Multi-scale modelling of the ageing kinetics of ultra-thin deposits

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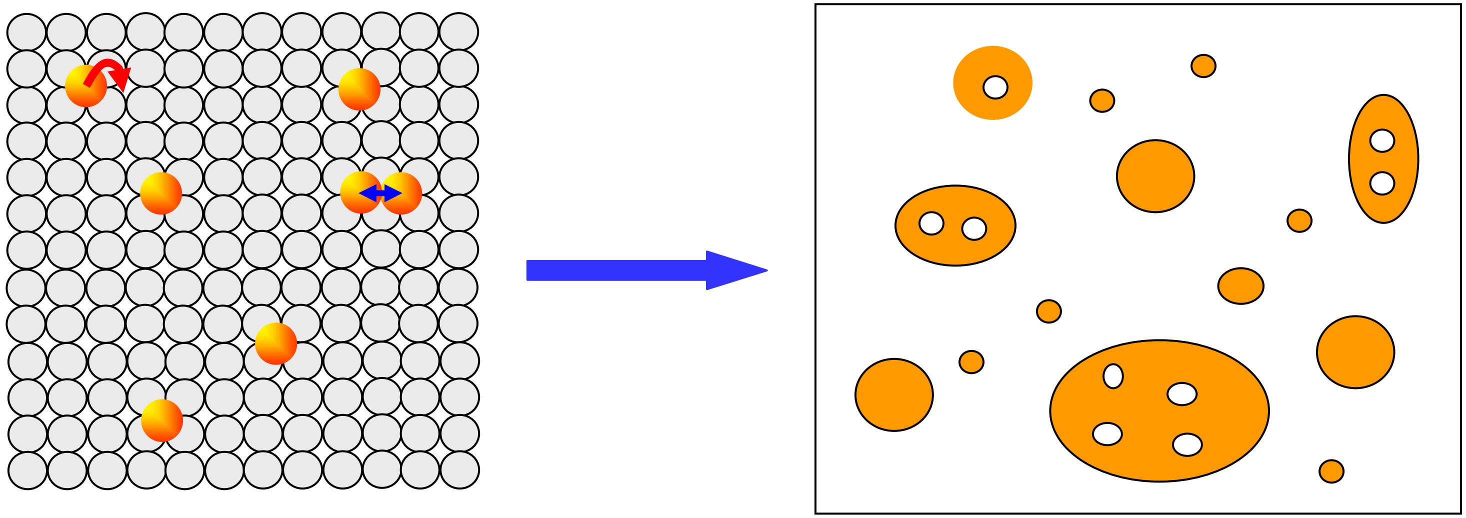
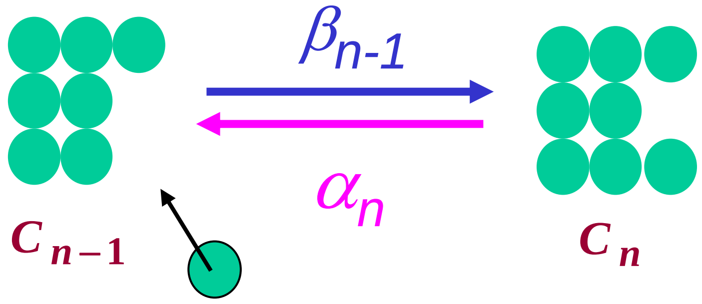


Mesoscopic scale : Cluster Dynamics



monomers are the only diffusing species

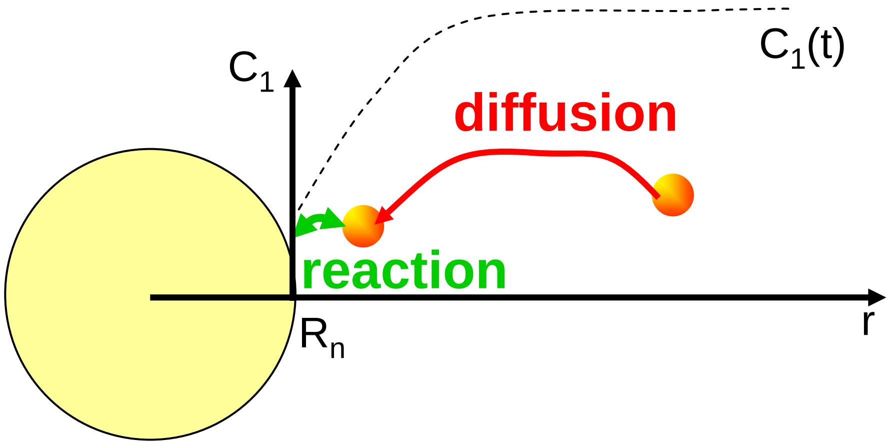
$$\frac{dC_n}{dt} = \beta_{n-1} C_{n-1} - (\alpha_n + \beta_n) C_n + \alpha_{n+1} C_{n+1}$$

emission **adsorption**

frequencies

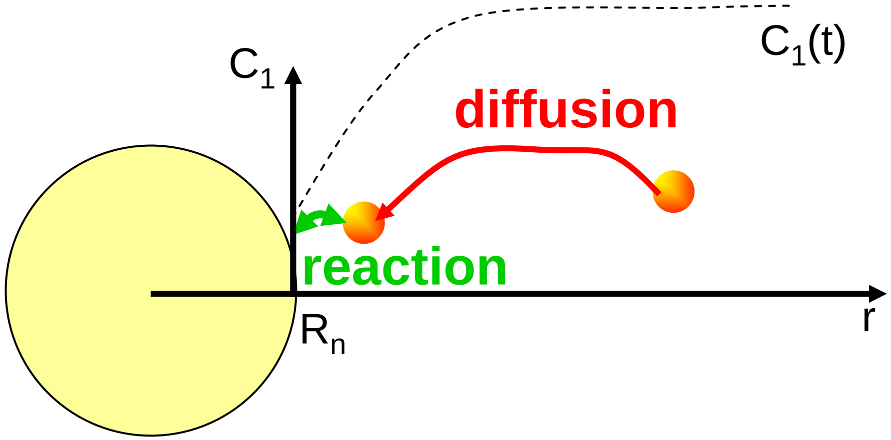
The consistency between the description at the atomic scale and this approach is related to the determination of β_n and α_n and their relation with the hopping frequencies at the atomic scale.

We consider the kinetics of monomer diffusion and the sticking reaction as two mechanisms in series



Kinetic controlled by diffusion ⇒

$$\begin{cases} \beta_n = m_n C_1(t) \\ \alpha_n = m_n C_1^{Inter,n} \end{cases}$$



$$\begin{cases} \beta_n = m_n C_1(t) \\ \alpha_n = m_n C_1^{Inter,n} \end{cases}$$

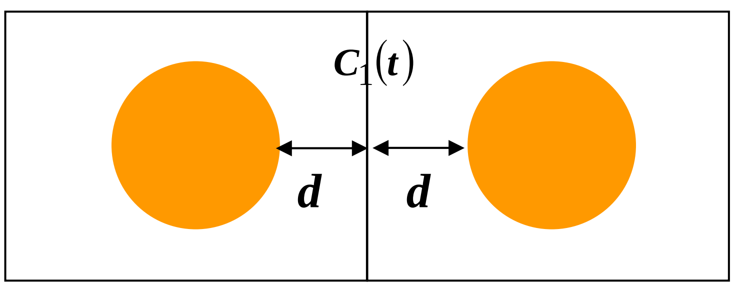
mesoscopic description

$$m_n = \frac{8D}{ln\left(r^{\infty} / R_n\right)}$$

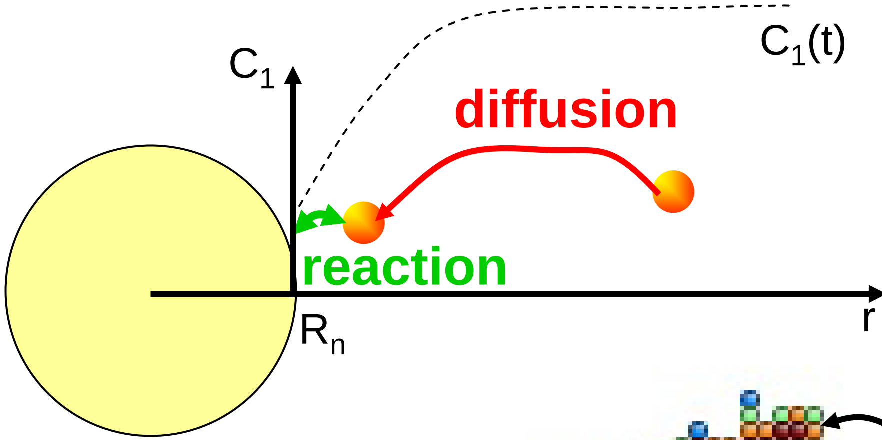
Diffusion coefficient

Radius at infinite distance =??
2D specific factor

Interactions of diffusion fields

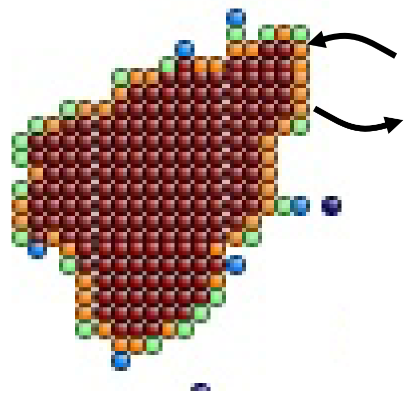


$$m_n(t) = \frac{8D}{ln\left(1 + 2\,d(t) / \sqrt{n}\right)}$$



$$\begin{cases} \beta_n = m_n C_1(t) \\ \alpha_n = m_n C_1^{Inter,n} \end{cases}$$

atomistic description



k_n^{det}, k_n^{att}

Assumption that clusters retain their equilibrium morpholgy

$$C_1^{Inter,n} = k_n^{det} / k_n^{att}$$

$$C_1^{Inter,n} = C_1^{eq} \exp\left(\frac{2\sigma}{kT\sqrt{n}}\right)$$

edge free energy

Concentration of monomers for saturated solution

Model of nucleation-growth-coarsening (MNGC)

Quasi-continuous model

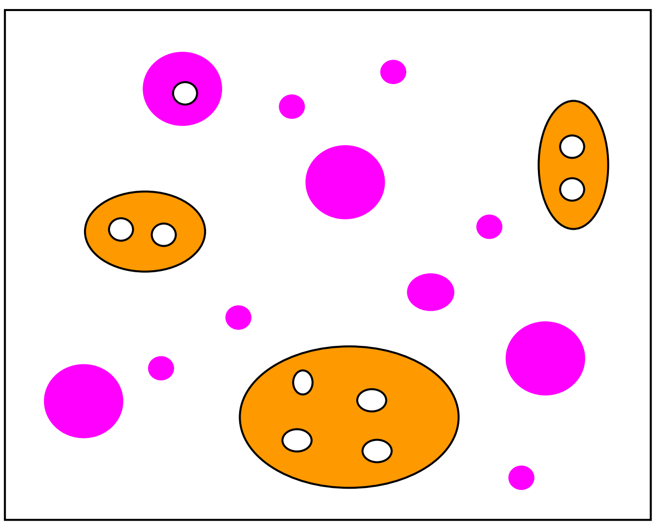
$$v_n = \frac{dn}{dt} = \beta_n - \alpha_n = m_n(t)\left(C_1(t) - C_1^{eq}\exp\left(\frac{2\sigma}{kT\sqrt{n}}\right)\right)$$

Ensures the coherence between CD and MNGC

n(t) → **n(t+Δt)** → $\theta_{PP}(t+\Delta T) = \sum_{n\in PP} nC_n(t+\Delta T)$

CNT: **J(t)**

***PP* : Precipitate Phase**



$$\theta_{SS}(t+\Delta t) = \theta - \theta_{PP}(t+\Delta t)$$

$$\theta_{SS}(t+\Delta t) = \sum_{n<n^*(t+\Delta t)} nC_n(t+\Delta t)$$

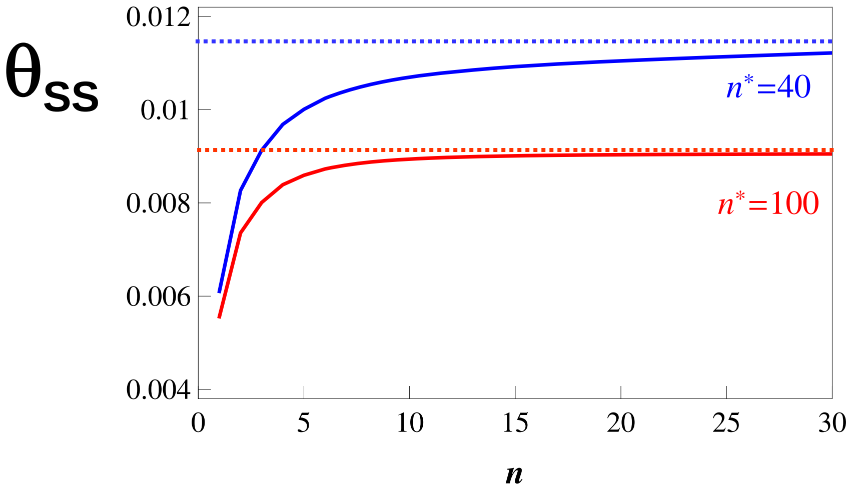
***SS* : Solid Solution**

constrained equilibrium

$$C_{n<n^*(t)}(t) = C_n^{eq}\, exp\left(\frac{2n\sigma}{kT\sqrt{n^*(t)}}\right)$$

Critical size
≡
Driving force

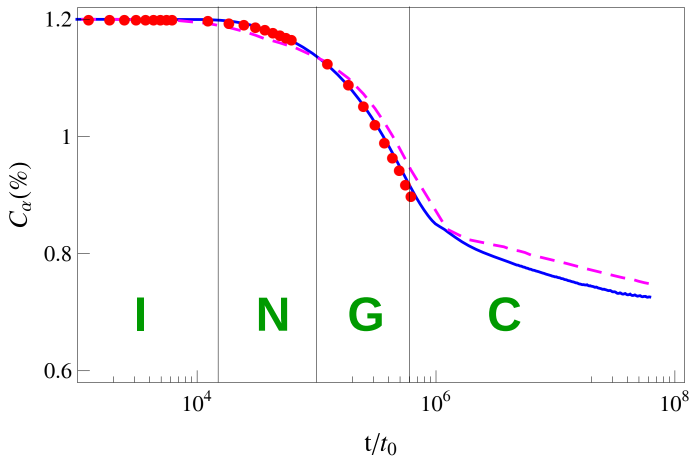
$$\theta_{SS}(t) = \sum_{n<n^*(t)} nC_n^{eq}\, exp\left(\frac{2n\sigma}{kT\sqrt{n^*(t)}}\right)$$



$$\begin{cases} C_1(t) \\ J(t) \end{cases} \rightarrow \theta_{PP}(t+\Delta t) \rightarrow \theta_{SS}(t+\Delta t) \rightarrow n^*(t+\Delta t) \rightarrow \begin{cases} C_1(t+\Delta t) \\ J(t+\Delta t) \end{cases}$$

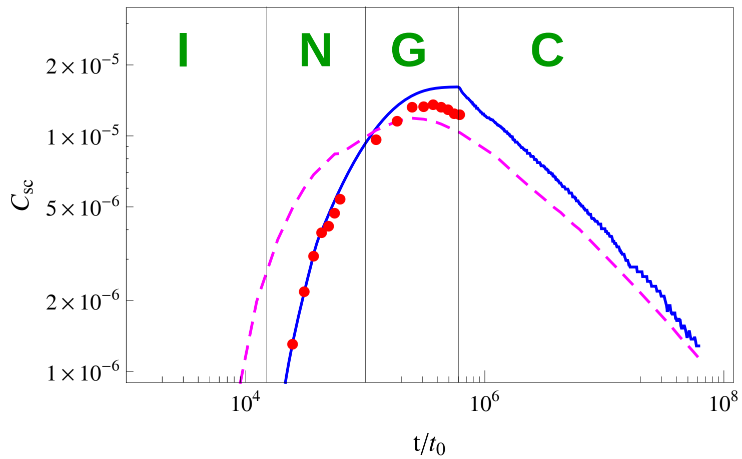
Iterative loop

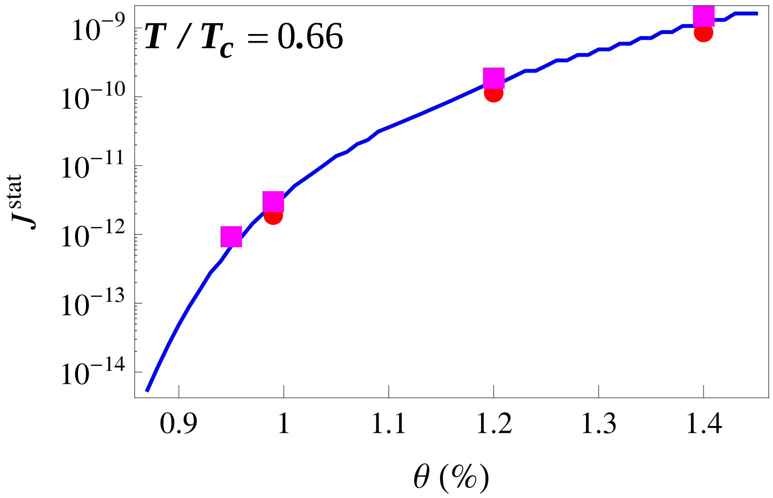
KMC / CD / MNGC



$\theta = 0.012$

$T\,/\,T_c = 0.66$

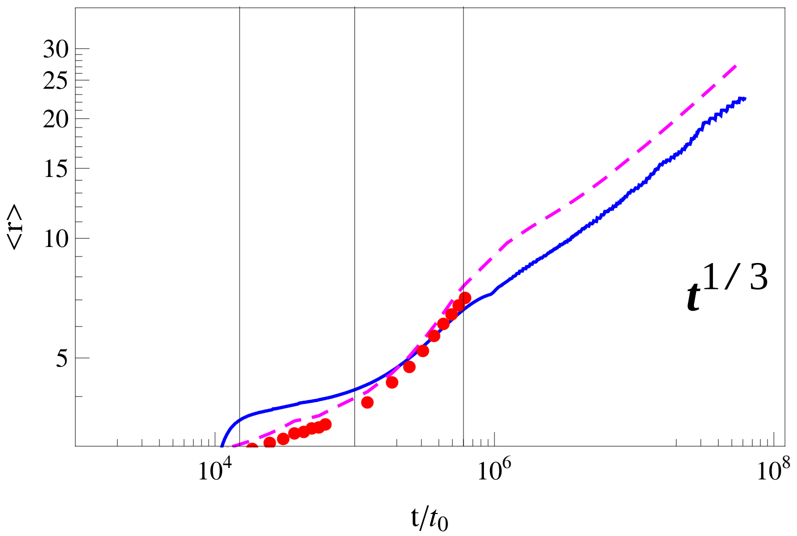


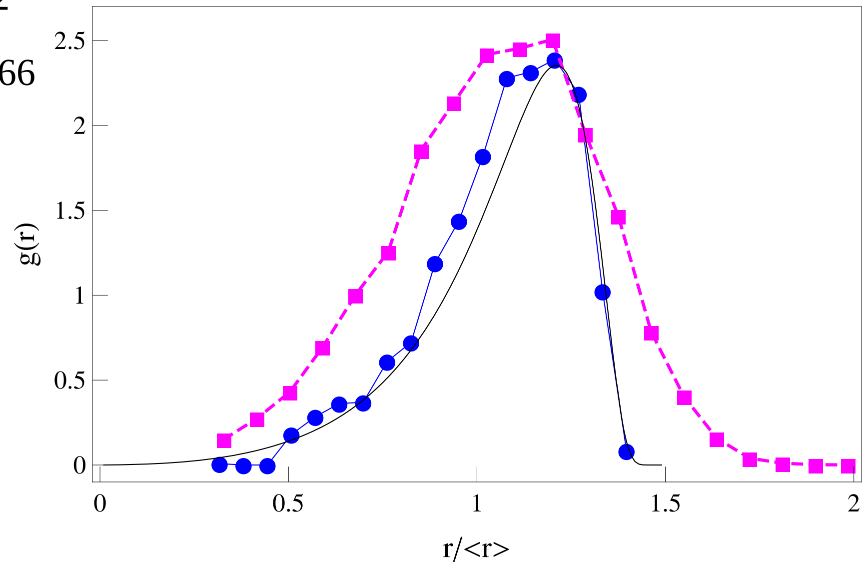


Coarsening : CD / MNGC / LSW

$\theta = 0.012$

$T / T_c = 0.66$





CD : «stochastic » model

MNGC : deterministic model

LSW : deterministic model

Conclusions

Consistency between KMC /CD / MNGC requires :

- σ : interfacial free energy
- interactions of diffusion fields
- constrained equilibrium

→ **Extension to concentrated alloys**