11

Magnetic interactions

Magnetism at the nanoscale

Magnetism in Condensed Matter
S. Blundell
Oxford University Press

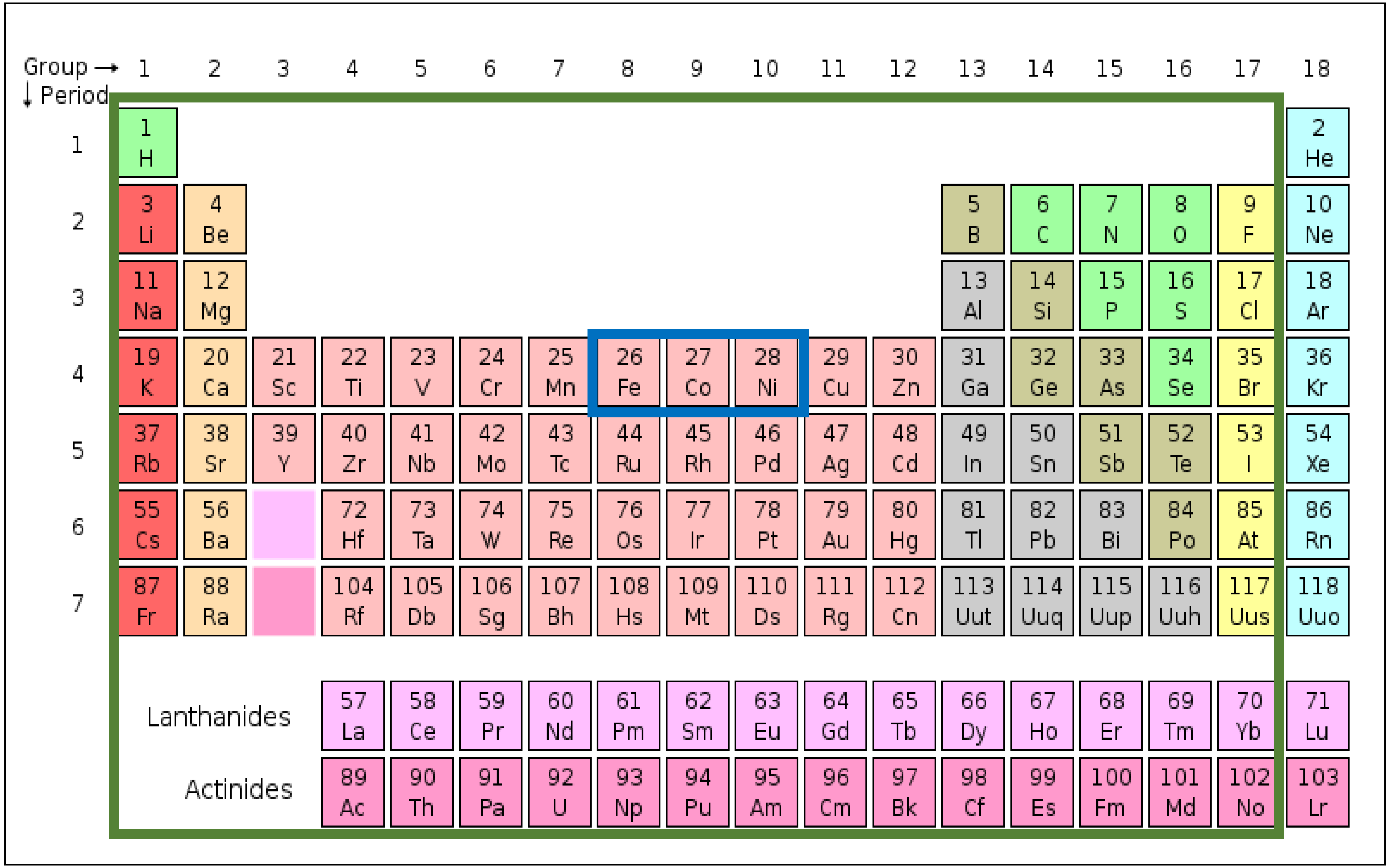
Fundamentals of Magnetism
M. Getzlaff
Springer

Magnetism and Magnetic Materials
J. M. D. Coey
Cambridge University Press

Magnetism - From Fundamentals to Nanoscale Dynamics
J. Stöhr and H.C. Siegmann
Springer

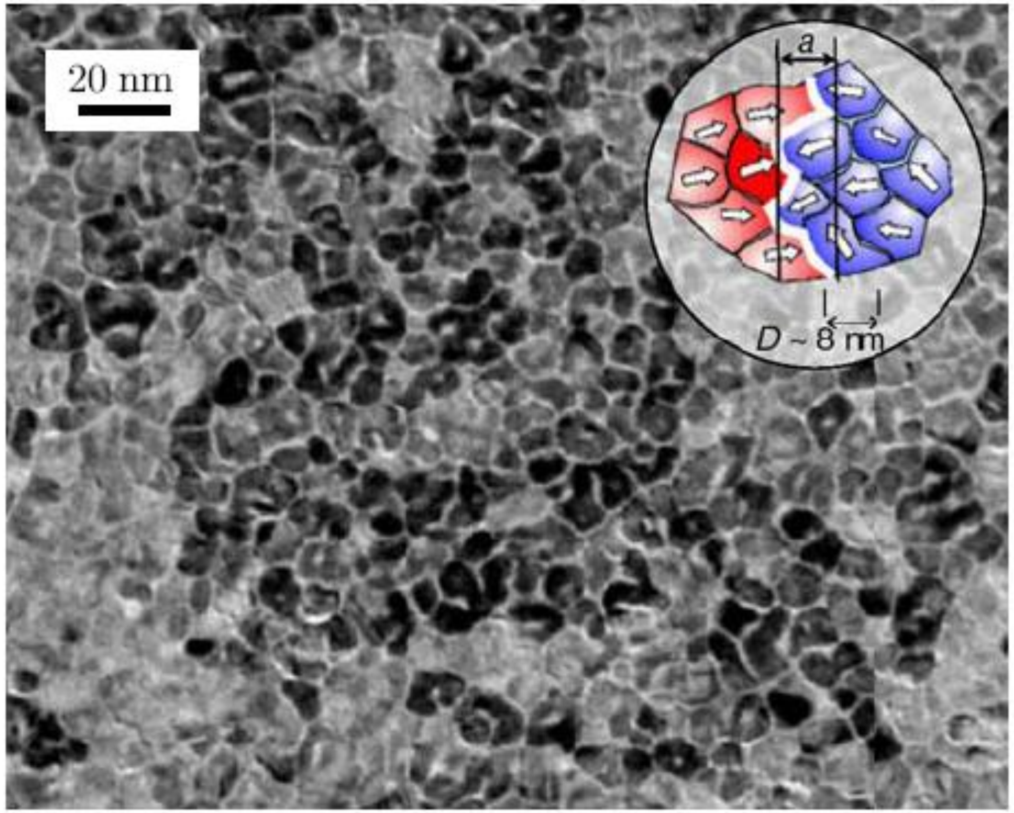
Atomic scale vs bulk

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



Group → ↓ Period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	1 H																	2 He
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba		72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra		104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Uut	114 Uuq	115 Uup	116 Uuh	117 Uus	118 Uuo
Lanthanides				57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
Actinides				89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

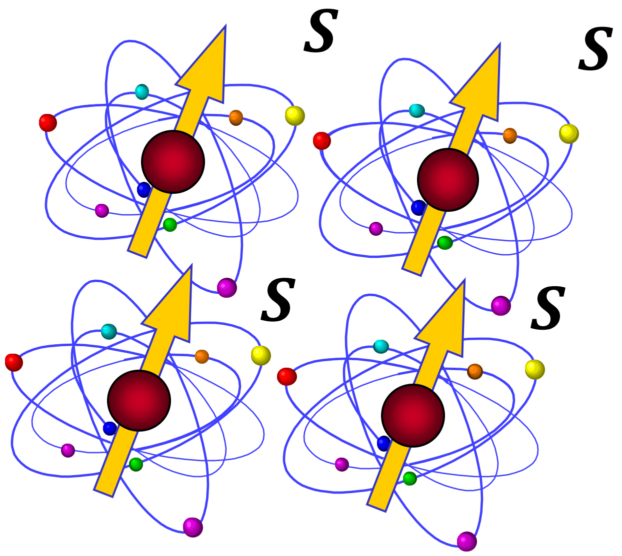
Some basic questions looking at this picture



CoCrPt recording layer

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

Exchange interaction



Orbitals overlapping between adjacent atoms (→ short ranged interaction)

We can describe the interaction via an **effective Hamiltonian** like the one developed for the H_2 molecule

Heisenberg Hamiltonian $\mathcal{H}_{exch} = -2J_{ex}\boldsymbol{S}_1 \cdot \boldsymbol{S}_2$

$J_{ex} > 0$ → ferromagnetic coupling ↑↑

$J_{ex} < 0$ → antiferromagnetic coupling ↑↓

Can be extended to more than two spins by summing over pairs

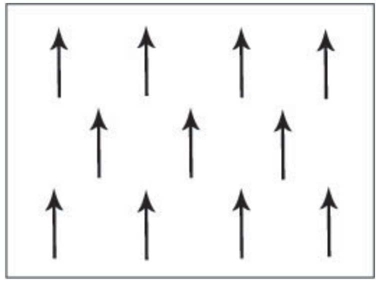
Origin of exchange interaction:

- **Coulomb repulsion** between electrons
- total anti-symmetric wave function (**Pauli exclusion principle**)

Ordering temperature

Exercise 11.1

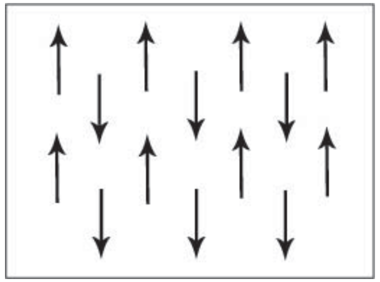
Ferromagnet



$T < T_C$

(T_C is the Curie temperature)

Antiferromagnet



$T < T_N$

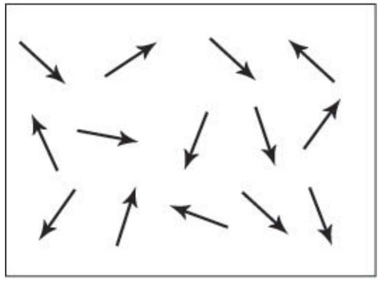
(T_N is the Néel temperature)

$T > T_C$

$T > T_N$

Paramagnet

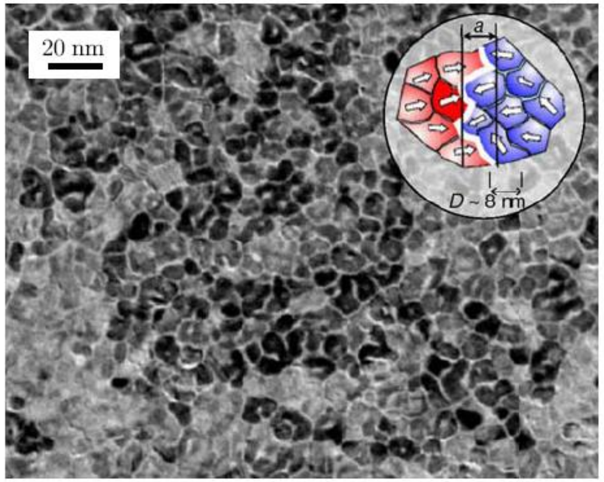
the magnetic moments are randomly oriented due to thermal fluctuations



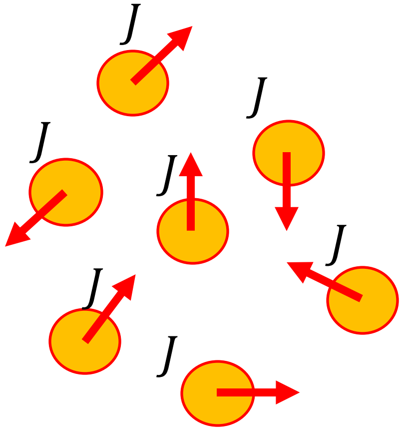
The Curie (Néel) temperature depends on the exchange coupling and on the number of nearest neighbors N

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

Spin of a grain: Macrospin



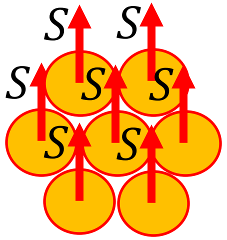
The grain (particle) magnetization $\boldsymbol{M}$ can be described as a single **macrospin**



Isolated atoms
with moment J

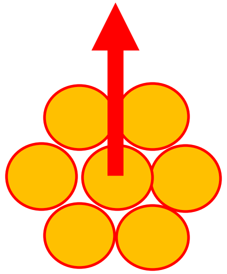
Grain (particle) formation:
quenching of L

$$J \approx S$$



All atomic spins in the grain are ferromagnetically aligned by the exchange interaction:

$$-2\, J_{ex} \sum_{i<j} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$



$$\boldsymbol{M} = \sum_i \boldsymbol{S}_i$$

Oxide layer for grain decoupling

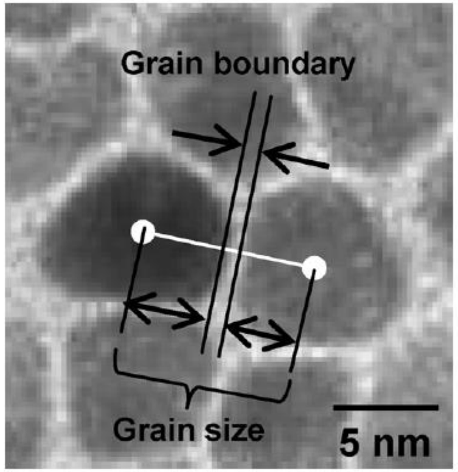


Fig. 1. Plan-view TEM image of CoCrPt-SiO_2 with definition of grain size and grain boundary width. White dots in the image show the centroids of each grain.

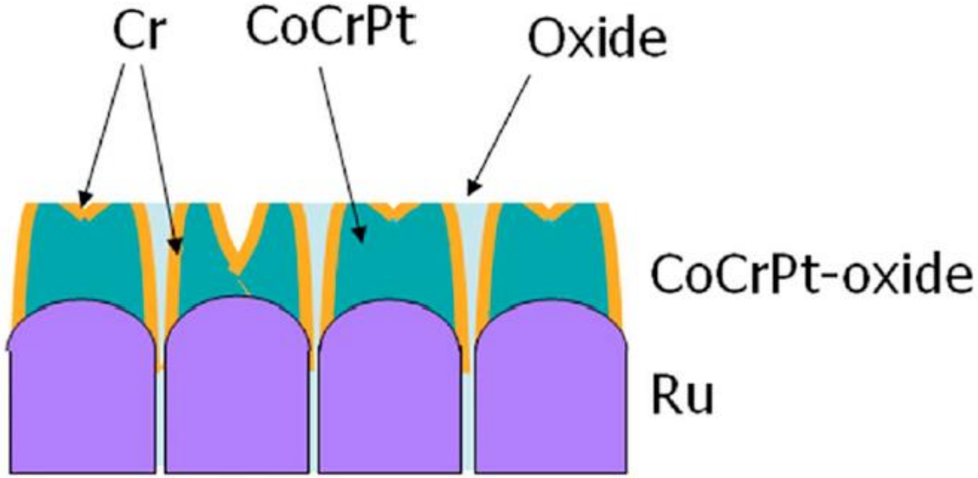


FIG. 3. (Color online) Schematic of a possible mechanism for tooth growth.

SiO_2 is non-magnetic ($S = 0$)

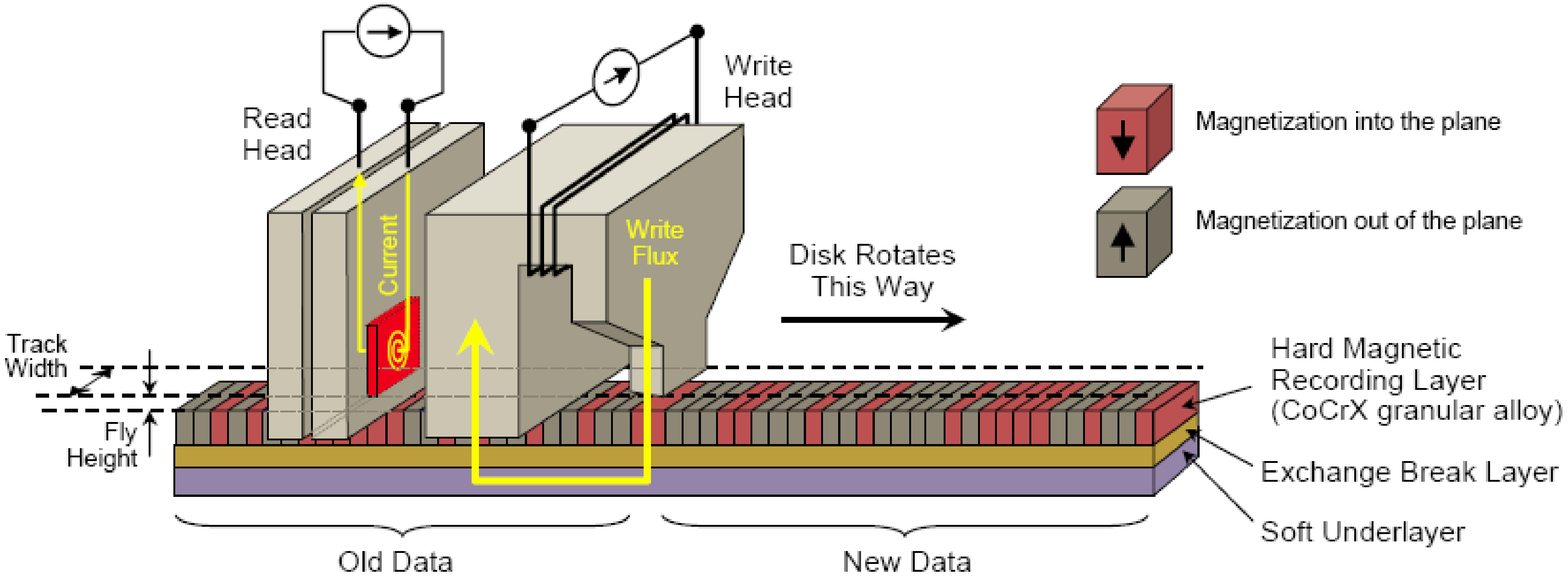
⬇

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



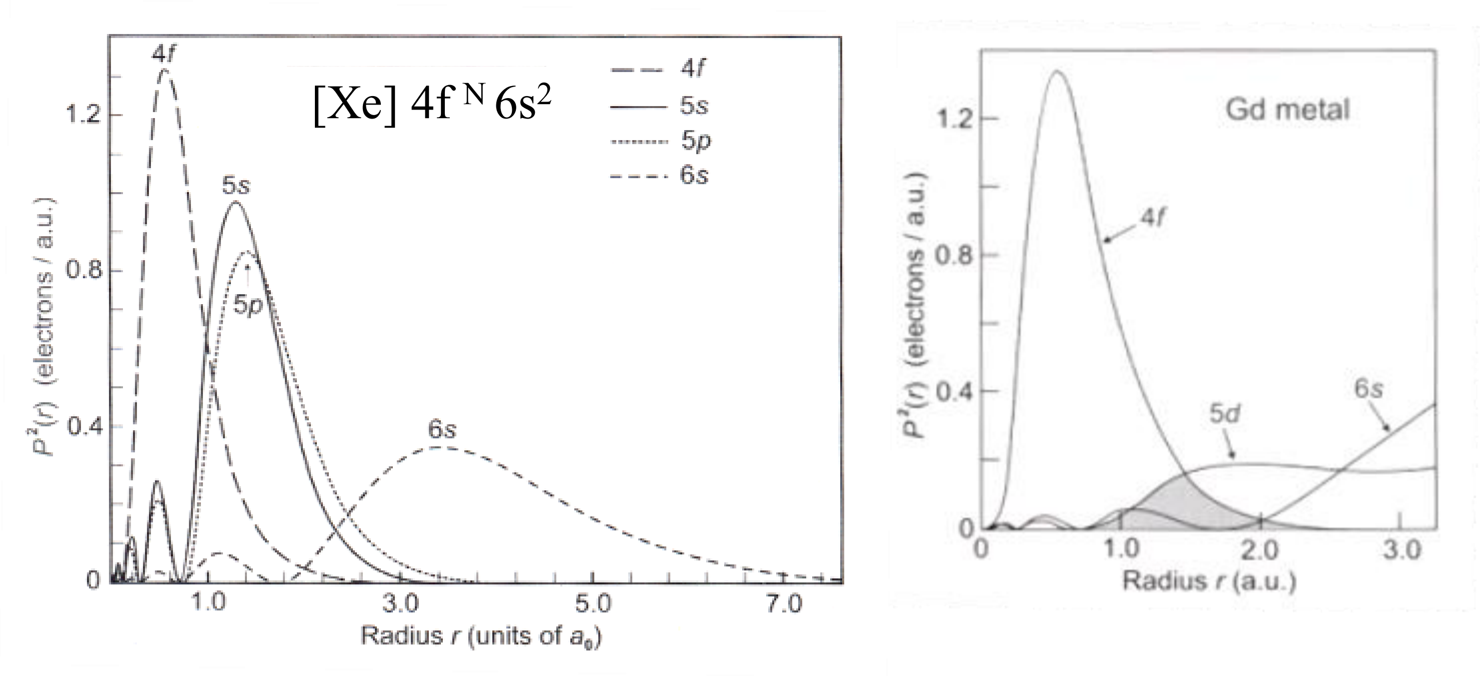
The exchange breaking layer is necessary to decouple the recording layer from the soft underlayer

The soft underlayer helps to close the magnetic flux lines

Valence orbitals in rare earths and consequences for exchange

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

In rare earths, magnetism comes from 4f states



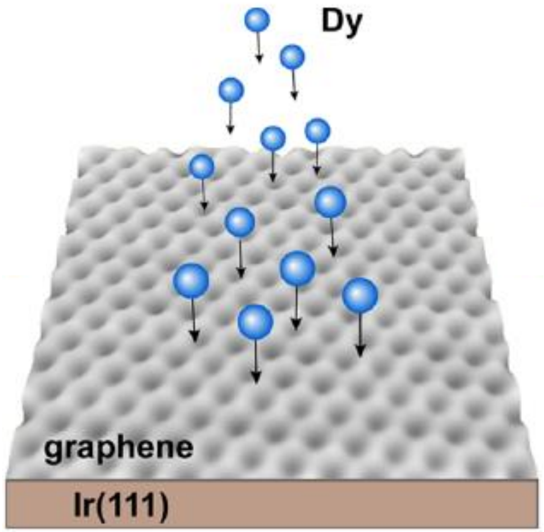
Radial distribution of the different orbitals

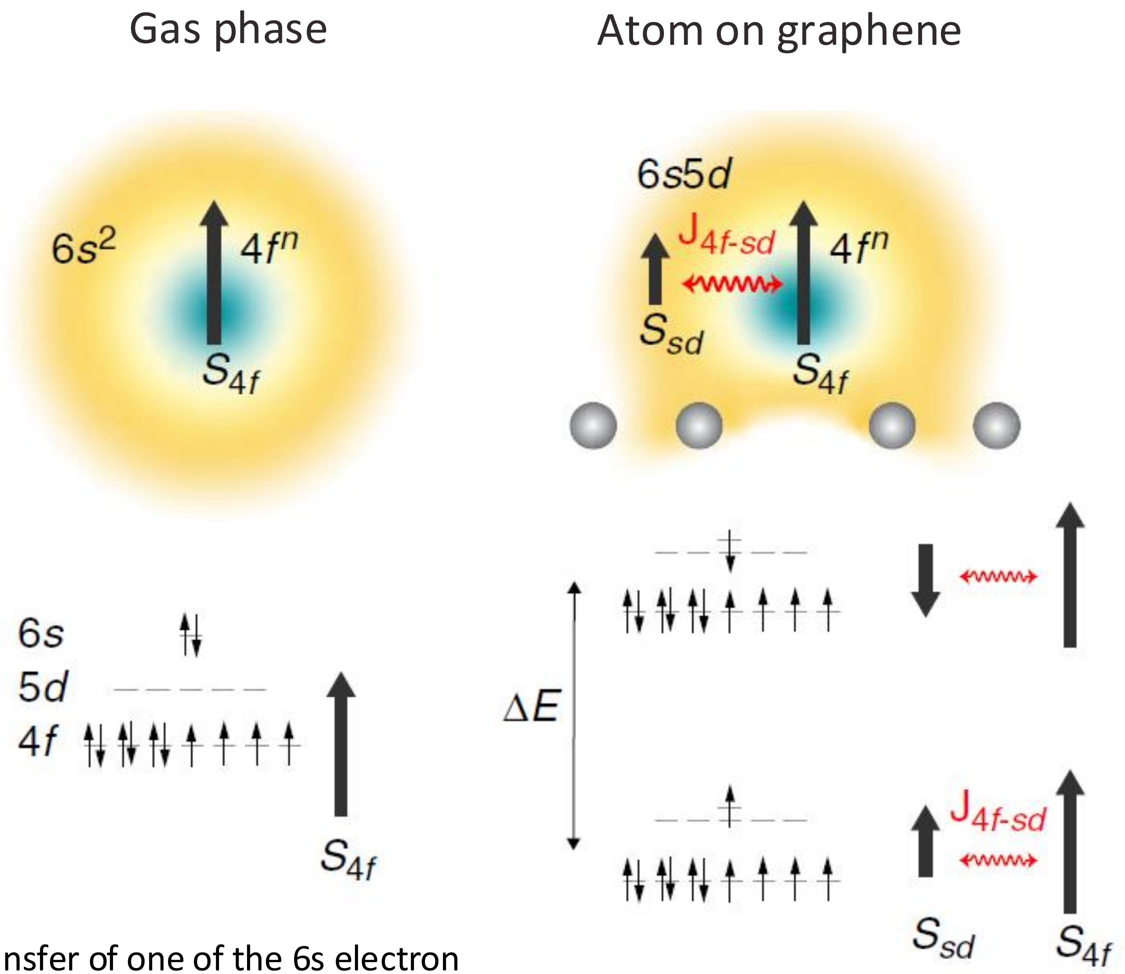
4f states are strongly localized →

do not overlap with wave functions of neighbouring atoms

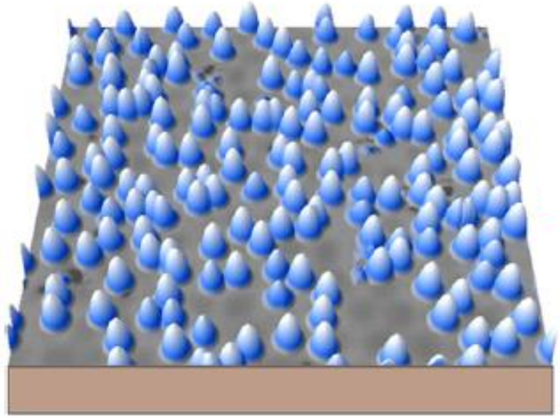
How is collective magnetism possible in rare earth compounds ?

Intra-atomic exchange in rare earths





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

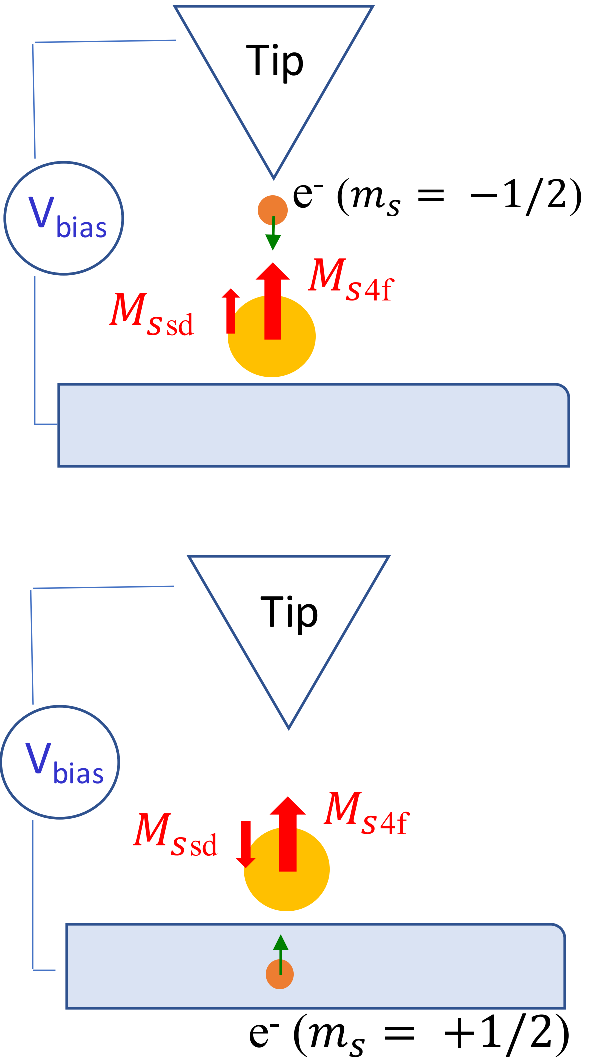


S_{sd} and S_{4f} are coupled by the intra-atomic exchange interaction $J_{4f\text{-}sd}$

$$\mathcal{H}_{ex} = -2\,J_{ex}\,\boldsymbol{S}_{4\mathrm{f}}\cdot\boldsymbol{S}_{\mathrm{sd}}$$

Measuring the intra-atomic exchange in RE with IETS

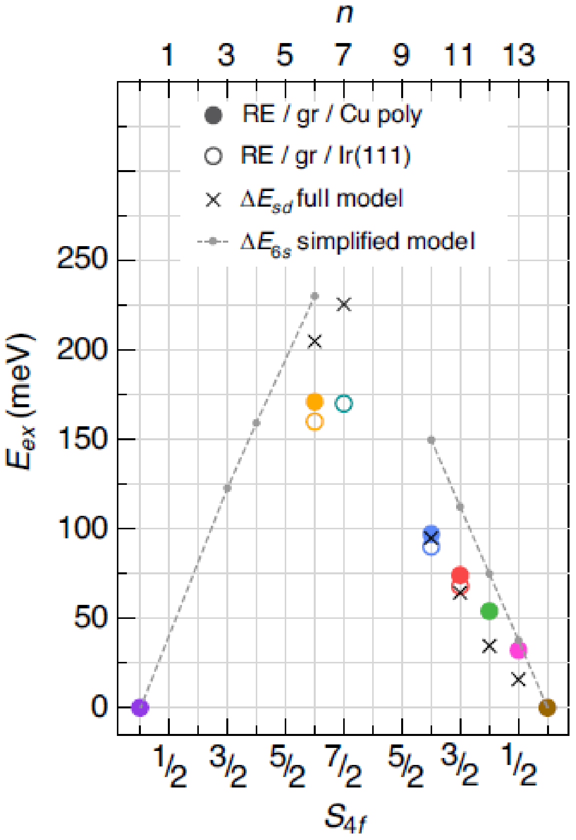
Exercise 11.2

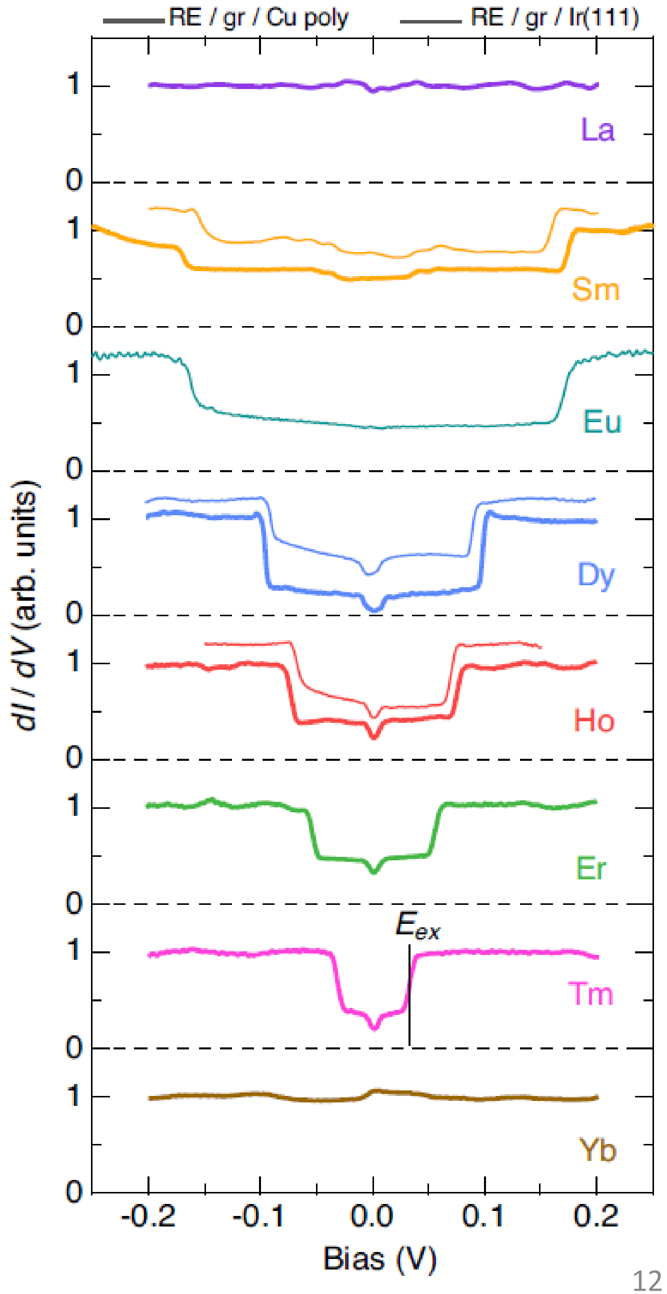


$$\mathcal{H}_{ex} = -2\,J_{ex}\,\boldsymbol{S}_{4f}\cdot\boldsymbol{S}_{sd}$$

$$\Delta m_s = +1$$
$$\downarrow$$
$$\Delta M_{s\,sd} = -1$$

$$|E_{ex}| = 2\,J_{4f-sd}S_{4f}\,\Delta S_{sd}$$





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

Inter-atomic exchange: 4f-3d strong magnets (SmCo$_5$, Nd$_2$Fe$_{14}$)

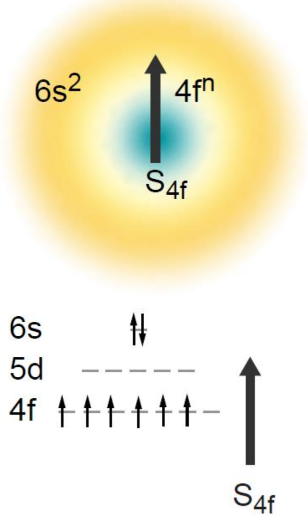
Gas phase

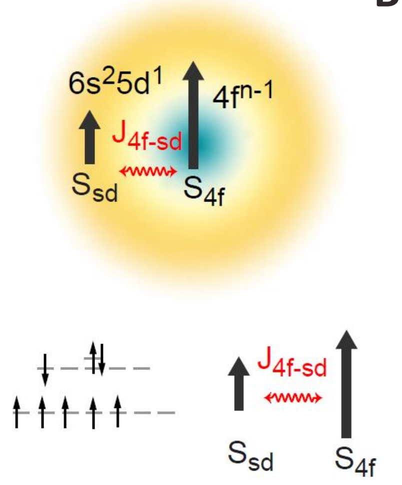
Bulk

Rare earths (RE)

[Xe] 4f^N 6s^2

$$S_{6s} = 0$$



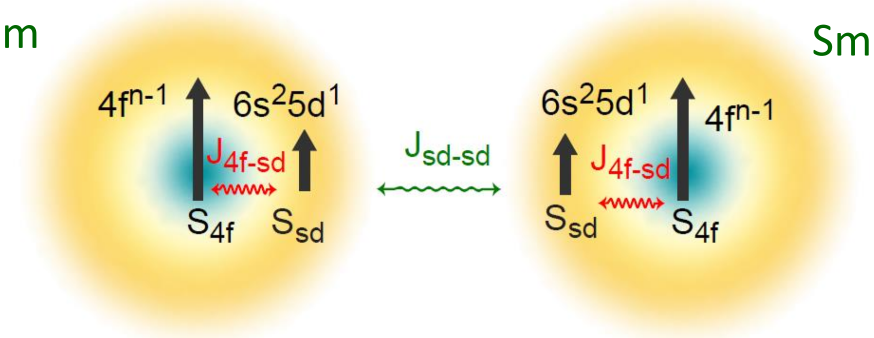


[Xe] 4f^{N-1} 6s^2 5d^1

$$S_{6s5d} \neq 0$$

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

Pure Sm

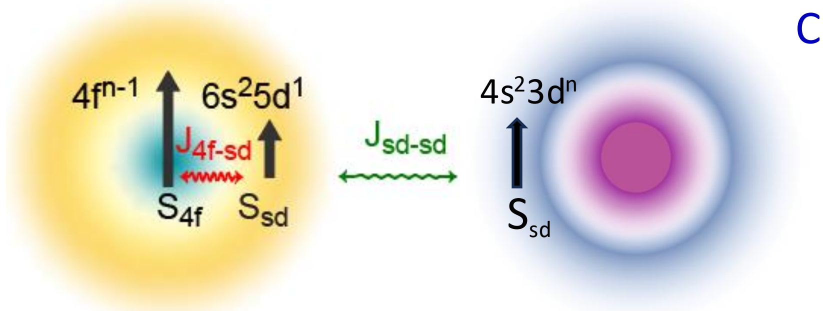


$$\mathcal{H}_{ex} = -2\, J^{\mathrm{Sm-Sm}}_{sd\_sd}\, \boldsymbol{S}^{\mathrm{Sm}}_{sd} \cdot \boldsymbol{S}^{\mathrm{Sm}}_{sd}$$

$T_C\ (T_N) < 300$ K

Gd: $T_C = 292$ K

SmCo$_5$



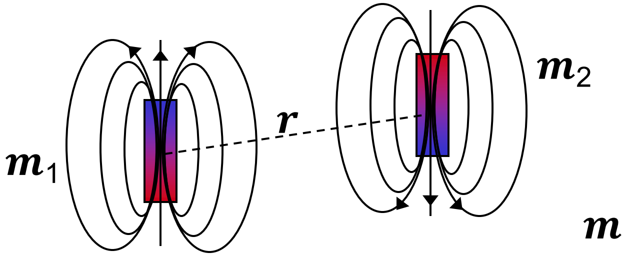
$$\mathcal{H}_{ex} = -2\, J^{\mathrm{Sm-Co}}_{sd\_sd}\, \boldsymbol{S}^{\mathrm{Sm}}_{sd} \cdot \boldsymbol{S}^{\mathrm{Co}}_{sd}$$

The 3d element increases the strength of the exchange interaction (mainly 5d-3d exchange), thus increasing T_C

SmCo$_5$: $T_C = 800$°C

Dipolar interaction

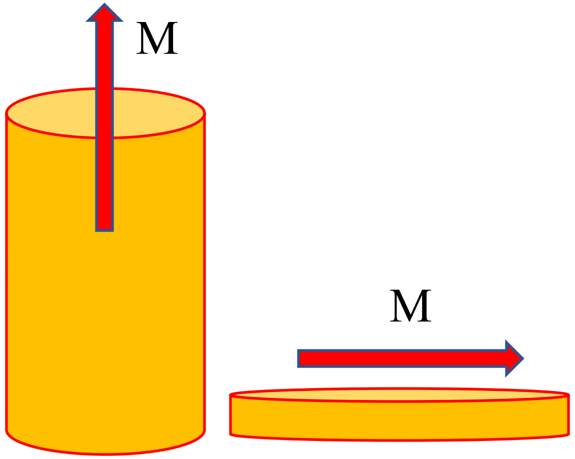
Long range interaction between magnetic moments



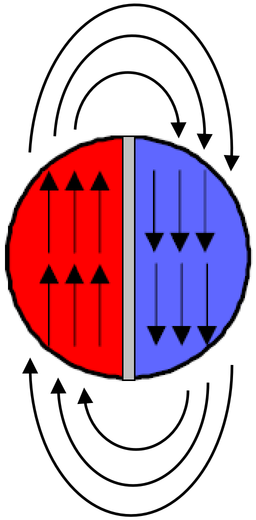
$$E_{dip} = \frac{\mu_0}{4\pi}\left[\frac{\boldsymbol{m}_1\cdot\boldsymbol{m}_2}{r^3} - 3\frac{(\boldsymbol{m}_1\cdot\boldsymbol{r})(\boldsymbol{m}_2\cdot\boldsymbol{r})}{r^5}\right]$$

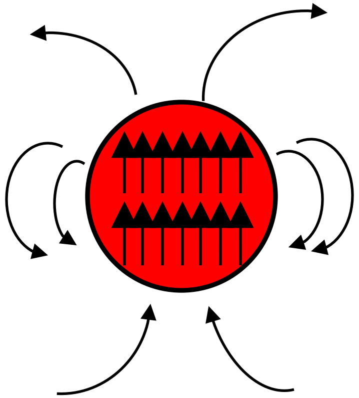
$\boldsymbol{m}_1$ and $\boldsymbol{m}_2$: magnetic moments of two atoms in a particle or moments of two particles

Magnetization orientation (shape anisotropy)

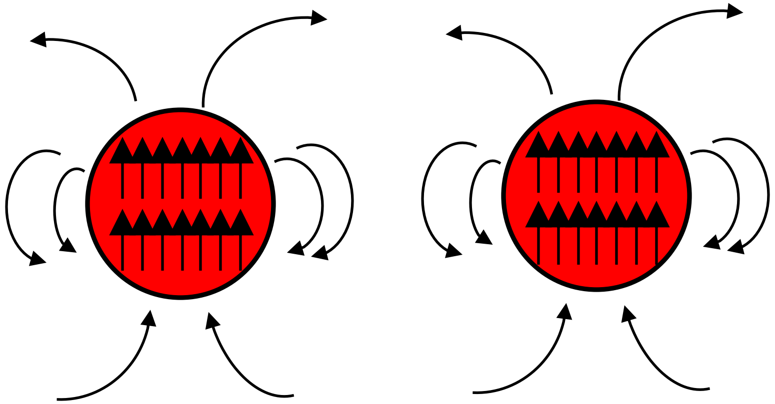


Domain formation:
competition between exchange and dipolar energy





Interaction between particles



Stray and demagnetizing field

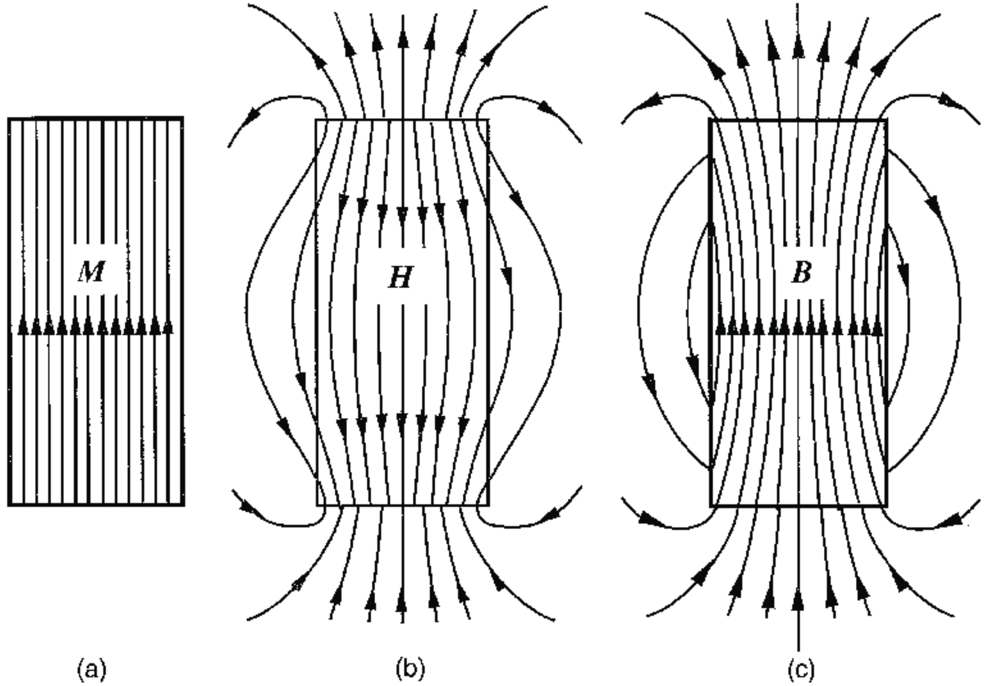
$\boldsymbol{B} = \mu_0(\boldsymbol{H} + \boldsymbol{M})$

In free space: $\boldsymbol{M} = 0$ and $\boldsymbol{B} = \mu_0 \boldsymbol{H}$
almost indistiguishable

Inside a magnet:
$\boldsymbol{B}$-field and the $\boldsymbol{H}$-field are oppositely directed;
$\boldsymbol{H}$ is oppositely directed to $\boldsymbol{M}$
$(\boldsymbol{\nabla} \cdot \boldsymbol{B} = 0 \quad \rightarrow \boldsymbol{\nabla} \cdot \boldsymbol{M} = -\boldsymbol{\nabla} \cdot \boldsymbol{H})$

Usual nomenclature:
Stray field $\boldsymbol{H}_s$ (outside) and demagnetizing field $\boldsymbol{H}_d$ (inside)

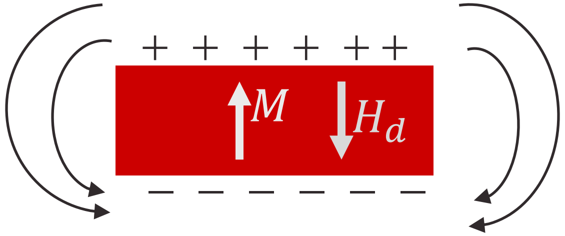
Microscopic origin: dipolar interaction
Each atom (= dipole) creates a magnetic field that interacts with all the other dipoles around it.
The demagnetizing field is the average field acting on each dipole.



Demagnetizing energy

Infinite plate

magnetic poles only at very far ends
$\rightarrow H_d = H_s = 0$



magnetic poles at the two faces
$\rightarrow H_d \neq 0, H_s \neq 0$

No demagnetizing field, no stray field outside the plate

The demagnetizing field H_d is equal and opposite to M, the flux lines extend outside the plate

Energy of a magnetic sample
(called demagnetization energy, magnetostatic energy or dipolar energy)

$$E_d = -\frac{\mu_0}{2}\int_V \boldsymbol{M}\cdot\boldsymbol{H}_d\,dV$$

The orientation of $\boldsymbol{M}$ (and therefore of $\boldsymbol{H}_d$) minimizes this energy (minimize demagnetizing and stray field)

Shape anisotropy

The demagnetizing field can be an extremely complicated function of position for a magnet of arbitrary shape.

$$E_d = -\frac{\mu_0}{2}\int_V \boldsymbol{M}\cdot\boldsymbol{H}_d dV$$

In simple, homogenous cases, it can be expressed with the help of a tensor, the demagnetizing tensor $\boldsymbol{\mathcal{D}}$

$$\boldsymbol{H}_d = -\boldsymbol{\mathcal{D}}\,\boldsymbol{M}$$

$$E_d = \frac{\mu_0}{2}\int_V \boldsymbol{M}\,\boldsymbol{\mathcal{D}}\boldsymbol{M}\,dV$$

Simple shapes

cylinder axis along z
plate parallele to z

Sphere:

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

∞-Cylinder:

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

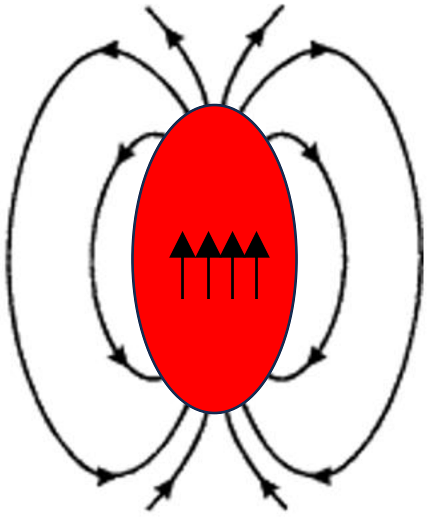
∞-Plane (thin film):

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

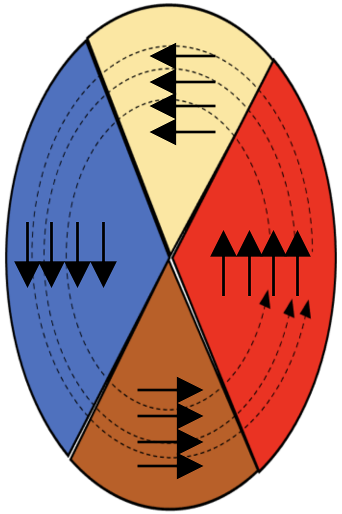
Sphere : all directions are equivalent
Cylinder : $\boldsymbol{M}$ // axis
Plate : $\boldsymbol{M}$ // surface

Magnetization state for in-plane magnetized particle

To minimize the dipolar energy, small particles/nanostructures exhibit shape anisotropy, while larger structures spontaneously form domains (energy balance between exchange and dipolar interaction)



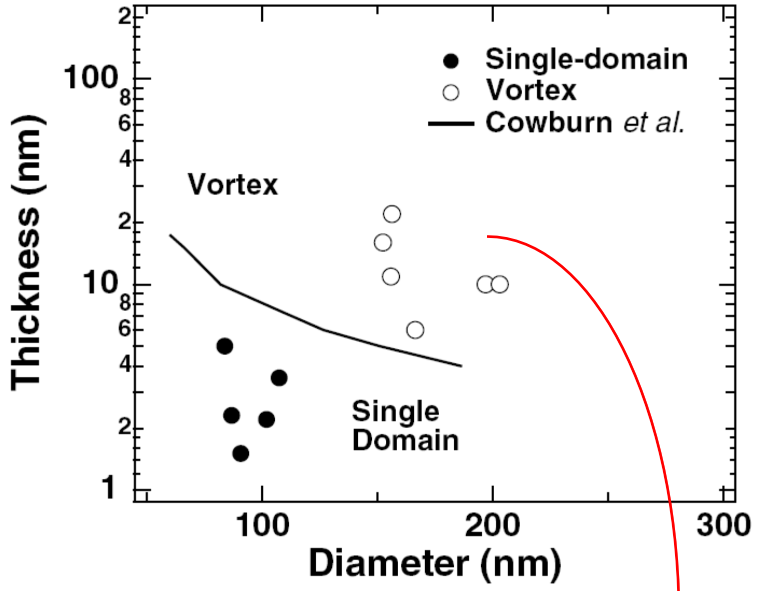
Flux lines extend outside the particle



Flux lines are confined inside the particle (magnetic closure domains, domain wall formation)

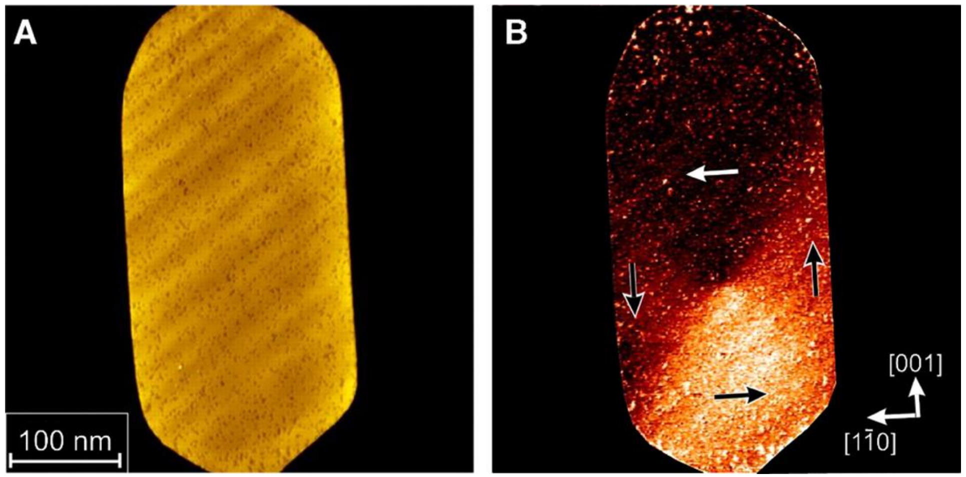
R. Skomski *et al*. Phys.Rev. Lett. **91**, 127201 (2003)
A. Wachowiak *et al*.. Science **298**, 577 (2002)

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



Topography

SP- STM



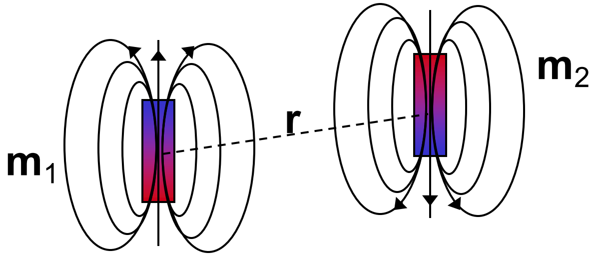
magnetic domain pattern of a 8 nm high Fe island grown on W(110)

Longitudinal *vs.* perpendicular recording media

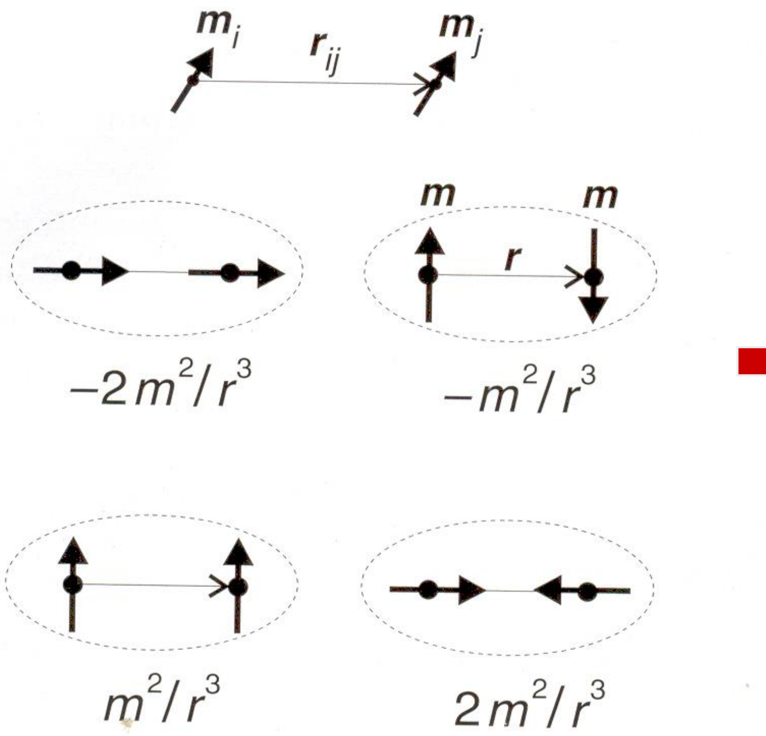
Exercise 11.3

Long range interaction between magnetic moments

$$E_{dip} = \frac{\mu_0}{4\pi}\left[\frac{\boldsymbol{m}_1 \cdot \boldsymbol{m}_2}{r^3} - 3\,\frac{(\boldsymbol{m}_1 \cdot \boldsymbol{r})(\boldsymbol{m}_2 \cdot \boldsymbol{r})}{r^5}\right]$$



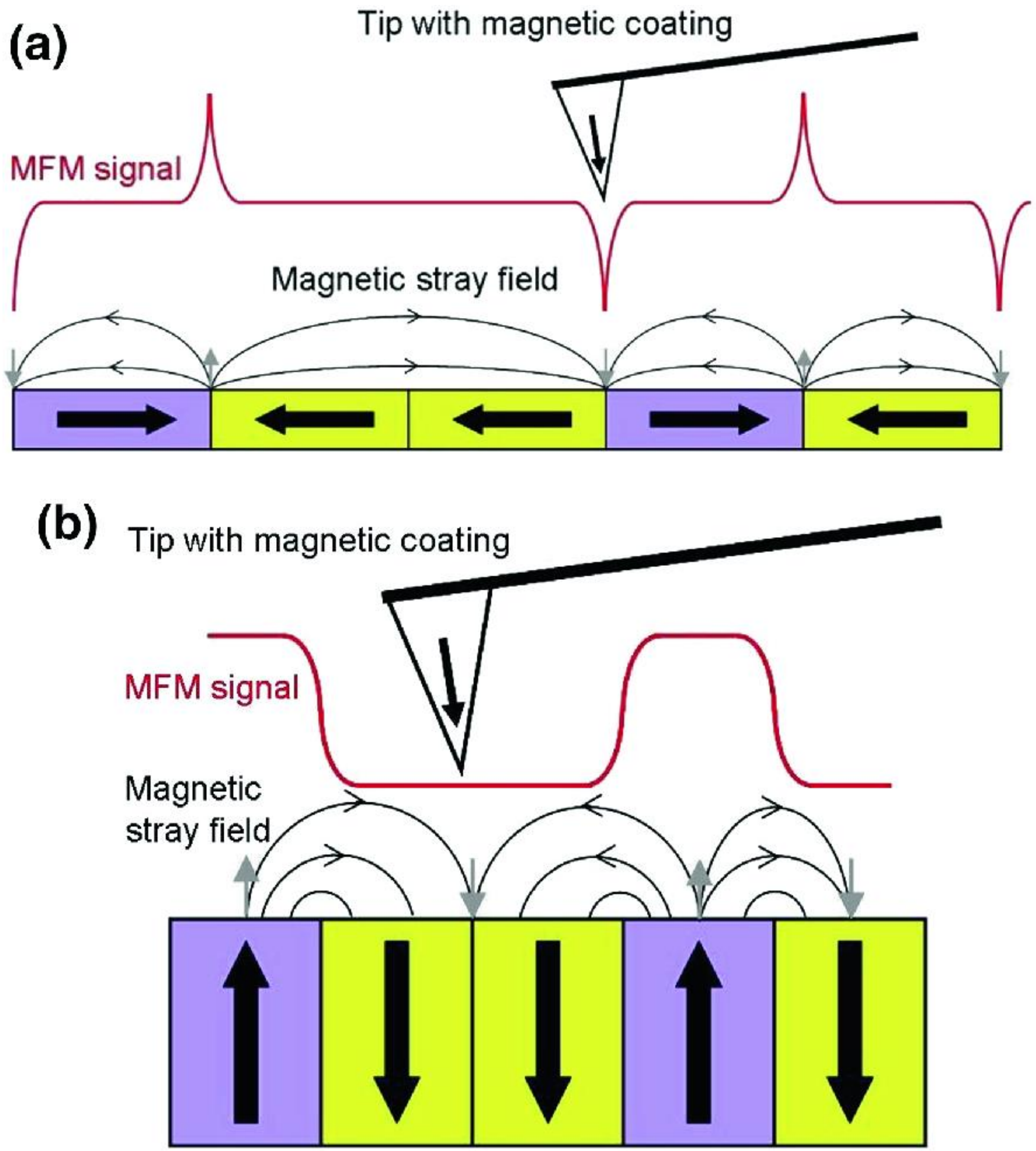
$\boldsymbol{m}_1$ and $\boldsymbol{m}_2$ the magnetic moments of two particles



The out-of-plane configuration reduces the dipolar interaction

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

MFM: magnetic force microscope



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 emanating from the surface (large gradient, grey arrows). 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)

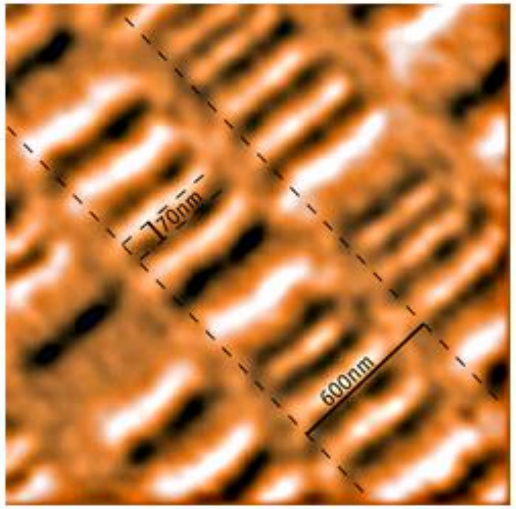
https://link.springer.com/chapter/10.1007/978-3-030-15612-1_8

AFM *vs* MFM

AFM

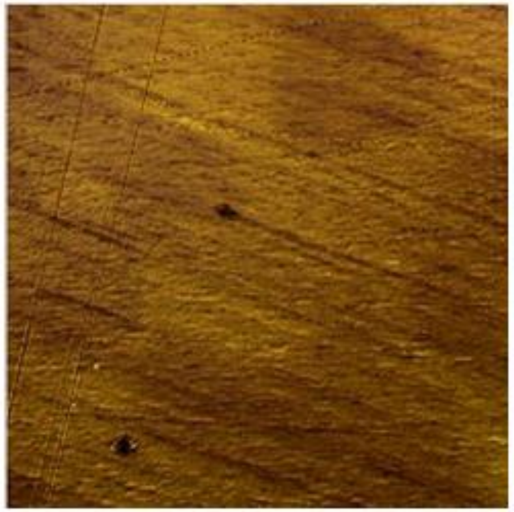
MFM

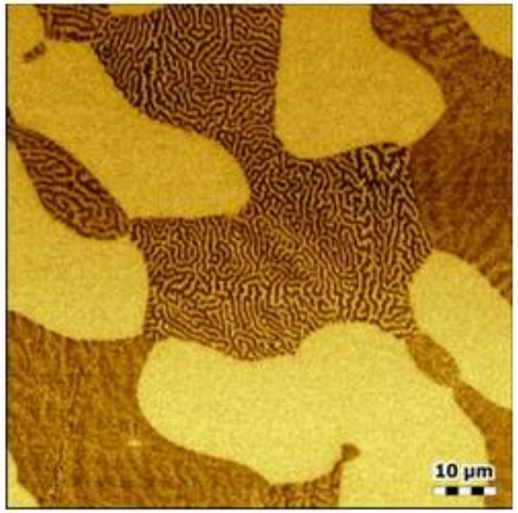
Hard-disk drive platter with **in-plane** magnetized media

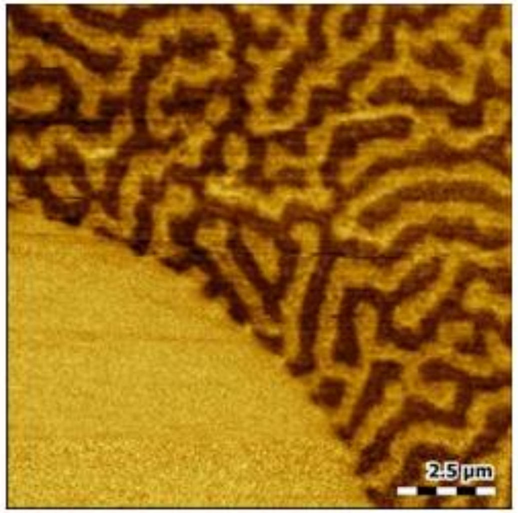


Black and white regions are the domain wall between in-plane magnetized domains

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