \delta^2 \sim \frac{1}{n^{1/2}}$$

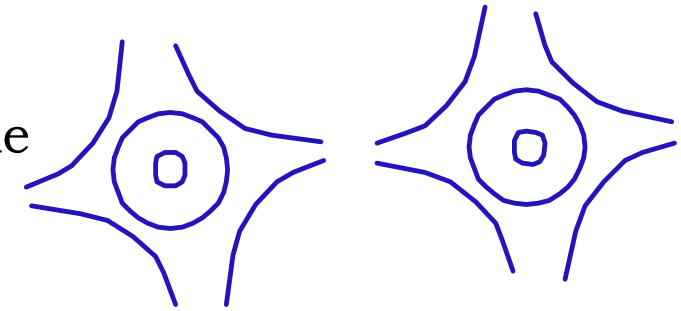
For d-dimensional integration the regular method meets impassable difficulty:

$$\Delta I_{reg} \to \sim \frac{1}{n^{2/d}}$$

MC method is more efficient at $d > 4$ and remained valid in a high-dimensional space

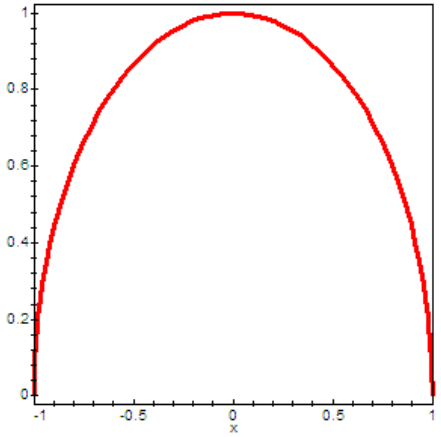
$$\Delta I_{MC} \sim \frac{1}{n^{1/2}}$$

But random sampling of configuration space is not the efficient method for the systems with restricted areas



Concept of importance sampling

Example: $f(x) = \sqrt{1-x^2}$



The region near x = 0 mainly contributes to the integral.

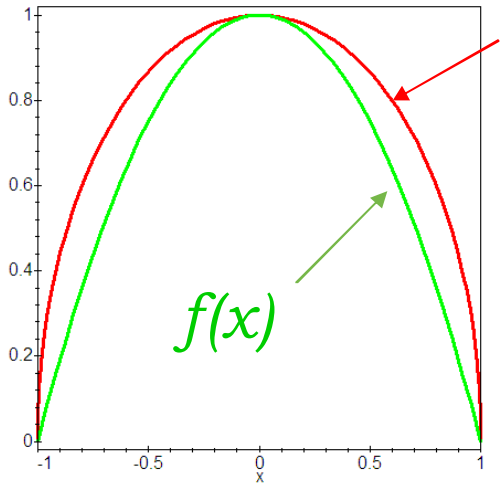
Introduce an appropriate non-uniform distribution

$$p(x) = \frac{3}{4}(1-x^2)$$

and approximate integral by

$$I \to \sum_{j=1}^{N} f(x_j)\cdot\delta_j, \qquad \delta_j = \frac{b-a}{N}\frac{1}{p(x_j)}$$

$$\to \frac{b-a}{N}\sum_{j=1}^{N}\frac{f(x_j)}{p(x_j)} \qquad d\xi_j \to p(x_j)dx_j$$



Metropolis MC

$$\langle Q \rangle = \int dr_1 ... dr_N Q(R) p(R) \qquad \text{where} \qquad p(R) = \frac{1}{Z} e^{-U(R)/kT}$$

Configurations are covered by random walk in the phase space sampling with desired distribution *p(R)*.

MMC uses random walk with transition probability to go from state ***m*** to ***n*** defined by the ratio

$$p_{m \to n} = p_n / p_m = e^{-(U_n - U_m)/kT}$$

if $U_n - U_m < 0$ we put $p_{m \to n} = 0$

Metropolis MC algorithm

1. Set up initial state (configuration) m and calculate U_m

2. Generate random displacement of any coordinate
   $r_n \to r_m + \Delta r_m$

3. Calculate energy U_n.
   if U_n-U_m < 0 then accept the new configuration
   else

   calculate $p_{m\to n} = \exp\left(-\frac{U_n - U_m}{kT}\right)$

   if $p_{m\to n} > \xi$ then accept the new configuration
   else stay at the same state.

4. Repeat from the step 2 accumulating sum for averaging

Example: order-disorder phase transformations

C. Abromeit, S. Matsumura, Phil. Mag. A82 (2002), p. 2287

***Kawasaki* approach:**

- An arbitrary atom is chosen,
- Energy change $\Delta E=-\Sigma V_n\{\Sigma\sigma_i\sigma_j\}$ due to an exchange of this atom with a neighbouring atoms is calculated.
- Both atoms change their sites according to the probability

$$W(\Delta E,T) = 0.5\,[1- \tanh(\Delta E/2k_BT)]$$

Parameters: A_3B alloy with a $L1_2$ structure

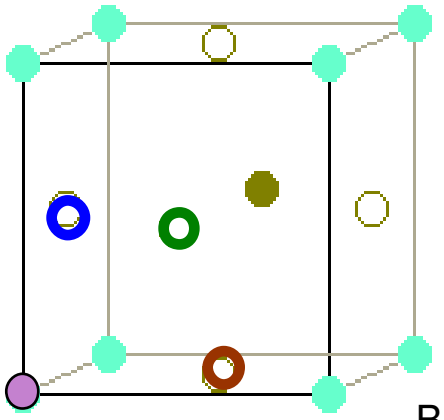
Interaction parameters: Poduri and Chen, Acta metall. **46** (1998) 1719

Determination of a local chemical order

Properties of $L1_2$ long range order transformation (A_3B)

Ground state: fourfold degenerated: α β γ δ

4 simple cubic sublattices: four type of ordered domains



B atom

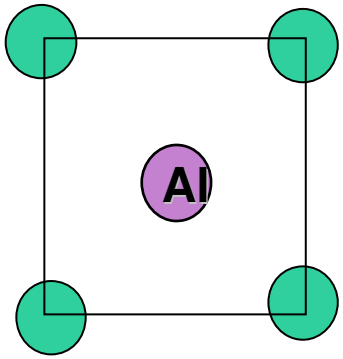
Atomic structure

Example:

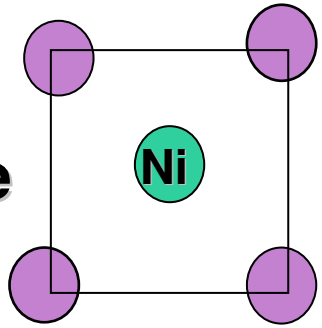
α Configuration

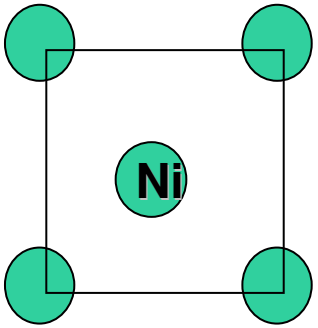
(100) plain

Al site



Ni site



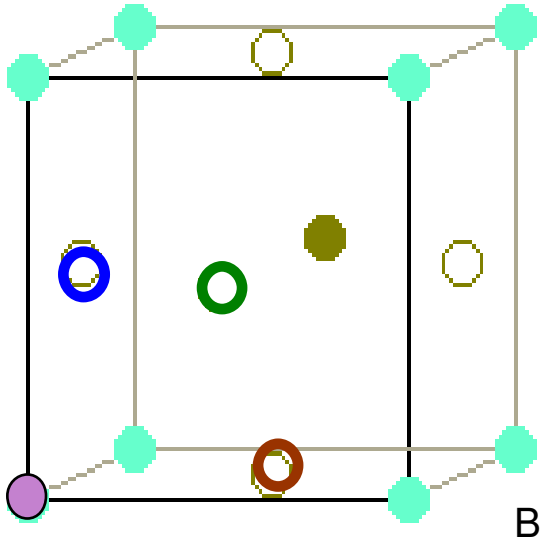


…. Etc.

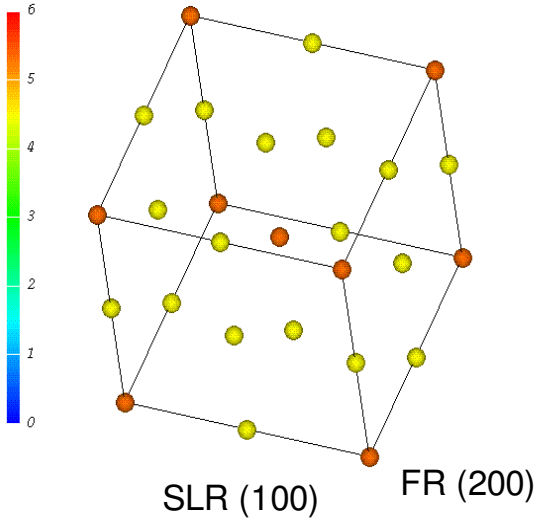
Properties of $L1_2$ Long Range Order transformation (A_3B)

Ground state: fourfold degenerated: α β γ δ

4 simple cubic sublattices: four type of ordered domains

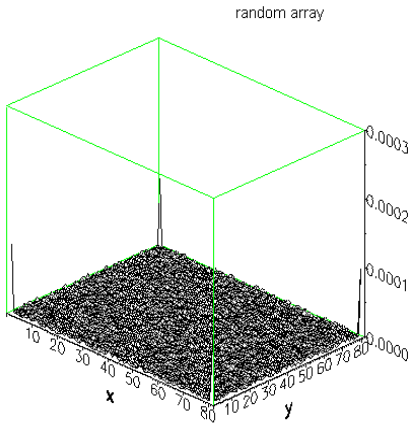


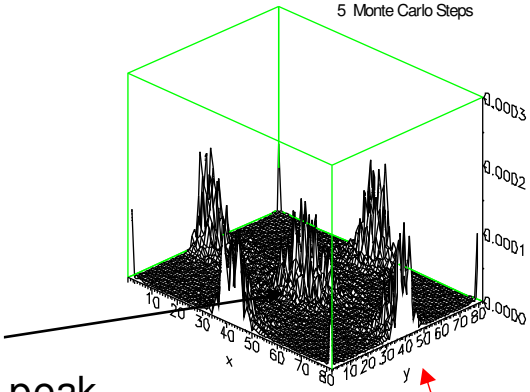
Atomic structure



Reciprocal k space:
3-d Fouriertransform

Thermal ordering of a $L1_2$ structure

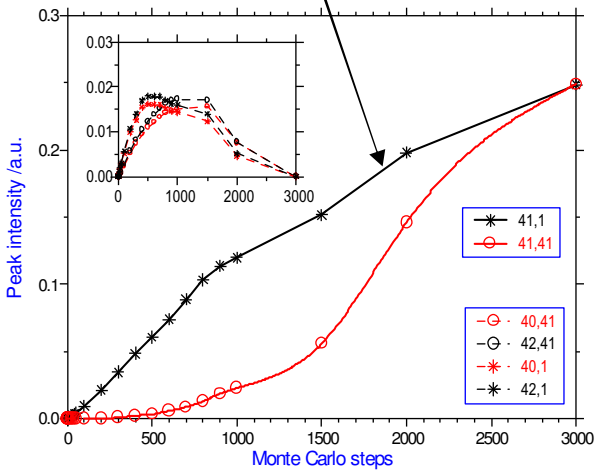




Central peak
110

100
SLR

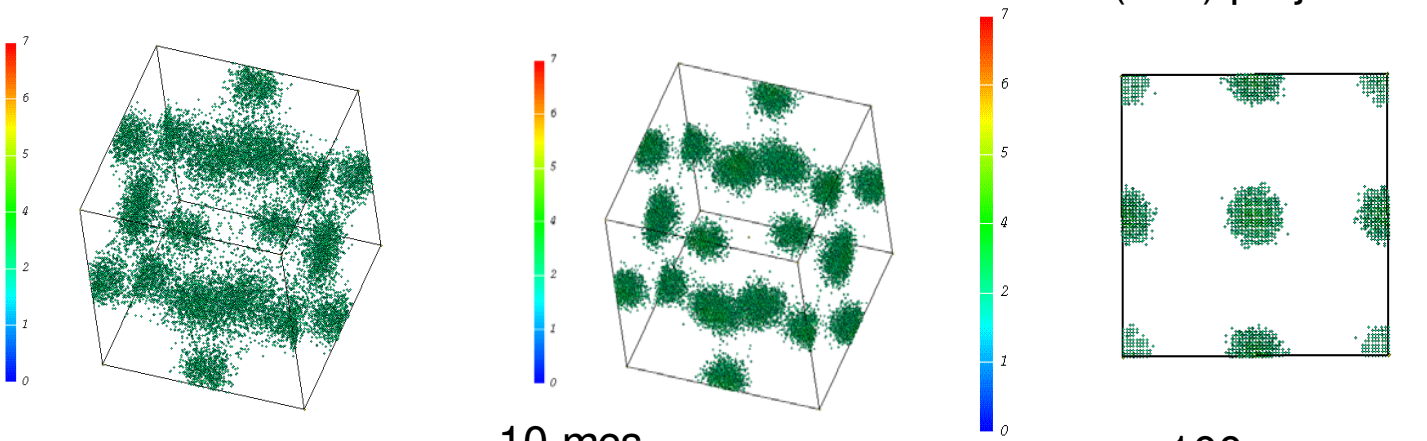
intensity of the
superlattice reflections



Thermal ordering of a $L1_2$ structure

3-d Fouriertransform

(100) projection

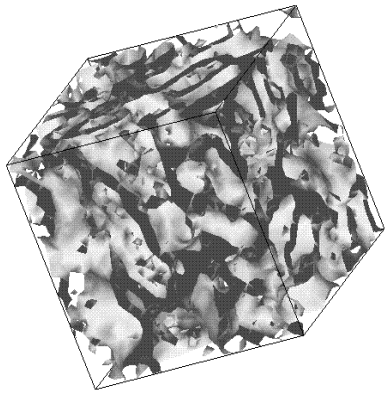


2 mcs

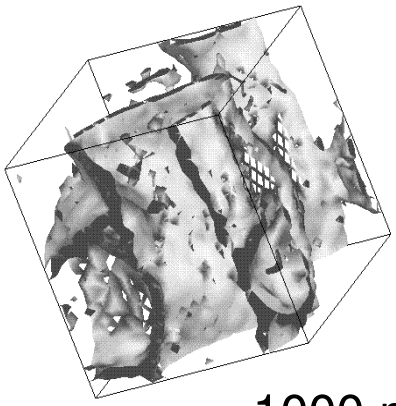
10 mcs

100 mcs

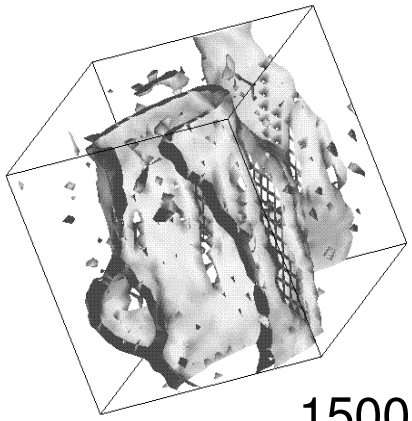
anti phase boundaries
in 3-d real space



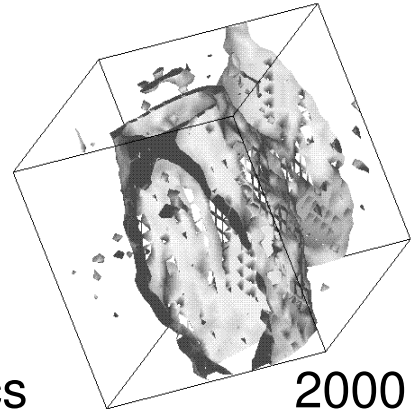
Thermal ordering of a $L1_2$ structure



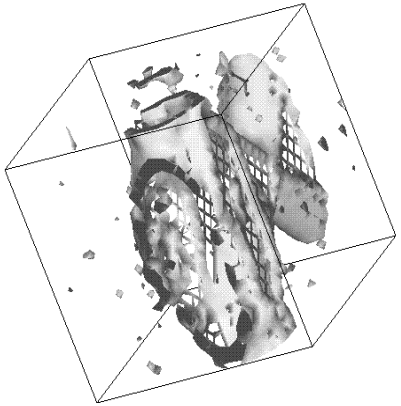
1000 mcs



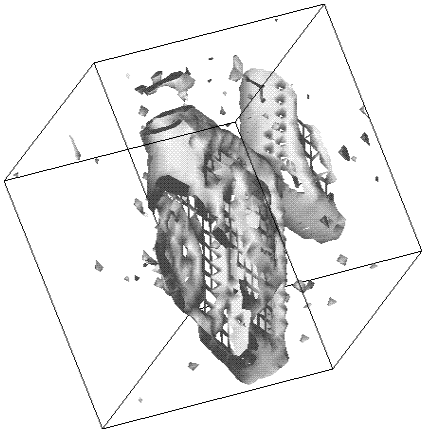
1500 mcs



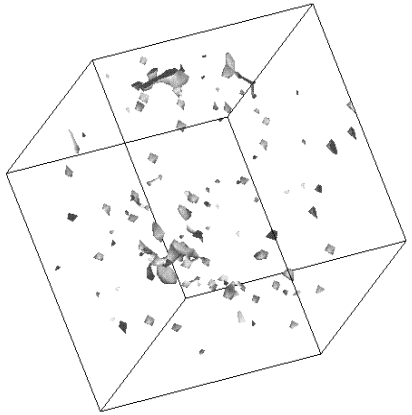
2000 mcs



3000 mcs



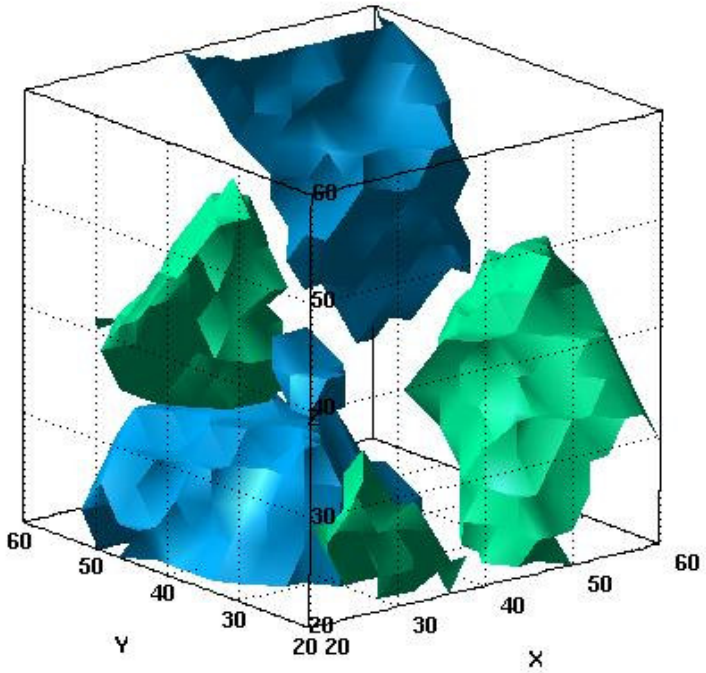
4000 mcs



5000 mcs

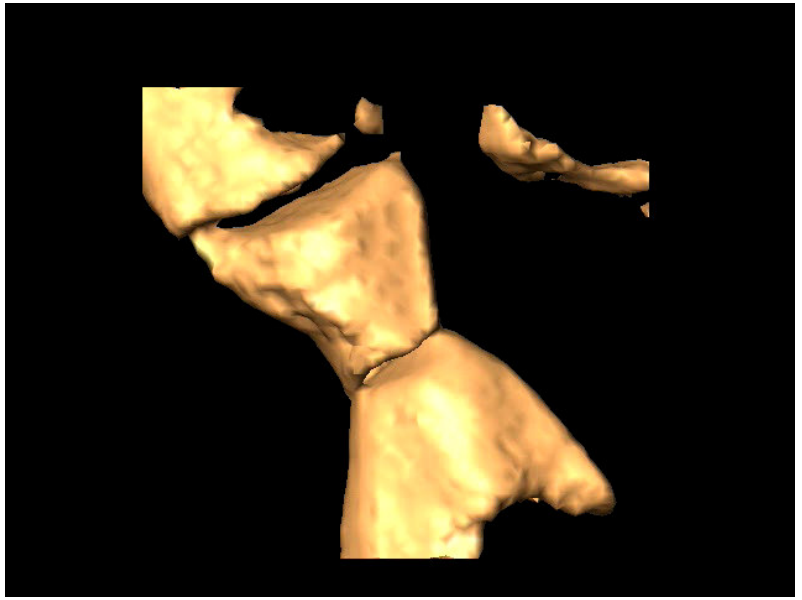
LRO order evolution in initially disordered Ni_4Mo

MC Simulation



D1a domains
(Variants 1 & 2)
@ 20000 mcs

TEM -tomography



S. Matsumura,
Kyushu University, Japan
MRS Boston 2006

Metropolis MC summary

Advantages:

- *Simple implementation*
- *High efficiency*
- *Can be easily adopted for the calculations in any ensembles: NVT, NPT, grand-canonic etc*
- *No problems with strongly different rates in system*

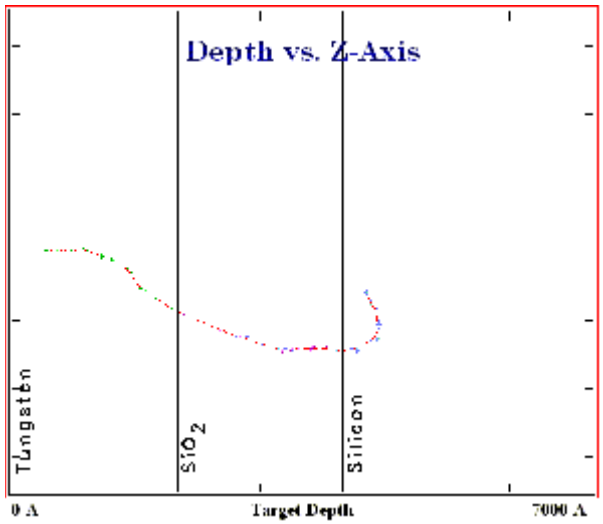
Restriction:

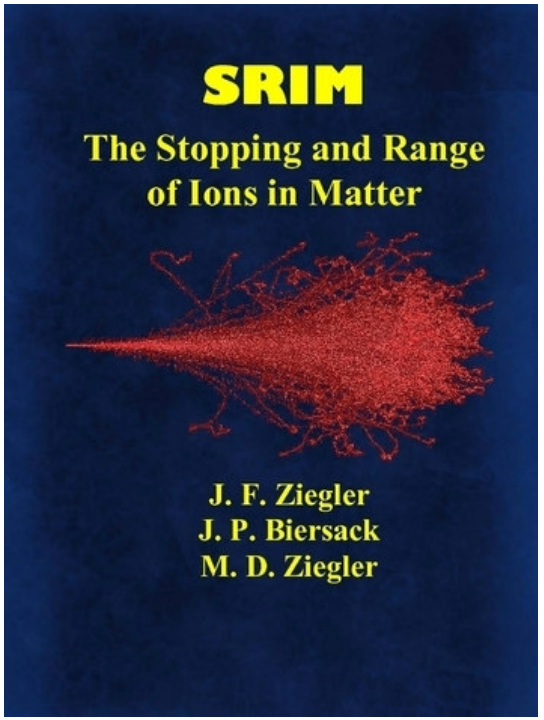
- *Cannot study kinetics or dynamical problems*
- *Poor count of cooperative rearrangements*

Dynamic Monte Carlo

The Stopping and Range of Ions in Matter

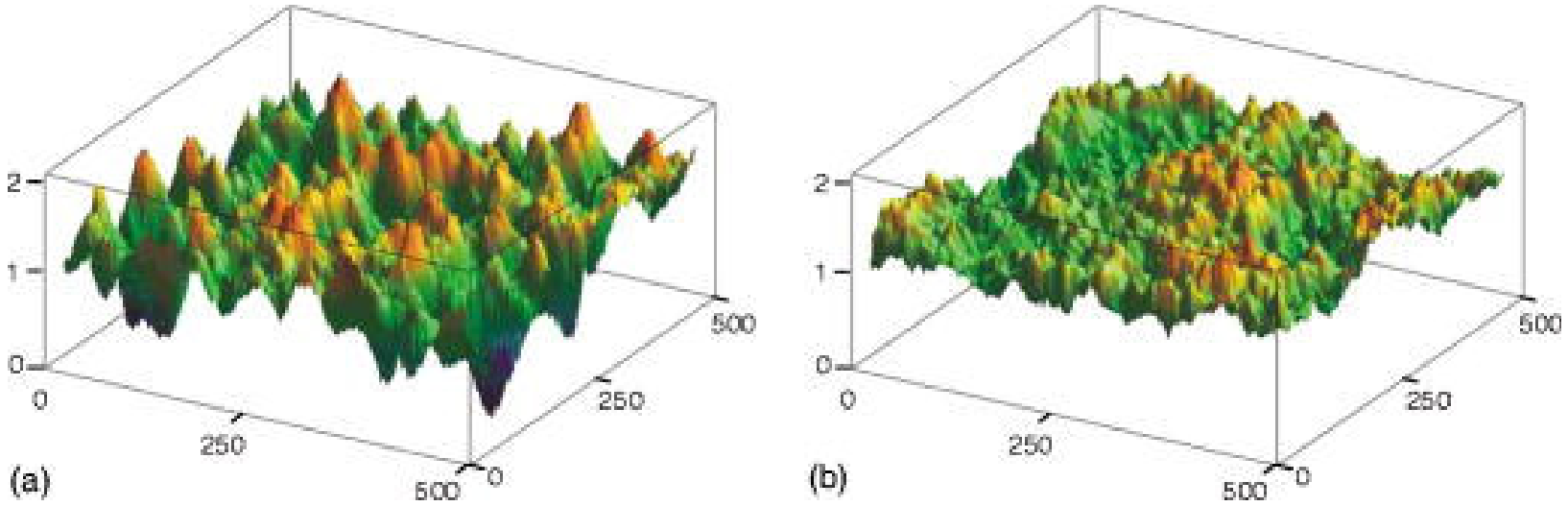
http://www.srim.org





Surface roughness evolution of nanocomposite thin films

A. A. Turkin et al, JAP 105 (2009) 013523



Typical examples of surface morphology: (a) the initial surface of the CrTi interlayer and (b) the surface of the TiC/a-CTiC/a-C coatings after 4 h deposition hours in pp-dc mode.

Film deposition: continuum linear growth model

$$h(\mathbf{r},t) = H(\mathbf{r},t) - \langle H(\mathbf{r},t) \rangle$$

$$\frac{\partial h(\mathbf{r},t)}{\partial t} = \eta(\mathbf{r},t) - div\mathbf{j} \qquad \mathbf{j} = -D_2 \nabla h(\mathbf{r},t) + D_4 \nabla \left(\nabla^2 h(\mathbf{r},t) \right)$$

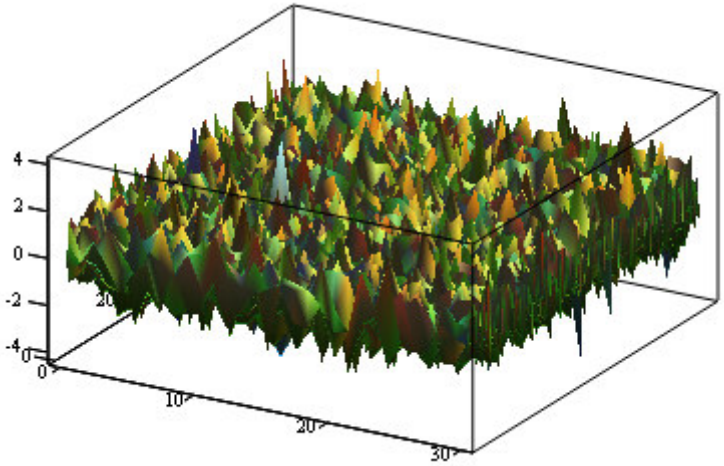
$\eta(\mathbf{r},t)$ = stochastic noise term due to local fluctuations of the deposition flux

$$\langle \eta(\mathbf{r},t)\eta(\mathbf{r}',t') \rangle = D\delta(\mathbf{r}-\mathbf{r}')\delta(t-t')$$

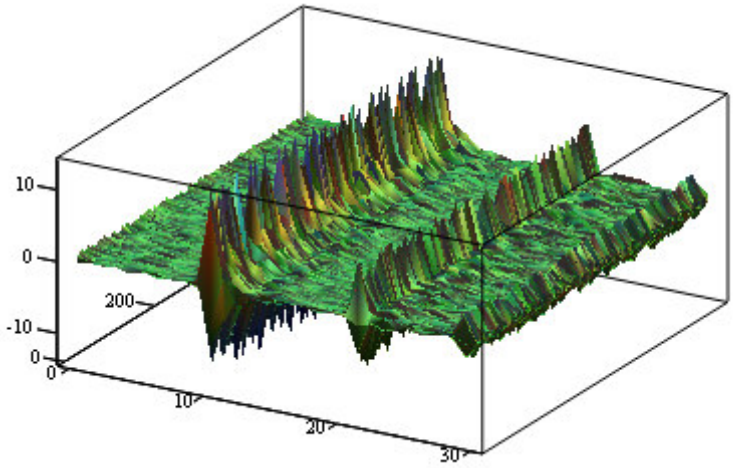
$\mathbf{j}$ = diffusion flux along the surface

Linear growth model

Results of numerical integration essentially depends on correct choice of random number generator



Good RNG



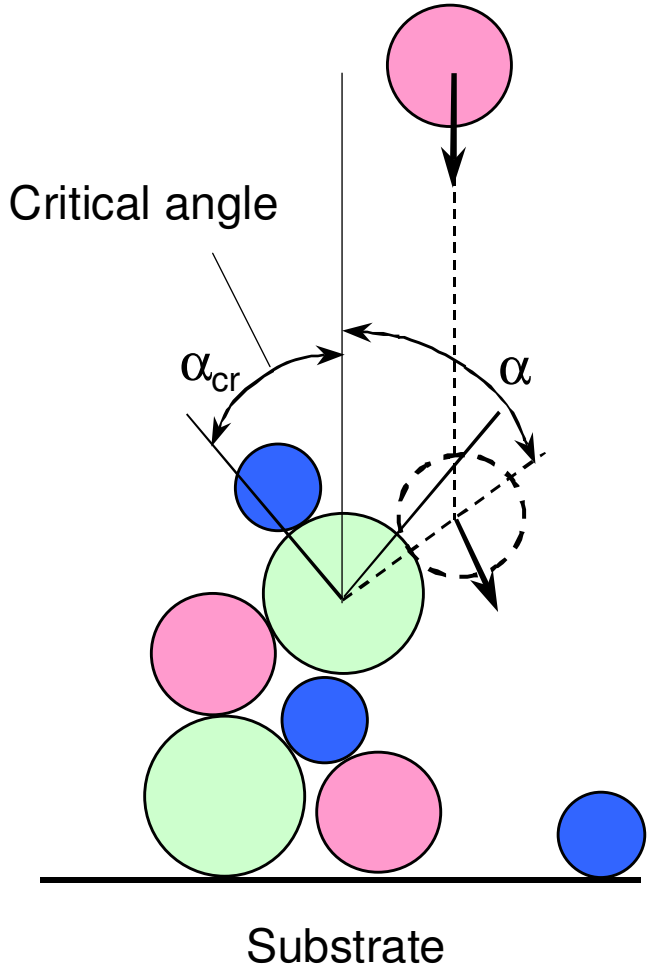
Bad RNG

Sample size NxN = 512x512, $2x10^7$ random numbers were generated

Surface topography depends on N (periodicity of RNG)

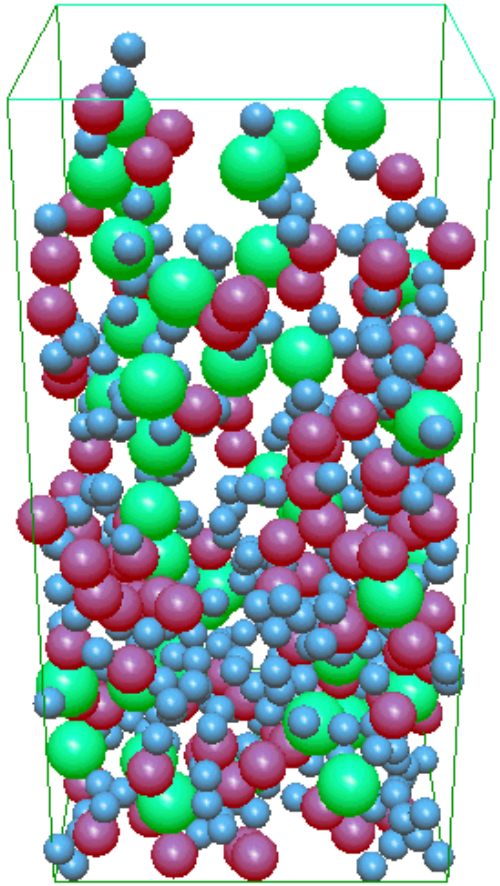
Synthesis of cluster-assembled nanostructured materials

M.S. Byshkin, A.S. Bakai, N.P. Lazarev, A.A. Turkin, Cond. Matter Physics, 6 (2003) 93.

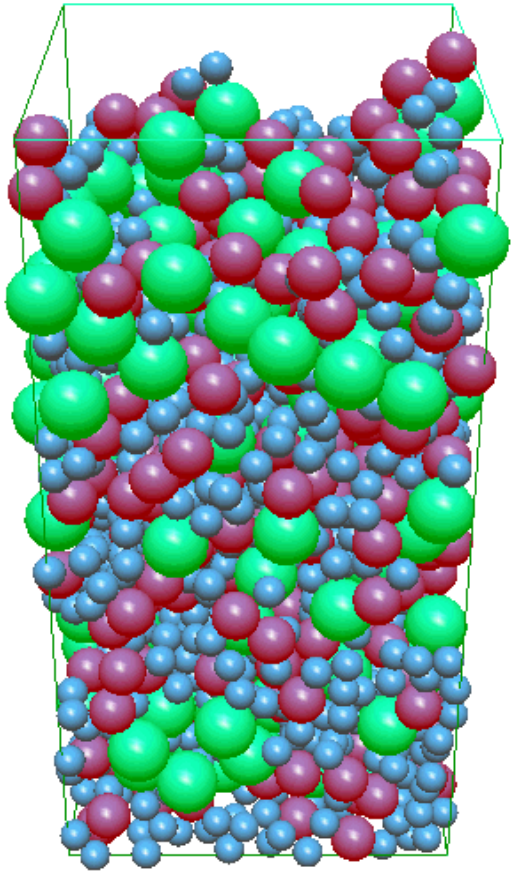


- Spherical clusters of various sizes fall at random to the substrate
- Cluster stops if $\alpha<\alpha_{cr}$, otherwise it rolls down till it meets at least two other balls

Synthesis of cluster-assembled nanostructured materials



$\alpha_{cr} = \pi/2$
Low surface mobility

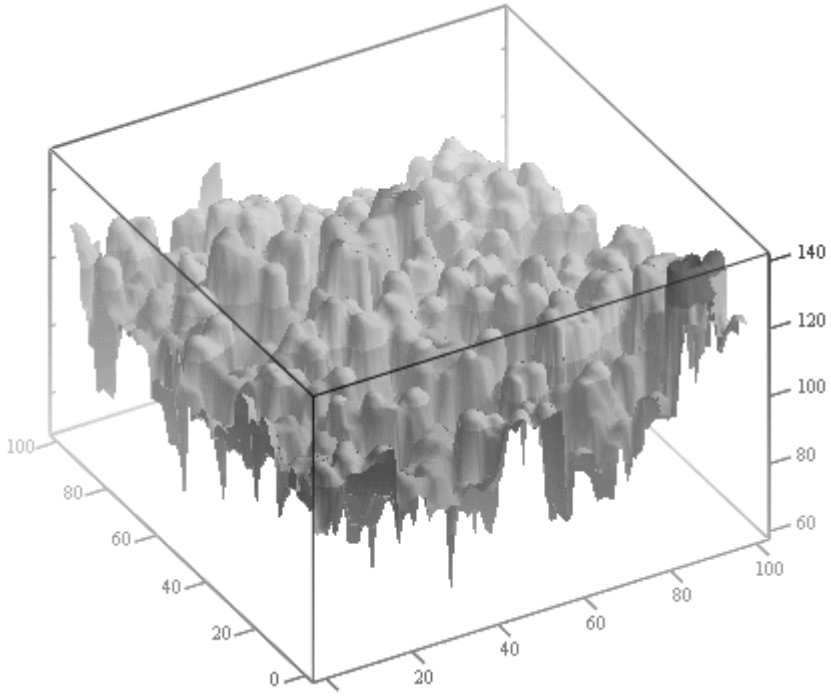


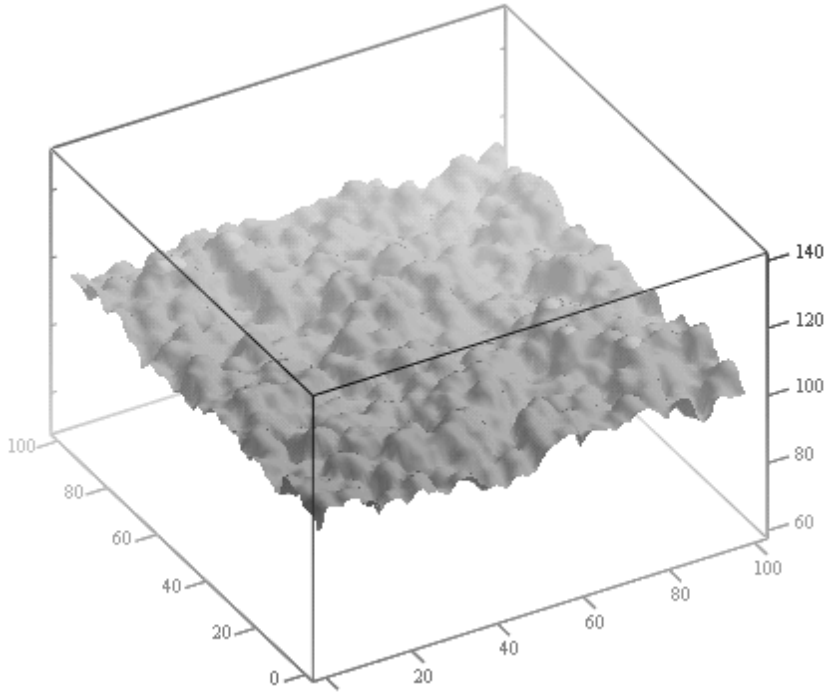
$\alpha_{cr} = 0$
High surface mobility

Large statistically representative samples can be generated with non-lattice MC

Non-lattice dynamic MC

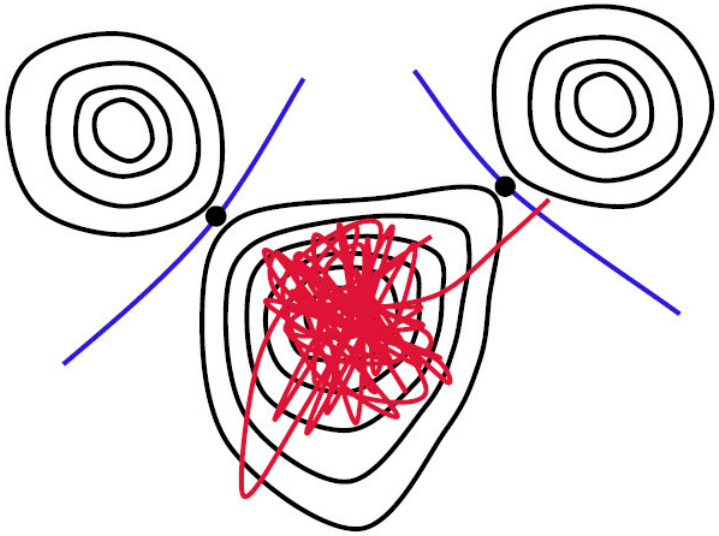
Images of the film surface scanned with a probe ball





Kinetic Monte Carlo

A. F. Voter (2005)



Potential energy surface for an energy-barrier-limited infrequent-event system. After many vibrational periods, the trajectory finds a way out of the initial basin, passing a ridgetop into a new state.

Kinetic Monte Carlo

Principles:

Relies on knowing the *rates* and *mechanisms* of all the relevant transitions from a given initial state.

The rates depend on the energy barrier between the states.

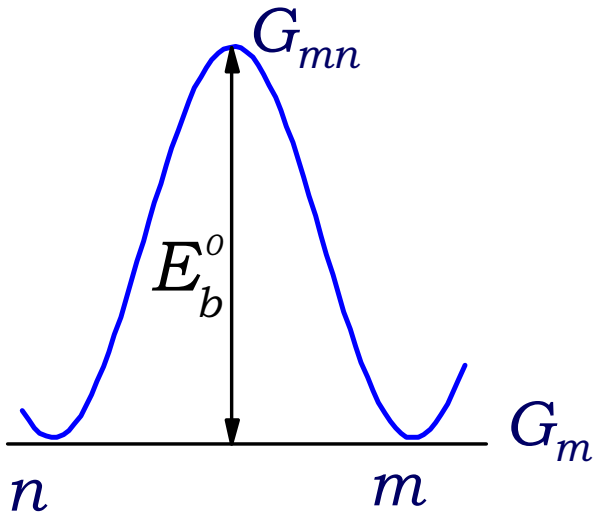
The main advantages of kinetic Monte Carlo is that *time* is defined and only a small number of elementary reactions are considered, so the calculations are fast.

Kinetic Monte Carlo

Jump over barrier occurs by thermal activation

$$W_{mn} = \omega_0 \exp\left[-\left(G_{mn} - G_m\right)/kT\right]$$

$$E_b = G_{mn} - G_m$$



Probability density for a jump: Poisson distribution

$$f(W) = W \exp(-Wt)$$

System with multiple rates

In system with multiple set of rates W_k, $k = 1,\ldots N$ we choose event randomly with a probability $P(k)$ proportional to its jump rate:

$$P(k) = \frac{W_k}{W_{tot}} \qquad W_{tot} = \sum_{i=1}^{N} W_i$$

After that simulation time is advanced by:

$$\Delta t \propto \frac{1}{W_{tot}}$$

Time step decreases with increasing N .

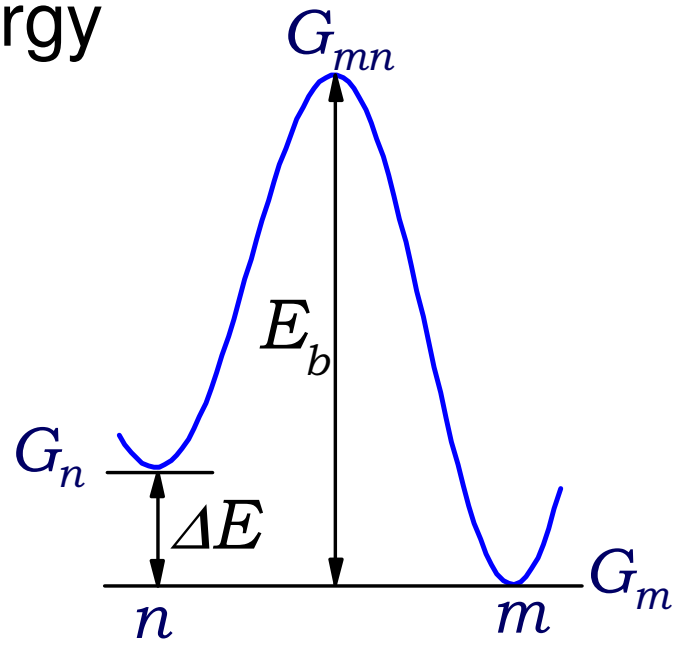
Kinetic MC algorithm

0. Set time $t = 0$
1. Form a list of all rates W_k of all possible events in the system
2. Calculate the cumulative rates $R_k = \sum_{i=1}^{k} W_i$, for $k = 1,...,N$
3. Get a uniform random number $0 \le \xi < 1$
4. Find the k for which $R_{k-1} < \xi R_N \le R_k$
5. Carry out event k
6. Recalculate all W_k which may have changed due to jump
7. Get a new uniform random number $0 \le \xi < 1$
8. Update the time with $t = t + \Delta t$ where $\Delta t = -\ln(\xi) / R_N$
9. Repeat from step 1

Heuristic calculation of barrier energy

$$E_b = E_b^0 + (G_m - G_n)/2$$

Only energies in sites are calculated



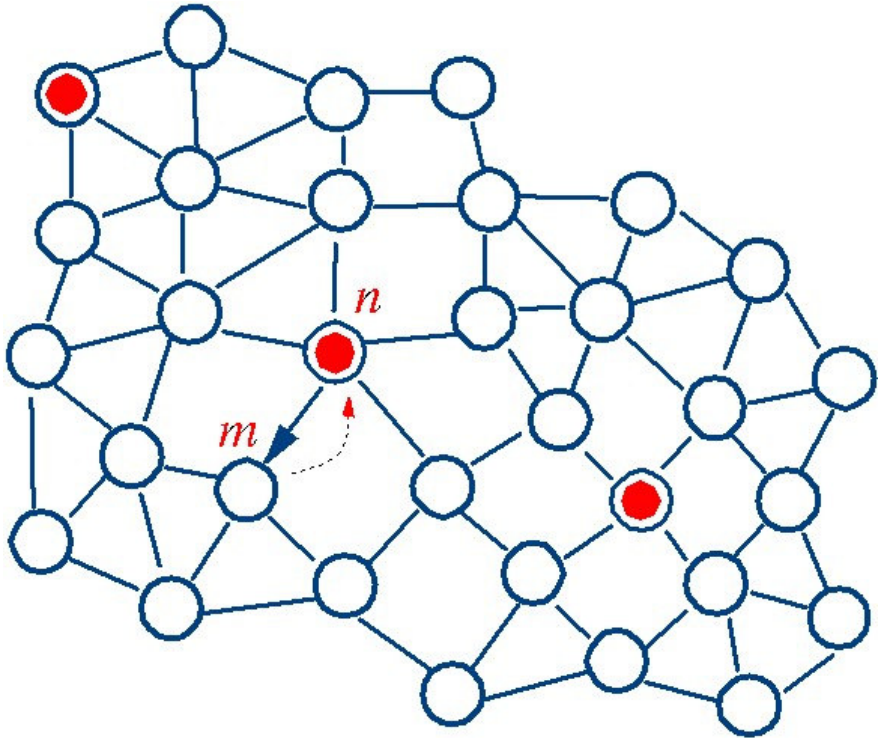
Imitate a local force:

$$\Delta E = F_{mn} \cdot a$$

$$G_{mn} = G_{mn}^0 + F_{mn} \cdot a/2$$

Diffusion in regular and disordered lattices

J.W.Haus, K.W.Kehr, Phys. Rep. **150**, 5&6, (1987) 263-406.
S.Havlin, D.Ben-Avraham, Adv. Phys. **36**, 6, (1987), 695-798
J.-Ph.Bouchand, A.Georges, Phys. Rep, 4&5 (1990).



Random walks on a random structure. Positions of sites are fixed in space and time.

Master equation

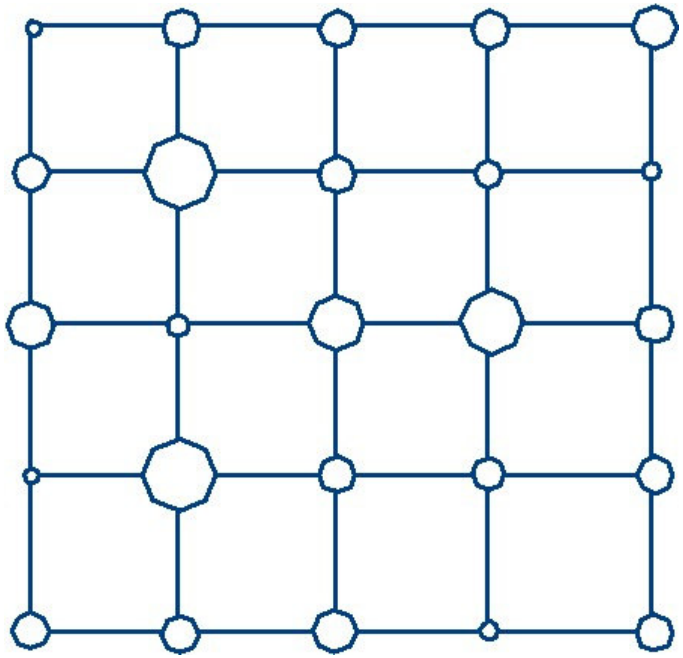
Jumps as Poisson process:

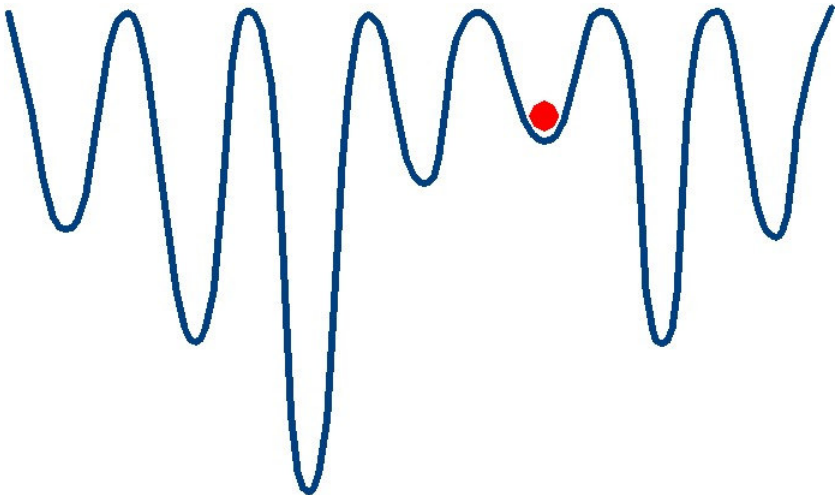
$$P(n,t'+\tau \mid m,t') = \begin{cases} W_{nm} & n,m \text{ nearest neighbours} \\ 1-\tau \cdot W_{mn} & n=m \\ 0 & \text{otherwise} \end{cases}$$

$$\dot{P}(n,t \mid m,0) = \sum_{n'} \left[W_{n'n} P(n',t \mid m,0) - W_{nn'} P(n,t \mid m,0) \right]$$

$$P(n,t) = \sum_{m} \rho_0(m) \cdot P(n,t \mid m,0)$$

Random traps



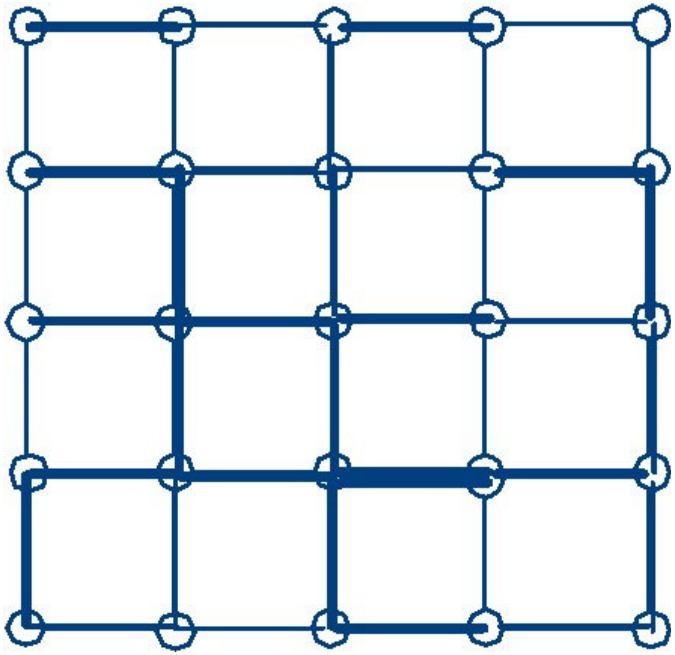


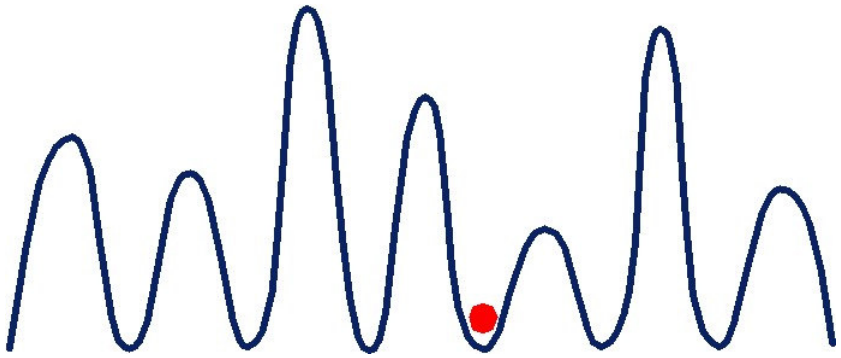
$$\dot{P}(n,t) = \sum_m \left[\Gamma_m P(m,t) - \Gamma_n P(n,t)\right]$$

$$P^{eq}(n) \propto \frac{1}{\Gamma_n} \qquad D = a^2 \left\langle \frac{1}{\Gamma_n} \right\rangle^{-1}$$

Deepest holes control the diffusion

Random barriers



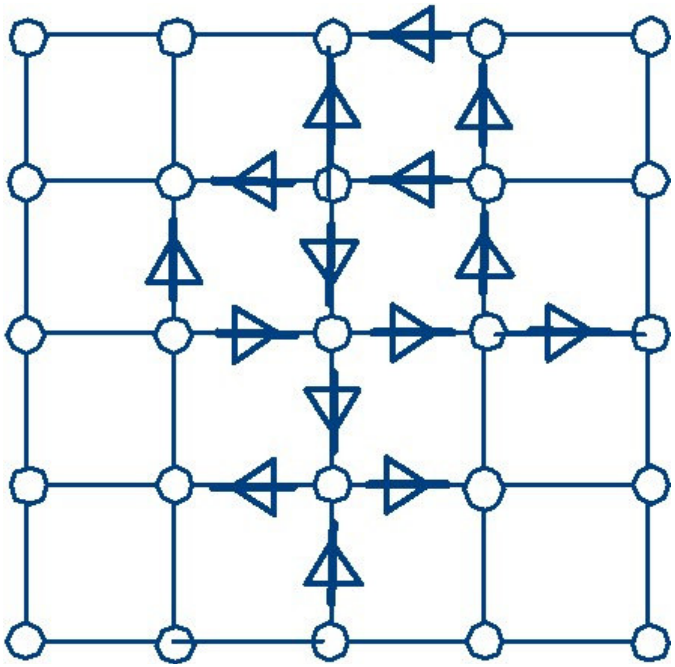


$$\dot{P}(n,t) = \sum_m W_{nm}\left[P(m,t) - P(n,t)\right]$$

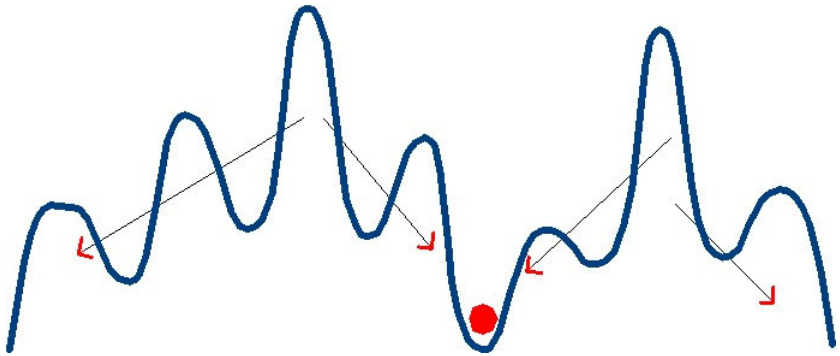
$$W_{nm} = W_{mn} \qquad P^{eq}(n) = const$$

Correlations and percolation on fast diffusion paths

Random forces



“Gradient” of a potential

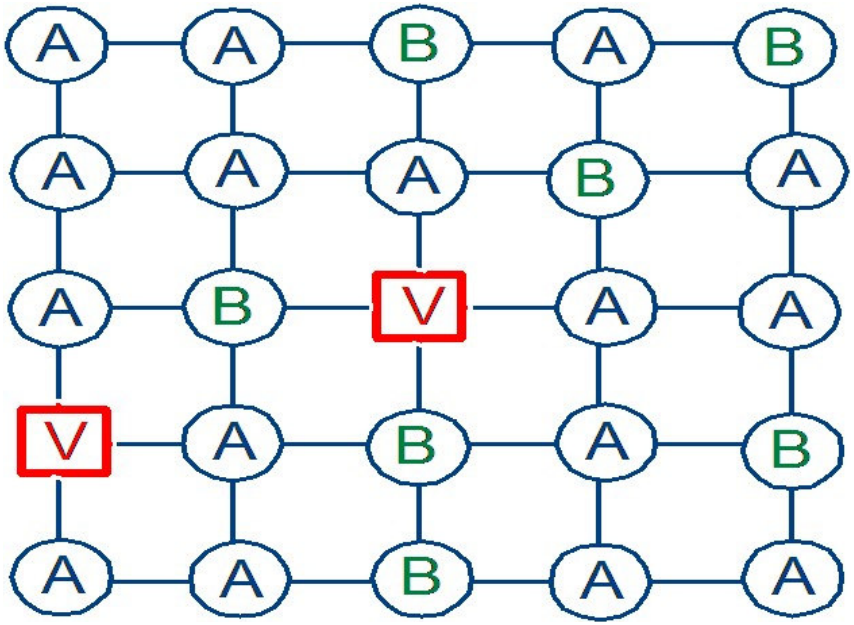


$$W_{nm} = W_0 \exp[-aF_{mn}/2kT]$$

$$W_{mn} = W_0 \exp[+aF_{mn}/2kT]$$

$$F_{nm} = (U_n - U_m)/a \qquad P_n^{eq} \sim \exp[-U_n/kT]$$

Diffusion in random multicomponent alloys via vacancies



,

Manning model

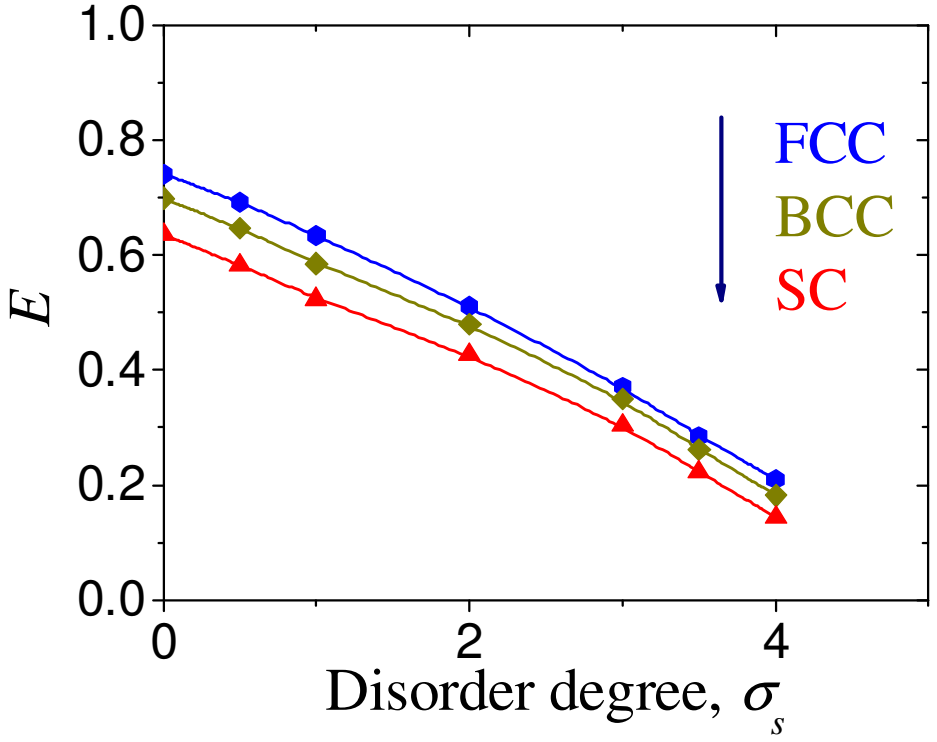
$$W_{mn} = \Gamma_j \exp\left[-\left(G_{mn} - G_m\right)/kT\right] \qquad j = A, B$$

Isotop-effect

N.P.Lazarev et al, Phys. Rev. Lett. 88, 045502 (2002).

$$E = (D_B / D_A - 1) / (\sqrt{m_A / m_B} - 1)$$

$$\rightarrow (D_B / D_A - 1) / (\Gamma_B / \Gamma_A - 1)$$



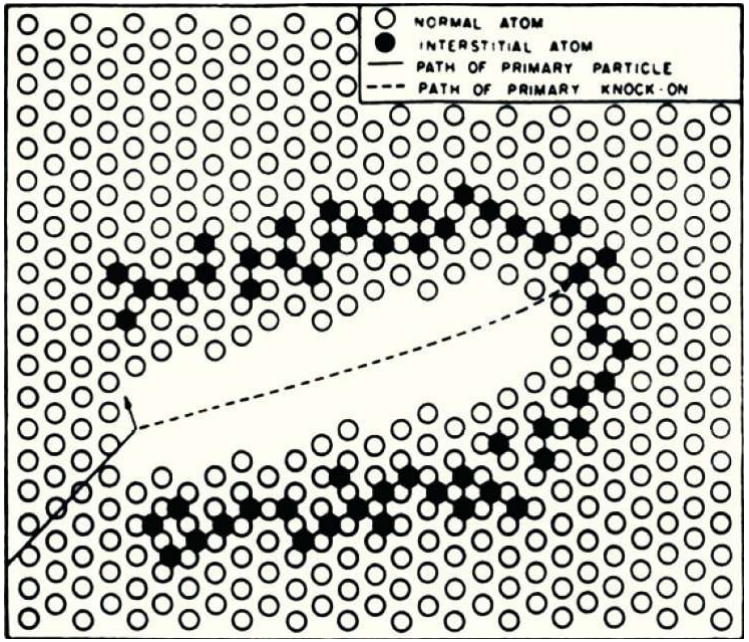
Manning:

$$f_{tr}^{B} = \frac{f_{tr}^{A}}{f_{tr}^{A} + (1 - f_{tr}^{A})(\Gamma_B / \Gamma_A)}$$

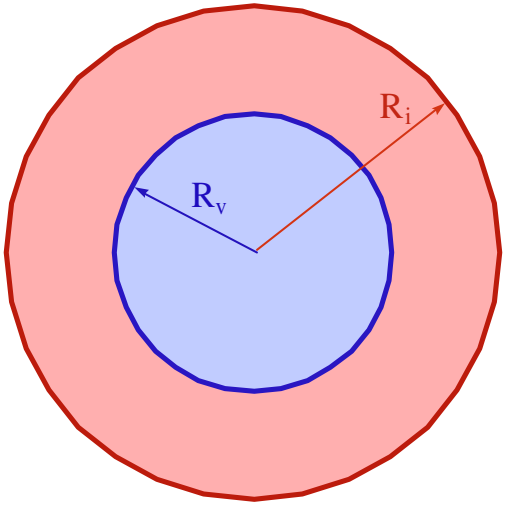
$$E \approx f_{tr}$$

$$\rho(G) = \frac{1}{\sqrt{2\pi\sigma}} \exp\left[-\beta(G - G_0) / 2\sigma^2\right]$$

Recombination in cascades: rate theory appr.



Schematic drawing of a displacement spike
J.A.Brinkman 1954



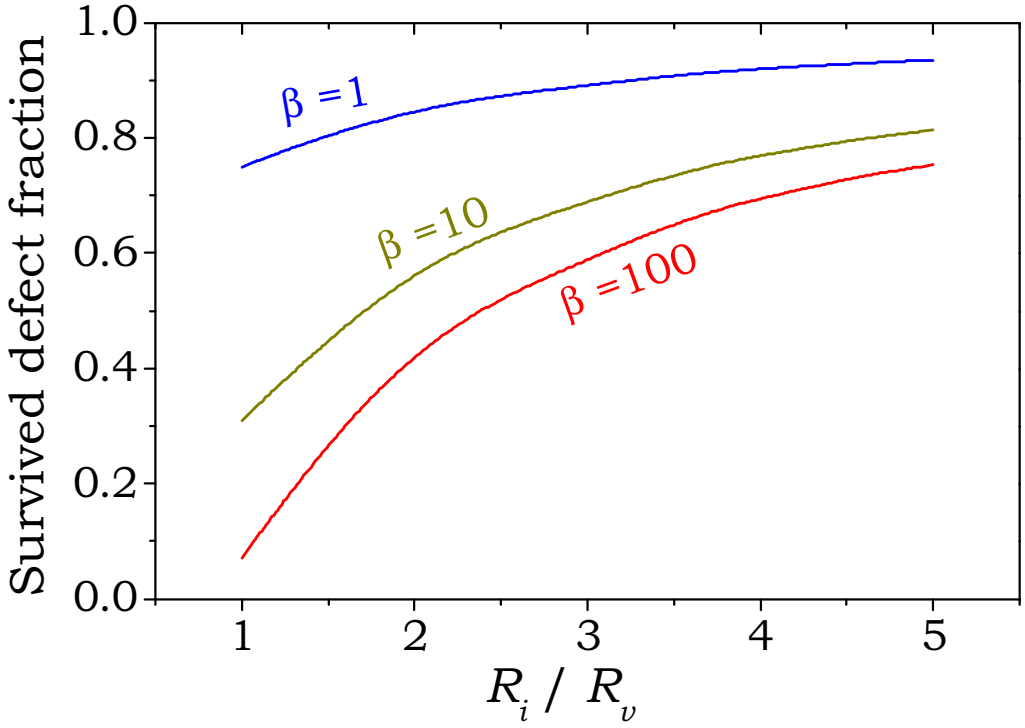
$$\frac{dC_{i,v}(r,t)}{dt} = -\alpha_{iv} C_v(r,t) C_i(r,t)$$

$$\alpha_{iv} \approx 4\pi r_{iv}(D_i + D_v)$$

$$C_{i,v}(r,0) = \begin{cases} C^0_{i,v}, r < R_{i,v} \\ 0, r > R_{i,v} \end{cases}$$

Recombination in cascades: rate theory appr.

$$\beta = R_v^2 / l_{r_{iv}}^2 \sim 3N_v \frac{r_{iv}}{R_v} \qquad l_{r_{iv}} = \frac{1}{\sqrt{4\pi r_{iv} C_v^0}}$$

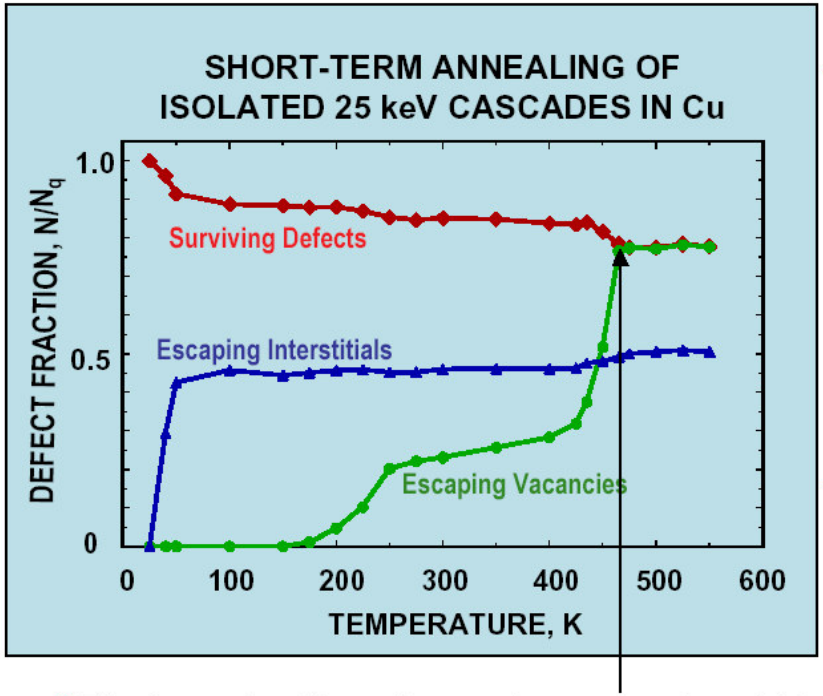


Recombination in cascades: kMC

Brian D. Wirth (2001)

Intra-cascade annealing in Cu

What happens to the cascade defects long after the first 10 ps?



KMC simulations of annealing individual cascade regions show the fractions of defects produced in a cascade that

- **survive recombination**
- **escape the cascade region**
- **form stable, sessile clusters**

as a function of temperature

❖ **The large fraction of escaping vacancies at high temperatures corresponds with the experimentally observed temperature dependence of void swelling.**

Low temperature evolution of precipitate platelets

EXPERIMENTAL

Fusion reactor material

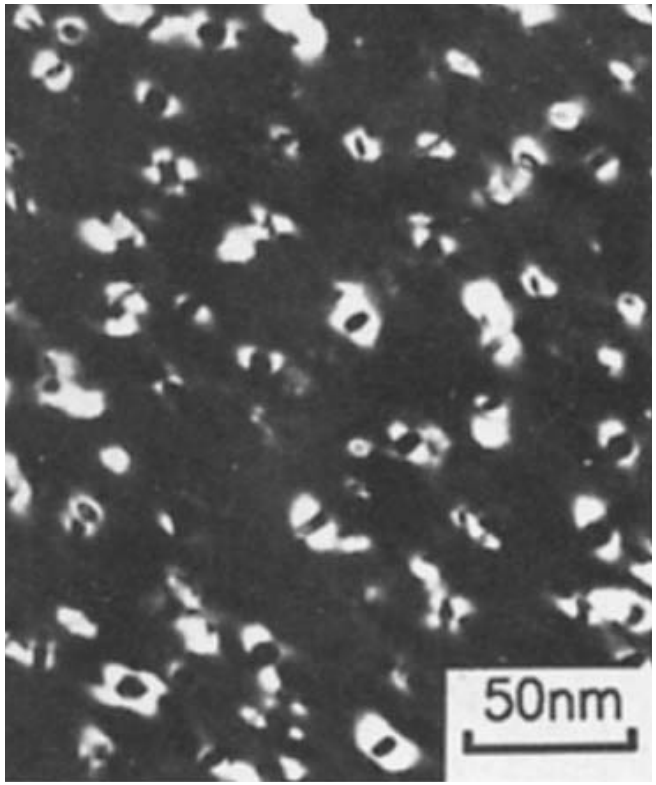
Cu-1.15at%Cr-0.03at%Zr-0.14at%Si alloy

coherent Cr platelets (fcc structure)

$\langle d_o \rangle$ = 12 nm $C_p = 10^{22} m^{-3}$

Irradiation conditions

300 KeV Cu^+ T = 80, 293 and 803 K



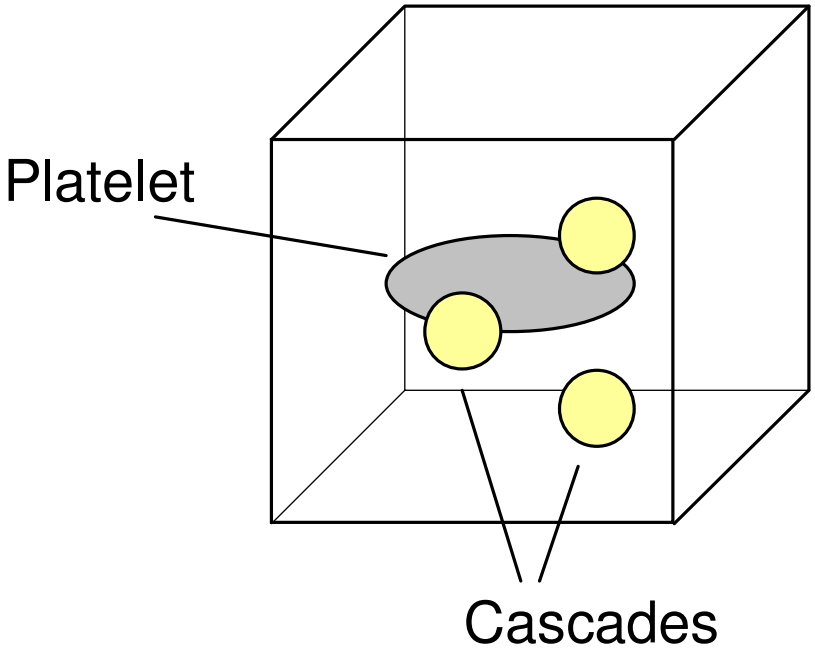
Results

- Mean diameter decreases with dose
- Size distribution function narrows
- Precipitate number density remains constant
- After 0.1 dpa precipitates disappeared completely below TEM resolution limit at 80 K and room temperature

N. Wanderka, C. Ramachandra, R. P. Wahi and H. Wollenberger, JNM, 1992

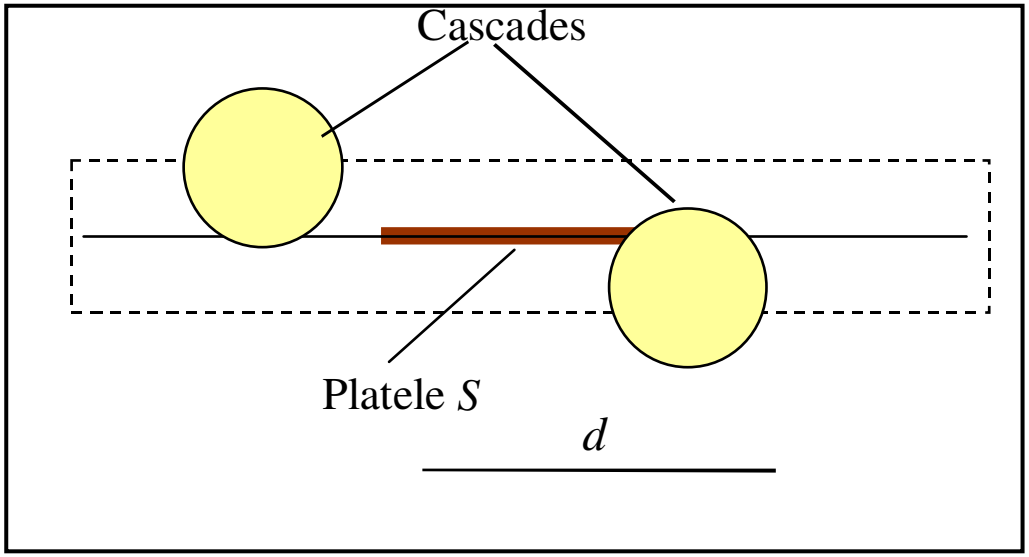
MC simulation of platelet destruction by cascades

A.A. Turkin et al, 1995



- The platelet thickness is smaller than its diameter and the cascade size
- The cascade intersecting the platelet dissolves the platelet in the cascade region
- No thermally-activated restoration process
- After each cascade impact the platelet restores the disk-shaped form

Evolution of platelet distribution function



Only one parameter controls evolution

$$\mu = \frac{4\pi r^3}{3\Omega n_d} = \frac{replacements}{displacements}$$

$$S(\phi) = S_0 \exp(-\mu\phi)$$

$$\phi = Kt$$

S_0 = initial area of the platelet
n_d = number of displacements per cascade
r = effective cascade radius(radius of mixed zone)

Distribution function

$$F(d,\phi) = F(d\exp(\mu\phi/2),0)\exp(\mu\phi/2)$$

Initial distribution

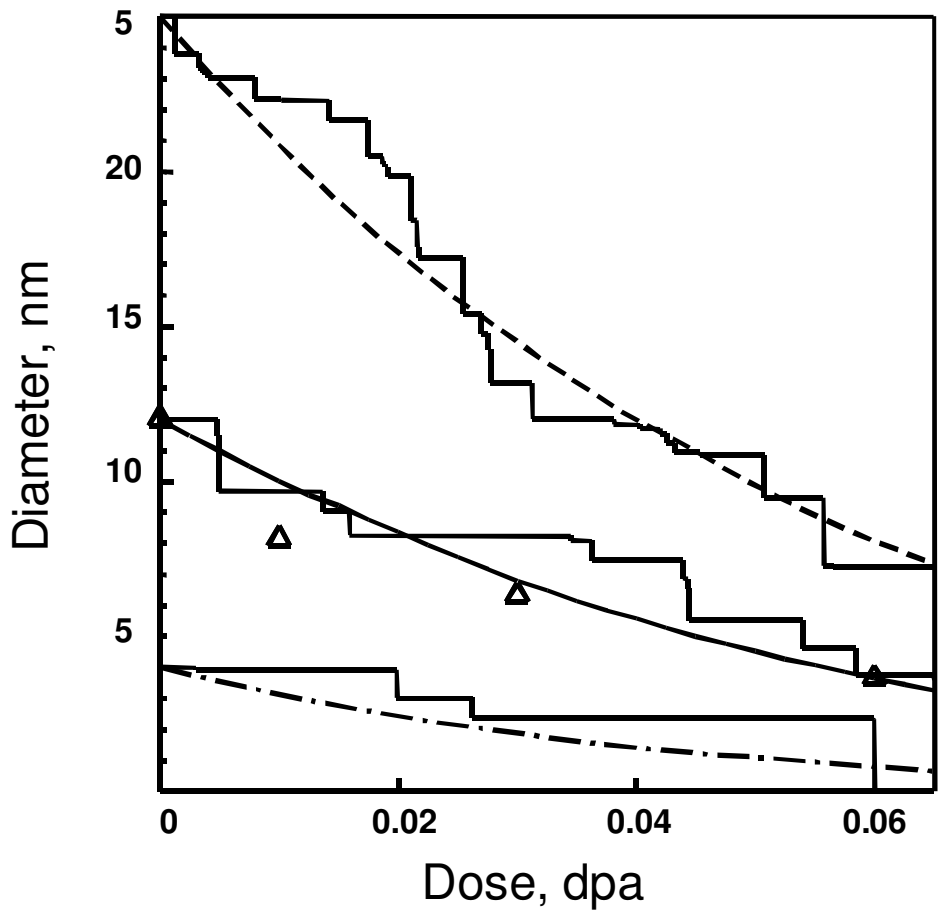
Mean diameter

$$\langle d \rangle = \langle d_0 \rangle \exp(-\mu\phi/2)$$

Platelet diameter vs dose

Dissolution curves for low temperature irradiation

Monte Carlo simulation & Analytical results



Platelets of initial diameters 4, 12 and 25 nm.

Smooth curves are the result of averaging over 500 run of the Monte Carlo simulation.

Stepped curves represent separate platelets (only one run).

Average number of cascades (number of steps) required for platelet dissolution depends on cascade size.

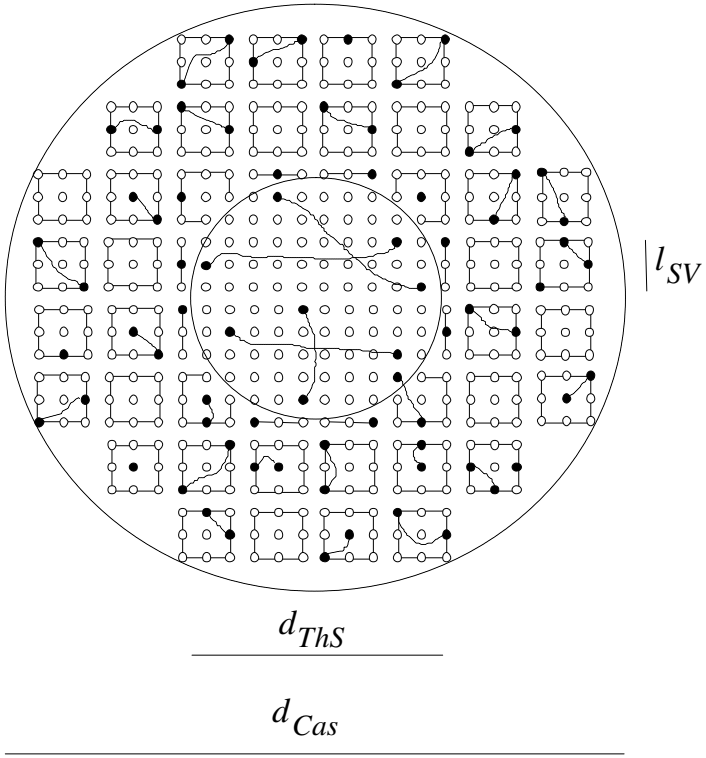
Open triangles show the experimental values of the mean diameter at room temperature

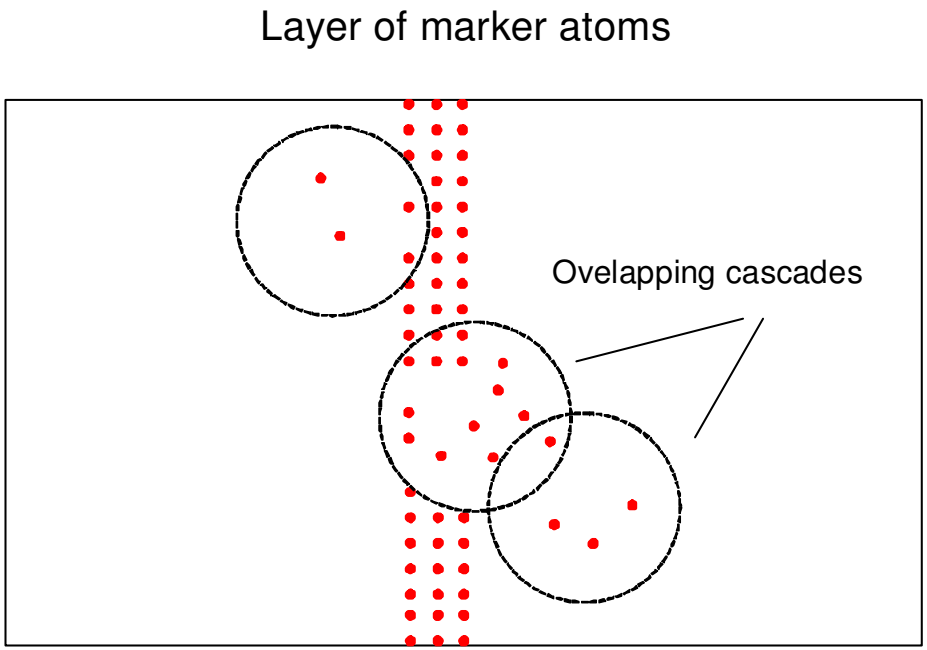
MC simulation of cascade-induced mixing

Model of atomic mixing inside cascade:

- ballistic mixing at the cascade periphery,
- thermal-spike mixing in the cascade core (liquid-like diffusion)

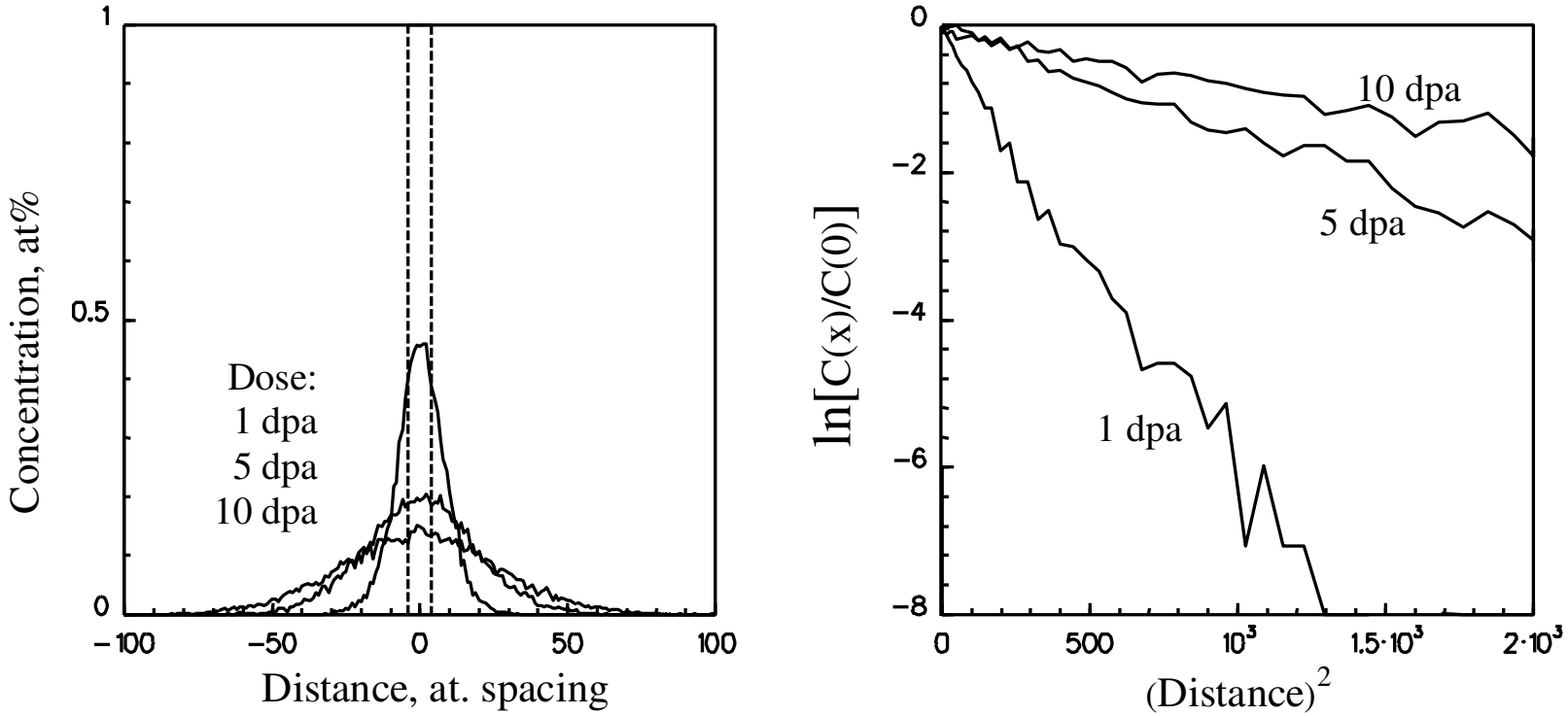
Cascade parameters correspond to MD simulations





A.A. Turkin, C. Abromeit, V. Naundorf, JNM,1996

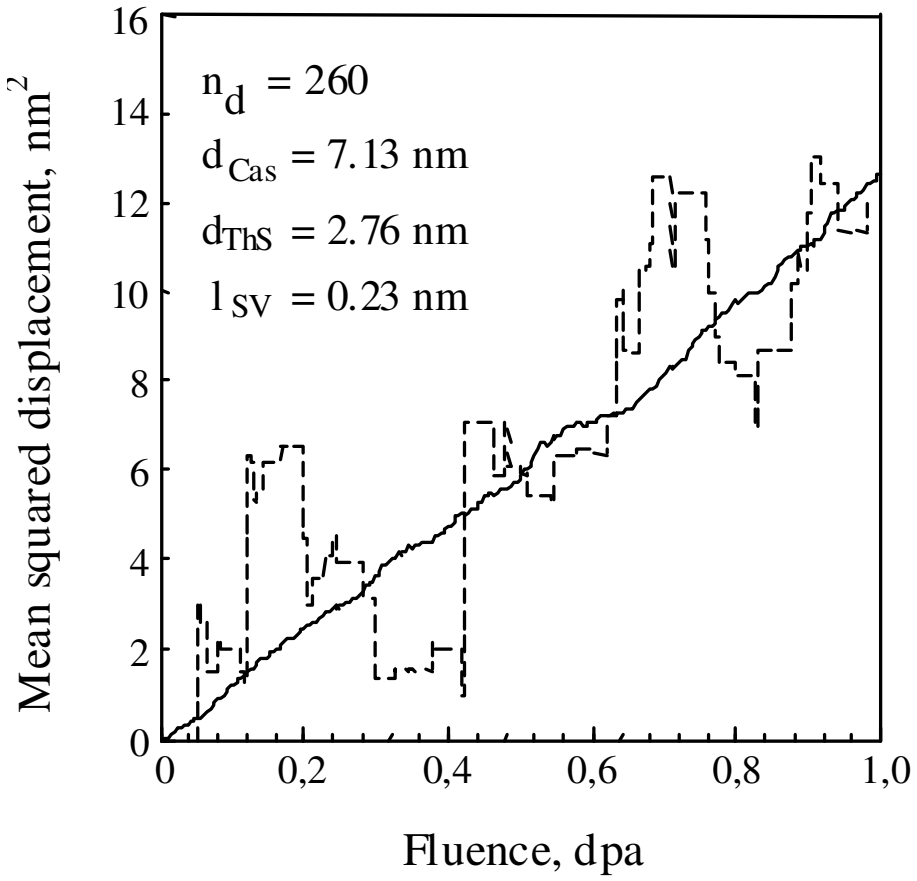
MC simulation of cascade-induced mixing



Due to overlapping of cascades the initial profile of marker atoms is smeared with time. At sufficiently high fluences the resulting profile is closely approximated by the Gaussian distribution

MC simulation of cascade-induced mixing

Fluence dependence of the mean squared displacement of a tracer atom. Dashed line - single run. Solid line - averaging over 200 runs.



Thank You.

I wish you every success!