Discretisation and numerical tests of a diffuse–interface model with Ehrlich–Schwoebel barrier

Felix Otto[1,2], Patrick Penzler[1,2], Tobias Rump[1,2]

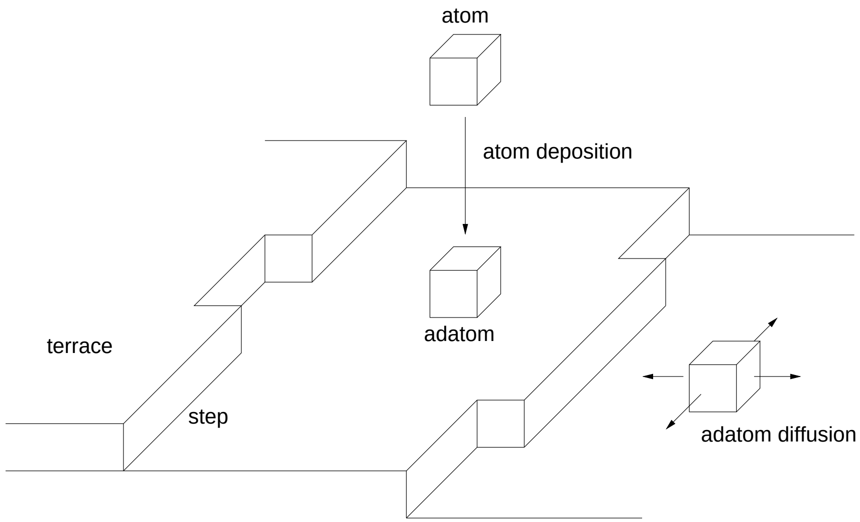
Based on joint work with Andreas Rätz[2], Axel Voigt[2]

[1] University of Bonn,

[2] caesar Bonn

Epitaxial growth

(i.e. layer–by–layer growth of a crystalline thin film on a substrate)



Microscopic processes:

- deposition of atoms on the terraces
- diffusion of adatoms on the terraces
- attachment of adatoms to steps
- detachment of adatoms from steps

Step flow model: Non–dimensionalised BCF model[1)]

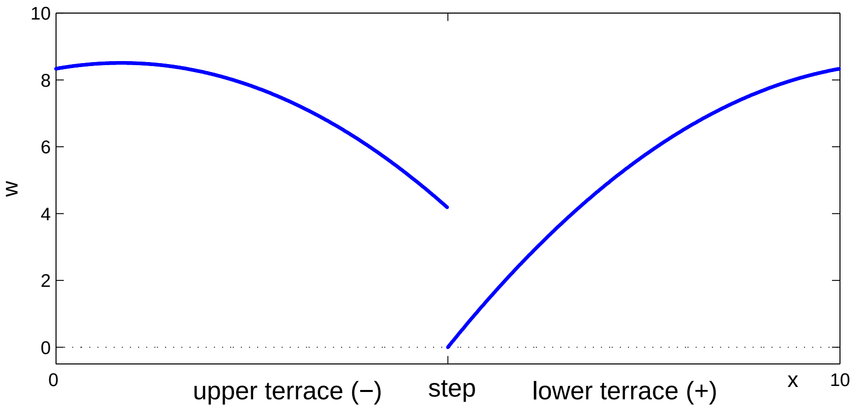
for excess adatom density $w = \rho - \rho^*$:

$$-\Delta w = f \qquad \text{on terraces}$$

$$V = \frac{\partial w^+}{\partial \nu} - \frac{\partial w^-}{\partial \nu} \qquad \text{at steps}$$

$$\begin{pmatrix} 0 & 0 \\ 0 & \zeta \end{pmatrix} \begin{pmatrix} \frac{\partial w^+}{\partial \nu} \\ \frac{\partial w^-}{\partial \nu} \end{pmatrix} = \begin{pmatrix} w^+ - \kappa \\ -(w^- - \kappa) \end{pmatrix} \qquad \text{at steps}$$

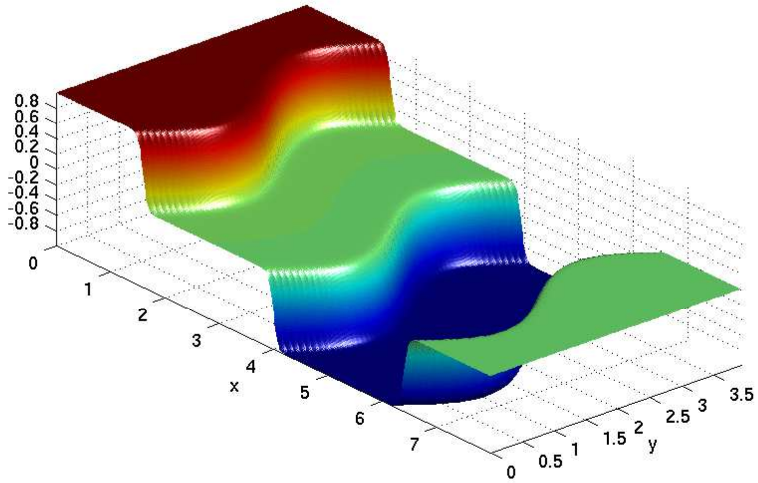
- infinite attachment rate at step up
- finite attachment rate $\sim 1/\zeta$ at step down



1) *[Burton, Cabrera, Frank; 1951]*

Diffuse–interface model

[OPRRV; Nonlin. 17(2), 2004]



- Cahn–Hilliard–type equation

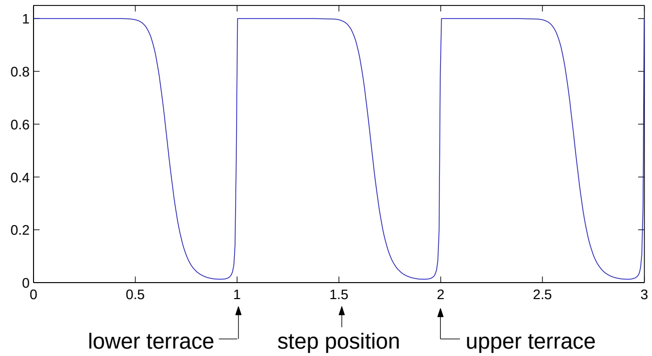
$$\begin{aligned} \partial_t \phi + \nabla \cdot J &= f \\ J &= -M(\phi)\nabla w \\ w &= -\varepsilon\Delta\phi + \varepsilon^{-1}G'(\phi) \end{aligned}$$

- asymmetric mobility function

$$M(\phi) = \frac{1}{1+\varepsilon^{-1}\zeta(\phi)},$$

$$\zeta(\phi) = \zeta_{(p+4)(p+5)}\phi^{p+2}(1-\phi)^2, \quad p \gg 1$$

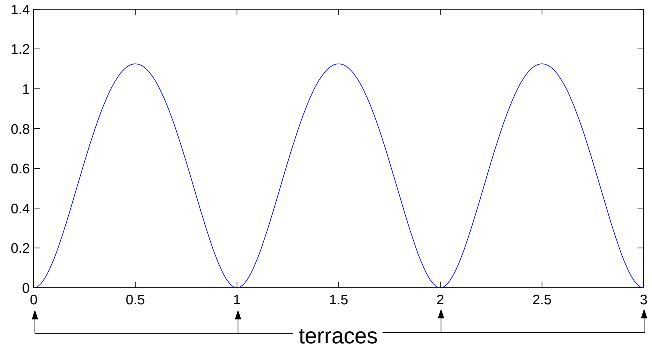
to model the Ehrlich–Schwoebel barrier.



- multiwell–potential

$$G(\phi) = 18\phi^2(1-\phi)^2$$

periodically extended.



Asymptotic analysis for $\varepsilon \to 0$ yields p–BCF model

$$-\Delta w = f \qquad \text{on terraces,}$$

$$V = \frac{\partial w^+}{\partial \nu} - \frac{\partial w^-}{\partial \nu} \qquad \text{at steps,}$$

$$\begin{pmatrix} \zeta^+ & \zeta^m \\ \zeta^m & \zeta^- \end{pmatrix} \begin{pmatrix} \frac{\partial w^+}{\partial \nu} \\ \frac{\partial w^-}{\partial \nu} \end{pmatrix} = \begin{pmatrix} w^+ - \kappa \\ -(w^- - \kappa) \end{pmatrix} \qquad \text{at steps,}$$

with

$$\zeta^- = \zeta, \qquad \zeta^+ = \mathcal{O}(p^{-2}) \quad \text{and} \quad \zeta^m = \mathcal{O}(p^{-1}).$$

➠ numerical parameter ε and p:

$$\boxed{\text{diffuse–interface} \xrightarrow{\varepsilon \downarrow 0} p\text{–BCF} \xrightarrow{p \uparrow \infty} \text{BCF}}$$

[OPRRV; 2004]

Time discretisation:

Model equation

$$\partial_t \phi + A\phi = 0, \qquad A \text{ positive semi–definite}$$

	order	theoretical Δt–restriction	amplification factor for (spectrum of A) $\to \infty$
Euler explicit	first order	high	unbounded
Euler implicit	first order	none	goes to 0
Crank–Nicholson	second order	none	goes to 1
θ–scheme[1)]	second order	none	uniformly bounded away from 1

$\Longrightarrow$ θ–scheme is right choice

[1)] *[Bristeau, Glowinski, Périaux; 1987], [Weikard; 2002]*

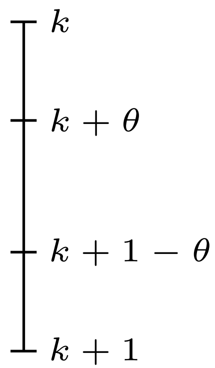
Time discretisation: θ–scheme

Write diffuse–interface model as

$$\partial_t \phi + F(\phi) = 0, \qquad F(\phi) = \nabla \cdot [-M(\phi)\nabla(-\varepsilon\Delta\phi + \varepsilon^{-1}G'(\phi))] - f.$$

The θ–scheme is given by

$$\begin{aligned}
\frac{\phi^{k+\theta} - \phi^k}{\theta\Delta t} &= -(\alpha F(\phi^{k+\theta}) + \beta F(\phi^k)) \\
\frac{\phi^{k+1-\theta} - \phi^{k+\theta}}{(1-2\theta)\Delta t} &= -(\beta F(\phi^{k+1-\theta}) + \alpha F(\phi^{k+\theta})) \\
\frac{\phi^{k+1} - \phi^k}{\theta\Delta t} &= -(\alpha F(\phi^{k+1}) + \beta F(\phi^{k+1-\theta}))
\end{aligned}$$



with

$$\theta = 1 - 1/\sqrt{2}, \quad \alpha = 2 - \sqrt{2}, \quad \beta = \sqrt{2} - 1.$$

➠ Newton's method in each intermediate step (two Newton steps)

Symmetrisation of time discrete problem via flux

One Newton step in semi–implicit Euler scheme:

$$\left(\frac{1}{\tau}\mathrm{id} + \nabla\cdot M(\phi^k)\nabla(\varepsilon\Delta - \varepsilon^{-1}G''(\phi^k))\right)(\phi^{k+1} - \phi^k) + F(\phi^k) = 0$$

$\rightsquigarrow$ non-symmetric fourth order problem due to $M(\phi^k)$

How do we get a symmetric problem?

Rewrite as

$$\boxed{\tfrac{1}{\tau}(\phi^{k+1} - \phi^k) + \nabla\cdot J^{k+1} = 0}$$

$$J^{k+1} := M(\phi^k)\nabla\left[\varepsilon\Delta\phi^{k+1} - \varepsilon^{-1}(G'(\phi^k) + G''(\phi^k)(\phi^{k+1} - \phi^k))\right]$$

J^{k+1} can be characterised as

$$\left(\frac{1}{M(\phi^k)}\text{id} + \tau\varepsilon(\nabla\nabla\cdot)^2 - \tau\varepsilon^{-1}\nabla G''(\phi^k)\nabla\cdot\right) J^{k+1} = -\nabla w^k.$$

$\leadsto$ symmetric fourth order problem

$\leadsto$ discrete (and regularised) version of

$$\tfrac{1}{M(\phi)} J = -\nabla w.$$

Advantages:

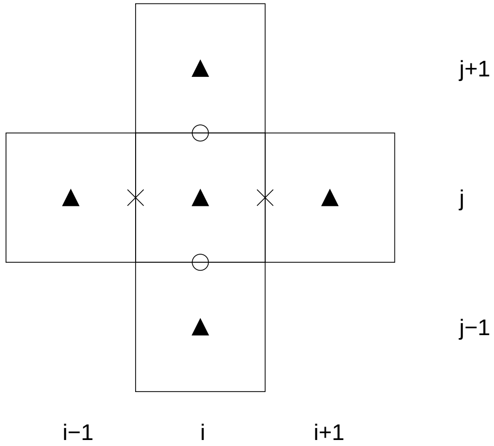
- periodic boundary conditions
- Solver: conjugate gradient method
  Preconditioner: $M \mapsto M(\text{terrace})$ and $G'' \mapsto G''(\text{terrace})$
  $\leadsto$ linear operator with constant coefficients
  $\leadsto$ easy to invert, e.g. by FFT on equidistant grid

Spatial discretisation

Goal: discrete volume conservation
preserve symmetric structure

Finite volume scheme in 2-d:

$$
\begin{aligned}
(\nabla^h \cdot J)(i,j) &= \tfrac{1}{h}(J_x(i+\tfrac{1}{2},j) - J_x(i-\tfrac{1}{2},j) \\
&\quad + J_y(i,j+\tfrac{1}{2}) - J_y(i,j-\tfrac{1}{2})) \\
(\nabla^h \phi)_x(i+\tfrac{1}{2},j) &= \tfrac{1}{h}(\phi(i+1,j) - \phi(i,j)) \\
(\nabla^h \phi)_y(i,j+\tfrac{1}{2}) &= \tfrac{1}{h}(\phi(i,j+1) - \phi(i,j))
\end{aligned}
$$



Discrete mobility $M = (M_x, M_y)$: average of friction ζ

$$
\begin{aligned}
M_x(i+\tfrac{1}{2},j) &= \frac{1}{1+\frac{1}{2\varepsilon}\left(\zeta(\phi(i+1,j)) + \zeta(\phi(i,j))\right)}, \\
M_y(i,j+\tfrac{1}{2}) &= \frac{1}{1+\frac{1}{2\varepsilon}\left(\zeta(\phi(i,j+1)) + \zeta(\phi(i,j))\right)}.
\end{aligned}
$$

[Gruen, Rumpf; 2000]

Outlook: Finite element formulation

Goal: local refinement at steps

Weak formulation is given by

$$\int_\Omega K \cdot \widetilde{J} = \int_\Omega (\nabla \cdot J^{k+1})(\nabla \cdot \widetilde{J})$$

$$\int_\Omega \frac{1}{M(\phi^k)} J^{k+1} \cdot \widetilde{J} + \int_\Omega \tau\varepsilon(\nabla \cdot K)(\nabla \cdot \widetilde{J}) + \int_\Omega \tau\varepsilon^{-1} G''(\phi^k)(\nabla \cdot J^{k+1})(\nabla \cdot \widetilde{J}) = \int_\Omega w^k (\nabla \cdot \widetilde{J}).$$

$$\Longrightarrow \boxed{\text{Find } \; J^{k+1} \in H(\nabla \cdot, \Omega) := \{J \in (L^2(\Omega))^2 \mid \nabla \cdot J \in L^2(\Omega)\}.}$$

In terms of the mass and stiffness matrices:

$$\left(B_1 + \tau\varepsilon A_0 B_0^{-1} A_0 + \tau\varepsilon^{-1} A_1\right) J^{k+1} = C w^k.$$

Raviart–Thomas element

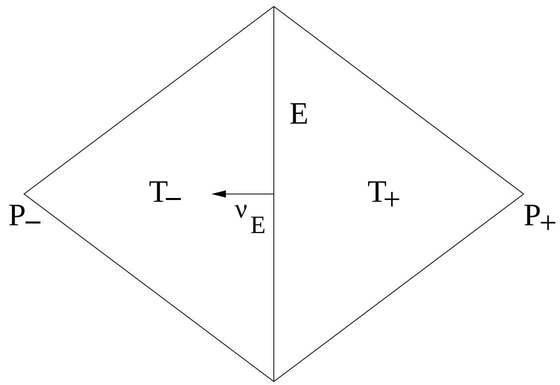
$H(\nabla\cdot,\Omega)$–conforming elements are the Raviart–Thomas elements[1] defined by

$$\mathrm{RT}(\mathcal{T}) := \{J \in (L^2(\Omega))^2 \mid \forall T \in \mathcal{T}\ \exists a_T \in \mathbb{R}^2 \text{ and } b_T \in \mathbb{R} : \forall x \in T\ J(x) = a_T + b_T x \text{ and } \forall \text{ edges } E : [J]_E \cdot \nu_E = 0\}.$$

Construction of basis functions:

Edge–oriented basis element ψ_E is defined by

$$\psi_E(x) := \begin{cases} \pm\frac{|E|}{2|T_\pm|}(x - P_\pm) & \text{for } x \in T_\pm \\ 0 & \text{elsewhere.} \end{cases}$$



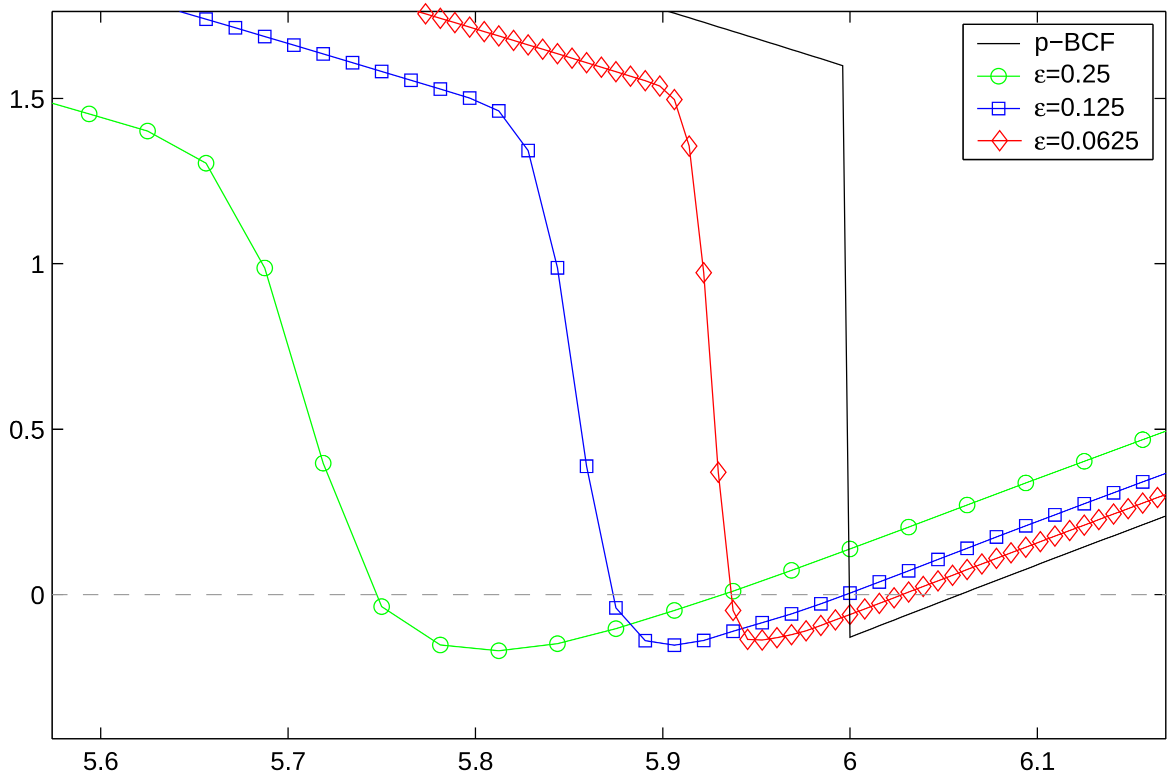
➠ Normal component is constant along an arbitrary edge F: $\psi_E \cdot \nu_F(x) \equiv \delta_{EF}$.

1) *[Raviart, Thomas; 1977]*

Accuracy of the diffuse–interface model

Outer solution

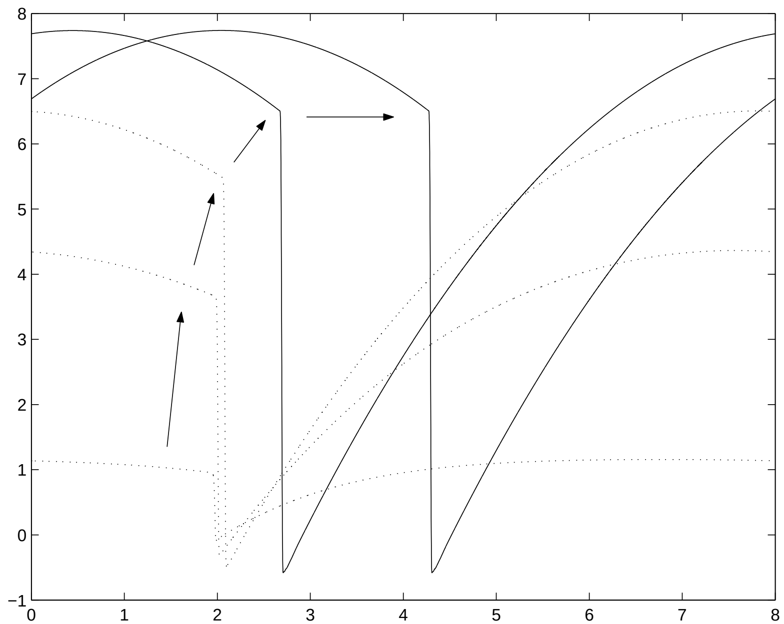
Linear convergence in ε:



Why does the diffuse–interface solution lag behind the BCF-solution?

Initial delay:

diffuse–interface density needs time to reach its travelling wave shape



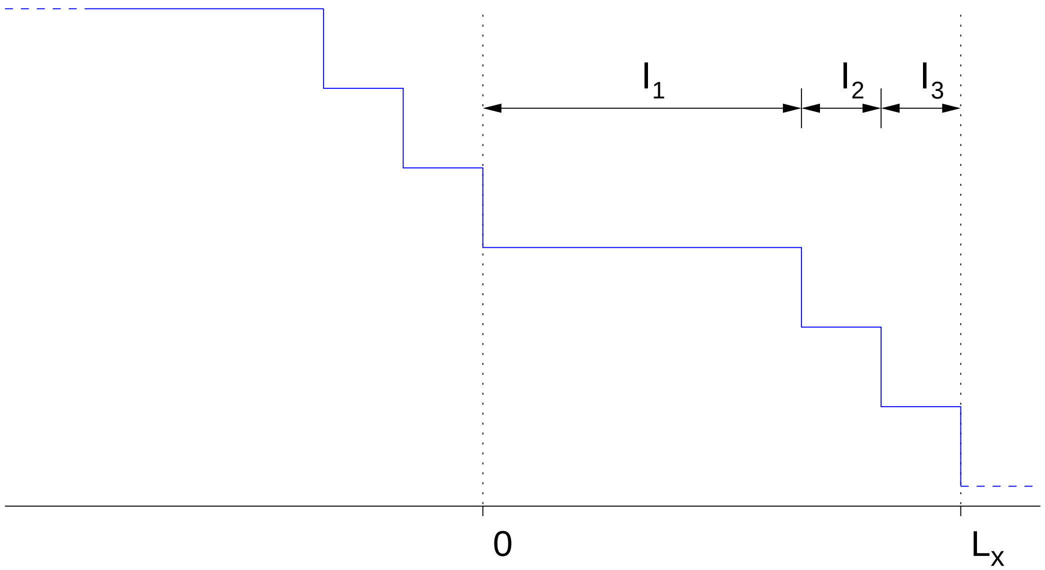
After initial layer, the step has the same speed as the BCF–solution: $V = fL_x$.

($\rightsquigarrow$ discrete volume conservation)

1d effect: step bunching

(i. e. width of a terrace goes to zero)

We consider a periodic *step train* with three terraces:



From BCF model:

ODE for the terrace width l_i is given by

$$\dot{l}_i = f\left(\frac{\zeta l_{i+1} + \frac{1}{2}l_{i+1}^2}{\zeta + l_{i+1}} - \frac{\zeta l_i}{\zeta + l_i} - \frac{\frac{1}{2}l_{i-1}^2}{\zeta + l_{i-1}}\right).$$

$\zeta = 0$

$$\dot{l}_i = \frac{f}{2}\left(l_{i+1} - l_{i-1}\right)$$

$\Rightarrow$ step bunching possible

$\zeta = +\infty$

$$\dot{l}_i = f(l_{i+1} - l_i)$$

$\Rightarrow$ no step bunching

$\Longrightarrow$ Ehrlich–Schwoebel barrier counteracts step bunching under growth.

The ODE system for 3 periodic terraces

$$(\dot{l_1}, \dot{l_2}, \dot{l_3}) = L(l_1, l_2, l_3)$$

preserves $l_1 + l_2 + l_3$ and has equidistant steps as stationary point

$$(l_1, l_2, l_3) = (l^*, l^*, l^*).$$

The eigenvalues of $DL(l^*, l^*, l^*)$ are

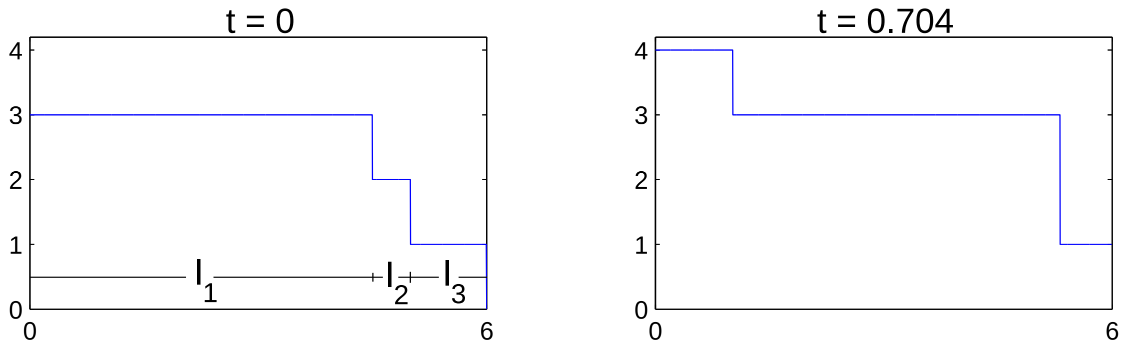
$$\begin{aligned} \lambda_1 &= 0 \qquad \text{(conservation of } l_1 + l_2 + l_3 = 3l^*) \\ \lambda_{2/3} &= -\frac{3f\zeta^2}{2(\zeta + 3l^*)^2} \pm i\frac{\sqrt{3}f}{2} \end{aligned}$$

$\zeta > 0 \quad \Rightarrow \quad \text{Re}\,(\lambda_{2/3}) < 0 \quad \Rightarrow \quad$ stable

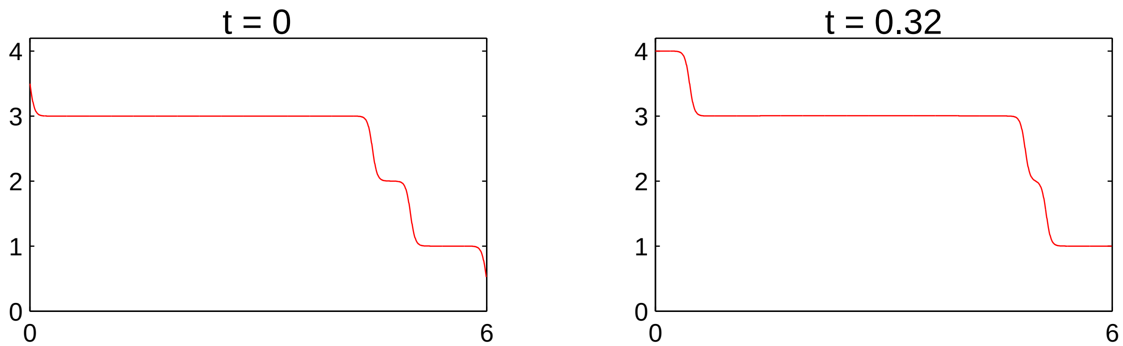
$\Longrightarrow$ Ehrlich–Schwoebel barrier favours equidistant steps under growth.

Vanishing of terraces for $\zeta = 0$:

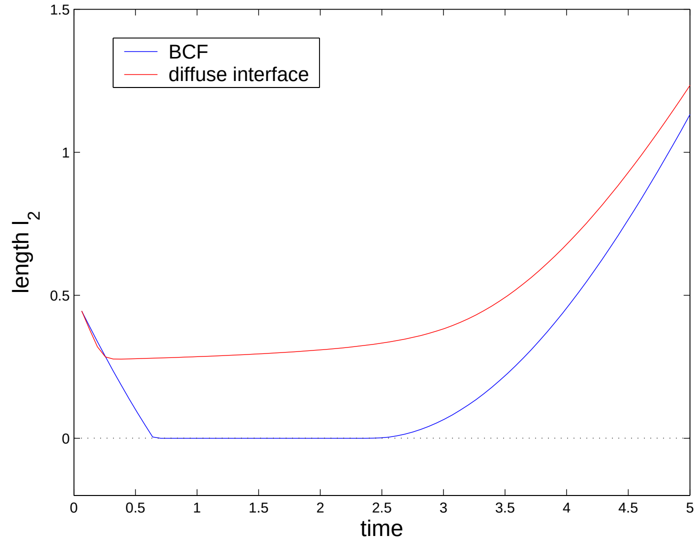
BCF model



diffuse–interface model



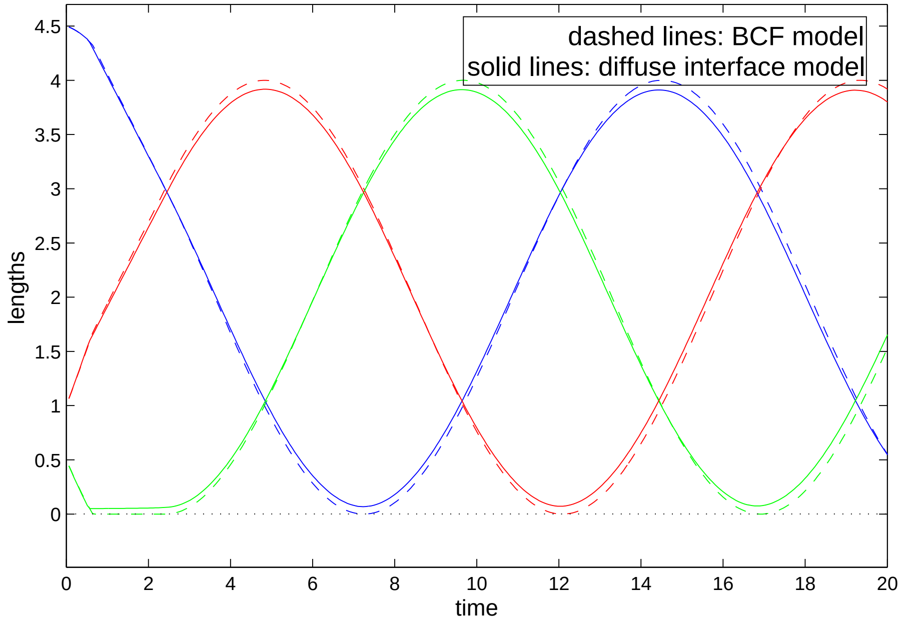
Comparison



Comparison with the measured lengths of our **simulation**:

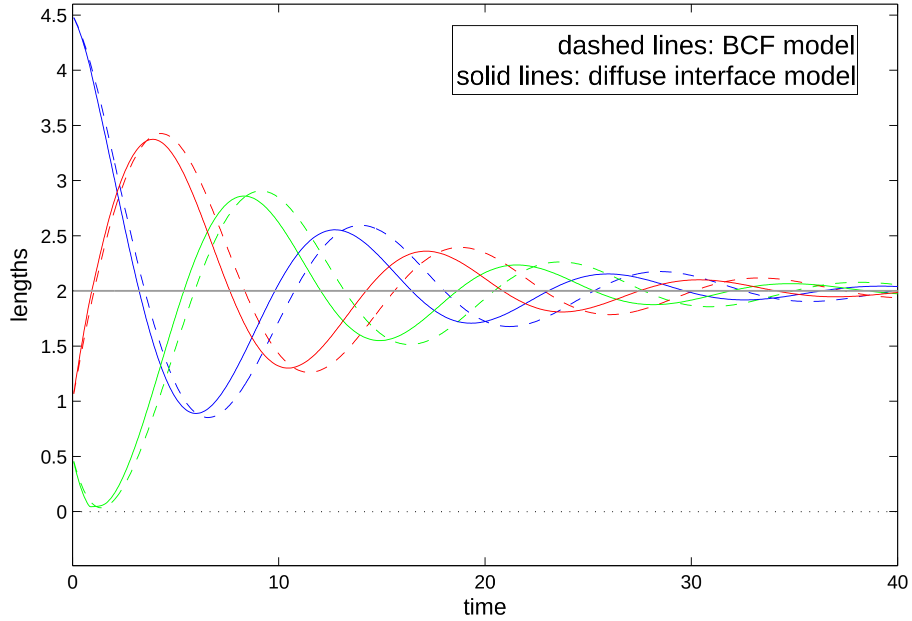
$\zeta = 0$:

time periodic
step bunching



$\zeta = 1$:

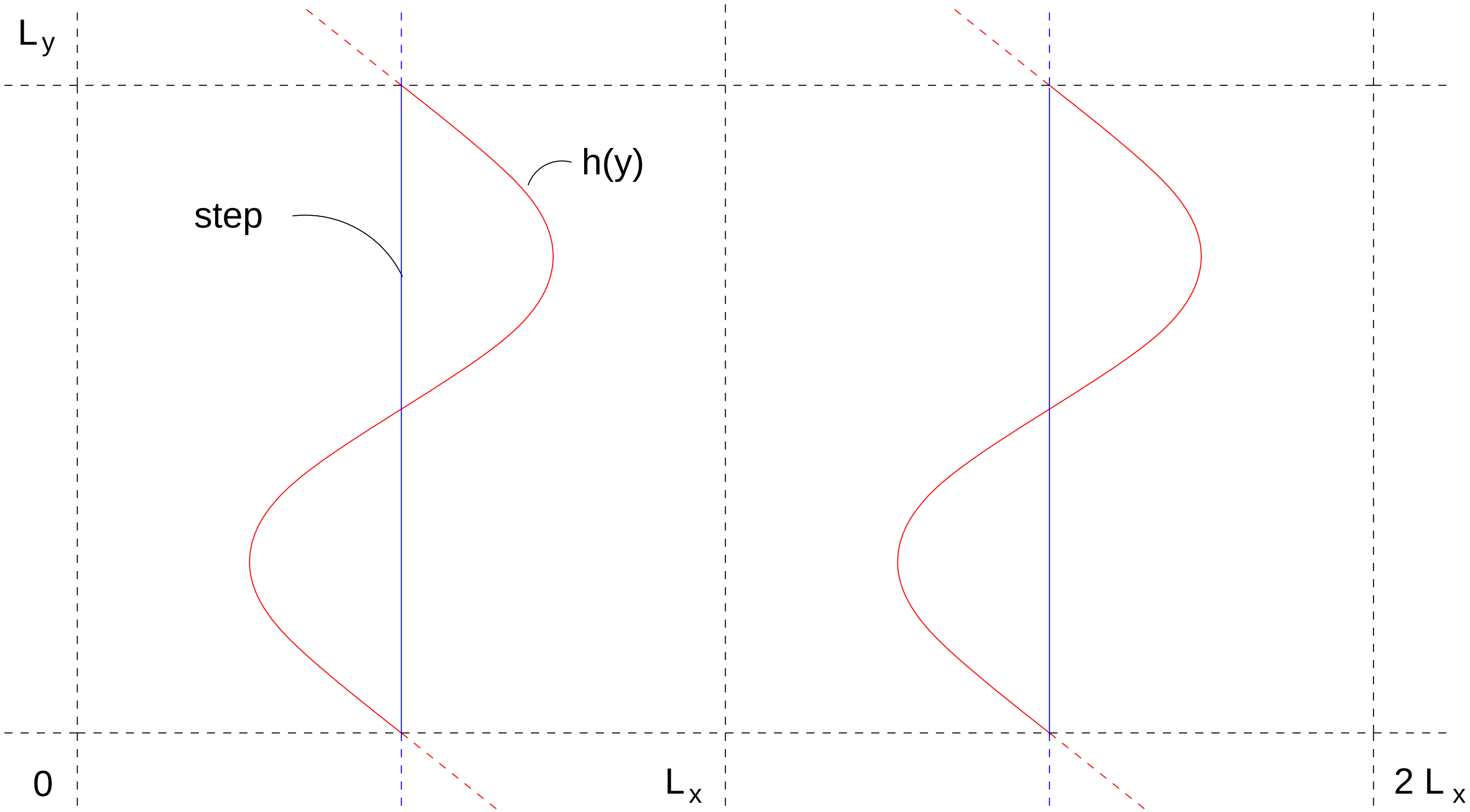
trend towards
equidistant steps



2d effect: step meandering

(i. e. steps do not stay straight)

Consider an equidistant step train, each step perturbed by the same function $h(y)$ of order ε.



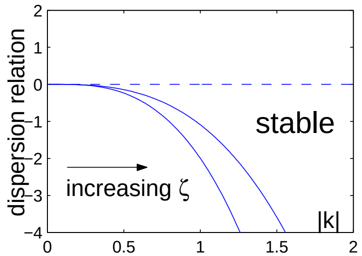
How does this perturbation develop in time?

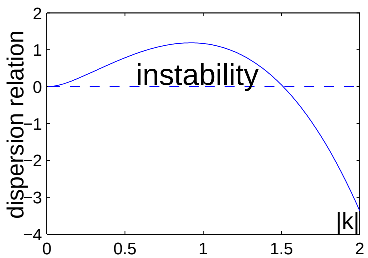
Linear stability analysis for the BCF model yields for each wave vector k

$$\frac{d}{dt}\hat{h}_k(t) = \omega(|k|)\hat{h}_k(t),$$

with the dispersion relation ω depending on f, ζ, L_x.

$\zeta = 0$ or $f = 0$		$\zeta > 0$ and $f > 0$
$\omega(\lvert k\rvert) < 0$ for all $\lvert k\rvert$	$\longleftrightarrow$	$\omega(\lvert k\rvert) > 0$ for small $\lvert k\rvert$
$\Rightarrow$ stability		$\Rightarrow$ instability occurs





$\Longrightarrow$ Ehrlich–Schwoebel barrier favours step meandering under growth.

How do we get the dispersion relation of the diffuse–interface model?

1. discrete Fourier transformation $(\hat{\phi}_{ij}(t_n))_{ij}$ of ϕ in each time step t_n
2. vector $H(t_n)$ with components

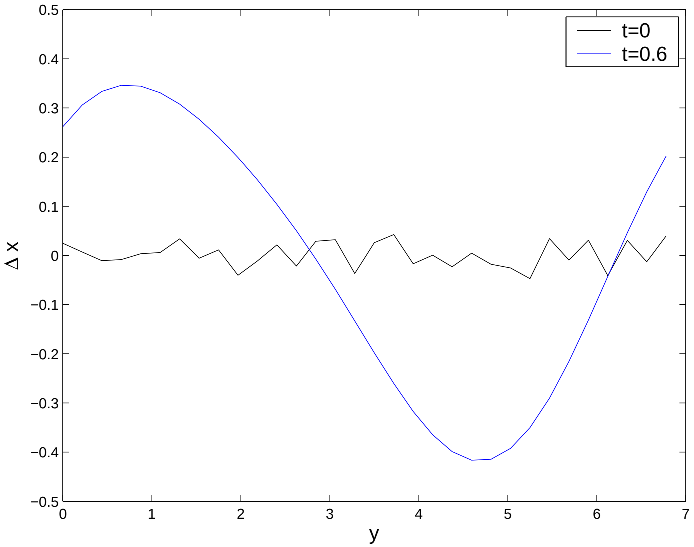
$$H_j(t_n) = \sum_i |\hat{\phi}_{ij}(t_n)|^2.$$

3. growth rate over time interval Δt:

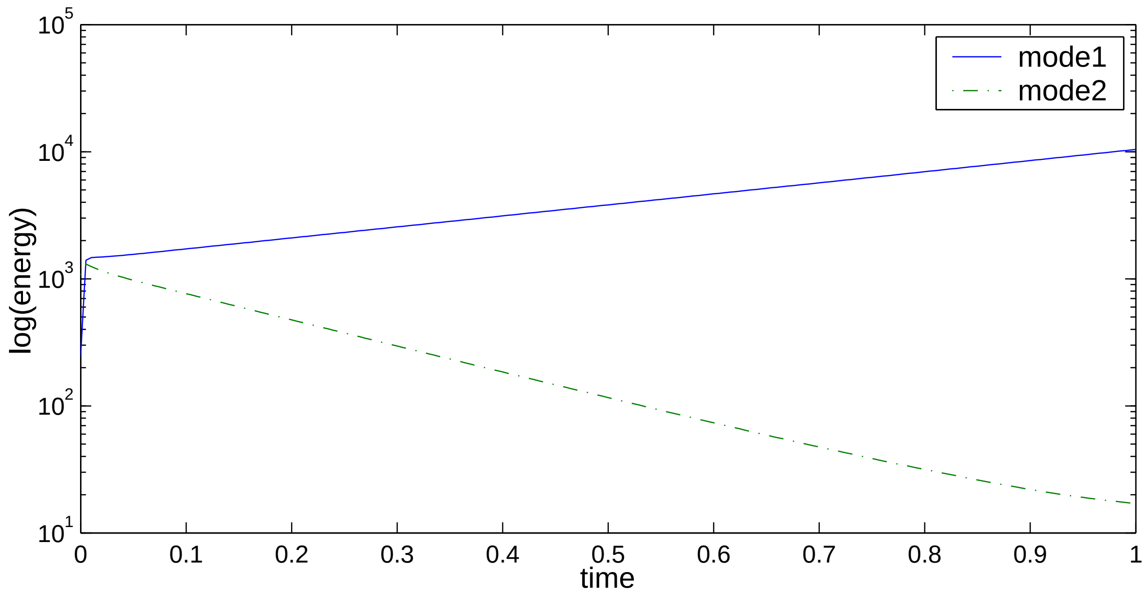
$$\omega_{t_n}(|k|) = \frac{\log\left(\frac{H_{|k|}(t_n)}{H_{|k|}(t_n-\Delta t)}\right)}{2\Delta t}$$

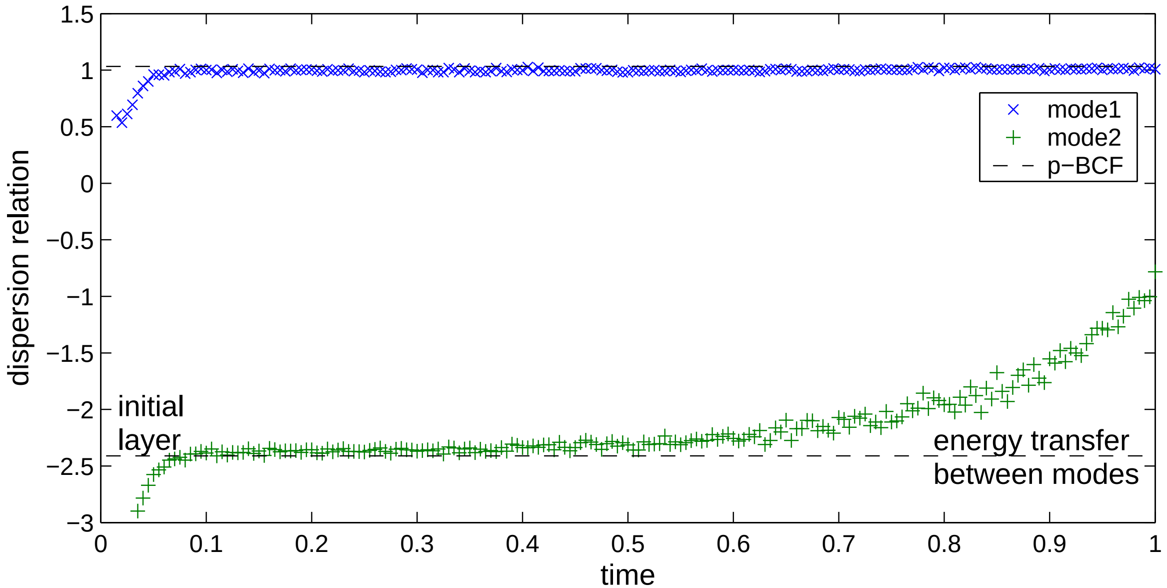
$h(y)$= "white noise"

most unstable mode: 1



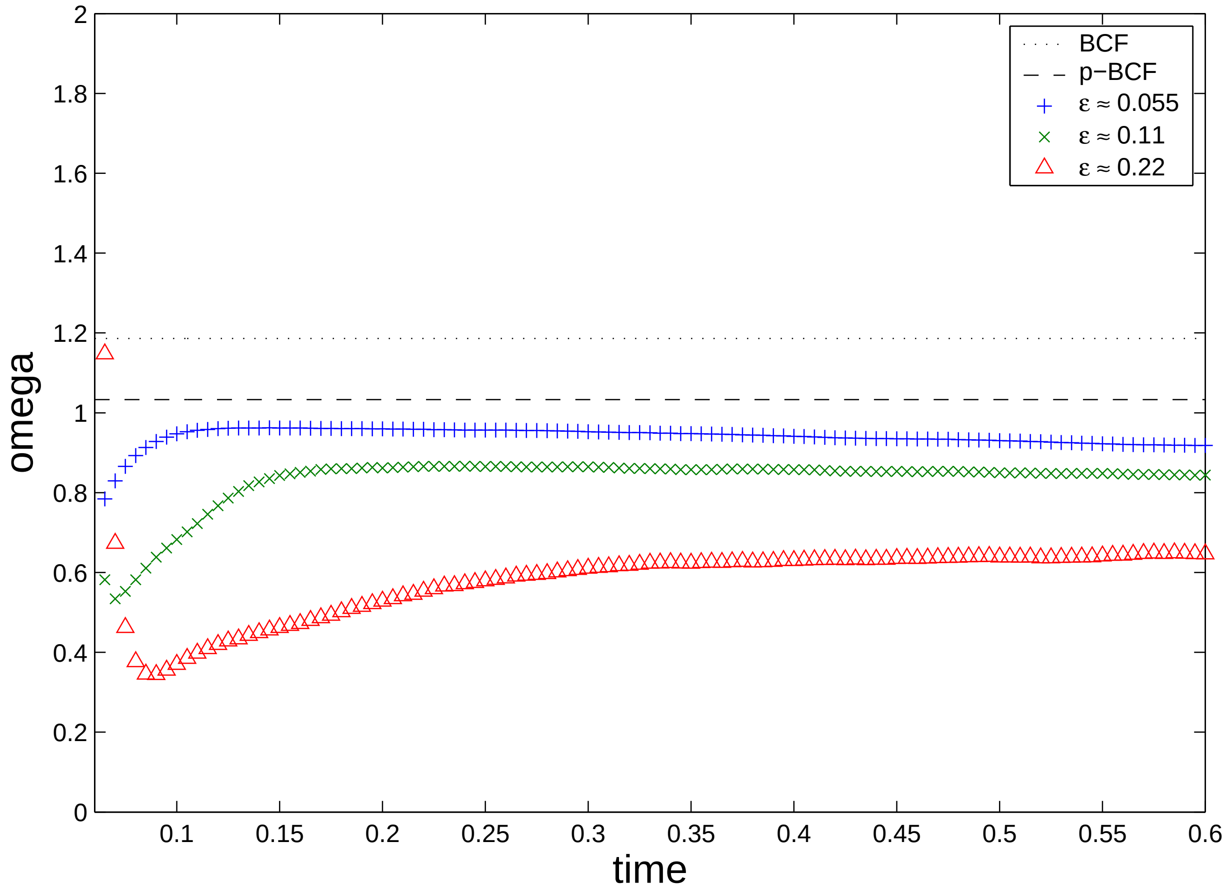
step for $t = 0$ and $t = 0.6$





$h(y) = \sin(2\pi/L_y)$, most unstable mode: 1

Linear convergence in ε:



$(f = 0.5, \zeta = 10, L_x = 7, n_x = 2048, n_y = 32)$

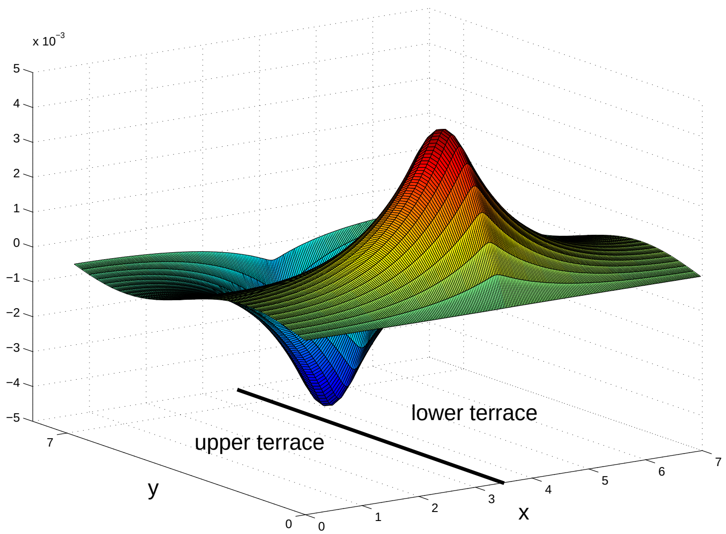
Boundary conditions

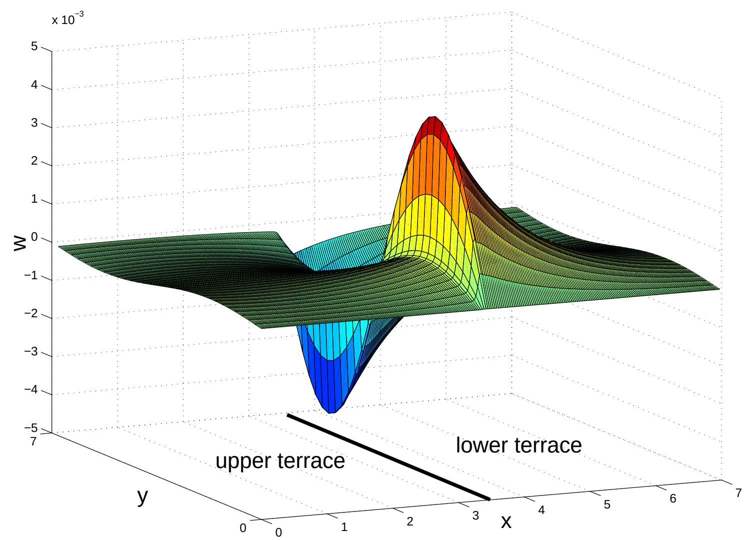
Recall that the BCF–w satisfies the boundary conditions

$$\begin{aligned} w^+ - \kappa &= 0 \qquad \text{at step up} \\ \zeta \frac{\partial w^-}{\partial \nu} + (w^- - \kappa) &= 0 \qquad \text{at step down.} \end{aligned}$$

So for the two extreme cases $\zeta = 0$ and $\zeta \gg 1$ we have

$$w^- = \kappa \qquad \text{and} \qquad \frac{\partial w^-}{\partial \nu} \approx 0.$$





With deposition ($f = 0.1$) and ES barrier ($\zeta = 5$):

- concave shape due to deposition
- jump at the step due to the ES barrier
- variation in jump height due to curvature