

Magnetic properties of self-organized systems

[Individual and collective aspects]



O.Fruchart

Laboratoire Louis Néel (CNRS-UJF-INPG)
Grenoble



Laboratoire Louis Néel, Grenoble, France.

<http://lab-neel.grenoble.cnrs.fr/themes/couches/ext/slides/>



Magnetic properties of self-organized systems : TOC

➡ **1. Introduction to magnetism**

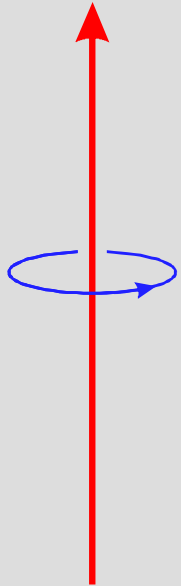
➡ **2. Individual properties**

➡ **3. Collective properties**

➡ **4. Towards materials?**

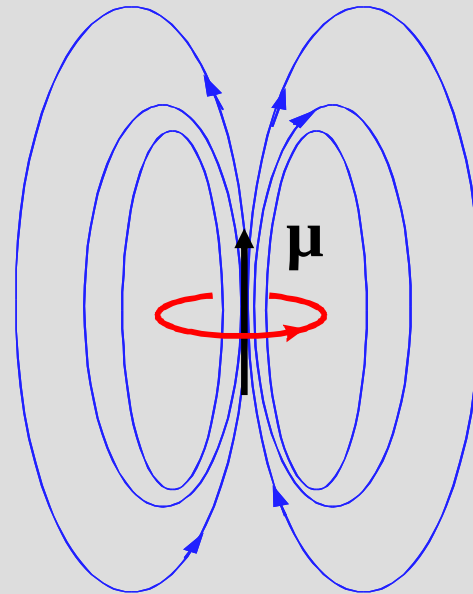


Oersted field



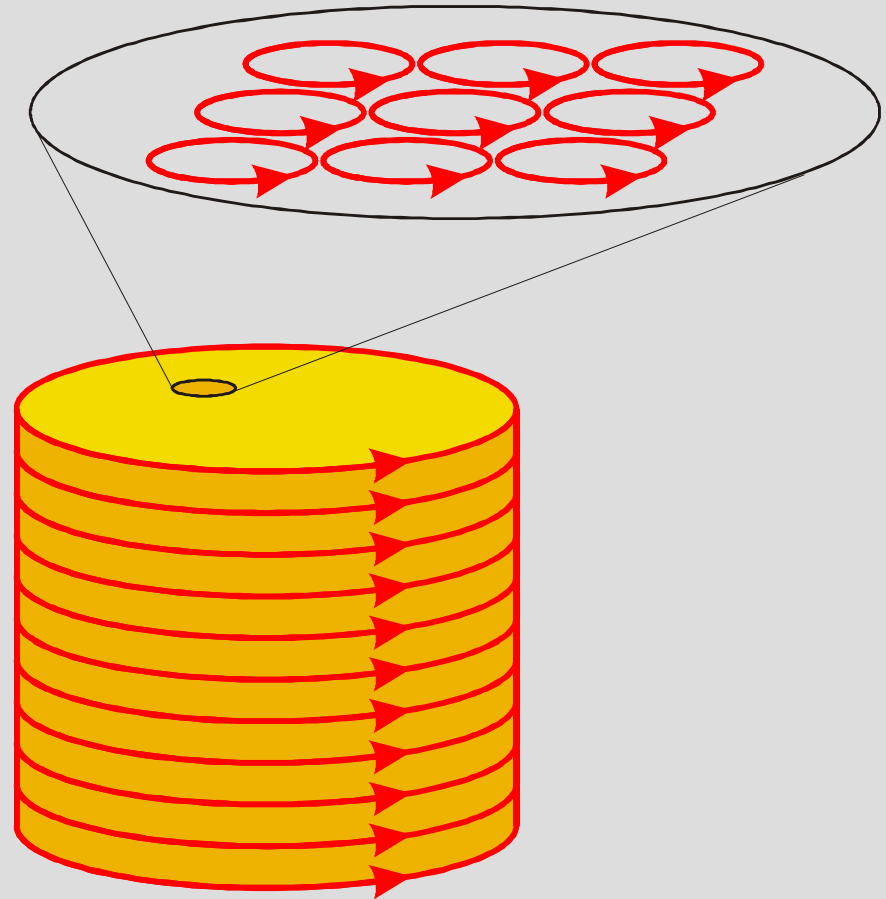
$$B = \frac{\mu_0 I}{2\pi r}$$

Magnetic moment



$$B = \frac{\mu_0}{4\pi r^3} \times \left[\frac{3}{r^2} (\boldsymbol{\mu} \cdot \mathbf{r}) \mathbf{r} - \boldsymbol{\mu} \right]$$

Magnetic material



Magnetization: $\text{A}\cdot\text{m}^{-1}$

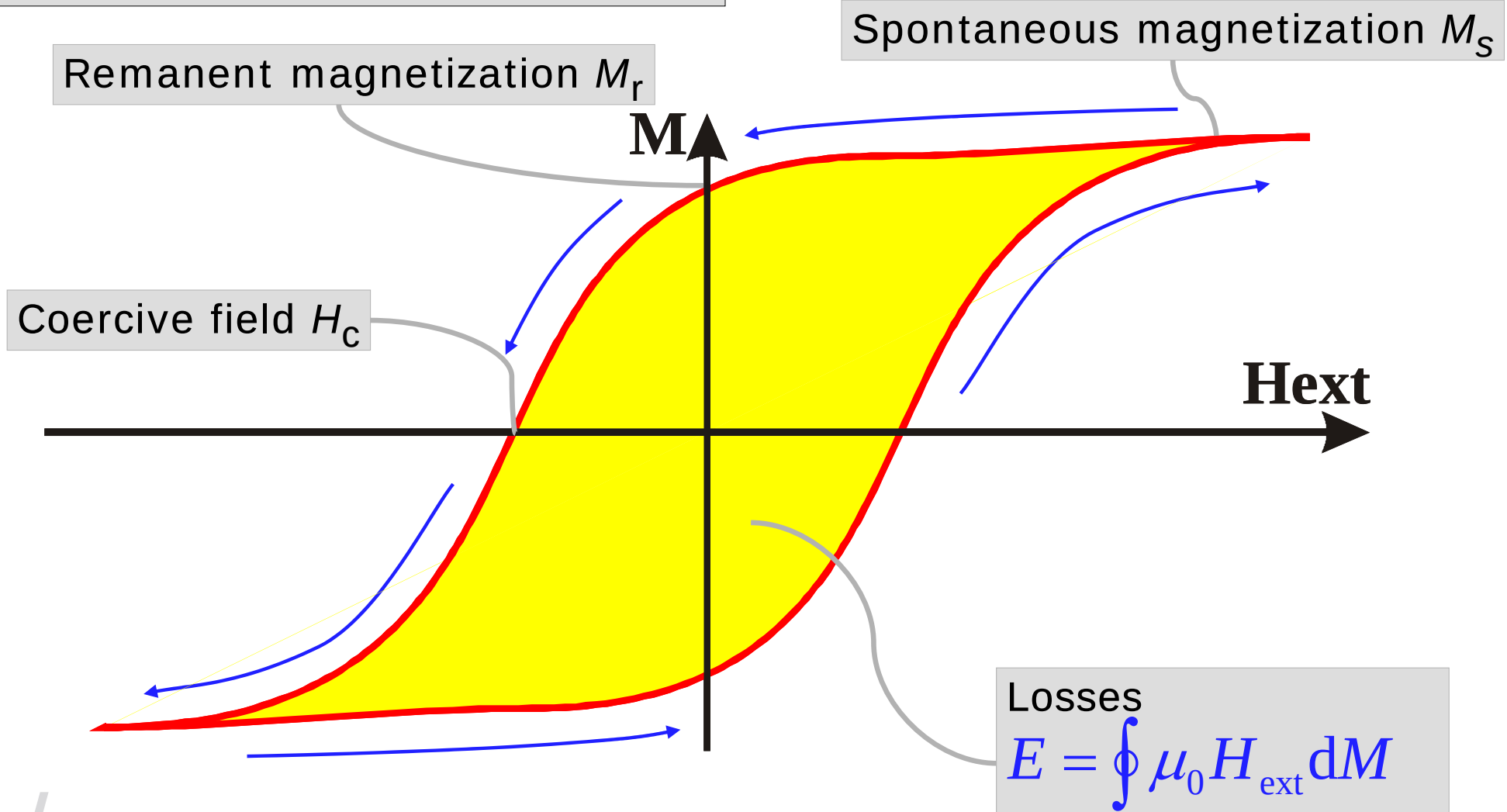
Magnetic moment: $\text{A}\cdot\text{m}^2$



Manipulation of magnetic materials:

↳ Application of a magnetic field

Zeeman energy: $E_Z = -\mu_0 \mathbf{H} \cdot \mathbf{M}$





Soft materials

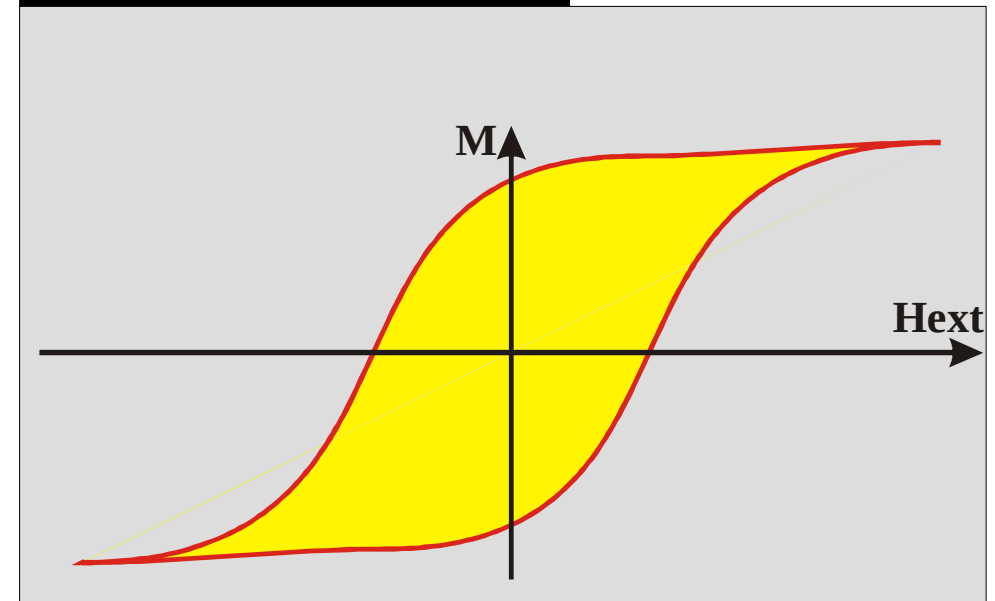


Transformers

Flux guides

Magnetic shielding

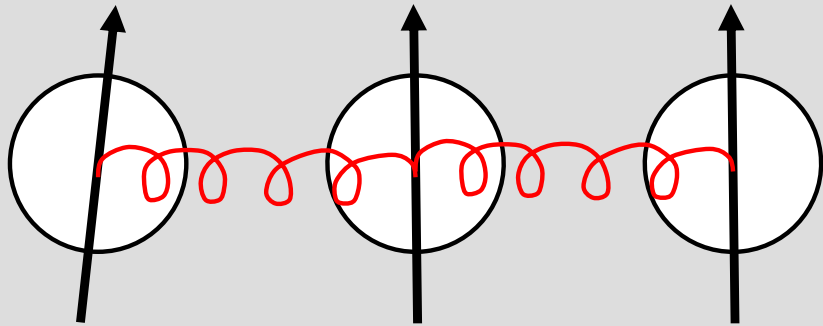
Hard materials



Permanent magnets, motors

Magnetic recording

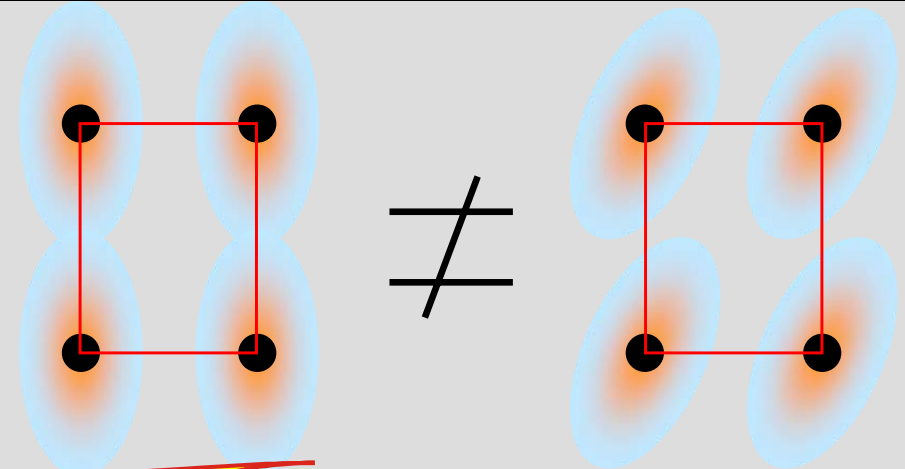
Exchange energy



$$E_{\text{Ech}} = -J_{1,2} \vec{S}_1 \cdot \vec{S}_2$$

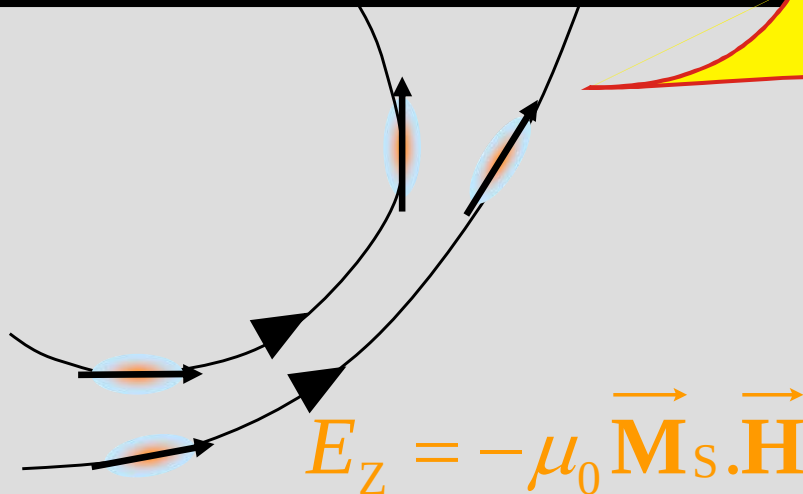
$$= A(\nabla \theta)^2$$

Magnetocrystalline anisotropy energy



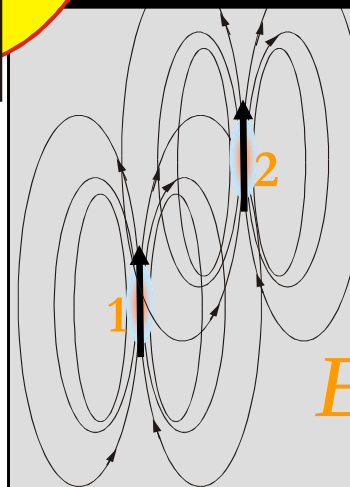
$$E_{\text{mc}} = K \sin^2(\theta)$$

Zeeman energy (enthalpy)



$$E_z = -\mu_0 \vec{M}_s \cdot \vec{H}$$

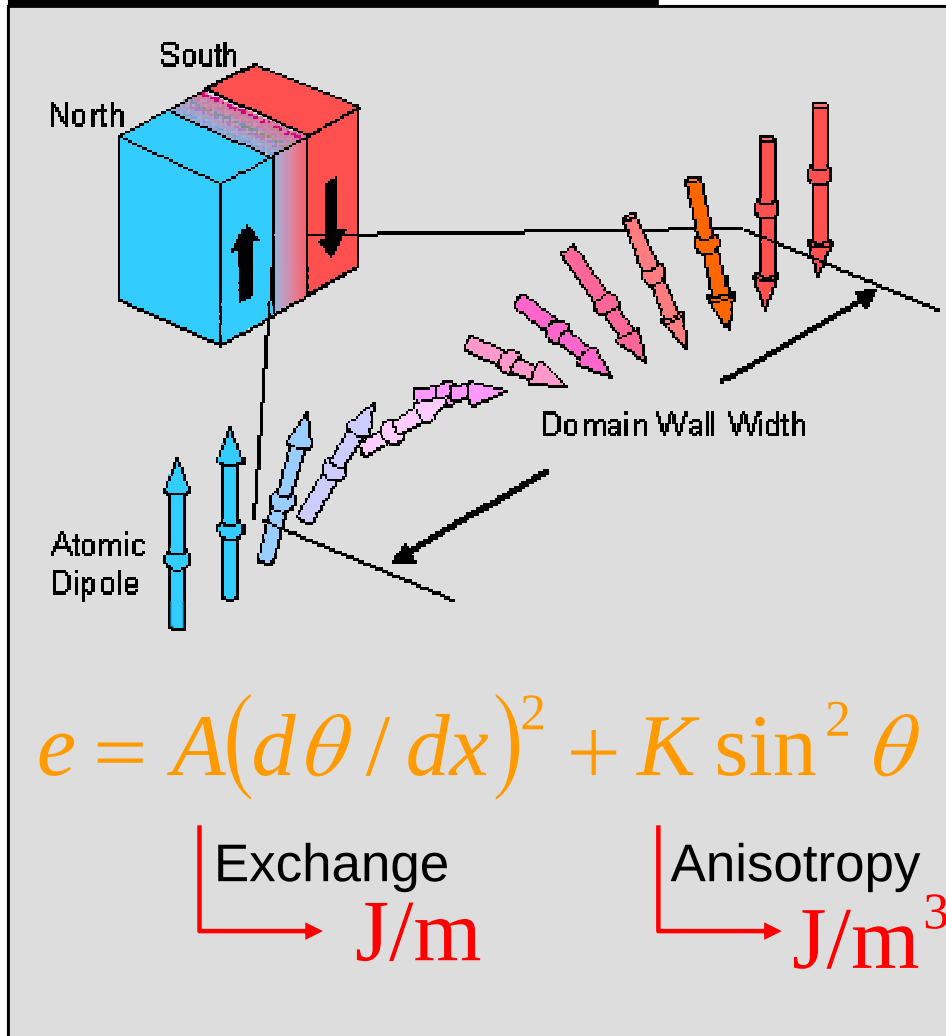
Dipolar energy



$$E_d = -\frac{1}{2} \mu_0 \vec{M}_s \cdot \vec{H}_d$$



Typical length scale:
Bloch wall width λ_B



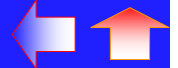
Numerical values

$$\lambda_B = \pi \sqrt{A/K}$$

$$\lambda_B = 2 - 3 \text{ nm} \longrightarrow \lambda_B \geq 100 \text{ nm}$$

Hard

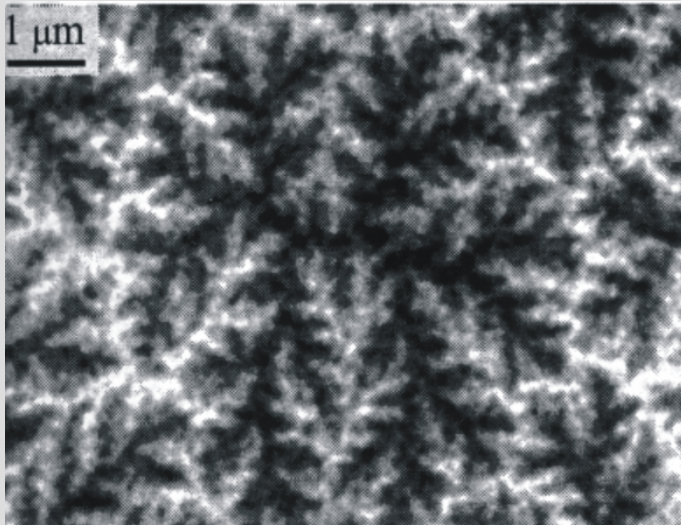
Soft



One possible definition: mesoscopic magnetism

Bulk material

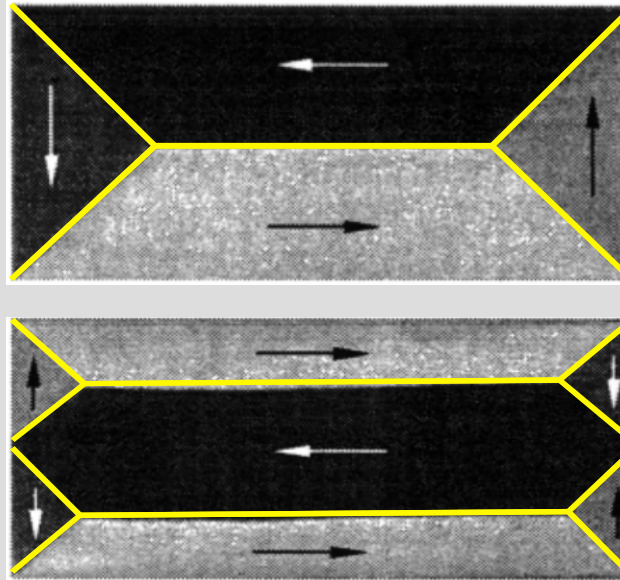
Numerous and complex magnetic domains



Co(1000) crystal – SEMPA
A. Hubert, *Magnetic domains*

Mesoscopic scale

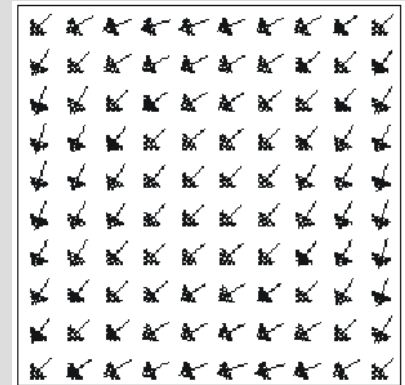
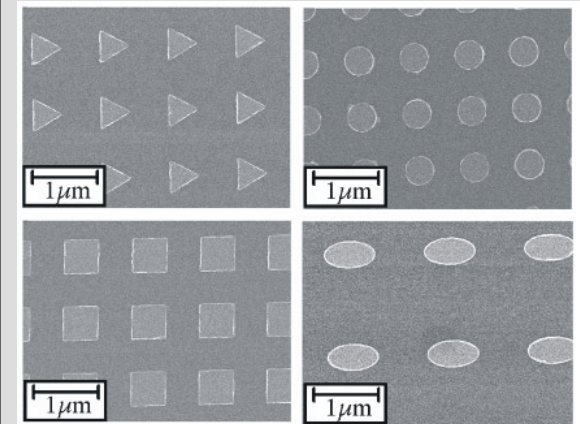
Small number of domains, simple shape



Microfabricated dots
Kerr magnetic imaging
A. Hubert, *Magnetic domains*

Nanometric scale

Magnetic single-domain



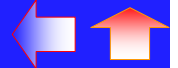
R.P. Cowburn,
*J.Phys.D:Appl.Phys.*33,
R1 (2000)

Magnetic recording essentially makes use of single-domain particles



Magnetic properties of self-organized systems : TOC

- ➡ **1. Introduction to magnetism**
- ➡ **2. Individual properties**
- ➡ **3. Collective properties**
- ➡ **4. Towards materials?**

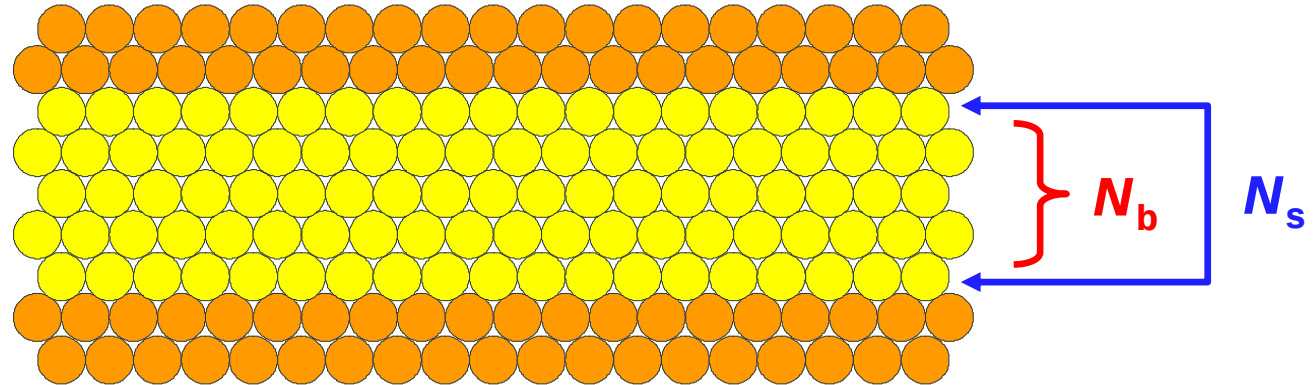


Naïve model

$$T_c = \mu_0 \cdot \frac{J+1}{3J} \cdot \frac{Nw_0 M_0 m_0}{k_B}$$

Molecular field

N neighbors



$$\bar{N} = N_b - \frac{2(N_b - N_s)}{t} \quad \longrightarrow \quad \Delta T_c(t) \sim t^{-1}$$

Less naïve...

Thickness-dependant molecular field

$$\Delta T_c(t) \sim t^{-\lambda}$$

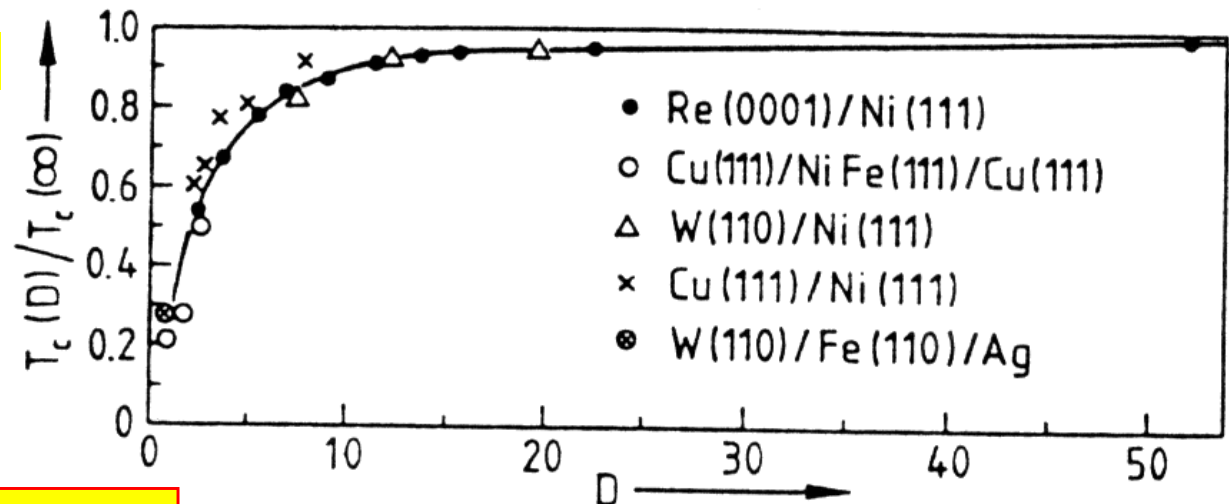
$$\lambda = 1$$

G.A.T. Allan, PRB1, 352 (1970)

Conclusion:

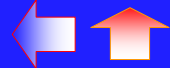
Naïve views are roughly correct

Experiments



U. Gradmann,
Handbook of Magn. Mater. Vol.7, ch.1 (1993)

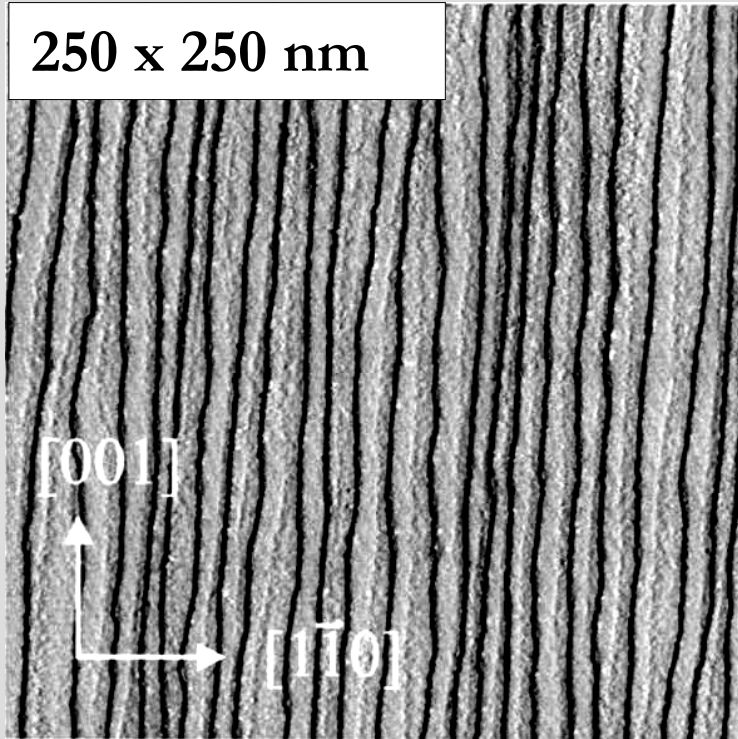




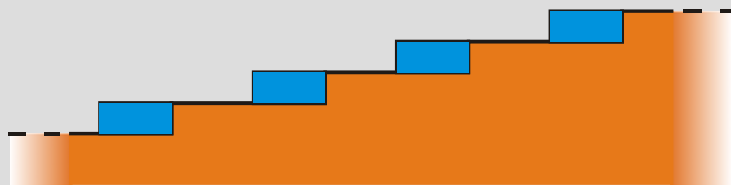
Stripes on vicinal surfaces

Fe(0.5ML) / W(110)

250 x 250 nm

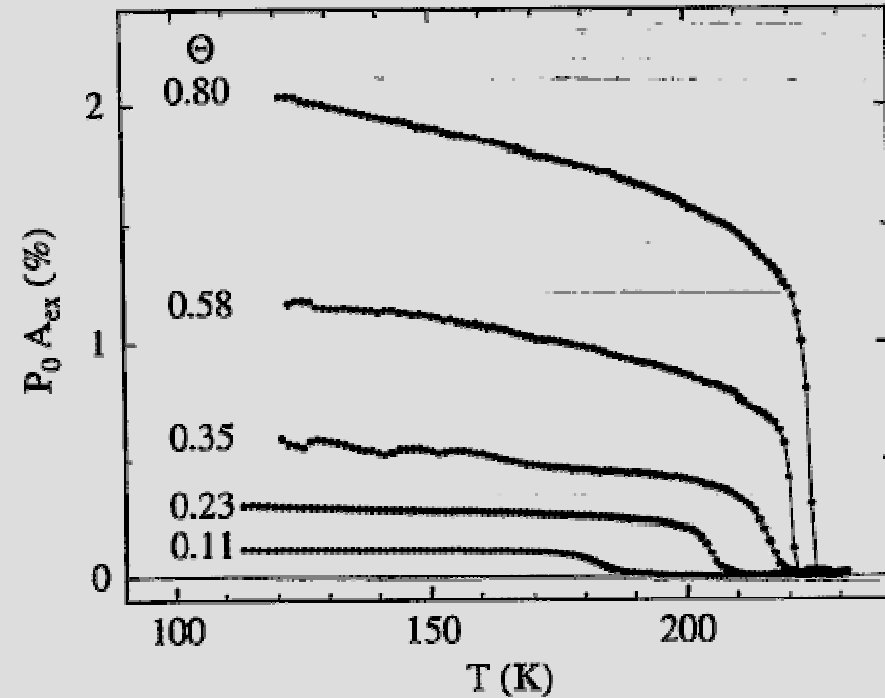


J. Hauschild *et al.*,
Phys.Rev.B57, R677(1998)



Magnetic order in 2D: finite-size scaling

Curie temperature as a function of stripe width



$$T_C(n)/T_C(\infty) \propto 1 - (n_0/n)^\lambda$$

$\lambda = 1.03$ in agreement with Ising model (=1)

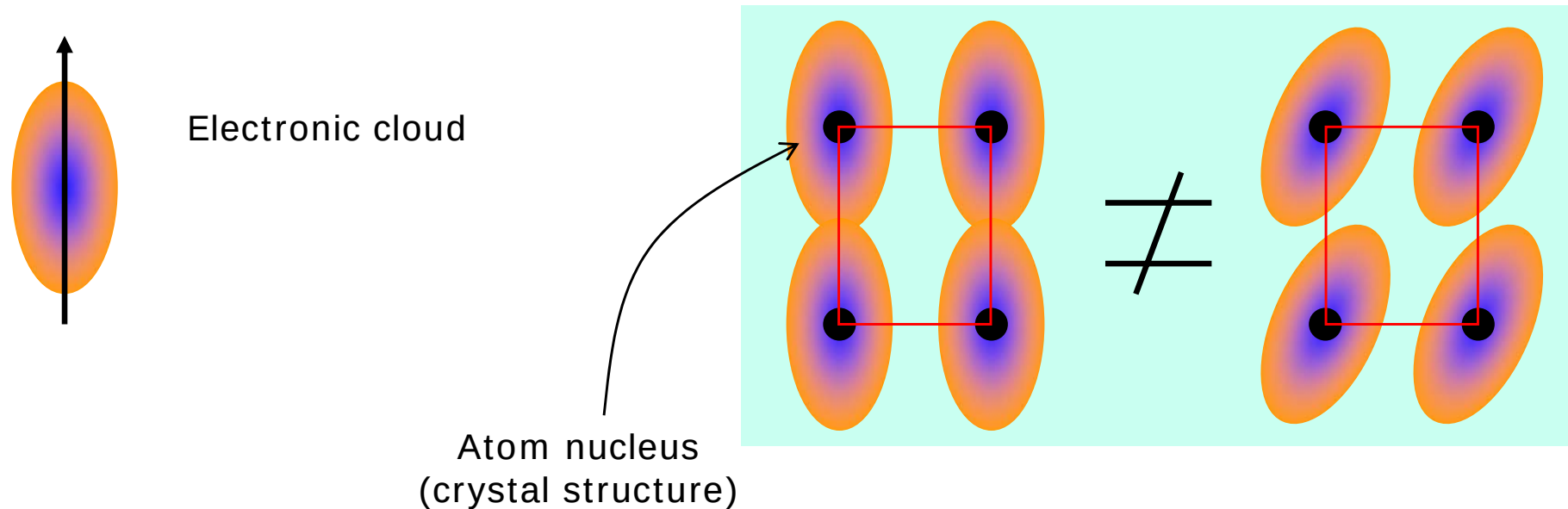
H. J. Elmers *et al.*, PRL73, 898 (1994)

$\lambda = 1.2 \pm 0.3$ for Fe stripes / Pd(110)

D. Li *et al.*, PRB64, 144410 (2001)



Magnetocrystalline anisotropy energy



Spin-orbit coupling $\Rightarrow$ the energy of both spin and orbital moment depends on orientation

Series development on an angular basis:

Anisotropy energy

Normalized magnetization components

$$E_{mc} = K_1 m_z^2 + K_2 m_z^4 + \dots \quad \text{Uniaxial}$$

$$E_{mc} = K_4 (m_x^2 m_y^2 + m_y^2 m_z^2 + m_z^2 m_x^2) + \dots \quad \text{Cubic}$$

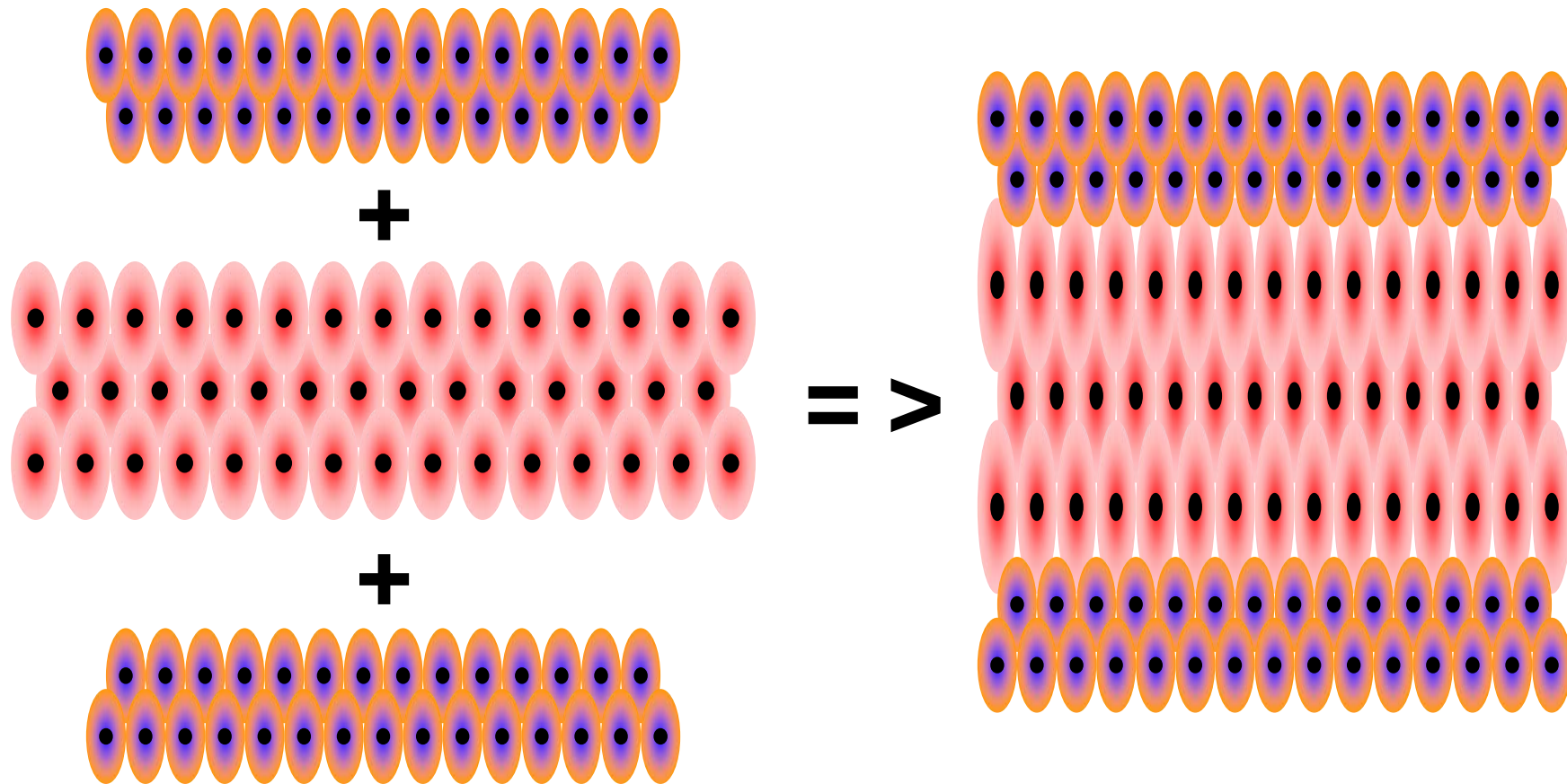
...

**Alignement of magnetization
is favored along
given axes of the crystal**

(Derived from slide of A. Thiaville - CNRS/Orsay)

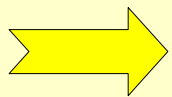


Magneto-elastic anisotropy



Origin

Deformation of orbitals



Correction to the
magneto-crystalline energy

Result

$$E_{\text{mel}} = K_{\text{mel},1} \cos^2(\theta) + \dots$$

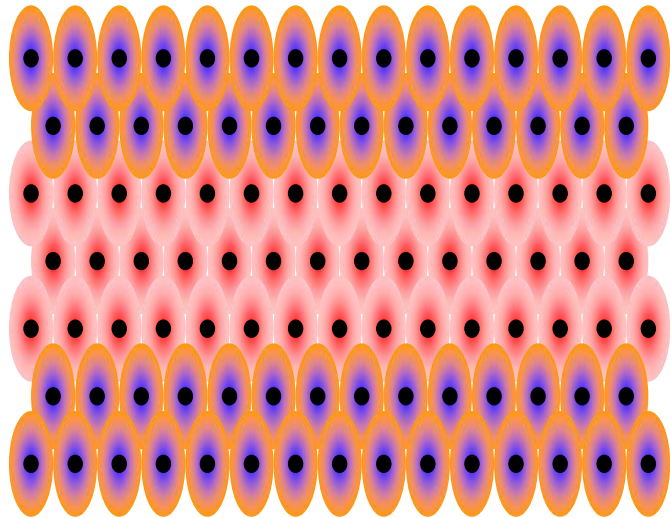
$$K_{\text{mel},i} \sim B_i \varepsilon$$



Surface anisotropy

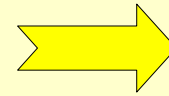
L. Néel,
J. Phys. Radium 15,
15 (1954)

« Anisotropie magnétique superficielle et surstructures d'orientation »
« Superficial magnetic anisotropy and orientational superstructures »



Overview

Breaking of symmetry for
surface/interface atoms



Correction to the
magneto-crystalline energy

$$E_s = K_{S,1} \cos^2(\theta) + K_{S,2} \cos^4(\theta) + \dots$$

« Cette énergie de surface, de l'ordre de 0.1 à 1 erg/cm², est susceptible de jouer un rôle important dans les propriétés des substances ferromagnétiques dispersées en éléments de dimensions inférieures à 100Å »

« This surface energy, of the order of 0.1 to 1 erg/cm², is liable to play a significant role in the properties of ferromagnetic materials spread in elements of dimensions smaller than 100Å »

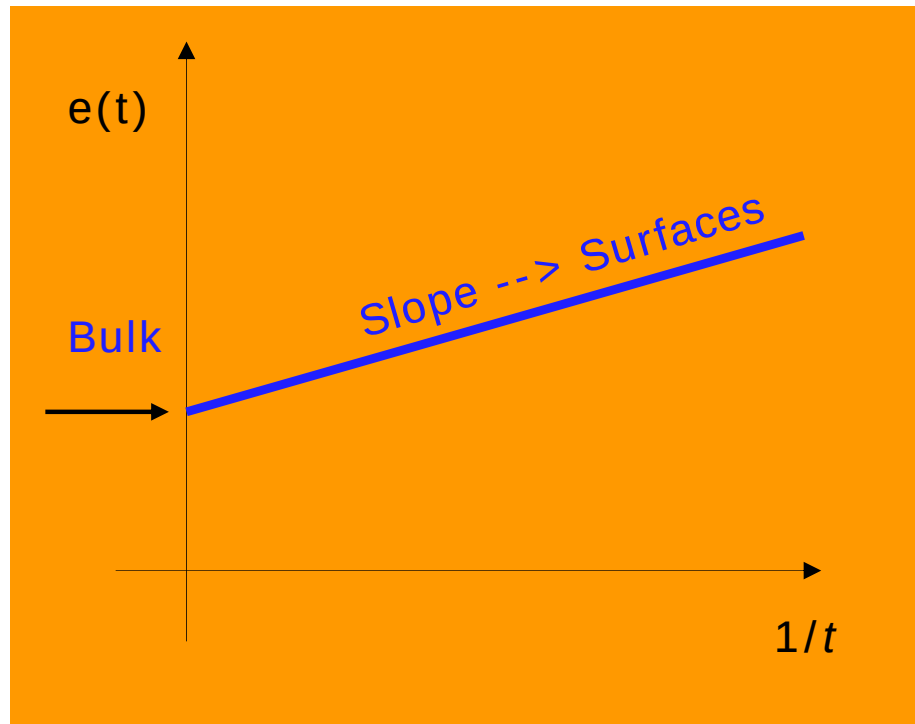
Pair model of Néel:

- K_s estimated from magneto-elastic constants
- Does not depend on interface material
- Yields order of magnitude only: correct value from experiments or calculations

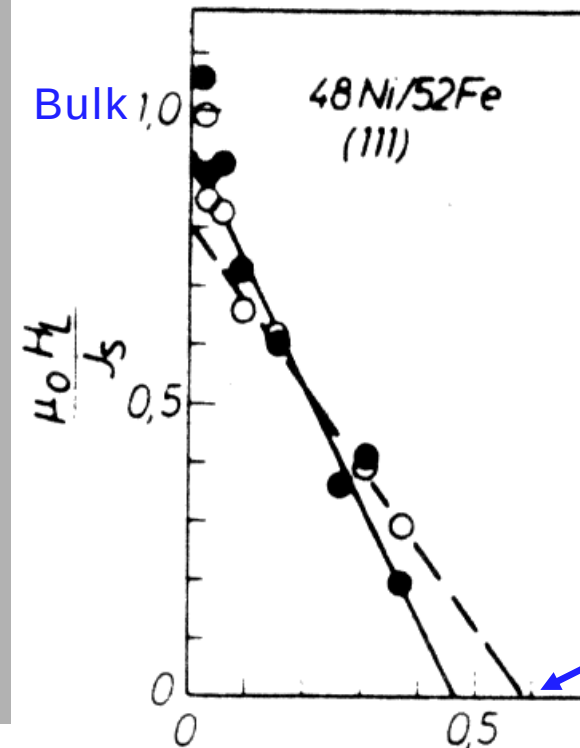


History of surface anisotropy : STEP 1 (1/t plot)

$$E_{\text{tot}}(t) = k_V t + 2k_S \quad \Rightarrow \quad e(t) = k_V + \frac{2k_S}{t}$$

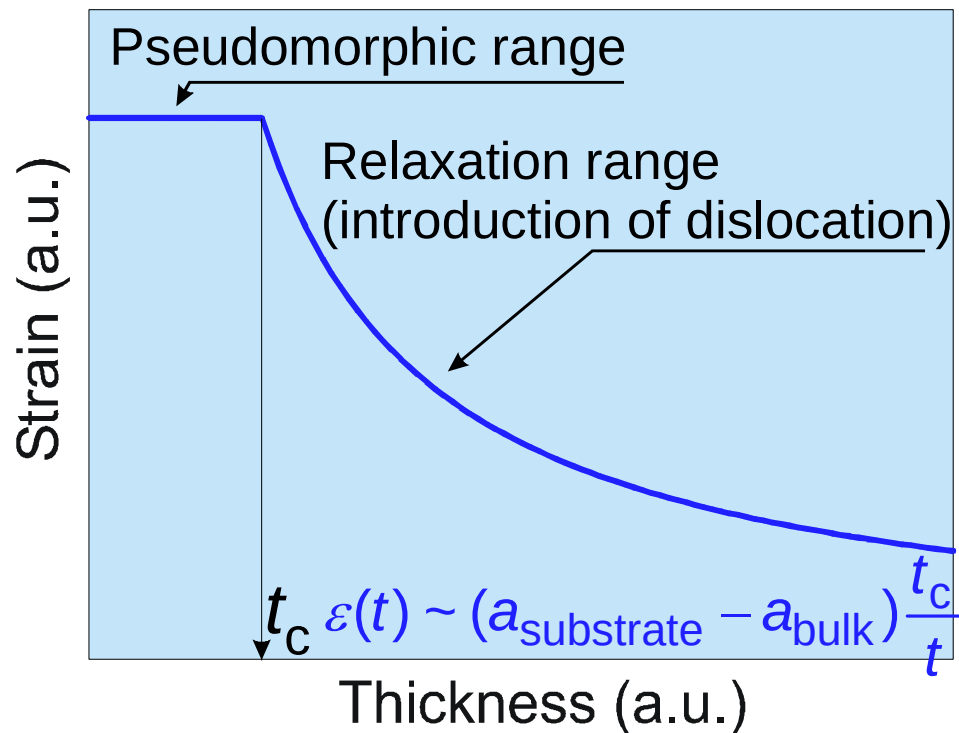


First example of perpendicular anisotropy

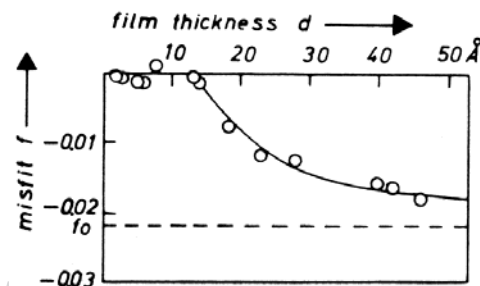


U. Gradmann and J. Müller,
Phys. Status Solidi 27, 313 (1968)

Structural relaxation



W. A. Jesser et al., Phys. Stat. Sol. 19, 95 (1967)



Co/Cu(111)

U. Gradmann, Appl. Phys. 3, 161 (1974)

Laboratoire Louis Néel, Grenoble, France.

Effect on anisotropy

Magneto-elastic anisotropy:

$$k_{\text{mel}} \sim B_{\text{mel}} \varepsilon$$

Strain relaxation regime:

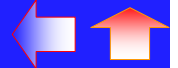
$$k(t) = k_{\text{bulk}} + \alpha B_{\text{mel}} / t$$

Conclusion:

Mixing of surface and magneto-elastic contributions

$$e(t) = k_V + \frac{2k_S}{t}$$

C. Chappert and P. Bruno., JAP64, 5736 (1988)



Magnetic Anisotropy Energy (MAE): Link with anisotropy of orbital moment

Theory

Perturbation theory for 3d metals:

$$MAE = \alpha \frac{\xi}{4\mu_B} \Delta\mu_L$$

P. Bruno,
PRB39, 865 (1989)

μ_L does not rotate in 3d metals
→ MAE reflects cost in ξ

Covers magnetocrystalline, magnetoelastic
and surface anisotropy

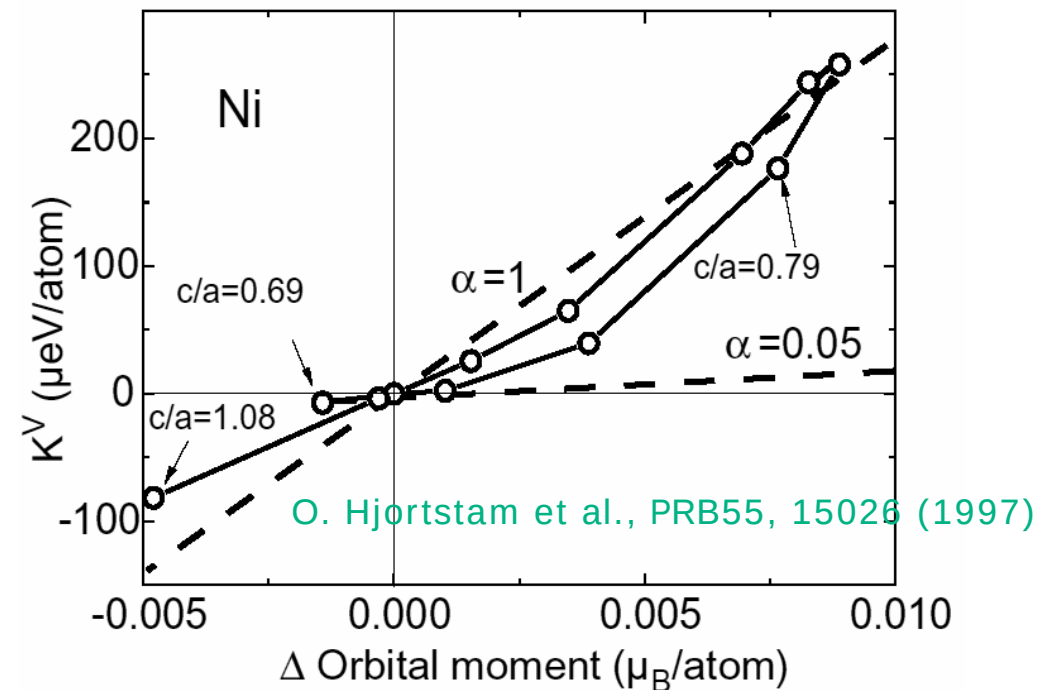
Experiments

Bulk (Fe, Ni, ...)

$\Delta\mu_L \approx 10^{-4} \mu_B / \text{atom}$ → $MAE \leq 1 \mu\text{eV}$

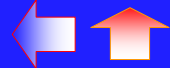
Ab initio calculations

High precision needed: $1 \mu\text{eV} \ll 10 \text{eV}$



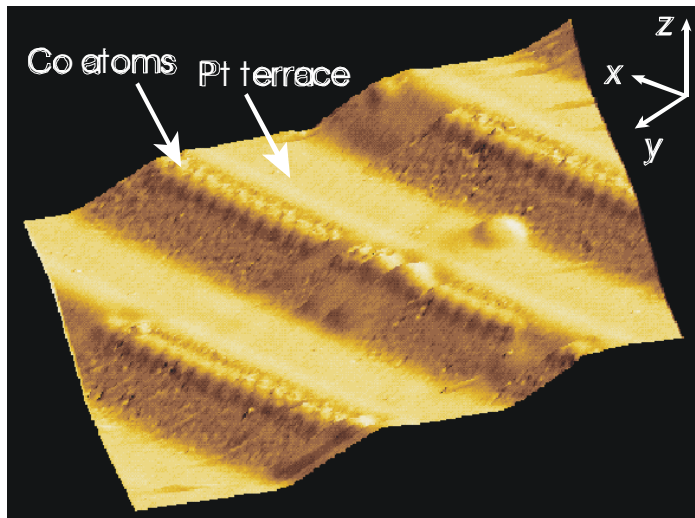
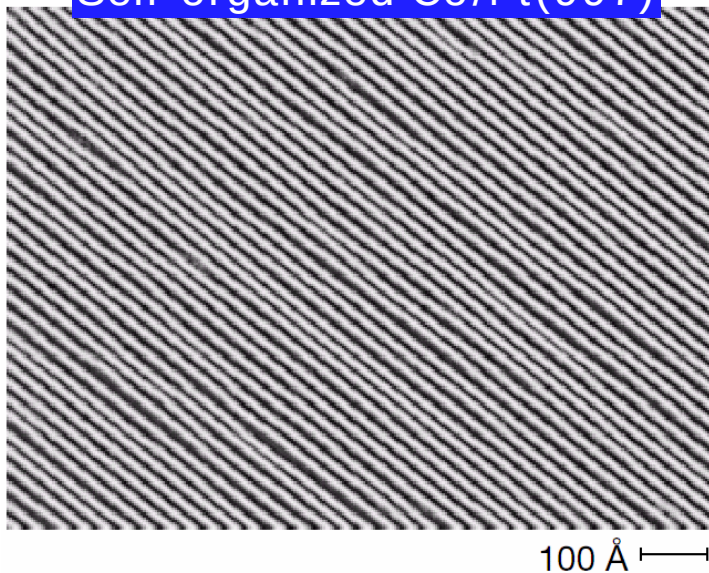
Conclusions

- Origin of MAE = anisotropy of orbital moment
- No strict linearity
- α may also depend on thickness in thin films (band structure)
→ Direct measurement of MAE preferable

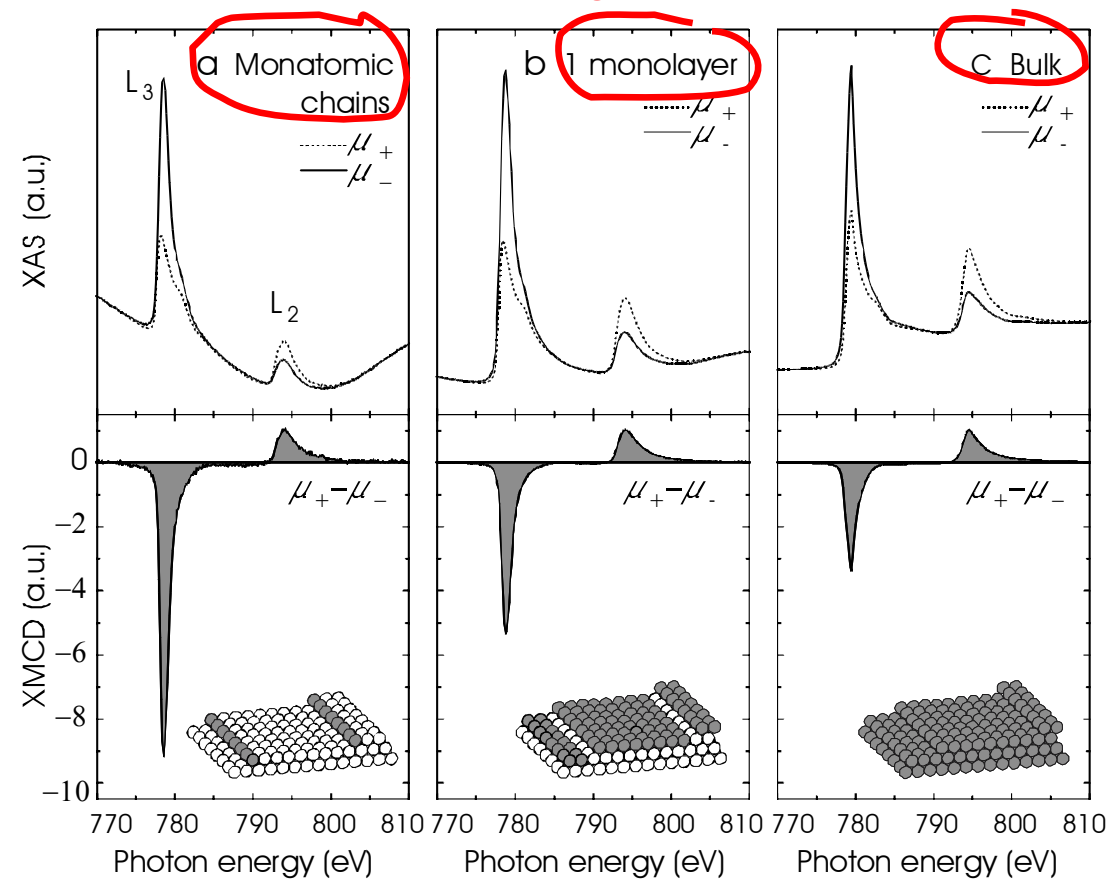


From surface to wires (1D)

Self-organized Co/Pt(997)



Magnetic circular dichroism

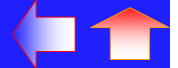


$$\mu_L \approx \int L_3 + \int L_2$$

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

Conclusion:

- Increase of orbital moment (necessary condition for anisotropy)



From surface to wires (1D)

Method

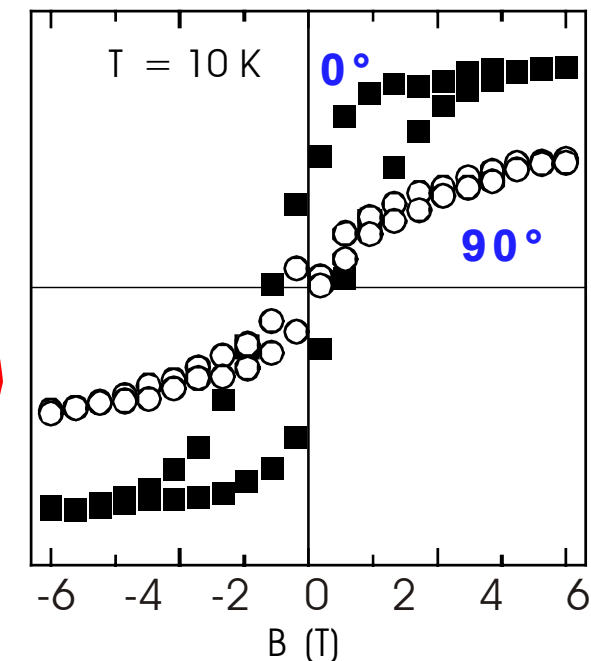
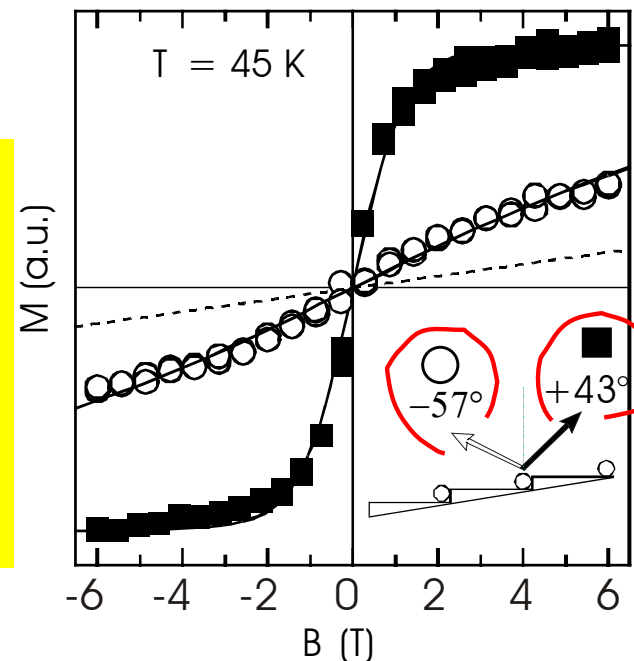
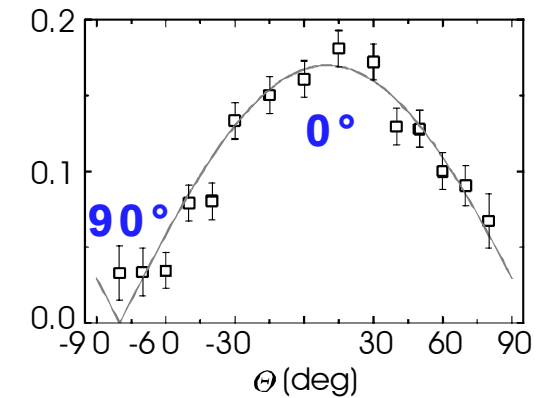
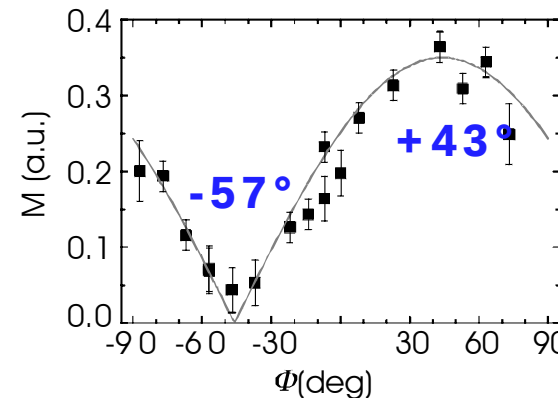
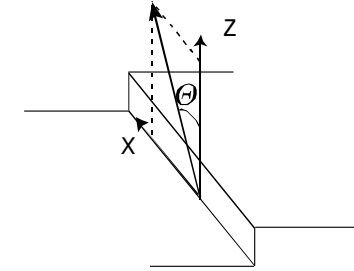
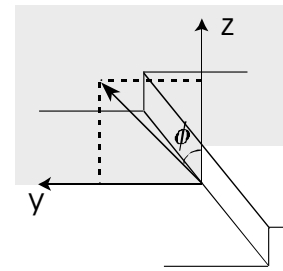
- XMCD > Orbital moment
- Fit magnetization curves > Anisotropy functional

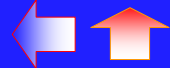
MAE

- Bulk Co: $40\mu\text{eV}/\text{atom}$
- Co ML: $140\mu\text{eV}/\text{atom}$
- Co bi-wire: $0.34\text{meV}/\text{atom}$
- Co wire: $2\text{meV}/\text{atom}$

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.



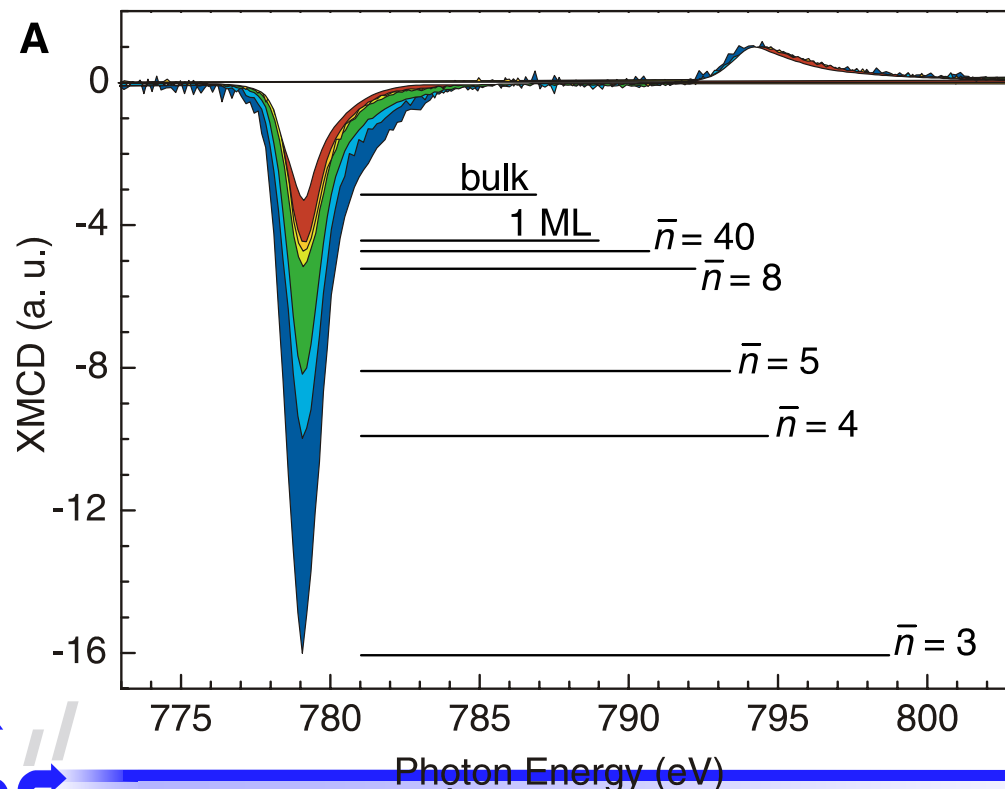


From surface to atoms (0D) **Co/Pt(111)**

Giant Magnetic Anisotropy of Single Cobalt Atoms and Nanoparticles

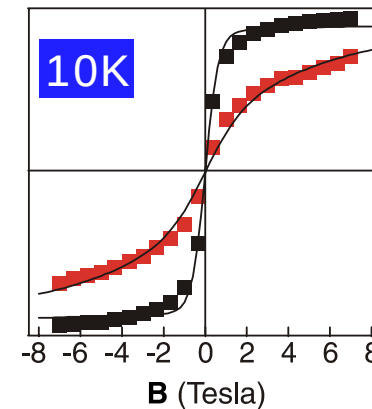
P. Gambardella,^{1,2*} S. Rusponi,^{1,2} M. Veronese,³ S. S. Dhesi,^{4,†}
C. Grazioli,³ A. Dallmeyer,⁵ I. Cabria,⁵ R. Zeller,⁵
P. H. Dederichs,⁵ K. Kern,^{1,2} C. Carbone,^{3,5} H. Brune¹

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

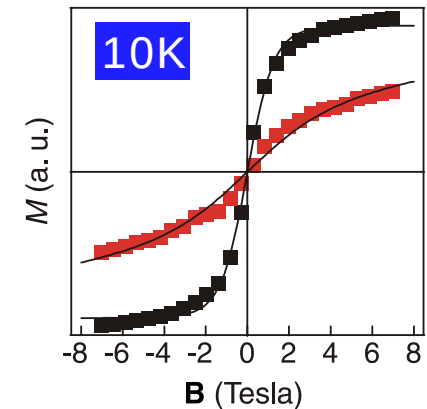


Laboratoire Louis Néel, Grenoble, France.

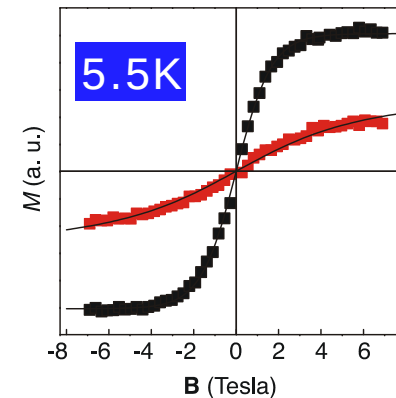
8 atoms



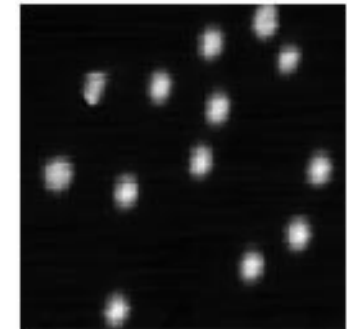
4 atoms



1 atom



STM, 8.5nm, 5.5K



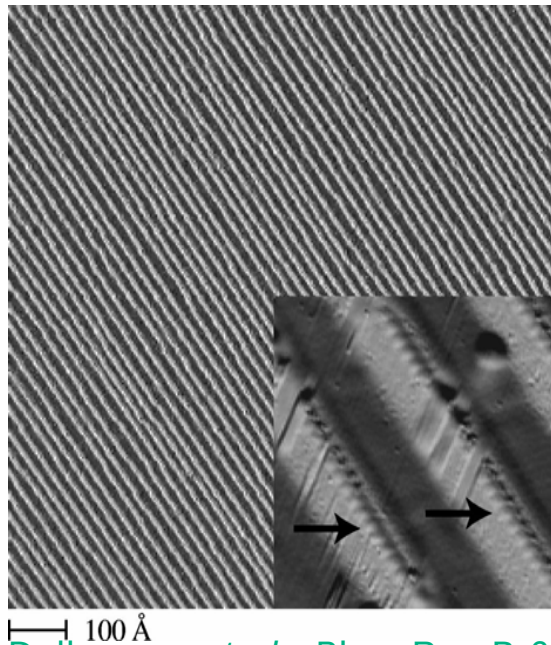
Qualitatively:

- Easy axis of magnetization perpendicular to Pt(111)

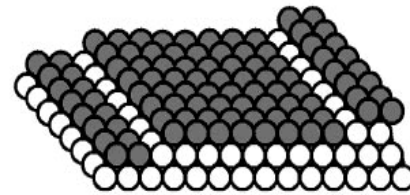




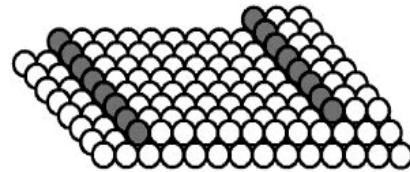
Co / Pt(997)



Terraces ~ 1 ML



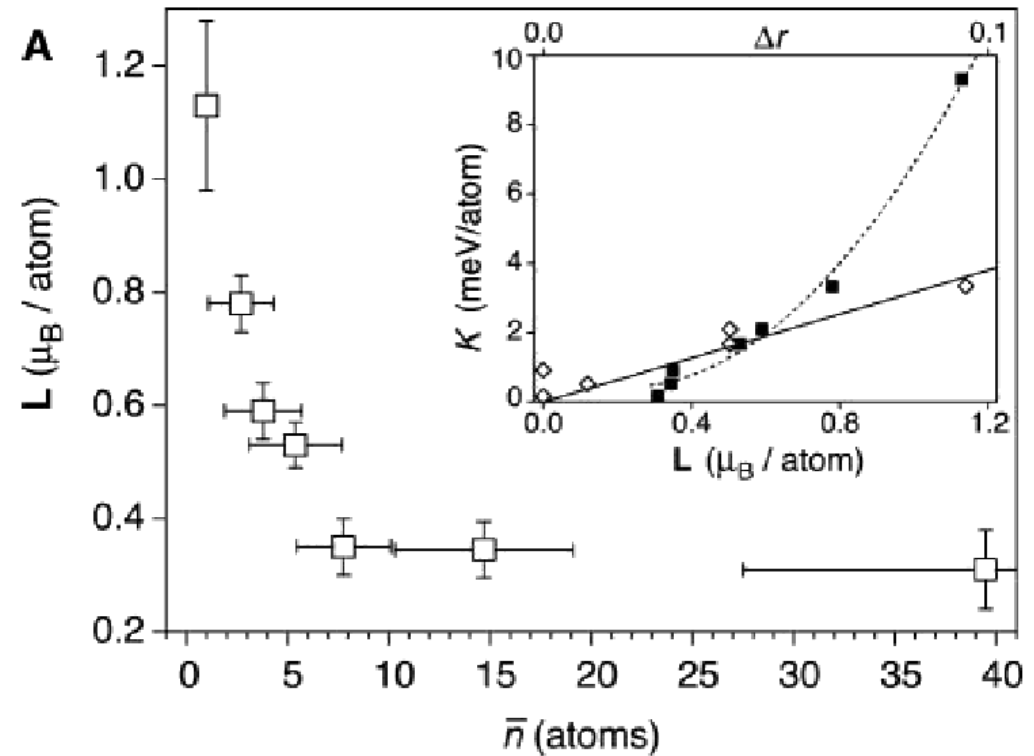
Chains ~ 0.12 ML

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

Conclusions

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

Co / Pt(111)

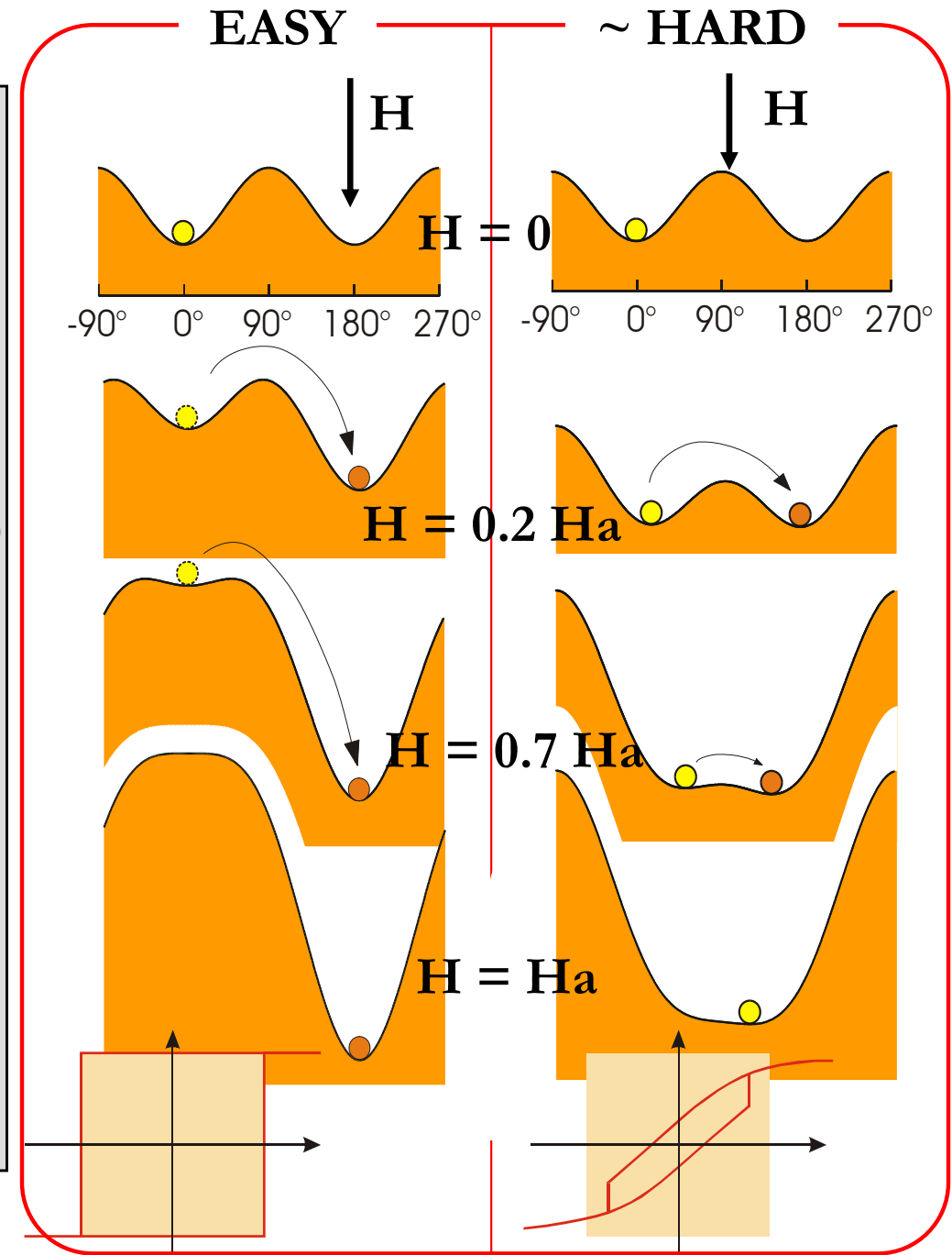
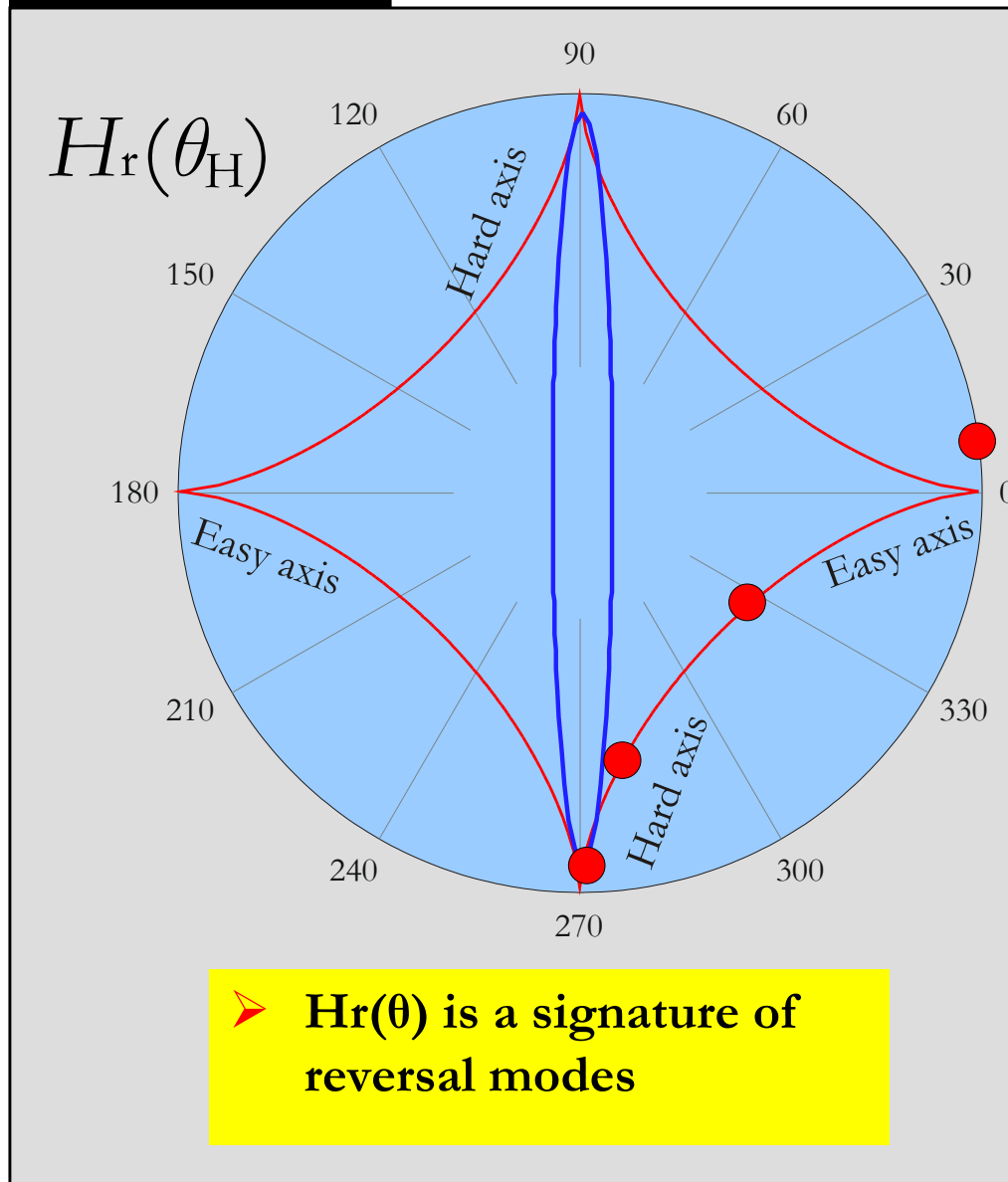
P. Gambardella *et al.*, Science 300, 1130 (2003)

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**

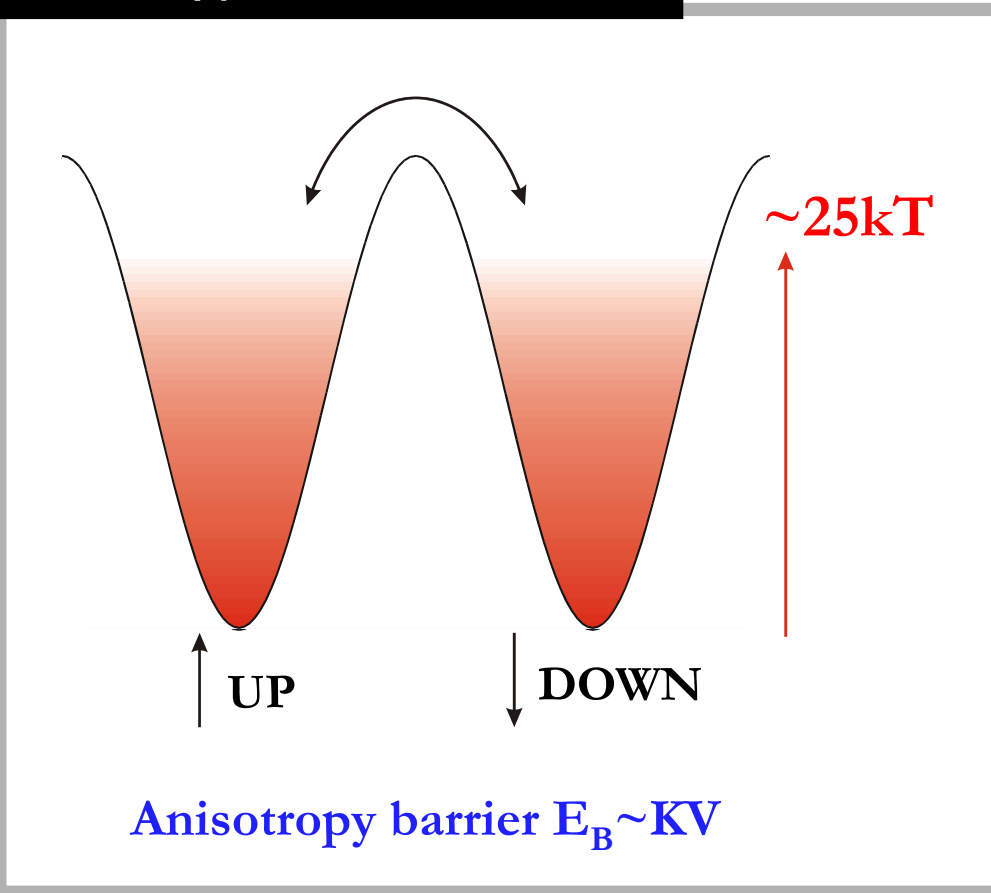


'Astroid' curve





Anisotropy barrier



Formalism for thermal excitations

Phenomenological model

Brown, Phys.Rev.130, 1677 (1963)

- Probability for non-reversal

$$P(t) = e^{-t/\tau}$$

- Mean waiting time for reversal

$$\tau = \tau_0 e^{E_B / k_B T} \quad \tau_0 \approx 10^{-9} \text{s}$$

- ↪ Energy barrier required to prevent magnetization reversal during duration t

$$E_B = k_B T \ln(t / \tau_0)$$

Orders of magnitude

Laboratory : $t = 1 \text{s}$

$$V_B \approx 25 k_B T / K$$

Recording : $t \gg 10^9 \text{s}$

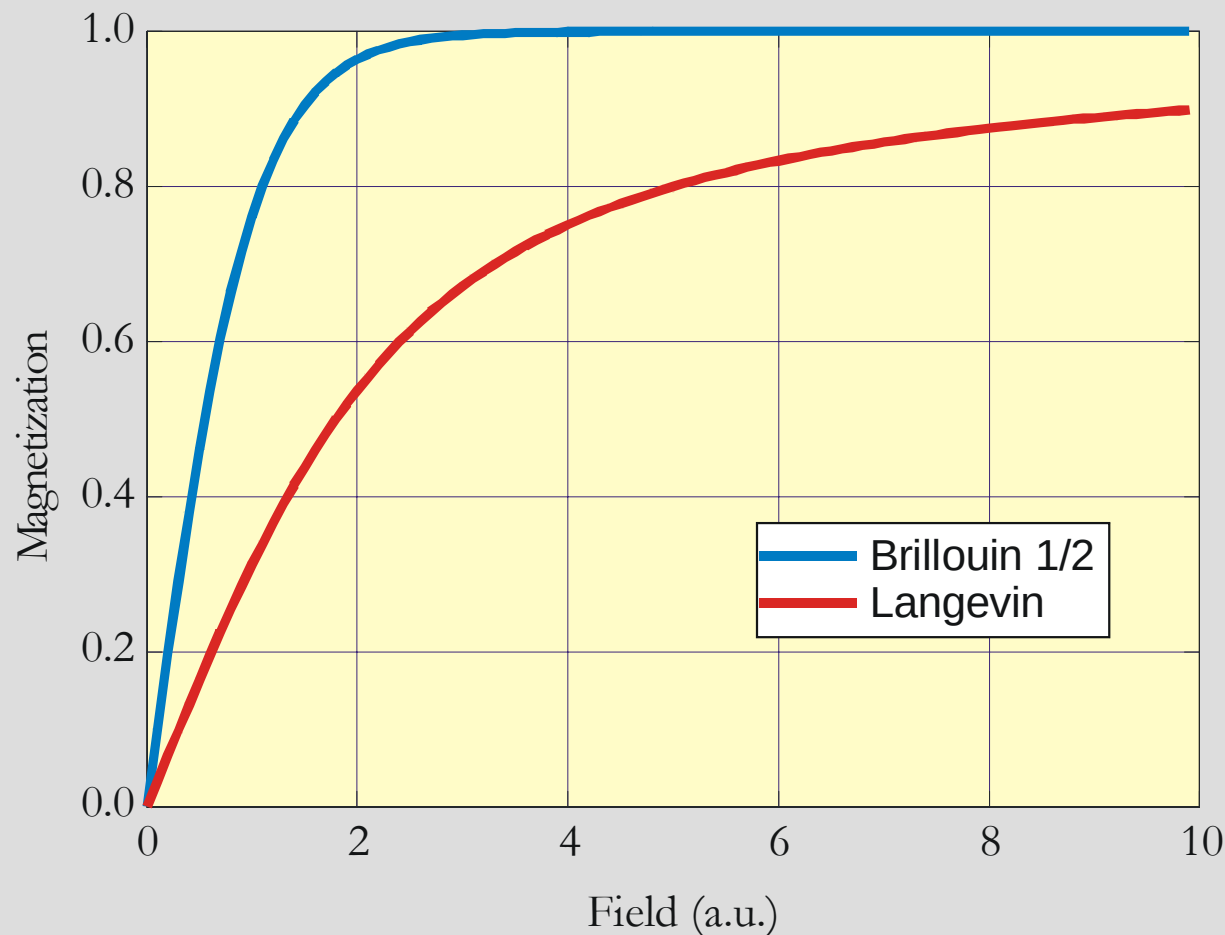
$$V_B \approx 40 - 60 k_B T / K$$

Magnetic recording with one grain per bit ($V = V_B$, discrete media) more favorable than $V = N V_B$





Which fitting function ?



Extremely weak anisotropy

Langevin function

$$m = \tanh(h), \quad h = \beta\mu_0\mu H$$

Strong uniaxial anisotropy

Brillouin 1/2 function

$$m = 1 / \tanh(h) - 1 / h$$

Other cases:

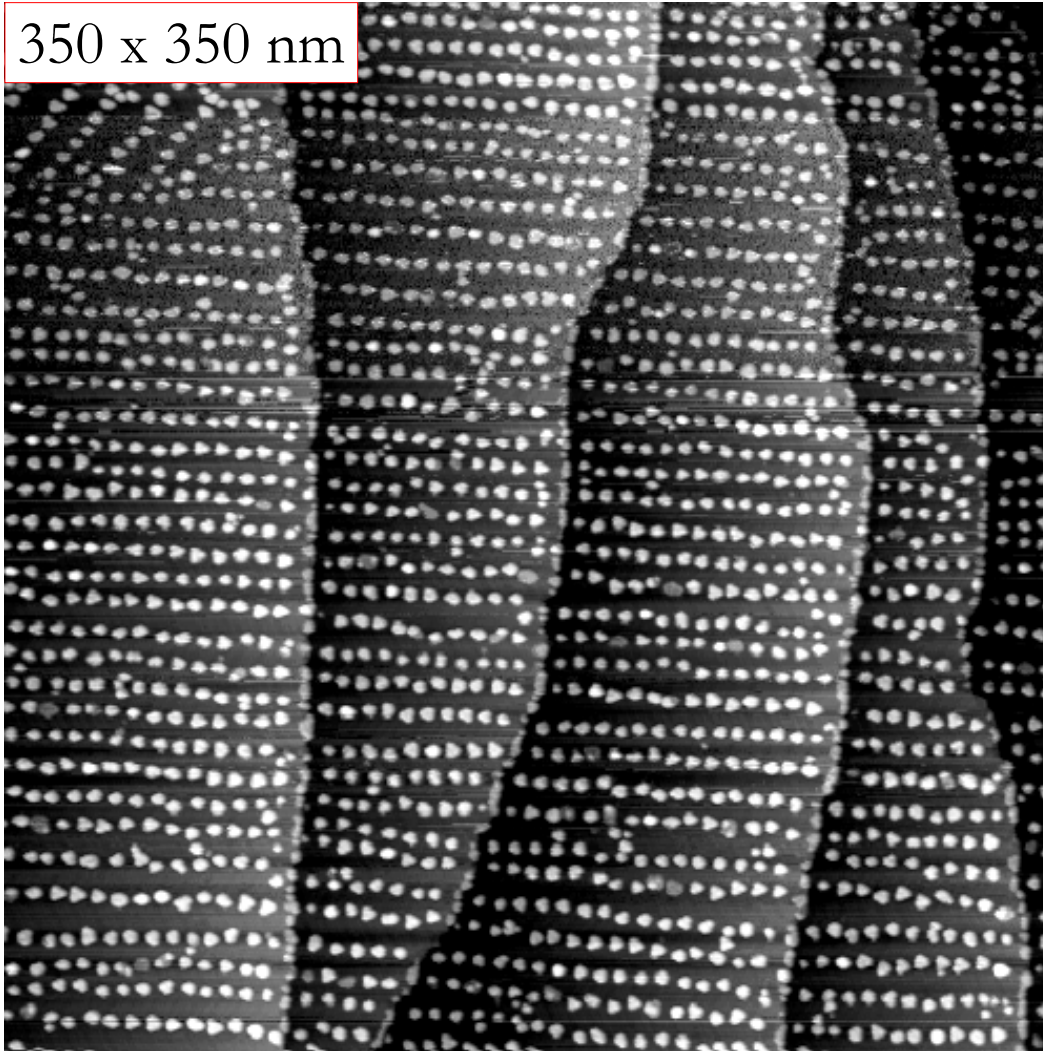
moderate or tilted anisotropy,
distributions etc.

Numerical fitting

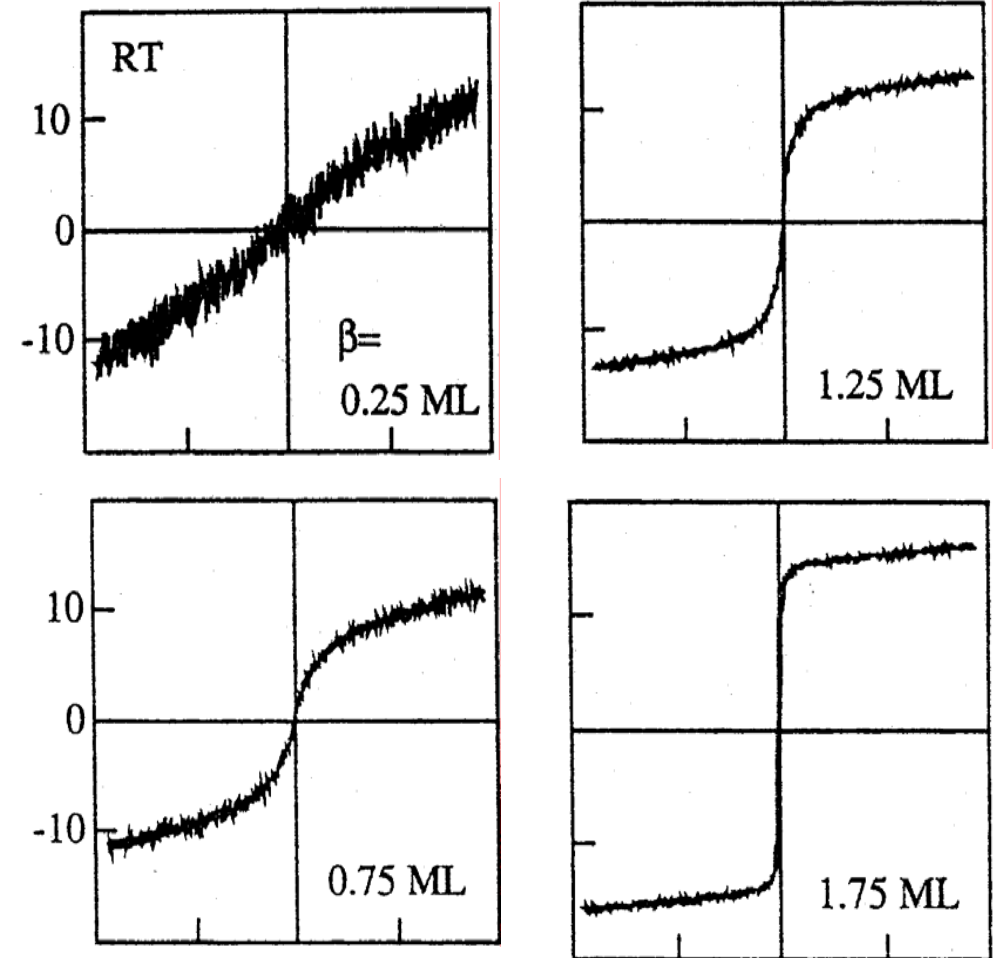
- Yields an estimation of the magnetic moment per particle μ
- Fitting with inadequate functions yields errors on μ

Example: Co/Au dots

350 x 350 nm



O. Fruchart et al.



H. Takeshita et al., JMMM **165**, 38 (1997)

Curie temperature versus blocking temperature?

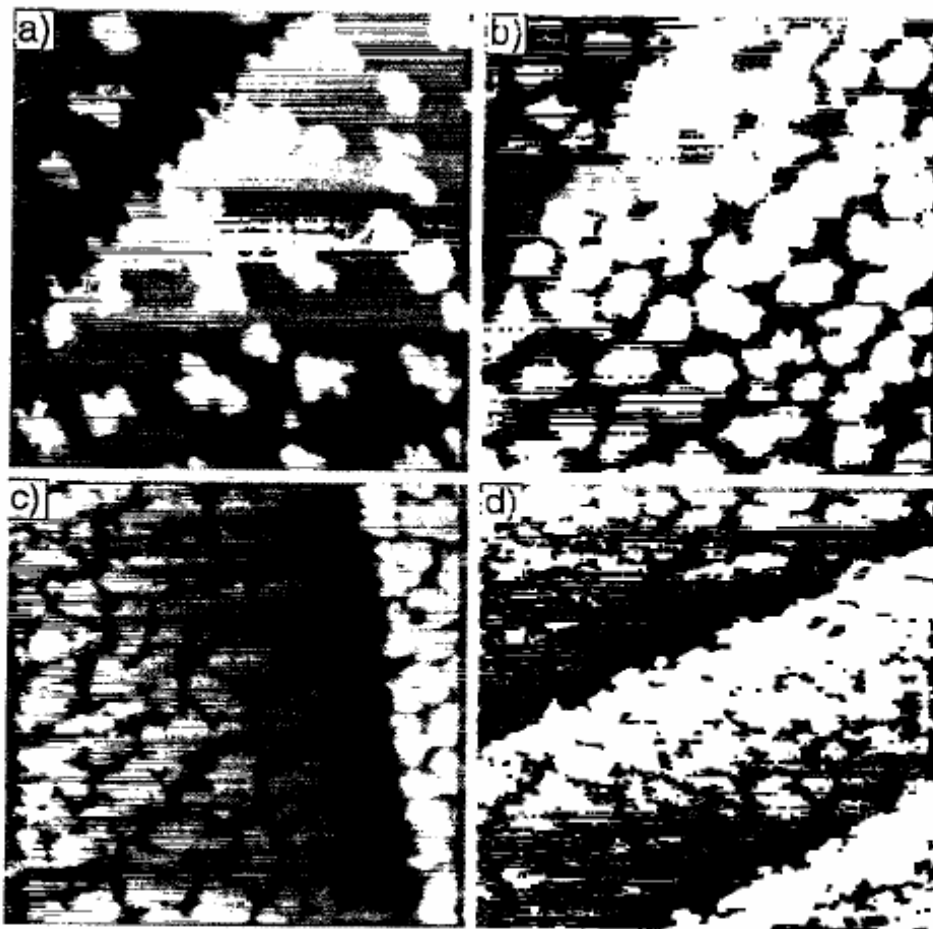
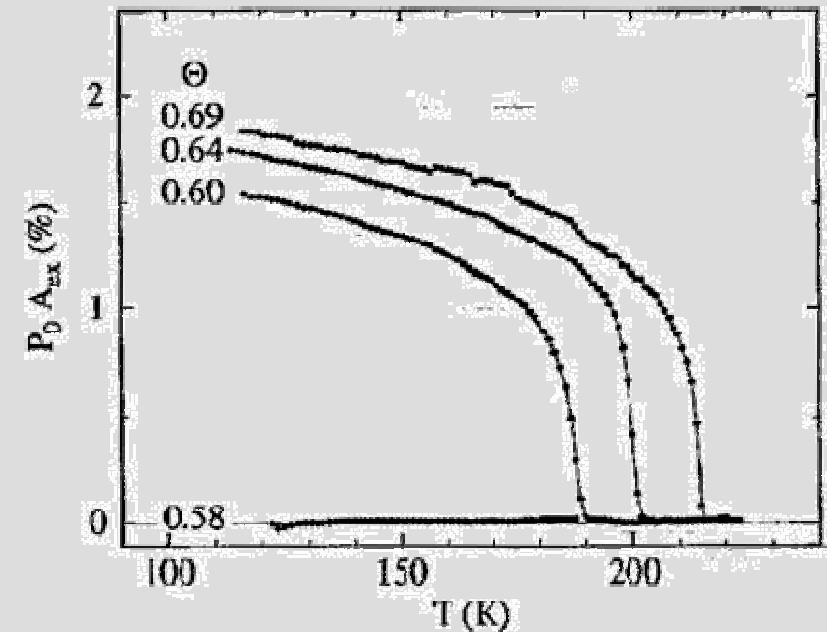


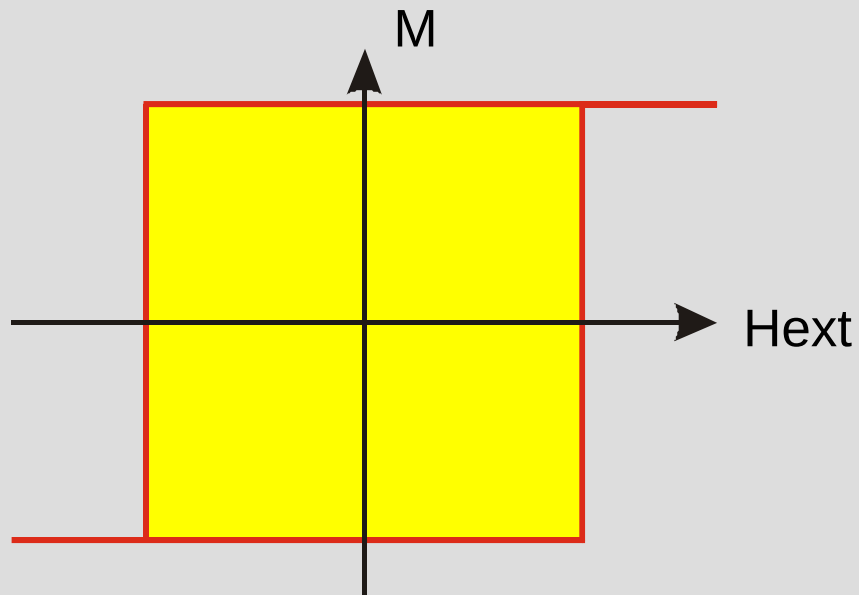
FIG. 1. STM images of Fe(110) films, prepared at RT on W(110), all $70 \text{ nm} \times 70 \text{ nm}$ in size, with [001] horizontal. (a) $\theta = 0.23$; (b) $\theta = 0.53$; (c) $\theta = 0.66$; (d) $\theta = 0.85$. Upper levels (Fe) bright; lower levels (W) dark.

Remanence plotted as a function of temperature

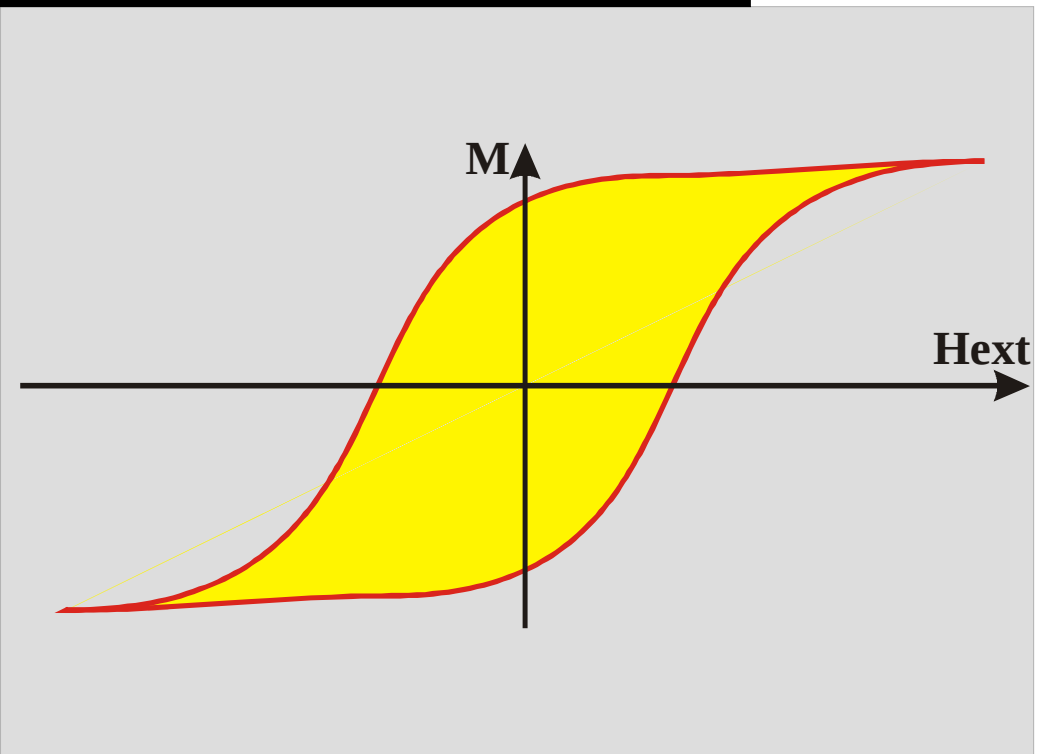


Transition from superparamagnetism to a blocked state

Expected hysteresis loop for macrospins



Hysteresis for assemblies of dots



Possible effects

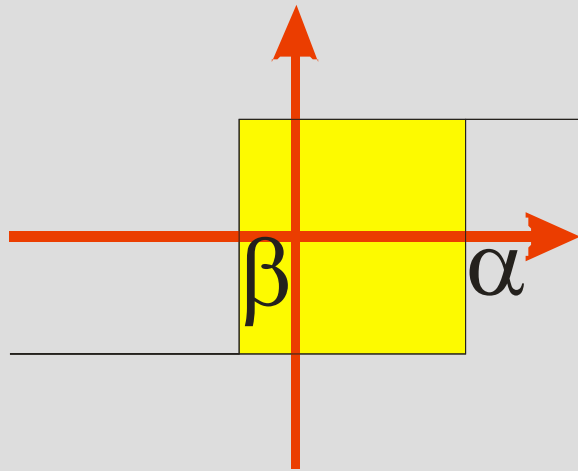
- Distribution of coercive fields
- (Dipolar) interactions



Preisach model

G. Biorci et al., Il Nuov. Cim. VII, 829 (1958)

I. D. Mayergoyz, Mathematical models of hysteresis, Springer (1991)

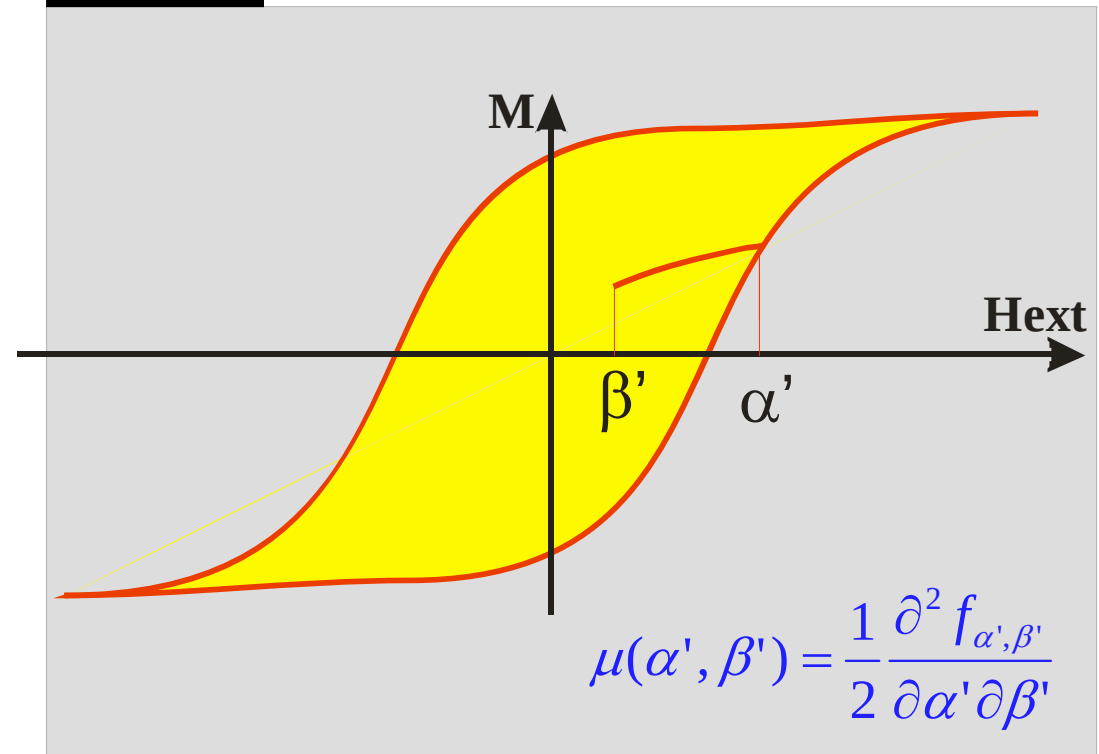


↪ Distribution function

$\mu(\alpha, \beta)$ with $\alpha > \beta$

↪ No true link between real particles and μ

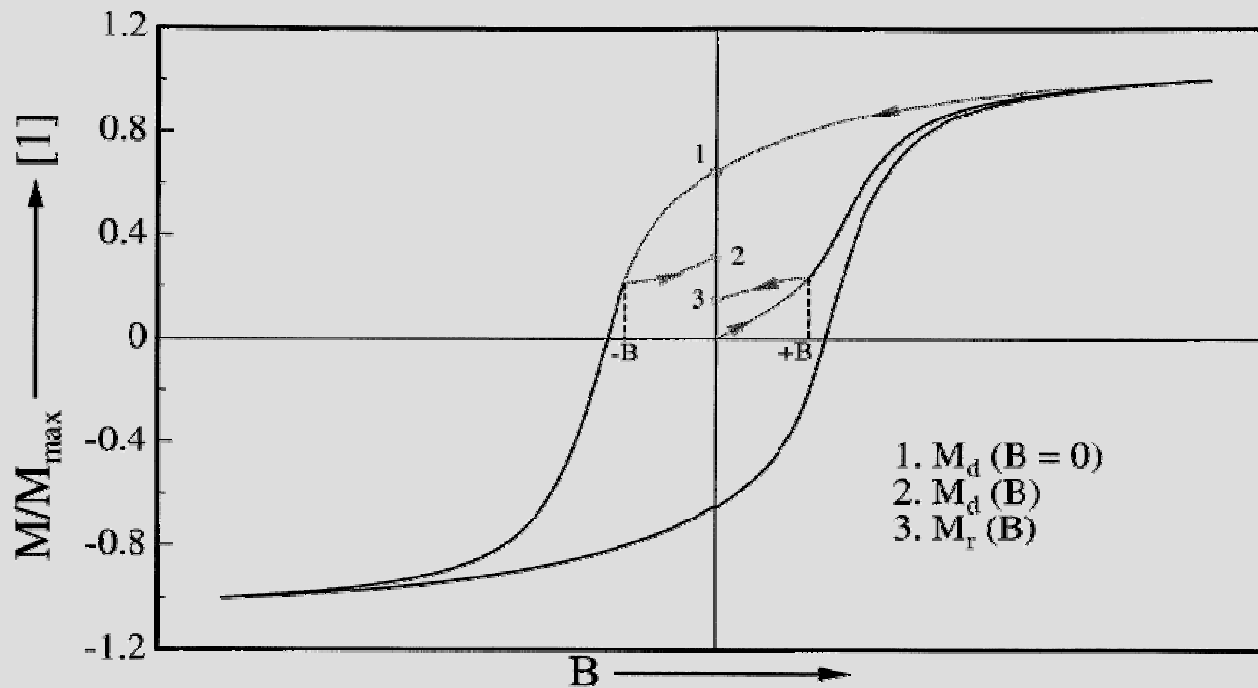
Solving



- Long experiments (1D set of hysteresis curves)
- Better suited to bulk materials with strong interactions



Henkel plots



O. Henkel,
Phys. Stat. Sol. 7, 919 (1964)

S. Thamm et al.,
JMMM184, 245 (1998)

Fig. 1. Explanation of how to measure the two different remanent magnetisations M_r and M_d .

Measure of dipolar interactions

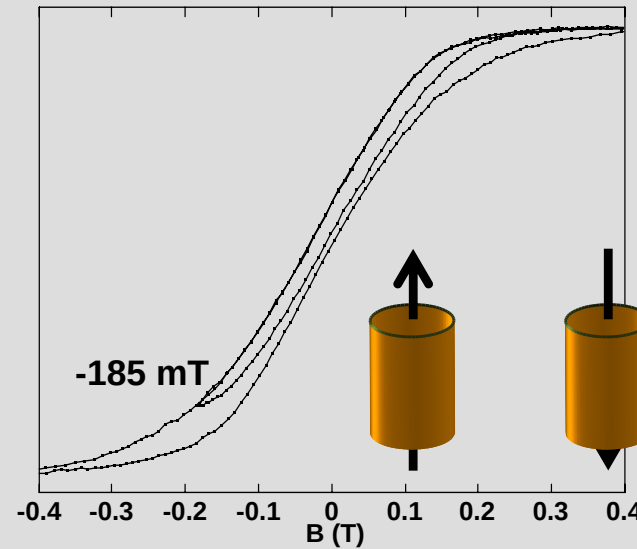
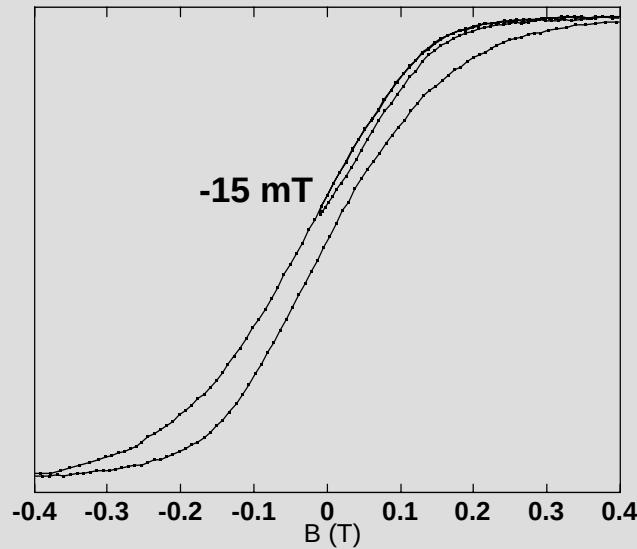
$$\Delta M_H(x) = M_d(x) - [1 - 2M_r(x)]$$

- Long experiments (ac demagnetization)
- Better physical meaning than Preisach

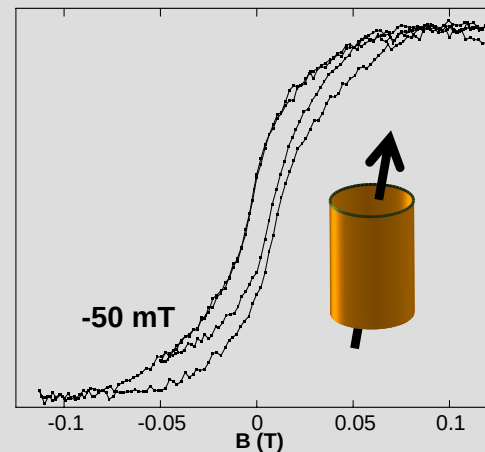
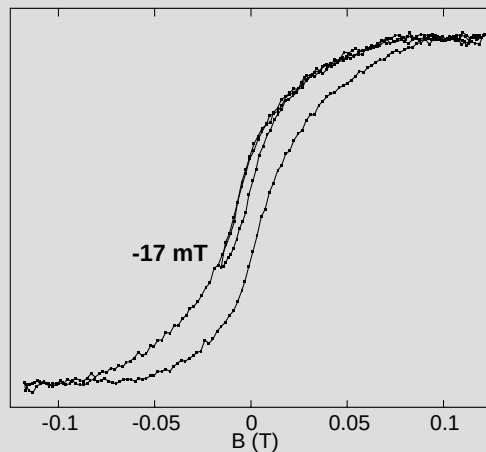


Minor loops: negative interactions

Example: dipolar interactions in arrays of Co/Au(111) pillars

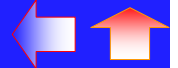


Minor loops: negligible interactions



- Faster than Henkel
- Other applications: characterization of exchange bias

O. Fruchart et al., unpublished



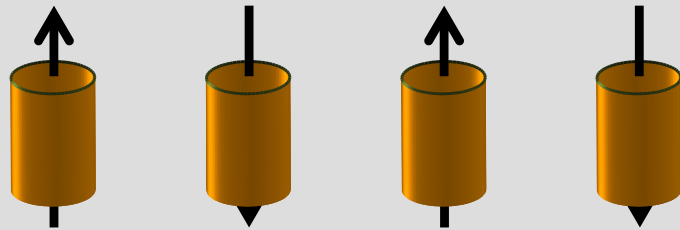
Superparamagnetic regime: plot of inverse susceptibility

- Brillouin 1/2 function

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

- Effective field

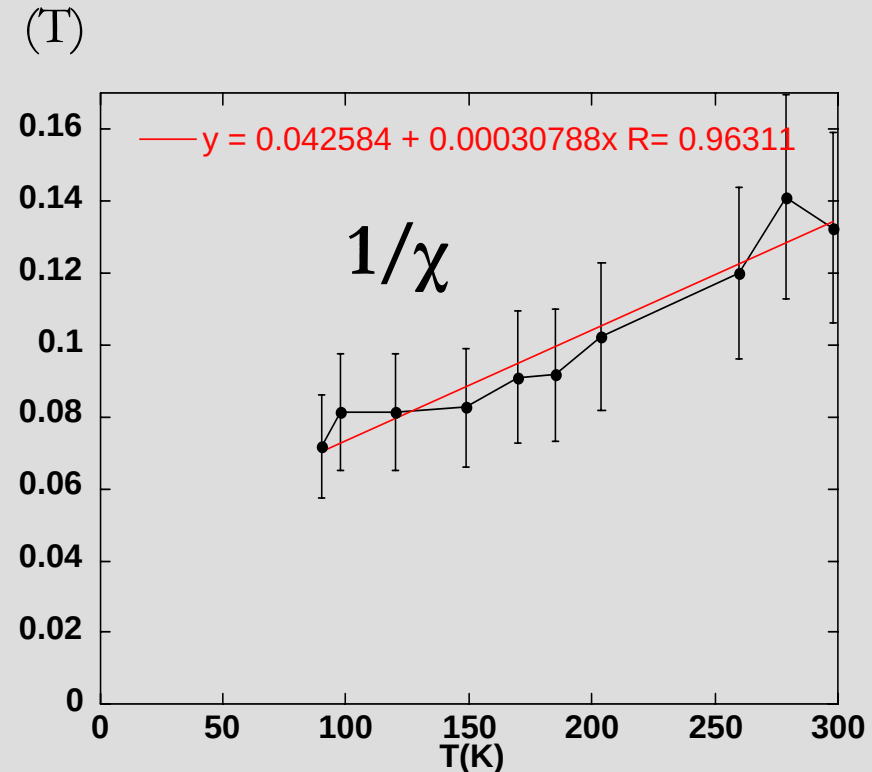
$$H_{eff.} = H + r M_s m$$



(Demagnetizing dipolar interactions)

- First order expansion:
susceptibility

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



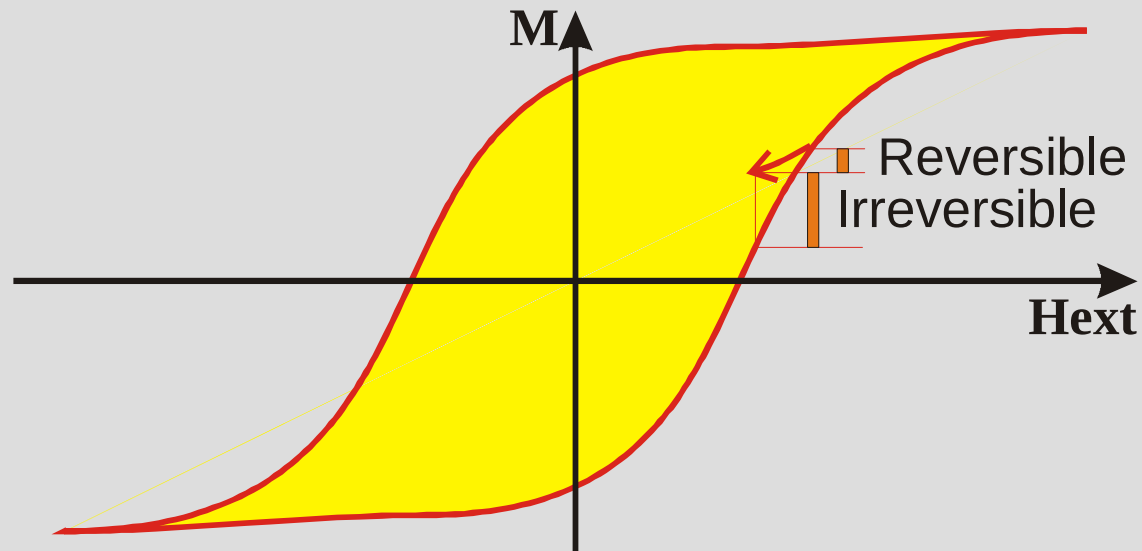
O. Fruchart *et al.*, PRL 23, 2769 (1999)

➤ No need of hysteresis

➤ Analogy with Curie-Weiss law



Distribution of properties

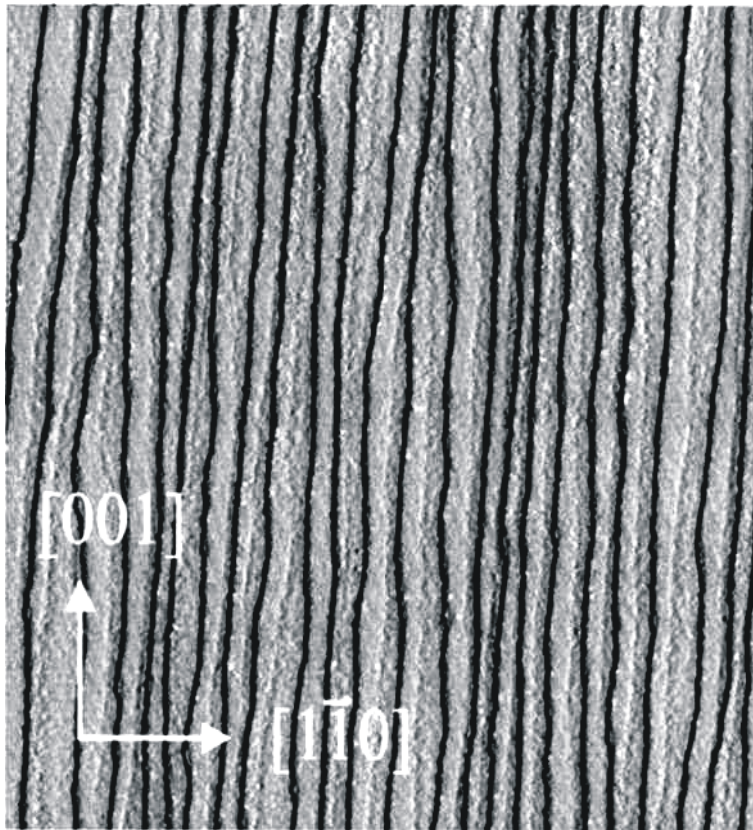


$$\rho(H_r) = \left. \frac{dm}{dH} \right|_{\text{irreversible}}$$

$H_c(T)$ for a given population of the distribution can be studied at a given stage of the reversal (10 %, 20 % etc.)

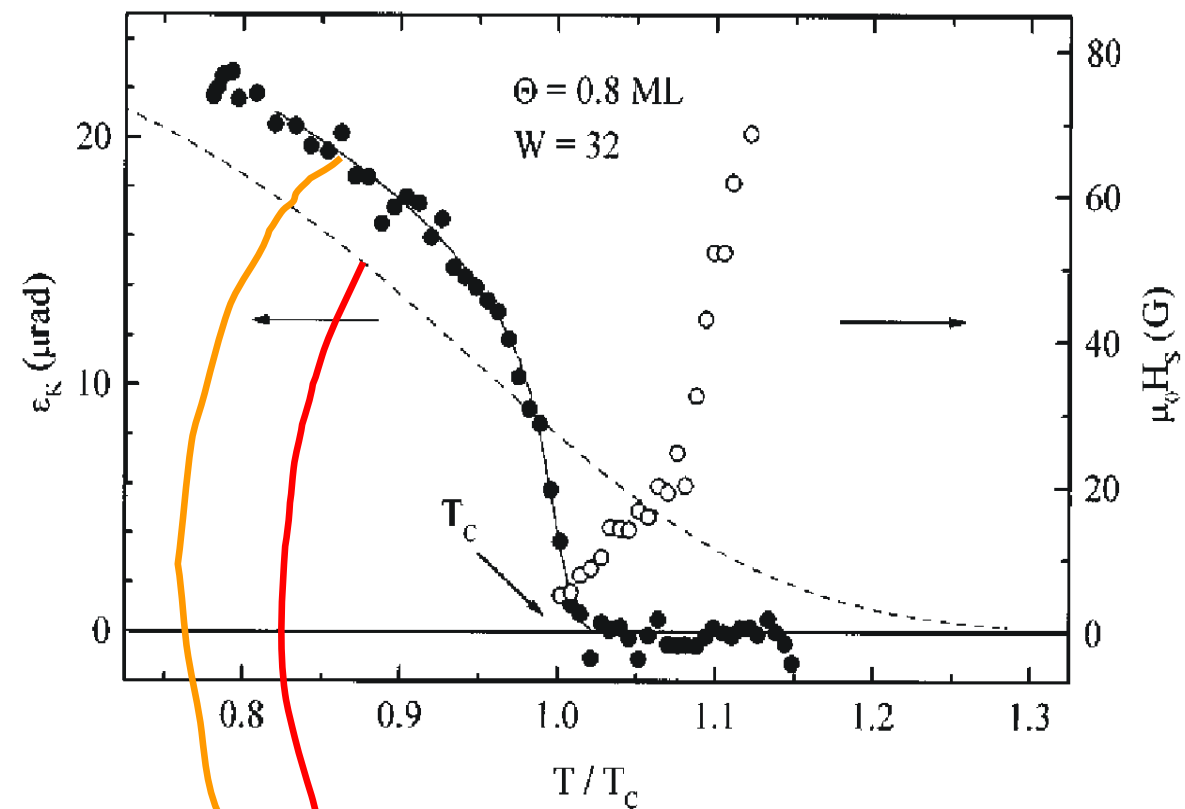
Effect of distributions and dipolar interactions
are sometimes difficult to disentangle

Ferromagnetic order stabilized by dipolar interactions



J. Hauschild et al., Phys. Rev. B 57, R677 (1998)

$T_c = 179\text{K}$



Simulation without dipolar interactions

Simulation with dipolar interactions

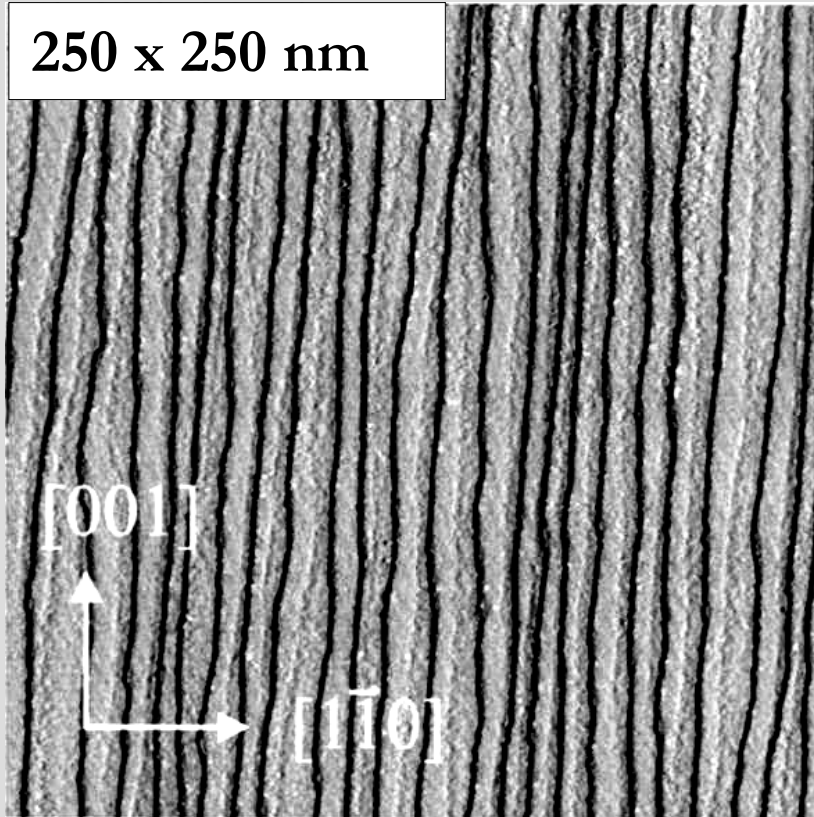
Although weak, dipolar interactions stabilize ferromagnetic order and sharpen the transition at T_c , owing to their long range



Magnetic order in 1D

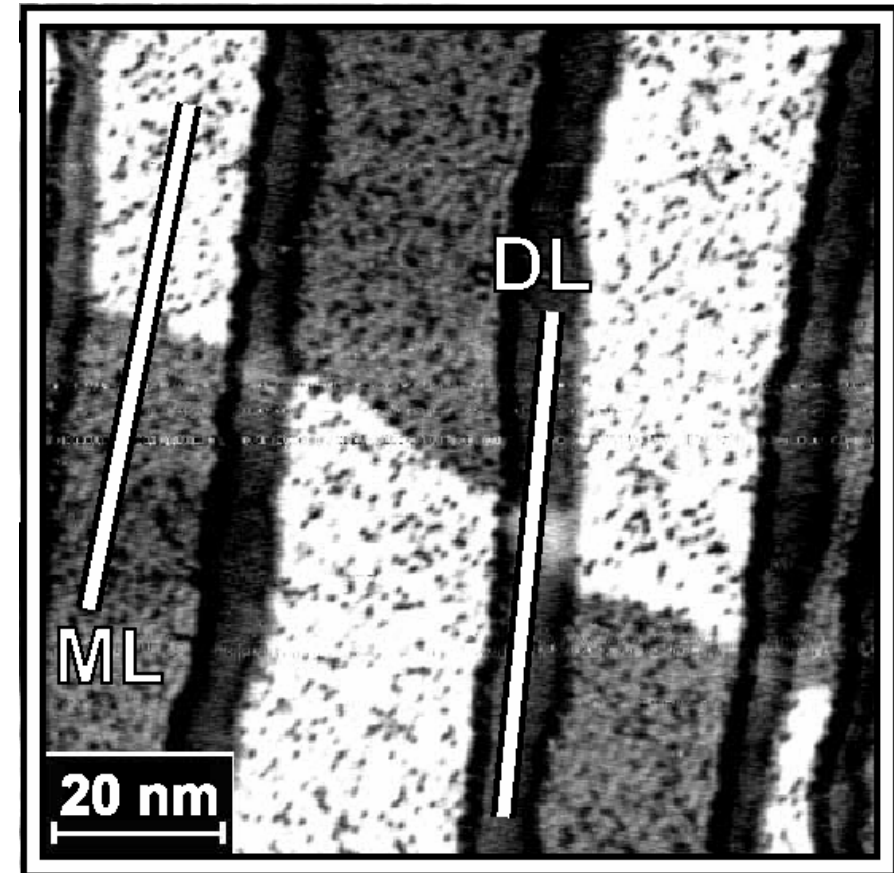
Fe(0.5ML) / W(110)

250 x 250 nm

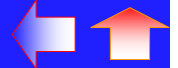


J.Hauschild *et al.*, Phys.Rev.B57, R677(1998)

Vicinal Fe(110)[1.2ML]/W(110)

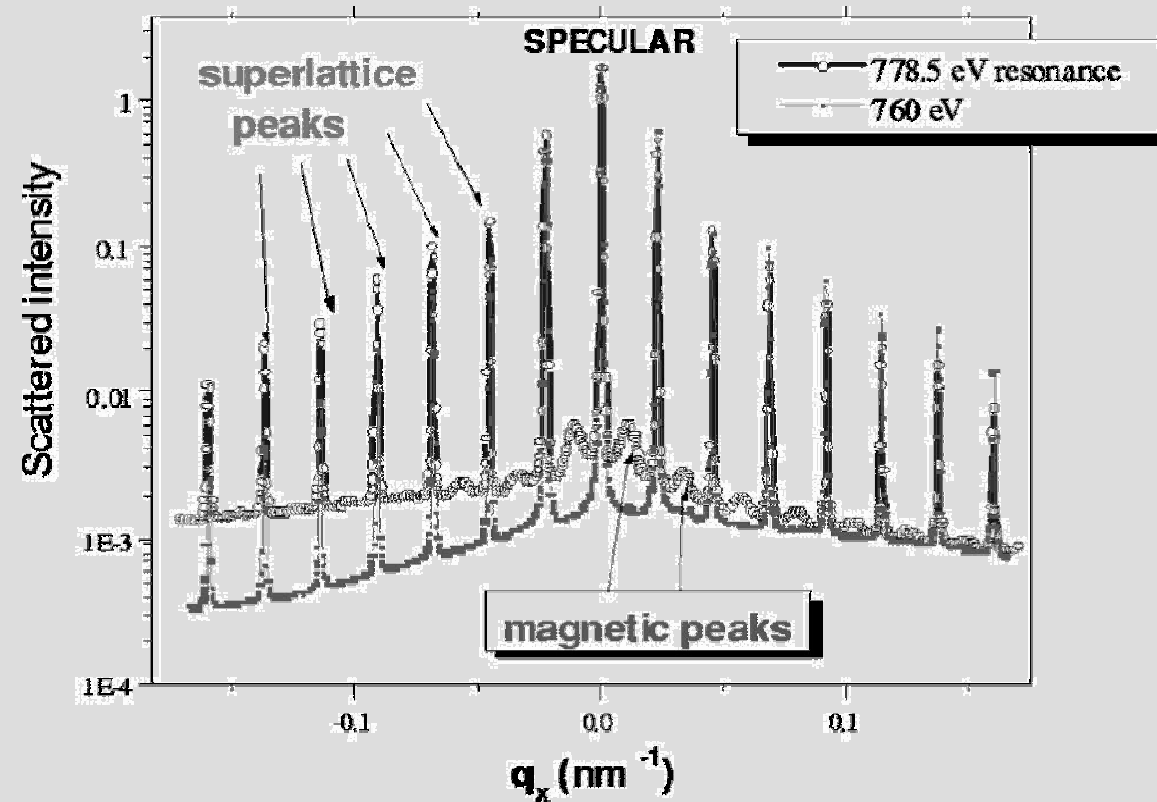
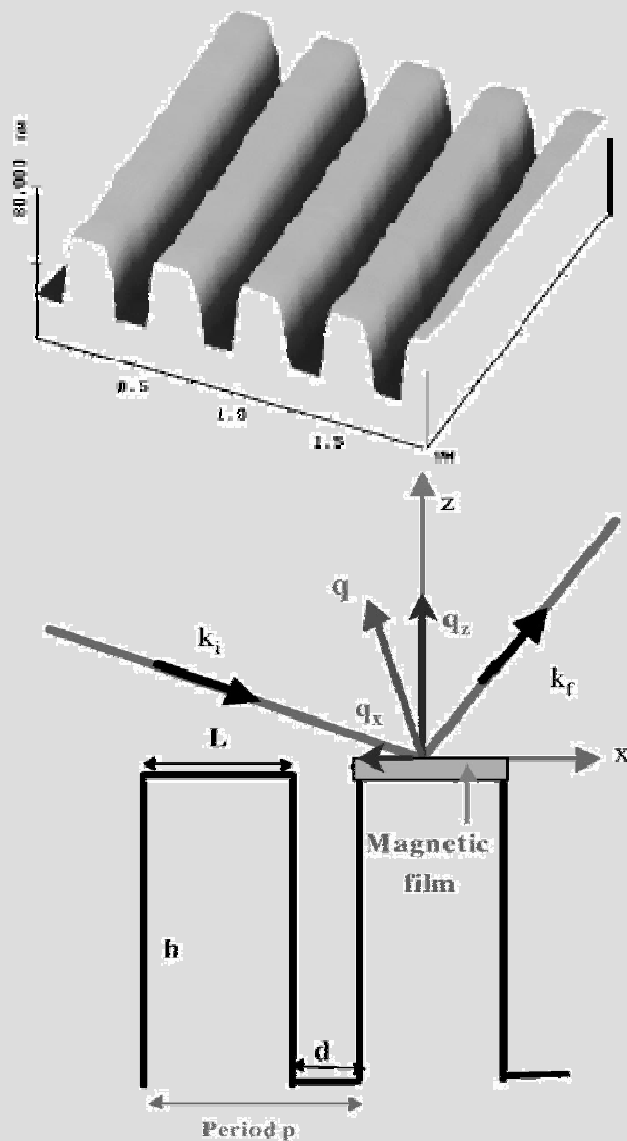


M. Pratzner *et al.*, PRL87, 127201 (2001)

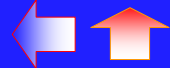


Magnetic scattering on nanofabricated arrays of lines

Co/Pt(111) multilayers

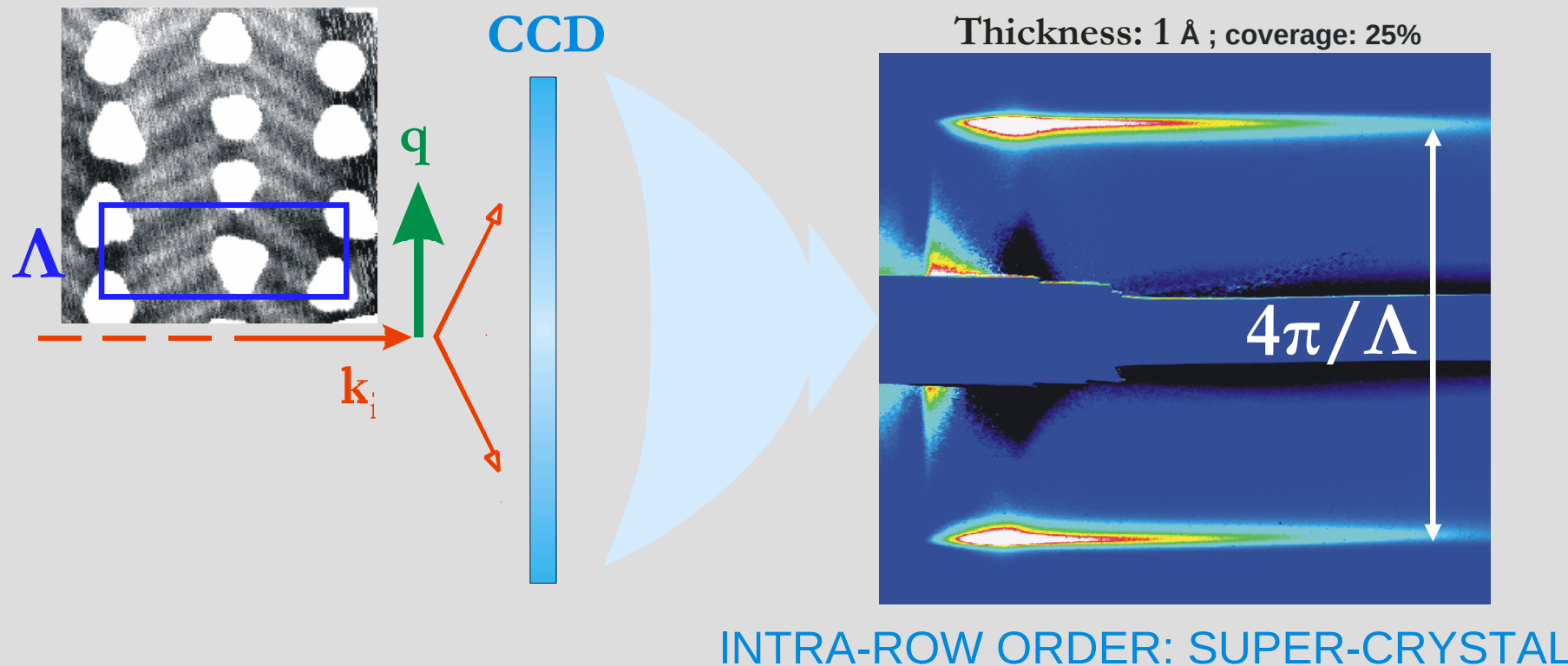


Informations about magnetic correlations
can be extracted from magnetic satellites



Structural scattering on self-organized systems

Co/Au(111) nanodots



O. Fruchart et al., Europhys. Lett. 63, 275 (2003)

Evidencing magnetic scattering is in principle possible, however:

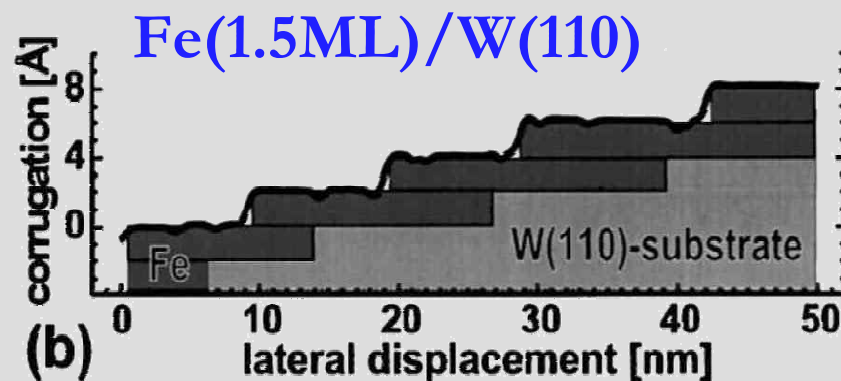
- Low temperature and weak signal
- Distribution of properties may prevent ordering to occur



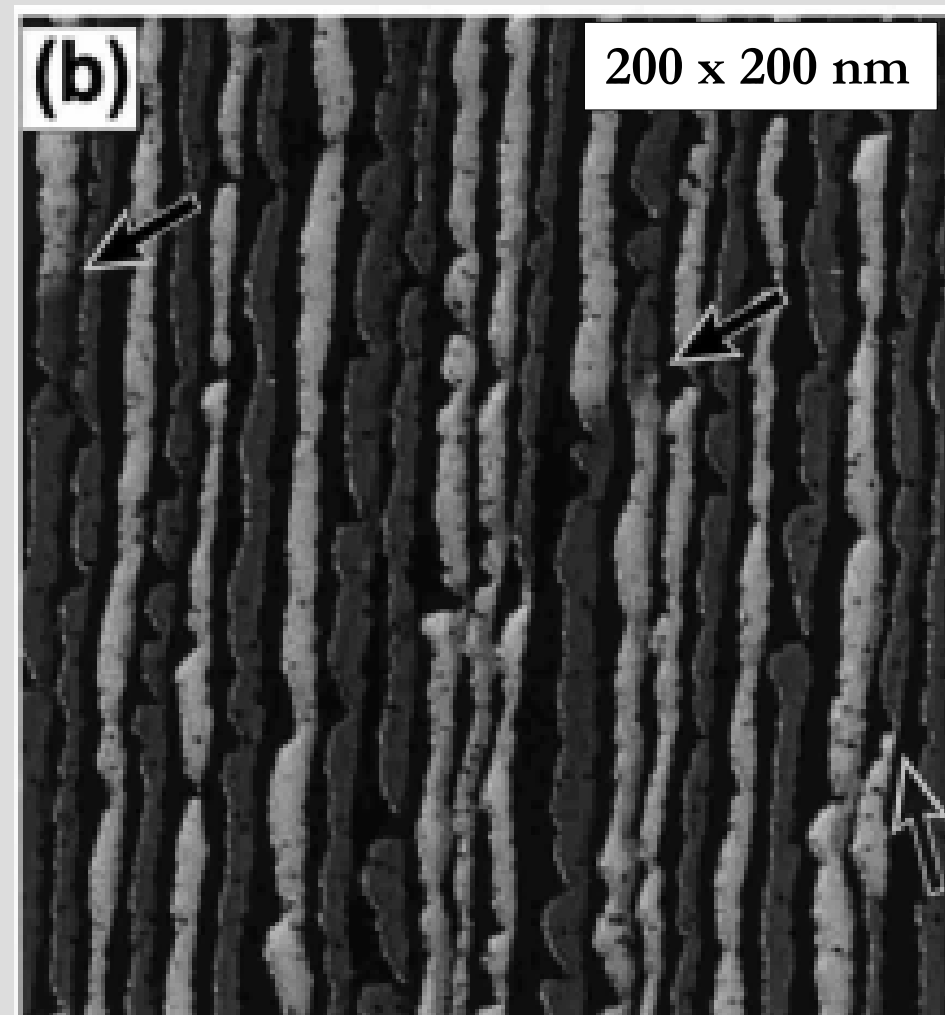
Magnetic properties of self-organized systems : TOC

- ➔ **1. Introduction to magnetism**
- ➔ **2. Individual properties**
- ➔ **3. Collective properties**
- ➔ **4. Towards materials?**

Domain walls in geometrical constriction



Spin-Polarized
Scanning
Tunneling
Spectroscopy



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

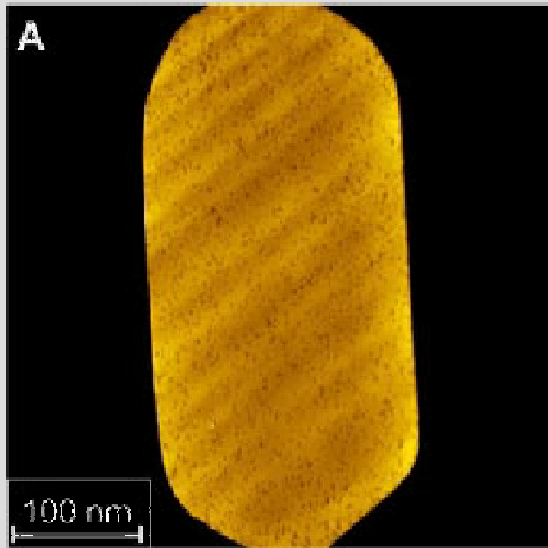
Theoretical prediction: P. Bruno, Phys. Rev. Lett. 83, 2425 (1999)



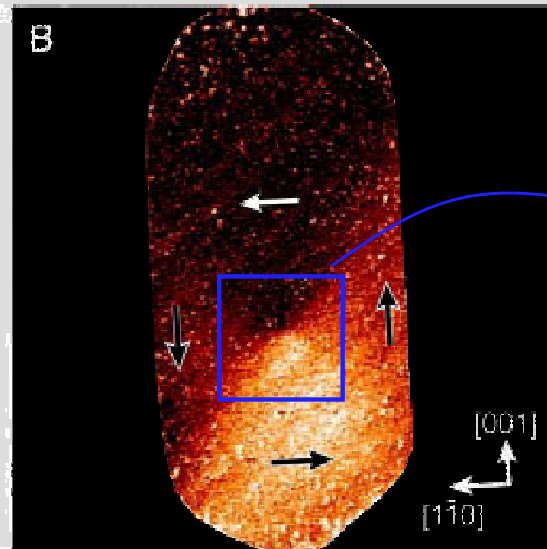
Magnetic vortex core

Spin-polarized Scanning Tunneling Microscopy on Fe/W(110) self-assembled dots

Structural image

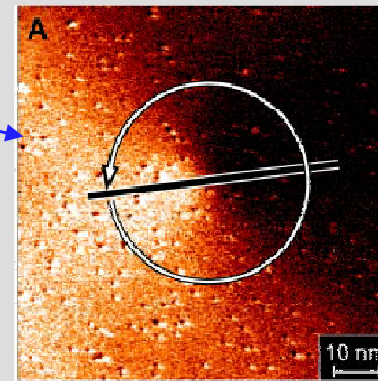


Magnetic image

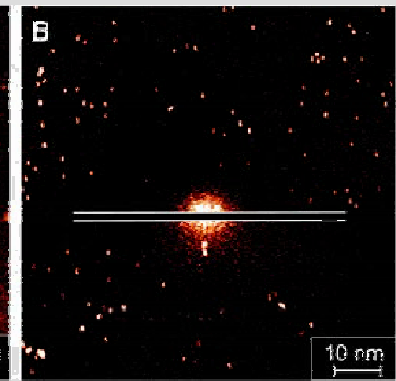


A. Wachowiak, Science 298, 577 (2002)

In-plane component of magnetization

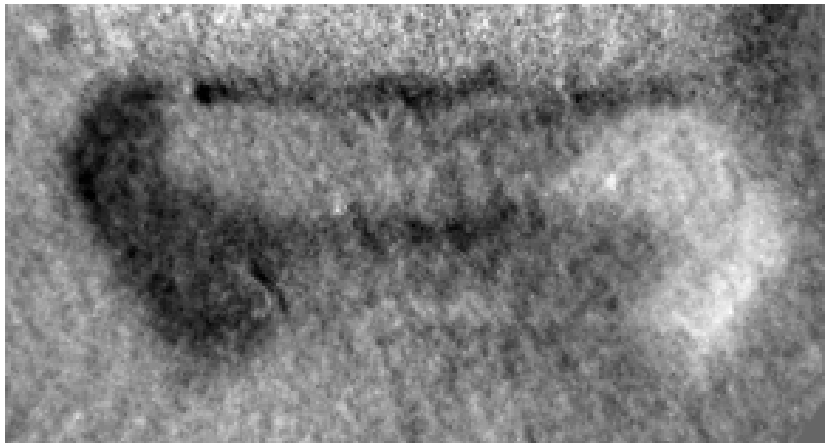
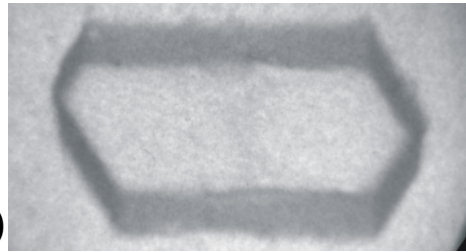


Out-of-plane component of magnetization

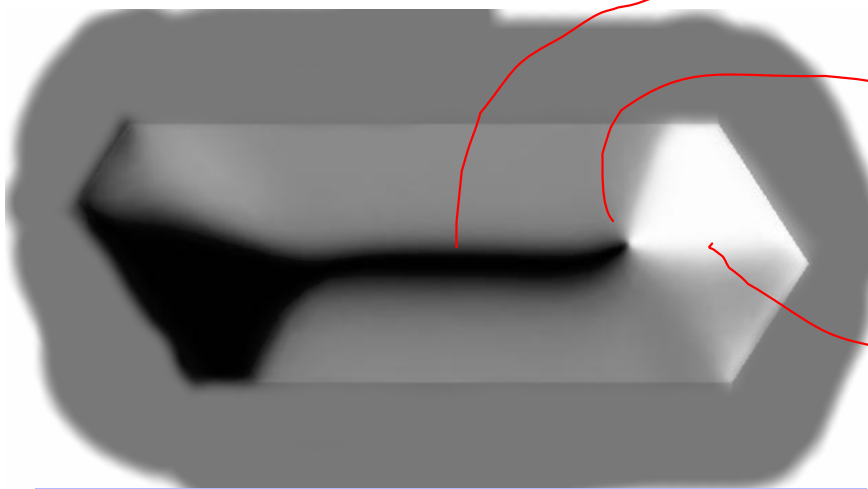


...also in-field study

Experiment (top)



Simulation (top with facets)



Néel cap displaced from median line

Asymmetry around vortex

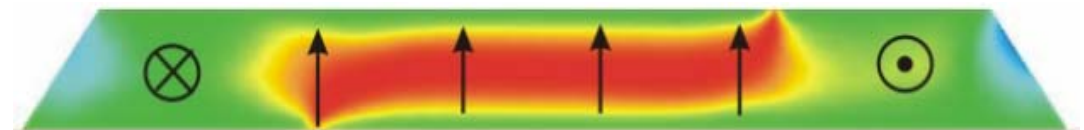
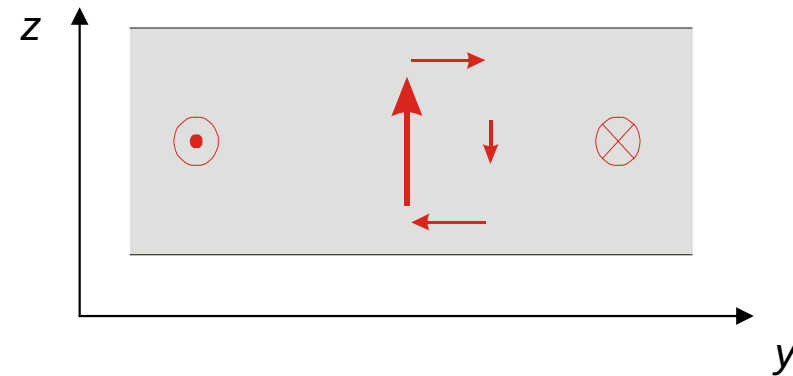
Strong asymmetry of end domains

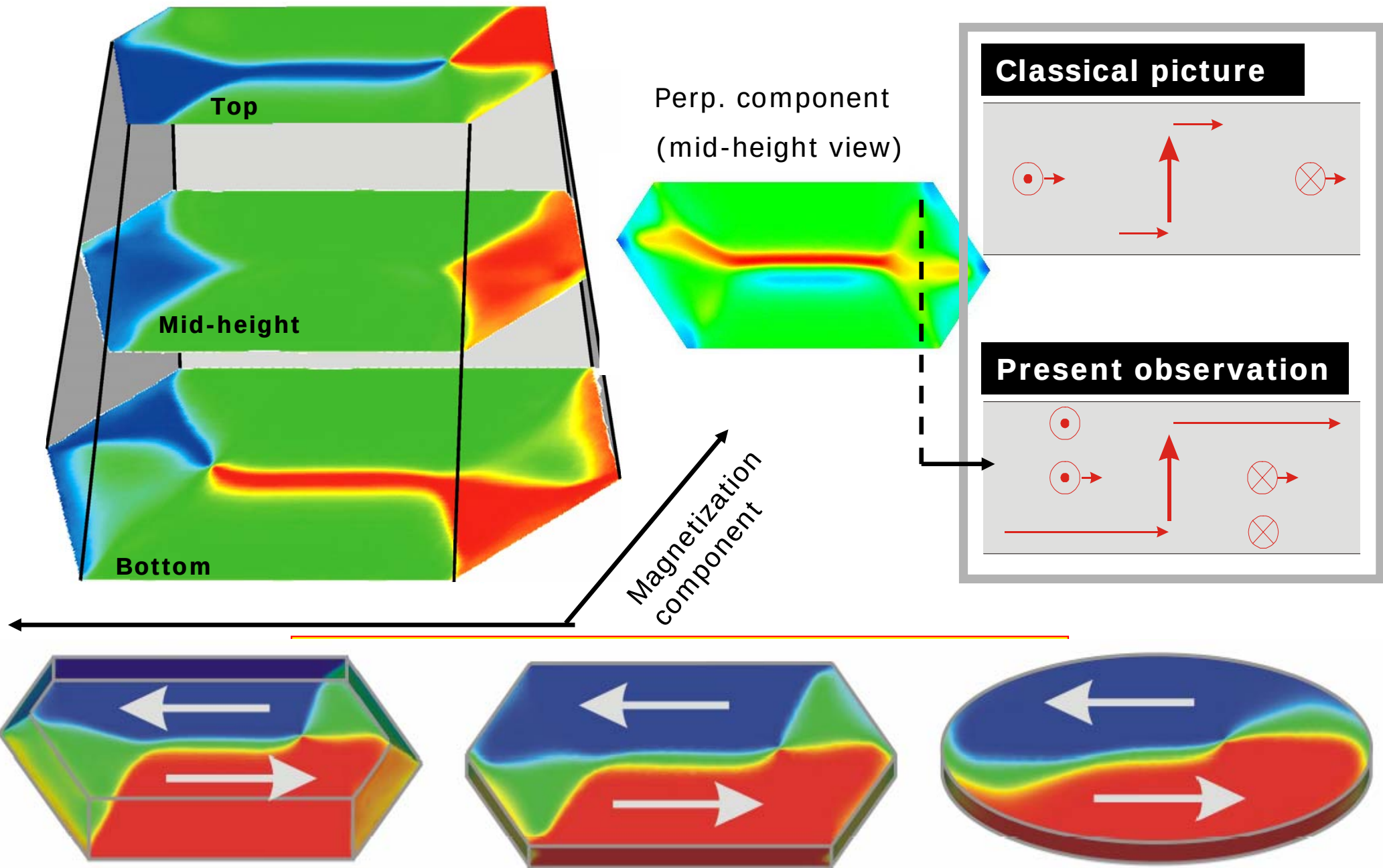
Pseudo-2D

3D



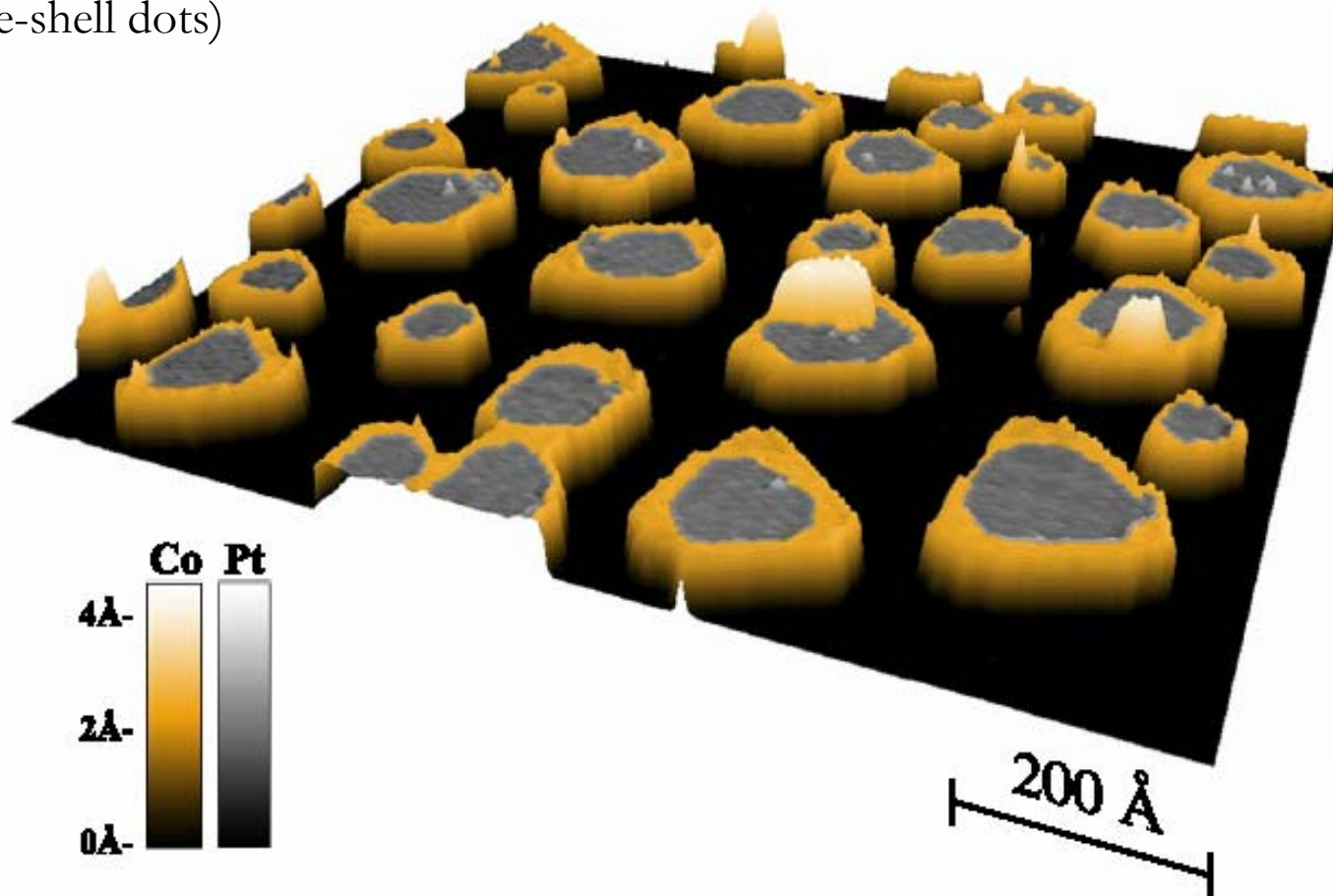
The main features are reproduced





Manipulation of edge anisotropy Pt brims around Co/Pt(111)

(core-shell dots)

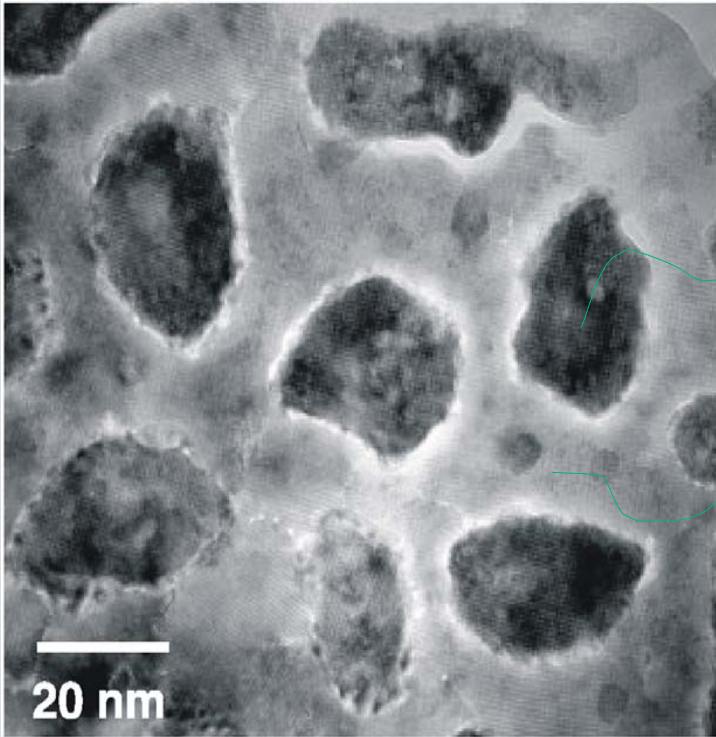


S. Rusponi et al., Nature Mater. (2003)

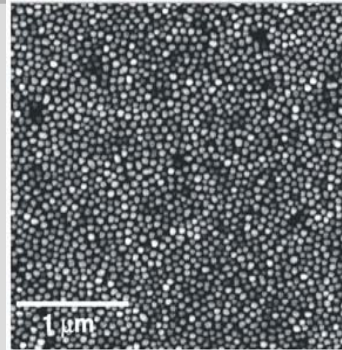
- **Self-organization : concepts**
- **Self-assembly : towards materials?**

Structure

CoFe₂O₄ in BaTiO₃ matrix



Deposition from a Ti-Ba-Co-Fe oxide target by pulsed laser deposition

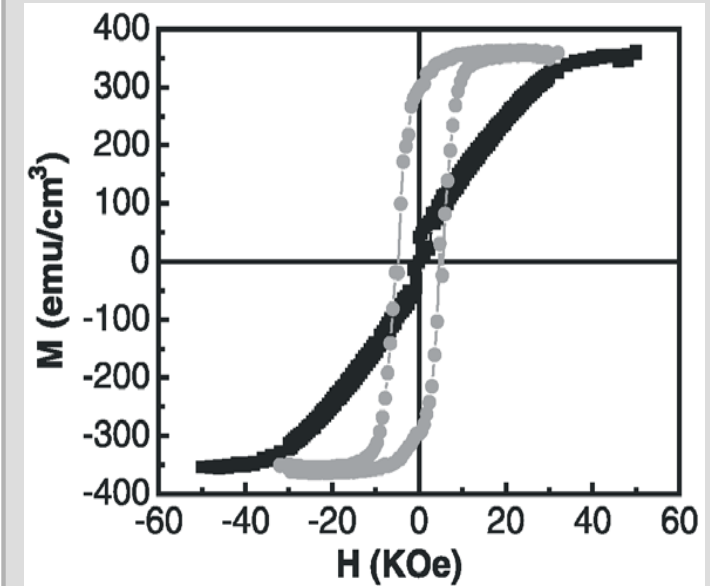


CoFe₂O₄
(ferrimagnetic)

BaTiO₃
(piezoelectric)

Magnetis

m

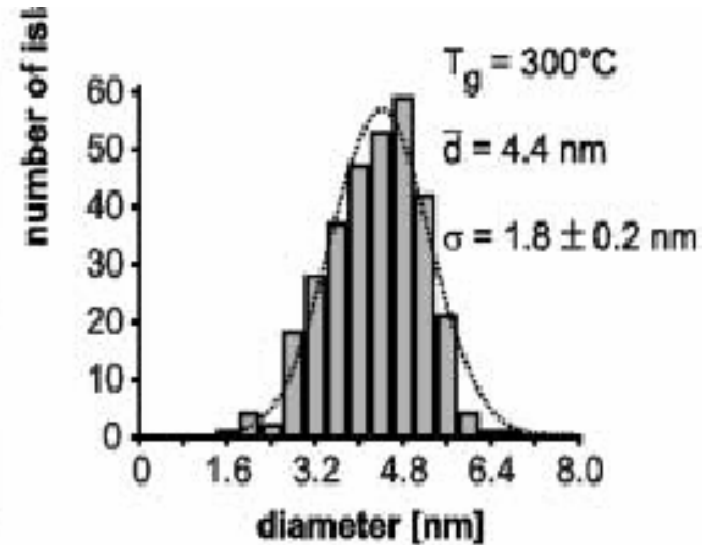
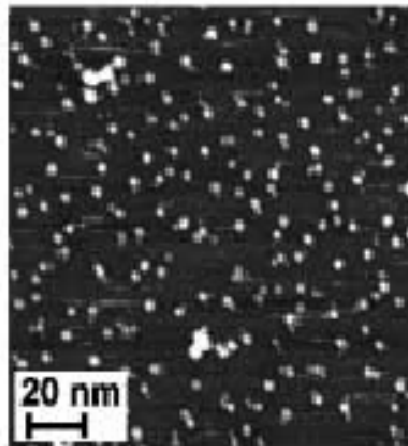


- Room-temperature functionality
- Perpendicular anisotropy owing to matrix-induced strain in the columns

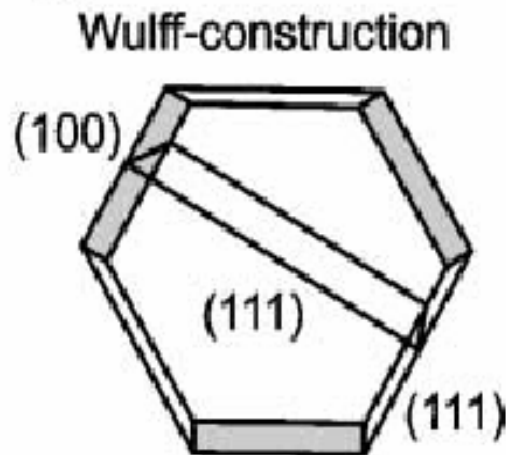
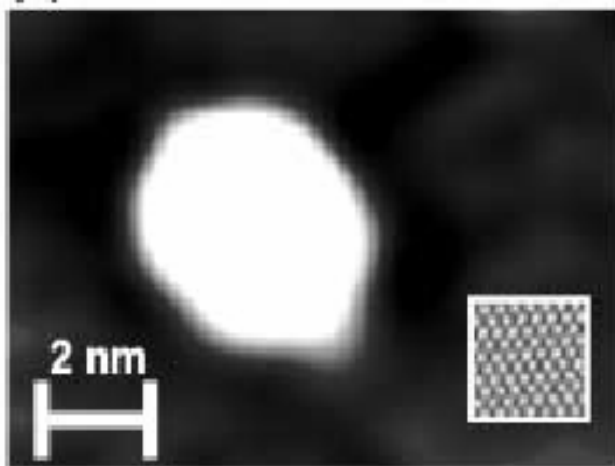
H. Zheng et al., Science 303, 661 (2004)



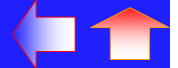
CoPt₃(0.8ML)/WSe₂



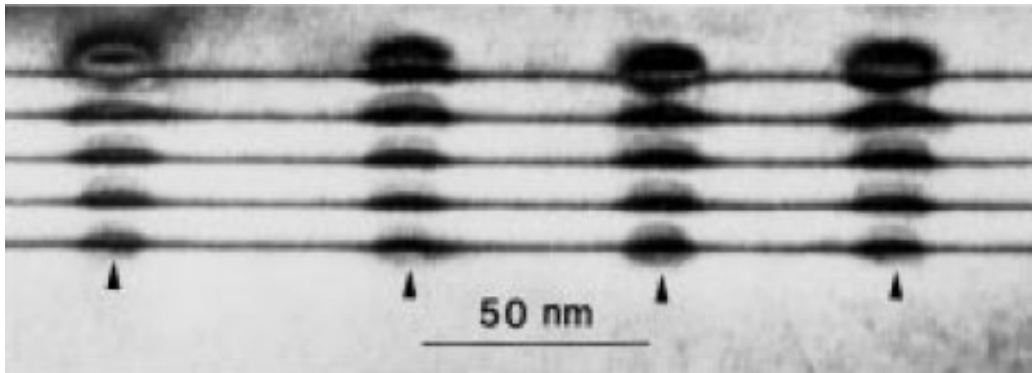
Chemical ordering (L1₂ phase) occurs from 150°C, against 500°C for continuous films
(search for high-anisotropy L1₀ phases)



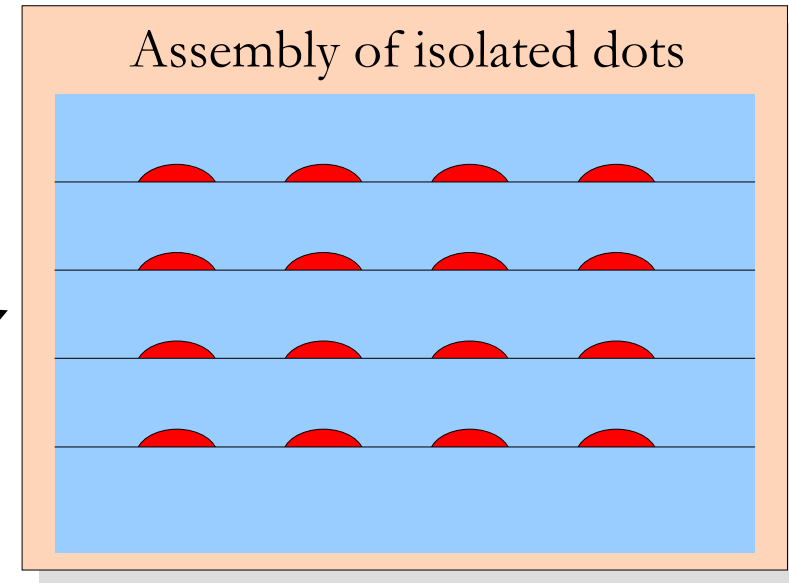
M. Albrecht et al., Europhys. Lett. 56, 884 (2001)



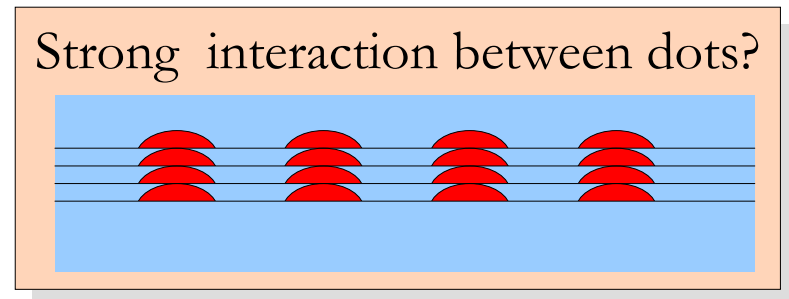
- Vertical 3D self-organization of $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$:



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



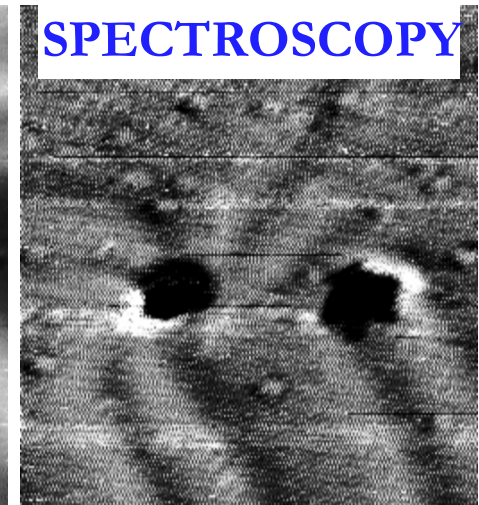
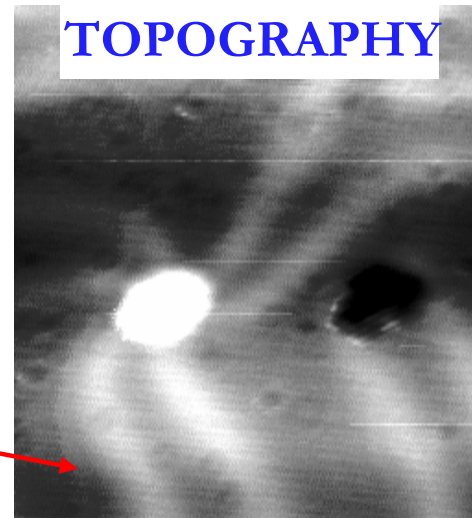
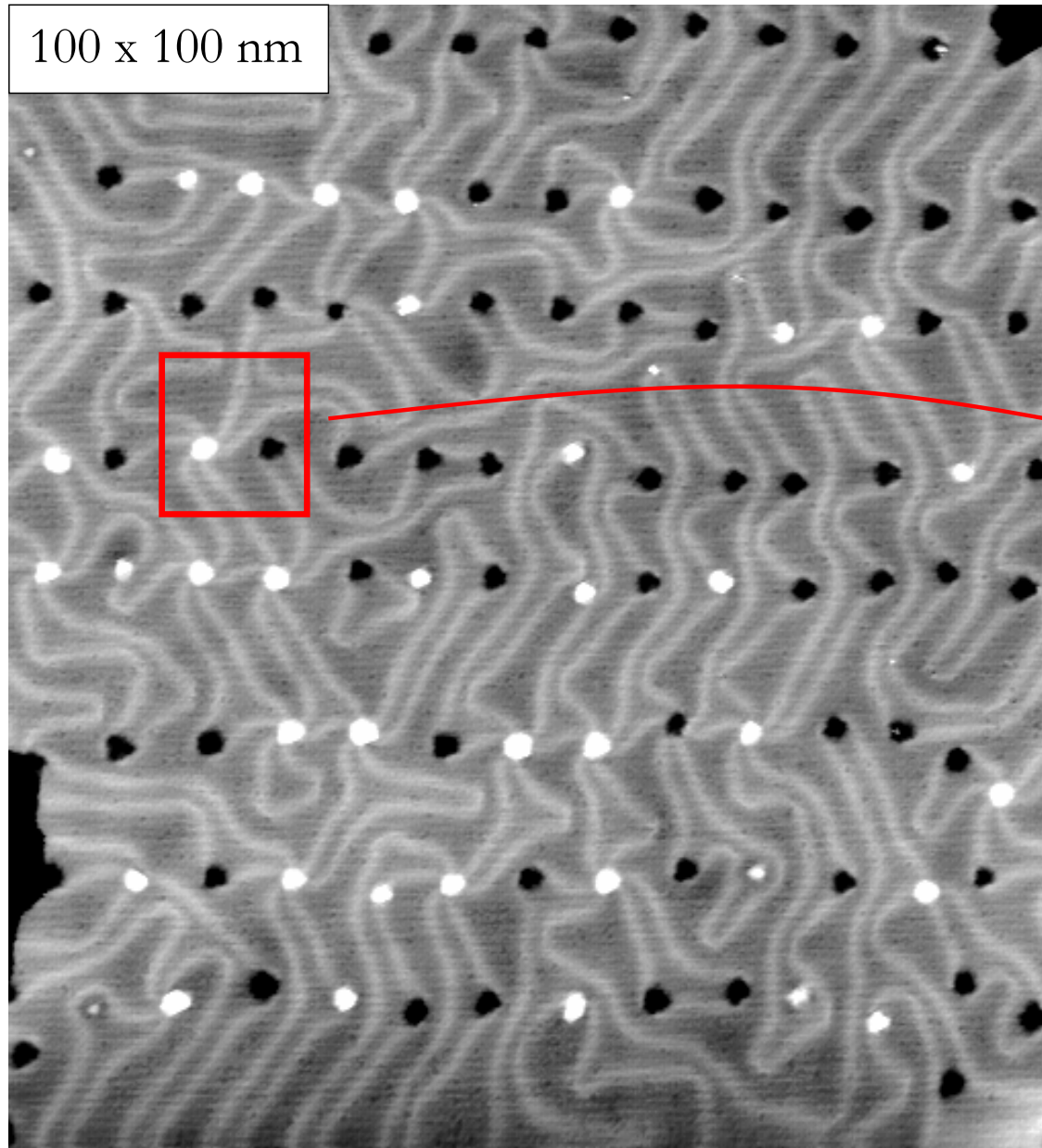
Thinning the spacer layer



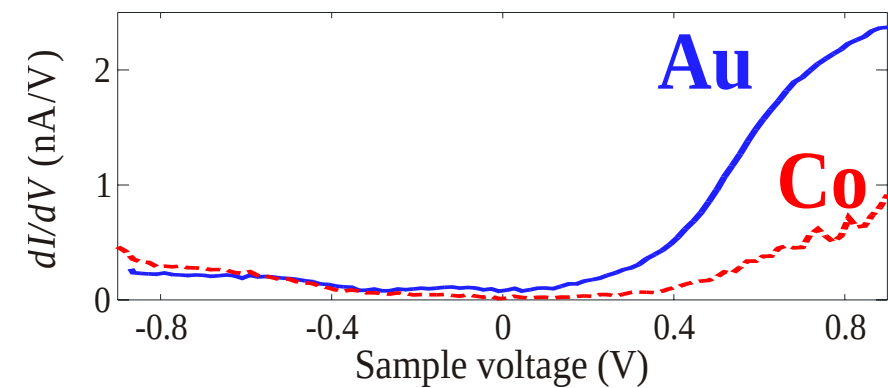
- superparamagnetism overcome ?
- Enhanced magnetic signal



Co/Au(111) pillars



SAME AREA

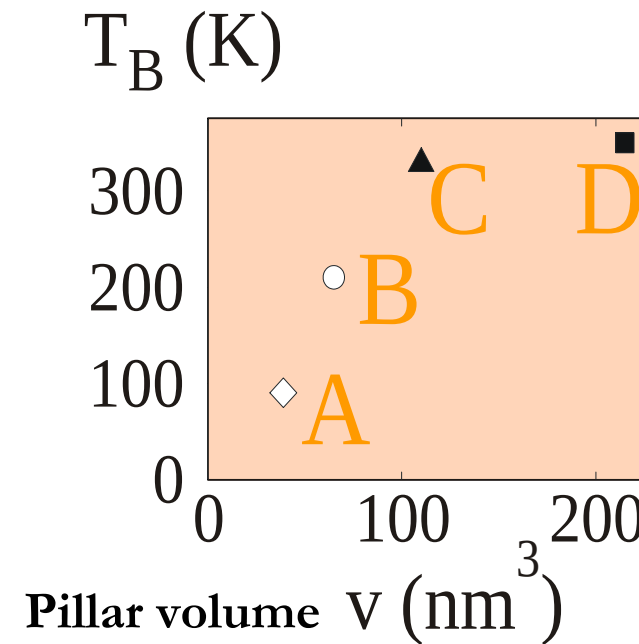
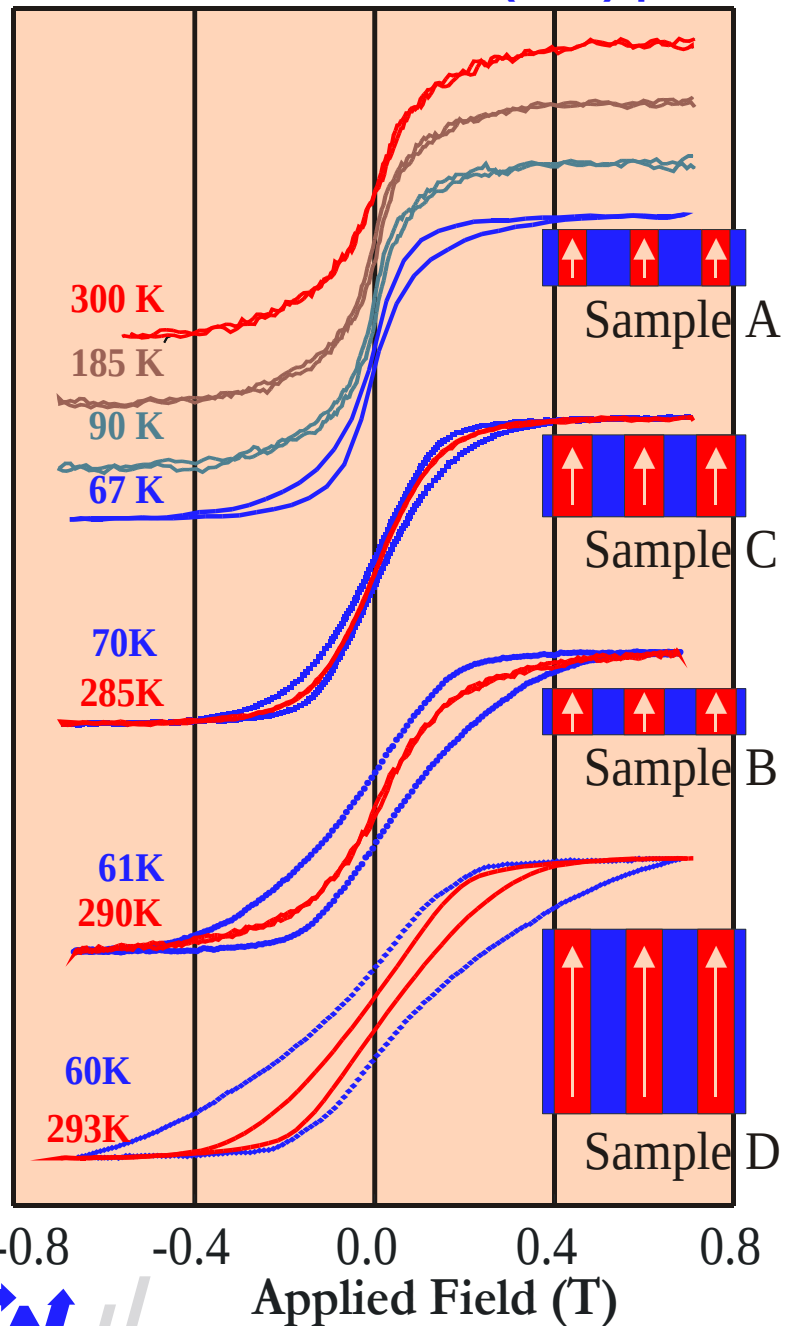


Co atoms grow only on existing dots

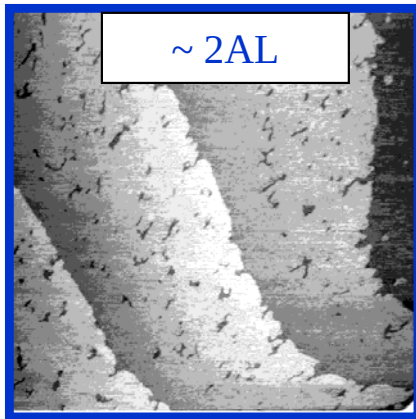
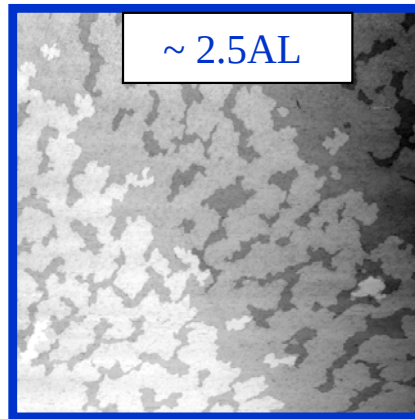
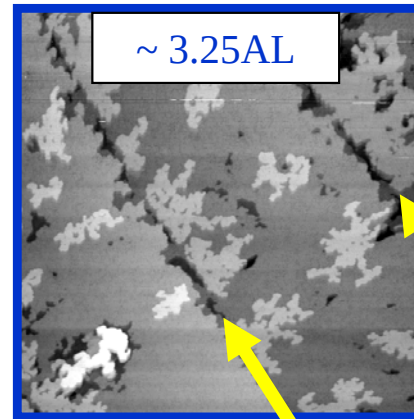
VERTICAL SELF-ORGANIZATION



Co/Au(111) pillars

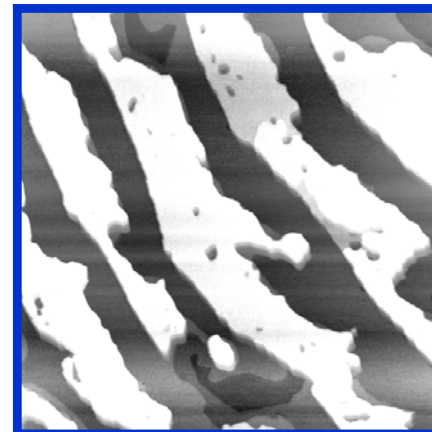
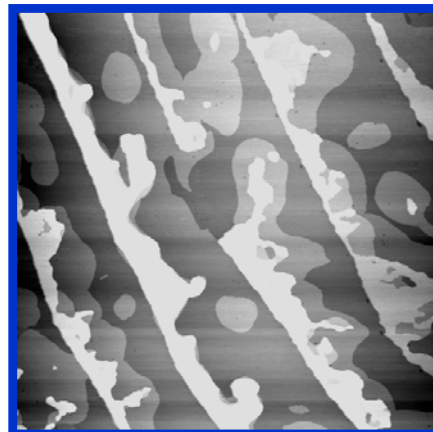
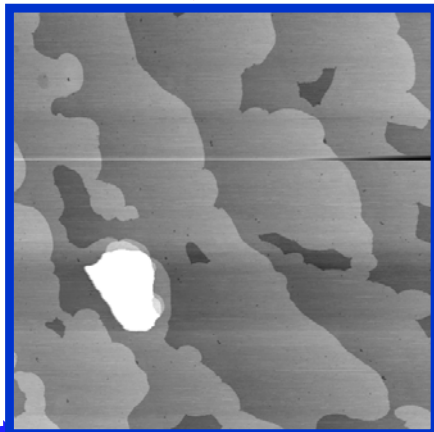


➤ **Blocking temperature > 300K**
(~ 30K for flat Co/Au dots)

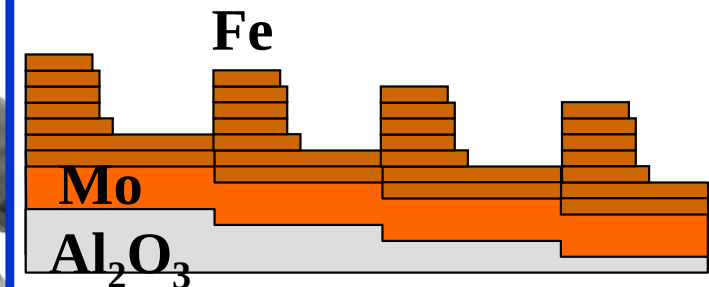
Step 1: layer-by-layer growth at 150°C**Fe / Mo(110) stripes** $\sim 2AL$  $\sim 2.5AL$  $\sim 3.25AL$

Fe films grown at 150°C

STM, 600nm x 600nm

Step 2: annealing at 500°C --> stripes along steps

STM, 750nm x 750nm



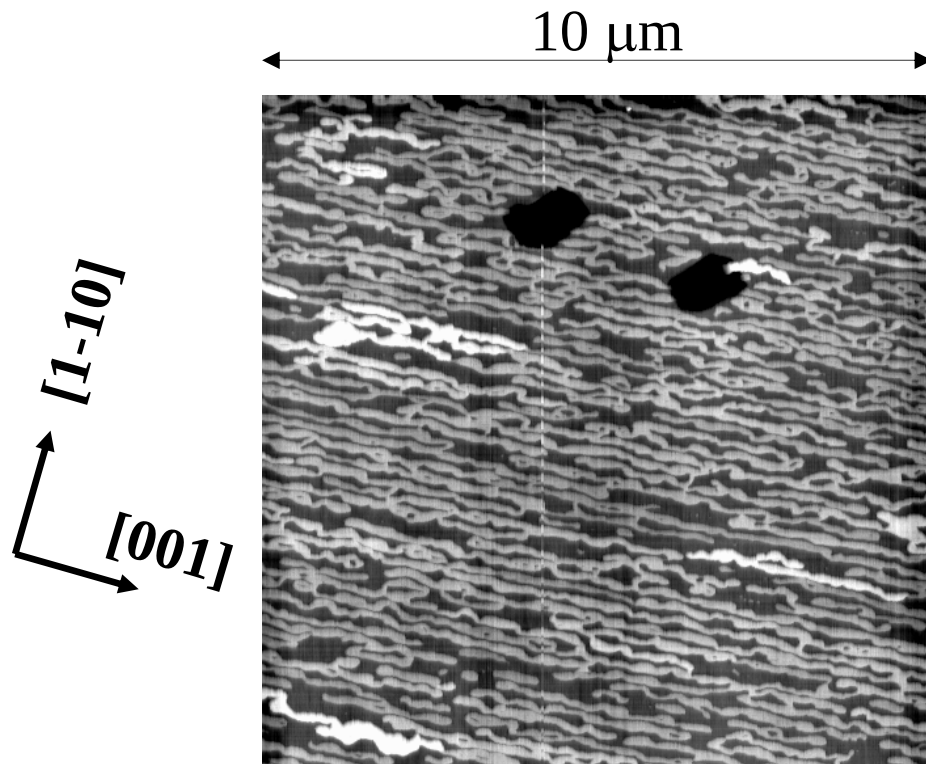


Coercivity and remanence at 300K

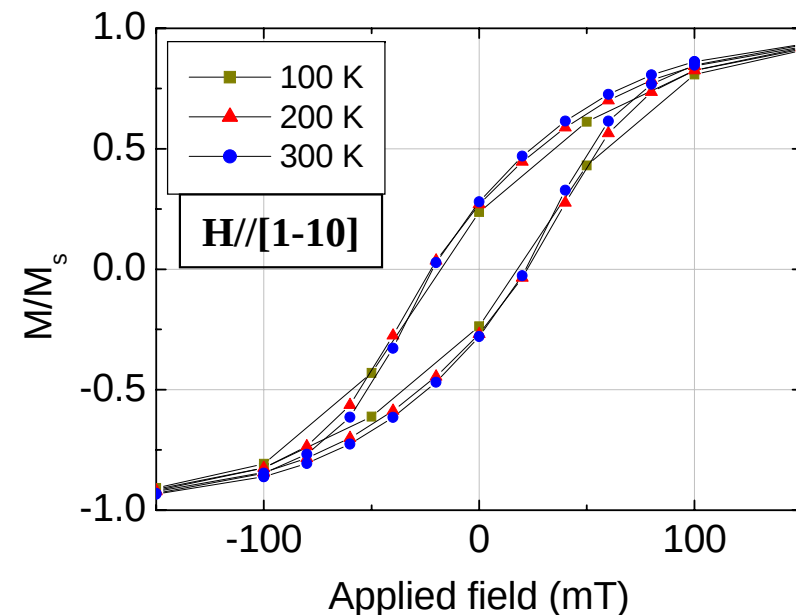
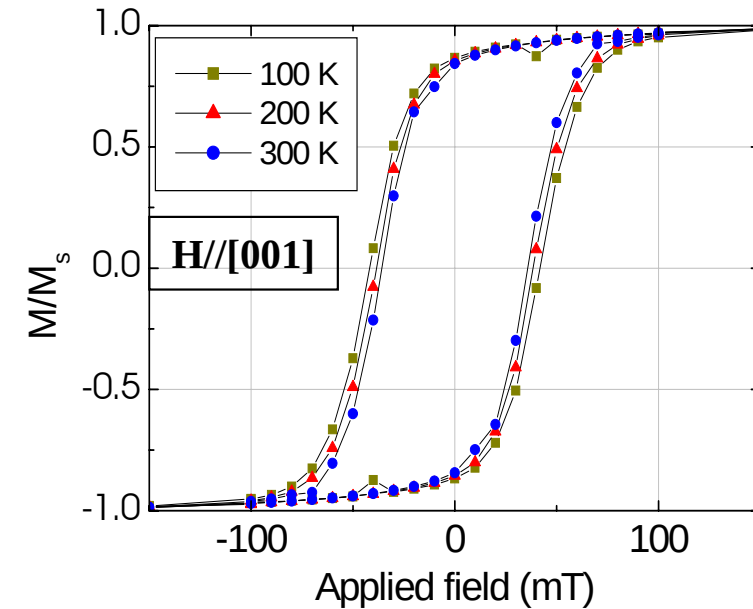
Fe/W(110) stripes

Sapphire \ W \ Fe(3nm) \ Mo

Stripe height = 5.5nm



- ↻ Coercivity and high remanence at 300K
- ↻ Weak temperature dependence
- > behaves like a conventional material



SOME LITERATURE

- [1] O. Fruchart, *Epitaxial self-organization: from surfaces to magnetic materials*, C. R. Physique **6** (1), 61-73 (2005).
- [2] J.P. Bucher, *Magnetism of free and supported metal clusters*, in: S.N. Khanna, A.W. Castleman Jr. (Eds.), Quantum Phenomena in Clusters and Nanostructures, in: Springer Series in Cluster Physics, Springer, Berlin, pp. 83–137 (2003).
- [3] J.P. Bucher, F. Scheurer, *Self-organized clusters and nanosize islands on metal surfaces*, in: J.S. Miller, M. Drillon (Eds.), Magnetism: Molecules to Materials III, Wiley–VCH, Weinheim, Germany, pp. 211–251 (2002).
- [4] J.I. Martin et coll., *Ordered magnetic nanostructures: fabrication and properties*, J. Magn. Magn. Mater. **256**, 449-501 (2003).
- [5] R. Skomski, *Nanomagnetics*, J. Phys.: Cond. Mat. **15**, R841–896 (2003).
- [6] J. Bansmann et al., *Magnetic and structural properties of isolated and assembled clusters*, Surf. Sci. Rep. **56**, 189 (2005).