Numerical study for controllable dendritic growth in Phase-field model

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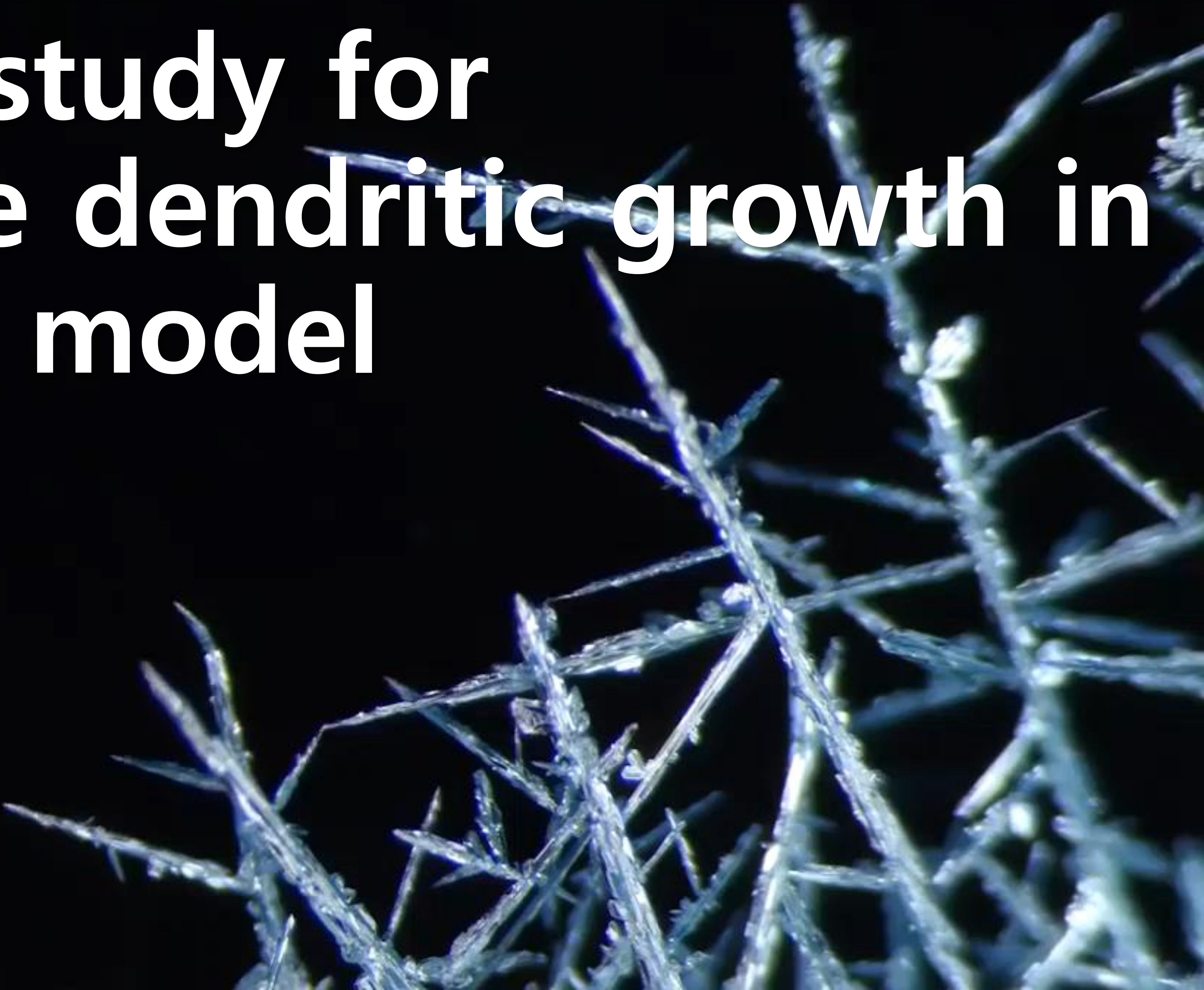
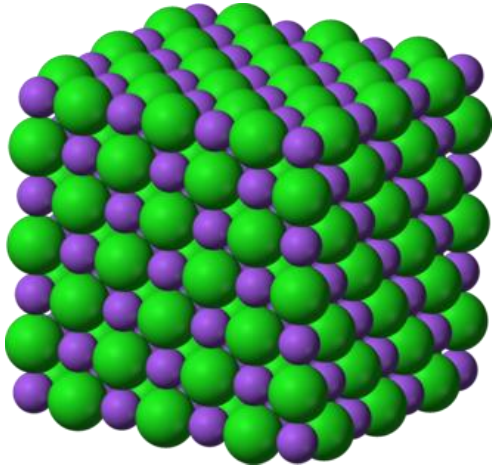


Crystal

A **crystal** is a solid material whose constituents (atoms, molecules, or ions) are arranged in a highly ordered microscopic structure.



Crystal structure

Dendritic structures of a silver crystal

Aluminum

Snowflakes

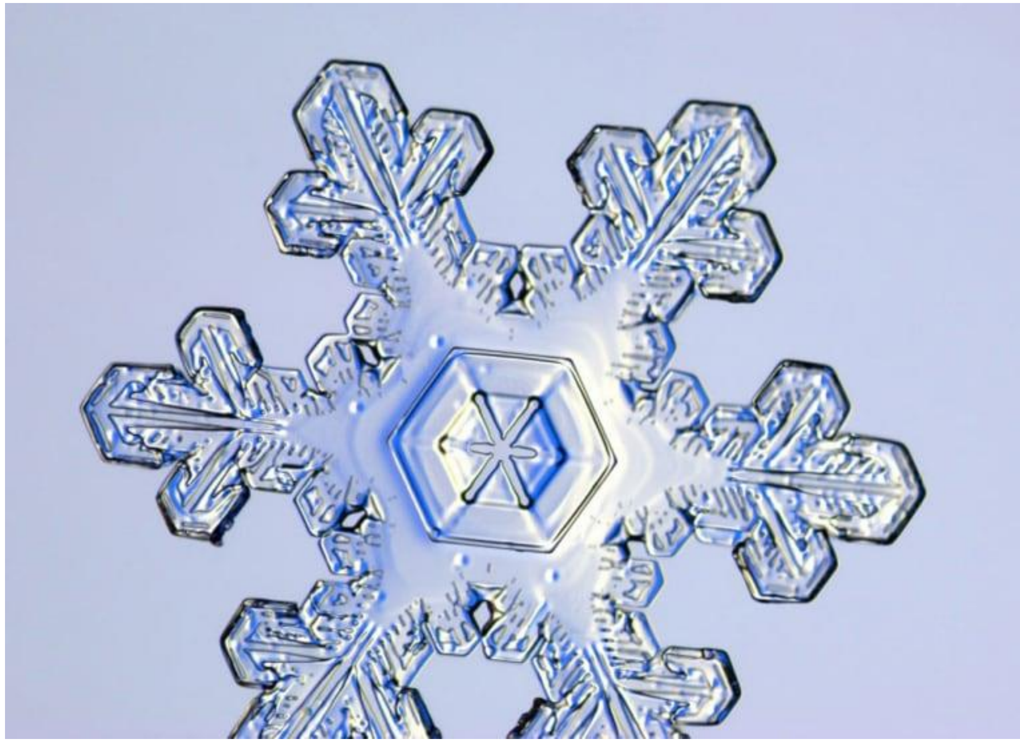
Crystal growth

Crystal growth is major stage of a crystallization process (Nucleation and growth)
Crystal growth is observed when a spherical solid nucleus grows in the supercooled melt.
In general, phase transformation from the liquid phase to the solid phase via heat transfer.

aluminum potassium sulfate

Complex shape of crystals

In ideal situation, the resulting dendrite tend to be perfectly symmetric shape.

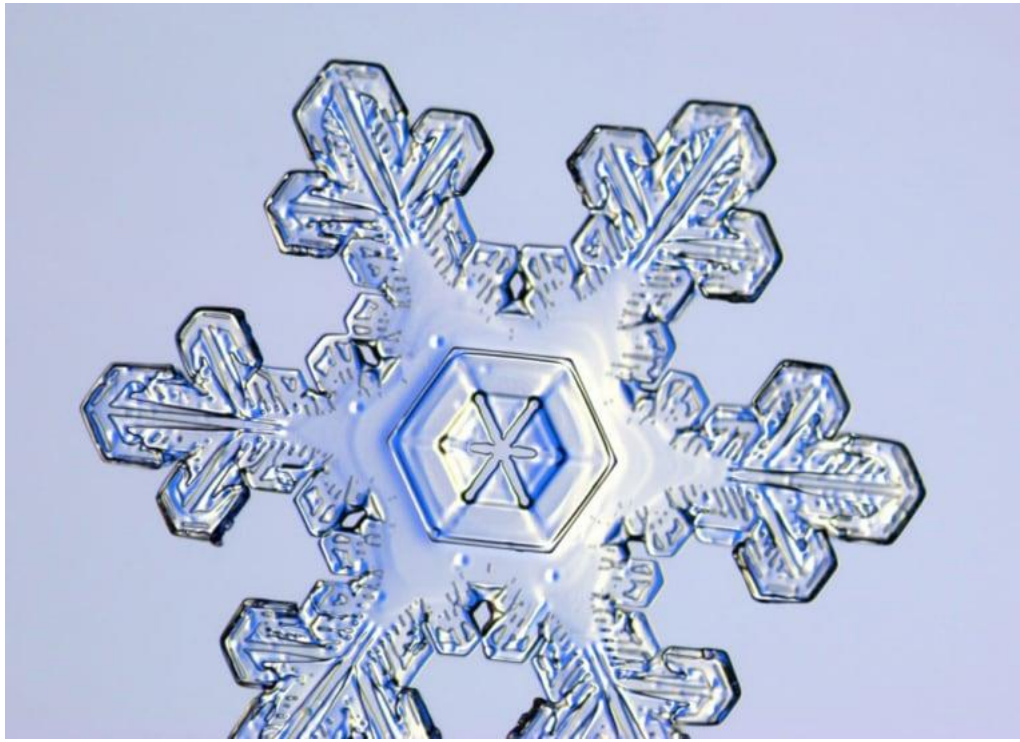


Symmetric snowflakes

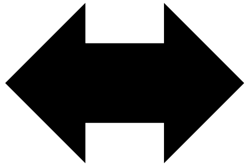
Complex shape of crystals

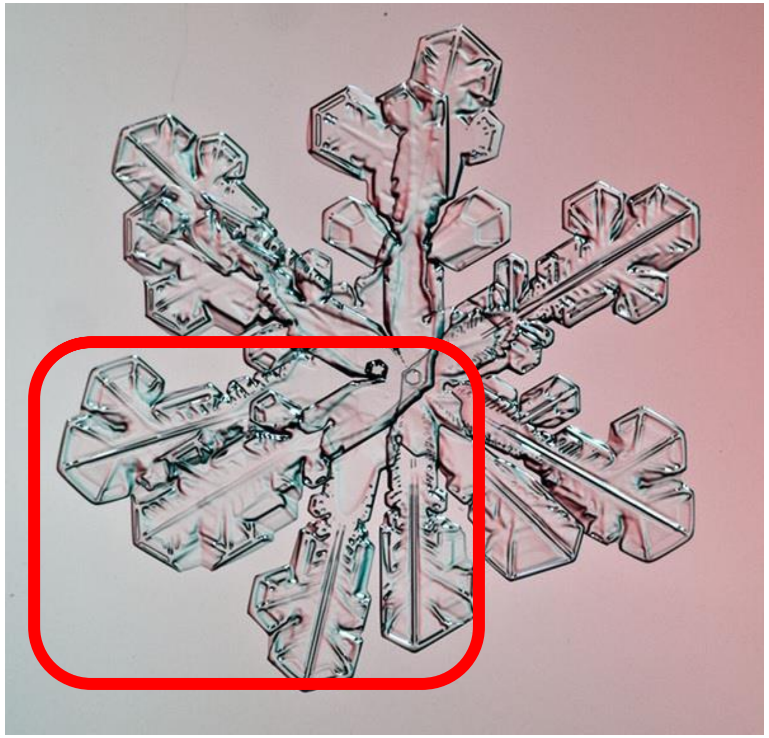
In ideal situation, the resulting dendrite tend to be perfectly symmetric shape.

However there are huge number of crystals, which have not perfectly symmetric structures in nature.



Symmetric snowflakes





Irregular snowflake

Previous work

Ryo Kobayashi

Professor of Mathematics, Hiroshima University

One of first present crystal growth model

Provides a description of the growth of single crystallites and subsequent grain boundary motion

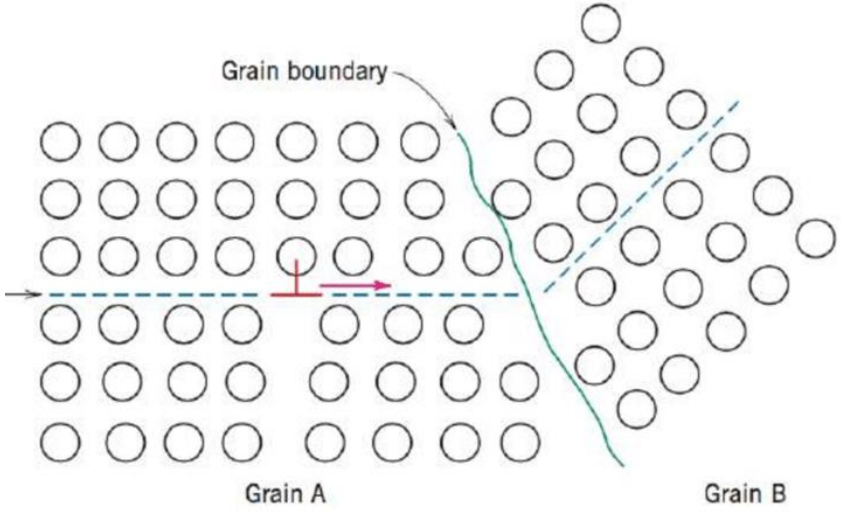
László Gránásy

Wigner Research Centre for Physics, Budapest, Hungary

Extend the work of 'Kobayashi' for grain boundary motion

A grain boundary is the interface between two grains, or crystallites, in a polycrystalline material.

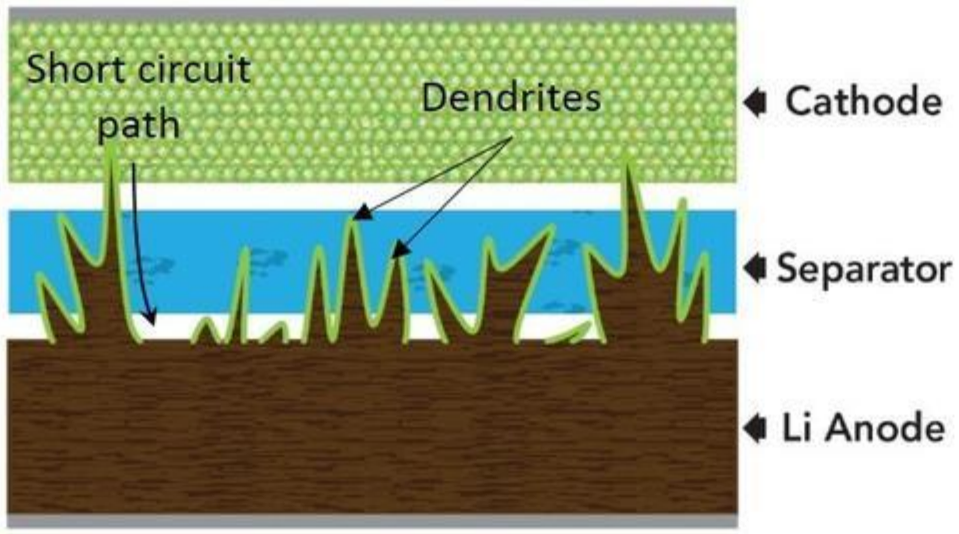
Presence of static heterogeneities (particles) can lead to polycrystalline growth patterns through a sequential deflection of the tips of growing dendrites.

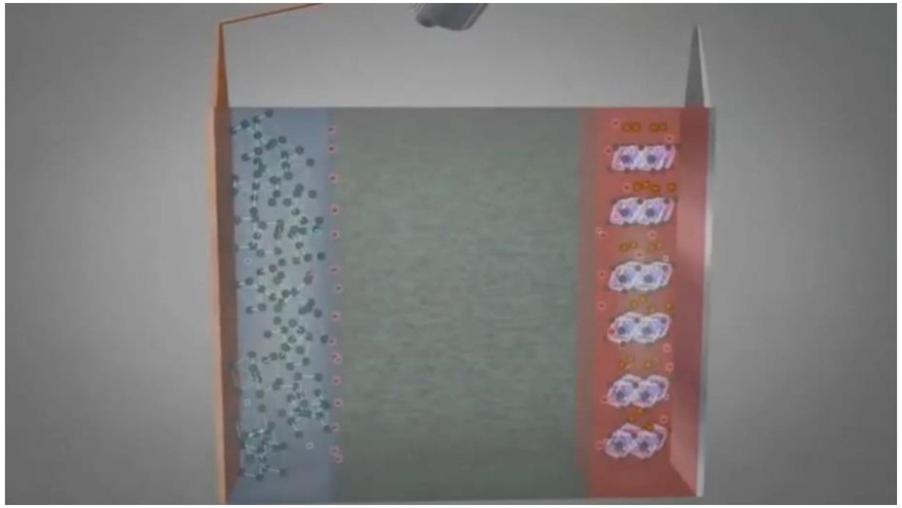


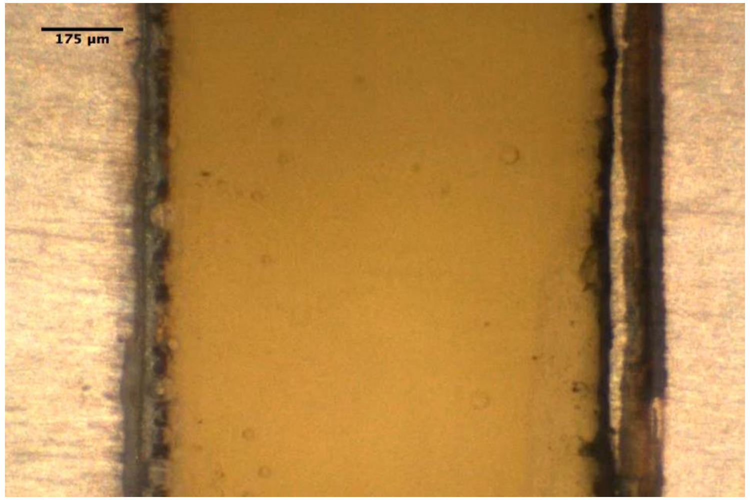
Motivation

Battery

Dendrites are structures that grow in the battery’s internal architecture and cause harm by breaking the barrier between the electrolyte and the material, which leads to battery failure.



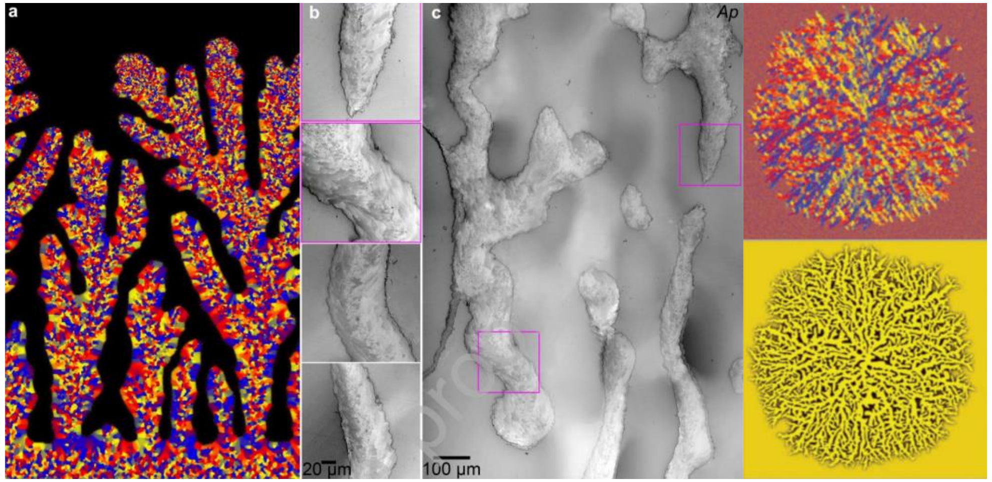


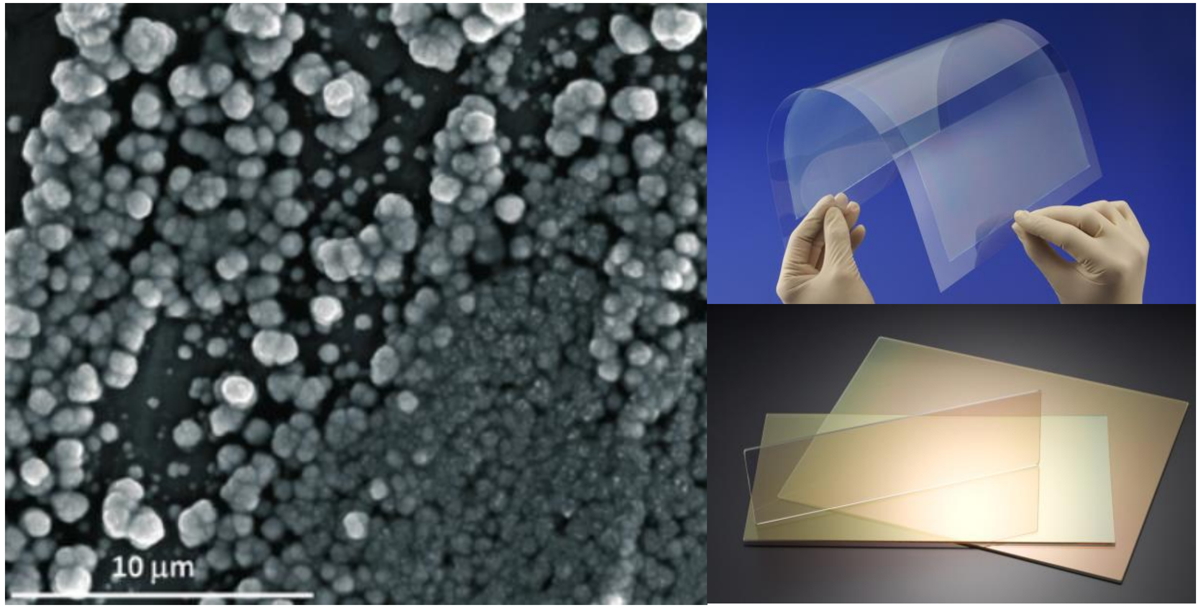


Motivation

Spherulites structure

Crystal growth of spherulites structure, which demonstrated by coral skeletons.





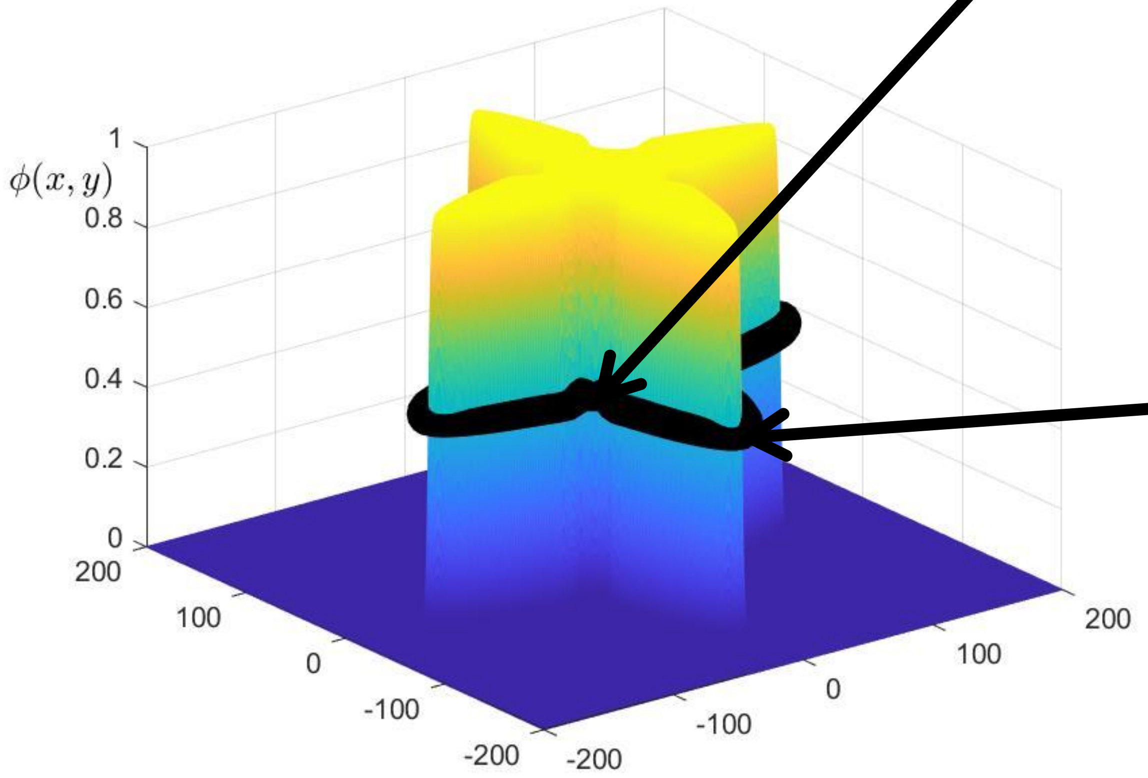
Thin films

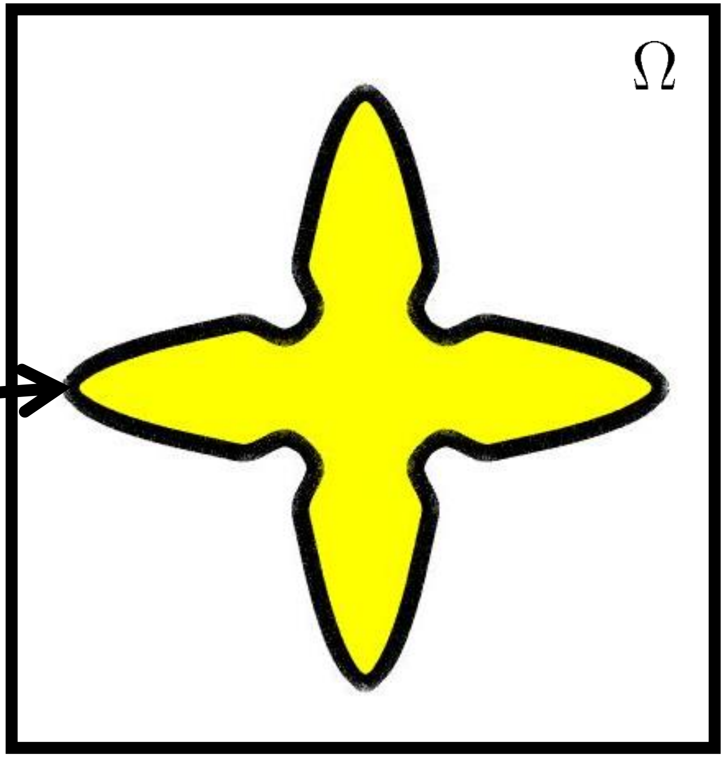
A wide range of patterns having intermediate complexity is found thin films of polymer blends, electrodeposited metal films and single-component polymer films.

Mathematical approach

Phase-field method

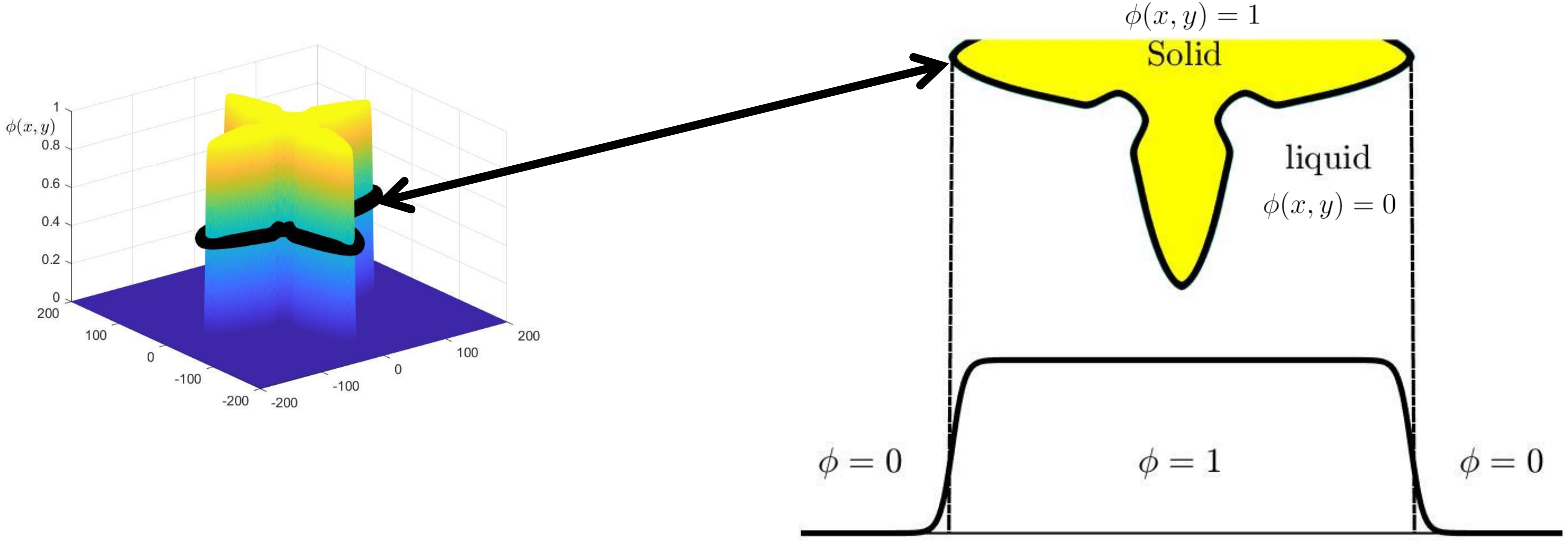
$$\phi(x, y) = 0.5$$





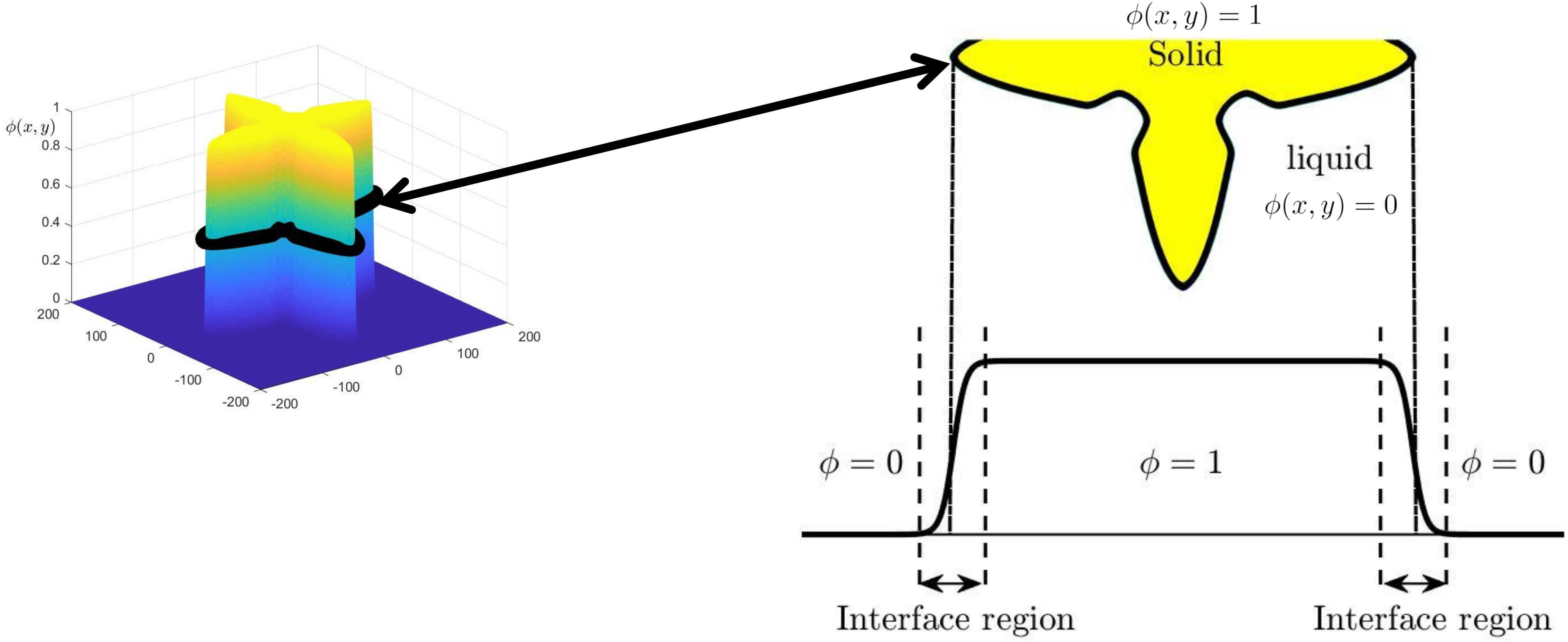
Mathematical approach

Phase-field method



Mathematical approach

Phase-field method



Crystal growth model

Ginzburg Landau type total energy

$$\mathcal{E}(\phi,\xi(U)) = \int \frac{1}{2}\boxed{\epsilon^2(\phi)}|\nabla\phi|^2 + \mathcal{F}(\phi,\xi(U))d\mathbf{x}$$

Anisotropy function

$$\epsilon(\phi) = \bar{\epsilon}(1+\delta\cos(j(\theta-\theta_0)))$$

$\theta = \arctan\left(\phi_y/\phi_x\right)$ θ_0 : Initial orientation

We describe later in detail

Crystal growth model

Ginzburg Landau type total energy

$$\boxed{\mathcal{E}(\phi,\xi(U)) = \int \frac{1}{2}\epsilon^2(\phi)|\nabla\phi|^2 + \boxed{\mathcal{F}(\phi,\xi(U))}\,d\mathbf{x}}$$

Free energy function of double well potential form

$$\mathcal{F}(\phi,\xi(U)) = \boxed{F(\phi)} - \boxed{\lambda}\xi(U)\boxed{p(\phi)}$$

$$F(\phi) = \frac{1}{4}\phi^2(\phi-1)^2$$: Double well potential

λ : Positive constant corresponding with $p(\phi)$

Tilting part of the potential (has two form)

$$p(\phi) = \phi^3(10-15\phi+6\phi^2)$$ or $$p(\phi) = \phi^2(3-2\phi)$$

Crystal growth model

Thermodynamic driving force

$$\mathcal{F}(\phi, \xi(U)) = F(\phi) - \lambda \boxed{\xi(U)} p(\phi)$$

Temperature functional

$$\xi(U) = \frac{\alpha}{\pi} \arctan(\beta(U_e - U)) \quad \text{or} \quad \xi(U) = \beta(U_e - U)$$

U_e : Equilibrium temperature(melting temperature)

β : Positive constant

α : constant in (0, 1)

Crystal growth model

Thermodynamic driving force

$$\xi(U) = \frac{\alpha}{\pi} \arctan(\beta(U_e - \boxed{U}))$$

U : Temperature evolution

The equation for temperature

$$\frac{\partial U}{\partial t} = D\nabla^2 U + K\frac{\partial \boxed{h(\phi)}}{\partial t}$$

D : Diffusion constant

K : Dimension less latent heat

The equation for latent heat

$$h(\phi) = \phi^3(10 - 15\phi + 6\phi^2) \quad \text{or} \quad h(\phi) = \phi$$

Free energy functional

Free energy functional

$$\boxed{\mathcal{F}(\phi, \xi(U)) = F(\phi) - \lambda \xi(U) p(\phi)}$$

We choose functions as

$$p(\phi) = \phi^2(3 - 2\phi) \qquad h(\phi) = \phi \qquad \xi(U) = \frac{\alpha}{\pi}\arctan(\beta(U_e - U))$$

With $F(\phi) = \frac{1}{4}\phi^2(\phi - 1)^2$ and $\lambda = 1/6$

Then we get,

$$\mathcal{F}(\phi, \xi(U)) = \frac{1}{4}\phi^2(\phi - 1)^2 - \frac{1}{6}\xi(U)\phi^2(3 - 2\phi)$$

Free energy functional

Taking fixed value at $\phi = 0$ and $\phi = 1$

$$\boxed{\mathcal{F}'(\phi, \xi(U)) = \phi(\phi - 1)(\phi - \frac{1}{2} + \xi(U))}$$

Local extremum point at
$$\begin{aligned} \phi &= 0, \\ \phi &= 1, \\ \phi &= \frac{1}{2} - \xi(U) \end{aligned}$$

Therefore we need that $|\xi(U)| < \frac{1}{2}$, because the leading coefficient is positive.

Since $\xi(U) = \frac{\alpha}{\pi} \arctan(\beta(U_e - U))$, we need to $0 < \alpha < 1$

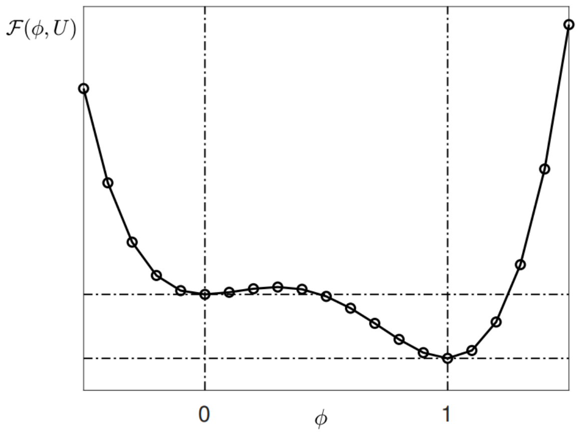
Free energy functional

Interface movement

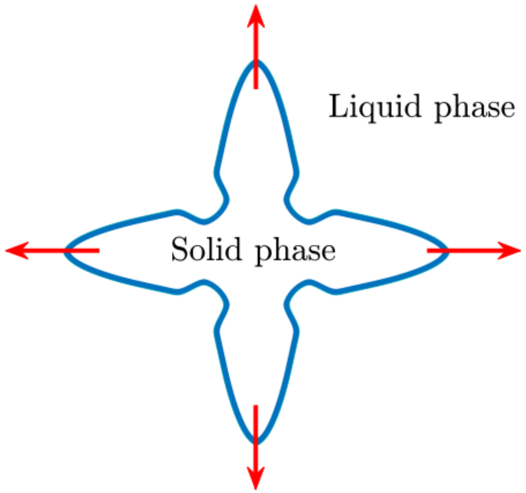
$$\boxed{\mathcal{F}(0,\xi(U)) - \mathcal{F}(1,\xi(U)) = \lambda\xi(U)}$$

If $\xi(U) > 0$, then solid phase is stable and liquid phase is metastable.

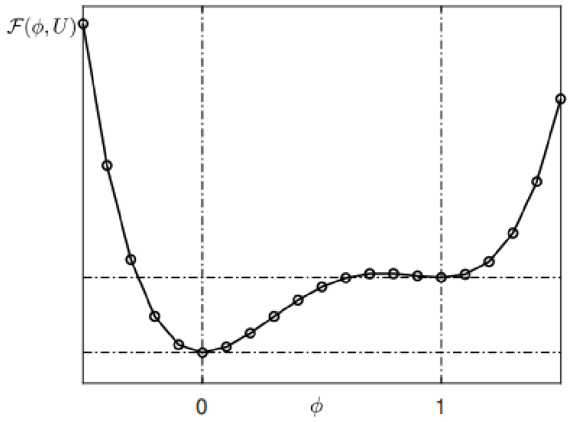
If $\xi(U) < 0$, then liquid phase is stable and solid phase is stable.



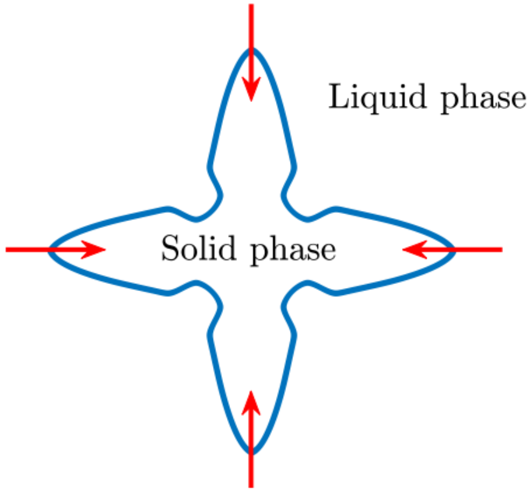
$\xi(U) > 0$



$\xi(U) > 0$



$\xi(U) < 0$



$\xi(U) < 0$

Crystal growth model

Phase field equation

$$\mathcal{E}(\phi,\xi(U)) = \int \frac{1}{2}\epsilon^2(\phi)|\nabla\phi|^2 + \mathcal{F}(\phi,\xi(U))d\mathbf{x}$$

$$\mathcal{F}(\phi,\xi(U)) \;=\; \frac{1}{4}\phi^2(\phi-1)^2 - \frac{1}{6}\xi(U)\phi^2(3-2\phi)$$

By gradient formula, $\tau(\phi)\frac{\partial\phi}{\partial t} = -\frac{\partial\mathcal{E}}{\partial\phi}$

$$\tau(\phi)\frac{\partial\phi}{\partial t} = \nabla\cdot(\epsilon^2(\phi)\nabla\phi) - \frac{\partial}{\partial x}(\epsilon(\phi)\frac{\partial\epsilon(\phi)}{\partial\theta}\frac{\partial\phi}{\partial y}) + \frac{\partial}{\partial y}(\epsilon(\phi)\frac{\partial\epsilon(\phi)}{\partial\theta}\frac{\partial\phi}{\partial x}) + \phi(1-\phi)(\phi - \frac{1}{2} + \xi(U))$$

$\tau(\phi)$ is also anisotropy function, which we define $\tau(\phi) = \epsilon^2(\phi)$

Controllable crystal growth model

Total free energy

$$\mathcal{E}(\phi, \xi(U), \boxed{\theta_{ori}}) = \int \frac{1}{2}\epsilon^2(\phi)|\nabla\phi|^2 + \mathcal{F}(\phi, \xi(U), \boxed{\theta_{ori}})d\mathbf{x}$$

The local free energy density

$$\mathcal{F}(\phi, \xi(U), \theta_{ori}) = F(\phi) + p(\phi)(g_s(U) + g_{ori}(|\nabla\theta_{ori}|)) + (1 - p(\phi))g_l(U)$$

By gradient formula,

$$\tau(\phi)\frac{\partial\phi}{\partial t} = \nabla\cdot(\epsilon^2(\phi)\nabla\phi) - \frac{\partial}{\partial x}(\epsilon(\phi)\frac{\partial\epsilon(\phi)}{\partial\theta}\frac{\partial\phi}{\partial y}) + \frac{\partial}{\partial y}(\epsilon(\phi)\frac{\partial\epsilon(\phi)}{\partial\theta}\frac{\partial\phi}{\partial x})$$
$$-f(\phi) - p'(\phi)(g_{ori}(|\nabla\theta_{ori}|) + g_s(U) - g_l(U))$$

Free energy functional

Free energy functional for controllable crystal growth

$$\mathcal{F}(\phi, \xi(U), \theta_{ori}) = \boxed{F(\phi)} + \boxed{p(\phi)}\left(\boxed{g_s(U)} + g_{ori}(|\nabla\theta_{ori}|)\right) + (1 - p(\phi))\boxed{g_l(U)}$$

$F(\phi) = \frac{1}{4}\phi^2(\phi - 1)^2$: Double well potential

$g_s(U)$: Local free energy function for solid phase

$g_l(U)$: Local free energy function for liquid phase

The function $p(\phi)$ switches contribution of each local free energy function.

$p(1) = 1$ and $p(0) = 0$

$$p(\phi) = \phi^3(10 - 15\phi + 6\phi^2)$$
$$p(\phi) = \phi^2(3 - 2\phi).$$

Orientation field

Orientation energy

$$\mathcal{F}(\phi, \xi(U), \theta_{ori}) = F(\phi) + p(\phi)(g_s(U) + \boxed{g_{ori}(|\nabla\theta_{ori}|)}) + (1 - p(\phi))g_l(U)$$

Misalignment energy function

$$g_{ori} = H|\nabla\theta_{ori}|$$

H : Constant for low angle grain boundaries

Also, by the formula $\dfrac{\partial\theta_{ori}}{\partial t} = -M_\theta \dfrac{\partial\mathcal{E}}{\partial\theta_{ori}}$

$$\frac{\partial\theta_{ori}}{\partial t} = -M_\theta H \nabla \cdot \left(p(\phi) \frac{\nabla\theta_{ori}}{|\nabla\theta_{ori}|} \right)$$

Orientation field

Orientation field

$$\frac{\partial \theta_{ori}}{\partial t} = \boxed{-M_\theta} H \nabla \cdot \left(p(\phi) \frac{\nabla \theta_{ori}}{|\nabla \theta_{ori}|} \right)$$

Orientation mobility

$$M_\theta = M_s + (M_l - M_s)(1 - p(\phi))$$

M_s : Mobility for pure solid

M_l : Mobility for pure liquid

Suppose that there is no mobility in solid phase : $M_s = 0$

Orientation field

$$\frac{\partial \theta_{ori}}{\partial t} = -M_l (1 - p(\phi)) H \nabla \cdot \left(p(\phi) \frac{\nabla \theta_{ori}}{|\nabla \theta_{ori}|} \right)$$

Orientation field

Anisotropy function

$$\epsilon(\phi) = \bar{\epsilon}(1 + \delta\cos(j(\theta - \boxed{\theta_{ori}})))$$

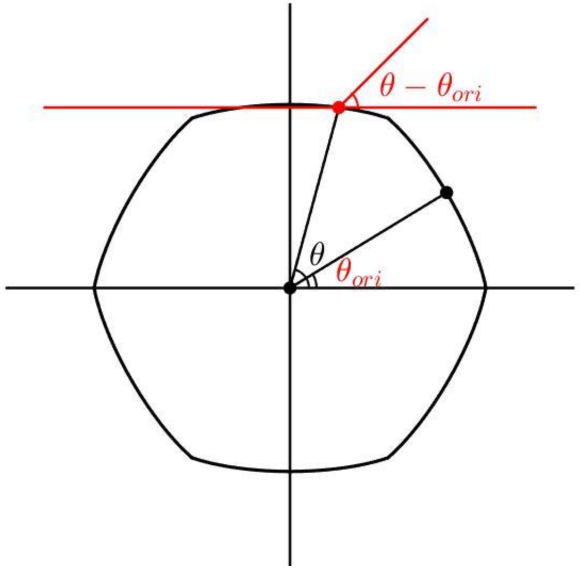
$\theta = \arctan\left(\phi_y/\phi_x\right)$

θ_0 : Initial orientation

$\bar{\epsilon}$: Interface thickness

δ : Anisotropy strength

j : Mode of anisotropy



$$\frac{\partial \theta_{ori}}{\partial t} = -M_l(1 - p(\phi))H\nabla \cdot \left(p(\phi)\frac{\nabla\theta_{ori}}{|\nabla\theta_{ori}|}\right)$$

Controllable crystal growth model

Free energy functional

$$\boxed{\mathcal{F}(\phi, \xi(U), \theta_{ori}) = F(\phi) + p(\phi)(g_s(U) + g_{ori}(|\nabla\theta_{ori}|)) + (1 - p(\phi))g_l(U)}$$

We choose functions as

$$F(\phi) \;=\; \frac{1}{4}\phi^2(\phi-1)^2 \qquad p(\phi) = \phi^2(3-2\phi) \qquad g_l \;=\; 0 \qquad g_s \;=\; -\frac{1}{6}\xi(U)$$

Then we get,

$$\mathcal{F}(\phi, \xi(U), \theta_{ori}) = F(\phi) - \frac{1}{6}p(\phi)(\xi(U) - 6g_{ori}(|\nabla\theta_{ori}|))$$

Controllable crystal growth model

Phase field equation

$$\mathcal{E}(\phi, \xi(U), \theta_{ori}) = \int \frac{1}{2}\epsilon^2(\phi)|\nabla\phi|^2 + \mathcal{F}(\phi, \xi(U), \theta_{ori})d\mathbf{x}$$

$$\mathcal{F}(\phi, \xi(U), \theta_{ori}) = F(\phi) - \frac{1}{6}p(\phi)(\xi(U) - 6g_{ori}(|\nabla\theta_{ori}|))$$

Therefore

$$\tau(\phi)\frac{\partial\phi}{\partial t} = \nabla\cdot(\epsilon^2\nabla\phi) - \frac{\partial}{\partial x}(\epsilon\frac{\partial\epsilon}{\partial\theta}\frac{\partial\phi}{\partial y}) + \frac{\partial}{\partial y}(\epsilon\frac{\partial\epsilon}{\partial\theta}\frac{\partial\phi}{\partial x})$$
$$+\phi(1-\phi)(\phi - \frac{1}{2} + \xi(U) - 6g_{ori}(|\nabla\theta_{ori}|))$$

Governing system

Phase field

$$\tau(\phi)\frac{\partial\phi}{\partial t} = \nabla\cdot(\epsilon^2\nabla\phi) - \frac{\partial}{\partial x}(\epsilon\frac{\partial\epsilon}{\partial\theta}\frac{\partial\phi}{\partial y}) + \frac{\partial}{\partial y}(\epsilon\frac{\partial\epsilon}{\partial\theta}\frac{\partial\phi}{\partial x})$$
$$+\phi(1-\phi)(\phi - \frac{1}{2} + \xi(U) - 6g_{ori}(|\nabla\theta_{ori}|))$$

Temperature field

$$\frac{\partial U}{\partial t} = D\nabla^2 U + K\frac{\partial\phi}{\partial t}$$

Orientation field

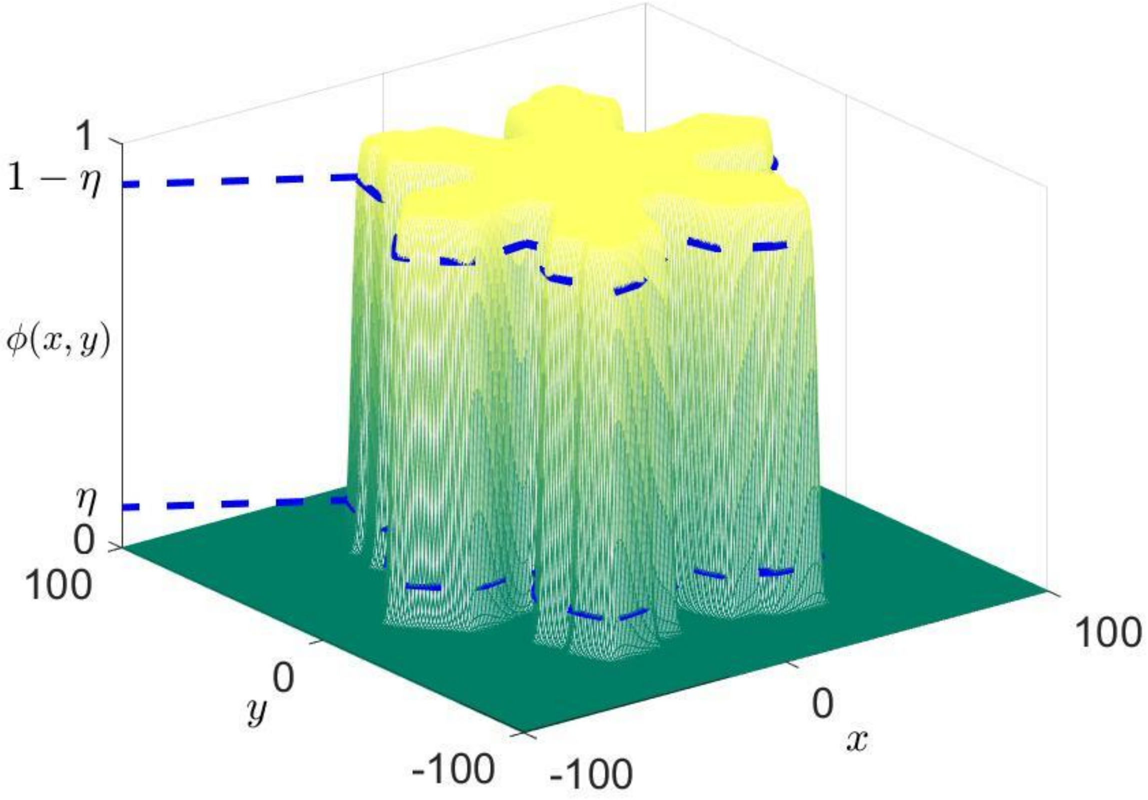
$$\frac{\partial\theta_{ori}}{\partial t} = -M_l(1-p(\phi))H\nabla\cdot\left(p(\phi)\frac{\nabla\theta_{ori}}{|\nabla\theta_{ori}|}\right)$$

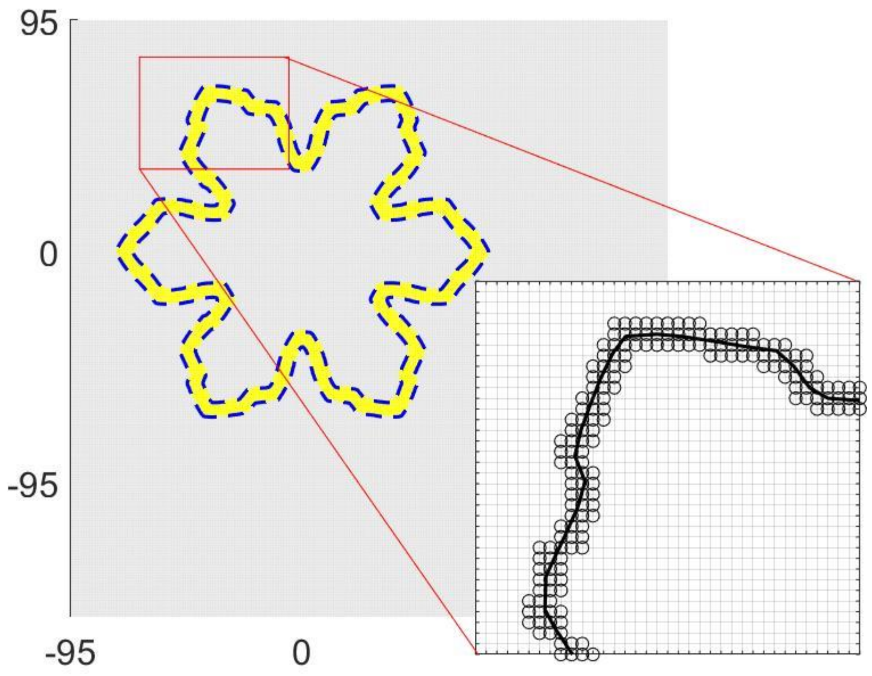
Numerical scheme

Temporal domain

$$\Omega_{tmp} = \{(x_i, y_i) |\, \eta \leq \phi_{ij} \leq 1-\eta, 1 \leq i \leq N_x, 1 \leq j \leq N_y\}$$

For positive integer, $0 < \eta << 1$

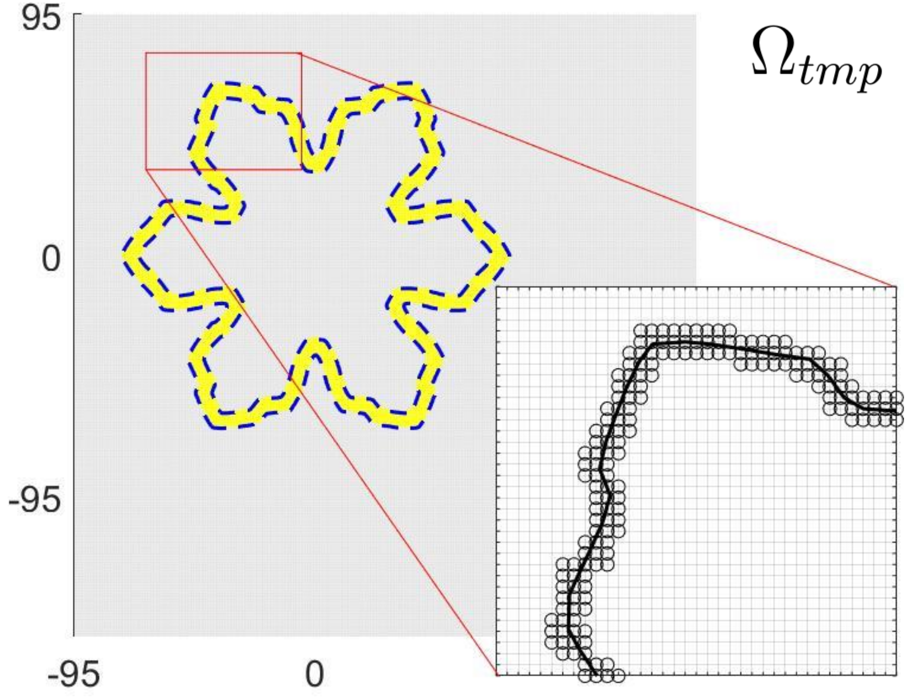


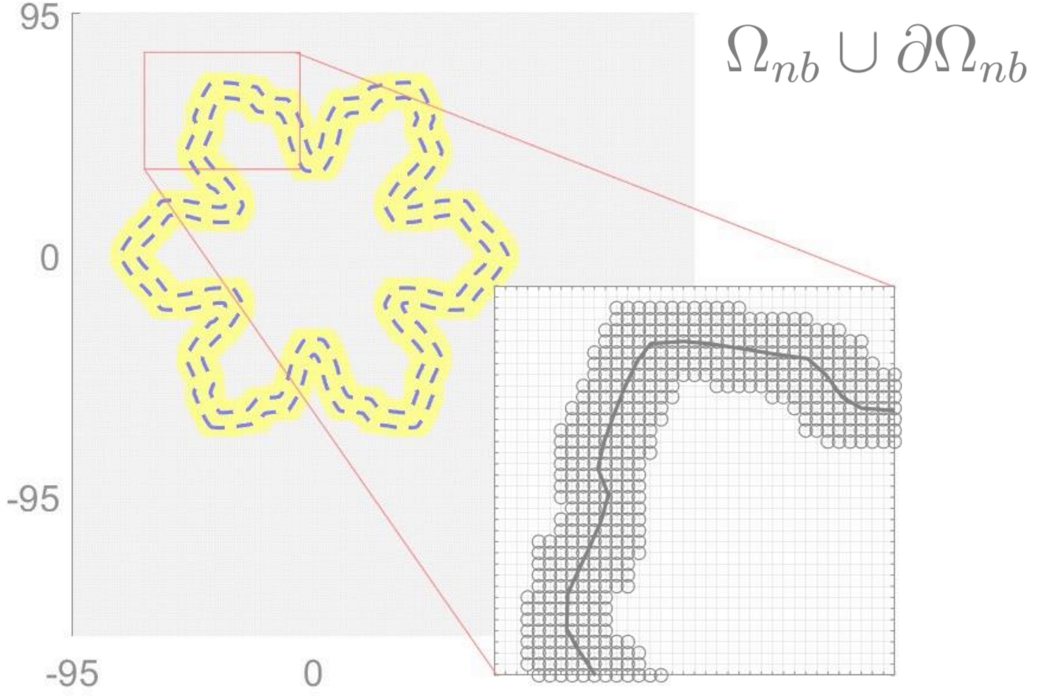


Numerical scheme

Narrow band domain

$$\Omega_{nb} \cup \partial\Omega_{nb} = \bigcup_{p=-m}^{m} \bigcup_{q=-m}^{m} \{(x_{i+p}, y_{j+q}) | \, (x_i, y_j) \in \Omega_{tmp}\}$$

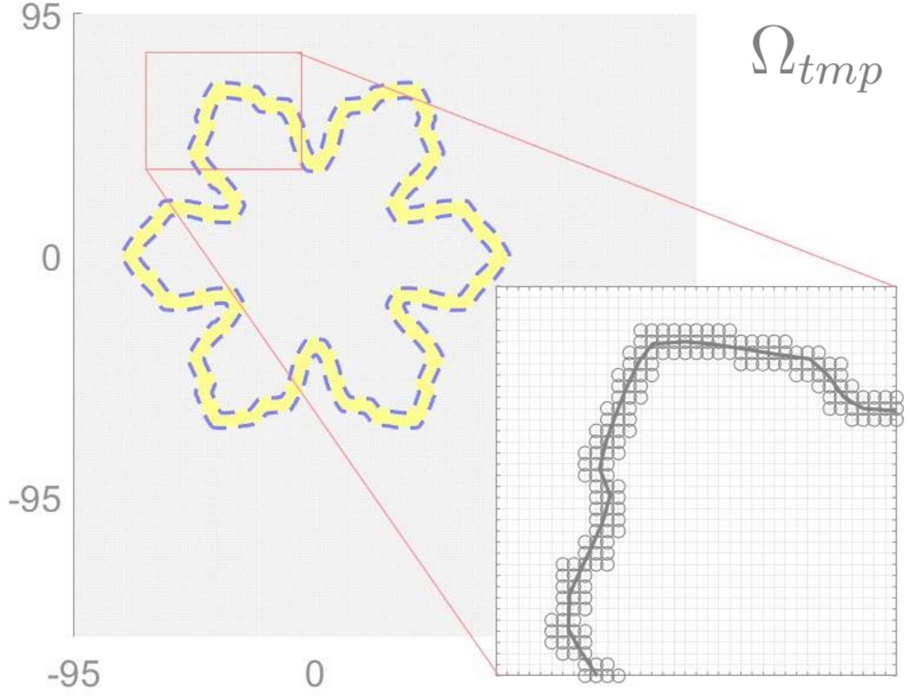


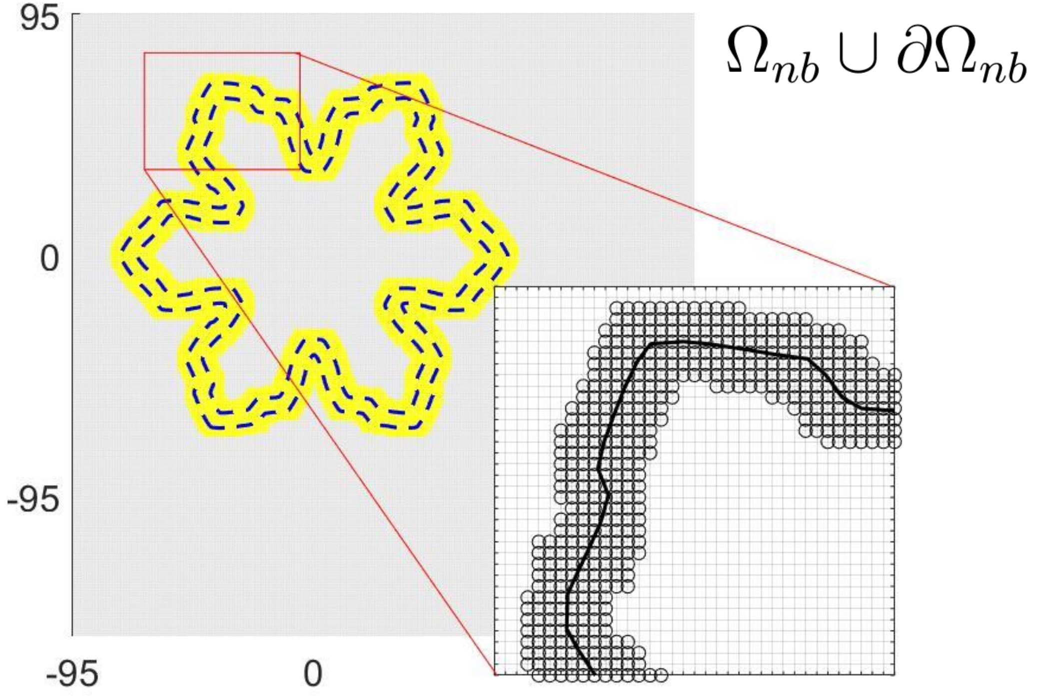


Numerical scheme

Narrow band domain

$$\Omega_{nb} \cup \partial\Omega_{nb} = \bigcup_{p=-m}^{m} \bigcup_{q=-m}^{m} \{(x_{i+p}, y_{j+q}) | (x_i, y_j) \in \Omega_{tmp}\}$$





Numerical scheme

Operator splitting scheme for phase field

[Phase field] $$\epsilon^2(\phi)\frac{\partial \phi}{\partial t} = \nabla\cdot(\epsilon^2\nabla\phi) - \frac{\partial}{\partial x}(\epsilon\frac{\partial \epsilon}{\partial \theta}\frac{\partial \phi}{\partial y}) + \frac{\partial}{\partial y}(\epsilon\frac{\partial \epsilon}{\partial \theta}\frac{\partial \phi}{\partial x})$$
$$+\phi(1-\phi)(\phi-\frac{1}{2}+\xi(U)-6g_{ori}(|\nabla\theta_{ori}|))$$

[Step 1] $$\epsilon^2(\phi)\frac{\partial \phi}{\partial t} = \nabla\cdot(\epsilon^2\nabla\phi) - \frac{\partial}{\partial x}(\epsilon\frac{\partial \epsilon}{\partial \theta}\frac{\partial \phi}{\partial y}) + \frac{\partial}{\partial y}(\epsilon\frac{\partial \epsilon}{\partial \theta}\frac{\partial \phi}{\partial x})$$
$$+\phi(1-\phi)(\xi(U)-6g_{ori}(|\nabla\theta_{ori}|))$$

[Step 2] $$\frac{\partial \phi}{\partial t} = \phi(1-\phi)(\phi-1/2)$$

Numerical scheme

Operator splitting scheme

[Step 1]

$$
\begin{aligned}
\epsilon^2(\phi_{ij}^n)\frac{\phi_{ij}^* - \phi_{ij}^n}{\Delta t} \quad &= \quad [\nabla \cdot (\epsilon^2(\phi)\nabla\phi)]_{ij}^n + \phi_{ij}^n(1-(\phi_{ij}^n))(\xi(U_{ij}^n) - 6g_{ori}^n(|\nabla\theta_{ori}|)) \\
&\quad - \left[\left(-\frac{12\bar{\epsilon}\delta\epsilon(\phi)\phi_y^2(3\phi_x^5 - 10\phi_x^3\phi_y^2 + 3\phi_x\phi_y^4)}{|\nabla\phi|^6}\right)_x\right]_{ij}^n \\
&\quad + \left[\left(-\frac{12\bar{\epsilon}\delta\epsilon(\phi)\phi_x^2(3\phi_y^5 - 10\phi_x^2\phi_y^3 + 3\phi_y\phi_x^4)}{|\nabla\phi|^6}\right)_y\right]_{ij}^n
\end{aligned}
$$

[Step 2]

$$
\frac{\phi_{ij}^{n+1} - \phi_{ij}^n}{\Delta t} \quad = \quad \phi_{ij}^*(1-\phi_{ij}^*)(\phi_{ij}^* - 1/2)
$$

Numerical scheme

Explicit scheme for other field

[Step 3]

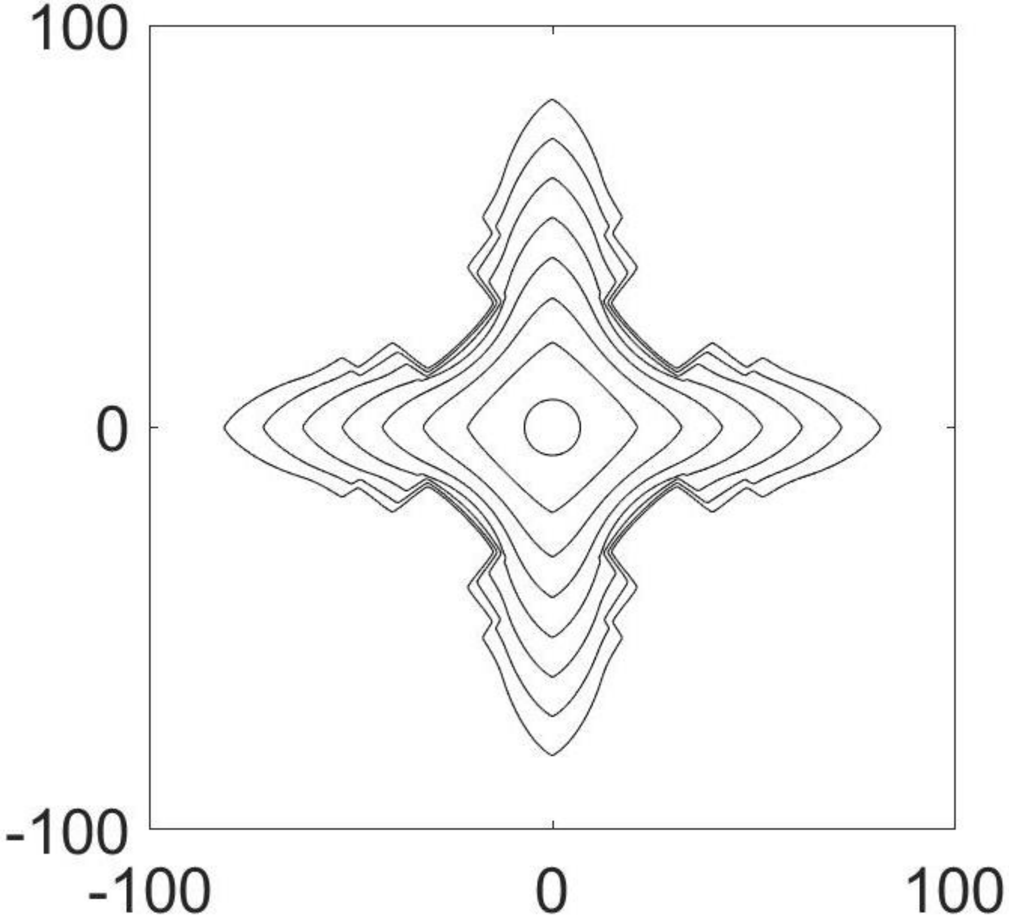
$$\frac{U_{ij}^{n+1} - U_{ij}^{n}}{\Delta t} = D\frac{U_{i-1,j}^{n} + U_{i+1,j}^{n} - 4U_{i,j}^{n} + U_{i,j-1}^{n} + U_{i,j+1}^{n}}{h^2} + \frac{\phi_{ij}^{n+1} - \phi_{ij}^{n}}{\Delta t}$$

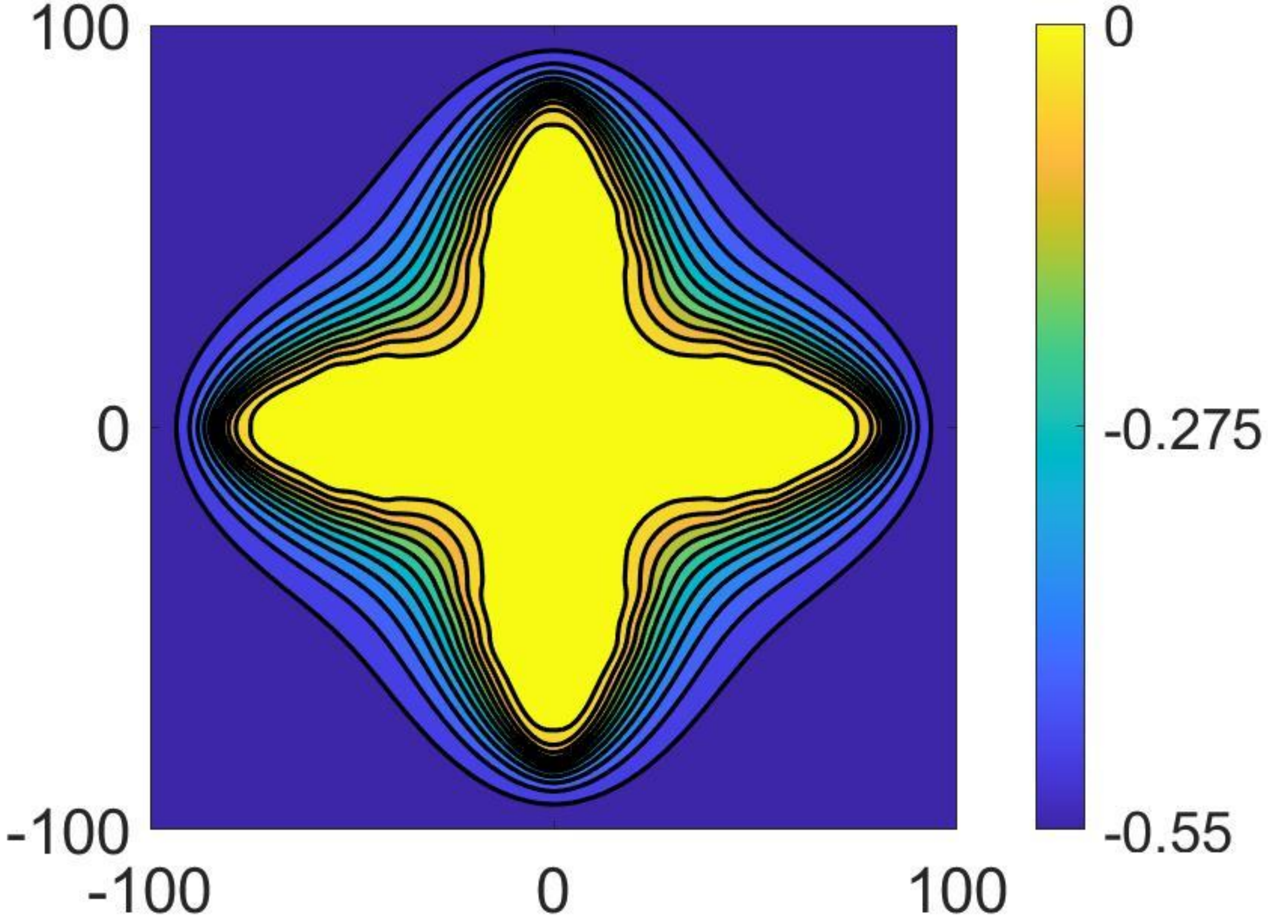
[Step 4]

$$\frac{\theta_{ij}^{n+1} - \theta_{ij}^{n}}{\Delta t} = -M_l(1 - p(\phi_{ij}^{n}))H\left[\nabla \cdot \left(p(\phi_{ij})\frac{\nabla\theta_{ori}}{|\nabla\theta_{ori}|}\right)\right]_{ij}^{n}$$

Numerical result

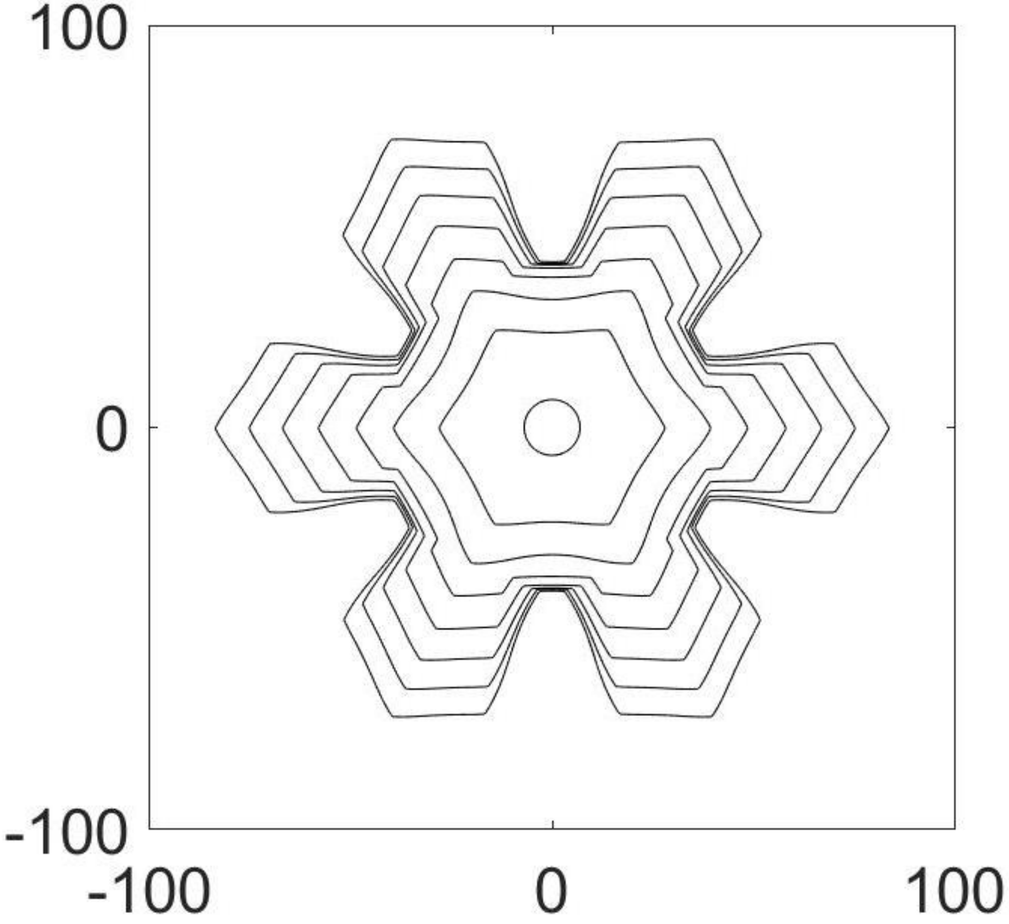
Growth history and temperature distribution

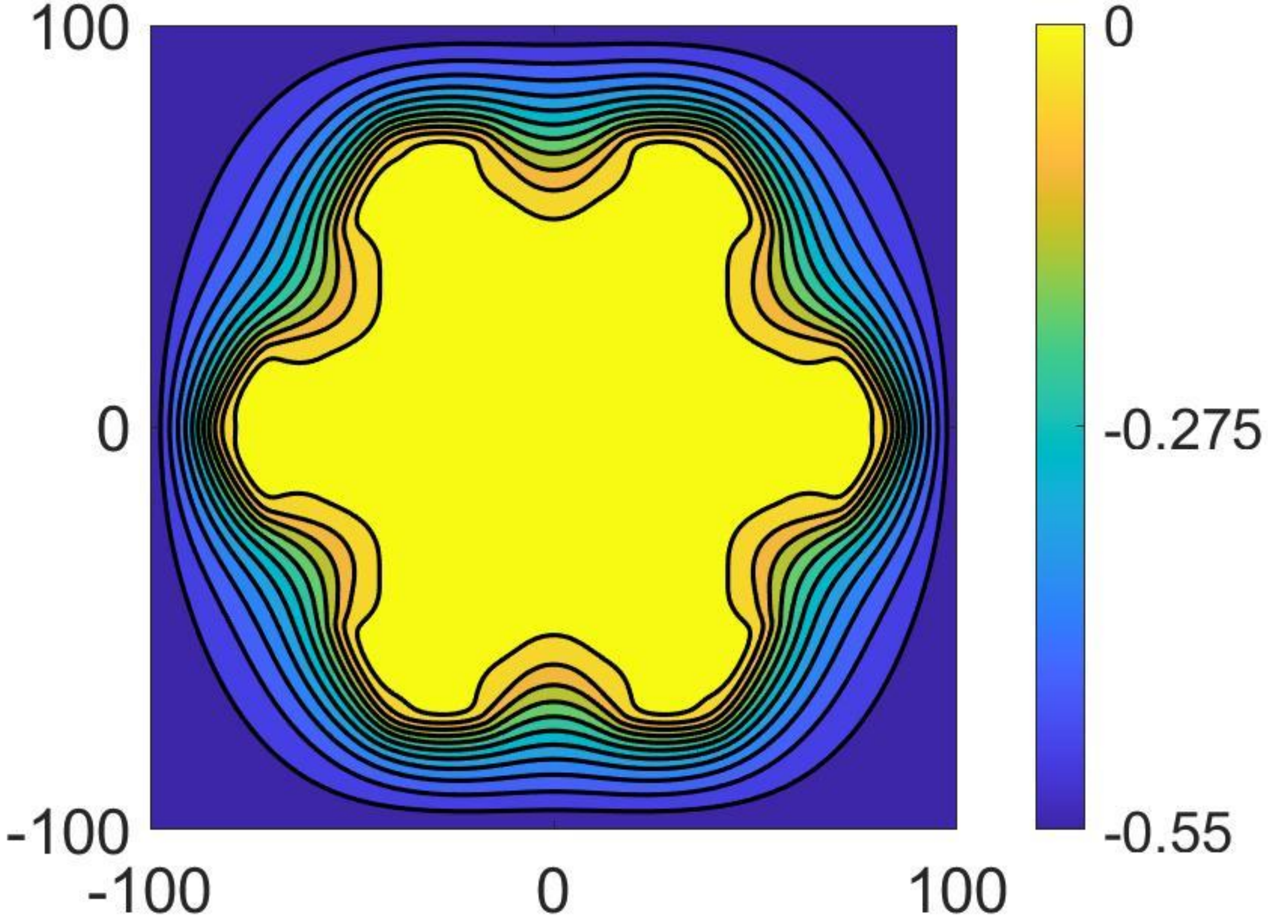




Numerical result

Growth history and temperature distribution

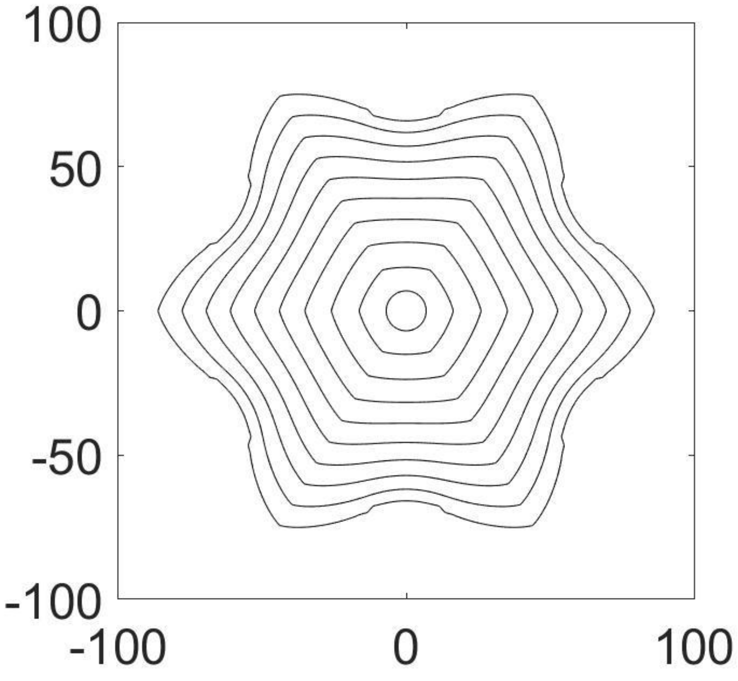




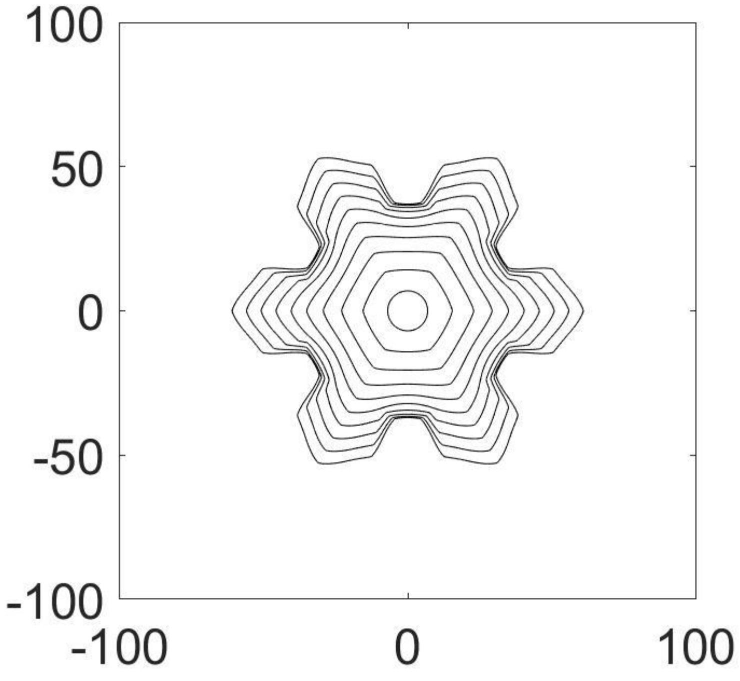
Numerical result

Effect of parameters K

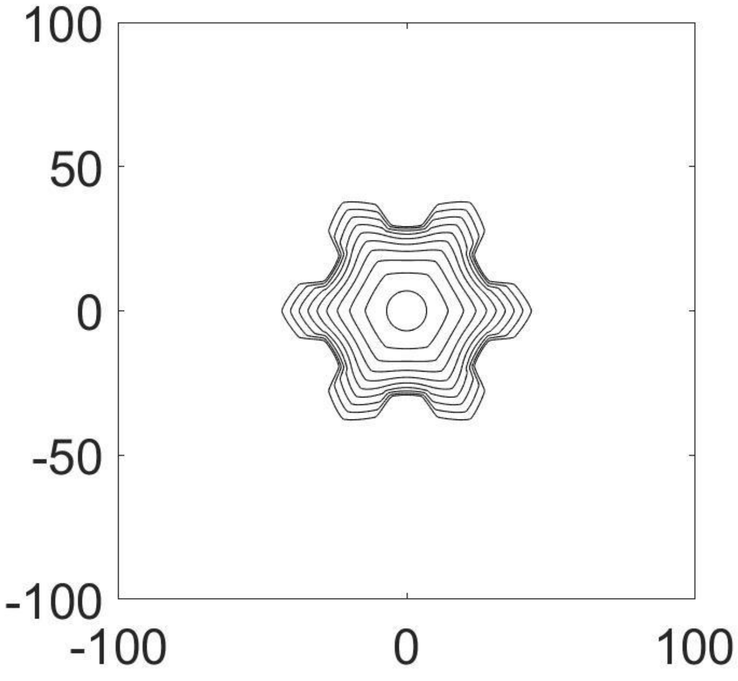
$$\frac{\partial U}{\partial t} = D\nabla^2 U + K\frac{\partial \phi}{\partial t}$$



$K = 0.75$



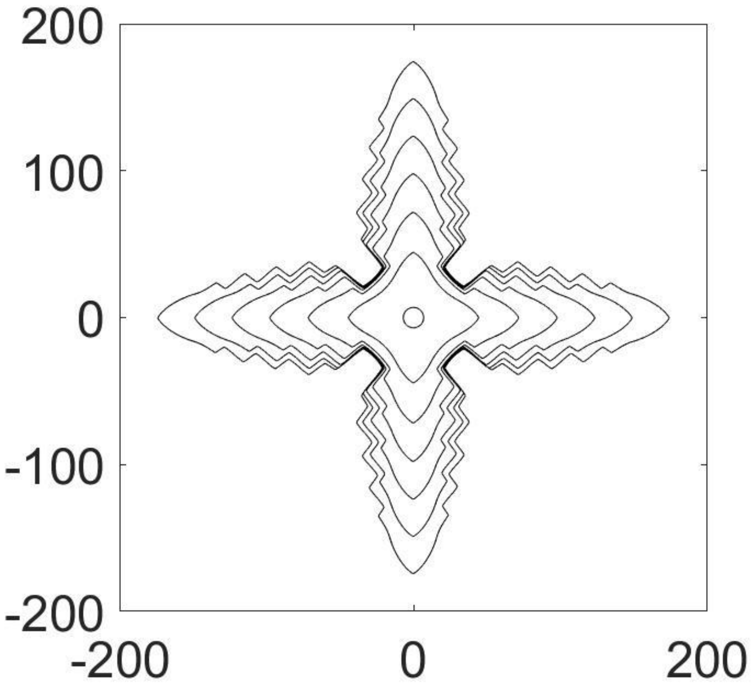
$K = 1.0$



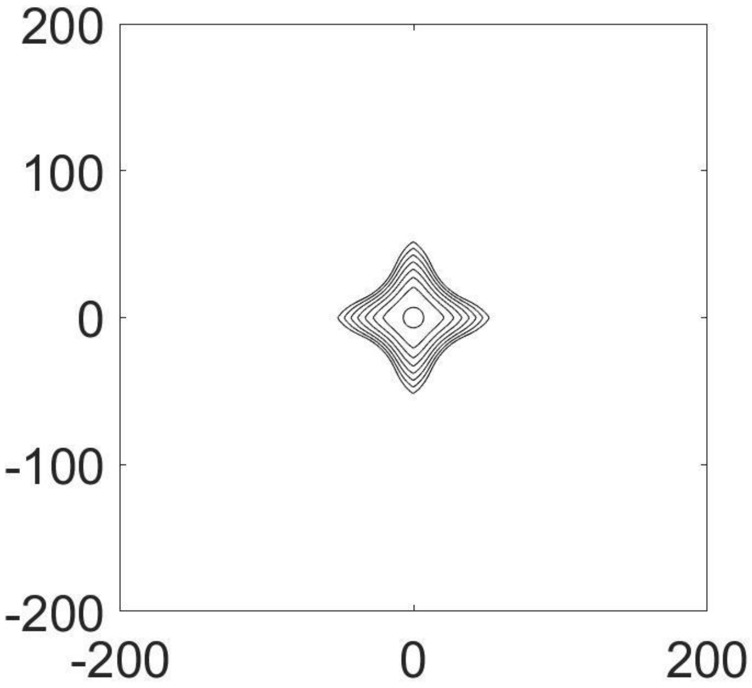
$K = 1.25$

Numerical result

Effect of parameters K



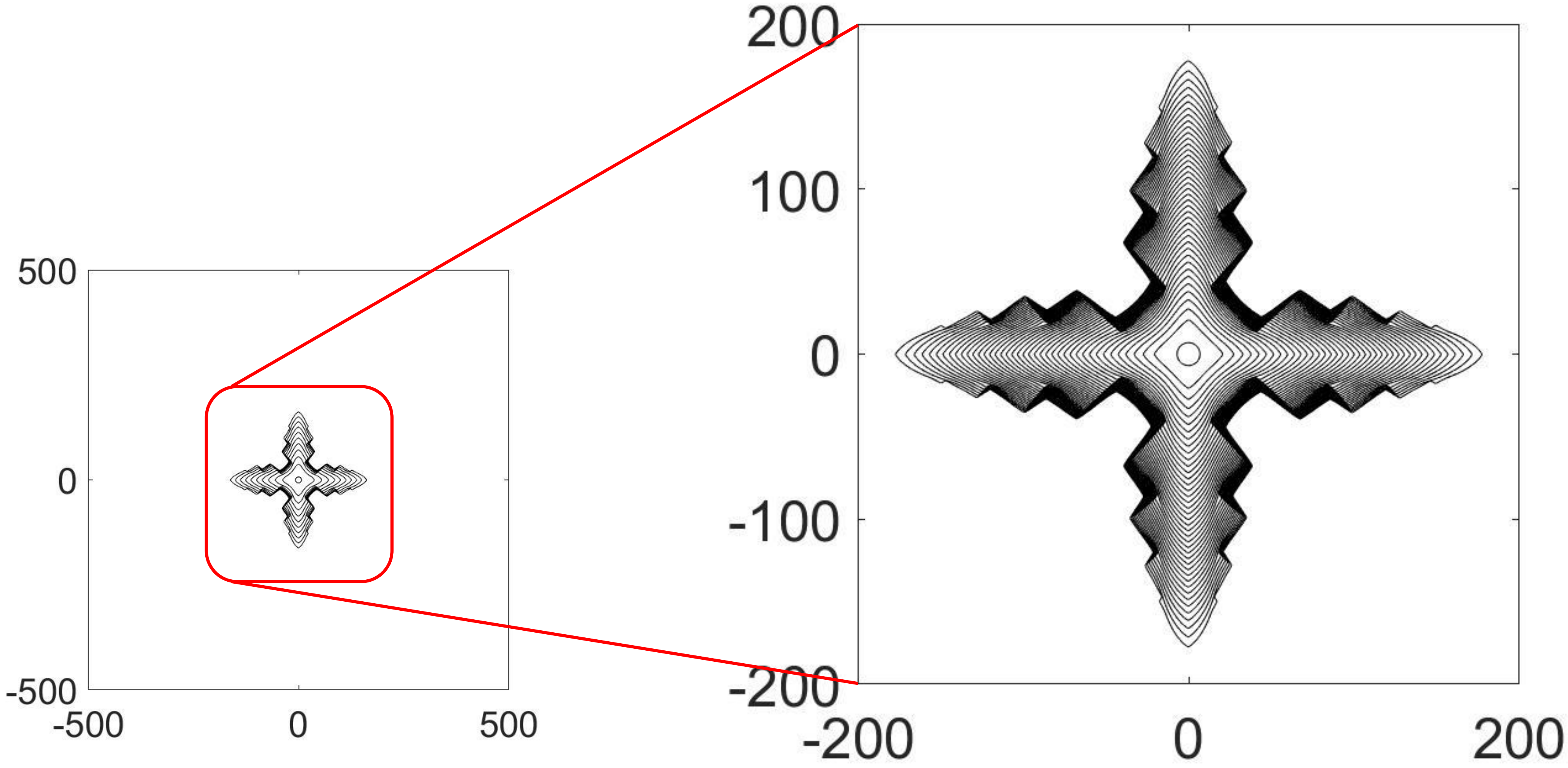
$K = 1.0$



$K = 2.0$

Numerical result

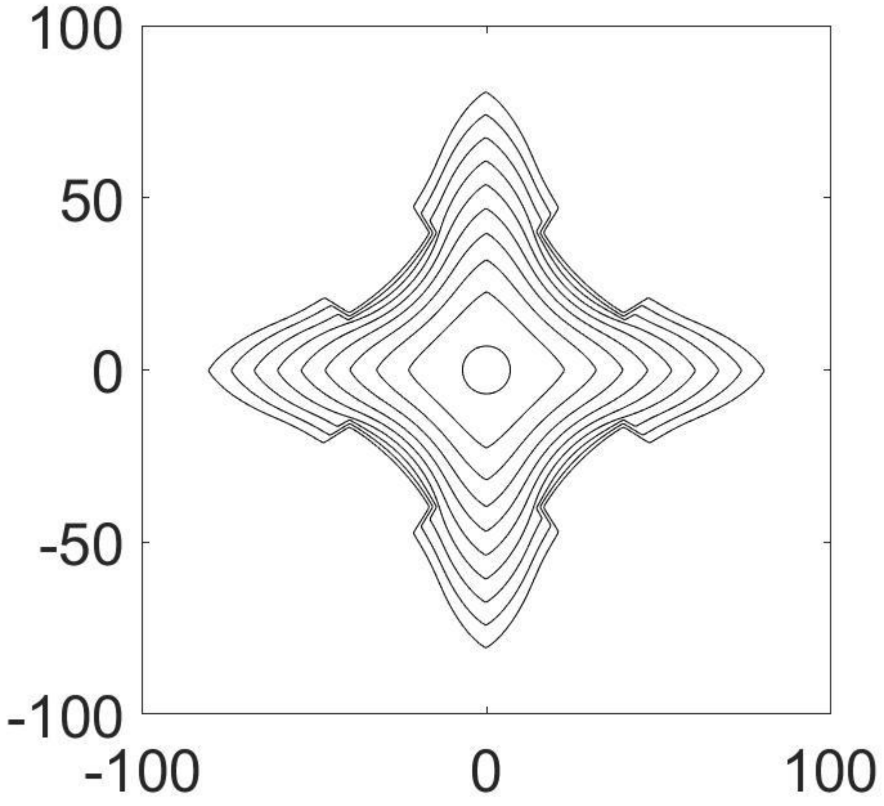
Long time Simulation



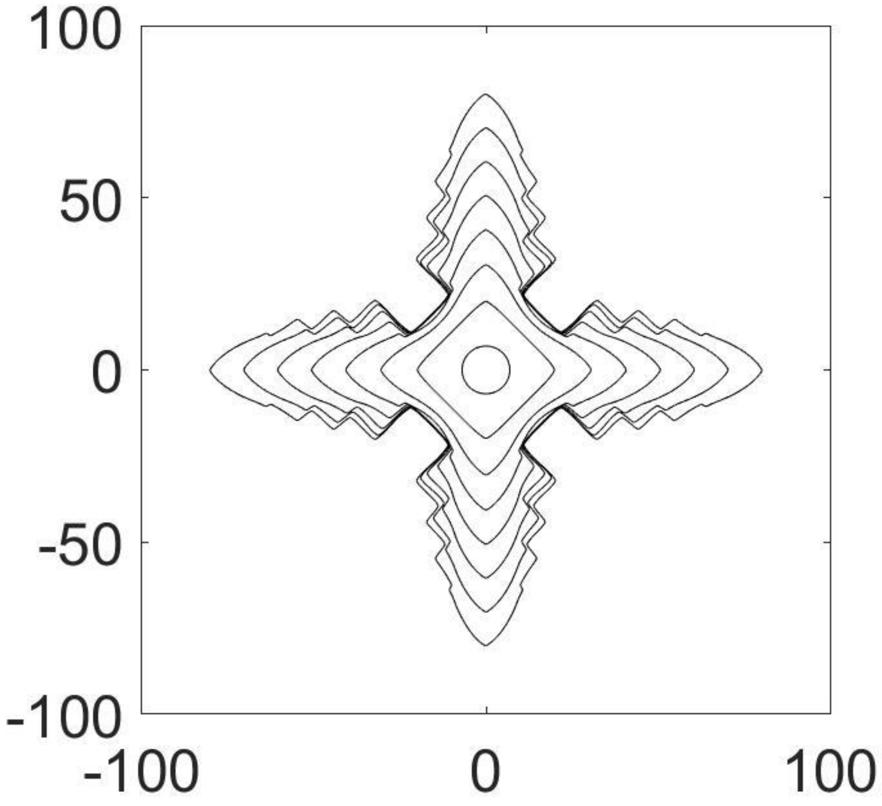
Numerical result

Effect of parameters α and β

$$\xi(U) = \frac{\alpha}{\pi}\arctan(\beta(U_e - U))$$



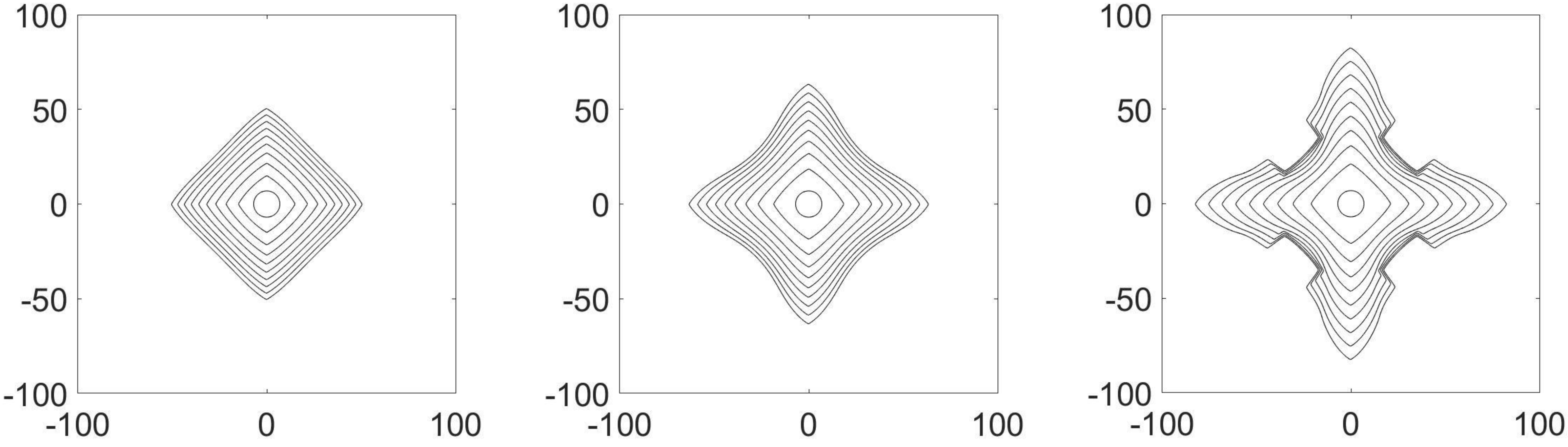
$\alpha = 0.9, \beta = 5$



$\alpha = 0.9, \beta = 10$

Numerical result

Effect of parameters α and β

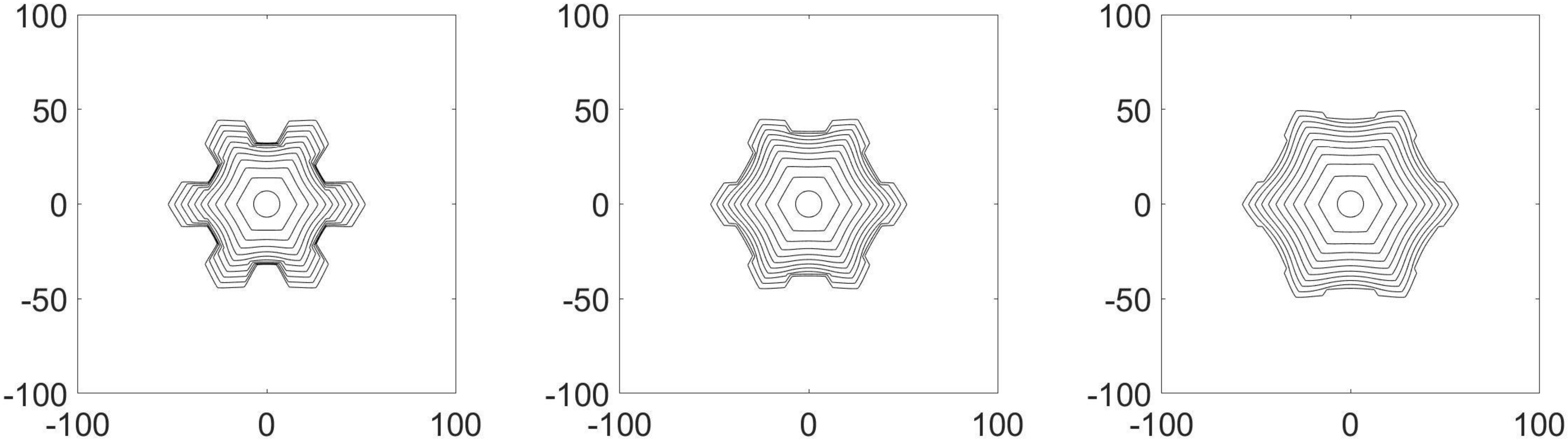


$\alpha = 0.45, \beta = 5$ $\alpha = 0.45, \beta = 10$ $\alpha = 0.45, \beta = 20$

Numerical result

Effect of parameters D

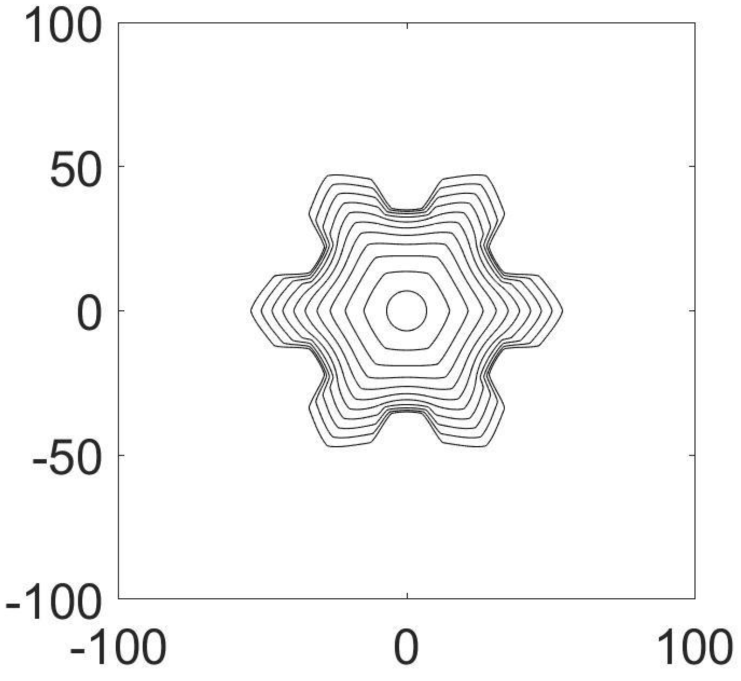
$$\frac{\partial U}{\partial t} = D\nabla^2 U + K\frac{\partial \phi}{\partial t}$$



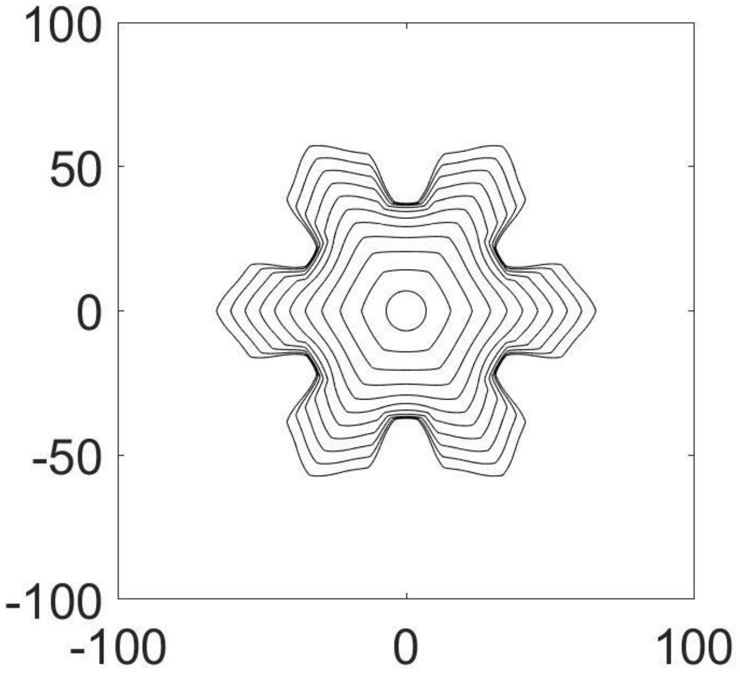
$D = 1.5$ $D = 2$ $D = 3$

Numerical result

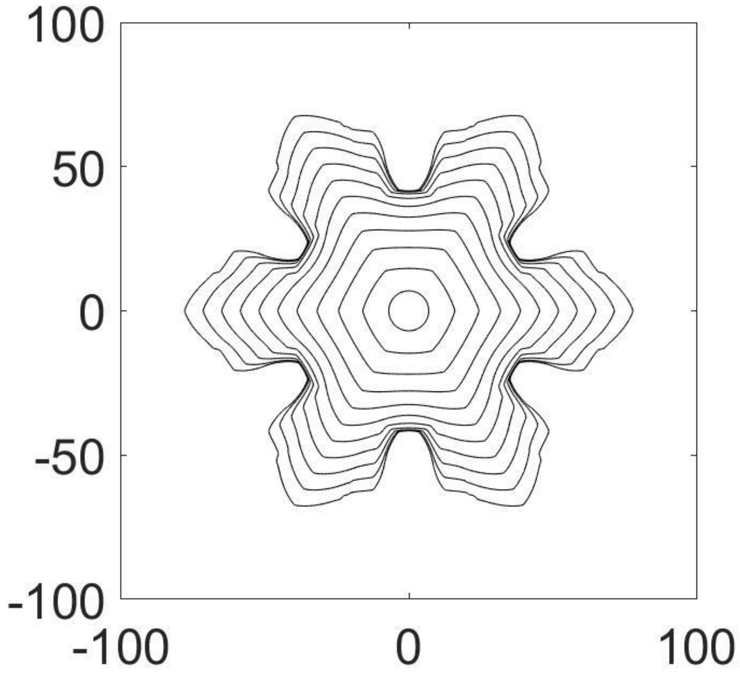
Effect of parameters Δ



$\Delta = -0.55$



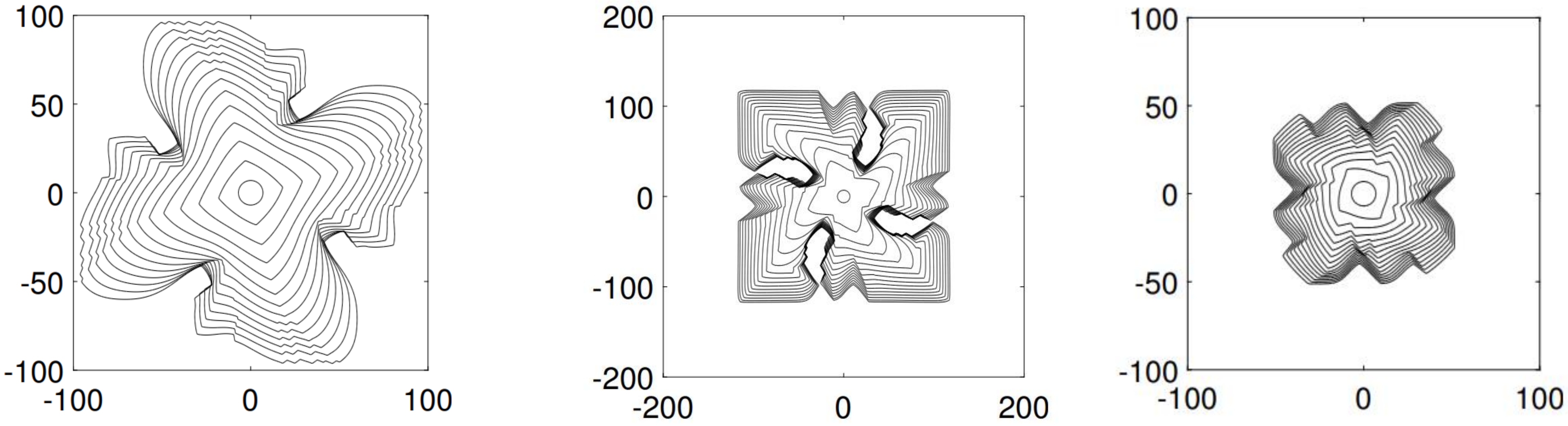
$\Delta = -0.60$



$\Delta = -0.65$

Numerical result

Oriented shape



Conclusions

- We explored the numerical solution for the established crystal growth model and controllable dendrite growth model.

- The presented scheme is effective to simulate complex model for long time simulation, due to it is simple to apply and does not use complicate adaptive data structure.

- Finally, we present richer and various shapes of dendrite by adjusting symmetric breaking patterns with many parameters.

감사합니다