Self-organized regular superconducting patterns in thin films

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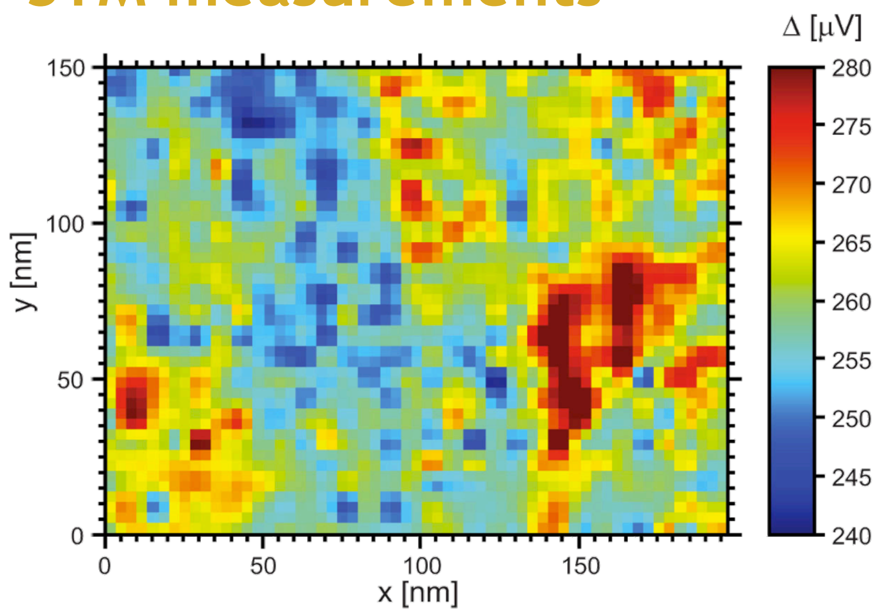
motivation

STM measurements



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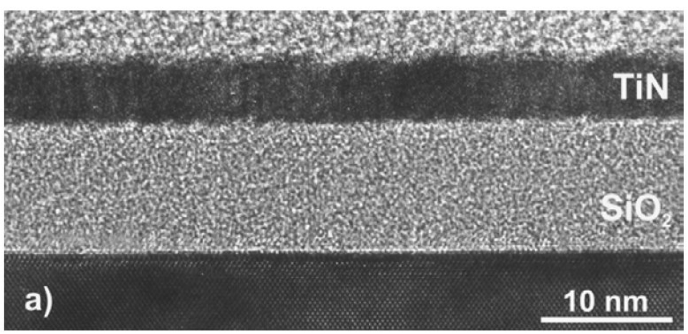
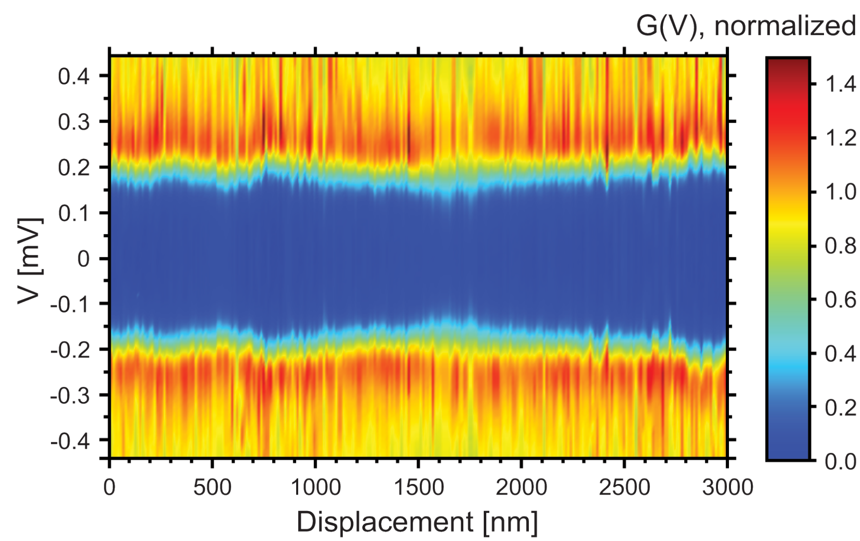


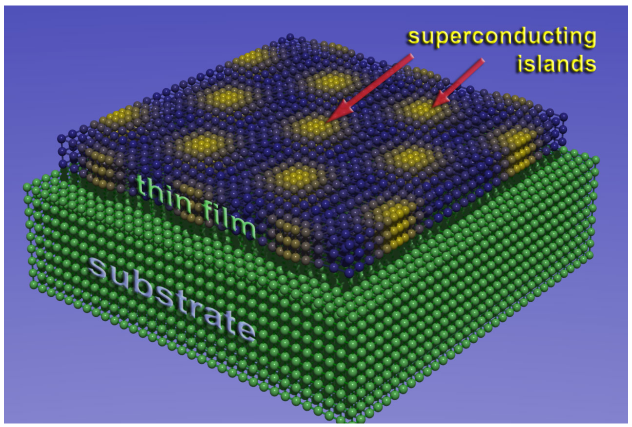
spatial fluctuations of Δ in TiN

- 3.3nm resolution, T=300mK
- sample TiN1: T_c=1.3K , 3.6nm thick
- sample TiN2: T_c=1.0K , 5.0nm thick



the question

How can an inhomogeneous superconducting state form?



One possibility: Effect of local pressure changes on T_c in thin films.

$$k_B T_c = 1.13 \hbar \omega_D \exp\left(-\frac{1}{\nu_0 V}\right)$$

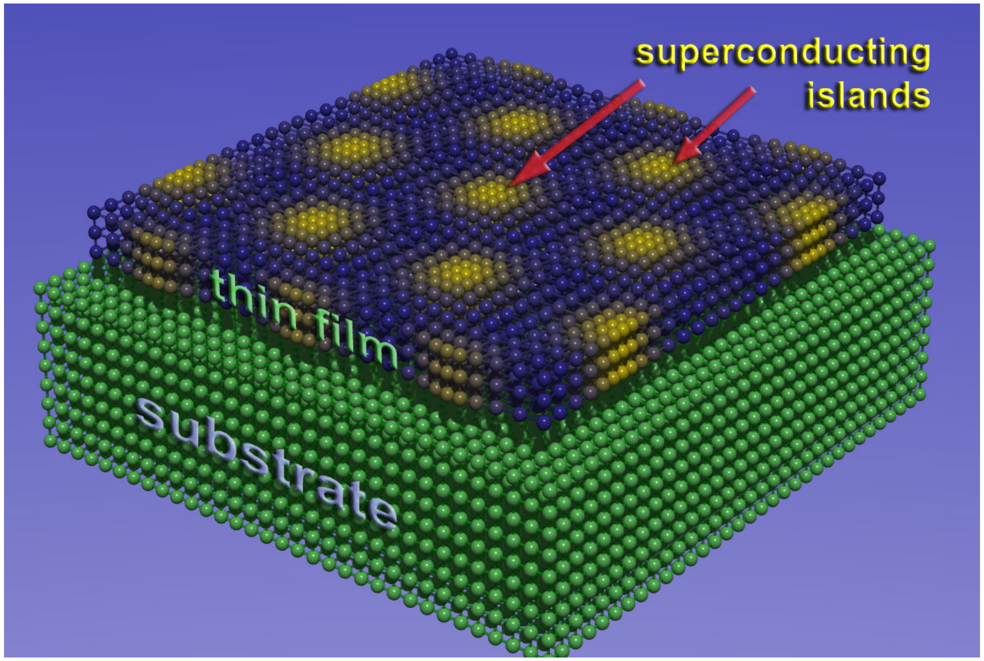
Bardeen-Cooper-Schrieffer (1957)

outline

- model
- Ginzburg-Landau equation
- Elastic equations
- **Time evolution of the “island” state**
- **Linear stability**
- **Phase diagram**
- outlook
- conclusion

model

Thin film coupled to a massive substrate with different elastic properties.



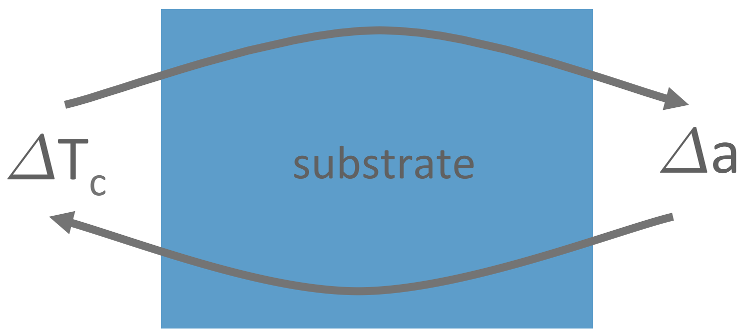
For example:
TiN on silicon oxide

→ **What is the influence of a rigid substrate on a soft film?**

positive feedback

Classic superconductivity rests on phonon-mediated effective attraction between electrons resulting in the formation of Cooper pairs [BCS].

Cooper condensate → change of elastic properties, i.e., the phonon spectrum



→ positive feedback for change in T_c and the (average) lattice constant a due to the substrate → *effective long-range interaction*

construction of the model

Two ingredients:

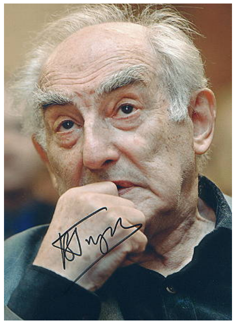
1. ***Ginzburg-Landau equation:***
   - ➔ ***free energy of the superconductor***
   - ➔ ***spatial dependence of the superconducting order parameter***
2. *Linear elasticity*

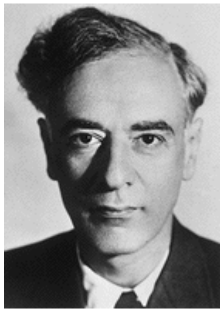
Ginzburg-Landau theory: an introduction

Superconducting order parameter

Microscopic origin: Coherent state of Cooper pairs

$$\begin{aligned} \psi(\mathbf{r}) &= |\psi| e^{i\varphi(\mathbf{r})} \\ n_s &= |\psi|^2 \\ \mathbf{p} &= \hbar \nabla \varphi \end{aligned}$$





Ginzburg and Landau, 1950

free energy of a superconductor *(expansion in ψ and its gradients)*

$$\mathcal{F}_{\mathrm{GL}} = \frac{1}{2} \int d^d x \left\{ \beta \left(\frac{\alpha}{\beta} + |\psi|^2 \right)^2 + \frac{\hbar^2}{m} \left| \left(i\nabla - \frac{2\pi}{\phi_0} \mathbf{A} \right) \psi \right|^2 + \frac{1}{4\pi} (\nabla \times \mathbf{A} - \mathbf{H})^2 \right\}$$

$\alpha(T) \sim (T_c - T)$

Correlation or coherence length: $\xi^2(T) = \frac{\hbar^2}{2m|\alpha(T)|}$ [$2\alpha(T) + \hbar^2 \xi^{-2}/m = 0$]

linear term diffusion term

→ Goal: Find solution showing the nucleation of superconductivity from the normal state

Ginzburg-Landau theory

kinetic equation: $$\frac{\partial \psi}{\partial t} = \frac{\delta \mathcal{F}_{GL}}{\delta \psi^\star}$$

steady state solution (no diffusion):

$$0 = \alpha\psi - \beta|\psi|^2\psi \quad \Rightarrow |\psi|^2 = \alpha/\beta \propto (T_c - T)$$

➔ the typical solution is a homogeneous superconducting state

"supercurrent": $$\mathbf{j} \propto \Im\left[\psi^\star \left(\nabla - \imath\mathbf{A}\right)\psi\right]$$

- *complex Ginzburg-Landau equation is one of the most-studied nonlinear equations in the physics*
- *describes vast variety of phenomena: from nonlinear waves to second-order phase transitions, from superconductivity, superfluidity, and Bose-Einstein condensation to liquid crystals and strings in field theory*
- often even on a quantitative level

Ginzburg-Landau equation

model: time dependent Ginzburg-Landau equation (TDGL)

$$\partial_t \psi = \alpha\psi - \beta|\psi|^2\psi + \gamma\nabla^2\psi - \delta|\psi|^4\psi$$

linear term $\propto$ (T_c-T)

diffusion term

higher order term for numerical stability

ψ: dimensionless order parameter

▶ typical values:

- **β=1/2**
- **γ=0.01**
- **δ=0.1**

How is α modified by the influence of the substrate?

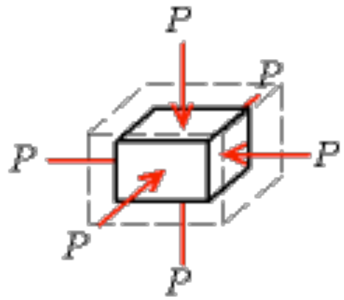
$$T_c(p = p_0 + \Delta p) = T_c(p_0) + \Delta p \frac{\partial T_c(p_0)}{\partial p}$$

Two ingredients:

1. Ginzburg-Landau equation ✔
2. Linear elasticity:
   - ➔ free energy of the superconductor
   - ➔ spatial dependence of the superconducting order parameter

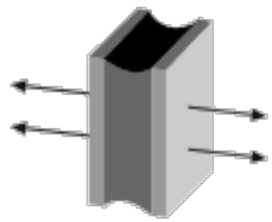
elastic moduli

- **uniform pressure:**



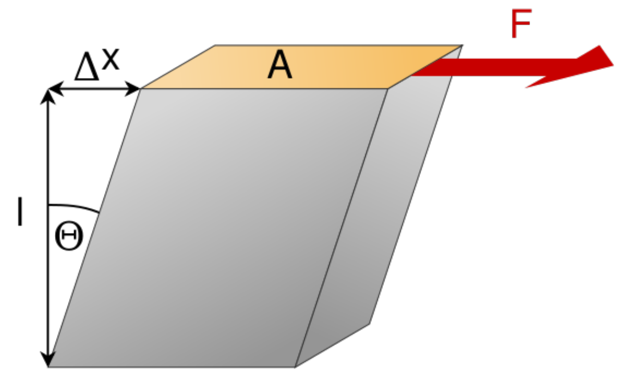
→ bulk modulus

- **tensile strain:**



→ Young's modulus

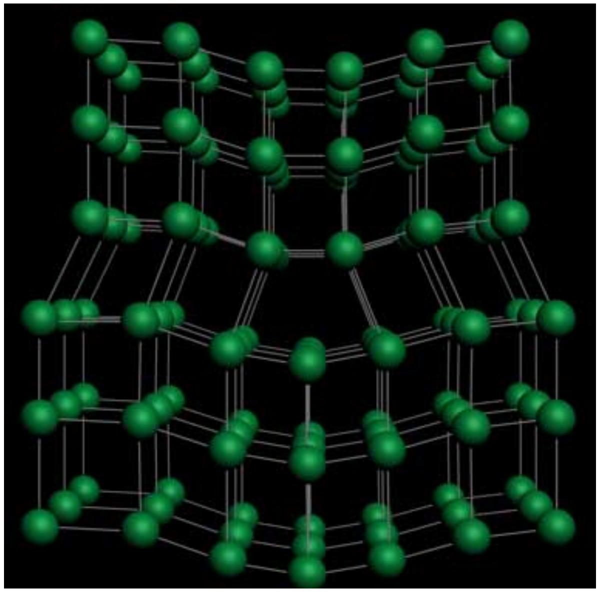
- **shear strain:**



→ shear modulus

typical material values: (1-100) GPa

linear elasticity



- elastic lattice deformations: displacement field ***u(r)***
- strain tensor: $\underline{\epsilon}$=1/2[∇**u**+(∇**u**)^T]
- Hooke's law, Cauchy stress tensor: $\underline{\sigma}$=E$\underline{\epsilon}$
- elastic constants/moduli:

- Young's modulus ***(E):*** tensile response to linear strain
- bulk modulus ***(K):*** volumetric response to shearing strains
- shear modulus ***(μ):*** response to shearing strains
- Poisson ratio: ν=1/2-E/6K

generalized Hooke's law:

$$\sigma_{ij} = c_{ijk\ell}\, \varepsilon_{k\ell}\,, \quad i,j,k,l \in x,y,z$$

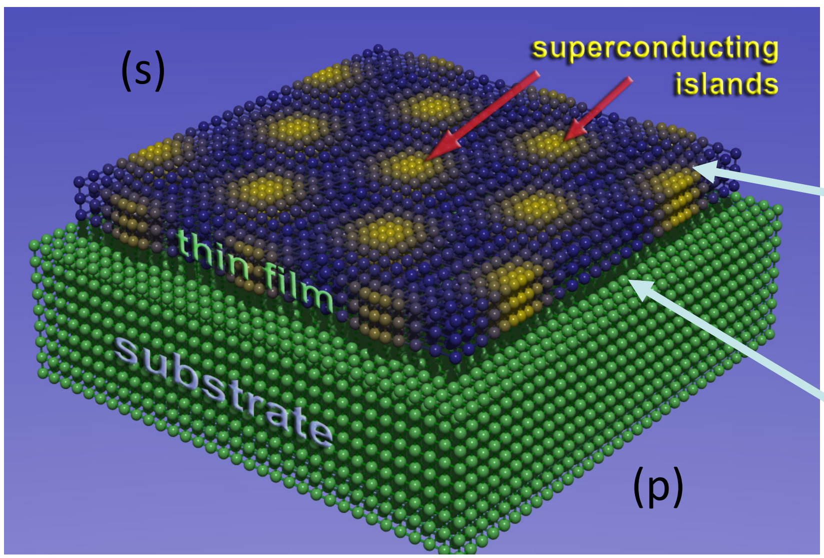
Using the symmetry of the Cauchy tensor σ_{ij}=σ_{ji}, one finds that the stiffness tensor C={c_{ijkl}} has 21 independent components!

$$\varepsilon_{ij} = \tfrac{1}{2}(\partial_i u_j + \partial_j u_i)$$

pressure corrections

pressure in the film due to elastic deformations:

$$p^{(s)}(q_x, q_y, q_z) = \frac{1}{3d} \int_0^d dz \left(\partial_x u_x^{(s)} + \partial_y u_y^{(s)} + \partial_z u_z^{(s)} \right)$$



stress balance & continuity equations at the interfaces for flat interfaces

$$\sigma_{iz}^{(s)}(0,0,d) = 0 \text{ for } i = x, y, z$$

$$\begin{cases} \sigma_{iz}^{(s)}(0,0,0) = \sigma_{iz}^{(p)}(0,0,0) \text{ for } i = x, y, z \\ u_i^{(s)}(0,0,0) = u_i^{(p)}(0,0,0) \text{ for } i = x, y, z \end{cases}$$

ansatz for elastic equation

plain wave ansatz with exponentially decay in z-direction

$$
\begin{aligned}
u_x^{(s)} &= e^{\imath q_x x + \imath q_y y}\left(e^{q_z z} A_{1,x} + A_{2,x} e^{-q_z z} + u_{0,x}^{(s)}\right) \\
u_y^{(s)} &= e^{\imath q_x x + \imath q_y y}\left(e^{q_z z} A_{1,y} + A_{2,y} e^{-q_z z} + u_{0,y}^{(s)}\right) \\
u_z^{(s)} &= e^{\imath q_x x + \imath q_y y}\left(e^{q_z z} A_{1,z} + A_{2,z} e^{-q_z z}\right) \\
u_x^{(p)} &= C_x e^{q_z z + \imath q_x x + \imath q_y y} \\
u_y^{(p)} &= C_y e^{q_z z + \imath q_x x + \imath q_y y} \\
u_z^{(p)} &= C_z e^{q_z z + \imath q_x x + \imath q_y y}
\end{aligned}
$$

→ **9 unknown complex parameters**

+ special solutions for the x & y coordinate due to interplay of superconductivity and elasticity (extended stress-strain balance to account for energy transfer from superconductivity to elasticity & vice versa)

$$u_{0,i}^{(s)} \propto -\imath \frac{|\psi|^2 q_i}{\mathbf{q}^2}$$

stress tensor & coupling parameter

- from the displacement fields we can derive the stress tensors:

$$
\begin{aligned}
\sigma_{xz}^{(s)} &= \mu^{(s)} \left(\partial_z u_x^{(s)} + (1-2\sigma_0)\,\partial_x u_z^{(s)}\right) \\
\sigma_{yz}^{(s)} &= \mu^{(s)} \left(\partial_z u_y^{(s)} + (1-2\sigma_0)\,\partial_y u_z^{(s)}\right) \\
\sigma_{zz}^{(s)} &= 2\mu^{(s)} \left[\left(1+\nu^{(s)}\right)\partial_z u_z^{(s)} + \nu^{(s)}\left(\partial_x u_x^{(s)} + \partial_y u_y^{(s)}\right)\right] \\
\sigma_{xz}^{(p)} &= \mu^{(p)} \left(\partial_z u_x^{(p)} + \partial_x u_z^{(p)}\right) \\
\sigma_{yz}^{(p)} &= \mu^{(p)} \left(\partial_z u_y^{(p)} + \partial_y u_z^{(p)}\right) \\
\sigma_{zz}^{(p)} &= 2\mu^{(p)} \left[\left(1+\nu^{(p)}\right)\partial_z u_z^{(p)} + \nu^{(p)}\left(\partial_x u_x^{(p)} + \partial_y u_y^{(p)}\right)\right]
\end{aligned}
$$

σ_o due to lattice mismatch

- final expression for T_c-correction due to elasticity in q-space:

$$U_0 \mathcal{K}(q) |\psi_q|^2$$

with $$U_0 = 3K\Delta\alpha_L[\partial T_c(p_0)/\partial p]$$

from $K = -V\partial p/\partial V$ $\rightarrow$ $\Delta p = -K\frac{\Delta V}{V} = -3K\frac{\Delta L}{L} \propto -3K\alpha_L$

elastic kernel $\mathcal{K}$(q)

- **FYI**

$$
\begin{aligned}
\mathcal{K}(q) \;=\; & \Big\{\mu_p^2\left(1+2\nu^{(p)}\right)\left[d\ q(1+e^{4d\ q})\vartheta_s-(e^{4d\ q}-1)\left(2\vartheta_s+1\right)\right] \\
& +d\ q\left[e^{4d\ q}\mu_s\vartheta^{(s)}\left(2\mu^{(p)}+2\mu^{(p)}\nu^{(p)}\sigma_0-\mu^{(s)}\vartheta^{(s)}\right)\right. \\
& \quad -\mu^{(s)}\vartheta^{(s)}\left(\mu^{(s)}\vartheta^{(s)}+2\mu^{(p)}\left[1+\nu^{(p)}+\nu^{(s)}-\nu^{(p)}\sigma_0\right]\right) \\
& \quad \left.-2e^{2d\ q}\left(\mu^{(p)2}\left[1+2\nu^{(p)}\right]\left[1+2\nu^{(s)}\sigma_0\right]-\mu^{(s)2}\vartheta^{(s)2}-\mu^{(p)}\mu^{(s)}\vartheta^{(s)}(\nu^{(p)}+\nu^{(s)}-2\nu^{(p)}\sigma_0)\right)\right] \\
& +\mu^{(s)}\left(e^{d\ q}-1\right)^2\Big[4\mu^{(s)}\nu^{(s)}\left(e^{2d\ q}-1\right)\vartheta^{(s)}\left[\sigma_0-1\right] \\
& \quad +\mu^{(p)}\Big[1+2\nu^{(s)}\left(2-2\vartheta^{(s)}-3\sigma_0\right)+2\nu^{(p)}\left[2\vartheta^{(s)}+1\right]\left[\sigma_0-1\right]-2e^{d\ q}\left(\nu^{(s)}\left(2\nu^{(p)}\left[\sigma_0-1\right]+2\sigma_0-3\right)-1\right) \\
& \quad -e^{2d\ q}\left(3\vartheta^{(s)}+2+2\nu^{(p)}\left[\sigma_0-1\right]\left(\nu^{(s)}\left[4\sigma_0-2\right]-1\right)\right)\Big]\Big] \\
& +4\mu^{(p)2}\nu^{(s)}e^{2d\ q}\left[1+2\nu^{(p)}\right]\left[4\sigma_0-3\right]\sinh\left(d\ q\right)\Big\}\Big/ \\
& \Big\{3d\ q\Big\{-\mu^{(s)2}\left(e^{2d\ q}-1\right)^2\vartheta^{(s)2} \\
& +2\mu^{(p)}\mu^{(s)}\left(e^{2d\ q}-1\right)\vartheta^{(s)}\left(1+\nu^{(p)}+\nu^{(s)}-\nu^{(p)}\sigma_0+e^{2d\ q}\left[1+\nu^{(p)}\sigma_0\right]\right) \\
& +\mu^{(p)2}(1+2\nu^{(p)})\left(e^{4d\ q}\vartheta^{(s)}+2\nu^{(s)}\left[\sigma_0-1\right]-1-2e^{2d\ q}\left[1+2\nu^{(s)}\sigma_0\right]\right)\Big\}\Big\}
\end{aligned}
$$

parameters

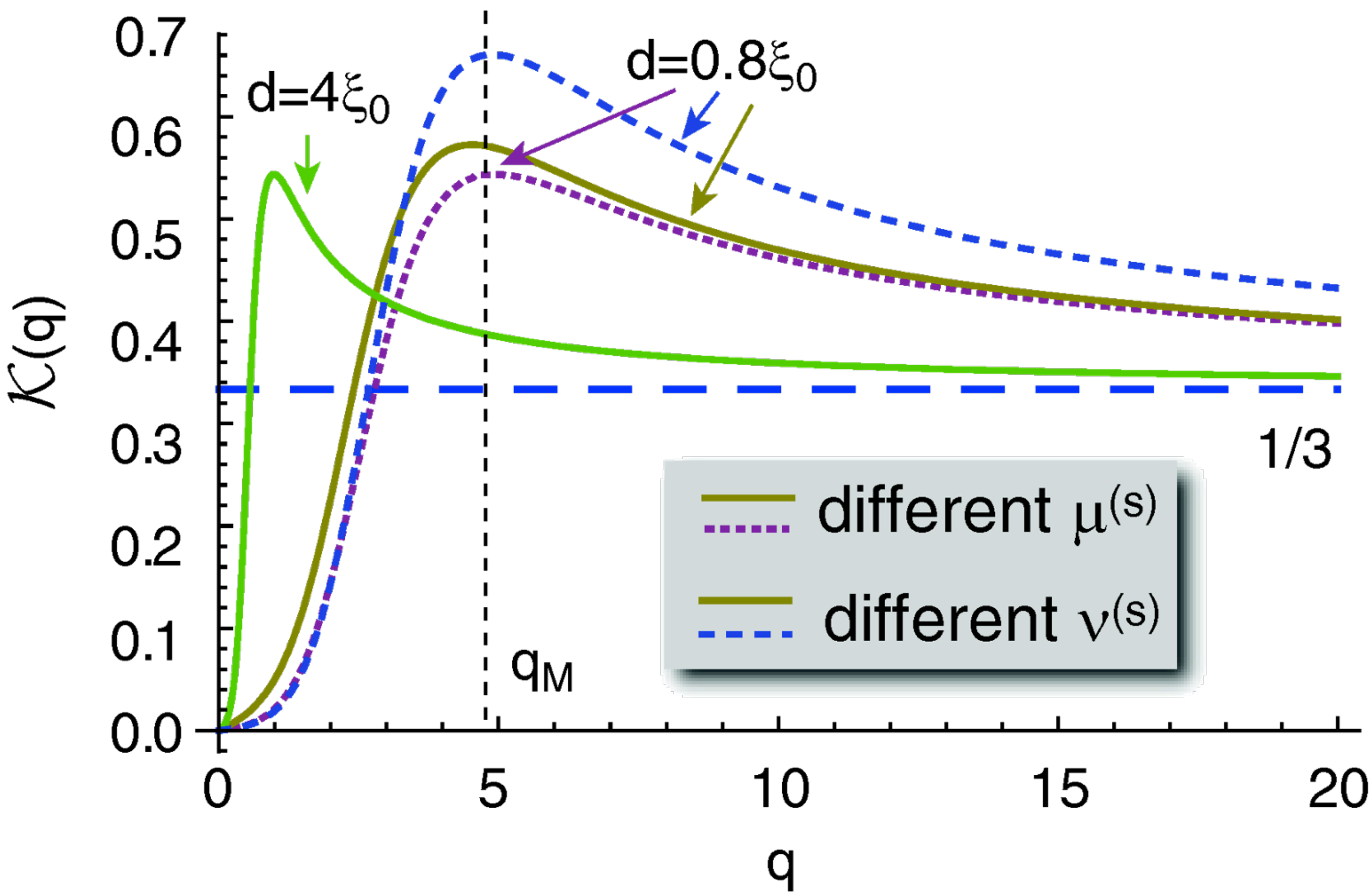
- **parameters of the elastic potential**

d	thickness of the film
$\mu^{(s,p)}$	shear modulus
σ_0	deformation stress
$\nu^{(s,p)}$	modified Poisson number
U_0	potential strength
K	bulk modulus
$\Delta\alpha_L$	linear expansivity
$\partial T_c/\partial p$	T_c change with pressure

with

$$U_0 = 3K\Delta\alpha_L[\partial T_c(p_0)/\partial p]$$

$\mathcal{K}$(q)



0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
$\mathcal{K}$(q)
d=4ξ_0
d=0.8ξ_0
q_M
1/3
different $\mu^{(s)}$
different $\nu^{(s)}$
0
5
10
15
20
q

final expression for α

$$\partial_t \psi = \alpha\psi - \beta|\psi|^2\psi + \gamma\nabla^2\psi - \delta|\psi|^4\psi$$

- The original linear coefficient in the TDGL equation has to be replaced by

$$\alpha \to \alpha_\psi(\mathbf{r}) = \alpha_0[T_c(\mathbf{r}) - T] + \int d\mathbf{r}' U(\mathbf{r} - \mathbf{r}')|\psi(\mathbf{r}', t)|^2$$

includes the effect of
(weak) quenched disorder

effective long-range potential

weak disorder effects are not important.

numerical realization

- **The TDGL equation is solved using a quasi-spectral "split-step" method**

the order parameter is discretized in x&y direction: ψ_{ij}

0. step

- calculate α in Fourier space (using FFT)

1. step

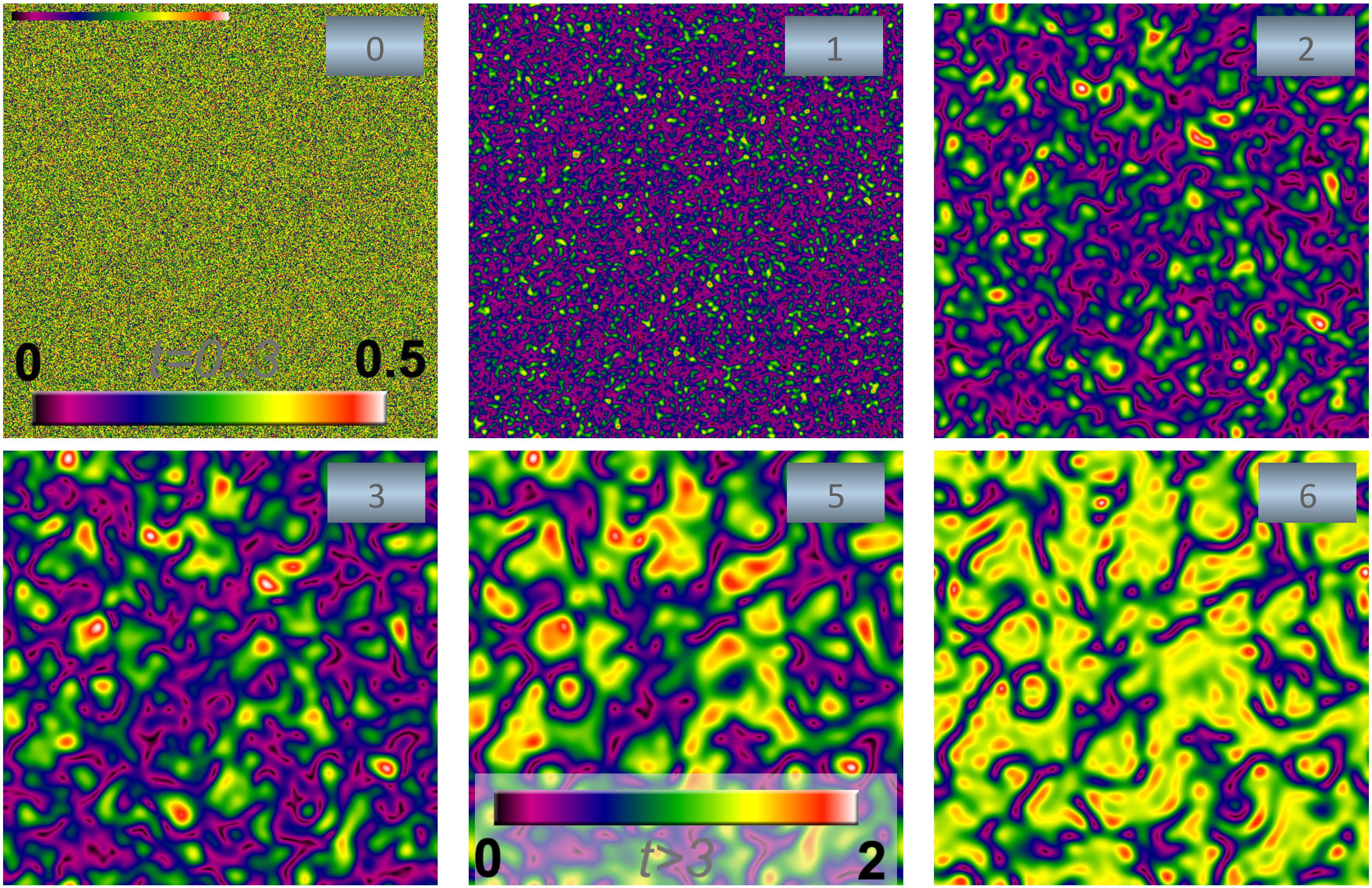
- in real space do:

$$\psi_{ij}(t+\Delta t) = e^{\Delta t(\alpha - \beta|\psi_{ij}|^2 + \delta|\psi_{ij}|^4)}\psi_{ij}(t)$$

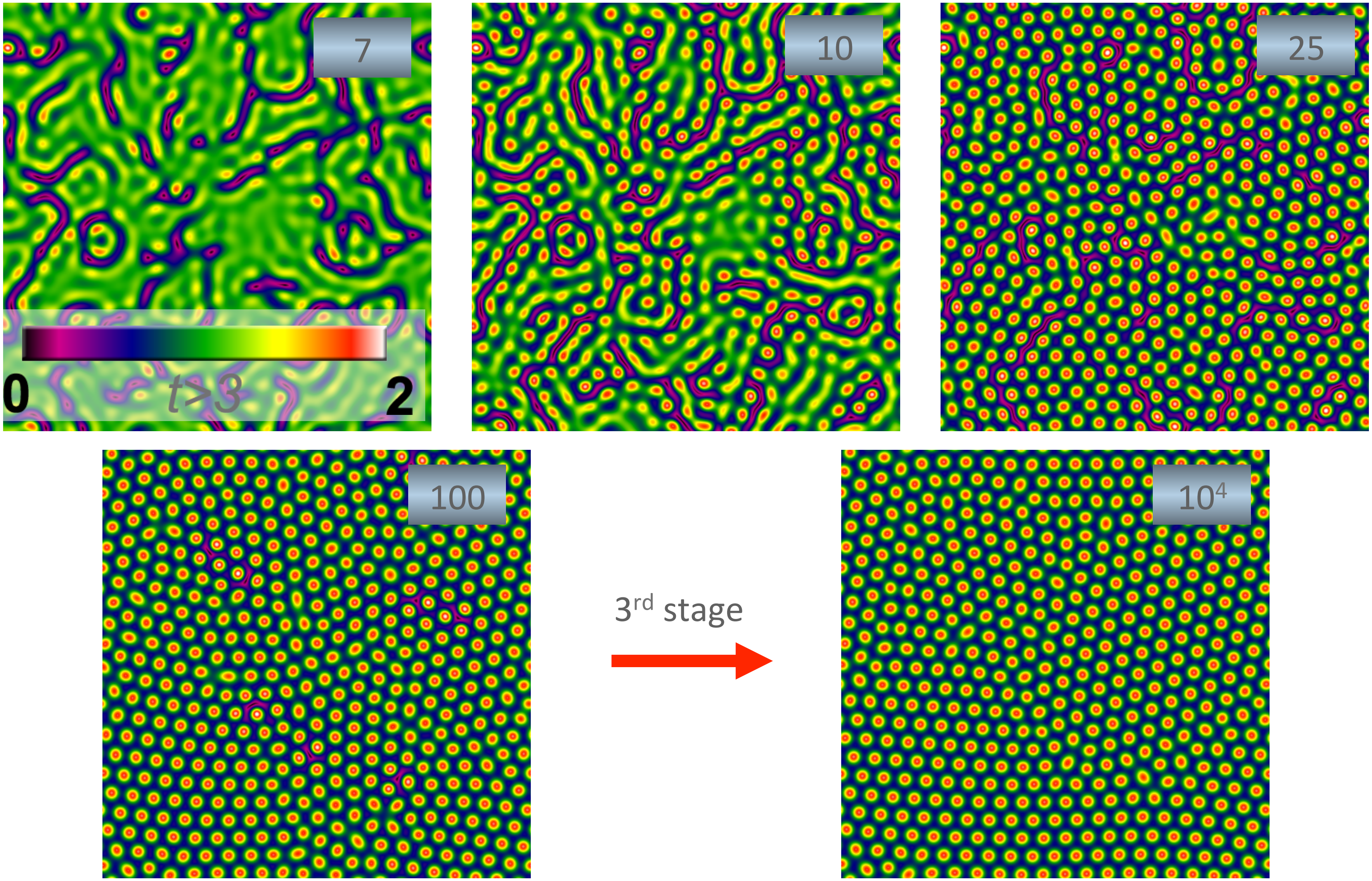
2. step

- apply the diffusion kernel $-\gamma q^2$ to ψ_q in Fourier space

First stage: formation of amorphous state

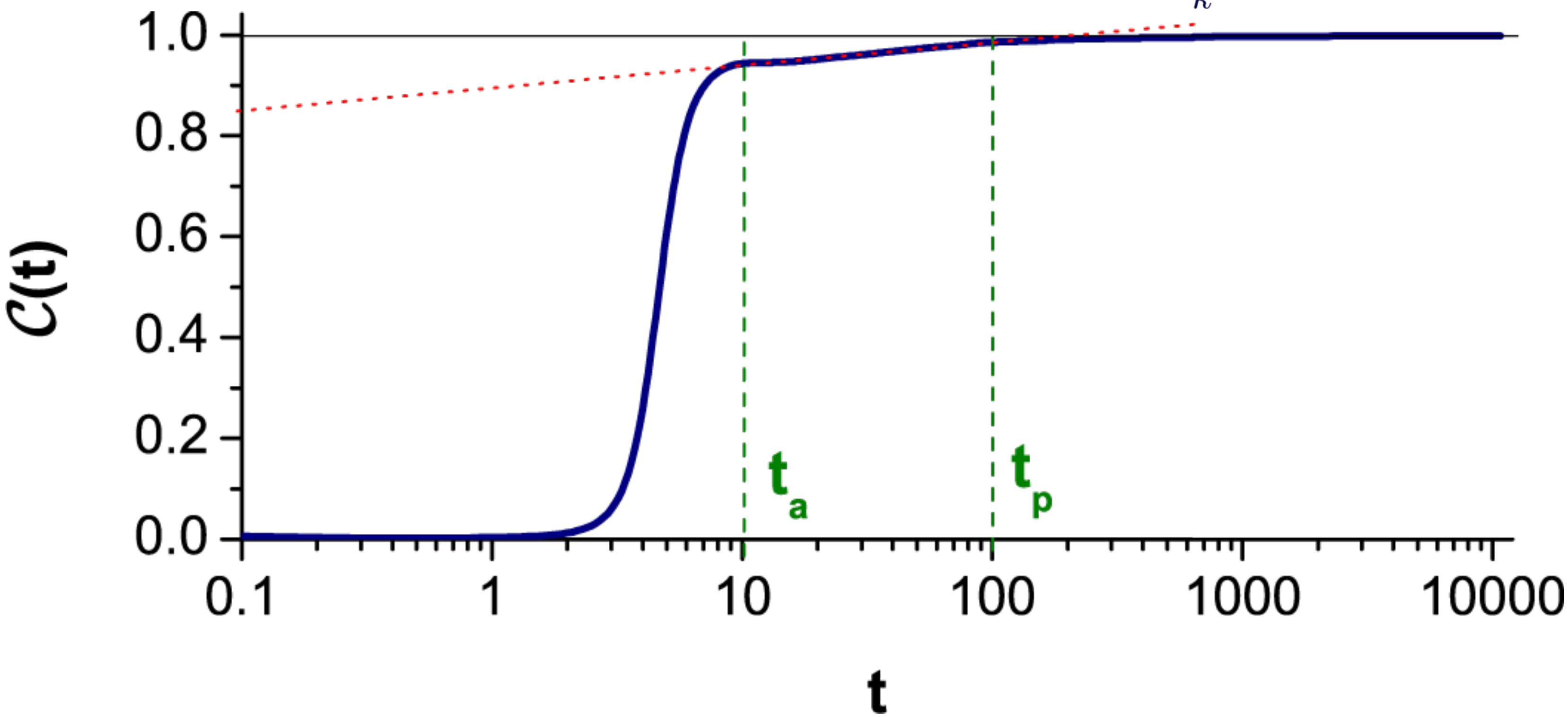


2nd stage: formation of a polycrystalline lattice



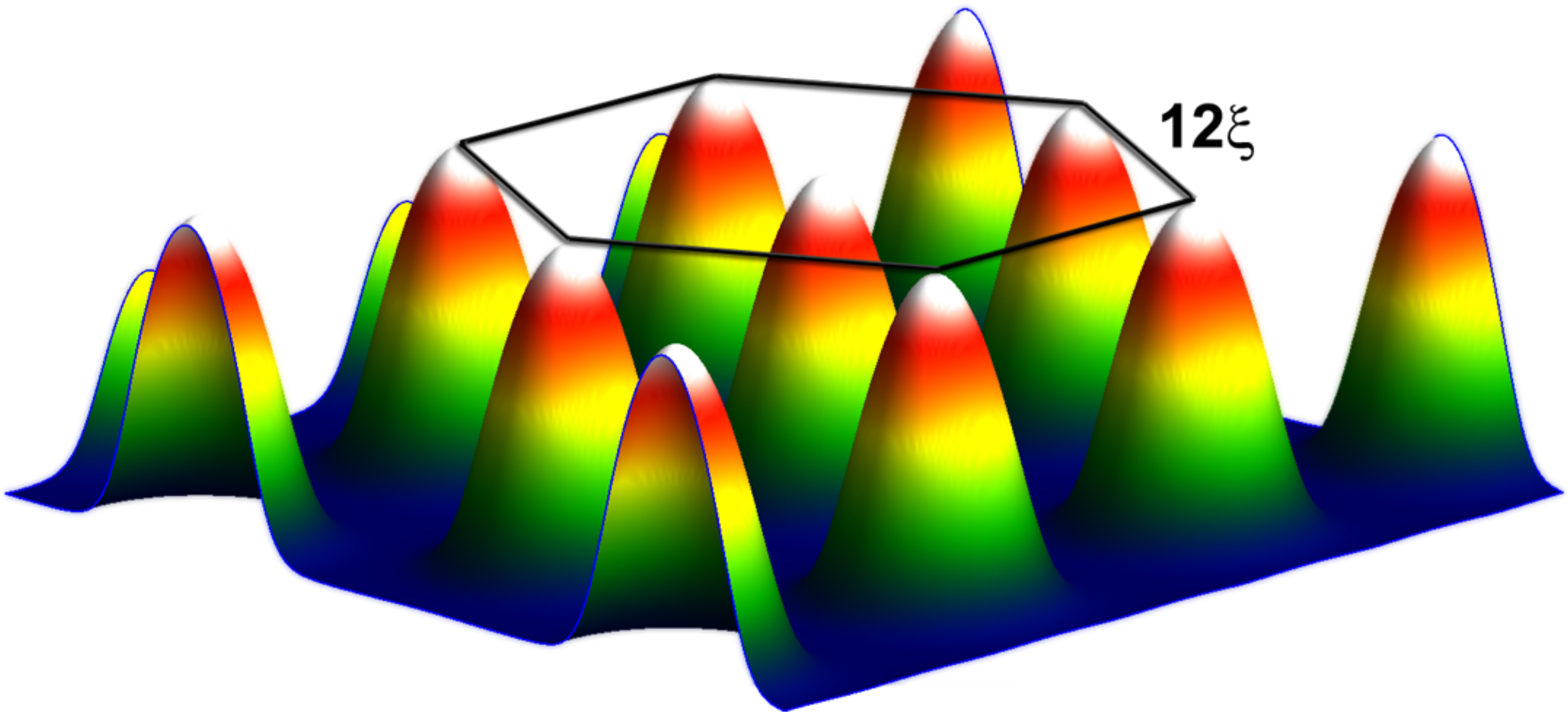
amplitude evolution over time

- order parameter correlation function: $\mathcal{C}(t) = \mathcal{N}^{-1} \sum_k |\psi_k|^2$



- temperature: $T = 0.8T_c$
- units of time: $\tau_{\rm GL} \simeq 10^{-11} s$
- coupling constant: $U_0 = 2.2U_c$
- thickness: $d = 0.8\xi_0$

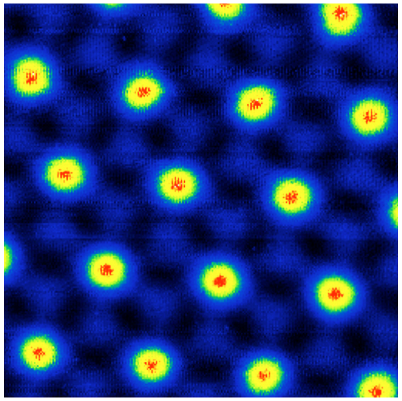
amplitude in the finial lattice configuration

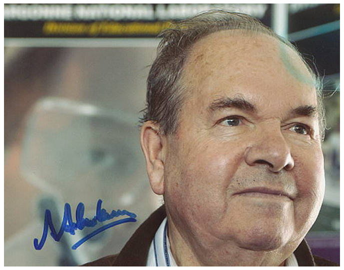


coherence length: $\xi(T) \propto (T_c - T)^{-1/2}$

comparison: vortex & SC island lattice

- Vortex lattice





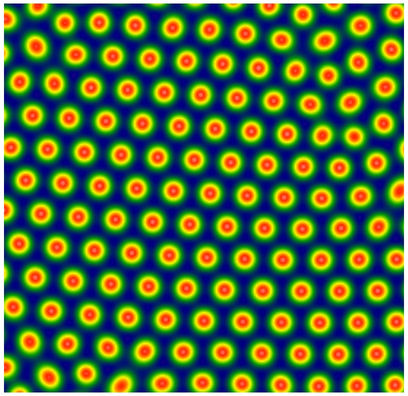
Abrikosov, 1957

STM of $NbSe_2$
T=400mK
B=0.5T

- ψ=0 in the vortex core
- ψ>0, H=0 outside

- hexagonal lattice
- depending on pinning R=0

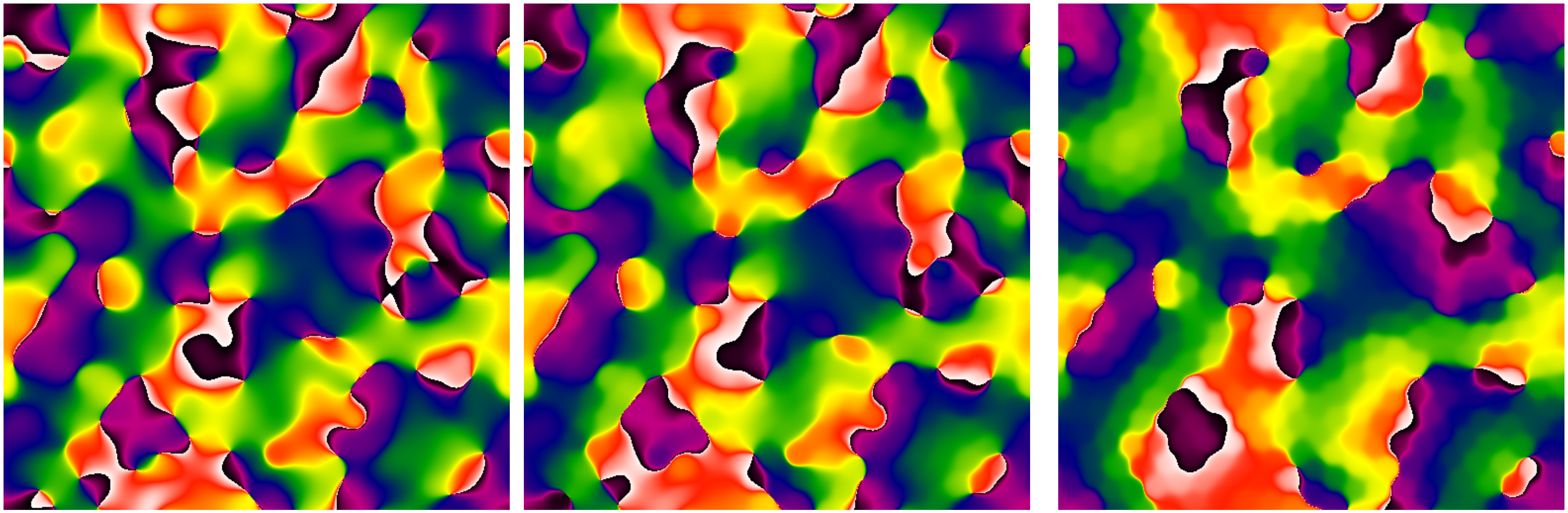
- superconducting islands

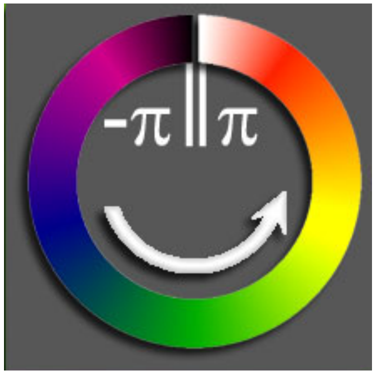


- ψ>0 in islands
- $\psi \rightarrow 0$, H>0, T_c<T between islands
- hexagonal lattice
- depending on coupling and material system is normal or insulator

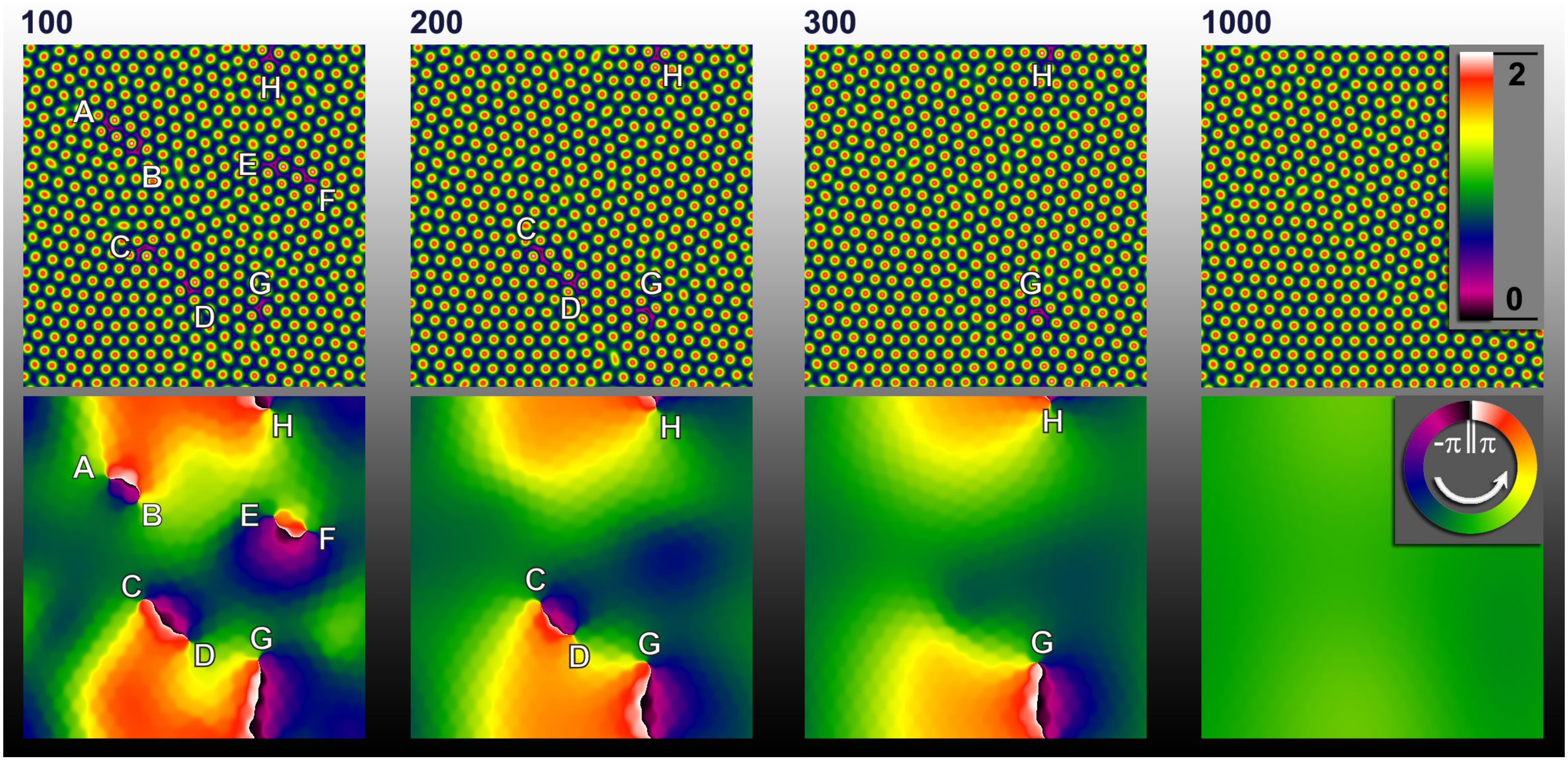
initial phases

$$\psi(\mathbf{r}) = |\psi| e^{i\varphi(\mathbf{r})}$$

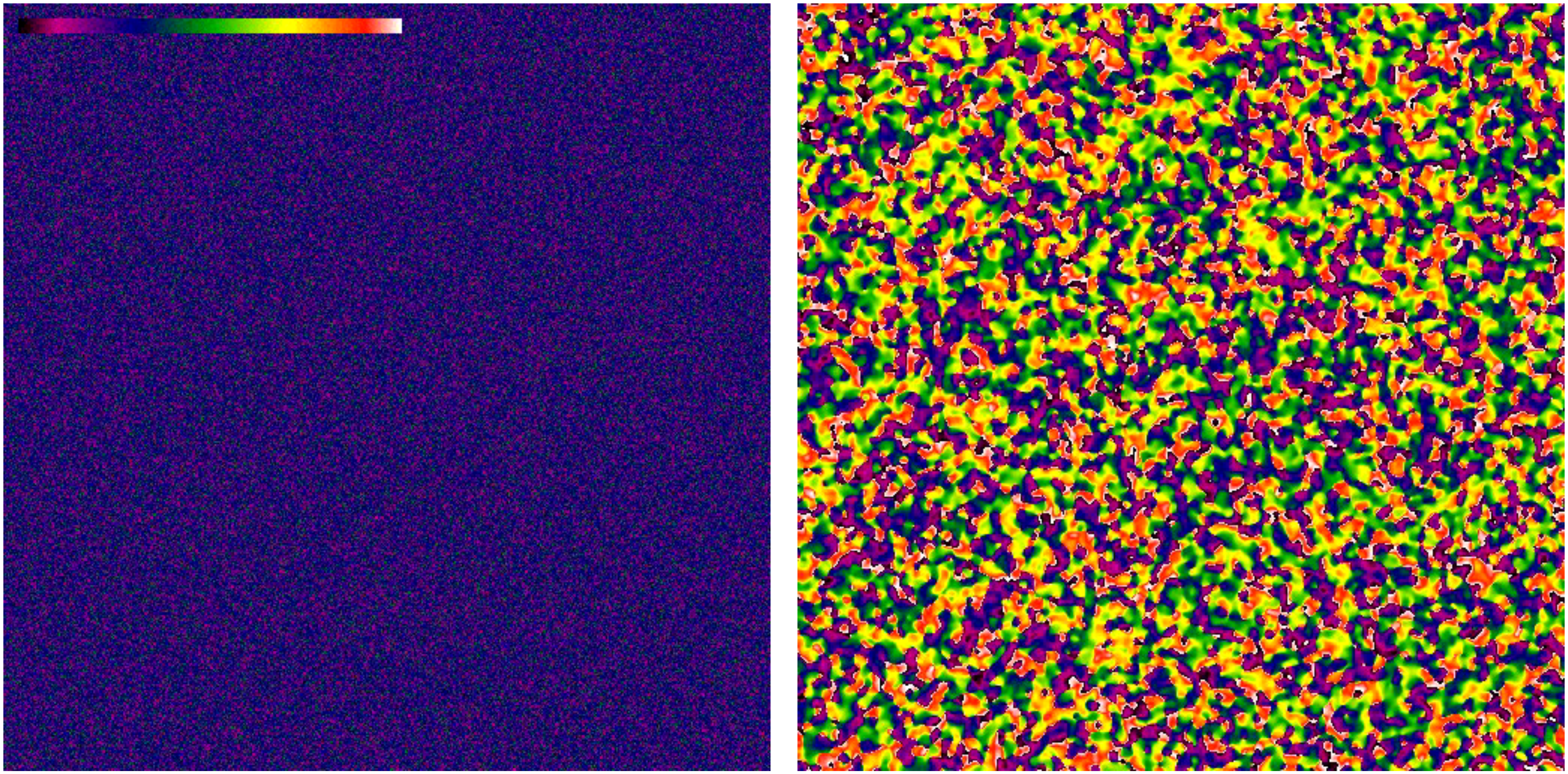




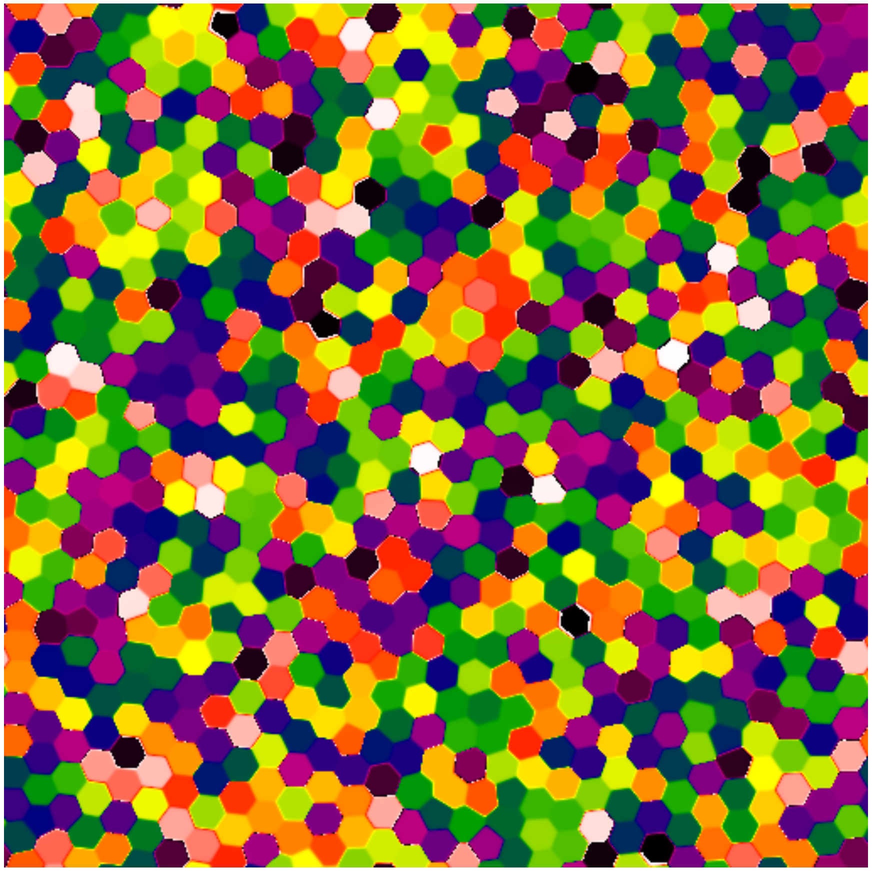
3rd stage: long-range order formation

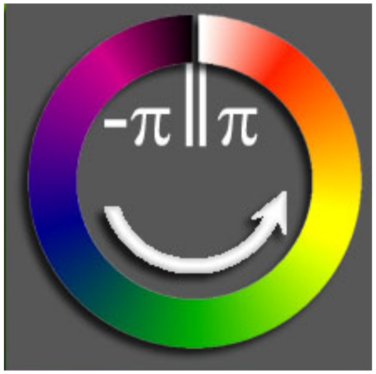


amplitude and phase evolution



stronger coupling constant





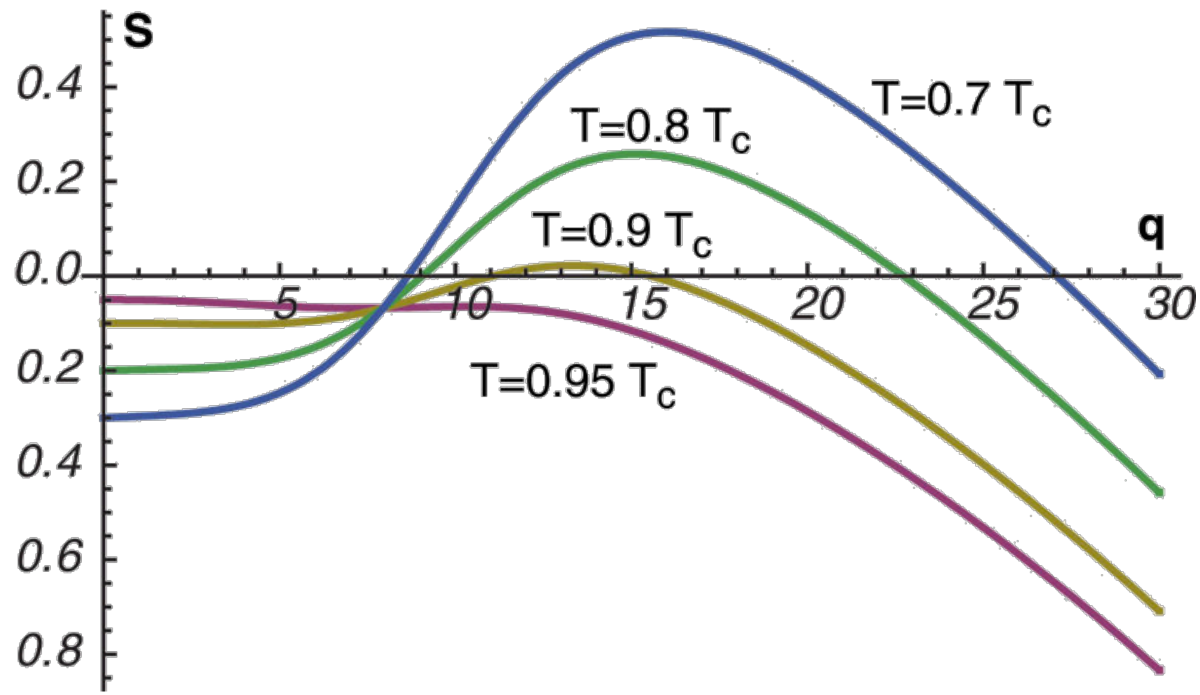
"intermediate state" for larger U_0 and lower temperature: phase equilibration very slow

linear stability

Question: Under which condition do islands form?

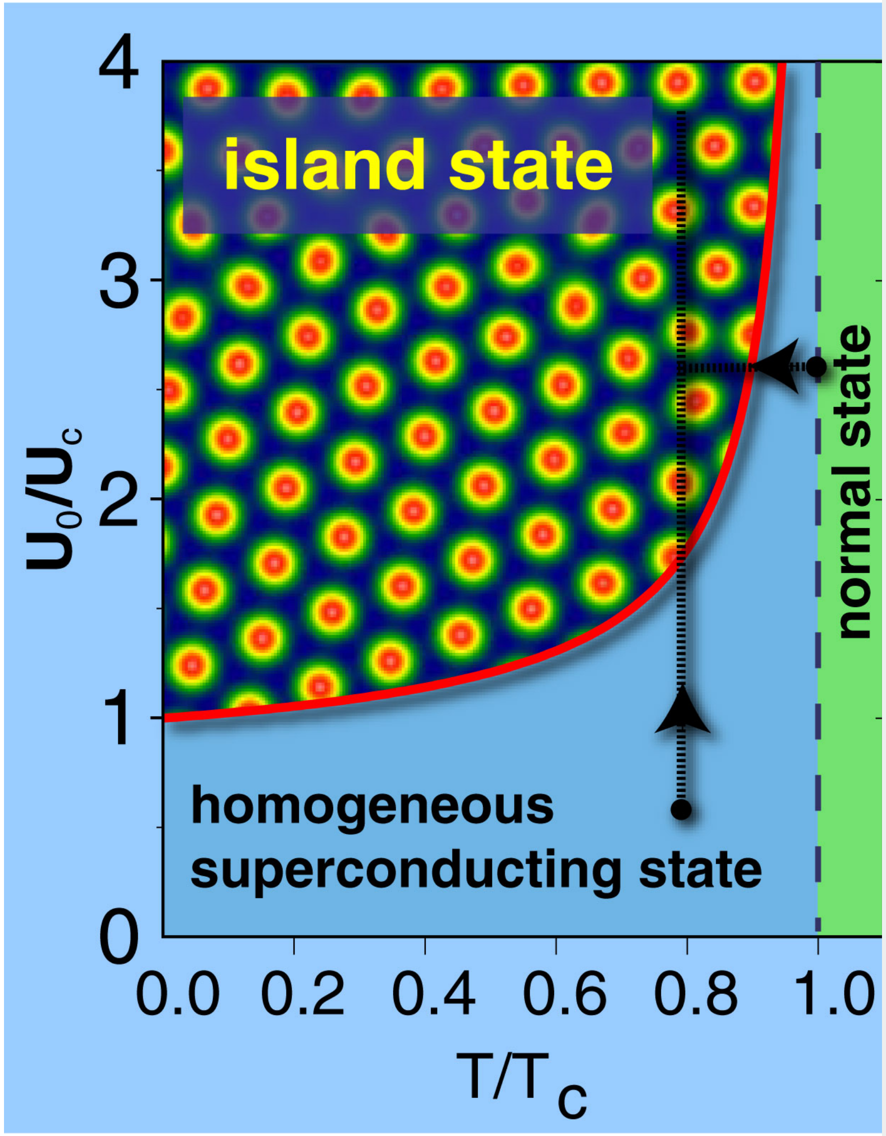
Answer: When the homogeneous solution of the GL equation becomes unstable!

$$S(q) = (2U_0\mathcal{K}(q) - 1)(1 - T/T_c) - \gamma q^2/9.38$$



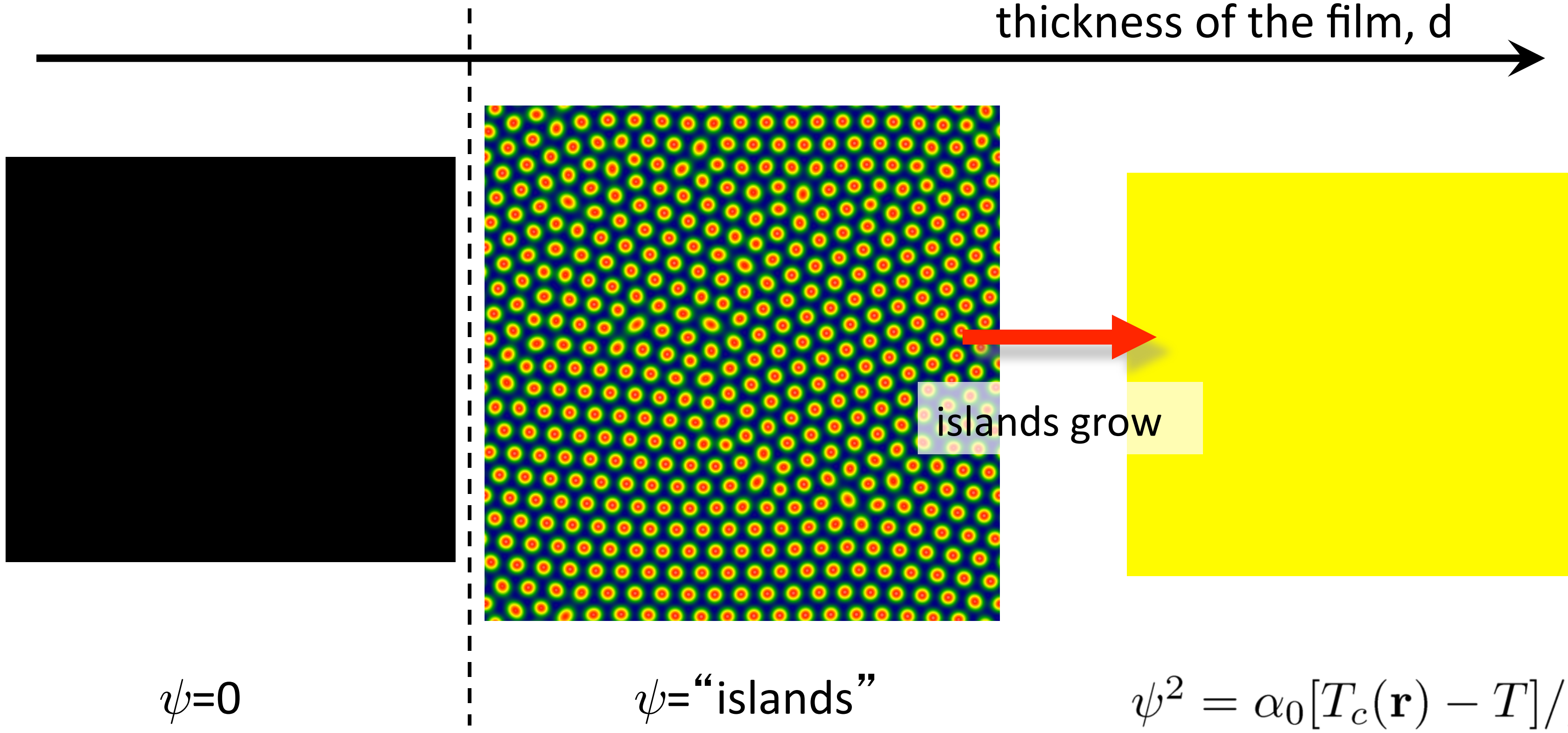
A long-range Coulomb potential does not show this instability!

phase diagram



Superconductor-Insulator transition

- possible scenario



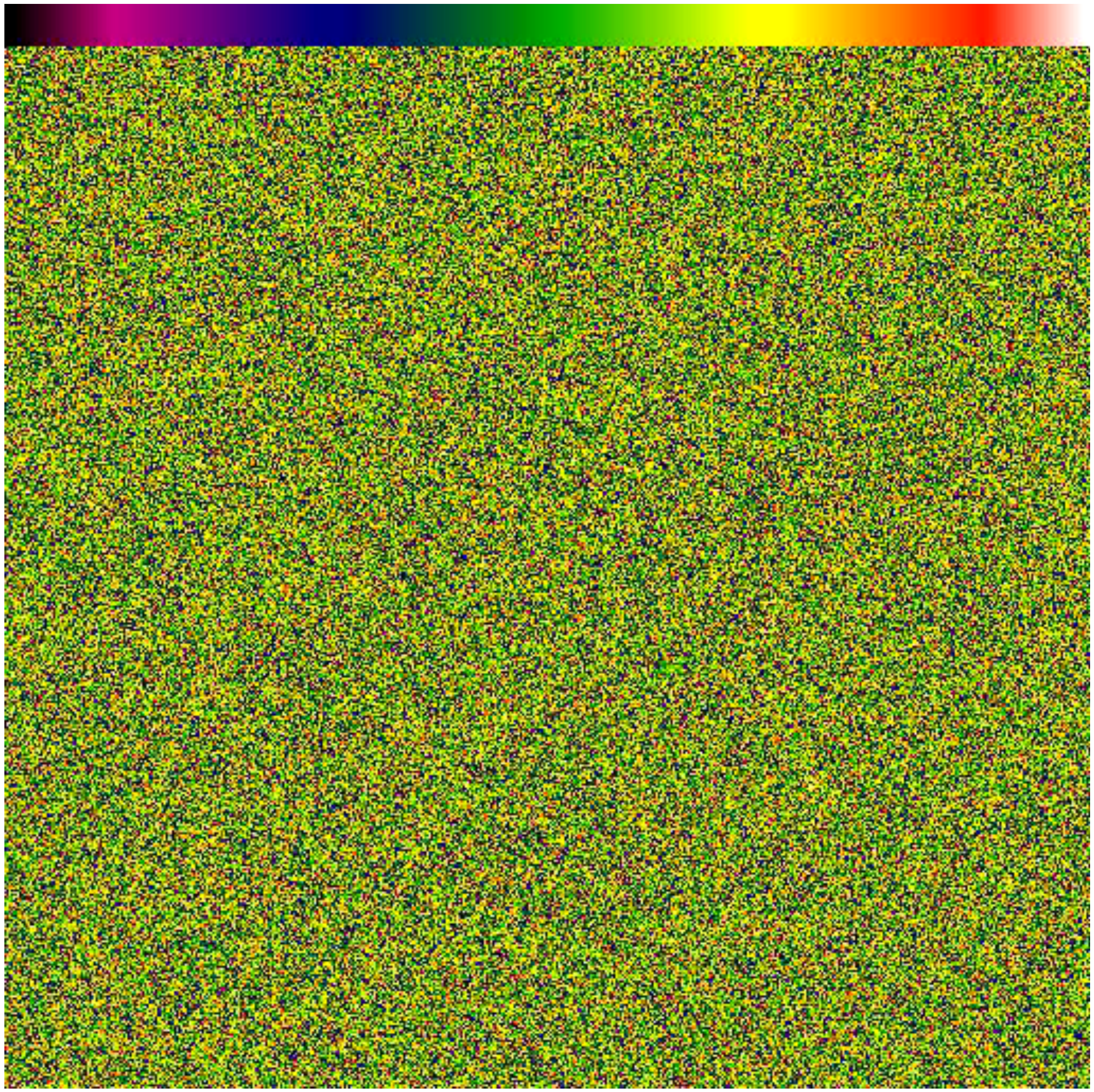
ψ=0 | ψ="islands" | $\psi^2 = \alpha_0[T_c(\mathbf{r}) - T]/\beta$

bulk values for U_0

Material	T_{c0}, K	$\frac{\partial T_c}{\partial p}$, K/GPa	α_L, 1/K	K, GPa	U_0
(a) $YBa_2Cu-3O_{7-\delta}$	90.9	1.9-2.2 (a,b-axis)	0.5-1 $\times 10^{-6}$	200-250	2×10^{-3}
(b) $Ba(Fe_{1-x}Co_x)_2As_2$	21	-26 (c-axis)	-11.7×10^{-6}	~ 250	~ 0.23
(c) κ-$(D_8\text{-ET})_2Cu(NCS)_2$	9	-30 (hydrostatic)	8×10^{-6}	12.2	-10^{-2}

Table 1: **Coupling constants.** Estimate of the coupling constant $U_0 = 3K\alpha_L \partial T_c/\partial p$ for different materials. The values are given for bulk materials and therefore define only the lower limit for thin films. (a) data from [**?**] for bulk, anisotropic material; (b) from [**?**]: bulk, $x = 0.074$; (c) from [**?**]: bulk, organic

… in electric field



summary and conclusions

- elastic coupling of a thin film to a rigid substrate introduces an effective long-range interaction
- **positive feedback of local change in T_c and lattice leads to the formation of a regular, hexagonal island structure**
- Solution showing the nucleation of superconducting islands in a normal background – in contrast to Abrikosov's solution
- transition into the island state show first order hysteretic behavior
- **pattern can be viewed as a self-assembled array of nano-scale Josephson junctions**
- depending on elastic coupling and pinning the structure might be either in the superconducting or insulation state!

→ ***can be a key scenario for the superconductor-to-insulator transition in thin homogeneous films***