Phase field modelling of microstructural evolution

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Outline

- Microstructure and its evolution
  (a) Solid-Solid transformation
  (b) Solid-Liquid transformation
- Phase field models as diffusion equations
- Other viewpoints on phase field models
- Elastic stress driven microstructural evolution – ATG instabilities
- Summary

Jet engine

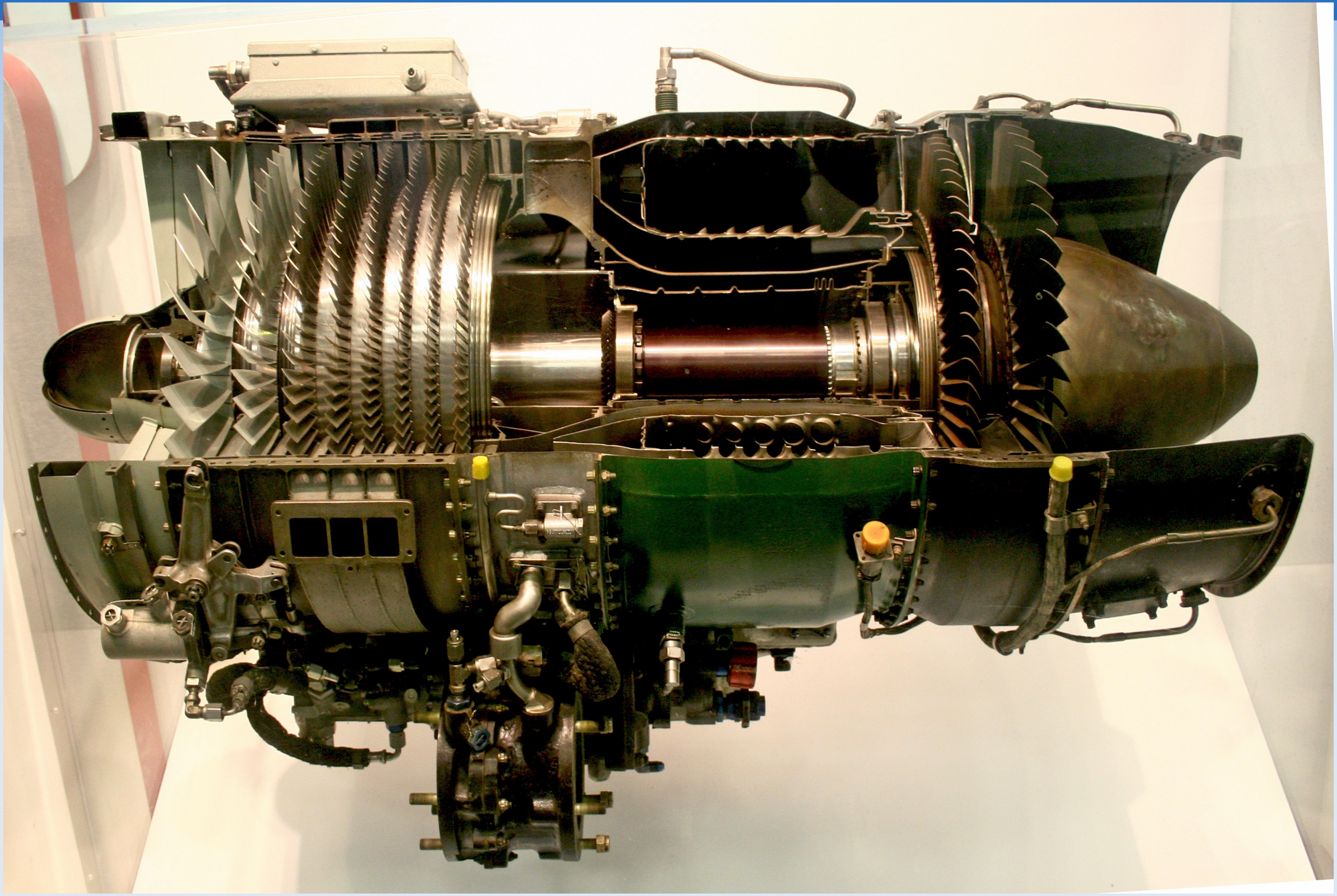


Photo courtesy: Sanjay Acharya -- http://en.wikipedia.org/wiki/File:J85_ge_17a_turbojet_engine.jpg

Turbine blade

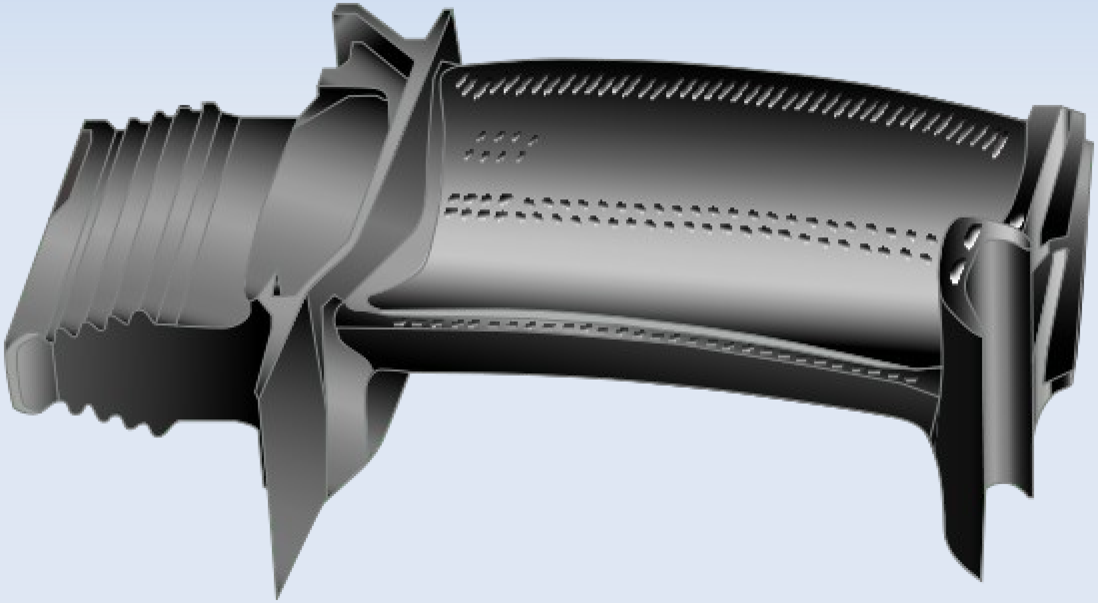


Photo courtesy: Tomeasy -- http://en.wikipedia.org/wiki/File:GaTurbineBlade.svg

Using materials above their T_m

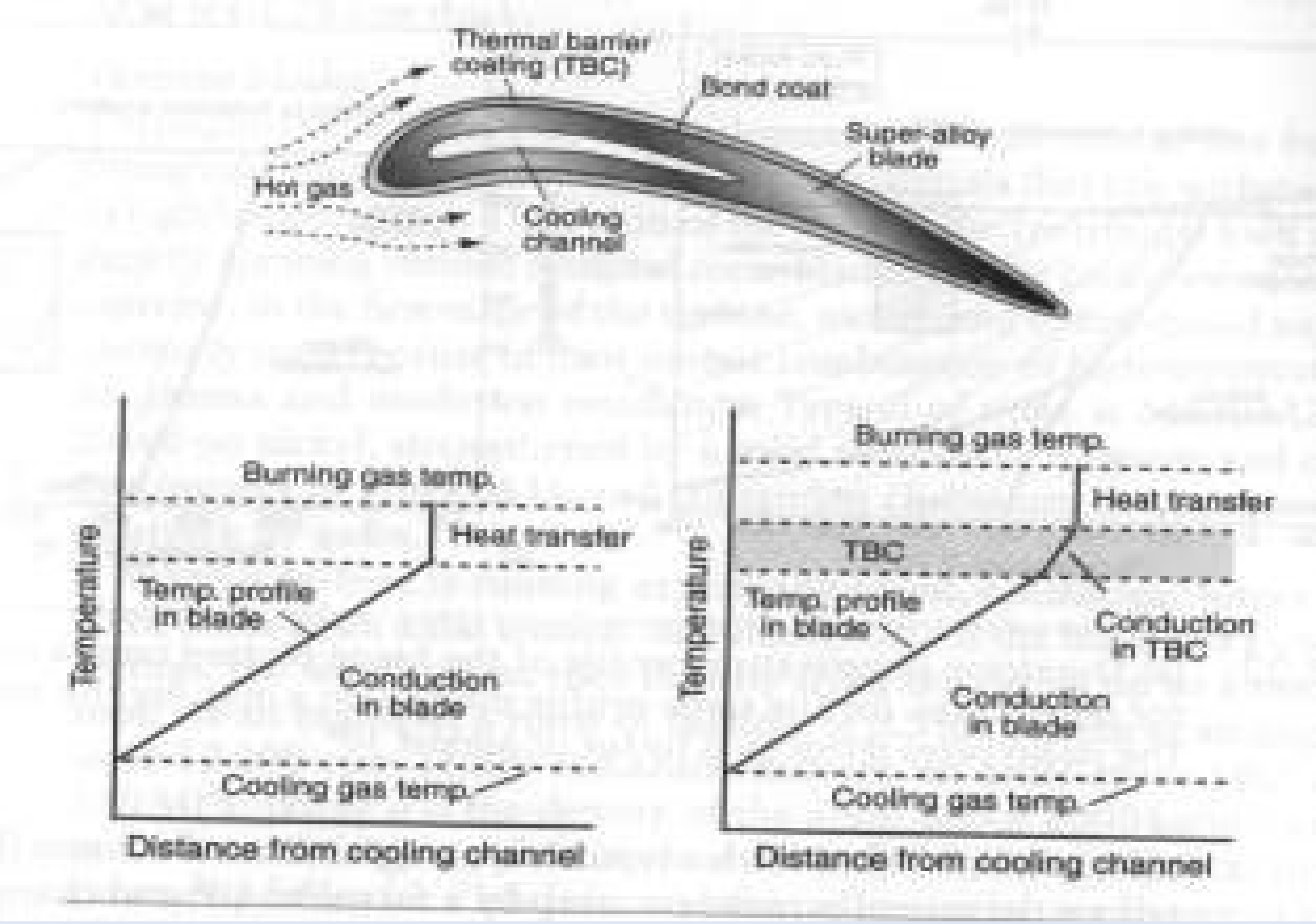
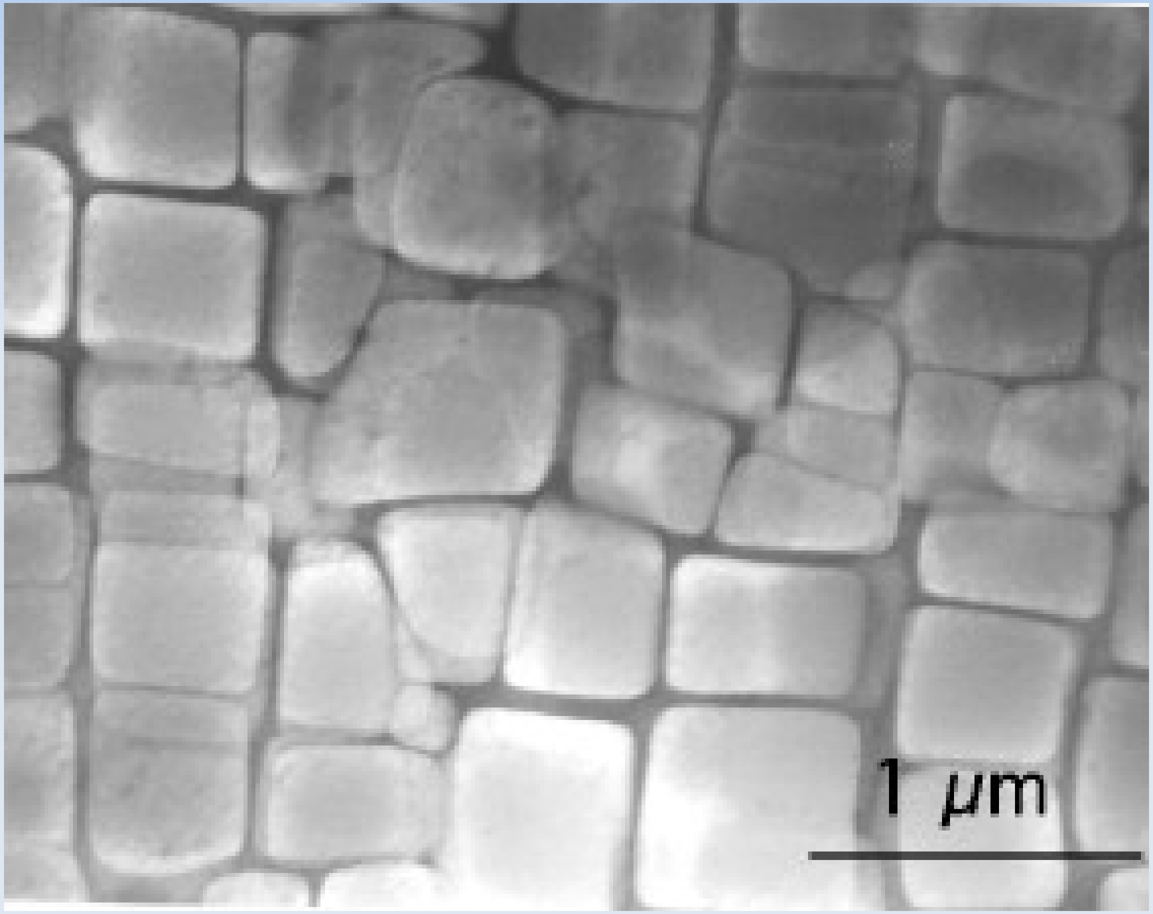


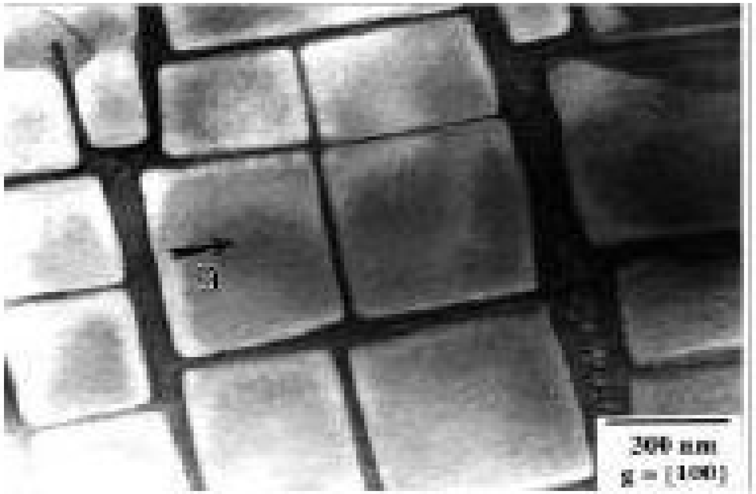
Image courtesy: Ashby, Shercliff and Cebon, Materials: engineering, science, processing and design, Butterworth-Heinemann, 2007

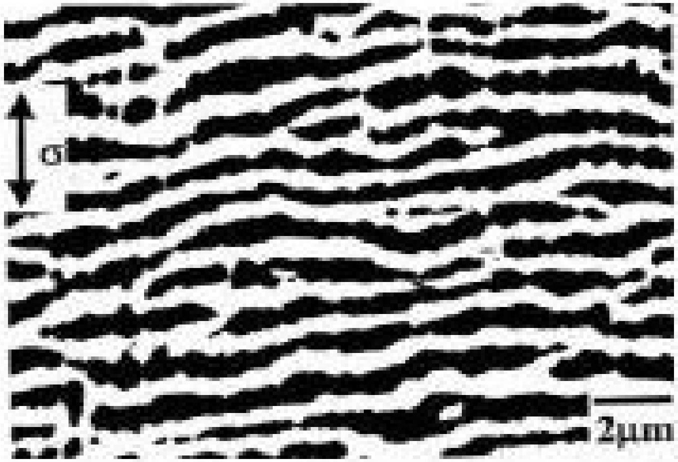
Microstructure



Micrograph courtesy: Hillier -- http://www.msm.cam.ac.uk/phasetrans/2003/Superalloys/superalloys.html

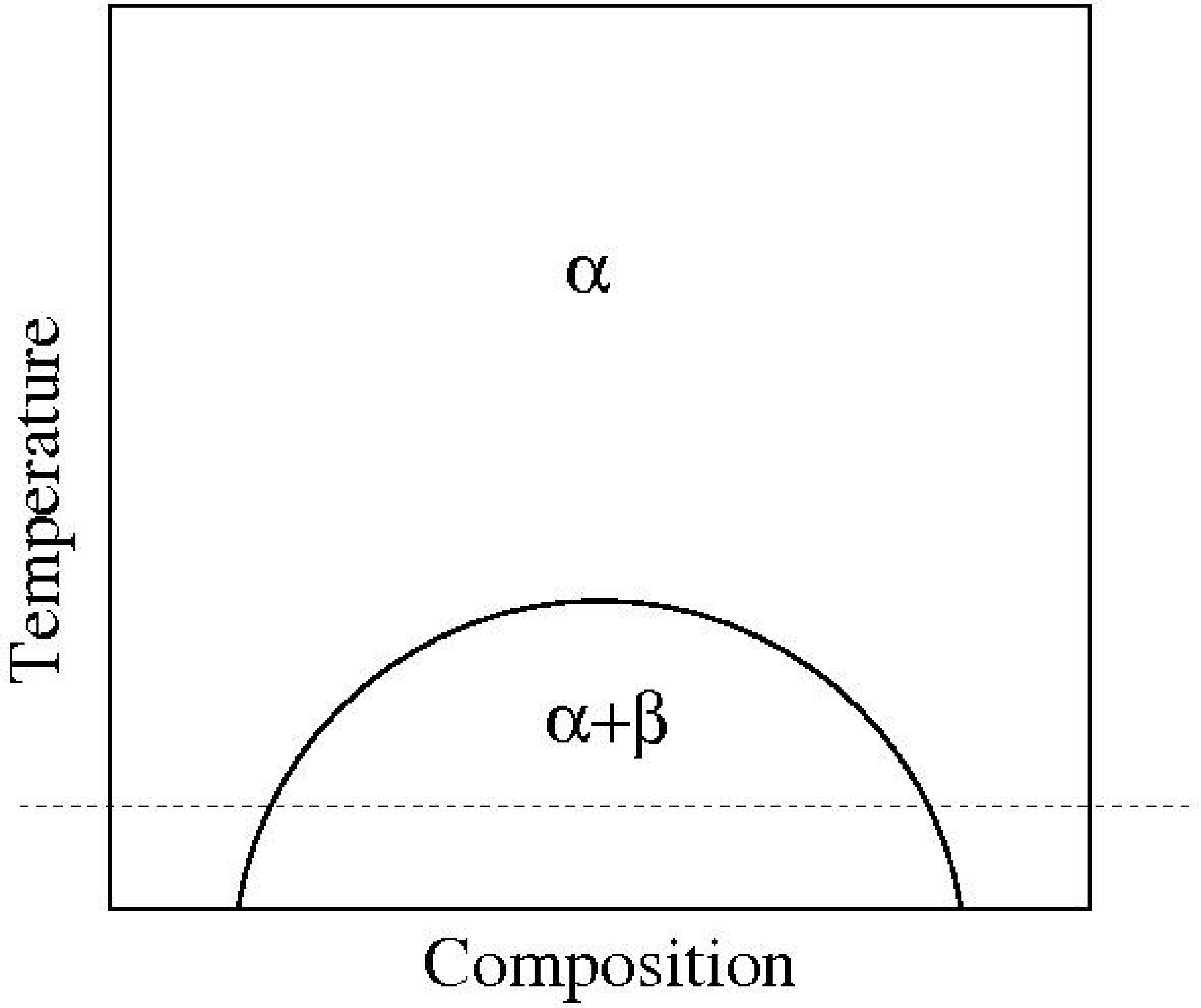
Rafting



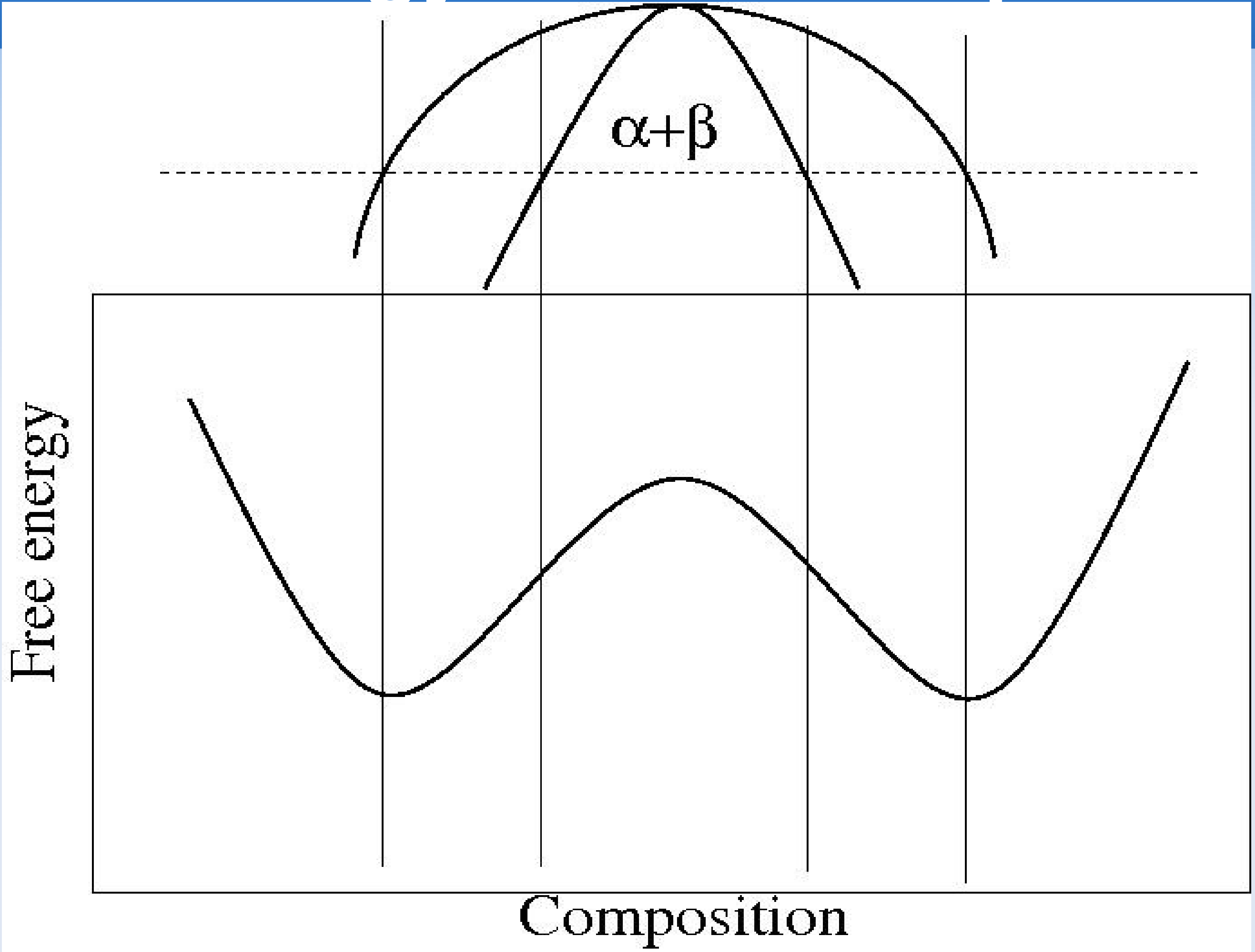


Micrographs courtesy: M Kamaraj, Sadhana, Vol. 28, pp. 115-128, 2003

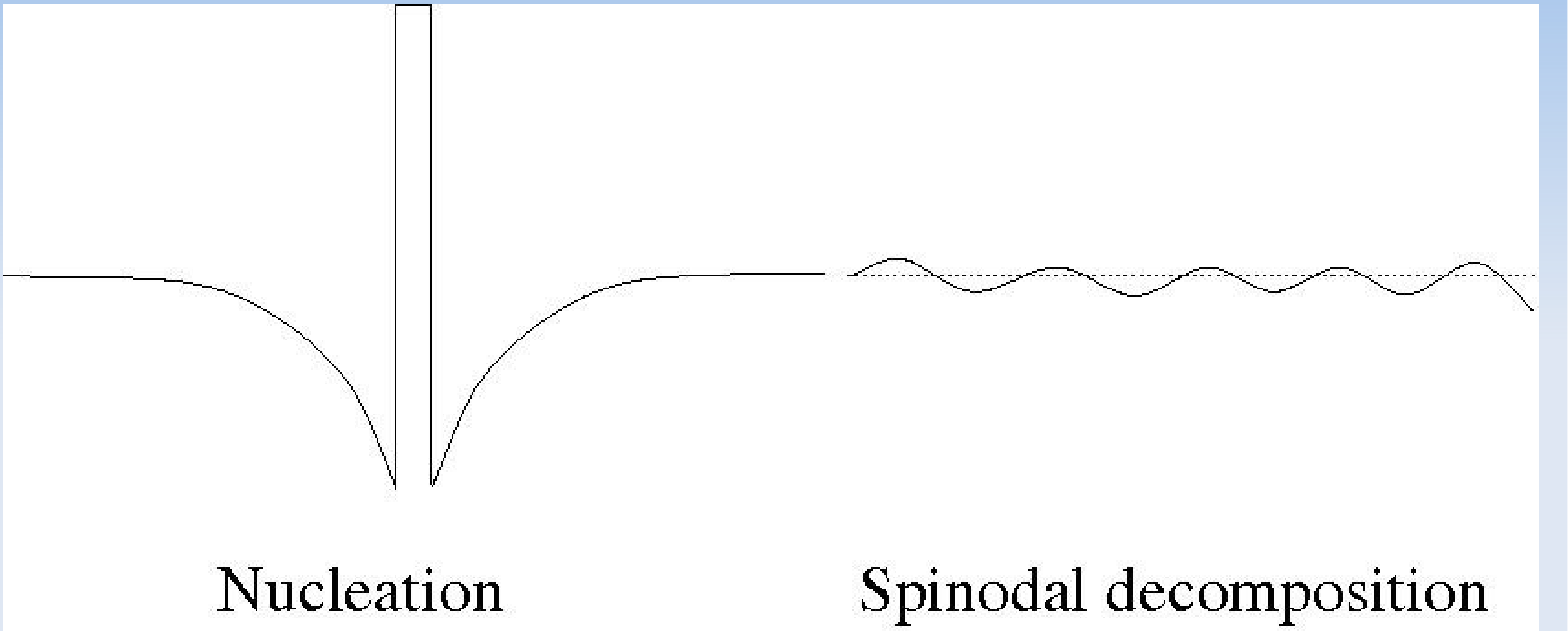
Miscibility gap



Free energy versus composition



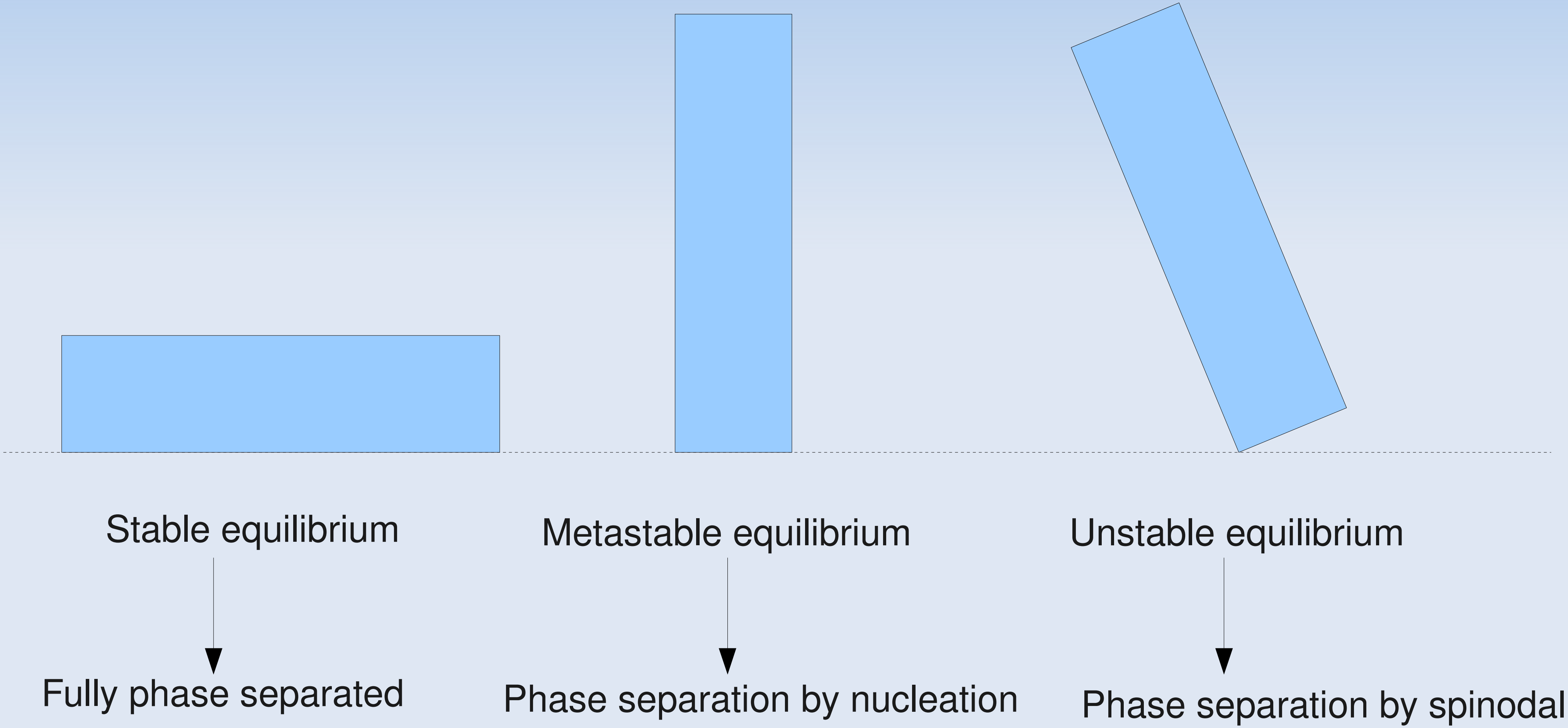
Mechanisms of phase separation



Large in degree and small in extent and small in degree but large in extent
-- J W Gibbs, Collected Scientific Papers

Mechanical analogue

Figure based on J W Cahn, Trans. Met. Soc. AIME, Vol. 242, pp. 166-180, 1968



Stable equilibrium → Fully phase separated

Metastable equilibrium → Phase separation by nucleation

Unstable equilibrium → Phase separation by spinodal

Diffusion equation

- Fick's first law: flux is proportional to concentration gradient

$$J = -M \nabla c$$

- Continuity equation:

$$\frac{\partial c}{\partial t} = -\nabla \bullet J$$

- Diffusion equation:

$$\frac{\partial c}{\partial t} = M \nabla^2 c$$

Diffusion equation: Alternate derivation

- Flux is proportional to chemical potential gradient

$$J = -M \nabla \mu$$

- Continuity equation:

$$\frac{\partial c}{\partial t} = -\nabla \bullet J$$

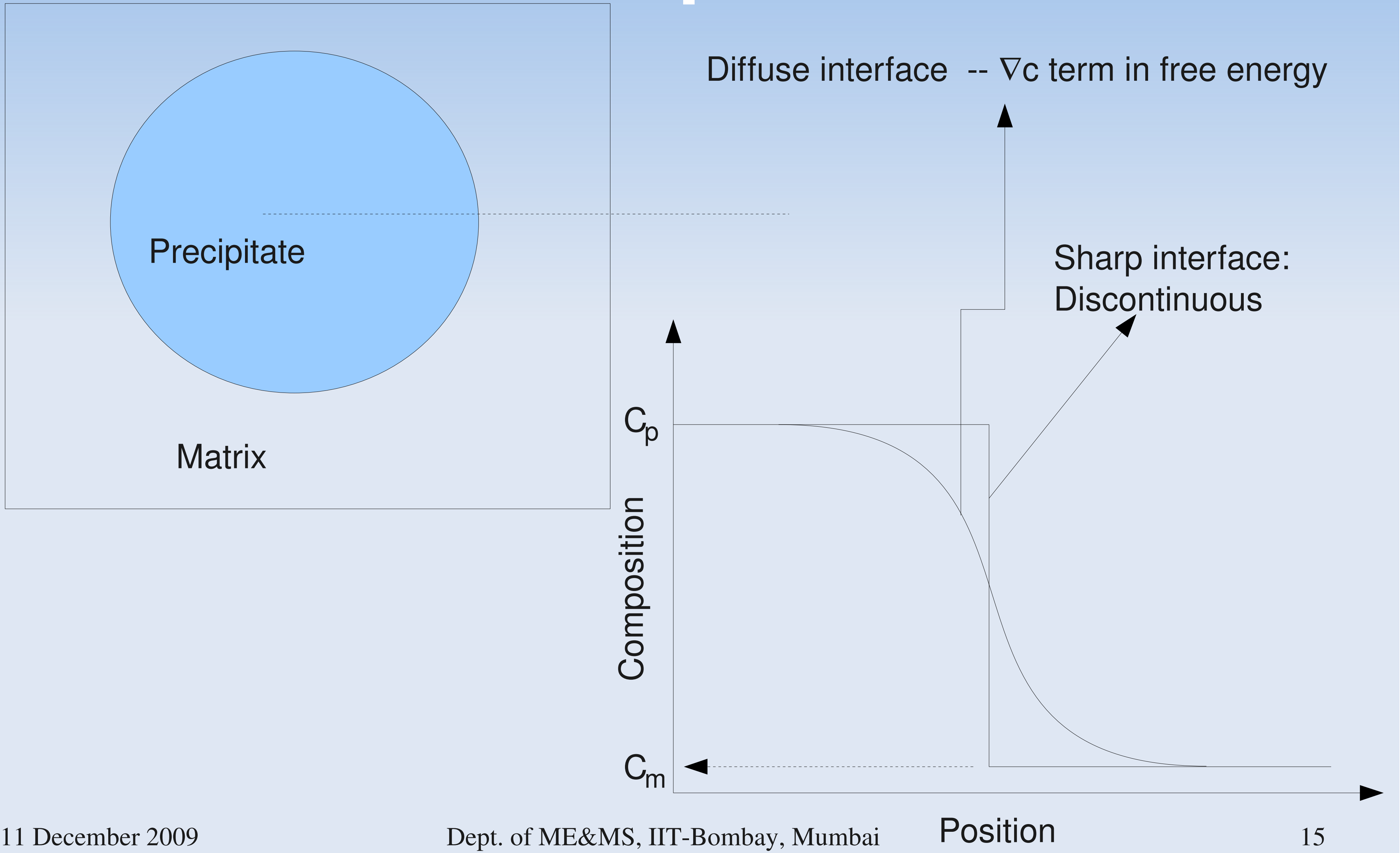
- Diffusion equation:

$$\frac{\partial c}{\partial t} = M \nabla^2 \mu$$

Modified diffusion equation

- Diffusion equation: $$\frac{\partial c}{\partial t} = M \nabla^2 \mu$$
- Chemical potential: $$\mu = \frac{1}{N_V} \left[\frac{\delta G}{\delta c} \right]$$
- G = Ḡ (c): Classical (or sharp interface) diffusion equation
- G = Ḡ (c, ∇c): Cahn-Hilliard (or diffuse interface) diffusion equation

Sharp versus diffuse interface description



Phase field model

Nothing but diffusion equation modified to account for interfaces!

Generalization

What is the general recipe for formulating a phase field model?

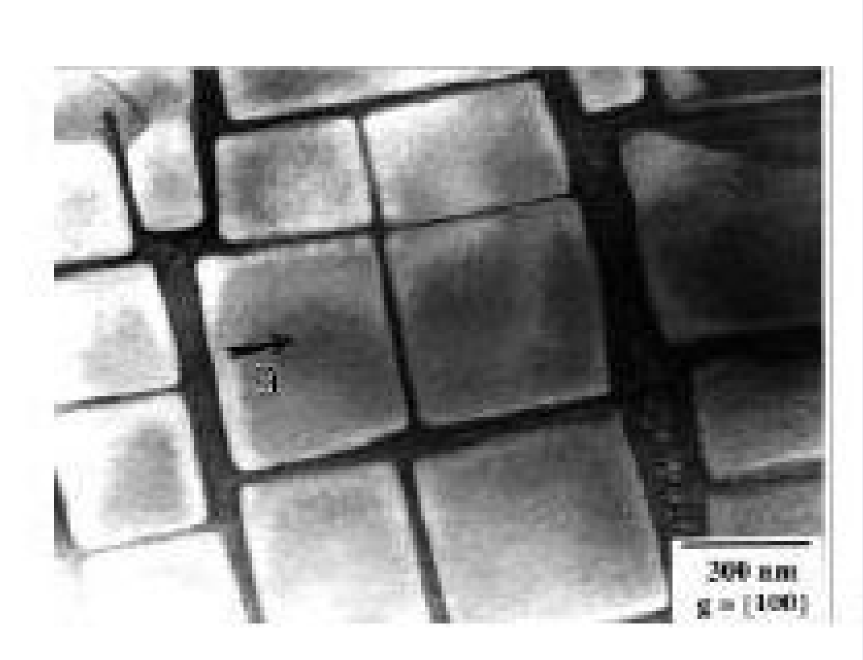
J E Hilliard, Chapter 12, Spinodal decomposition, in Phase transformations, AMS, 1970

A very nice pedagogical review!

“Derivations of the important expressions are given in full, on the premise that it is easier for a reader to skip a step than it is for another to bridge the algebraic gap between “it is easily shown that” and the ensuing equation.”

Step 1: Microstructure

- Describe microstructure (in terms of field variables -- order parameters): composition, for example



Step 2: Free energy

- Write the free energy of the system as a functional of order parameters and their gradients: Thermodynamics

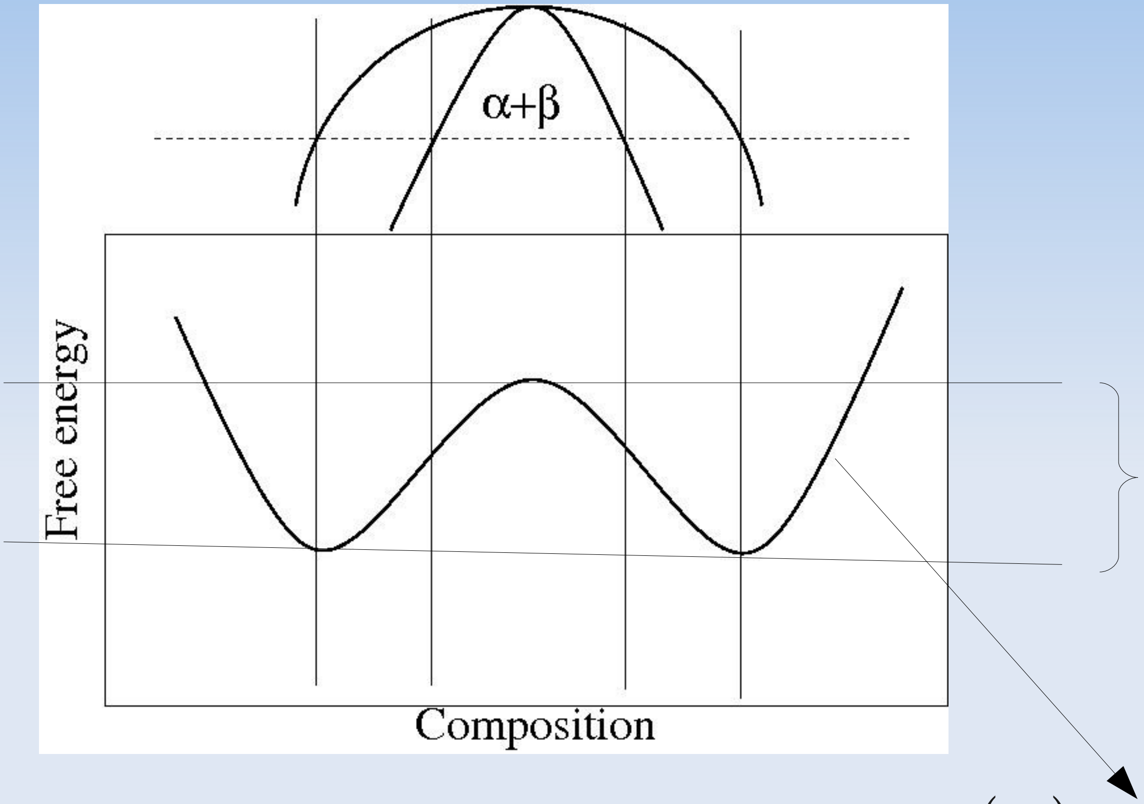
$$G=\bar{G}(c,\nabla c)$$

$$G=\frac{1}{N_V}\int\left[f_0(c)+\kappa(\nabla c)^2\right]dV$$

Double-well potential

Gradient energy

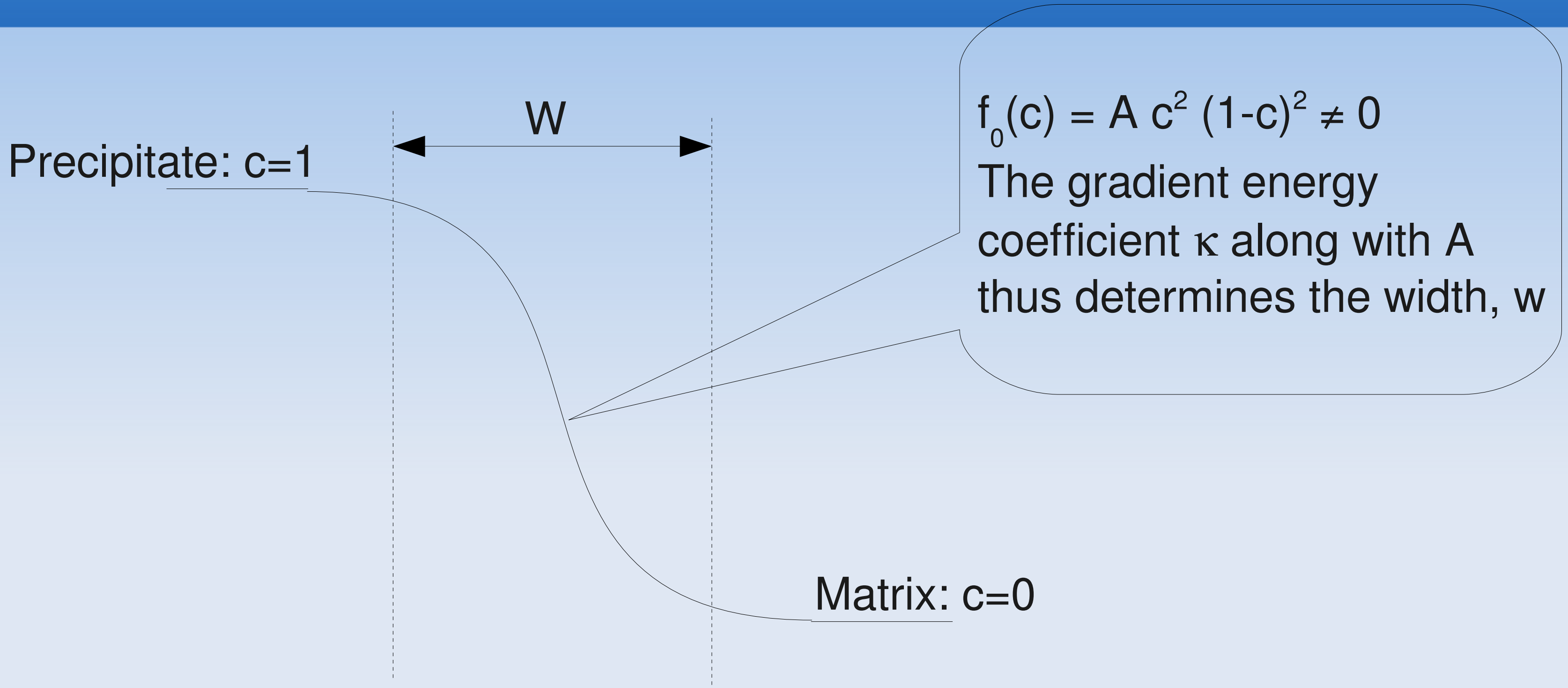
Double well potential



A, Barrier height

$$f_0(c)=A\,c^2(1-c)^2$$

Interface width



$f_0(c) = A\, c^2\, (1-c)^2 \neq 0$

The gradient energy coefficient κ along with A thus determines the width, w

$f_0(c) = A\, c^2\, (1-c)^2 = 0$ for matrix (c=0) and precipitate (c=1)

Step 3: Extremisation

- Minimise the free energy (incorporating the constraints if any)
- Extremise $G=\frac{1}{N_V}\int\left[f_0(c)+\kappa(\nabla c)^2\right]dV$

  subject to the constraint $\int(c-c_0)\,dV=0$
  where c_0 is the average alloy composition
- Euler-Lagrange equation

$$\mu=\left[\frac{\partial f_0(c)}{\partial c}\right]-2\kappa(\nabla^2 c)$$

- μ: Lagrangian multiplier (Chemical potential)

Step 4: Kinetics

- At equilibrium, the gradients in chemical potential are zero
- When the system evolves towards its equilibrium, the flux is proportional to the gradient in chemical potential

$$J = -M \nabla \left\{ \left[\frac{\partial f_0(c)}{\partial c} \right] - 2\kappa (\nabla^2 c) \right\}$$

- Continuity equation

$$\frac{\partial c}{\partial t} = -\nabla \bullet J = M \nabla^2 \left\{ \left[\frac{\partial f_0(c)}{\partial c} \right] - 2\kappa (\nabla^2 c) \right\}$$

Cahn-Hilliard: non-linear diffusion equation

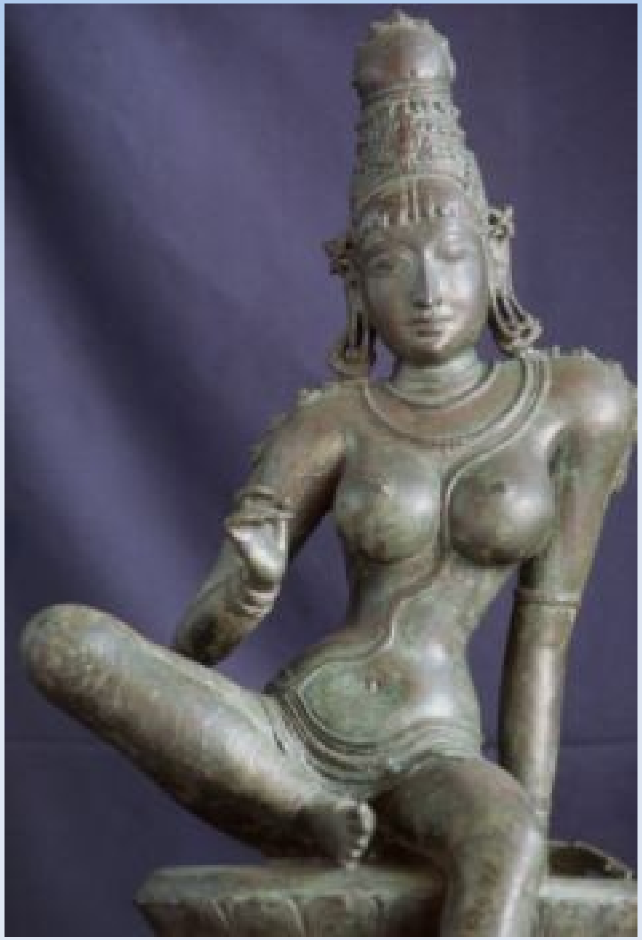
$$\frac{\partial c}{\partial t} = -\nabla \bullet J = M \nabla^2 \left\{ \left[\frac{\partial f_0(c)}{\partial c} \right] - 2\kappa \left(\nabla^2 c \right) \right\}$$

$$\frac{\partial c}{\partial t} = M \nabla^2 c$$

Allen-Cahn or Time Dependent Ginzburg-Landau equations

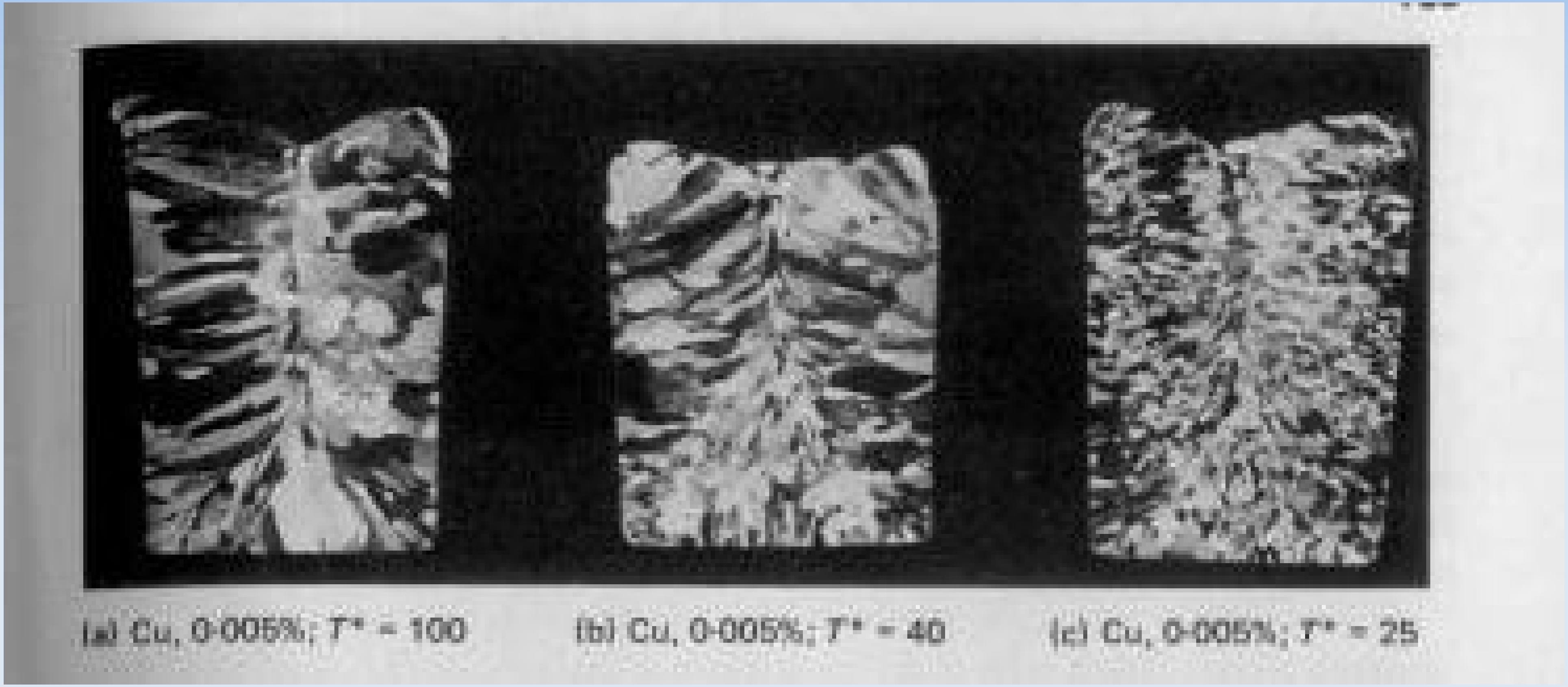
Casting

Cast turbocharger turbine
Image courtesy: MarkBolton
http://en.wikipedia.org/wiki/File:Investment_casting_-_turbocharger_turbine.jpg



Parvati: Chola Bronze
Image courtesy: Benoy K Behl,
Frontline, Aug11-24, 2007

Microstructure: undercooling



(a) Cu, 0·005%; $T^* = 100$ (b) Cu, 0·005%; $T^* = 40$ (c) Cu, 0·005%; $T^* = 25$

Grain structure in cast Al-Cu alloys: Chadwick, Metallography of phase transformations, Butterworths, 1972

Microstructure: composition

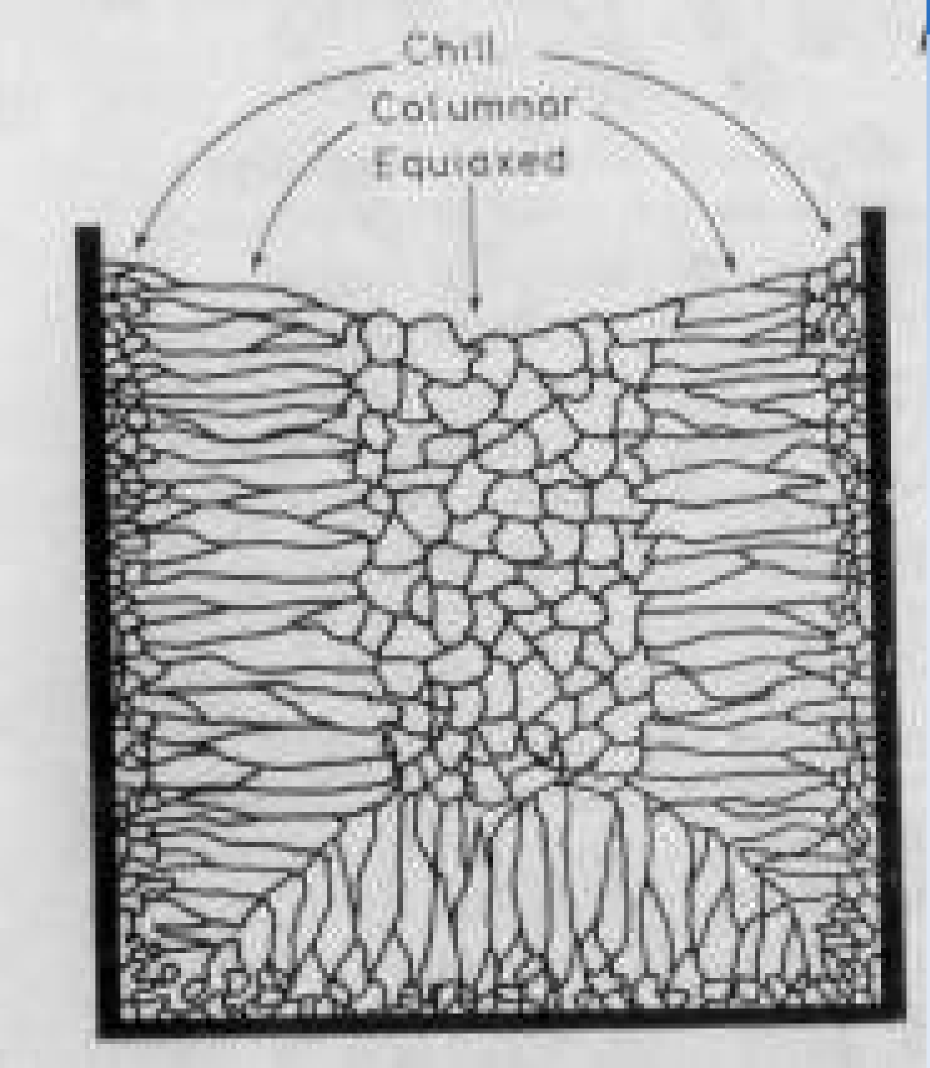
(a) Cu, 0·005%; T^* = 100

(d) Cu, 0·05%; T^* = 100

(g) Cu, 0·5%; T^* = 100

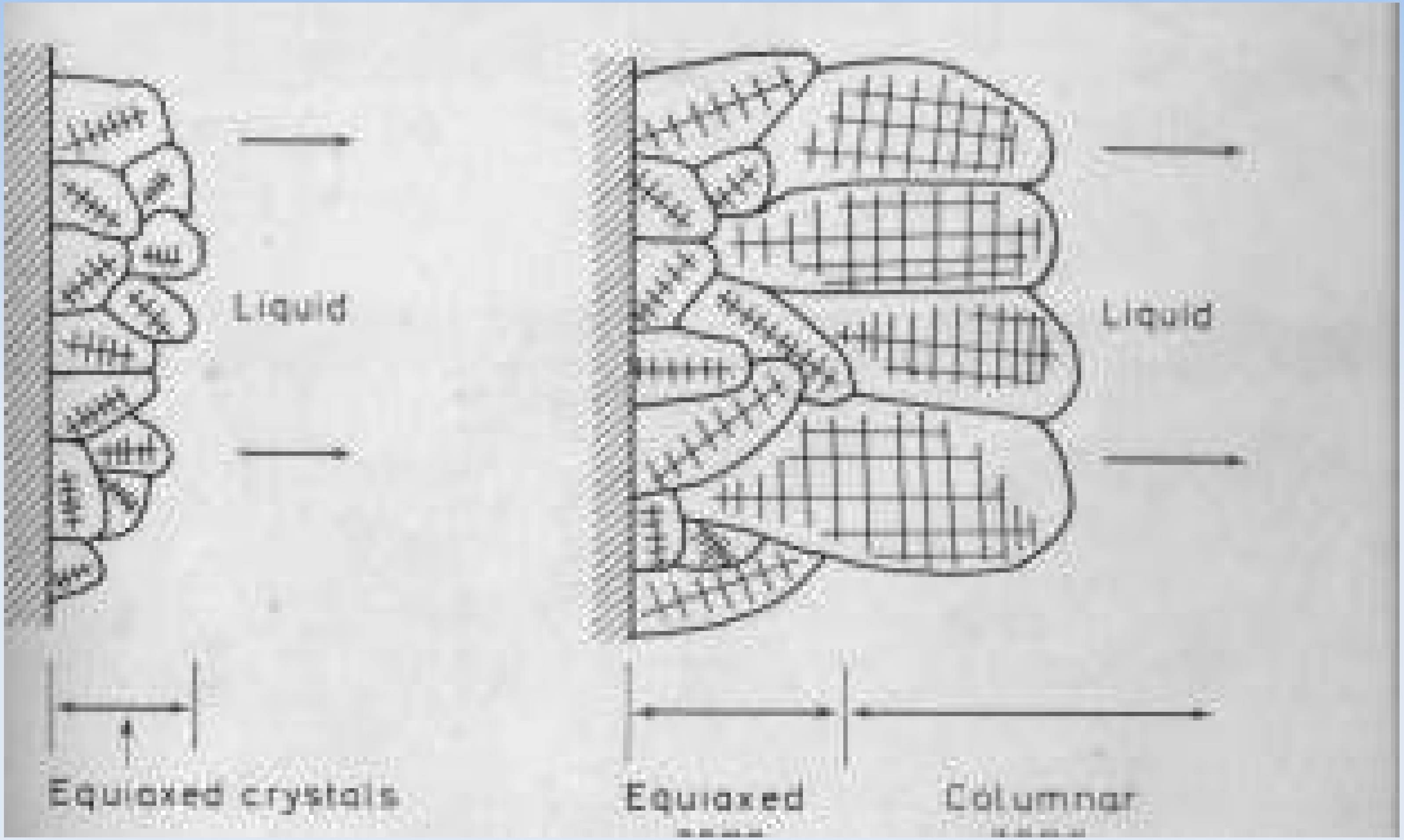
Grain structure in cast Al-Cu alloys: Chadwick, Metallography of phase transformations, Butterworths, 1972

Casting: microstructure



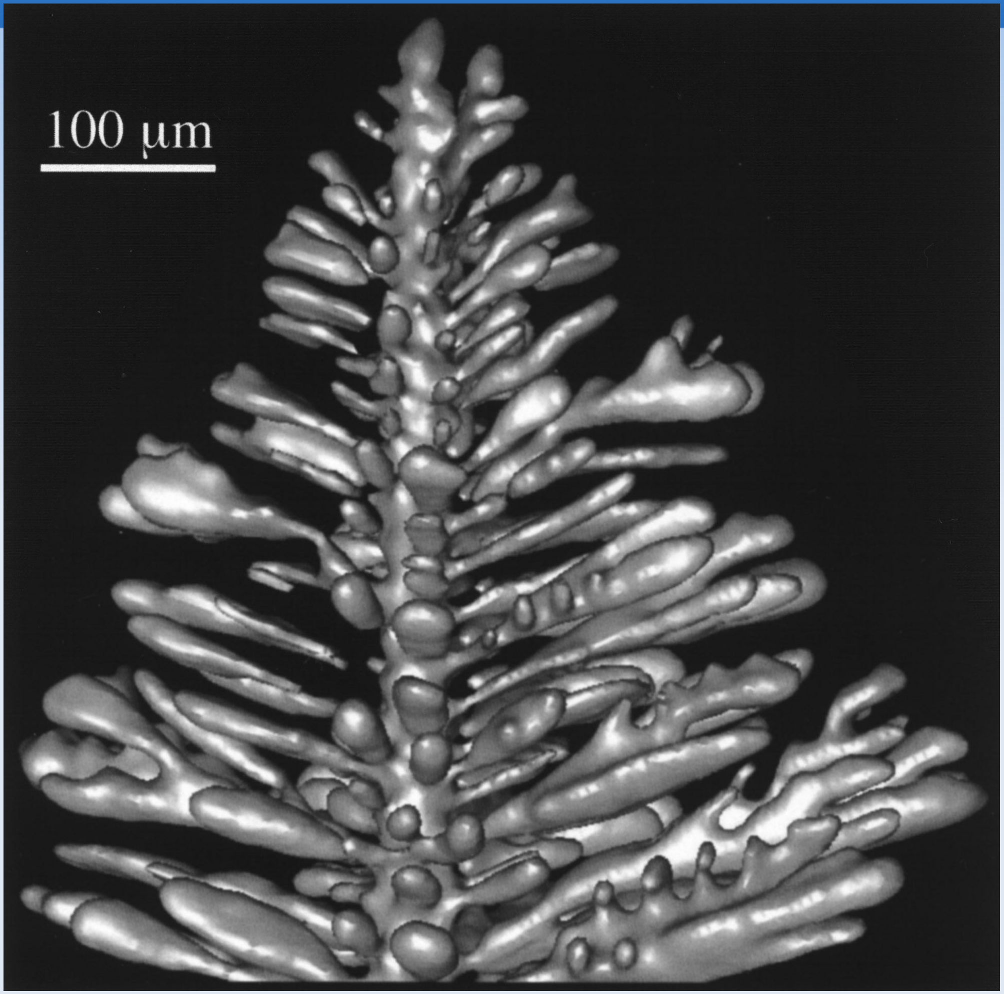
Cast microstructures - Schematic: Chadwick, Metallography of phase transformations, Butterworths, 1972

Dendritic structure



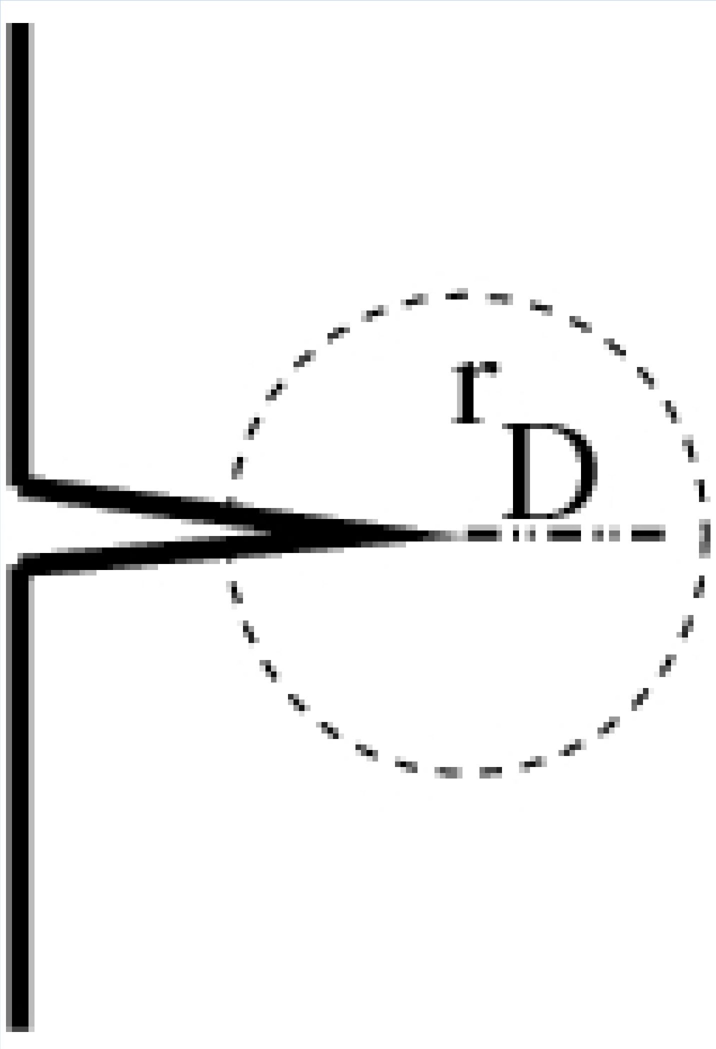
Cast microstructures - Schematic: Chadwick, Metallography of phase transformations, Butterworths, 1972

Dendritic microstructure



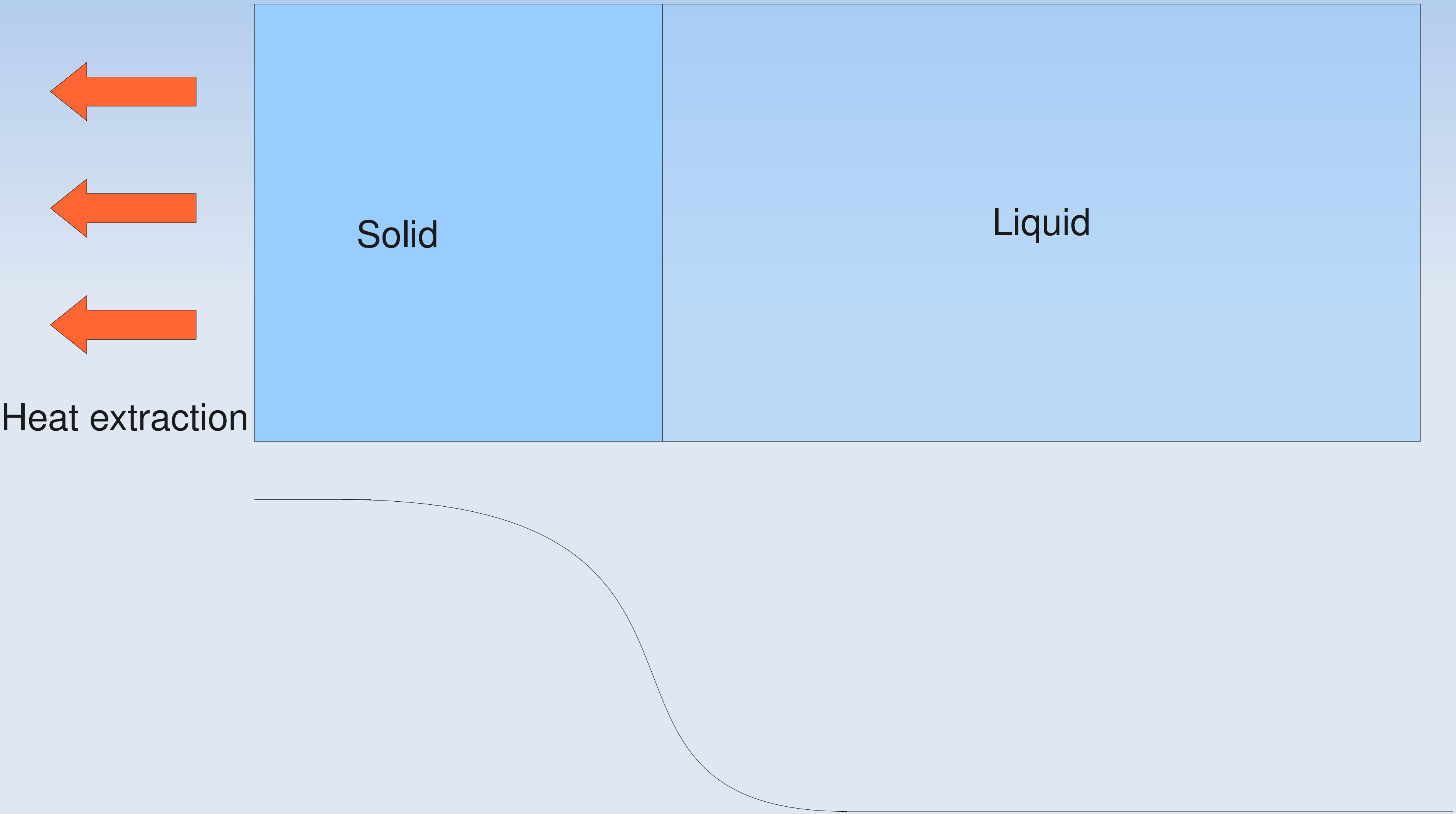
Al dendrite in Al-Cu alloy: Mendoza et al, Metallurgical and Materials Transactions A, Vol. 34A, p.481-489, 2003.

Why dendrites?



- Bunching of isothermal lines ahead of the interface
- Point effect of diffusion – interface instability
- Length scale: set by interfacial energy and the point effect

Order parameter – not conserved



Solid

Liquid

Heat extraction

Evolution equation for non-conserved order parameters

$$\frac{\partial \phi}{\partial t} = -L\left\{\left[\frac{\partial f_0(\phi)}{\partial \phi}\right] - 2\kappa\left(\nabla^2 \phi\right)\right\}$$

Reaction-Diffusion equation

A bit of history of Cahn-Hilliard equation

- Mats Hillert: atomistic (mean-field model) Incipient interfaces are important – MIT – PhD Thesis
- J E Hilliard
- J W Cahn: continuum version of Hillert's theory
- Mean field theory for phase separating systems: Cahn-Hilliard equation (Model B)

Phase field as coarse grained description

- Mean field theory for ordering systems: Allen-Cahn equation
- Microscopic theory for superconductors: Limiting case is Ginzburg-Landau theory (Gor'kov) – See Tinkham for example
- Ising Model: Glauber model, TDGL (Model A) – See Principles of condensed matter physics, Chaikin and Lubensky for example

Classical DFT and phase field models

- Classical density functional theory for the description of crystal-liquid interfaces: can be used to derive the Allen-Cahn equation
- *Phase-field model of interfaces in single-component systems derived from classical density functional theory,* G Pruessner and A P Sutton, Physical Review B 77, 054101, 2008

Phase field models as gradient flows w.r.t. inner products

- W C Carter et al, Variational methods for microstructural-evolution theories, JOM, pp. 30-36, December 1997.
- Take Lyapunov functions
- Use inner-products and norms on fields to measure kinetic distances between microstructures (Gradient flow)
- Evolve the microstructure in such a way that the Lyapunov functions change as fast as possible in a given time interval

Elastic stress driven microstructual evolution

ATG Instabilities

- History
- Experiments
- Stability analysis
- Modelling
- Phase field modelling of ATG instabilities in thin film assemblies
- Summary

History

- Gibbs (1876), Bridgman (1916) – Equilibrium between stressed solids and liquids
- Asaro-Tiller (1972) – Stressed solid and melt (Linear stability analysis)
- Grinfeld (1982) – Variational ideas of Gibbs (Thermodynamic result)
- ATG instability
- Srolovitz and co-workers: Solid-vapour and solid-solid interfaces
- Review in Solid state physics, Vol. 59 by Johnson and Voorhees

ATG instability: what do we mean by that?

- **ATG instabilities**

  The nominally flat surface/interface

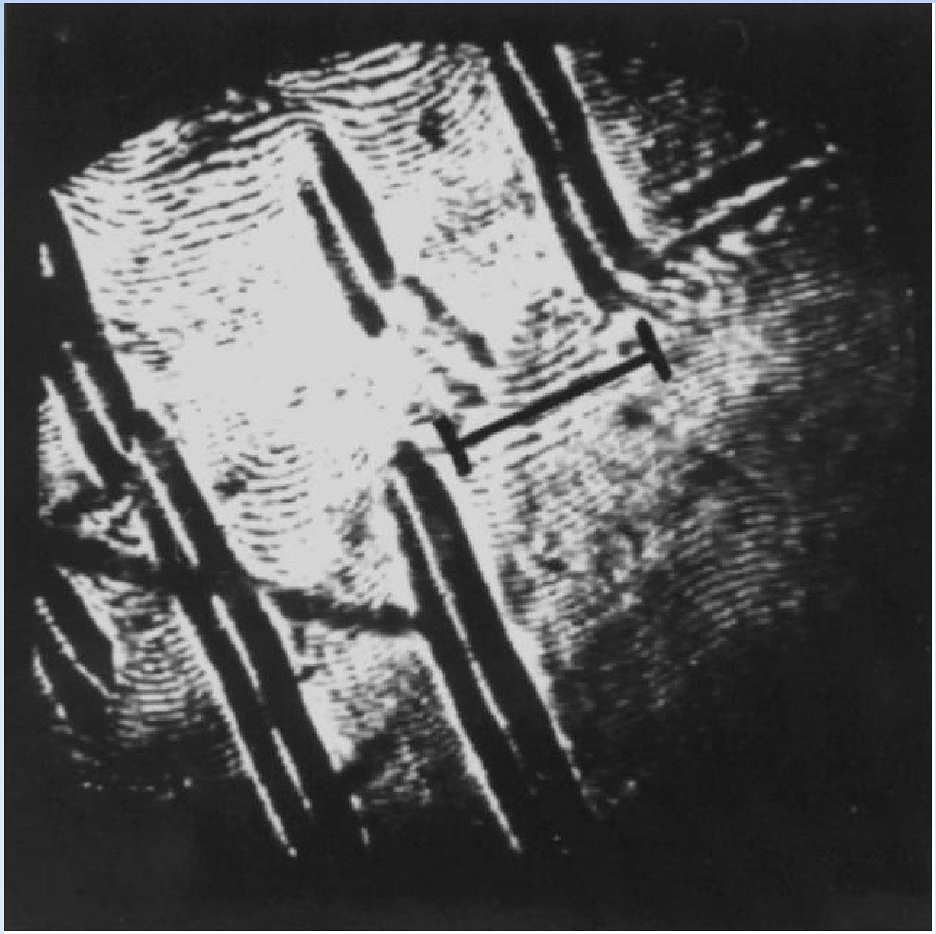
  of any non-hydrostatically stressed solid

  in contact with a compliant phase

  is unstable with respect to perturbations of wavelengths greater than a critical wavelength

Experiments!

Experiments: Solid-liquid interface in Helium IV



- Bondensohn et al, Z. Phys. B: Condens. Mat., 1986
- Length of scale bar on the image: $2\pi l_c$
- l_c: Capillary length
- Torii and Balibar, 1992: controlled experiments to conclusively prove Grinfeld instabilities

Image courtesy: Balibar et al, Rev. Mod. Phys., 2005

Experiments: Rippling of SiGe films on Silicon

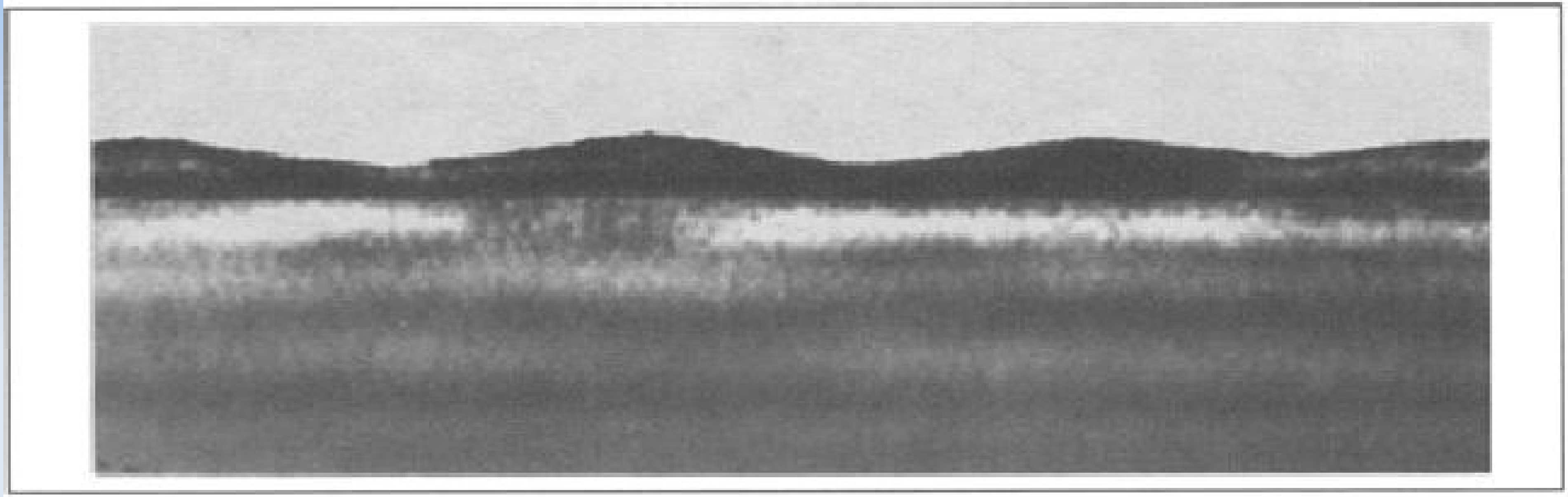
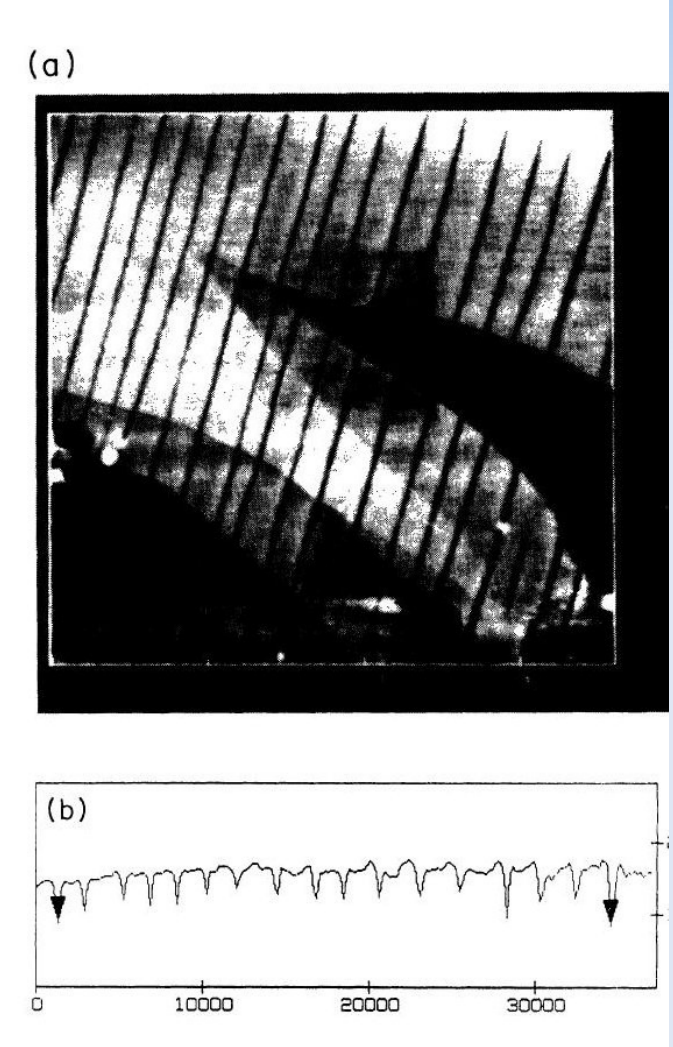


Fig. 1. Cross-sectional TEM image showing a typical rippled surface of a 10 nm thick $Si_{0.5}Ge_{0.5}$ film grown on Si (100) at 400°C and annealed in-situ at 560°C for 1 min.

- Note: no dislocations
- Stranski-Krastanov growth patterns leading to quantum dots!

Image courtesy: Jesson et al, J. Electron. Mat., 1997

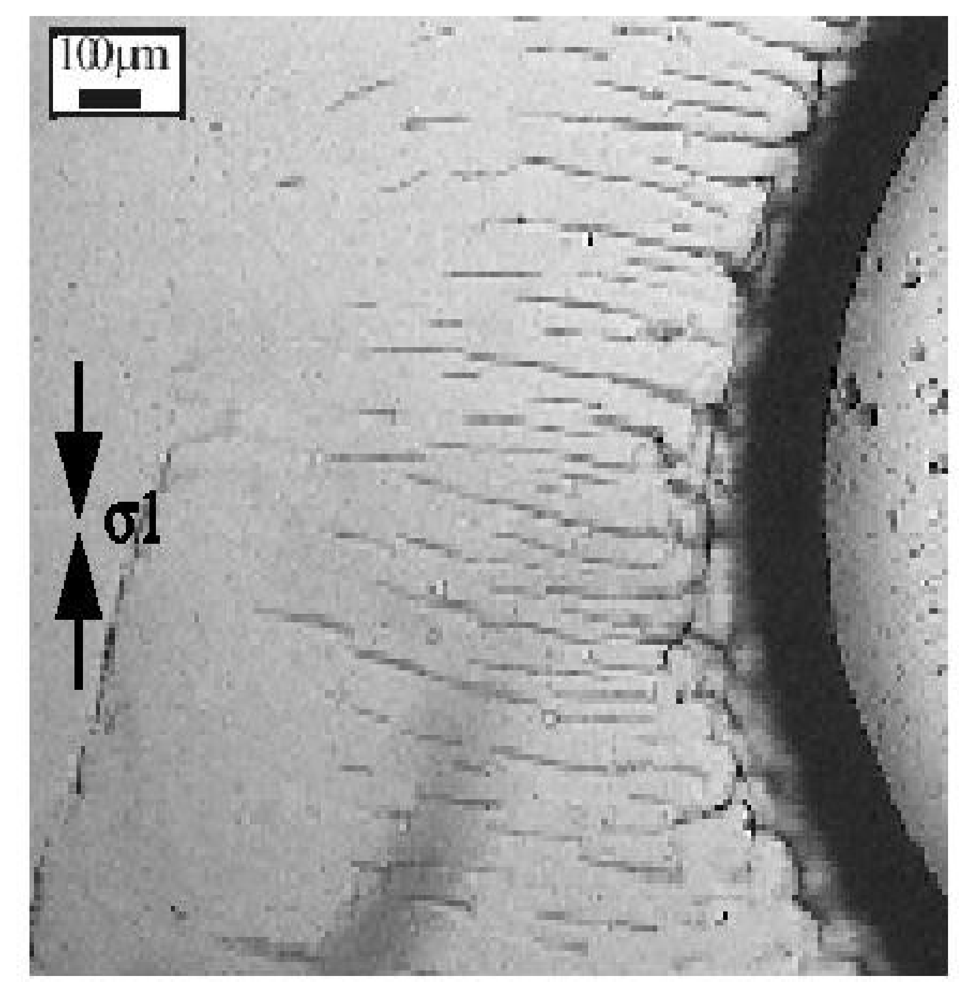
Experiments: Cracking in polymeric thin films



- Single crystal polymer film of polydiacetylene
- 310 nm thick film
- Stress due to polymerisation
- Distance between two arrows: 33.5 microns

Image courtesy: Berrehar et al, Phys. Rev. B, 1992

Experiments: stress induced cracking in minerals



- Stressed salt crystal with a hole (filled with a liquid)
- Den Brok et al, Deformation mechanisms, rheology and tectonics: Current status and Future Perspectives, *Geological Society,*

Image courtesy: D Koehn et al, American Journal of Science, 2003

Experiments: summary

- Wide range of materials undergo (static) ATG
- ATG – great practical interest: cracking of polymeric thin films, rippling and growth mechanisms in semi-conductor films, minerals in contact with fluids under stress, ...

Why ATG Instability?

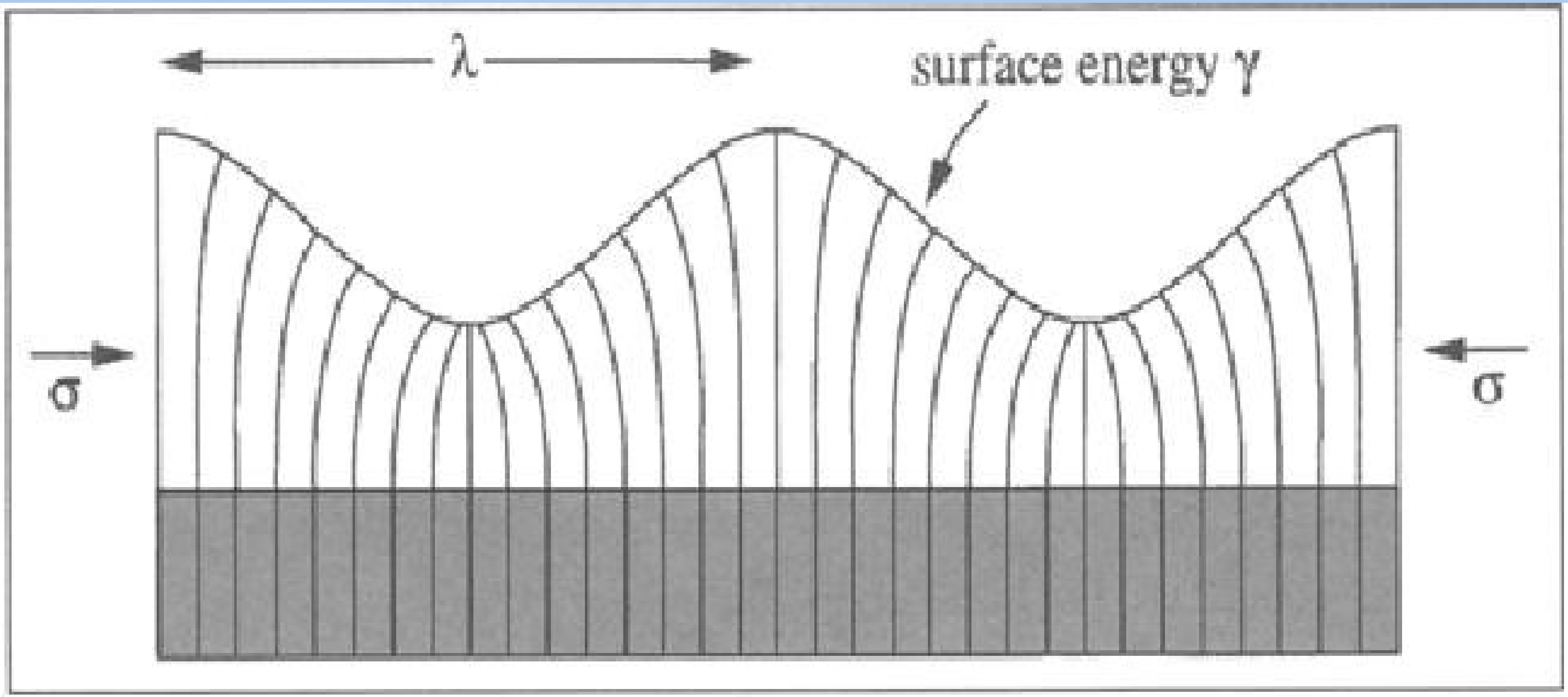
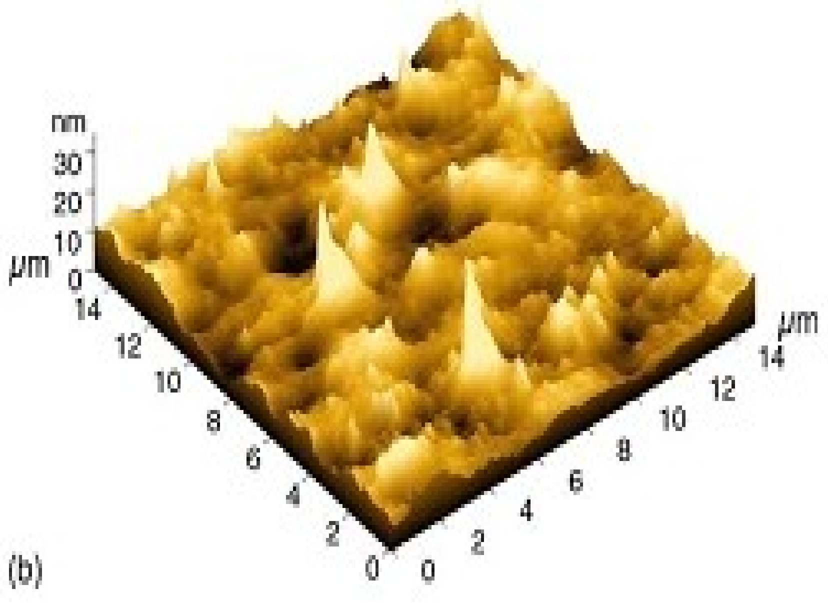


Fig. 2. Cross-sectional schematic illustrating the physical basis of the ATG instability.

Image courtesy: Jesson et al, J. Electron. Mat., 1997

Modelling

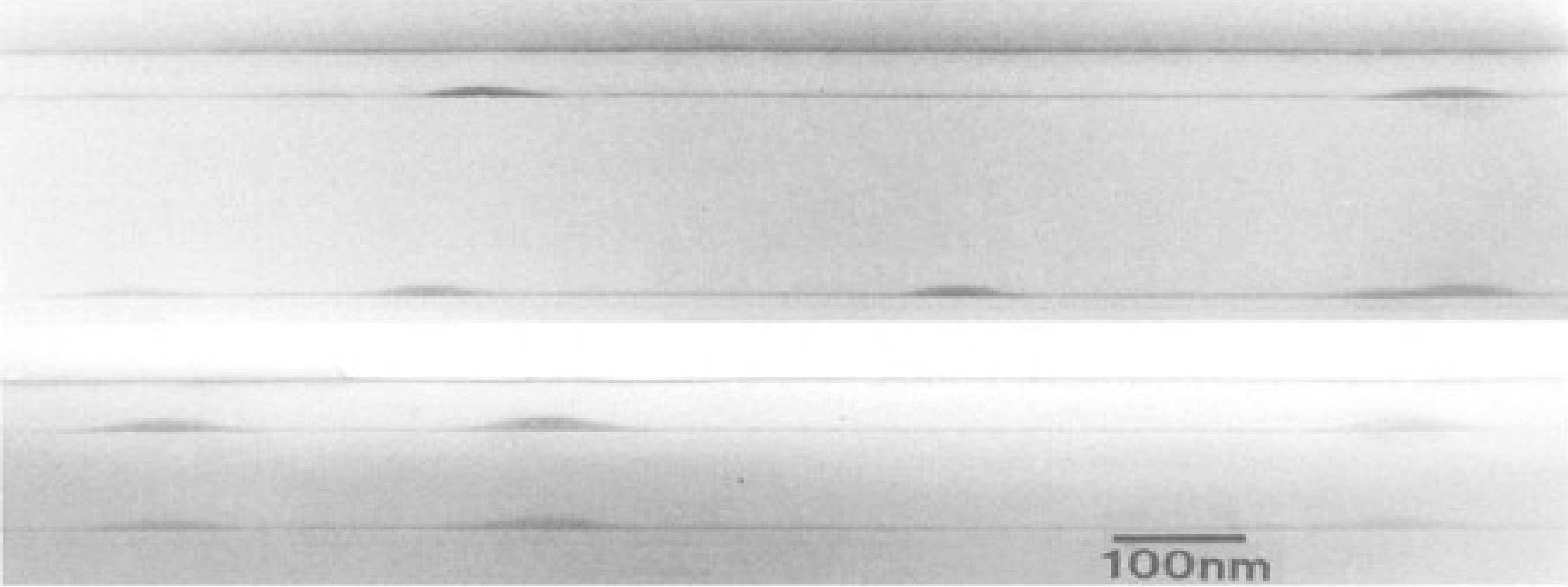
Gadolinia-silica multilayers



- Post-growth topographic changes
- Known to be sensitive to number of layers and layer geometry

Image courtesy: Sahoo et al, Appl. Surf. Sci., 2005

Ge films separated by Si



- Interlayer interactions
- Separation: 300 and 100 nm Si

Image courtesy: Rahmati et al, Appl. Phys. A, 1996

Why modelling?

- Linear stability analysis -- ideal for studying only the onset of instability
- Typically, our interest -- the evolution of the instability, break-up of the film and the eventual coarsening of the microstructure
- Correlations, for example: far too complicated to be treated in the linear stability analysis paradigm!

Advantages of diffuse interface description

- No tracking of interface
- Merger, splitting, disappearance, etc -- topological singularities -- naturally accounted for

Cahn-Hilliard equation

$$(\partial c/\partial t) = M\nabla^2\mu$$

$$\mu = (1/N_V)[\delta F/\delta c]$$

$$F = F^{ch} + F^{el}$$

Elastic term

$$F^{el} = (1/2) \int \sigma \epsilon \, dV$$

$$\nabla . \sigma = 0$$

- Solving equation of mechanical equilibrium under prescribed traction boundary conditions: homogenisation problem

Implementation

- Semi-implicit Fourier spectral technique for both the diffusion and homogenisation problem
- Details can be found in

  Phase field study of precipitate rafting under a uniaxial stress, M P Gururajan and T A Abinandanan, Acta Materialia, Vol. 55, 15, pp. 5015-5026 (September 2007)

  and at

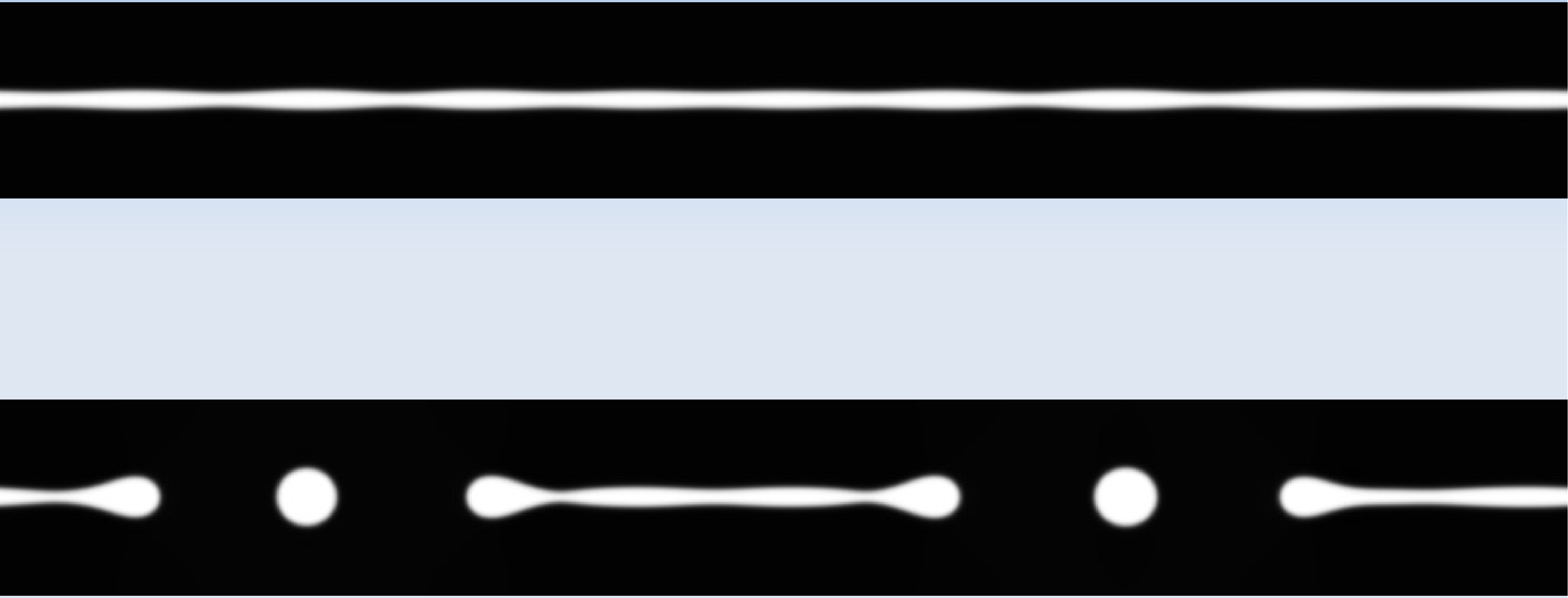
  http://imechanica.org/node/440
- Cahn-Hilliard and Allen-Cahn (without elasticity): can be downloaded from http://sites.google.com/site/gurusofficialhomepage/research/downloads/write-ups-and-codes/the-art-of-phase-field-modelling

Parameters

- δ: Elastic inhomogeneity
  Ratio of shear modulus of the film to that of the matrix
- h: thickness of the film
- H: inter-film spacing
- f: volume fraction of the film phase (h/H)
- A_Z : Zener anisotropy parameter

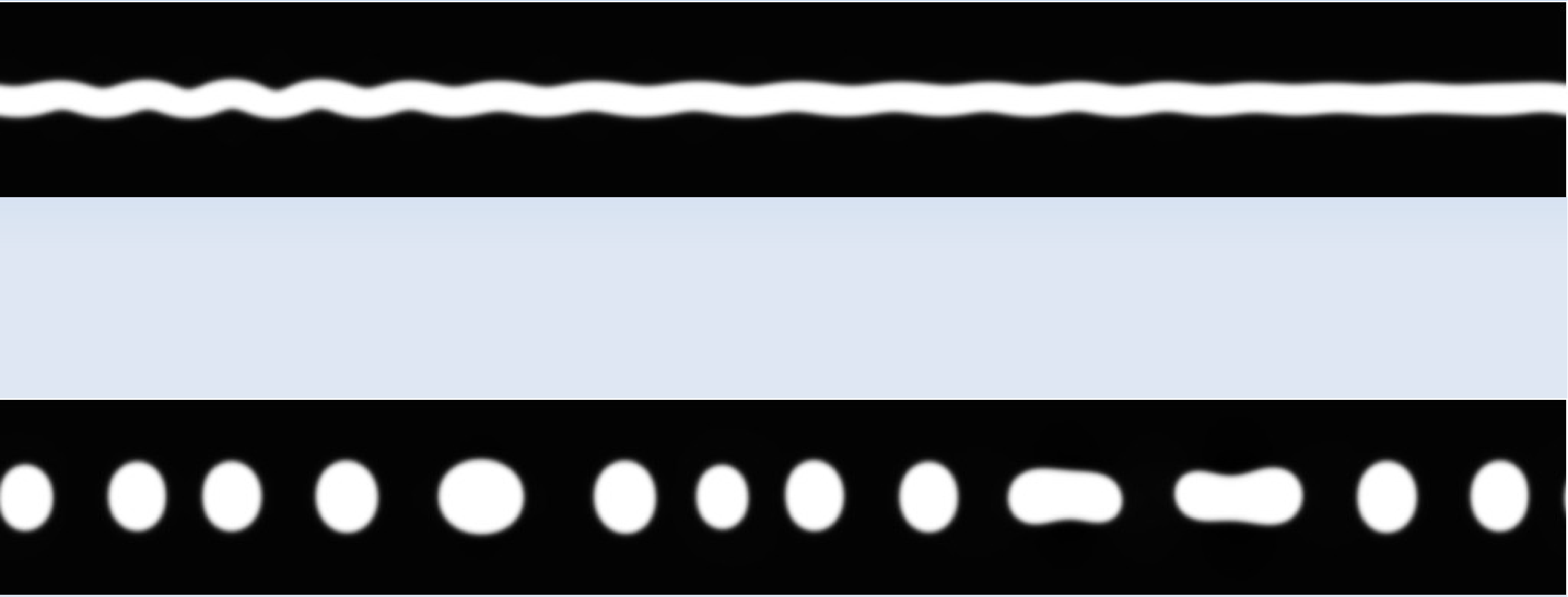
  A measure of elastic anisotropy

Symmetric break-up



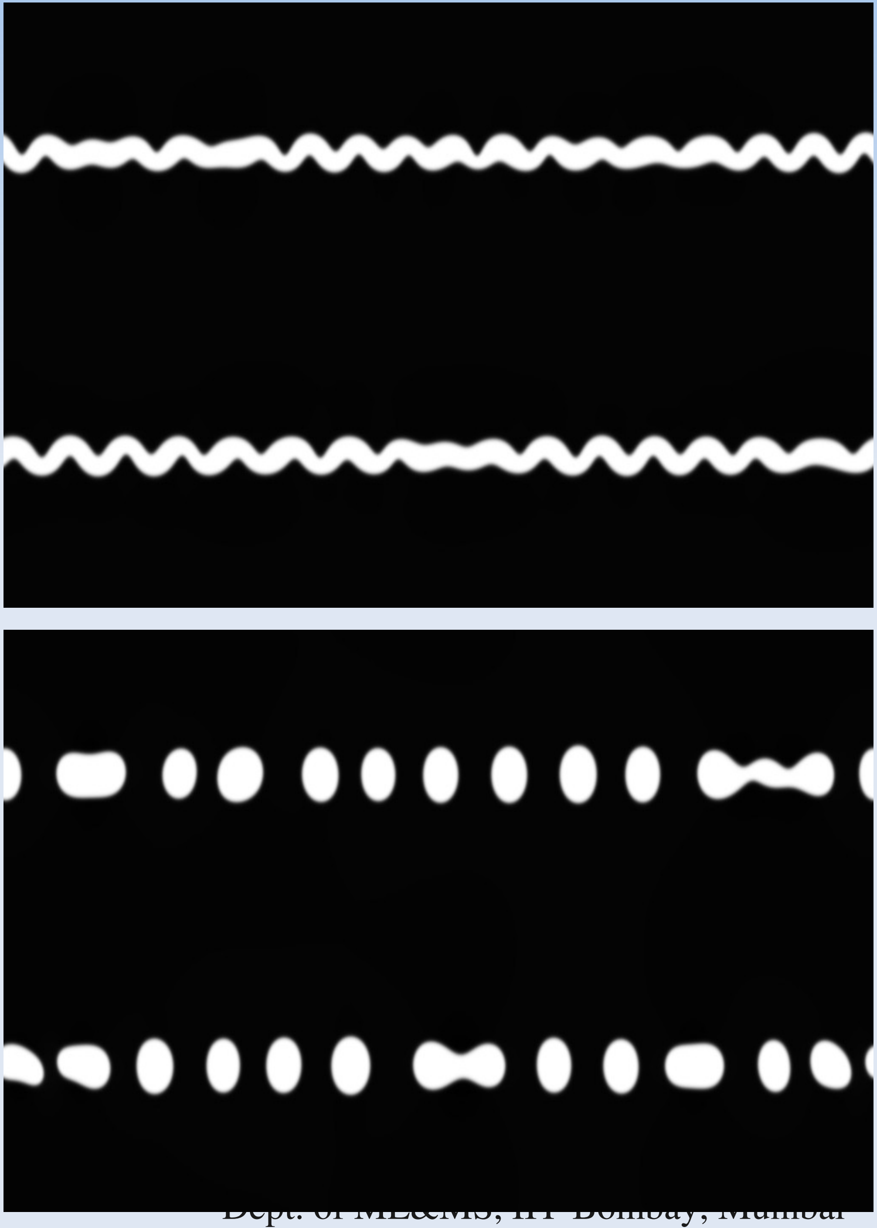
δ = 2; h = 11; times: 1,28,000 and 1,44,000

Anti-symmetric break-up



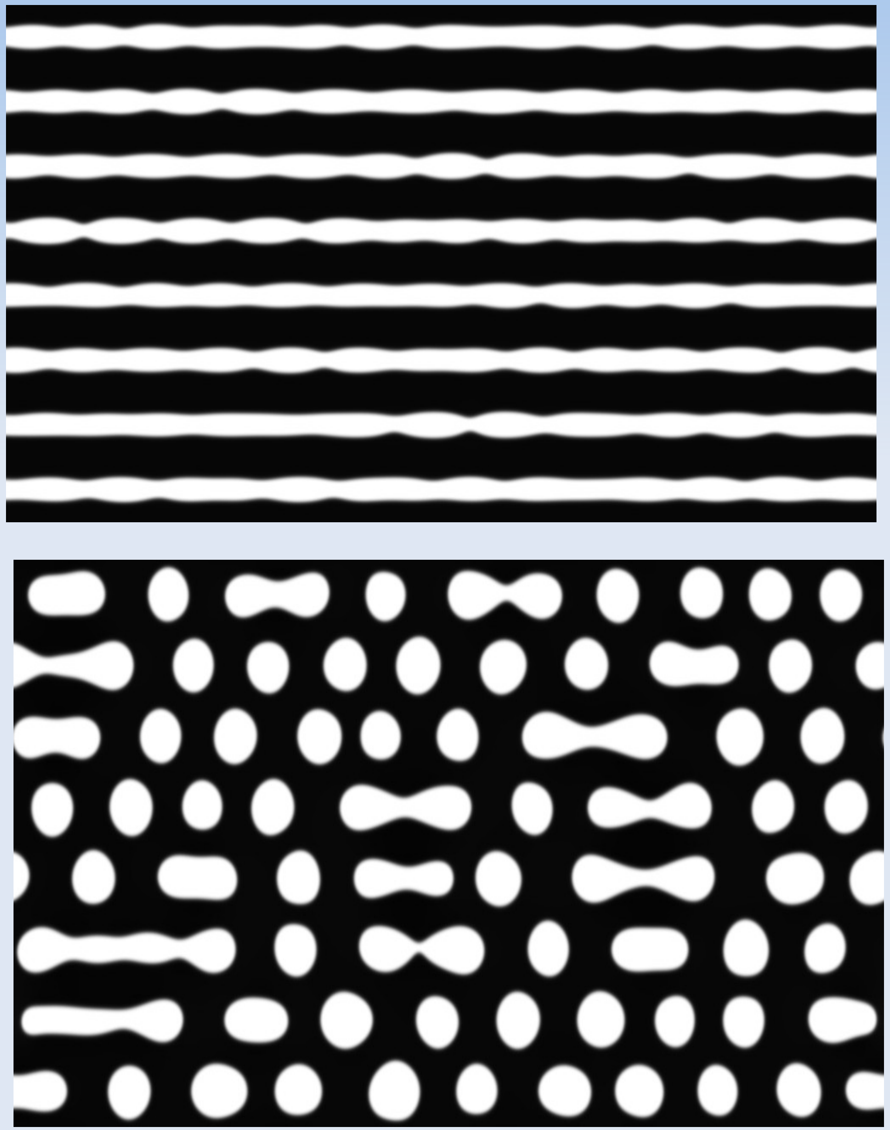
δ = 4; h = 22; times: 20,000 and 35000

Multilayers: two layers



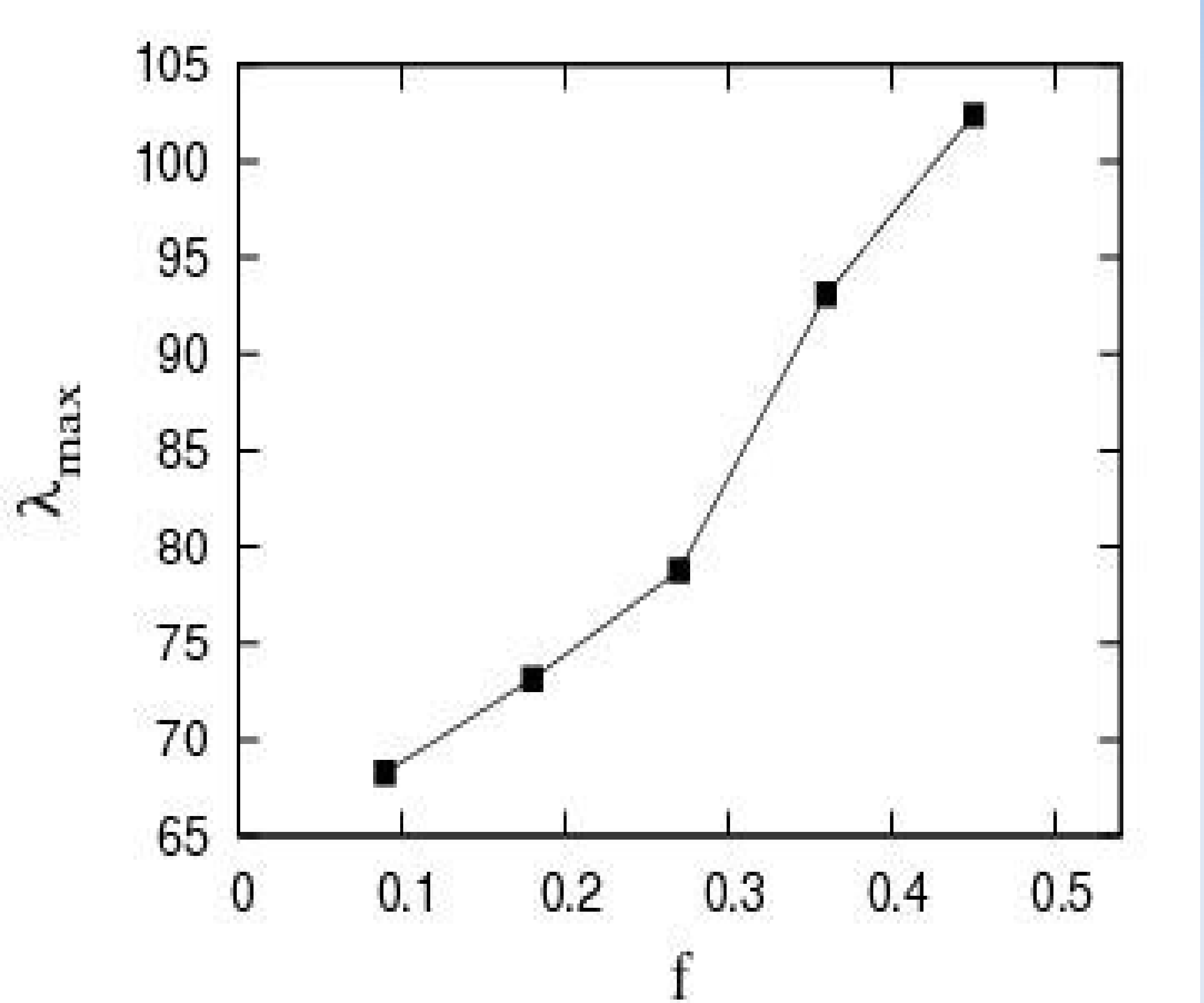
δ = 4; f = 0.086; times: 25,000 and 36,000

Multilayers: eight layers



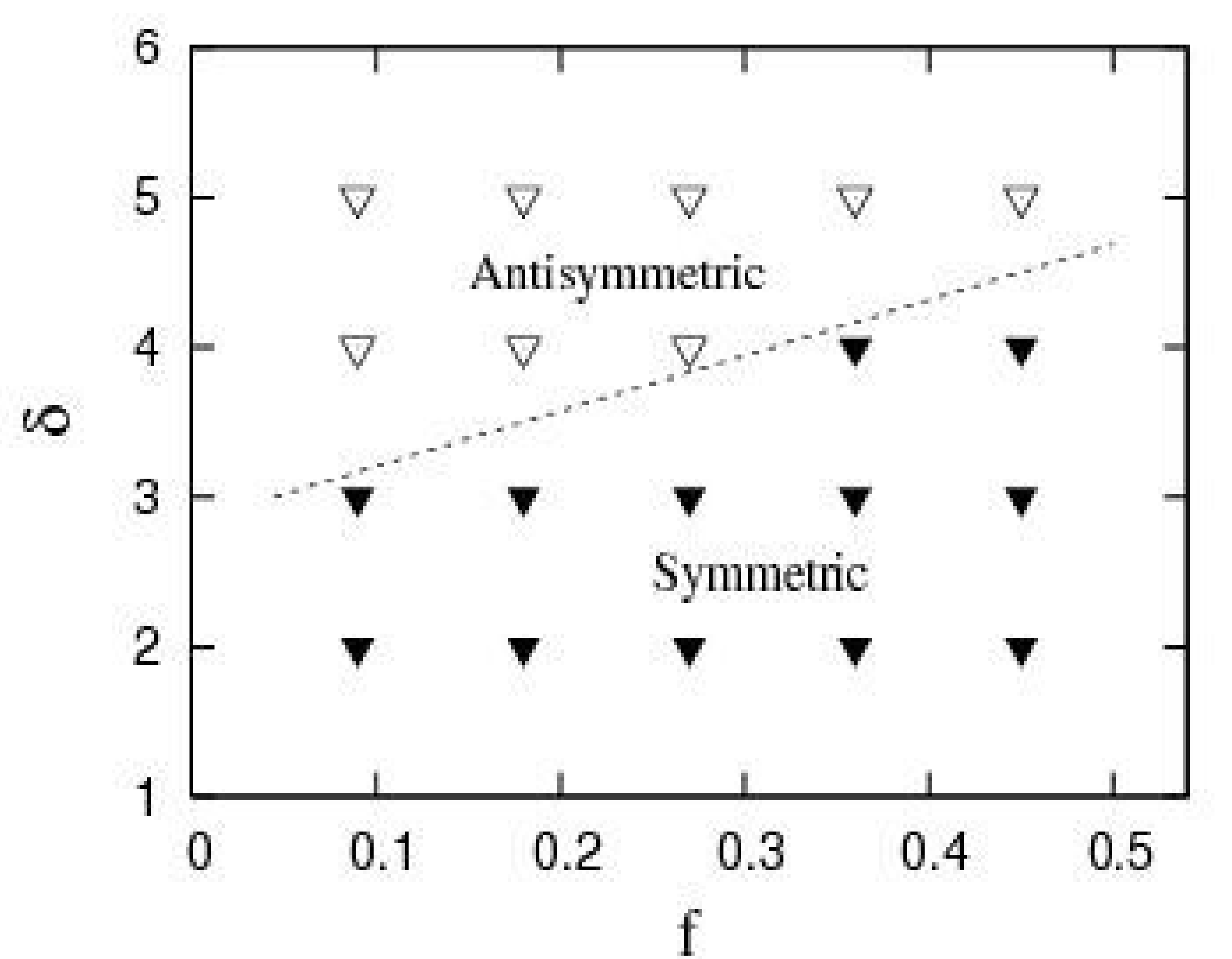
$\delta = 4$; f = 0.34; times: 36,000 and 49,000

Effect of f on λ_{max}



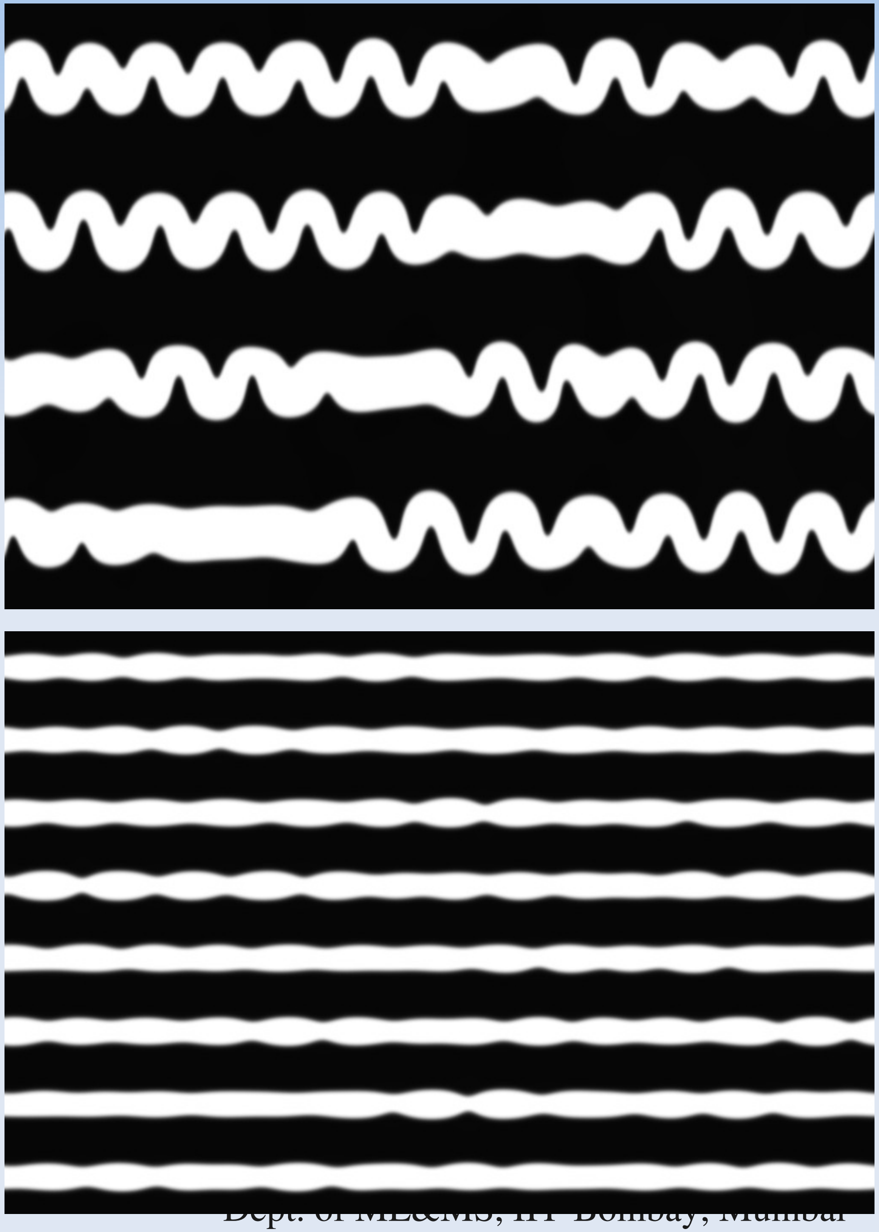
$\delta = 4; h = 22$

Stability diagram



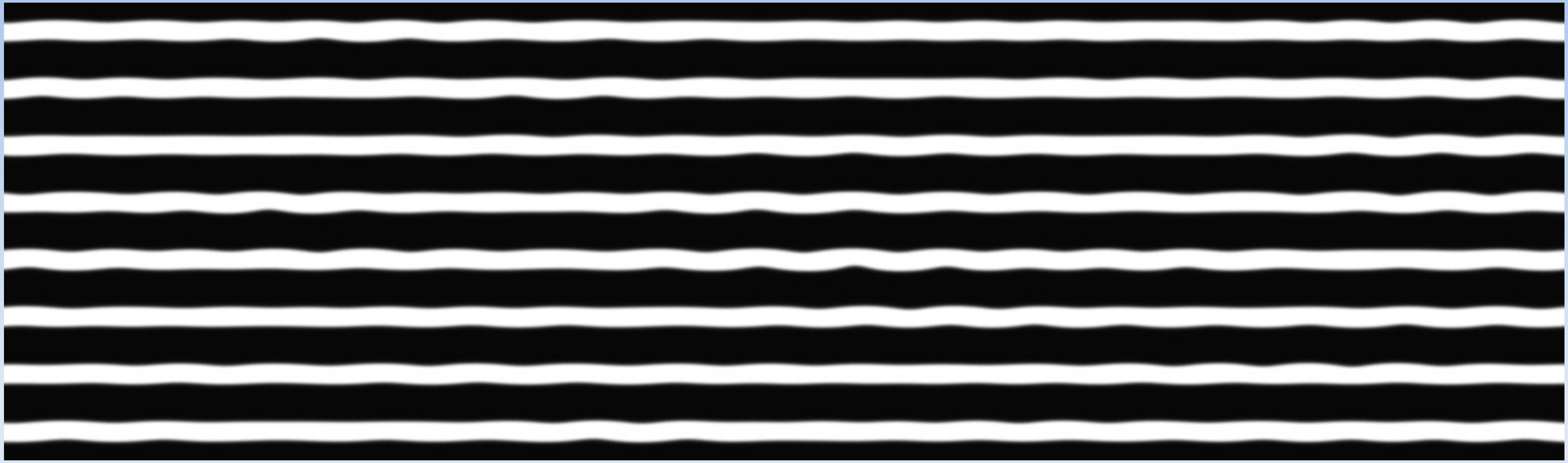
Systems with h = 22

Effect of h

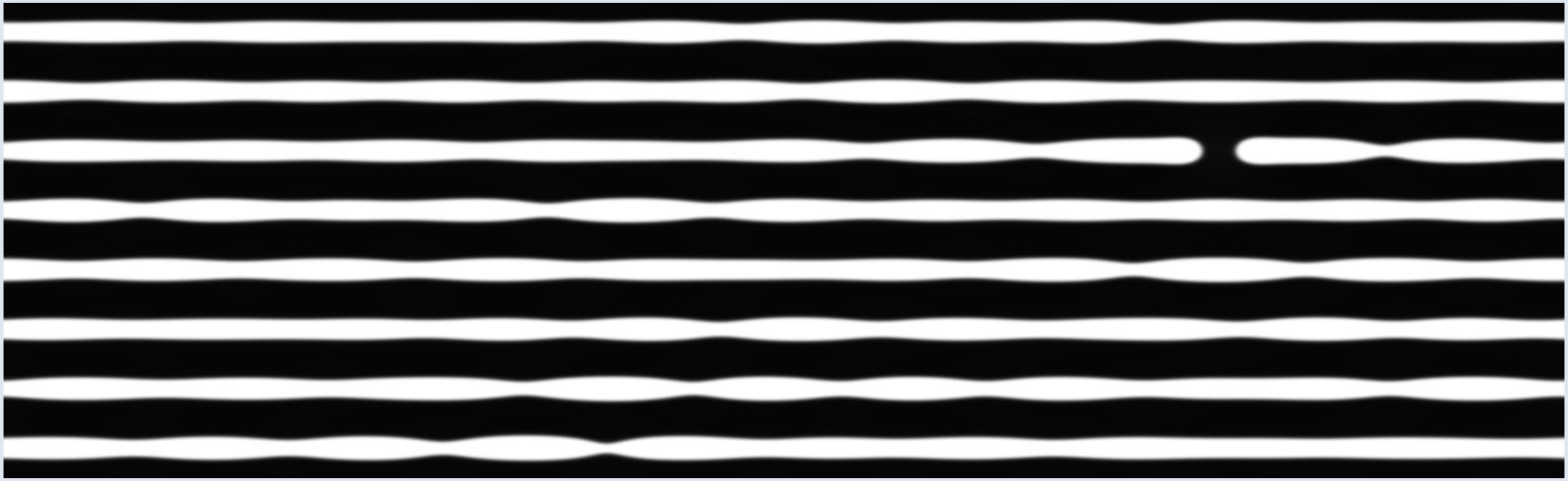


δ = 4; f = 0.35; times: 59,000 (h=45) and 36,000 (h=22)

Effect of A_Z



$A_Z = 0.8$



$A_Z = 1.2$

δ = 4; f = 0.34; h = 22; times: 16,000 and 64,000

Summary

- Phase field models: ideal for the study of microstructural evolution
- Phase field modelling: strictly speaking is a way of looking at problems
- Very good for quick and qualitative studies
- Integration with atomistic and macroscopic models: in progress

Thank you!

- Abi
- Shankara
- Ram
- Deep-da
- Sasoto
- Chirru
- Rajdip
- Peter
- Kuo-An
- Ferdi
- Volkswagen, CSIR-UGC, DAAD

For the sake of completion ...

Where do we obtain such pretty physical pictures as was shown in the last slide from?

A six slide detour into stability analysis

Perturbative analysis

- Consider the diffusion equation:

$$J=-M\nabla\Phi$$

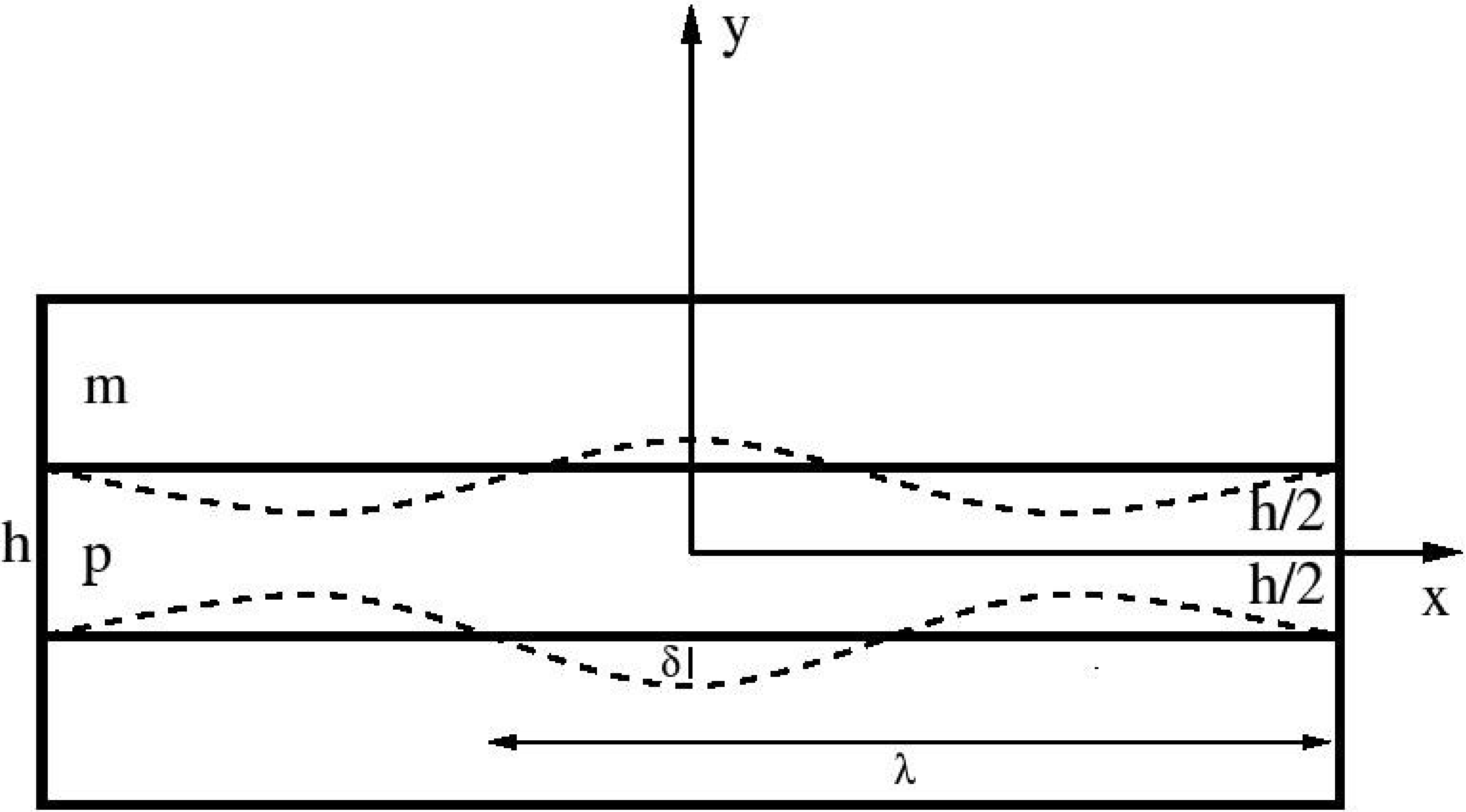
J: flux; M: mobility; Φ: diffusion potential

- Conservation of mass: kinematic constraint for the velocity of the interface normal to itself, namely, v_i:

$$v_i=-V_a\nabla J=MV_a\nabla^2\Phi$$

V_a : atomic volume

Geometry (perturbation)



Perturbative analysis

- Perturbed interface profile:

$$y_i(x)=\pm[(h/2)+\delta\cos(kx)]$$

- Another expression for interface velocity

$$v_i\simeq(\partial\delta/\partial t)\cos(kx)$$

- Final equation to be solved

$$(\partial\delta/\partial t)\cos(kx)=M V_a \nabla^2\Phi$$

Potential

$$\Phi = V_a(-\chi\gamma + \Phi^{elastic})$$

$$\Phi^{elastic} = (1/2)[\sigma^m \epsilon^m - \sigma^p(\epsilon^p - \epsilon^T)] - \sigma^m[\epsilon^p - \epsilon^m]$$

$$\Phi \propto \delta \cos(kx)$$

Growth rate

$$(\partial \delta / \partial t) \cos(kx) = G \delta \cos(kx)$$

$$\boxed{\delta(\tau) = \delta(0) \exp(\phi \tau)}$$

Non-dimensional time

Non-dimensional growth rate parameter

Growth rates

$$\phi = \phi_c + \phi_e$$

	Surface diffusion	Volume diffusion
ϕ_c	$-(kh)^4$	$-(kh)^3$
ϕ_e	$\theta f(\alpha, \omega)(kh)^3$	$\theta f(\alpha, \omega)(kh)^2$

$$\alpha = (E_p - E_m)/(E_p + E_m) \qquad \omega = kh/2$$

$$\theta = E_p h [\epsilon^T]^2 / \gamma$$

Growth rates

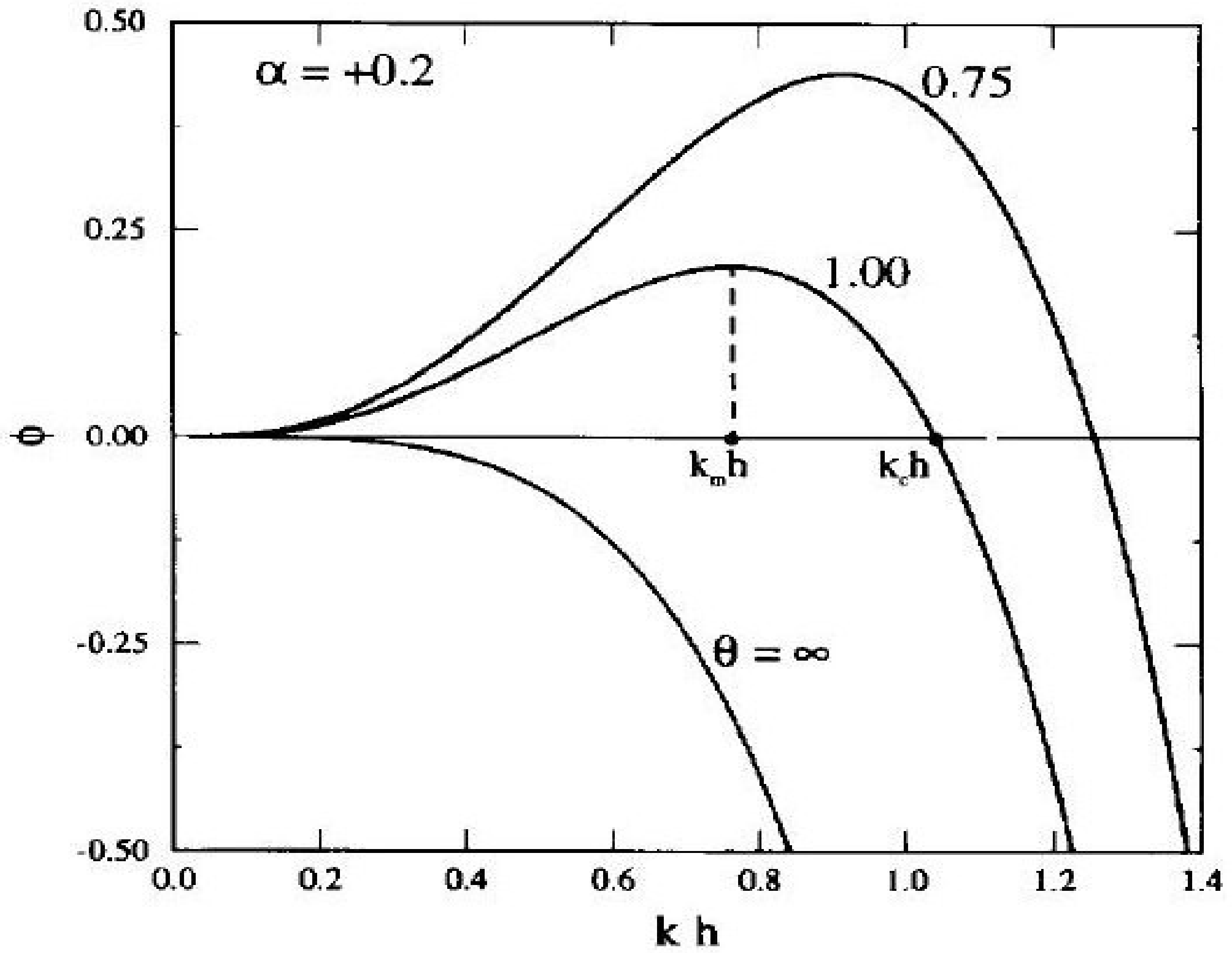


Fig. 2. The dimensionless perturbation amplitude growth rate ϕ as a function of the perturbation wavenumber kh for different values of the normalized interface energy $\theta(=\gamma/E_p h(\epsilon^*)^2)$.

Image courtesy: Sridhar et al, Acta Materialia, 1997

Types of instabilities

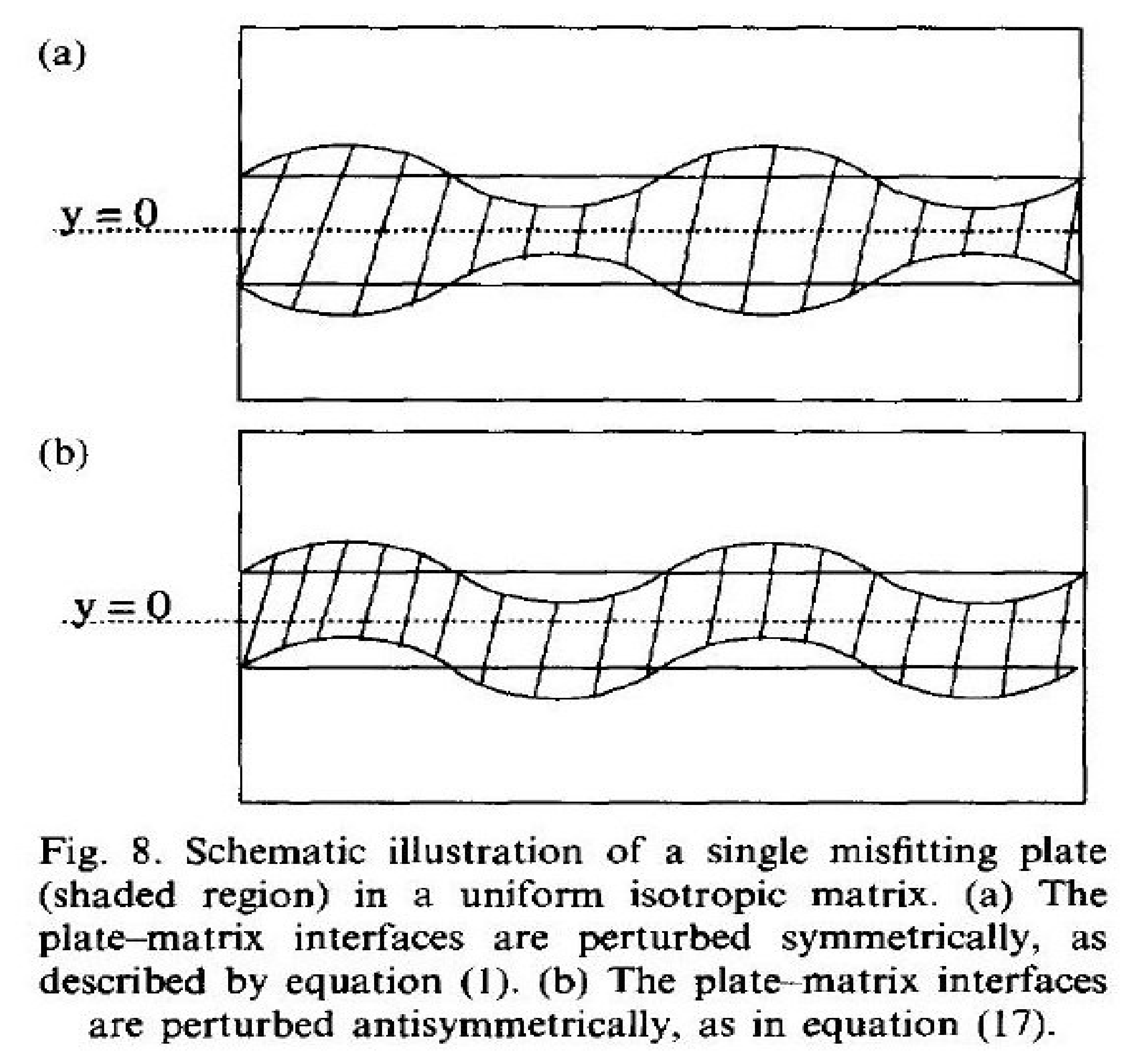


Fig. 8. Schematic illustration of a single misfitting plate (shaded region) in a uniform isotropic matrix. (a) The plate–matrix interfaces are perturbed symmetrically, as described by equation (1). (b) The plate–matrix interfaces are perturbed antisymmetrically, as in equation (17).

Image courtesy: Sridhar et al, Acta Materialia, 1997

Stability diagrams

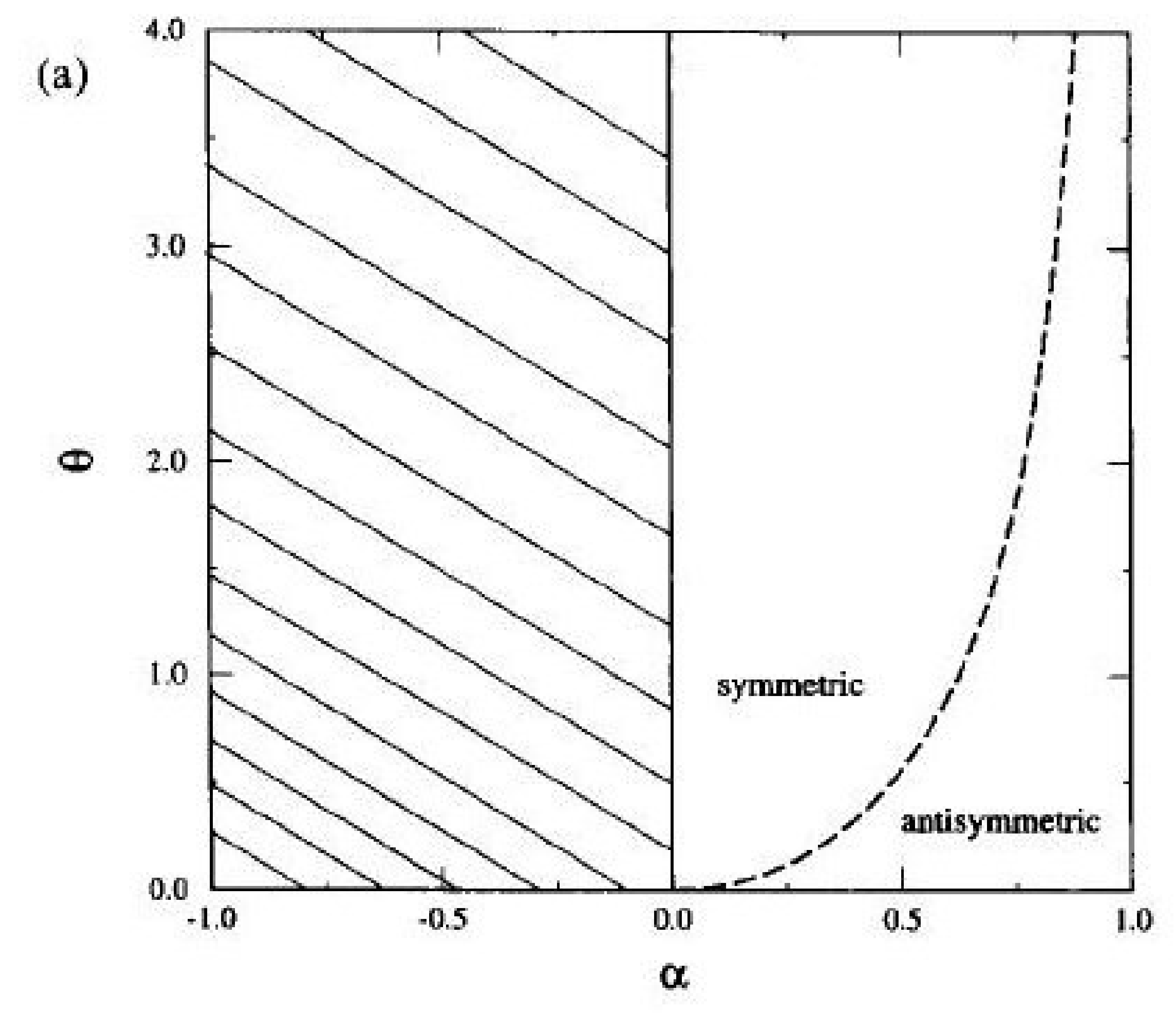


Image courtesy: Sridhar et al, Acta Materialia, 1997