Quantitative phase field simulations of anisotropic grain growth in columnar thin films with a fiber texture

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Introduction

- Thin films with columnar grains and a fiber texture
- Grain boundary energy as a function of $\theta, \gamma(\theta)$
  - Low-angle: $\theta < \theta_m$
    - Read-shockley
  - High-angle:
    - $\gamma = \gamma_m$
    - Cusps
- MDF? $R = k t^n$?
Grain growth model

- **Phase field variables**
  \[ \eta_1, \eta_2, \ldots, \eta_i(r, t), \ldots, \eta_p \]
  
  with for orientation \( i \)
  
  \[ (\eta_1, \eta_2, \ldots, \eta_i, \ldots, \eta_p) = (0, 0, \ldots, 1, \ldots, 0) \]

- **Total grain boundary energy**
  
  \[
  F = \int_V \left[ m \left( \sum_{i=1}^{p} \left( \frac{n_i^4}{4} - \frac{n_i^2}{2} \right) + \sum_{i=1}^{p} \sum_{j<i} \gamma_{i,j} n_i^2 n_j^2 + \frac{1}{4} \right) + \frac{k(\eta)}{2} \sum_{i=1}^{p} (\nabla n_i)^2 \right] dV
  \]

- **Ginzburg-Landau equation**
  
  \[
  \frac{\partial \eta_i(\vec{r}, t)}{\partial t} = -L(\eta) \left[ \frac{\partial f(\eta_1, \ldots, \eta_p)}{\partial \eta_i(\vec{r}, t)} - k(\eta) \nabla^2 \eta_i \right]
  \]

Chen and Yang, *PRB*, 50, 15752 (1994)

Kazaryan et al., *PRB*, 61, 14275 (2000)

Grain growth model—misorientation dependence

- Misorientation dependence: \( \gamma_{i,j}, \kappa(\eta), L(\eta) \)

- with
  \[ \kappa(\eta) = \frac{\sum_{i=1}^{p} \sum_{j<i}^{p} \kappa_{i,j} \eta_i^2 \eta_j^2}{\sum_{i=1}^{p} \sum_{j<i}^{p} \eta_i^2 \eta_j^2} \]
  and
  \[ L(\eta) = \frac{\sum_{i=1}^{p} \sum_{j<i}^{p} L_{i,j} \eta_i^2 \eta_j^2}{\sum_{i=1}^{p} \sum_{j<i}^{p} \eta_i^2 \eta_j^2} \]

- For each grain boundary
- Individual parameters
  \[ \eta_i^2 \eta_j^2 \neq 0 \]

- Misorientation
  \[ \theta_{i,j}(\eta_i, \eta_j) \]
Model parameters

- **Parameter relations**
  - **Grain boundary energy**
    \[
    \gamma_{gb,\theta_{i,j}} = g(\gamma_{i,j}) \sqrt{m\kappa_{i,j}}
    \]
  - **Grain boundary mobility**
    \[
    \mu_{gb,\theta_{i,j}} = L_{i,j} \sqrt{\frac{\kappa_{i,j}}{m(g(\gamma_{i,j}))^2}}
    \]
  - **Diffuse grain boundary width**
    \[
    l_{gb} = \frac{4}{3} \sqrt{\frac{\kappa_{i,j}}{m(g(\gamma_{i,j}))^2}}
    \]

Numerically calculated function values for \(g(\gamma_{i,j})\)

Fitting: \(g^{-1}(\gamma_{i,j}) \approx 5th\) order polynomial
Model parameters

- Diffuse grain boundary width
  - Measure of largest gradient
    \[ l_{gb} = \frac{1}{\max \left| \frac{d\eta_i}{dx} \right|} = \frac{1}{\max \left| \frac{d\eta_j}{dx} \right|} \]
  - \( l_{gb} = ct \rightarrow \) high controllability of numerical accuracy

- Iterative algorithm
  - \( \ell_{gb}, [\gamma_{gb,\theta}, [\mu_{gb,\theta}] \rightarrow m, [K_{i,j}, [\gamma_{i,j}, [L_{i,j}]] \]
Calculation model parameters

- Physical quantities:
  \[ \gamma_{gb, m} = 0.25, \mu_{gb} = 1 \times 10^{-6} \]

- Diffuse grain boundary width
  \[ \ell = 1 \times 10^{-6} \]

- Model parameters
  \[ m = 2.25 \times 10^6, L = 2 \]
  \[ \kappa_{\text{max}} = 0.25, \gamma_{\text{max}} = 1.5 \]

- 4-fold symmetry

- Discrete orientations
  \[ \eta_1, \eta_2, ..., \eta_i(r, t), ..., \eta_{60} \Rightarrow \Delta \theta = 1.5^\circ \]

\[ \gamma_{gb, \theta} = \gamma_{gb, m} \frac{\theta}{\theta_m} \left( 1 - \ln \left( \frac{\theta}{\theta_m} \right) \right), \quad 0 < \theta < 15^\circ \]

\[ \gamma_{gb} = \gamma_{gb, m}, \quad \theta > 15^\circ \]
Calculation model parameters

- **Physical quantities:**
  \[ \gamma_{gb,m} = 0.25, \mu_{gb} = 1 \times 10^{-6} \]

- **Diffuse grain boundary width**
  \[ \ell = 1 \times 10^{-6} \]

- **Model parameters**
  \[ m = 2.25 \times 10^6, L = 2 \]
  \[ \kappa_{max} = 0.25, \gamma_{max} = 1.5 \]

- **4-fold symmetry**
- **Discrete orientations**
  \[ \eta_1, \eta_2, \ldots, \eta_i(r,t), \ldots, \eta_{60} \Rightarrow \Delta \theta = 1.5^\circ \]

**Read-Shockley + cusp**
\[ \gamma_{gb,\theta} = (\gamma_{gb,m} - 0.1) \frac{|\theta - 37.5^\circ|}{10^\circ} \left( 1 - \ln \left( \frac{|\theta - 37.5^\circ|}{10^\circ} \right) \right) + 0.1, \]
\[ 27.5^\circ < |\theta| < 45^\circ \]
Simulations: no high-angle cusp

- Initially random grain orientations and grain boundary types
- High-angle grain boundaries form independent network
- Low-angle grain boundaries follow movement of high-angle grain boundaries → elongate
- No stable quadruple junctions

White: \( \theta = 1.5 \)
Gray: \( \theta = 3 \)
Black: \( \theta > 3 \)

1024x1024 g.p. (256x256 are shown)
System size: 102.4x102.4 \( \mu \)m\(^2\) (\( \Delta x = 0.1 \mu \)m)
Simulations: no high-angle cusp

- Initially random grain orientations and grain boundary types
- High-angle grain boundaries form independent network
- Low-angle grain boundaries follow movement of high-angle grain boundaries → elongate
- No stable quadruple junctions
Simulations: 1 high-angle energy cusp

- Grain boundary energy with extra cusp at $\theta = 37.5^\circ$

White: $\theta = 1.5$
Gray: $\theta = 3$
Black: $\theta > 3$, $\theta \neq 37.5$
Red: $\theta = 37.5$
Grain growth kinetics

- Grain growth exponent
- PFM: steady-state growth with
  \[ n \approx 1 \]
- Previous findings:
  \[ n = 0.6...1 \]

\[ A - A_0 = kt^n \]

Read-Shockley (\( \theta_m = 15^\circ \)) + cusp at \( \theta = 37.5^\circ \)
Misorientation distribution function (MDF)

- Area weighted MDF

- Reaches a steady-state
- Low energy boundaries lengthen + their number increases

Read-Shockley + cusp at $\theta = 37.5^\circ$
Mean field analysis

- Assume parabolic growth for high-angle grains
  \[ A_h(t) = A_h(0) + kt \]
  \[ A_h = \frac{A_{\text{tot}}}{N} \Rightarrow N(t) = \frac{A_{\text{tot}}}{A_h(t)} \]

- Assume \( N' \) low energy boundaries result in \( N' \) extra grains
  \[ A_{\text{eff}} = \frac{A_{\text{tot}}}{N + N'} \]

- Growth coefficient
  \[ A_{\text{eff}}(t) = k_{\text{eff}} t^{n_{\text{eff}}}, t \to \infty \]
  \[ n_{\text{eff}} = \frac{d \log(A_{\text{eff}})}{d \log(t)} = \frac{kt}{A_h(0) + kt} - \frac{t}{1 + \frac{N'}{N}} \frac{d(N'/N)}{dt} \]

- Constant \( N'/N \)
  \[ n_{\text{eff}} = \frac{kt}{A_h(0) + kt} \to 1, t \to \infty \]
  \[ k_{\text{eff}} = \frac{dA_{\text{eff}}}{dt} = \frac{k}{1 + N'/N} \]

Moelans et al., Phil. Mag., to be published
Conclusions

- Phase field formulation for anisotropic systems with high controllability of the numerical accuracy

- Phase field simulations of grain growth in thin columnar films with fiber texture
  - High-energy grain boundaries form independent network
  - Steady-state is reached after finite amount of time
    - Effective growth exponent equal to 1
    - Final MDF correlated with $\gamma(\theta)$
    - Quantitatively, the MDF is very sensitive to numerical aspects

- Mean field analysis confirms existence of steady-state regime in fiber textured films
Thank you for your attention!

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More information on http://nele.studentenweb.org
Numerical validation

- Shrinking grain:

\[
\frac{dA_\alpha}{dt} = -2\pi \mu_{\alpha\beta} \sigma_{\alpha\beta}
\]

- Triple junction angles:

\[
\sigma_{\alpha\gamma} = \sigma_{\beta\gamma}, \mu_{\alpha\gamma} = \mu_{\beta\gamma}
\]

- Observations
  - Accuracy controlled by \( \frac{l_{\text{num}}}{\Delta x} \)
  - Diffuse interface effects for \( \frac{l_{\text{num}}}{R}>5 \)
  - Angles outside [100°-140°] require larger \( \frac{l_{\text{num}}}{\Delta x} \) for same accuracy
Misorientation distribution function (MDF)

- **Effect of orientation discretization**

- **Qualitatively MDF correlated with g(q)**

- **Quantitative results very sensitive for numerical aspects**
  - $\Delta \theta$
  - $\Delta x$

![Graph showing misorientation distribution function (MDF) with different discretization values: $\Delta \theta = 3^\circ, t = 479$, $\Delta \theta = 3^\circ, t = 719$, $\Delta \theta = 1.5^\circ, t = 418$, $\Delta \theta = 1.5^\circ, t = 490$. The graph plots length density against misorientation $\theta$ (°) with peaks indicating the distribution.](image)