

Auto-organisation épitaxiale : des surfaces aux matériaux magnétiques

O.Fruchart





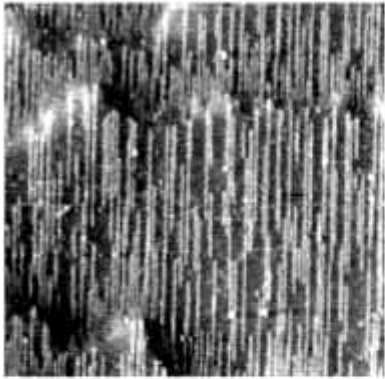
- **1. Introduction**
- **2. Co/Au(111) nano-columns**
- **3. Fe/bcc(110) stripes**
- **4. Conclusion and perspectives**

From surfaces to
functional materials at 300K

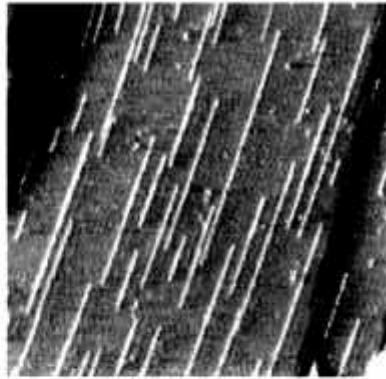


Self-assembly

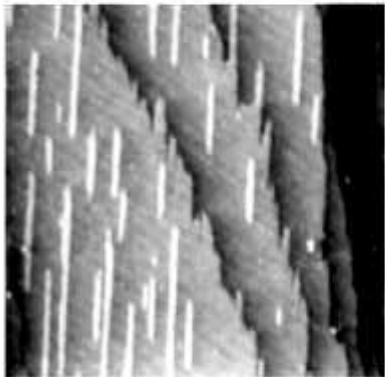
a) T = 265 K



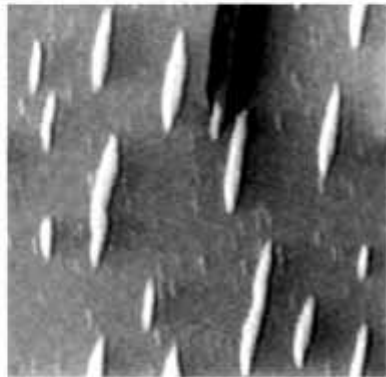
b) T = 300 K



c) T = 320 K



d) T = 350 K



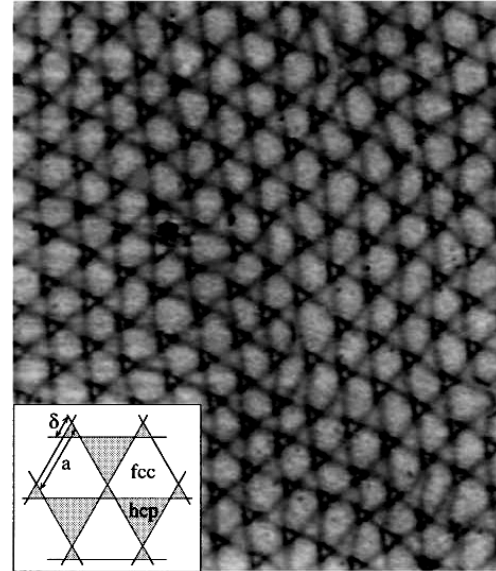
Cu/Pd(110)

500 Å

J.P.Bucher *et al.*, *Europhys.Lett.* **27**, 473(93)

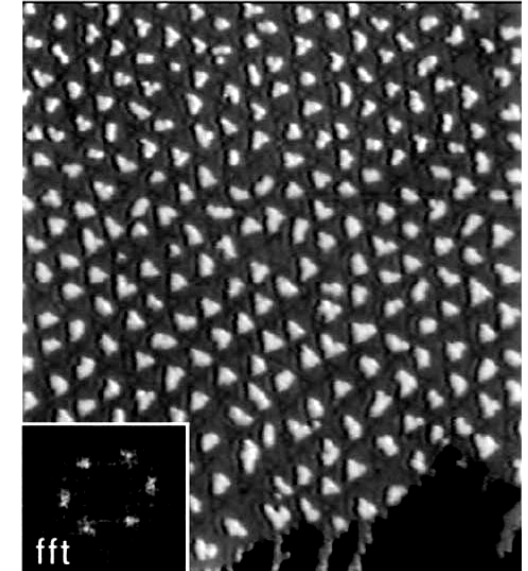
Self-organization

a)



200 Å

b)



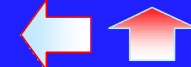
250 Å

Ag(110K)/Ag(2ML, 400K annealed at 800K)/Pt(111)

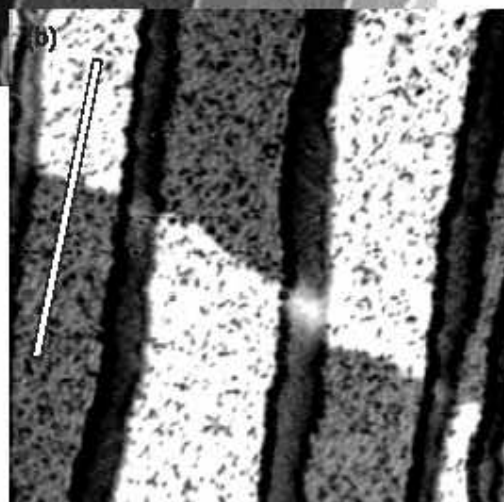
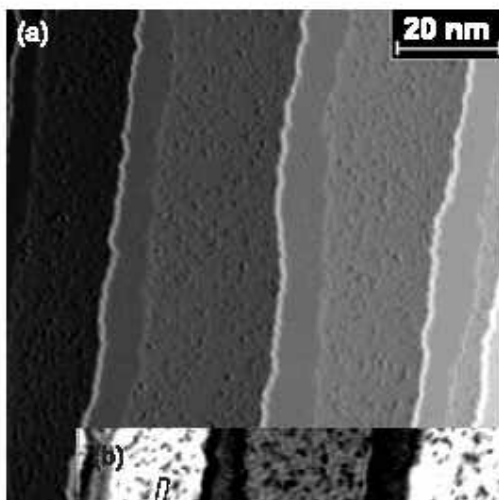
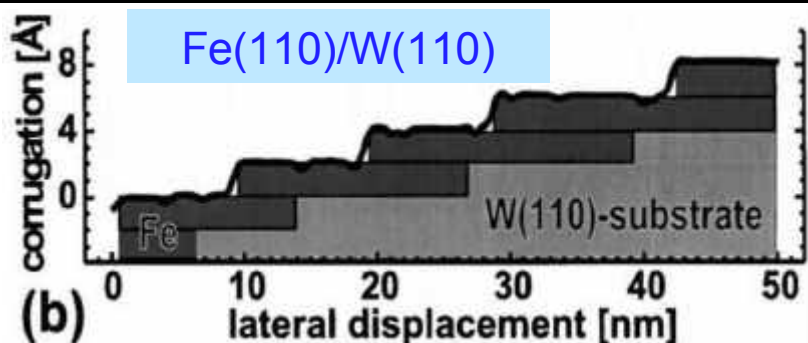
H.Brune *et al.*, *Nature* **394**, 451 (1998)



Any use for magnetism ?



Ordering in 1D; micromagnetism



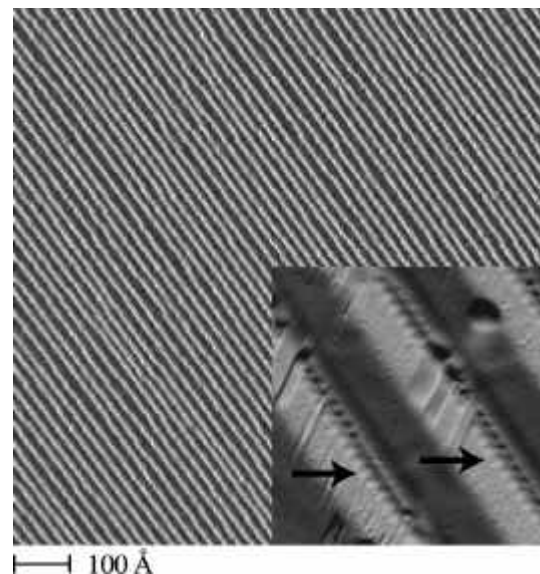
Spin-Polarized Scanning Tunneling Spectroscopy

Thickness = 1.25ML

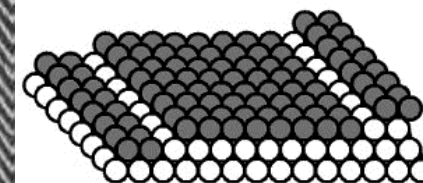
M. Bode et al, Rep. Prog. Phys. 66 523 (2003)

Ordering in 1D; anisotropy

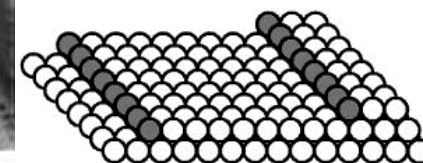
Co(250K - 0.12ML)/Pt(997)



Terraces ~ 1 ML



Chains ~ 0.12 ML



A. Dallmeyer et al., Phys.Rev.B 61(8), R5153 (2000).

Conclusion

- * Fundamental physics
- * Low temperature $T_c < 300K$
- * Difficult and expensive experiments

Open question

Any use for common magnetic materials?





From surfaces to magnetic materials

Model systems

Study the intrinsic microscopic details of phenomena that are too complex in common systems: hysteresis, ...

Demonstrate functionality at 300K

From fundamental physics at low temperature to ferromagnetic systems at 300K



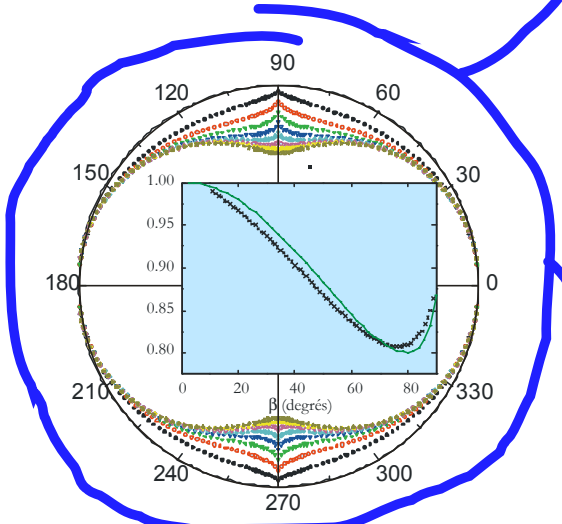
Model systems

CEA (Y.Samson)
LLN
(J.C.Toussaint,
W.Wernsdorfer)



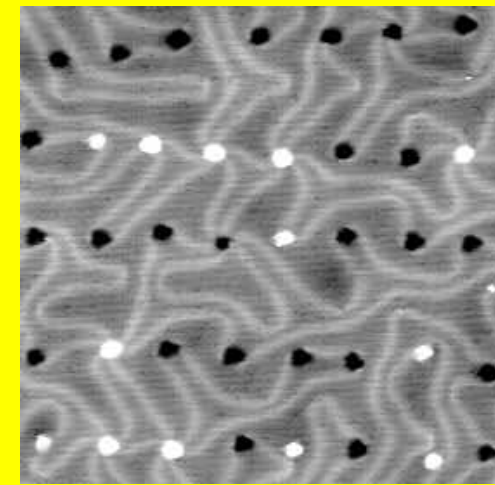
Self-assembled Fe/Mo(110) dots

LLN
(J.C.Toussaint)



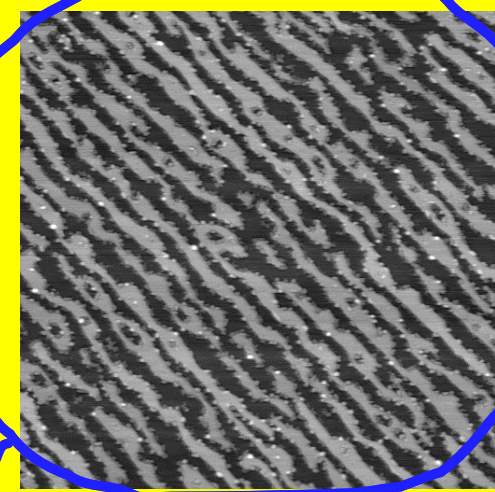
Micromagnetic modelling

Demonstrate functionality at 300K



~100nm
MPI-Halle
Cristallo (J.M.Tonnerre...)
IPCMS (F. Scheurer...)
CEA
(G.Renaud, A.Barbier...)
GPS-Jussieu
(S.Rousset...)

Self-organized Co/Au(111) pillars



~10µm
IPCMS (F.Scheurer)
ELETTRA (A.Locatelli...)
LLN (J.Vogel)

Self-organized Fe/bcc(110) stripes

PhD students

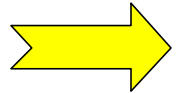
P.-O. Jubert (1998-2001)

M. Eleoui (2001-2004)





1. Introduction



2. Co/Au(111)

● 2.1 Literature

● 2.2 Self-organization of vertical pillars

● 2.3 Room-temperature coercivity

● 2.4 'Super-diffraction' on arrays of dots

3. Fe/bcc(110) stripes

4. Conclusion and perspectives



Co/Au(111) : 0.4ML

in-situ STM

350 x 350 nm



O. Fruchart *et al.*, Phys. Rev. Lett. 23, 2769 (1999)

FACTS

Co, Ni, Fe, Cu, Rh growth on Au(111)
> self-organized arrays of dots

ORIGIN ?

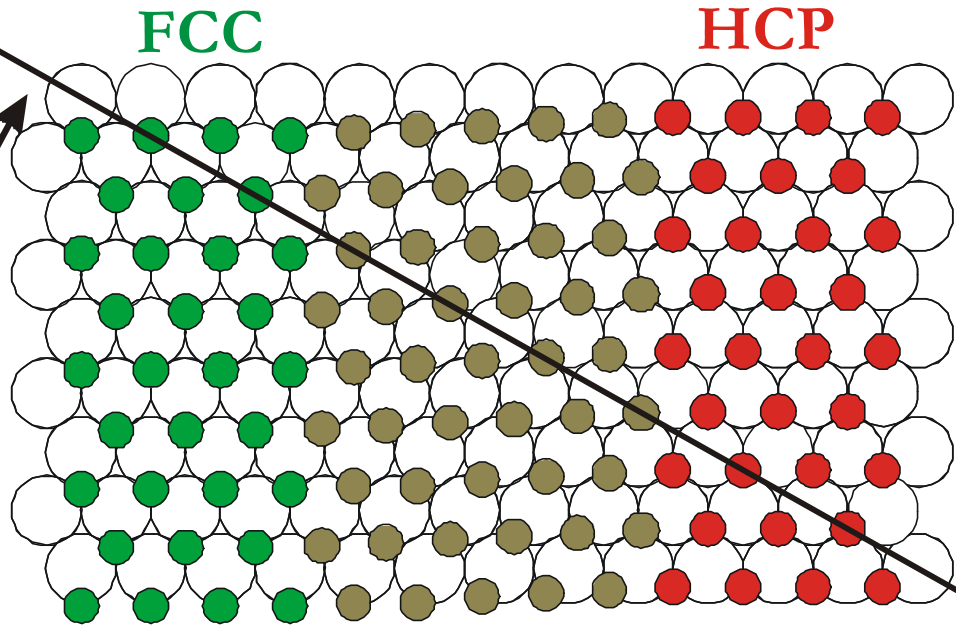
First observations:

D.D. Chambliss *et al.*, PRL 66, 1721 (1991)

B.Voigtlander *et al.*, PRB 44, 10354 (1991)

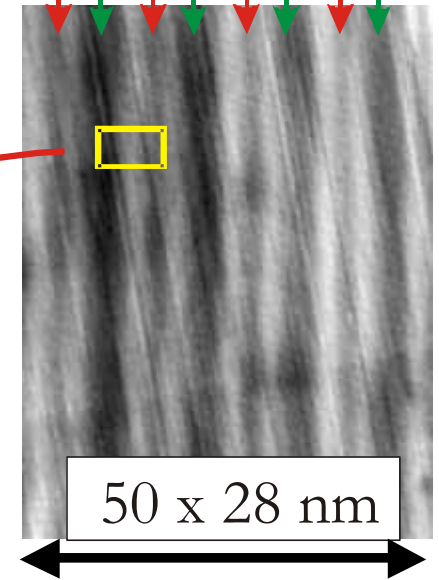


Top view of Au(111)



FCC HCP

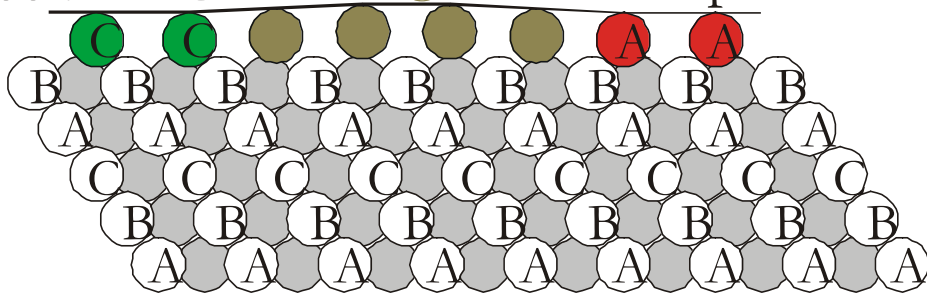
ZOOM



50 x 28 nm

Cross-sectional view

fcc : ABC *Corrugation* hcp : ABA



Atoms of the **second layer**

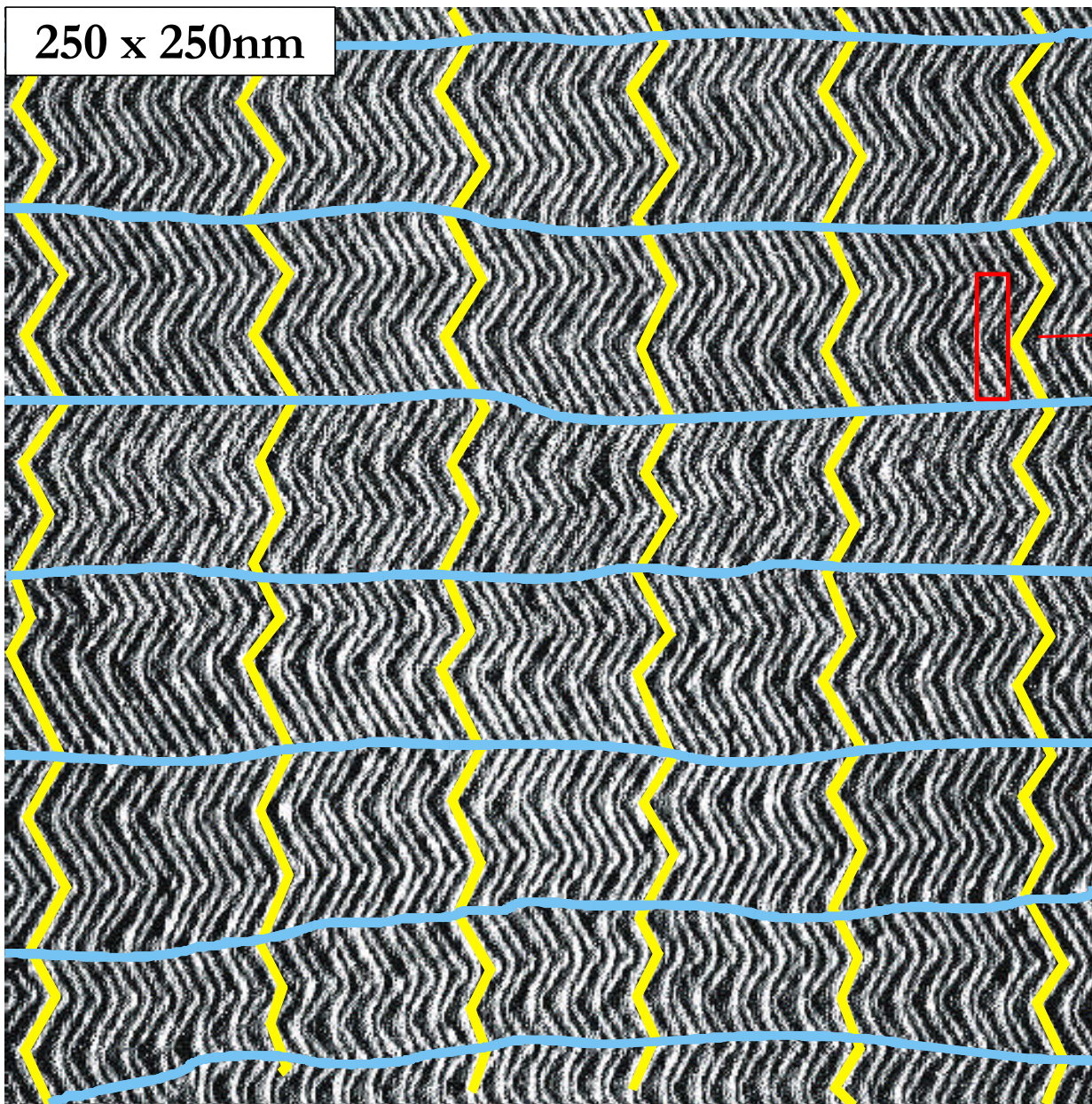


Atoms of the **topmost layer**

- Site A
- Site C
- Bridge position

(smaller diameter for clarity only)





Corrugation $\sim 0.2 \text{ \AA}$

Unit cell size :
 $\sim 7.5 \times 25 \text{ nm}$

J. V. Barth et al., Phys. Rev. B 42, 9307 (1990)

A.R. Sandy et al., Phys. Rev. B 43, 4667 (1991)

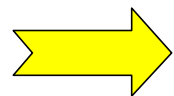
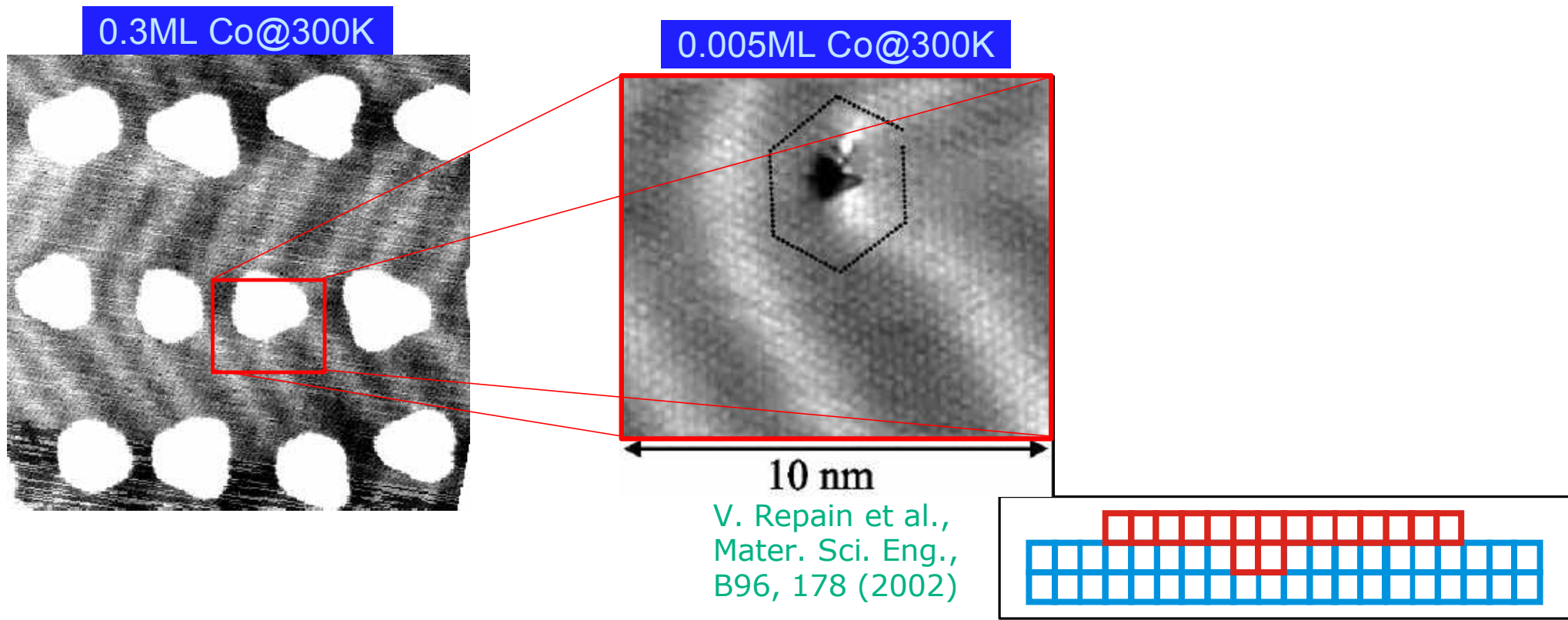
Olivier Fruchart - HDR - 25/09/2003 - p.10



Isotropic surface relaxation



Fe, Co, Ni (etc.) nucleation: atomic place exchange mechanism with Au atoms



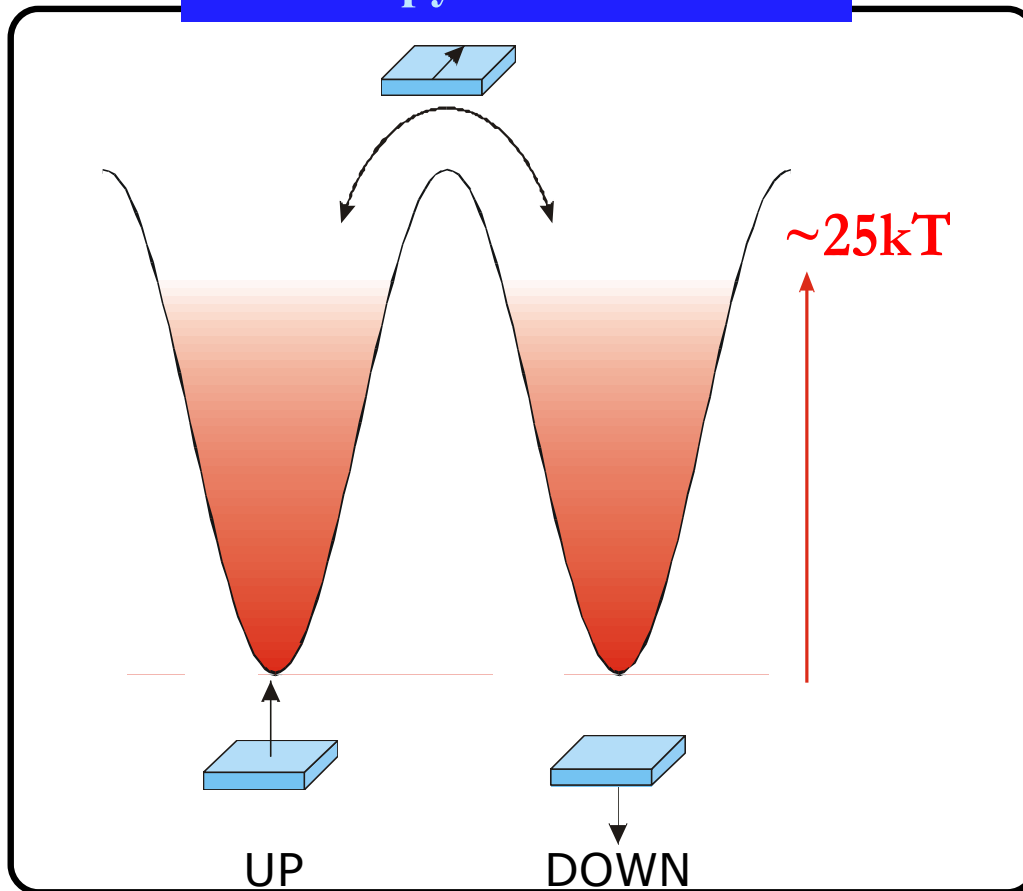
Leading parameter :
deposit has a higher surface energy than Au.
(and Au atoms stress near chevrons)

FIRST REPORT (Ni/Au(111]):
J.A.Meyer et al., Surf.Sci.365, L647 (1996)





Anisotropy barrier $\Delta E \sim KV$



Arrhenius law

Phenomenological thermal activation

$$t = t_0 \exp\left(\frac{\Delta E}{k_B T}\right)$$

Precession

$$\Rightarrow \Delta E = k_B T \ln(t / t_0) \sim 25 k_B T$$

$$t_0 \sim 10^{-9} \text{ s}$$

$$t \sim 1 \text{ s}$$

Experiments

Blocking temperature

$$T_b = KV / 25 k_B$$

For $T > T_b$:

- zero remanence
- zero coercivity



$$T_b = KV / 25k_B$$

TWO ROUTES TO OVERCOME SUPERPARAMAGNETISM

- **Increase K**

Problem: K does not increase as fast as V decreases

P. Gambardella et al., Nature 416, 301 (2002)

P. Gambardella et al., Science 300, 1130 (2003)

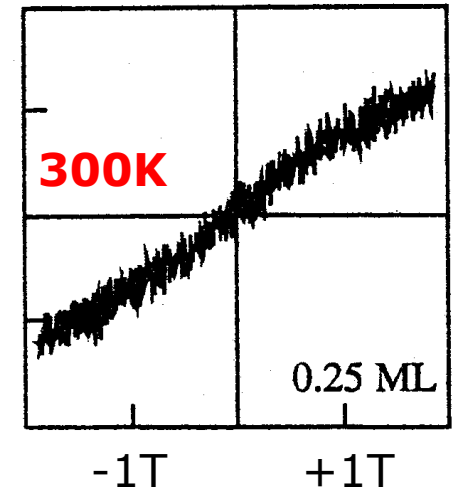
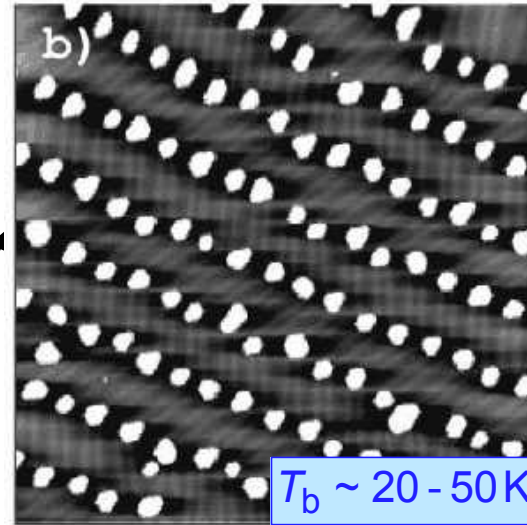
- **Increase V**

Problem : lateral coalescence occurs

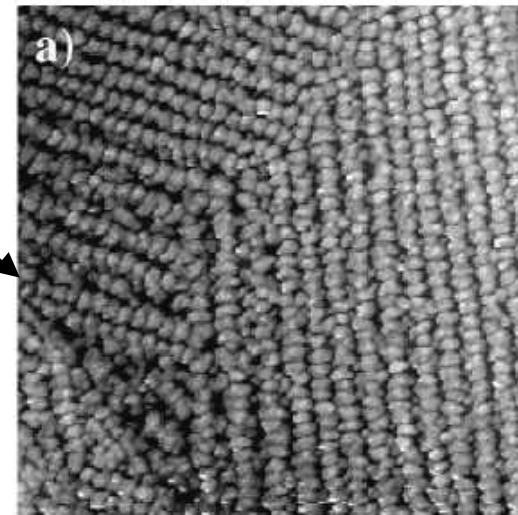
S. Padovani et al., PRB59, 11887 (1999)

Co/Au(111)

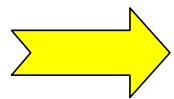
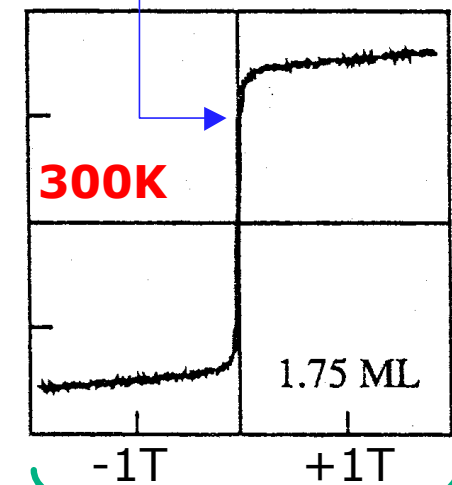
0.25ML



1.75ML



Onset of remanence



HOW INCREASE THE HEIGHT OF THE NANOSTRUCTURES ?

H.Takeshita et al., JMMM165, 38 (1997)

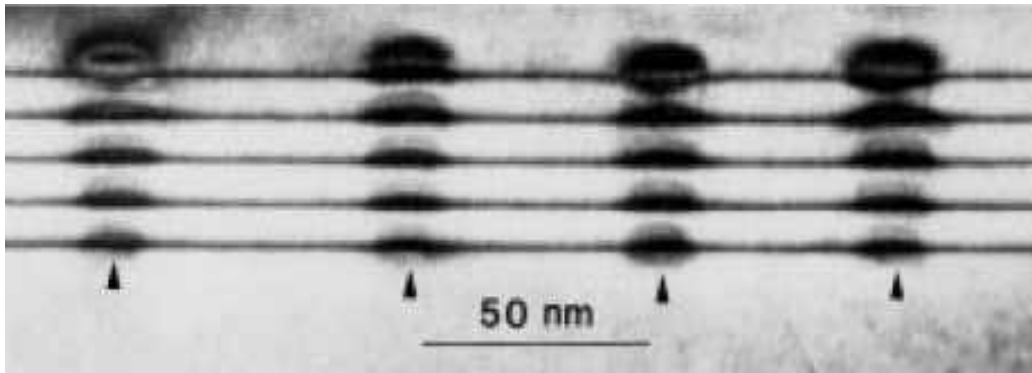
Olivier Fruchart - HDR - 25/09/2003 - p.13



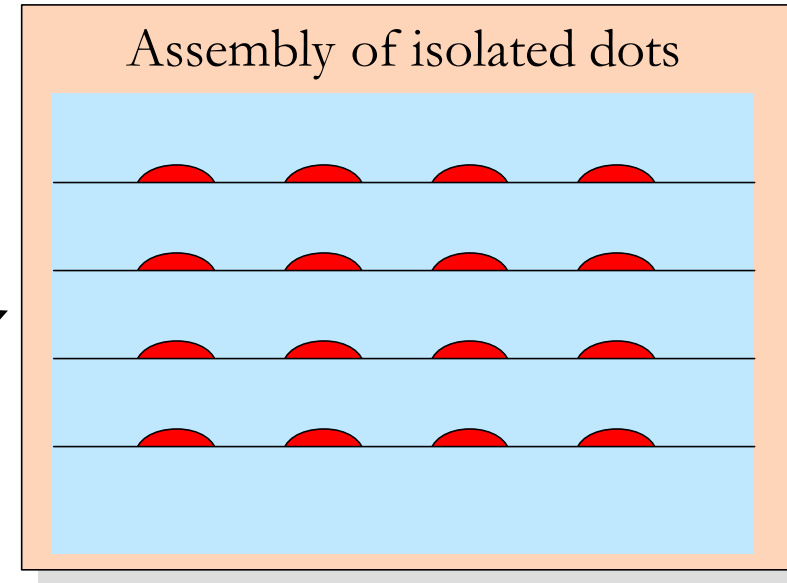


Vertical 3D self-organization

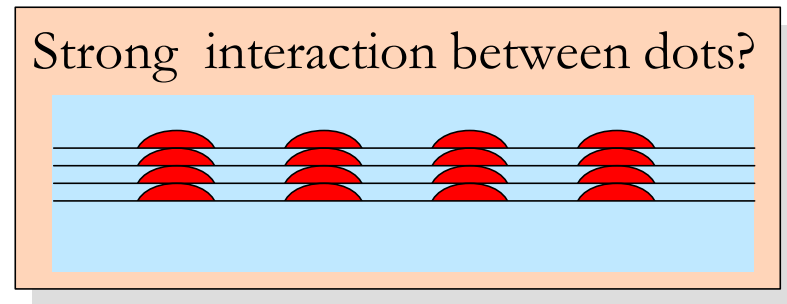
$In_xGa_{1-x}As/GaAs$



Q.Xie et al., Phys.Rev.Lett.75(13), 2542 (1995)



Thinning the spacer layer

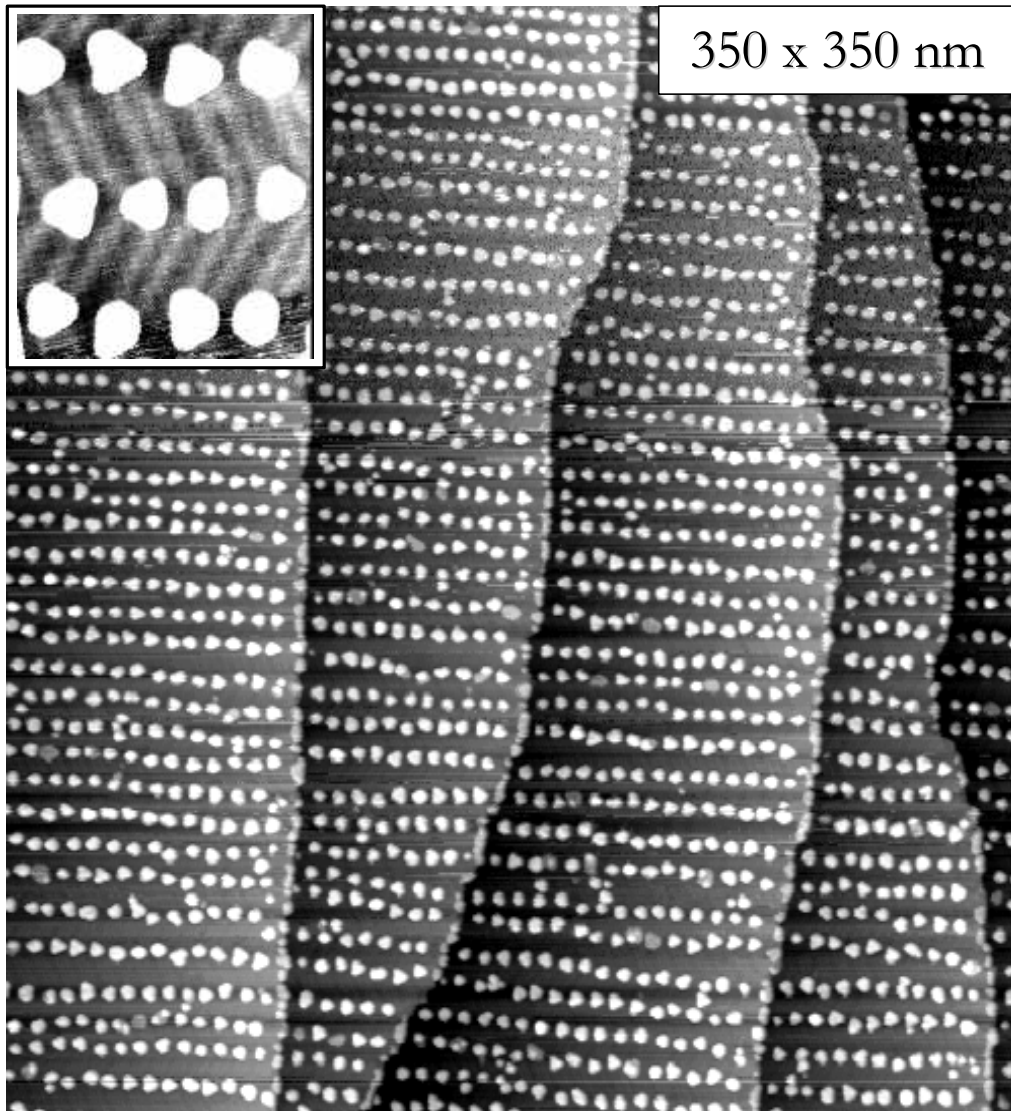


- ↪ Superparamagnetism overcome ?
- ↪ Enhanced magnetic signal

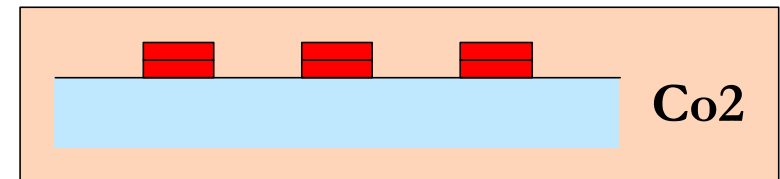
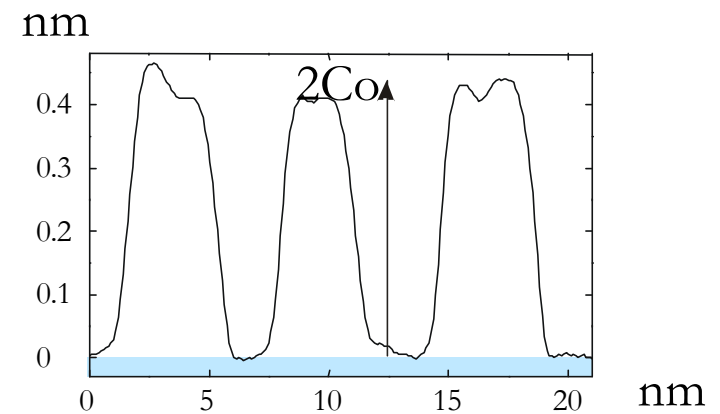




➤ Step 1 : 0.2ML Co @300K *in-situ* STM



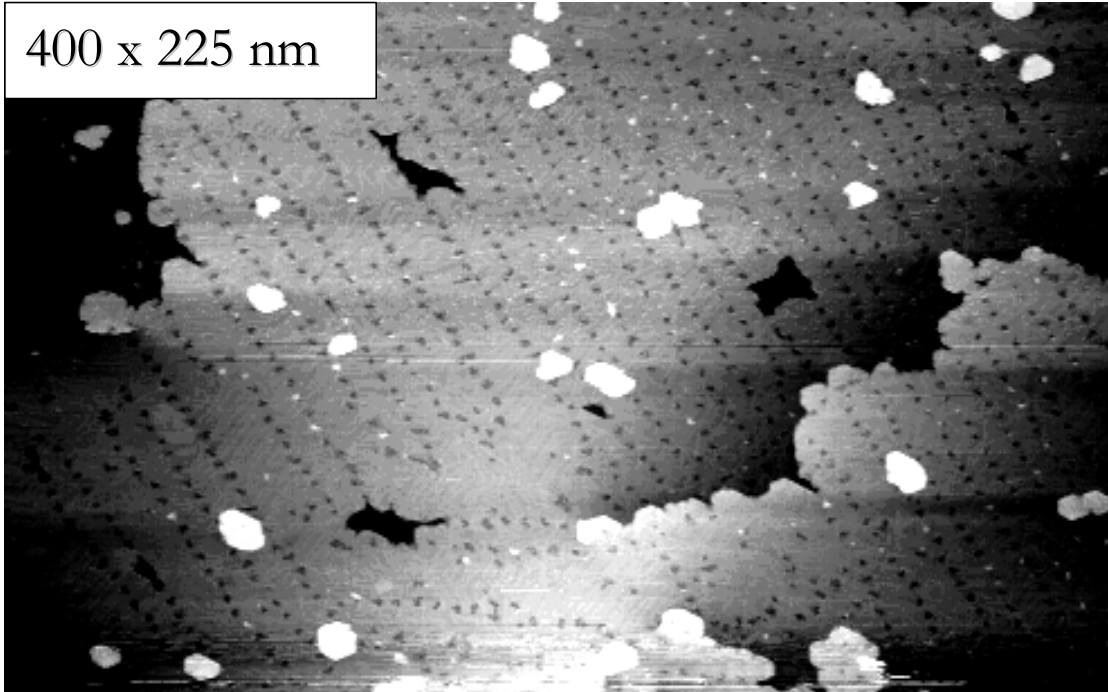
Typical cross-section :





➤ Step 2 : 3.8AL Au @375 → 410K

400 x 225 nm

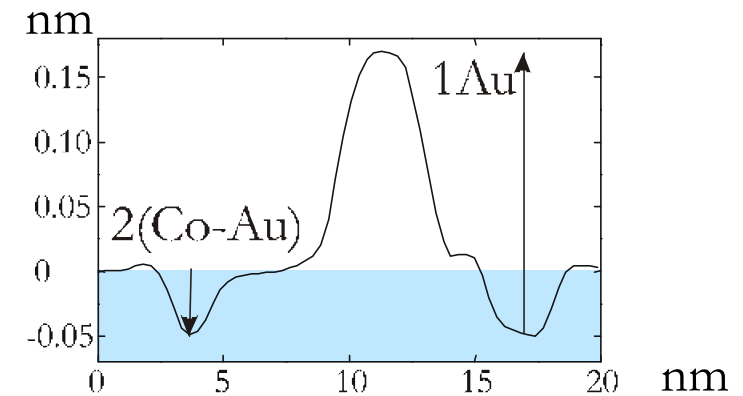


See also : [Wollschläger et al., Surf.Sci.277, 1 \(1992\)](#)

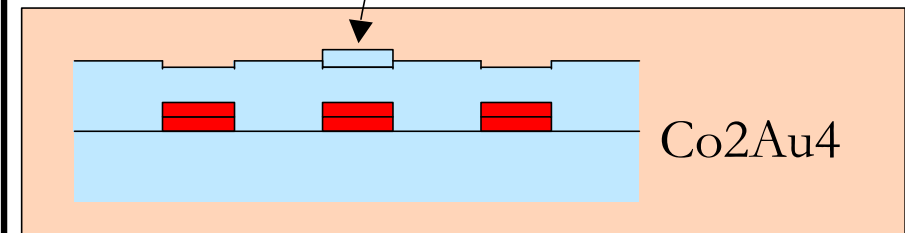
1AL Co hcp \approx 0.205nm

1AL Au fcc \approx 0.235nm

Typical cross-section :



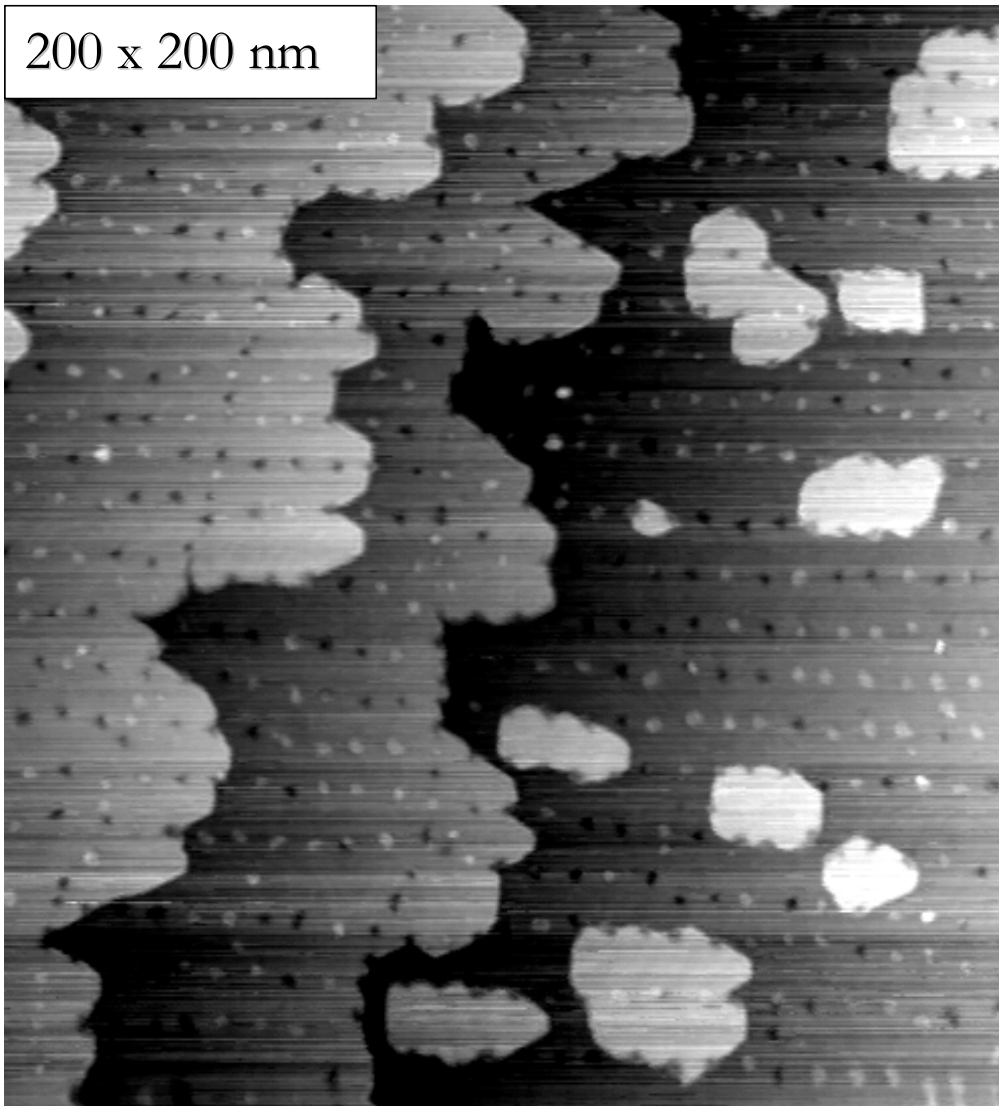
1AL Au
decoration



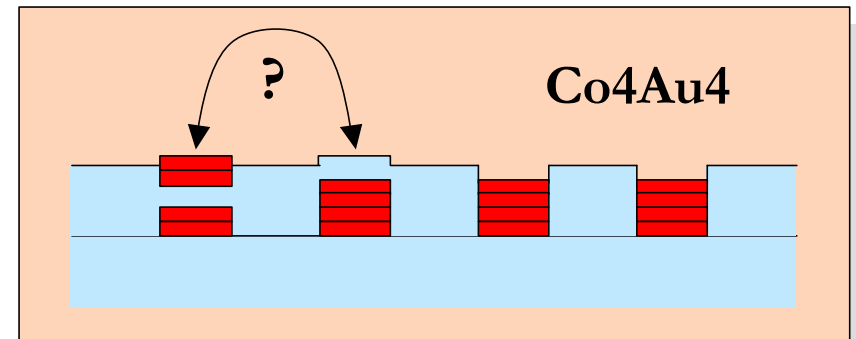
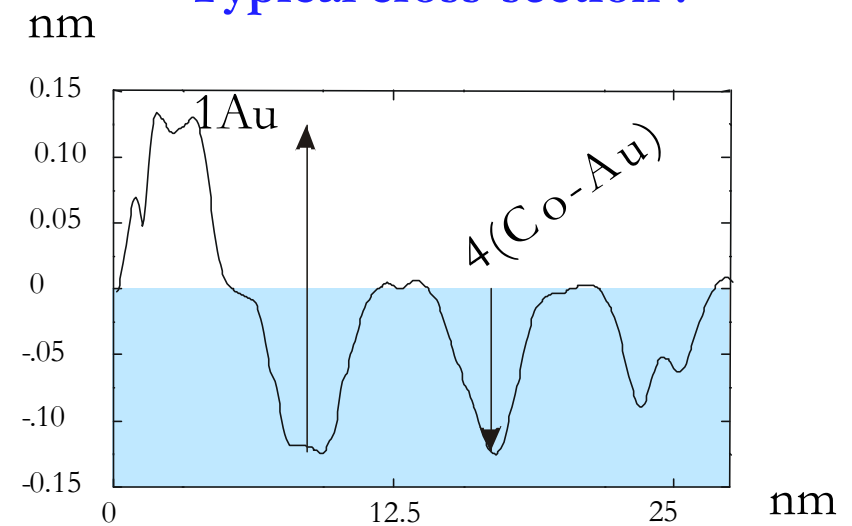
- Surface smoothing
- Unaffected Co dots



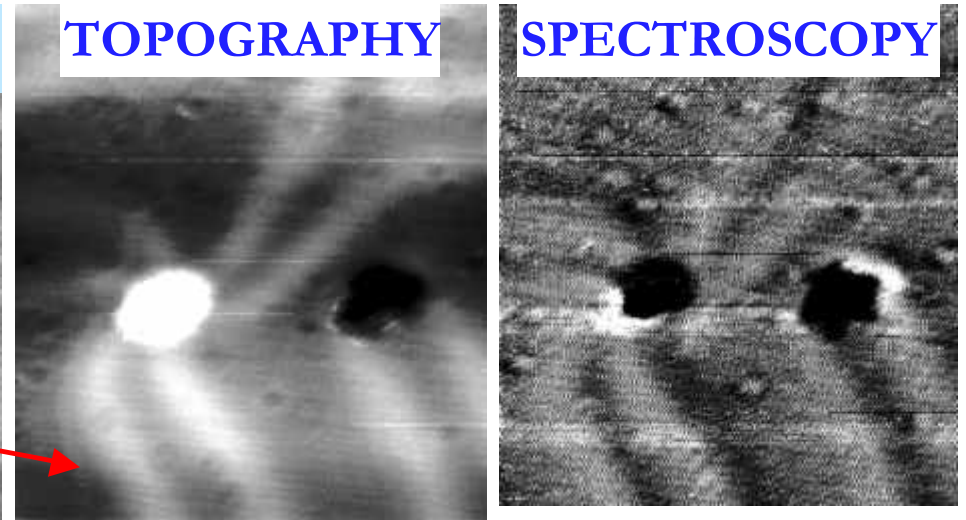
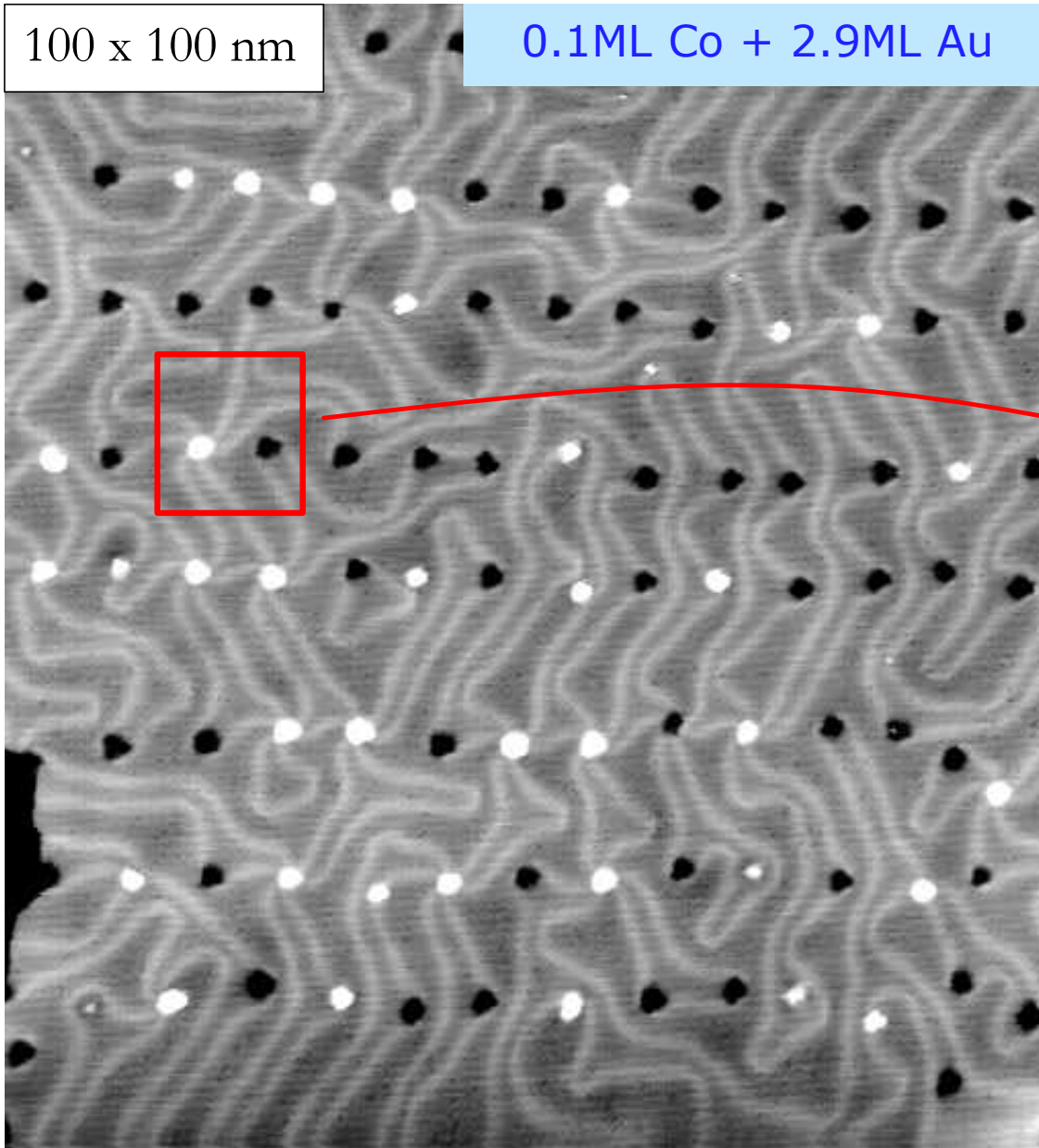
➤ Step 3 : 0.1AL Co @500K



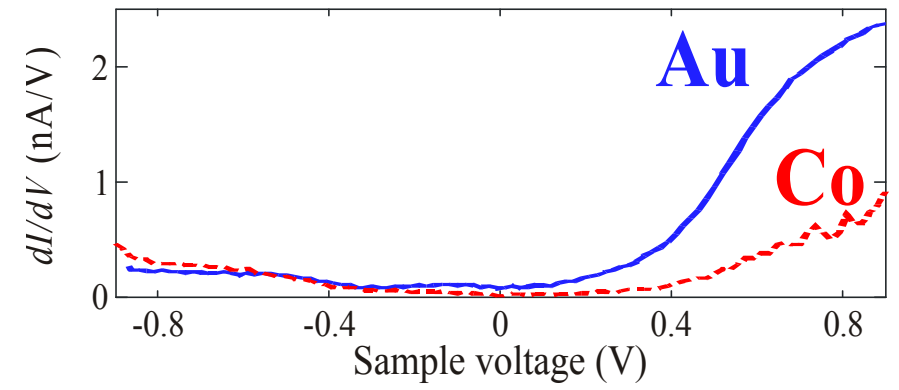
Typical cross-section :



- **Co/Au exchange mechanism**
- **The dots are now 4ML high**



SAME AREA



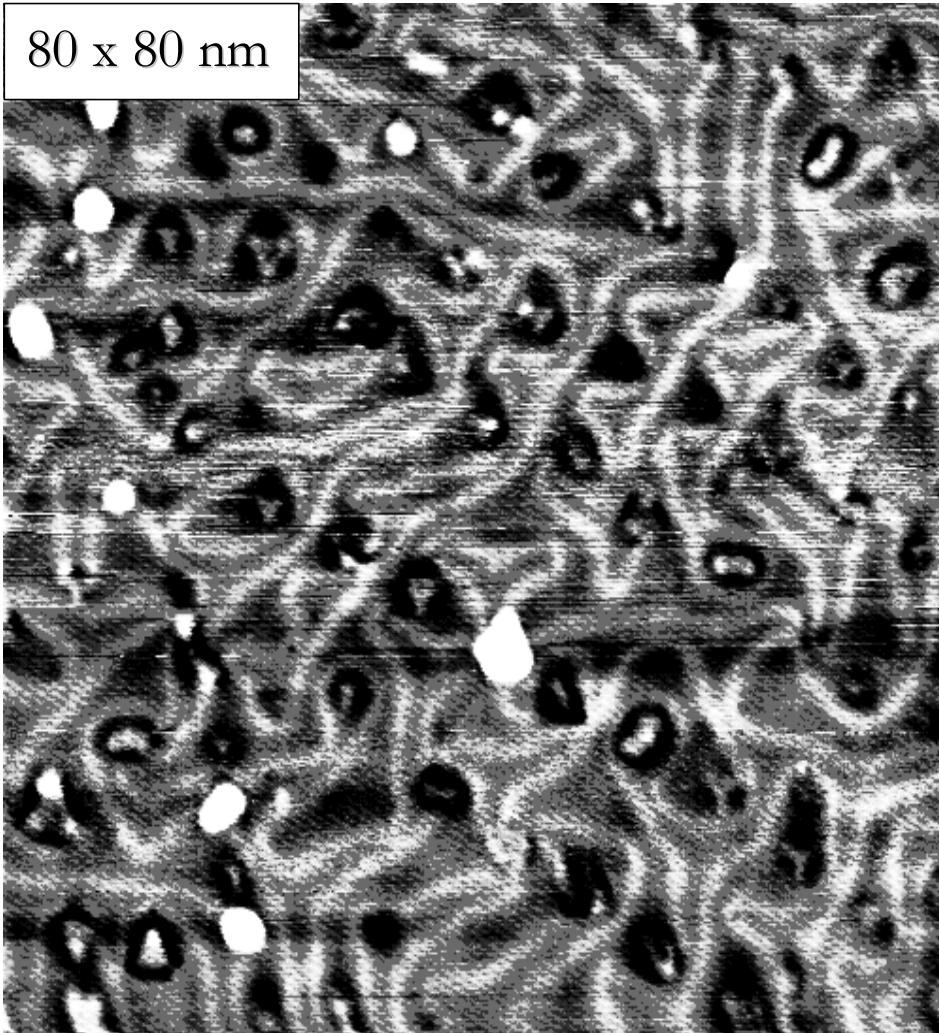
Co atoms grow only on existing dots
↪ **Vertical self-organization**



One step = (0.1ML Co) + (0.9ML Au) @500K

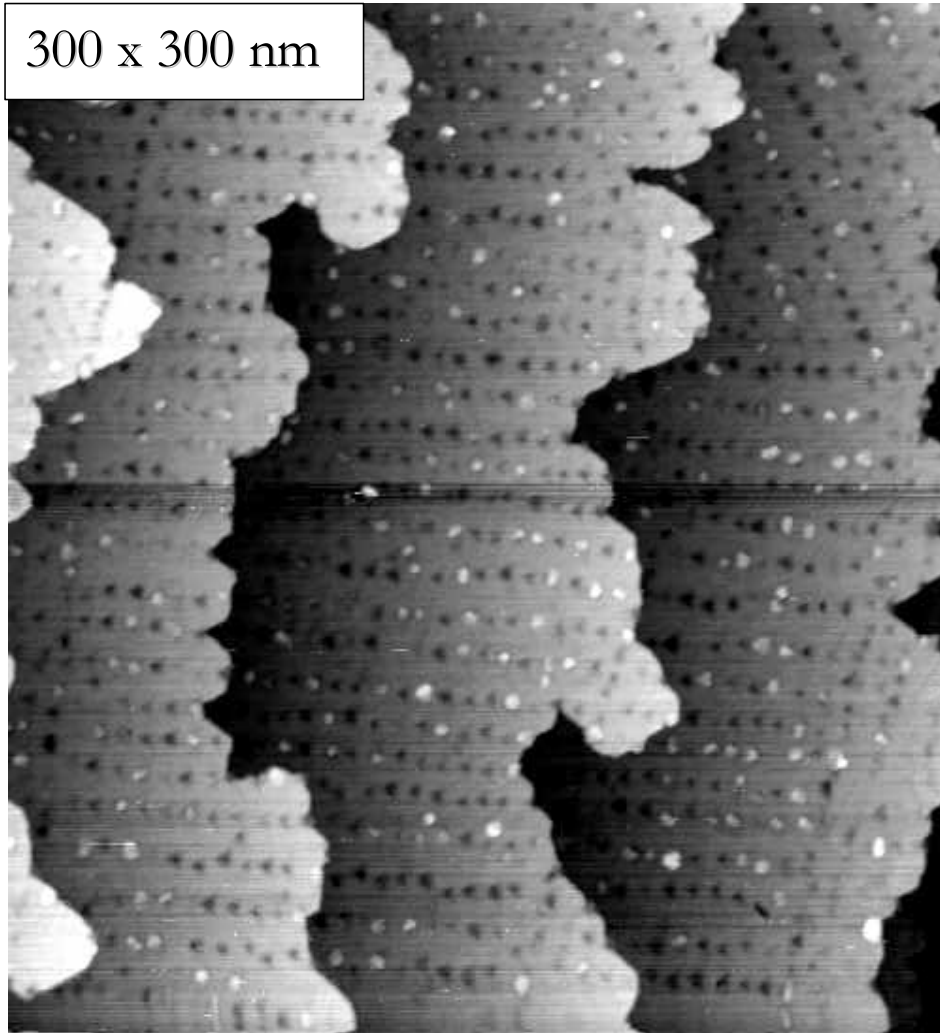
Co6Au6

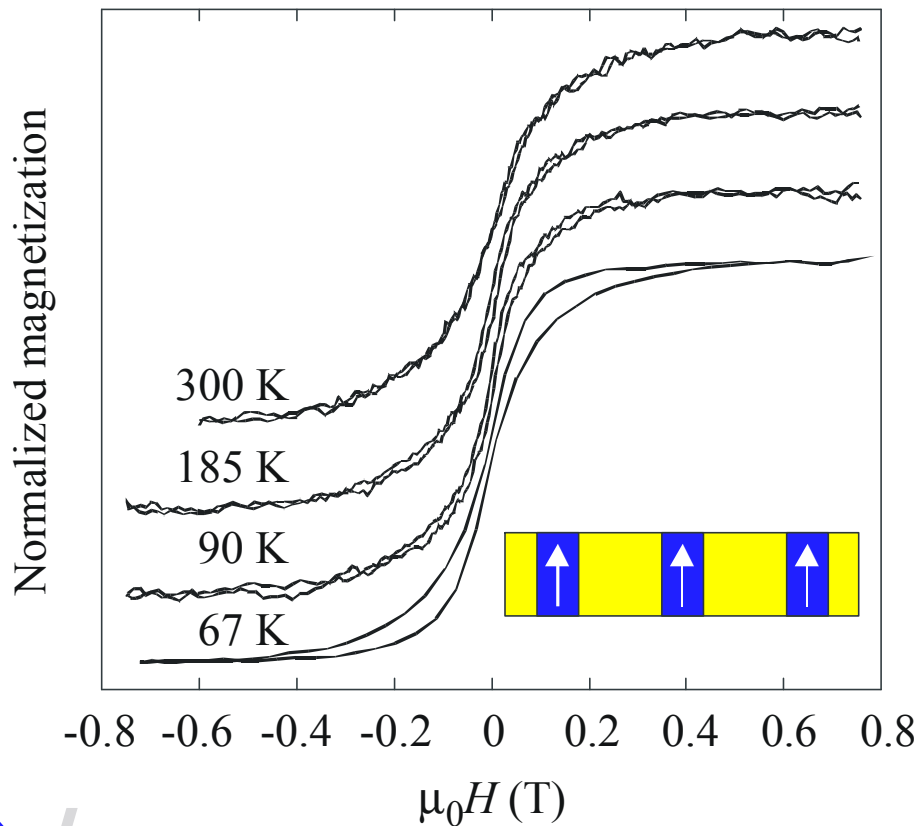
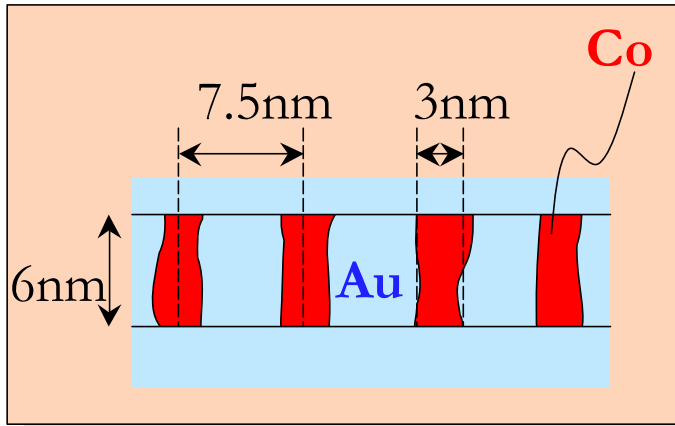
80 x 80 nm



Co10Au10

300 x 300 nm

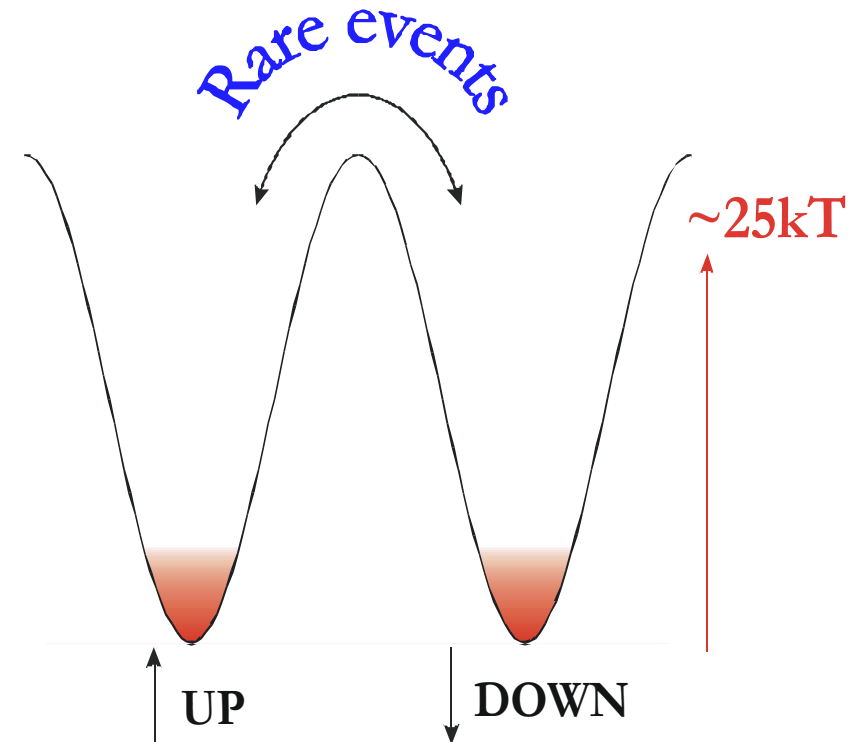




➤ **Magnetization essentially perpendicular**

↳ **2 states: up and down** (Ising macrospin)

↳ Superparamagnetism fitted using **Brillouin 1/2 function**



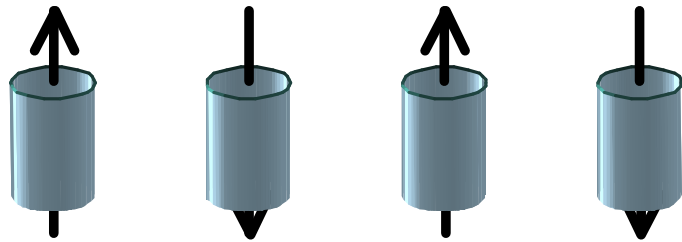


➤ Brillouin 1/2 function

$$m = B_{1/2}(\mu_0 \mu_{Co} N H_{eff.} / kT)$$

➤ Effective field

$$H_{eff.} = H + r M_s m$$



(Demagnetizing dipolar interactions)

➤ First order expansion:

susceptibility

$$\frac{d(\mu_0 H)}{dm} = \frac{1}{?} = -\mu_0 M_s r + \frac{k}{\mu_{Co} N} T$$

a + b . T

... from magnetism

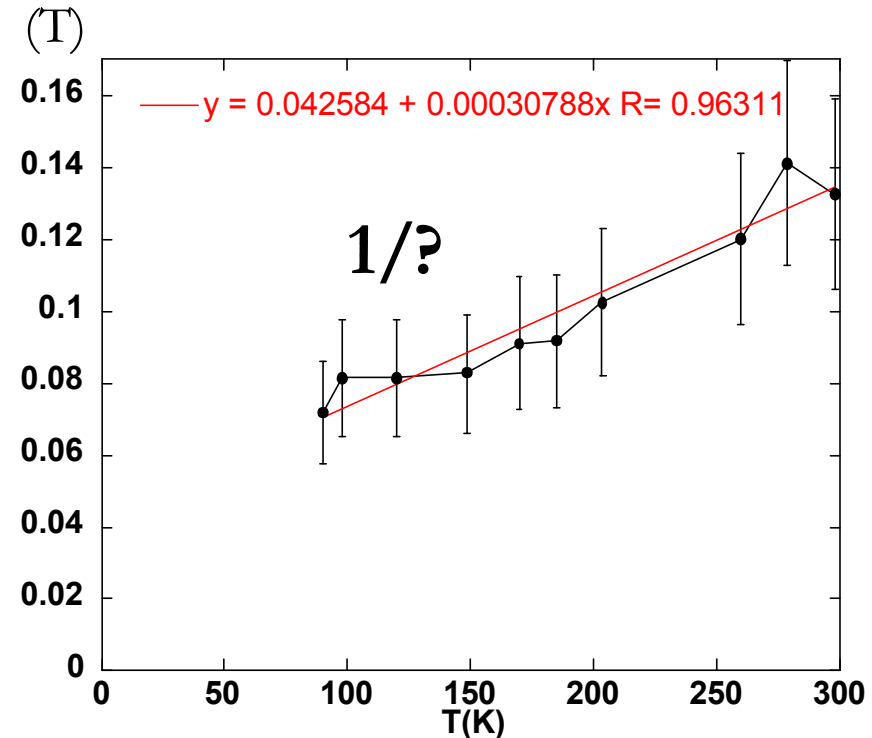
↪ N=2800 atoms

↪ Hdip= -42 mT

Deduced from STM

↪ N=3300 atoms

↪ Hdip= -32 mT

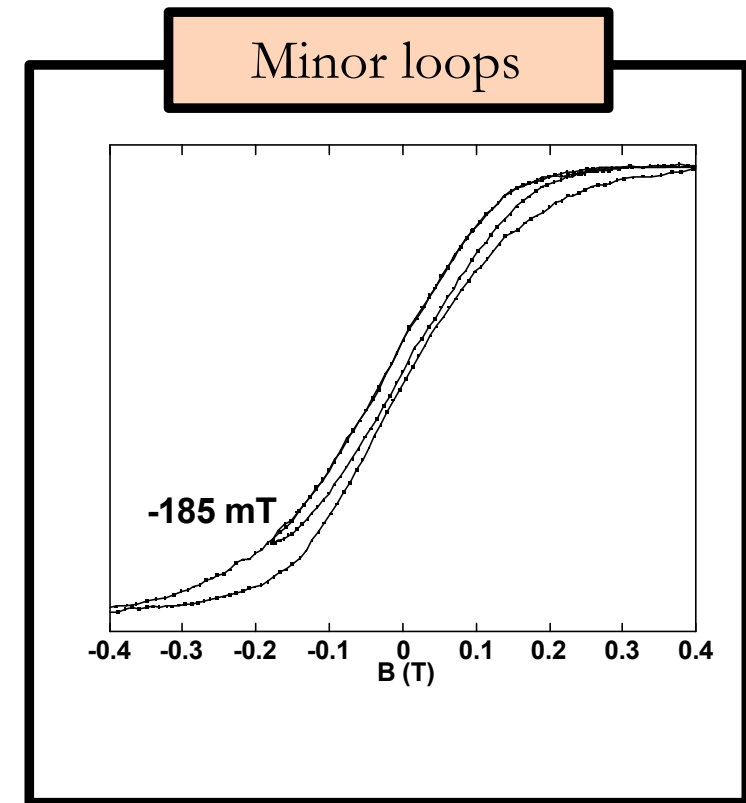
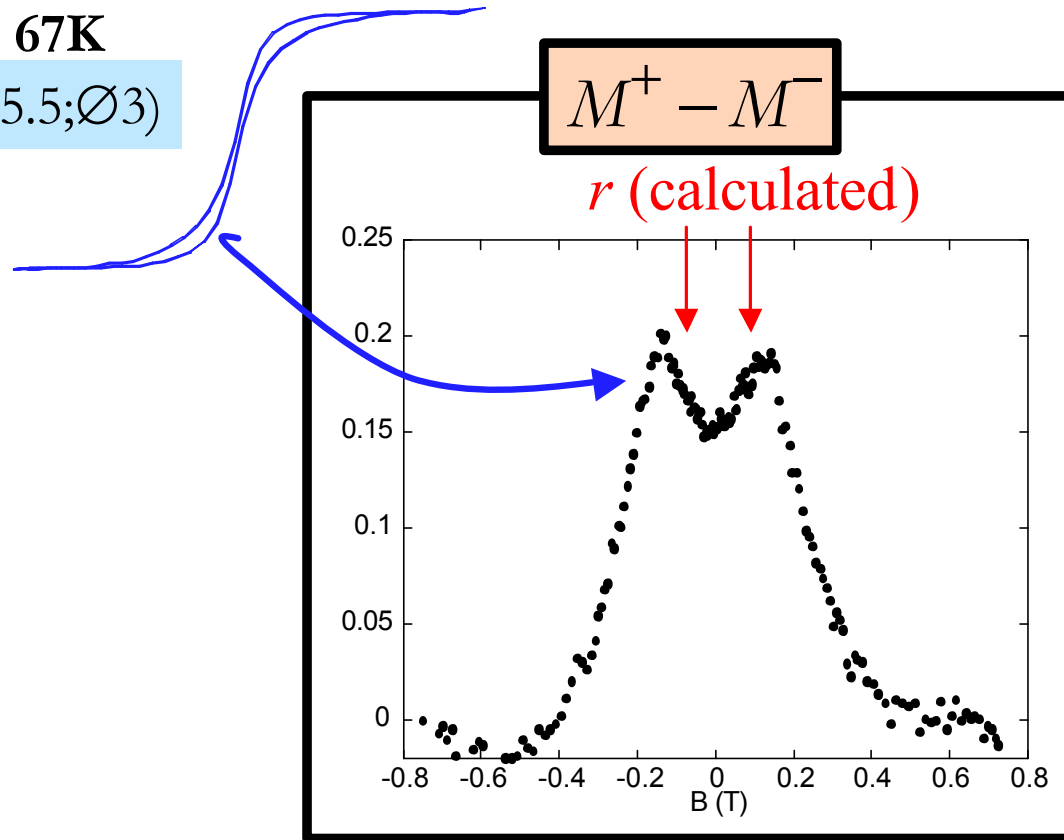


➤ Good quantitative agreement ↪ 1 pillar = 1 magnetic entity

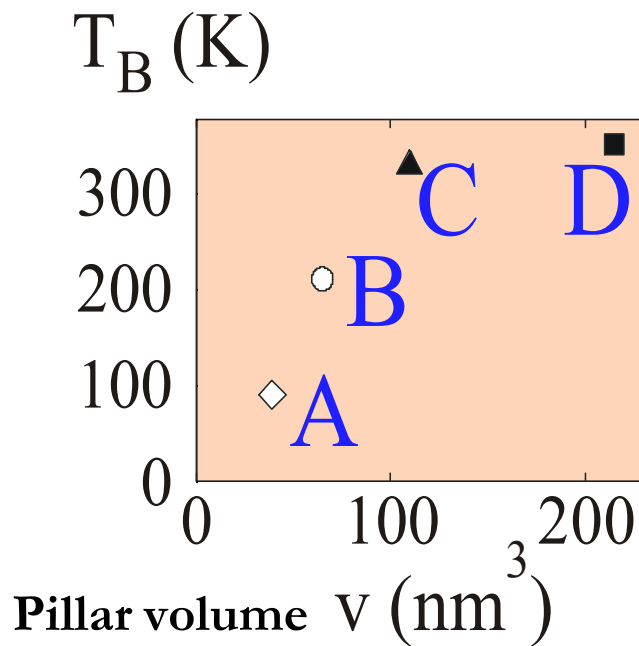


$T = 67\text{K}$

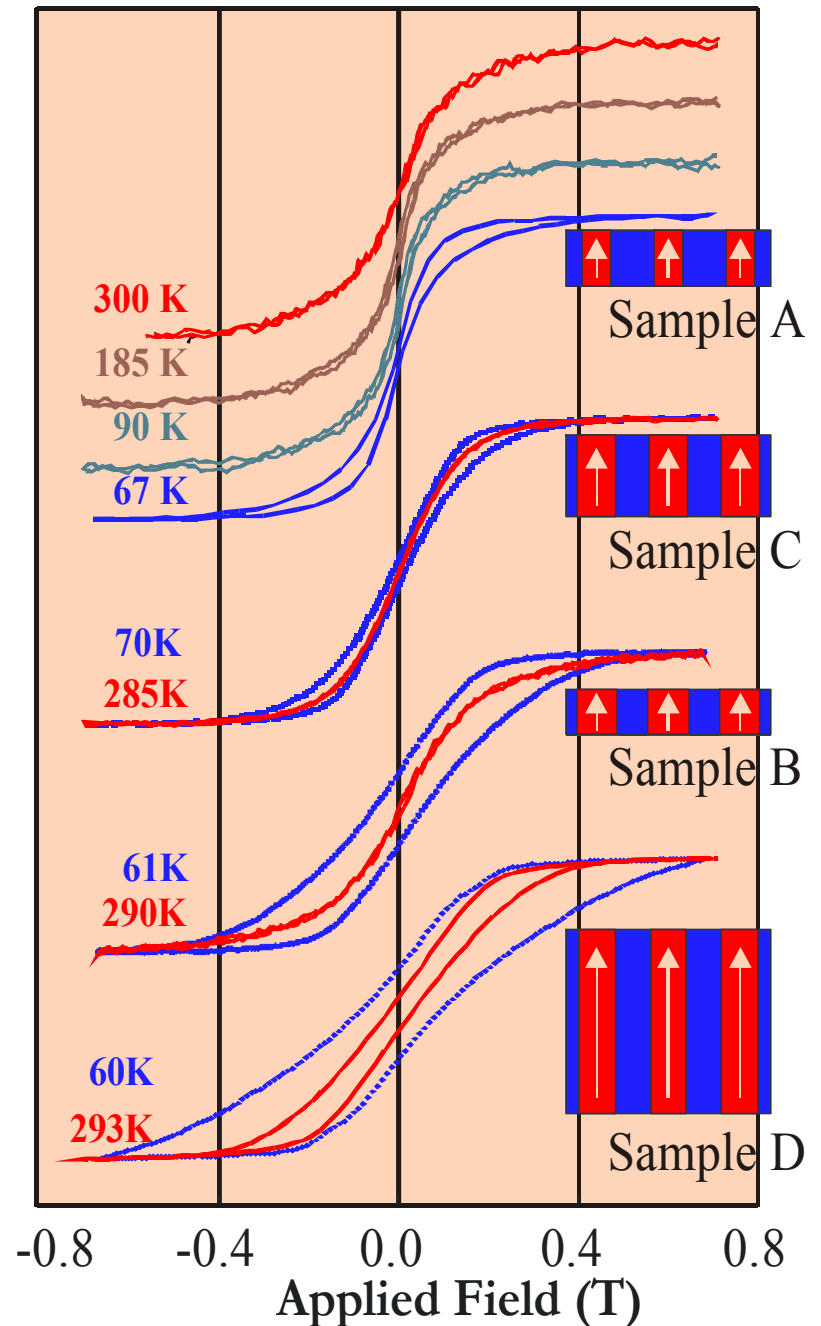
($\uparrow 5.5; \emptyset 3$)



- Signature of demagnetizing dipolar interactions: agrees with model



➤ **Blocking temperature > 300K**
 (~ 30K for flat Co/Au dots)

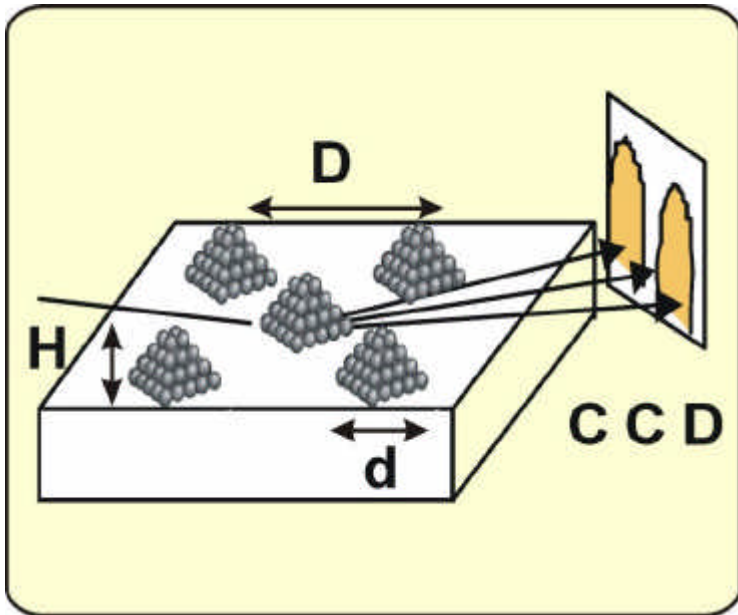


GISAXS: Grazing Incidence Small Angle X-ray Scattering

Size >> atoms

Surface sensitive

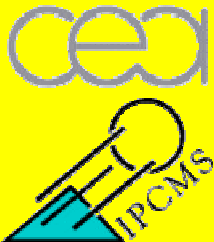
PRINCIPLE



TECHNICAL ASPECTS

- ID32 beamline at ESRF
- In-situ under UHV > real time

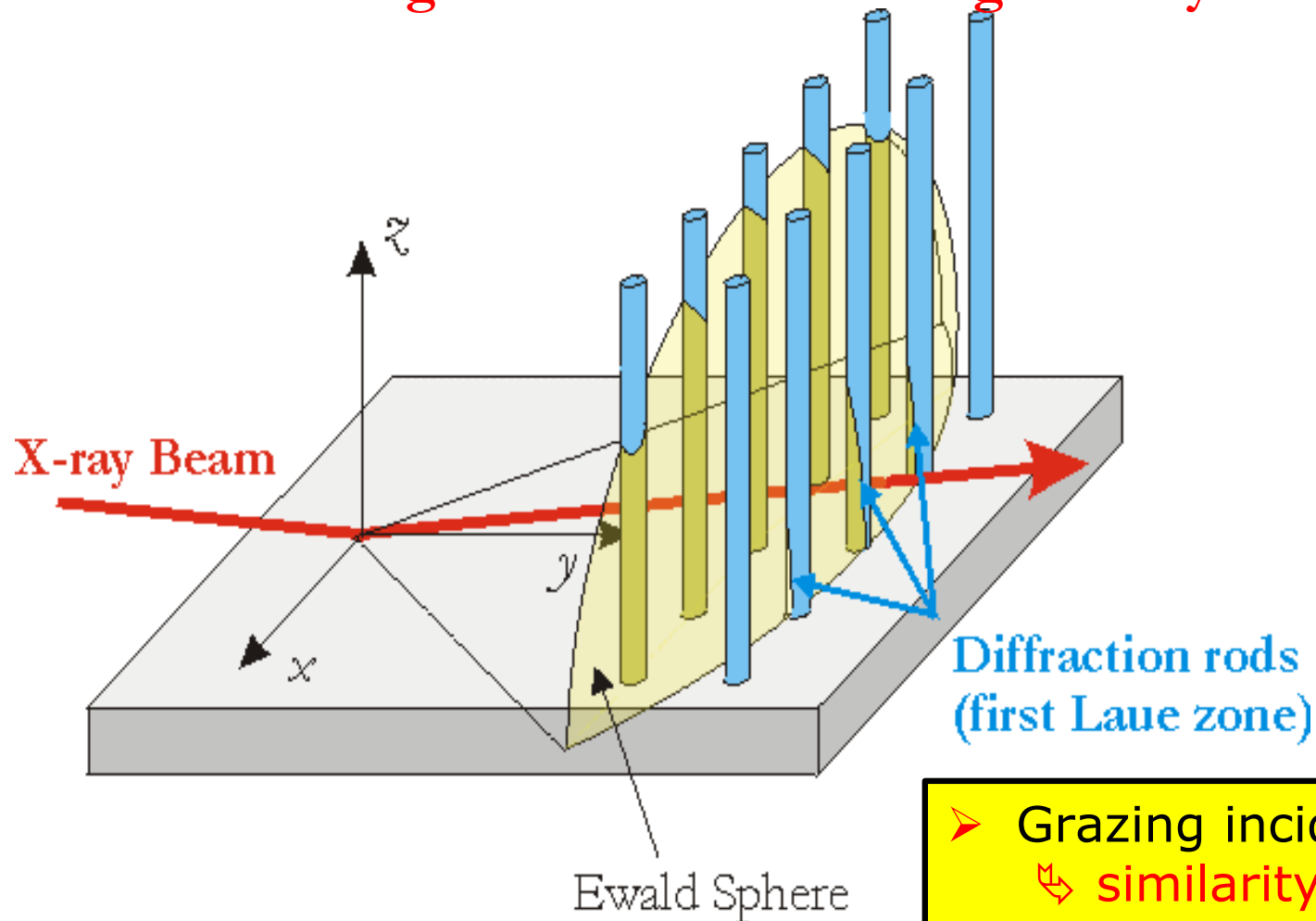
First step: dots and films, not pillars



G. Renaud, M. Noblet, O. Ulrich / A. Barbier
 DRFMC/SP2M/IRS (CEA-Grenoble) / CEA-Saclay
J.-P. Deville, F. Scheurer, J. Mané-Mané
 IPCMS (CNRS/ULP/ECPM), Strasbourg



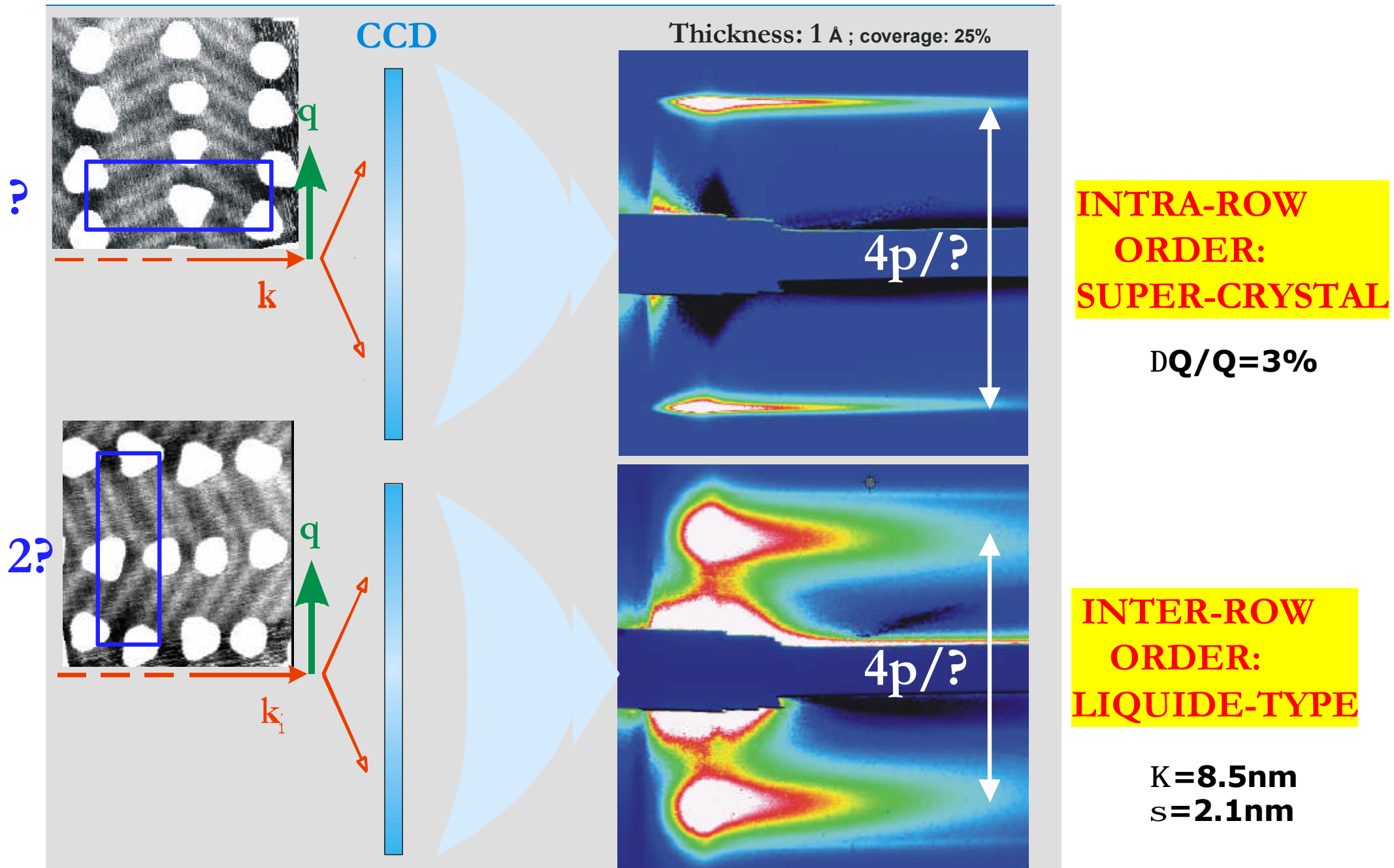
GISAXS: Grazing Incidence Small Angle X-ray Scattering



- Grazing incidence and 2D detector
↳ similarity with RHEED
- Ordered sample ↳ 'super' reciprocal space ?

RHEED: Reflection High Energy Electron Diffraction



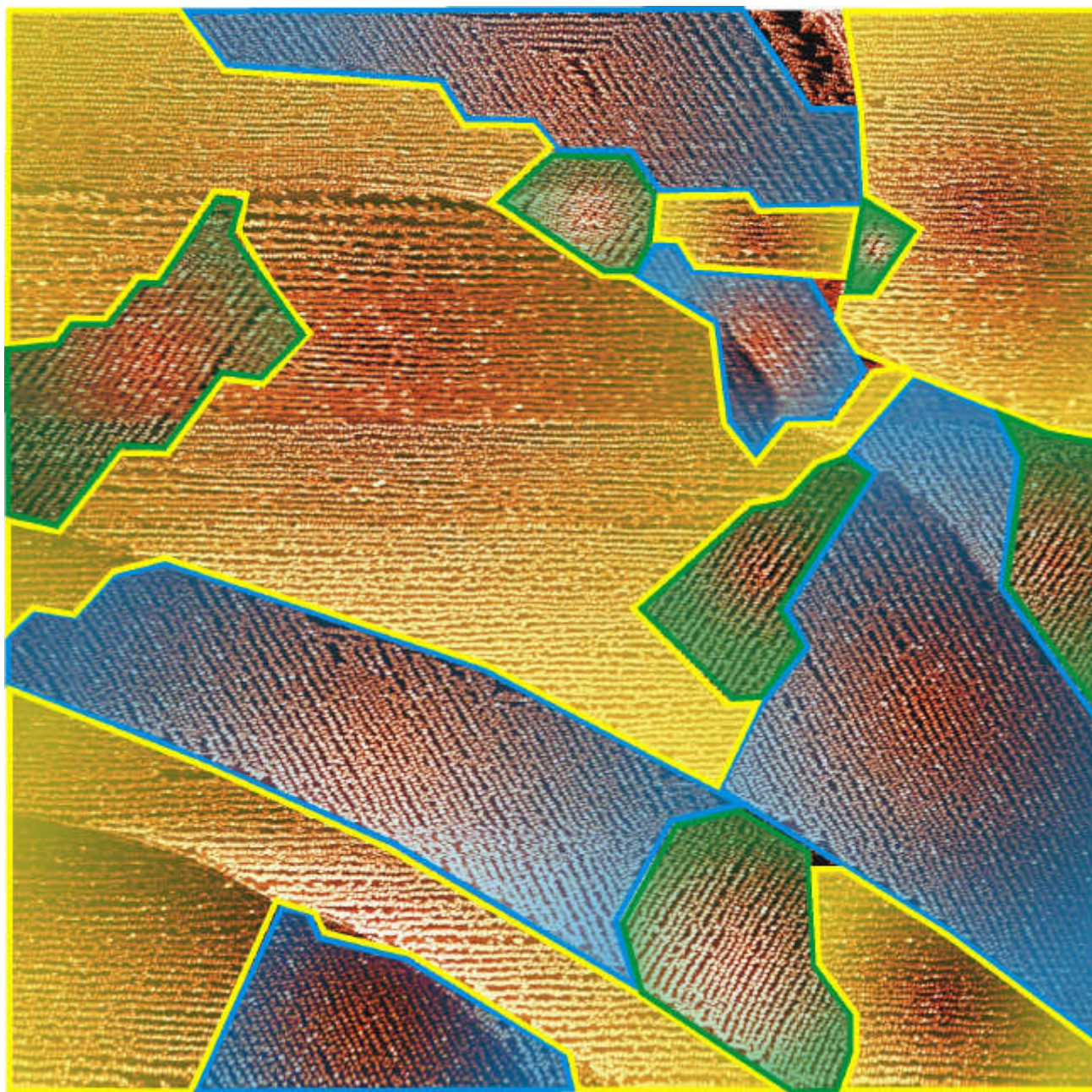


1.5mm x 1.5mm STM image

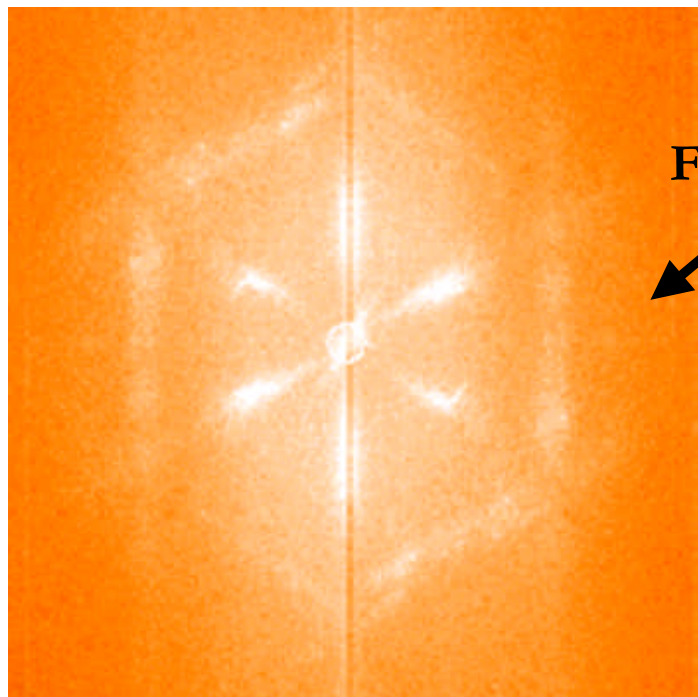


3-fold symmetry of Au(111) crystal

➤ 3 'equivalent domains'

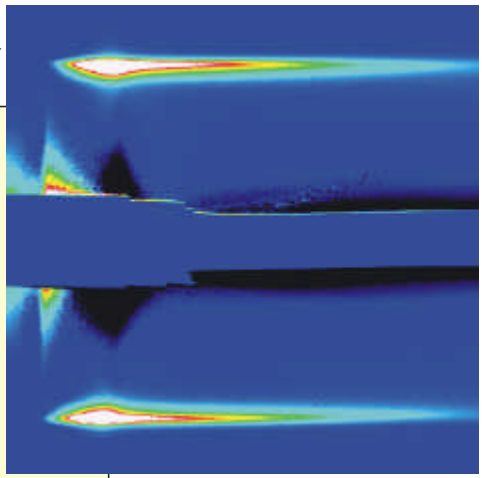
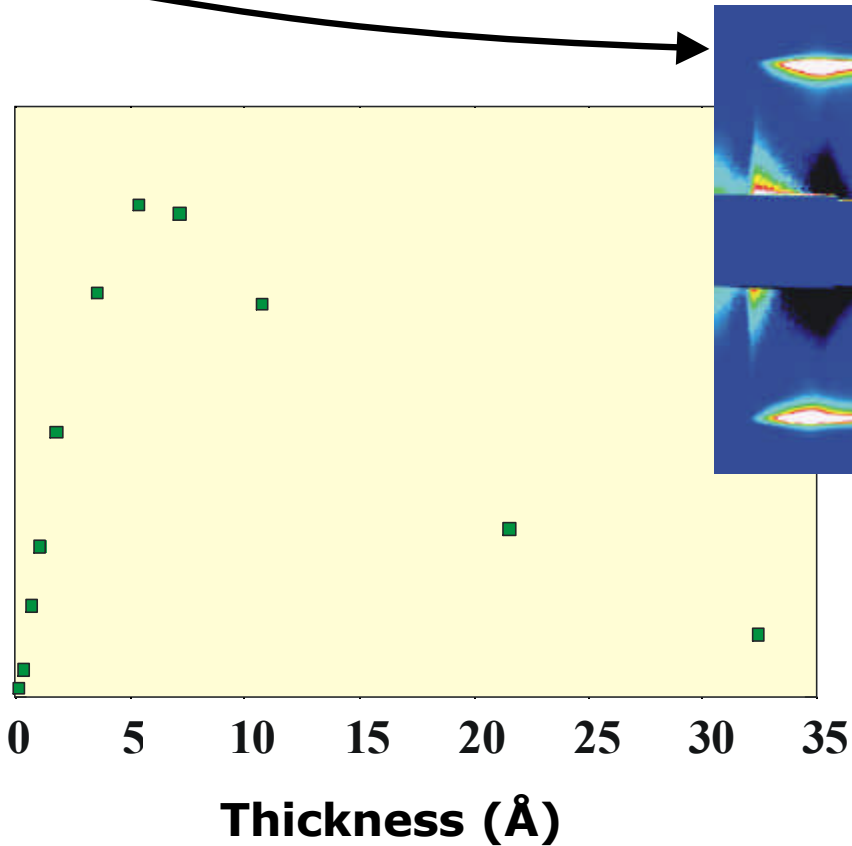


FFT

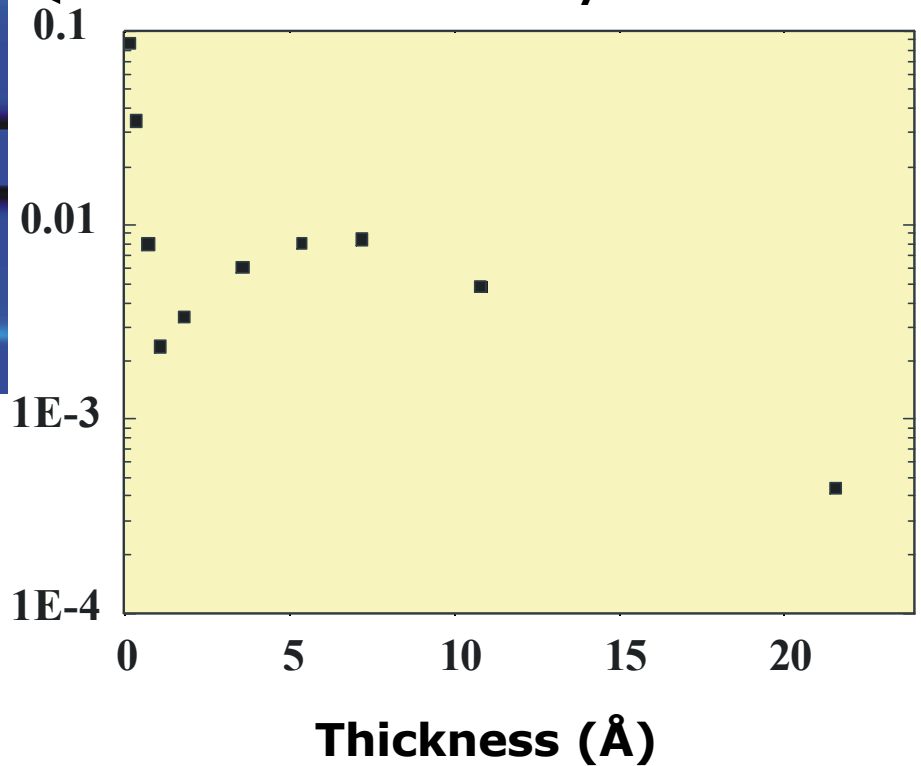




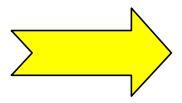
First order peak intensity



Second /First order



Quantitative analysis



Analysis of peaks of several orders
Information about dot's size, shape, coalescence, etc.
Organization used as a tool, not as a goal





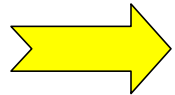
Co/Au(111) overview

- **New process for vertical growth from flat dots**
- **Coercivity at room temperature**
- **Information through X-ray scattering**
Generalization to pillars and magnetic studies?..



1. Introduction

2. Co/Au(111)



3. Fe/bcc(110) stripes

 **3.1 Background**

 **3.2 Growth of thick self-organized Fe(110) stripes**

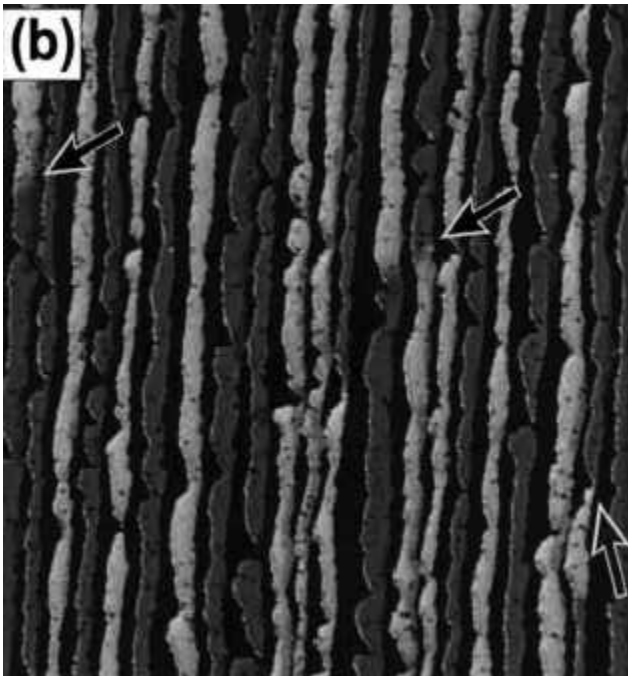
 **3.3 Magnetic functionality at 300K**

4. Conclusion and perspectives



MONOLAYER RANGE : WETTING

↳ nanometer-world / surface physics



1.5ML on vicinal Fe/W(110)

M. Bode et al, J. Electr. Spectr. Rel. Phenom. 114– 116, 1055 (2001)

THICK DEPOSITS : NO WETTING

↳ micrometer-world / materials physics

Fe/Mo(110)

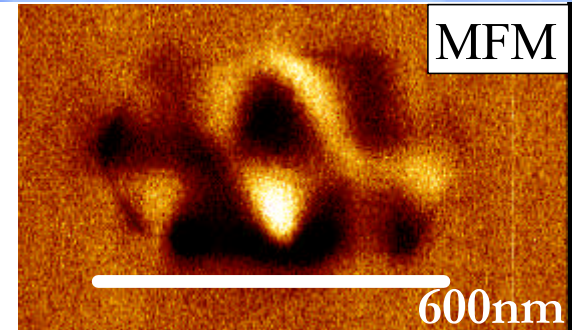
AFM, ~1 μm



MFM:

Y. Samson

(CEA/France)



P.-O.Jubert et al., JMMM 226, 1842 (2002)

P.-O.Jubert et al., PRB64, 115419 (2002)

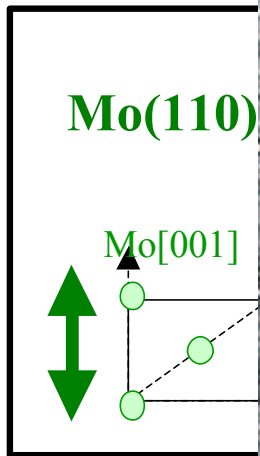
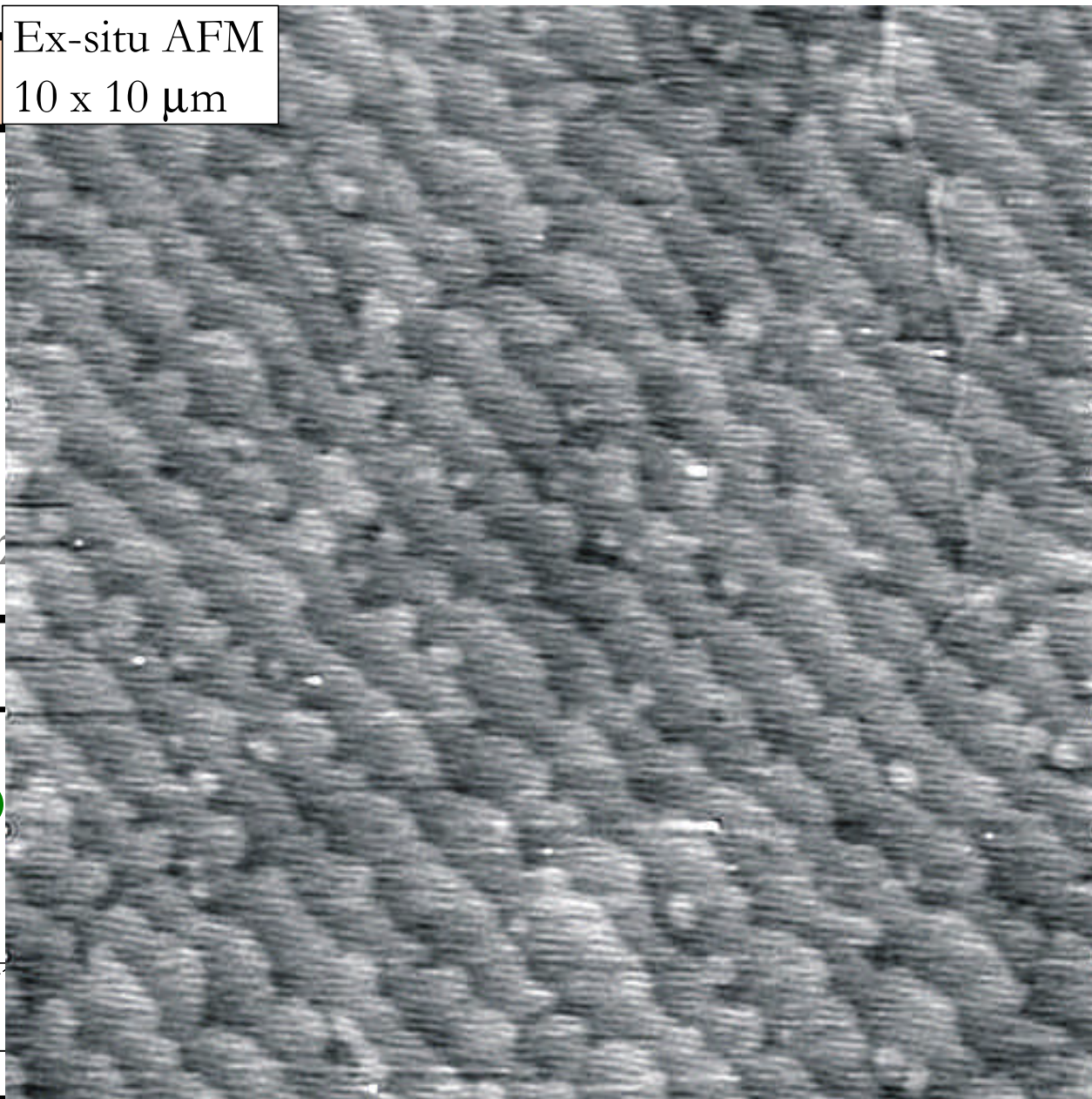
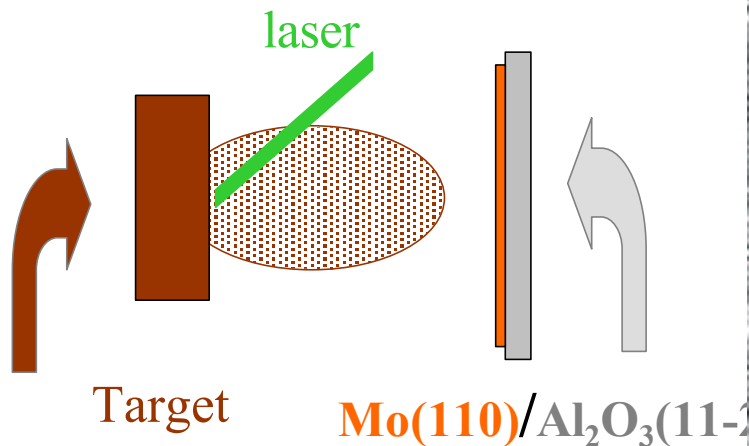
P.-O.Jubert et al., Europhys. Lett. (2003)

↳ Is there an intermediate world ?



PULSED LASER DEPOSITION,

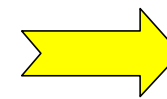
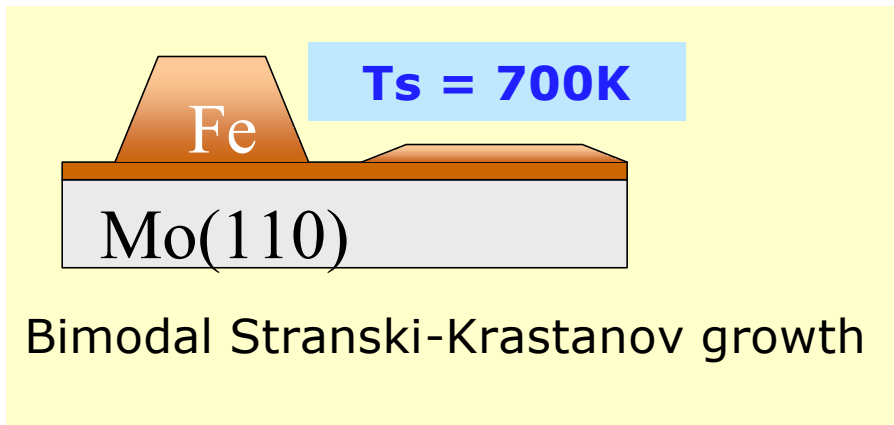
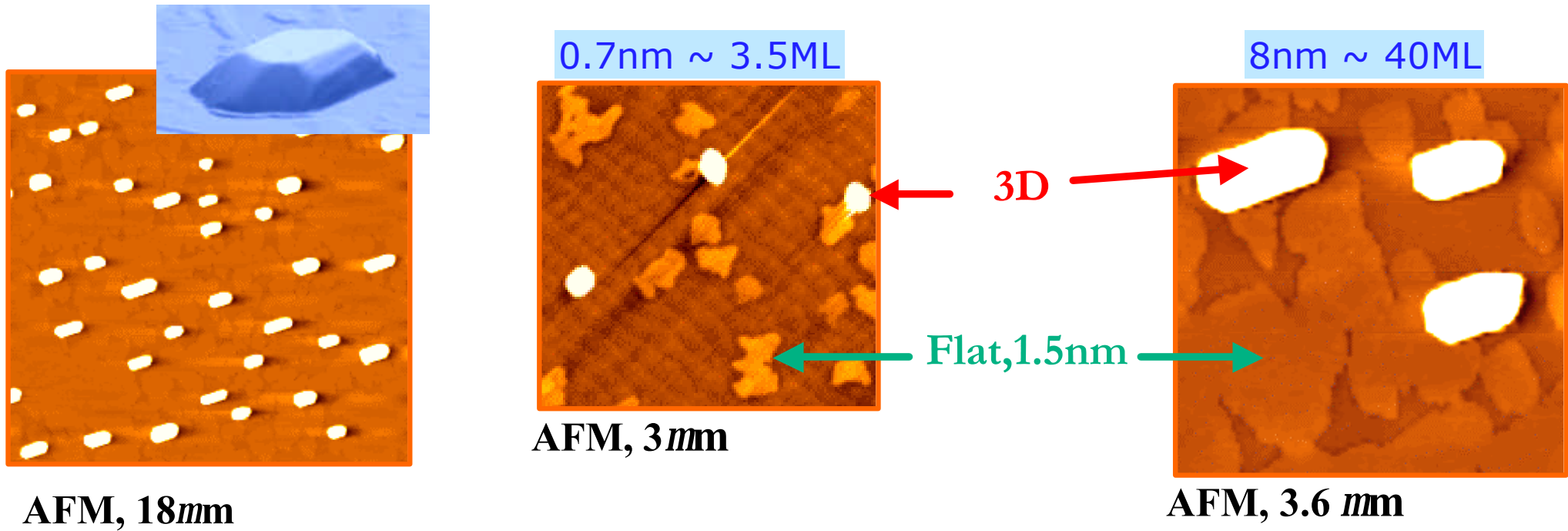
Ex-situ AFM
10 x 10 μm



Buffer layer growth : **O. Fruchart, S. Jaren, J. Rothman, Appl. Surf. Sci. 135, 218 (1998)**



Growth of Fe/Mo(110) at 450°C



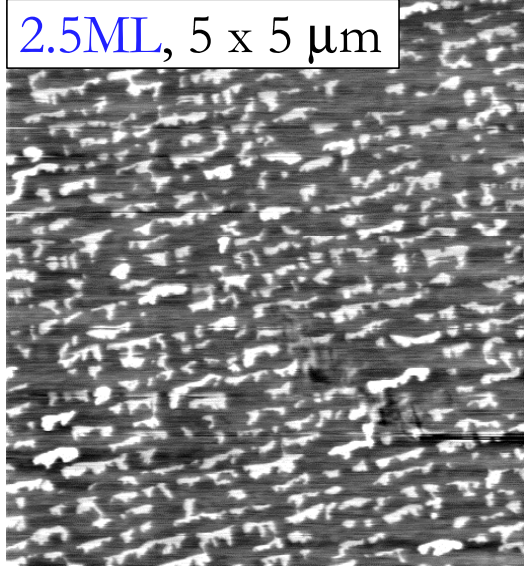
Reason ?
Can it be used ?



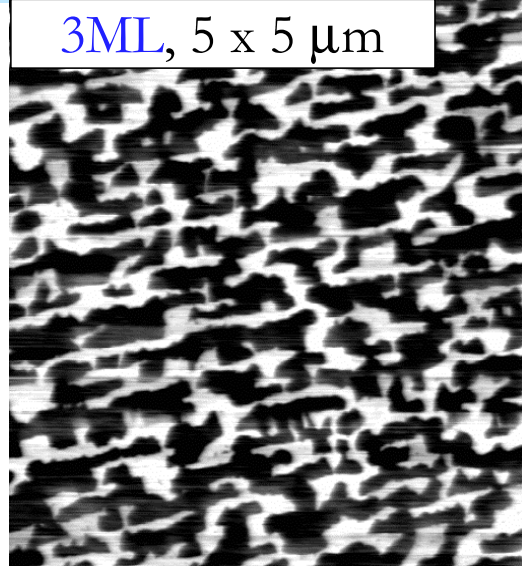
Metastable thickness for Fe/Mo(110)

25°C growth + 450°C Annealing - Ex-situ AFM

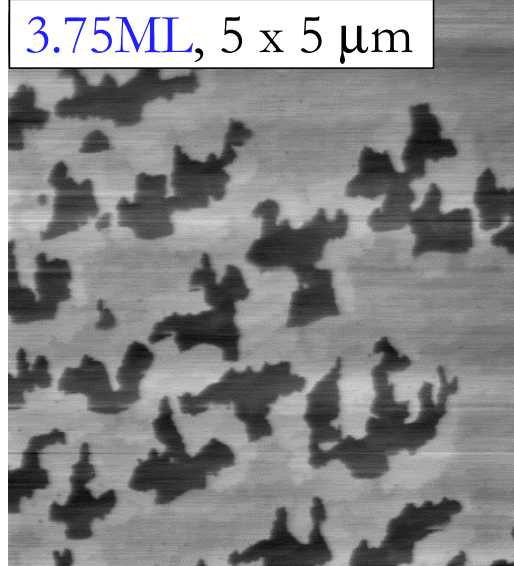
2.5ML, 5 x 5 μm



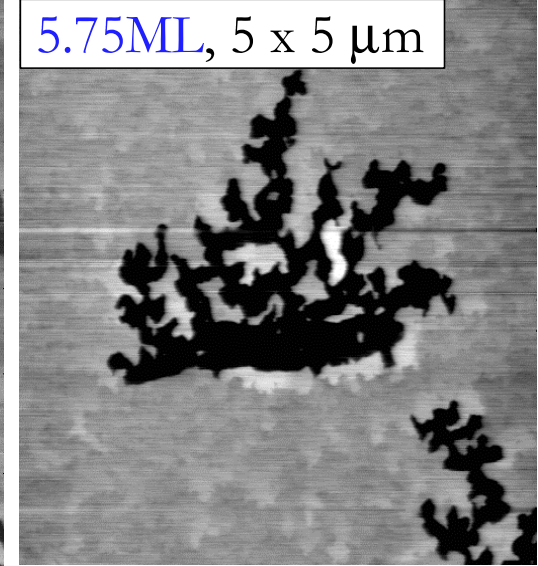
3ML, 5 x 5 μm



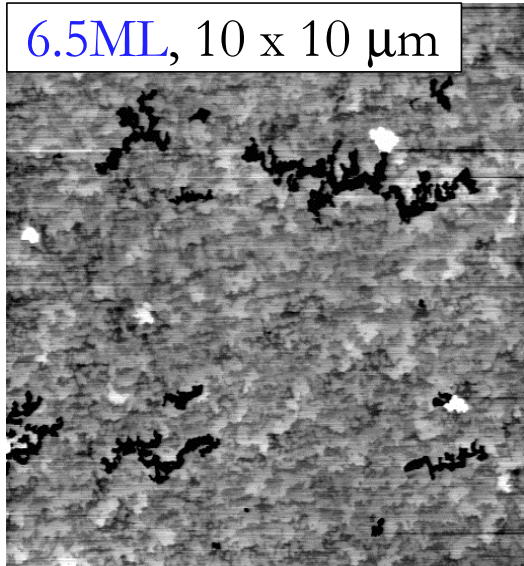
3.75ML, 5 x 5 μm



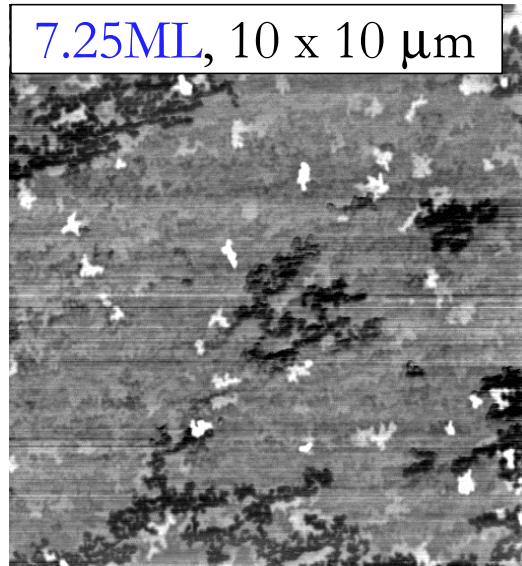
5.75ML, 5 x 5 μm



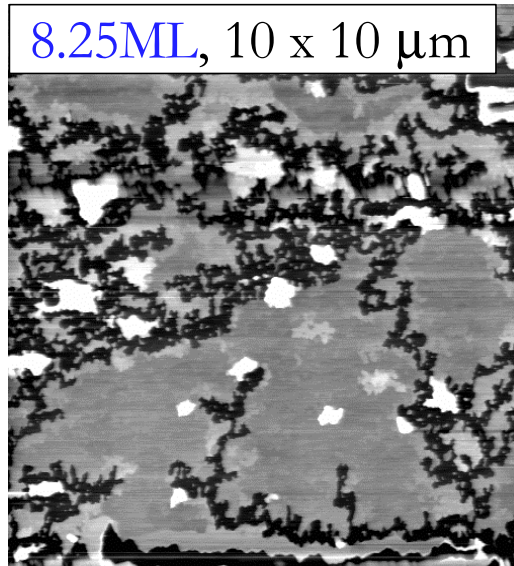
6.5ML, 10 x 10 μm



7.25ML, 10 x 10 μm



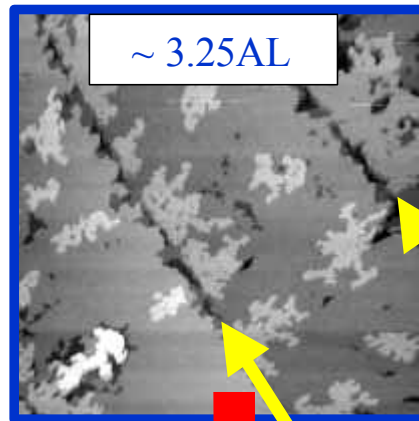
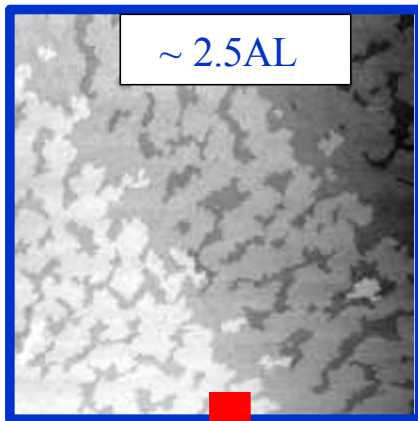
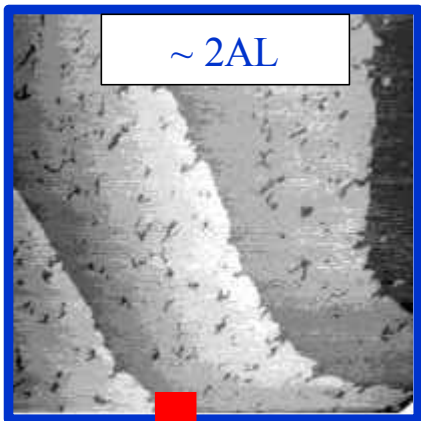
8.25ML, 10 x 10 μm



- $t < 7AL$:
7AL metastable patches
- $t > 7AL$:
patches are 'sucked'
- ↪ 3D dots more stable than 7AL patches.



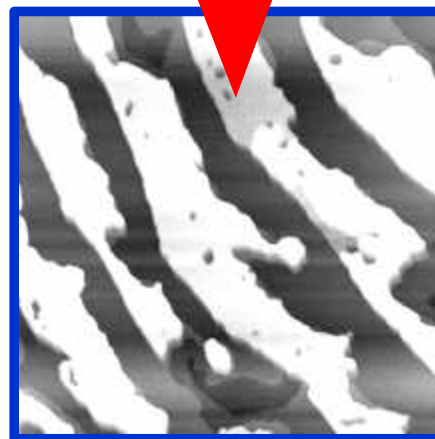
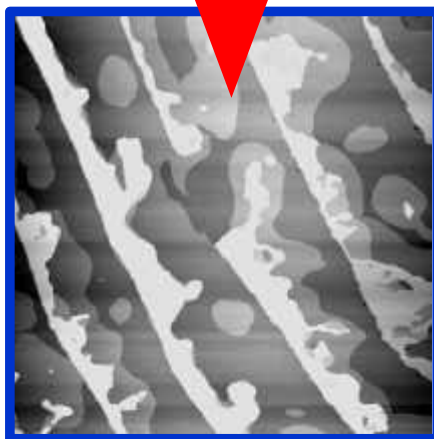
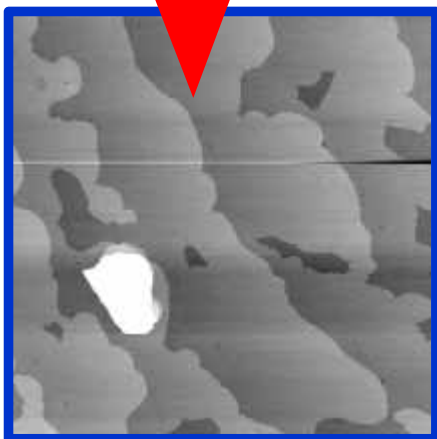
Nucleation along edges



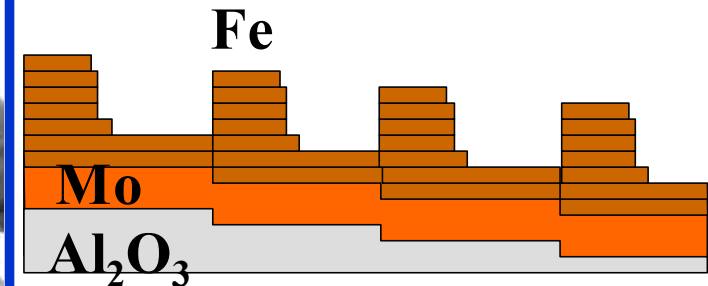
Growth @ 150°C

STM, 750nm x 750nm

Annealing : 450°C
- stress relaxation : stripe formation
- metastable thickness : 7ML



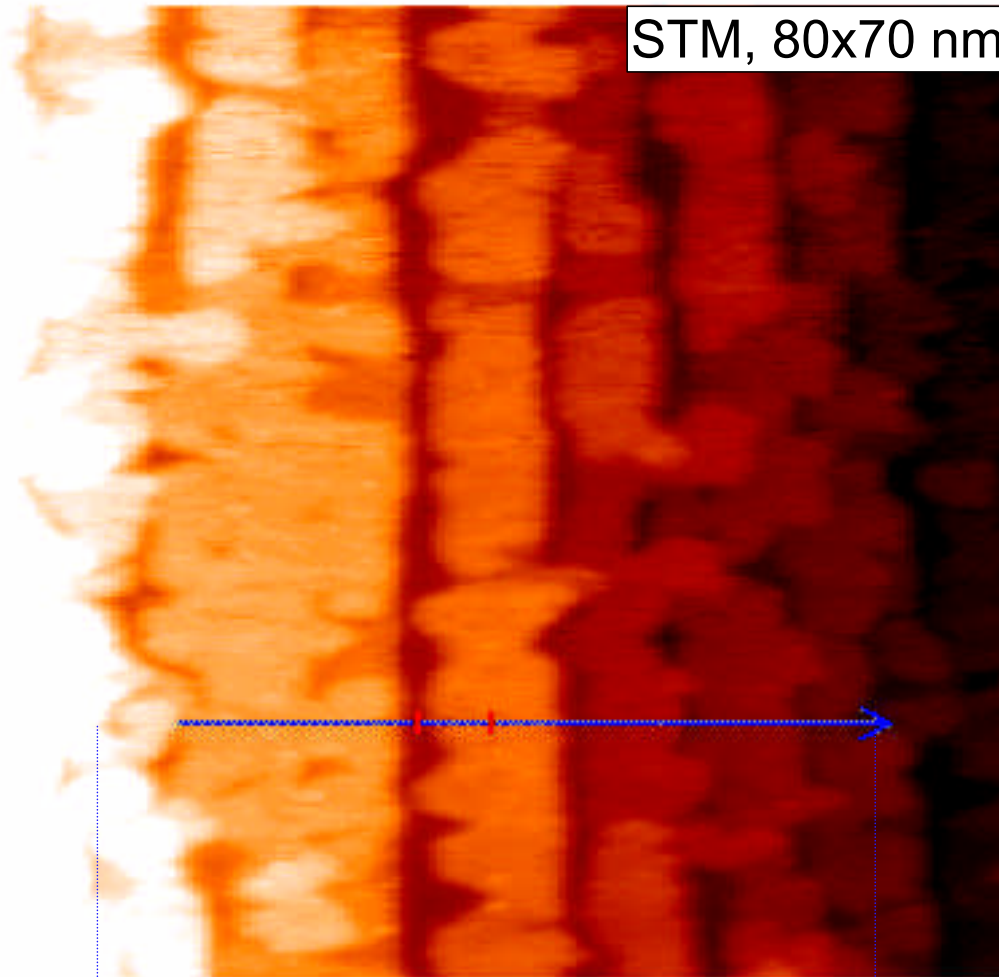
STM, 950nm x 950nm



Role of stress at edges



Valid for other systems ?

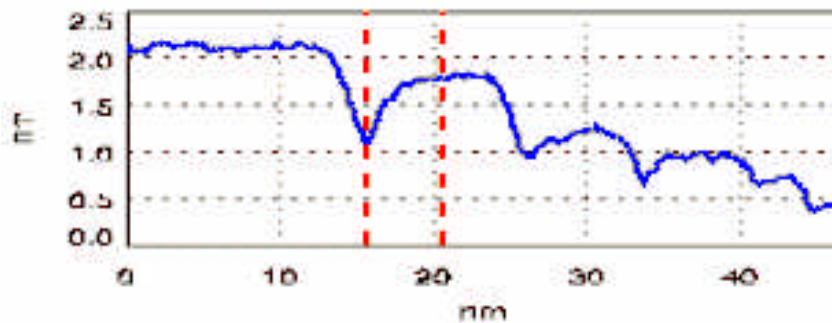


STM, 80x70 nm

3 ML Fe₆₅Ni₃₅ /
Cu(111)-vic 1.2°

↪ Grooves along steps

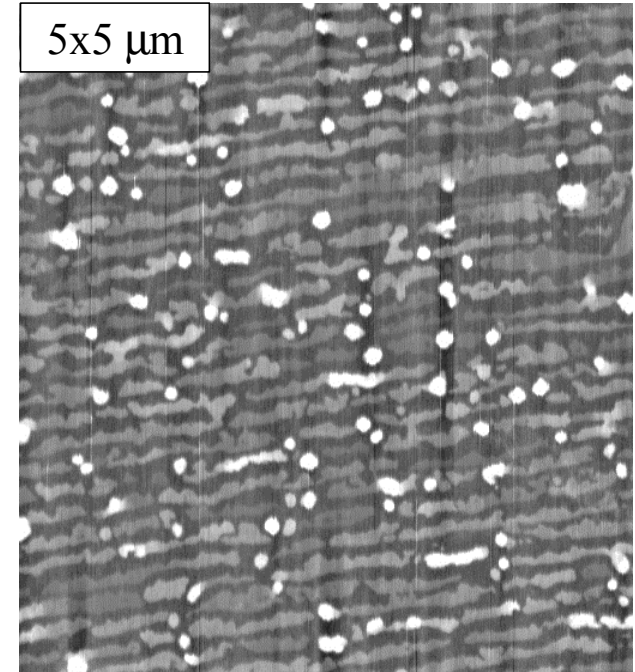
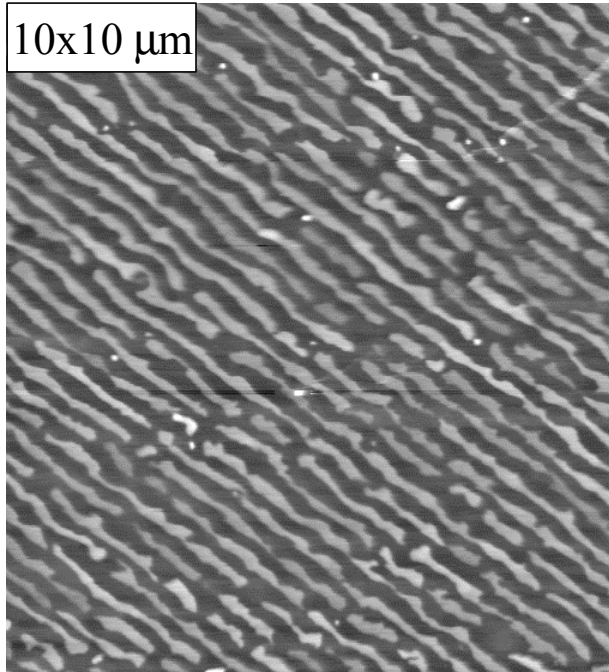
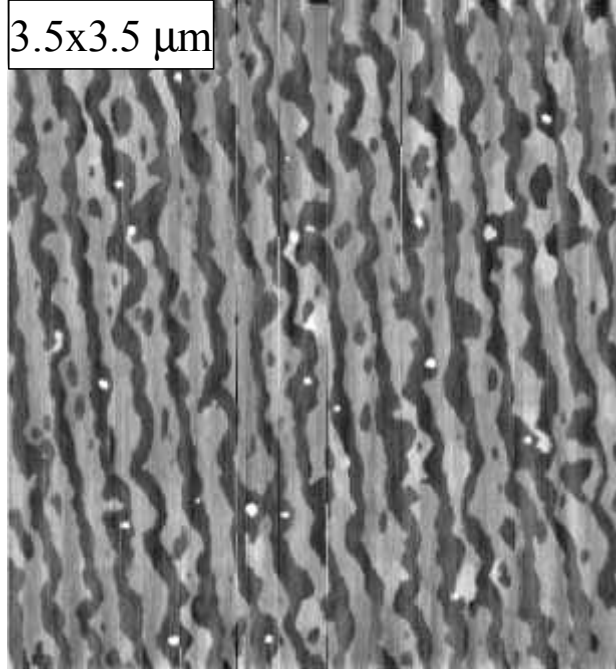
General phenomenon
that may be exploited ?



S. Cherifi *et al.*, PRB 64 (2001) 184405

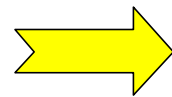
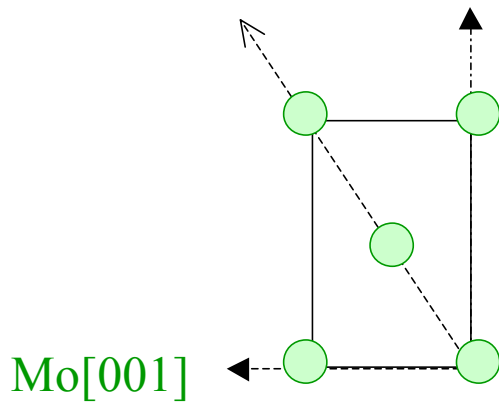


Ex-situ AFM



(Slightly too high temperature for this sample...)

Mo[1-10]

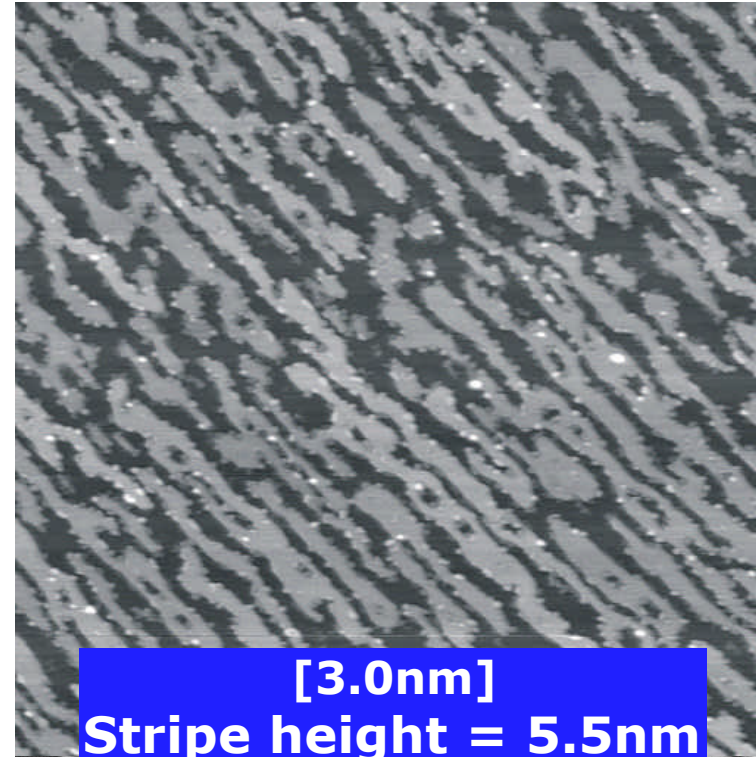
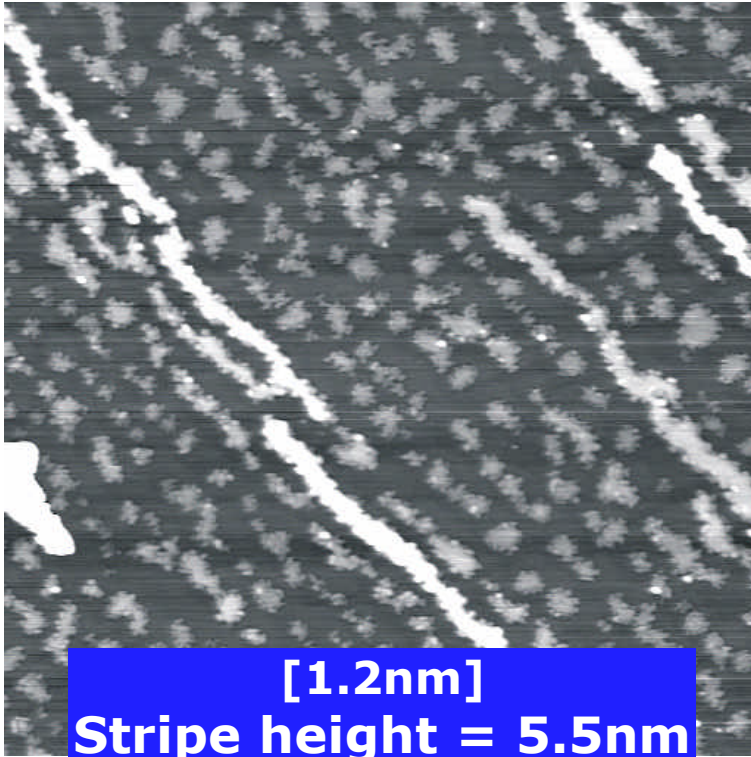


 Tunable orientation and period

Thickness still too small for quasistatic coercivity at 300K

Sapphire\W(8nm)\Fe(150°C, 450°C annealing)

Ex-situ AFM

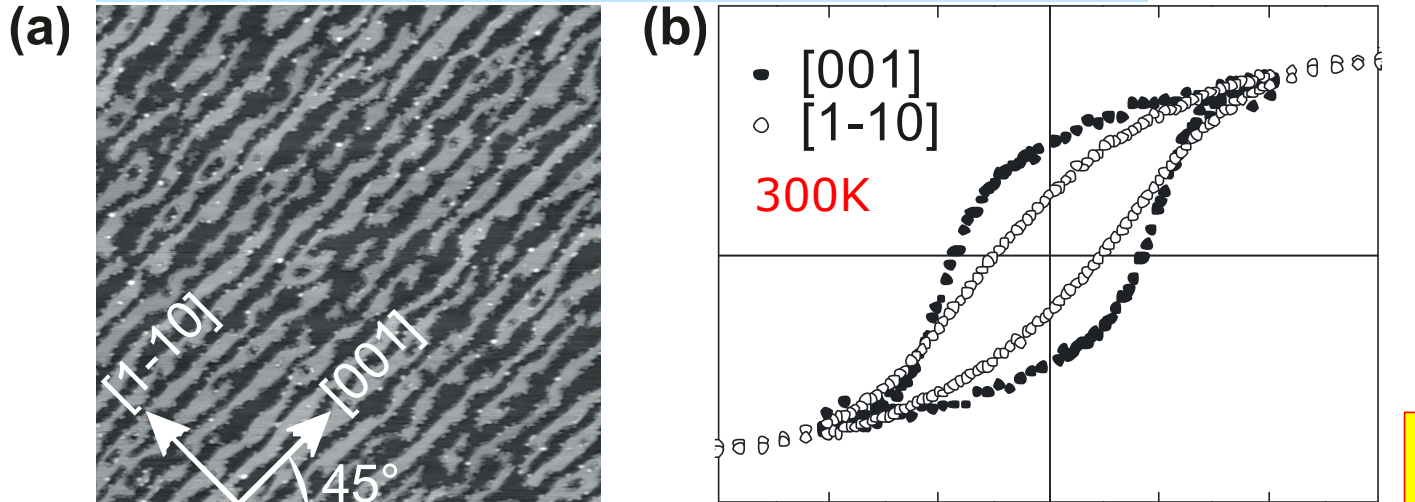


- ↪ Stripes for Fe/W(110)
- ↪ Increased height : 5nm
- ↪ Why difference with Mo ?

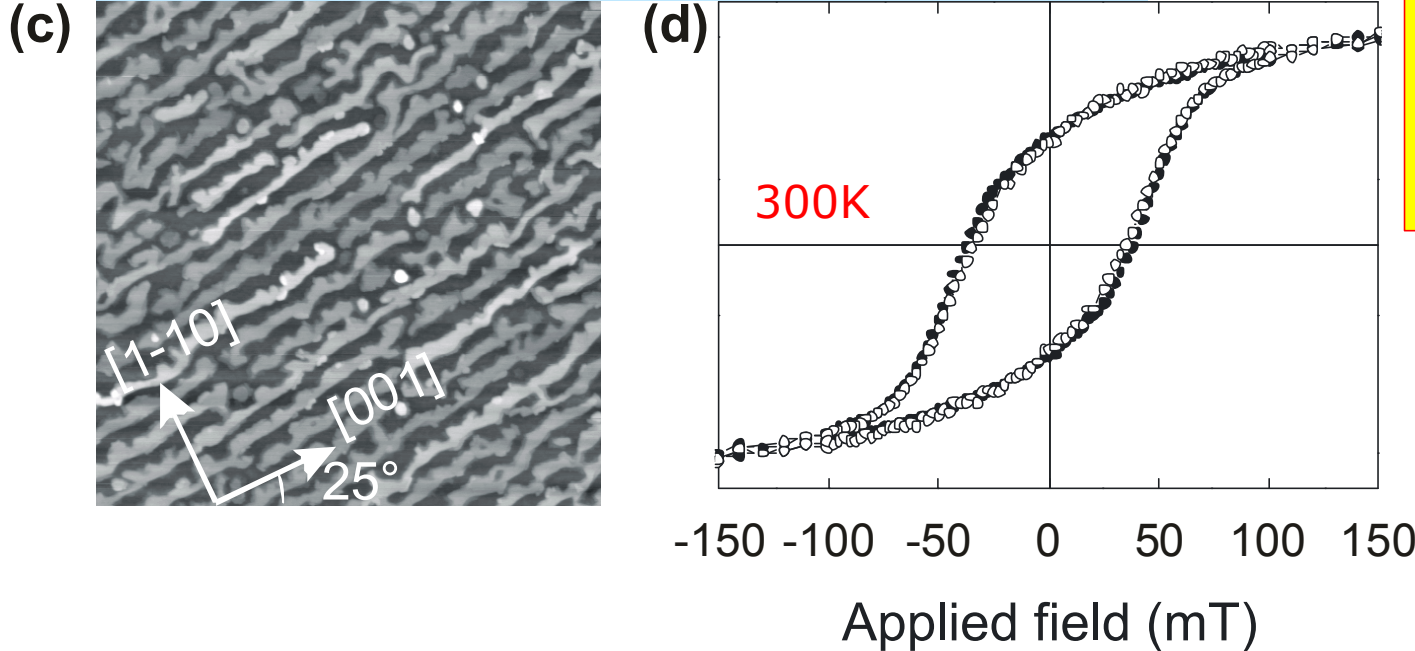


Coercivity at 300K

Sapphire\Mo(1nm)\W(8nm)\Fe(2.5nm)\Mo(1nm)\Al(3nm)



Sapphire\Mo(8nm)\W(1nm)\Fe(2.5nm)\Mo(1nm)\Al(3nm)



↪ **Coercivity and remanence at 300K**
 > behaves like a **conventional hard magnetic material**



PEEM = PhotoElectron Emission Microscope

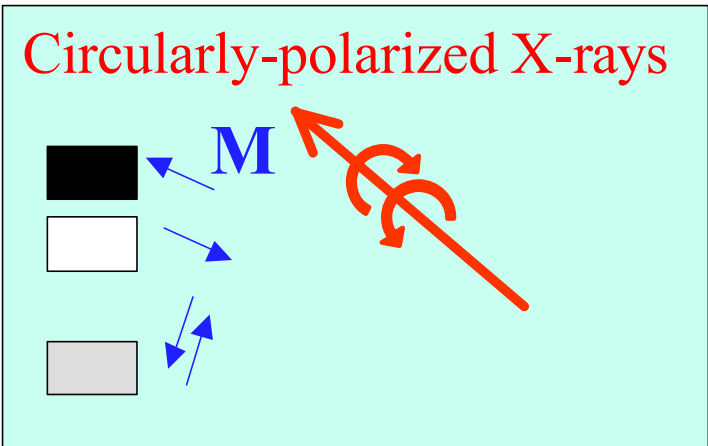
Combine XMCD and imaging with secondary electrons

X-ray Magnetic Circular Dichroism

Element selectivity

Magnetic information

Imaged using electromagnetic lenses

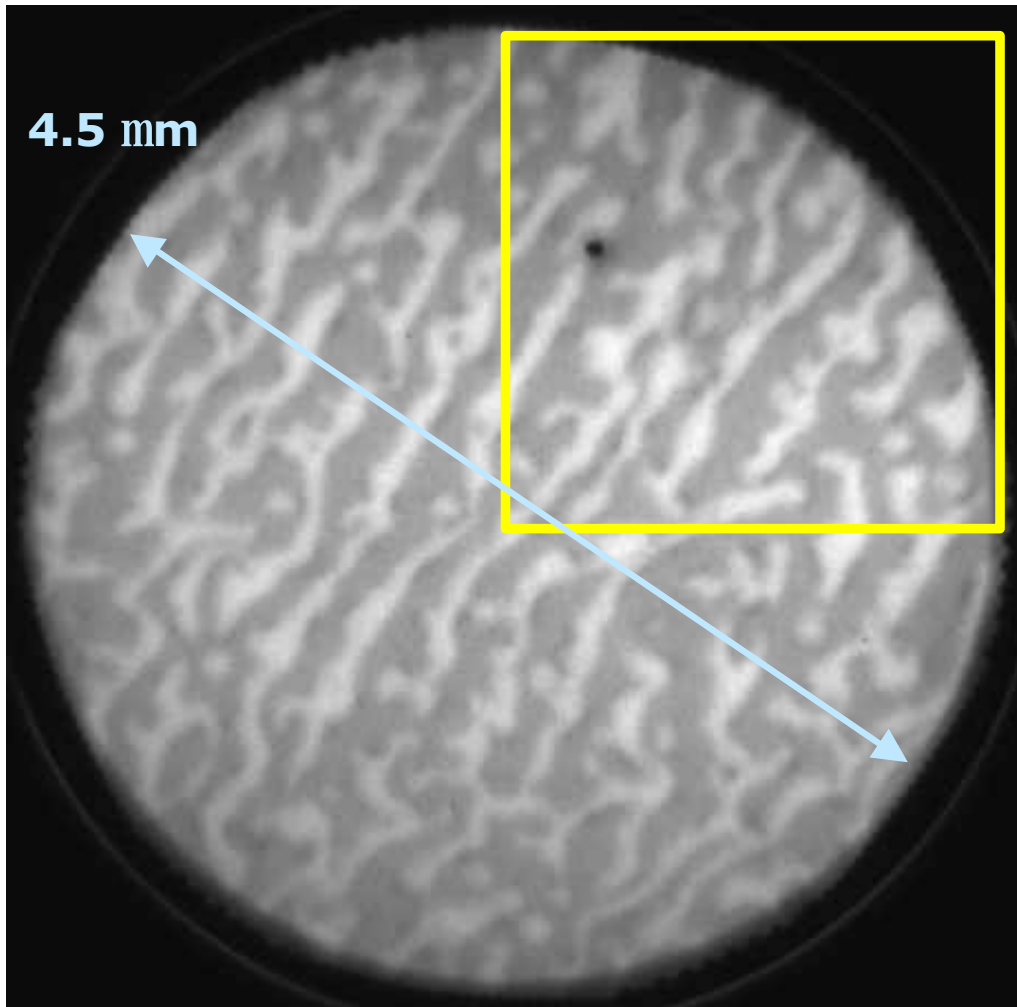


Probes directly the magnetization direction, not stray fields like in MFM (Magnetic Force Microscope)



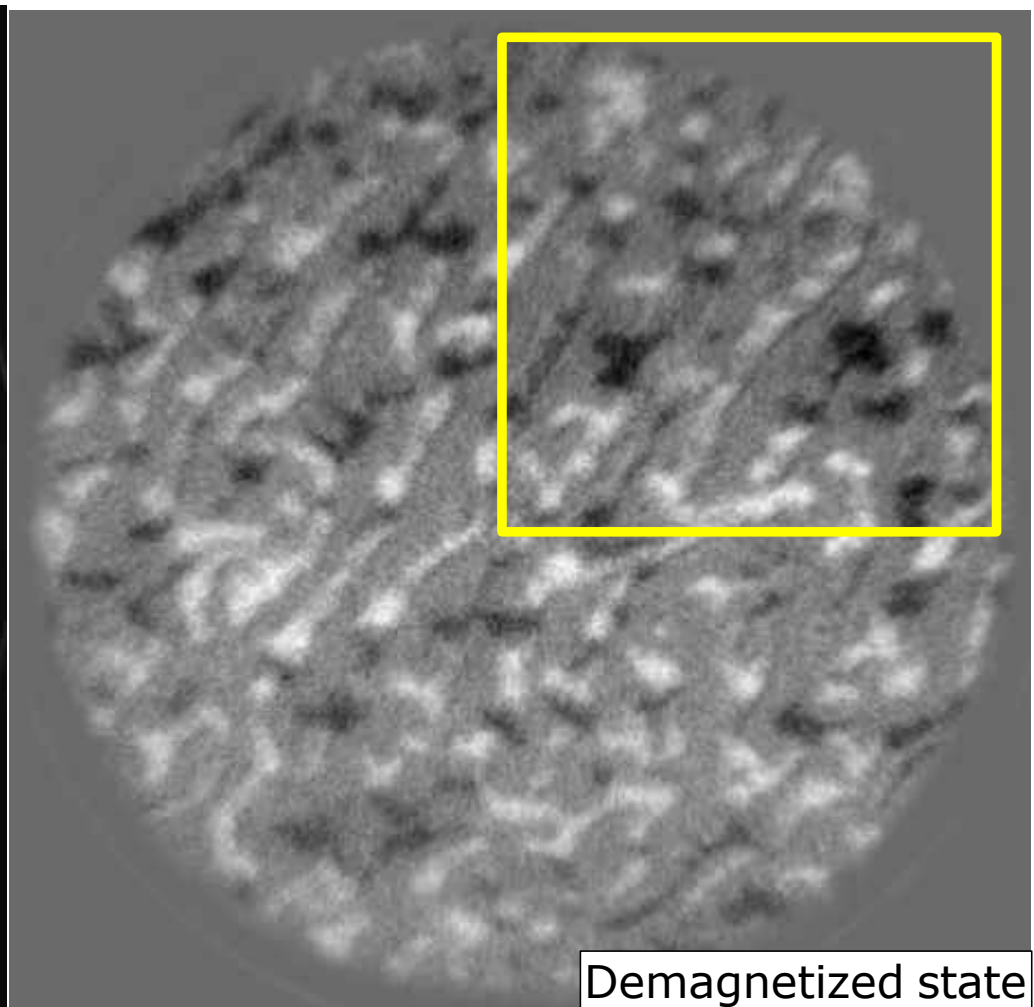
PEEM = PhotoElectron Emission Microscope

Fe L3 edge

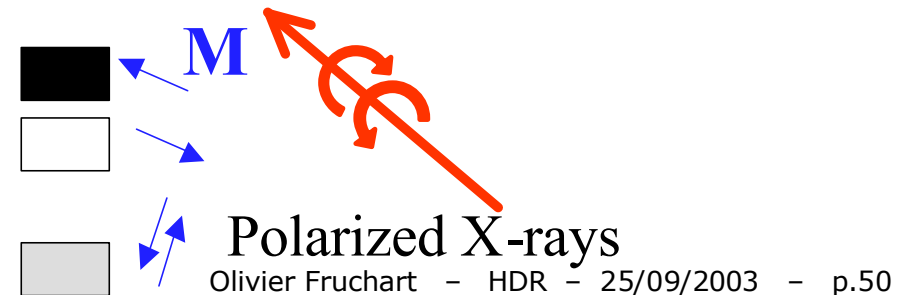


4.5 mm

Sample: Sapphire\Mo(8nm)\W(1nm)\
Fe(2.5nm)\Mo(1nm)\Al(3nm)



Demagnetized state

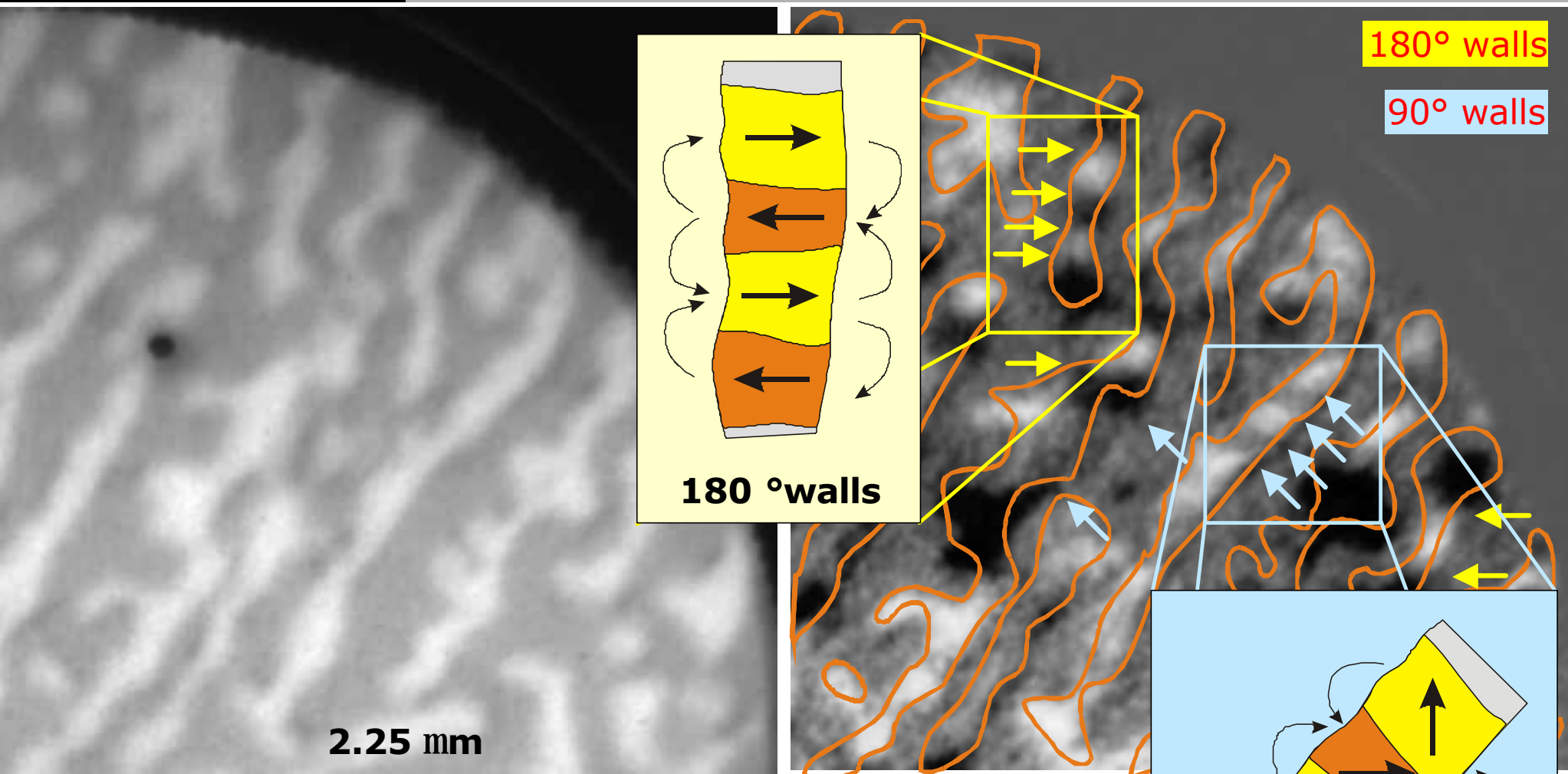


ELETTRA Synchrotron, Trieste
Coll. Andrea Locatelli, Jan Vogel





Domain patterns

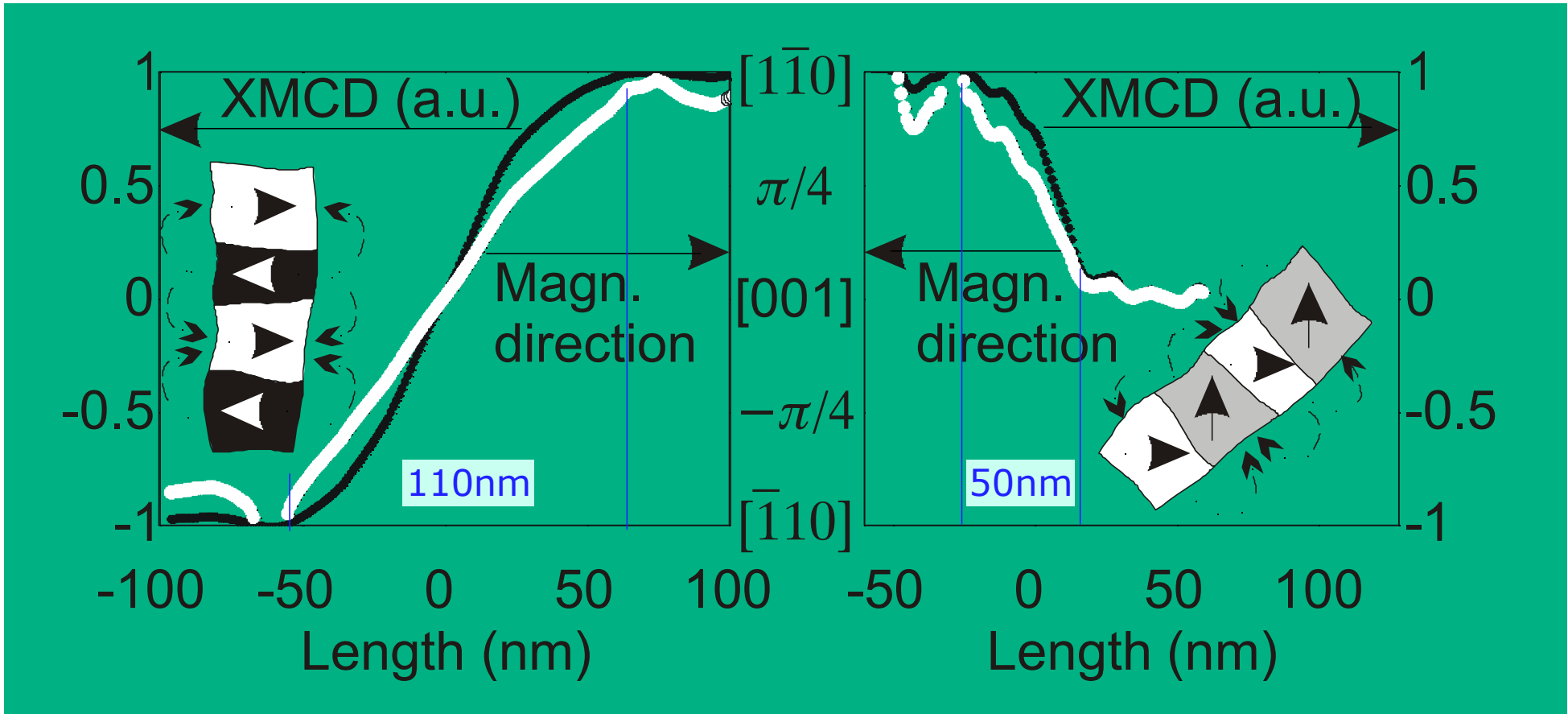


↪ **Stable domain patterns at 300K**
 > behaves like a **conventional soft material**



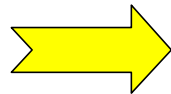


PEEM resolution



$180^\circ > \lambda = 110\text{nm}$

$90^\circ > \lambda = 50\text{nm}$



PEEM resolution better than 50nm

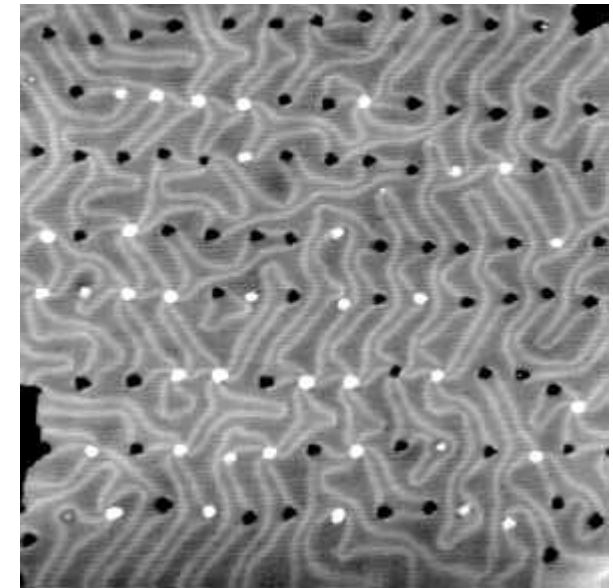
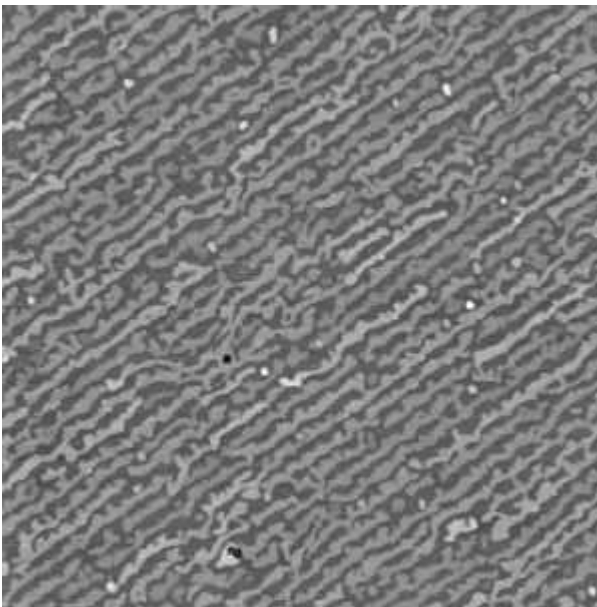


- 1. Introduction
 - 2. Co/Au(111) nano-columns
 - 3. Fe/bcc(110) stripes
 - ➔ ● **4. Conclusion and perspectives**
-

FROM SURFACES TO MAGNETIC MATERIALS

➤ Original growth processes:

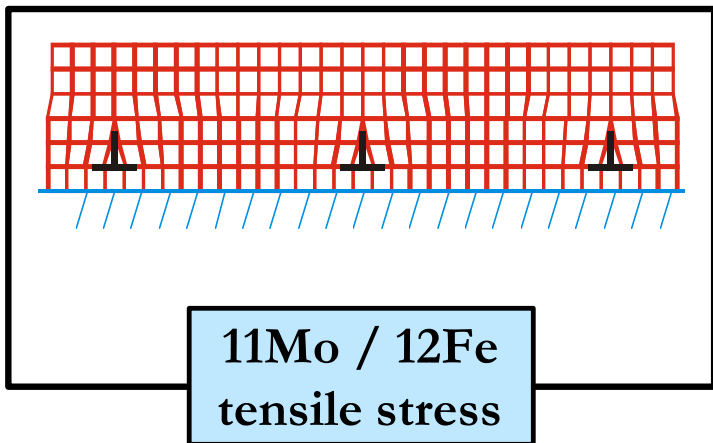
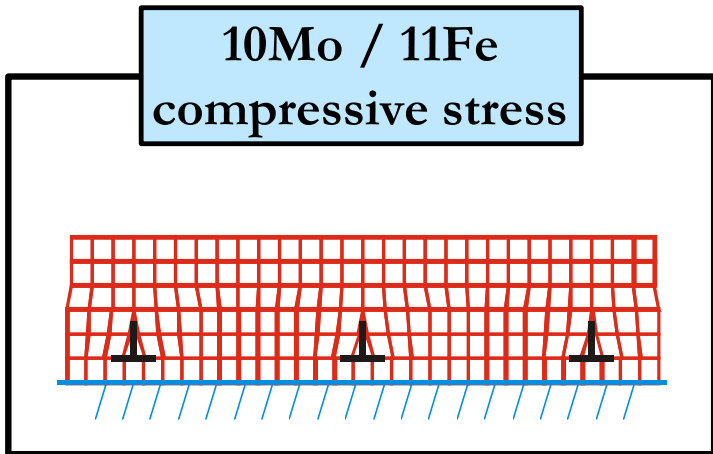
- ↳ Use surface science effects:
dislocations, atomic steps, reconstructions, etc.
- ↳ Self-organization with multi-atomic-layers height



➤ Magnetism:

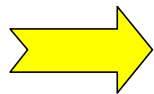
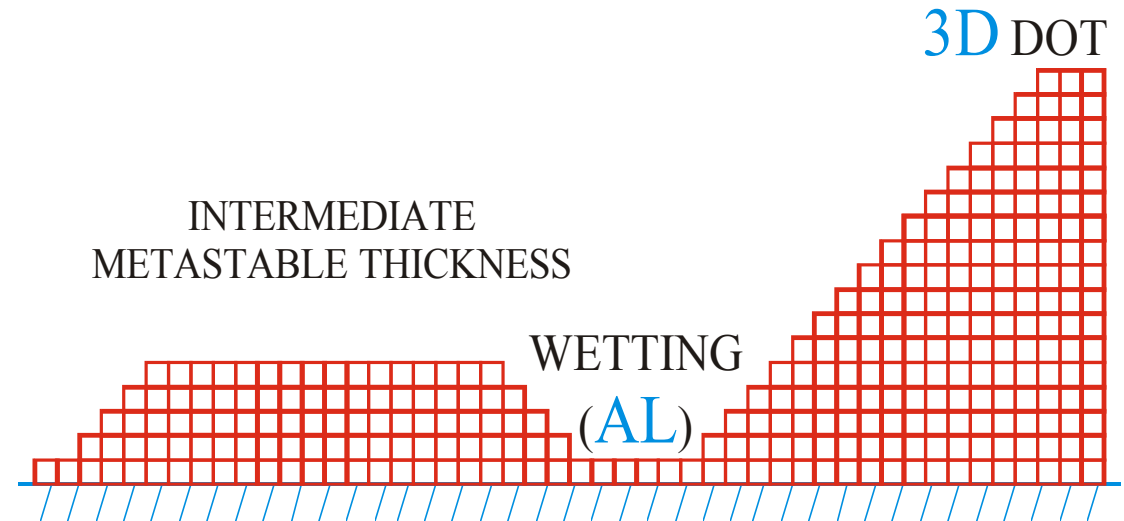
- ↳ Rise Curie and blocking temperature
> coercivity and stable domains
- ↳ Increase amounts of material

Origin of metastable thickness



UNUSUAL BEHAVIOR.

Possible explanation = stress minimization
 commensurate / incommensurate transition of
 interface dislocation array



Requirement to demonstrate this:
 Control independently surface energy and
 lattice parameters mismatch



Sapphire\Mo(8nm)\interface\Fe(150°C, 450°C annealing)

Ex-situ AFM

1nm

2.5nm

Average thickness

**Mo\
W(<1nm)**

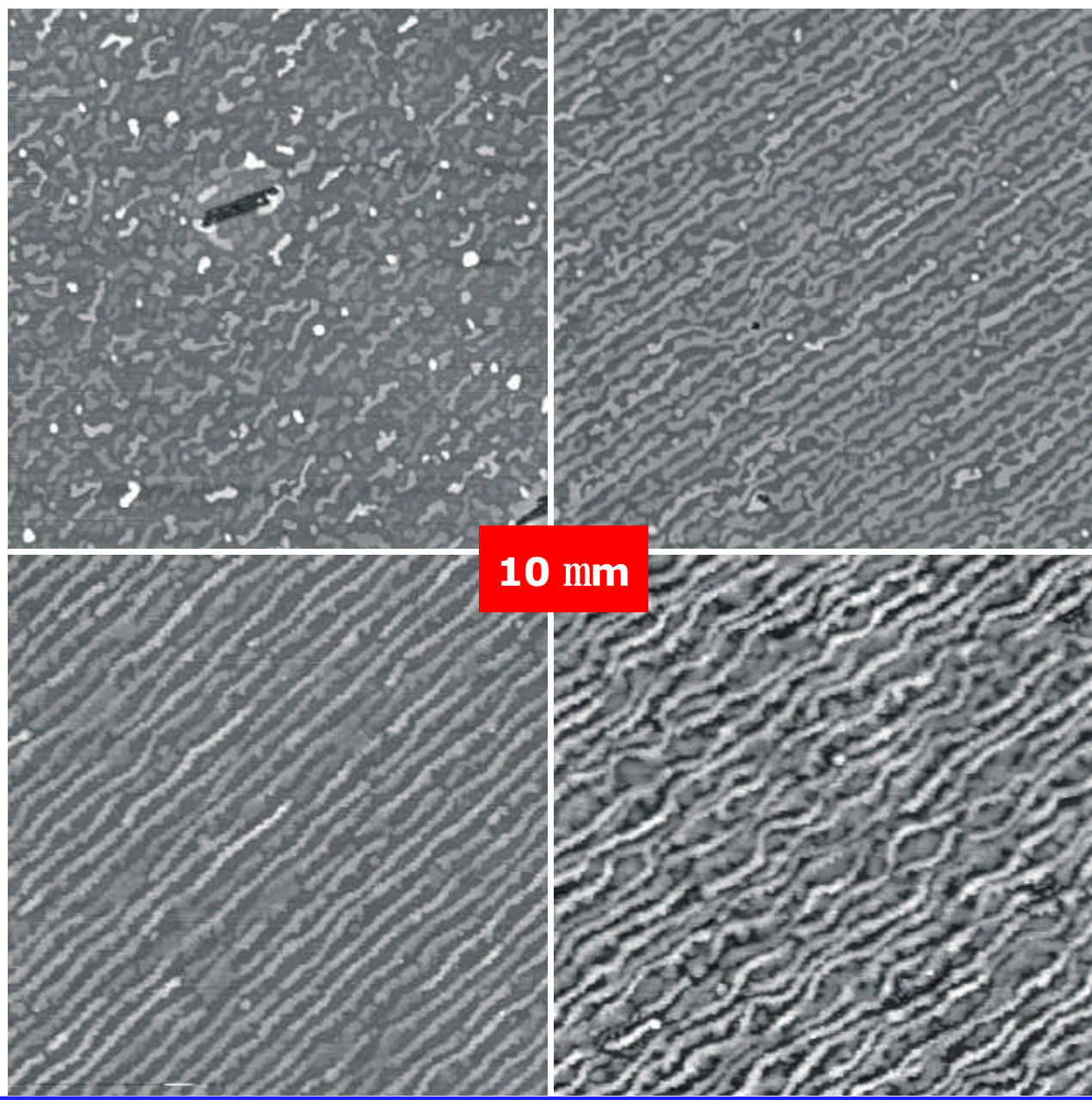
Height : 5nm

Mo

**Height : bimodal
1.5/4nm**

**Chemical
interface**

10 mm

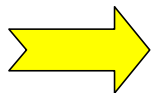
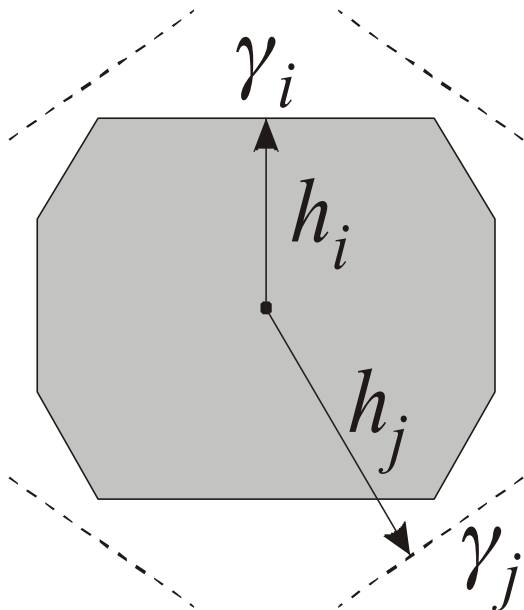




Wulff's theorem

Free crystal

$$\frac{\gamma_i}{h_i} = \text{Constant}$$

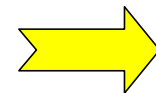
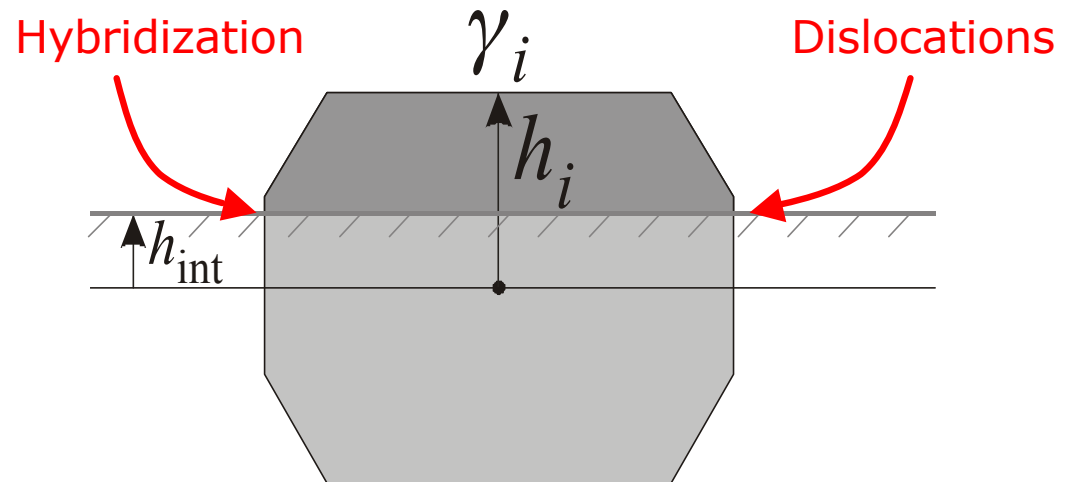


No facets with high surface energy

Wulff Kaishev's theorem

Supported crystal (growth on surfaces)

$$\frac{\gamma_i}{h_i} = \frac{\gamma_S - \gamma_{int}}{h_{int}} = \text{Constant}$$

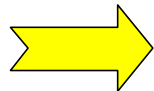
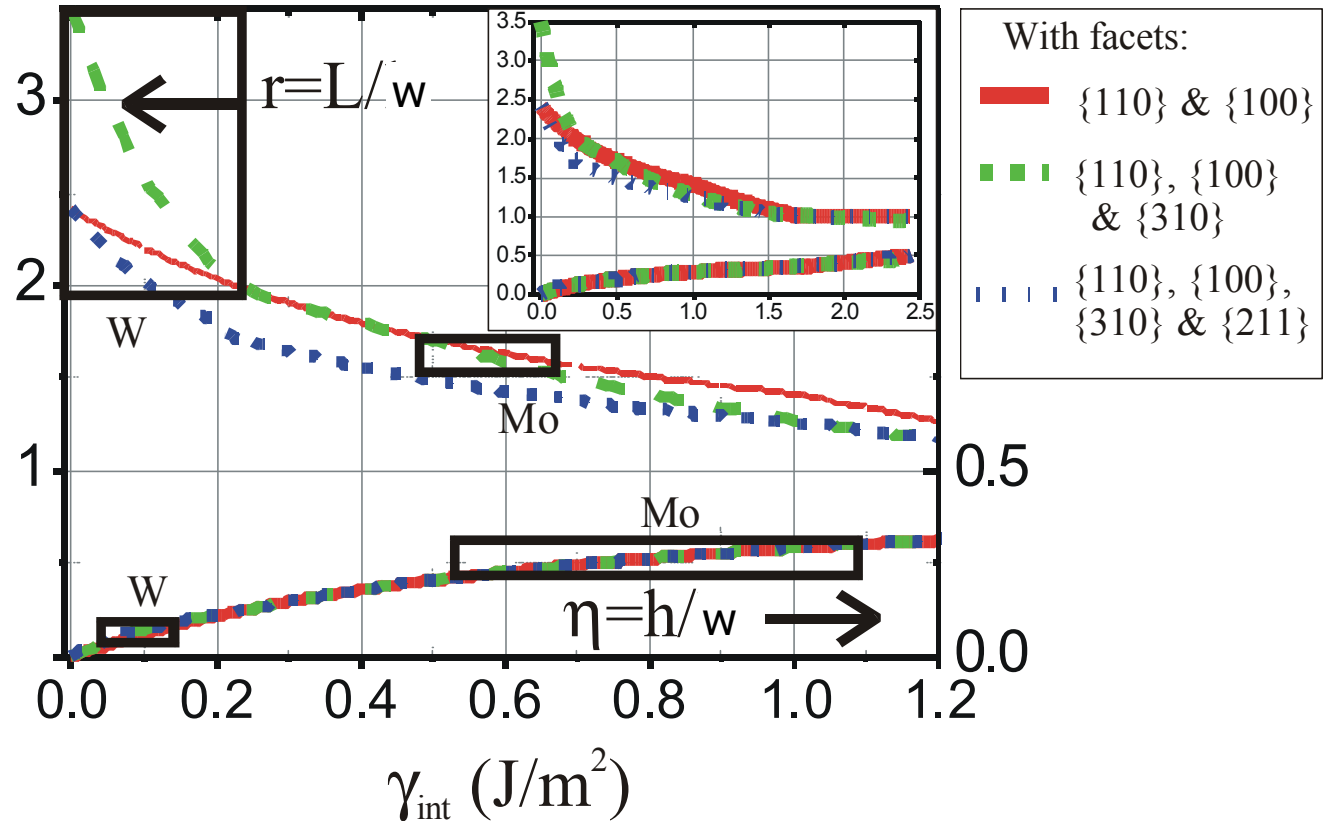
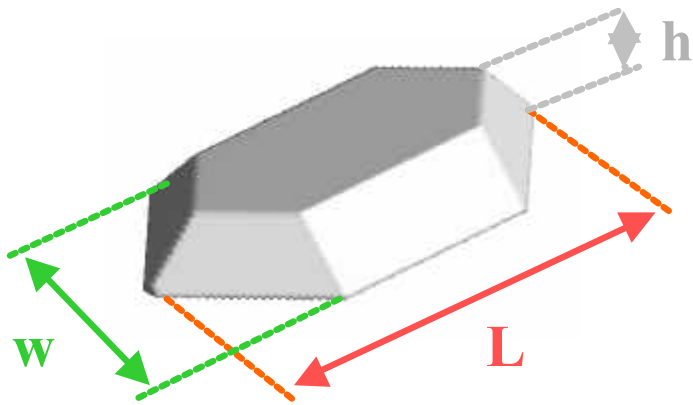


Truncated crystal



Influence of both surface energy and lattice mismatch (>dislocations)

Fe/bcc(110) 3D dots



NEED:
Control independently surface energy and lattice parameters mismatch



FROM SURFACES TO MAGNETIC MATERIALS ... and back

GROWTH ENGINEERING

A tool: independant control of lattice parameter and interface

- Ultrathin interface layers
- Solid solutions of bcc elements

New 3D self-organized systems

- CoAg/bcc(110) or FeAg/bcc(110)
- ...

CHARACTERIZATION TECHNIQUES

- PEEM Microscopy
- X-ray magnetic scattering

APPLICATION TO MAGNETISM

Fundamental properties:

- Interface anisotropy, non-linear magneto-elasticity
- 'Dislocation' magnetic anisotropy

Micromagnetism:

- Magnetization reversal, nucleation
- Walls and vortex manipulation

Transport:

- GMR: cpp configuration with in-plane current
- Polarized current-assisted magnetization reversal



PhD students

P.-O. Jubert (1998–2001) / M. Eleoui (2001- 2004)

Jean-Christophe Toussaint

Laboratoire Louis Néel (numerical micromagnetism)

Dominique Givord

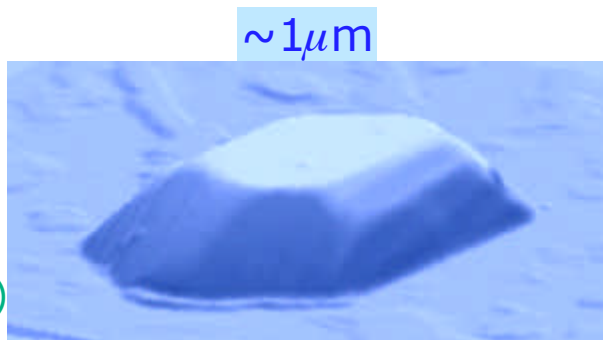
Director of LLN and advisor during PhD

And many many other people !

Model systems

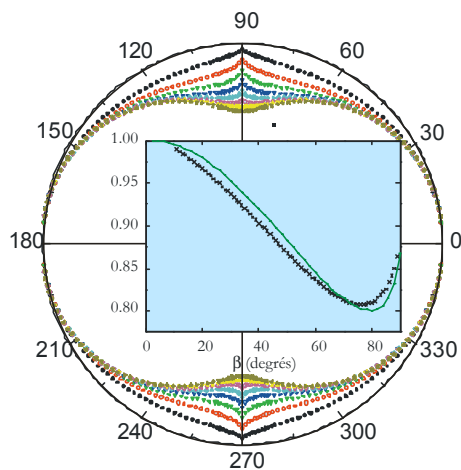
CEA (Y.Samson)

LLN
(J.C.Toussaint,
W.Wernsdorfer)



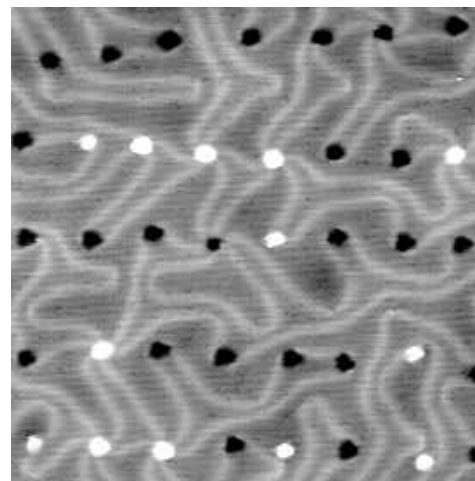
Self-assembled Fe/Mo(110) dots

LLN
(J.C.Toussaint)



Micromagnetic modelling

Demonstrate functionality at 300K



~100nm

MPI-Halle

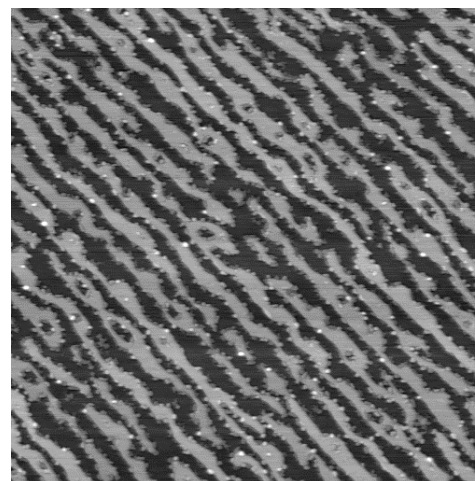
Cristallo (J.M.Tonnerre...)

IPCMS (F. Scheurer...)

CEA
(G.Renaud, A.Barbier...)

GPS-Jussieu
(S.Rousset...)

Self-organized Co/Au(111) pillars



~10μm

IPCMS (F.Scheurer)

ELETTRA (A.Locatelli...)

LLN (J.Vogel)

Self-organized Fe/bcc(110) stripes

PhD students

P.-O. Jubert (1998–2001) / M. Eleoui (2001- 2004)

Olivier Fruchart - HDR - 25/09/2003 - p.61