

# Magnetism and pattern formation

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## **Abstract**

We are interested in pattern formation, mainly 2D ramified patterns resulting from interface instabilities. We are studying experimental systems where magnetism, through dipolar and Zeeman energy, and magnetic field effects, affects the growth processes. We will present three different systems, where magnetism acts in a different way.

The first one is “electrodeposition in a thin cell”. Magnetic field effects on the growth of a non-magnetic deposit result essentially from magnetohydrodynamic effects induced by Lorentz forces. On a magnetic deposit, dipolar interactions and Zeeman energy affect the growth processes.

The second system is “viscous fingering of a ferrofluid in a Hele-Shaw cell”. The growth is modified by a magnetic field in a way which depends on the orientation of the field with respect to the growth direction. The last system is a magnetic domain wall in a field gradient. When the wall is destabilized, ramified patterns may appear, but energy minimization maintains a well-defined domain size.

The main conclusion is that magnetism leads to an increased complexity in this kind of ramified patterns. The field effects can be understood in terms of stabilizing or destabilizing, depending on its orientation, and in terms of energy minimization leading to a characteristic length competing with the one defined by the instabilities.

## **References**

### **Electrodeposition**

- [1] V. Heresanu et al., *Magnetohydrodynamics*, 39 (2003) 459
- [2] S. Bodea et al., *Phys. Rev. Lett.* 83 (1999) 2612

### **Ferrofluids and viscous fingering**

- [3] R. E. Rosensweig, *Ferrohydrodynamics*, Dover, Mineola, NY, 1997
- [4] C. Flament et al., *Physics of Fluids* 10 (1998) 2464
- [5] E. Ben-Jacob et al., *Phys. Rev. Lett.* 55 (1985) 1315

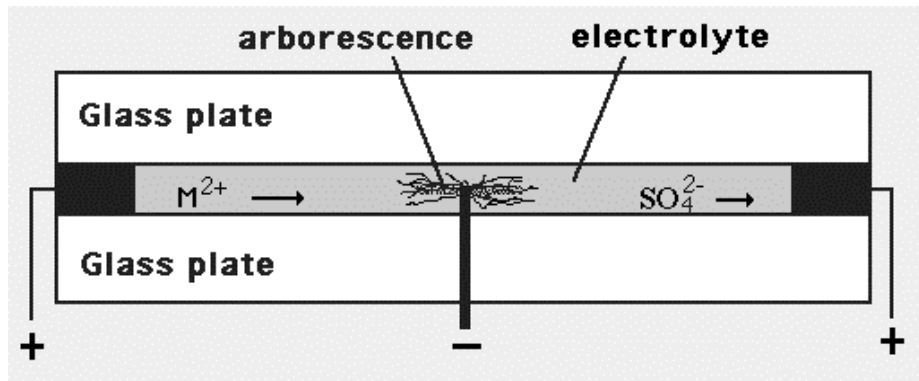
# Introduction

- Ramified, fractal-like patterns: common in nature
- Numerical simulations: DLA
- Experimental realizations:
  - solidification
  - electrodeposition
  - viscous fingering
  - ...
- Magnetism:
  - magnetic properties
  - magnetic field as control parameter
  - effects on the growth processes

# Outline

- **Electrodeposition experiments**
  - parameters, morphologies
  - Zn growth in normal field
  - Fe growth in in-plane field
- **Viscous fingering of ferrofluid**
  - Viscous fingering
  - Ferrofluid
  - Normal and in-plane field

# Electrodeposition in thin gap geometry



Electrolyte =  $M(SO_4) + H_2O$

$M = Zn \text{ or } Fe$

parameters



- ⇒ initial concentration
- ⇒ applied voltage (current)
- ⇒ thickness of the cell

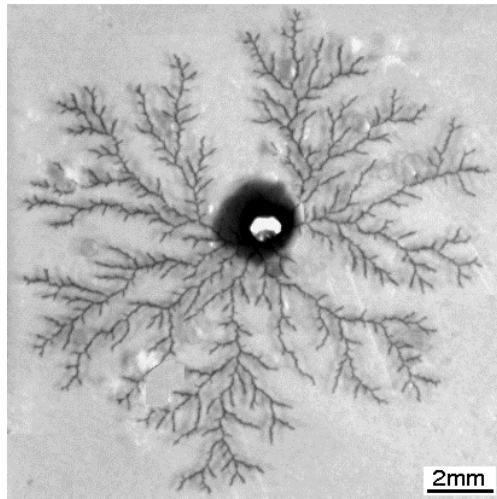
- ⇒ small thickness, 10-300  $\mu m$
- ⇒ cell diameter 20-150 mm

Quasi 2D

# Obtained morphologies for Zn

## DLA-like

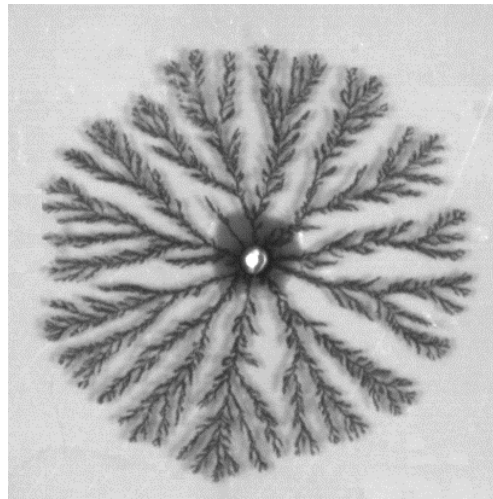
(Diffusion Limited Aggregation)



0.01 M; 2.6V; 50 $\mu$ m

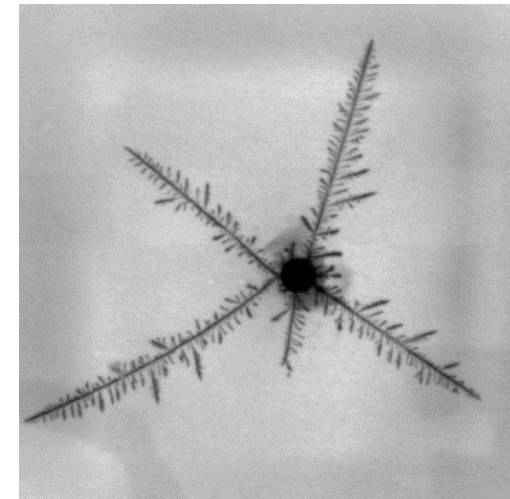
## DBM

(Dense Branching Morphology)



0.01 M; 4V; 200 $\mu$ m

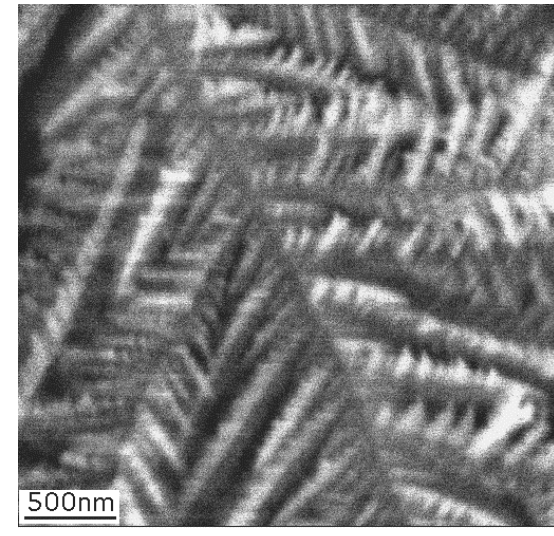
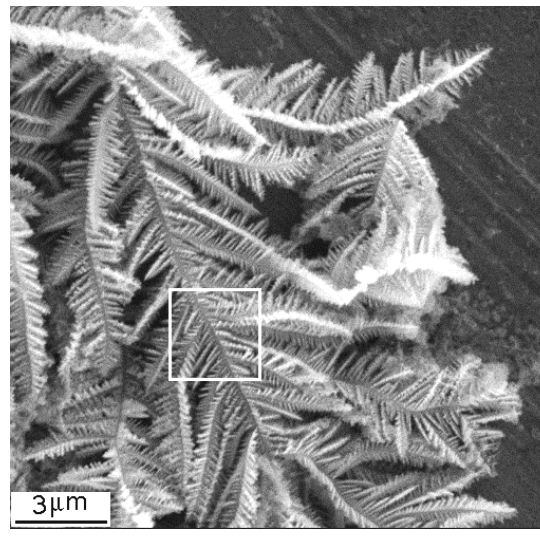
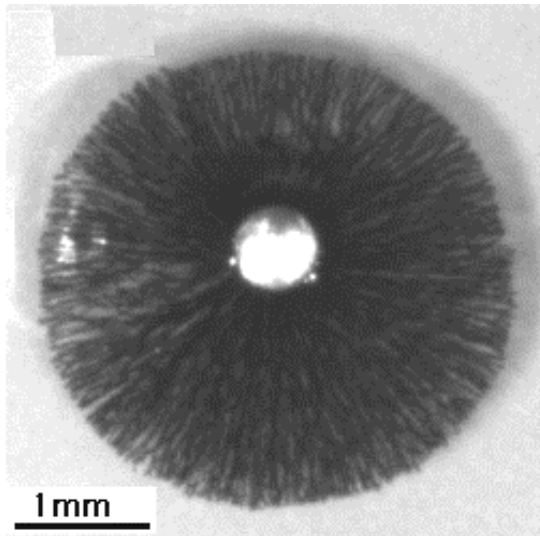
## Dendritic



0.3 M; 10V; 100 $\mu$ m

# Obtained morphologies for Fe

generally  $\longrightarrow$  DBM morphology



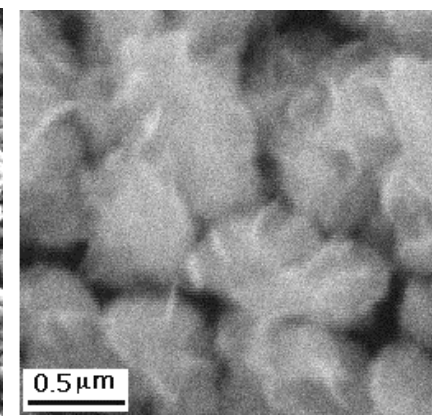
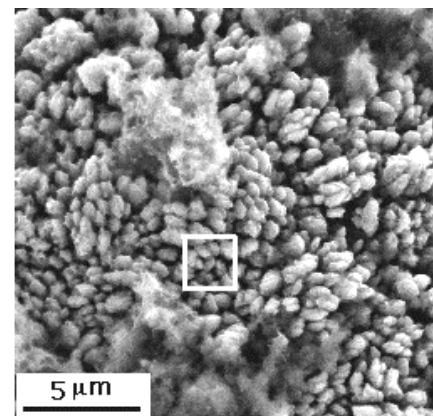
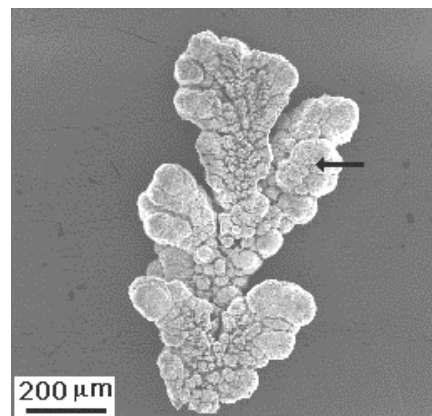
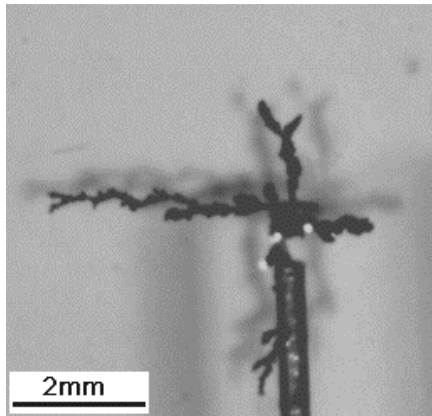
Dendrites at a small scale

Limitation due to H<sub>2</sub> bubbles

less often



ramified morphology



grains at a small scale

# Effects of a magnetic field on Zn arborescences

Effects which can be expected:

## Electrochemical effects

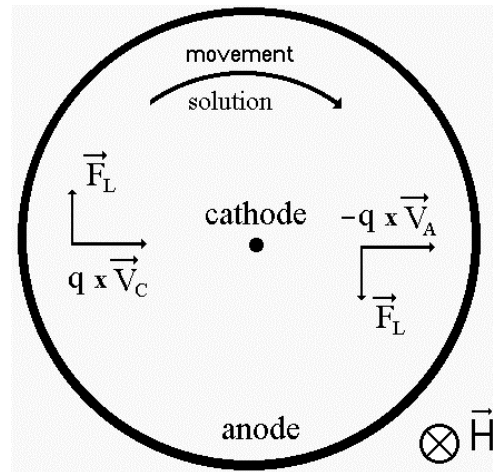
- ⇒ Lorentz forces on ions: negligible
- ⇒ effect of convection on ion transport
- ⇒ charge transfer at the interface electrolyte / electrode

## Specific to arborescences

- ⇒ fragile character of the aggregates
- ⇒ Laplace force on the branches

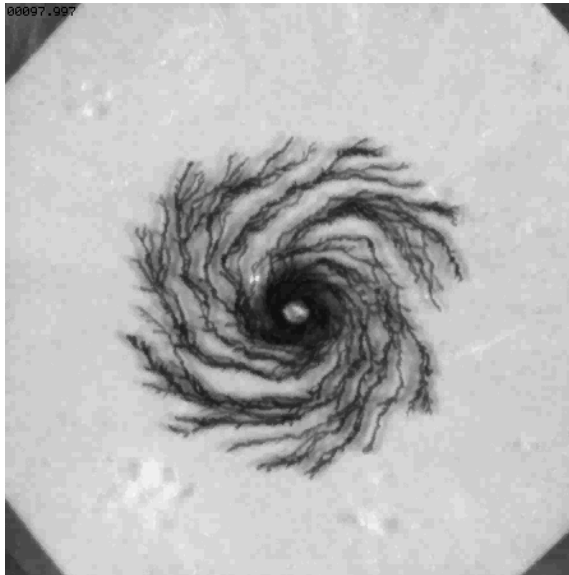
# Normal magnetic field

Lorentz force

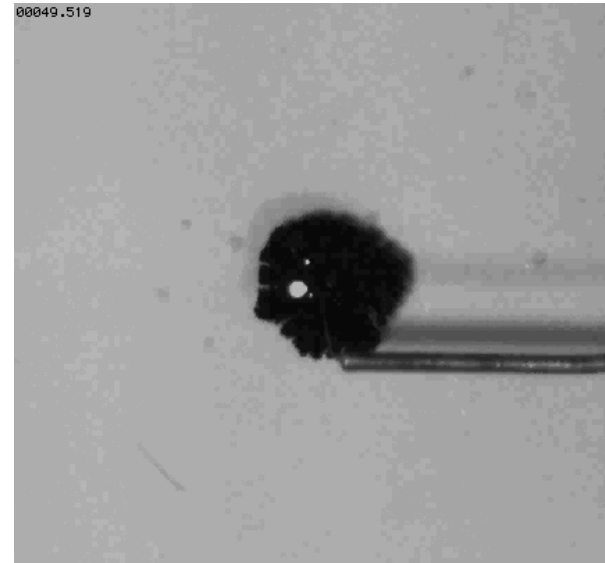


Observed effects

- $\Rightarrow$  spirals
- $\Rightarrow$  compactification
- $\Rightarrow$  dissymmetry



spiralling



compactification

## Origin of spiraling:

⇒ bias in ion motion?

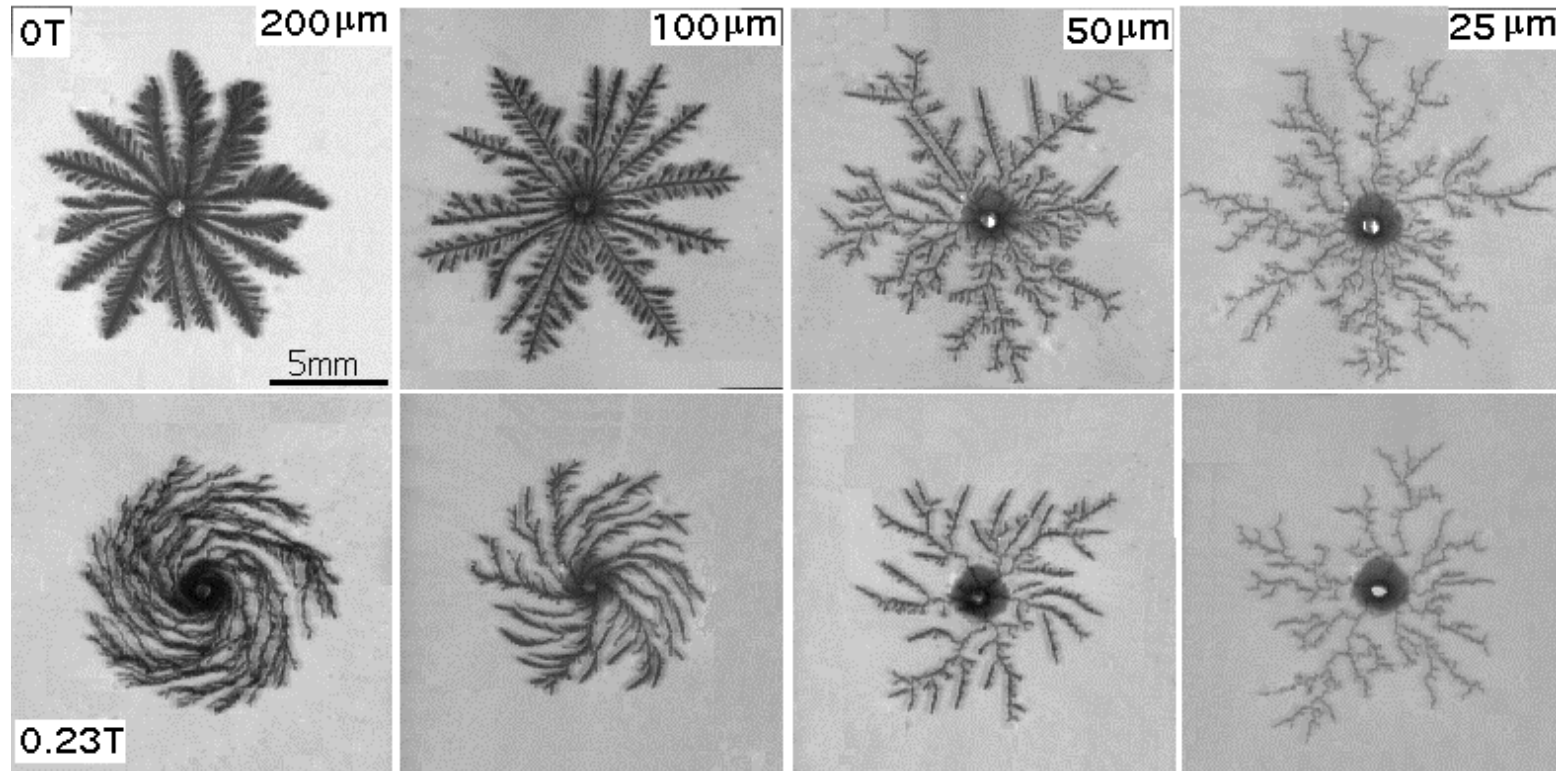
⇒ simulations and experiments ⇒ spirals in opposite sense

⇒ effects too large with respect to involved forces

mechanical action of the rotating fluid on the aggregate

in order to check, reduction of the fluid motion

# Cell thickness reducing

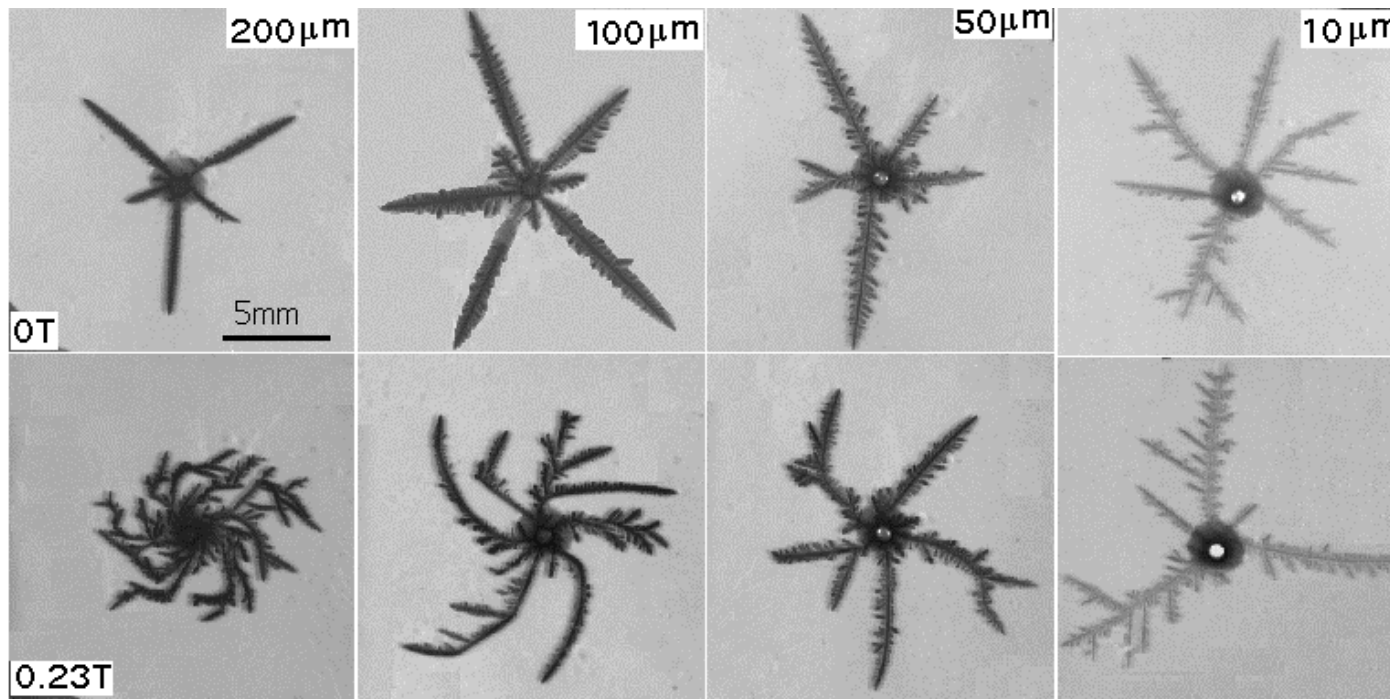


H

Zinc arborescences; concentration 0.06 M, voltage 10V

The spiral is inhibited by reducing the thickness of the cell  
*but*  
the morphology also changes

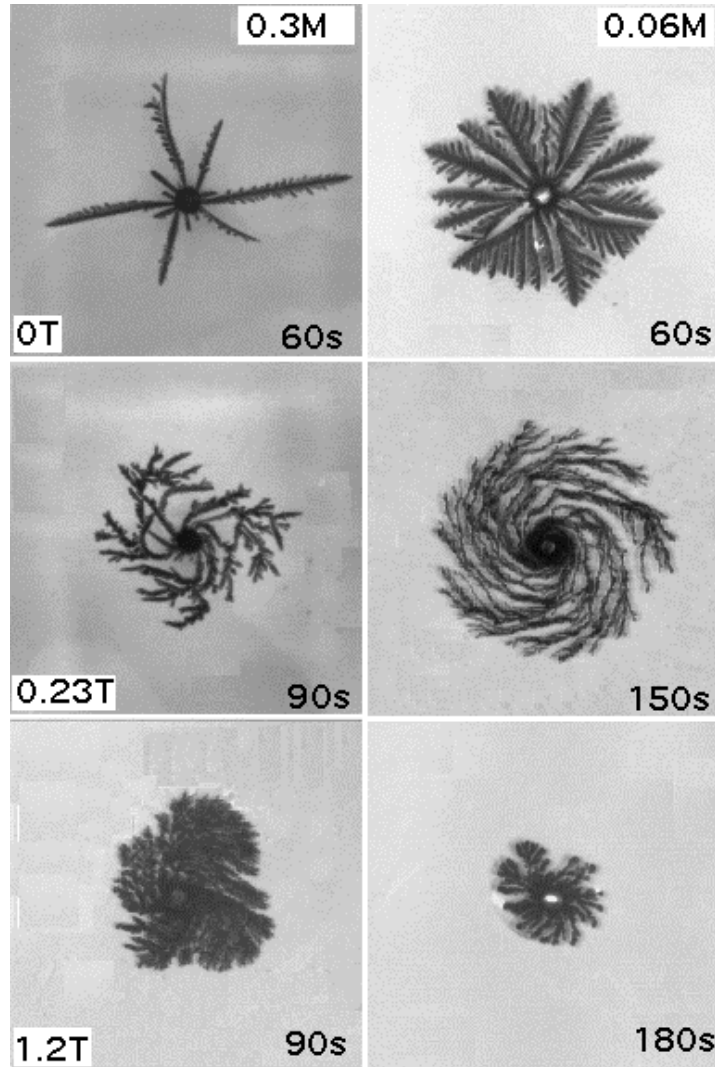
Conditions (0.3 M, 10 V) where the morphology is dendritic



H

No convective motion → no spiral

# Effects of the magnetic field amplitude



## Concentration **0.3 M**

⇒ spiral at 0.23 T

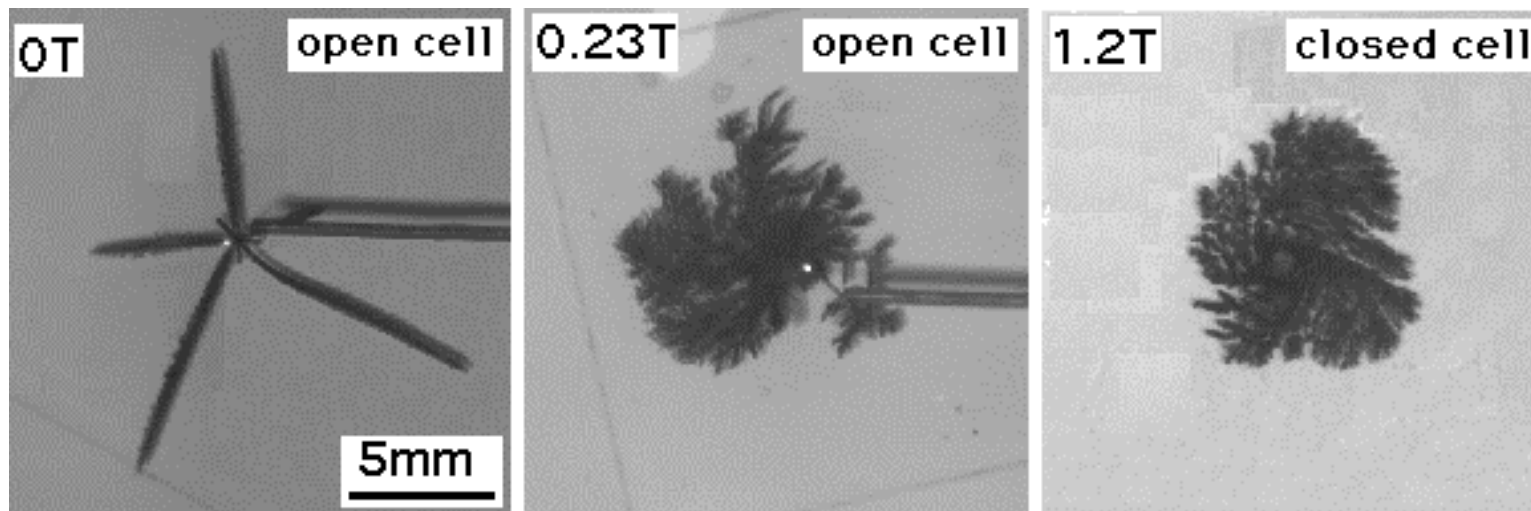
⇒ more compact at 1.2 T

## Concentration **0.06 M**

⇒ spiral at 0.23 T

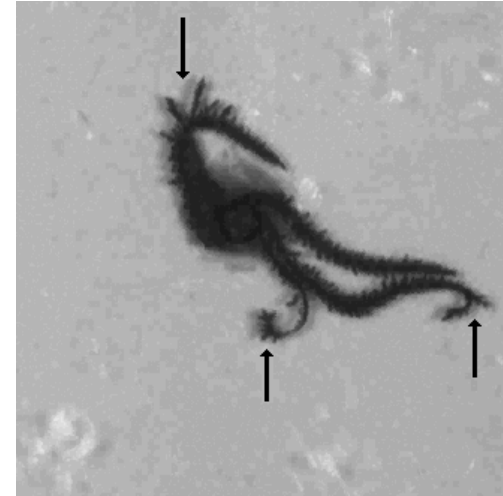
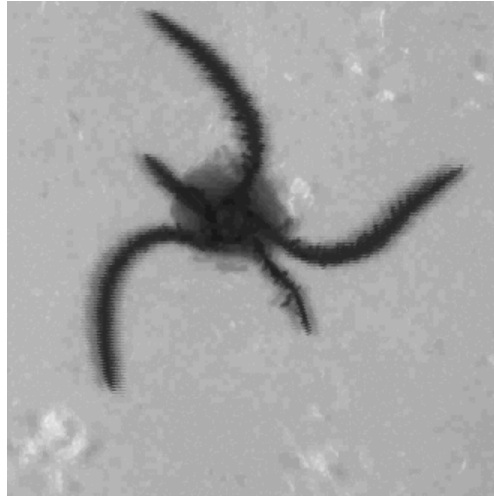
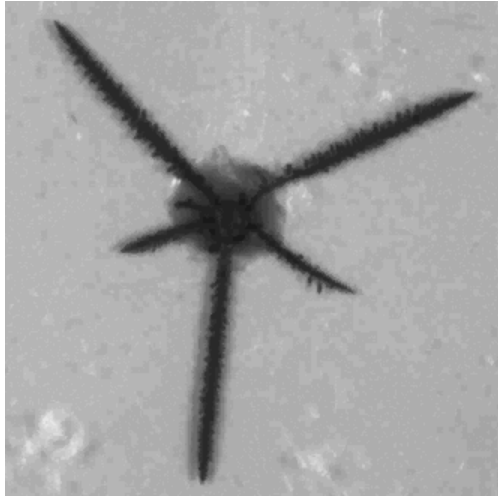
⇒ inhibition of the growth at 1.2 T

# Growth in open cell



same effects for lower values of the field

# H applied on a growth obtained without field



Bending zone

⇒ still electric contact

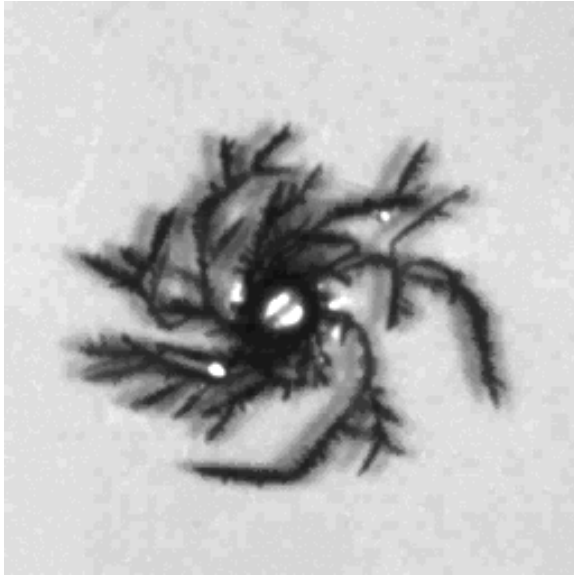
⇒ apparition of new branches

Many bending



More compact growth

# Comparison with mechanical rotation



field 0.23 T  
(real time = 40 s)



mechanical rotation (16 rpm)  
(real time = 40 s)

H effects are reproduced by the mechanical rotation

(but velocity profiles are not the same)

# Conclusion

## **Magnetic field effects on zinc arborescences**

### **Normal** magnetic field

- ⇒ effects due to the mechanical action on the aggregate of the convective motion generated by Lorentz force *and to the Laplace force on branches*
- ⇒ weak effects at a small scale for 0.3 M but morphological changes for 0.06 M
- ⇒ no effects on the crystalline structure

### **In-plane** magnetic field

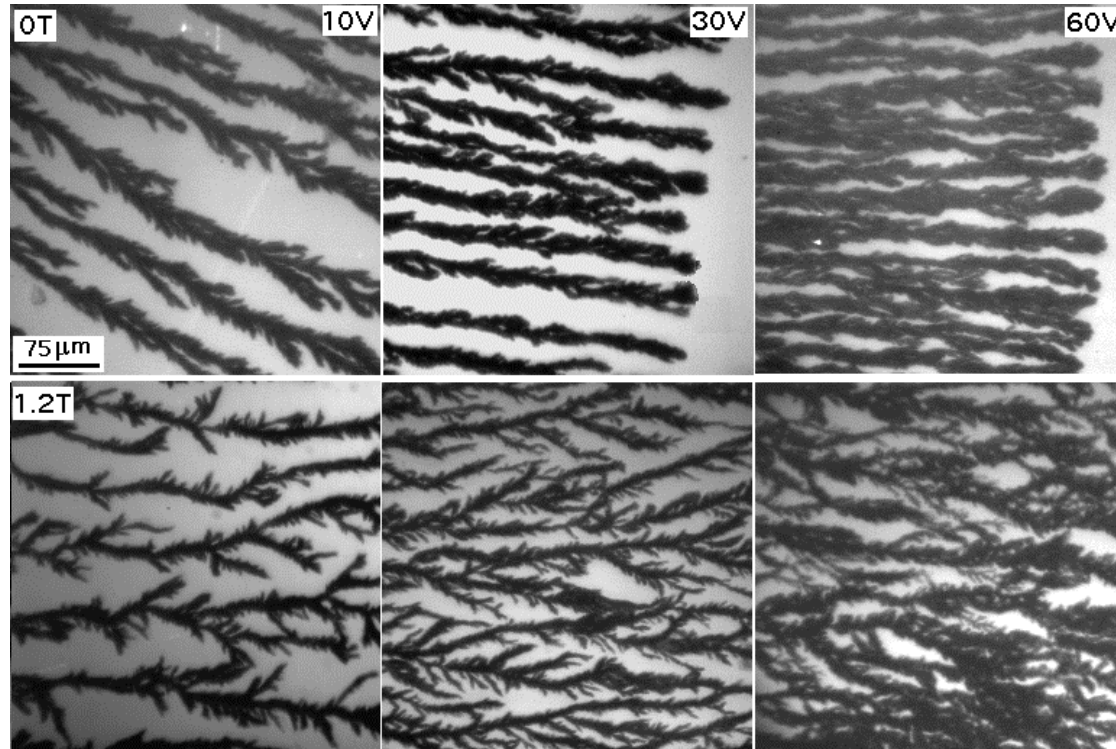
- ⇒ weak effects
- ⇒ no specific effects with respect to field direction

# Magnetic field effects on iron arborescences

Magnetic aggregate

Normal field effect: very weak



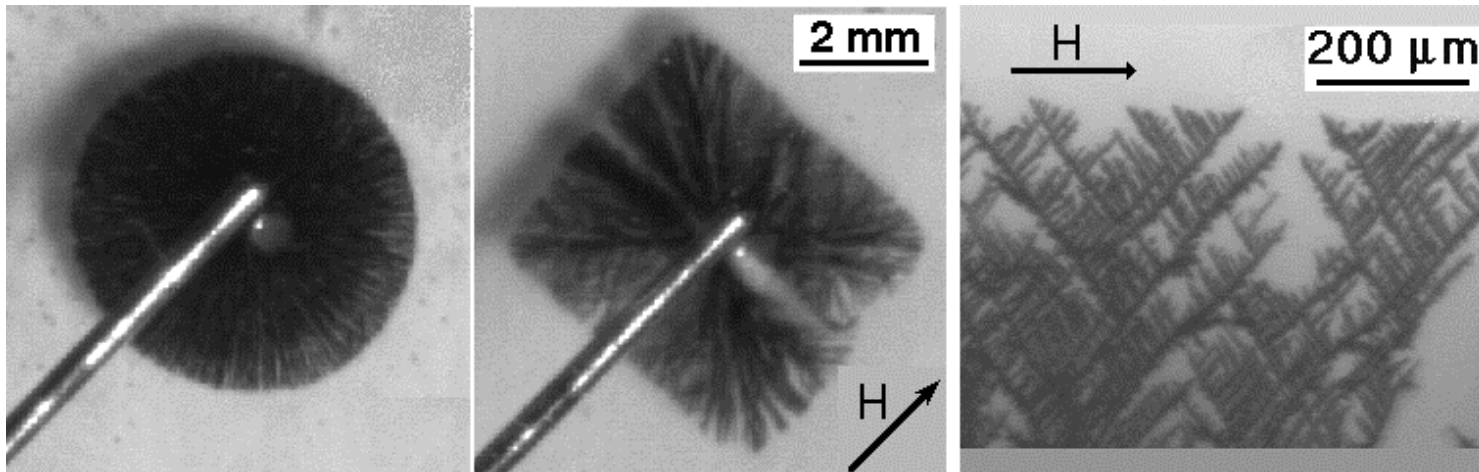


Concentration 0.3 M; cell thickness 10μm

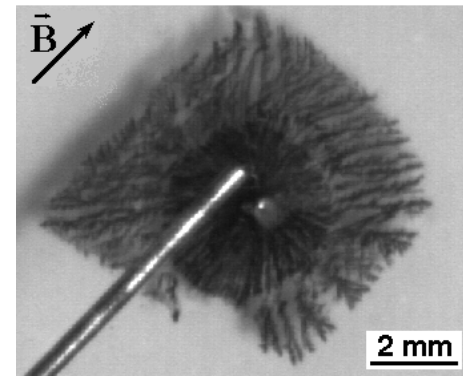
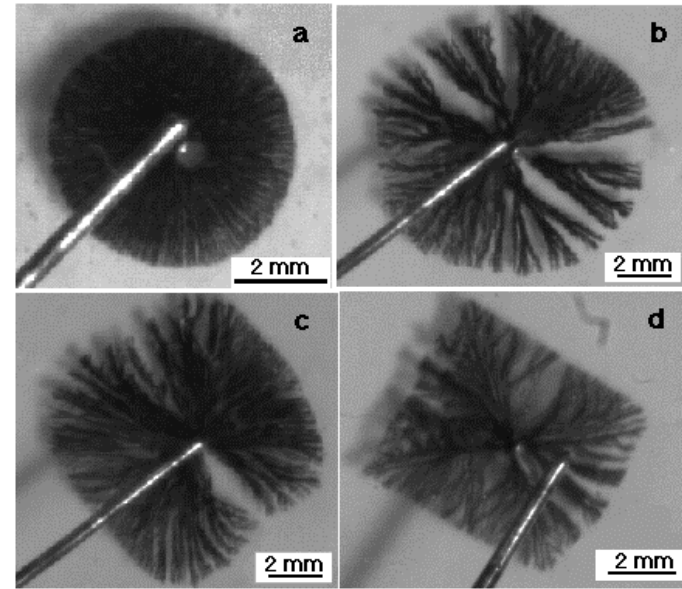
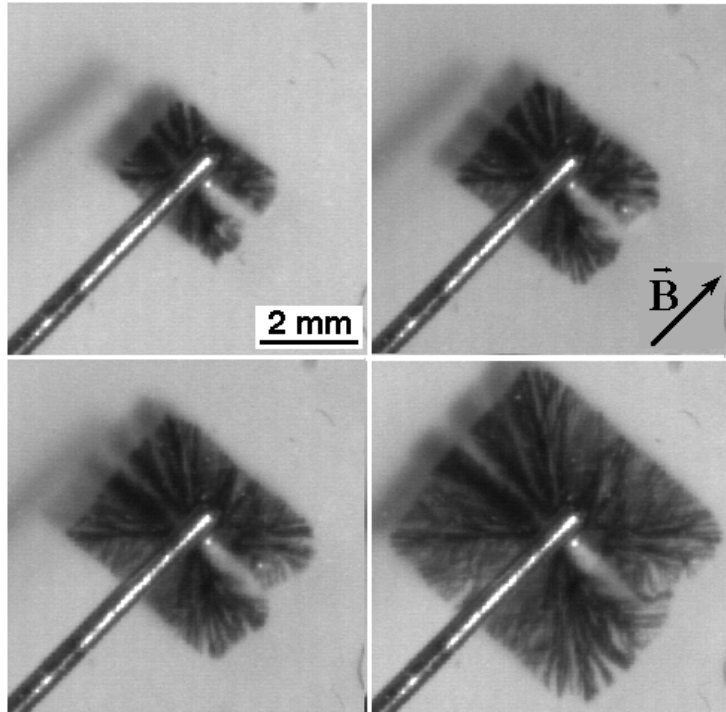
Rigidity induced by magnetism  
(dipolar interactions) and morphology

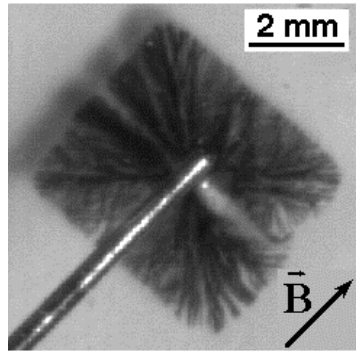
# In-plane magnetic field

Symmetry breaking: circular  $\Rightarrow$  rectangular

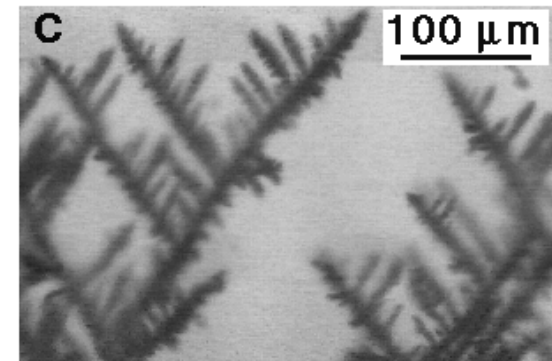
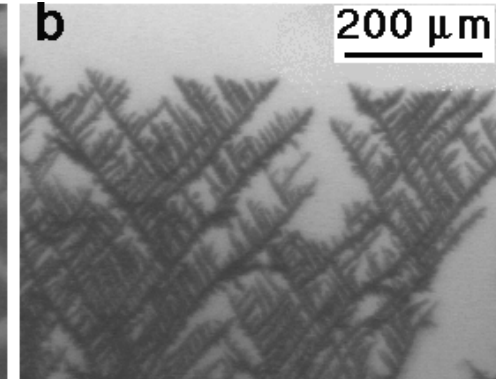
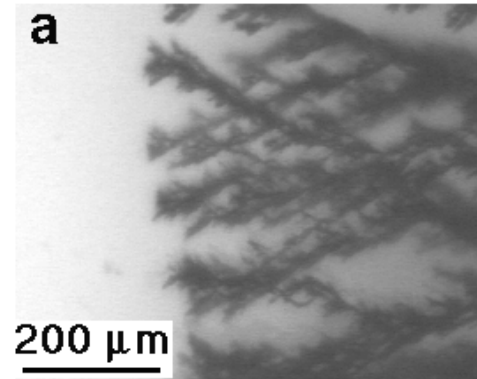


H selects two directions of growth, symmetrical with respect to its direction and to its perpendicular

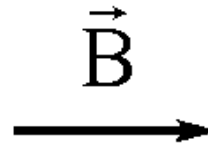
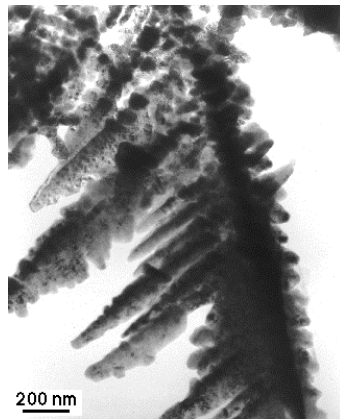




optical microscopy



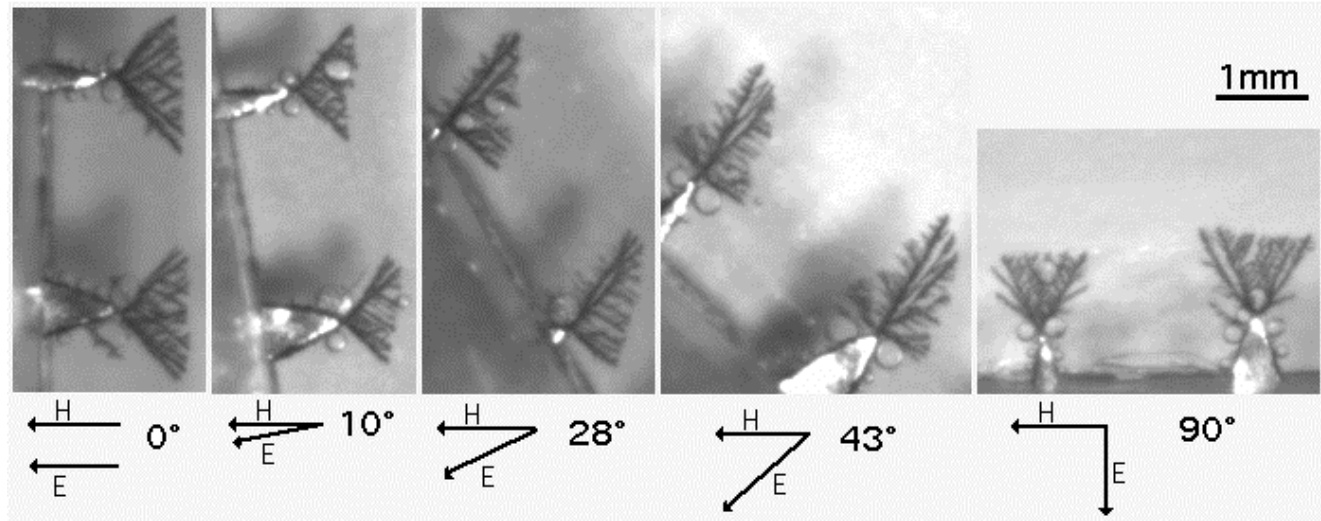
TEM:  
singlecrystalline  
dendrites



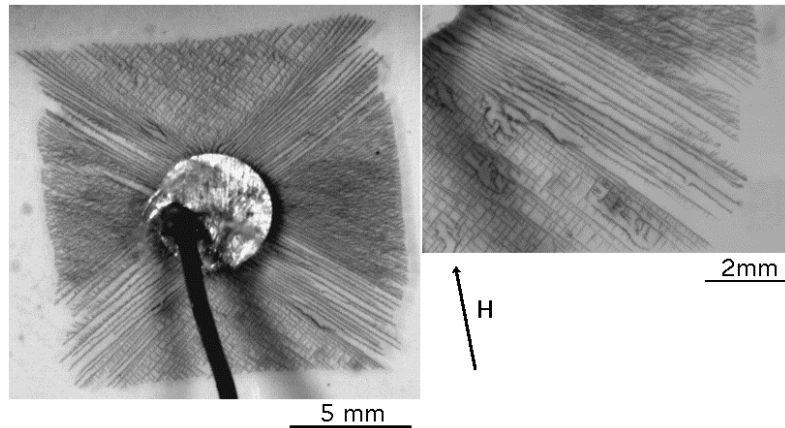
selection of growth directions

# Angle variation between E and H

Growths in quasi - parallel cell



Growths from  
a large  
cathode



Straight branches  
in selected directions

**Two selected  
directions**

# Conclusion

## **In-plane magnetic field effects on iron arborescences**

Branches orientation function of their structure :

⇒ parallel to H for granular branches

⇒ at a definite angle from H for dendritic branches

Order induced by the magnetic field seems related to the crystalline anisotropy of the dendrites

⇒ Idea of viscous fingering experiments to compare

# Viscous fingering experiments

## Experimental setup

Hele-Shaw cell

anisotropy

ferrofluid and oils

## Experimental results

growth with no field

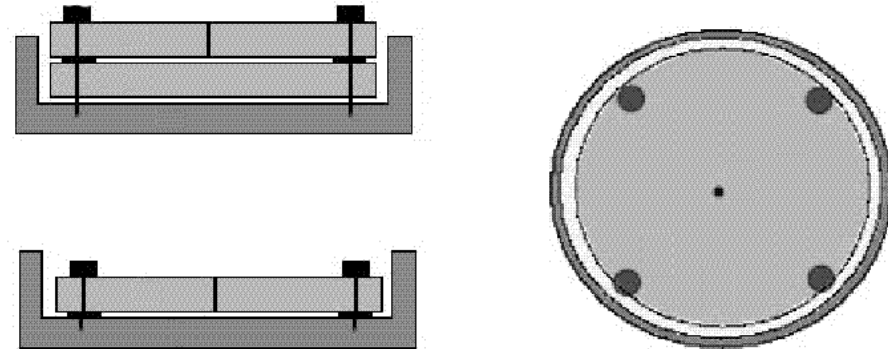
growth in a field normal to the cell

growth in a field parallel to the cell

# Experimental setup

## Hele-Shaw cell

- Plexiglas plates 5mm thick
- Cell diameter 50 mm
- Cell thickness 0.15 to 0.5 mm



## Anisotropy

Square or hexagonal symmetry  
by engraving circular elements  
diameter, distance, depth: 0.5 mm



**Ferrofluid :** colloidal suspension of nanoparticles  
surfacted or charged particles  
superparamagnetic  
no sedimentation, no aggregation  
  
mixture of water and glycerol  
viscosity 10 mPa.s

**Silicon oil :** viscosity between 200 and 30000 mPa.s

**Many parameters:** cell thickness, viscosity, pressure  
anisotropy, magnetic field

**Qualitatively:**

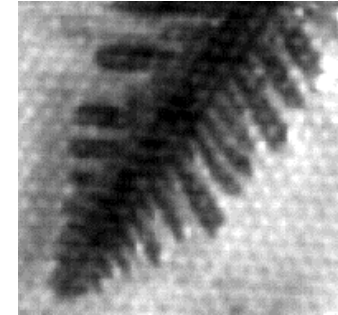
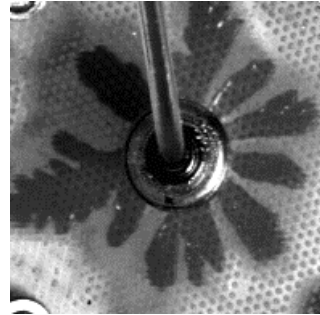
- slow growth regime, faster, ...
- no field, normal or parallel field
- with or without anisotropy

# Experimental results

## I - With no field

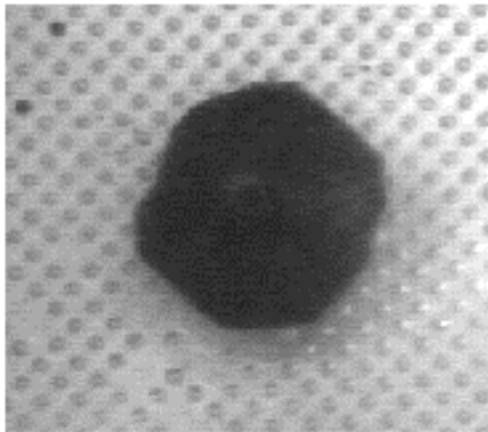
Like usual liquid

water  
into oil

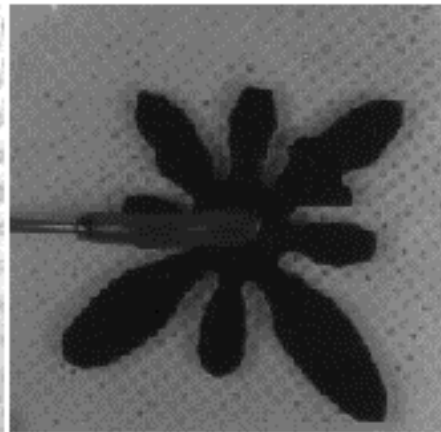


**slow injection:** equilibrium  
shape, circular or faceted

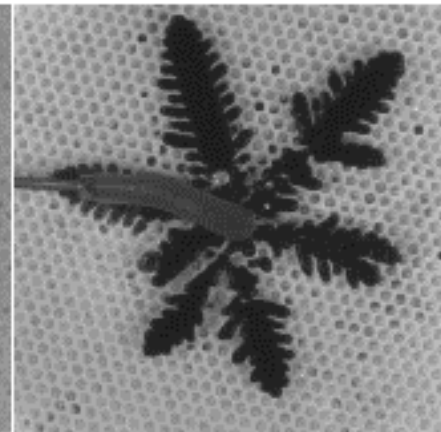
**injecting faster:** oriented  
fingers or dendrites



[0.30, sq\*, 200]



[0.20, sq , 1000]



[0.30, hex , 29100]

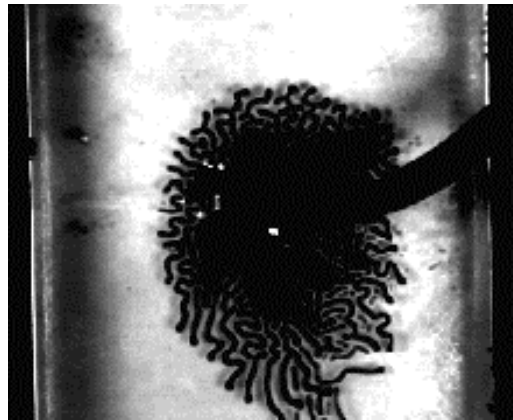
**II - With a field normal to the cell plane ( $H = 0.23 \text{ T}$ )  
no anisotropy**

**no injection:** equilibrium shape, labyrinth

[0.50, - , 200]

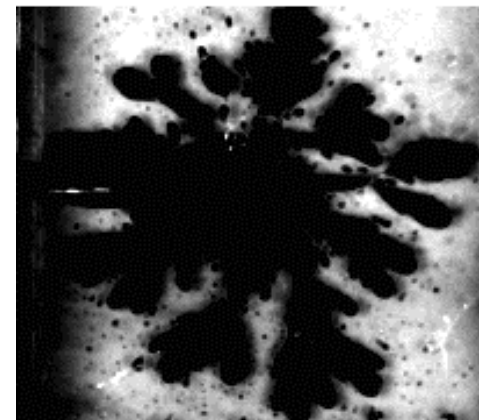


**slow injection:**  
homogeneous labyrinth



[0.30, - , 200]

**faster injection:**  
disordered fingering

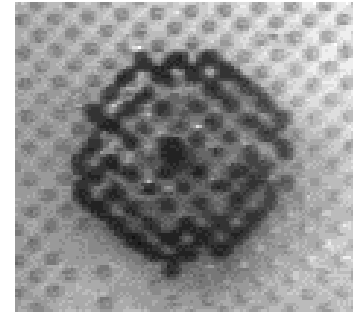
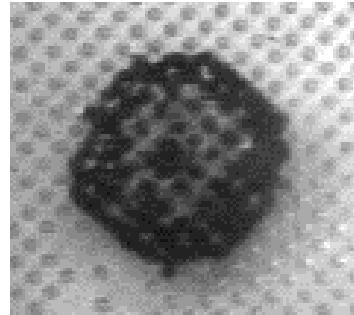
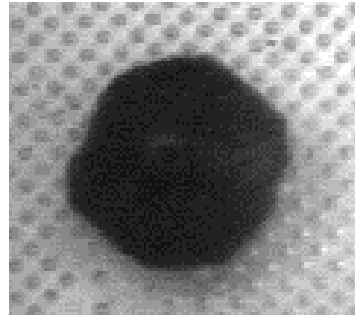


[0.15, - , 1000]

## with anisotropy

normal field: filling of the holes with ferrofluid

**no injection:**  
preserved envelope  
discrete pattern



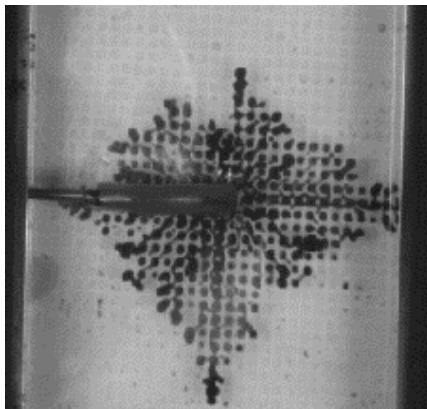
[0.30, sq\*, 200]

homogeneous filling  
overall symmetry

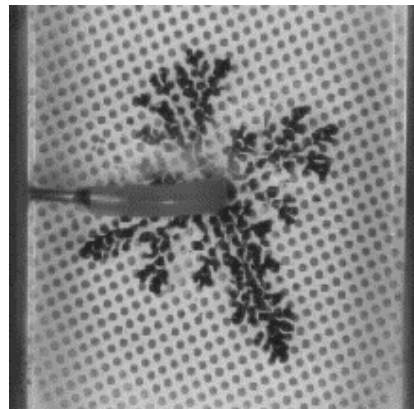
random filling: DLA

dendrite skelton

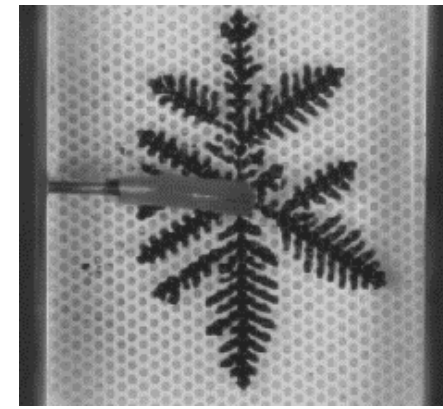
**injection:**



[0.20, sq , 1000]



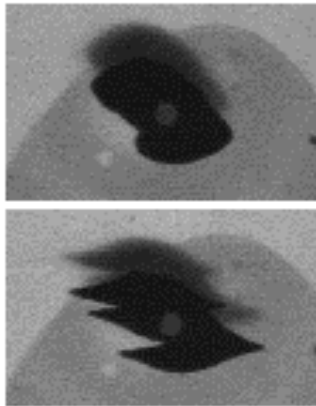
[0.15, sq , 29100]



[0.15, hex , 29100]

### III - With a field parallel to the cell plane

no injection or injecting slowly:  
equilibrium shape, spikes



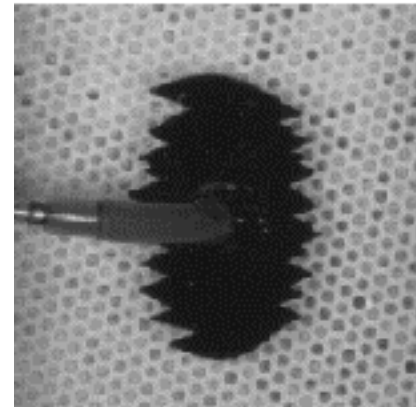
[0.15, - , 1000]

1.2 T



[0.15, sq , 200]

0.23 T



[0.15, hex , 200]

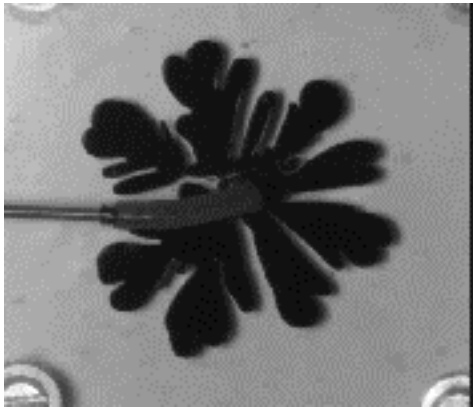
0.23 T

## no anisotropy

**injecting faster:**

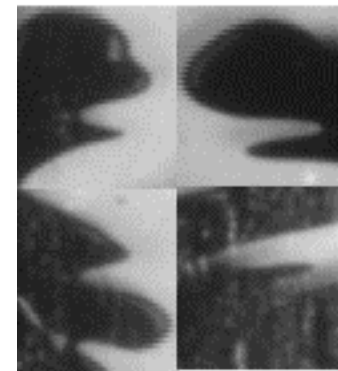
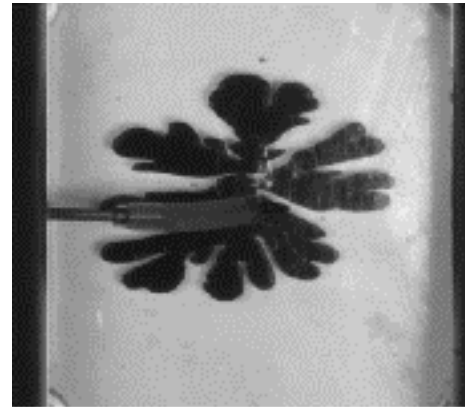
- fingering, modification of zero field pattern
- envelope becomes more elliptic
- fingers depend on their orientation
- spikes

zero field



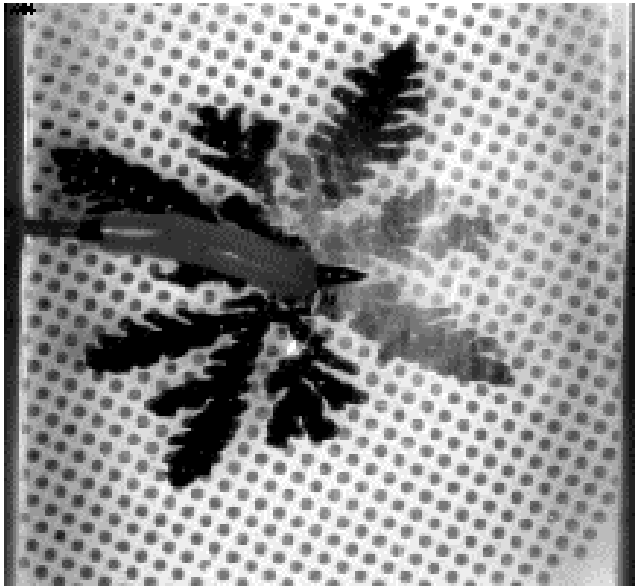
[0.30, -, 29100]

0.23 T

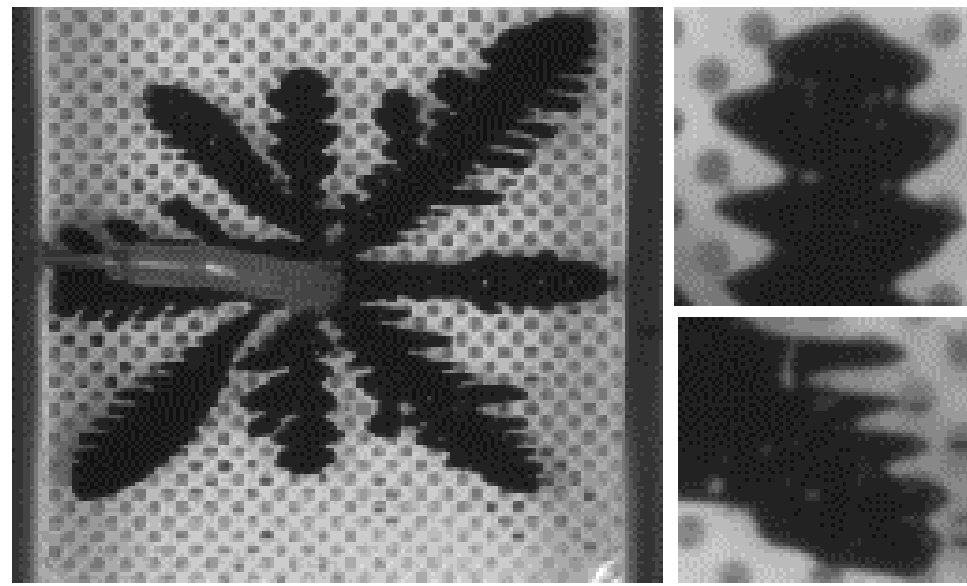


## with anisotropy

- again different effect depending on growth directions
- competition between anisotropy and field direction
- normal field instability, spikes, dissymmetry



[0.20, sq , 29100]



[0.20, sq , 29100]

# Conclusion

## Effects of the magnetic field on viscous fingering

Changes the equilibrium shape of a drop of ferrofluid

⇒ labyrinth in normal field

⇒ « eye-shaped » in parallel field

When superimposed to injection:

⇒ stabilizes interfaces parallel to the field

⇒ destabilizes interfaces normal to the field

This leads to local morphological modifications and more complex patterns, even more when anisotropy, which allows to obtain dendrites, is added.

# General conclusion

## **Case of Zn :**

- Indirect effect of the magnetic field
- Complex situation where hydrodynamics is added to electrochemical growth

## **Case of Fe :**

- The magnetic field induces some order in the pattern by giving a reference to the local anisotropy via dipolar interactions

## **Case of viscous fingering :**

- While the global anisotropy gives ordered patterns, the magnetic field induces disorder since its effect are different for branches growing in different directions