Virtual Cyclic Phase Transformation Dilatometer Experiments for Fe-Mn-C by means of Phase Field Simulations

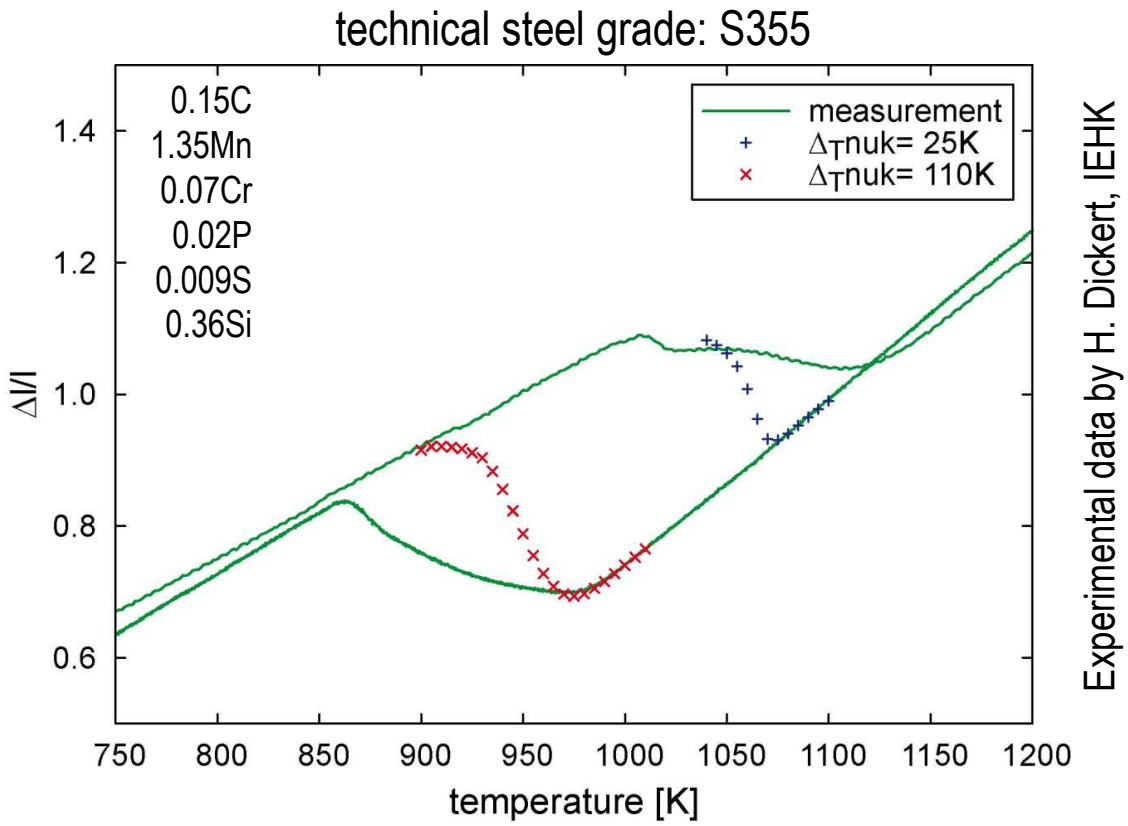
M. Apel, G.Laschet, B.Böttger

Motivation

Phase field model for the computation of dilatometer curves: Apel, Benke, Steinbach, Comp. Mat. Sci. 2009

Simulation input:

- databases TCFE5, MOB2
- cooling rate: 0.5K/s
- high interface mobility for diffusion limited growth



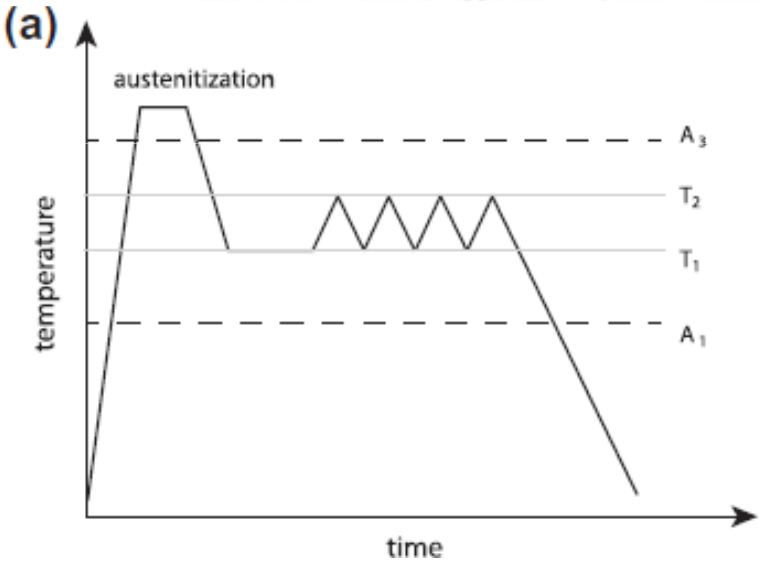
➡ interface mobility and nucleation undercooling needs calibration

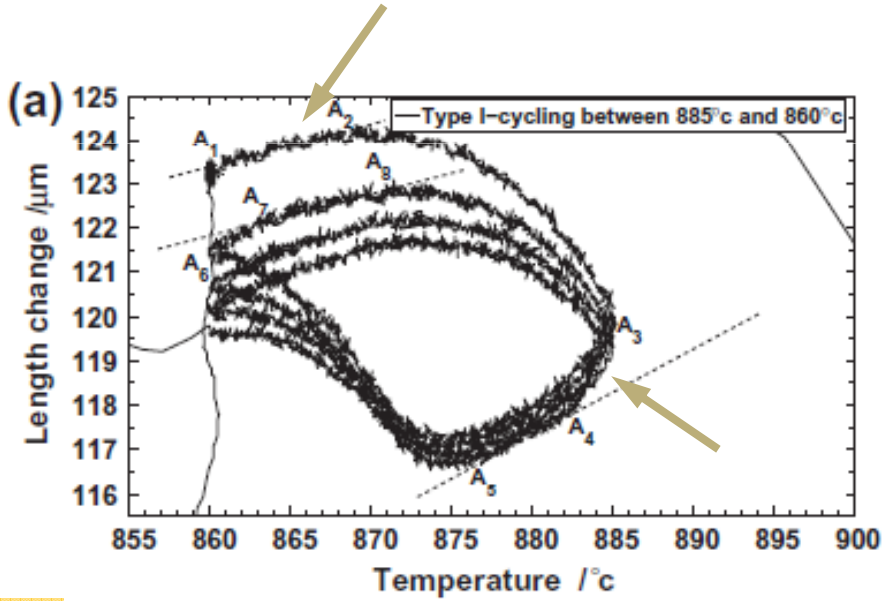
Motivation

Available online at www.sciencedirect.com
ScienceDirect
Acta Materialia 59 (2011) 6751–6760
Acta MATERIALIA
www.elsevier.com/locate/actamat

Application of cyclic partial phase transformations for identifying kinetic transitions during solid-state phase transformations: Experiments and modeling

Hao Chen [a,*], Benoît Appolaire [b], Sybrand van der Zwaag [a]



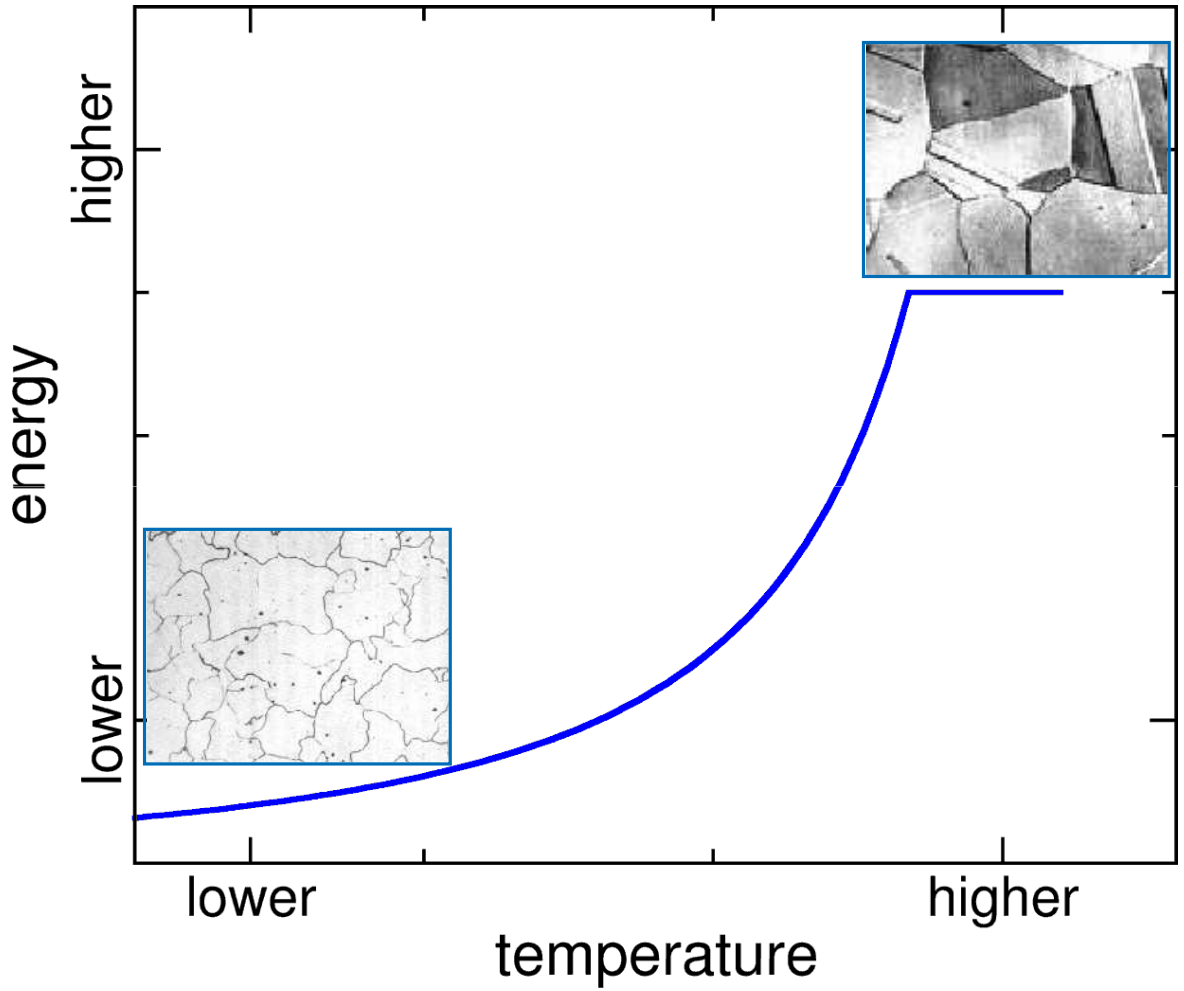


+ more papers and contributions this meeting !!!

Outline

- Introduction
- Phase field simulations (3D)
  - initial microstructure
  - cyclic transformation
- Driving force models – detailed discussion (1D)
- Conclusion

Introduction

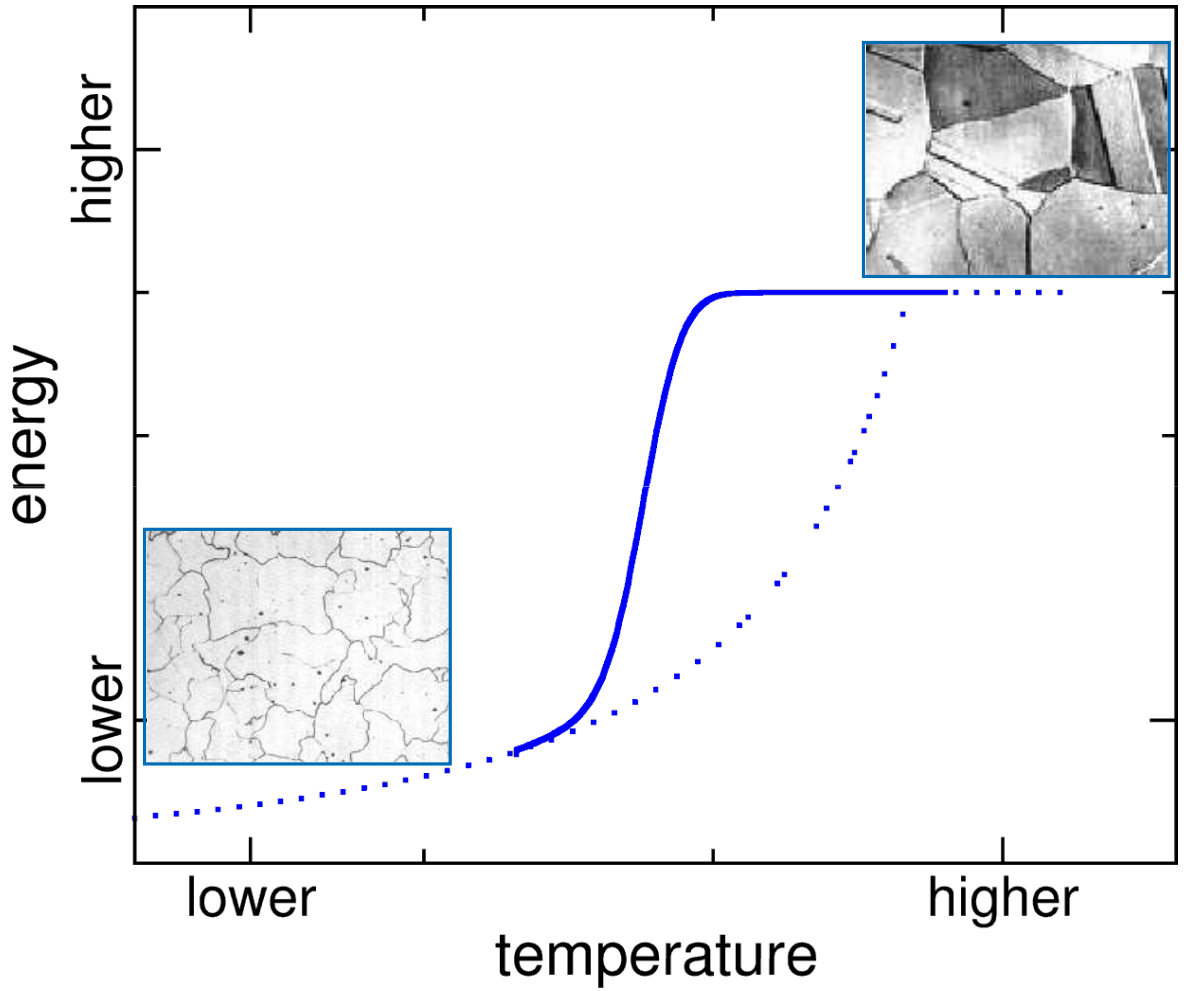


transformation $\gamma \rightarrow \alpha$

needs energy dissipation !

- $\Delta G_{diffusion}$
- $\Delta G_{interface\ motion}$
- …

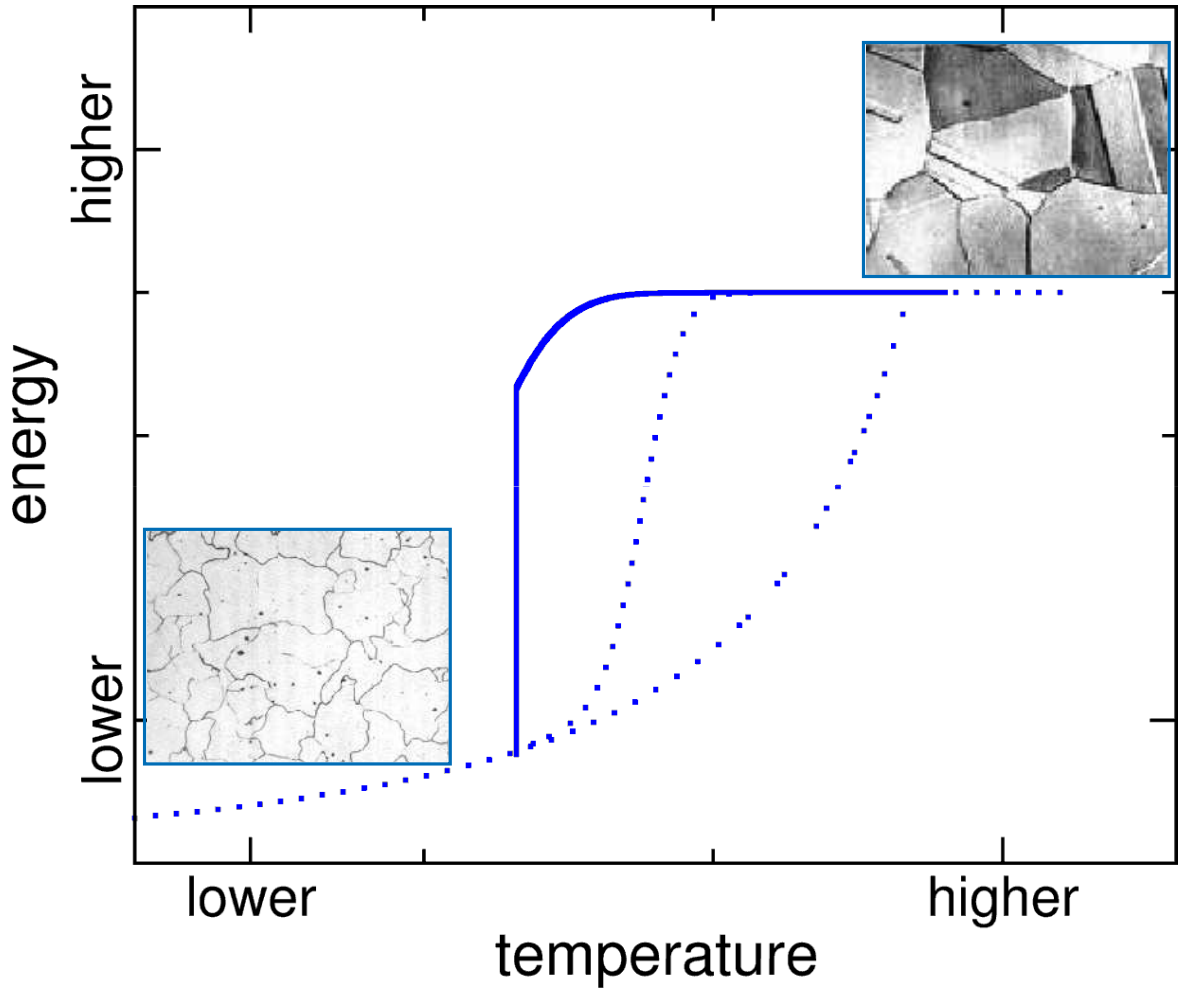
Introduction



Energy dissipation takes time: hysteresis is a natural consequence of kinetic processes

Which way does the transformation choose?

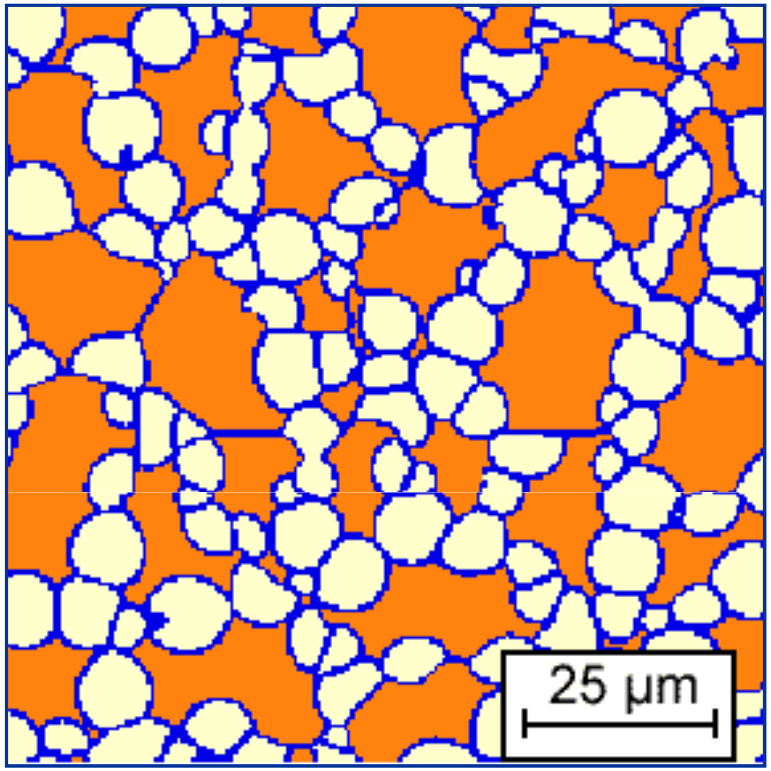
Introduction



Energy dissipation takes time: hysteresis is a natural consequence of kinetic processes

the reaction path reveals the energy dissipation

Introduction



interface velocity = interface mobility × driving force:

$$v = \mu \cdot \Delta G_{local}$$ (not necessarily a linear relationship)

v can be measure precisely, but:

How to decompose v into µ and ΔG?

- sharp interface models: LE, PE, mixed mode, …
- diffuse interface models: PF, MPF, TEP, …

$$\Delta G_{tot} = v/\mu + \Delta G_{diffusion} + \ldots$$

Estimate from thin interface assymptotics: $\mu > 10^{-3}$ cm^4/Js for diffusion controlled limit
Consistent with experimental findings: $\mu \approx 10^{-5}$ cm^4/Js „mixed mode“ transformation

Diffuse interface discription of a phase field model

Gibbs-Thomson condition

$$T = T_m - m_l c_l - \Gamma\kappa - \beta v$$

Stefan Condition

$$c_l(1-k)v_n = D_s(\partial_n c)_s - D_l(\partial_n c)_l$$

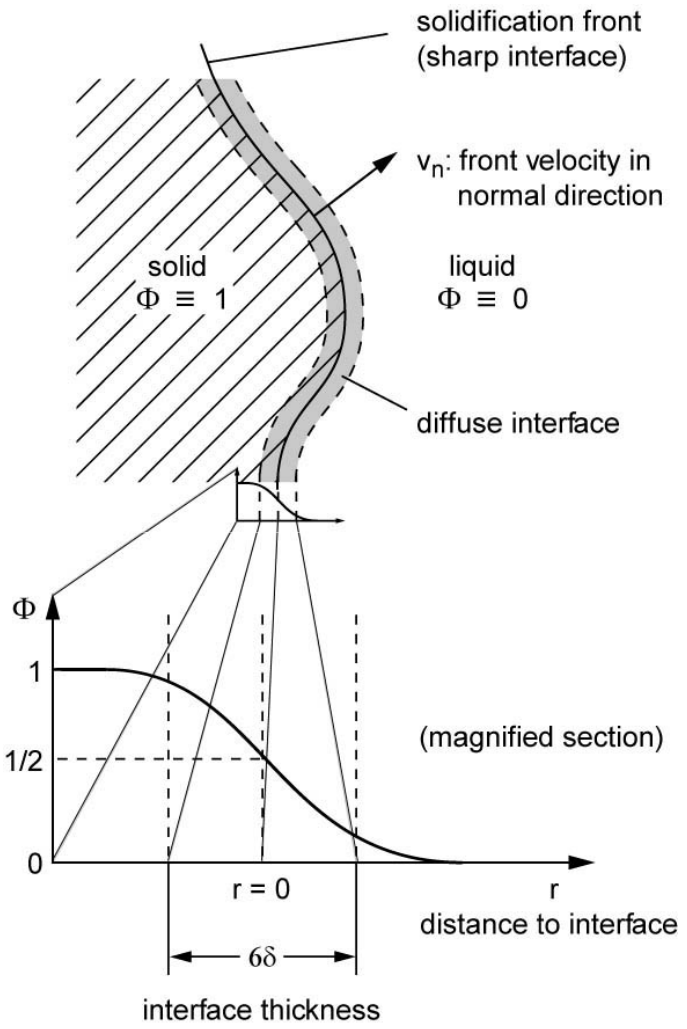
PFM: Order parameter for state of phase

ϕ:=0 solid ϕ :=1 liquid 0< ϕ<1 interface

Free energy relaxes into minimum

$$\tau\frac{d}{dt}\phi = \frac{\delta}{\delta\phi}F[\phi,\nabla\phi,T,\mu(c),...]$$

Aim: recover BC for sharp interface e.g. by matching assymptotics



Multiphase field model (MPF)

Phase-Field

$$\dot{\phi}_\alpha = \frac{1}{n}\sum_\beta \mu_{\alpha\beta}\left(\frac{\delta f}{\delta\phi_\alpha} - \frac{\delta f}{\delta\phi_\beta}\right) = \frac{1}{n}\sum_\beta \mu_{\alpha\beta}\left(l_{\alpha\beta} + \Delta G^{chem}_{\alpha\beta} + \Delta G^{el}_{\alpha\beta}\right)$$

Eiken, Böttger, Steinbach: Phys. Rev. E (2006)

- Free energies $\left(f = f^i + f^{ch} + f^{el} + \ldots\right)$ $f^i = \sum_{\alpha,\beta} \frac{4\sigma_{\alpha\beta}}{\eta_{\alpha\beta}}\left\{\frac{\eta^2_{\alpha\beta}}{\pi^2}\left|\nabla\phi_\alpha\nabla\phi_\beta\right| + \phi_\alpha\phi_\beta\right\}$

$$f^{ch} = \sum_\alpha \phi_\alpha f^{ch}(c_\alpha) + \mu\left(c - \sum_\alpha \phi_\alpha c_\alpha\right)$$

Multicomponent diffusion

$$\dot{c} = \nabla\left(\sum_\alpha \phi_\alpha \vec{\vec{D}}_\alpha \nabla\vec{c}_\alpha\right)$$

The Multiphase-Field Model: Extension towards Elasticity

Phase-Field

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$$f^{el} = \sum_\alpha \phi_\alpha \frac{1}{2}\left(\varepsilon_\alpha - \varepsilon^*_\alpha - \varepsilon^{th}_\alpha(T - T_0)\right)C_\alpha\left(\varepsilon_\alpha - \varepsilon^*_\alpha - \varepsilon^{th}_\alpha(T - T_0)\right)$$

Multicomponent diffusion

$$\dot{\vec{c}} = \nabla\left(\sum_\alpha \phi_\alpha \vec{\vec{D}}_\alpha \nabla\vec{c}_\alpha\right)$$

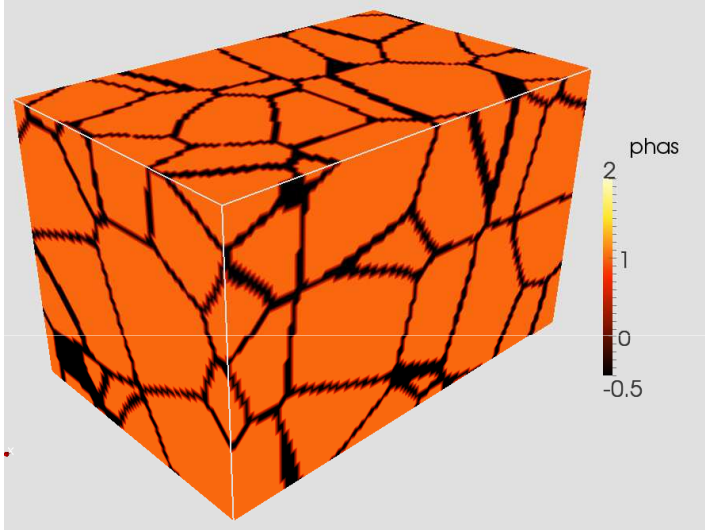
Mechanical equilibrium

$$0 = \nabla\sigma = \sum_\alpha \nabla\phi_\alpha C_\alpha\left(\varepsilon_\alpha - \varepsilon^*_\alpha - \varepsilon^{th}_\alpha(T - T_0)\right)$$

Steinbach, Apel: Physica D 54 (2006)

Virtual phase field experiment

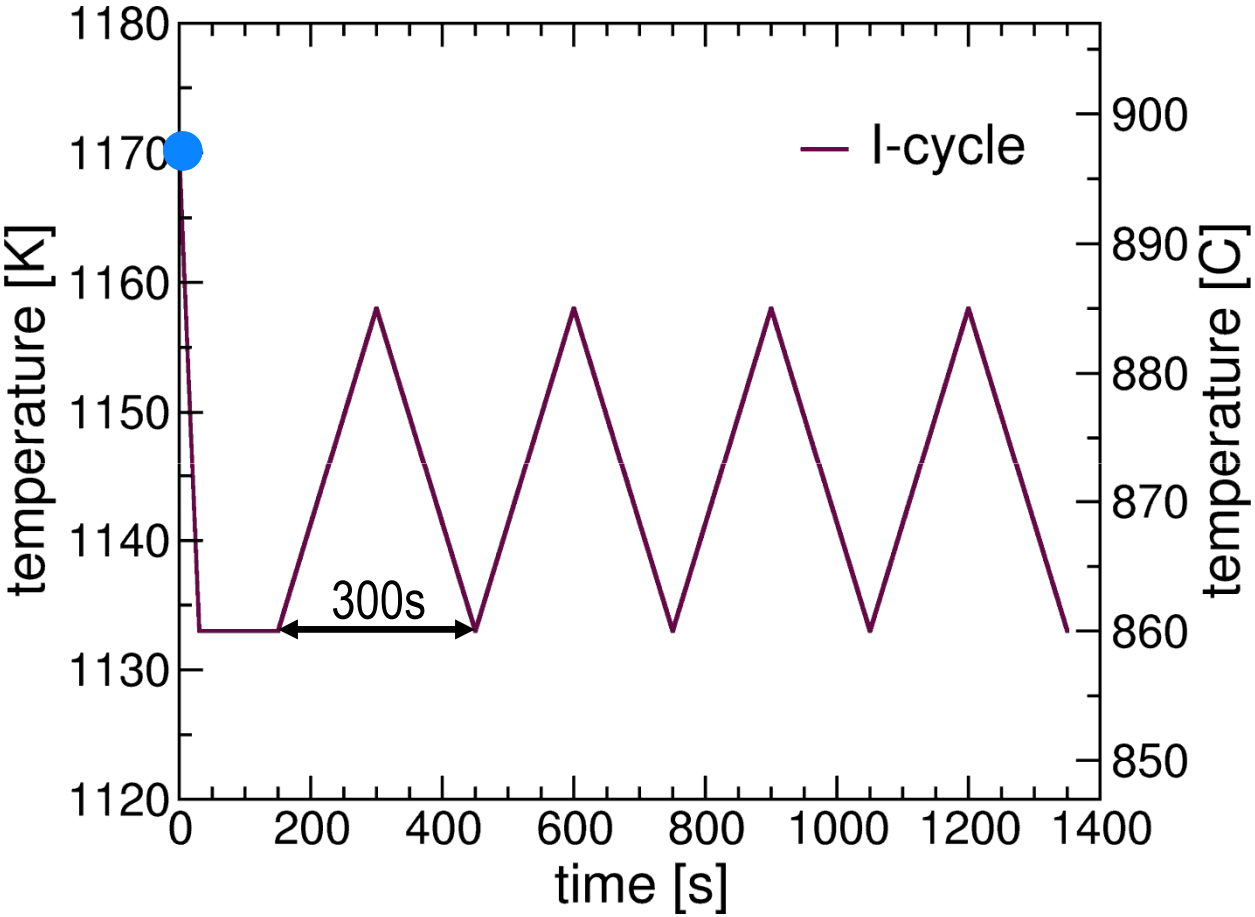
t=0s, T=895°C



domain size: 55µm × 35µm × 35µm

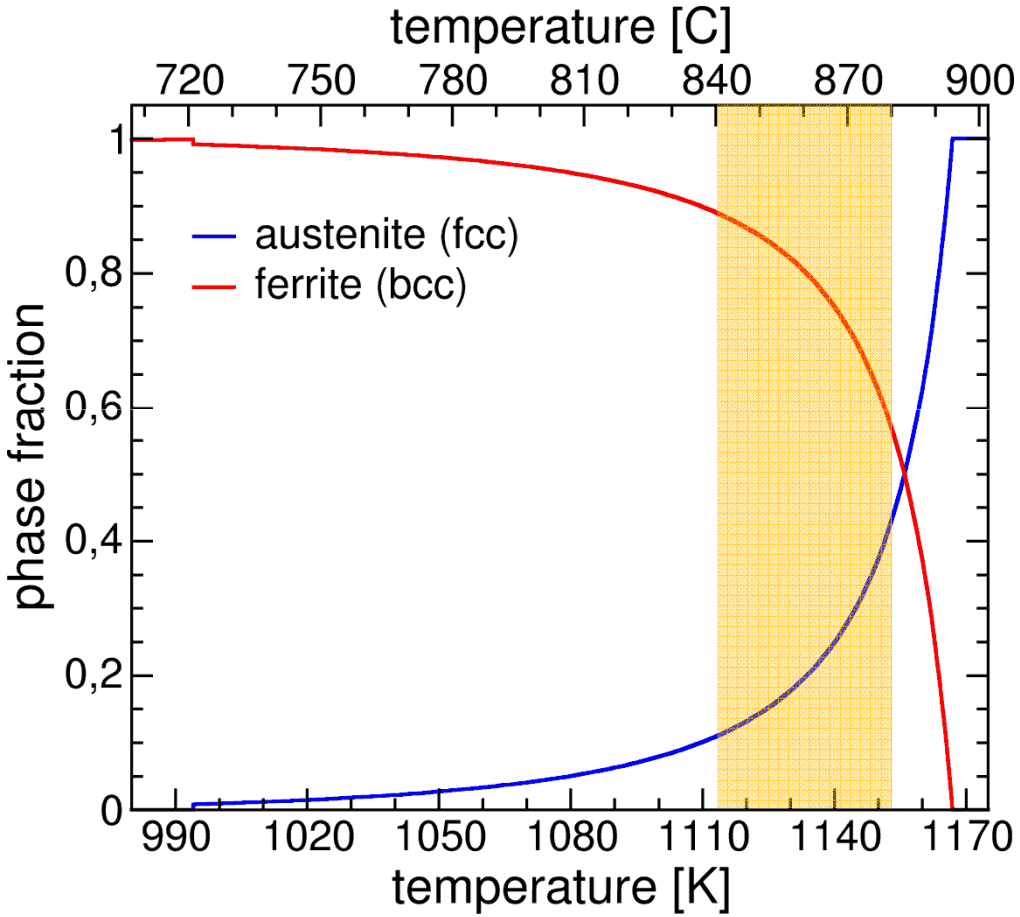
30 austenite grains

$\langle r_\gamma \rangle$ = 8.1µm



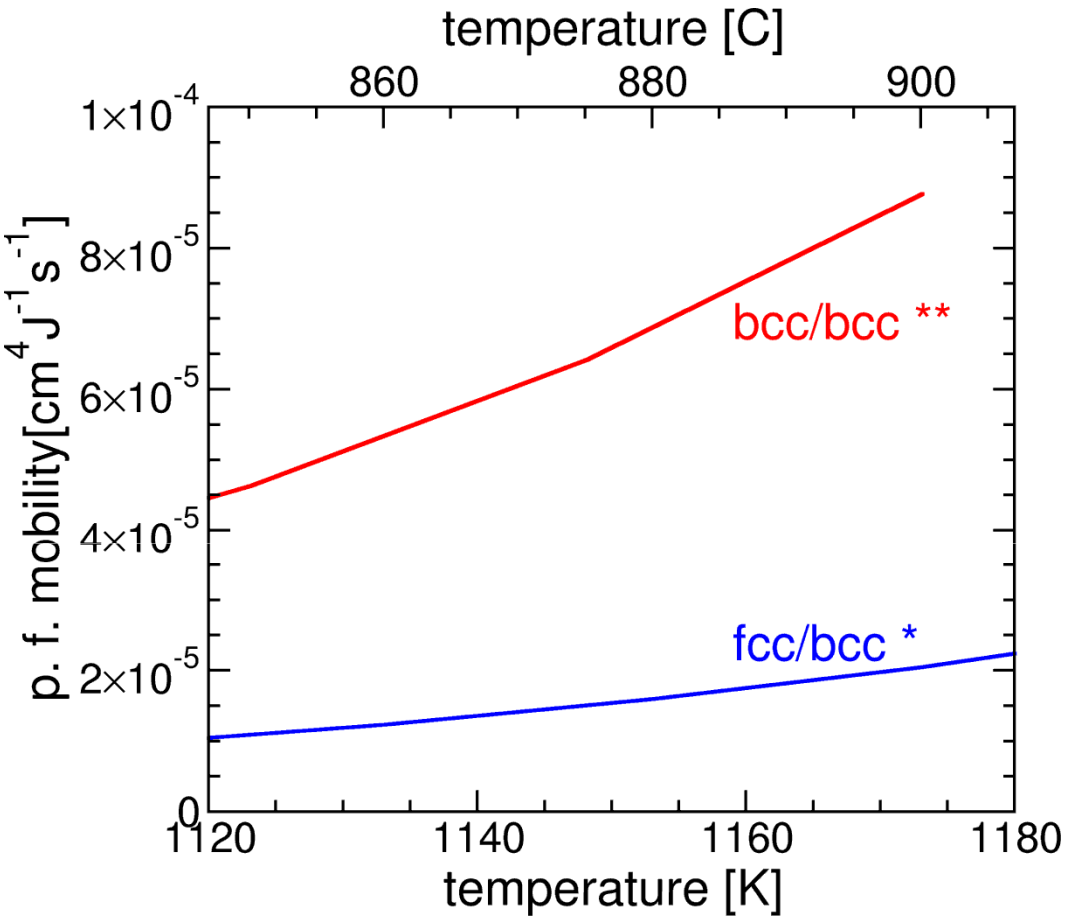
Thermodynamic equilibrium

- Database TCFE6 (Thermocal AB)



Fe	balance
C [wt %]	0.023
Mn [wt %]	0.17
Si [wt %]	0.009

Interface mobility (temperature dependent)



- μ(γ/γ) set identical to μ(γ/α)

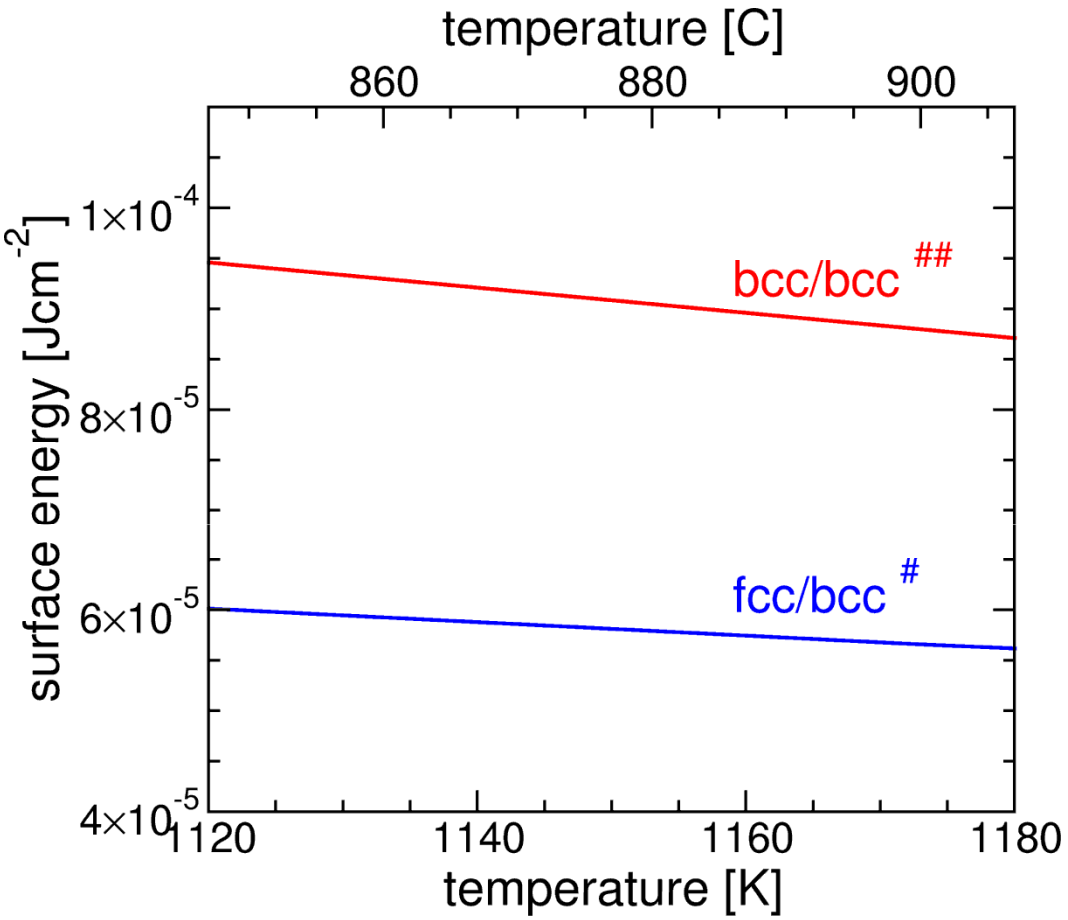
Estimate for kinetic undercooling:

ΔG_{kin} = v/μ = 10μm/100s / 10^{-5}cm^{4}J^{-1}s^{-1}
= 1 Jcm^{-3}

energy dissipation by interface „friction“

[#] γ/α: Mecozzi et al., Comp. Mat. Sci. 50 (2011) p.1846
[##] α/α: Rudnitzki et al

Interfacial energy (temperature dependent)

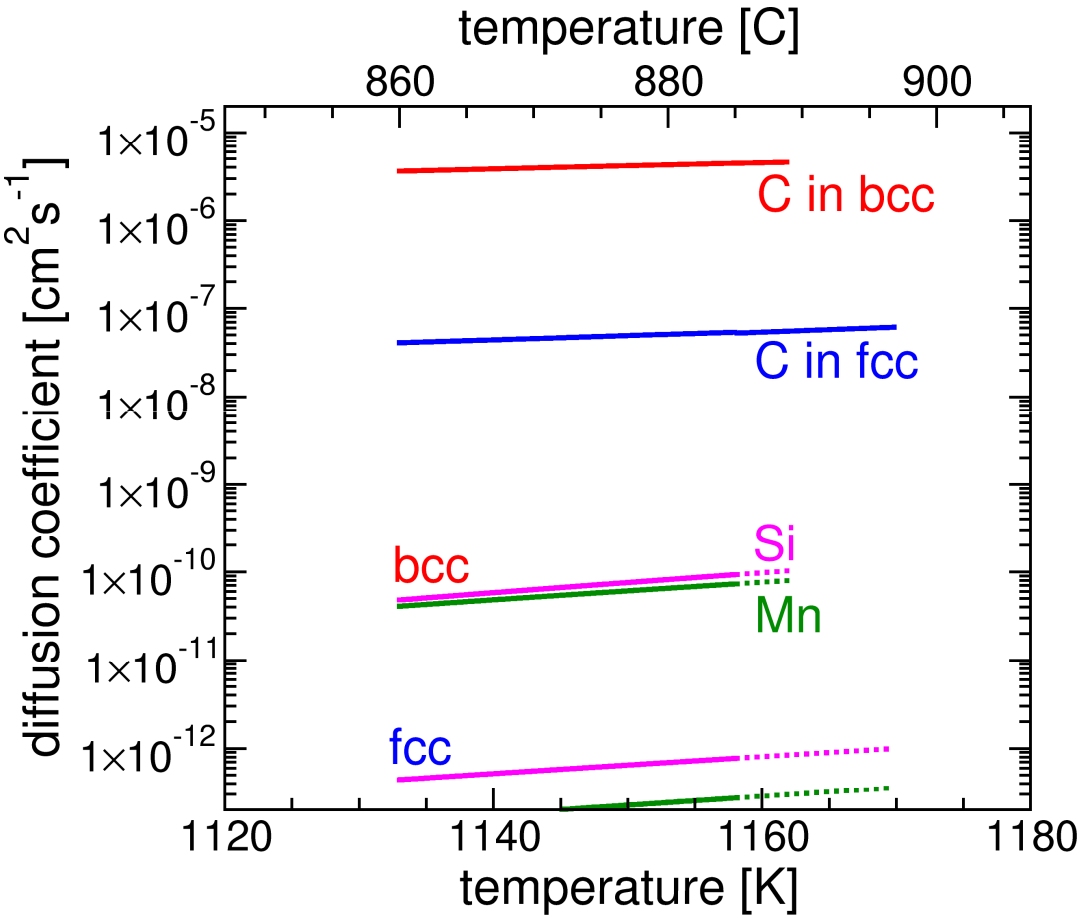


- μ(γ/γ) set identical to μ(γ/α)

„dislocation“ based model

Diffusion coefficients

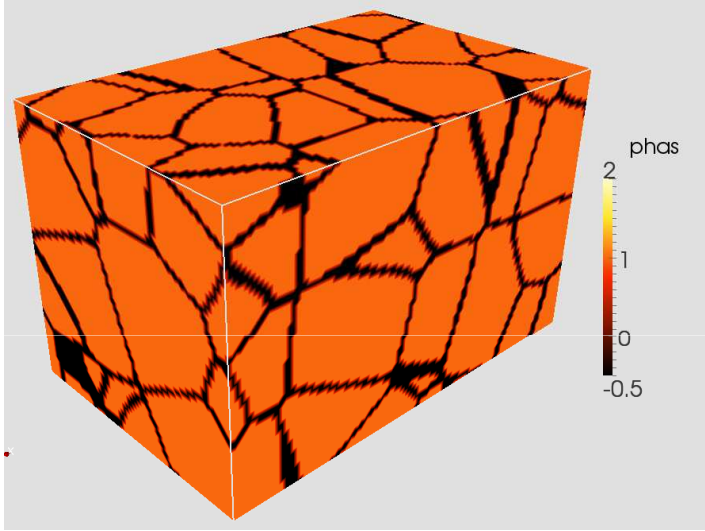
- Mobility data from MOBFE2 (Thermocalc)



approx. diffusion length for 150s @ 880°C		
	bcc	fcc
C	250µm	25µm
Mn	1µm	0.1µm
Si	1µm	0.1µm

Virtual phase field experiment

t=0s, T=895°C



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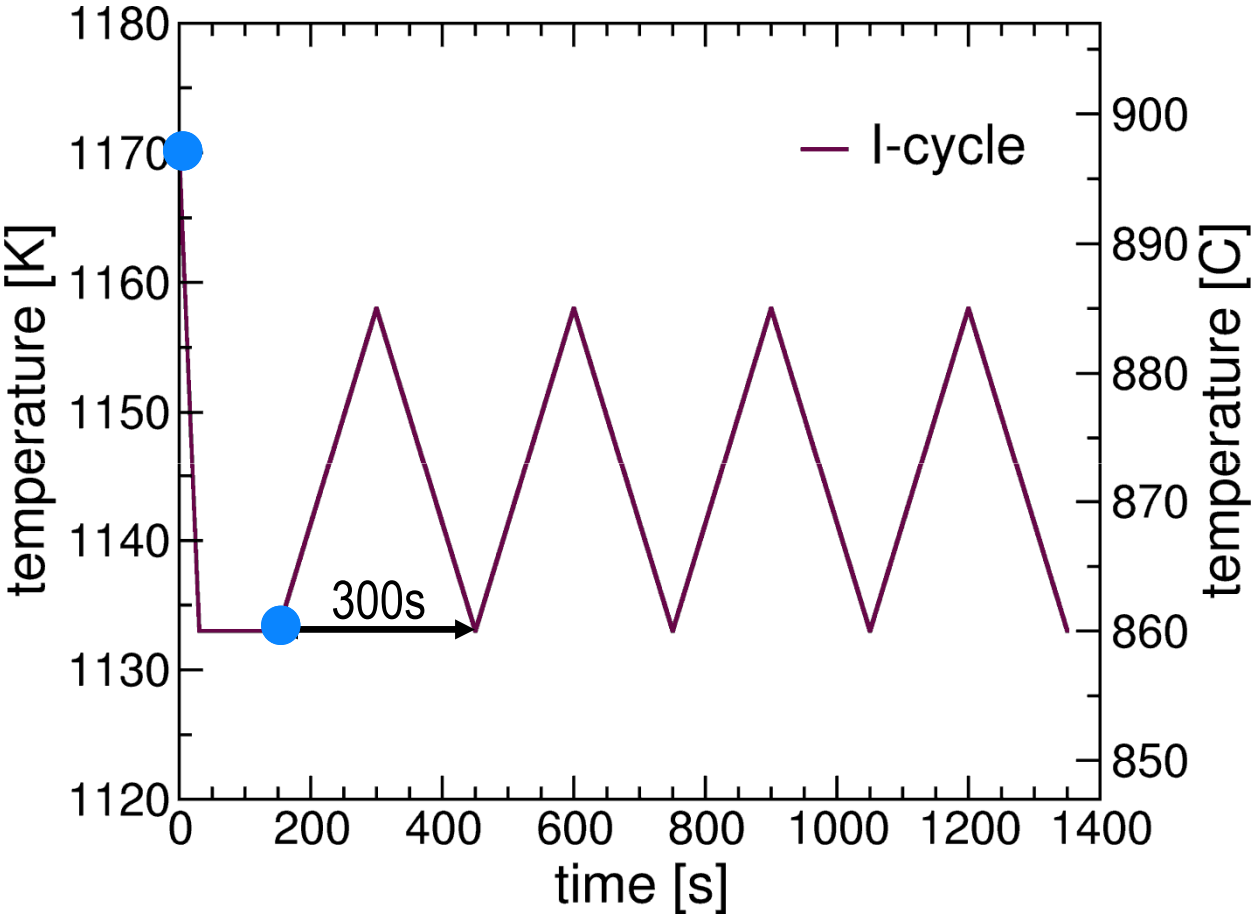
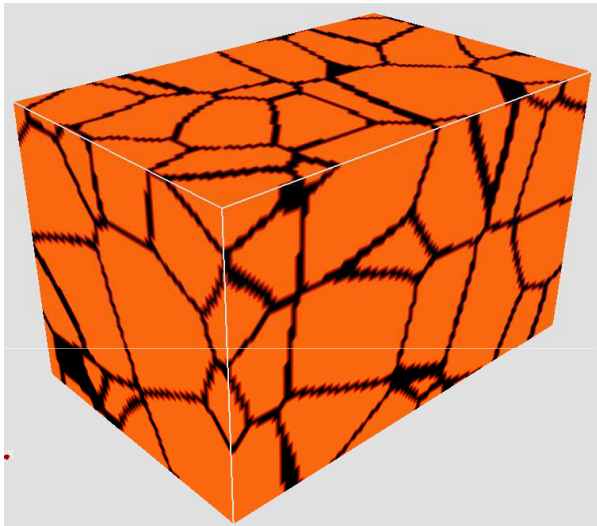


Initial grain structure (895°C → 860°C)

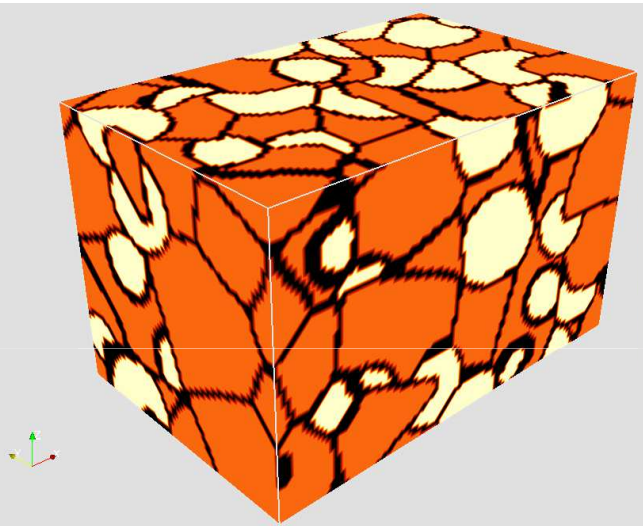
t=0s, T=895°C



30 austenite grains

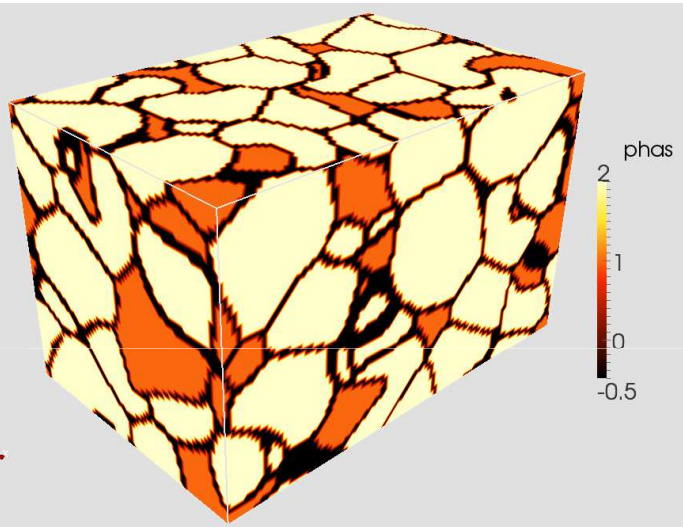
$\langle r_\gamma \rangle$ = 8.1µm

t=30s, T=860°C



ferrite nucleation at quadrupel points,
fix orientation relationship

t=150s, T=860°C



30 austenite + 44 ferrite grains

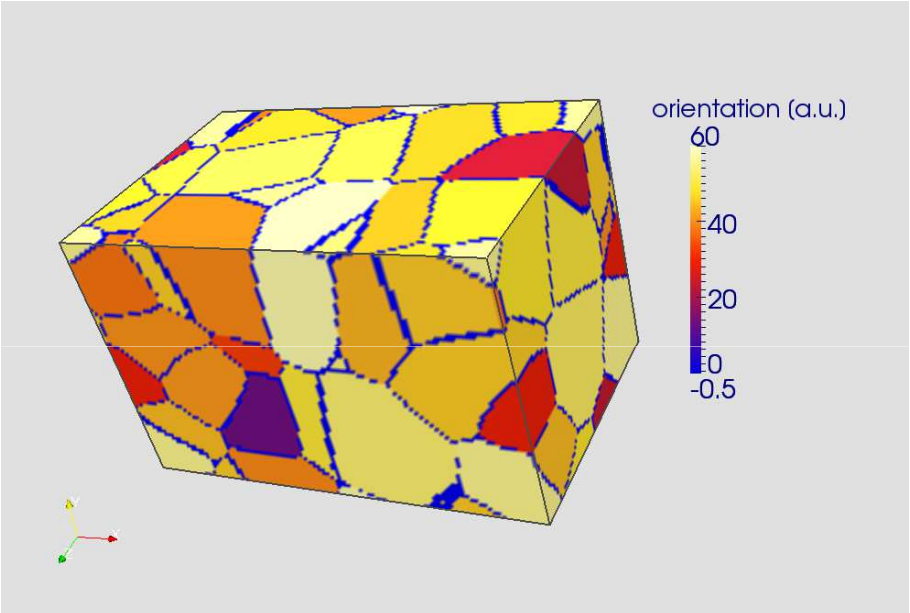
$\langle r_\gamma \rangle$ = 4.7µm

$\langle r_\alpha \rangle$ = 6.7µm

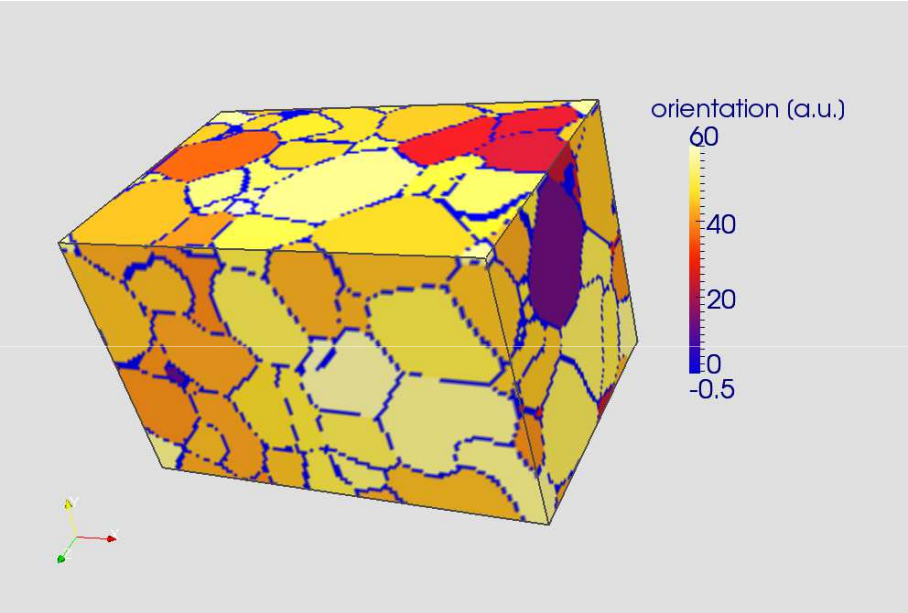
$\langle r \rangle$ = 6.1µm

Initial grain structure: orientation

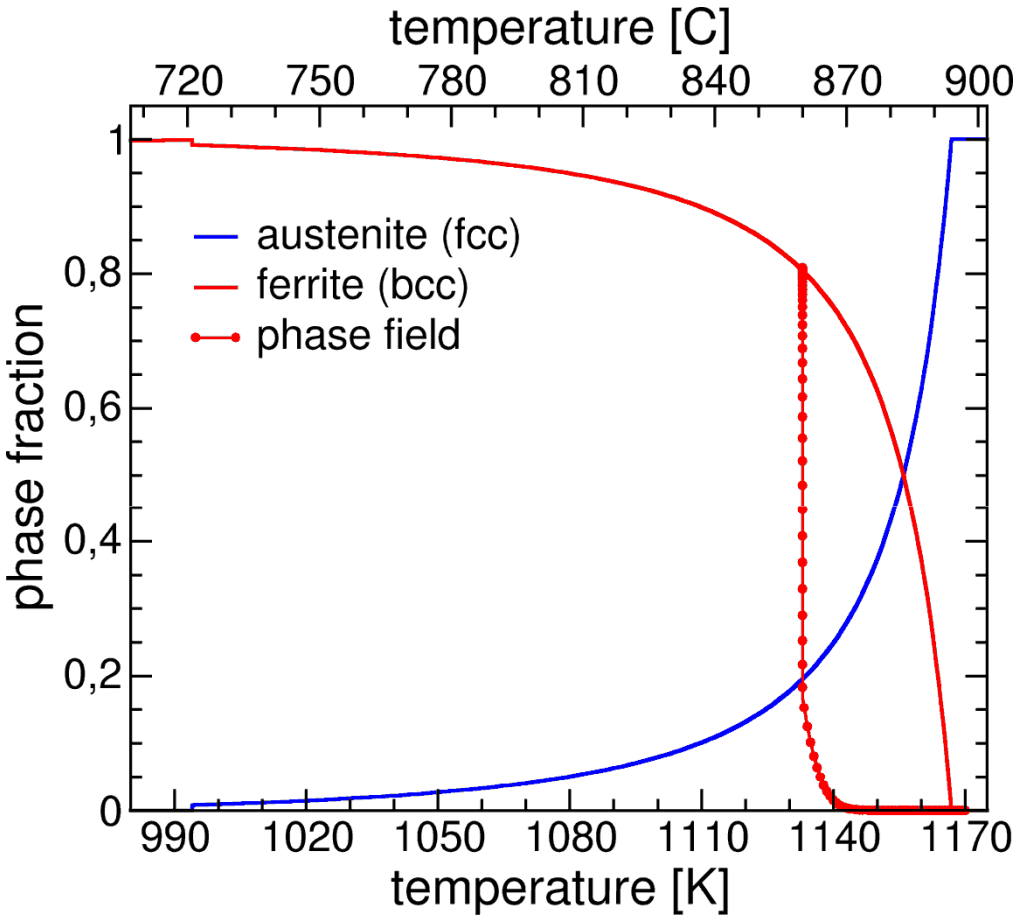
t=0s, T=895°C



t=150s, T=860°C

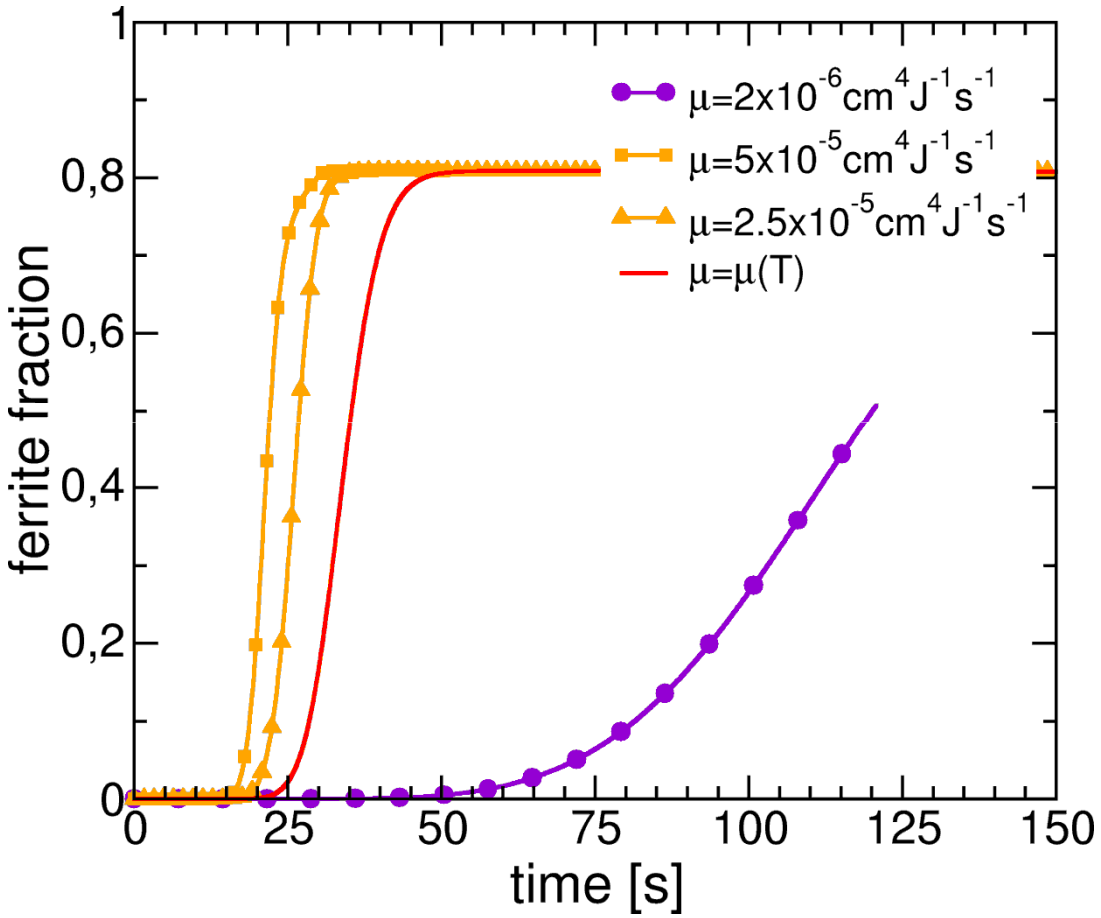


Initial cooling into 2-phase region



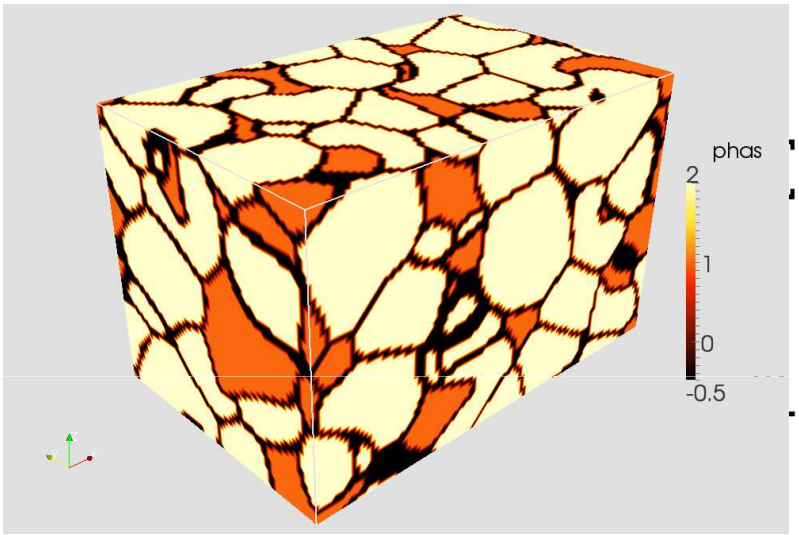
Equilibrium phase fraction reached

Initial cooling into 2-phase region: phase fraction over time



Virtual phase field experiment

t=150s, T=860°C

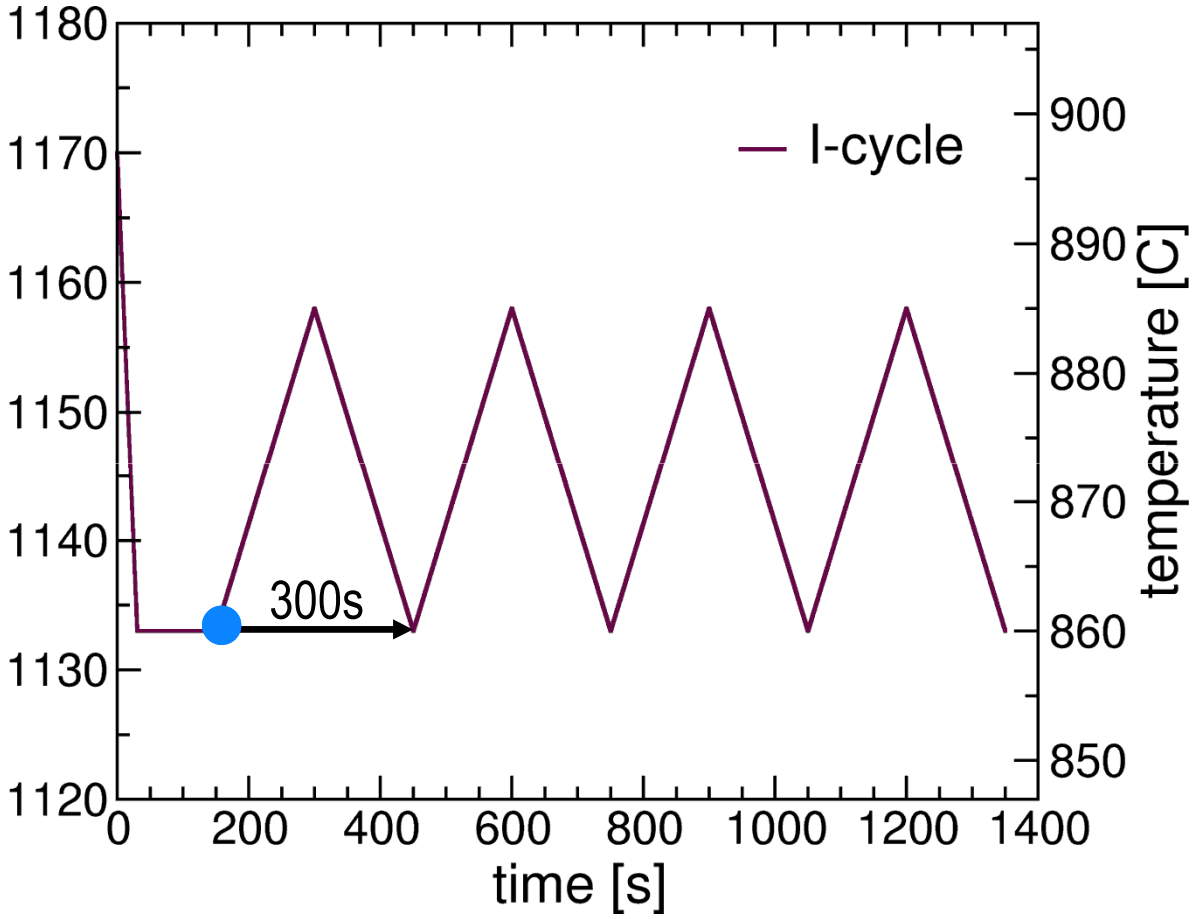


30 austenite + 44 ferrite grains

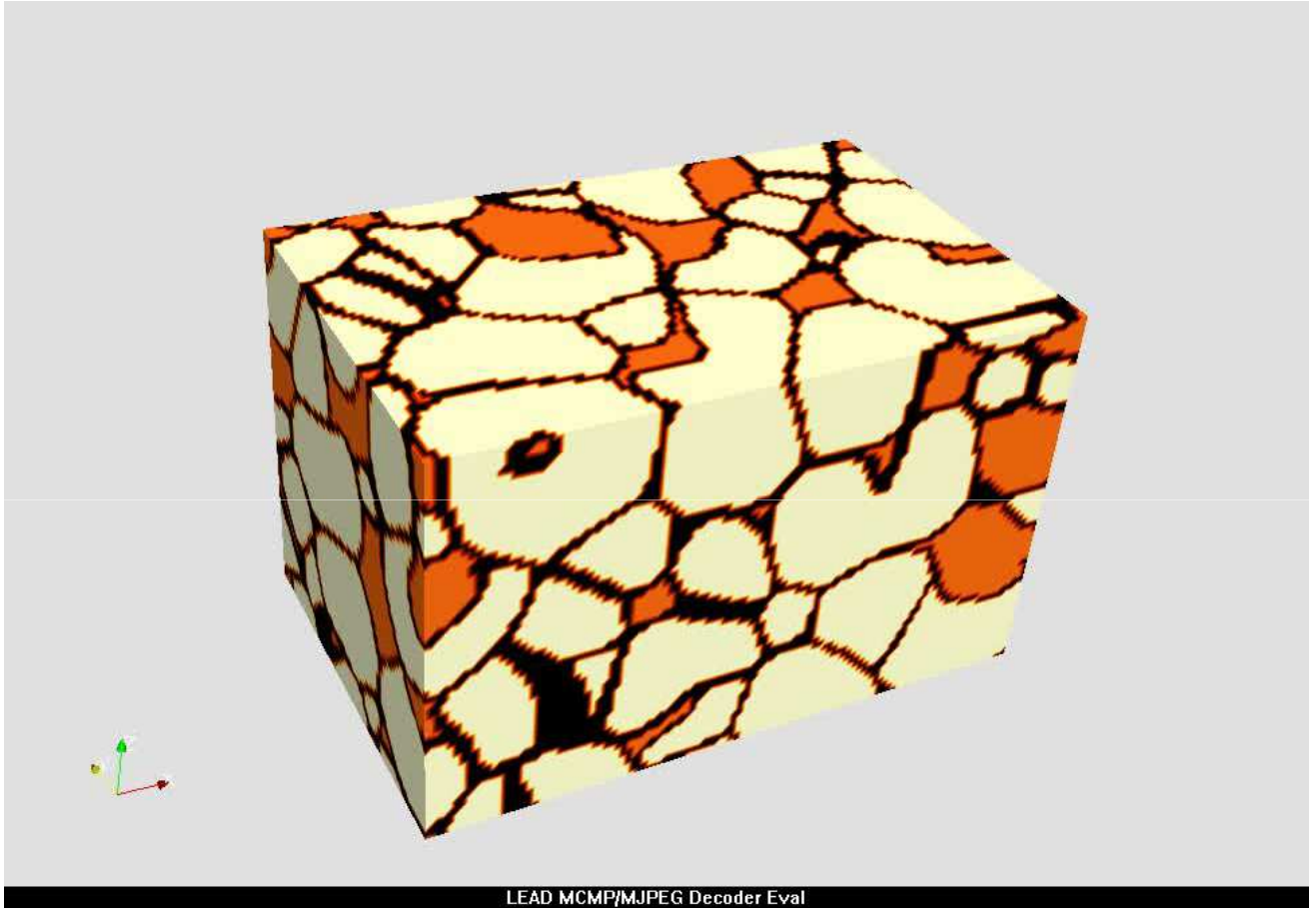
$\langle r_\gamma \rangle$ = 4.7µm

$\langle r_\alpha \rangle$ = 6.7µm

$\langle r \rangle$ = 6.1µm



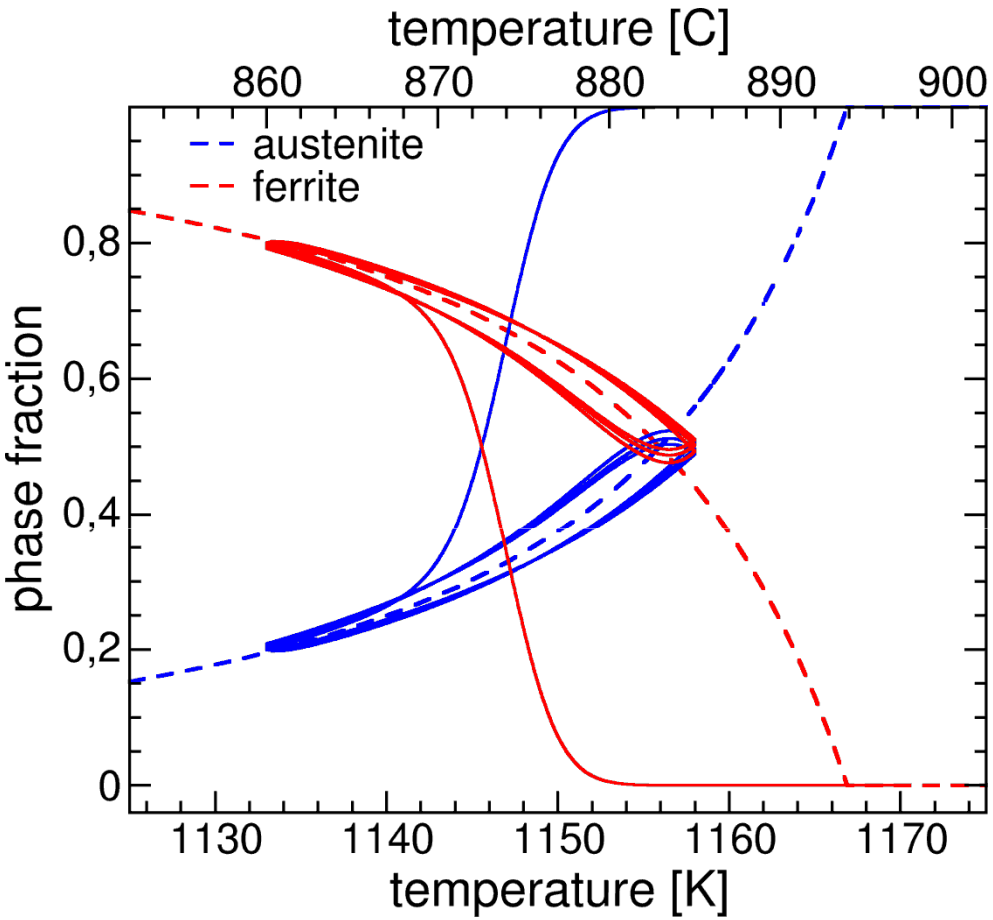
Cyclic transformation



austenite

ferrite

I-cycle: MPF model



Multiphase field model (MPF): computation of length changes

Phase-Field

$$\dot{\phi}_\alpha = \frac{1}{n}\sum_\beta \mu_{\alpha\beta}\left(\frac{\delta f}{\delta\phi_\alpha} - \frac{\delta f}{\delta\phi_\beta}\right) = \frac{1}{n}\sum_\beta \mu_{\alpha\beta}\left(l_{\alpha\beta} + \Delta G^{chem}_{\alpha\beta} + \Delta G^{el}_{\alpha\beta}\right)$$

Eiken, Böttger, Steinbach: Phys. Rev. E (2006)

- Elastic free energy

$$f^{el} = \sum_\alpha \phi_\alpha \frac{1}{2}\left(\varepsilon_\alpha - \varepsilon^*_\alpha - \varepsilon^{th}_\alpha (T - T_0)\right) C_\alpha \left(\varepsilon_\alpha - \varepsilon^*_\alpha - \varepsilon^{th}_\alpha (T - T_0)\right)$$

Mechanical equilibrium

$$0 = \nabla\sigma = \sum_\alpha \nabla\phi_\alpha C_\alpha \left(\varepsilon_\alpha - \varepsilon^*_\alpha - \varepsilon^{th}_\alpha (T - T_0)\right)$$

Steinbach, Apel: Physica D 54 (2006)

➡ here we do not consider the contribution to the driving force

Mechanical material properties

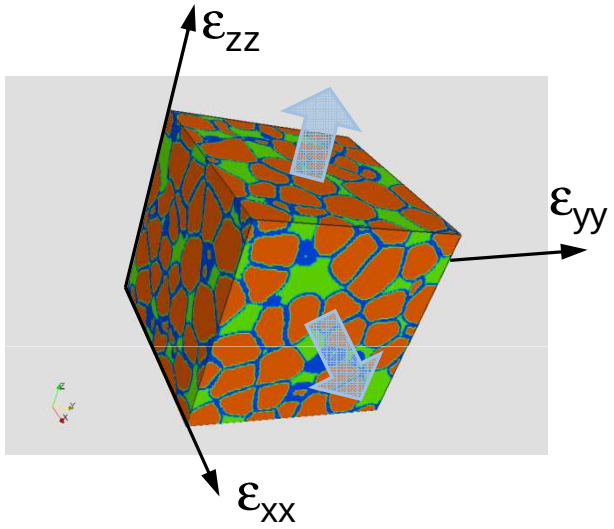
- temperature and composition dependent literature data

Fe	fcc (@950°C)	bcc (@850°C)
C_{11} [GPa]	188.3#	133.9#
C_{12}[GPa]	162.6#	77.3#
C_{44}[GPa]	95.00#	67.7#
molar volume [cm^3]	7.2589##	7.3495##
thermal expansion coeff.##	$2.192 \cdot 10^{-5}$	$1.285 \cdot 10^{-5}$

Ghosh & Olson, Acta Mat. 50 (2002) p. 2655 and Rayne & Chandrasekhar, Phys. Rev 122 (1961) p.1714
Cho et al. Met.& Mater. Trans. A, vol 42A (2011), p. 2094

Virtual dilatometer signal

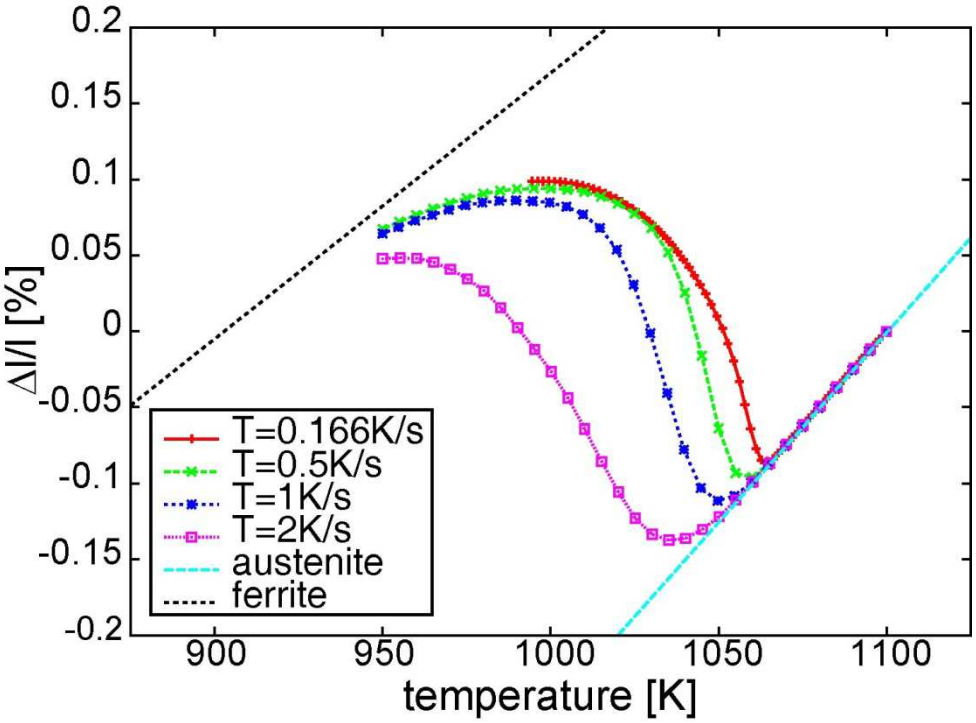
- Calculating the volume expansion of the 2-phase microstructure during phase transformation



$\sigma_{xx} = \sigma_{yy} = \sigma_{zz} = 0$

Boundary conditions:

free expansion but preserving a cubic shape



- Uniform expansions ⇨ isotropic behavior of the polycrystalline cube

The Multiphase-Field Model: Extension towards Elasticity

Phase-Field

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- Free energies $\left(f = f^i + f^{ch} + f^{el} + \ldots\right)$

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$$f^{ch} = \sum_\alpha \phi_\alpha f^{ch}(c_\alpha) + \mu\left(c - \sum_\alpha \phi_\alpha c_\alpha\right)$$

$$f^{el} = \sum_\alpha \phi_\alpha \frac{1}{2}\left(\varepsilon_\alpha - \varepsilon^*_\alpha - \varepsilon^{th}_\alpha(T - T_0)\right)C_\alpha\left(\varepsilon_\alpha - \varepsilon^*_\alpha - \varepsilon^{th}_\alpha(T - T_0)\right)$$

Multicomponent diffusion

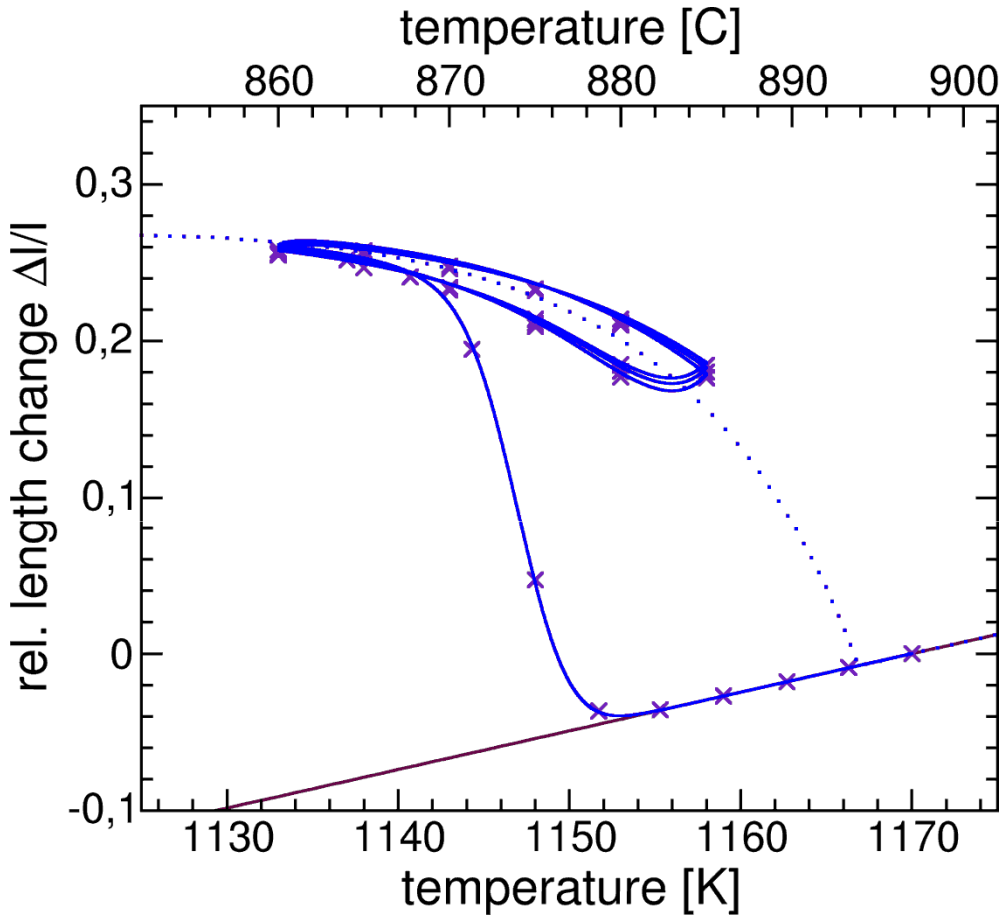
$$\dot{\vec{c}} = \nabla\left(\sum_\alpha \phi_\alpha \vec{\vec{D}}_\alpha \nabla\vec{c}_\alpha\right)$$

Mechanical equilibrium

$$0 = \nabla\sigma = \sum_\alpha \nabla\phi_\alpha C_\alpha\left(\varepsilon_\alpha - \varepsilon^*_\alpha - \varepsilon^{th}_\alpha(T - T_0)\right)$$

Steinbach, Apel: Physica D 54 (2006)

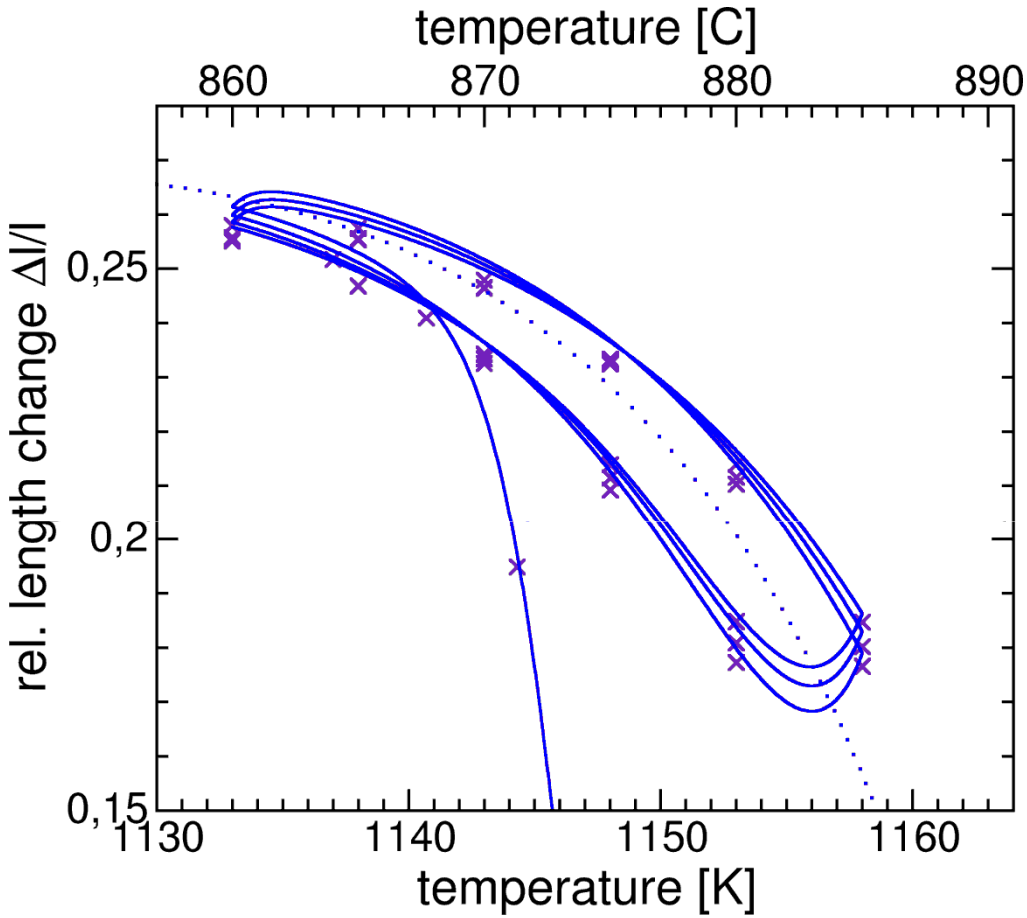
I-cycle: MPF model



$$\Delta l/l = f_\alpha \cdot \alpha_\alpha + f_\gamma \cdot \alpha_\gamma + f_\alpha \cdot \Delta_{mol}$$

linear relationship between phase fraction and dilatation is reasonable

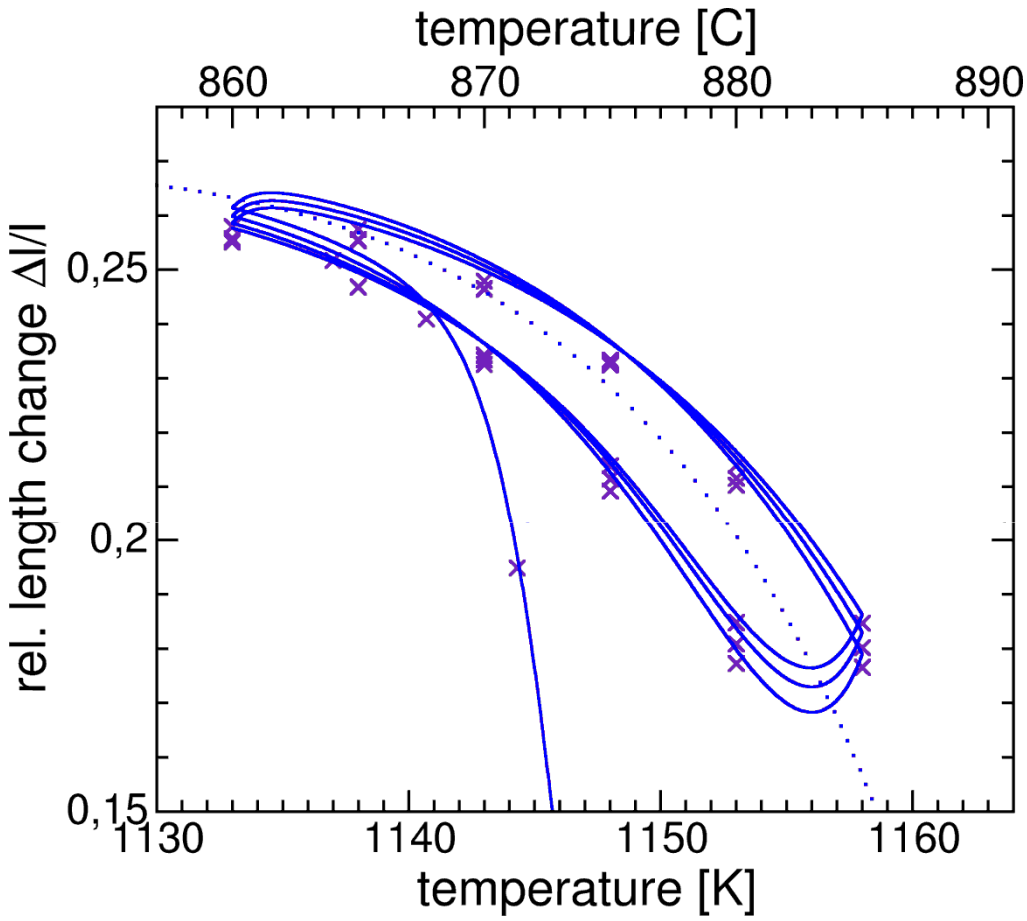
I-cycle: MPF model



$$\Delta l/l = f_\alpha \cdot \alpha_\alpha + f_\gamma \cdot \alpha_\gamma + f_\alpha \cdot \Delta_{mol}$$

minor differences caused by the different mechanical properties of austenite and ferrite

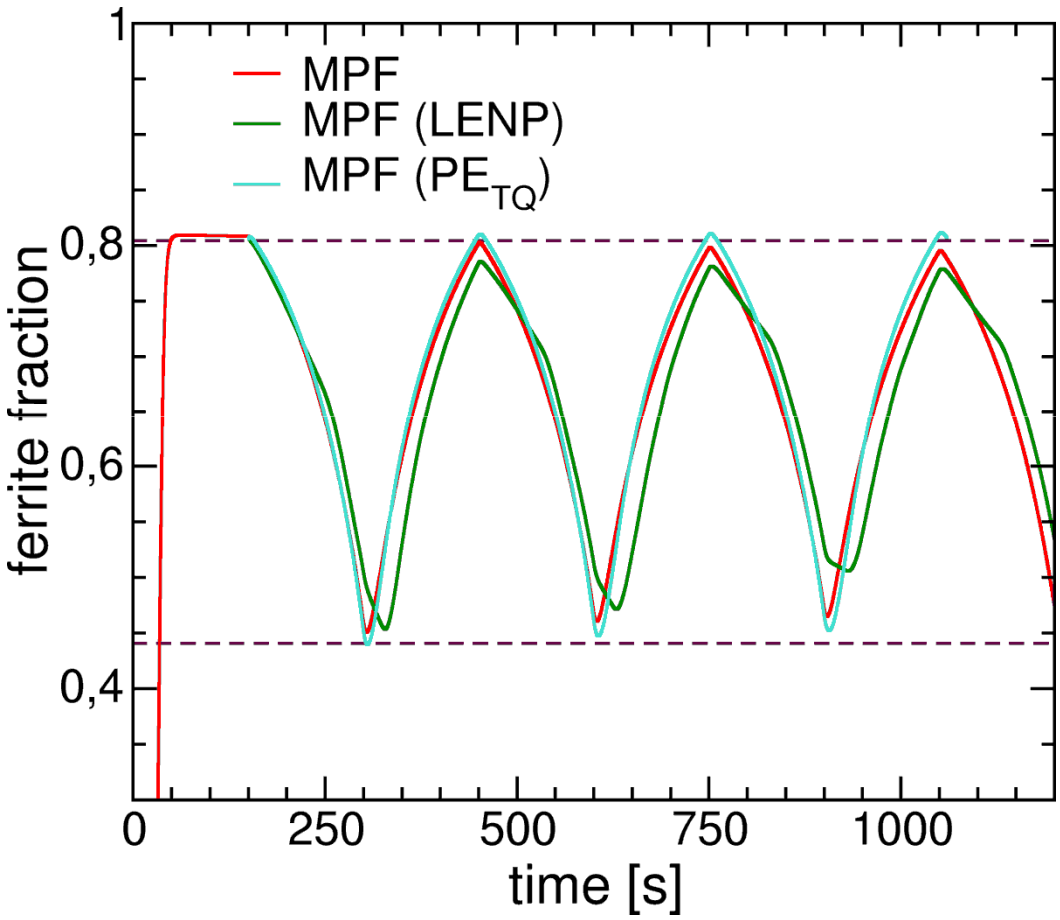
I-cycle: MPF model

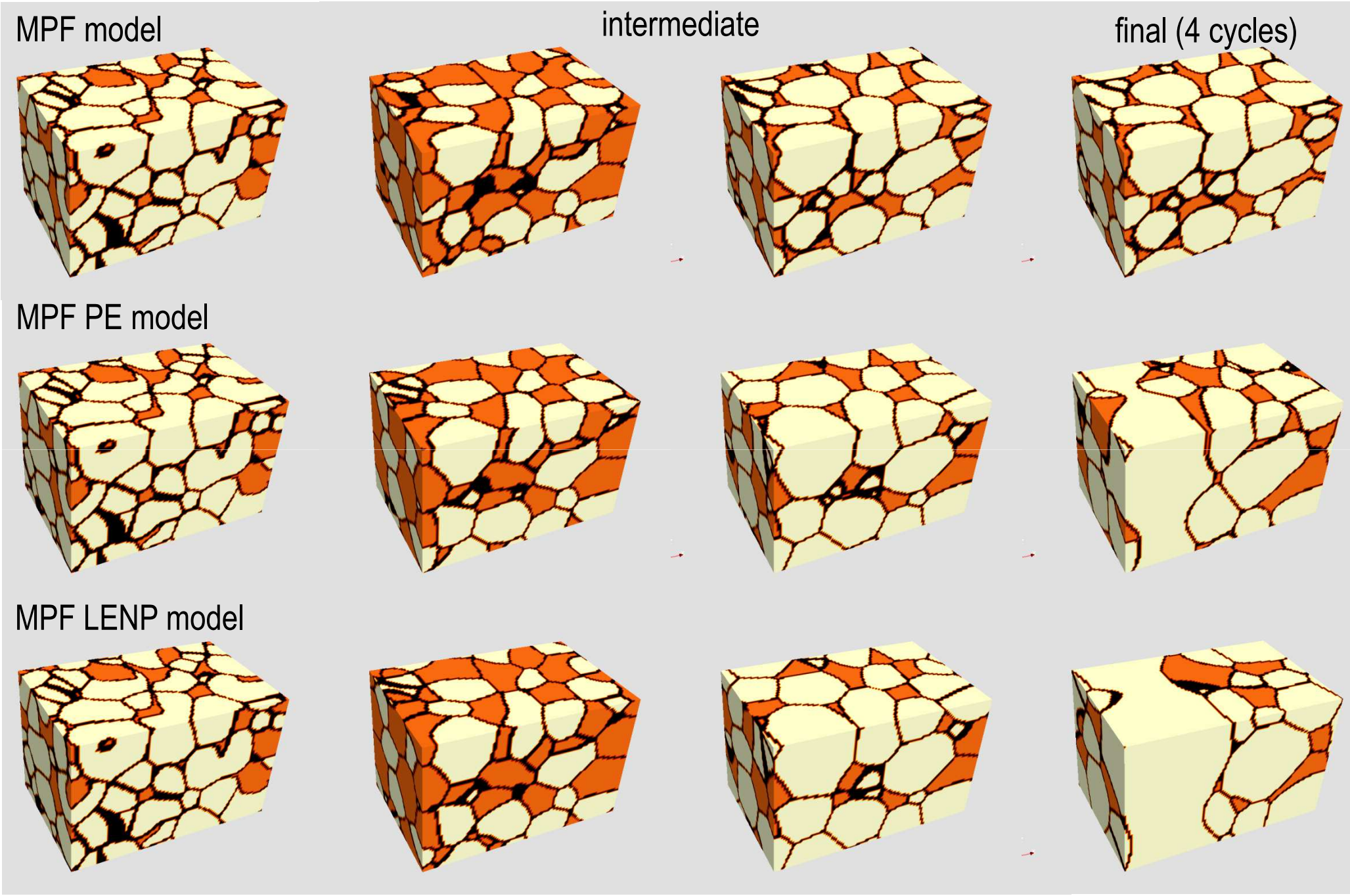


No stagnant stage ???

I-cycle: alternative driving force models

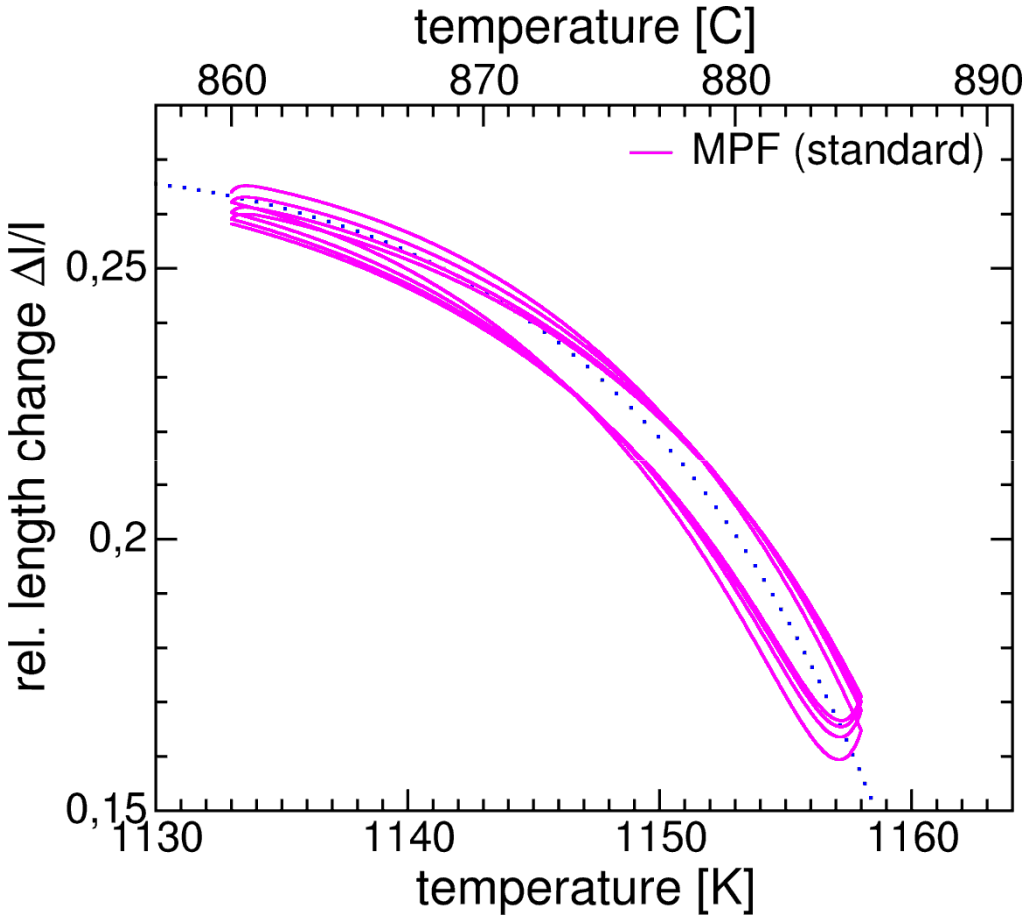
- MPF with LENP or Pe_{td} approximation




MPF model
intermediate
final (4 cycles)
MPF PE model
MPF LENP model

I-cycle: driving force model MPF (standard)

- 3D simulation: length change computed from phase fraction



I-cycle: driving force model MPF (standard + LENP)

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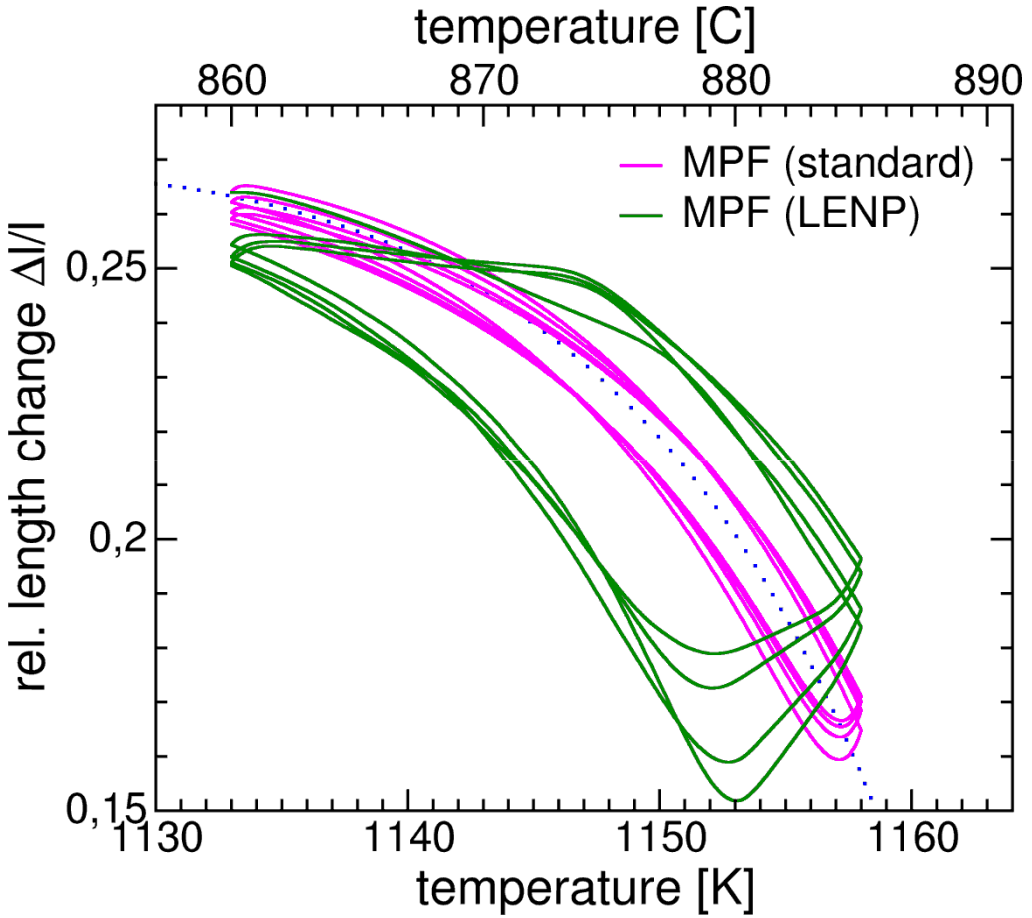
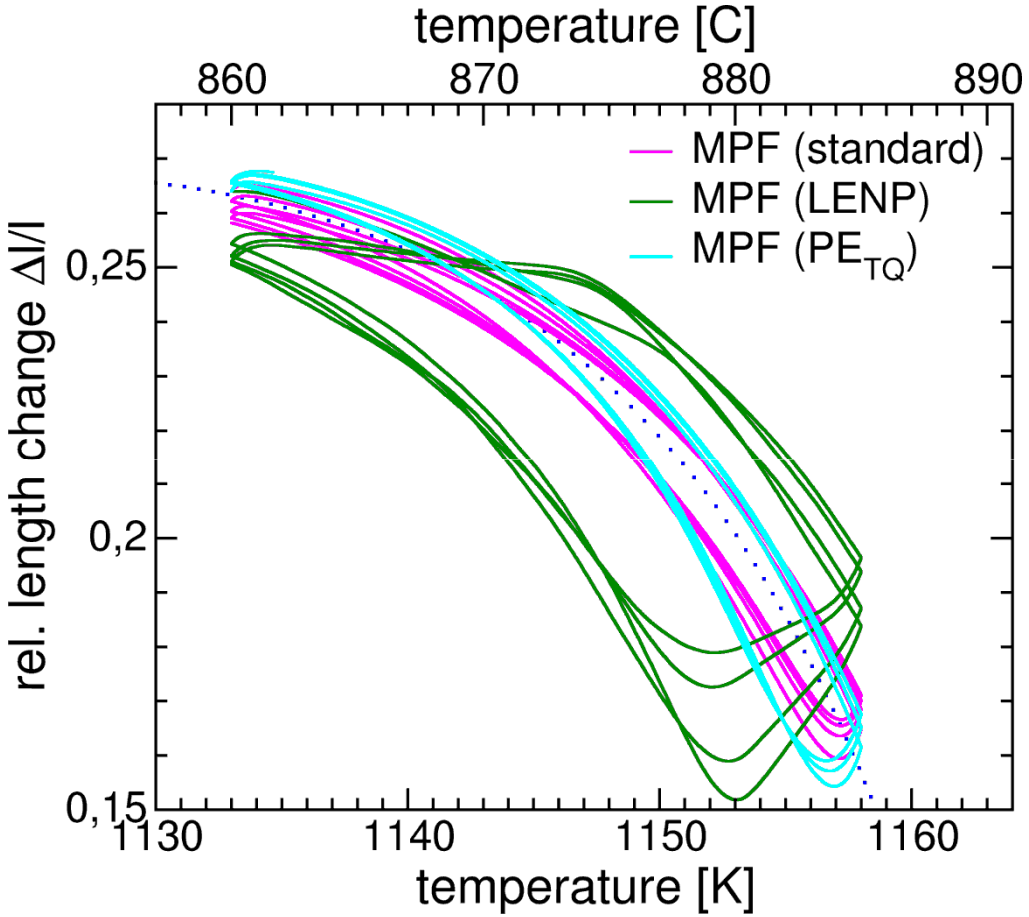


I-cycle: driving force model MPF (standard + LENP + PE)

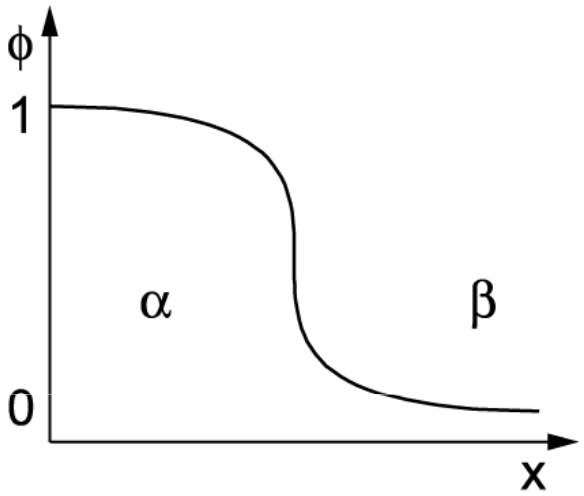
- 3D simulation: length change computed from phase fraction



➡ standard MPF and PE show similar behavior

Element composition in the diffuse interface

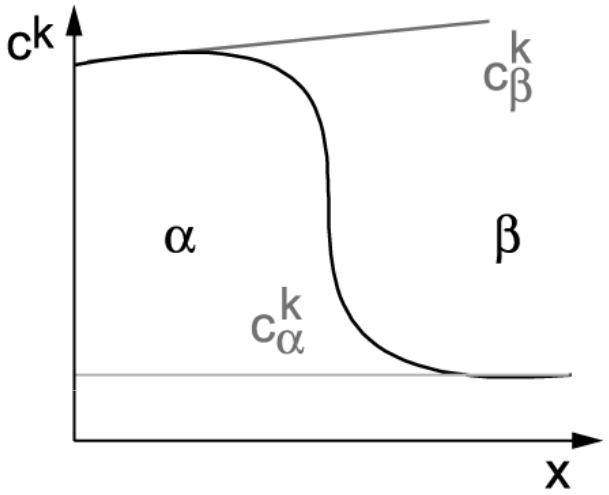
Wheeler-Boettinger-McFadden type models



Continuous concentration

natural choice when η is on the atomistic scale

Multiphase field models



split into partial concentrations

allows to separate bulk and interfacial properties and thus mesoscopic simulations

$$c^i = \sum_\alpha c^i_\alpha \phi_\alpha$$

$$f = \sum_\alpha \phi_\alpha f_\alpha(c_\alpha)$$

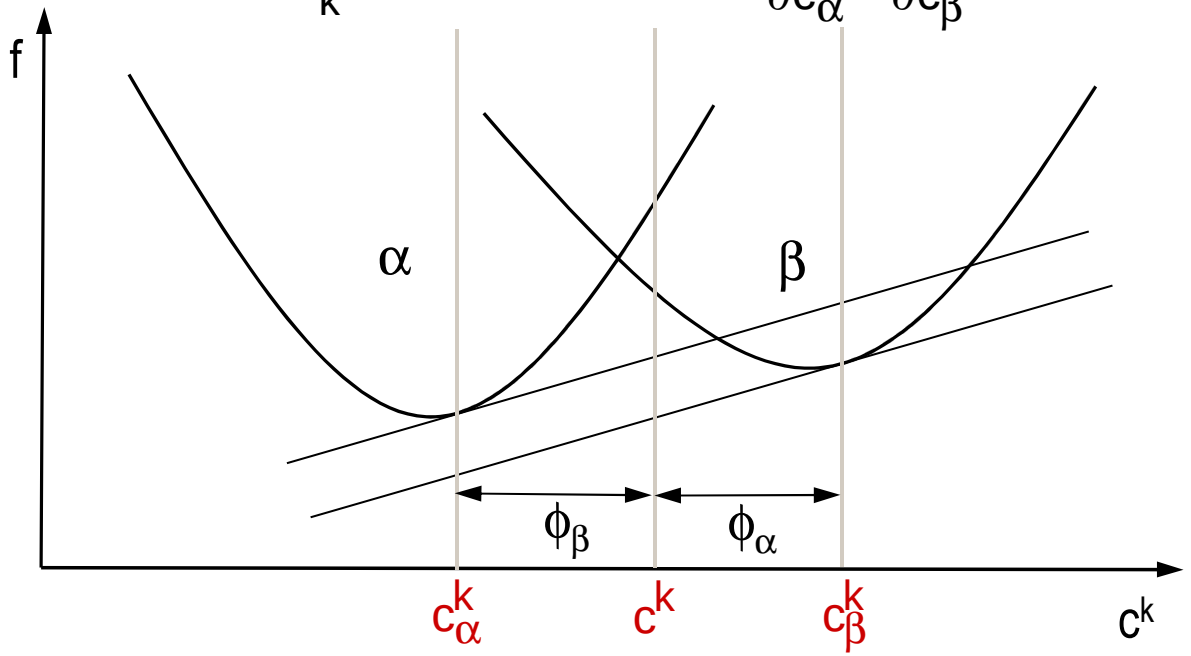
➡ diffuse interface region considered as “mixture” of bulk phases

➡ addition degree of freedom needs to be fixed

Phase-field and thermodynamics: quasi-equilibrium

- minimization of free energy $F = \sum_{\alpha} \phi_\alpha f_\alpha$ at constant phase fractions ϕ_α and total composition c gives:

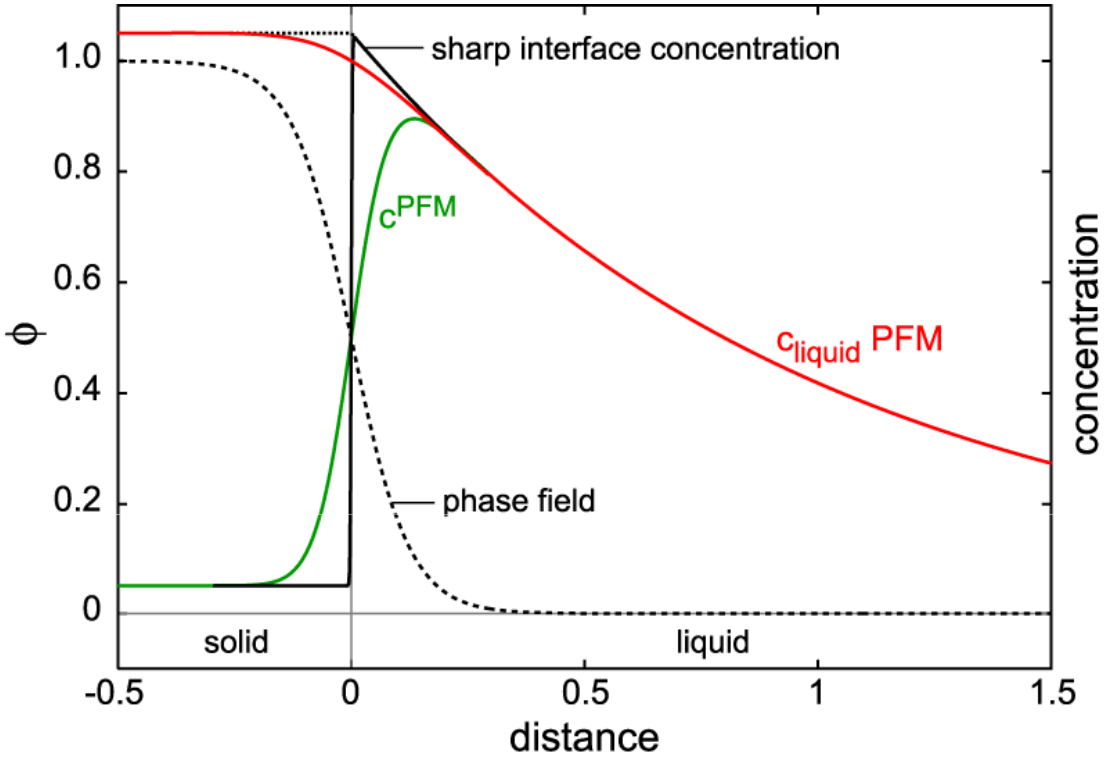
driving force $\Delta G_{\alpha\beta} = f_\alpha - f_\beta + \sum_k \tilde{\mu}^k \left(c_\alpha^k - c_\beta^k \right)$, $\tilde{\mu}^k = \frac{\partial f_\alpha}{\partial c_\alpha^k} = \frac{\partial f_\beta}{\partial c_\beta^k}$



and partitioning c_α^k, c_β^k,

calculated iteratively including the mass balance: $\phi_\alpha c_\alpha^k + \phi_\beta c_\beta^k = c^k$

Diffuse interface „artifacts“



processes scaling with interface width

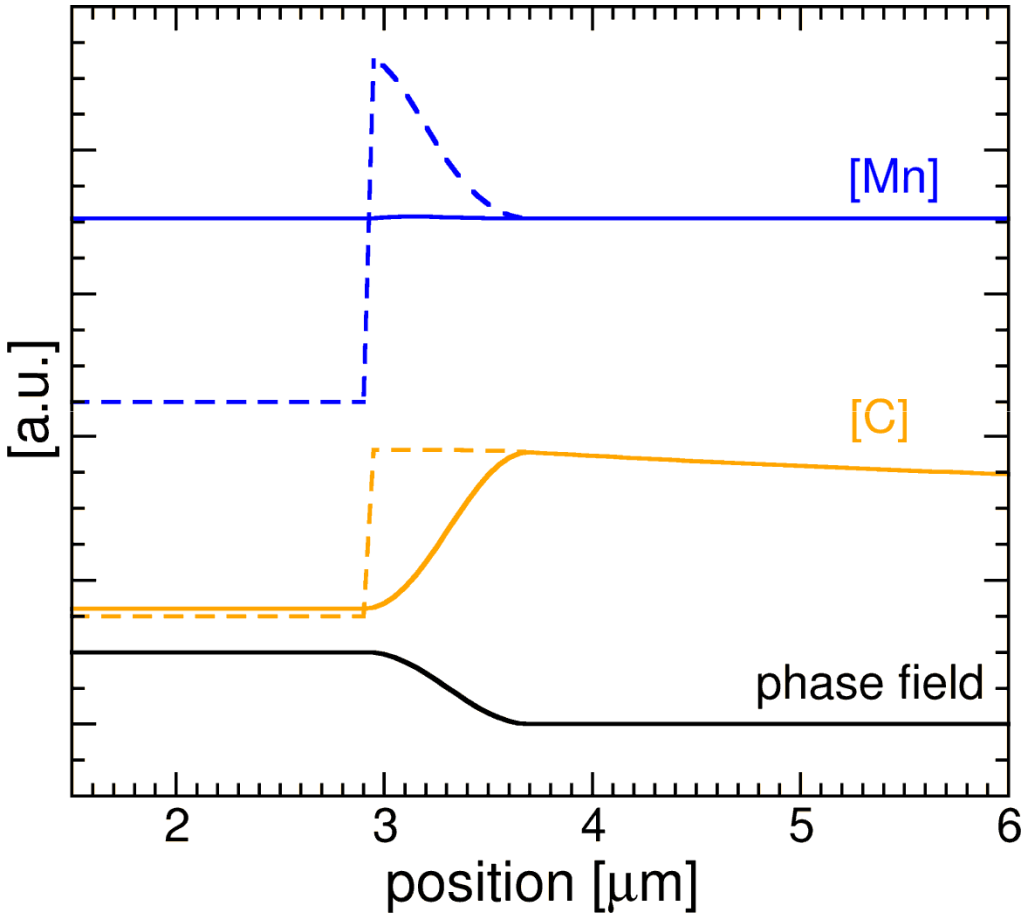
- interface kinetics
- solute trapping
- surface diffusion
- surface stretching

measures against unwanted artifacts:
thin interface limit, anti trapping current, …
(e.g. A. Karma, Phys Rev. Lett 87, 115701 (2001))

physical interface ~ d_0[nm]
numerical width in p.f. model ~ η [µm]
condition for “quantitative” phase field: η << diffusion length l_D
OK for C, but necessarily violated for substitiutional elements like Mn

Element profiles within the diffuse interface: standard MPF

- Due to the small diffusion length (“spike”) the solutal pile up is completely within the diffuse interface



- „solute trapping“ leads to a (partial) overrunning of the pile-up
- interface kinetics / driving force depends on the solute distribution within the interface
- calibration of the interface mobility would avoid the problem

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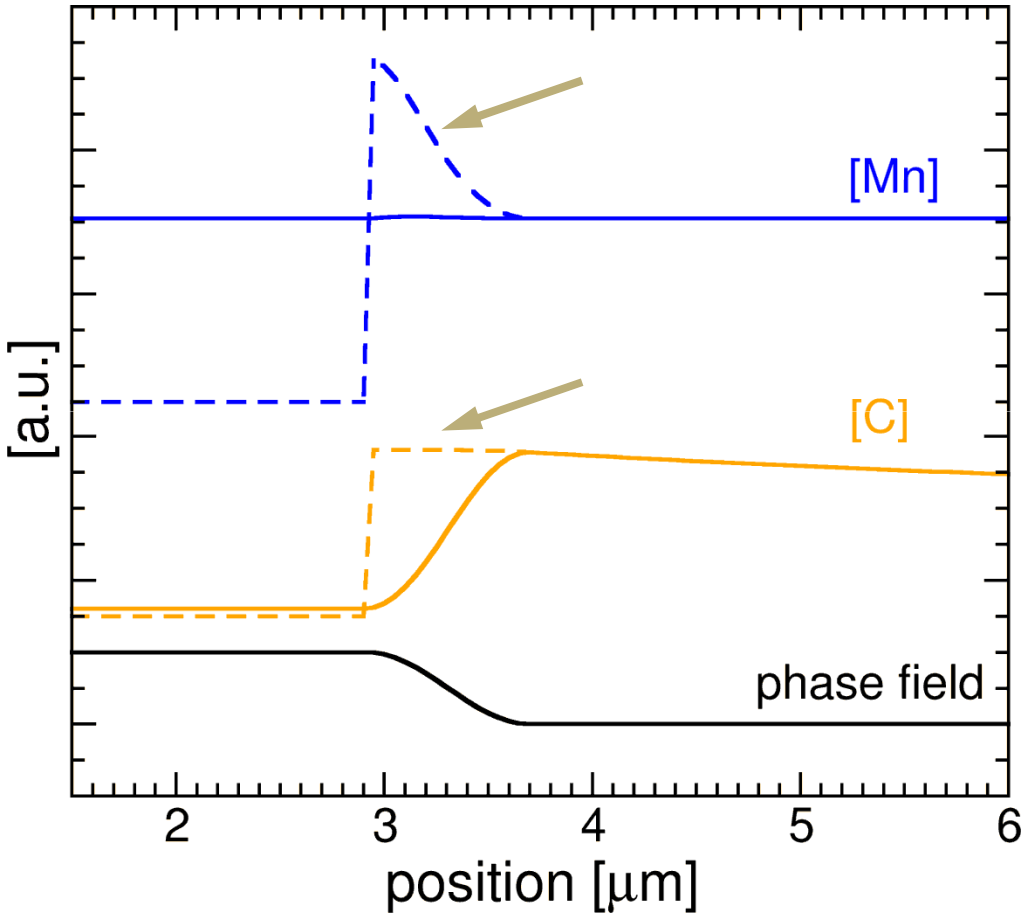

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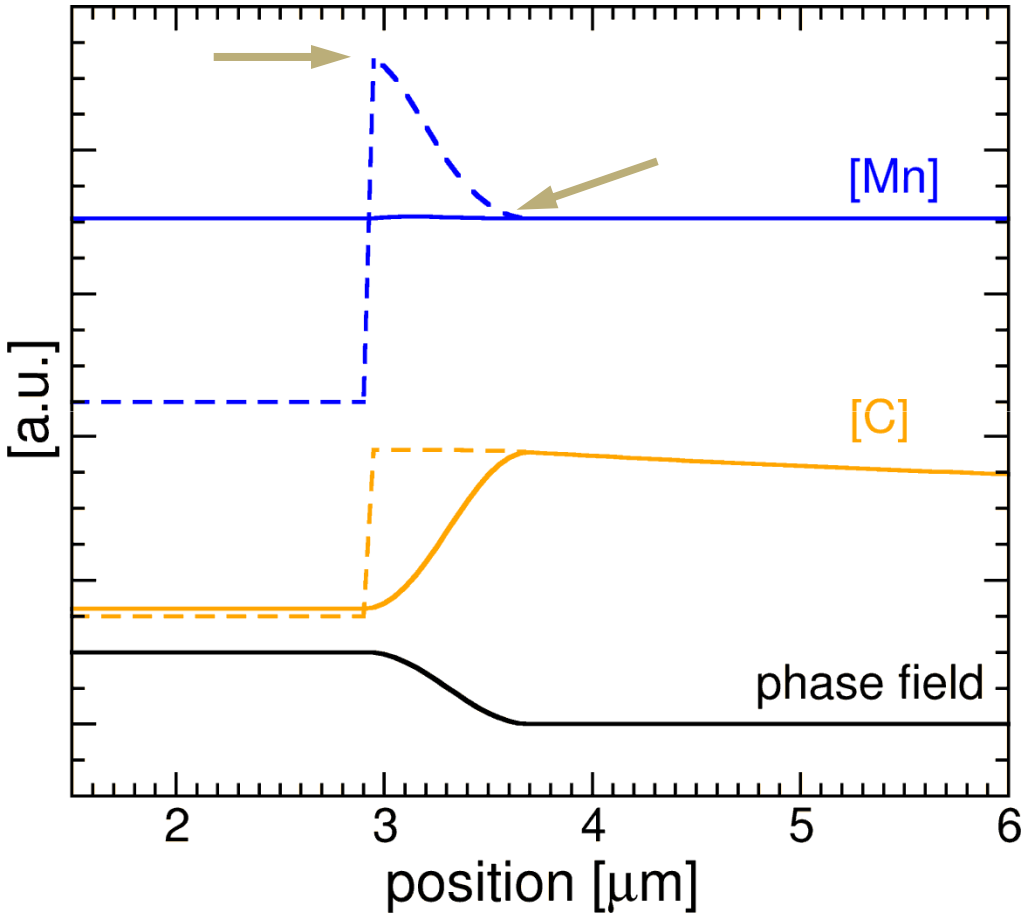

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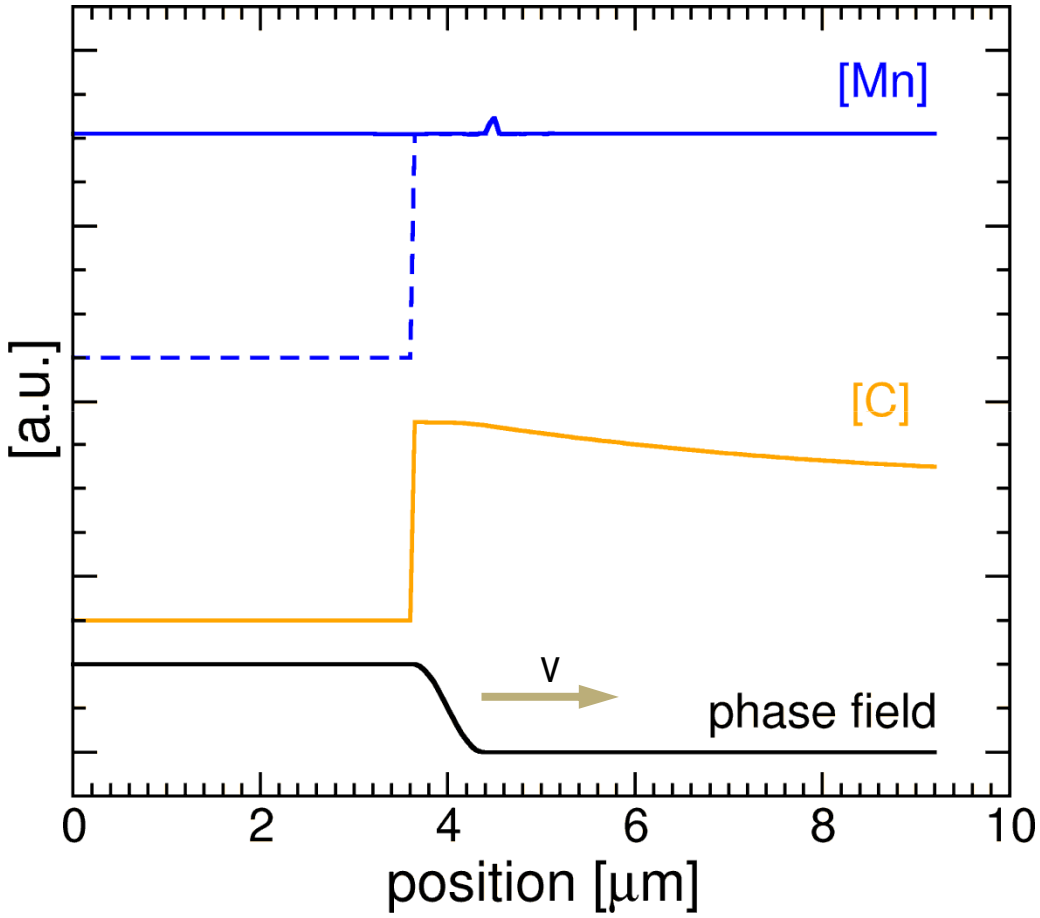
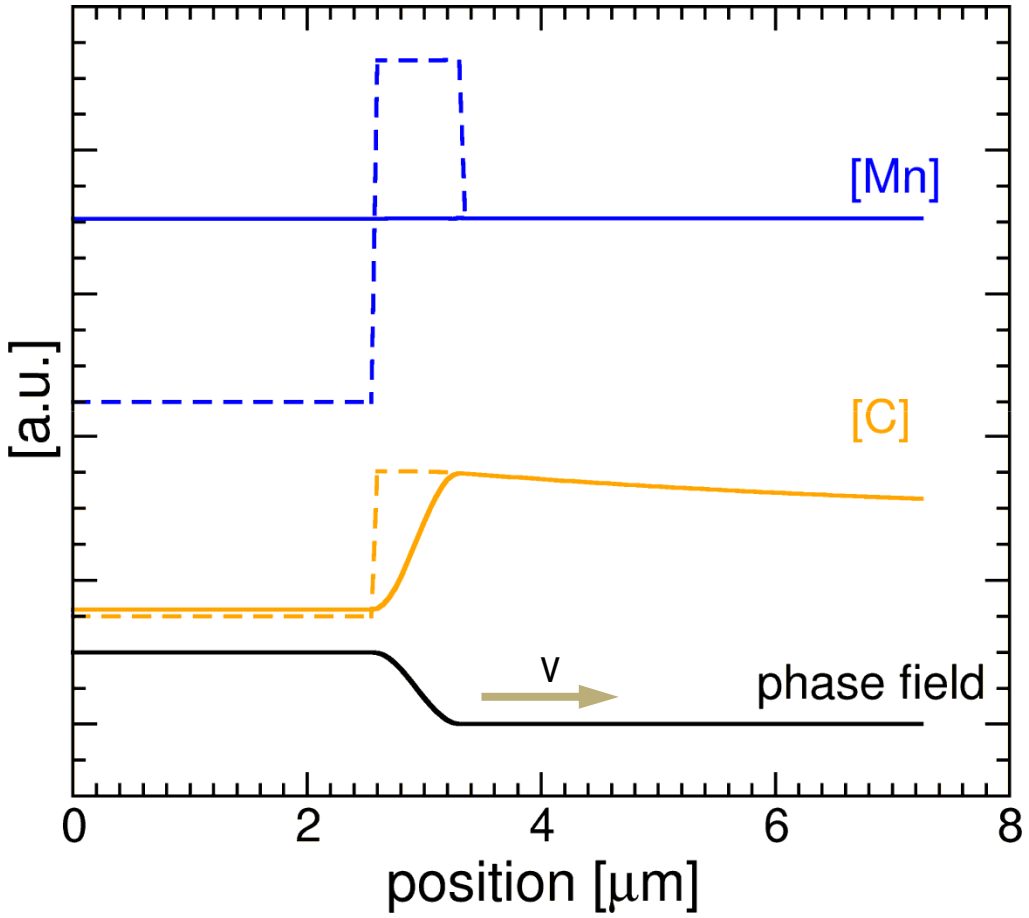
Element profiles within the diffuse interface: PE approximation

- composition of substitutional element taken from the “upstream” side

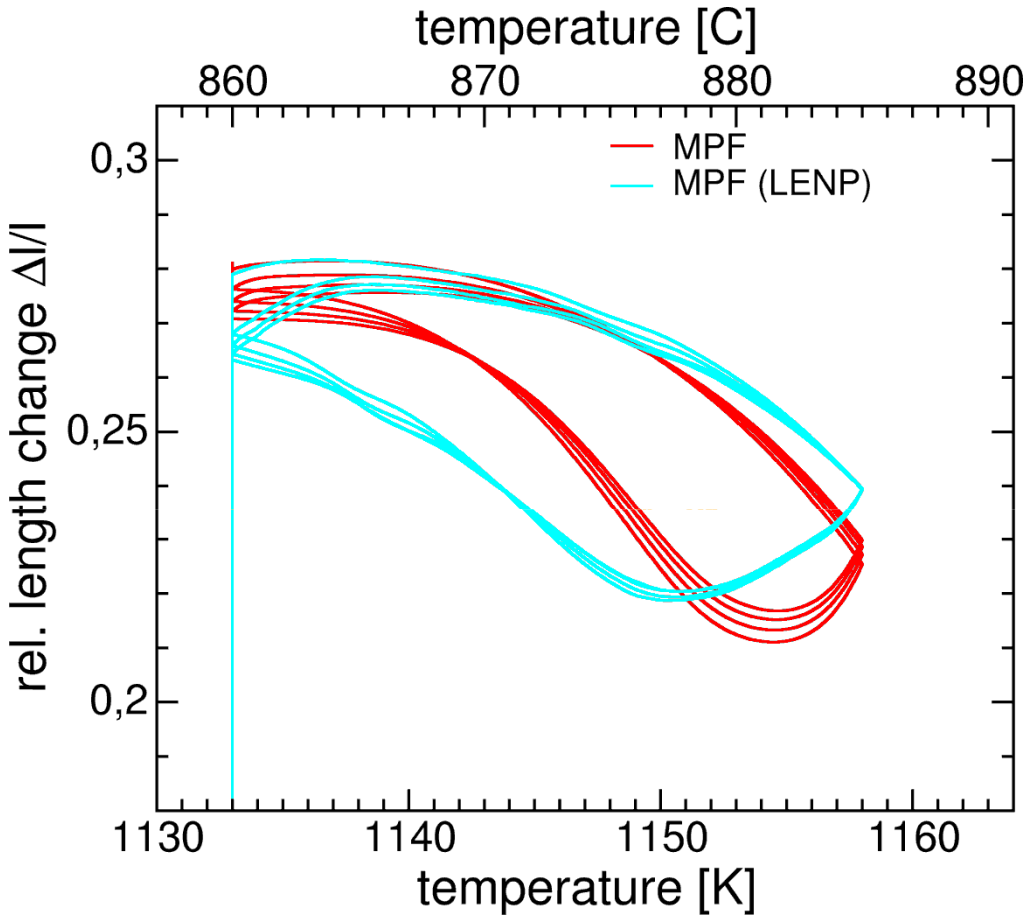


Element profiles within the diffuse interface: PE approximation

- composition of substitutional element taken from the “downstream” side

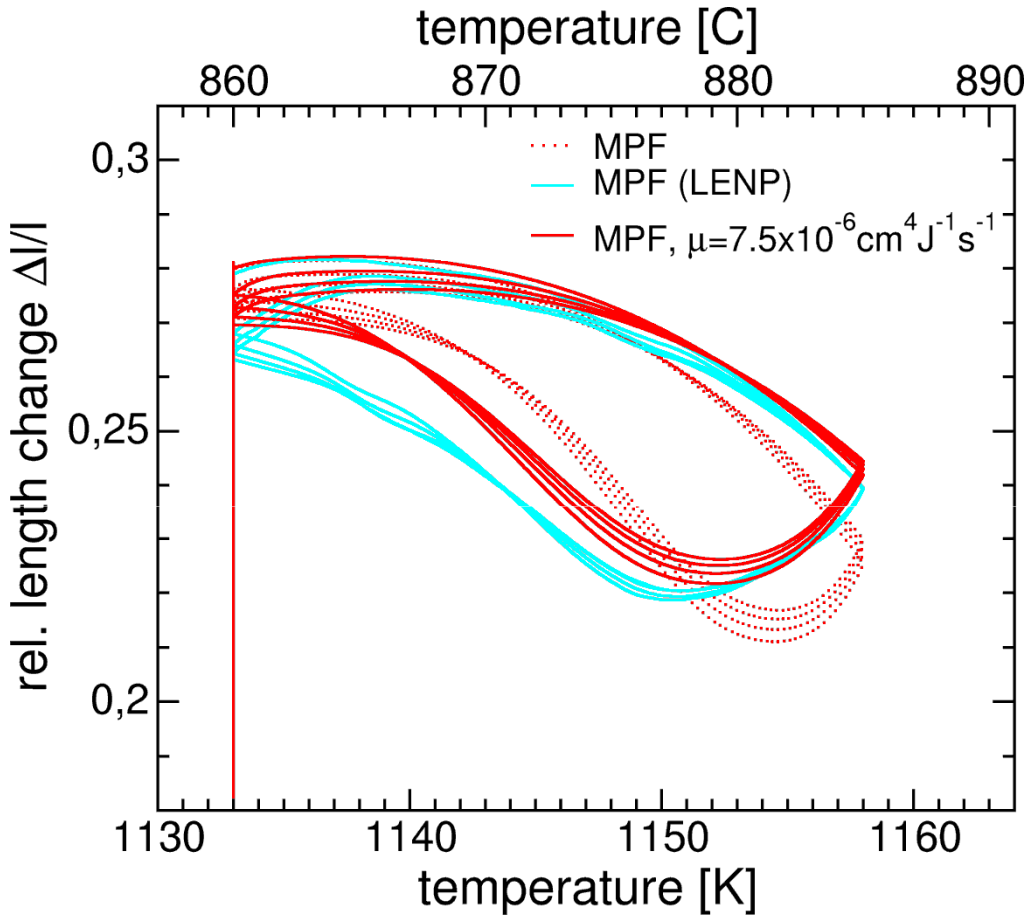


1D simulation: model comparison



- Can we exclude standard MPF (and PE-models) from the absence of the stagnant stage?

1D simulation: model comparison



- Maybe not if the interface mobility is significantly reduced (mixed mode)

  (A detailed investigation of the shape may allow a decision)

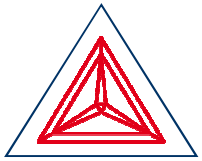
Conclusion

- Transformation according standard MPF predictions behaves similar to PE
- The stagnant stage can be observed for a MPF-LENP model
- Standard MPF can show a stagnant stage for mixed mode conditions
- Cycles show „aging“ cause by a Mn „wall“

Quantitative comparison betwee sharp interface, phase field and experiments should be the next step

Acknowledgment

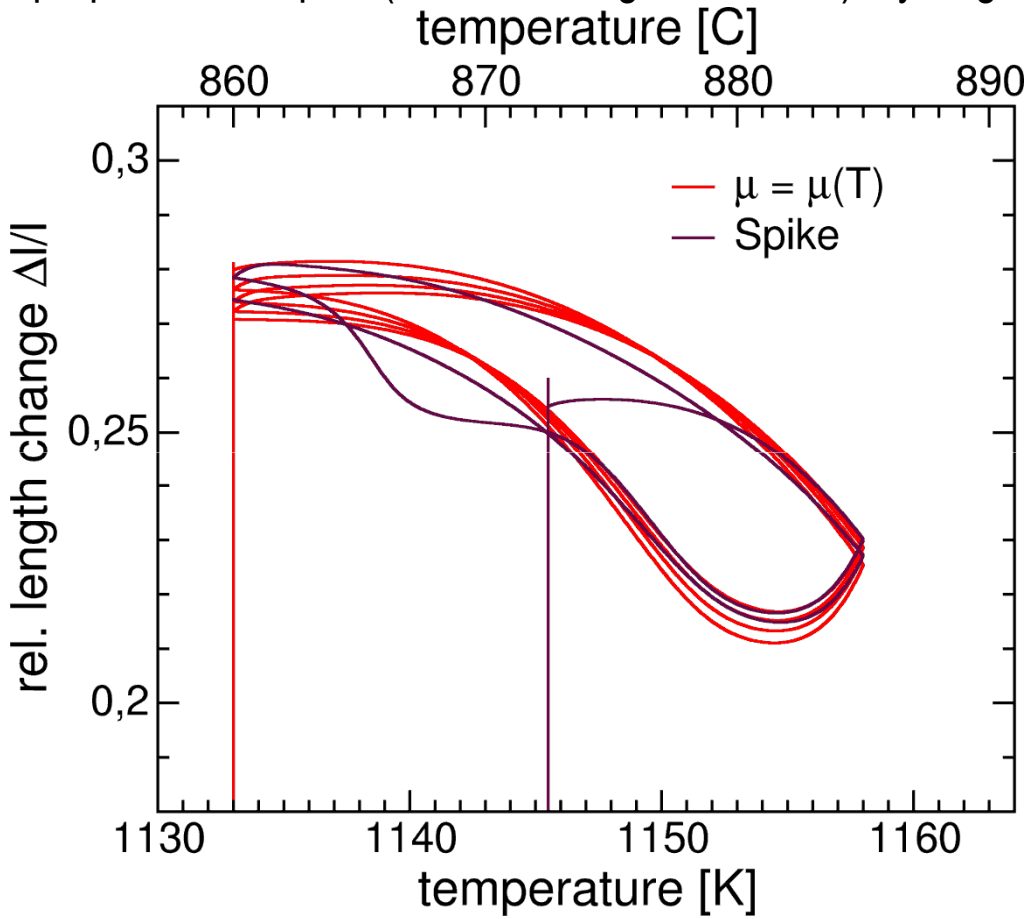
German Research Foundation (DFG)
Financial support in the framework of the Cluster of Excellence
"Integrative Production Technology for High-Wage Countries“

thermodynamic computations by  www.thermocalc.com

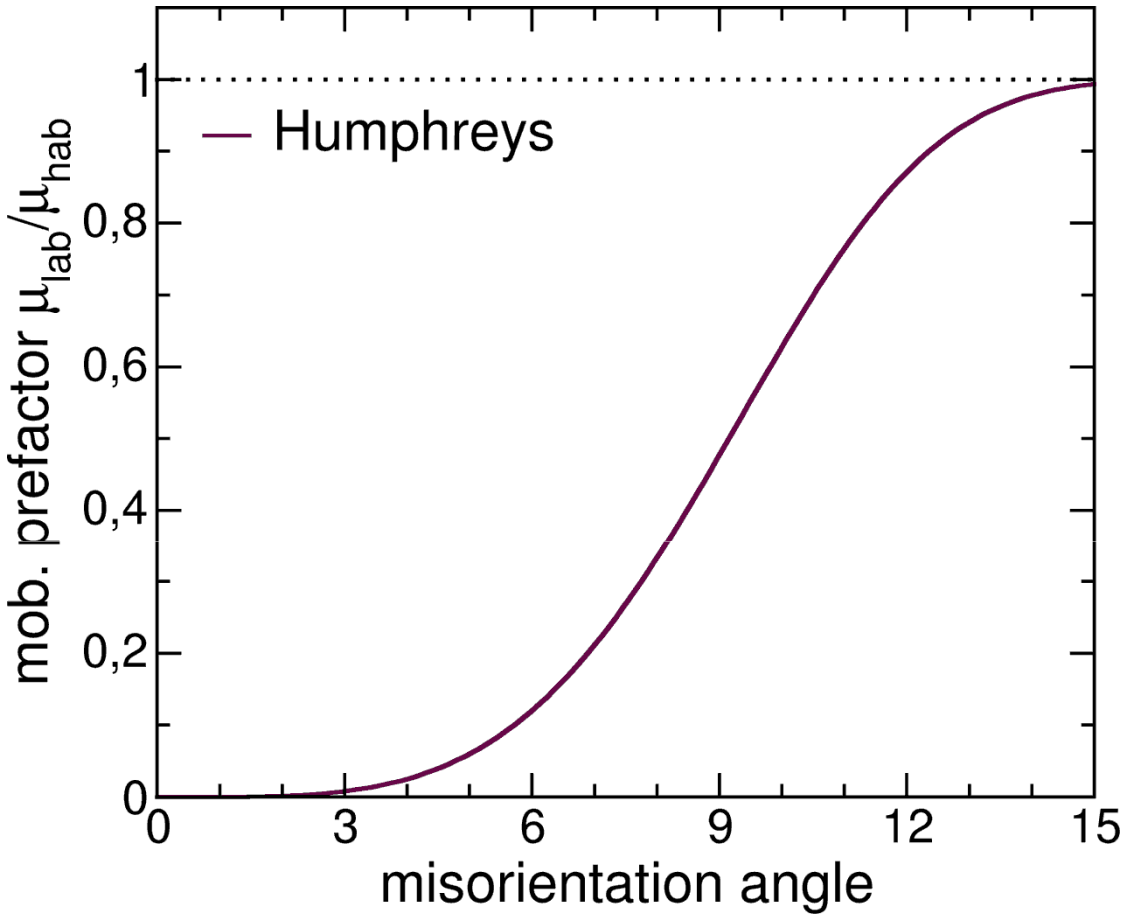
phase field simulations using  V6.1, www.micress.de

The Mn spike

- Virtual experiment: prepare a Mn spike (10min holding at 872.5°C), cycling between 860°C and 885°C.



Interface mobility (temperature dependent)



- $\mu(\gamma/\gamma)$ set identical to $\mu(\gamma/\alpha)$

[#] γ/α: Mecozzi et al., Comp. Mat. Sci. 50 (2011) p.1846
[##] α/α: Rudnitzki et al

Coupling to solute diffusion: The multiphase-field model

$$c^i = \sum_\alpha c^i_\alpha \, \phi_\alpha$$

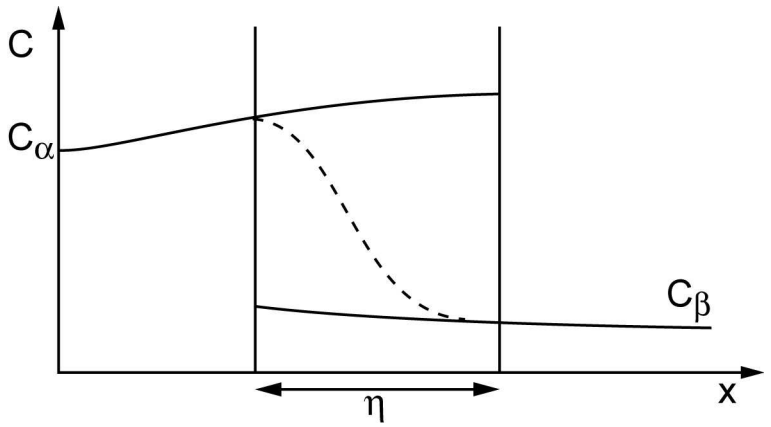
$$f = \sum_\alpha \phi_\alpha \, f_\alpha(c_\alpha)$$

$$\dot{c}^i = \sum_{k=1}^{m} \nabla M^{ik} \nabla \frac{\delta f}{\delta c^i}$$

$$\vdots$$

$$= \sum_{k=1}^{m} \nabla D^{ik} \left[\nabla c^k + \sum_{\alpha=1}^{n} c^k_\alpha \nabla \phi_\alpha \right]$$

bulk free energies $f_\alpha(c^k_\alpha)$ from thermodynamic databases



$$\frac{\delta f_\alpha}{\delta c^k_\alpha} = \frac{\delta f_\beta}{\delta c^k_\beta}$$

$$\text{or } c^k_\alpha = \text{const}$$

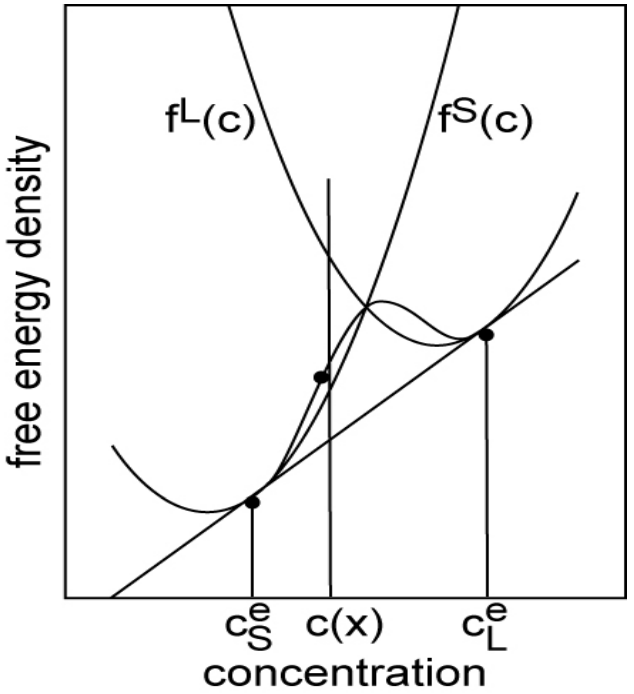
for all phases β, α and components k in stoichiometric phases

Wheeler-Boettinger-McFadden type models

$$\tilde{f}(\phi,c) = \frac{\varepsilon}{2}|\nabla\phi|^2 + (cW_A + (1-c)W_B)\phi^2(1-\phi^2)$$
$$+ (c\beta_A + (1-c)\beta_B)\phi^2(3-2\phi)$$
$$+ c\,ln\,c + (1-c)\,ln(1-c)$$

$$W_A, W_B \sim \frac{\sigma}{\eta}$$

$$\beta_A,\ \beta_B \sim \Delta G \cdot \eta$$



- **Energetic contribution of the interface scales with its (numerical) thickness**

  ➡ **spurious effects for simulations on mesoscopic length scale**

- **Problems in handling stoichometric phases, free energy for the interface diverges**

Multiphase field model: PE approximation

- Phase field, LENP, PE_{ff}, PE_{el}