Contact conditions at migrating interfaces

E. Gamsjäger[1], J. Svoboda[2] and F. D. Fischer[1,3]

[1]Montanuniversität Leoben, Institut für Mechanik, Austria
[2]IPM, Academy of Sciences of the Czech Republic, Brno, Czech Republic
[3]Österreichische Akademie der Wissenschaften, ESI, Austria

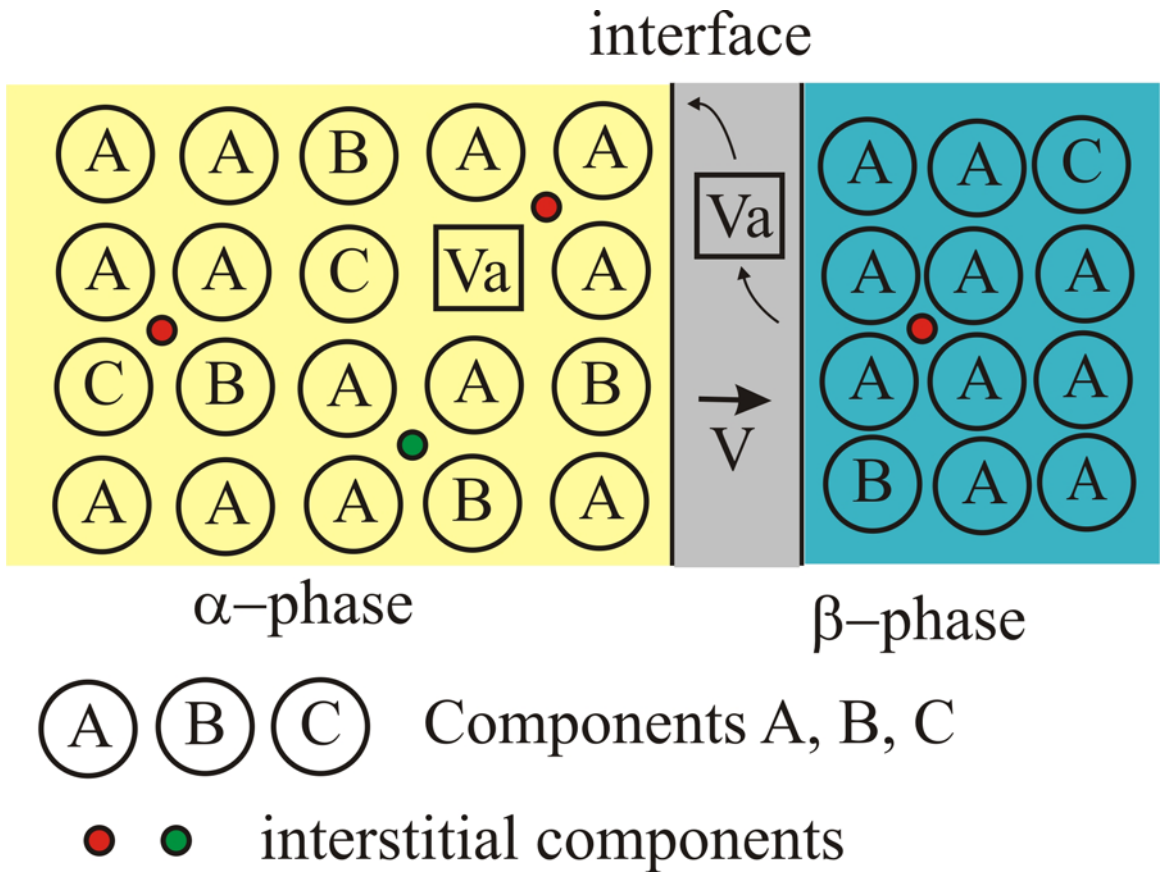
ALEMI (Workshop on alloying element effects on migrating interfaces),
18.-19. June 2007, Mc-Master University, Hamilton, Canada

Contents and Structure

- **Kinetics of diffusional phase transformations (sharp interface)**
  - Total Gibbs energy in the system
  - Balance relations
  - Dissipation at the sharp interface
  - Contact conditions
  - “Equal jump” contact conditions
- **Limitations of the sharp interface approach**
  - Intrinsic and effective interface mobility
  - Comparison with experiments
  - “Quasi-thick” interface model
  - Results of the quasi-thick interface model
  - Solid-liquid phase transformation
- **Thick interface**
  - Diffusion processes inside the interface
  - Comparison to the sharp interface model
- **Conclusions and Outlook**

Kinetics of diffusional phase transformations

One-dimensional system of unit cross-sectional area



Total Gibbs energy in the system

Chemical potential of component *i* in phase φ:

$$\mu_i^{\varphi} = \mu_i^{\varphi}(x_1, \ \ldots \ , x_N, T)$$

Total chemical Gibbs energy in a system of φ = α, β, γ, ... phases, each consisting of *i* = 1, .. , *N* components.

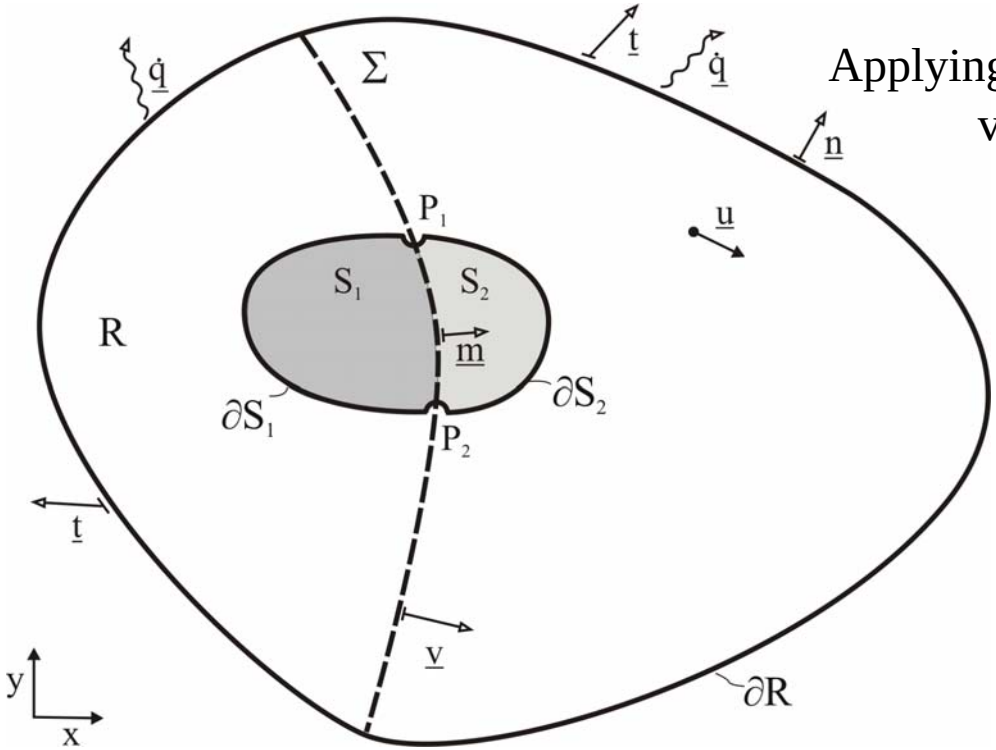
$$g_{\text{chem}}^{\varphi} = \frac{1}{V_{\text{m}}} \cdot \sum_{i=1}^{N} \int_{z_1}^{z_2} x_i^{\varphi} \mu_i^{\varphi} \, \mathrm{d}z$$

The total Gibbs energy of the system generally also consists of a mechanical part related to the elastic energies U'.

$$U' = \frac{1}{2} \underline{\underline{\sigma}} : \underline{\underline{\varepsilon}}$$

- **Incoherent** or partly coherent **interfaces**, no external stresses
- **Relaxation by a vacancy mechanism**.

Balance relations



Applying divergence and transport theorem and volumes of $S_1 \to 0$ and $S_2 \to 0$.

Mass balances

Interface:

$$[\![x_i / V_{\mathrm{m}}]\!] v = [\![j_i]\!], i = 1,2 ,..., N$$

$$\sum_{i=1}^{s} [\![x_i / V_{\mathrm{m}}]\!] v = -[\![j_{va}]\!]$$

Bulk:

$$\frac{\partial \left(x_i / V_{\mathrm{m}} \right)}{\partial t} = -\underline{\nabla} \cdot \underline{j}_i$$

F.D. Fischer, N.K. Simha: ”Influence of material fluxes on the jump relations at a singular interface in a multicomponent solid” *Acta Mech.* **171** (2004) 213.

Dissipation at the sharp interface

$$Q_{\text{int}} = \sum_{i=1}^{N} [\![\mu_i]\!] \cdot \left\langle \frac{x_i}{V_{\text{m}}} \cdot v - j_i \right\rangle \geq 0$$

mass balance

F.D. Fischer, N.K. Simha: "Influence of material fluxes on the jump relations at a singular interface in a multicomponent solid" *Acta Mech.* **171** (2004) 213.

$$Q_{\text{int}} = \sum_{i=1}^{N} \frac{x_i^{\text{tf}}}{V_{\text{m}}} [\![\mu_i]\!] \cdot v$$

$$x_i^{\text{tf}} = x_i^{\text{o}} - \left(V_{\text{m}} / v \right) \cdot j_i^{\text{o}} = x_i^{\text{n}} - \left(V_{\text{m}} / v \right) \cdot j_i^{\text{n}}$$

M. Hillert, M. Rettenmayr: "Deviation from local equilibrium at migrating phase interfaces" *Acta Mater.* **51 (**2003) 2803.

x_i^{tf} … mathematical construct – mole fraction?

Is the concept of **trans**-interface diffusion in the framework of a sharp interface model required (allowed)?!

M. Hillert: "Overview No.135. Solute drag, solute trapping and diffusional dissipation of Gibbs energy" *Acta Mater.* **47 (**1999) 4481.

Contact conditions I

A diffusional phase transformation with ***N* components** is a problem that comprises **(2*N*-1) indepenent variables** at the interface.

N-1 … independent fluxes or mole fractions in the parent (old) phase
N-1 … independent fluxes or mole fractions in the new phase
1 … interface velocity

Equilibrium concepts $M \to \infty$ $\Delta f = \frac{v}{M} = 0$

- **Local equilibrium contact conditions**

N c.c. $[[\mu_i]] = 0,\ i = 1,\ \ldots,\ N$ (*N*-1) m.b. $[\![x_i / V_{\mathrm{m}}]\!] v = [\![j_i]\!], i = 1,2,\ldots, N$

- **Paraequilibrium contact conditions**

(*N*-*s*-1) c.c. $[[\mu_i]] = 0$ (*N*-*s*-1) m.b. $[\![x_i / V_{\mathrm{m}}]\!] v = [\![j_i]\!]$ $i = s+1,\ \ldots,\ N$(Interstitials)

2s c.c. $K_i = y_i^{\mathrm{o}} \Big/ \sum_{i=1}^{s} y_i^{\mathrm{o}} = y_i^{\mathrm{n}} \Big/ \sum_{i=1}^{s} y_i^{\mathrm{n}},\ i = 1,\ldots, s$ (Substitutionals) 1 pe. c. $\Delta f = \sum_{i=1}^{s} x_i^{\mathrm{n}} \cdot \frac{[[\mu_i]]}{V_{\mathrm{m}}} = 0$

Contact conditions II

Non-equilibrium concepts M (finite, intrinsic interface mobility?!)

$$\Delta f = \frac{v}{M}$$

- **No substitutional diffusion**

(N-s-1) c.c. $[[\mu_i]] = 0$ (N-s-1) m.b. $[\![x_i / V_\mathrm{m}]\!] v = [\![j_i]\!]$ $i = s+1, \ldots, N$ (Interstitials)

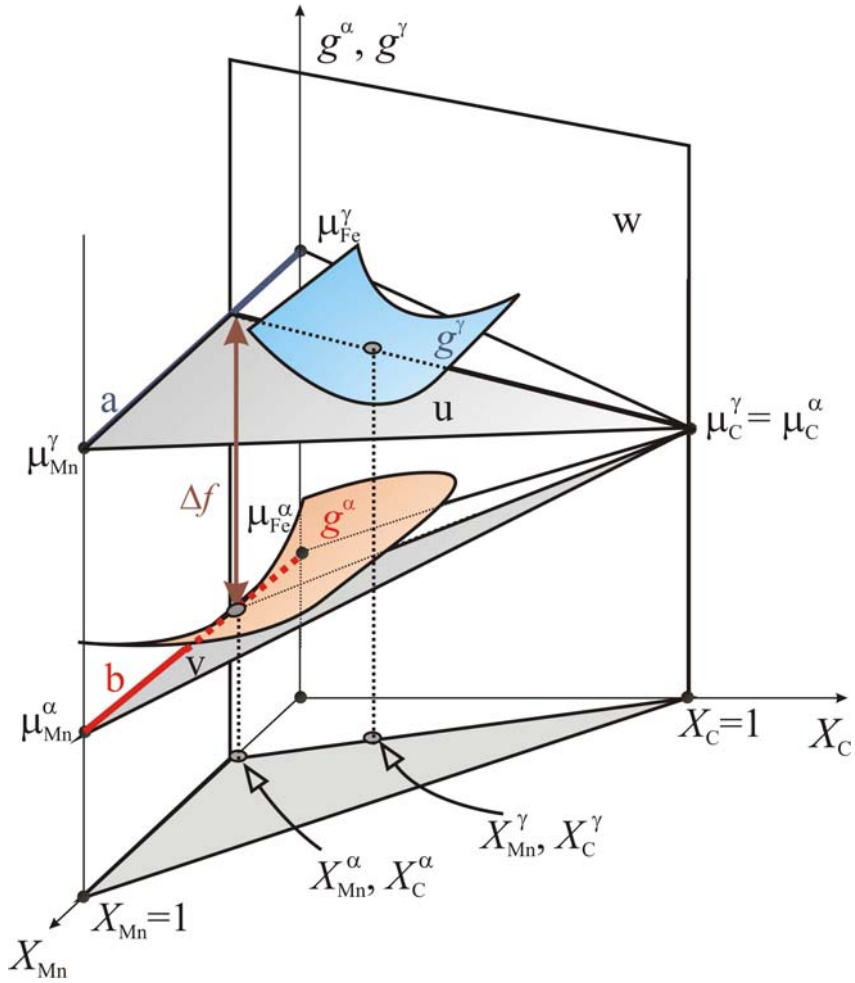
$2s$ c.c. $K_i = y_i^\mathrm{o} \Big/ \sum_{i=1}^{s} y_i^\mathrm{o} = y_i^\mathrm{n} \Big/ \sum_{i=1}^{s} y_i^\mathrm{n}$, $i = 1, \ldots, s$ (Substitutionals) 1 kinetic c. $\Delta f = \sum_{i=1}^{s} x_i^\mathrm{n} \cdot \frac{[[\mu_i]]}{V_\mathrm{m}}$

- **„Equal jump“ contact condition**

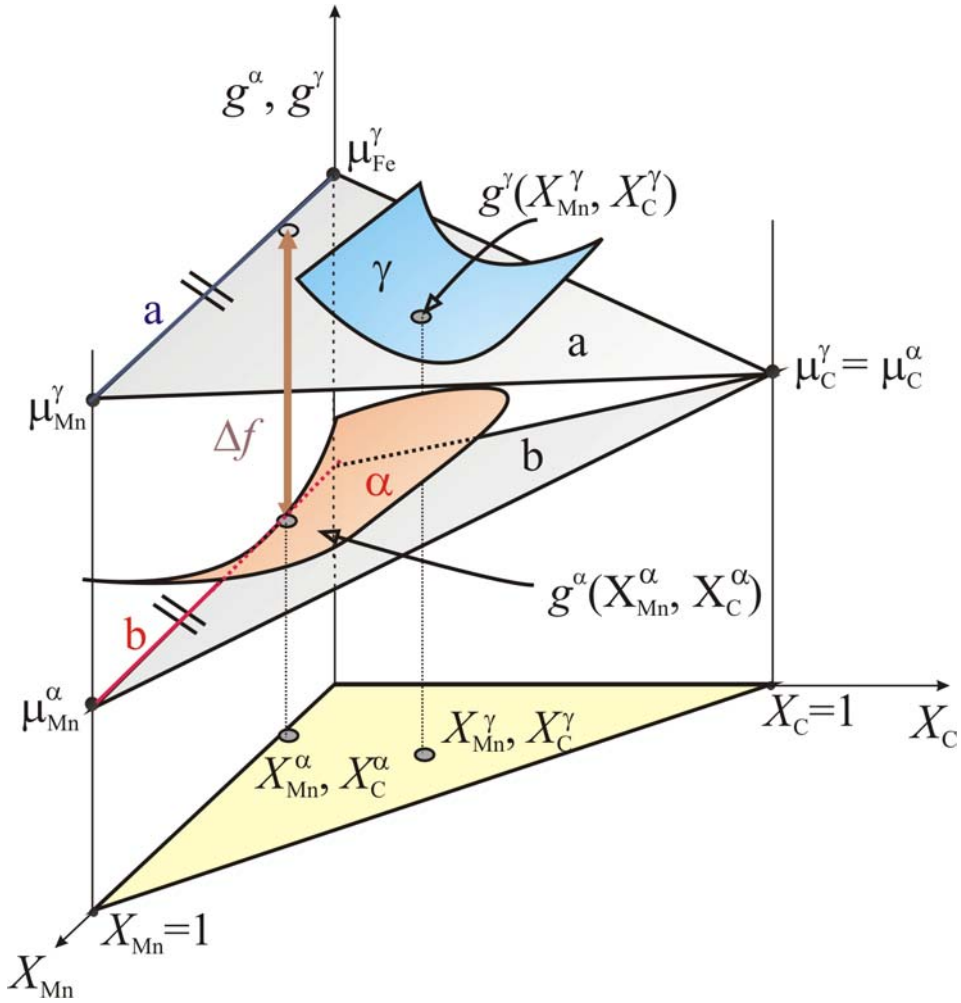
(N -1) c.c.
$$[\![\mu_i]\!] = 0,\ i = s+1, \ldots, N \text{ (interstitials)},$$
$$[\![\mu_i]\!] = [\![\mu_N]\!], i = 1, \ldots, s \text{ (substitutionals)}.$$

1 kinetic c. $\Delta f = \sum_{i=1}^{s} x_i^\mathrm{n} \cdot \frac{[[\mu_i]]}{V_\mathrm{m}}$ (N-1) m.b. $[\![x_i / V_\mathrm{m}]\!] v = [\![j_i]\!], i = 1,2,\ldots, N$

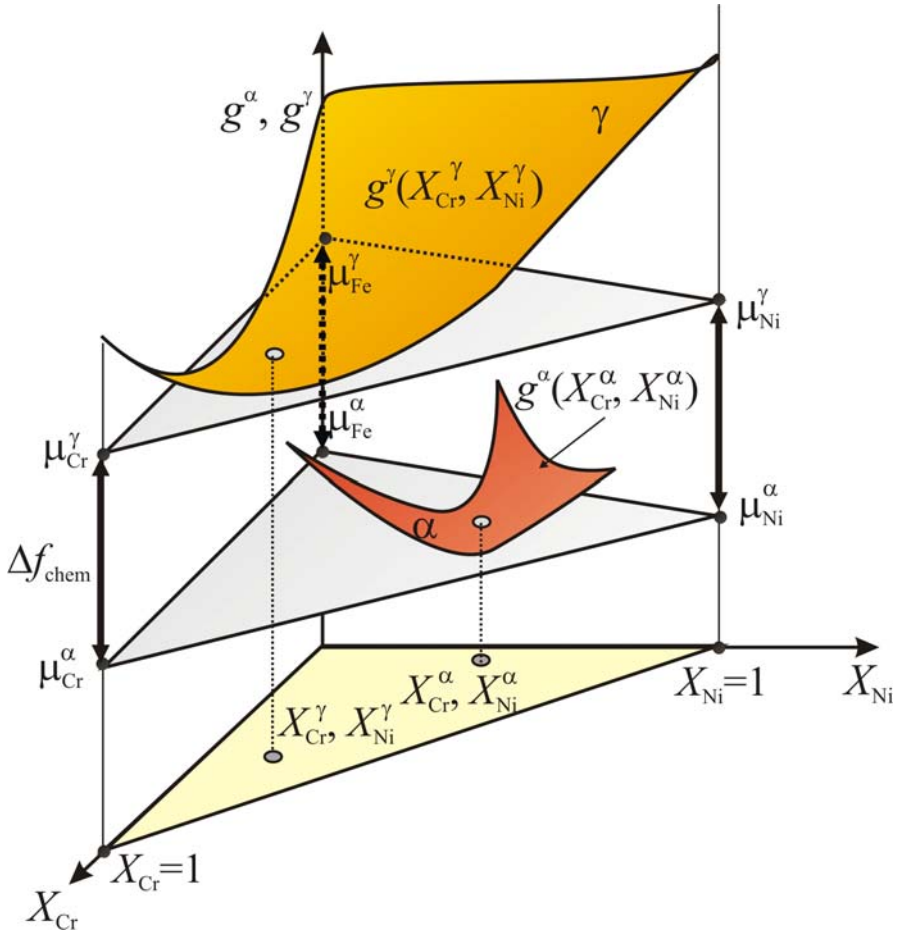
Non-equilibrium (No substitutional diffusion)



„Equal jump“ contact conditions



„Equal jump“ contact conditions



Derivation of the „equal jump“ conditions by the thermodynamic extremal principle

Energy balance

$$Q_{\text{int}} = Q = -\dot{G} \qquad Q_{\text{int}} = v^2 / M$$

Volumes of $S_1 \to 0$ and $S_2 \to 0$.

$$\dot{G} = \sum_{i=1}^{N} j_i^{\text{o}} \cdot [[\mu_i]] - \sum_{i=1}^{N} \frac{x_i^{\text{o}}}{V_{\text{m}}} v \cdot [[\mu_i]]$$

Thermodynamic extremal principle

$$\frac{\partial}{\partial \dot{q}_i}\left[Q + \eta(\dot{G} + Q) \right] = 0 \rightarrow \eta = -2 \rightarrow \frac{\partial \dot{G}}{\partial \dot{q}_i} = -\frac{1}{2}\frac{\partial Q}{\partial \dot{q}_i}$$

Contact conditions

$$v = \frac{M}{V_{\text{m}}} \cdot \sum_{i=1}^{N} x_i^{\text{o}} [[\mu_i]] = \frac{M}{V_{\text{m}}} \cdot \Delta f$$

$$\frac{\partial \dot{G}}{\partial j_i^{\text{o}}} = [[\mu_i]] - [[\mu_N]] = \frac{\partial Q}{\partial j_i^{\text{o}}} = 0$$

Contents and Structure

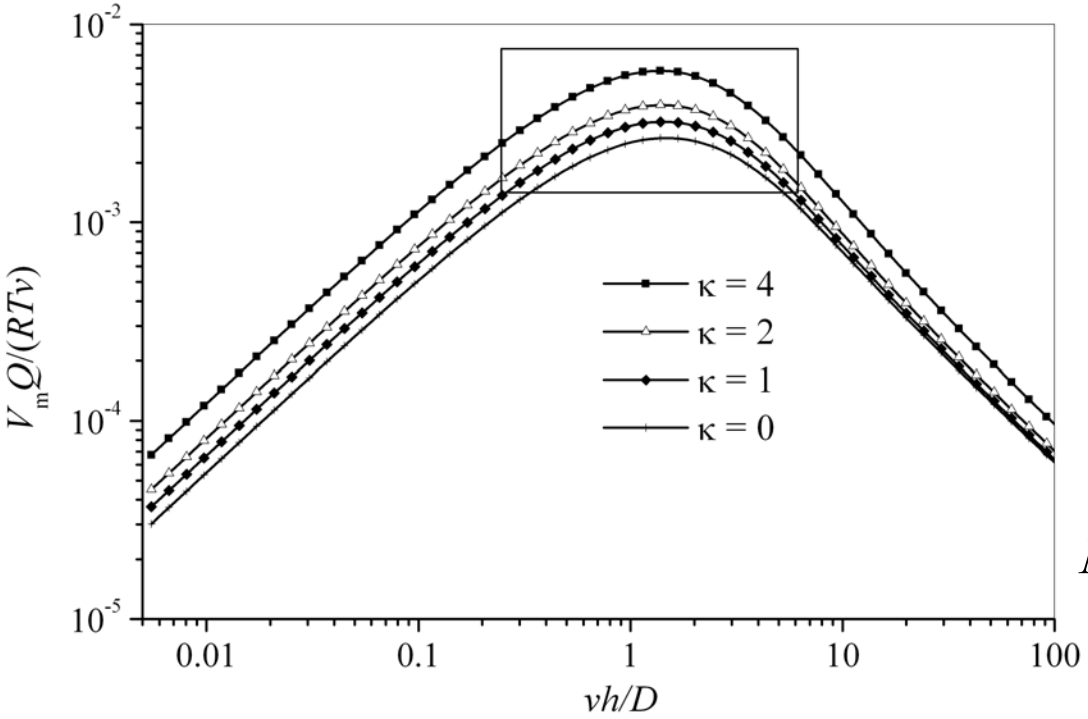
- **Kinetics of diffusional phase transformations (sharp interface)**
  - Total Gibbs energy in the system
  - Balance relations
  - Dissipation at the sharp interface
  - Contact conditions
  - “Equal jump” contact conditions

- **Limitations of the sharp interface approach**
  - Intrinsic and effective interface mobility
  - Comparison with experiments
  - “Quasi-thick” interface model
  - Results of the quasi-thick interface model
  - Solid-liquid phase transformation

- **Thick interface**
  - Diffusion processes inside the interface
  - Comparison to the sharp interface model

- **Conclusions and Outlook**

Intrinsic and effective interface mobility



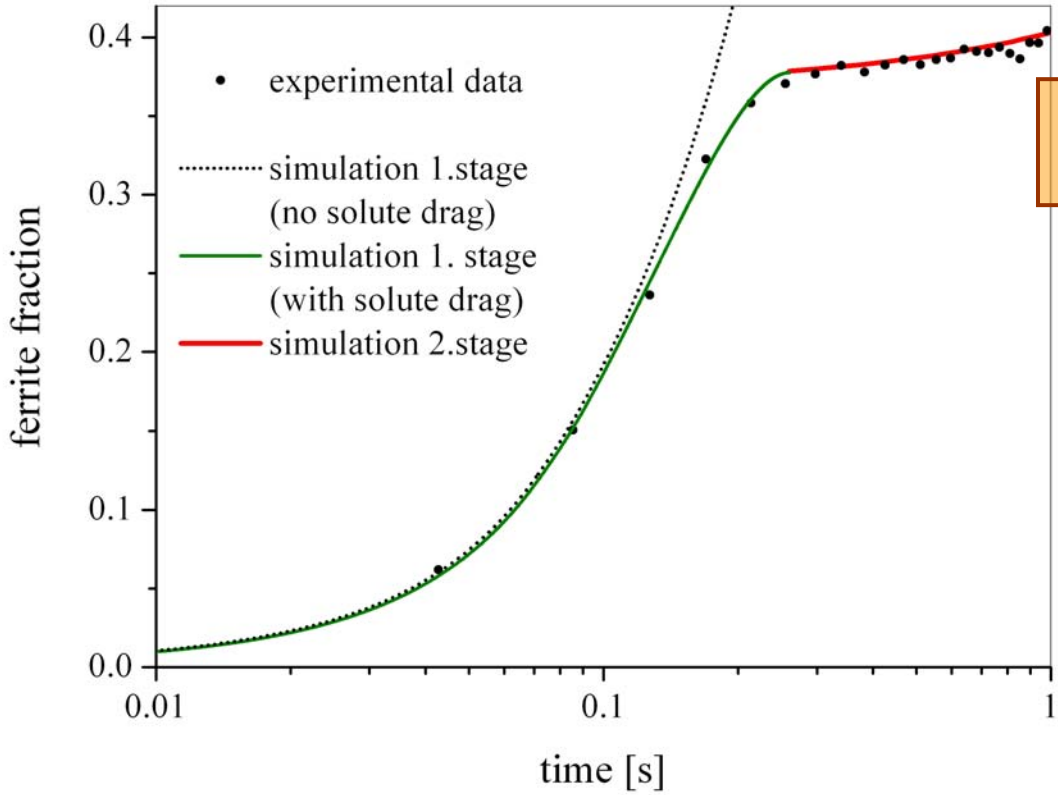
J. Svoboda, F. D. Fischer, E. Gamsjäger: "Influence of solute segregation and drag on properties of migrating interfaces", *Acta mater.*, **50**, 967-977, 2002.

$$M_{\text{eff}} = \left(\frac{1}{M_{\text{intr}}} + \frac{Q_{\text{sd}}}{v^2} \right)^{-1}, \quad \frac{Q_{\text{sd}}}{v} = \text{const.} \Rightarrow$$

$$M_{\text{eff}} = \left(\frac{1}{M_{\text{intr}}} + \frac{C}{v} \right)^{-1}$$

C depends linearly on time

Comparison: Theory and experiment

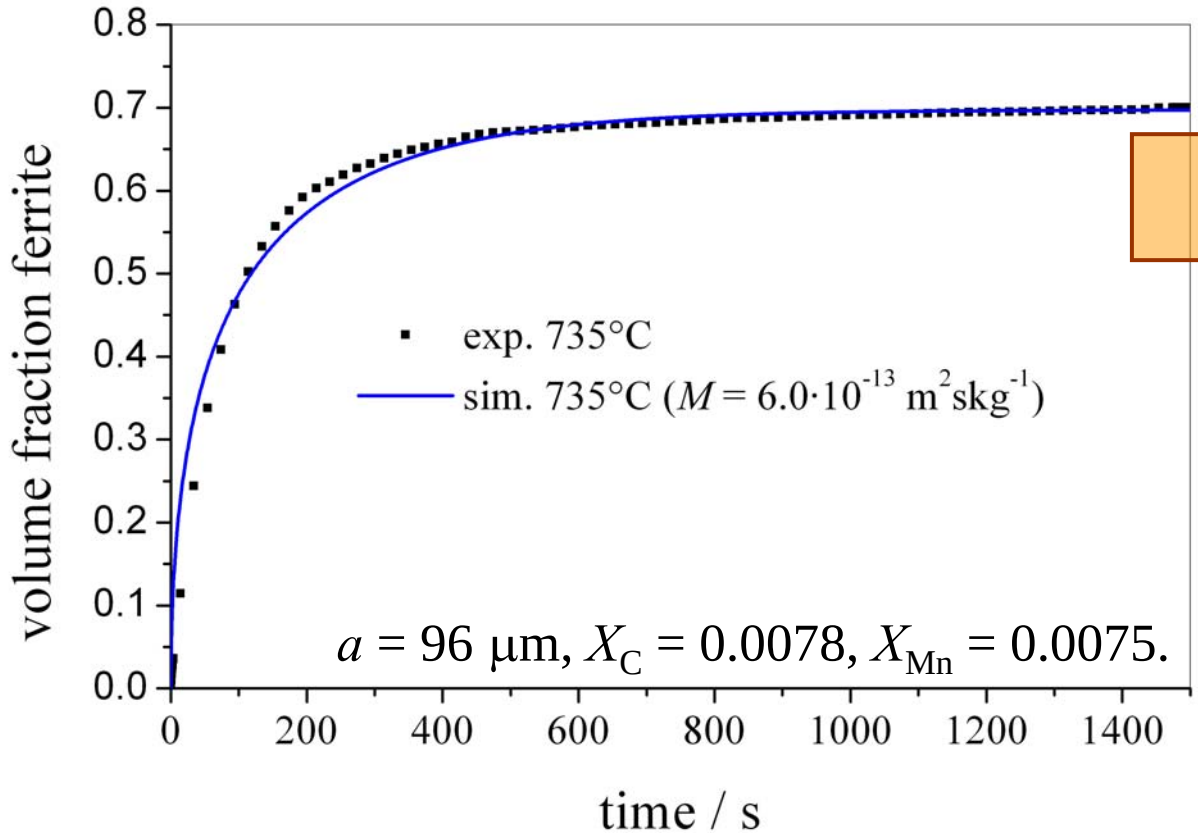


Non – equilibrium
No substitutional diffusion

E. Kozeschnik, E. Gamsjäger, "High-Speed Quenching Dilatometer Investigation of the Austenite to Ferrite Transformation in low to ultra low carbon steel", *Metall. Mater. Trans.* **37A** (2006) 1791-1797.

E. Gamsjäger: "Kinetics of the austenite-to-ferrite phase transformation – From the intrinsic to an effective mobility", *Materials Science Forum* **539-543** (2007) 2570-2575.

Comparison with experimental data*
Fe-C-Mn (ϑ = 735°C)

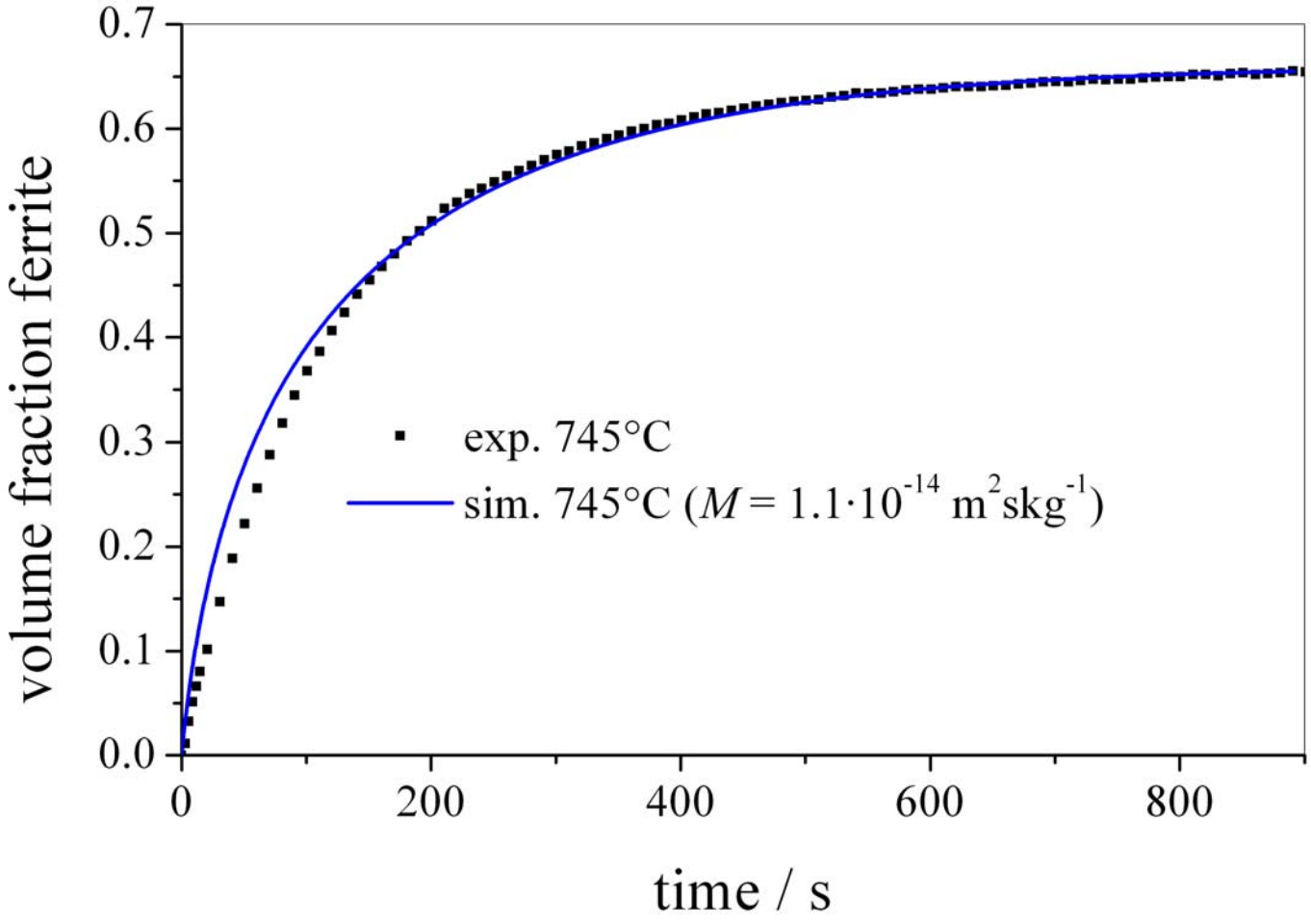


Non – equilibrium
No substitutional diffusion

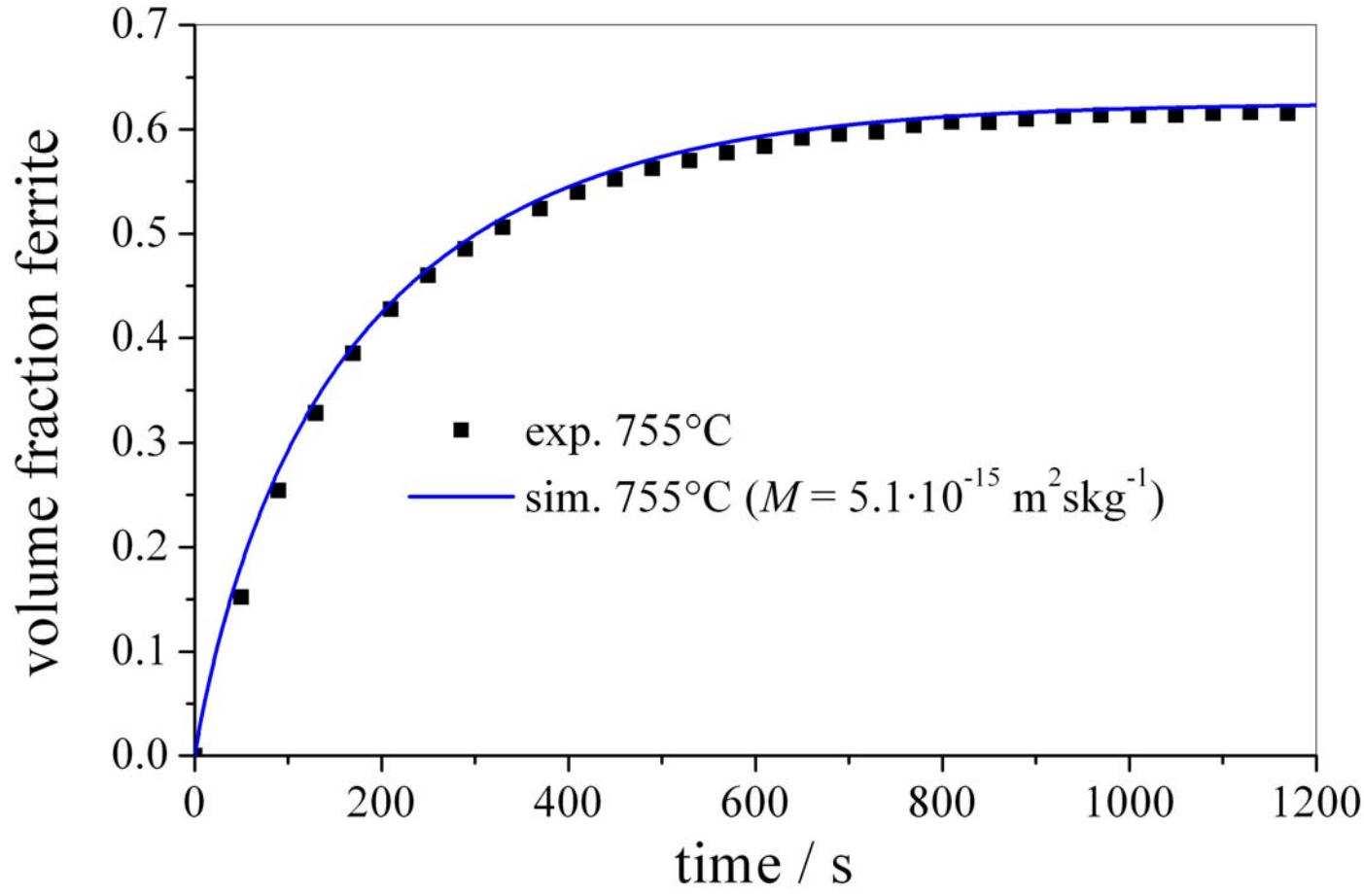
*F. Fazeli, M. Militzer; Modelling the effects of alloying elements on the ferrite transformation kinetics, 44th Mechanical Working and Steel Processing Conf. Proc., 2002.

E. Gamsjäger, J. Svoboda, F.D. Fischer: "Austenite-to-ferrite phase in low-alloyed steels", *Comp.Mat. Sci.* **32** (2005) 360-369.

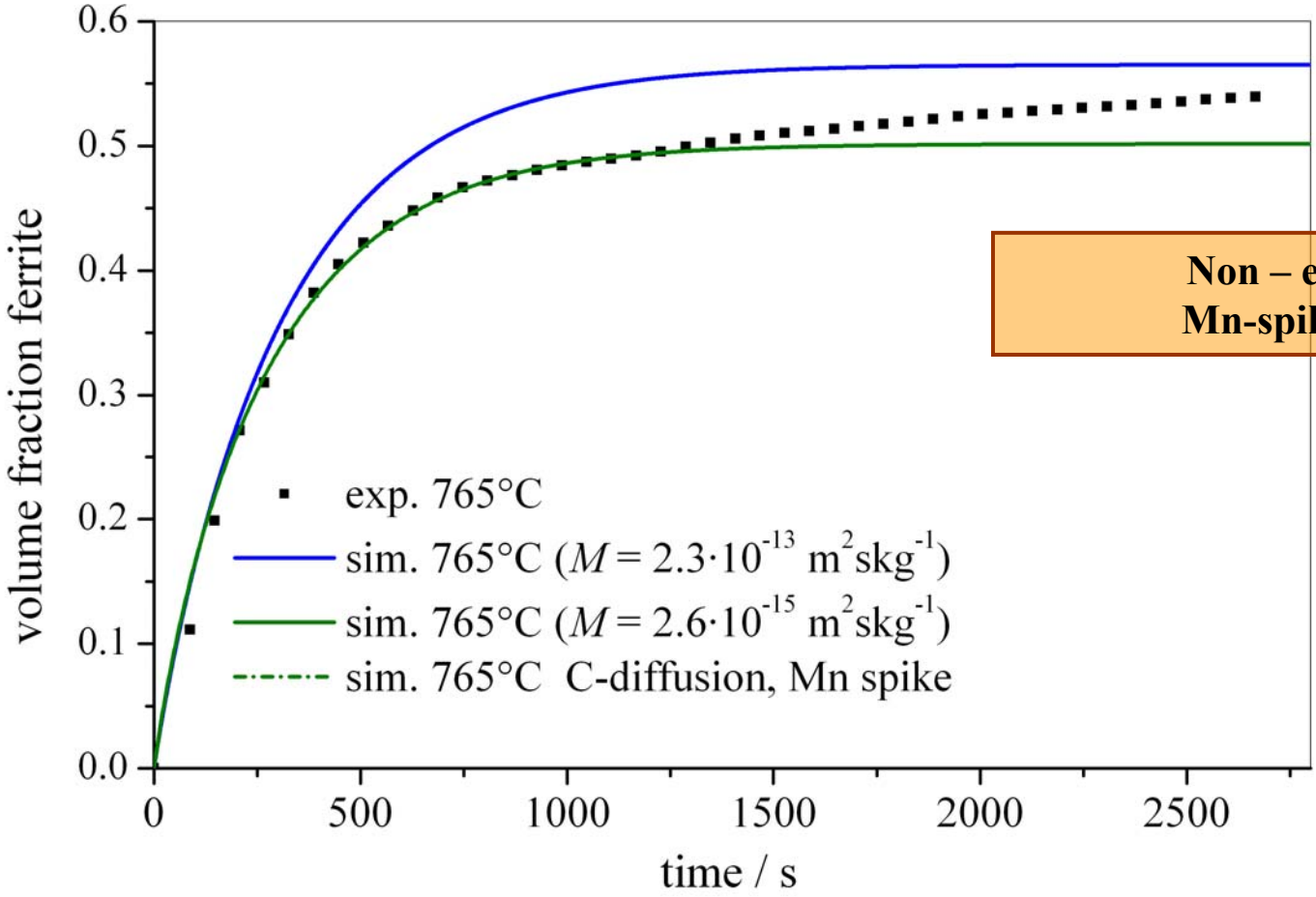
Comparison with experimental data
Fe-C-Mn (ϑ = 745°C)



Comparison with experimental data
Fe-C-Mn (ϑ = 755°C)

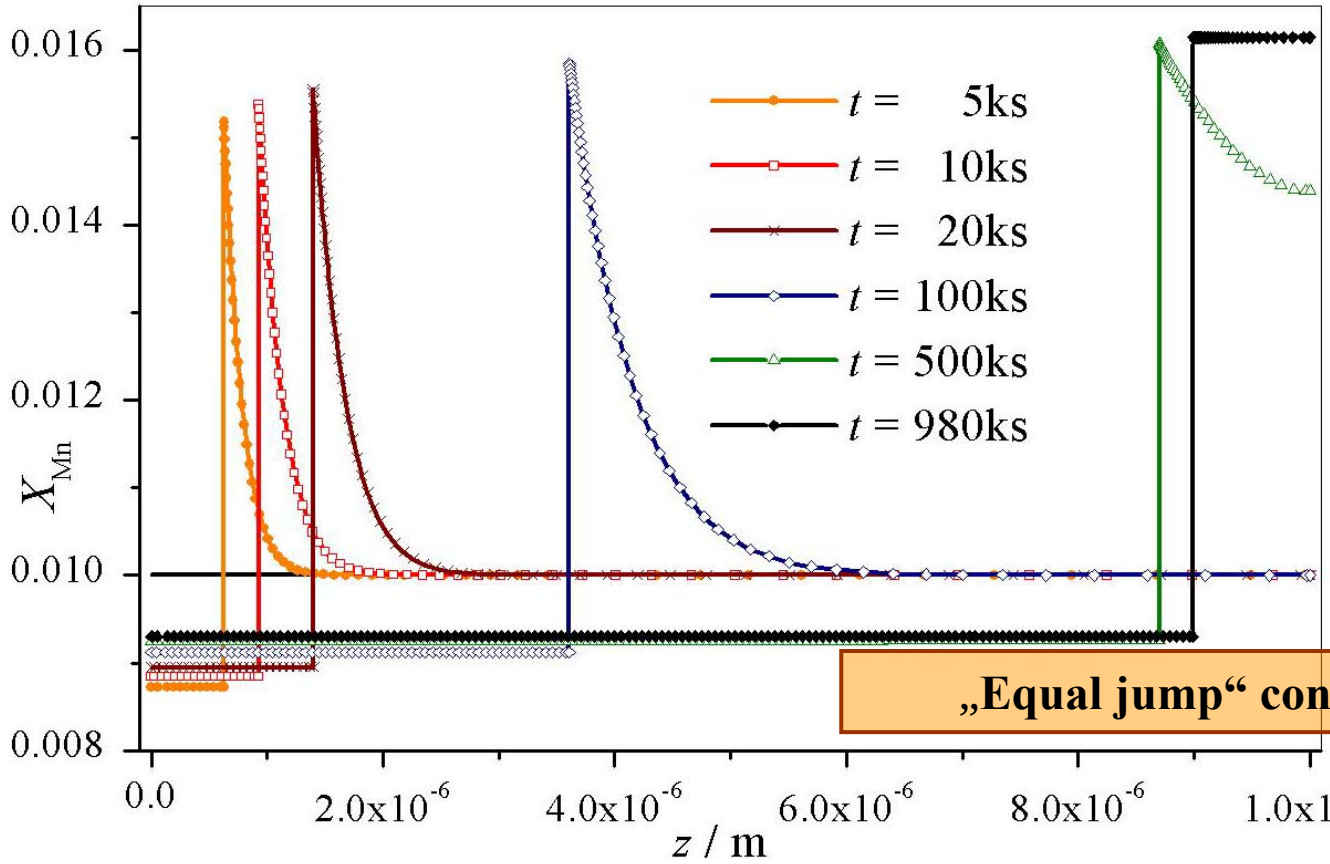


Comparison with experimental data
Fe-C-Mn (ϑ = 765°C)



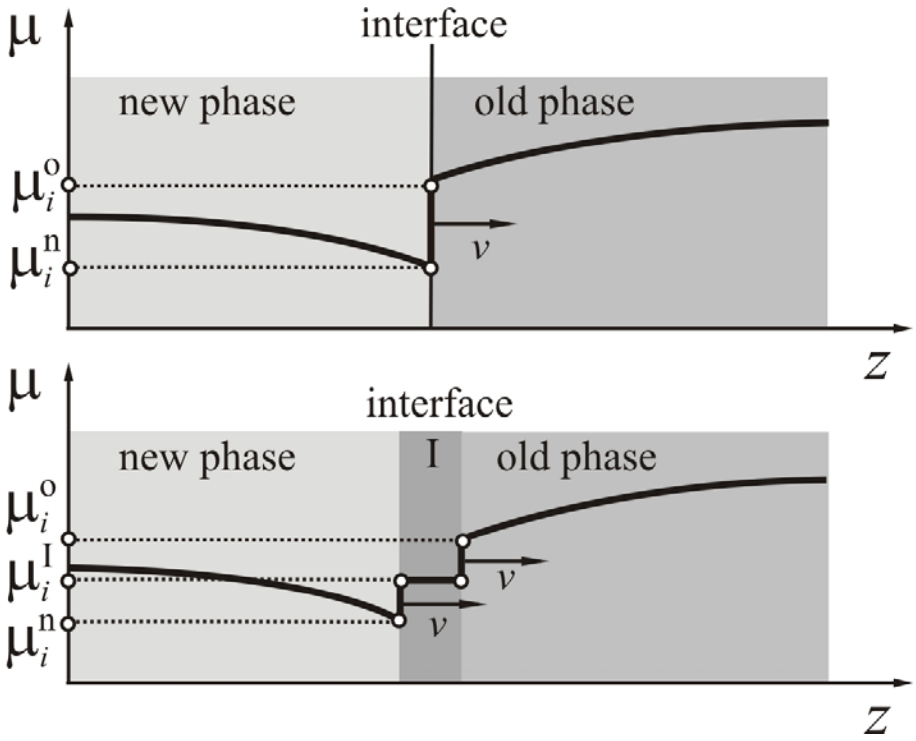
Non – equilibrium
Mn-spike diffusion

Mobility and diffusion controlled growth: Fe-Mn-Profiles (T = 1123K)



„Equal jump“ contact condition

„Quasi-thick“ interface



Model $\boxed{j_i^{\mathrm{I}} = 0}$

$$d_{\mathrm{chem,n/I}} = \sum_{i=1}^{N} [\![\mu_i]\!]_{\mathrm{n/I}} \left(x_i^{\mathrm{I}} / V_{\mathrm{m}}\right) v \,, \quad [\![\mu_i]\!]_{\mathrm{n/I}} = \mu_i^{\mathrm{I}} - \mu_i^{\mathrm{n}}$$

$$d_{\mathrm{chem,I/o}} = \sum_{i=1}^{N} [\![\mu_i]\!]_{\mathrm{I/o}} \left(x_i^{\mathrm{I}} / V_{\mathrm{m}}\right) v \,, \quad [\![\mu_i]\!]_{\mathrm{I/o}} = \mu_i^{\mathrm{o}} - \mu_i^{\mathrm{I}}$$

$$v = M_{\mathrm{n/I}} \cdot \Delta f_{\mathrm{n/I}} = M_{\mathrm{I/o}} \cdot \Delta f_{\mathrm{I/o}} \qquad \frac{1}{M} = \frac{1}{M_{\mathrm{I/o}}} + \frac{1}{M_{\mathrm{n/I}}}$$

$$v = \frac{M_{\mathrm{I/o}} \cdot M_{\mathrm{I/n}}}{M_{\mathrm{I/o}} + M_{\mathrm{I/n}}} \cdot \sum_{i=1}^{N} [\![\mu_i]\!]_{\mathrm{n/o}} \cdot x_i^{\mathrm{I}} = M \cdot \sum_{i=1}^{N} [\![\mu_i]\!] \cdot x_i^{\mathrm{I}}$$

E. Gamsjäger: “A Note on the Contact Conditions at Migrating Interfaces”, accepted for publication, *Acta mater.*, 2007.

Results of the „quasi-thick“-interface approach

- **“Quasi-thick”-interface – descriptive way to visualize the situation at a sharp interface.**

$$x_i^{\mathrm{tf}} = x_i^{\mathrm{o}} - \left(V_{\mathrm{m}} / v\right) \cdot j_i^{\mathrm{o}} = x_i^{\mathrm{n}} - \left(V_{\mathrm{m}} / v\right) \cdot j_i^{\mathrm{n}} = x_i^{\mathrm{I}}$$

x_i^{tf} ... mathematical construct

x_i^{I} ... mole fraction inside the interface $(0 \le x_i^{\mathrm{I}} \le 1)$

- **“Quasi-thick”-interface – is consistent with the “equal jump”-condition.**

Limitations of the sharp interface approach

Only valid for $(0 \le x_i^{\mathrm{tf}} \le 1)$

Fluxes inside the interface cannot be considered.

$\Rightarrow$

Contact conditions at real interfaces ≠
Contact conditions at ideally sharp interfaces

Applicability of the sharp interface model

Solid-liquid phase transformations – *yes*

Solid-solid transformation – *yes,*
if dissipation by diffusion processes in the interface is negligible.

Solid-liquid phase transformation

Experimental set-up

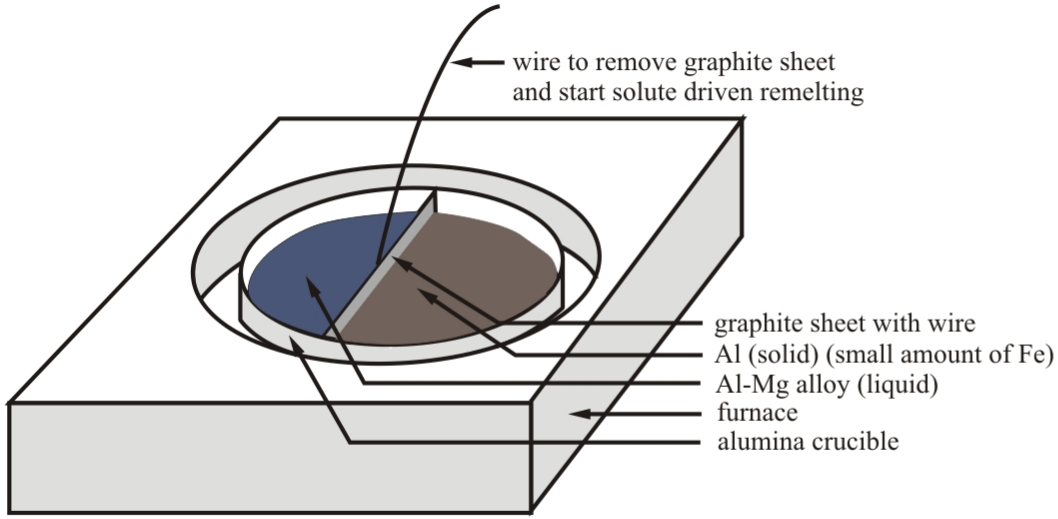


Fig 1: Liquid Al-Mg and solid Al half cylinder separated by a graphite sheet.

Phase diagram

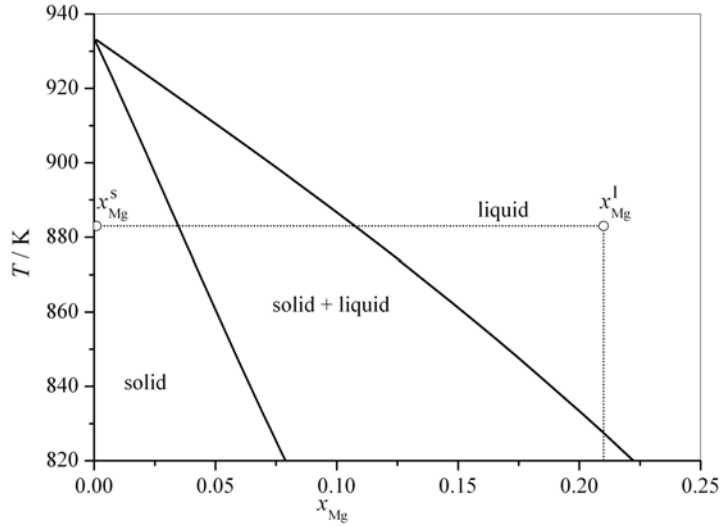
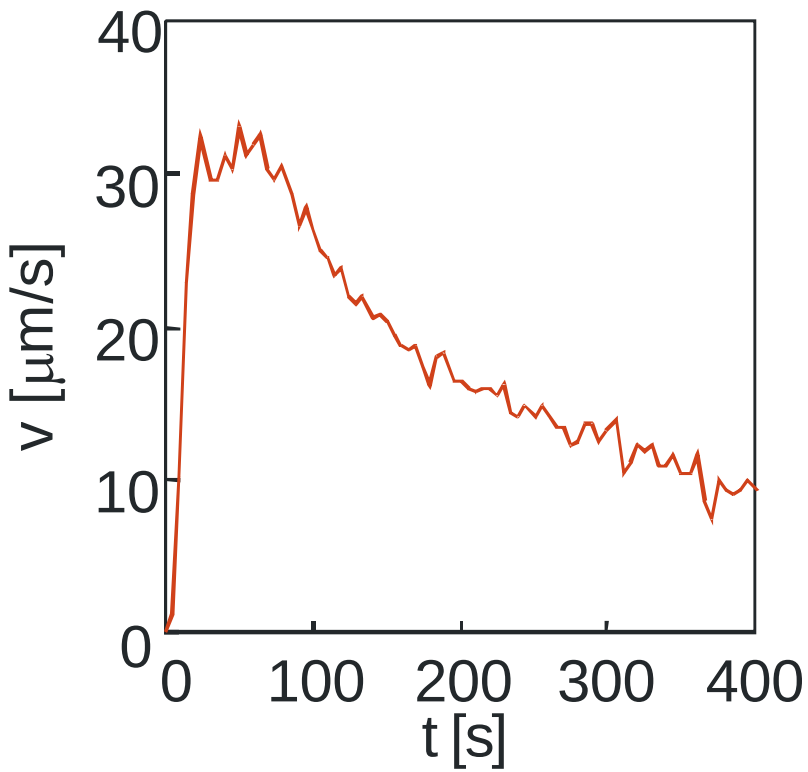


Fig 2: Al-rich side of the Al-Mg phase diagram.
Initial mole fraction in the solid and liquid phase.

M. Rettenmayr, O. Warketin, M. Rappaz, H. E. Exner: "Simulation of solutal remelting", *Acta mater.*.**49** (2001) 2499-2510.

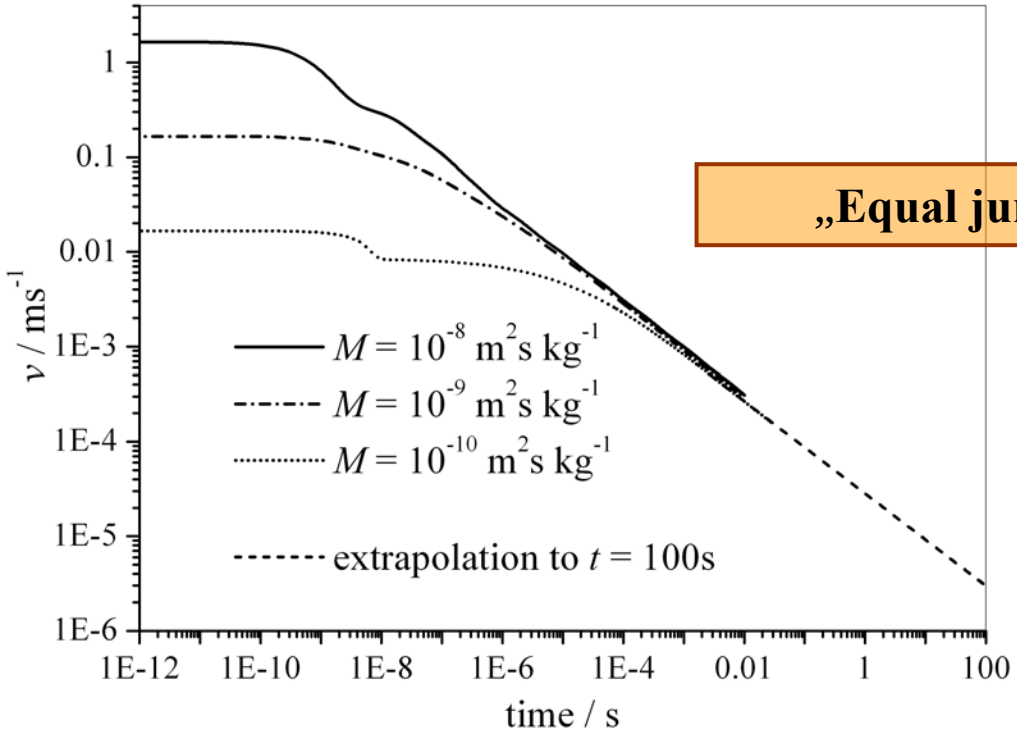
Experimental data

Time-temperature data ⇒ heat balance equation ⇒ time vs. velocity



Interface velocity: $x_{Mg} = 0.204$, $T = 883$K.

Calculations



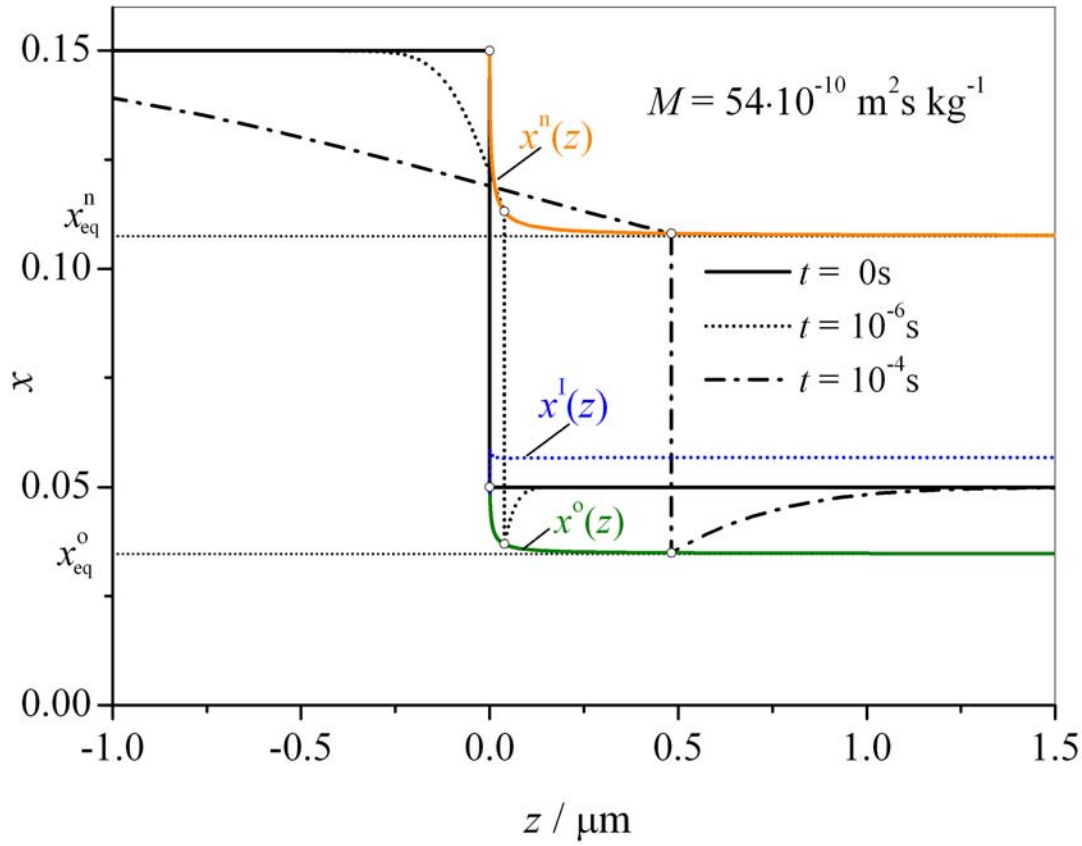
„Equal jump“ contact condition

Interface velocity for three different interface mobilities.

Local equilibrium is reached at very small transformation times.

Solid-liquid transformation in the Al-Mg system

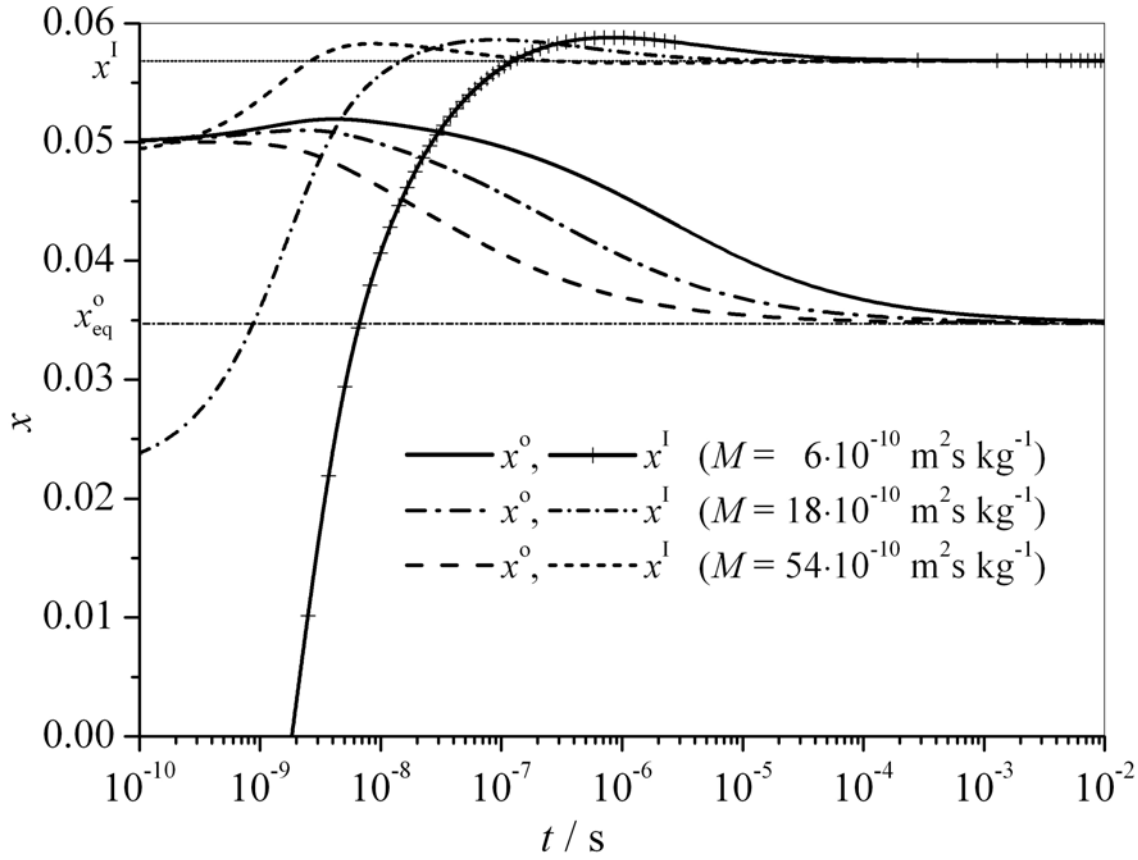
$x^{n} = 0.15$, $x^{o} = 0.05$, $T = 883$K



The mole fractions x^{o}, x^{n} and x^{I} are depicted versus the spatial variable z (grey lines).
The mole fraction profiles are plotted at different transformation times.

Solid-liquid transformation in the Al-Mg system

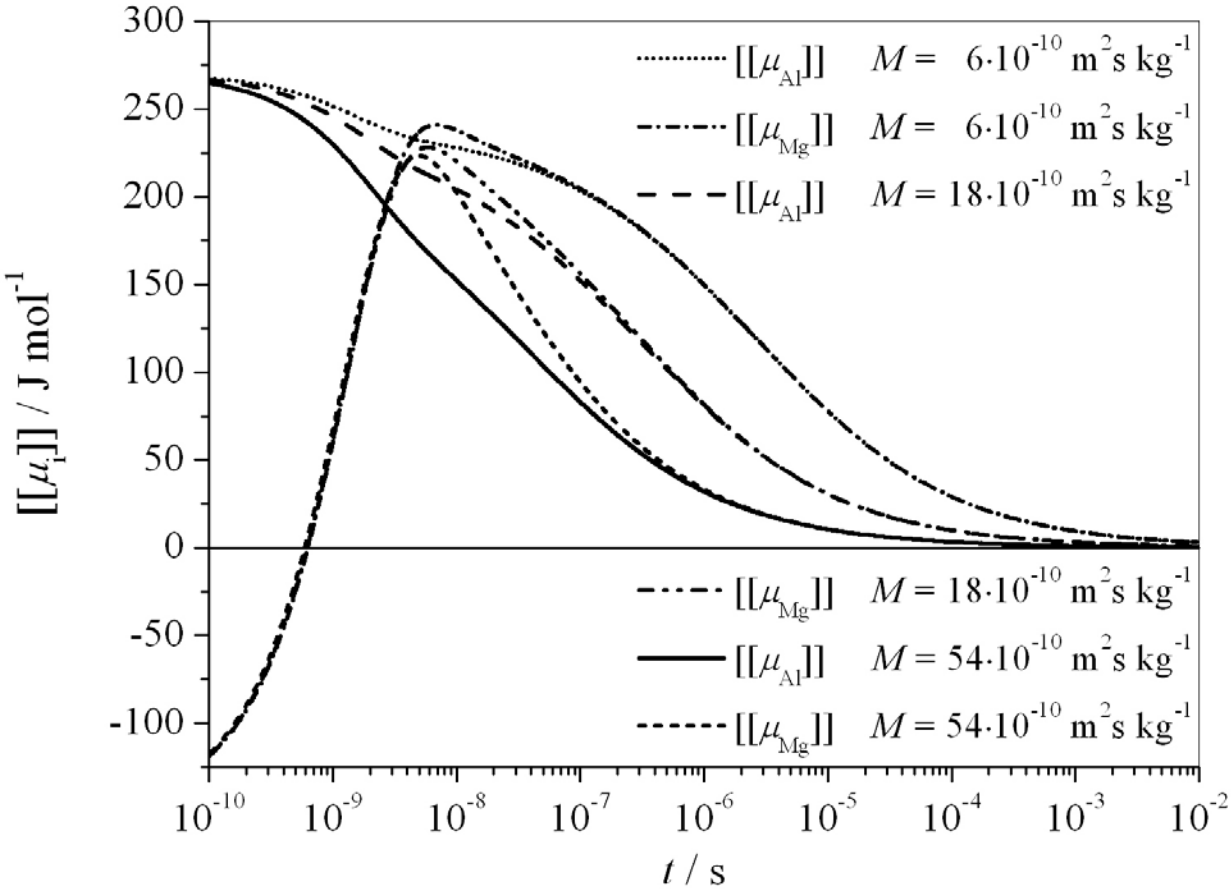
$x^{n} = 0.15$, $x^{o} = 0.05$, $T = 883$K



The time-dependent evolution of the mole fractions x^{o} and x^{I} at three interface different interface mobilities M.

Solid-liquid transformation in the Al-Mg system

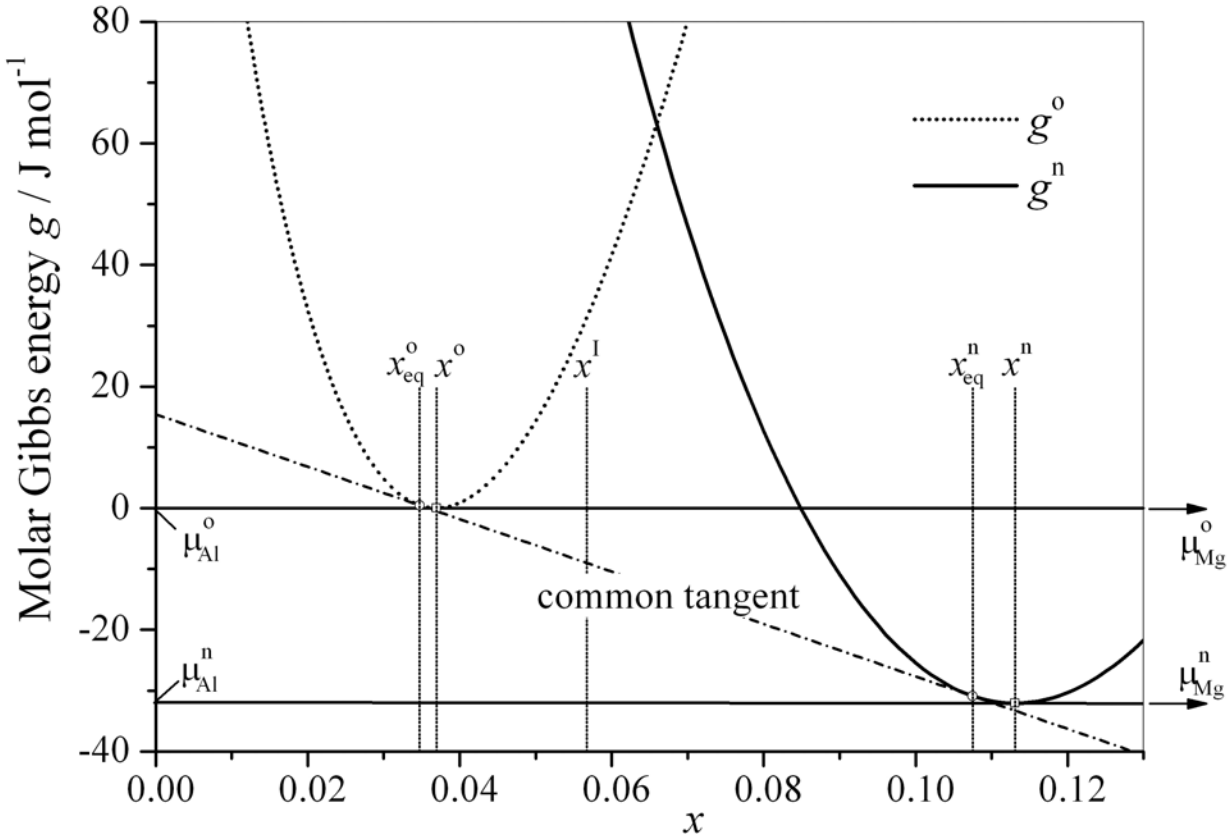
$x^n = 0.15$, $x^o = 0.05$, $T = 883$K



The jumps of the chemical potentials, [[μ_{Al}]] and [[μ_{Mg}]] vs. time.

Solid-liquid transformation in the Al-Mg system

$x^{n} = 0.15$, $x^{o} = 0.05$, $T = 883$K

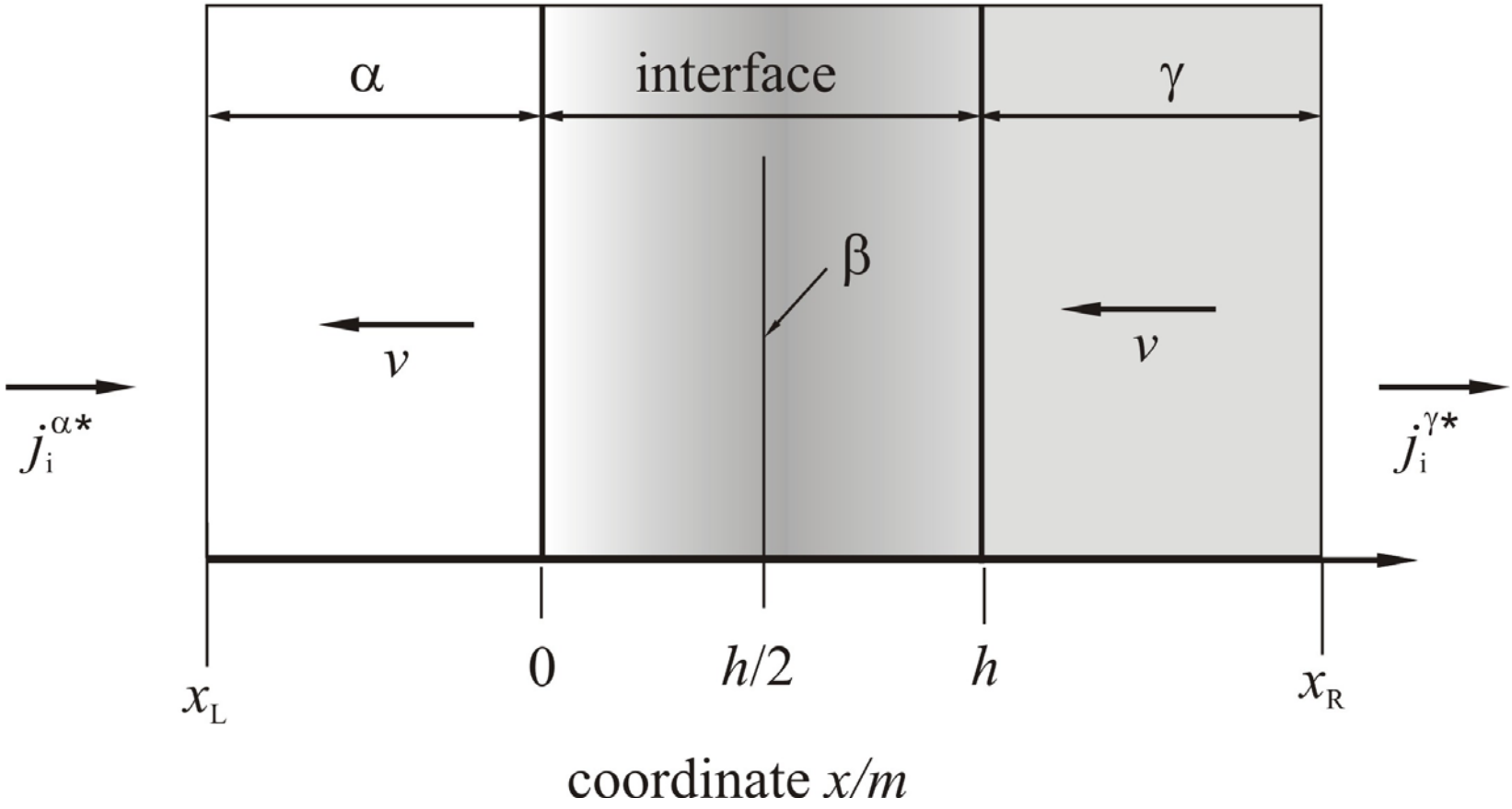


Calculated molar Gibbs energy diagram for a certain deviation from local equilibrium (t = 1μs).

Contents and Structure

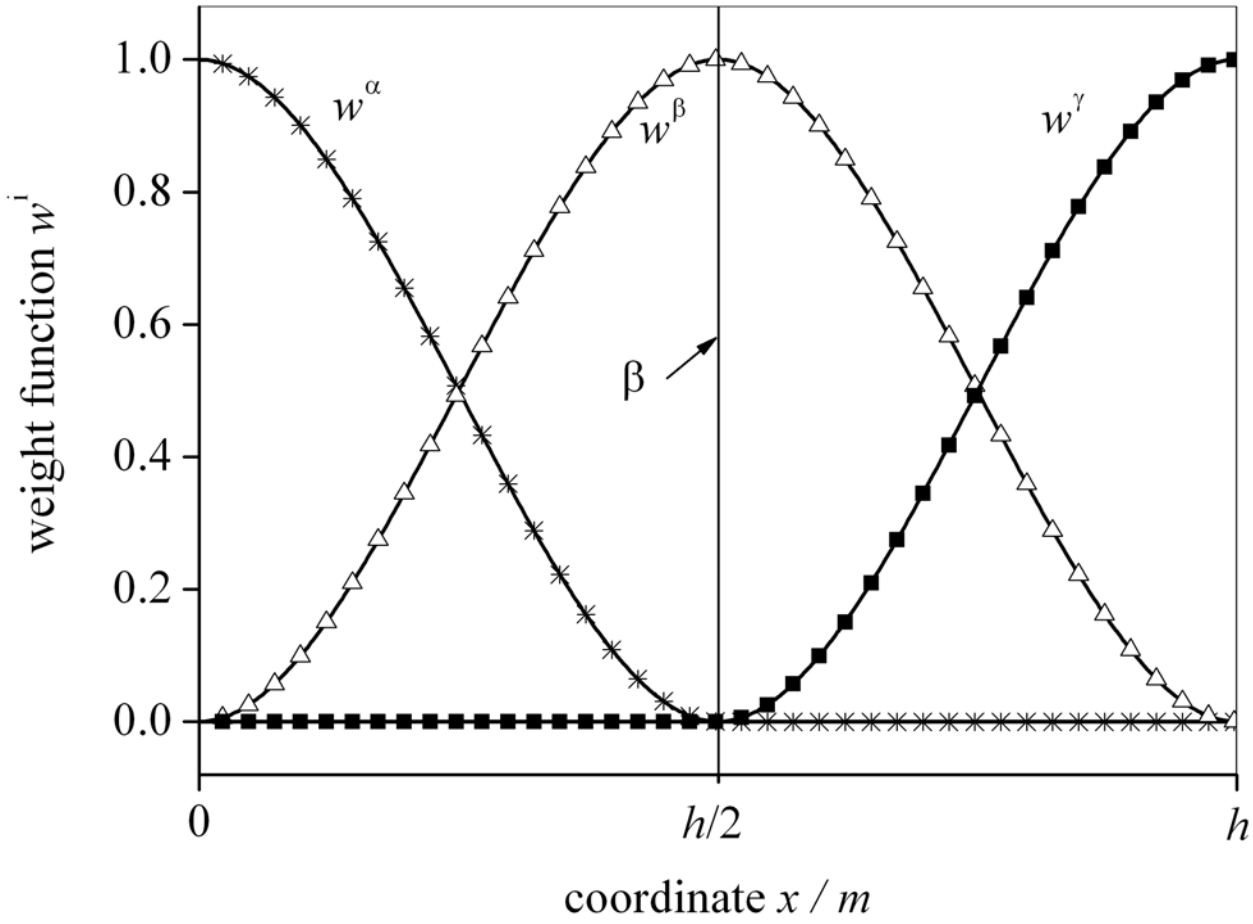
- **Kinetics of diffusional phase transformations (sharp interface)**
  - Total Gibbs energy in the system
  - Balance relations
  - Dissipation at the sharp interface
  - Contact conditions
  - “Equal jump” contact conditions
- **Limitations of the sharp interface approach**
  - Intrinsic and effective interface mobility
  - Comparison with experiments
  - “Quasi-thick” interface model
  - Results of the quasi-thick interface model
  - Solid-liquid phase transformation
- **Thick interface**
  - Diffusion processes inside the interface
  - Comparison to the sharp interface model
- **Conclusions and Outlook**

Thick interface - steady state solutions

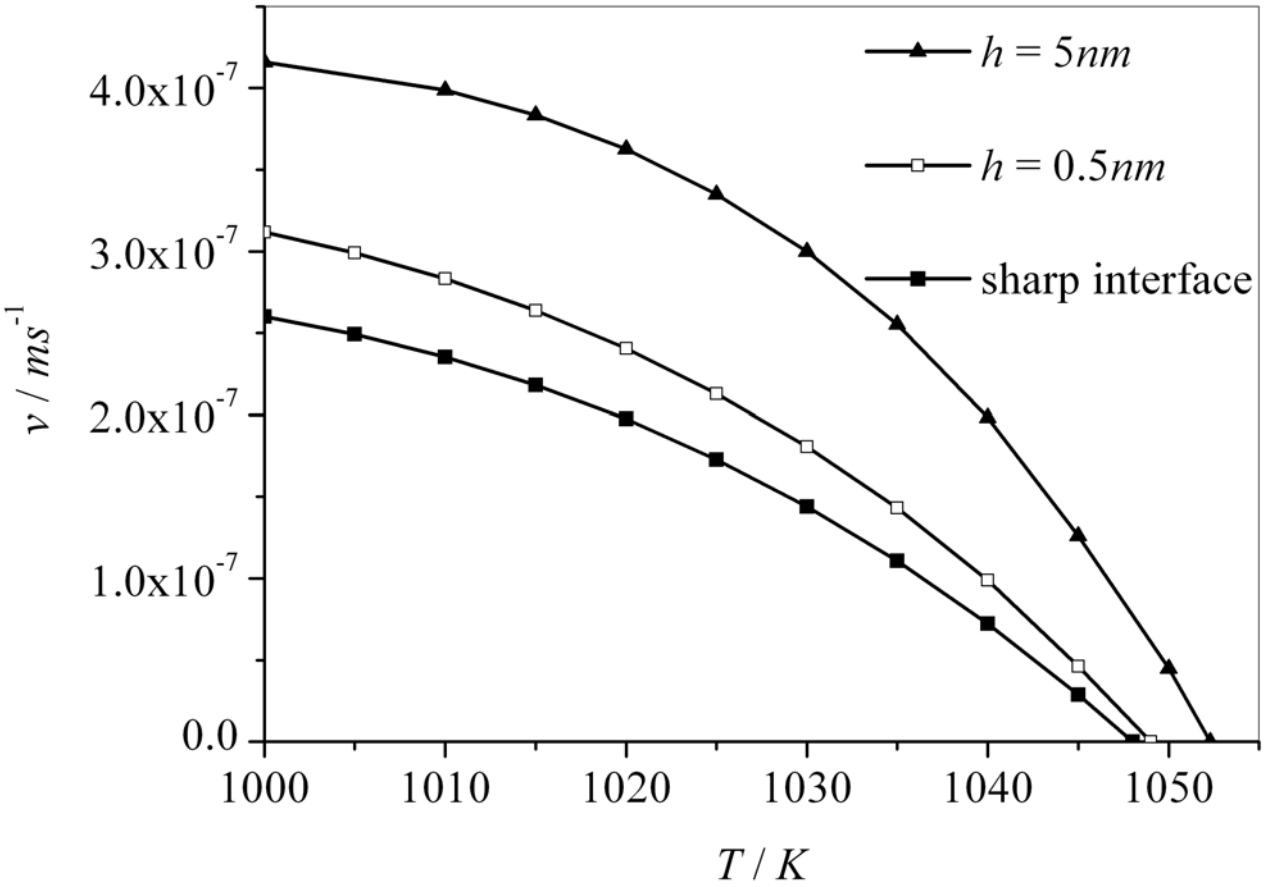


J. Svoboda, J. Vala, E. Gamsjäger, F.D. Fischer, *Acta mater.* **54** (2006) 3953-3960.

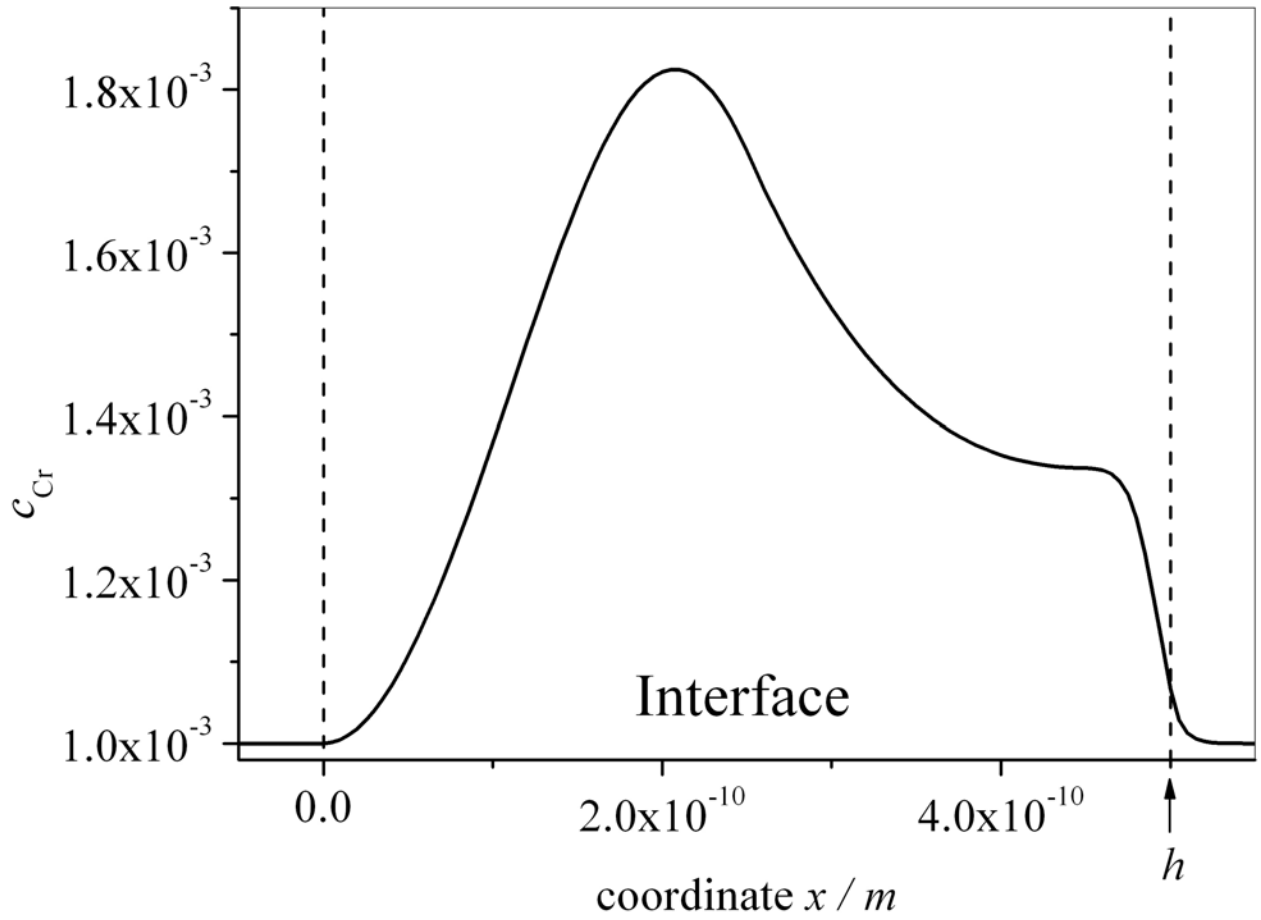
Thick interface – weight functions



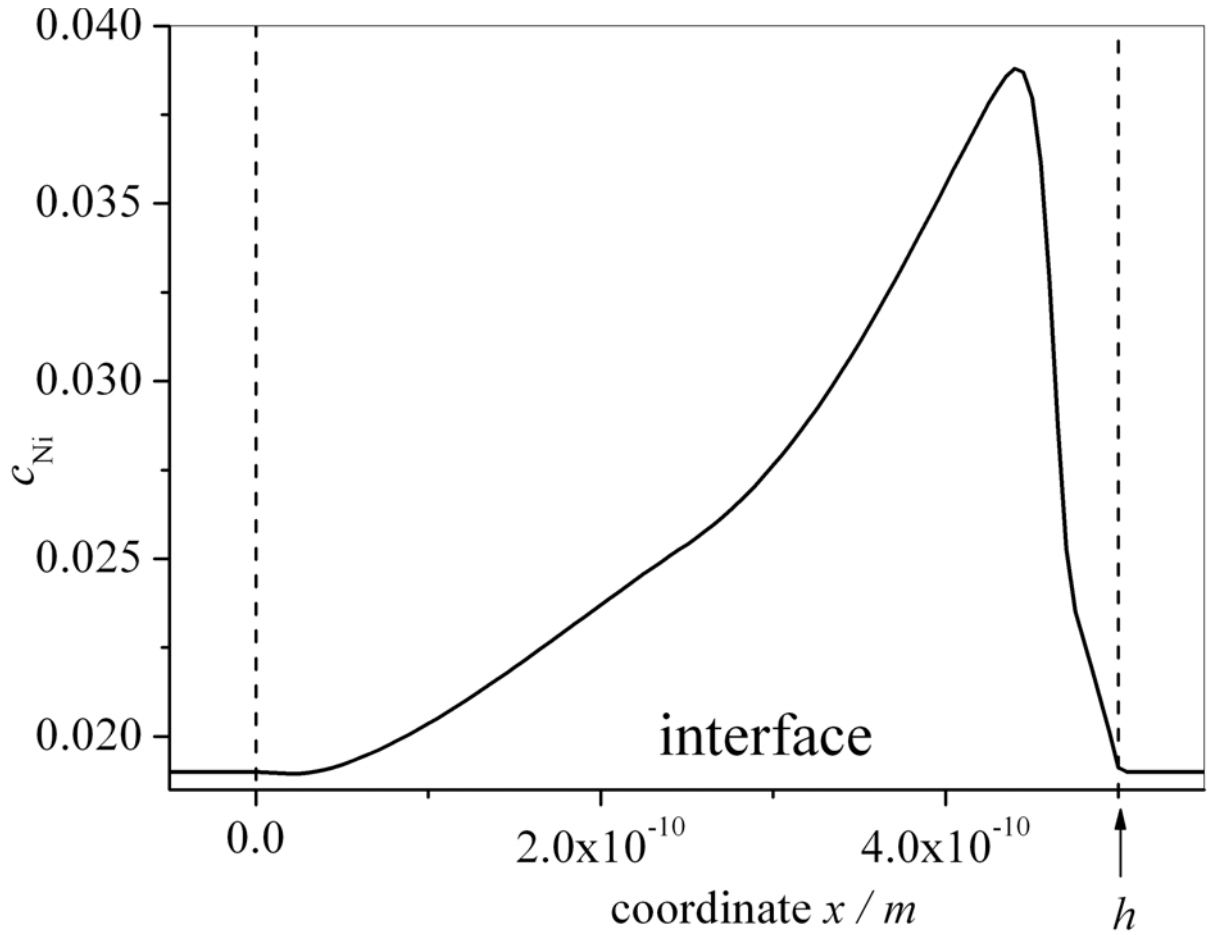
Temperature dependence of the interface velocity *v* for different values of the interface thickness *h*.



Thick interface – Cr-Profile



Thick interface – Ni-Profile



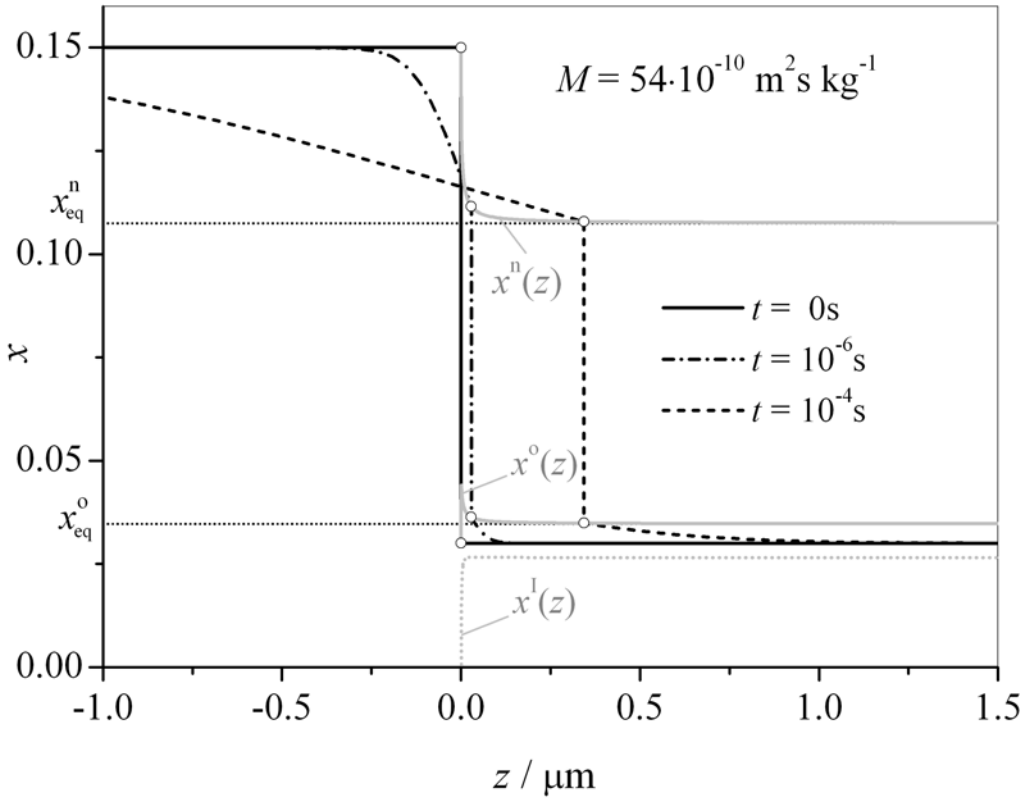
Conclusions and Outlook

- Sharp interface – equal jump contact conditions
- Thick interface with zero fluxes can describe a sharp interface $\Rightarrow$ Trans-interface diffusion is not required for a sharp interface
- Comparison of modelling results with experimental data
- Thick interface – steady state solutions

Next goal:

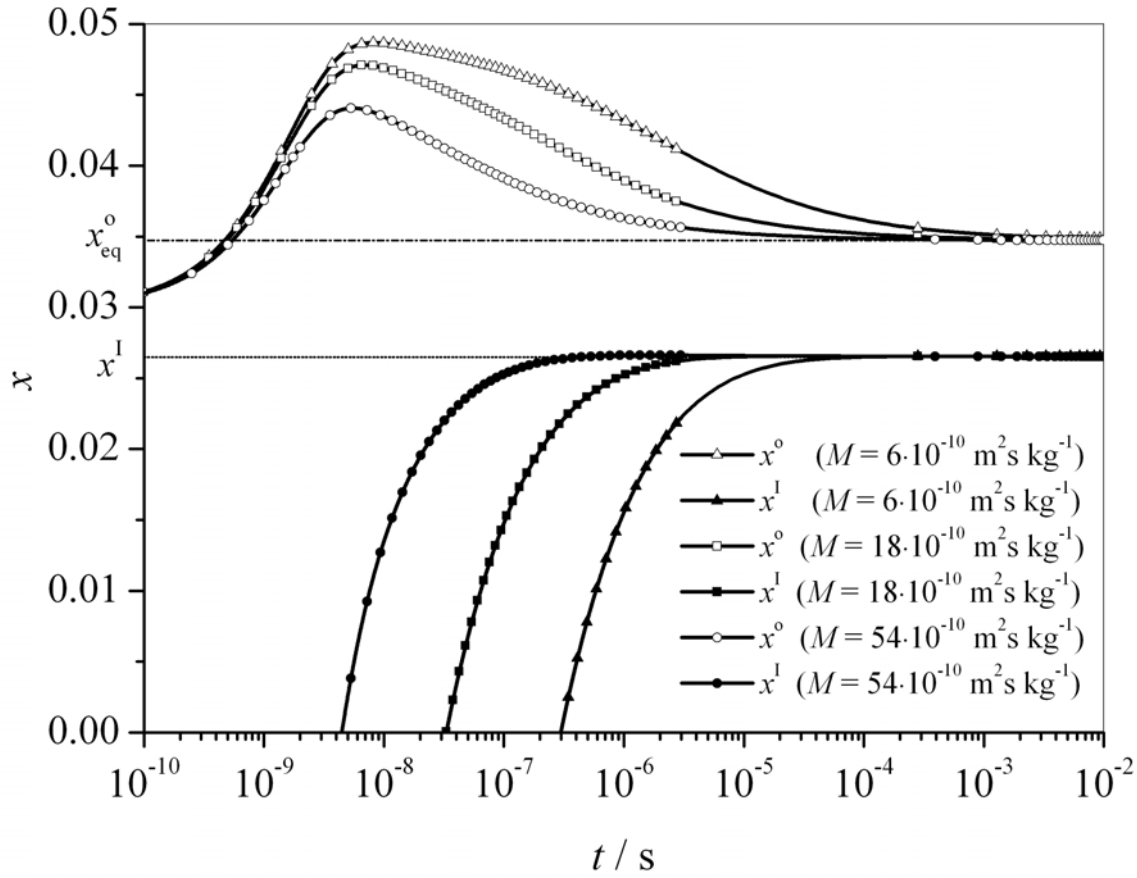
- Thick interface model – general solution

Solid-liquid transformation in the Al-Mg system
Case I: $x^n = 0.15$, $x^o = 0.03$, $T = 883$K



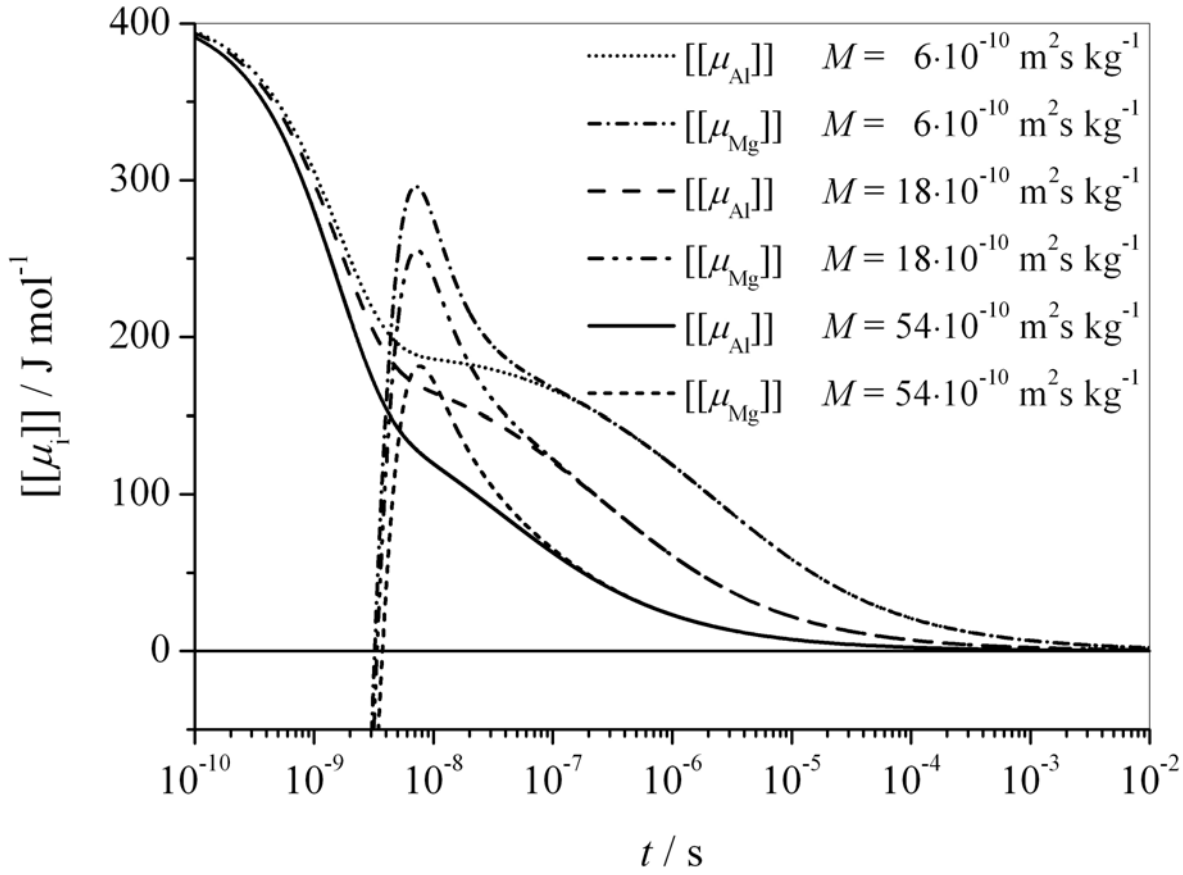
The mole fractions x^o, x^n and x^I are depicted versus the spatial variable z (grey lines).
The mole fraction profiles are plotted at different transformation times.

Solid-liquid transformation in the Al-Mg system
Case I: $x^{n} = 0.15$, $x^{o} = 0.03$, $T = 883$K



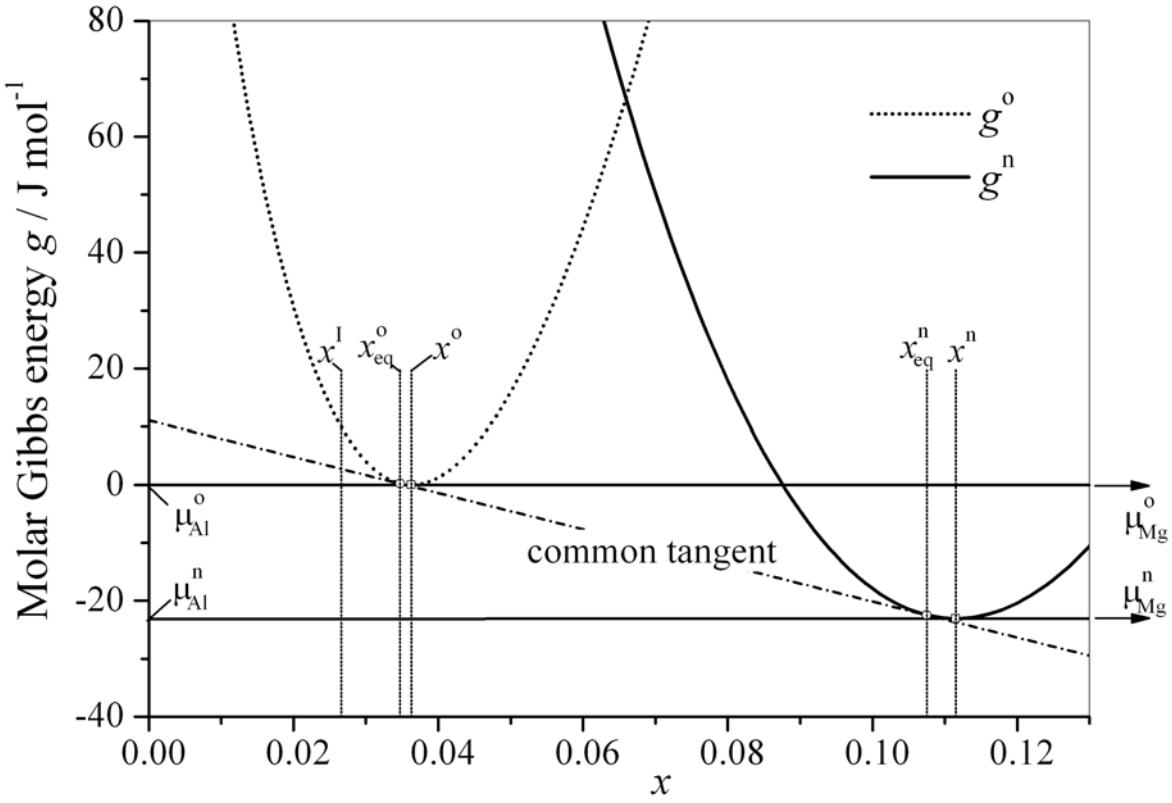
The time-dependent evolution of the mole fractions x^{o} and x^{I} at three interface different interface mobilities M.

Solid-liquid transformation in the Al-Mg system
Case I: $x^n = 0.15$, $x^o = 0.03$, $T = 883$K



The jumps of the chemical potentials, $[[\mu_{Al}]]$ and $[[\mu_{Mg}]]$ vs. time.

Solid-liquid transformation in the Al-Mg system
Case I: $x^n = 0.15$, $x^o = 0.03$, $T = 883$K



Calculated molar Gibbs energy diagram for a certain deviation from local equilibrium (t = 1μs).