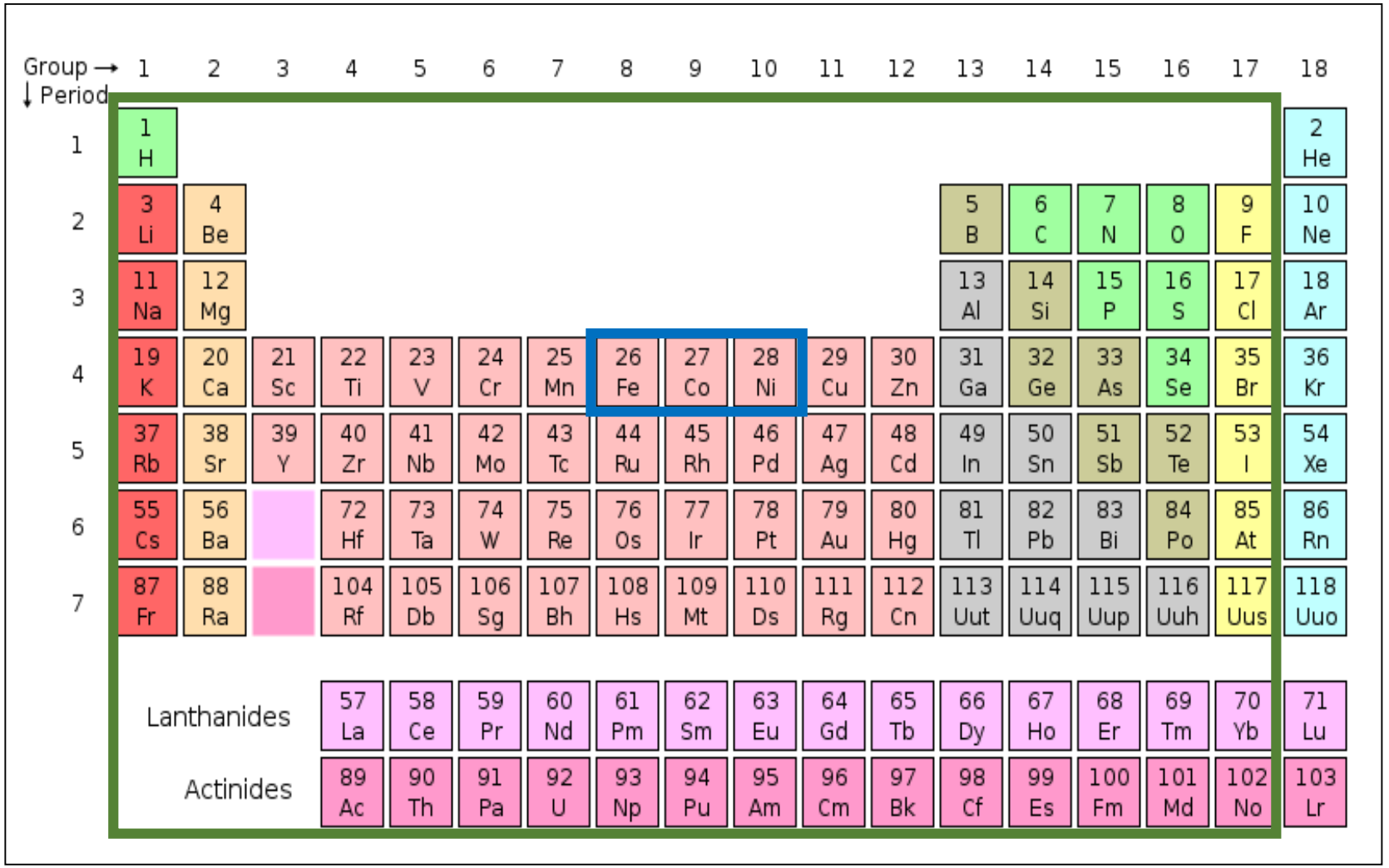
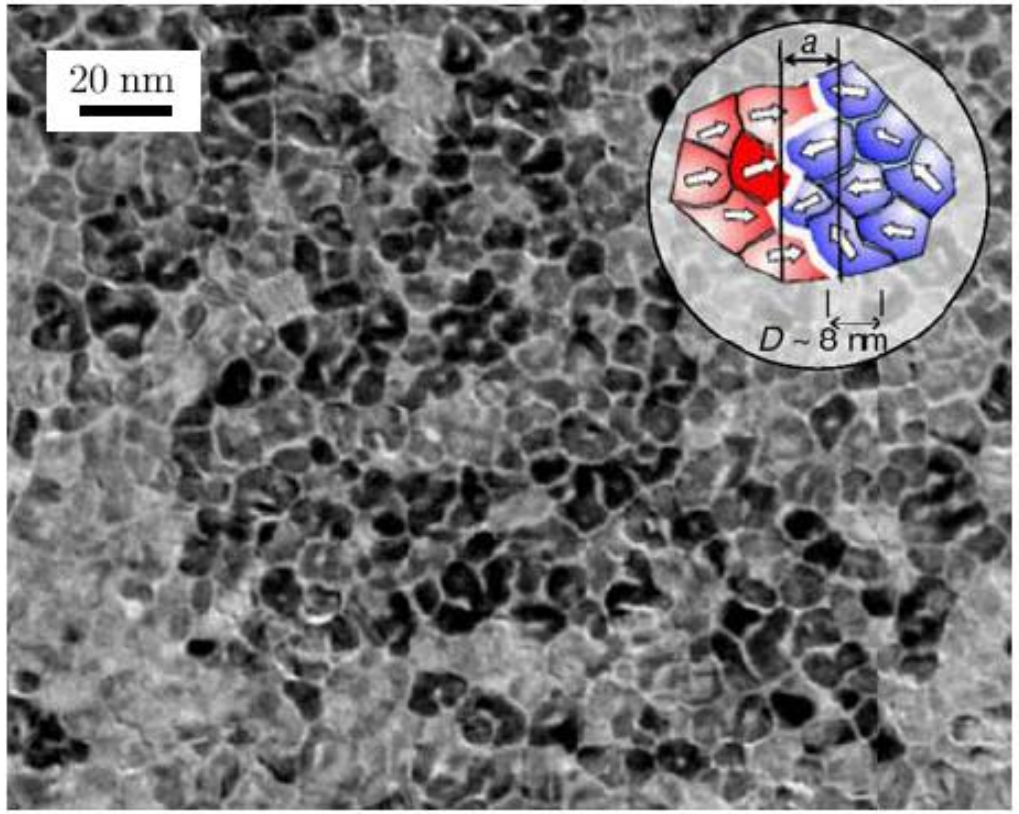
Atomic scale *vs* bulk

- Bulk (at room T): only a few elements (Fe, Co, and Ni) have a magnetic moment
- Atomic scale: all atoms except noble gases have a magnetic moment which has a **spin** and **orbital** contribution



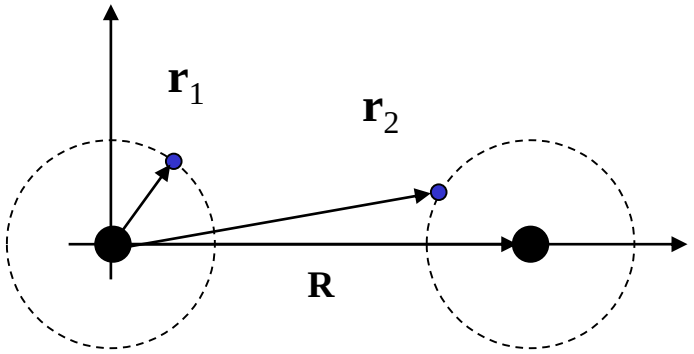
Some basic questions …..
… simply looking at this picture



CoCrPt recording layer

1) Why every grain has a magnetic moment?
2) How the material is chosen?
3) Why every grain is magnetically decoupled from the neighbors
4) Why the grain magnetization is pointing in one specific direction?
5) Why the magnetization direction changes from grain to grain?
6) Why the grain magnetization does not fluctuate with time?
7) …

Exchange energy : the H_2 molecule



H atom 1
Electron described
by $\psi_1(r_1)\ \sigma_1$

H atom 2
Electron described
by $\psi_2(r_2)\ \sigma_2$

Hamiltonian $H = H_1 + H_2 + H_{12}$, where

$$\begin{cases} H_1 = -\dfrac{\hbar^2}{2m}\Delta_1 - \dfrac{Ze^2}{r_1} \\ H_2 = -\dfrac{\hbar^2}{2m}\Delta_2 - \dfrac{Ze^2}{r_2} \\ H_{12} = +\dfrac{e^2}{r_{12}} \end{cases}$$

$$\sigma \rightarrow \alpha = \frac{1}{2}; \beta = -\frac{1}{2}$$

Because electrons are fermions, the overall wave function must be antisymmetric

$$\Psi_s = 1/\sqrt{2}[\Psi_1(r_1)\,\Psi_2(r_2) + \Psi_1(r_2)\,\Psi_2(r_1)] * [\alpha(r_1)\beta(r_2) - \beta(r_1)\alpha(r_2)]$$

Radial part symmetric; spin part antisymmetric

Singlet S=0

$$\Psi_{T1} = {}^{1}/_{\sqrt{2}}[\Psi_1(r_1)\,\Psi_2(r_2) - \Psi_1(r_2)\,\Psi_2(r_1)] * [\alpha(r_1)\beta(r_2) + \beta(r_1)\alpha(r_2)]$$

$$\Psi_{T2} = {}^{1}/_{\sqrt{2}}[\Psi_1(r_1)\,\Psi_2(r_2) - \Psi_1(r_2)\,\Psi_2(r_1)] * [\alpha(r_1)\,\alpha(r_2)]$$

$$\Psi_{T3} = {}^{1}/_{\sqrt{2}}[\Psi_1(r_1)\,\Psi_2(r_2) - \Psi_1(r_2)\,\Psi_2(r_1)] * [\beta(r_2)\,\beta(r_1)]$$

Radial part antisymmetric; spin part symmetric

Triplet S=1

Exchange energy: the H_2 molecule

Total spin S ($S^2 = \hbar^2 s(s+1)$)

$$\boldsymbol{S}^2 = \boldsymbol{\sigma}_1^2 + \boldsymbol{\sigma}_2^2 + 2\boldsymbol{\sigma}_1 \cdot \boldsymbol{\sigma}_2$$

$$2\boldsymbol{\sigma}_1 \cdot \boldsymbol{\sigma}_2 = \boldsymbol{S}^2 - \boldsymbol{\sigma}_1^2 - \boldsymbol{\sigma}_2^2 = \boldsymbol{S}^2 - \frac{3}{4} - \frac{3}{4}$$

$$S = 0 \rightarrow \boldsymbol{S}^2 = 0 \quad \rightarrow 2\,\boldsymbol{\sigma}_1 \cdot \boldsymbol{\sigma}_2 = -\frac{3}{2}$$ singlet

$$S = 1 \rightarrow \boldsymbol{S}^2 = 2 \quad \rightarrow 2\,\boldsymbol{\sigma}_1 \cdot \boldsymbol{\sigma}_2 = +\frac{1}{2}$$ triplet

$$E_S = \int \Psi_s{}^* \, H \, \Psi_s \, dr_1 dr_2$$

$$E_T = \int \Psi_T{}^* \, H \, \Psi_T \, dr_1 dr_2$$

$$\Delta E_{ST} = E_S - E_T = 2 \int \Psi_1{}^*(r_1)\Psi_2{}^*(r_2)\; H \;\Psi_1(r_2)\,\Psi_2(r_1)\, dr_1 dr_2$$

$$H = \frac{1}{4}(E_S + 3E_T) - \Delta E_{ST}\,\boldsymbol{\sigma}_1 \cdot \boldsymbol{\sigma}_2 = \frac{1}{4}(E_S + 3E_T) - 2\,J_{ex}\,\boldsymbol{\sigma}_1 \cdot \boldsymbol{\sigma}_2 \qquad J_{ex} = \Delta E_{ST}/2$$

$S = \sigma_1 + \sigma_2 = 1$ $E = (E_S + 3E_T)/4 - 1/2\ J_{ex}$

$S = \sigma_1 + \sigma_2 = 0$ $E = (E_S + 3E_T)/4 + 3/2\ J_{ex}$

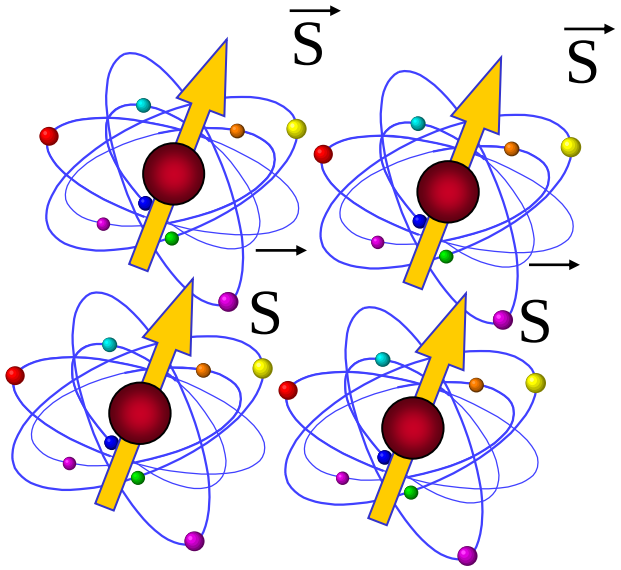
The energy depends on the relative orientation of the two spins and on the sign of J_{ex}

Origin of Exchange interaction:

→ Coulomb repulsion between electrons

→ total anti-symmetric wave function (Pauli exclusion principle)

Exchange in a cluster



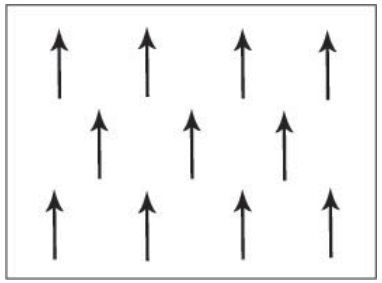
Orbital overlapping between adjacent atoms (similarly to the H_2 molecule)

We can describe the interaction via an effective Hamiltonian similar to the one developed for the H_2 molecule

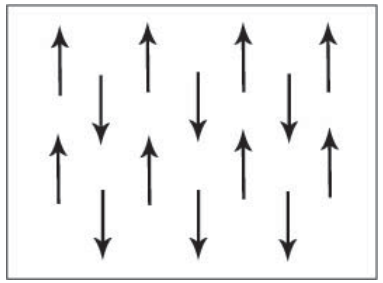
Heisenberg Hamiltonian

$$H = -2 J_{ex} \vec{S}_1 \vec{S}_2$$

$J_{ex} > 0$ Ferromagnetic coupling

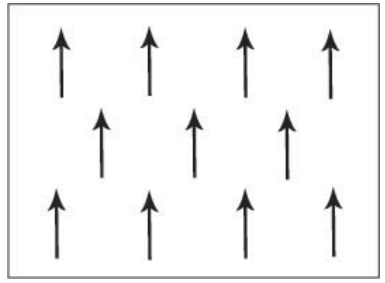


$J_{ex} < 0$ Antiferromagnetic coupling



Ordering temperature

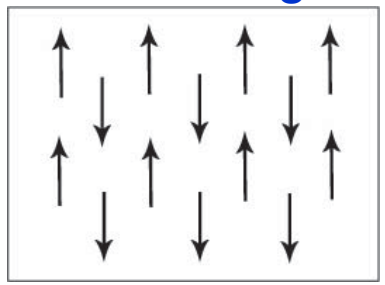
Ferromagnet



$T<T_C$

(T_C is the Curie temperature)

Antiferromagnet



$T<T_N$

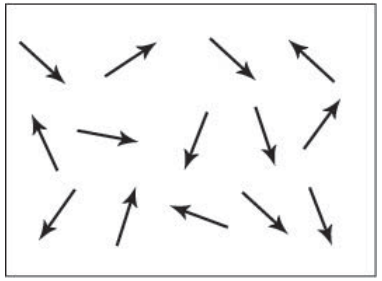
(T_N is the Néel temperature)

$T>T_C$

$T>T_N$

Paramagnet

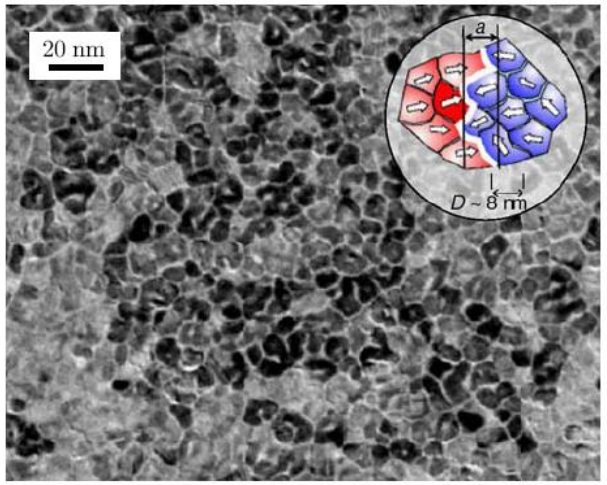
the magnetic moments are randomly oriented due to thermal fluctuations



The Curie (Néel) temperature depends on the exchange coupling and on the number of nearest neighbors N

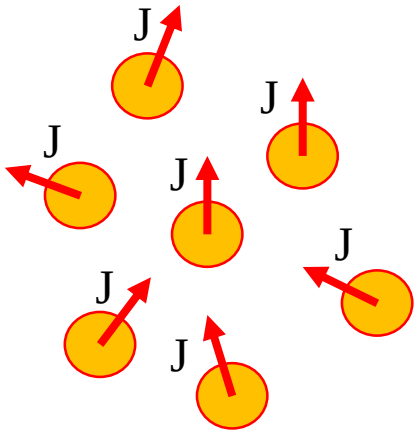
$$T_{C(N)} = \frac{2\,S(S+1)\,N\,J_{ex}}{3\,k_B}$$

Spin of a grain: macrospin



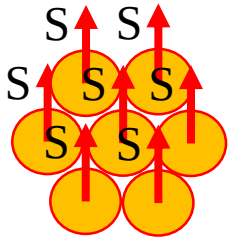
The grain (particle) can be described as a single **macrospin**

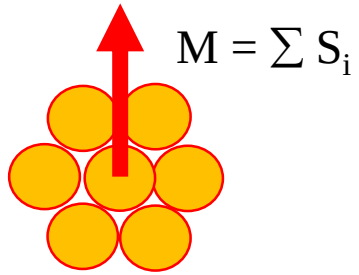
$$\mathbf{M = \Sigma_i S_i}$$



Isolated atoms with moment J

Grain (particle) formation:
1) Quenching of L
2) J ≈ S





All atomic spins in the grain are ferromagnetically aligned:

Exchange energy:

$$\mathbf{-2\, J_{ex}\, S_i\, S_j}$$

Exchange breaking layer: oxide phase for grain decoupling

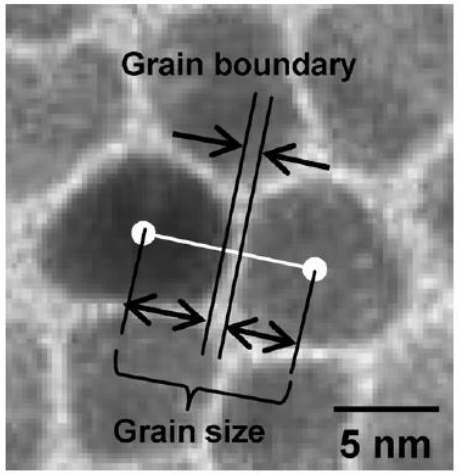


Fig. 1. Plan-view TEM image of CoCrPt-SiO_2 with definition of grain size and grain boundary width. White dots in the image show the centroids of each grain.

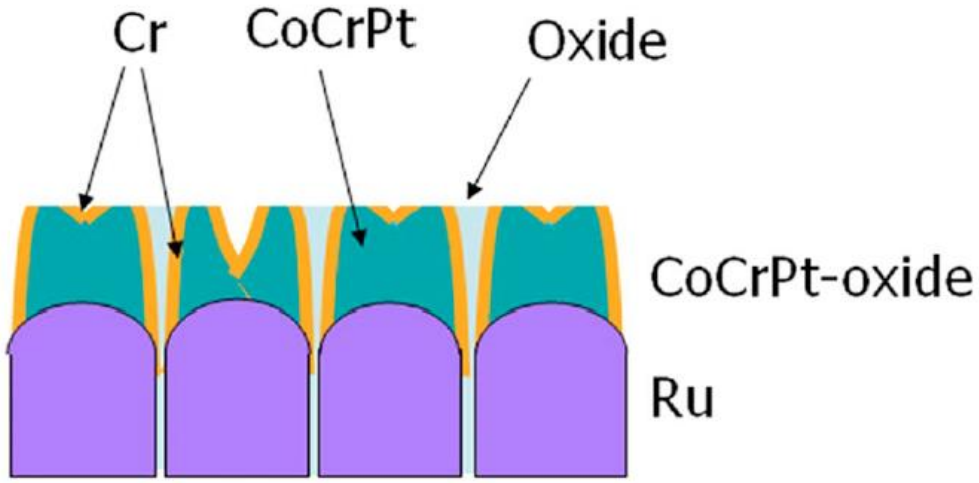


FIG. 3. (Color online) Schematic of a possible mechanism for tooth growth.

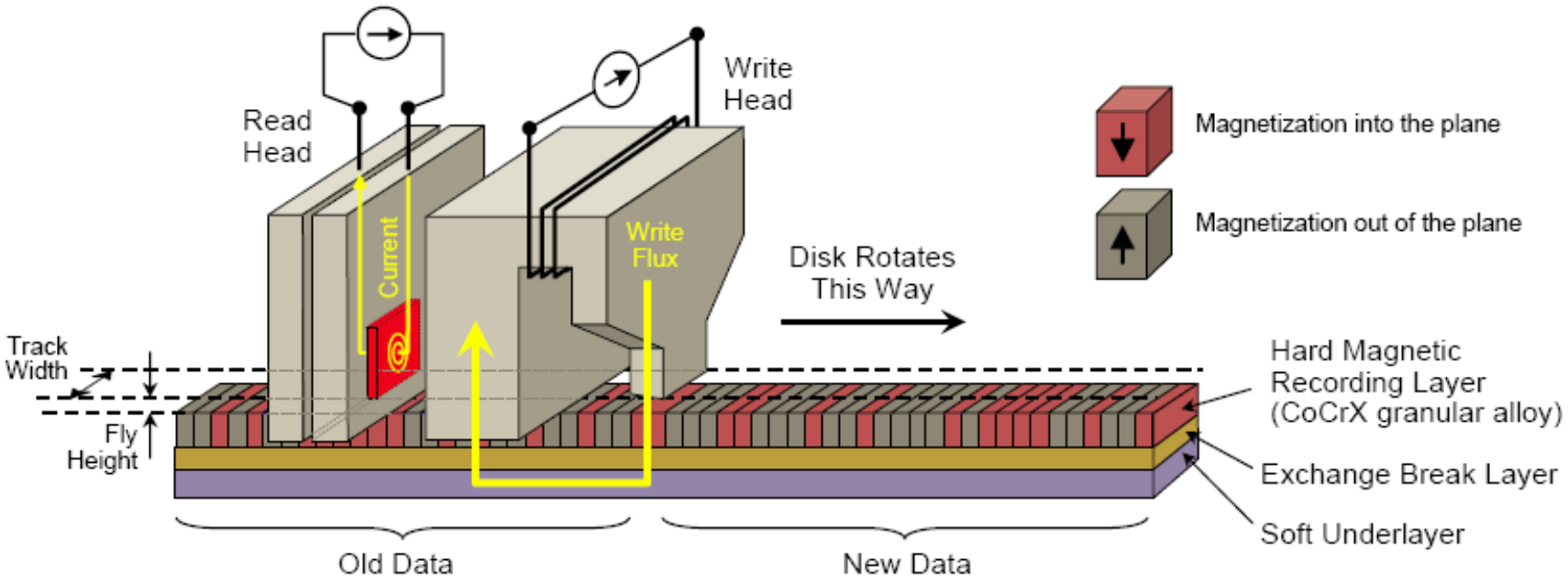
SiO_2 is non magnetic (S = 0)

The inter-grain exchange interaction is stopped by the oxide layer: every grain is independent of the others

R. Araki, *et al.* IEEE Trans. Magn. **44**, 3496 (2008).

D. E. Laughlin, *et al.* J. Appl. Phys. **105**, 07B739 (2009).

Exchange breaking layer: Magnetic recording technology

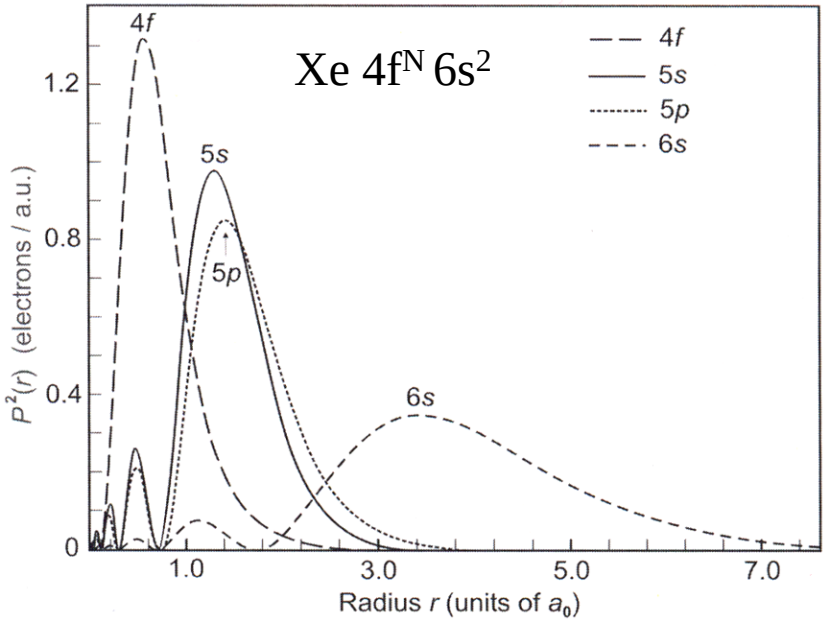


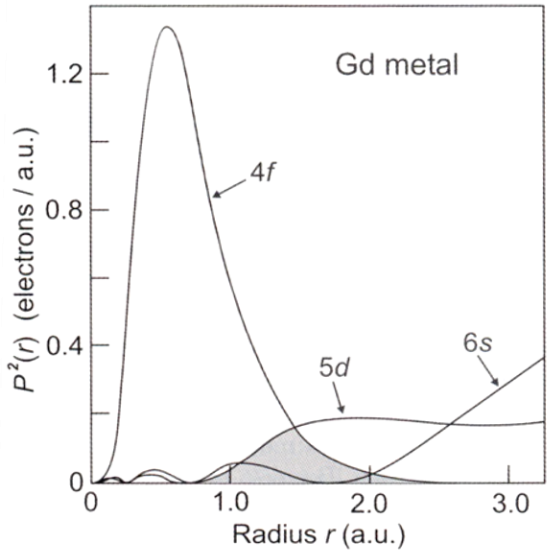
The exchange breaking layer is necessary to decouple the recording layer from the soft underlayer

The soft underlayer helps to close (focalize) the magnetic flux lines

Valence orbitals in rare earth and consequences for the Exchange

Electronic configuration in atomic case: Xe $4f^N 6s^2$
(exception for Gd: Xe $4f^7 5d^1 6s^2$)





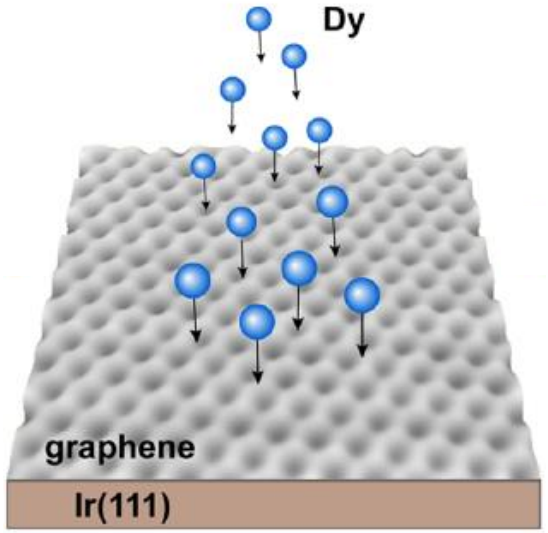
Radial distribution of the different orbitals

In rare earth, magnetism comes from 4f states

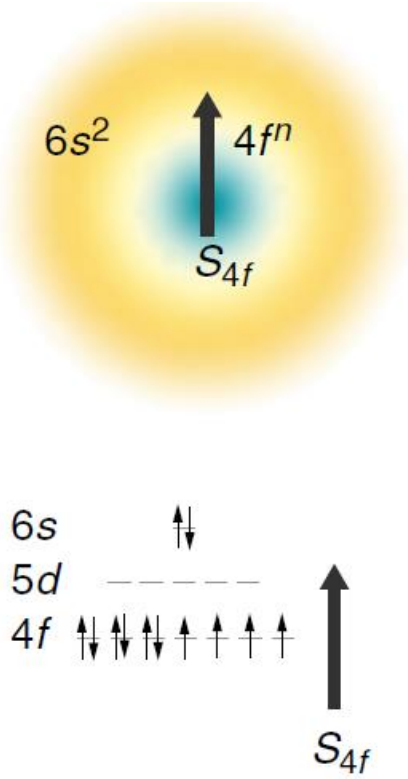
4f states are strongly localized →
do not overlap with wave functions of neighbouring atoms
but
overlap with 5d and 6s states

How collective magnetism is possible in rare earth compounds ?

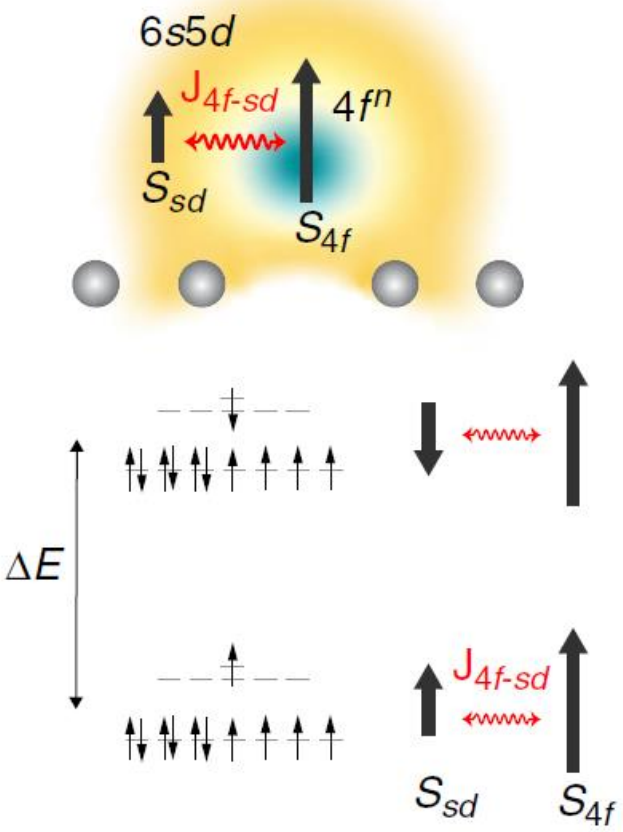
Intra-atomic Exchange in rare earths

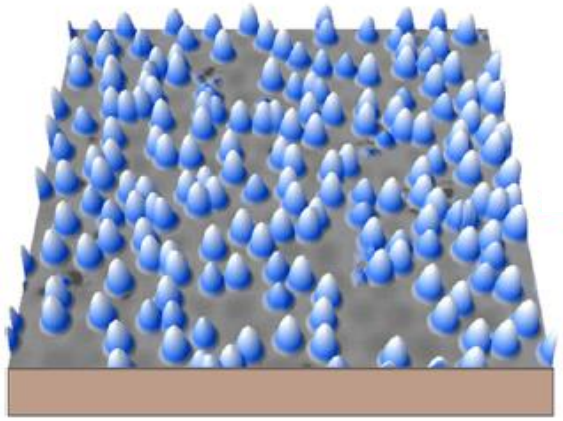


Gas phase



Atom on graphene

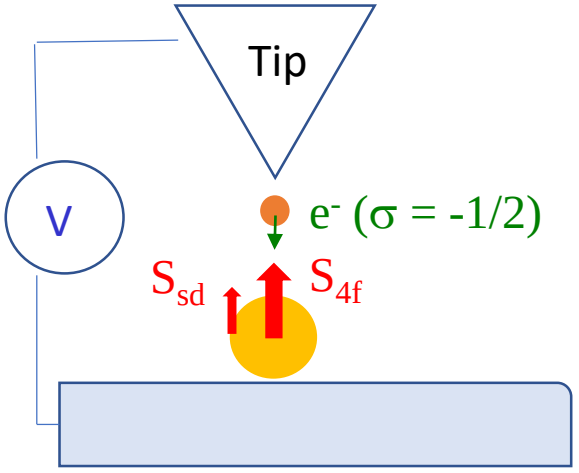


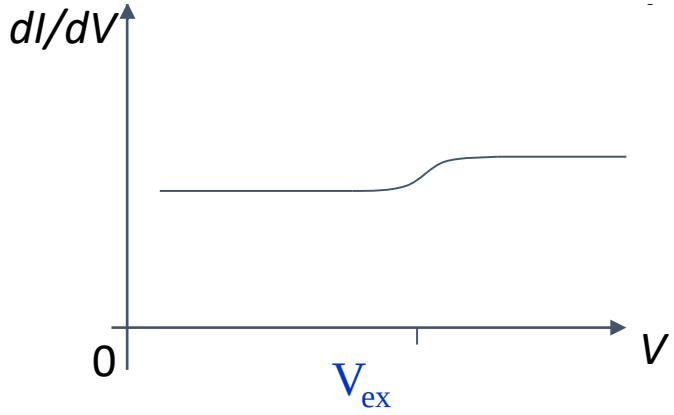


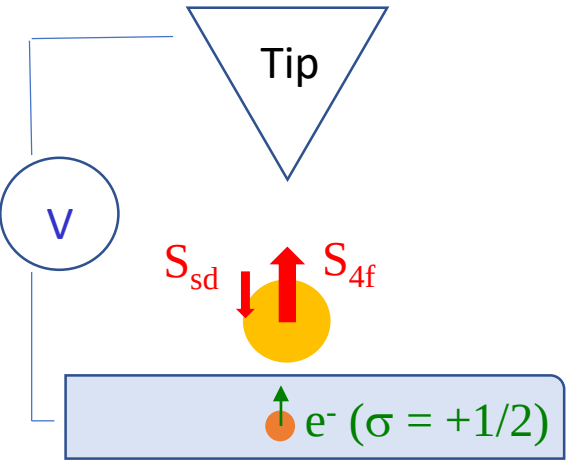
S_{sd} and S_{4f} are coupled by the intra-atomic exchange interaction $J_{4f\text{-}sd}$

Adsorption on graphene induces the transfer of one of the 6s electrons to graphene while the remaining one is delocalized on the 6s5d shells

Measuring the intra-atomic exchange with STM





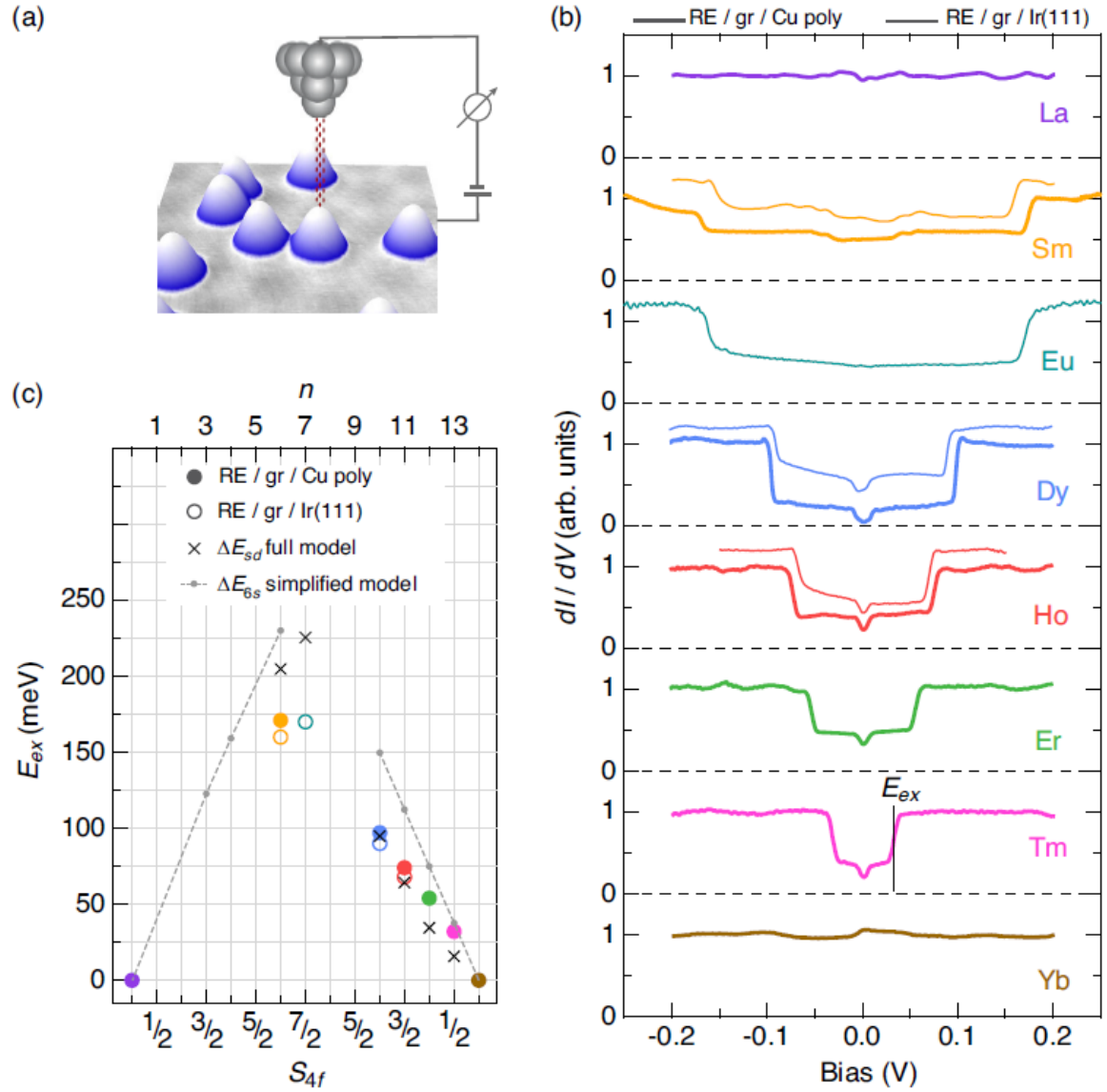


$E_{ex} = eV_{ex} = 2\, J_{4f\text{-}sd}\, S_{sd}\, S_{4f}$

Energy conservation

$\Delta\sigma = 1 \rightarrow \Delta S_{sd} = -1$

Total momentum conservation



M. Pivetta, *et al*. Phys. Rev. X **10**, 031054 (2020)

Inter-atomic Exchange: 4f-3d strong magnets ($SmCo_5$, Nd_2Fe_{14})

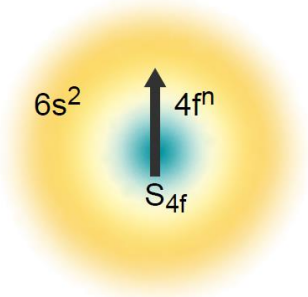
Gas phase

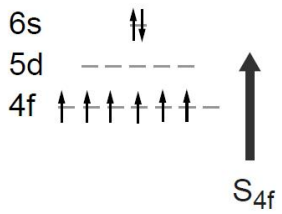
Bulk phase

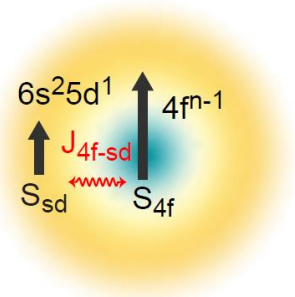
Rare earths (RE)

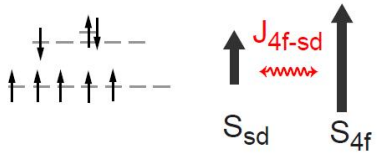
Xe 4f ^{N} 6s^2

$S_{6s} = 0$









Xe 4f $^{N-1}$ 6s^2 5d^1

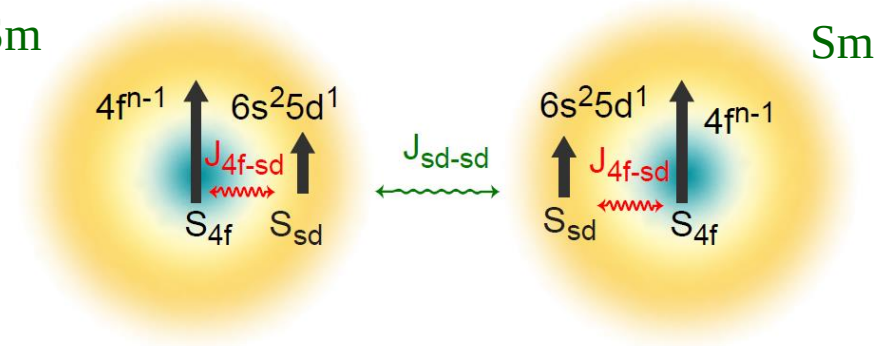
$S_{6s5d} \neq 0$

S_{sd} and S_{4f} are coupled by the intra-atomic exchange interaction J_{4f-sd}

Pure Sm

Sm

Sm



$H_{ex} = -2\ J^{Sm\text{-}Sm}{}_{sd\text{-}sd}\ S^{Sm}{}_{sd}\ S^{Sm}{}_{sd}$

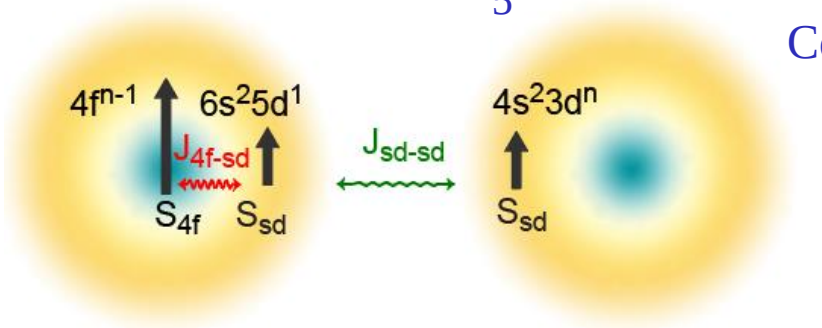
T_C (T_N) < 300 K

Highest T_C for Gd T_C = 292 K

$SmCo_5$

Sm

Co



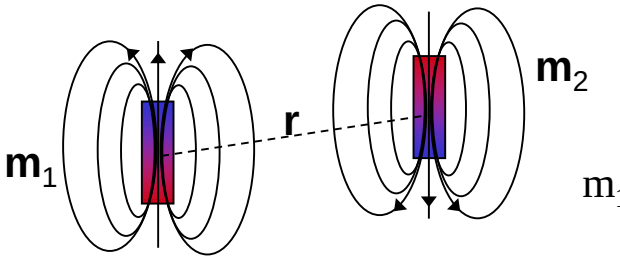
$H_{ex} = -2\ J^{Sm\text{-}Co}{}_{sd\text{-}sd}\ S^{Co}{}_{sd}\ S^{Sm}{}_{sd}$

The 3d element increases the strength of the exchange interaction thus increasing T_C

$SmCo_5$ -> T_C = 800°C

Dipolar interaction

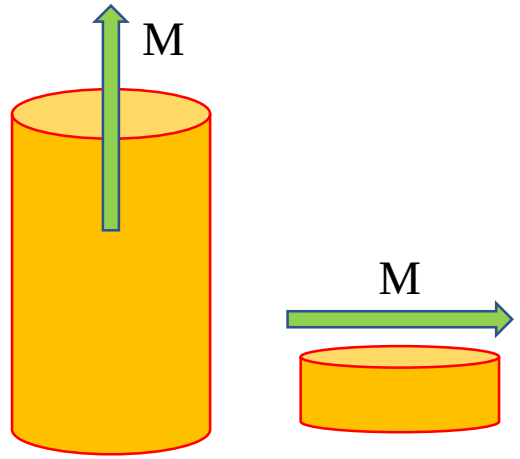
Long range interaction between magnetic moments



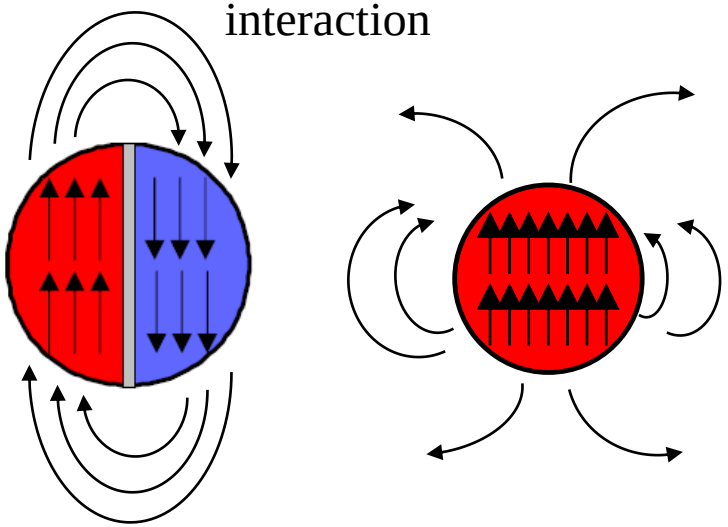
$$H_{dip} = \frac{\mathbf{m}_1 \cdot \mathbf{m}_2}{r^3} - 3\frac{(\mathbf{m}_1 \cdot \mathbf{r})(\mathbf{m}_2 \cdot \mathbf{r})}{r^5}$$

m_1 and m_2 : magnetic moments of two atoms in a particle or moments of two particles

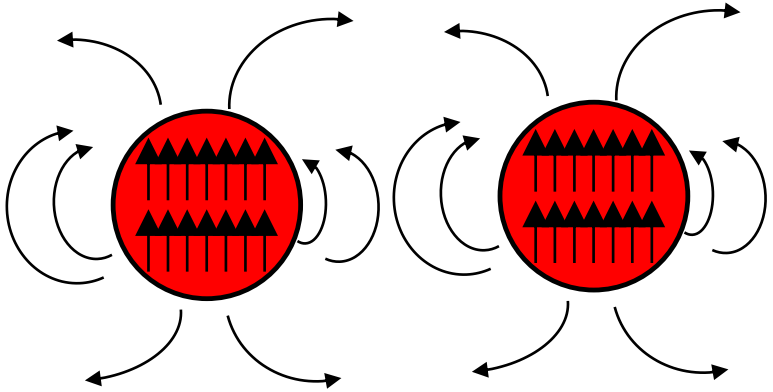
Magnetization orientation



Domain formation:
competition between exchange and dipolar interaction



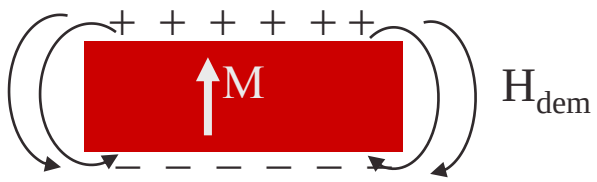
Interaction between close particles



Demagnetizing field: shape anisotropy

Infinite plate

poles only at very far ends ->
no magnetic field outside the plate $H_{dem} \approx 0$



poles at the two faces -> $H_{dem} \neq 0$

H_{dem} is the demagnetizing field

$$E_{dip} = -\frac{\mu_0}{2}\int \mathbf{M}\cdot\mathbf{H}_{dem}\,dV$$

$$\mathbf{H}_{dem} = -\mathbf{D}\,\mathbf{M}$$

Sphere:

$$D = \begin{bmatrix} \frac{1}{3} & 0 & 0 \\ 0 & \frac{1}{3} & 0 \\ 0 & 0 & \frac{1}{3} \end{bmatrix}$$

∞-Cylinder:

$$D = \begin{bmatrix} \frac{1}{2} & 0 & 0 \\ 0 & \frac{1}{2} & 0 \\ 0 & 0 & 0 \end{bmatrix}$$

∞-Plane (thin film):

$$D = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{bmatrix}$$

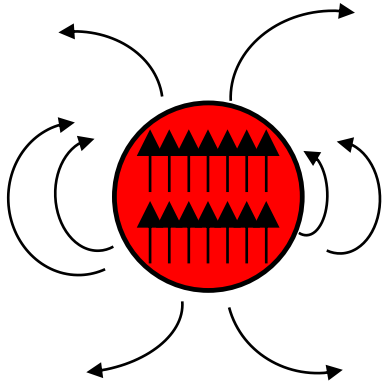
H_{dem} pushes the magnetization M along the longer side of the nanostructure:

Sphere → all directions are equivalent
Cylinder → M // axis
Disk → M // disk surface

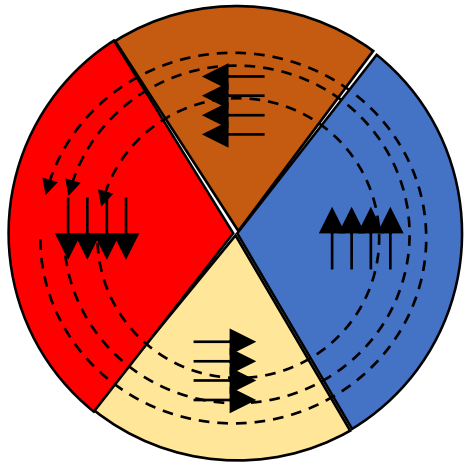
Shape anisotropy:
the shape determines
the magnetization easy axis

Magnetization state for in-plane magnetized particle

Increasing the particle size, domain formation minimizes the magneto-static energy due to the dipolar field

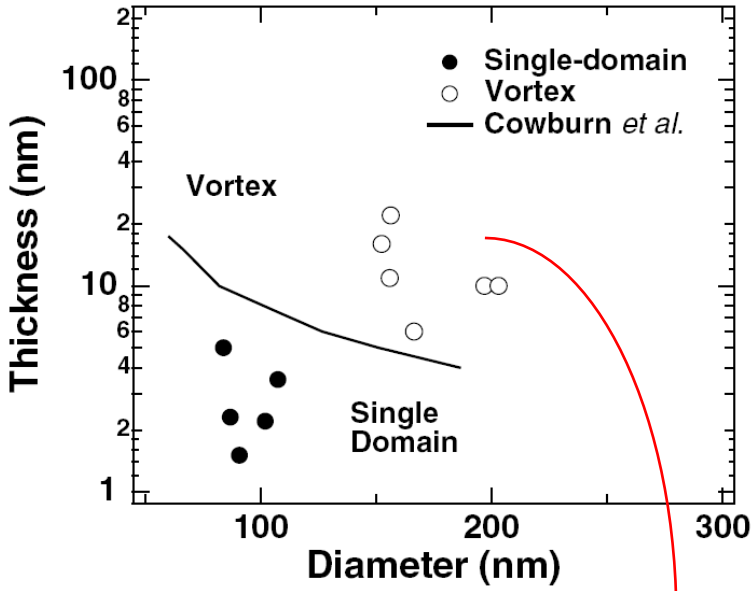


Flux lines extend outside the particle



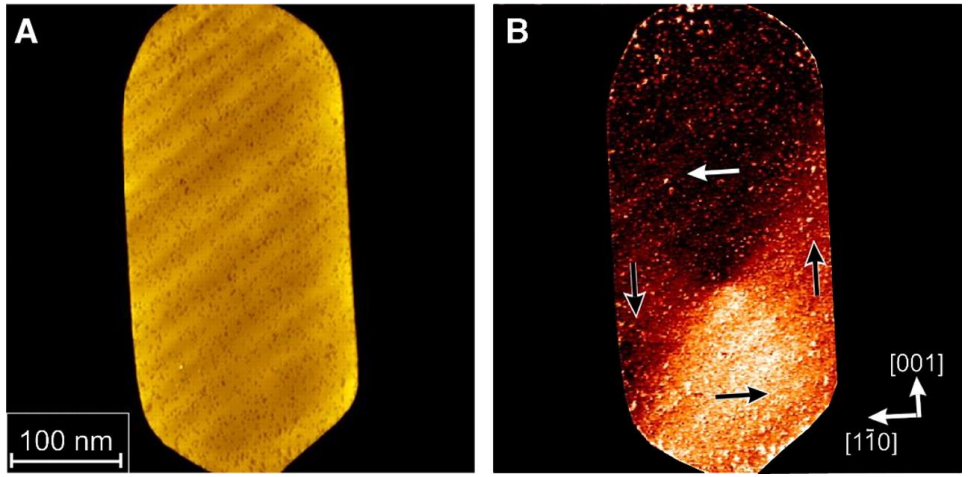
Flux lines are confined inside the particle

Magnetic phase diagram for ultrathin particles with in-plane anisotropy (Fe/W(001))



Topography

SP- STM



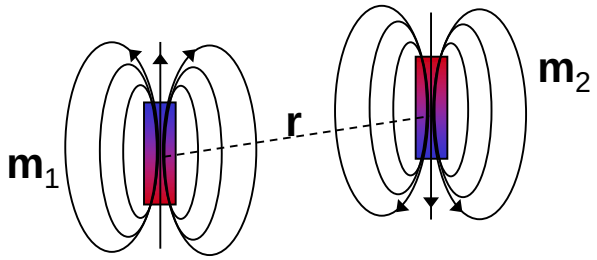
magnetic domain pattern of a 8 nm high Fe particle grown on W(110)

R. Skomski *et al*. Phys.Rev. Lett. **91**, 127201 (2003)
A. Wachowiak *et al*.. Science **298**, 577 (2002)

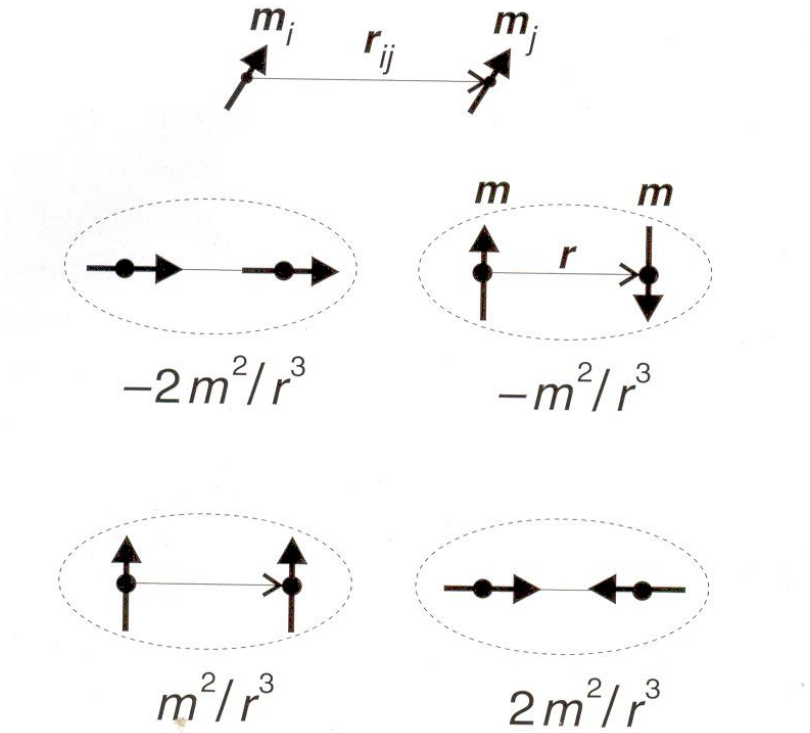
Longitudinal *vs.* perpendicular recording media

Long range interaction between magnetic moments

$$H_{dip} = \frac{\mathbf{m}_1 \cdot \mathbf{m}_2}{r^3} - 3\frac{(\mathbf{m}_1 \cdot \mathbf{r})(\mathbf{m}_2 \cdot \mathbf{r})}{r^5}$$



m_1 and m_2 the magnetic moments of two particles

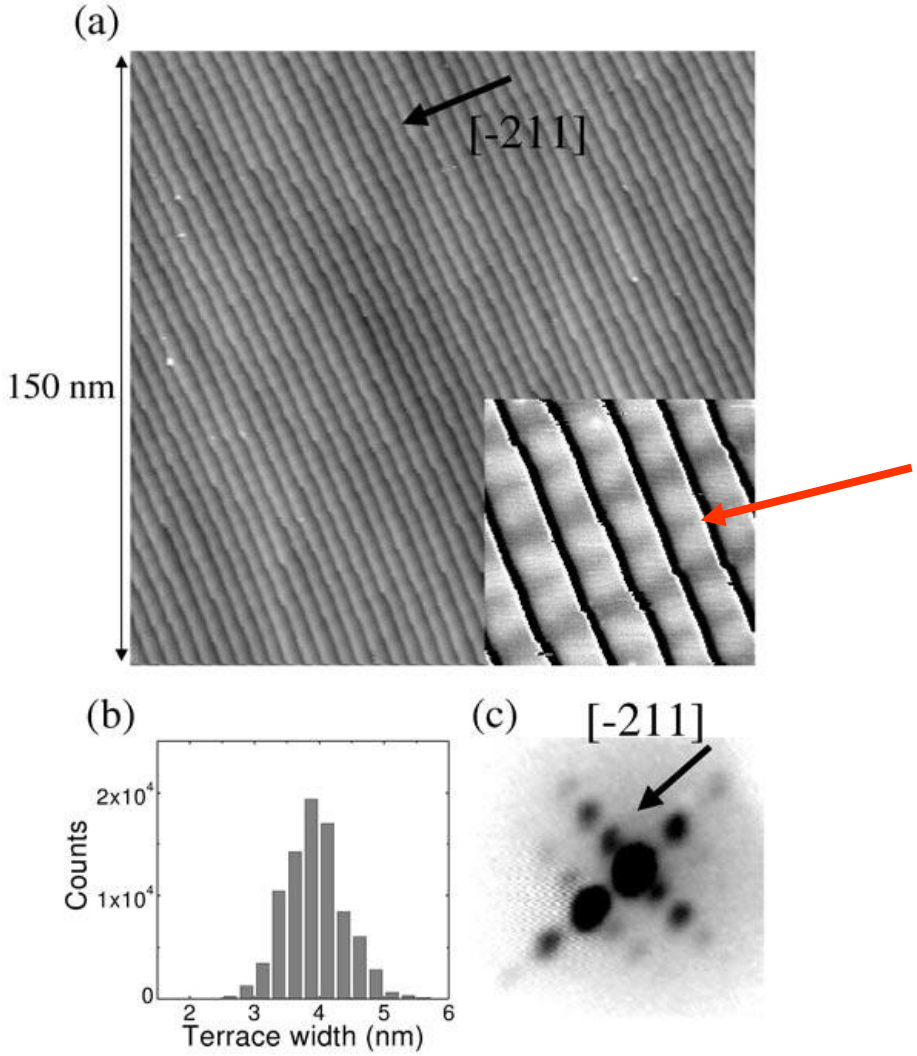


In the last decade, to overcome the 1Tbit/in^2 limit, the storage media has adopted perpendicular magnetized media in place of the longitudinal media

The out-of-plane configuration reduces the dipolar interaction

Dense lattice of magnetic dots: dipolar interaction

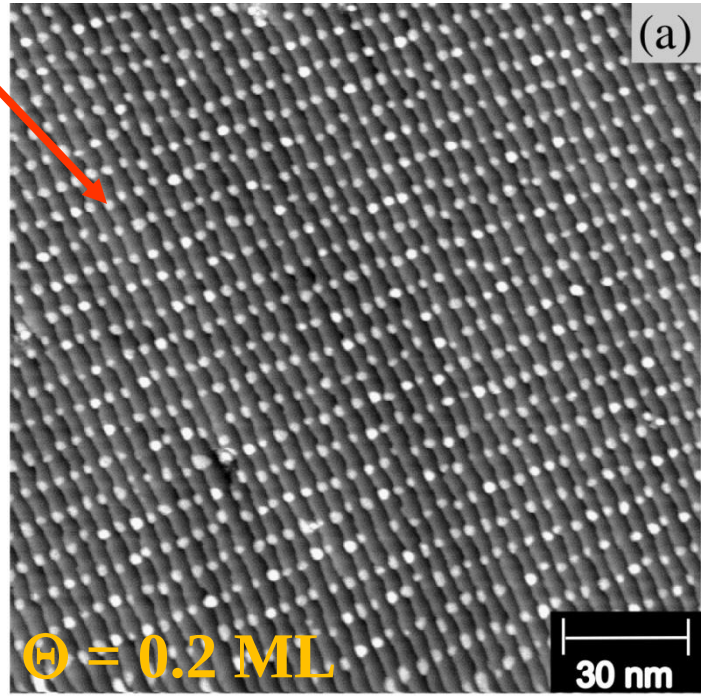
Au(788) vicinal surfaces



(111)-oriented terraces with reconstruction lines perpendicular to step edges

Surface reconstructions on two consecutive terraces are coherent

Co nucleates in bi-layer dots at the positions where the reconstruction lines cross the step edges

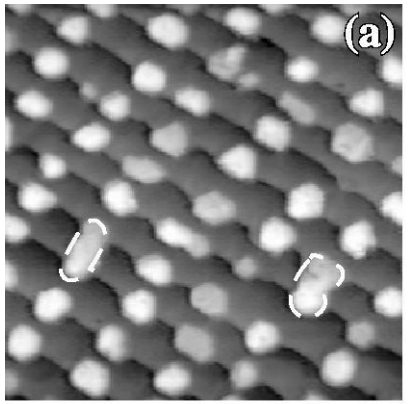


V. Repain et al. Europhys. Lett. **58**, 730 (2002)

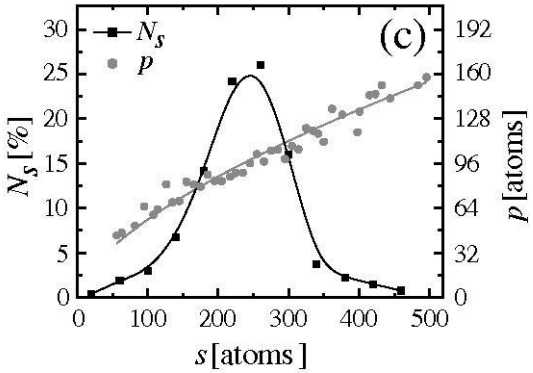
Negligible dipolar interaction at 26 Tdots/in^2

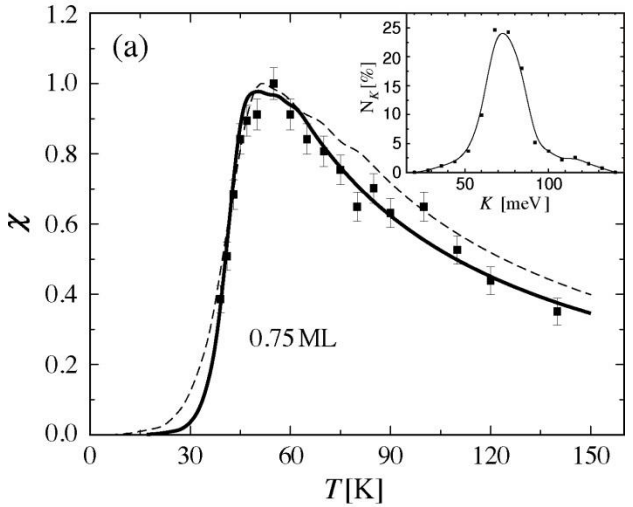
χ = dM/dB is the magnetic susceptibility
(B = B_0 cos(ωt))

0.75 ML Co



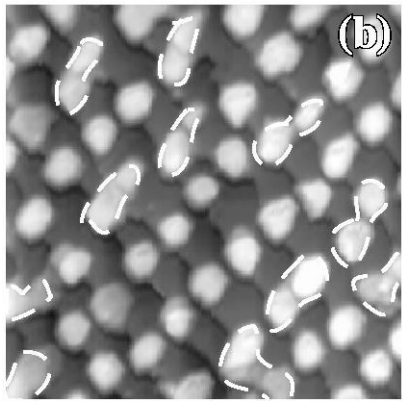
monodisperse size distribution



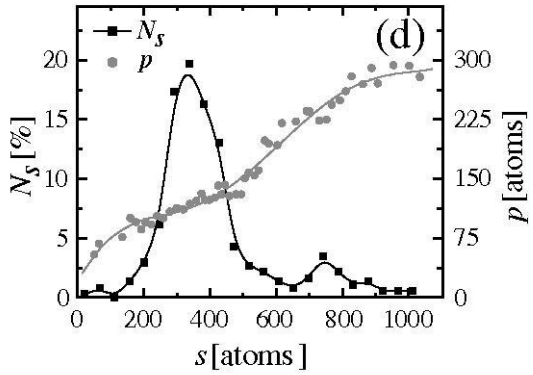


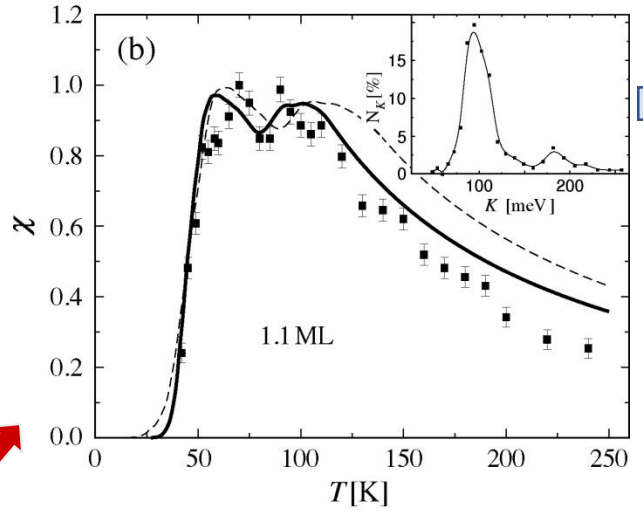
χ(T) is maximal at the temperature at which the island magnetic moment oscillates in phase with B

χ(T) shows a single peak



1.1 ML Co





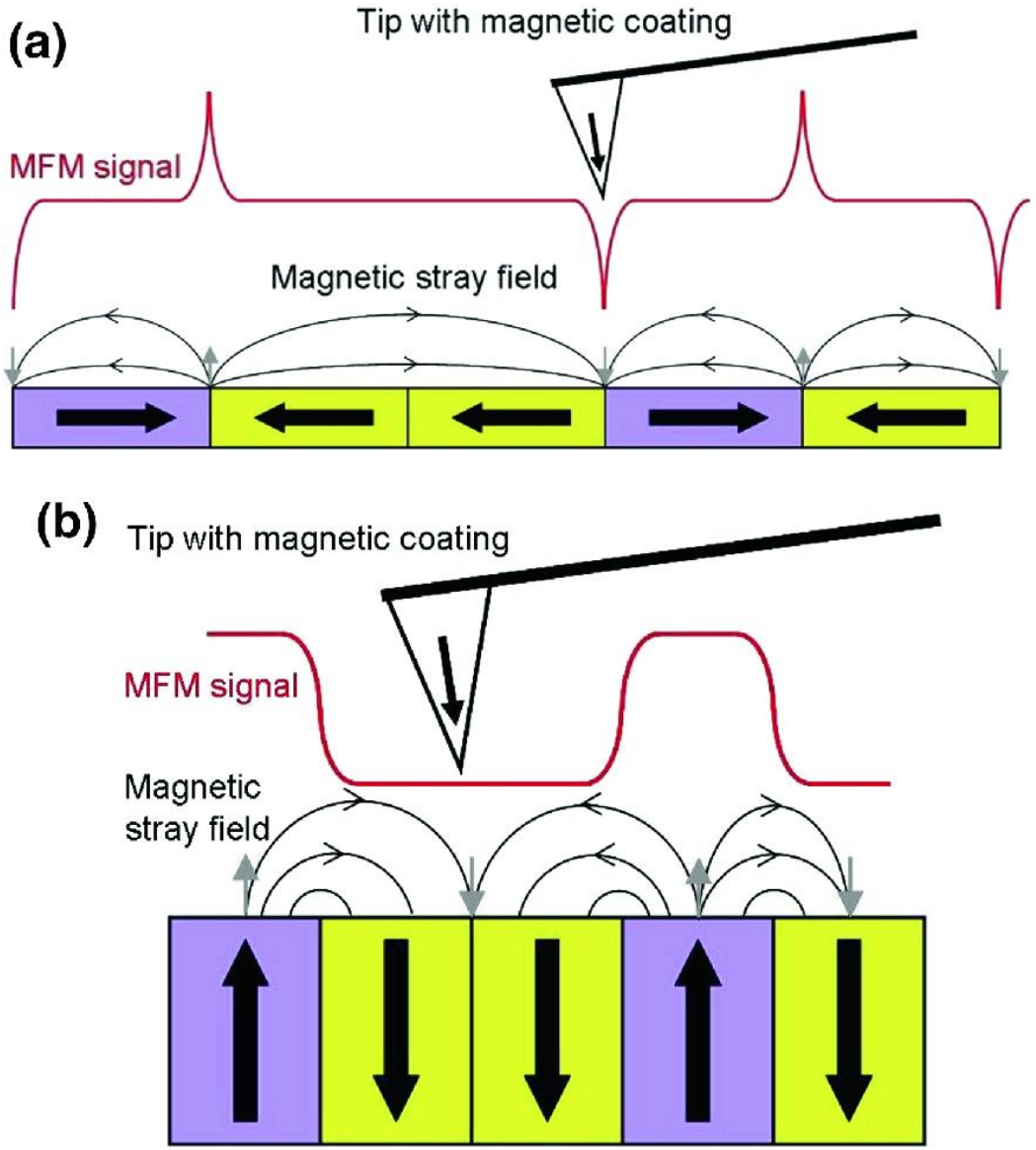
χ(T) shows two peaks

Bimodal size distribution ⟷ Bimodal χ vs. T

Big and small particles are independent: No coupling

N. Weiss, *et al*. Phys. Rev. Lett. **95**, 157204 (2005).

MFM: magnetic force microscope



Dipolar interaction between the magnetic moment of the tip and the magnetic moment of the bit

Principle of magnetic force microscopy over a sample surface with (**a)** in-plane and (**b)** perpendicular magnetization. The magnetically-coated tip detects the vertical component of the stray field (grey arrows) emanating from the surface. Hence, the MFM signal (in red) exhibits peaks at the domain boundaries in (a) and high (low) signal for anti-parallel (parallel) alignment of tip and domain magnetizations in (b)

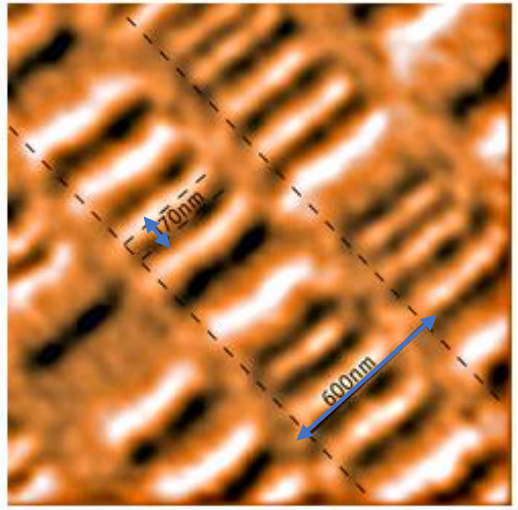
https://link.springer.com/chapter/10.1007/978-3-030-15612-1_8

AFM *vs* MFM

AFM

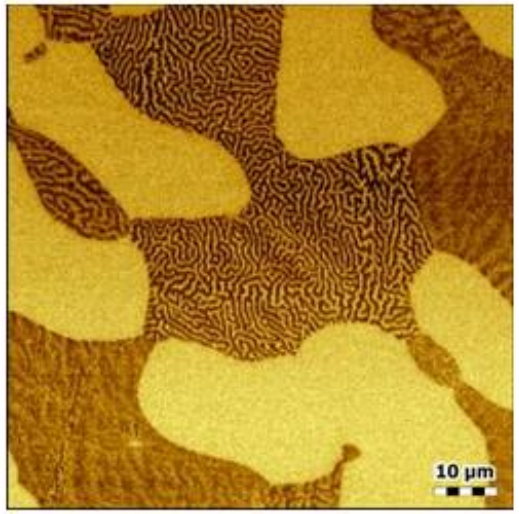
MFM

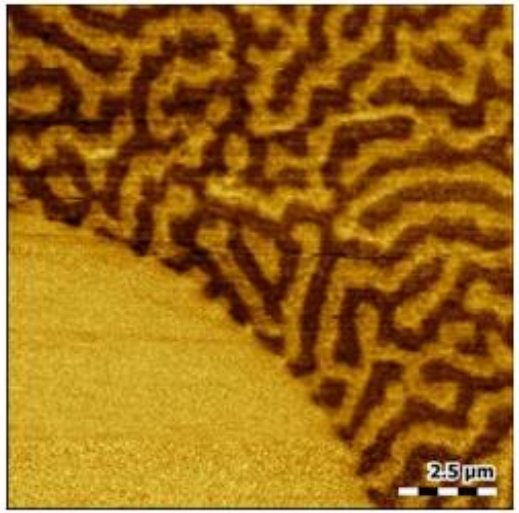
Hard disk drive platter with in-plane magnetized media



Black and white regions are the domain wall between in-plane magnetized domains
Each bit has a size of about 70 x 600 nm^2

Stainless steel with out-of-plane magnetized domain





Dark and white regions are up and down out-of-plane magnetized domains

https://www.nanosurf.com