Computational materials science: From needle crystals to complex polycrystalline forms

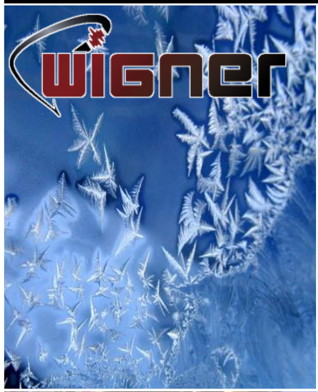
L. Gránásy[a,b]

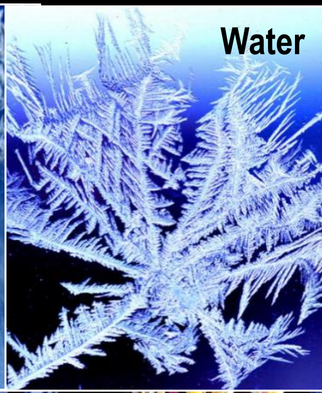
[a]Wigner Research Centre for Physics, H-1525 Budapest, P. O. Box 49, Hungary
[b]BCAST, Brunel University, Uxbridge, Middlesex UB8 3PH, U.K.

Inaugural presentation as elected member of Academia Europaea (Academy of Europe, London), Hungarian Academy of Sciences, 3 September 2017, Budapest, Hungary

0p

I. Introduction: Complex polycrystalline structures





Water

Gin

Polycystalline matter:
- technical alloys
- ceramics
- polymers
- minerals
- food products, etc.

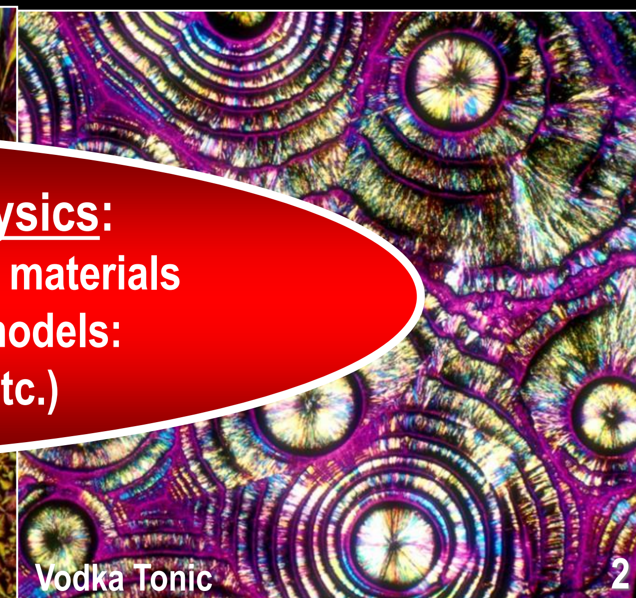
In biology:
- bones, teeth
- kidney stone
- cholesterol in arteries
- amyloid plaques in Alzheimer's disease

Complex patterns evolve due to the interplay of **nucleation** and **growth**.

Also frozen drinks:

American Pale Ale

Dirty Martini



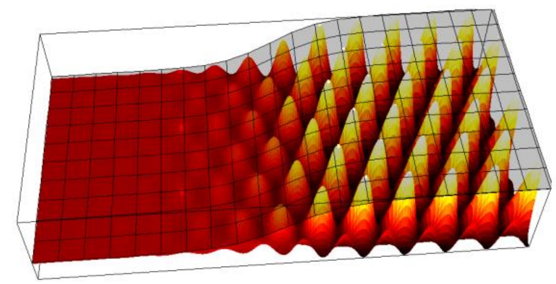
Vodka Tonic

Aim of <u>Computational Materials Physics</u>:
To understand and predict the behavior of materials
Tools: micro-, mezo- and macroscale models:
ab initio, DFT, MD, PFC, <u>PFT</u>, CFD, etc.)

II. Modeling of crystalline microstructure (μm, μs ↔ cm, min)

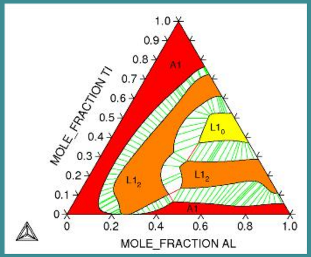
- **Mathematical model** ⇒ <u>PF theory:</u> EOMs are coupled nonlinear stochastic PDEs
- **Numerical solution** (finite diff., spectral, …)
- **Input data:** free energies, diffusion coefficients, interfacial free energies, anisotropies (⇐ micr. models, data bases)
- **Computation facilities:** CPU and GPU clusters

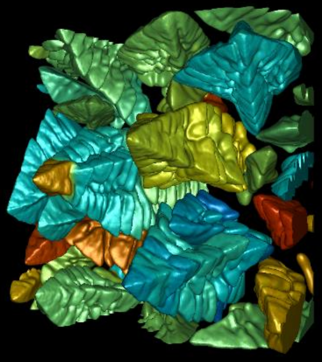
Structural order parameter [phase field: $\phi(r, t)$]



Model

Numerical solver



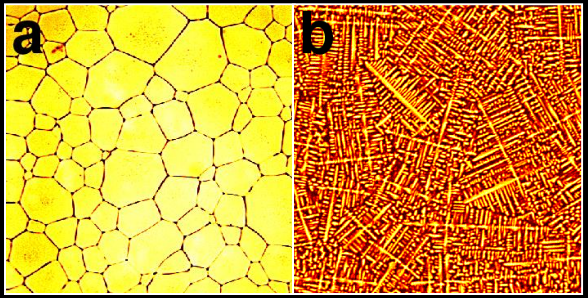


Microstructure

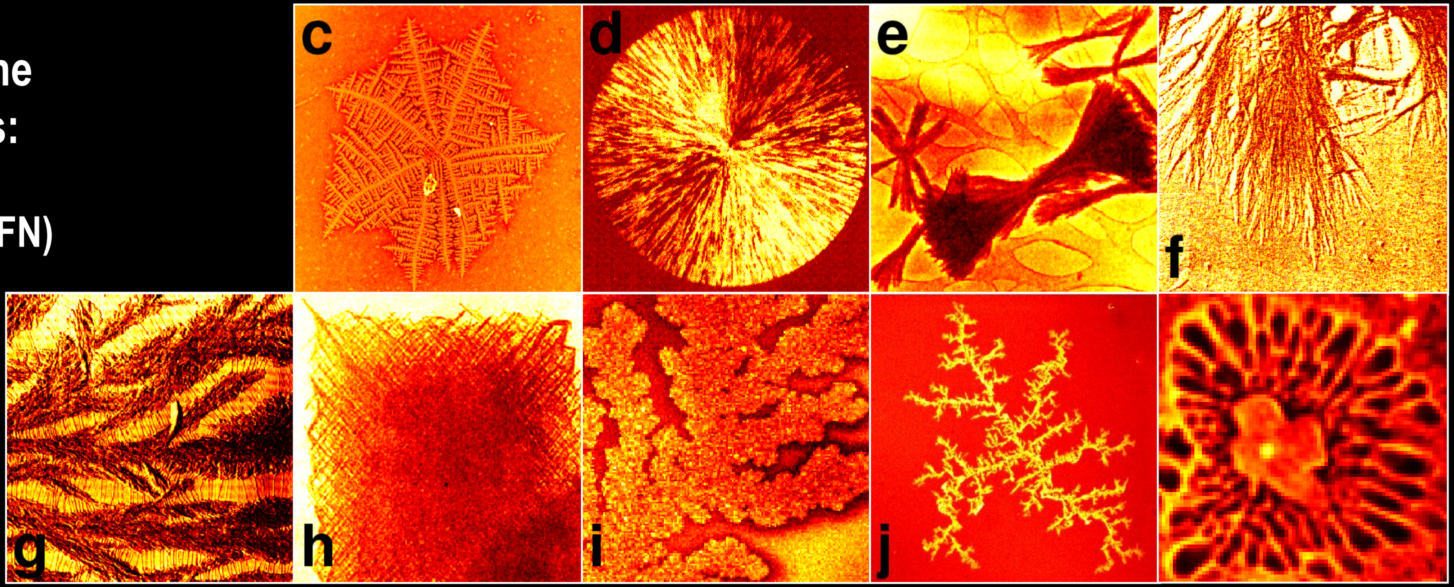
In a few cases (metal alloys):
Knowledge-based Materials Design

Classification of polycrystalline microstructures

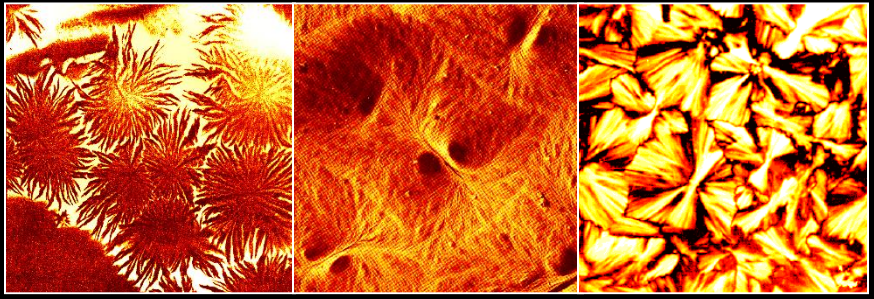
1. **Impinging** single crystals:



2. **Polycrystalline growth** forms:
(Growth Front Nucleation = GFN)

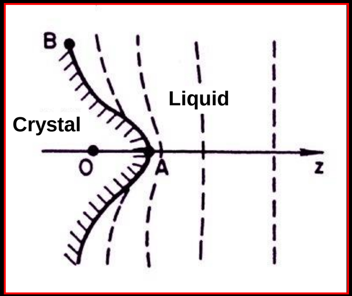


3. **Impinging** polycrystalline particles:

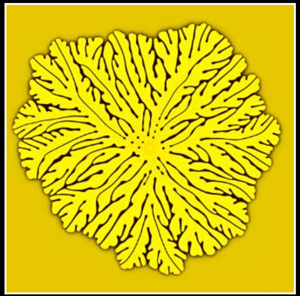


Contributing phenomena?

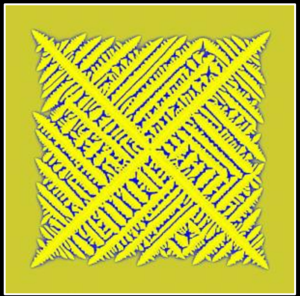
1. Diffusional instabilities:



Mullins-Sekerka instability



isotropic



anisotropic

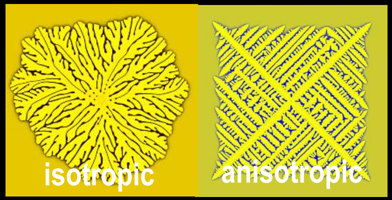
2. Nucleation

- *of growth centers*
  - homogeneous
  - heterogeneous (on particles or walls)
- *of new grains at the growth front* (Growth Front Nucleation = GFN)
  - heterogeneous (particle-induced)
  - homogeneous (???)

    with specific misorientation (fixed branching angle)

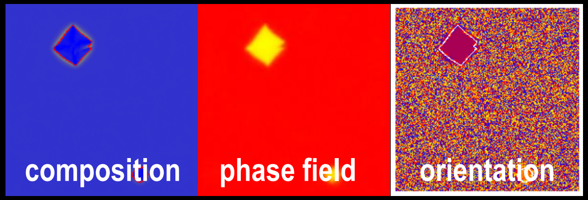
Summary: Phenomena incorporated in 2D & 3D:

1. Diffusional instabilities:

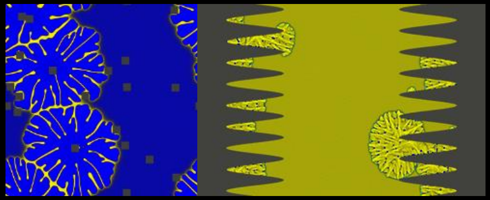


2. Nucleation of growth centers

- homogeneous — adding noise to EOM (Phys. Rev. Lett. 2002)

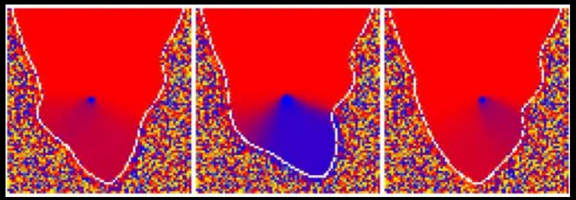


- heterogeneous — noise + appropriate BC (Phys. Rev. Lett. 2007)

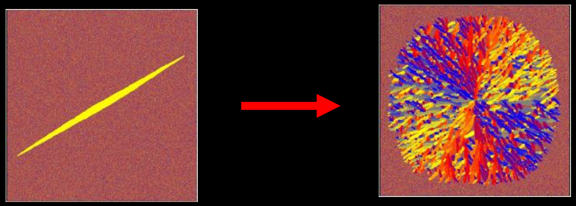


3. Nucleation of new grains at the growth front

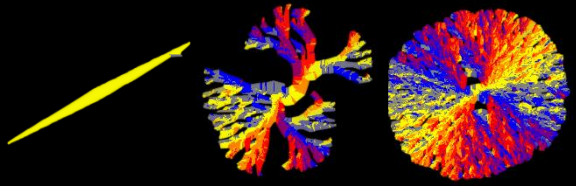
- heterogeneous — particle-induced tip-deflection (2D: Nature Mater. 2003, 3D: Europhys. Lett. 2005)



- homogeneous I. — reduced M_θ (2D: Nature Mater. 2004, 3D: Europhys. Lett. 2005)

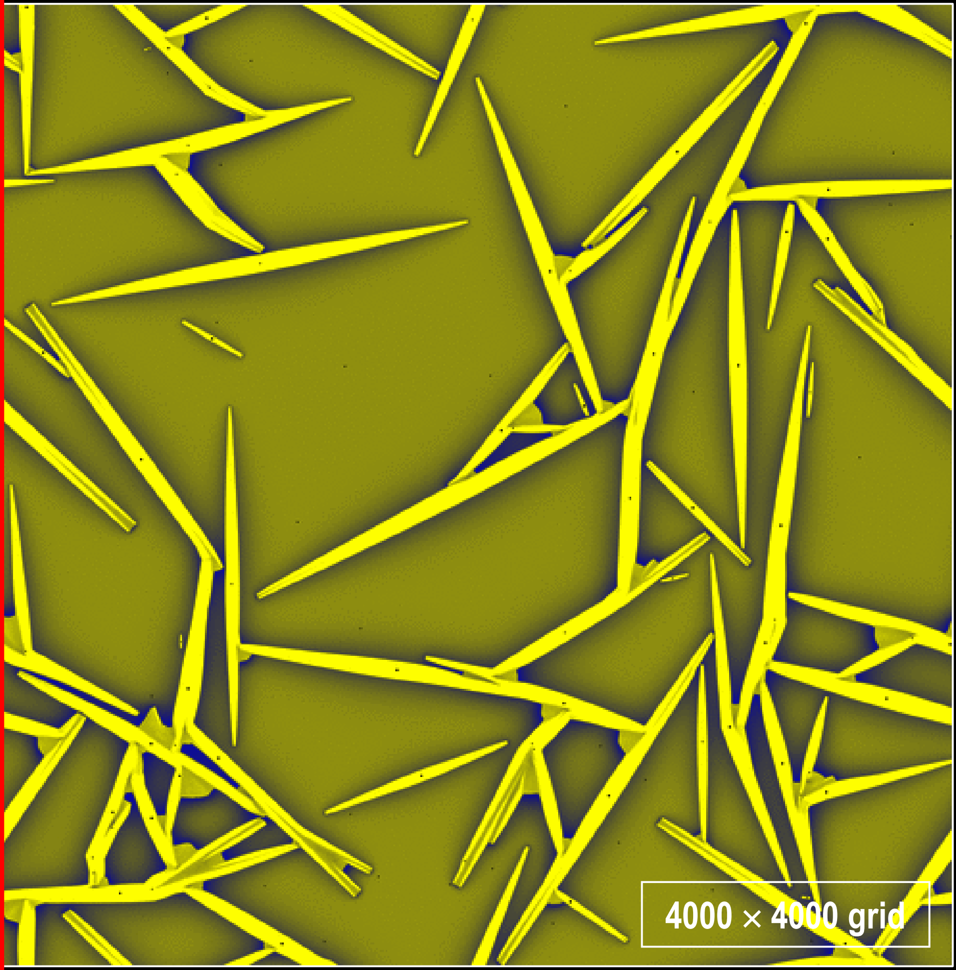


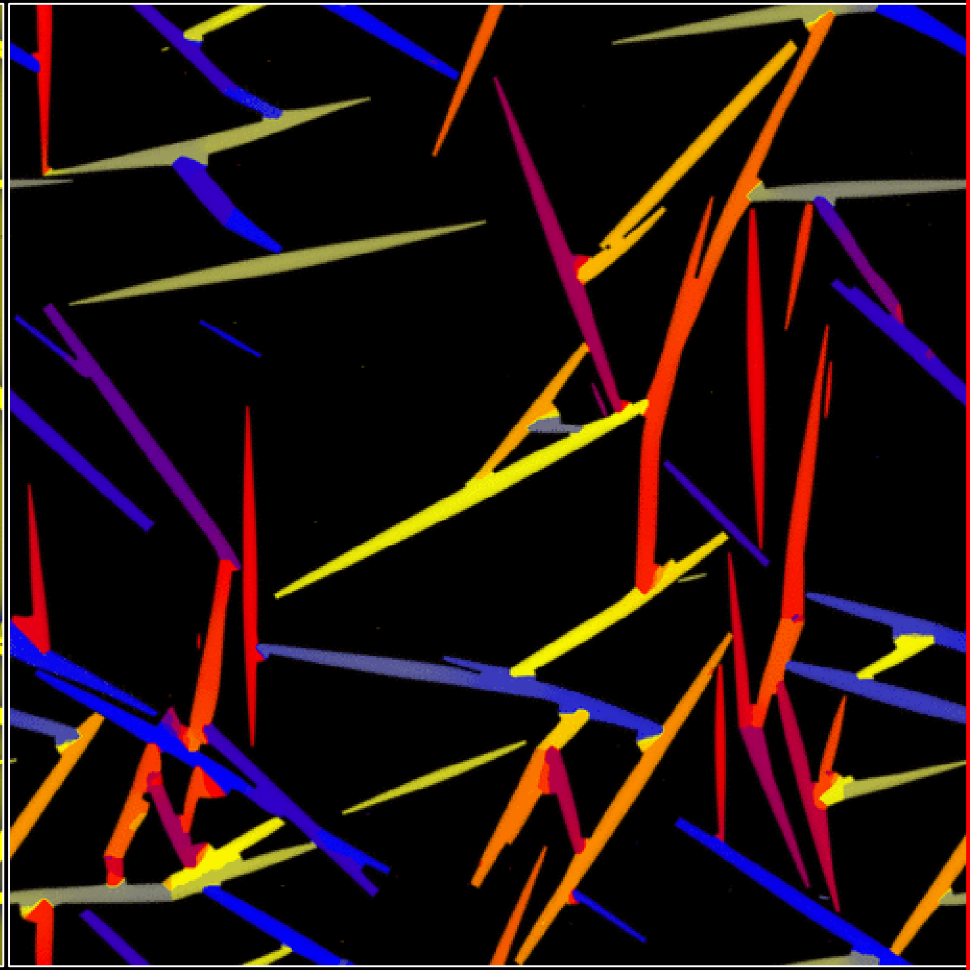
- homogeneous II. — MS minimum in f_{ori} (Phys. Rev. E 2005)



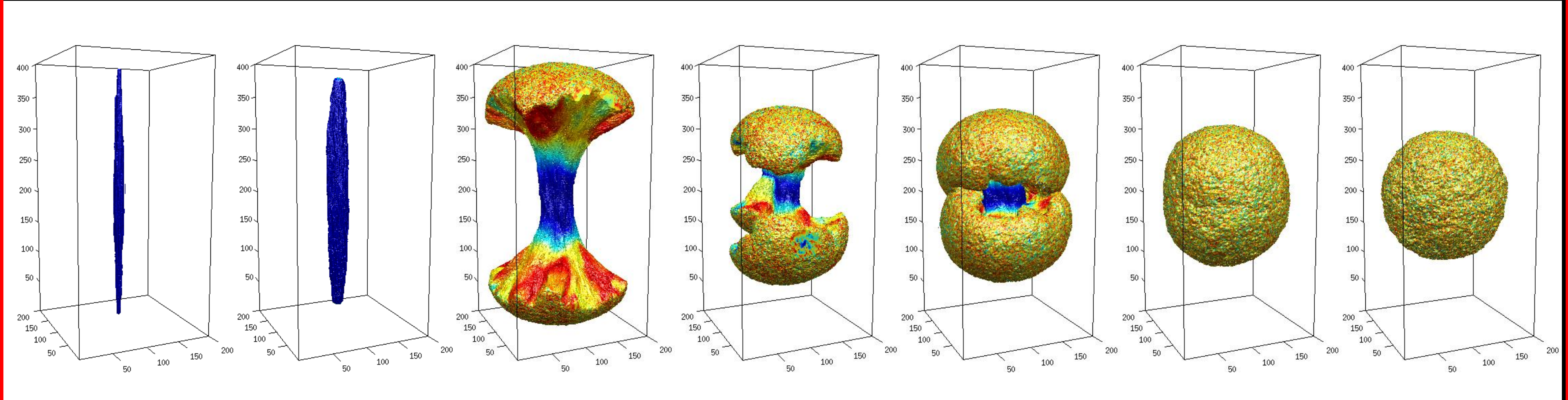
III. Applications

A. Needle crystals in 2D: (kinetic & interface free energy anisotropy)

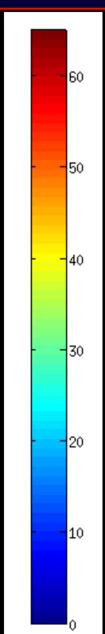




B. From needle crystal to polycrystalline spherulite:



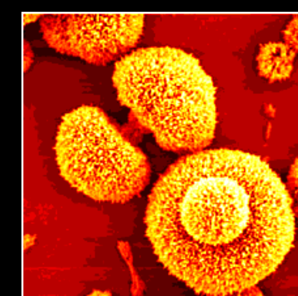
S = 1.5 | 1.8 | 1.9 | 1.95 | 2.0 | 2.1 | 2.2



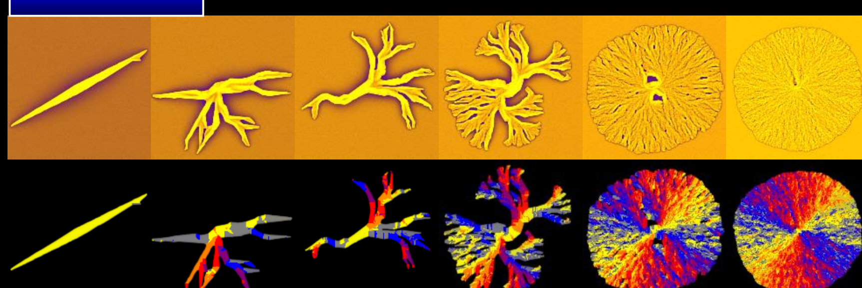
Coloring:
Inclination relative to nucleated direction in deg.

200×200×400 grid

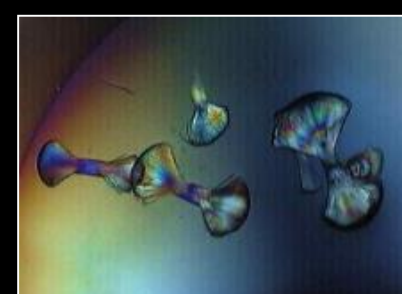
Triclinic crystal symmetry
Ellipsoidal symmetry of kinetic anisotropy

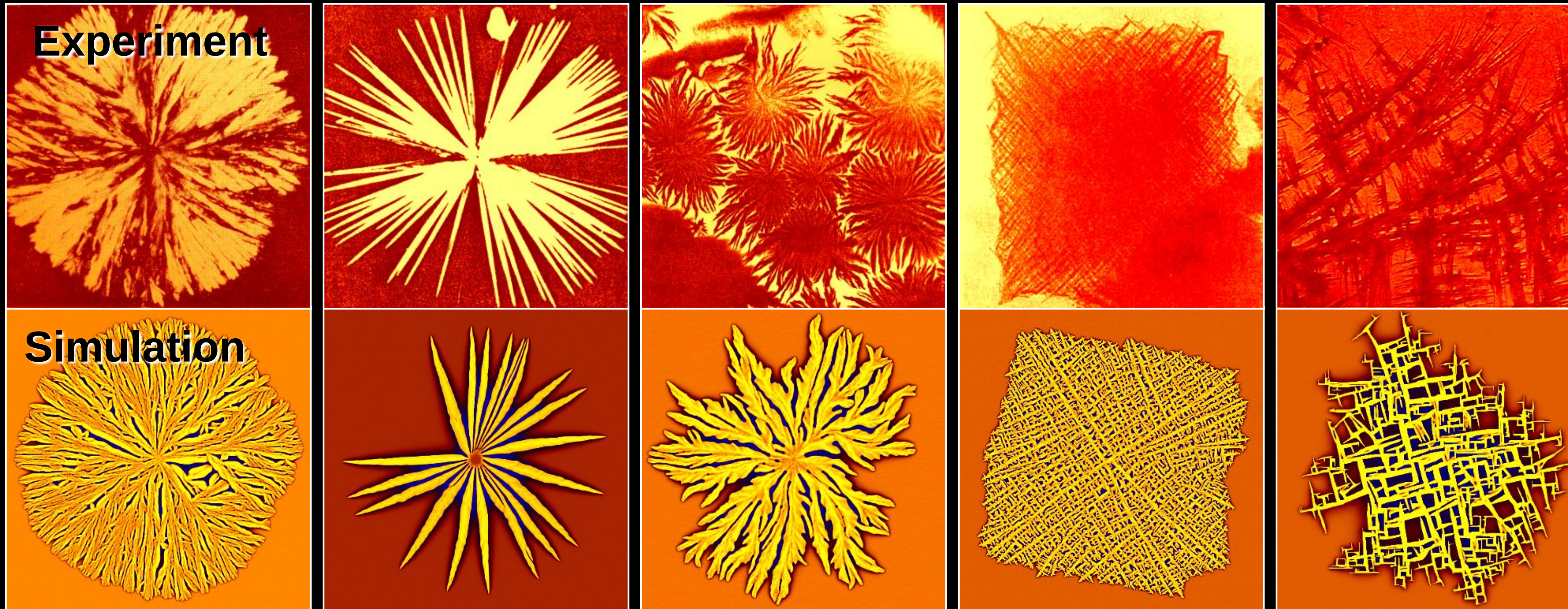


2D



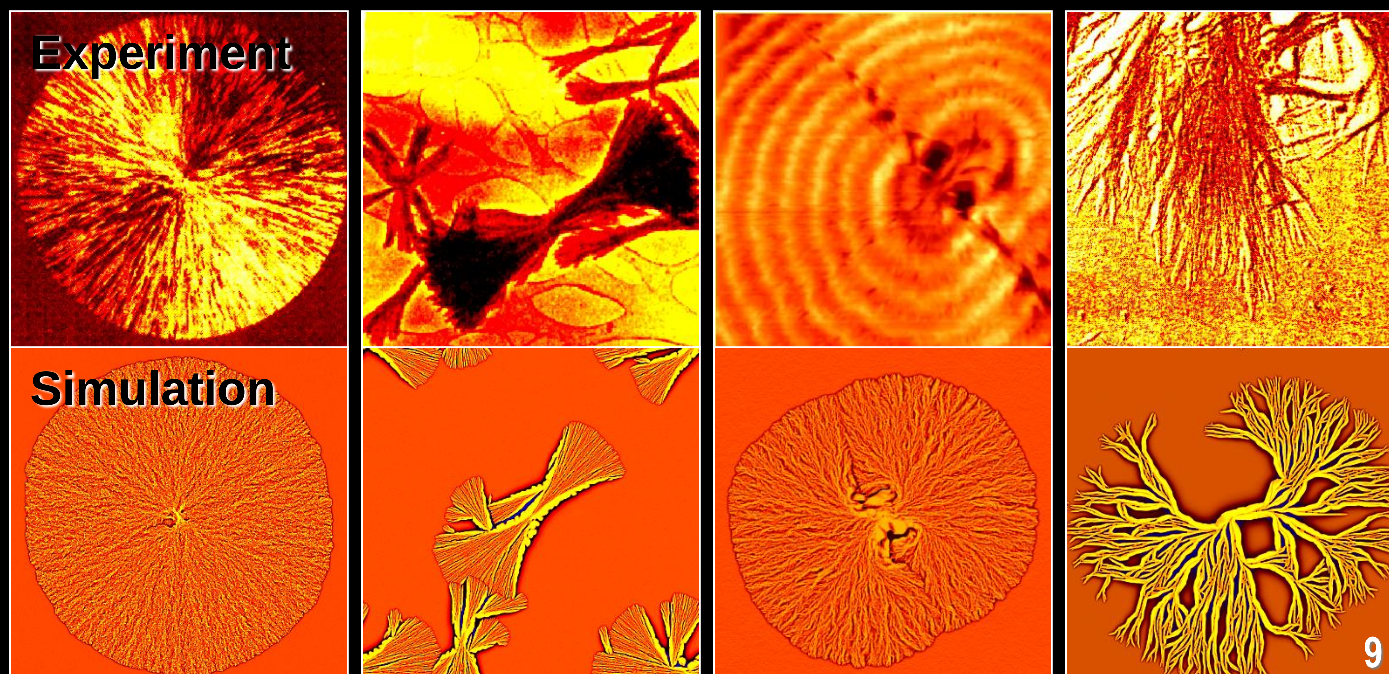
S = 0.75 | 0.85 | 0.90 | 0.95 | 1.00 | 1.10



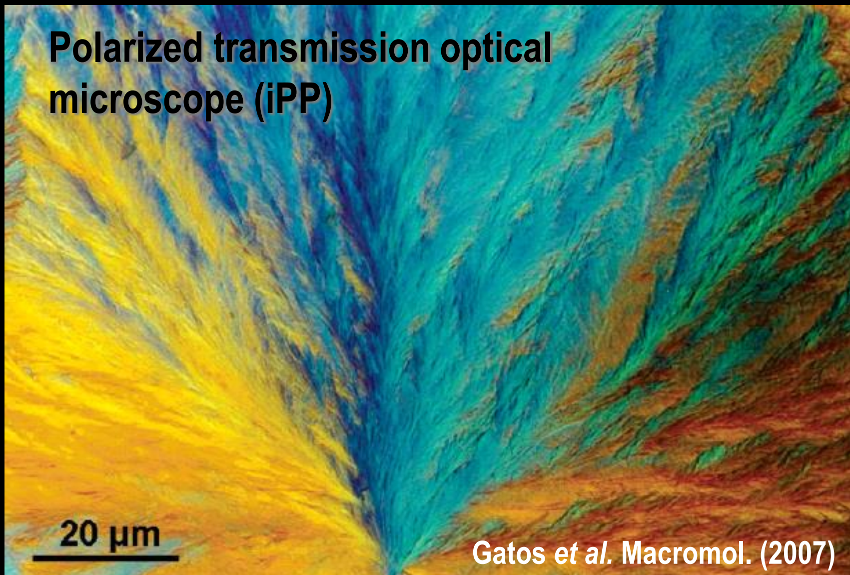


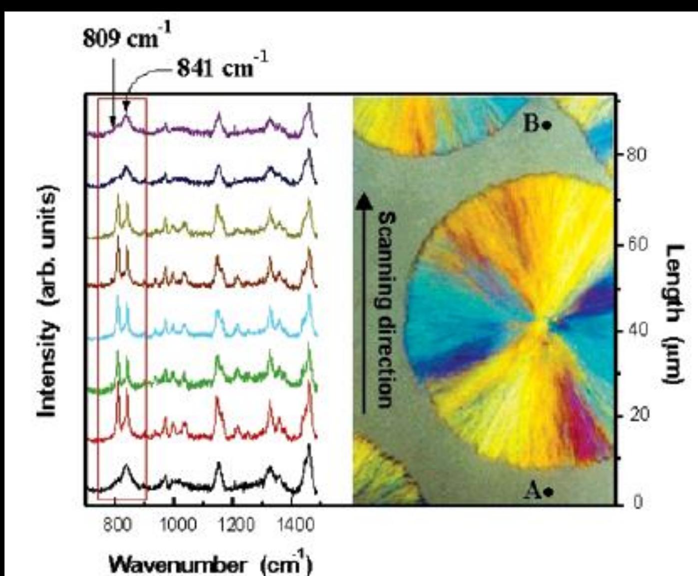
C. Morphological variability

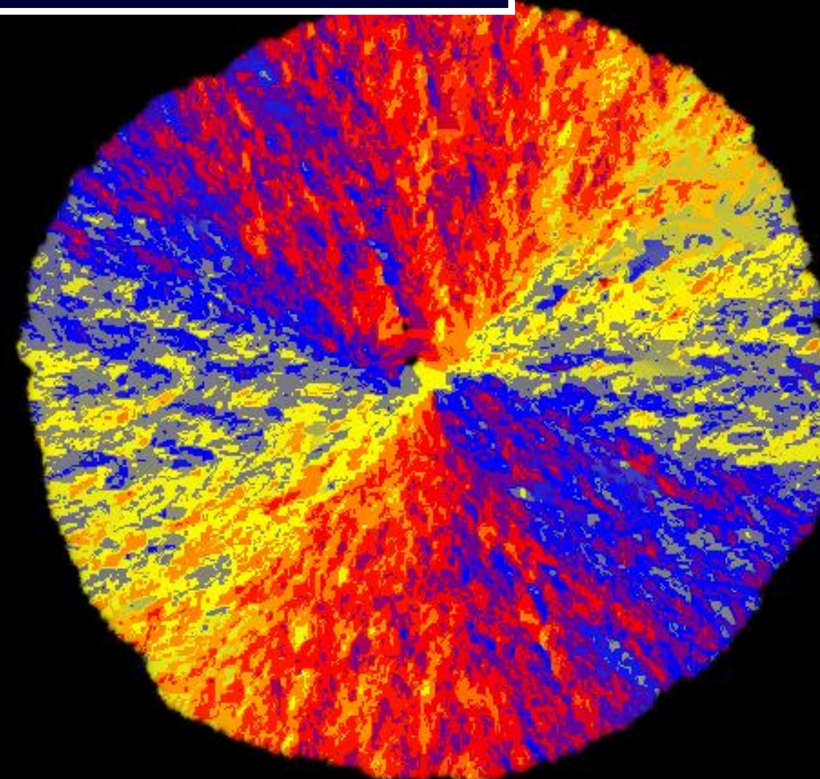
Description with only a few model parameters
(anisotropies, branching angle, MS well depth, …)



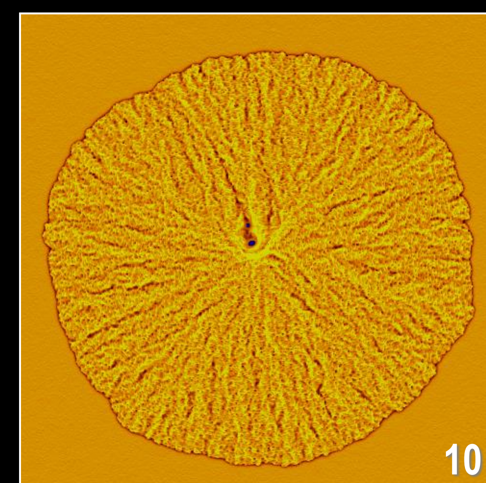
D. Comparison with experiment on orientation





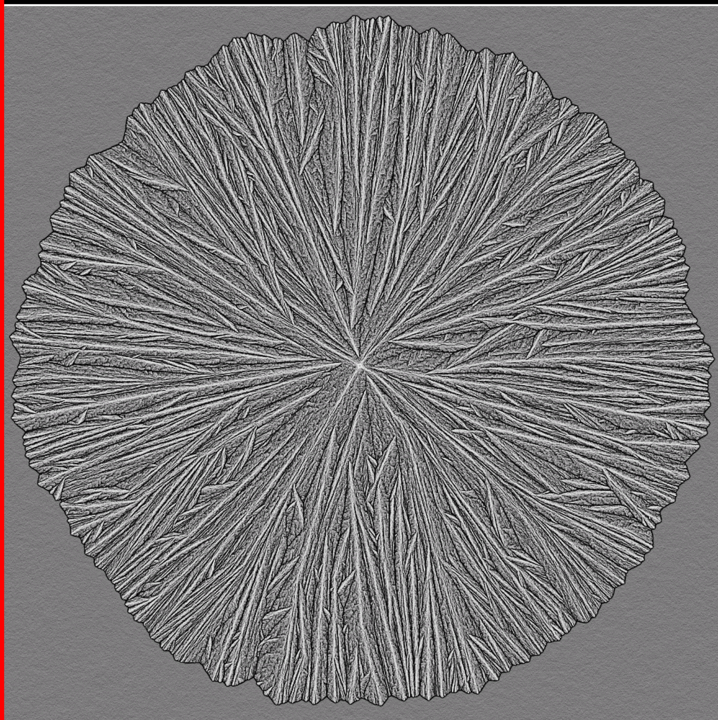


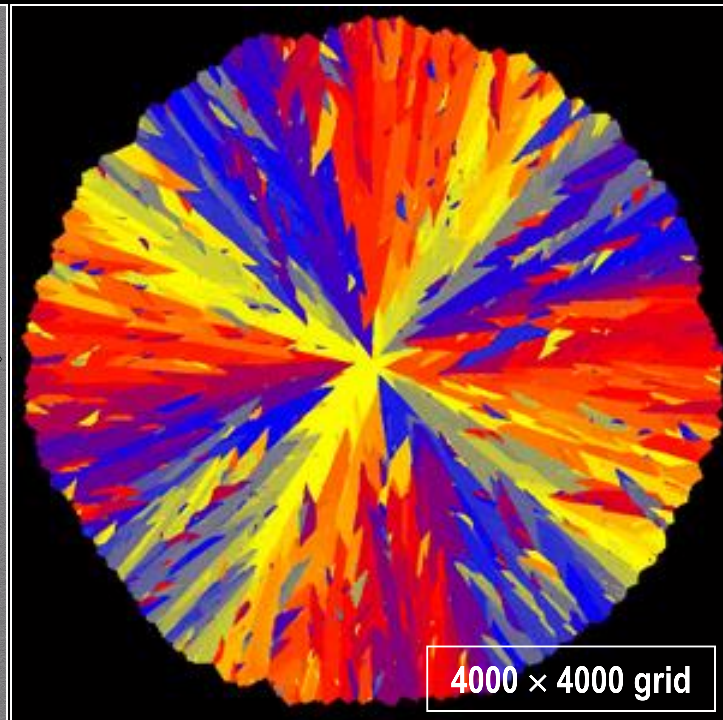
Phase-Field simulation

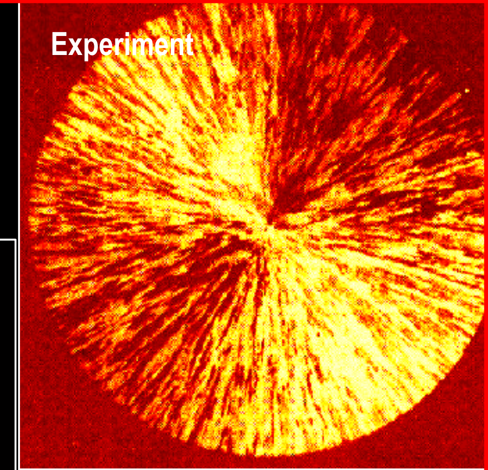


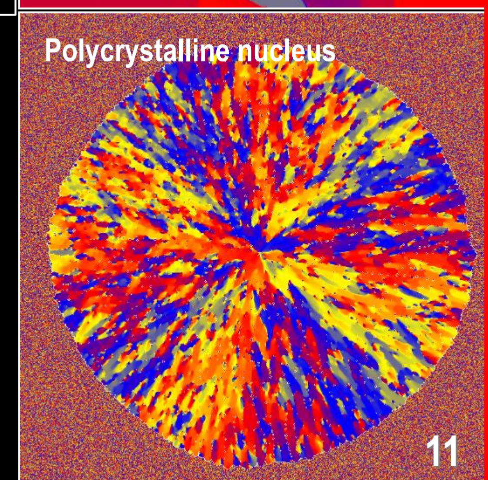
D. Formation of spherulite by GFN

Gradual transition from single crystal nucleus to Category 1 spherulite:



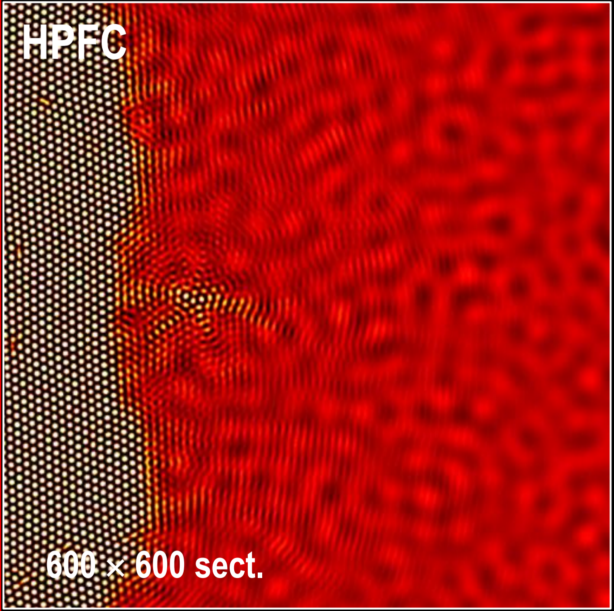


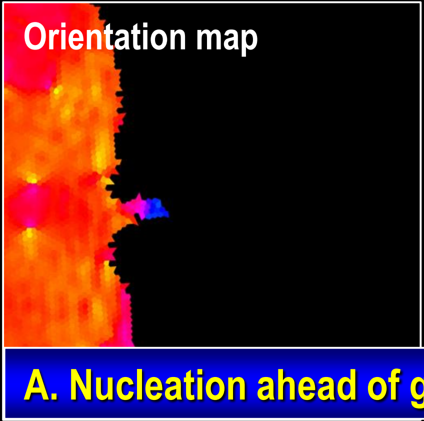


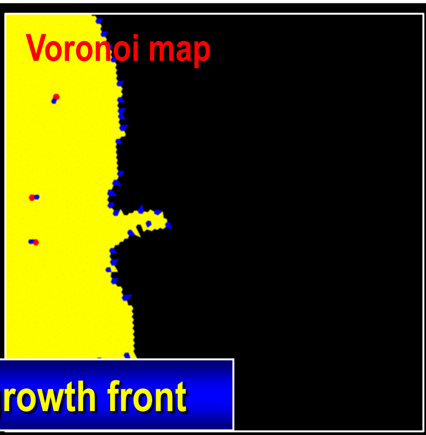


Atomistic view for GFN?

E. Two modes of GFN in hydrodynamic Phase-Field Crystal simulations:





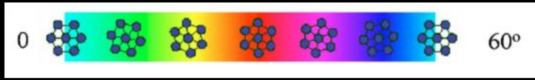


A. Nucleation ahead of growth front

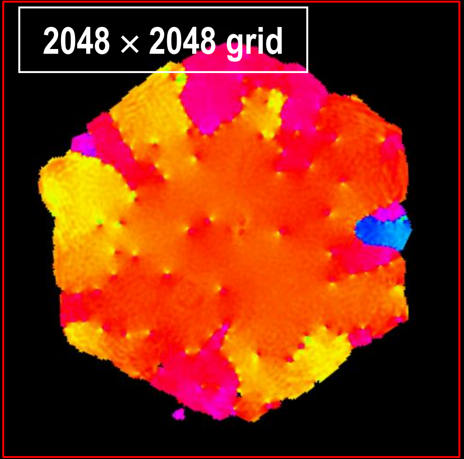
Structural analysis (complex bond oder parameter):

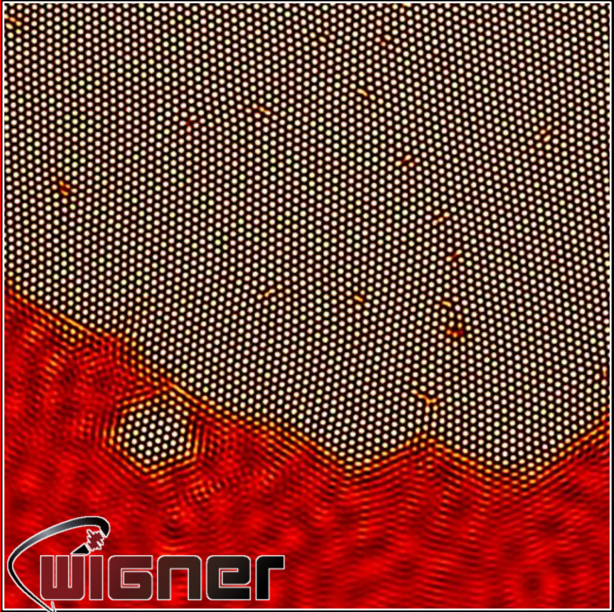
$$g_6 = \sum_j \exp\{i6\theta_j\}$$

- θ_j : angle towards *j*-th neighbor in lab. frame
- $|g_6|$: → degree of order
- phase: → local crystallographic orientation

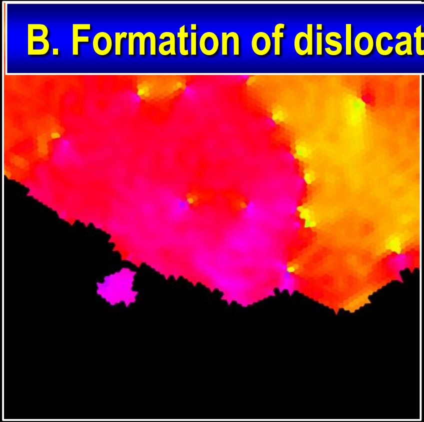


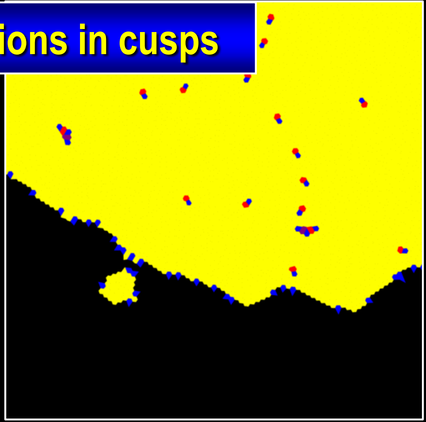
Voronoi analysis: 4 - grey; 5 - blue; 6 - yellow; 7 - red

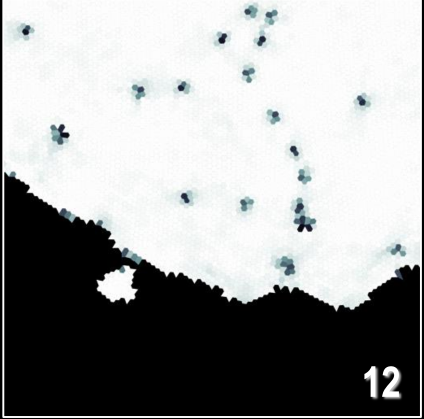




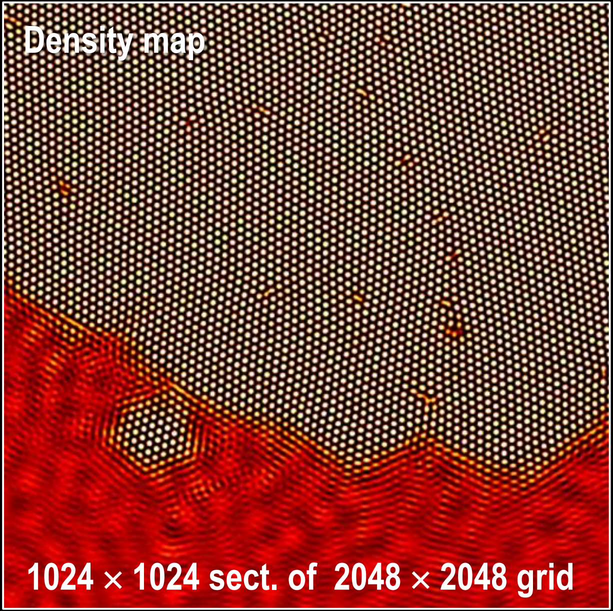
B. Formation of dislocations in cusps

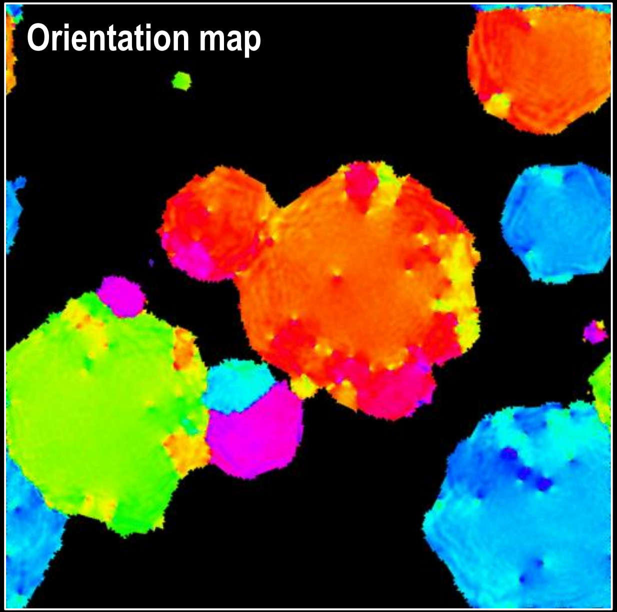


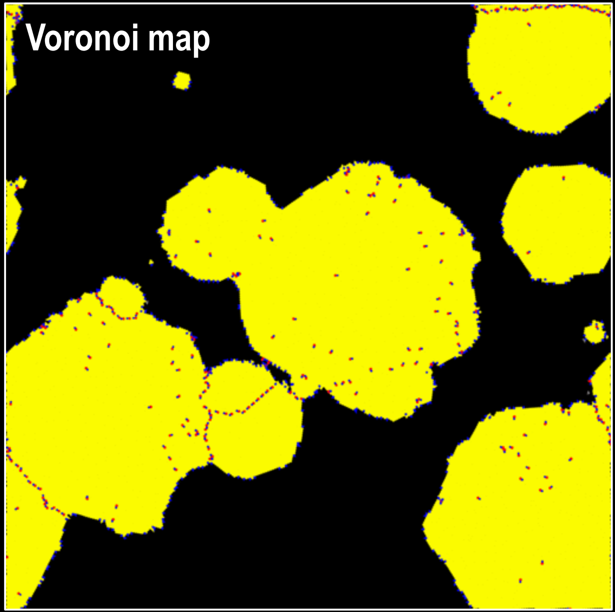




F. GFN by interference of density waves at front in the HPFC simulation:

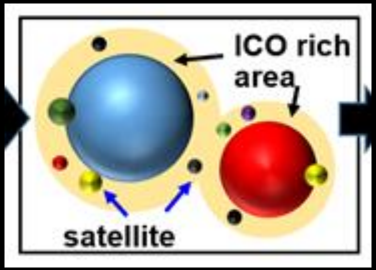


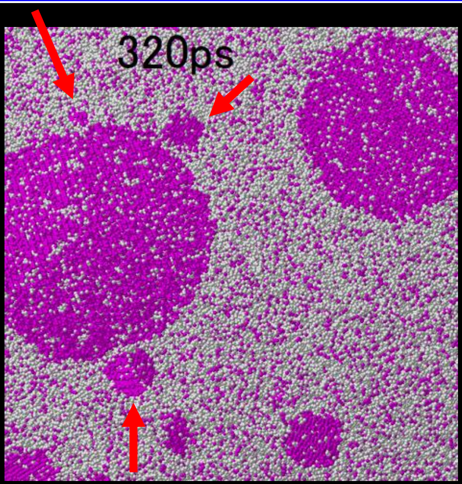




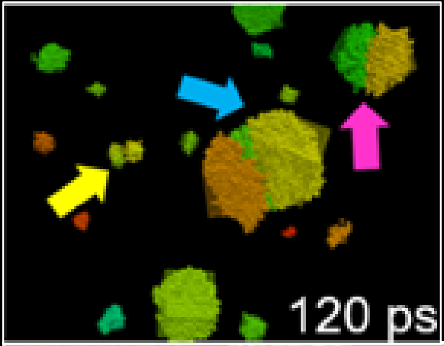
MD for 1 billion Fe atoms: ***Shibuta et al. Nature Comm. (2017)***

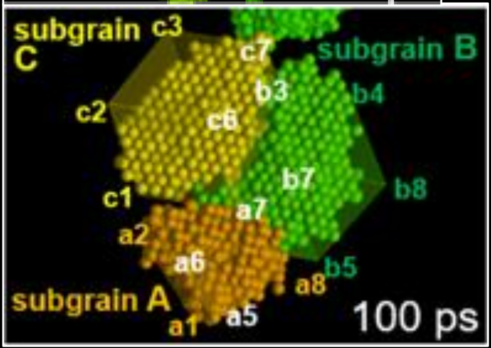
Satellite grains:
(red arrows)





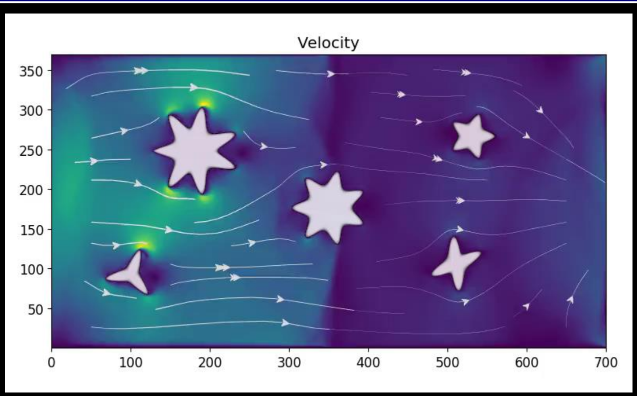
Multi-orientation crystallites:

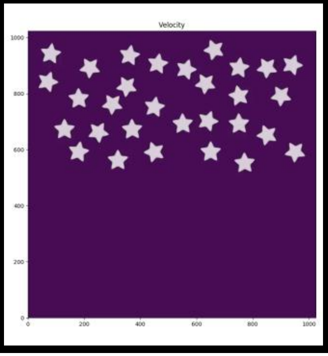


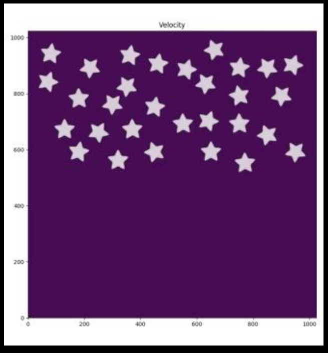


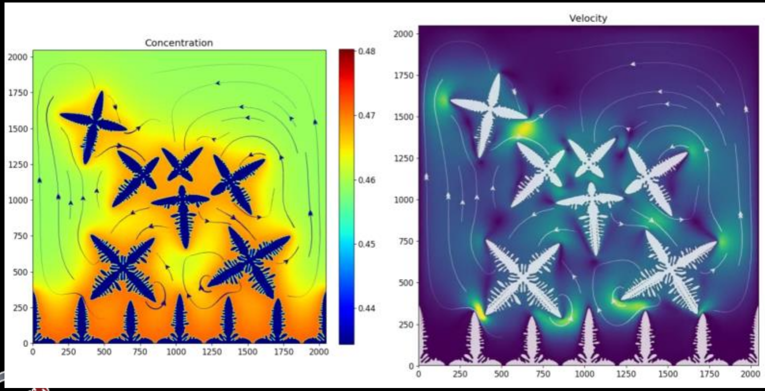
G. Other recent works:

I. Floating dendrites *(L. Rátkai et al.)*

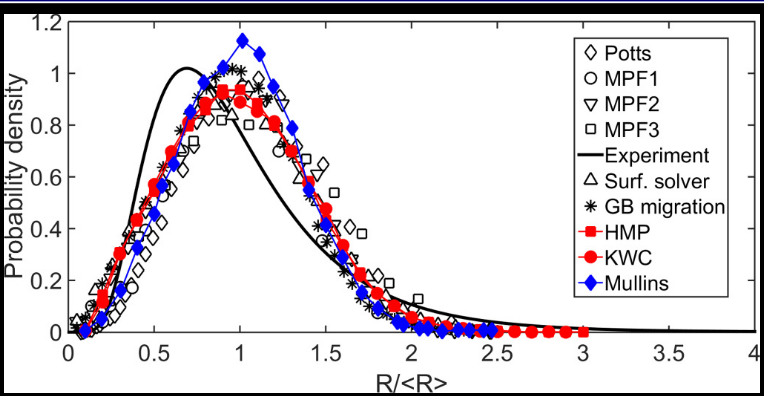




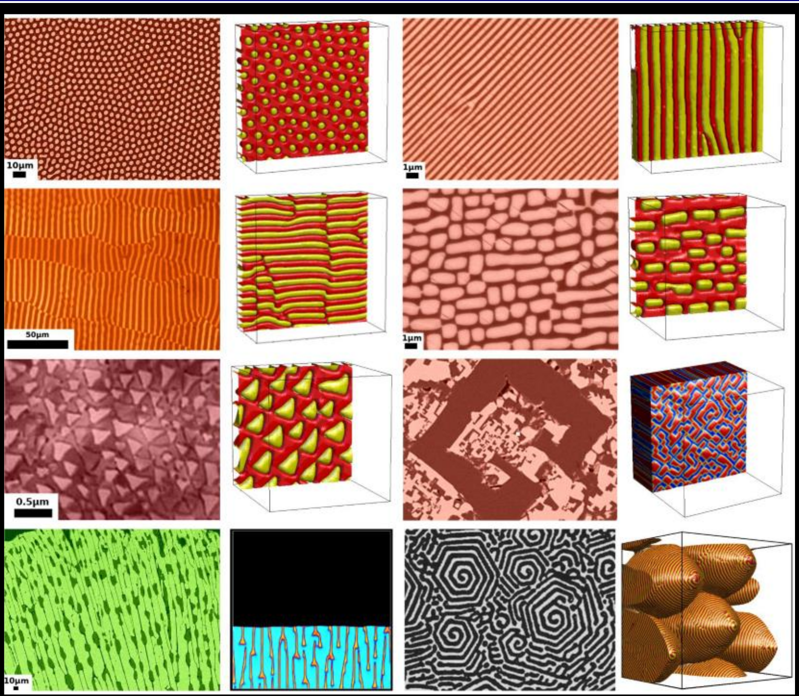




II. Grain boundary dynamics *(B. Korbuly et al. PRE 2017)*



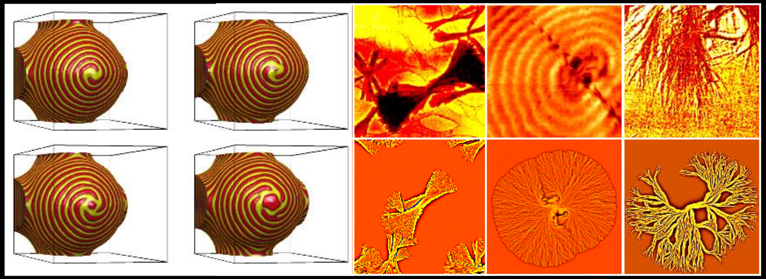
III. Anisotropic eutectics *(L. Rátkai et al. JMS 2017)*



IV. Summary: Main research directions

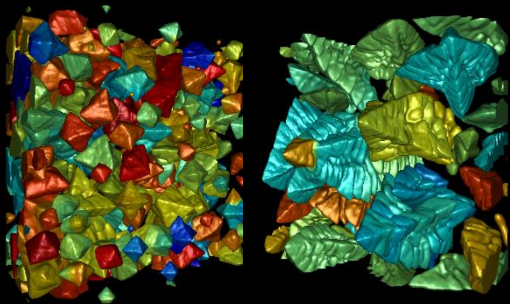
1. Modeling of exotic microstructures:

Phys. Rev. Lett. 2002; Nat. Mater, 2003, 2004 (IF = 10,8; 13,5);
Mater. Sci. Eng. Rep. 2004 (IF = 14,2); Europhys. Lett. 2005;
Phys. Rev. E 2013; Metall. Mater. Trans. A 2014;
J. Chem. Phys. 2015



2. Application of the Phase-Field (PF) model to materials of industrial interest:

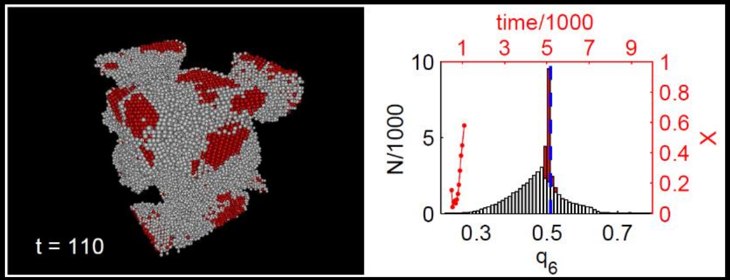
- optimization of soft magnetic alloys via phase selection (ESA Prodex/PECS)
- lead-free self lubricating bearing materials (ESA Prodex)
- high melting point alloys for gas turbine blades (EU FP 6)
- in-situ composites, particle-front interaction (ESA Prodex/PECS)
- production of metamaterials via eutectic solidification (EU FP7)



ESA website: “Space in videos”

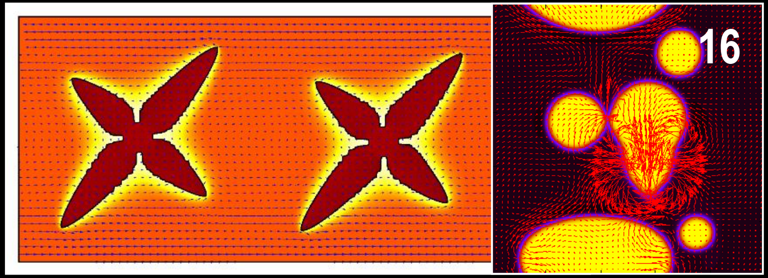
3. Molecular scale simulation of crystal nucleation (CDFT):

PRL 2011, 2012
Adv. Phys. 2012 (IF = 34,3)
Chem. Soc. Rev. 2014 (IF = 33,4)
Nat. Phys. 2014 (IF = 20,6)



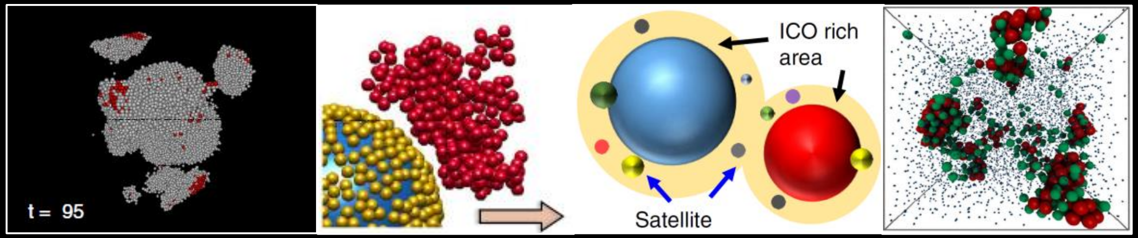
4. Modeling of multi-phase flow (PF + NS, PF + LB, HPFC):

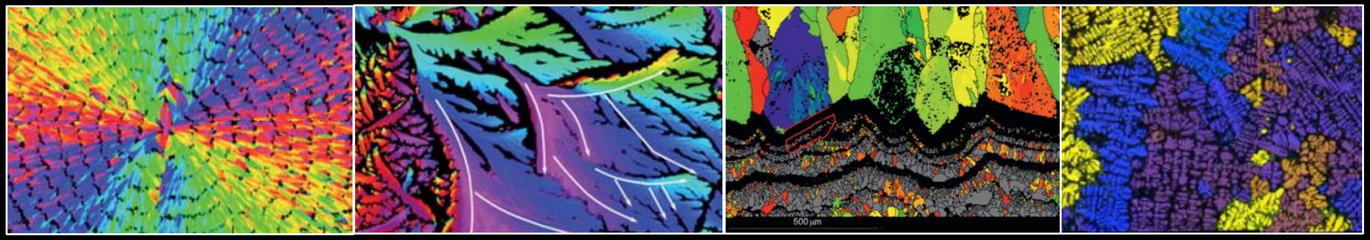
MSEA 2005; JPCM 2014; (ESA Prodex/PECS contracts)

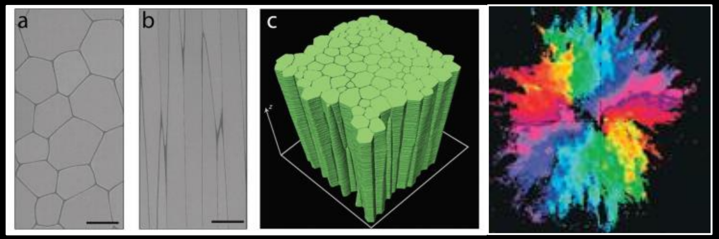


V. Future directions

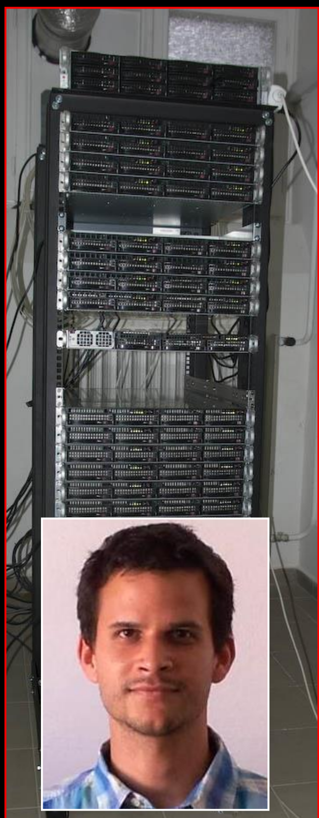
Molecular scale modeling of nucleation phenomena (HPFC)
Modeling of systems of more complex orientation maps
Modeling of crystallization in biological systems







Institute for Solid State Physics and Optics
WIGNER RESEARCH CENTRE FOR PHYSICS
Hungarian Academy of Sciences
H-1121 Budapest, Konkoly-Thege u. 29-33



László Gránásy
Sci. Advisor

Tamás Pusztai
Sci. Advisor

György Tegze
Senior Scientist

Gyula I.Tóth
Lecturer in Appl. Mathematics
Loughborough

Frigyes Podmaniczky
PhD student

Bálint Korbuly
PhD student

László Rátkai
PhD student

Computational Materials Science Group in WRCP:

László Gránásy	Prof.	- team leader nucleation, PF, DFT, …
Tamás Pusztai	Sci. Adv..	- nucleation, PF, topological defects
György Tegze	Sen. Sci.	- CFD, num. methods
Gyula I Tóth	Lecturer	- continuum models
Frigyes Podmaniczky	PhD student	- DFT, anisotropy, nucleation
László Rátkai	PhD student	- eutectics, LB flow
Bálint Korbully	PhD student	- grain coarsening, top. defects