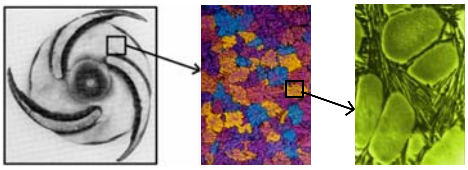
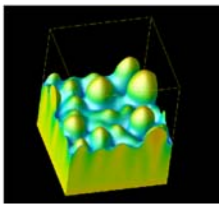
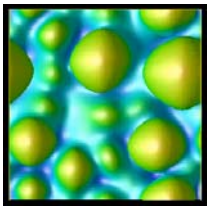
Part 2. Modelling micostructure formation and phase transitions – an introduction how to start with a simple phase-field model







Coworkers at ICE

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Content:

A solid-liquid phase-field model: Case study

Ginzburg-Landau entropy density functional

$$\mathcal{S}(\phi) = \int_\Omega s(\phi) - \left(\epsilon a(\nabla\phi) + \frac{1}{\epsilon} w(\phi)\right) dx$$

Entropy density contributions:

$s = -f_{,T}$ — bulk entropy density

with $f(\phi) = \tilde{\eta}\, h(\phi) = \tilde{\eta}\, \phi^2(3 - 2\phi)$ — free energy density

$w(\phi) = 9\gamma\phi^2(1-\phi)^2$ — potential entropy density

$a(\nabla\phi) = \gamma|\nabla\phi|^2$ — gradient entropy density

Variational derivative of the entropy functional

$$\epsilon\partial_t\phi = \frac{\delta\mathcal{S}(\phi)}{\delta\phi} = \frac{\partial\mathcal{S}}{\partial\phi} - \nabla\cdot\frac{\partial\mathcal{S}}{\partial(\nabla\phi)}$$

⇒ Phase-field equation

$$\epsilon\partial_t\phi \quad = \quad \epsilon\nabla\cdot a_{,\nabla\phi}(\nabla\phi) - A\,\frac{1}{\epsilon}w_{,\phi}(\phi) - B\, f_{,\phi}(\phi)$$

to switch on/off the terms (A, B)

with derivatives

$$a_{,\nabla\phi}(\nabla\phi) = 2\gamma(\nabla\phi)$$

$$w_{,\phi}(\phi) = 18\gamma(2\phi^3 - 3\phi^2 + \phi)$$

$$f_{,\phi}(\phi) = 6\eta\phi(1-\phi)$$

⇒ Phase-field equation with specific choice of entropy densities

$$\epsilon\partial_t\phi \quad = \quad \underbrace{\epsilon\nabla\cdot\Big(2\gamma(\nabla\phi)\Big)}_{2\epsilon\gamma(\triangle\phi)} - A\,\frac{1}{\epsilon}18\gamma(2\phi^3-3\phi^2+\phi) - B\,6\eta\phi(1-\phi)$$

with e.g. natural boundary condition

$$\nabla\phi\cdot n_{\partial\Omega} = 0.$$

Discretization on a rectangular region in two dimensions

$$\Omega = [0,a]\times[0,b]\subset \mathbb{R}^2,$$

Cell spacing $\delta x = \dfrac{a}{Nx} \qquad \delta y = \dfrac{b}{Ny}.$

Grid points

$$x_{i,j} = (i\delta x, j\delta y), \quad i = 0,\dots Nx, \quad j = 0,\dots,Ny$$

Finite difference expression of the laplace operator: $\triangle\phi = \dfrac{\partial^2\phi}{\partial x^2}+\dfrac{\partial^2\phi}{\partial y^2}$

$$(\triangle\phi)_{i,j} = \frac{\phi_{i+1,j}-2\phi_{i,j}+\phi_{i-1,j}}{\delta x^2} + \frac{\phi_{i,j+1}-2\phi_{i,j}+\phi_{i,j-1}}{\delta y^2}$$

Euler's method for the time discretization

$$(\partial_t \phi)^{n+1} = \frac{\phi^{n+1} - \phi^n}{\delta t}$$

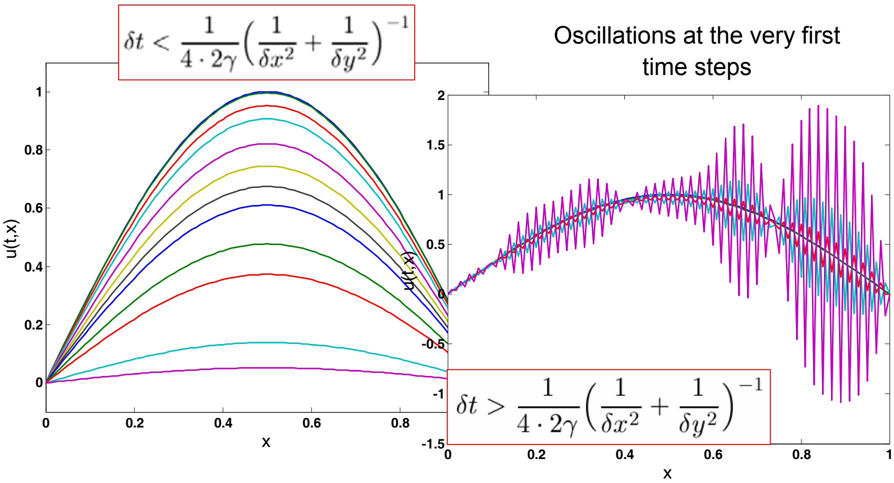
Explicit finite difference algorithm of the phase-field equation

$$\phi_{i,j}^{n+1} = \phi_{i,j}^n + \delta t \left\{ 2\gamma \left(\frac{\phi_{i+1,j}^n - 2\phi_{i,j}^n + \phi_{i-1,j}^n}{\delta x^2} + \frac{\phi_{i,j+1}^n - 2\phi_{i,j}^n + \phi_{i,j-1}^n}{\delta y^2} \right) - A\frac{1}{\epsilon^2} w_{,\phi}(\phi_{i,j}^n) - B\frac{1}{\epsilon} f_{,\phi}(\phi_{i,j}^n) \right\}$$

Stability condition $\delta t < \frac{1}{4 \cdot 2\gamma} \left(\frac{1}{\delta x^2} + \frac{1}{\delta y^2} \right)^{-1}$

Instability for the explicit difference method

Oscillations at the very first time steps



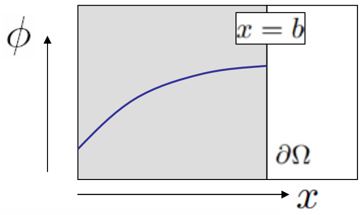
$$\delta t < \frac{1}{4 \cdot 2\gamma} \left(\frac{1}{\delta x^2} + \frac{1}{\delta y^2} \right)^{-1}$$

$$\delta t > \frac{1}{4 \cdot 2\gamma} \left(\frac{1}{\delta x^2} + \frac{1}{\delta y^2} \right)^{-1}$$

Boundary values for the discrete equation

(i) Neumann condition (isolation):

$$\phi_{0,j} = \phi_{1,j}, \quad \phi_{Nx,j} = \phi_{Nx-1,j}$$
$$\phi_{i,0} = \phi_{i,1}, \quad \phi_{i,Ny} = \phi_{i,Ny-1}$$



(ii) Periodic condition:

$$\phi_{0,j} = \phi_{Nx-1,j}, \quad \phi_{Nx,j} = \phi_{1,j}$$
$$\phi_{i,0} = \phi_{i,Ny-1}, \quad \phi_{i,Ny} = \phi_{i,1}$$

(iii) Dirichlet condition:

$$\phi_{0,j} = \phi_W, \quad \phi_{Nx,j} = \phi_E$$
$$\phi_{i,0} = \phi_S, \quad \phi_{i,Ny} = \phi_N$$

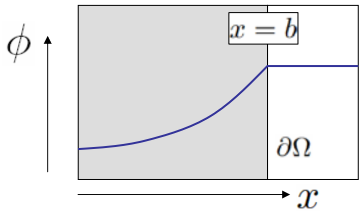
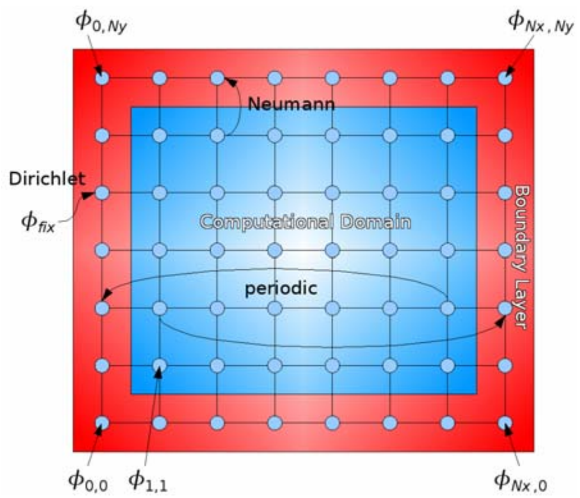


Illustration of boundary conditions



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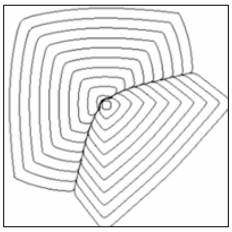
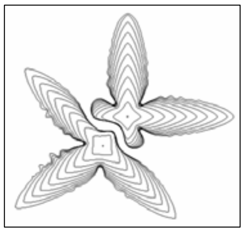
$$\mathcal{S}(e,\phi) = \int_\Omega \left(s(e,\phi) - \left(\varepsilon a(\nabla\phi) + \frac{1}{\varepsilon} w(\phi)\right)\right) dx$$

Anisotropic gradient entropy densities

$$a(\nabla\phi) = \gamma A(\nabla\phi)|\nabla\phi|^2$$





with e.g.

2D $A(\nabla\phi) = 1 + \delta\cos(4\theta)$

θ: angle between interface normal and some fixed direction

3D $A(\nabla\phi) = 1 - \delta_c\left(3 - 4\frac{1}{|\nabla\phi|^4}\sum_i\left(\frac{\partial\phi}{\partial x_i}\right)^4\right)$

[J. Bragard, A. Karma, Y. H. Lee, M. Plapp, Interface Science 10 (2-3) pp. 121-136 (2002)]

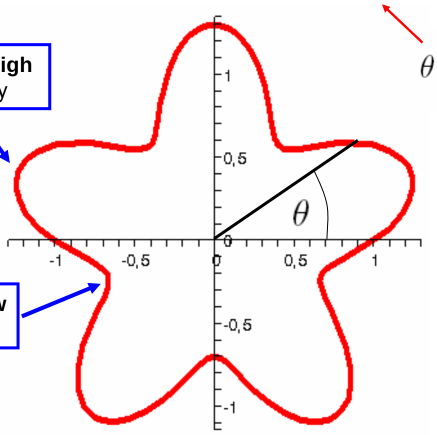
Anisotropic gradient entropy densities

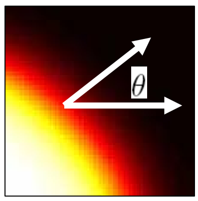
in 2D: $A(\nabla\phi) = 1 + 0.3\sin(5\theta)$

$$\theta = \arccos\left(\frac{\nabla\phi}{|\nabla\phi|}\cdot e_1\right)$$

directions of **high** surface energy

directions of **low** surface energy



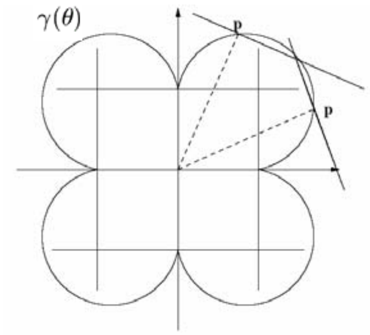


Equilibrium crystal shapes

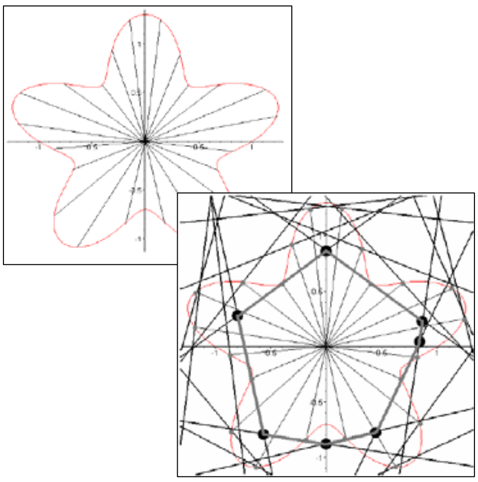
are determined by angular dependance of surface energy $\rightarrow$ $\gamma(\theta)$ - plot

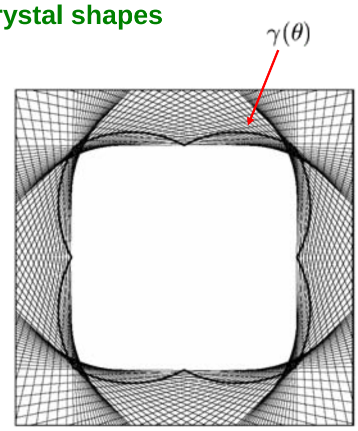
Wulff construction:

drop perpendiculars to all radius vectors on the curve $\gamma(\theta)$. The inner envelope of them gives the crystal shape.

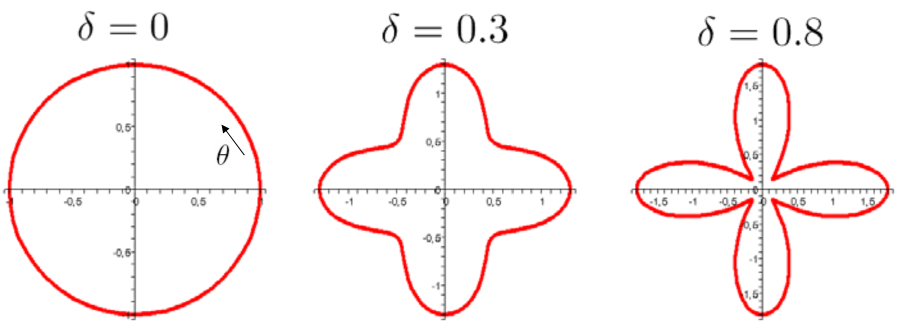


Wulff Construction of crystal shapes



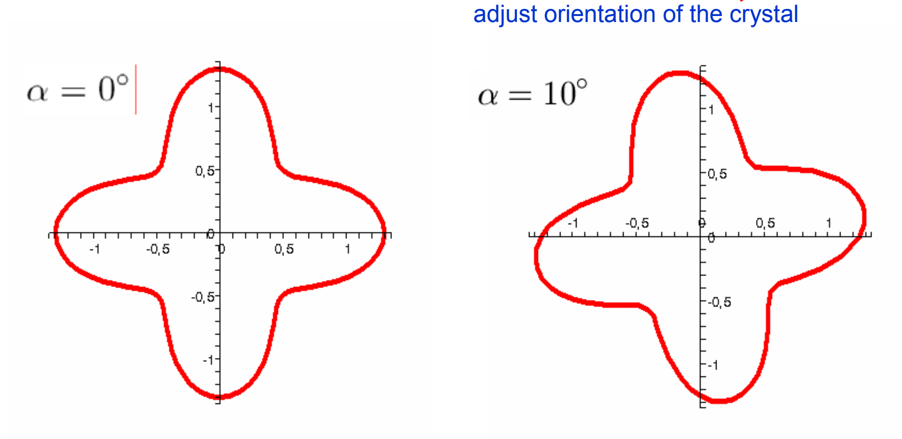


Strength of the anisotropy $A(\nabla\phi) = \quad 1 + \delta\cos(4\theta)$



Orientation of the crystal $A(\nabla\phi) = 1 + \delta cos(4(\theta - \alpha))$

adjust orientation of the crystal



Crystal shapes in 3D, multi-phase

examples for anisotropy function $A(\vec{q}) = A(\phi_\alpha \nabla \phi_\beta - \phi_\beta \nabla \phi_\alpha)$

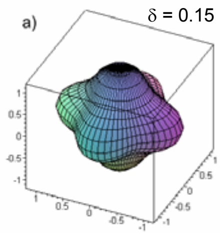
a) cubic (smooth)

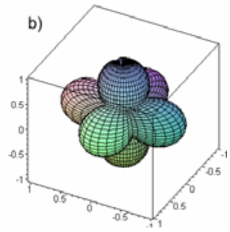
$$A(\vec{q}) = 1 - 3\delta + 4\delta \sum_{i=1}^{3} (\frac{q_i}{|q|})^4$$

b) cubic (facetted)

$$A(\vec{q}) = \left(\max \{ \frac{\vec{q}}{|q|} \cdot \vec{\eta_k},\ k = 1, \ldots, n \} \right)^2$$

η^k: k^{th} vector in the Wulff diagram





Anisotropic gradient entropy densities

$a(\nabla\phi) = \gamma A(\nabla\phi)|\nabla\phi|^2$ instead of $a(\nabla\phi) = \gamma|\nabla\phi|^2$

it is more convenient to discretize the divergence

$\nabla \cdot \frac{\partial S}{\partial(\nabla\phi)} = \nabla \cdot (a_{,\nabla\phi}(\nabla\phi))$ by $\nabla^l \cdot (a_{,\nabla\phi}(\nabla^r\phi))$

backward and forward differences

Forward differences

$$(\nabla^r\phi)_{i,j} = \left(\frac{\phi_{i+1,j} - \phi_{i,j}}{\Delta x}, \frac{\phi_{i,j+1} - \phi_{i,j}}{\Delta y} \right)$$

The discretization of the divergence gives

$$\left(\nabla^l \cdot (a_{,\nabla\phi}(\nabla^r\phi)\right)_{i,j} = \frac{(a_{,\nabla\phi}(\nabla^r\phi))_{i,j} - (a_{,\nabla\phi}(\nabla^r\phi))_{i-1,j}}{\Delta x} + \frac{(a_{,\nabla\phi}(\nabla^r\phi))_{i,j} - (a_{,\nabla\phi}(\nabla^r\phi))_{i,j-1}}{\Delta y}$$

Isotropic case

$$A(\nabla\phi) = 1 \quad \Rightarrow \quad a(\nabla\phi) = \gamma|\nabla\phi|^2$$

it is easy to show that $(\triangle\phi)_{i,j} = (\nabla^l \cdot (\nabla^r\phi))_{i,j}$

Energy equation (pure substance solidification)

Complex patterns arise when transport of conserved quantity (energy, solute) is limited by diffusion.

$$\frac{\partial e}{\partial t} = -\nabla \cdot \left\{ L_{00}(T,\phi)\nabla \cdot \frac{\delta\mathcal{S}}{\delta e} \right\} \quad (*)$$
$$\tau\varepsilon\frac{\partial\phi}{\partial t} = \frac{\delta\mathcal{S}}{\delta\phi}$$

mobility coefficient related to heat conductivity $\kappa(\phi) = \kappa$: $L_{00} = \kappa T^2$

with thermodynamic relation $e = f + Ts \Rightarrow \delta\mathcal{S}/\delta e = T^{-1}$

and introduce thermal diffusivity $k = \kappa/c_v$

and adiabatic temperature $T_Q = L/c_v$

Temperature equation (pure substance solidification)

We put all this into the energy equation (*) and use the Ansatz for e:

$$e = -L\,h(\phi) + c_v T \quad \text{(entropy of ordering + heat)}$$

to change to the temperature equation

$$\frac{\partial T}{\partial t} = k\,\nabla^2 T + T_Q\,h'(\phi)\,\frac{\partial \phi}{\partial t}$$

standard Laplacian discretisation

use the result from the recent phase-field update

use Euler scheme with new stability condition for both eqns.

$$\delta t < min\left\{ \frac{1}{4 \cdot 2\gamma}\left(\frac{1}{\delta x^2} + \frac{1}{\delta y^2}\right)^{-1}, \quad \frac{1}{4k}\left(\frac{1}{\delta x^2} + \frac{1}{\delta y^2}\right)^{-1} \right\}$$

and update synchronously with phase-field eqn.

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Phase-field equation for pure substance solidification

still necessary: use the correct temperature dependant term for the driving force in the PF eqn.

$$\tau\varepsilon \frac{\partial \phi}{\partial t} = \frac{\delta S}{\delta \phi}$$

with thermodynamic relation $e = f + Ts$ and

$$f(\phi, T) = L\frac{T - T_M}{T_M}\,h(\phi) - c_v\,T\,ln\left(\frac{T}{T_M}\right)$$

$$\Rightarrow \quad \varepsilon\tau\frac{\partial \phi}{\partial t} = \varepsilon\,\nabla \cdot a_{,\nabla\phi}(\nabla\phi) - \frac{1}{\varepsilon}w_{,\phi}(\phi) - \frac{f_{,\phi}}{T}$$

→ dendrites ...

frank.wendler@hs-karlsruhe.de ▪ Marie Curie Summer School 2007 ▪ Estremoz ▪ August 20th - August 29th

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Content:

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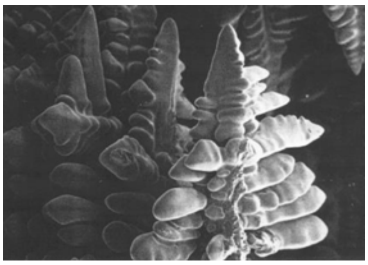
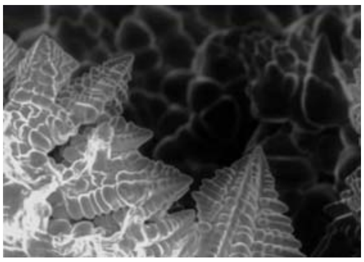
Directional solidification – Experiments

Microstructure images of alloy surfaces



Co-Cr-alloy

Labo de Micro-Analyses des Surfaces, Besançon



Ti-alloy

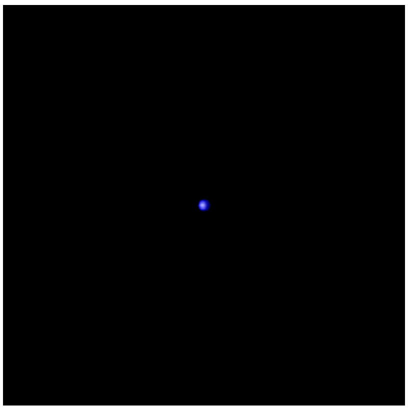
Iowa State University, Dept. of Mat. Science and Engineering

frank.wendler@hs-karlsruhe.de ▪ Marie Curie Summer School 2007 ▪ Estremoz ▪ August 20th - August 29th

Simulations of dendritic solidification in pure Nickel

Parameter	Symbol	Dimension	Ni - Data
Melting temperature	T_M	K	1728
Latent heat	L	J/m^3	8.113×10^9
Specific heat	c_v	$J/(m^3K)$	1.939×10^7
Thermal diffusivity	k	m^2/s	1.2×10^{-5}
Interfacial free energy	σ_0	J/m^2	0.326
Strength of interfacial energy	δ_c	—	0.018
Growth kinetics in $< 100 >$ −crystallographic direction	μ_{100}	$m/(sK)$	0.52
Growth kinetics in $< 110 >$ −crystallographic direction	μ_{110}	$m/(sK)$	0.40

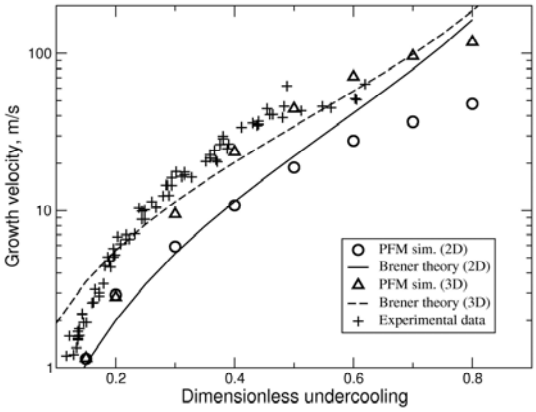
Simulation of equiaxial dendritic growth of pure Nickel



Undercooling: ΔT =130 K
Grid: 400 × 400 × 400 (6 μm)

Comparison of with experiments and analytical predictions in pure Nickel

together with D. Herlach, P. Galenko, DLR Köln
and Efim Brener, FZ Jülich



grid 400 × 400 × 400 (6 μm)

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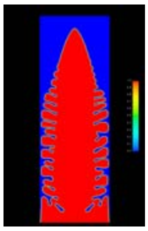
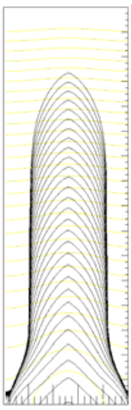
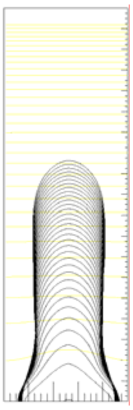
Dendritic arrays: Effect of undercooling

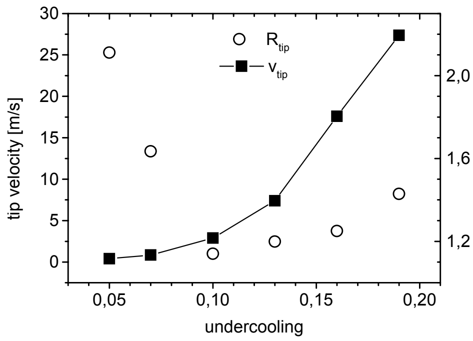


T=0.87: Sharp tip
V_{tip} =7.38 m/s,
R_{tip} = 1.20 μm

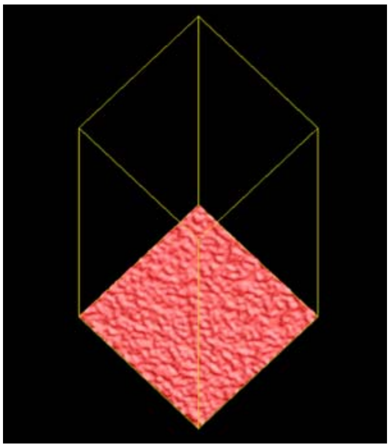


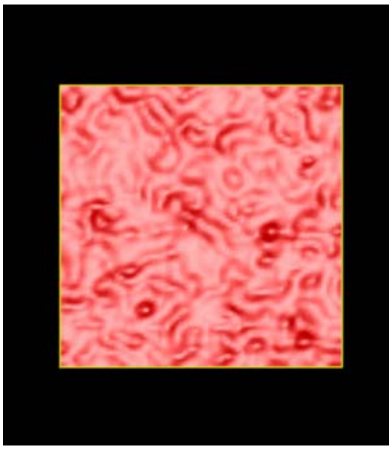
T=0.93: parabola
V_{tip} =0.85 m/s,
R_{tip} = 1.63 μm





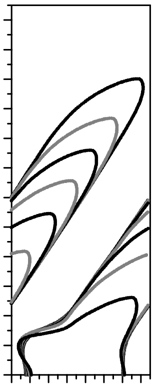
Ni - planar front, 15° off [001] normal, T = 0.85, grid 80 × 80 × 300

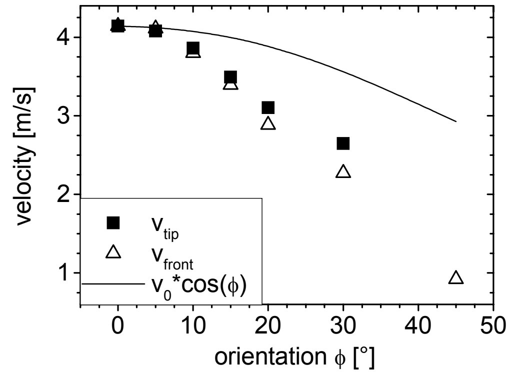




Dendritic arrays: orientation dependance

rotated 30°

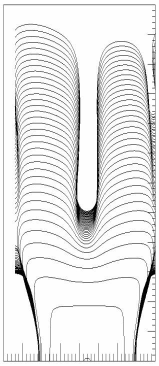




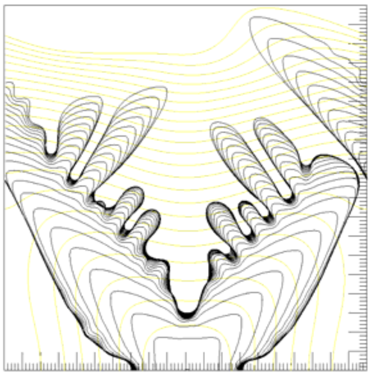
Ni Dendritic arrays: 45° orientation

At small interdendritic distances, a transition to cellular growth occurs

grid 75×250



grid 250×250



Content:

Dendritic growth in Ni-Cu

Data set:

Melting temperatures:
T_m^{Ni}=1728 K T_m^{Cu}=1358 K

Latent heats:

L^{Ni} =2.350·10^9 J/m^3

L^{Cu} =1.728·10^9 J/m^3

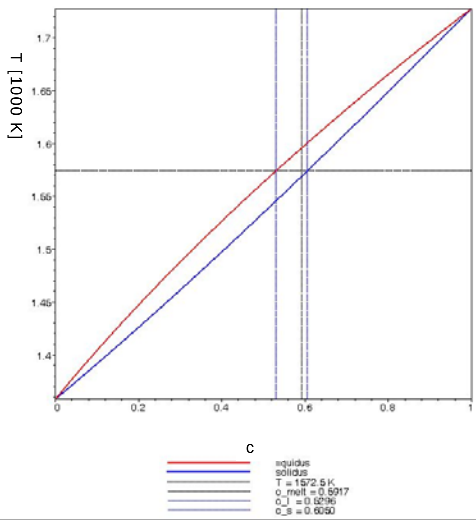
Surface tension: γ = 0.37 J/m^2

Surface anisotropy: δ = 0.04

Diffusion coefficients:
D_L = 10^{-9} m^2/sK D_S = 10^{-13} m^2/sK

Mobility μ = 3.3 ·10^{-3} m/sK

Initial condition:
c^{Ni}=0.5917 c^{Cu}=0.4083



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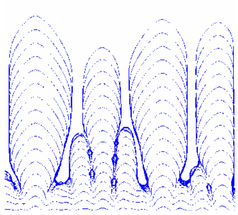
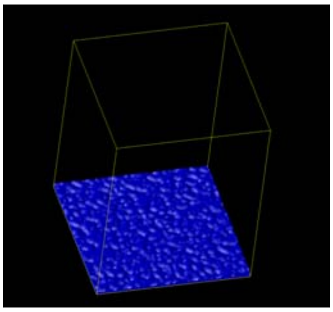
Ni/Cu cellular growth (movies)

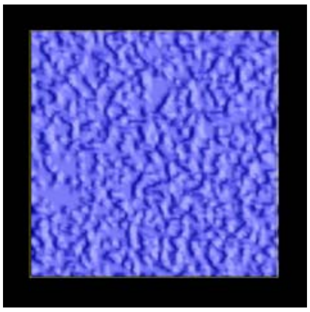
$Ni_{0.5917}Cu_{0.4083}$
Initial condition: rough surface,
undercooling ΔT = 20 K

2D simulation
L = 13.5 μm



Phase-field isosurface with mapped Ni conc. (10×10×12 μm, moving box)





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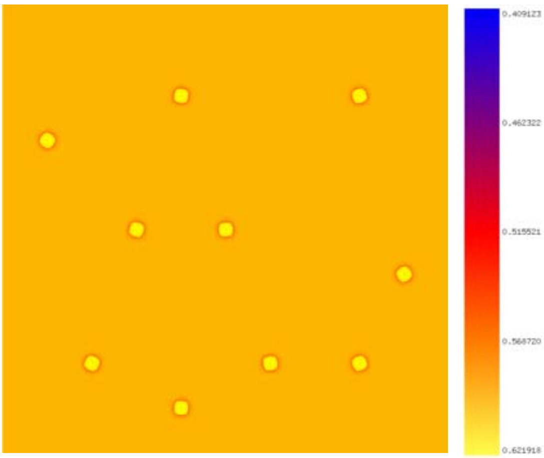
Simulations: Ni/Cu – evolution of polycrystals (2D)



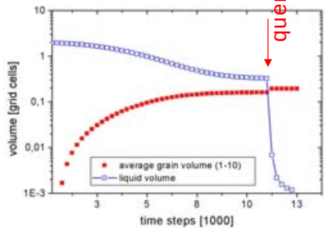
grid: 800×800 (160 μm)

10 orientations,

10 grains

S = 0.8, $c_{Ni,melt}$=0.592

grain volume:



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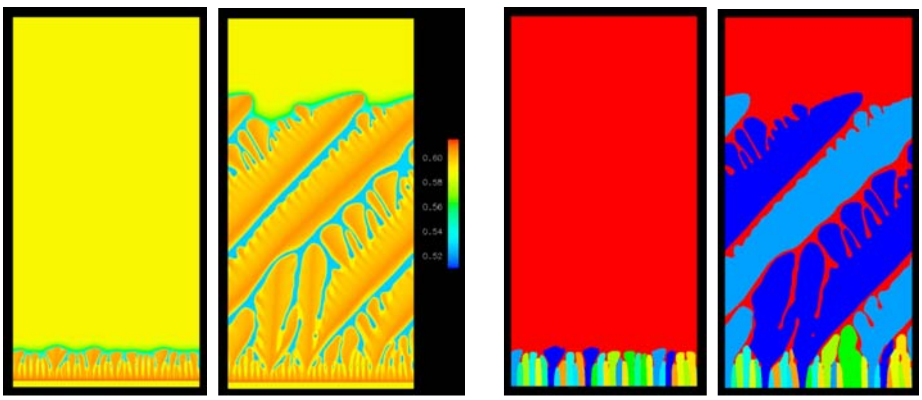
Directional solidification in Ni/Cu: Selection of orientations

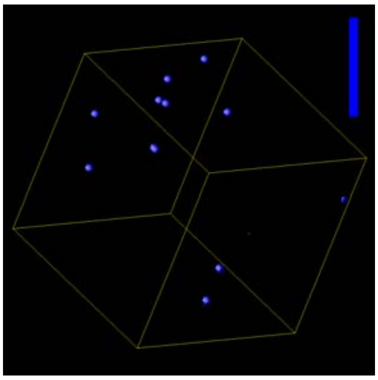


concentration field orientations

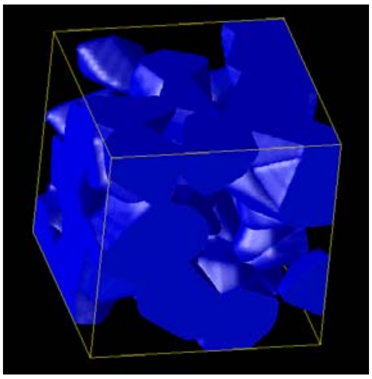
Simulation of growth and melting of polycrystals (3D)

approx. 100 Ni-Cu grains, Ni concentration is shown

Anisotropic growth

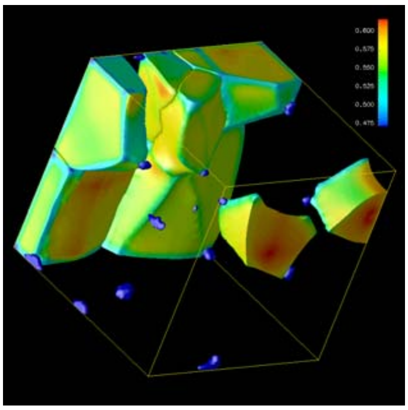


Melting (15 K superheated)



Simulations: Ni/Cu – annealing of polycrystals (3D)

Annealing and partial melting along grain boundaries



Content:

Phase-field model with fluid flow

Substitution: Total time derivative

$$\partial/\partial t \Longrightarrow D/Dt = \partial/\partial t + \vec{u}\cdot\nabla$$

General stress tensor $\boldsymbol{m} = \boldsymbol{\sigma} + \boldsymbol{\theta_{cap}}$

irreversible viscous stress + reversible stress tensor

Evolution equations with fluid flow:

$$\frac{De}{Dt} = -\nabla\cdot\left(L_{00}\nabla\frac{1}{T} + \sum_{j=1}^{K} L_{0j}\nabla\left(\frac{-\mu_j}{T}\right)\right) + \boldsymbol{m} : \nabla\vec{u}$$

$$\frac{Dc_i}{Dt} = -\nabla\cdot\left(L_{i0}\nabla\frac{1}{T} + \sum_{j=1}^{K} L_{ij}\nabla\left(\frac{-\mu_j}{T}\right)\right)$$

$$\omega\varepsilon\frac{D\phi_\alpha}{Dt} = \varepsilon\nabla\cdot\left(\rho\, a_{,\nabla\phi_\alpha}(\phi,\nabla\phi) - \rho\, a_{,\phi_\alpha}\right) - \frac{\rho}{\varepsilon} w_{,\phi_\alpha}(\phi) - \rho\frac{f_{,\phi_\alpha}}{T} - \lambda$$

Navier-Stokes Equations

$$\rho \frac{D\vec{u}}{Dt} = \nabla \cdot (\boldsymbol{\sigma} + \boldsymbol{\theta_{cap}}) = \nabla \cdot \Big[-p\boldsymbol{I} + \mu(\phi)\big(\nabla\vec{u} + (\nabla\vec{u})^T\big) + \boldsymbol{\theta_{cap}} \Big]$$

$\nabla \cdot \vec{u} = 0$ continuity equation

Viscosity

$$\mu(\phi) = \sum_{\alpha=1}^{N} \mu_\alpha \phi_\alpha \quad \text{(arithmetic)} \qquad \frac{1}{\mu(\phi)} = \sum_{\alpha=1}^{N} \frac{\phi_\alpha}{\mu_\alpha} \quad \text{(harmonic)}$$

Tensor representing capillary forces in the interfacial region

$$\boldsymbol{\theta_{cap}} = \left[a(\phi, \nabla\phi)\boldsymbol{I} - \sum_{\alpha=1}^{N} \left(\frac{\partial a(\phi, \nabla\phi)}{\partial(\nabla\phi_\alpha)} \otimes \nabla\phi_\alpha \right) \right]$$

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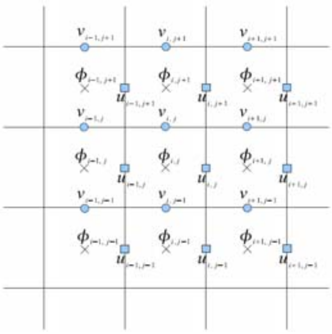
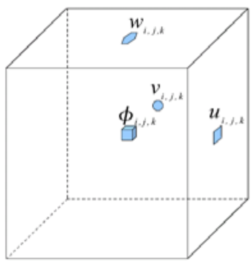
Discretization on a staggered grid with SOR method for the continuity equation





Stability condition for the time step

$$\delta t := \tau \, min \left(\frac{Re}{2\mu(\phi_\alpha)_{max}} \left(\frac{1}{\delta x^2} + \frac{1}{\delta y^2} + \frac{1}{\delta z^2} \right)^{-1}, \frac{\delta x}{|u_{max}|}, \frac{\delta y}{|v_{max}|}, \frac{\delta z}{|w_{max}|} \right) \quad \text{with} \quad \tau \in]0, 1]$$

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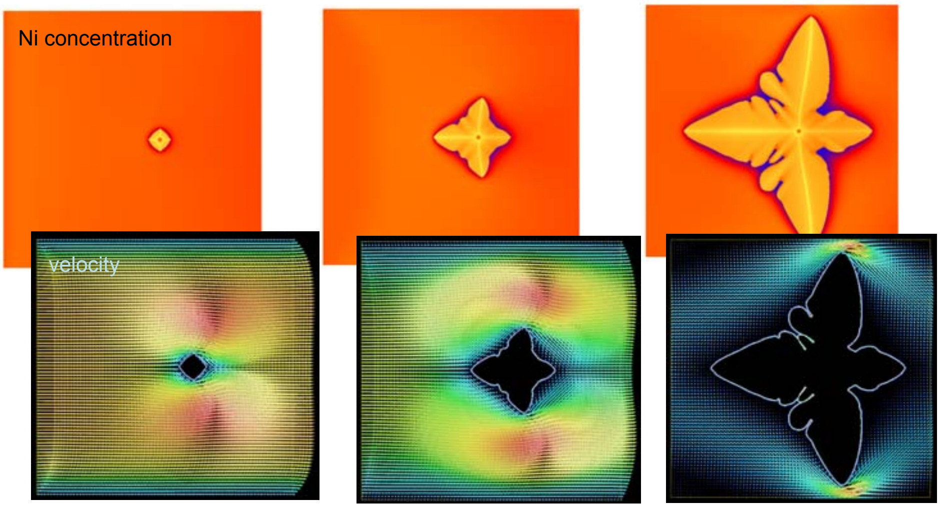
Growth of an equiaxed Ni-Cu dendrite into an undercooled melt



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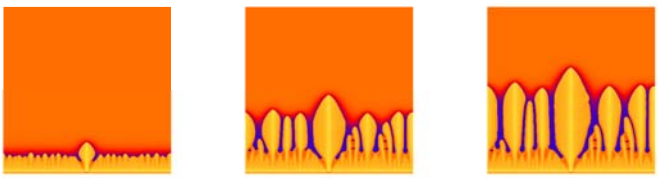
Dendritic solidification in Ni-Cu

without fluid flow



with fluid flow →

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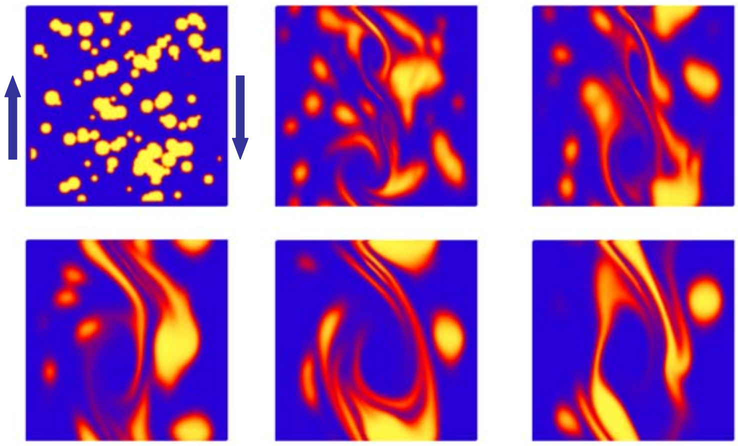
Ripening of liquid droplets in a fluid medium within an induced flow field (shear flow)



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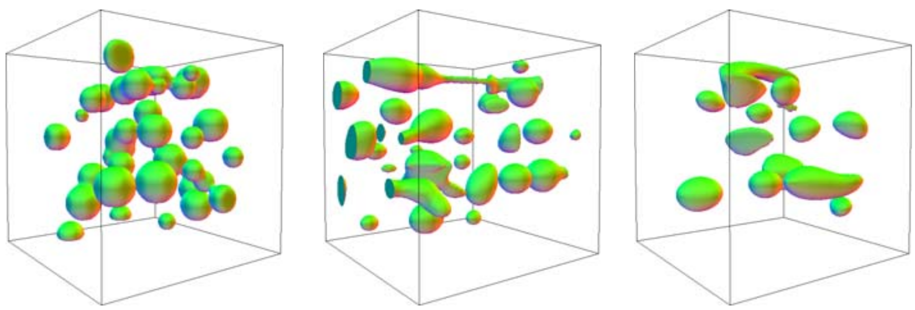
Flow of liquid droplets in a fluid matrix phase



The colours indicate the interfaces between the droplets and the matrix phase

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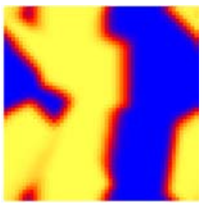
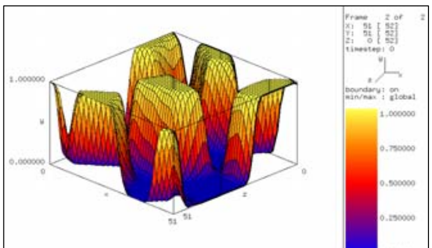
Content:

Postprocessing and data analysis What can phase-field data be used for ?

• Visualisation





simply assign a colour or grey scale and plot it. (2D)

`xsimview` (proprietary)

calculate isosurfaces. (3D)

`openDX` (www.opendx.org)

Postprocessing and data analysis

- Volume $V = \int_\Omega \phi \, dV$ (ϕ = volume fraction of the phase)

- Interfacial area $A = \int_\Omega |\nabla\phi| \, dV$

- Normal vectors of interface $\hat{n} = -\frac{\nabla\phi}{|\nabla\phi|}$

- Normal velocity of interface $v_n = \vec{v_i} \cdot \hat{n} = \vec{v_i} \cdot \left(-\frac{\nabla\phi}{|\nabla\phi|}\right) = \frac{\partial\phi/\partial t}{|\nabla\phi|}$

Postprocessing and data analysis

- Interface curvature $\kappa = \nabla \cdot \hat{n} = -\nabla \cdot \frac{\nabla\phi}{|\nabla\phi|} = -\frac{1}{|\nabla\phi|}\left[\nabla^2\phi - \frac{(\nabla\phi\nabla)|\nabla\phi|}{|\nabla\phi|}\right]$

with $(\nabla\phi\nabla)|\nabla\phi| = \frac{\nabla\phi}{|\nabla\phi|} \cdot \begin{pmatrix} \phi_x\,\phi_{xx} + \phi_y\,\phi_{xy} + \phi_z\,\phi_{xz} \\ \phi_x\,\phi_{xy} + \phi_y\,\phi_{yy} + \phi_z\,\phi_{yz} \\ \phi_x\,\phi_{xz} + \phi_y\,\phi_{yz} + \phi_z\,\phi_{zz} \end{pmatrix}$

- Interface and bulk free energies

$$\mathcal{F}_{surf} = \int_{\Omega(t)} T\left(\varepsilon\, a(\phi, \nabla\phi) + \frac{1}{\varepsilon} w(\phi)\right) dV$$

$$\sum_{\alpha<\beta} \dots$$

$$\mathcal{F}_{bulk} = \int_{\Omega(t)} f(\phi, c, T) \, dV$$

choose specific α/β interfaces for solid / liquid energies etc.