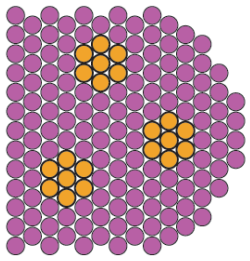
The kinetic pathway of coarsening morphology of a Ni-Al-Cr alloy by Lattice Kinetic Monte Carlo simulation

Zugang Mao[1], Georges Martin[1,2] and David N. Seidman[1]

Department of Materials Science

[1]Northwestern University, **[2]CEA, Paris, FRANCE**

NUCAPT
Northwestern University
Center for Atom-Probe
Tomography



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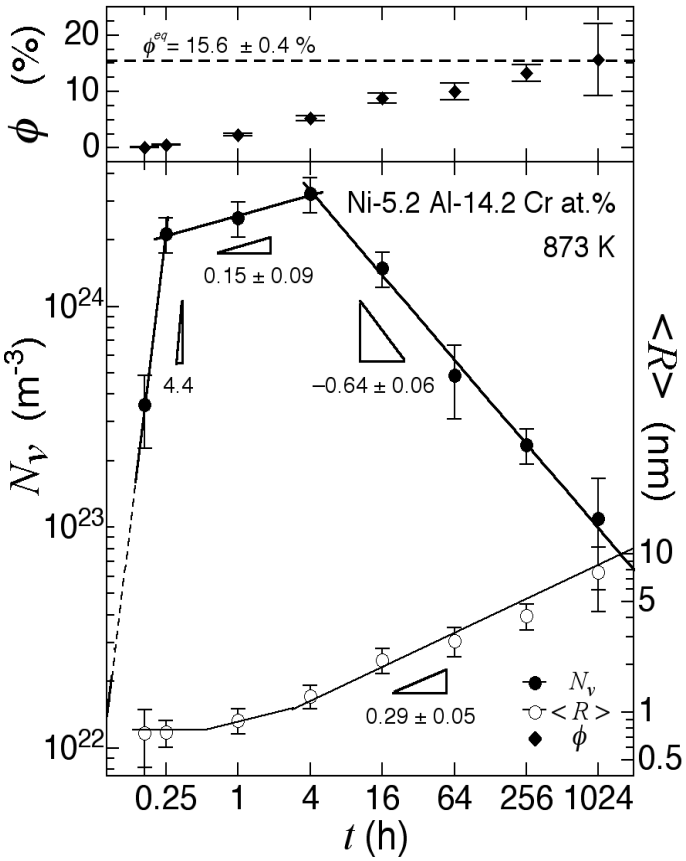
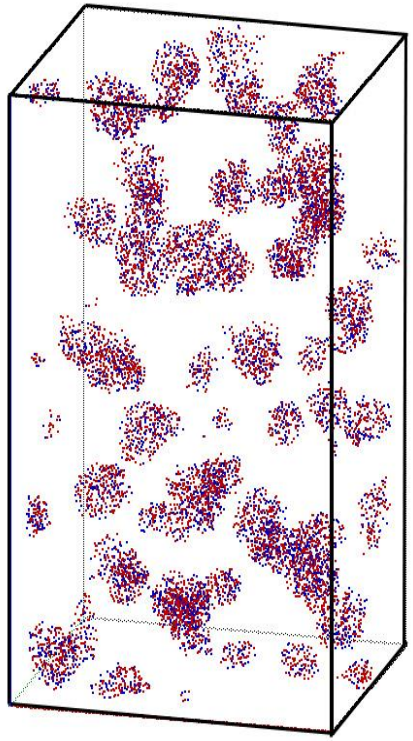
Coherent Phase Transformations with atomic resolution

Quantitative with good statistics & qualitative features

Ni- 5.24 Al-14.24 Cr at. % at 600ºC *(Sudbrack, Seidman et al. 2004)*





Necked precipitates;
density peaks at ≈30% at ≈4 hours

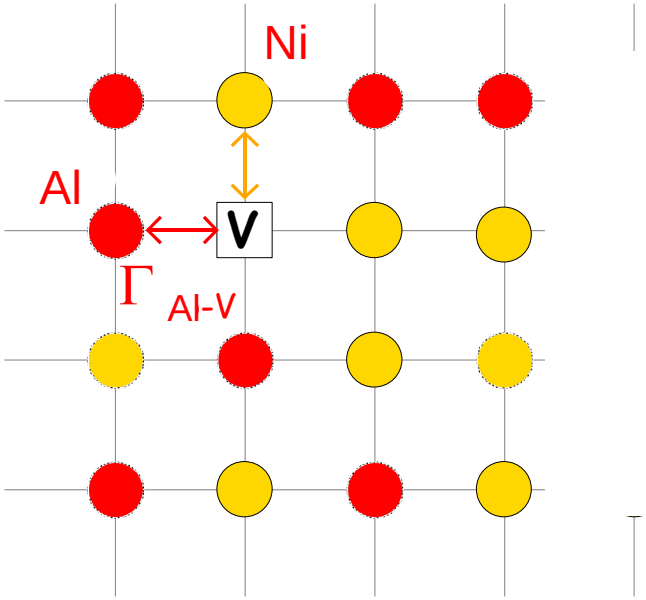
Misfit ≈ 6 10^{-4}

Nothing but vacancy jumps

Kinetic Monte Carlo & Residence time algorithm

$N_{sites} = N_A + N_B + 1_V$

$N_{chan} \approx Z$ channels out of {i}



$$\tau_i = \left(\sum_{j=1}^{N_{chan}} \Gamma_{ij} \right)^{-1}$$

$$P(j, t+\tau_i; i, t) = \Gamma_{ij} \times \tau_i$$

$$t = t + \tau_i$$

Physical time (C_v)

Set of attempt frequencies Γ_{ij} ?

Set of attempt frequencies Γ_{ij} ?

The jump frequency of a vacancy is:

$$\Gamma_{ij} = \nu_j \exp(-\Delta E_a / k_B T)$$

$$\Delta E_a = E_{sp} - E_j$$

The configurational energy:

$$E_j = \sum_{k \in nn(j)} \varepsilon_{jk} + \sum_{j \in nn(V),\, j \neq k} \varepsilon_{jV}$$

Parameterization of LKMC

Statistics: from first principle DFT-LDA (CASTEP + Chen MÖbius inversion lattice technique)

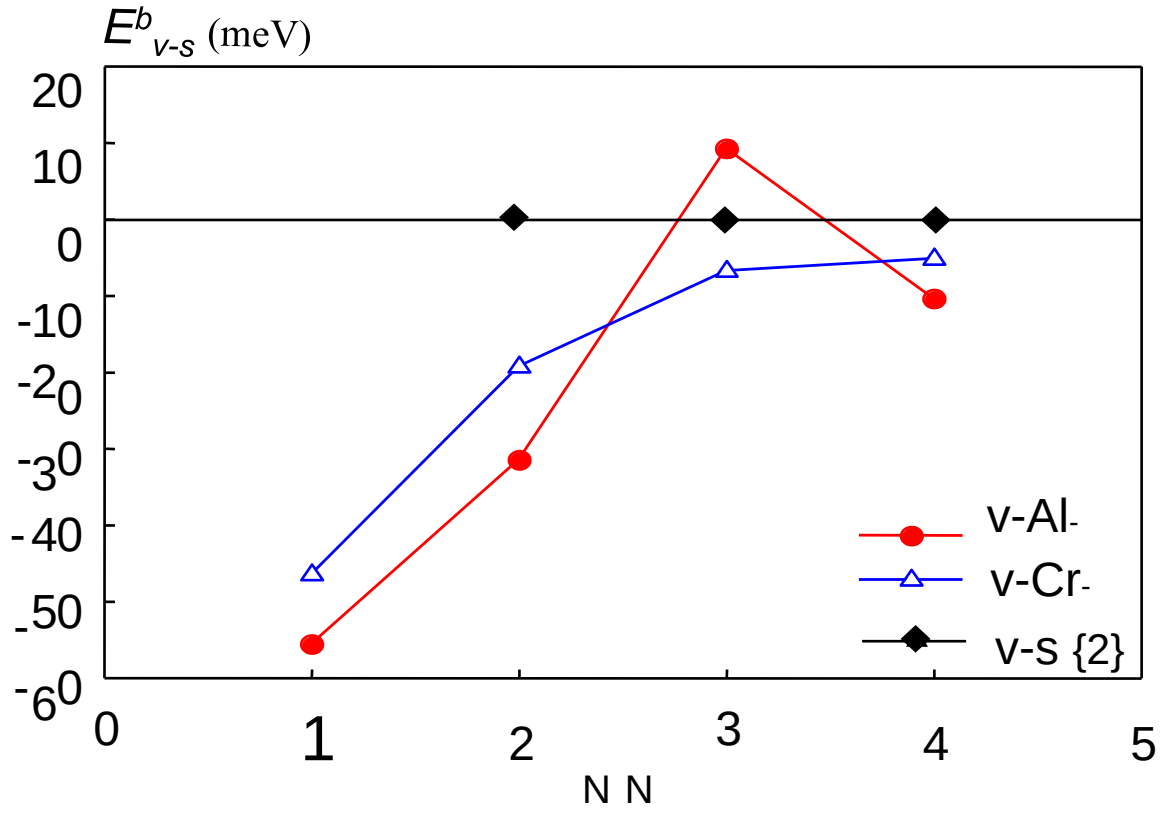
$\varepsilon^{\alpha,\alpha'}$ (eV)	Ni-Ni	Al-Al	Cr-Cr	Ni-Al	Ni-Cr	Al-Cr	V-Ni (1)/(2)	V-Al (1)/(2)	V-Cr (1)/(2)
1st NN	-0.7485	-0.5786	-0.6845	-0.7495	-0.7582	-0.6963	-0.178	-0.221	-0.223
2nd NN	-0.0135	-0.0265	-0.0112	0.0349	0.0257	0.0225	0/ $\varepsilon^{Ni,Ni}$	0/ $\varepsilon^{Al,Ni}$	0/ $\varepsilon^{Cr,Ni}$
3rd NN	0.0142	0.0084	-0.0185	-0.0285	0.00526	0.0211	0/ $\varepsilon^{Ni,Ni}$	0/ $\varepsilon^{Al,Ni}$	0/ $\varepsilon^{Cr,Ni}$
4th NN	-0.00664	-0.0121	-0.00945	0.0125	-0.0166	0.115	0/ $\varepsilon^{Ni,Ni}$	0/ $\varepsilon^{Al,Ni}$	0/ $\varepsilon^{Cr,Ni}$

Kinetics: fit to impurity diffusion coefficient in Ni (same as Pareige et.al. Acta Mater. 1999)

	Ni	Al	Cr
$E^{\alpha}_{sp-i.j}$ (eV)	-9.750	-9.412	-9.862
ν^{α} (s^{-1}) (1)/(2)	1.10×10^{15}/1.28×10^{15}	1.10×10^{15}/2.26×10^{15}	8.7×10^{14}/1.84×10^{15}

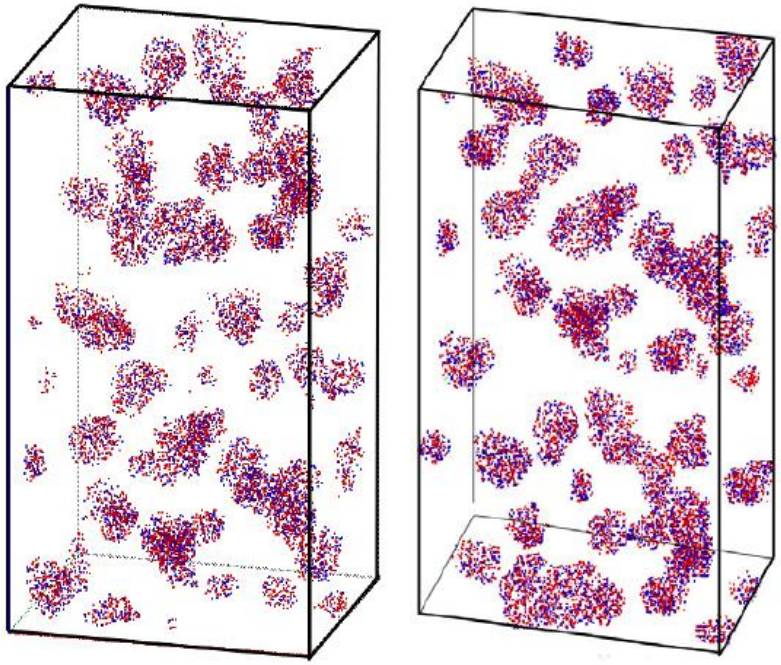
Vacancy-solute binding energies LKMC

First principle DFT-LDA => ***Long range vacancy solute binding*** **{1}**

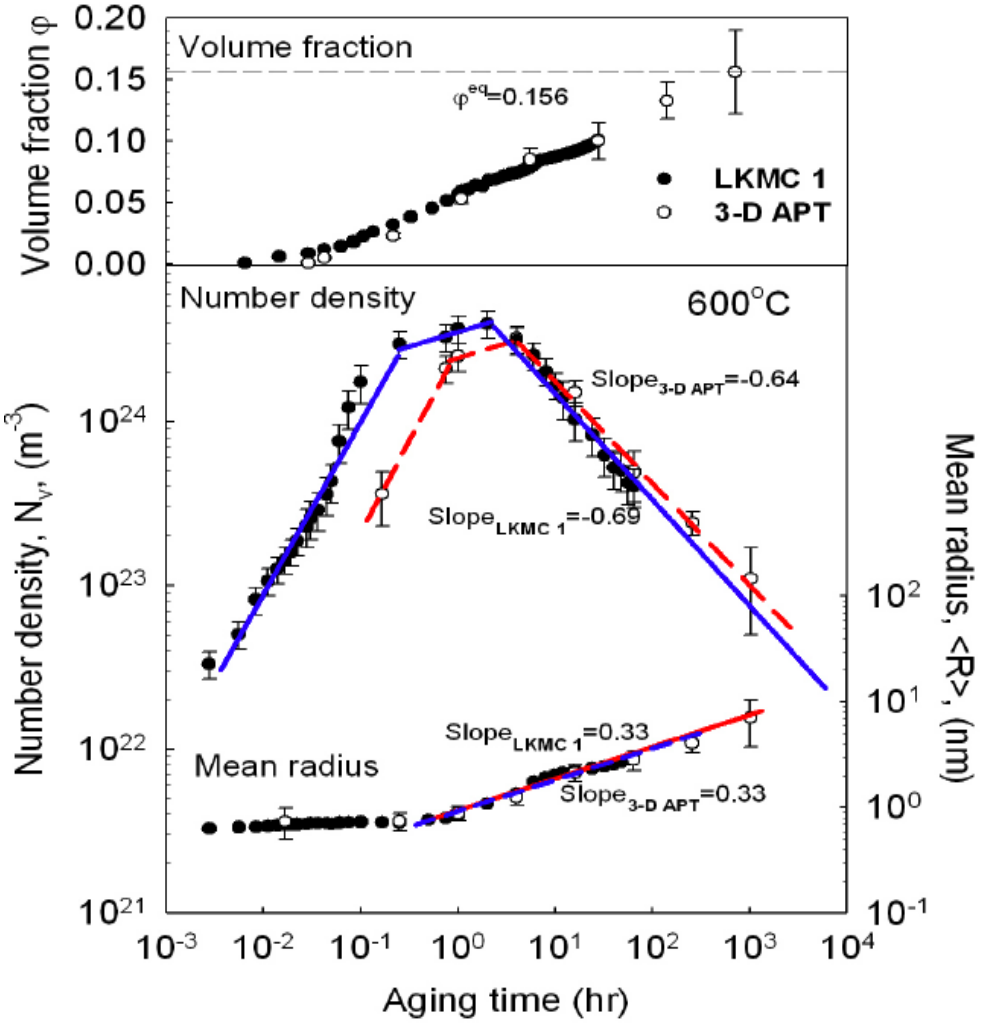


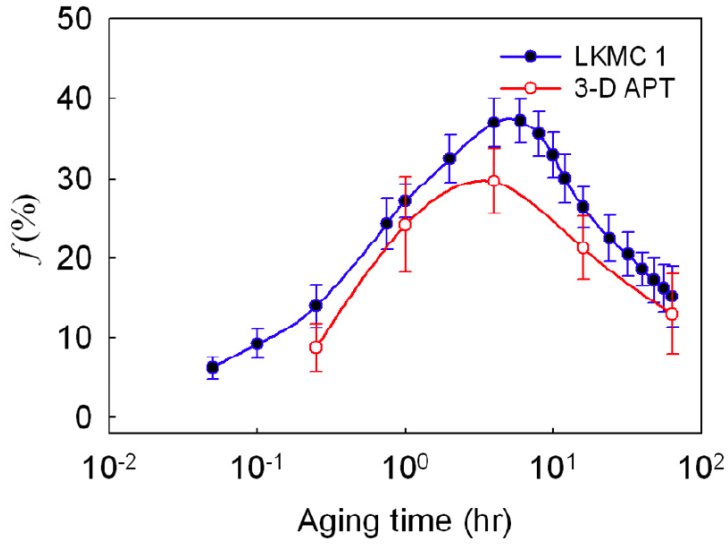
LKMC-1 => morphological features & quantitative OK

3D APT LKMC 1



3D-AP / KMC-1 (long range v-s binding)





LKMC-2 : no long range s-v binding => no necking

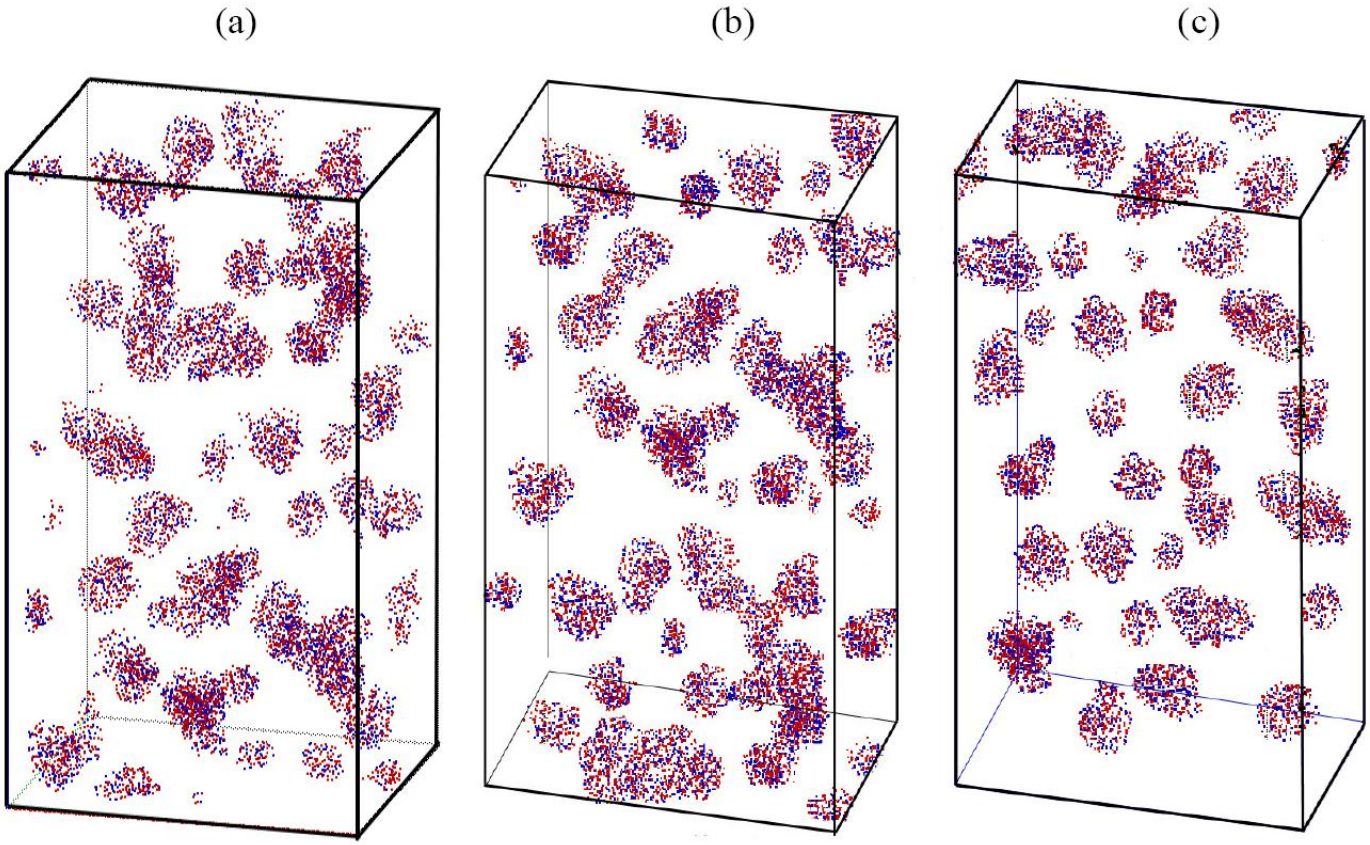
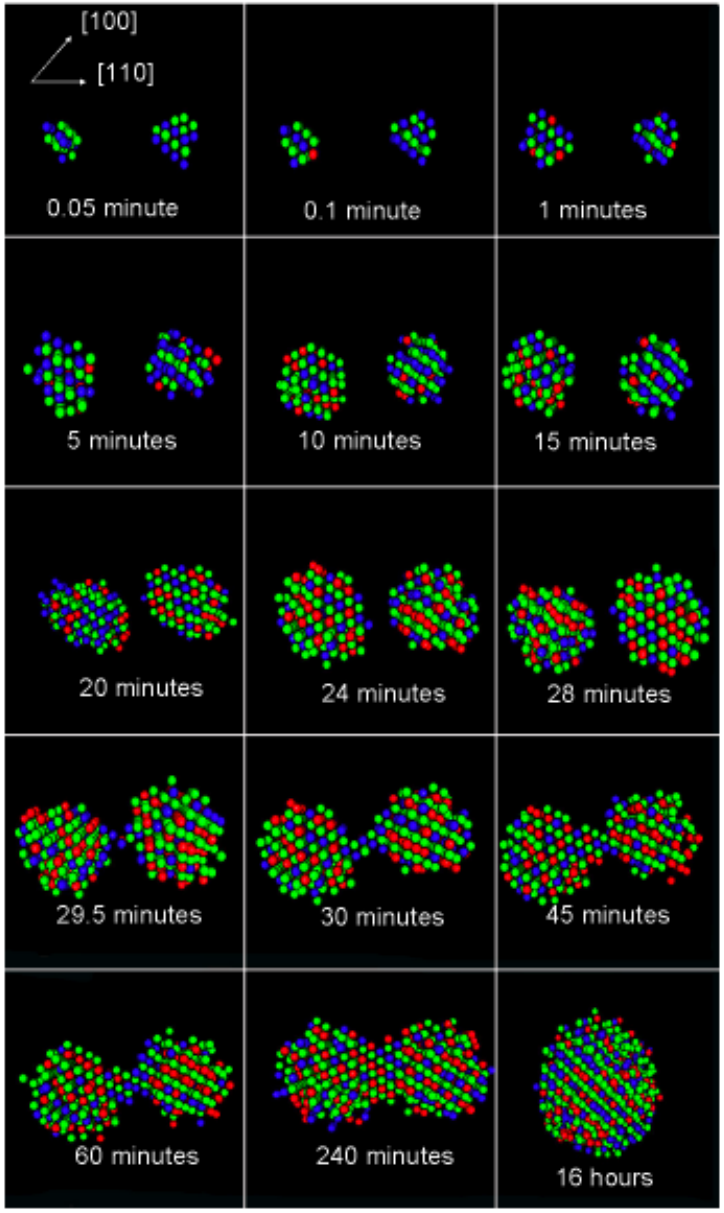


Figure 1:The morphology of γ'-precipitates in Ni 5.2 Al 14.2 Cr at.% after aging at 873 K: (**a)** As obtained from 3-D APT experiments after 4 hours;
(**b)** as simulated by LKMC with parameter set 1;
(**c)** as simulated by LKMC with parameter set 2.

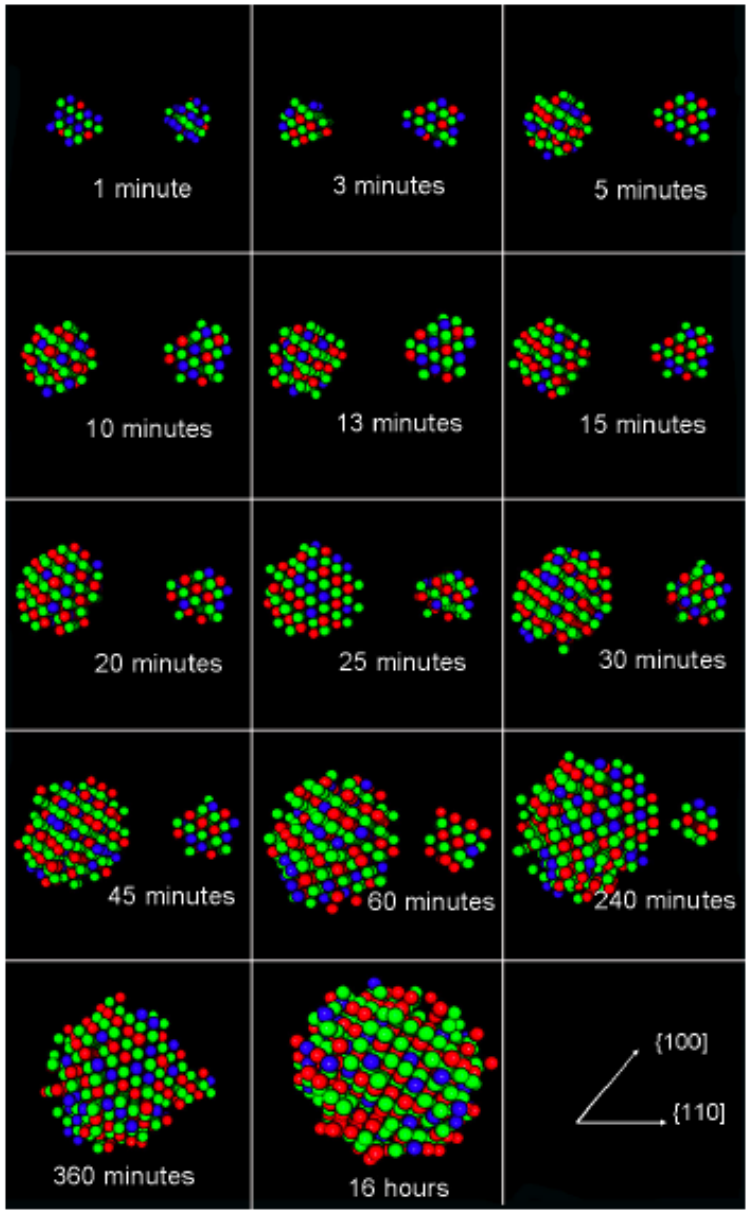
Necking is kinetics



Long range s-v binding

ON

OFF



Necking is triggered by kinetics, not by thermodynamics

dilute solutions — migration of clusters (Soisson, Bellon)

coagulation

concentrated solutions — *correlated diffusion*

Diffusion?=> L, D, fast / medium

$$\tilde{J} = -\overline{\overline{L}}\,\nabla\tilde{\mu} \quad = -\overline{\overline{D}}\,\nabla\tilde{C}\;\Omega^{-1} \quad \overline{\overline{D}} = \overline{\overline{L}}\;\overline{\overline{\chi}}$$

$$L_{\alpha\beta} = (\Omega kT)^{-1}\sum_{m,n}\frac{<\Delta\mathbf{r}_\alpha^m\cdot\Delta\mathbf{r}_\beta^n>}{6t}$$

KMC equilibrium terminal solid solution

$$\chi_{\alpha\beta} = \frac{\partial(\mu_\alpha - \mu_v)}{\partial C_\alpha}$$

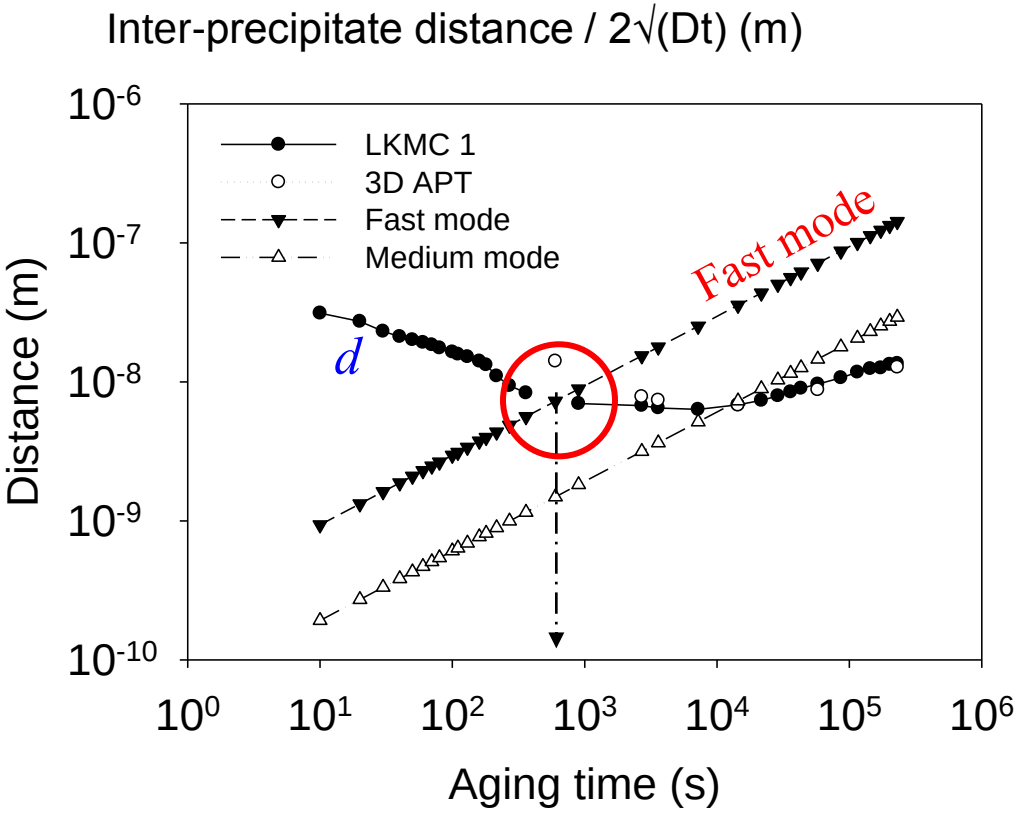
$$\overline{\overline{D}} = \begin{pmatrix} D_{fast} & 0 & 0 \\ 0 & D_{medium} & 0 \\ 0 & 0 & D_{slow} \end{pmatrix}$$

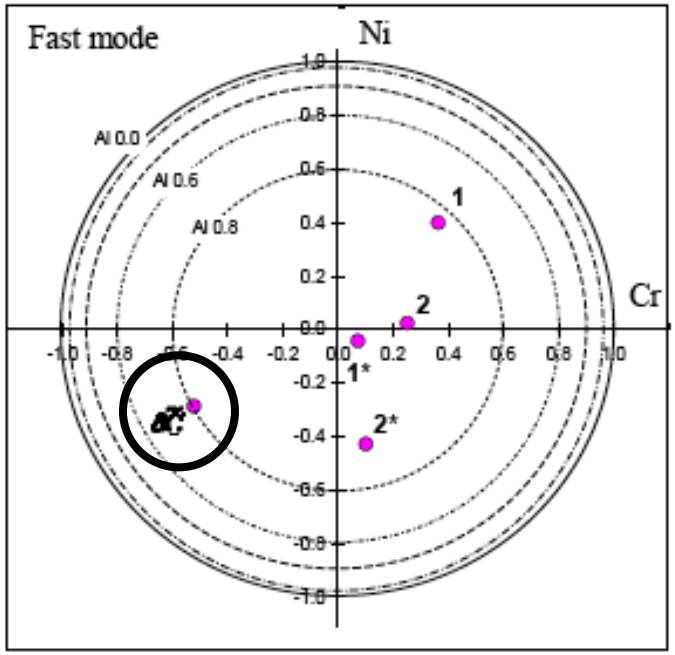
$D_{fast} \approx 2 \;\; 10^{-20}\ m^2s^{-1}$

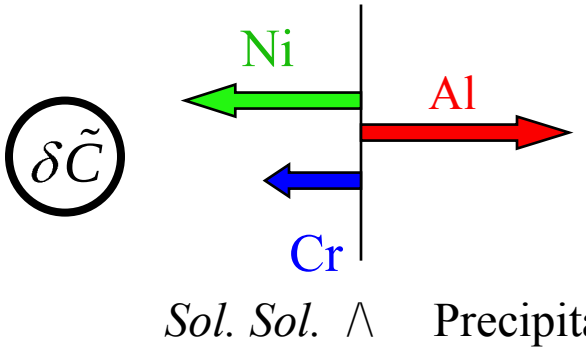
$D_{medium} \approx 9 \;\; 10^{-22}\ m^2s^{-1}$

$D_{slow} << D_{medium}$

Fast diffusion mode dominates early stage morphogenesis

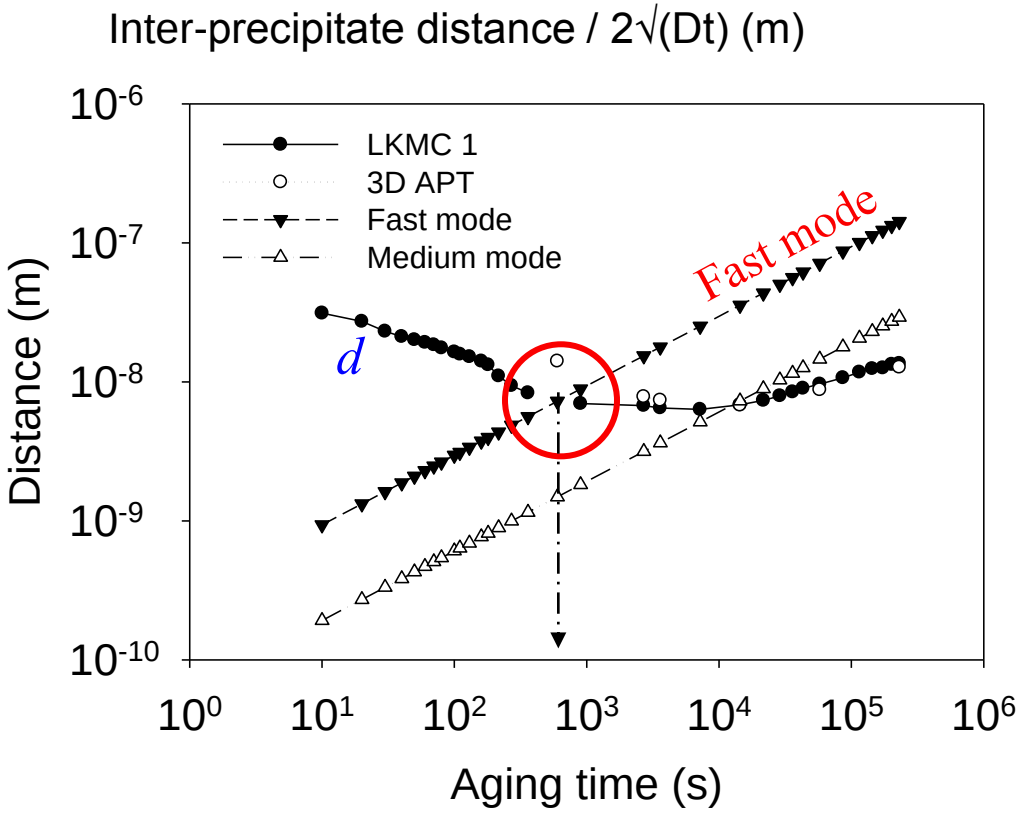


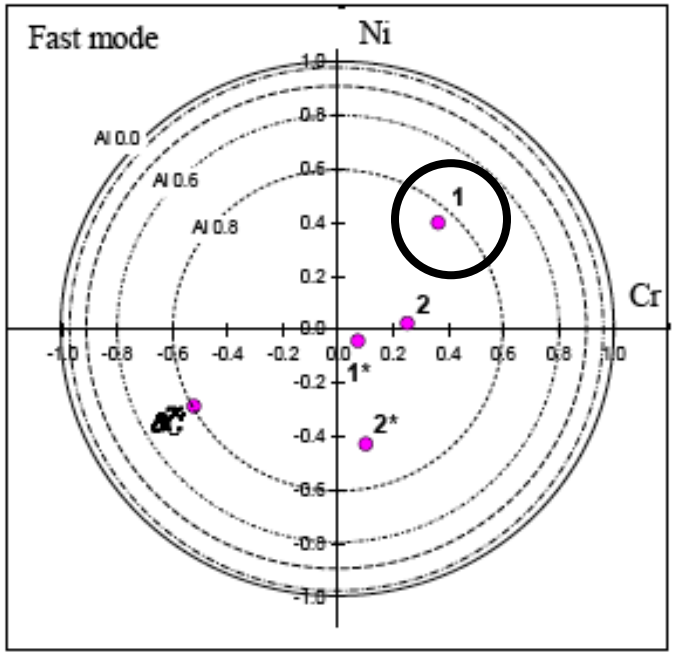




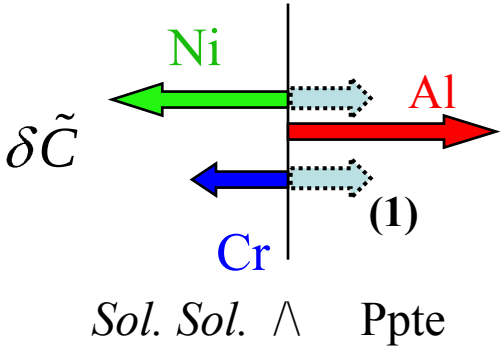
Sol. Sol. /\ Precipitates

Fast diffusion mode dominates early stage morphogenesis



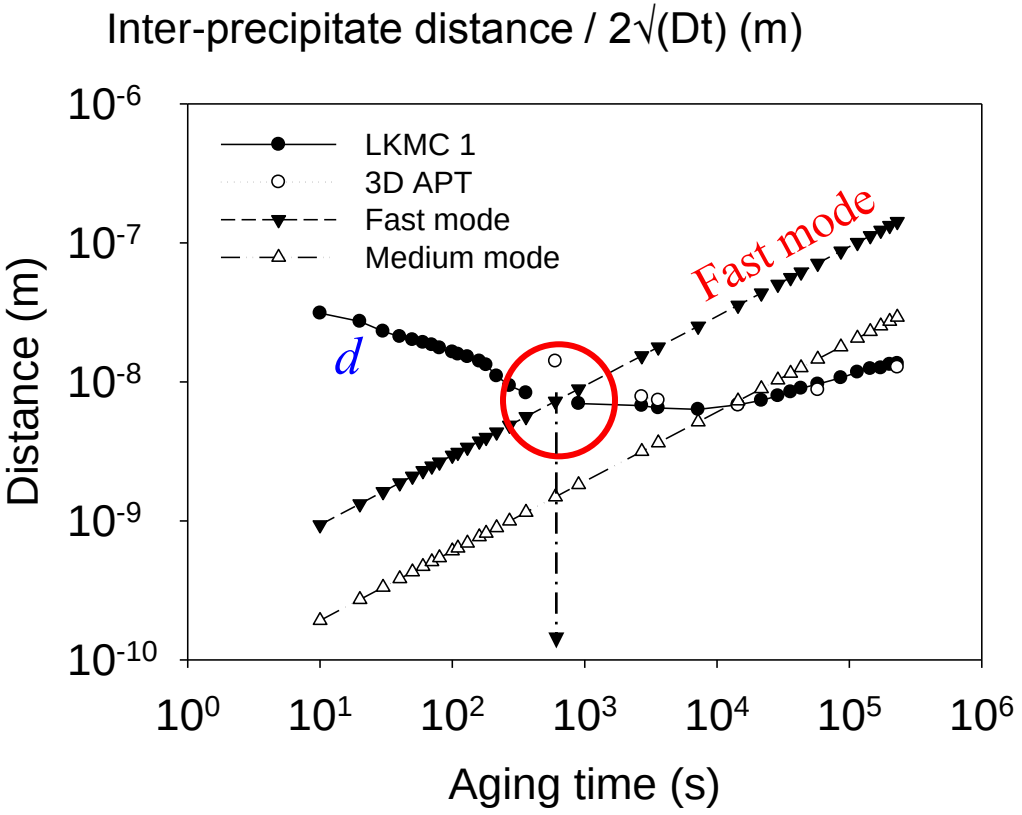


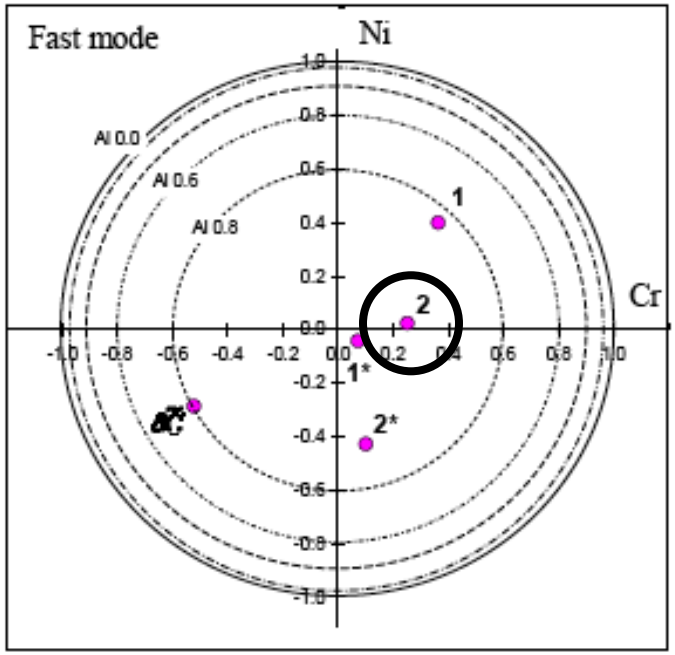
KMC-(**1**)
Long range
v-s binding



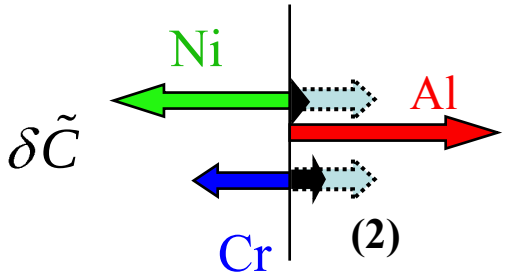
Sol. Sol. /\ Ppte

Fast diffusion mode dominates early stage morphogenesis





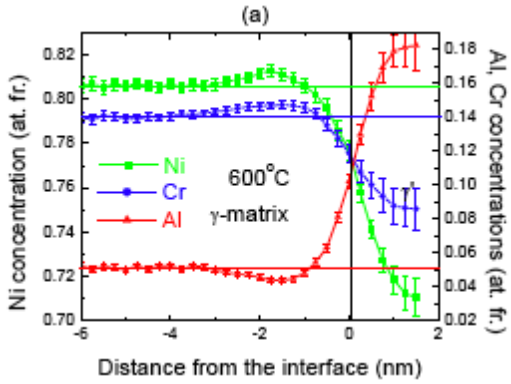
KMC-**(2)**
Zero long range
v-s binding



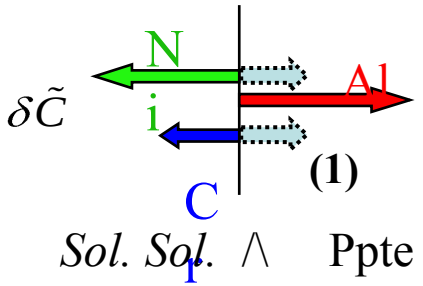
$\delta\tilde{C}$

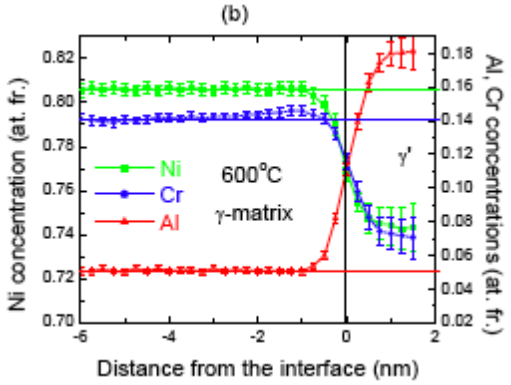
Sol. Sol. /\ Ppte

Kinetic correlations in fast mode oppose optimum coupling

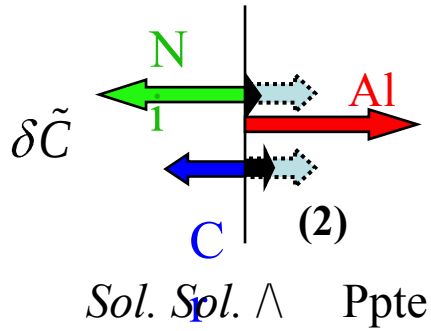


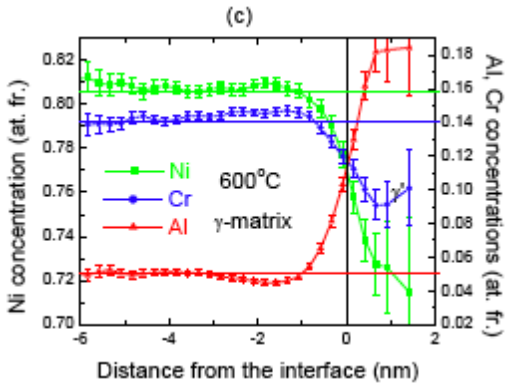
KMC-1
Long range v-s binding





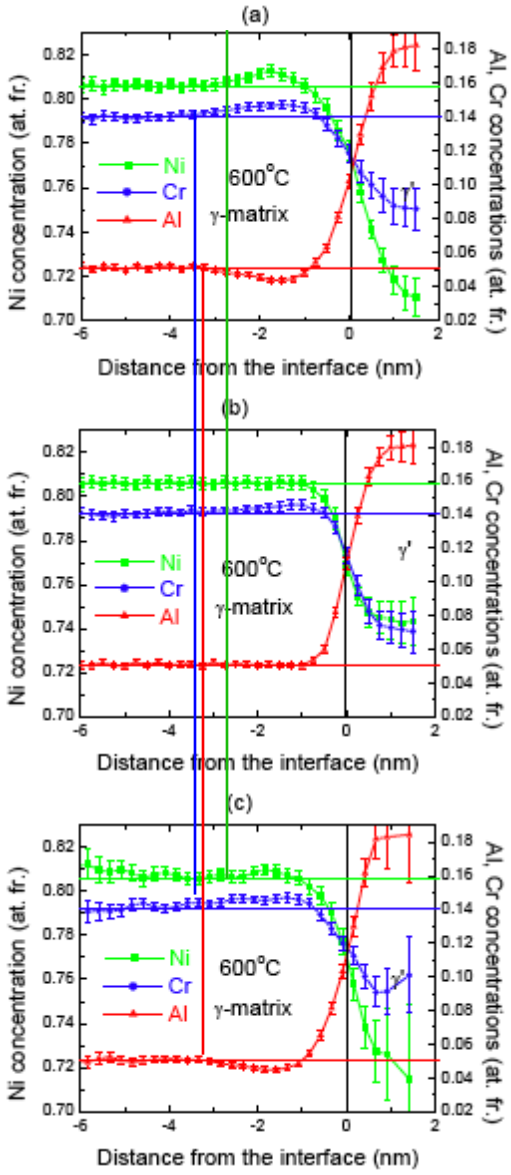
KMC-2
Zero long range v-s binding





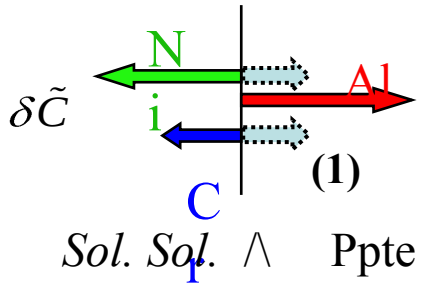
3D-APT

Kinetic correlations in fast mode oppose optimum coupling



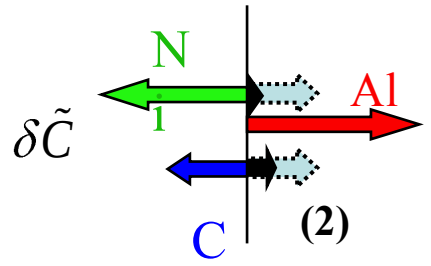
KMC-1

Long range v-s binding



KMC-2

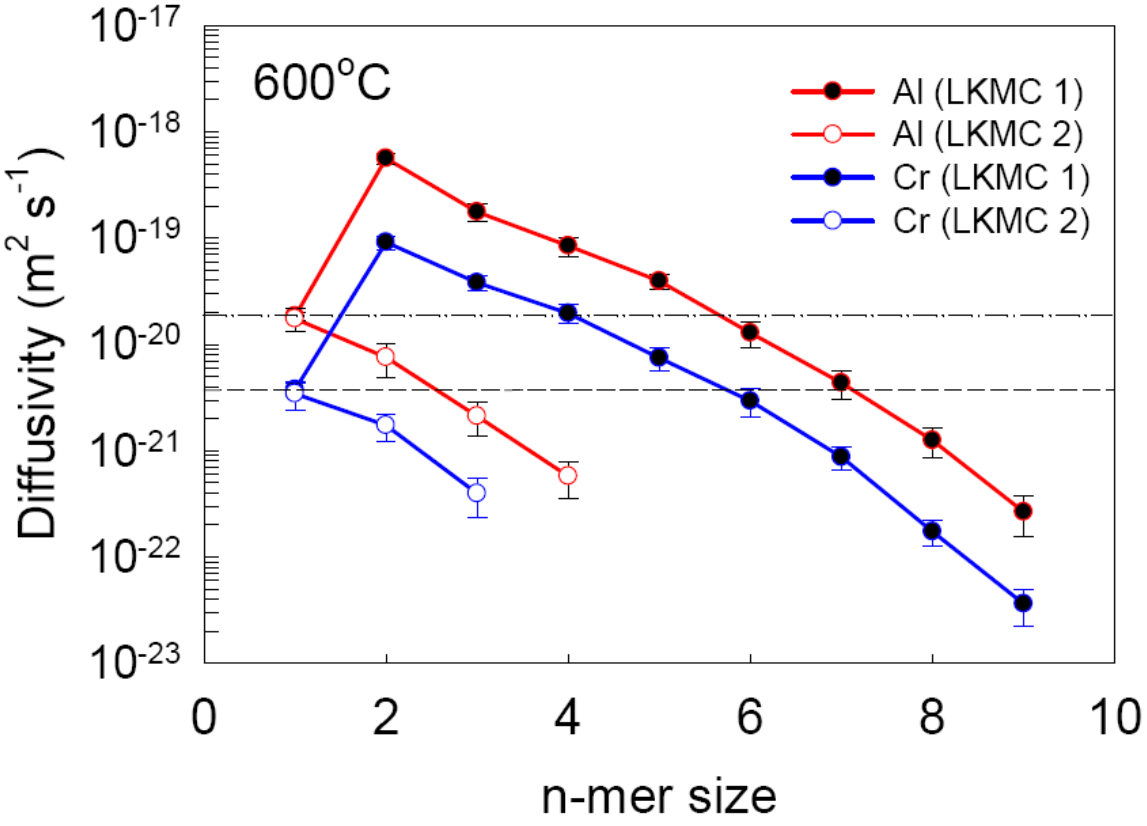
Zero long range v-s binding



3D-AP

Kinetic correlations broaden interface % profiles
⇒Inter-Ppte chemical interactions
⇒Necking

Highly correlated solute cluster diffusion



The significance of the diffusion of solute clusters (n-mers) in a Ni-Al or a Ni-Cr alloy. Diffusion coefficient (m^2 s^{-1}) of Al- and Cr-clusters (n-mers), as a function of the number of atoms (n) in the cluster: black lines for parameter set 1and red lines for parameter set 2.

Conclusions

Using the very same LKMC
to study correlation effects in diffusion
and to simulate coherent phase separation
+
Comparison with 3D-APT

Reveals :

$\Rightarrow$ New mechanisms (necking, elimination of APB's…)
$\Rightarrow$ Excellent quantitative agreement with observations in real alloys
$\Rightarrow$ Role of Off-diagonal terms of Onsager matrix in the morphogenetic process

This work has been published in Nature Materials, March 2007, and thanks C. Sudbrack and K. Yoon provide APT results.