Phase field simulations for grain growth and thermal grooving in thin films

Nele Moelans, Bart Blanpain, Patrick Wollants

Research group: Thermodynamics in materials engineering

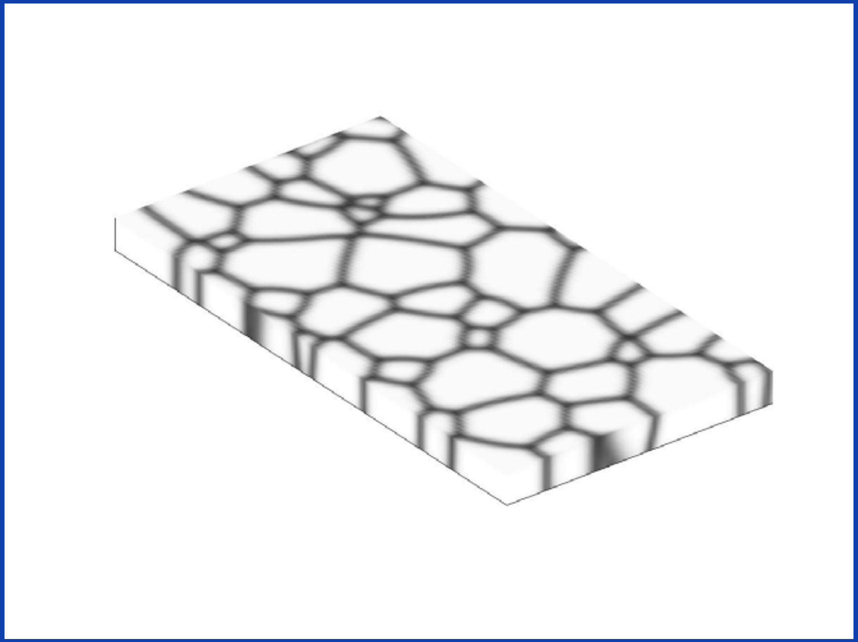
Department of Metallurgy and Materials Engineering
K.U. Leuven, Belgium

E-MRS 2006, September 4-8, 2006
Warsaw, Poland

Contents

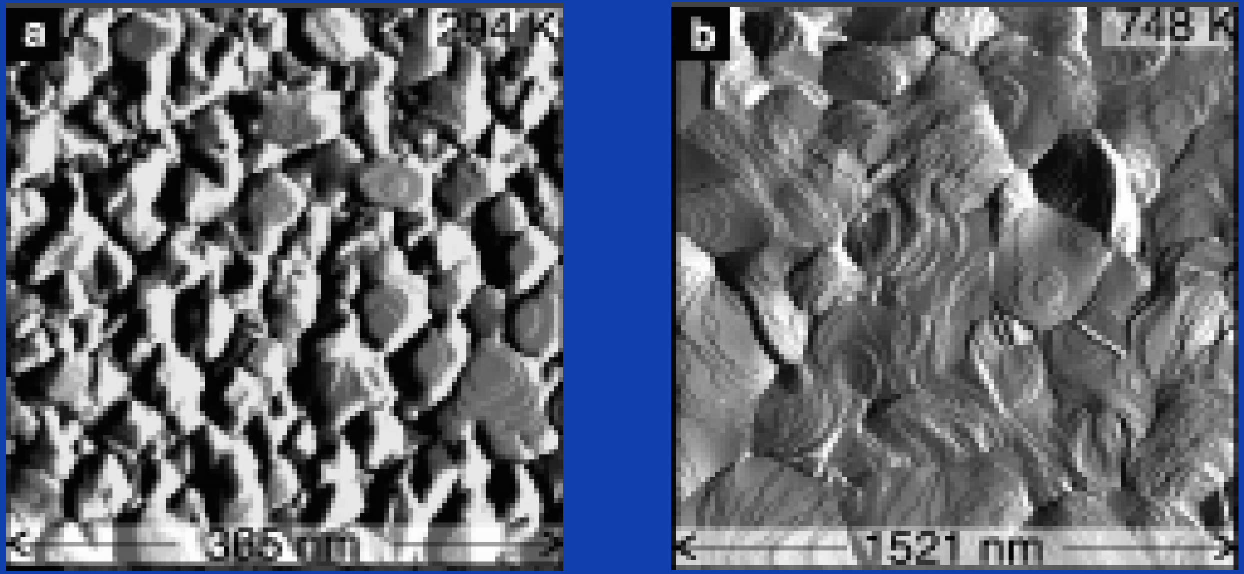
Grain growth in thin films

- Bamboo or columnar grain structure
  - => 2-D grain growth
- 3-D effects
  - Surface energy
  - Film-substrate interactions
  - Pinning precipitates



Importance of surface tension

- **Drag effect due to thermal grooving**



In-stiu STM image for a gold film (Rost et al. 2003)

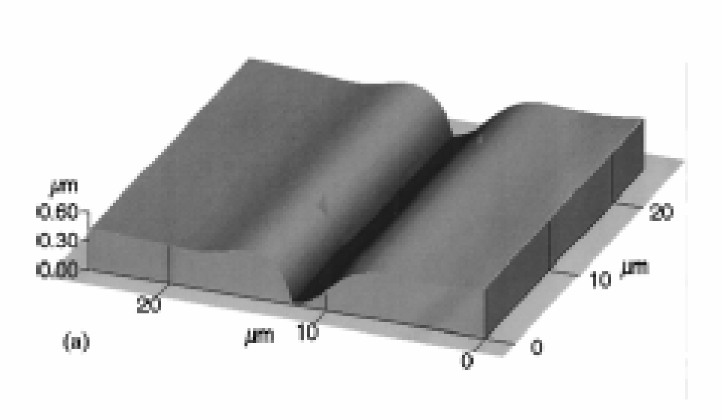
- **Abnormal grain growth induced by orientation dependence of surface energy**

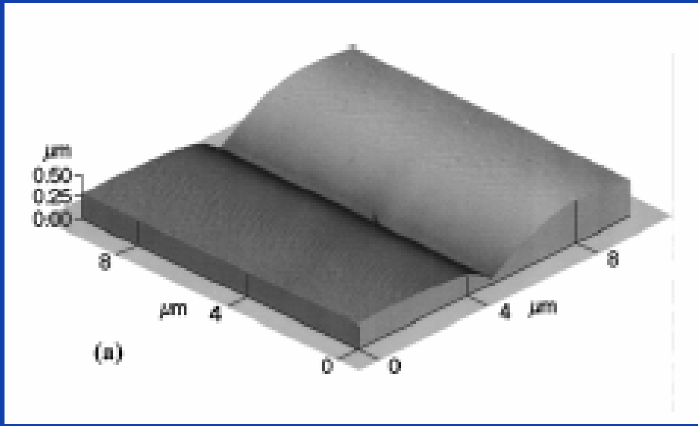
  => huge grain size

Thermal grooving

- Different mechanisms for groove formation (Mullins 1957, Zhang 2004)
  - Volume diffusion
  - Surface diffusion
  - Evaporation-condensation

=> groove morphology during steady-state growth





AFM image for a NiAl-alloy film (Klinger and Rabkin 2001)

Phase field model

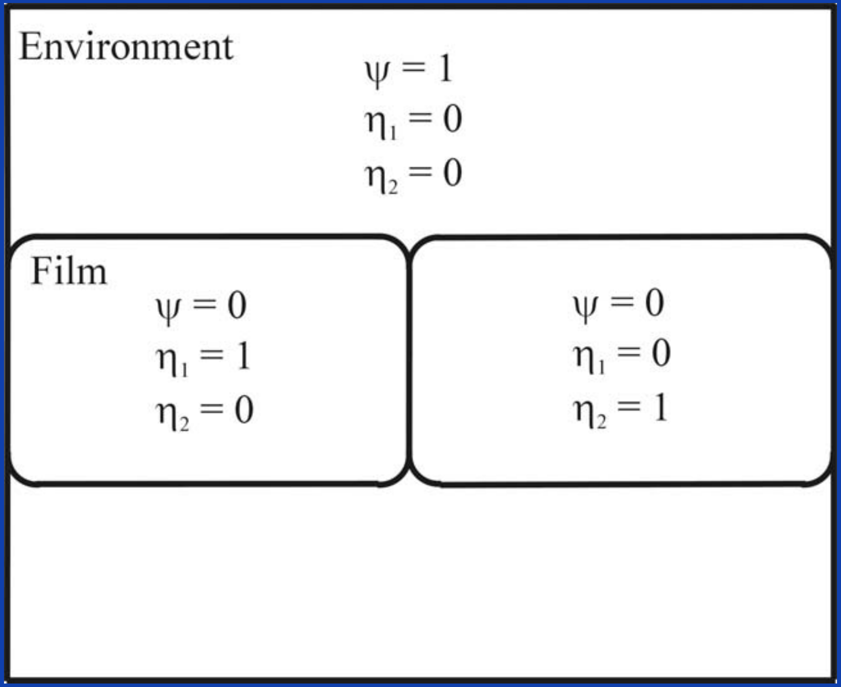
- **Simulation system**
  - **Polycrystalline film**
  - **Environment, e.g. atmosphere,film substrate**

- **Phase field variables**
  - **Grain orientations (Chen1994, Fan1997)**

    $$\eta_1, \eta_2, \ldots, \eta_i(\vec{r}, t), \ldots, \eta_p$$

  - **Environment** $\psi(\vec{r}, t)$
  - **Scaled compostion variable**

    $$\psi = \frac{c - c_{film}}{c_{env} - c_{film}}$$



Phase field model

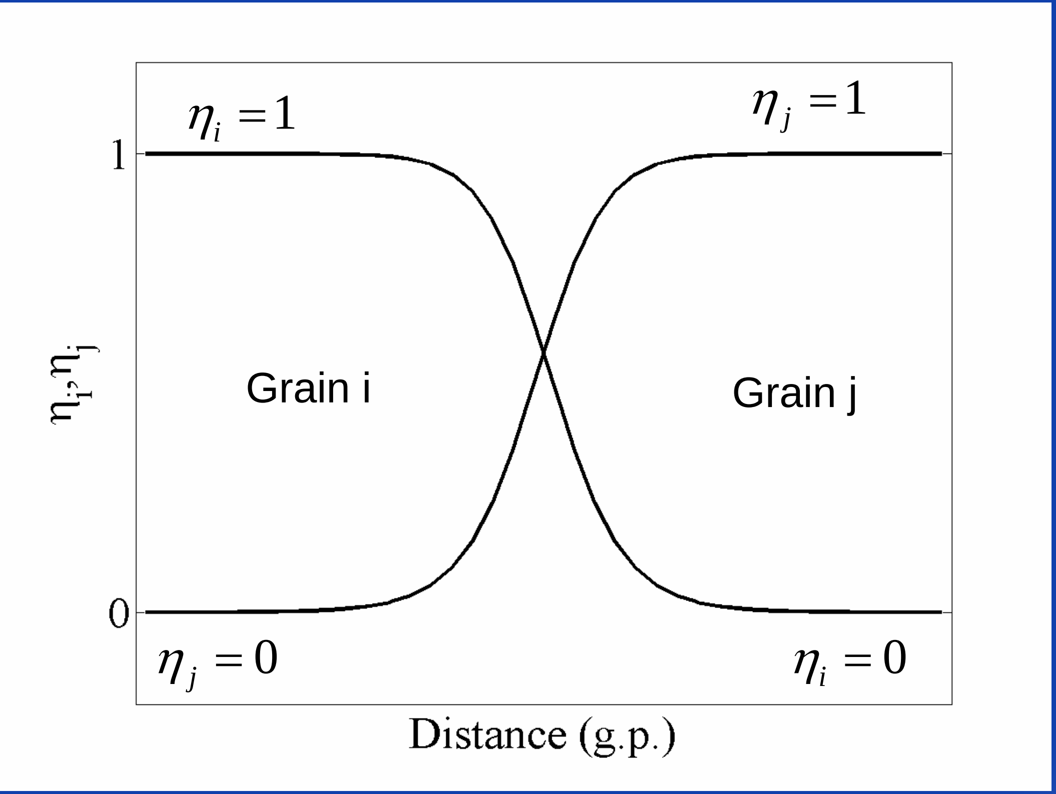
- Representation of an interface

  - Grain boundary between grain i and grain j:

$$\eta_i^2 \eta_j^2 \neq 0$$

  - Surface of grain i:

$$\eta_i^2 \psi^2 \neq 0$$



Phase field model

- **Free energy**

$$F = \int_V \left[f_0(\eta_1, \eta_2, \ldots, \eta_p, \psi) + \frac{\kappa}{2} \sum_{i=1}^{p} (\vec{\nabla} \eta_i)^2 + \frac{\kappa}{2} \sum_{i=1}^{p} (\vec{\nabla} \psi)^2 \right] dV$$

With

$$f_0 = m \left(\sum_{i=1}^{p} \left(\frac{\eta_i^4}{4} - \frac{\eta_i^2}{2} \right) + \left(\frac{\psi^4}{4} - \frac{\psi^2}{2} \right) + \sum_{i=1}^{p} \sum_{j<i}^{p} \gamma_{i,j} \eta_i^2 \eta_j^2 + \sum_{i=1}^{p} \gamma_{i,\psi} \eta_i^2 \psi^2 \right)$$

- **Orientation dependence of surface energy**

$$\gamma_{i,j}, \gamma_{i,\psi}$$

and

$$\kappa = \frac{\sum_{i=1}^{p} \sum_{j<i}^{p} \kappa_{i,j} \eta_i^2 \eta_j^2 + \sum_{i=1}^{p} \kappa_{i,\psi} \eta_i^2 \psi^2}{\sum_{i=1}^{p} \sum_{j<i}^{p} \eta_i^2 \eta_j^2 + \sum_{i=1}^{p} \eta_i^2 \psi^2}$$

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Phase field model

$$F = \int_V \left[f_0(\eta_1, \eta_2, \ldots, \eta_p, \psi) + \frac{\kappa}{2} \sum_{i=1}^{p} (\vec{\nabla} \eta_i)^2 + \frac{\kappa}{2} \sum_{i=1}^{p} (\vec{\nabla} \psi)^2 \right] dV$$

$$f_0 = m \left(\sum_{i=1}^{p} \left(\frac{\eta_i^4}{4} - \frac{\eta_i^2}{2} \right) + \left(\frac{\psi^4}{4} - \frac{\psi^2}{2} \right) + \sum_{i=1}^{p} \sum_{j<i}^{p} \gamma_{i,j} \eta_i^2 \eta_j^2 + \sum_{i=1}^{p} \gamma_{i,\psi} \eta_i^2 \psi^2 \right)$$

- **Interfacial energy**

$$\sigma_{i,j} = \frac{1}{\sqrt{3}} \sqrt{m \kappa_{i,j} (1 + 2\gamma_{i,j})}$$

- **Interfacial width**

$$l_{i,j} \propto \sqrt{\frac{32 \kappa_{i,j}}{m(1 + 2\gamma_{i,j})}}$$

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Phase field model

- **Temporal evolution η_i**

$$\frac{\partial \eta_i(\vec{r},t)}{\partial t} = -L\frac{\partial F(\eta_1,\ldots,\eta_p,\psi)}{\partial \eta_i(\vec{r},t)}$$

- **Temporal evolution ψ**

$$\frac{\partial \psi(\vec{r},t)}{\partial t} = \vec{\nabla} \bullet M\vec{\nabla}\frac{\partial F(\eta_1,\ldots,\eta_p,\psi)}{\partial \psi(\vec{r},t)}$$

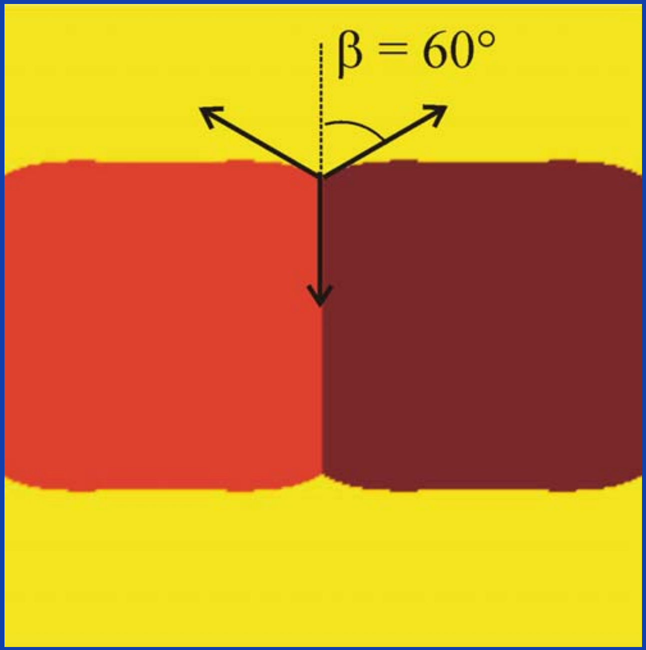
- **Bulk versus interface diffusion**

$$M = \frac{M_{film}\left(\sum_{i=1}^{p}\eta_i^2\right) + M_{env}\psi^2}{\sum_{i=1}^{p}\eta_i^2 + \psi^2} \quad for \quad \sum_{i=1}^{p}\sum_{j<i}^{p}\eta_i^2\eta_j^2 + \sum_{i=1}^{p}\eta_i^2\psi^2 < 10e-6$$

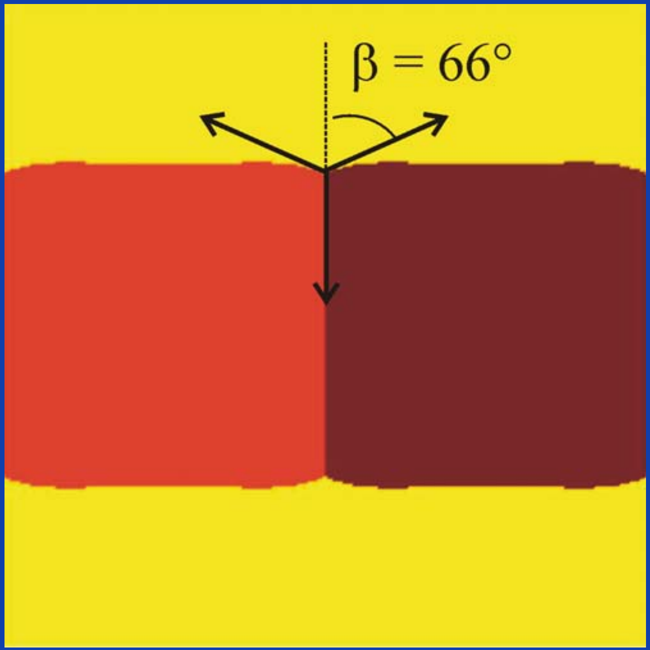
$$M = \frac{M_{gb}\left(\sum_{i=1}^{p}\sum_{j<i}^{p}\eta_i^2\eta_j^2\right) + M_{surf}\sum_{i=1}^{p}\eta_i^2\psi^2}{\sum_{i=1}^{p}\sum_{j<i}^{p}\eta_i^2\eta_j^2 + \sum_{i=1}^{p}\eta_i^2\psi^2} \quad for \quad \sum_{i=1}^{p}\sum_{j<i}^{p}\eta_i^2\eta_j^2 + \sum_{i=1}^{p}\eta_i^2\psi^2 > 10e-6$$

Groove angle

- **Grain boundary versus surface energy:** $\sigma_{gb} = 2\sigma_{surf}\cos(\beta)$

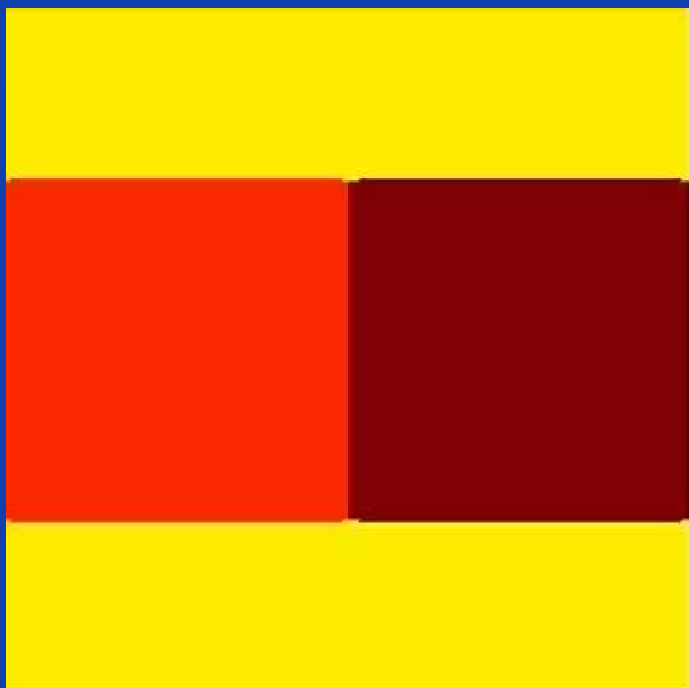


$\sigma_{surf} = \sigma_{gb}$



$\sigma_{surf} = (5/4)\sigma_{gb}$

Orientation dependent surface energy

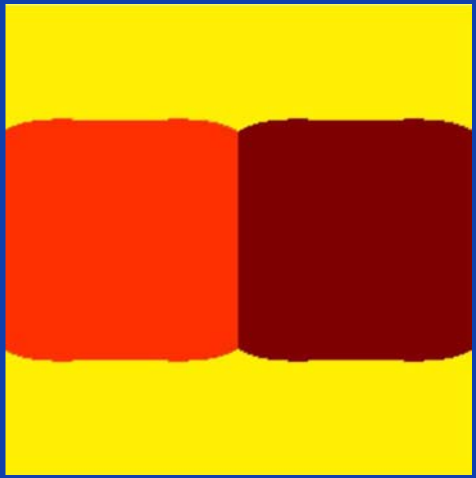


$$\sigma_{surf,2} = (5/4)\sigma_{surf,1}$$

- **Columnar grain boundary becomes curved to balance unequal surface tensions**
- **Grain boundary migration towards center of curvature**
- **=> favourably oriented grain grows**

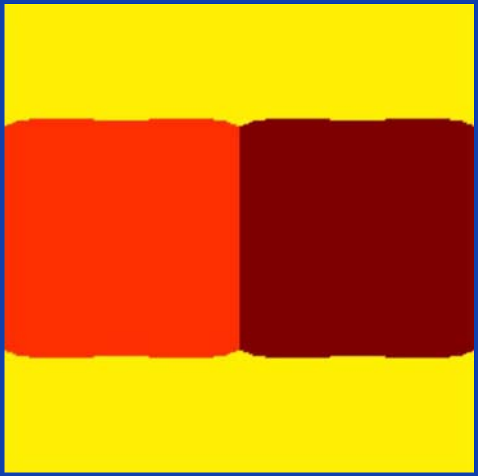
Groove formation mechanisms

- **Bulk diffusion**



$M = cte$
solubility ≈ 0

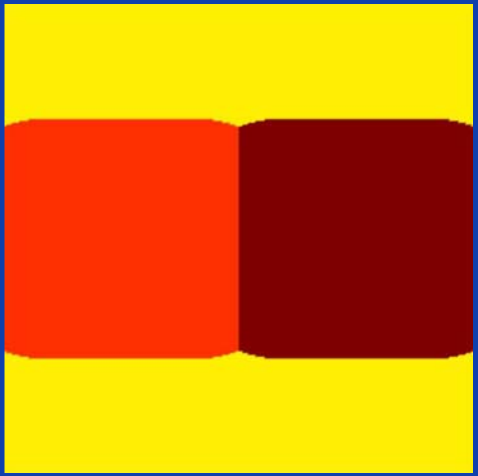
- **Surface diffusion**



$M_{surf} = 1$
solubility ≈ 0

$M_{film} = M_{env} = M_{gb} = 0$

- **Evaporation-condensation**

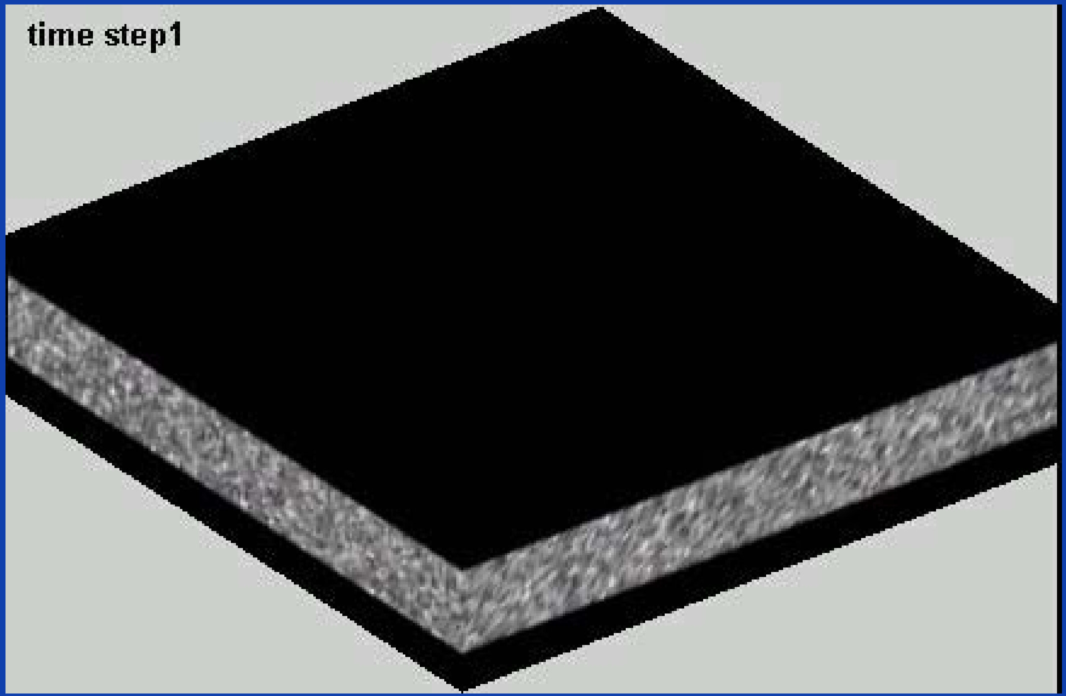


$M_{env} = M_{surf} = 1$
solubility = 0.0001

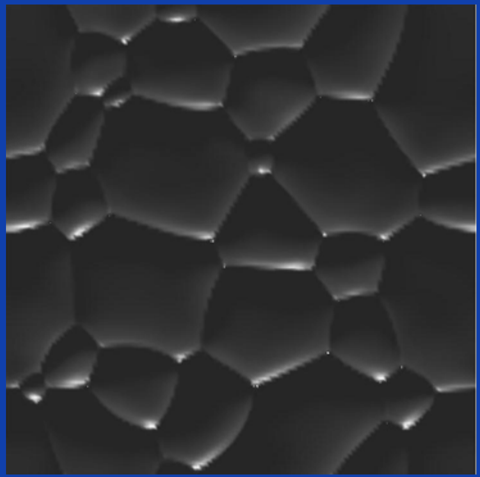
$M_{film} = M_{gb} = 0$

Outlook

- 3-D simulations for polycrystalline films



Efficient implementation for κ and M constant



Top view

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Conclusions

- **Phase field model for grain growth and grooving in thin films**
  - **Simulation system includes film + environment**
  - **Orientation dependent surface energy and misorientation dependent grain boundary energy**
  - **Bulk and interfacial diffusion can be distinguished**

- **More information about my research: http://www.ulyssis.org/~nele/**

- ***Acknowledgements:*** *This research was funded by the Institute for the Promotion of Innovation through Science and Technology in Flanders (IWT-Vlaanderen).*