Self-organized growth on the Au(111) surface

Olivier Fruchart

02/03/2002

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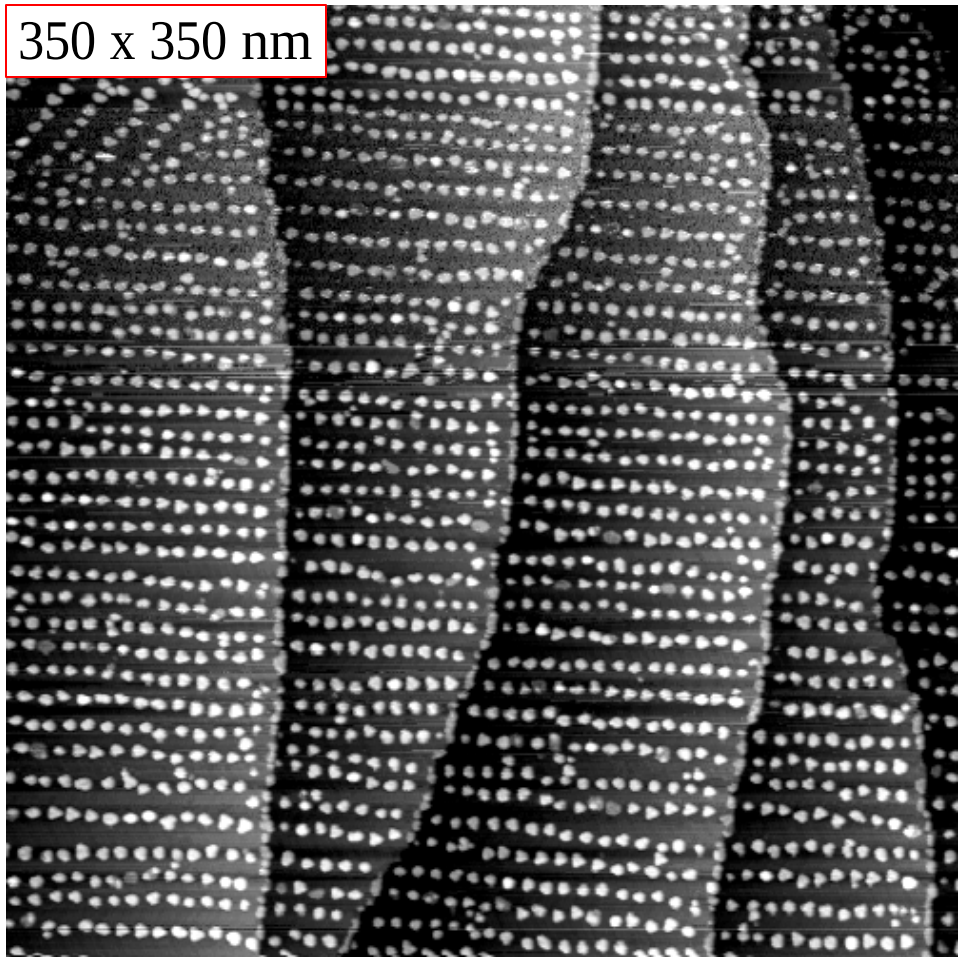
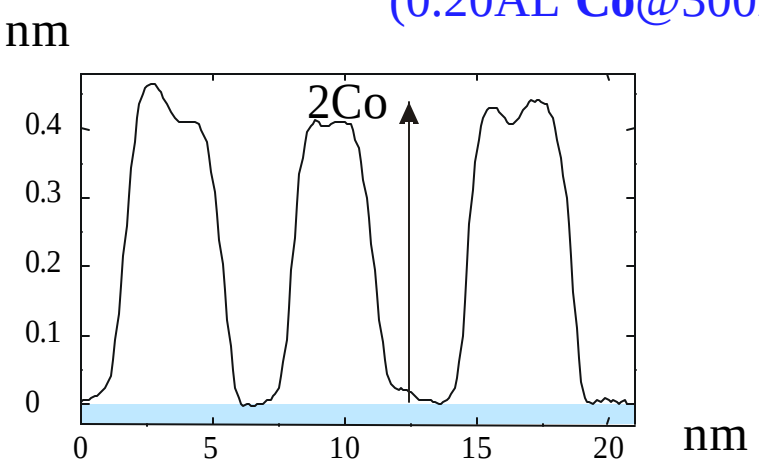
FACTS:

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

ORIGIN ?



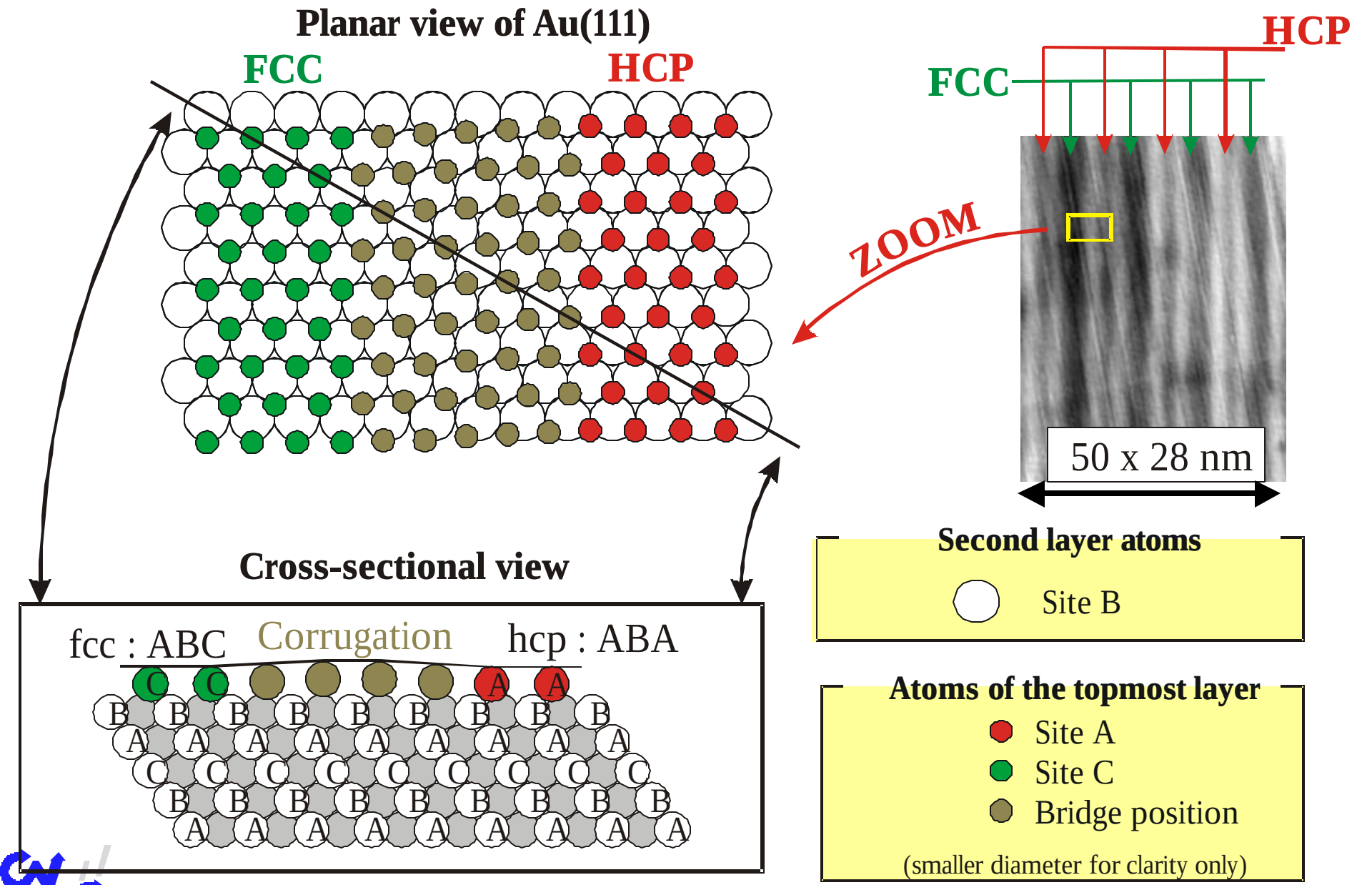
(0.20AL **Co**@300K)

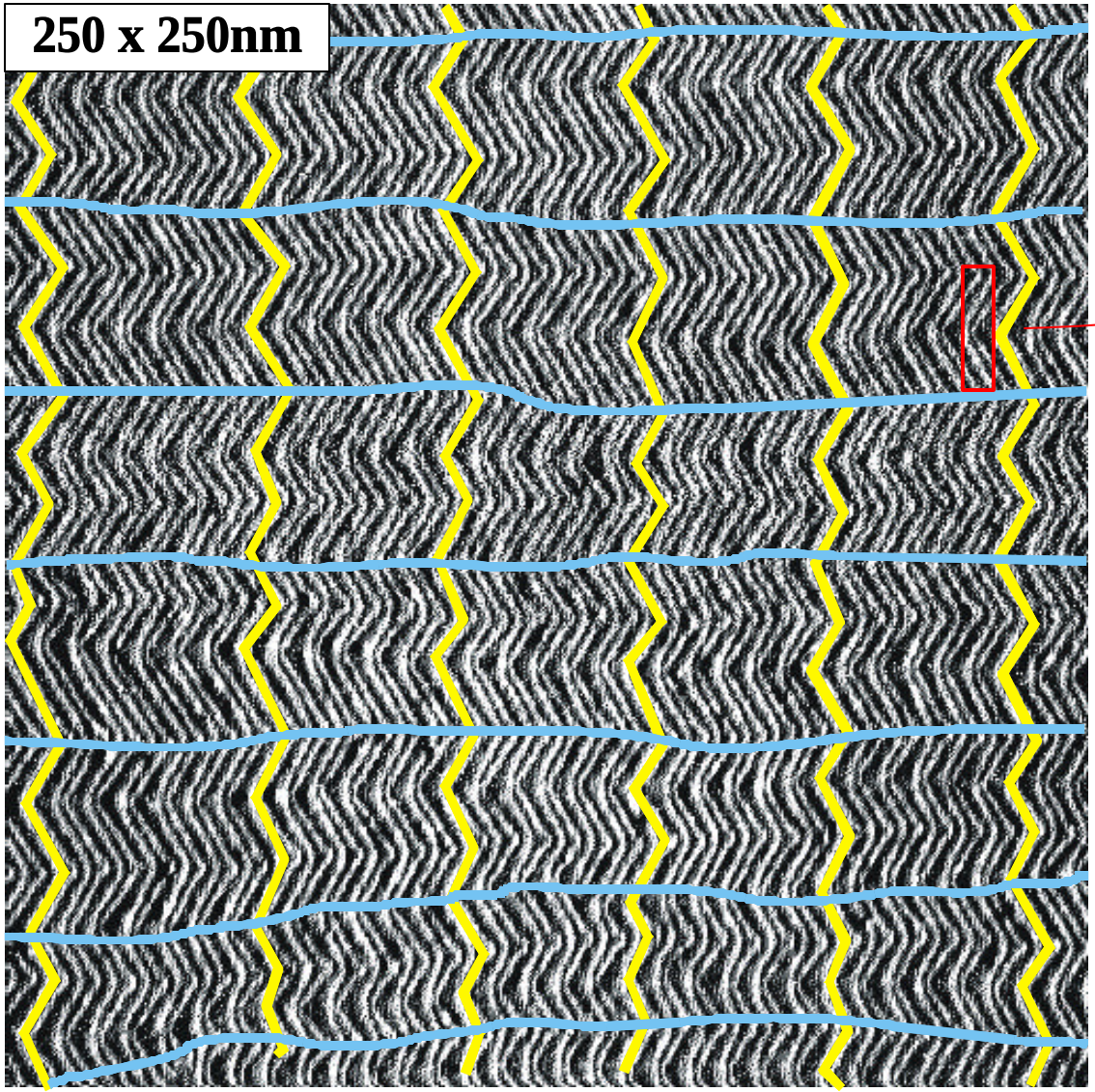


Fe, Ni : 1 AL-high dots

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

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


Planar view of Au(111)
FCC
HCP
HCP
FCC
ZOOM
50 x 28 nm
Cross-sectional view
fcc : ABC
Corrugation
hcp : ABA
Second layer atoms
Site B
Atoms of the topmost layer
Site A
Site C
Bridge position
(smaller diameter for clarity only)



Corrugation ~ 0.2 Å

Unit cell size :
~ 7.5 x 25nm

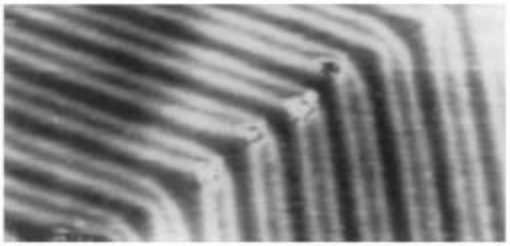
↳ Isotropic surface relaxation

J. V. Barth et al., Phys. Rev. B 42 (15), 9307 (1990)

A.R. Sandy et al., Phys. Rev. B 43 (6), 4667 (1991)

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

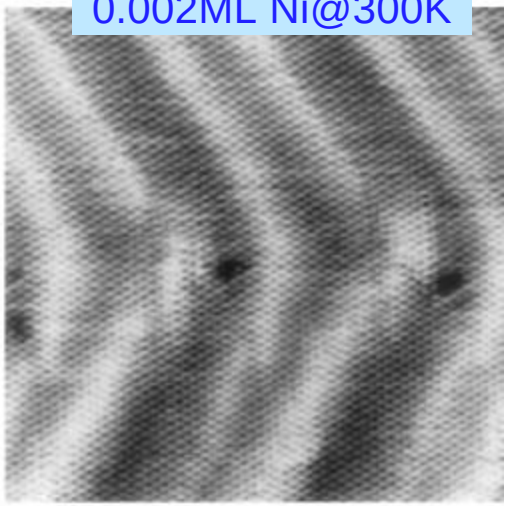
a)



Element	Surface free energy (eV)	Heat of sublimation (eV)
Ag	0.50	2.95
Al	0.56	3.39
Cu	0.69	3.51
Au	0.72	3.79
Ni	0.90	4.45
Co	0.94	4.40
Fe	0.96	4.32

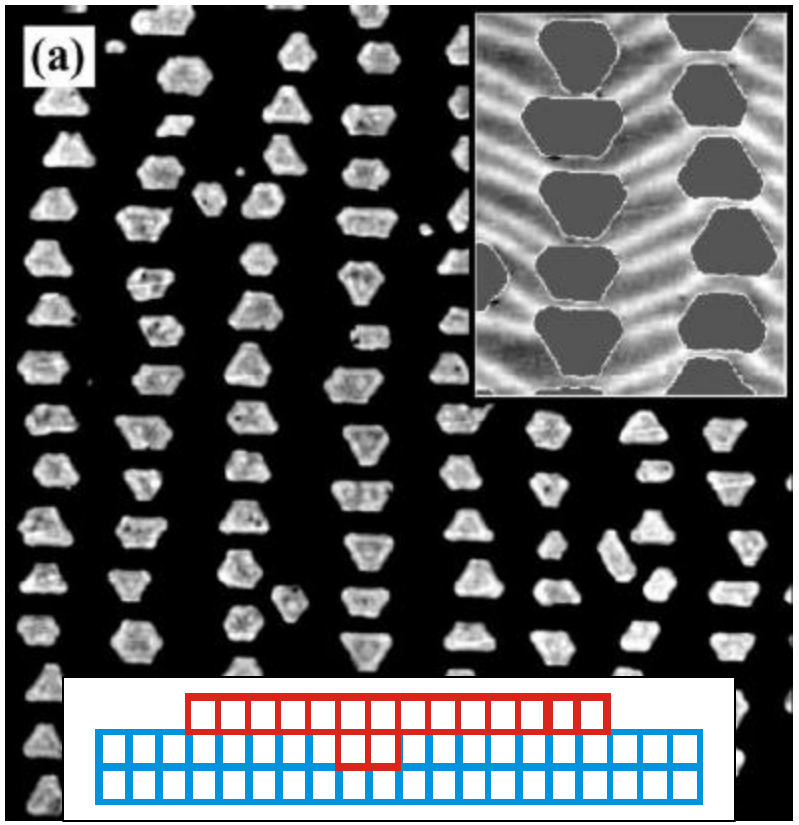
b)

0.002ML Ni@300K

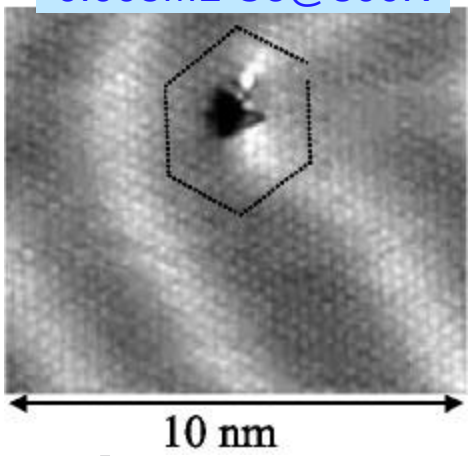


J.A.Meyer et al., Surf.Sci.**365**, L647 (1996)

0.25ML Ni@300K : 1ML-high dots



0.005ML Co@300K

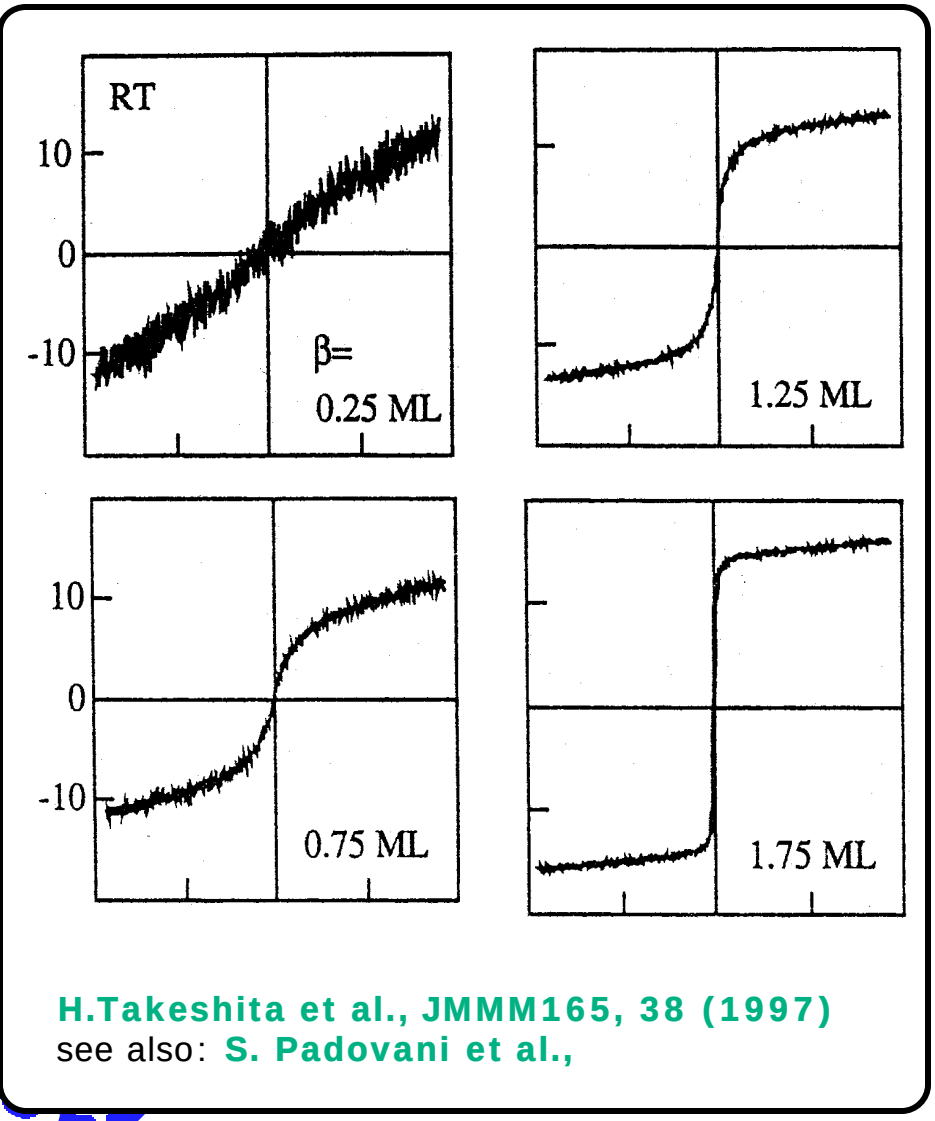


V. Repain et al., Mater. Sci. Eng., B96, 178 (2002)

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

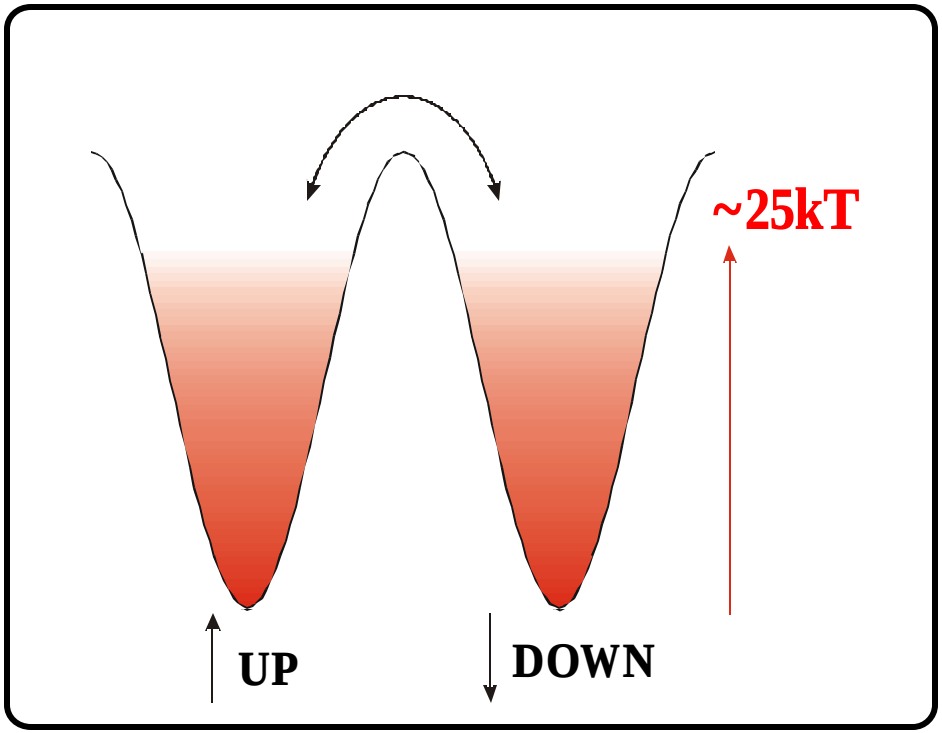
Spontaneous magnetization is perpendicular to the plane [similar to Co/Au(111) films]

Co/Au(111)



H.Takeshita et al., JMMM165, 38 (1997)
see also: S. Padovani et al.,

Anisotropy barrier ~KV



Blocking temperature Tb ~ 20K

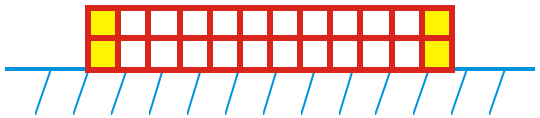
H.Dürr et al., PRB59, R701 (1999)
K. Koide et al., PRL87, 257201 (2001)
Ph. Ohresser, F. Scheurer et al., private comm.

Spin and orbital moments at films interfaces:

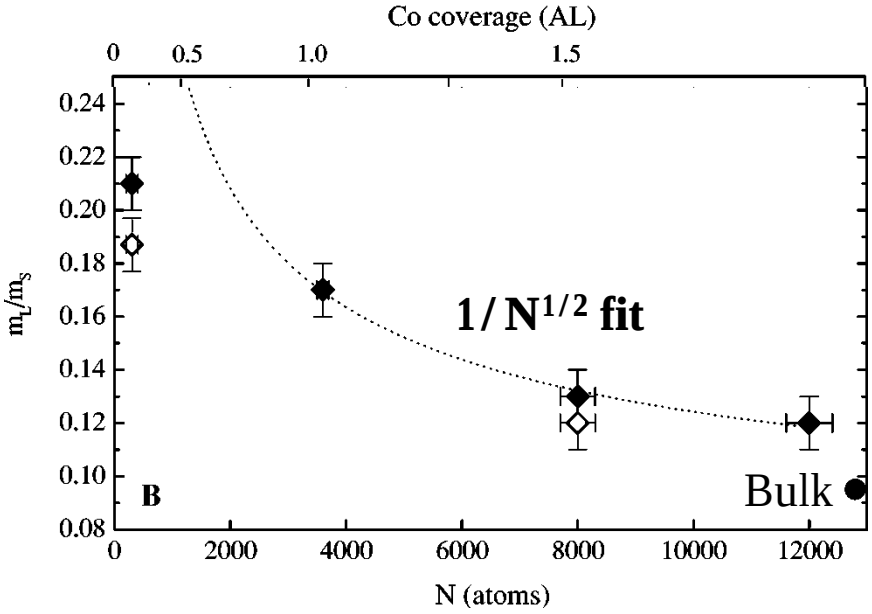
- Spin moment :
  - ↳ slightly increased because of band narrowing (up to 30%)
- Orbital moment : quenched in bulk 3d (~0). Symmetry breaking at the interface
  - ↳ Non-zero orbital moment (up to some $0.1\mu_B$/atome)

Probe: XMCD (X-ray Magnetic Circular Dichroism) and sum rules.

Expected in dots: further symmetry breaking at the dots edges: enhanced effects ?
(similarity with atomic steps)



H.Dürr et al., PRB59, R701 (1999)

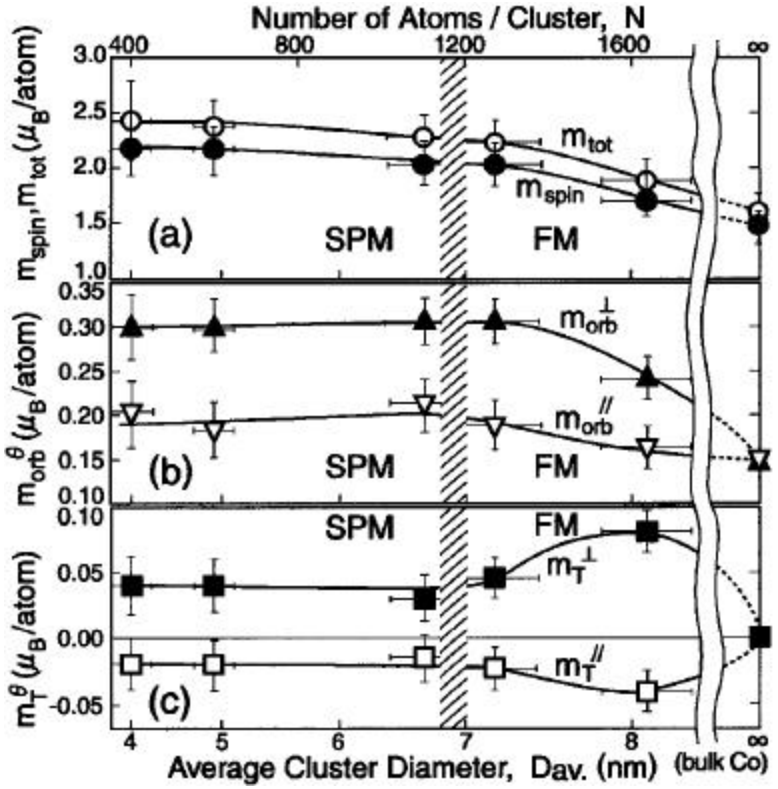


Conclusion: extra orbital $2\mu_B$/edge atome

Problems:

- dots coalescence above 1300 atoms: non-valid fit…
- Estimation of dot size by Langevin function (Brillouin ½ better suited)

K. Koide et al., PRL87, 257201 (2001)



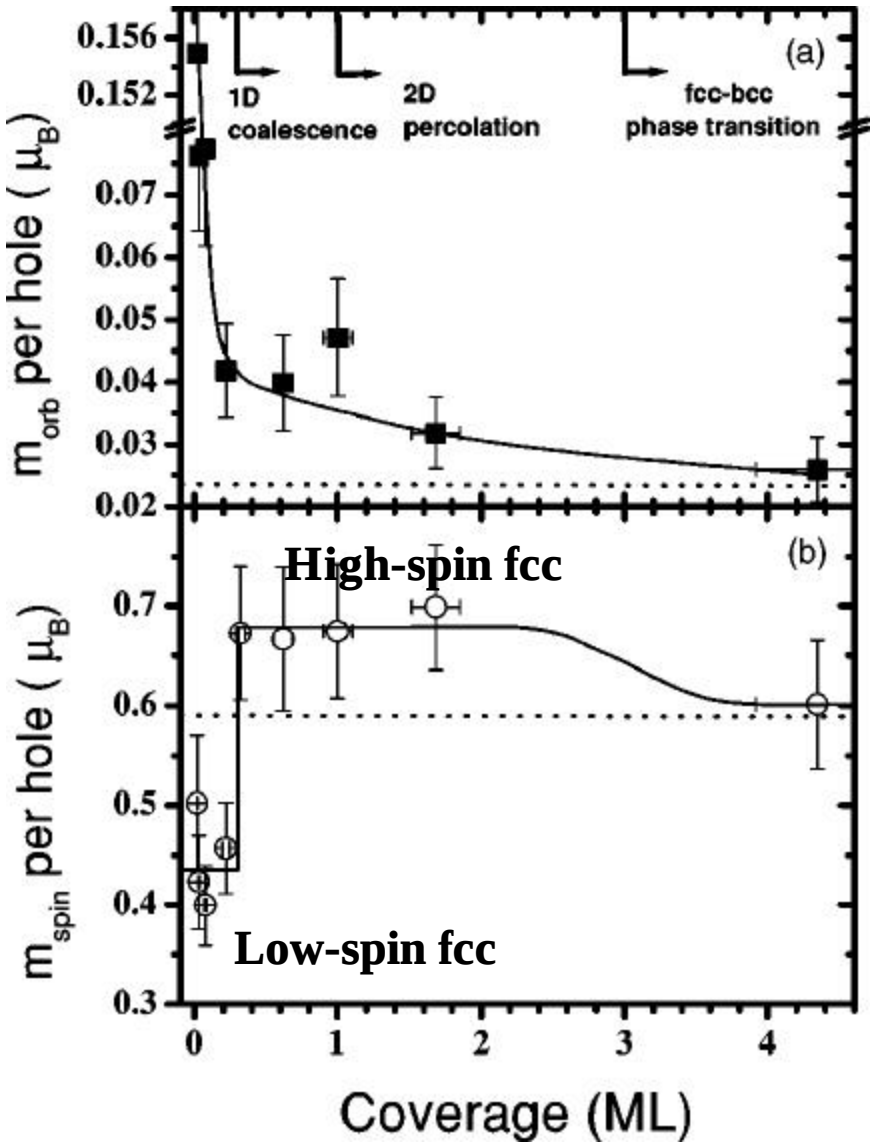
Conclusion: no extra orbital moment for edge atoms (dot='small thin film').

Problems:

- dot size is still large.

Need smaller systems !

P. Ohresser et al., PRB64, 104429 (2001)

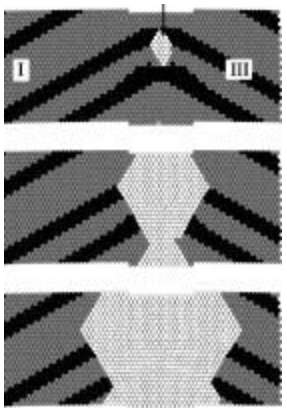


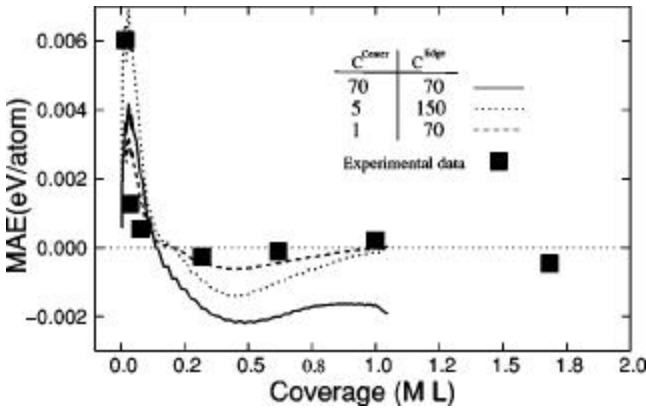
Conclusions:

- Spin moment not modified at edges (spin more influenced by deformation)
- Edge orbital moment ~ 0.5μ_B, similar to steps on vicinal Fe.
- Orbital moment anisotropy: not constant during growth.

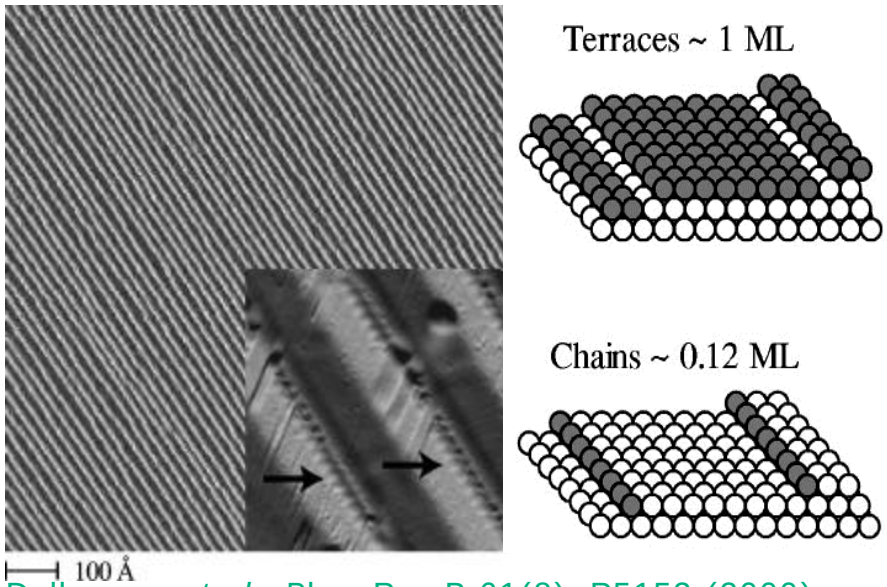
Strain-dependent interface anisotropy ?

Problem = not even solved in thin films…



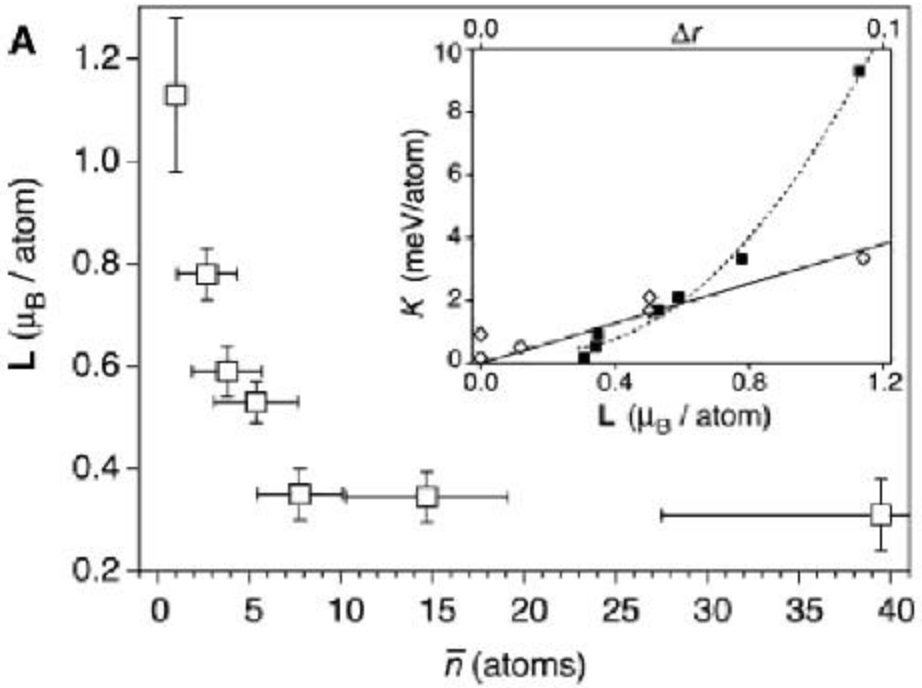


Co/Pt(997)



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

Co/Pt(111)



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

Conclusions

- Bulk: $m_L = 0.14\mu_B$/at.
- Surface: $m_L = 0.31\mu_B$/at.
- Bi-atomic wire: $m_L = 0.37\mu_B$/at.
- Mono-atomic wire: $m_L = 0.68\mu_B$/at.
- bi-atom: $m_L = 0.78\mu_B$/at.
- atom: $m_L = 1.13\mu_B$/at.

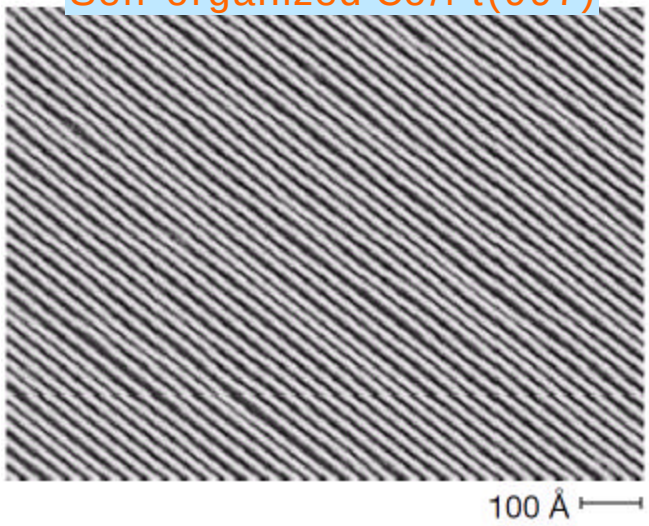
P. Gambardella et al., Nature 416, 301 (2002)

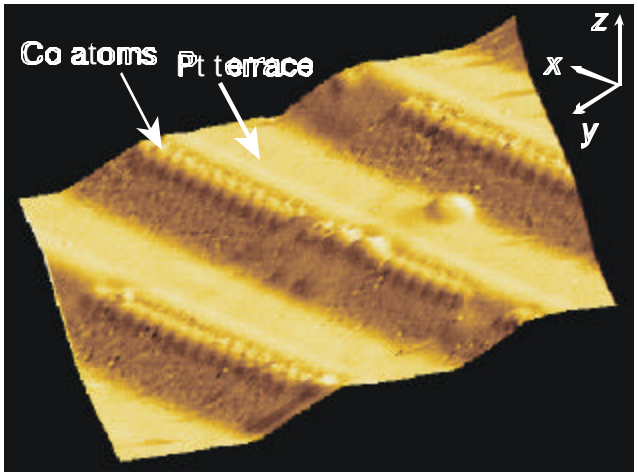
Conclusions

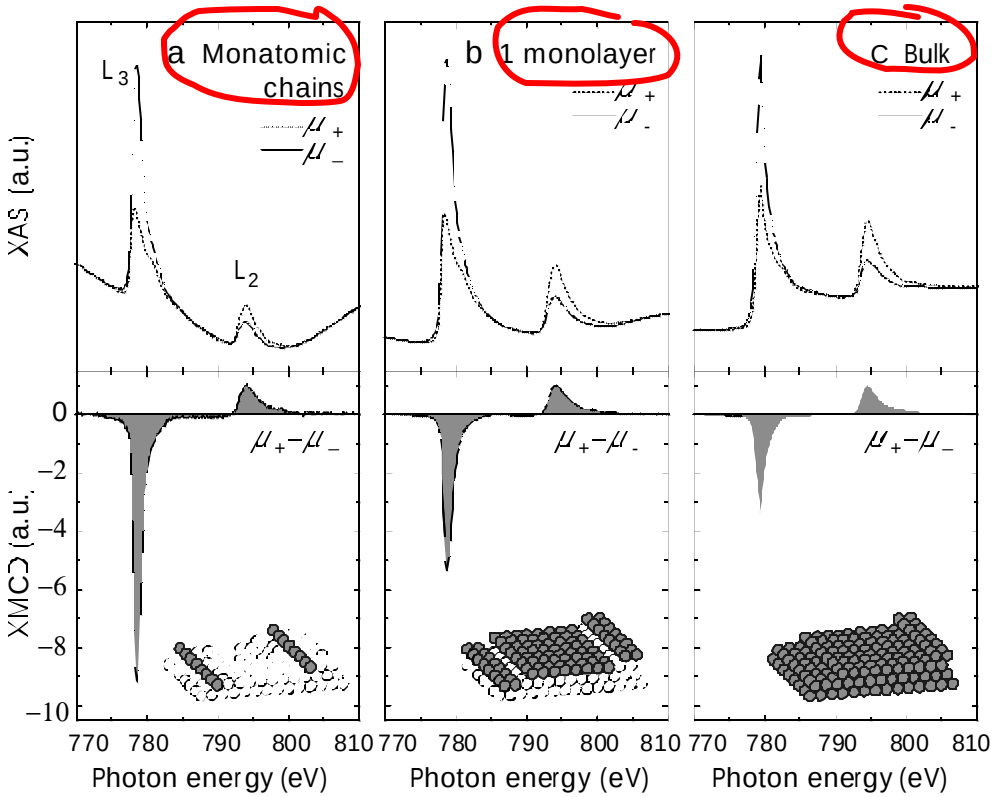
- From bulk to atoms:
considerable **increase of orbital moment**
- **2 atoms closer to wire than 1 atom**
- **bi-atomic wire closer to surface than wire**

From surface to wires (1D)

Self-organized Co/Pt(997)







$$m_L \approx \int L_3 + \int L_2$$

$$m_s^{eff} \approx -\int 2L_3 + 4\int L_2$$

Conclusion:

- Increase of orbital moment (necessary condition for anisotropy)
- Anisotropy of orbital moment?

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

From surface to wires (1D)

Method

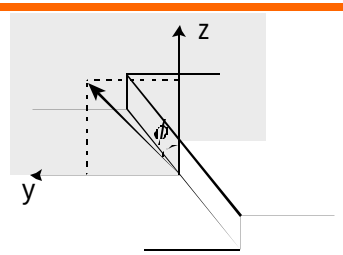
- XMCD > Orbital moment
- Fit magnetization curves > Anisotropy functional

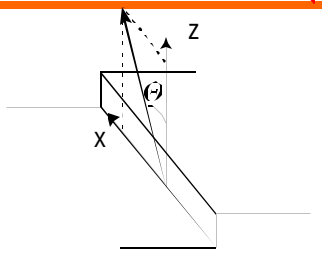
MAE

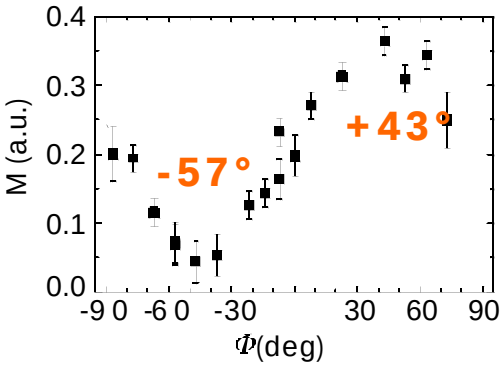
- Bulk Co: 40μeV/atom
- Co ML: 140μeV/atom
- Co bi-wire: 0.34meV/atom
- Co wire: 2meV/atom

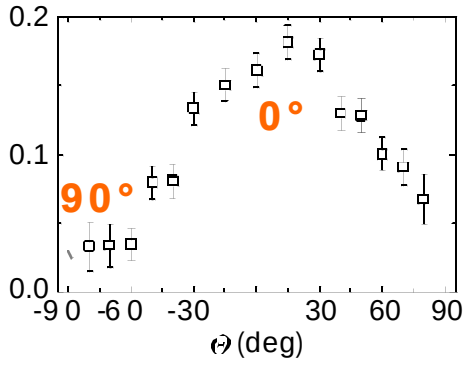
Conclusions:

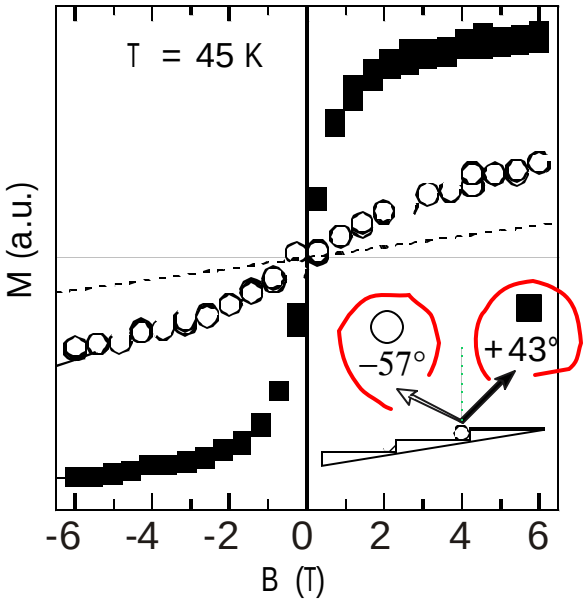
- Easy axis of magnetization perpendicular to the wires, but not the the mean film surface, nor to Pt(111)
- See anisotropy of orbital moment on the saturation XMCD.

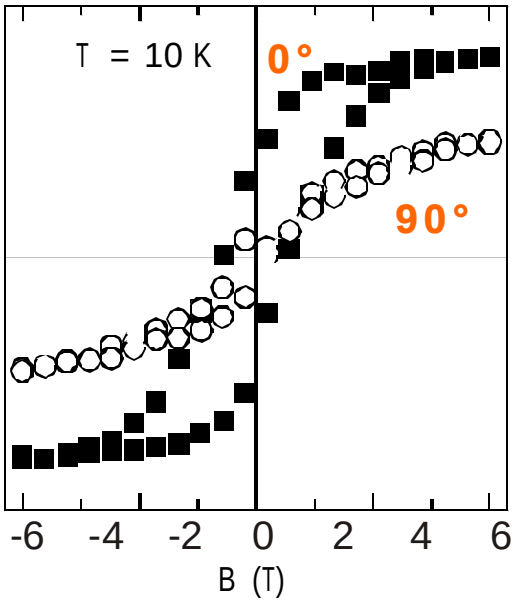












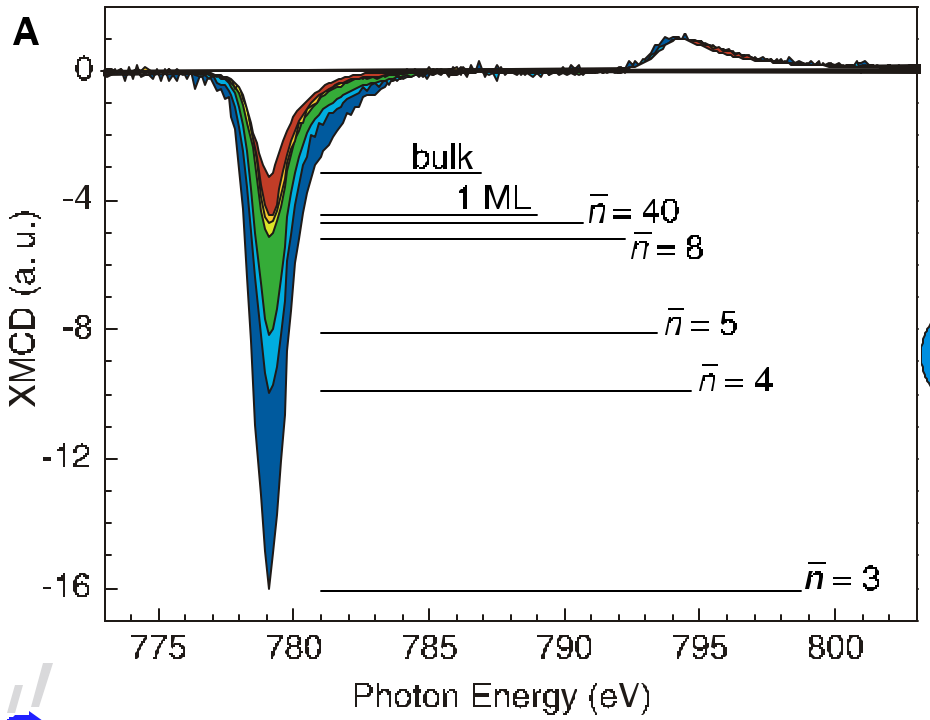
From surface to atoms (0D)

Co/Pt(111)

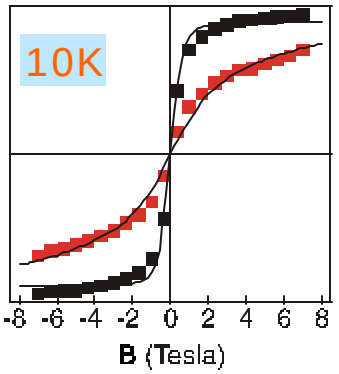
Giant Magnetic Anisotropy of Single Cobalt Atoms and Nanoparticles

P. Gambardella,[1,2*] S. Rusponi,[1,2] M. Veronese,[3] S. S. Dhesi,[4†]
C. Grazioli,[3] A. Dallmeyer,[5] I. Cabria,[5] R. Zeller,[5]
P. H. Dederichs,[5] K. Kern,[1,2] C. Carbone,[3,5] H. Brune[1]

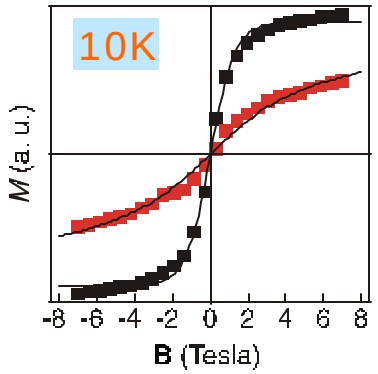
P. Gambardella et al., Science 300, 1130 (2003)



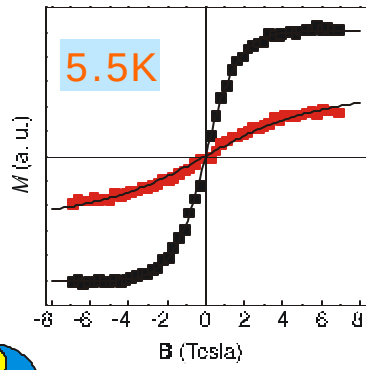
8 atoms



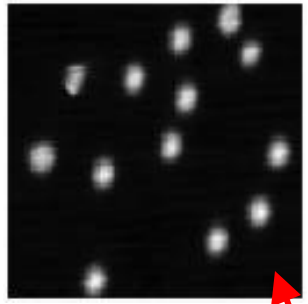
4 atoms



1 atom



STM, 8.5nm, 5.5K



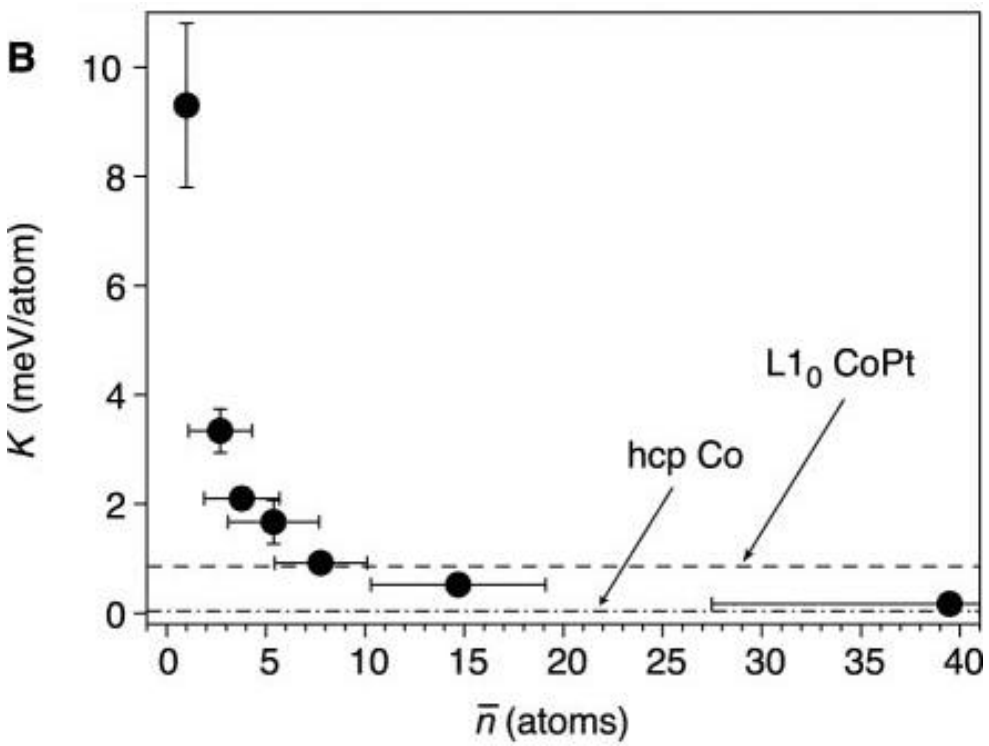
Cf question by Dominique GIVORD

Qualitatively:

- Easy axis of magnetization perpendicular to Pt(111)
- See anisotropy of orbital moment on the saturation XMCD.

From surface to atoms (0D)

Co/Pt(111)

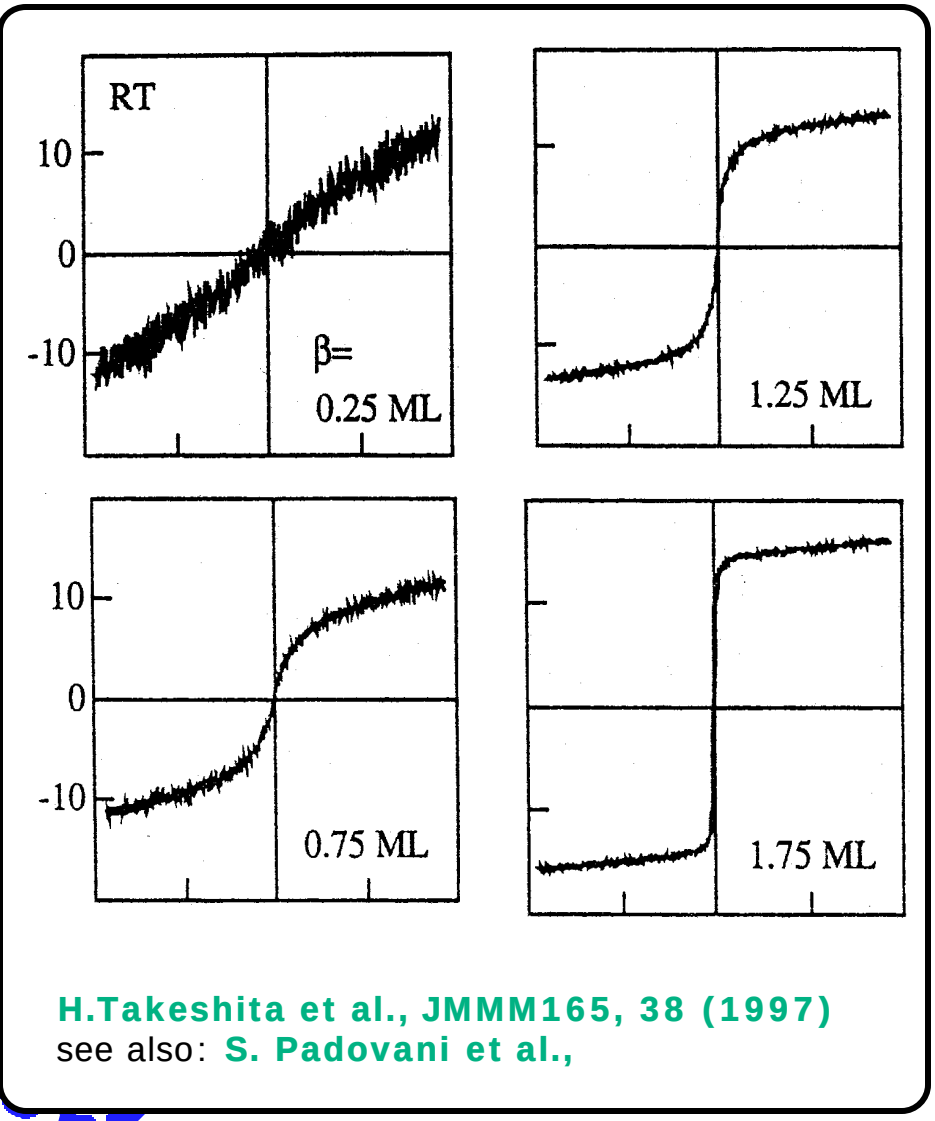


MAE

- Bulk Co: 40μeV/atom
- Co ML: 140μeV/atom
- Co bi-wire: 0.34meV/atom
- Co wire: 2meV/atom
- Co bi-atom: 3.4meV/atom
- Co atom: 9.2meV/atom

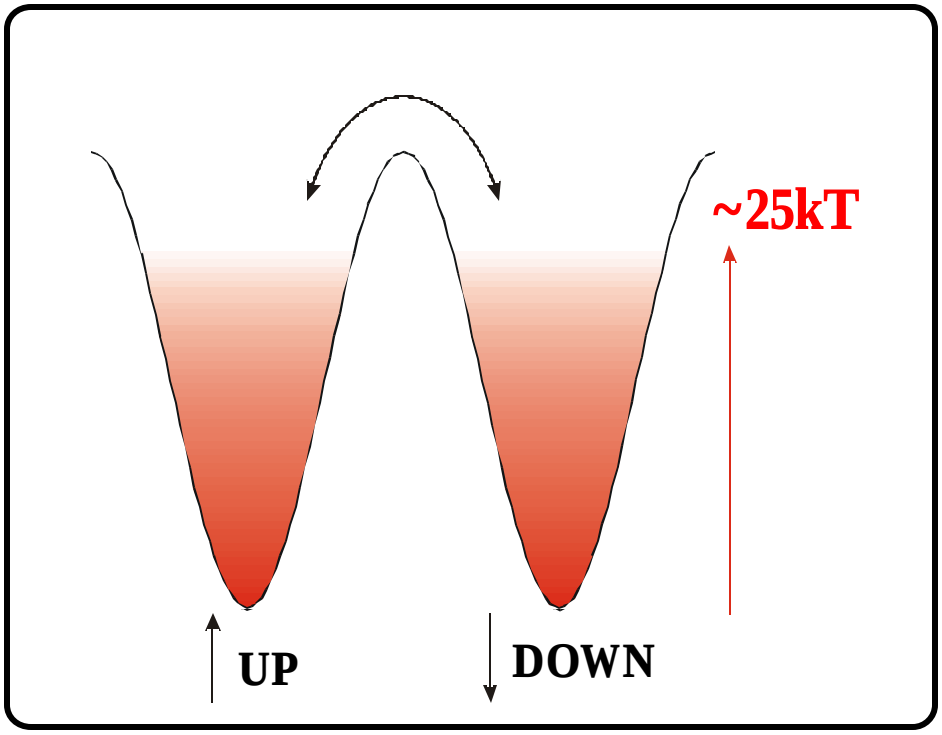
Spontaneous magnetization is perpendicular to the plane [similar to Co/Au(111) films]

Co/Au(111)



H.Takeshita et al., JMMM165, 38 (1997)
see also: S. Padovani et al.,

Anisotropy barrier ~KV



Blocking temperature Tb ~ 20K

H.Dürr et al., PRB59, R701 (1999)
K. Koide et al., PRL87, 257201 (2001)
Ph. Ohresser, F. Scheurer et al., private comm.

$T_b = KV / 25k_B$

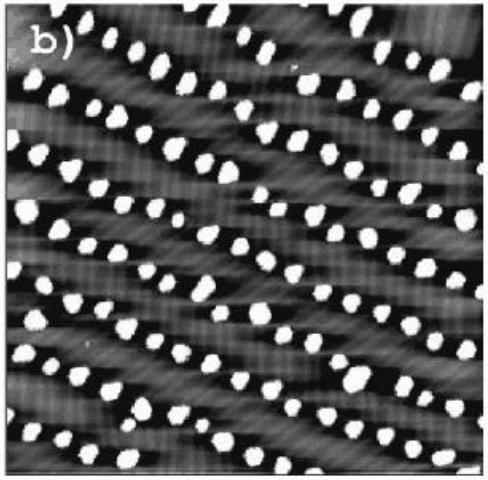
TWO ROUTES TO OVERCOME SUPERPARAMAGNETISM in SO

- Increase *K*. Problem: *K* does not increase as fast as *V* decreases

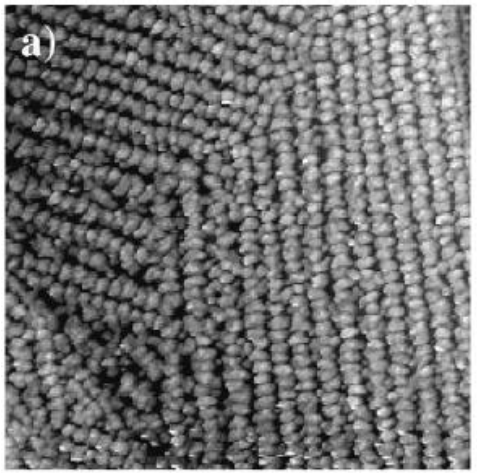
- Increase *V*. Problem : lateral coalescence occurs

Co/Au(111)

0.25AL

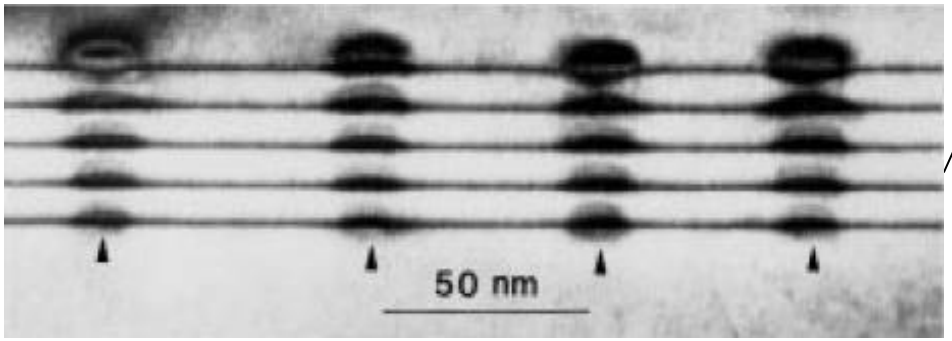


1.75AL

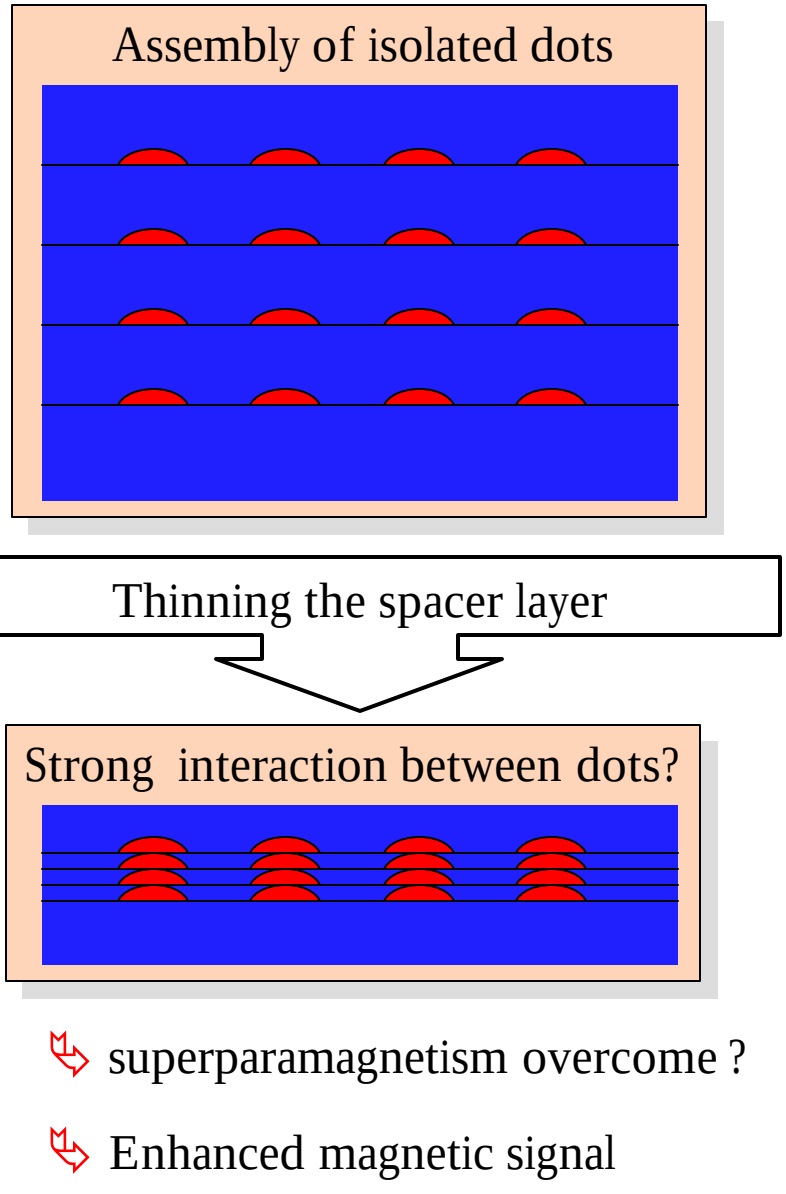


INCREASE THE HEIGHT OF NANOSTRUCTURES ?

- Vertical 3D self-organization of $In_xGa_{1-x}As/GaAs$:

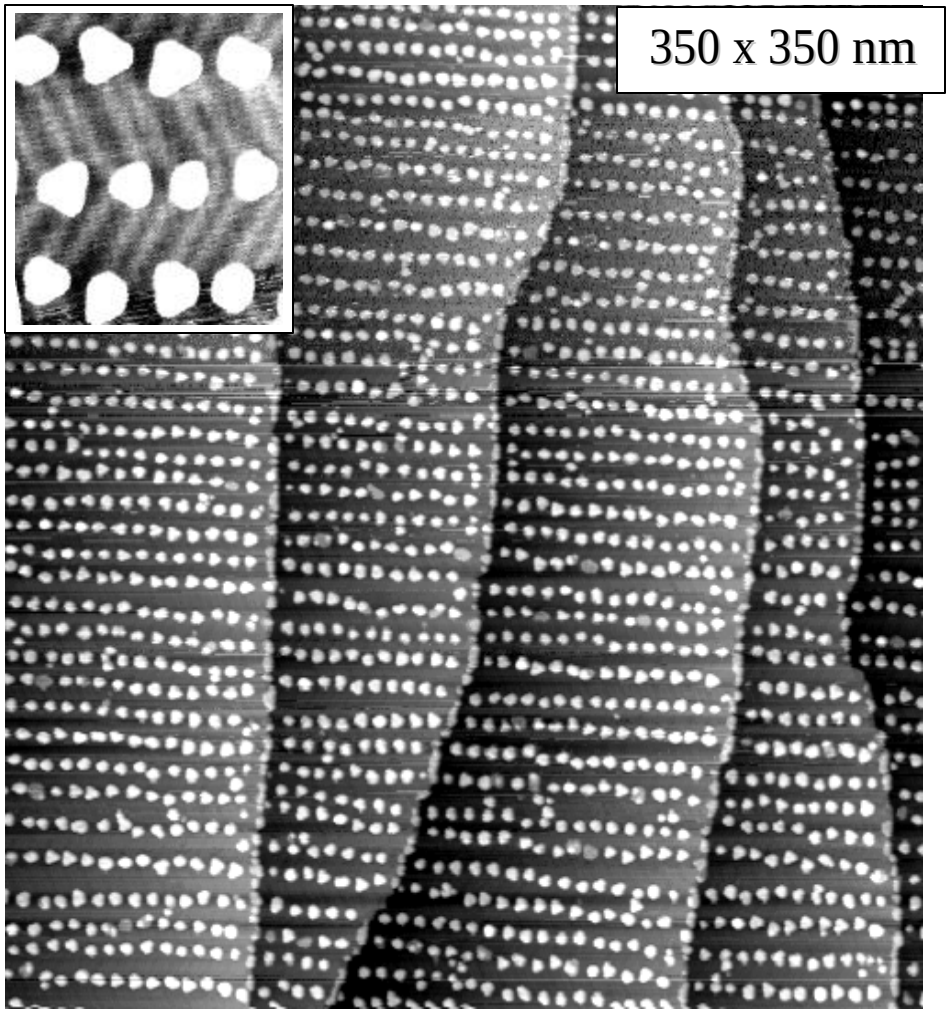


Q.Xie et al., Phys.Rev.Lett.75(13), 2542 (1995)

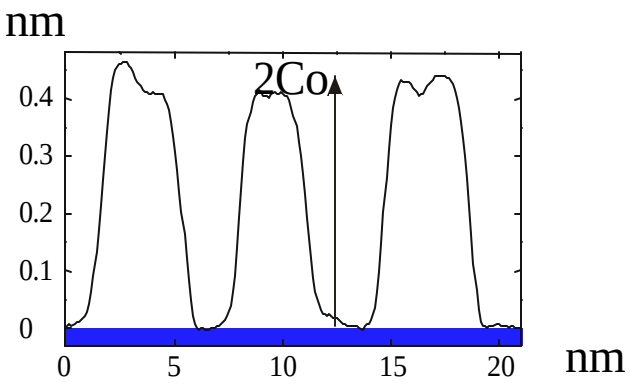


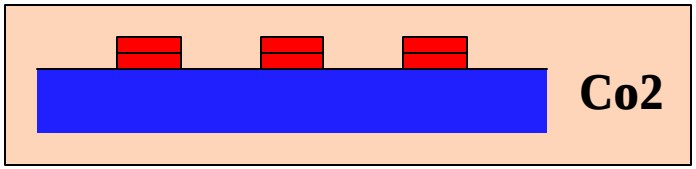
- superparamagnetism overcome ?
- Enhanced magnetic signal

➢ **Step 1** : 0.2AL Co @300K

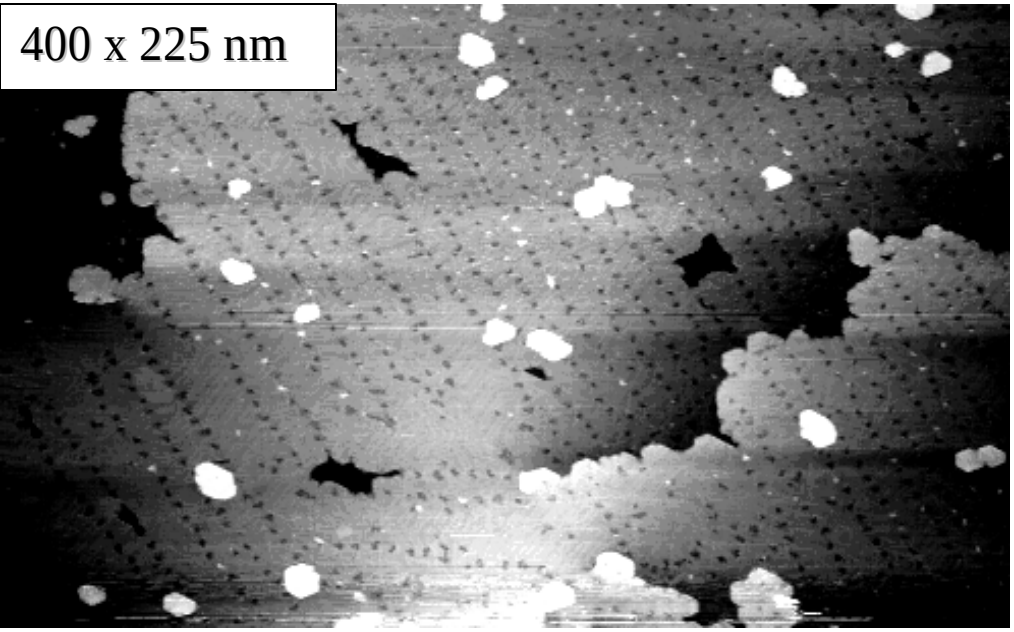


Typical cross-section :





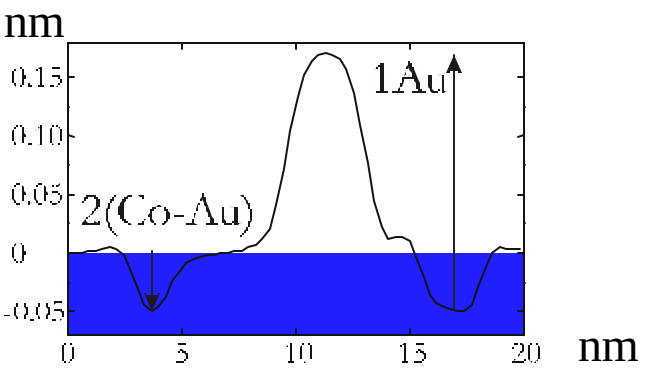
➢ **Step 2** : 3.8AL Au @375➚410K

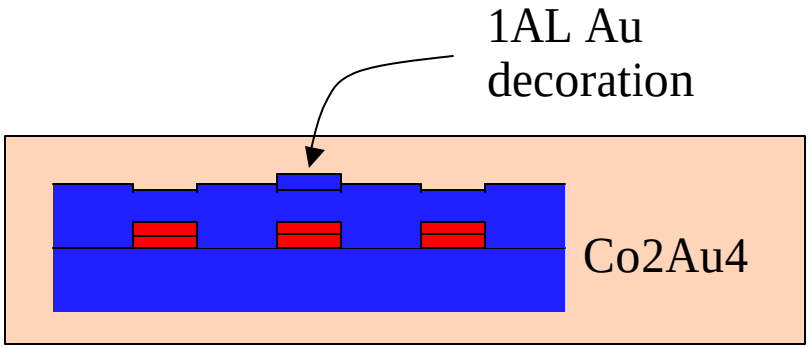


See also : **Wollschläger *et al.*, Surf.Sci.277, 1 (1992)**

1AL Co hcp ≈ 0.205nm
1AL Au fcc ≈ 0.235nm
} Array of hollows

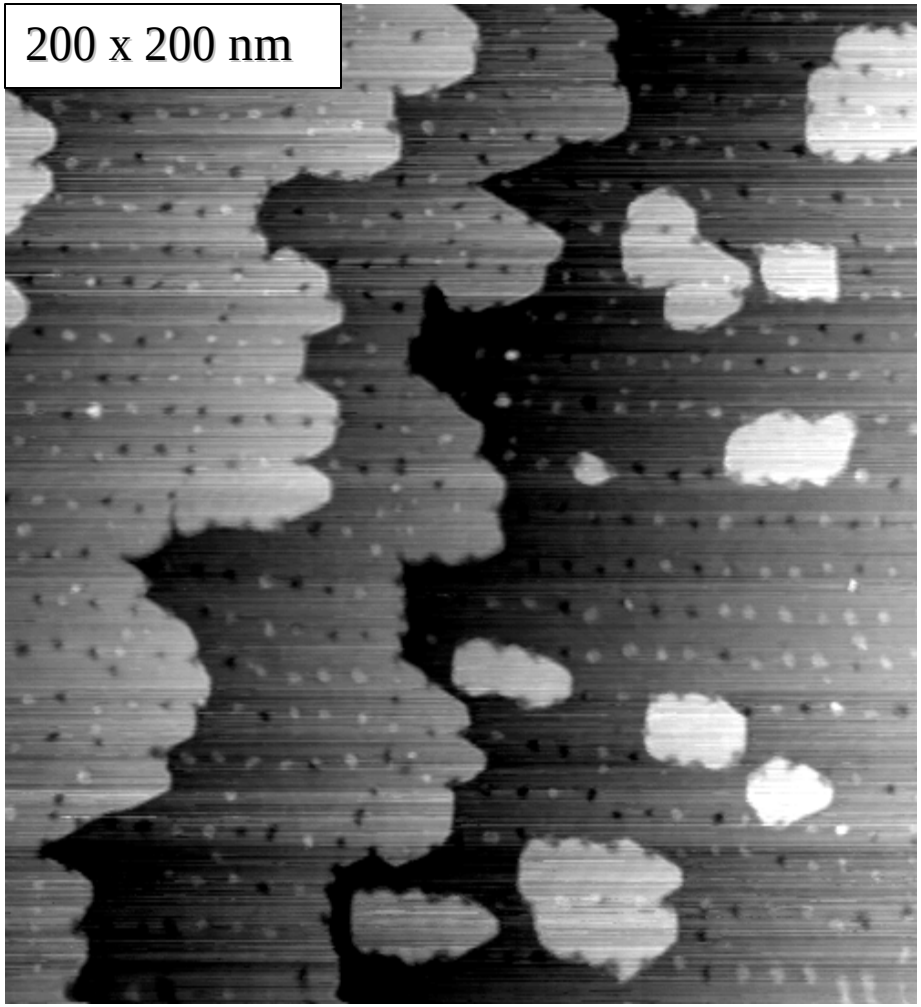
Typical cross-section :



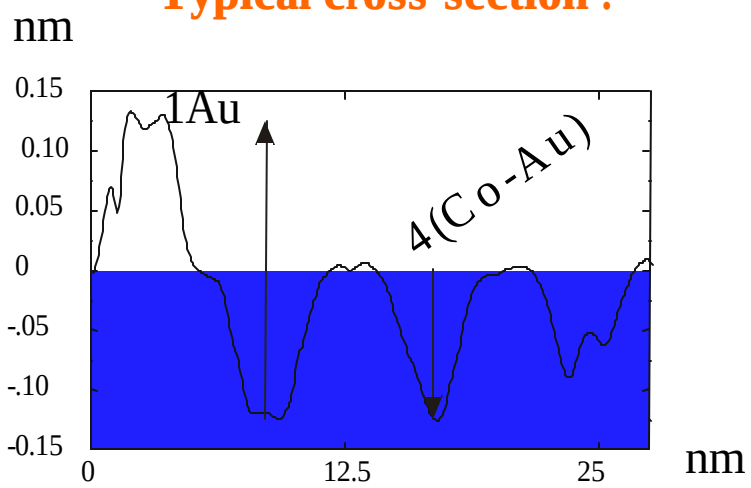


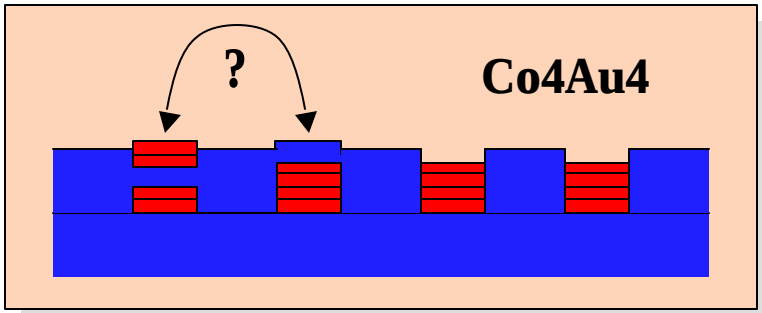
- Surface smoothing
- Unaffected Co dots

➢ **Step 3** : 0.1AL Co @500K

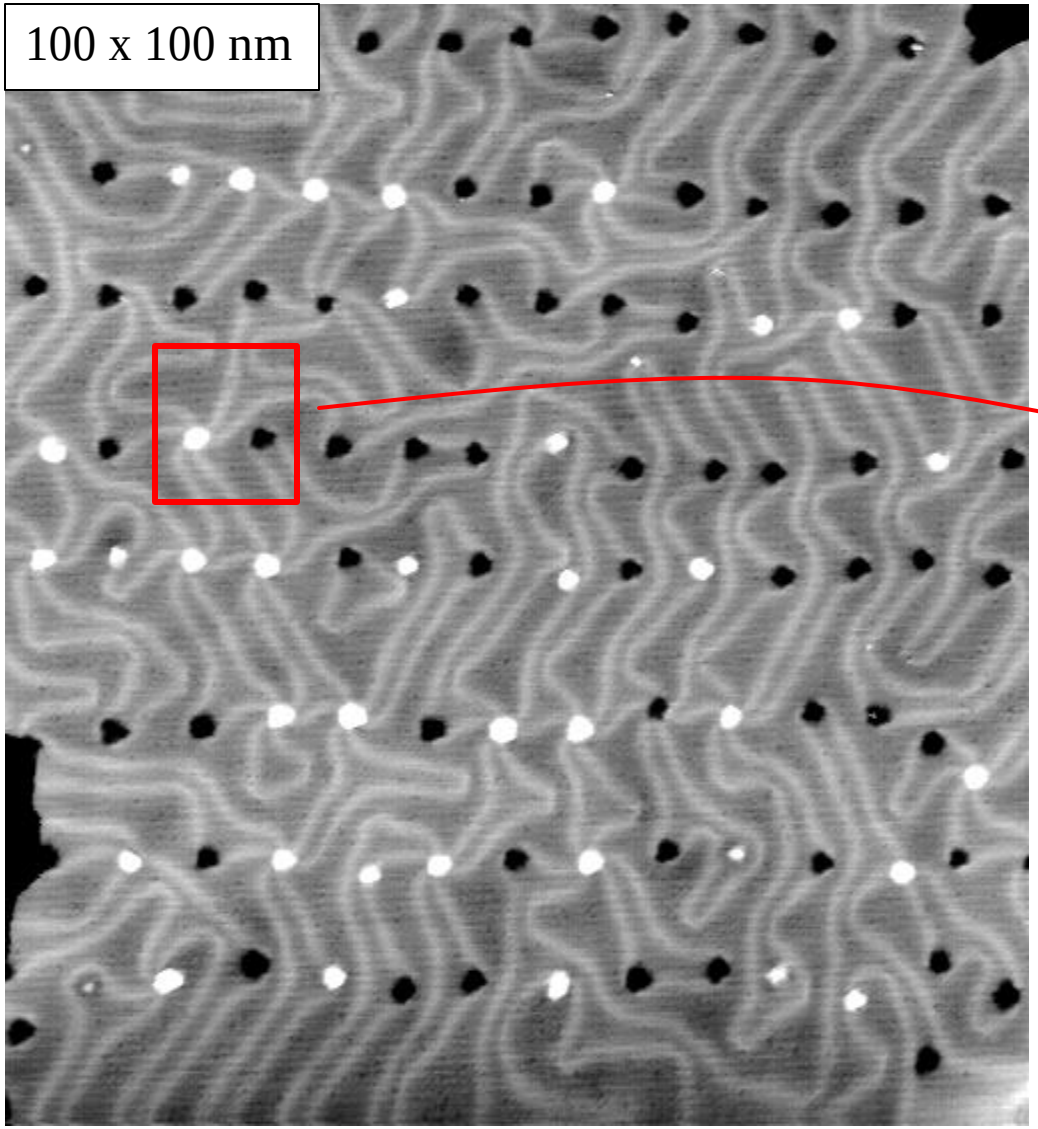


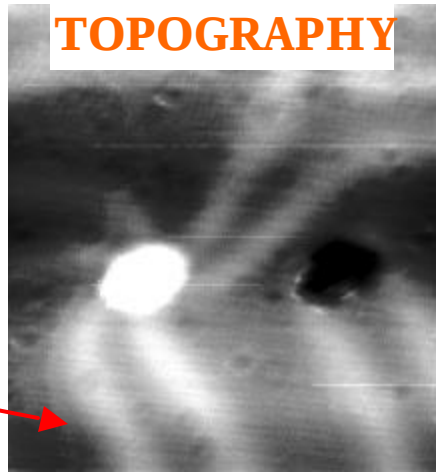
Typical cross-section :

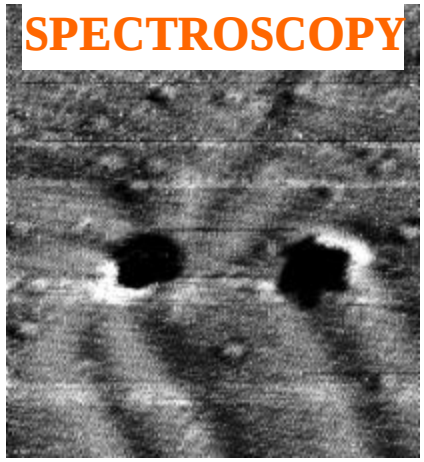




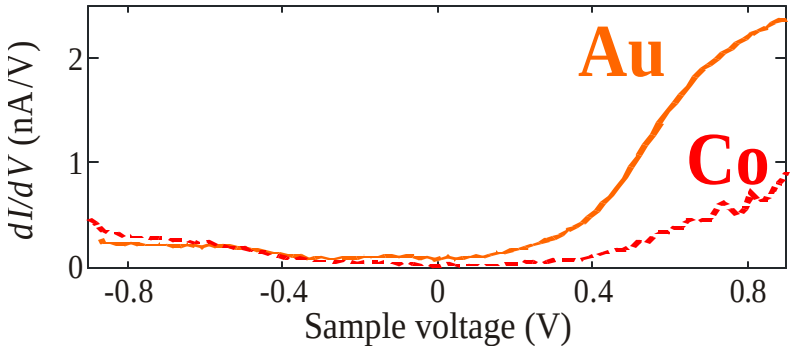
- Co/Au exchange mechanism
- The dots are now **4ML high**







SAME AREA



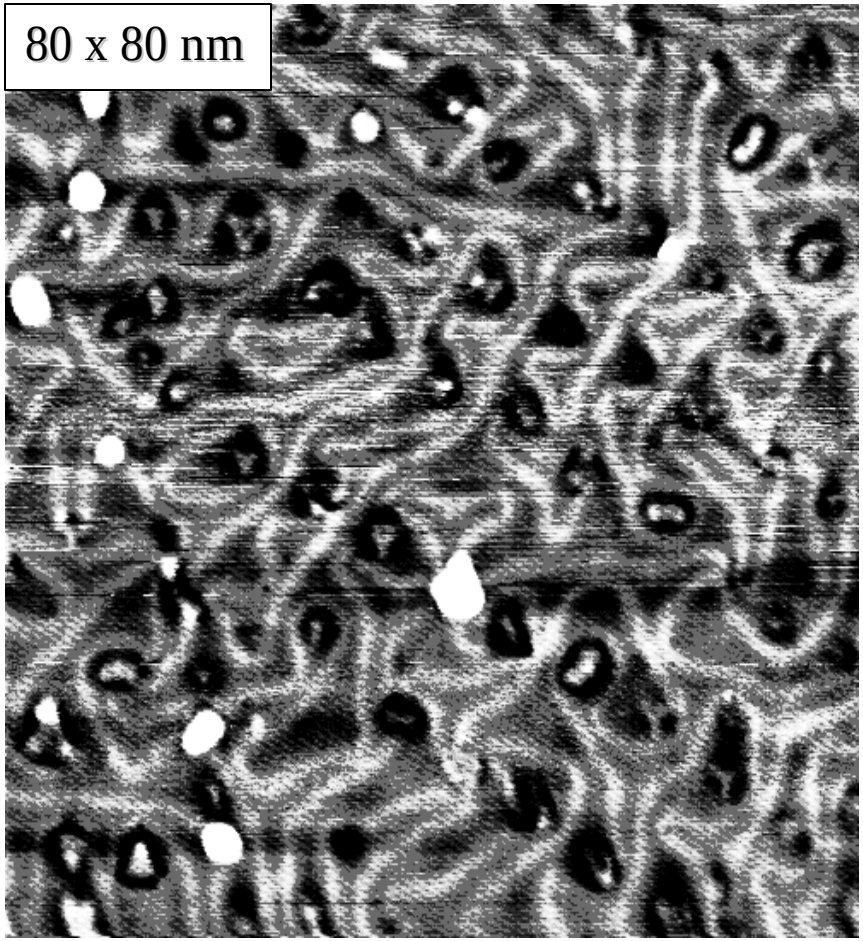
Co atoms grow only on existing dots

↳ **VERTICAL SELF-ORGANIZATION**

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

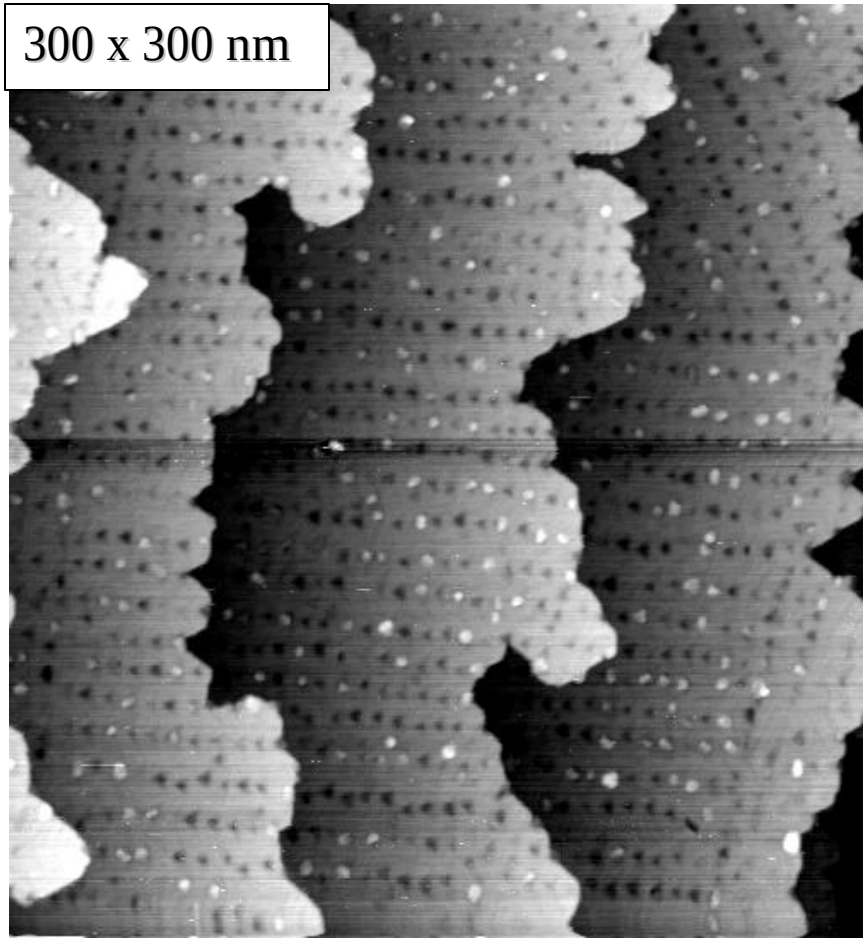
Co6Au6

80 x 80 nm

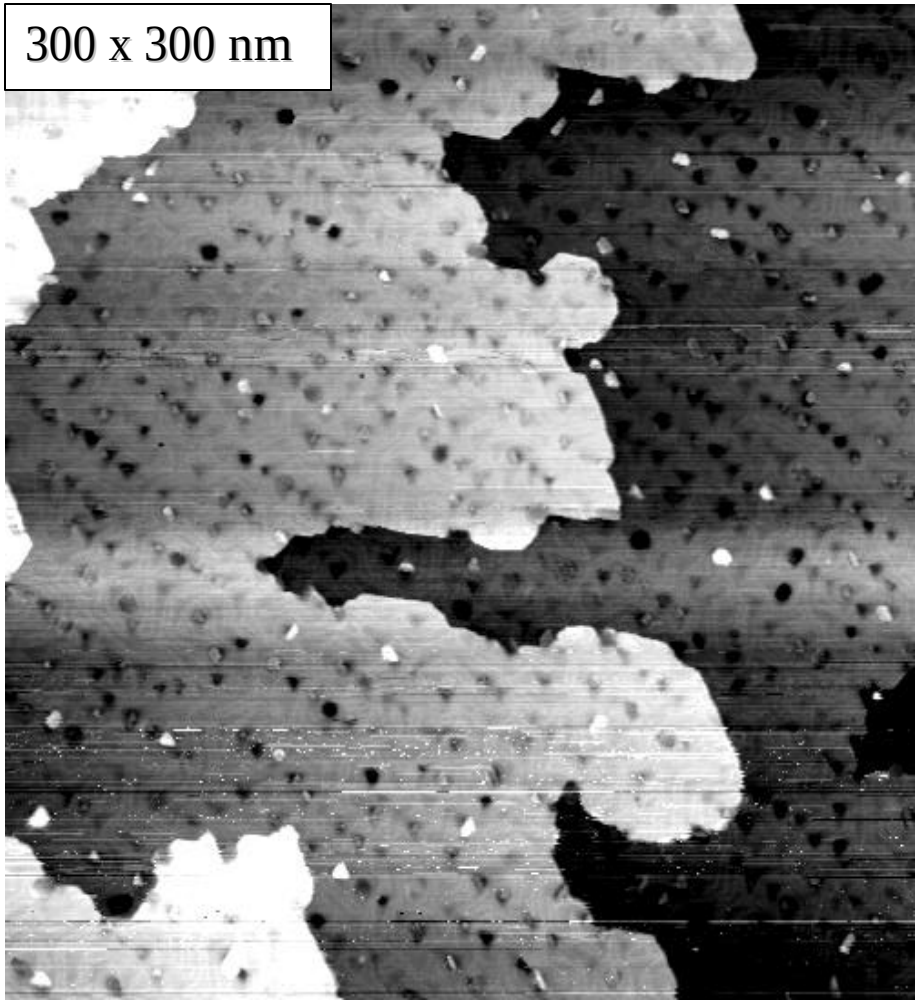


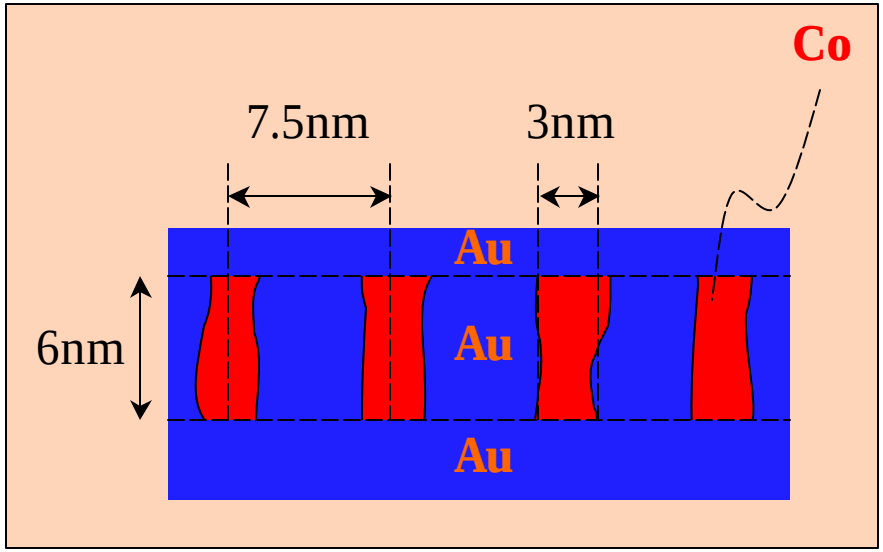
Co10Au10

300 x 300 nm



Co pillars of height 20ML





- Self-organization nearly undisturbed
- Pillars with 2:1 vertical aspect ratio
- Unclear to this point :
  - Exchange mechanism
  - Limitating factors ?
  - Composition, microstructure ?

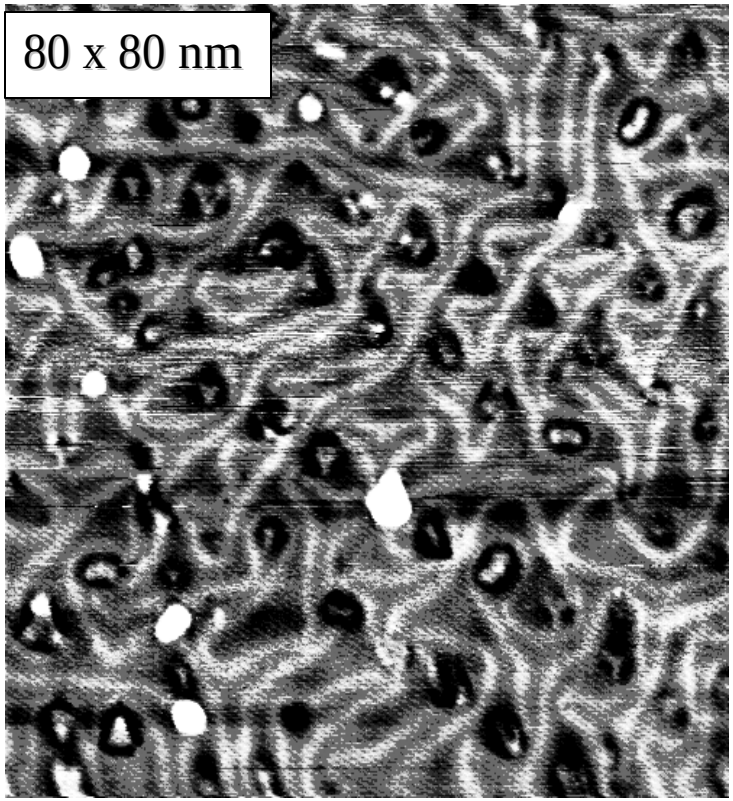
- **Sequential deposition process** : from flat dots to vertical pillars.
  - **vertical replication** of the sub-AL flat pattern
  - Partial control of the **pillars geometry**:
    lateral size (% of AL per cycle), height (number of cycles).

- **Requirements on materials** : lattice parameter mismatch, surface energies ?
  - **May be OK for other elements** as well.

- **Open questions** : composition, microstructure, destabilizing factors.

O. Fruchart *et al.*, Phys. Rev. Lett. 23 (14), 2769 (1999)

O. Fruchart *et al.*, Appl. Surf. Science 162-163, 529 (2000)



Facts

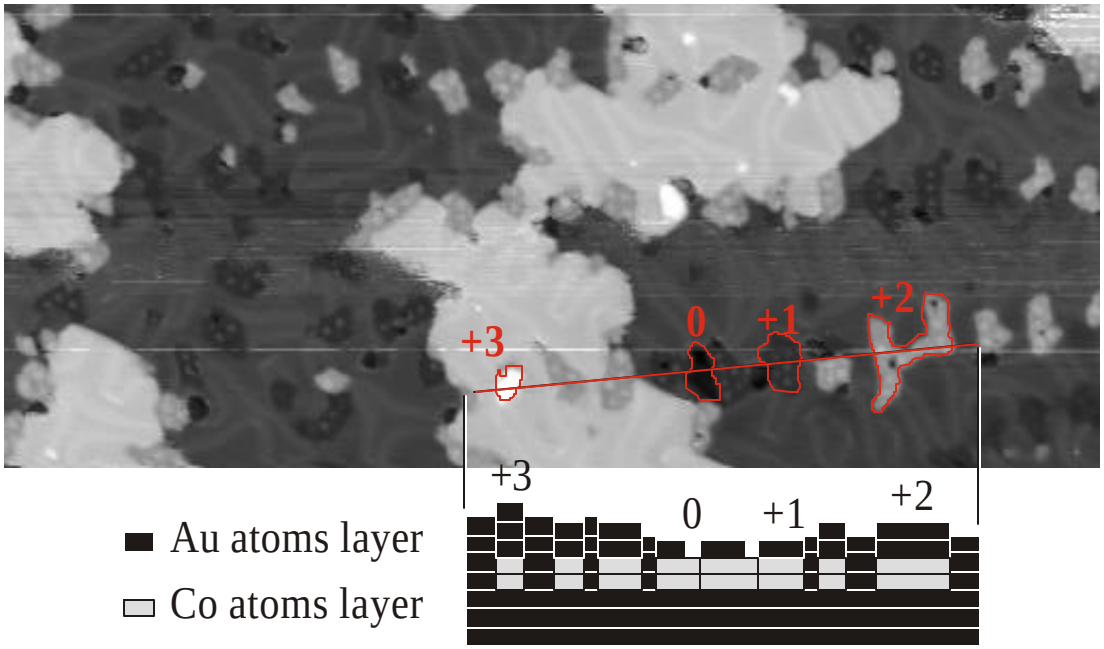
- **First Co layer** : nucleation on reconstructions
- **Further layers**: reconstructions disturbed

Questions

- What drives nucleation over buried dots ?
- Why no nucleation on reconstructions ?

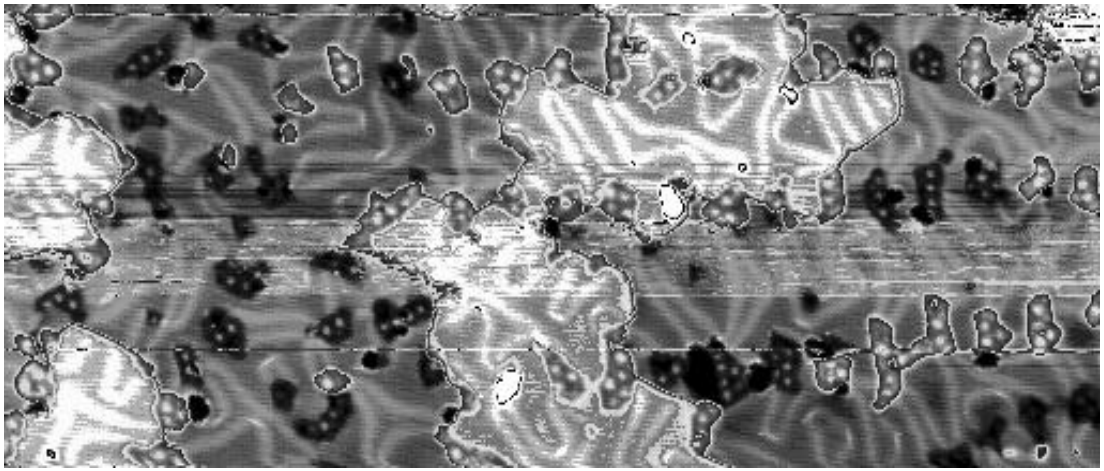
Understanding

- **Driving parameters**: lattice parameter mismatch, immiscibility, surface energy.
- Nucleation on reconstructions still possible, but energetically less favorable.

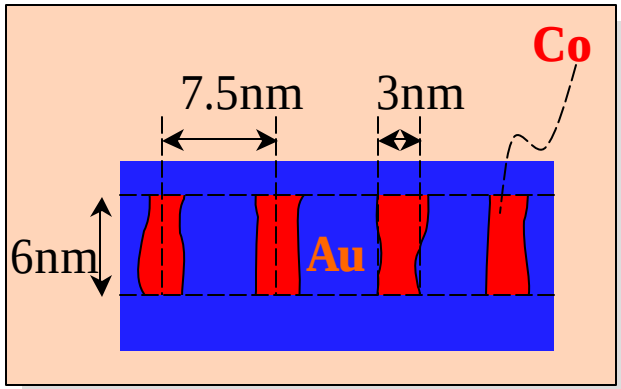


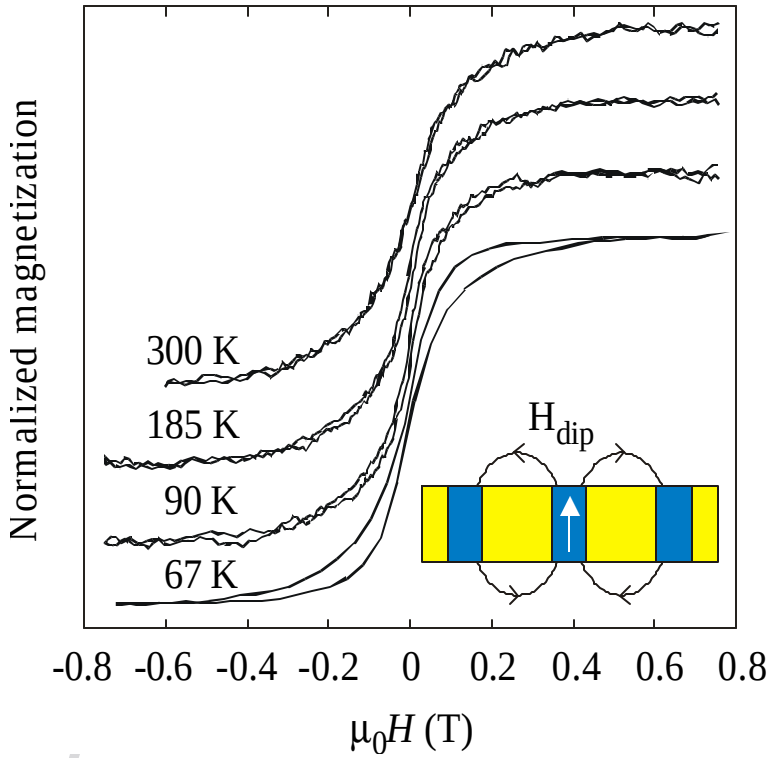
Au(2.5AL) / Co(0.65AL) / Au(111)

- **Au dislocations on Co**
  - Au easily expelled by incoming Co atoms
  - Role of lattice parameter mismatch

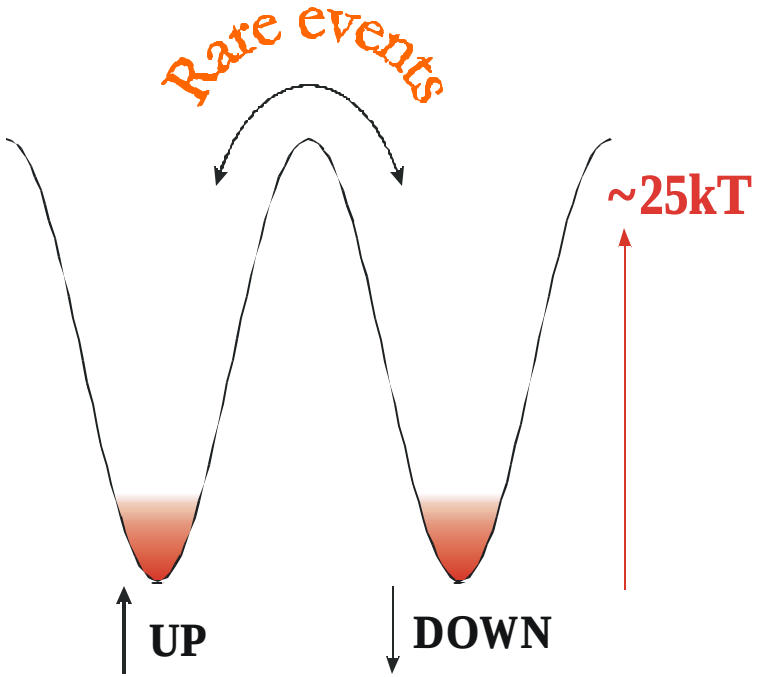


O. Fruchart *et al.*, J. Cryst. Growth 237-239, p. 2035 (2002) ;
Proceedings of ICCG13 / ICVGE11.





- **Magnetization essentially perpendicular**
  - 2 states: up and down (Ising macrospin)
  - Superparamagnetism fitted using Brillouin 1/2 function



Classical spin

Uniaxial anisotropy

H // anisotropy axis

$$bE = -dm^2 - hm$$

$$d = bK \qquad \text{Anisotropy}$$

$$K = K_V \times v \qquad \text{Anisotropy}$$

$$h = bm_0 mH \qquad \text{Zeeman}$$

Exact solution

Partition function $$Z = \int_{-1}^{1} \exp(dm^2 + hm)\mathrm{d}m$$

Obstacle (?) $$\int_0^t \exp(x^2)\mathrm{d}x = ?$$ ⟸ Imaginary Error function, Erfi(t)

Magnetization $$m = -h/2d + (2/\sqrt{p\,d}) \times \frac{\exp(d + h^2/4d)\sinh(h)}{\mathrm{Erfi}(\sqrt{d} + h/2\sqrt{d}) + \mathrm{Erfi}(\sqrt{d} - h/2\sqrt{d})}$$

Zero field susceptibility $$c = -1/2d + \exp(d)/(\sqrt{p\,d}\,\mathrm{Erfi}\sqrt{d})$$

Asymptotic behavior

High temperature $c = 1/3 + 4d/45$ Langevin-like

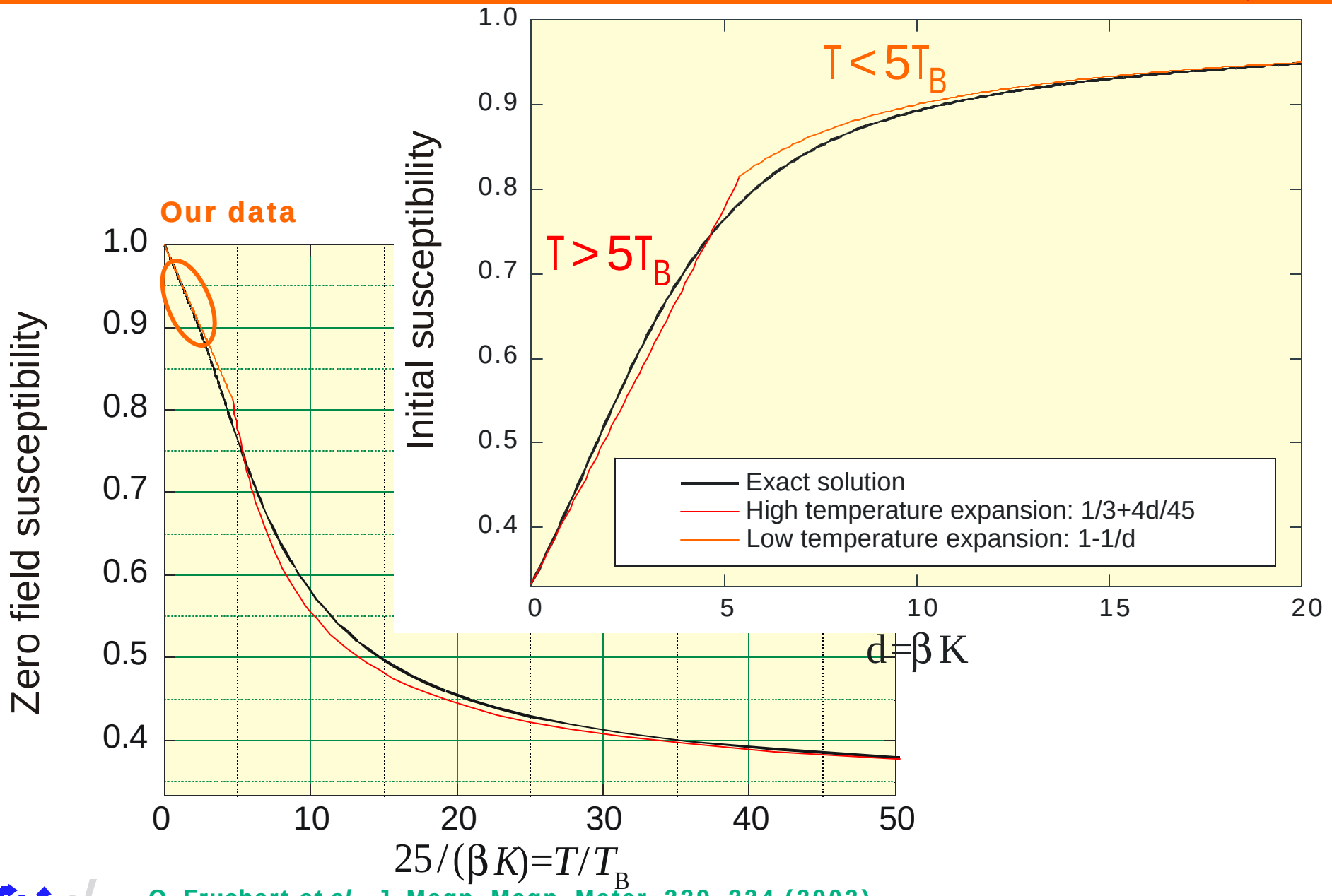
Low temperature $c = 1 - 1/d$ Brillouin ½ -like

Blocking temperature

$$T_B = K / 25 k_B$$

$$t = t_0 \exp(b_B E)$$

1s 10^{-9} – 10^{-12} s



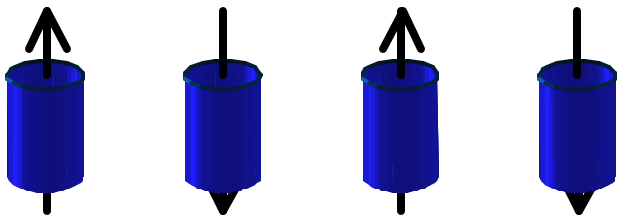
O. Fruchart *et al.*, J. Magn. Magn. Mater. 239, 224 (2002)

- **Brillouin 1/2 function**

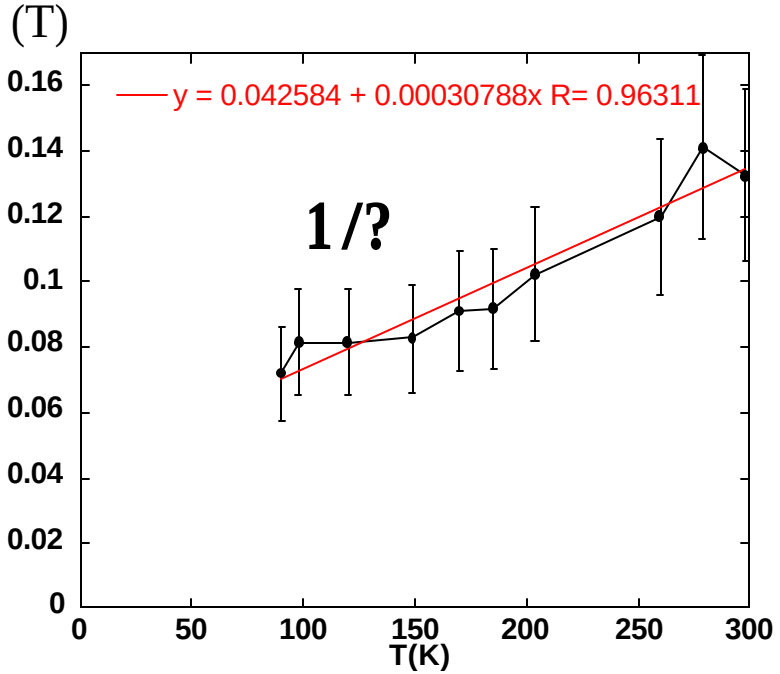
$$m = B_{1/2}\left(\mu_0 \mu_{Co} N H_{eff.} / kT\right)$$

- **Effective field**

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



(Demagnetizing dipolar interactions)



- **First order expansion**: susceptibility

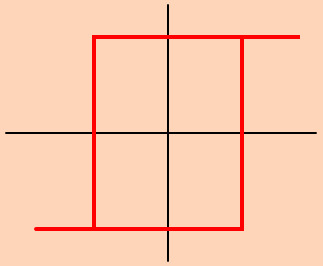
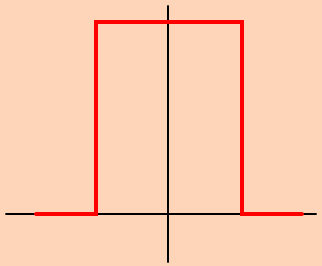
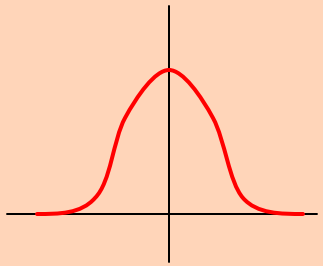
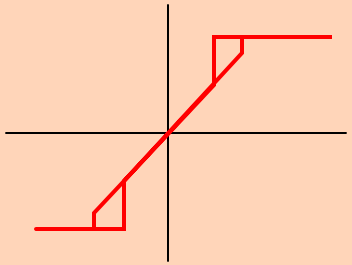
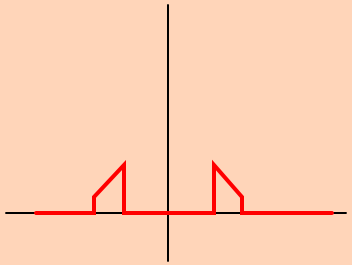
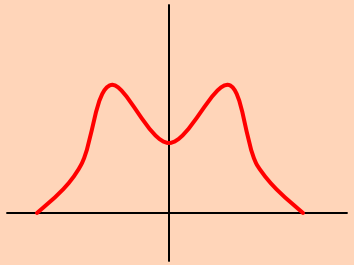
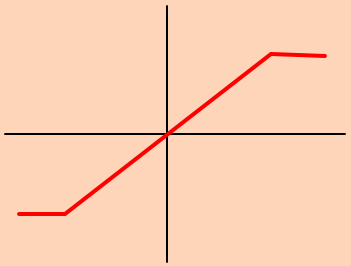
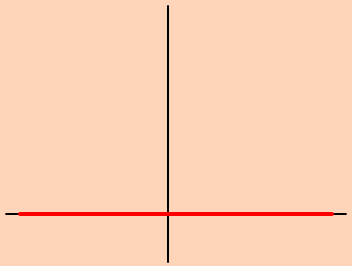
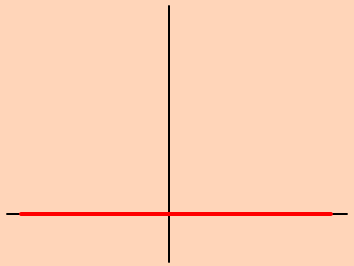
$$\frac{d(\mu_0 H)}{dm} = \frac{1}{?} = -\mu_0 M_S r + \frac{k}{\mu_{Co} N} T$$

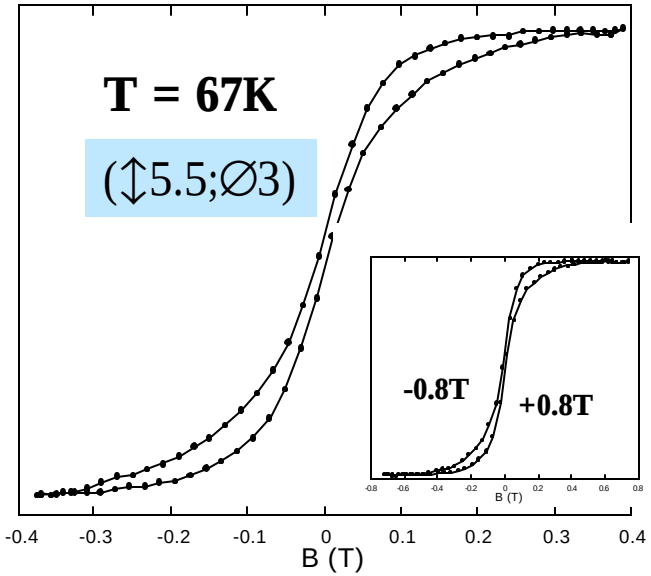
$$a + b \,.\, T$$

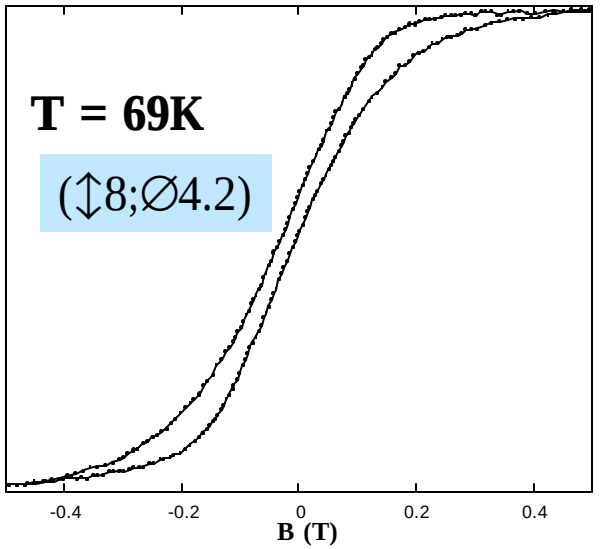
Deduced from STM	**... from magnetism**
N = **3300** atoms	N = **2800** atoms
Hdip = **-32** mT	Hdip = **-42** mT

- Good quantitative agreement ⇨ 1 pillar = 1 magnetic entity

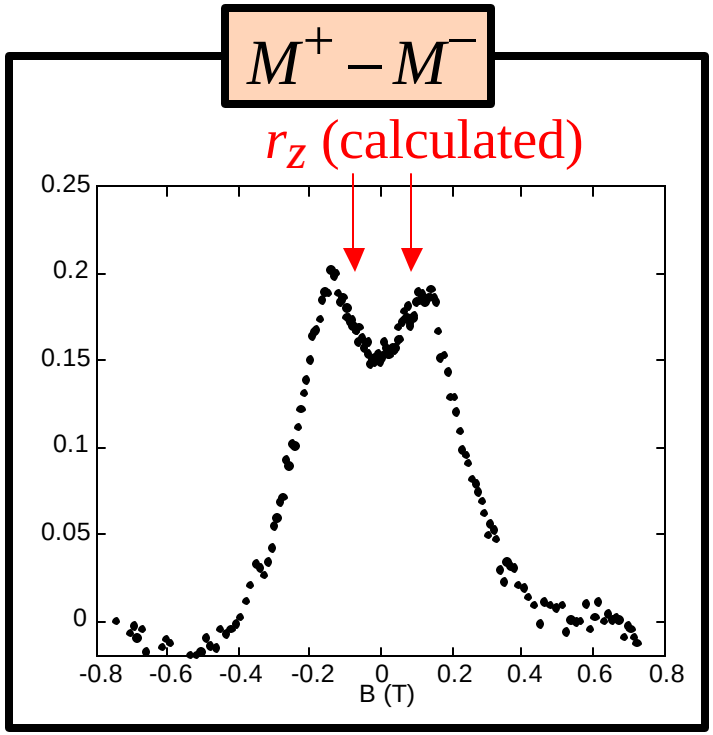
O. Fruchart *et al.*, Phys. Rev. Lett. 23 (14), 2769 (1999)

	Loop	$M^+ - M^-$	Distributed assembly	Minor loops
Easy-axis ‘ single domain ’				Symmetrical
‘ Easy ’ axis ‘ domains ’				Non-symmetrical
Hard axis				Symmetrical

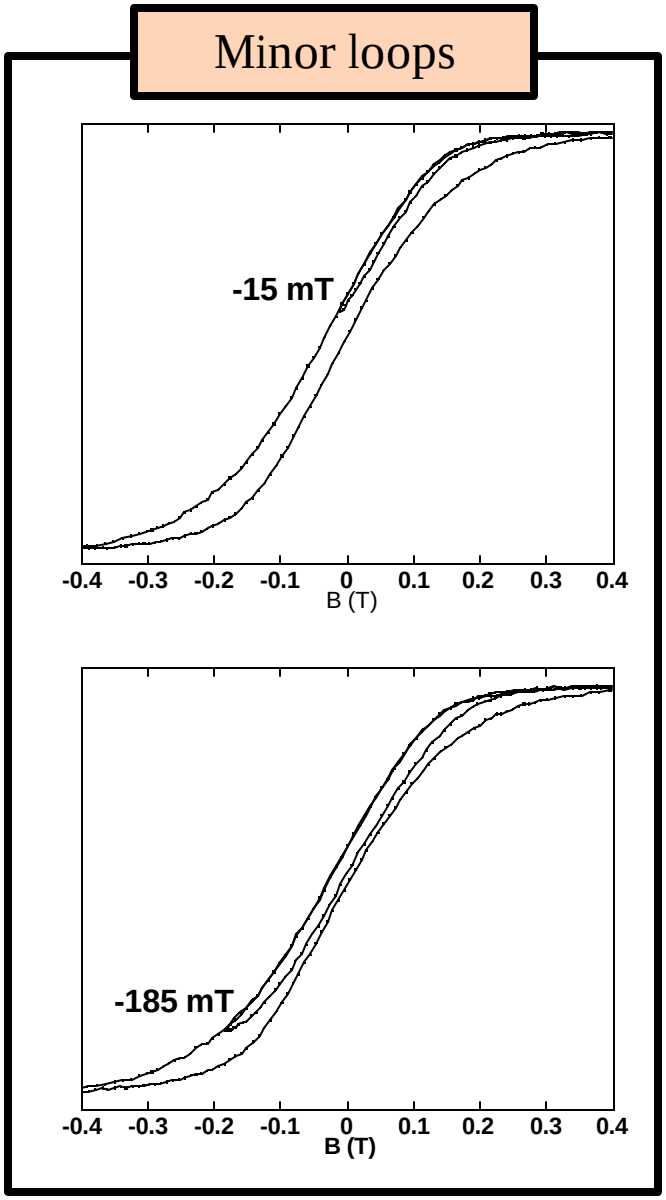




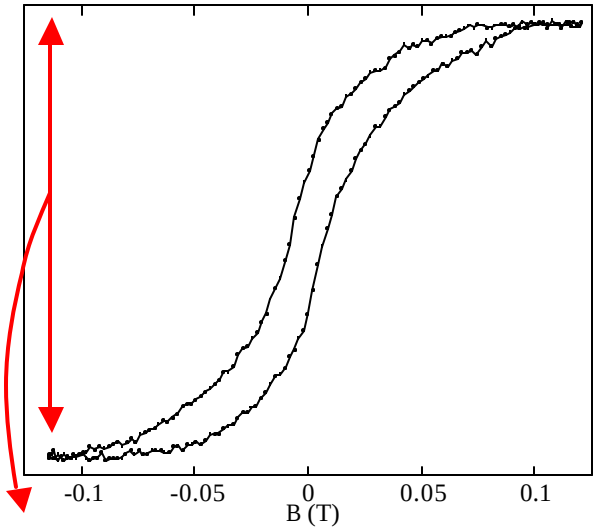
- Similar to « nucleation-annihilation » : ‘ domain ’ structure, with zero wall energy.
- Larger $H_{sat.}$ and more ‘ model ’ hard-axis loop for (↕8;∅4.2) : stronger dipolar interactions.
- ~ agreement with calculated r_z (32 mT and 77 mT, respectively).



- Signature of demagnetizing dipolar interactions: agrees with model
- ‘ Antiferromagnetic ’ ground state not reached: Frustration, metastability.



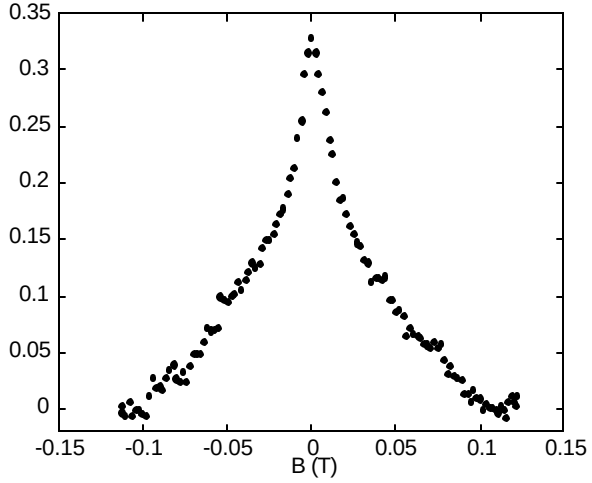
Loop



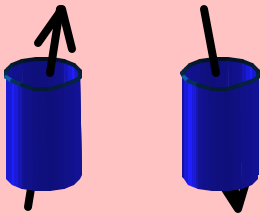
Less than 10% of awaited signal (large reversible part substracted)

$M^+ - M^-$

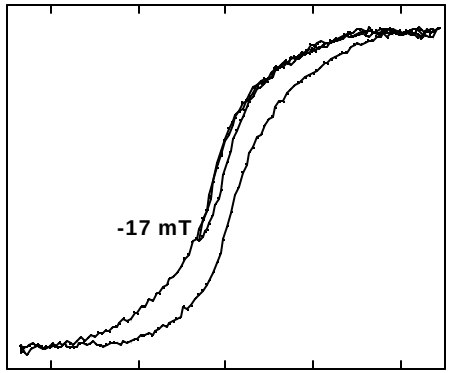
% of $M_{irr.}$

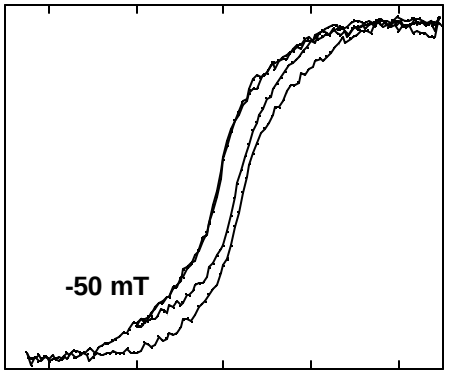


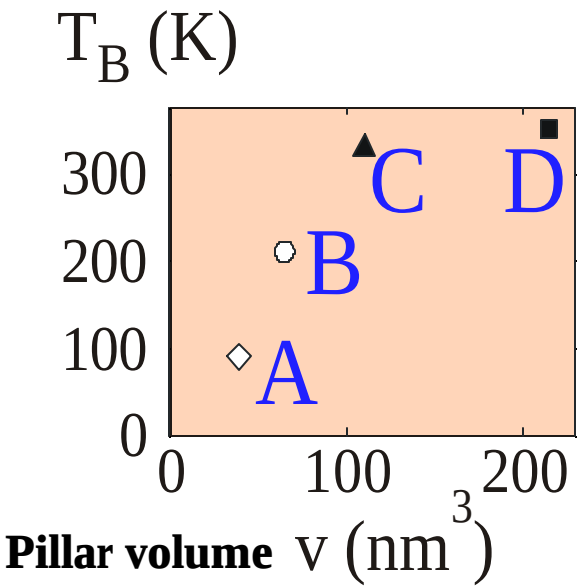
- Very weak hysteresis : perpendicular magnetization.
- Very weak in-plane inter-dots dipolar fields :



Minor loops



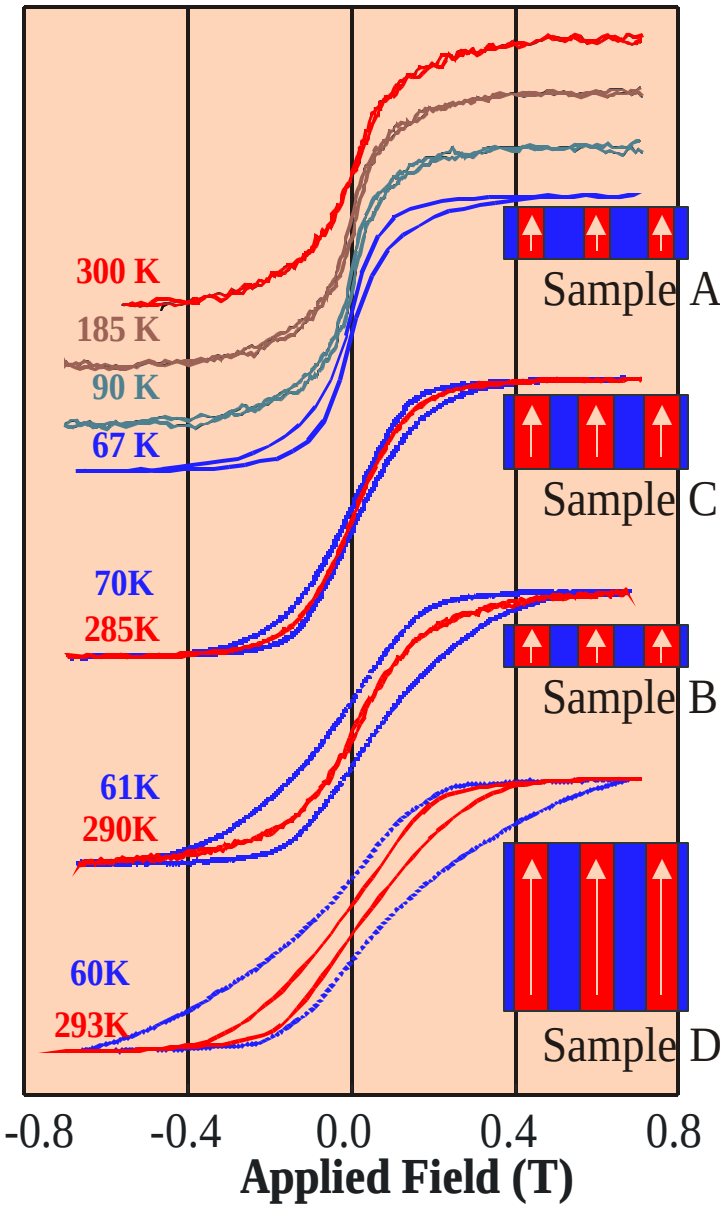




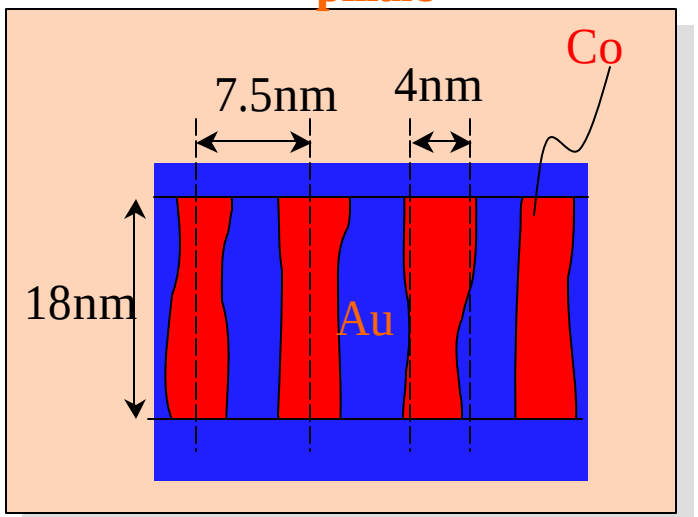
- **Blocking temperature > 300K**
  (~ 30K for flat Co/Au dots)

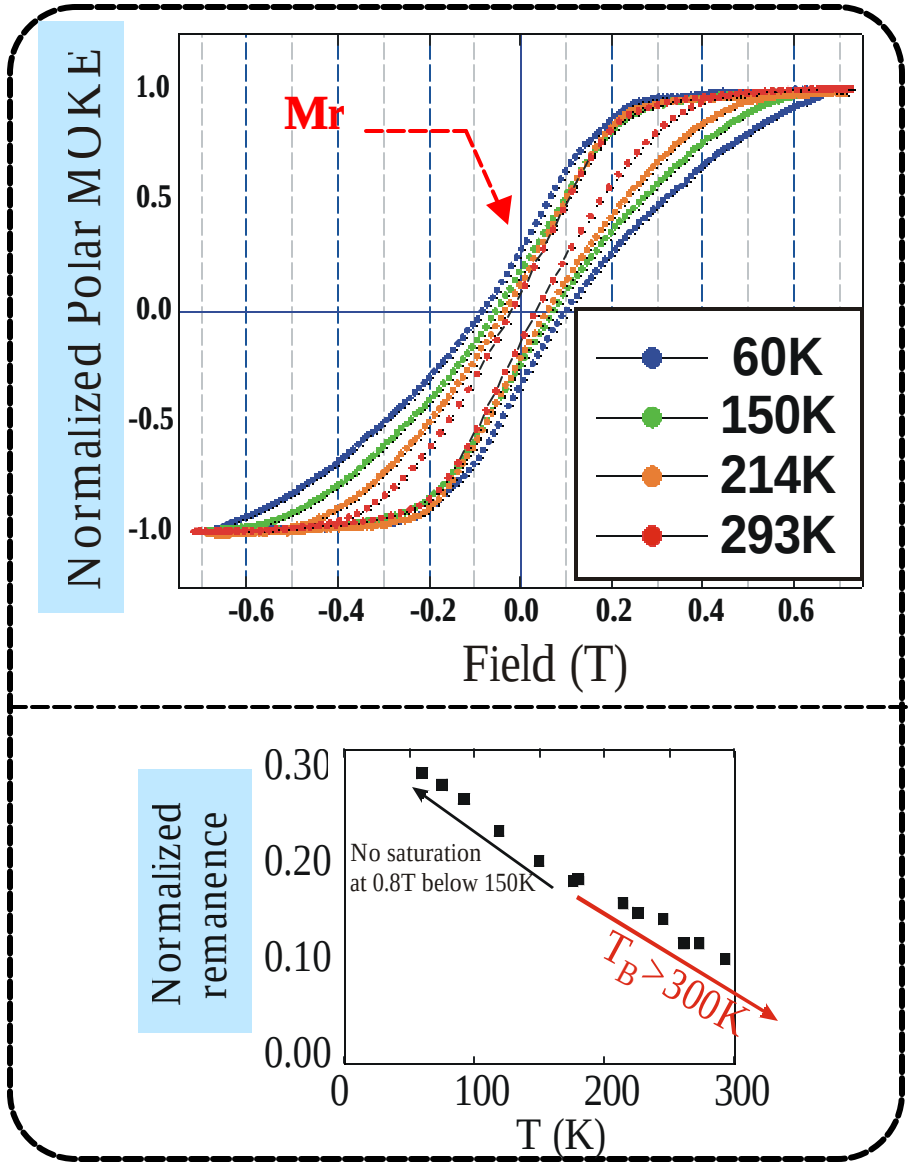
Expected: KV~25kTb

- **K decreases for the largest pillars**



➢ **Sample with bigger pillars**





- Moderate temperature effect
- **Blocking temperature > 300K**

(~ 20K for flat Co/Au dots)

O. Fruchart *et al.*, Proceedings of ISPMM/ISAMT 2001, to appear in J. Magn. Magn. Mater.

- **New growth process**:
  - **vertical replication** of sub-ML flat pattern
  - **self-organized pillars** with vertical aspect ratio

- **Driving forces**:
  - **lattice parameter mismatch**, **immiscibility**, surface energy.
  - validity for other elements than Co/Au ?

- **Motivation for magnetism**:
  - **dramatic rise of Tb**
  - **rise of magnetic signal** (stray fields, etc.)