BASICS AND MAGNETIC MATERIALS

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History of magnetism

- the names magnetism, magnets etc. go back to ancient Greeks: magnetite = loadstone (Fe_3O_4)

- The magnetism of magnetite was also known in ancient China
  - spoon-shaped compass 2000 years ago
  - long time used for geomancy
  - open-see navigation since 1100

European traditions in magnetism

- **A. Neckham** (1190): describes the compass
- **P. Peregrinus** (1269): *Epistola Petri Peregrini … de Magnete*
  - terrella, poles

- **E.W. Gilbert** (1544 – 1603)
  de Magnete

- **R. Descartes** (1569 – 1650)
  divorced physics from metaphysics

Modern developments in the 19th century

H.C. Oersted (1777-1851)
- electric currents are magnets

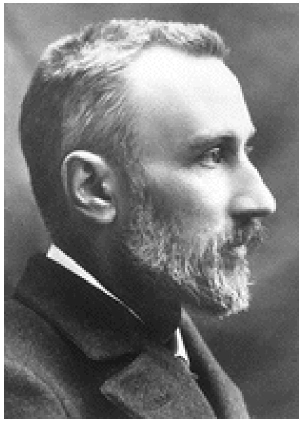
A.M. Ampère (1755-1836)
- $\nabla \times \mathbf{H} = \mathbf{j}$
- molecular currents

M. Faraday (1791-1867)
- magnetic field
- $\nabla \times \mathbf{E} = -\dot{\mathbf{B}}$

J.C. Maxwell (1831-1871)
- unification of light, electricity, magnetism
- $\mathbf{j} \rightarrow \mathbf{j} + \varepsilon_0 \dot{\mathbf{E}}$
- prediction of radio waves

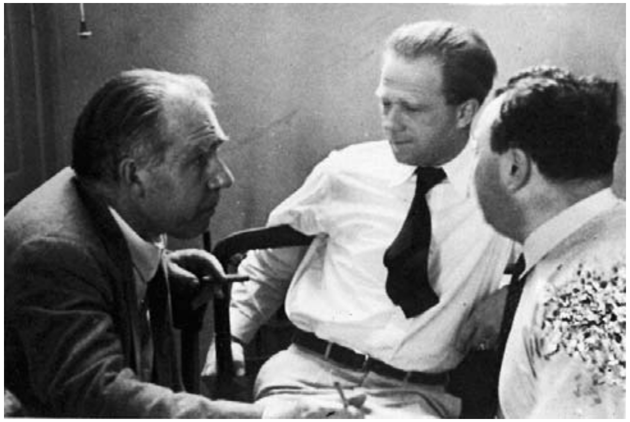
H.A. Lorentz (1853-1928)
- $m\dot{\mathbf{v}} = e(\mathbf{E} + \mathbf{v} \times \mathbf{B})$
- Lorentz transformation

The revolutionary 20th century

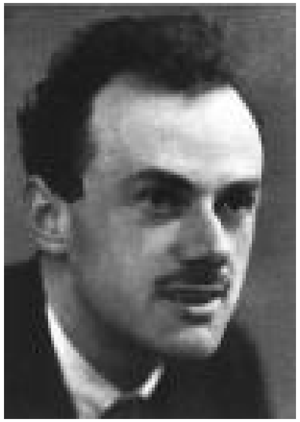


P. Curie (1859-1906)
- paramagnetism and ferromagnetism

P. Weiss
- the molecular field
- domains



N. Bohr, W. Heisenberg, W. Pauli
- quantum theory
- the need of spin



P. Dirac
- relativistic quantum theory
- explains the spin

→ exchange interaction ←

Magnetism and magnetic materials in our daily life

- earth's field
- natural electromagnetic waves
- TV
- portable phone
- telecommunication
- home devices
- 50 permanent magnets in an average home
- up to 100 permanent magnets in a modern car
- soft magnetic materials in power stations and motors and high frequency devices
- traffic
- medicine
- information technology

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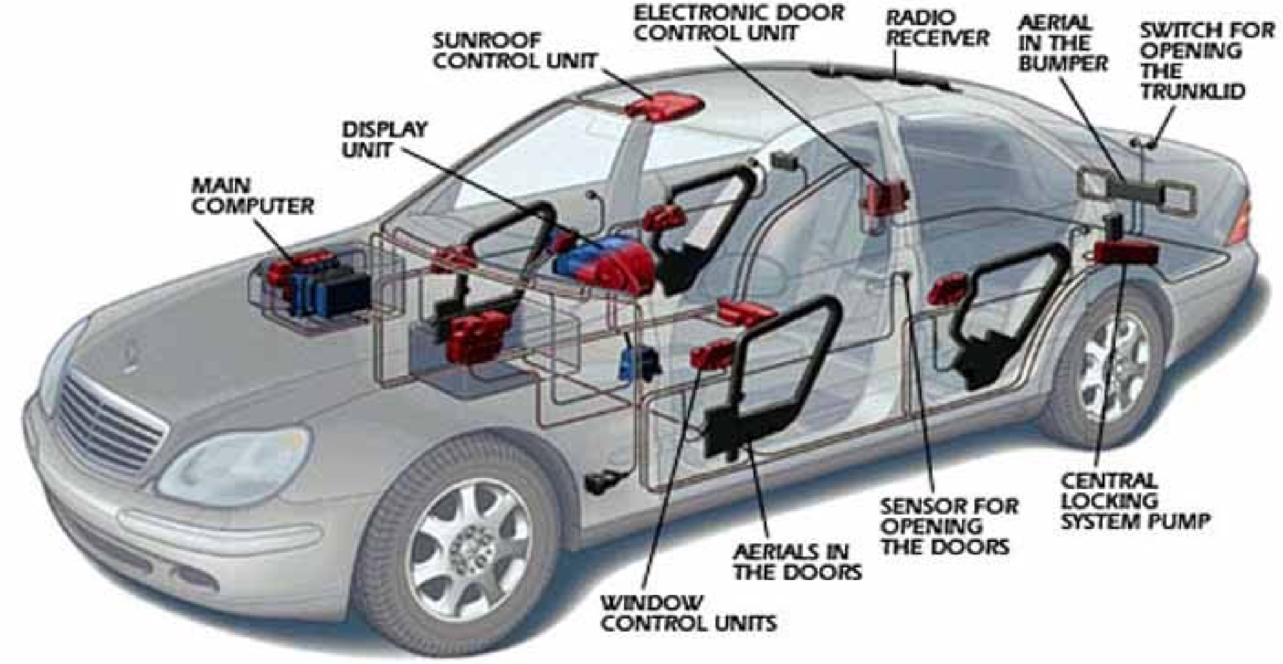
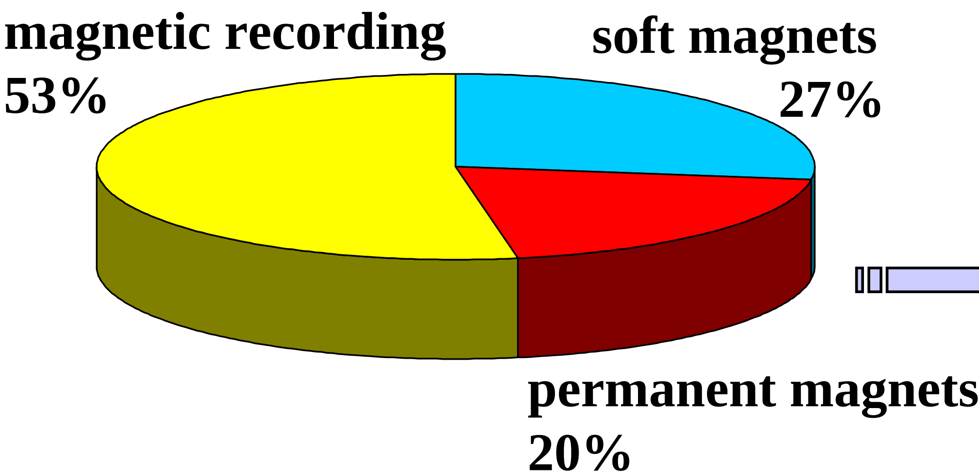


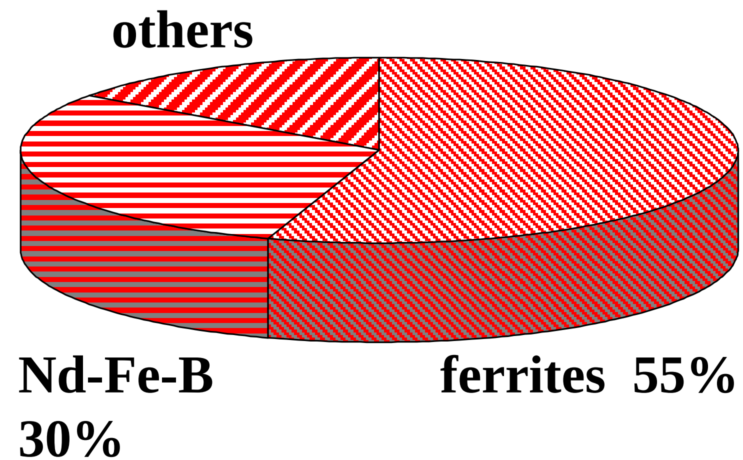
Magnet materials – world market

- in the late 20th century -

all magnetic materials
total 30B$

permanent magnet materials





Fields in nature, engineering and science

in Tesla

Brain ; Intergalactic space	10^{-13}		
Heart	10^{-10}		
Galaxy	10^{-9}		
urbanic noise	$10^{-6}…10^{-8}$		
Surface of earth	$5·10^{-5}$		
near power cable (in home)	10^{-4}		
Surface of sun	10^{-2}		
surface of magnetite	$5·10^{-1}$	simple resistive coil	10^{-1}
		permanent magnets	**1**
		supercond. permanent magnets	**16**
		superconducting coils	**20**
		high performance resistive coils (stationary)	**26**
		hybrid magnets (resistive + supercond.)	**40**
		long pulse coil (100ms)	**60**
		short pulse coil (10 ms)	**80**
Anisotropy fields in solids	$\leq 10^{2}$	one-winding coil	$3·10^{2}$
Exchange fields in solids	$\leq 10^{3}$	explosive flux compression	$3·10^{3}$
Neutron stars	10^{8}		

2 – Magnetic moment and magnetization

2-1 Magnetization in Maxwell's equations

microscopic form of Maxwell's equ.

$$\nabla \times \mathbf{h} = \varepsilon_0 \dot{\mathbf{e}} + \mathbf{i} \qquad \nabla \mathbf{h} = 0$$

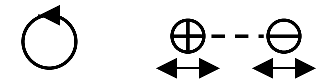
$$\nabla \times \mathbf{e} = -\mu_0 \dot{\mathbf{h}} \qquad \nabla \varepsilon_0 \mathbf{e} = r$$

$$\dot{r} + \nabla \mathbf{i} = 0$$

magnetization **M** and polarization **P**

$$r = \rho - \nabla \mathbf{P}$$

$$\mathbf{i} = \mathbf{j} + \nabla \times \mathbf{M} + \dot{\mathbf{P}}$$



$$\Rightarrow \qquad \nabla \times (\mathbf{h} - \mathbf{M}) = \varepsilon_0 \dot{\mathbf{e}} + \dot{\mathbf{P}} + \mathbf{j} \qquad \nabla(\varepsilon_0 \mathbf{e} + \mathbf{P}) = \rho$$

macroscopic fields

$$\overline{\mathbf{h}} \equiv \mathbf{B}/\mu_0 \quad \Rightarrow \quad \mathbf{H} \equiv \mathbf{B}/\mu_0 - \mathbf{M}$$

$$\overline{\mathbf{e}} = \mathbf{E} \quad \Rightarrow \quad \mathbf{D} \equiv \varepsilon_0 \mathbf{E} + \mathbf{P}$$

macroscopic form of Maxwell's equ.

$$\nabla \times \mathbf{H} = \dot{\mathbf{D}} + \mathbf{j} \qquad \nabla \mathbf{B} = 0$$

$$\nabla \times \mathbf{E} = -\mu_0 \dot{\mathbf{H}} \qquad \nabla \mathbf{D} = \rho$$

$$\rho + \nabla \mathbf{j} = 0$$

magnetic moment and magnetization

$$\Rightarrow \nabla \mathbf{H} = -\nabla \mathbf{M} \quad \Rightarrow \quad \mathbf{m} \equiv -\int_V d\tau\, \mathbf{r} \nabla \mathbf{M} = \ldots = \int_V d\tau\, \mathbf{M}$$

$\Rightarrow$ **M** is a density of magnetic moments $\equiv$ "magnetization"

2-2 Magnetic moment and angular momentum

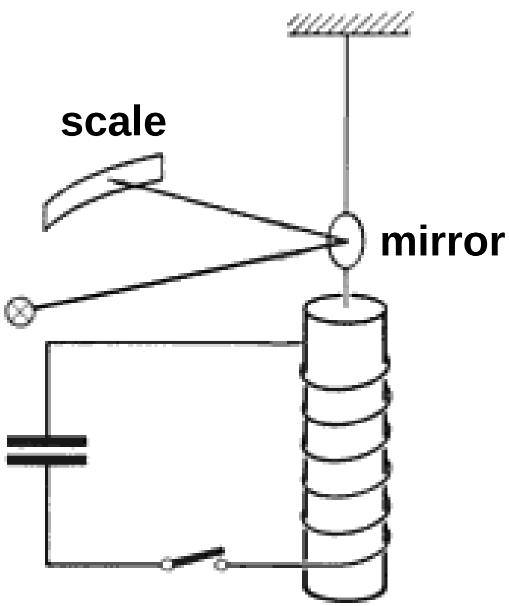
$$\mathbf{m} = -\int_V d\tau\, \mathbf{r}\,(\nabla \mathbf{M}) = \ldots = \frac{1}{2}\int_V d\tau\, \mathbf{r} \times (\nabla \times \mathbf{M})$$

$$\Rightarrow \quad \mathbf{m} = \frac{e}{2m}\int_V d\tau\, \rho_m\, \mathbf{r} \times \mathbf{v} = \frac{e}{2m}\mathbf{L} = g\,\frac{e}{2m}\mathbf{L}$$

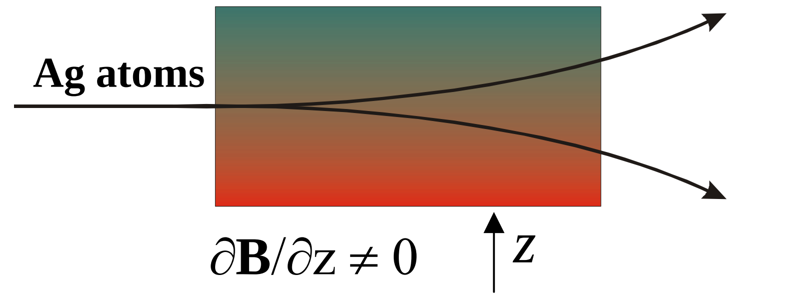
A. Einstein and W.J. de Haas (1915): $g \approx 2$ (instead of 1)

The spin of the electron and its g-factor

A. Einstein and W.J. de Haas (1915)



W. Gerlach and O. Stern (1922)



G. Uhlenbeck and S. Goudsmit (1925): atomic spectra at $\mathbf{B} \neq 0$ (Zeeman effect)

spin of the electron: $\mathbf{S} \to \pm \hbar/2$ $\qquad g = 2$

2-3 Quantization and relativity; diamagnetism

H.A. Lorentz $m\dot{\mathbf{v}} = e(\mathbf{E} + \mathbf{v} \mathrm{x} \mathbf{B})$

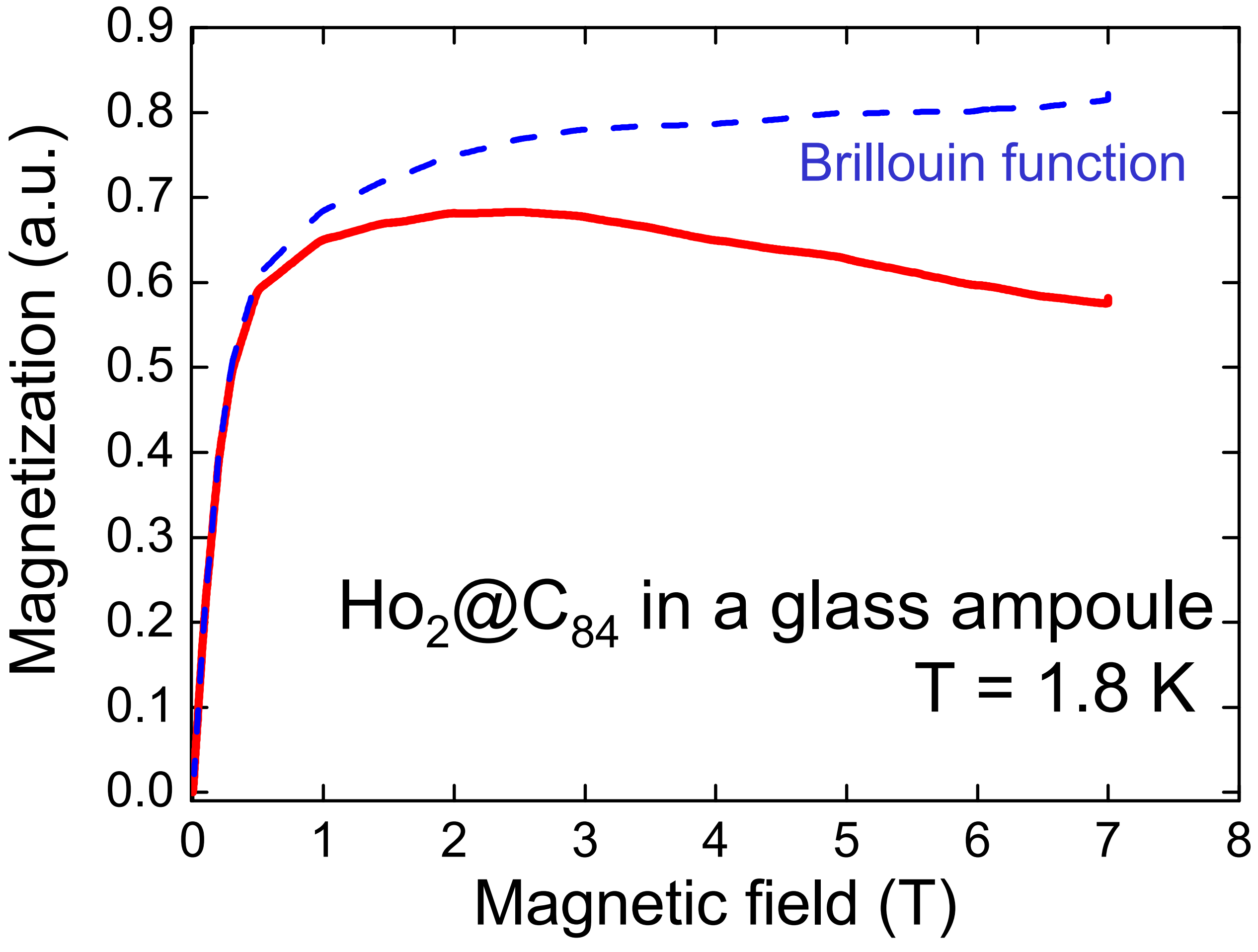
Dirac's Hamiltonian $\mathcal{H} = \mathcal{H}_0 - \frac{e}{2m}(\mathbf{L} + 2\mathbf{S})\mathbf{B} + \frac{e^2}{2m}\frac{\mathrm{B}^2}{4}(\mathrm{x}^2 + \mathrm{y}^2) - \lambda(\mathbf{LS})$

$\Rightarrow$ $\mathbf{m} = <\mu> = -\,\partial<\mathcal{H}>/\partial\mathbf{B}$ $\mathbf{M} = \mathcal{N}<\mu>$

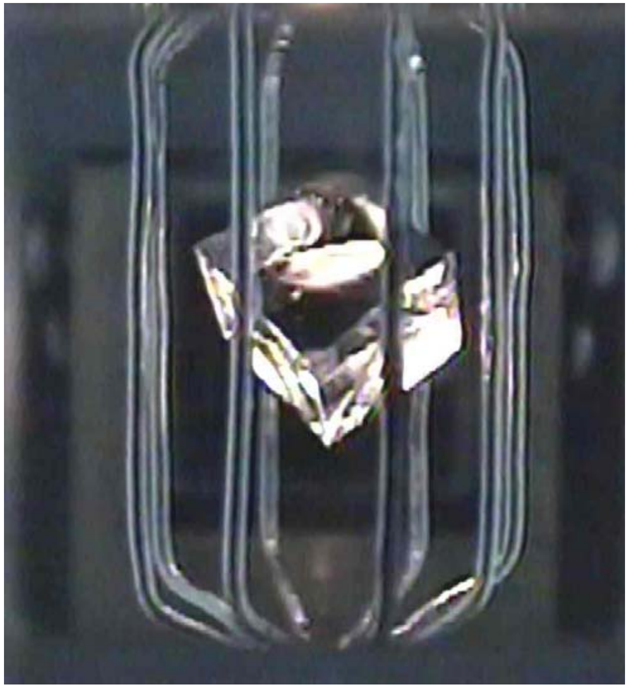
$$\chi_{ij} = \mu_0 \partial \mathrm{M}_i / \partial \mathrm{B}_j = -\,\mu_0 \mathcal{N} \partial^2 < \mathcal{H} > / \partial \mathrm{B}_i \partial \mathrm{B}_j$$

diamagnetic susceptibility (*in* 10^{-6}): H_2O: –9, alcohol: –7.2

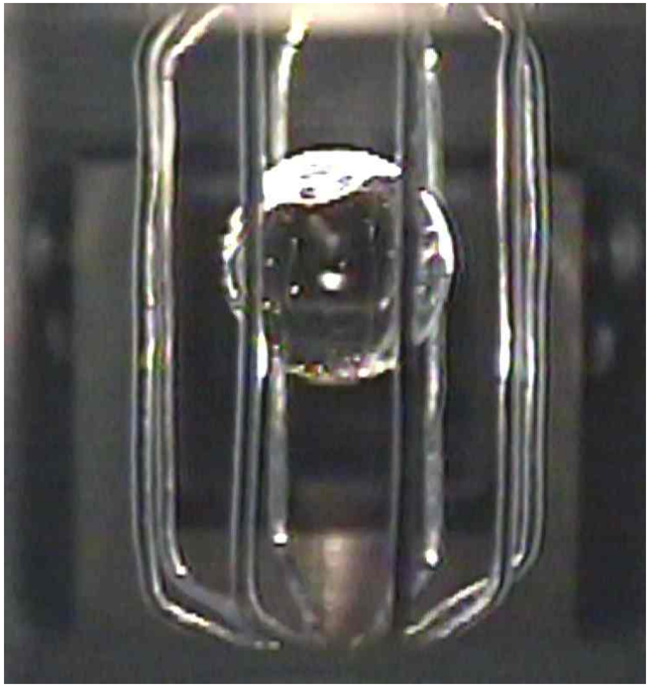
Omnipresence of diamagnetism



Levitation of a diamagnetic body



levitated glass cube



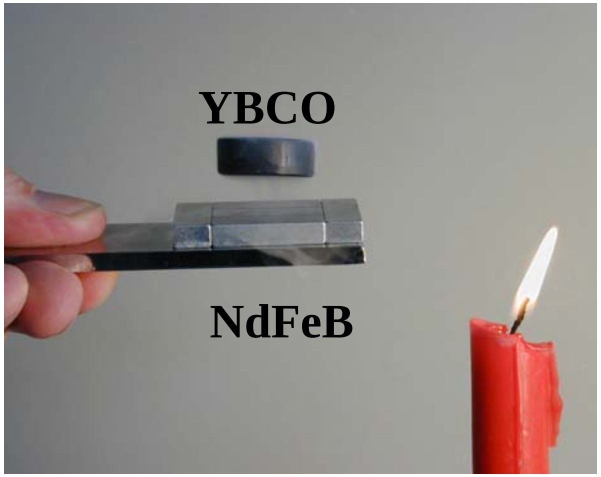
melted by a laser beam

- $\mu_0 H \approx 23$ Tesla
- $F \sim \chi \cdot H \cdot (dH/dz)$
- stable only for $\chi < 0$

M. Kitamura et al., Jpn. J. Appl. Phys. 39 (2000) L324

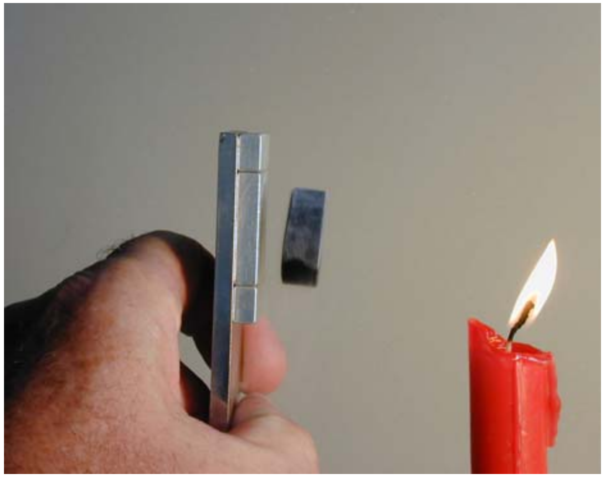
Stable position of a superconducting permanent magnet

Levitation



Suspension

Cross stiffness



- by varying the spatial distribution of the external field the position of the superconducting magnet may have different degrees of freedom :

  0 D – as in the examples above
  1 D – as a train on a rail

- the same holds for rotational degrees of freedom

2-4 Magnetization in thermodynamics

problems:

(i) thermodynamically metastable states

(ii) the field generated by the samples own magnetization

(iii) how to define a correct expression for magnetic work

- Quantum statistical thermodynamics with the Hamiltonian $\mathcal{H}$

$$d\langle\mathcal{H}\rangle = \delta Q - \mu_0 \mathbf{m}\, d\mathbf{H} = \delta Q - \mu_0 \int_V d\tau\, \mathbf{M}\, d\mathbf{H}$$

- alternative definition of internal energy

$U = \langle\mathcal{H}\rangle + \mu_0\, \mathbf{H}\,\mathbf{m}$ (a Legendre transformation)

$$\Rightarrow \quad dU = \delta Q + \mu_0 \mathbf{H}\, d\mathbf{m} = \delta Q + \mu_0 \int_V d\tau\, \mathbf{H}\, d\mathbf{M}$$

$\Rightarrow$ both expressions are correct: work done on different systems !

(iv) magnetostatic interaction is long range

Magnetism in atoms and condensed matter

Periodic System of Elements

1 **H**																	2 **He**
3 **Li**	4 **Be**											5 **B**	6 **C**	7 **N**	8 **O**	9 **F**	10 **Ne**
11 **Na**	12 **Mg**											13 **Al**	14 **Si**	15 **P**	16 **S**	17 **Cl**	18 **Ar**
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**
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**
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**

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**

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2-5 Localized vs. itinerant electron magnetism in solids

- isolated atoms or ions with incompletely filled electron shells

- magnetization: $M \sim B_J(H/T)$

$\Rightarrow$ *Curie*'s law $M = \chi H$ with $\chi = C/T$

- Two main types of solids
  - large electron density $\Rightarrow$ delocalized (itinerant) electrons – e.g. in Li- or Na-metal
    $\Rightarrow$ Hund's rule magnetic moment disappears
    $\Rightarrow$ a small, (nearly) temperature independent susceptibility
  - small electron densities
    $\Rightarrow$ strongly correlated electrons
    $\Rightarrow$ they can be localized and carry a magnetic moment
    $\Rightarrow$ examples: MnO, FeO, CoO, CuO (antiferromagnets); EuO, $CrCl_3$ (ferromagnets)

- In 4f materials localized and itinerant electrons coexist

2-6 Itinerant electron magnetism

- weakly interacting Landau quasiparticles

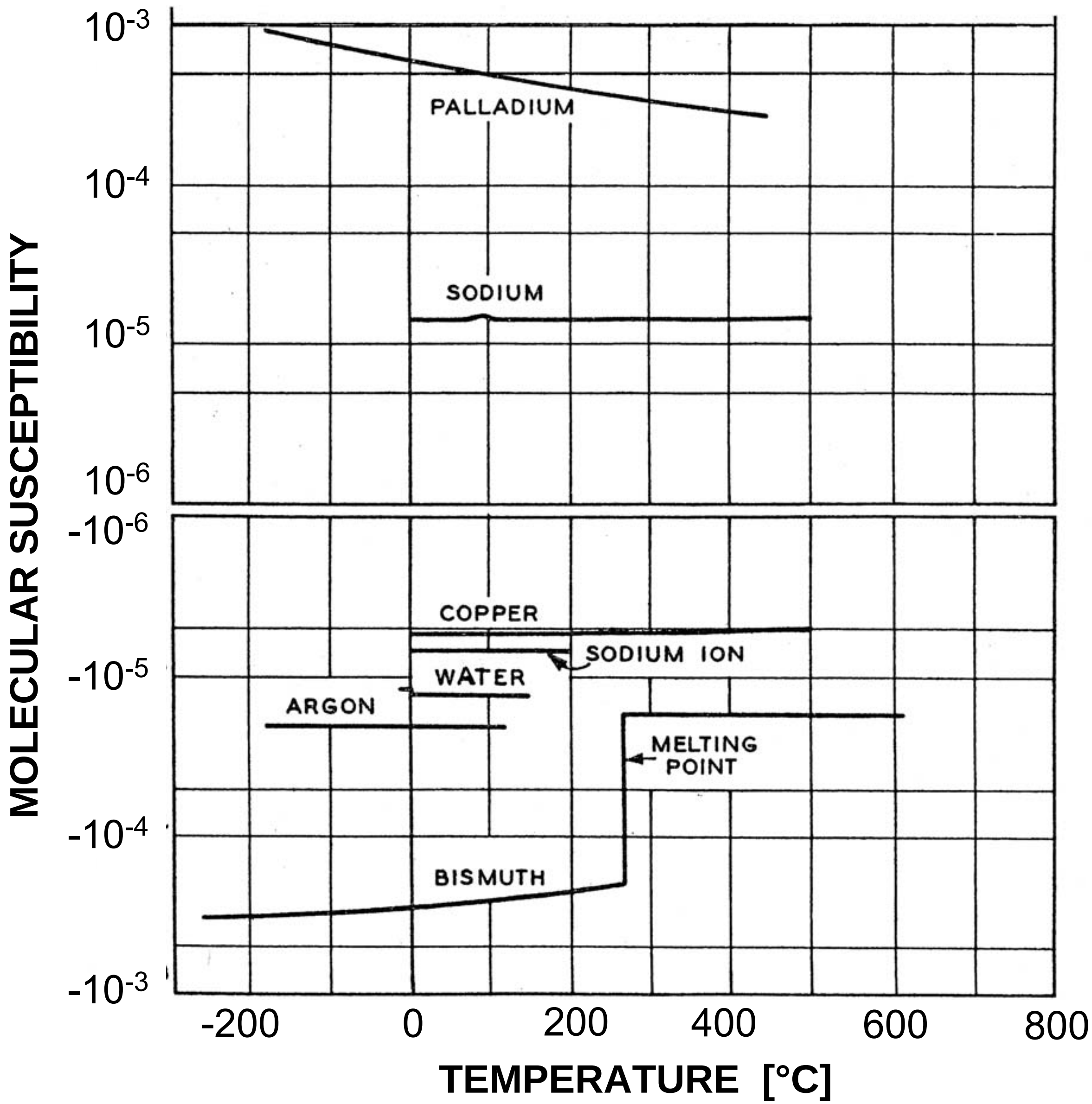
$$\chi = 2\mu_0\mu_B^2 N(E_F)\,(1 - \frac{1}{3}\frac{m^2}{m^{*2}}) \qquad \text{with} \qquad \mu_B = \frac{|e|\,\hbar}{2m}$$

$\Rightarrow$ metals with large or moderate m^* (e.g. Na: $m/m^* \approx 1$)
are Pauli paramagnets, $\chi > 0$

$\Rightarrow$ those with small m^* (e.g. Bi $m/m^* \approx 10^2$)
are Landau diamagnets, $\chi < 0$

Susceptibility *vs*. temperature

after *M. Bozort* 1951



Itinerant electron magnetism

- large fields and low temperatures

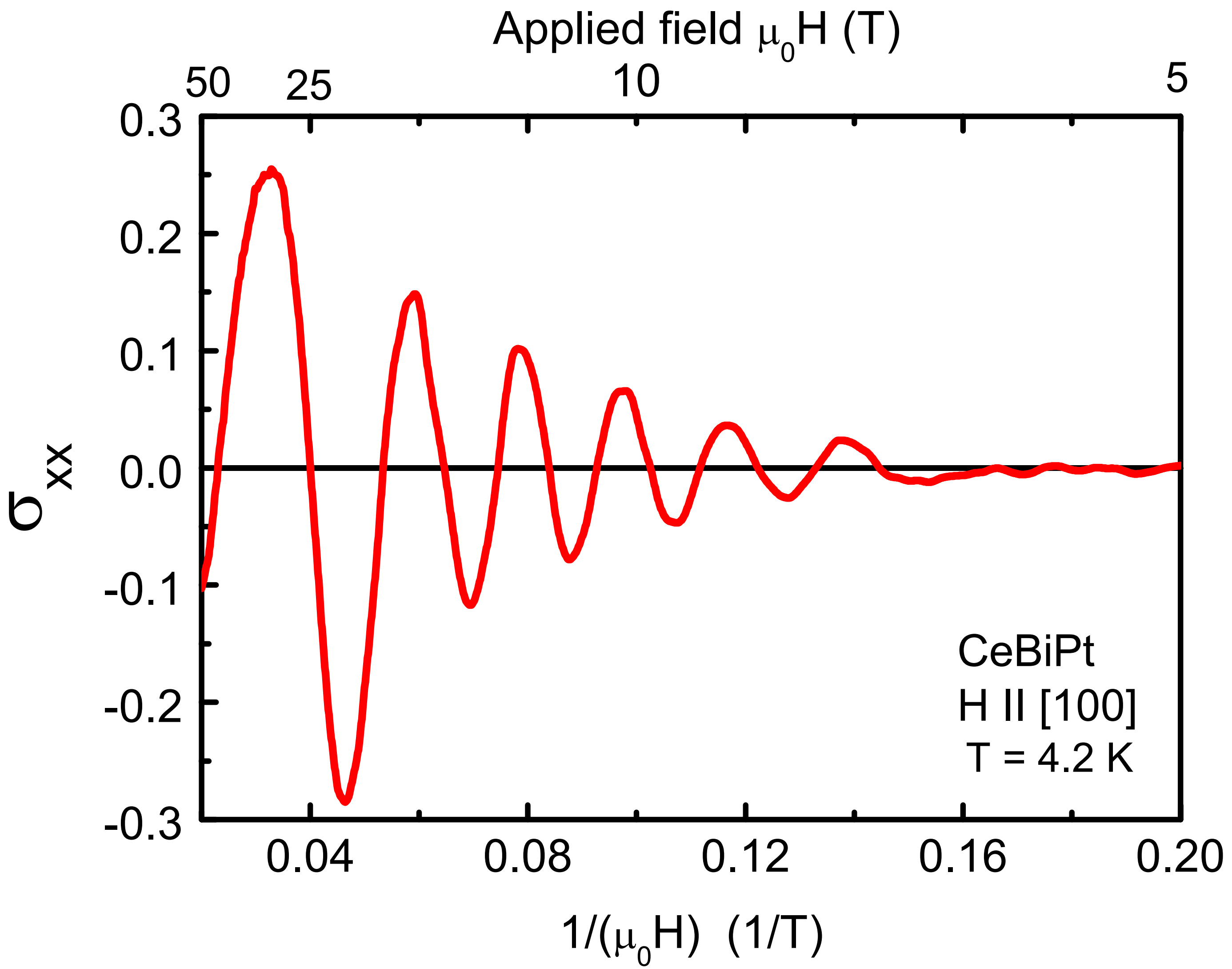
  $\Rightarrow$ discrete Landau levels:

  $$E_n = (n + 1/2)\hbar \frac{\mu_0 |e| H}{m^*} + \frac{(\hbar k_z^2)}{2m^*} \qquad \text{with } \mathbf{H} \,||\, \mathbf{z}$$

  $\Rightarrow$ an oscillation of $\mathbf{M(H)}$

  $\Rightarrow$ de Haas-van Alphen effect, Shubnikov-de Haas effect

Shubnikov-de Haas effect in CeBiPt



Itinerant electron magnetism

- Slater (1936) and Stoner (1938) the interaction between itinerant electrons

$$E = -\mu_0 \mathbf{MH} + \frac{1}{4\mu_B^2 N(E_F)}(1 - I N(E_F))M^2 + cM^4 + O[M^6]$$

$\Rightarrow$ finite value of $\mathbf{M}$, even for **H** = 0, if I $N(E_F) > 1$ (Stoner condition)

- I $N(E_F) < 1 \Rightarrow$ the paramagnetic state remains stable

$\Rightarrow$ exchange enhancement

$$\chi = \frac{2\mu_0 \mu_B^2 N(E_F)}{1 - I N(E_F)} \qquad \Rightarrow \quad \text{e.g. in Pd}$$

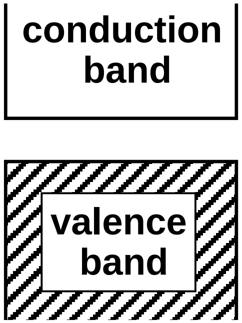
- *itinerant antiferromagnetism* and *spin density waves* (by $\chi_0(\mathbf{Q})$)

$\Rightarrow$ modified theory of spin fluctuations (T. Moriya 1978)

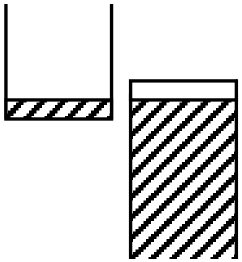
$\Rightarrow$ e.g. the afm in Cr

A dictionary

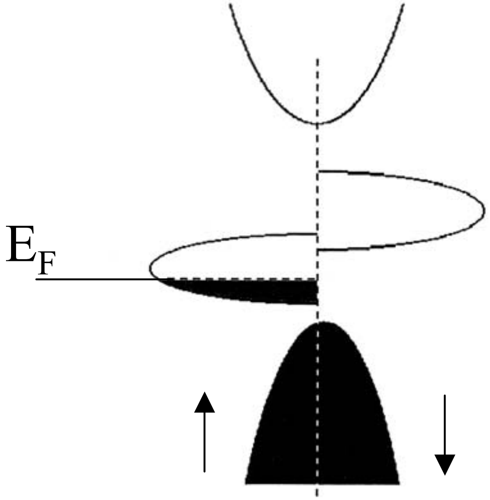
Semiconductor
Ge, Si, …



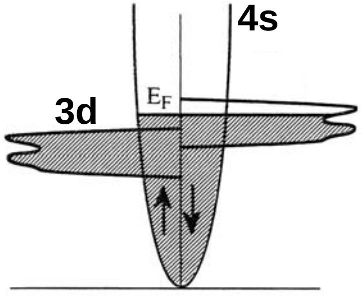
Semimetal
Bi, Sb, …



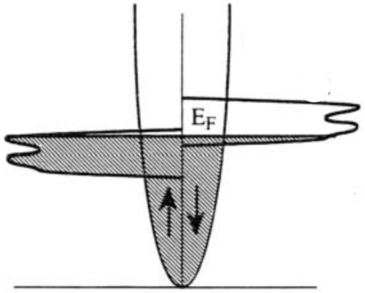
Half-metal
CrO_2, Mn_2VAl



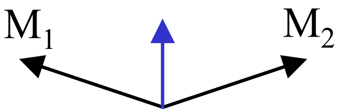
strong ferromagnet
Co, Ni, …



weak ferromagnet
Fe, …



weak ferromagnetism
α-Fe_2O_3, La_2CuO_4

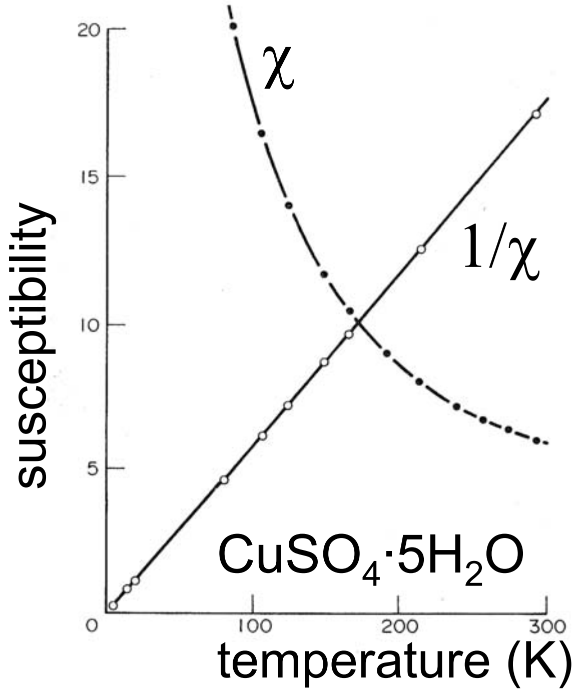


- localized electron spin canting in AFM
- by DM interaction

3 – Localized electron magnetism

- electrons of partially filled shells: localized by correlation
  $\Rightarrow$ carrying a magnetic moment

- mostly from 3d or 4f electrons in alloys and compounds $\Rightarrow$ $\Rightarrow$ $\Rightarrow$



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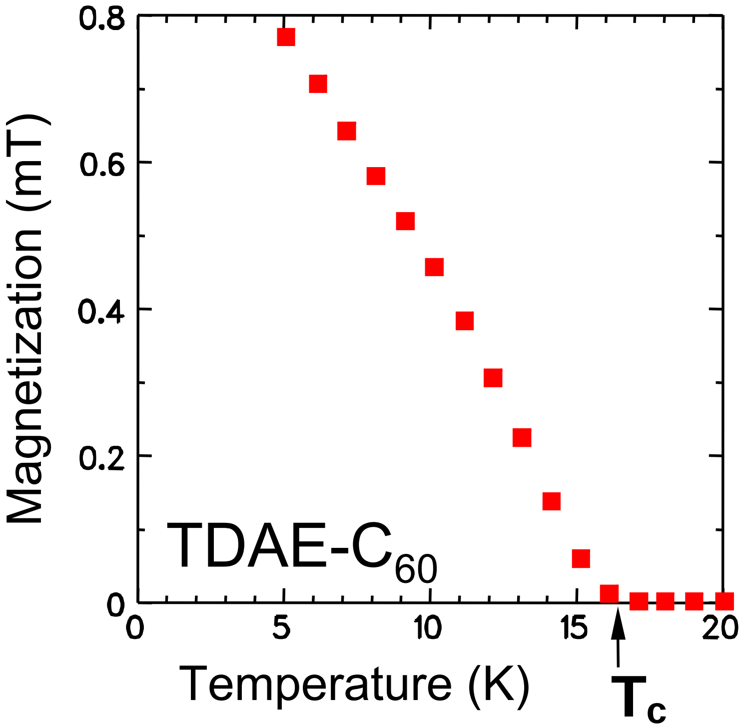
- electrons of partially filled shells:
  localized by correlation
  $\Rightarrow$ carrying a magnetic moment

- mostly from 3d or 4f electrons
  in alloys and compounds

- also 5f or 2p
  $\Rightarrow$ solid O_2 – an AFM: $T_N \approx 30K$
  $\Rightarrow$ TDAE-C_{60} – an organic FM $\Rightarrow$
  TDAE $\equiv C_2N_4(CH_3)_8$



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- electrons of partially filled shells:
  localized by correlation
  $\Rightarrow$ carrying a magnetic moment

- mostly from 3d or 4f electrons
  in alloys and compounds

- also 5f or 2p
  $\Rightarrow$ solid O_2 (T_s = 54K) – an antiferromagnet: T_N = (25…40)K
  $\Rightarrow$ TDAE-C_{60} – an organic ferromagnet
  TDAE $\equiv C_2N_4(CH_3)_8$

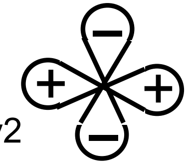
- interactions:
  $\Rightarrow$ total quenching of $\langle \mathbf{L} \rangle \Rightarrow \mathbf{M}_L = 0$
  $\Rightarrow$ cooperative phenomena of magnetic ordering

3-1 Effects of crystalline electric fields (CEF)

- atomic cores on the neighbour sites $\Rightarrow$ electrostatic potential

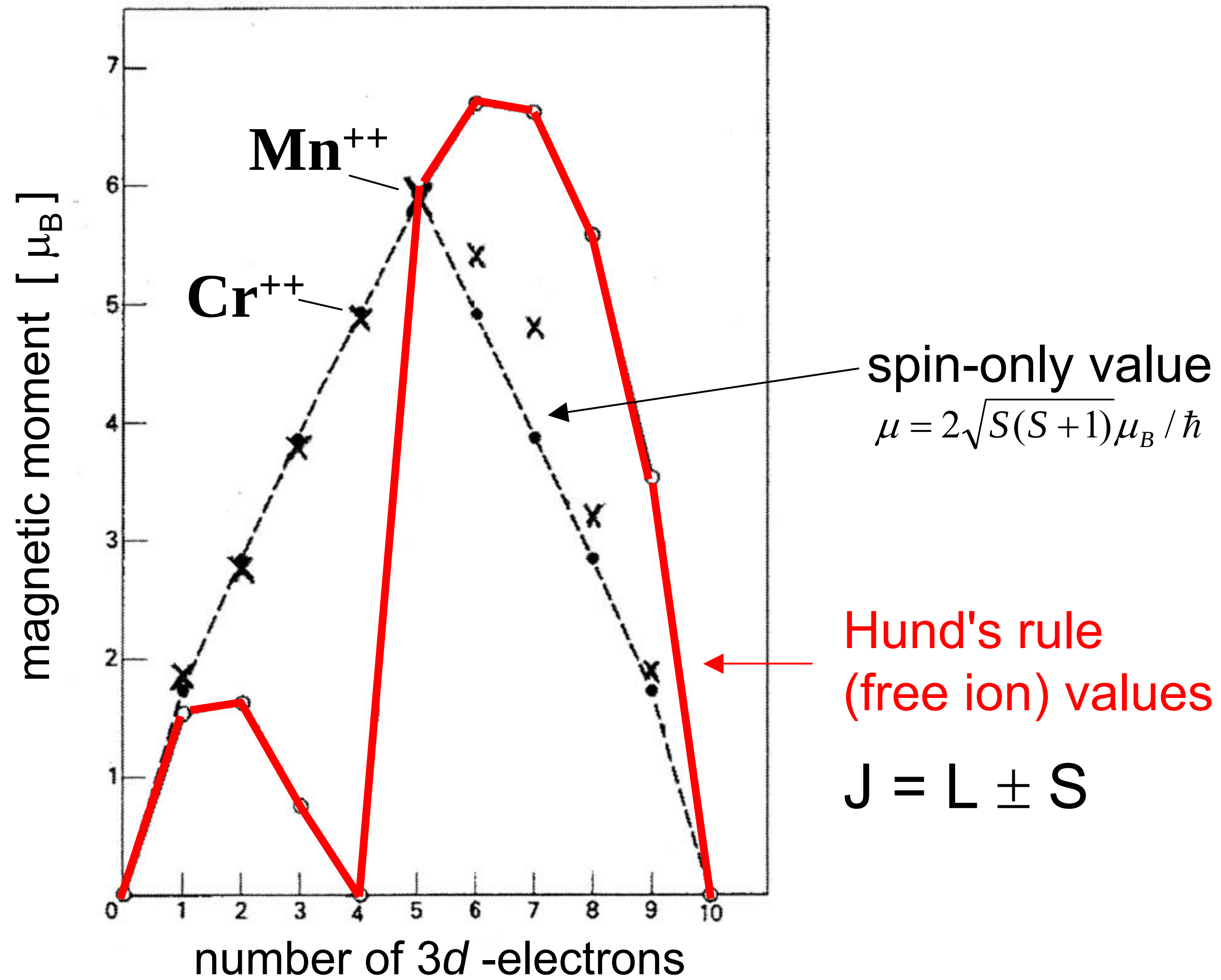
  $\Rightarrow$ no rotational symmetry

  $\Rightarrow$ L_z no good quantum number $\Rightarrow$ L_z-mixing : $L_z/\hbar = +2, -2$ $\Rightarrow$ d_{x2-y2}



- formalism of K.H.W. Stevens (1952)
- contribution **L** to **M** reduced or even totally "quenched"

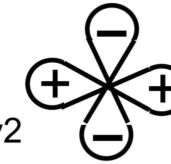
L-quenching in salts in two-valent *3d* ions



- the experimental values (x) are close to the spin only values
- e.g. Cr^{++} : $L/\hbar = 2 + 1 + 0 + (-1) = 2$, $S/\hbar = 4 \times 1/2 = 2$, $J = L - S = 0$

Effects of crystalline electric fields (CEF)

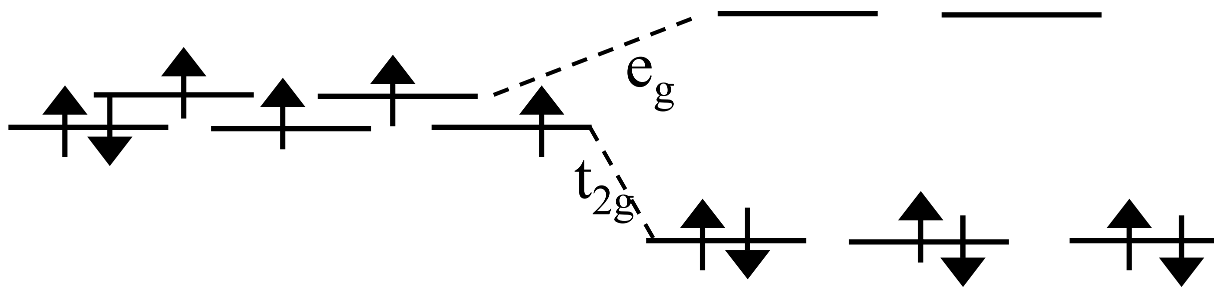
- atomic cores on the neighbour sites ⇒ electrostatic potential
  - ⇒ no rotational symmetry
  - ⇒ L_z no good quantum number ⇒ L_z-mixing : $L_z/\hbar$= +2, -2 ⇒ d_{x2-y2}



- formalism of K.H.W. Stevens (1952)
- contribution **L** to **M** reduced or even totally "quenched"

- if CEF-splitting >> Hund's-rule interaction
  ⇒ "High-spin–low-spin transition" or "spin quenching"

e.g. Fe^{++} in octahedral anionic environments

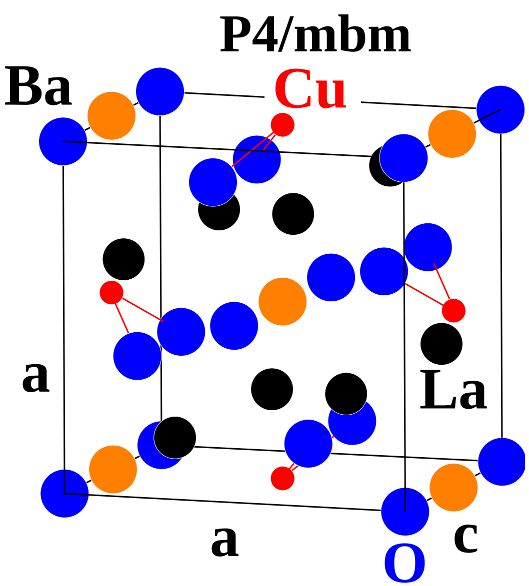


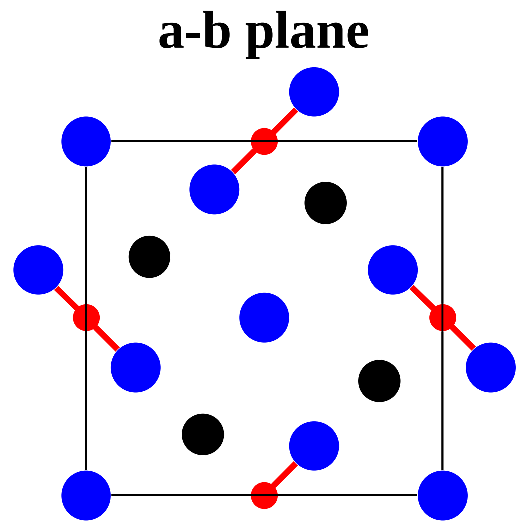
Δ(CEF) < Δ(HR)
⇒ S = 2ħ (high spin)
Hund's rule value

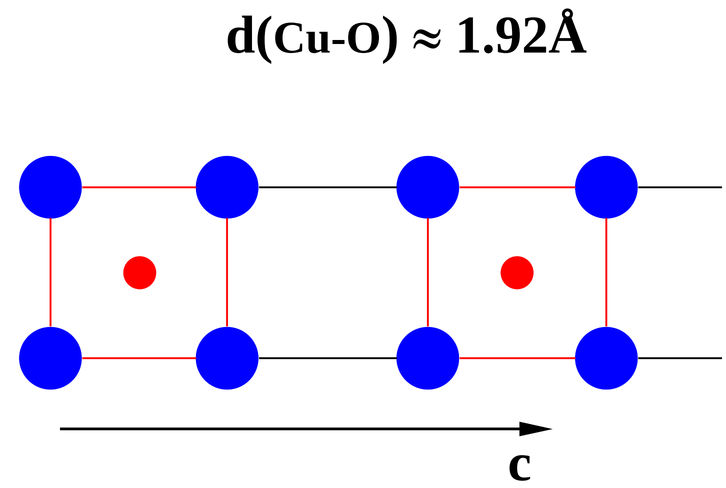
Δ(CEF) > Δ(HR)
⇒ S = 0 (low spin)

The ferromagnetic cuprate $La_2BaCu^{II}O_5$

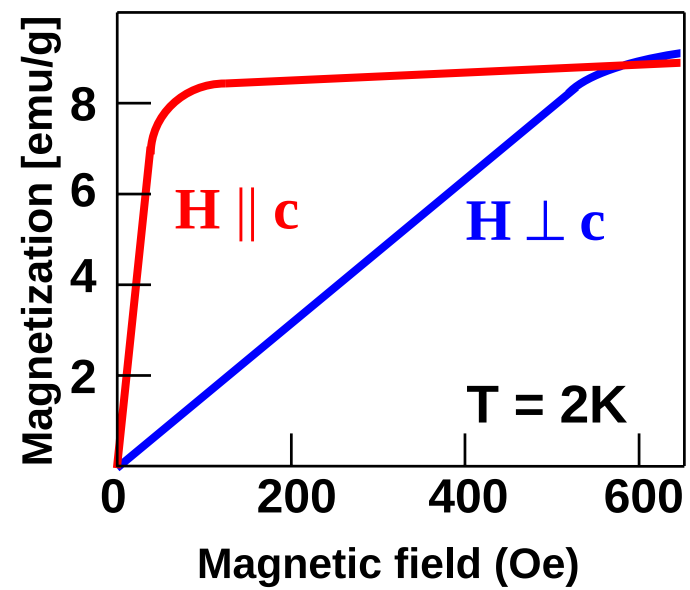
F. Mizuno et al., Nature 345 (1990) 788







- CuO_4 plaquettes as typical for Cu^{II} cuprates
- the plaquettes are nearly isolated $\Rightarrow$ d ≈ 0
- $\mu_p \approx 1.8\ \mu_B$; $\mu_s \approx 1\ \mu_B$ $\Rightarrow$ $S = \hbar/2$
- $T_c \approx 6$ K ;
- $\Rightarrow$ magnetic coupling between the Cu^{II} spins
- relatively strong magnetic anisotropy



3-2 Spin orbit interaction and CEF

$$\mathcal{H} = \mathcal{H}_0 - \frac{e}{2m}(\mathbf{L} + 2\mathbf{S})\mathbf{B} + \frac{e^2}{2m}\frac{B^2}{4}(x^2 + y^2) - \lambda(\mathbf{LS})$$

- spin orbit interaction is magnetic in its nature
- governs the third Hund's rule
- opposes L quenching $\Rightarrow$ e.g. in 4f-elements, alloys, compounds : $J = L \pm S$

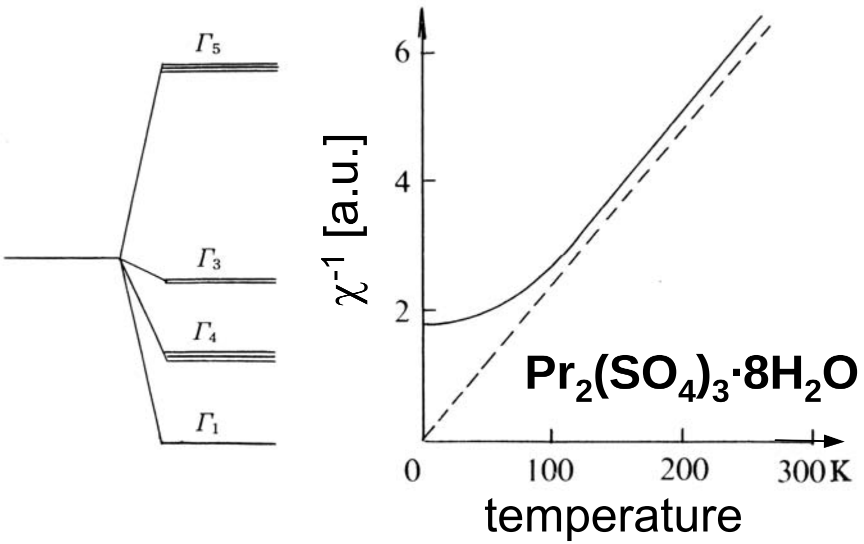
$\Rightarrow$ zero field splitting (simplest case): $\mathcal{H}_{CEF} = \tilde{D} J_z^2$, J/ħ integral $\Rightarrow$ $J_z = 0$

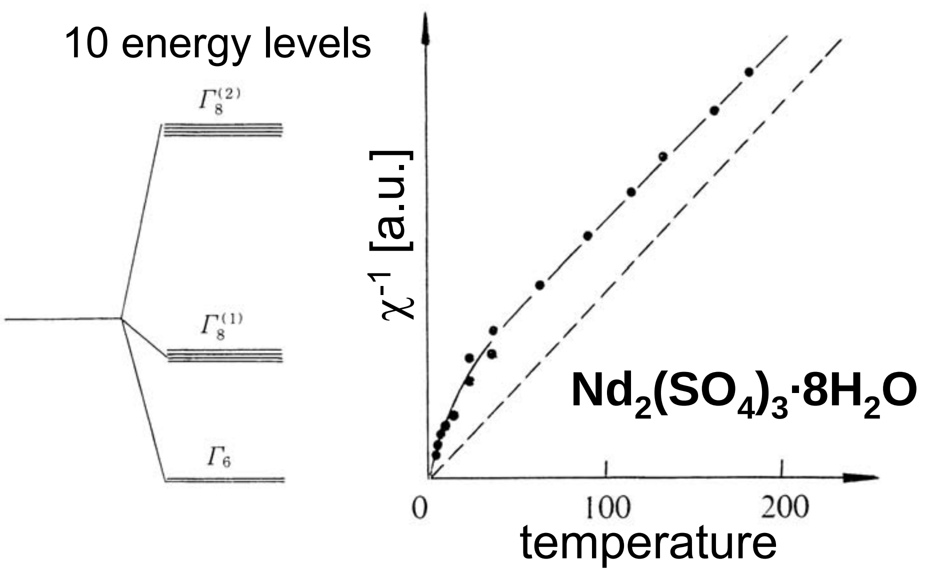
- H.A. Kramers (1930): odd number of electrons $\Rightarrow$ even degeneracy

$\Rightarrow$ "Kramers ions" (e.g. Cu^{++}, Nd^{3+})
$\Rightarrow$ "non-Kramers ions" (even numbers of electrons as in Fe^{++}, Pr^{3+})
$\Rightarrow$ singlets possible

- **H** mixes the ground state singlet of non-Kramers ions with excited CEF states
$\Rightarrow$ *Van Vleck paramagnetism*

Behaviour of Kramers and non-Kramers ions





- **non-Kramers ion Pr^{3+}** : 2 electrons
  $S = \hbar, L = 5\hbar, J = 4\hbar$

  $\chi(T \rightarrow 0)$ finite
  $\Rightarrow$ Van Vleck paramagnetism

- **Kramers ion Nd^{3+}** : 3 electrons
  $S = 3\hbar/2, L = 6\hbar, J = 9\hbar/2$

  $\chi(T \rightarrow 0) \rightarrow \infty$
  $\Rightarrow$ Curie-Langevin paramagnetism

Spin orbit interaction and CEF

$$\mathcal{H} = \mathcal{H}_0 - \frac{e}{2m}(\mathbf{L} + 2\mathbf{S})\mathbf{B} + \frac{e^2}{2m}\frac{B^2}{4}(x^2 + y^2) - \lambda(\mathbf{LS})$$

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$\Rightarrow$ singlets possible

- **H** mixes the ground state singlet of non-Kramers ions with excited CEF states
$\Rightarrow$ *Van Vleck paramagnetism*

- **S-L**-interaction mediates magnetic anisotropy from the lattice to the spin (or to **J**)

3-3 Dipolar interaction

- interaction energy of two dipoles $\mathbf{m}_i$, $\mathbf{m}_j$

$$E_{dip}(i,j) = \mu_0 \frac{(\mathbf{m}_i\mathbf{m}_j)r^2 - 3(\mathbf{m}_i\mathbf{r})(\mathbf{m}_j\mathbf{r})}{4\pi r^5}$$

$\Rightarrow$ is omnipresent in magnetic materials
$\Rightarrow$ results in magnetic ordering temperatures of typically 1 K
$\Rightarrow$ is long range
$\Rightarrow$ anisotropic

- *self-energy of a magnetization* $\mathbf{M(r)}$ in volume V

$$E_{self} = -\frac{\mu_0}{2}\int_V d\tau\, \mathbf{H'(r)M(r)} \quad \text{with} \quad \nabla\mathbf{H'}(\mathbf{r}) = -\nabla\mathbf{M}(\mathbf{r}) \quad , \quad \nabla \times \mathbf{H'}(\mathbf{r}) = 0$$

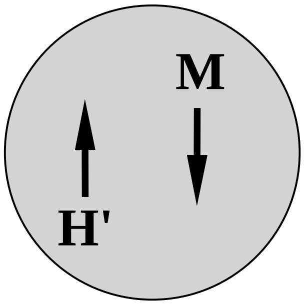
- is only semiconvergent
- in homogenously magnetized samples, **M(r) = M** = const

$$E_{self} = \frac{\mu_0}{2} V \sum_{i,j} D_{i,j} M_i M_j \qquad \sum_i D_i = 1 \qquad D_i \geq 0$$ demagnetization factors

$\Rightarrow$ "demagnetizing fields" $\quad \langle H'_i(\mathbf{r})\rangle_V = -D_i M \quad$ (bodies of arbitrary shape!)

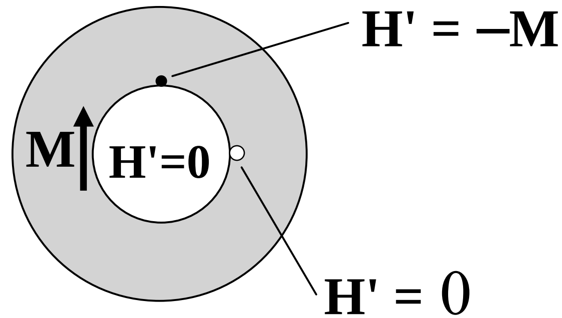
Examples of demagnetizing fields

sphere D =1/3



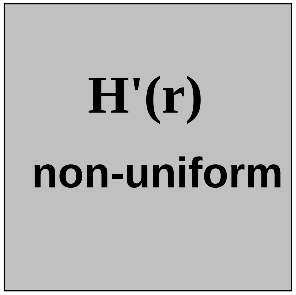
$H' = -M/3$

hollow sphere D =1/3



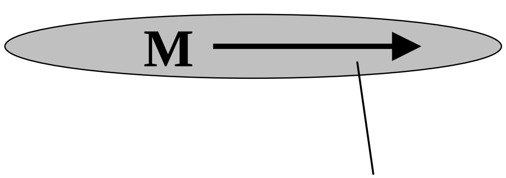
$\langle H' \rangle = -M/3$

cube D = 1/3



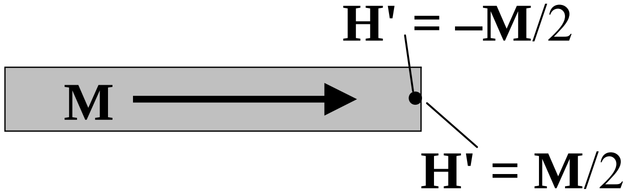
$\langle H' \rangle = -M/3$

prolate spheroid



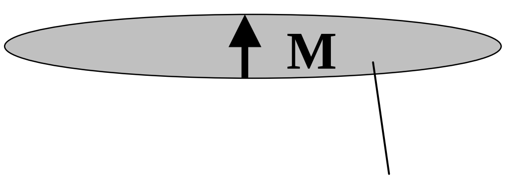
$D = 0 \Rightarrow H' = 0$

long cylinder



$D = 0 \Rightarrow \langle H' \rangle = 0$

prolate spheroid



$D = 1/2 \Rightarrow H' = -M/2$

Dipolar interaction

- interaction energy of two dipoles $\mathbf{m}_i$, $\mathbf{m}_j$

$$E_{dip}(i,j) = \mu_0 \frac{(\mathbf{m}_i\mathbf{m}_j)r^2 - 3(\mathbf{m}_i\mathbf{r})(\mathbf{m}_j\mathbf{r})}{4\pi r^5}$$

$\Rightarrow$ is omnipresent in magnetic materials
$\Rightarrow$ results in magnetic ordering temperatures of typically 1 K
$\Rightarrow$ is long range

- *self-energy of a magnetization* $\mathbf{M(r)}$ in volume V

$$E_{self} = -\frac{\mu_0}{2}\int_V d\tau\, \mathbf{H'(r)M(r)}$$ with $$\nabla\mathbf{H'}(\mathbf{r}) = -\nabla\mathbf{M}(\mathbf{r}) \quad , \quad \nabla \times \mathbf{H'}(\mathbf{r}) = 0$$

- is only semiconvergent
- in homogenously magnetized samples, **M(r) = M** = const

$$E_{self} = \frac{\mu_0}{2} V \sum_{i,j} D_{i,j} M_i M_j \qquad \sum_i D_i = 1 \qquad D_i \geq 0$$ demagnetization factors

$\Rightarrow$ "demagnetizing fields" $\langle H'_i(\mathbf{r})\rangle_V = -D_i M$ (bodies of arbitrary shape!)

$\Rightarrow$ tensor character of $(D_{i,j})$ $\Rightarrow$ shape anisotropy

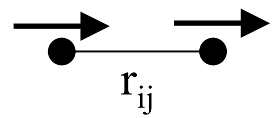
Effects of dipole-dipole interaction

$$E_{dip}(i,j) = \mu_0 \frac{(\mathbf{m}_i \mathbf{m}_j) r_{ij}^2 - 3(\mathbf{m}_i \mathbf{r}_{ij})(\mathbf{m}_j \mathbf{r}_{ij})}{4\pi r_{ij}^5}$$

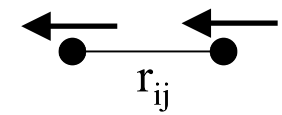
- is magnetic in its nature and anisotropic

$\Rightarrow$ its effect is sensitive to to the presence of other interactions

1.) two dipoles governed by E_{dip} only $\Rightarrow$

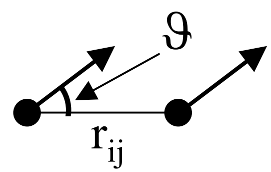


or



2.) if additionally $\mathbf{m}_1 \parallel \mathbf{m}_2$ required (strong exchange interaction)

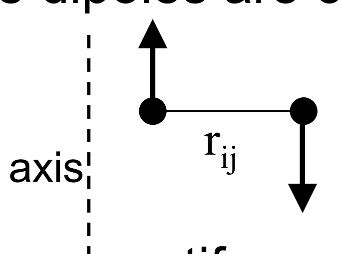
$$E_{dip} = \frac{\mu_0 m^2}{4\pi r_{ij}^3}(1 - 3\cos^2\vartheta)$$



typical easy-axis magnetic anisotropy

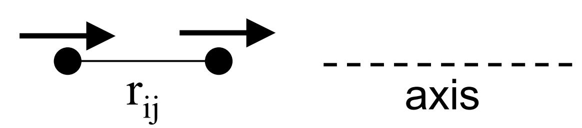
3.) if the dipoles are confined to a certain axis (by strong CEF)

a)



antiferromagnetic alignment

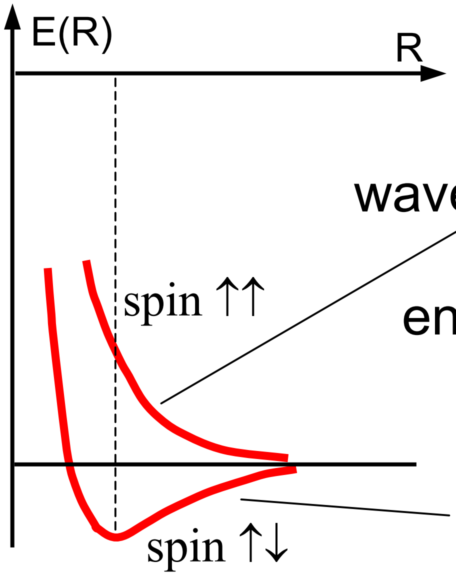
b)



ferromagnetic alignment

3-4 Exchange interaction

- example:
  H_2-molecule



wave function antisymmetric in $\mathbf{r}_1$ and $\mathbf{r}_2$

energy difference at R_0 : $E_{ant} - E_{symm} \equiv -J$

symmetric in $\mathbf{r}_1$ and $\mathbf{r}_2$

- the Pauli principle requires antisymetric total wave functions
- spin space

$$\Psi(s=0) = \{(\chi_1(\uparrow)\,\chi_2(\downarrow) - \chi_1(\downarrow)\,\chi_2(\uparrow)\}/\sqrt{2}$$

$\mathbf{s} = \mathbf{s}_1 + \mathbf{s}_2$

$$\Psi(s=1, s_z=1) = (\chi_1(\uparrow)\,\chi_2(\uparrow)$$

$\mathbf{s}_i = 1/2$

$$\Psi(s=1, s_z=0) = \{(\chi_1(\uparrow)\,\chi_2(\downarrow) + \chi_1(\downarrow)\,\chi_2(\uparrow)\}/\sqrt{2}$$

(with $S = \hbar s$ etc.)

$$\Psi(s=1, s_z=-1) = (\chi_1(\downarrow)\,\chi_2(\downarrow)$$

$\Rightarrow$ $\mathcal{H}_{ex}(i,j) = -J\,\mathbf{s}_i\,\mathbf{s}_j$

$\Rightarrow$ description in spin space although purely electrostatic in its nature

$\Rightarrow$ isotropic

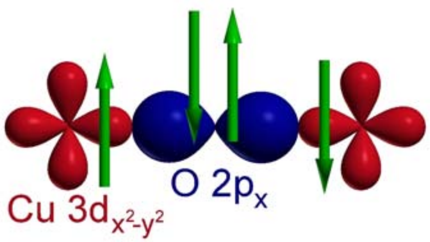
Exchange interaction

$$\mathcal{H}_{ex}(i,j) = -J\,\mathbf{s}_i\,\mathbf{s}_j$$

⇒ high ordering temperatures (molecular field of *P. Weiss*: $\langle JS_i \rangle$)
⇒ isotropic

- *direct exchange* (W. Heisenberg 1928):
  overlap of electron wave functions of neighbours ⇒ $J \gtrless 0$

- *superexchange:*
  in ionic compounds (e.g. Cu^{++} ⇒ $k_B J \approx -2000$ K)
  mediated by anions (e.g. O^{--}) ⇒ mostly $J < 0$



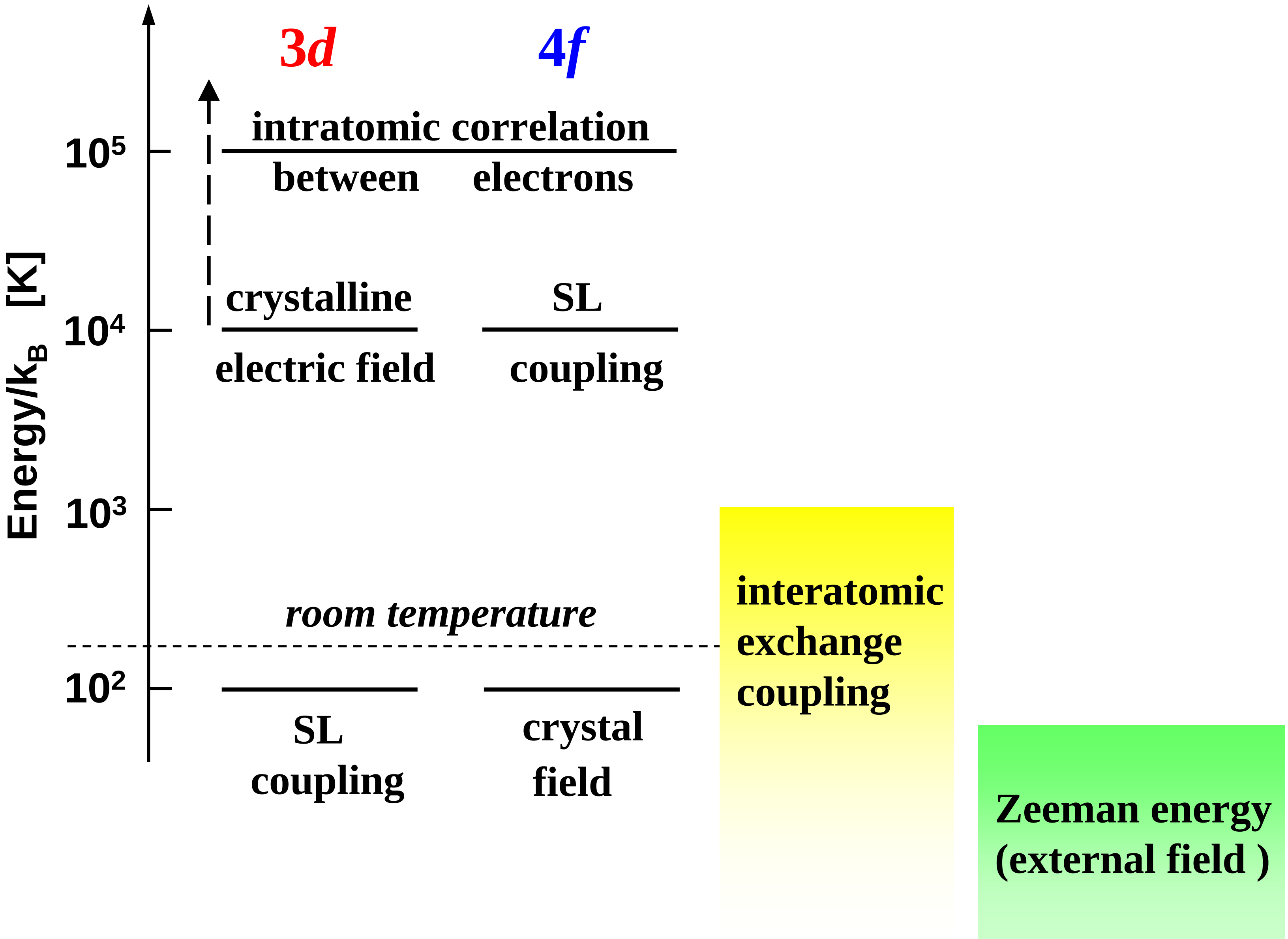
- *RKKY interaction:*
  of localized electrons mediated by itinerant electrons
  ⇒ long range and J oscillating in magnitude and sign, e.g. in 4f-elements

- *double exchange:* in mixed valence materials e.g. $(La,Sr)MnO_3$
  mobile Mn-3d electrons mediate the exchange between neighbouring
  Mn magnetic ions ⇒ ferromagnetic metals

- *exchange induced–moment magnetism*
  in materials with singlet CEF ground states of non-Kramers ions
  e.g. the ferromanget PrPtAl : $T_c \approx 6$ K Δ(CEF) = 21 K
  the antiferromagnet $PrNi_2B_2C$: $T_N = 4$ K

Energy scales for magnetic phenomena

dominated by localized *3d* or *4f* electrons



4-Anisotropy and dimensionality

4-1 Types of magnetic anisotropy

• most common : *single ion anisotropy* $\mathcal{H}_{sia} = \tilde{D} S_z^2$ caused by CEF

⇒ continuum description (micromagnetism)

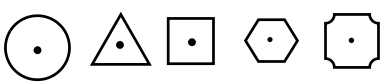
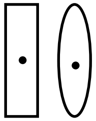
$$F = \int_V d\tau \, [A \frac{(\nabla \mathbf{M})^2}{M_s^2} - K \frac{(\mathbf{nM})^2}{M_s^2} - \frac{\mu_0}{2} \mathbf{MH'} - \mu_0 \mathbf{MH}]$$

simplest form of magnetic anisotropy in uniaxial *materials*

• *shape anisotropy* is due to the anisotropy of the dipolar interaction

⇒ represented by the demagnetization tensor (D_{ij}):

$$E_{self} = \frac{\mu_0}{2} V \sum_{\mathbf{i,j}} M_i D_{i,j} M_j$$

⇒ uniaxial bodies  excluded:  and spheres, cubes etc.

$$(D_{i,j}) = \begin{pmatrix} D_\perp & & 0 \\ & D_\perp & \\ 0 & & 1 - 2D_\perp \end{pmatrix}$$

$$\Rightarrow \quad E_{self} = \frac{\mu_0 V M^2}{2} [D_\perp - (3D_\perp - 1) \cos^2 \vartheta] = \text{const} - K_{sh} \cos^2 \vartheta$$

⇒ $K = K_{CEF} + K_{sh}$ with $K_{sh} = (3D_\perp - 1)\, \mu_0 V M^2/2$

e.g. **ALNICO** : needles of Fe-Co (1 μm x 1 nm; $\mu_0 M_s \approx 2.4$ T ⇒ $D_\perp = 0.5$)
embedded in low-M_s Ni-Al

further types of magnetic anisotropy

- *anisotropic exchange:*

combination of exchange interaction with CEF and **S**-**L**-interaction

$$\mathcal{H}_{ae} = \mathbf{S}_i \hat{D}_{ij} \mathbf{S}_j \qquad \text{symmetric tensor } \hat{D}_{ij}$$

⇒ "pseudodipolar interaction"

- *antisymmetric or Dzyaloshinsky-Moriya* exchange

$$\mathcal{H}_{DM} = \mathbf{d}\,(\mathbf{S}_i \times \mathbf{S}_j)$$

⇒ tends to orient spins $\mathbf{S}_i$ and $\mathbf{S}_j$ perpendicular to each other and to **d**
⇒ causes canting of the sublattice magnetizations
⇒ weak ferromagnetism for sufficiently low symmetry (e.g. α-Fe_2O_3)

- magnetic moments with **S** = ħ/2 (e.g. Cu^{++}) cannot experience single ion anisotropy because $S_x^2 = S_y^2 = S_z^2 = \hbar^2/4$ are constants

⇒ *anisotropy of the g-factor* in

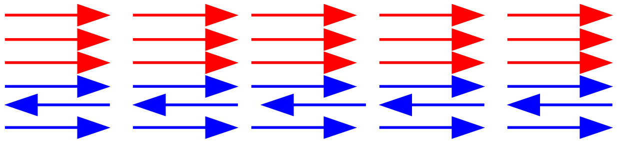
$$\mathcal{H}_z = g\,\mu_0 \mu_B \hbar^{-1} \mathbf{H}\,\mathbf{S}$$

e.g. $CuSO_4 \cdot 6H_2O$: $g_z = 2.20$, $g_y = g_z = 2.08$

further types of magnetic anisotropy

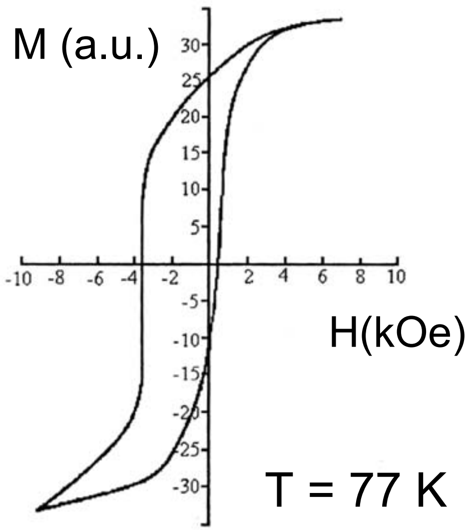
- *unidirectional magnetic anisotropy:*

  e.g. W.H. Meiklejohn and C-P. Bean 1957: exchange anisotropy

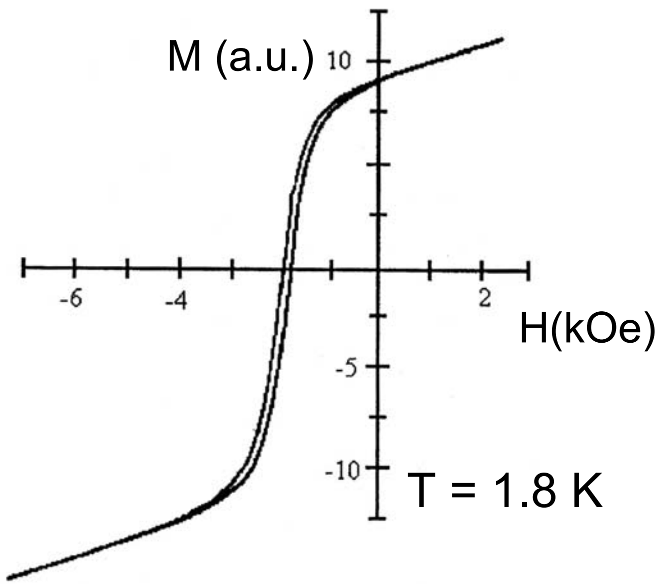


ferromagnet

antiferromagnet



CoO coated Co particles



$Ag_{80}Mn_{20}$ spin glass

- similar effects in "spring magnets" (e.g. $Nd_4Fe_{77}B_{19}$)

both curves obtained after field-cooling

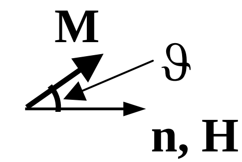
$\Rightarrow$ exchange bias in information storage technology

- *diamagnetism* can also be strongly anisotropic (e.g. graphite : $\chi_{||}/\chi_{\perp} \approx 53$)

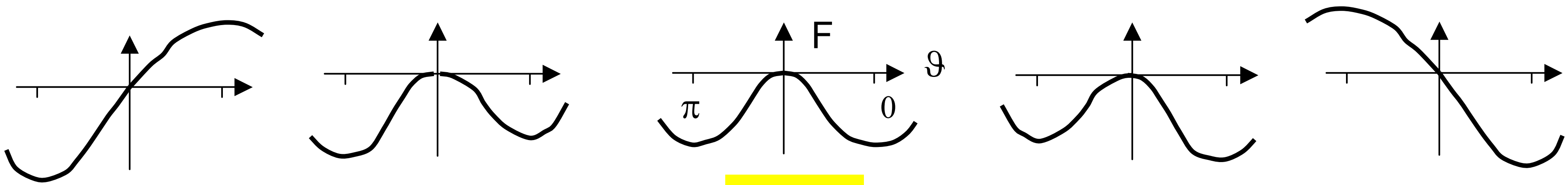
4-2 Magnetic anisotropy and coercivity

- minimization of the free energy

$$F = \int_V d\tau \,[\, A\frac{(\nabla \mathbf{M})^2}{M_s^2} - K\frac{(\mathbf{nM})^2}{M_s^2} - \frac{\mu_0}{2}\mathbf{MH'} - \mu_0 \mathbf{MH} \,]$$



simplest form of magnetic anisotropy in uniaxial *materials*



$H < -H_A$ | $-H_A < H < 0$ | ← $H = 0$ → | $0 \leq H < H_A$ | $H_A < H$

⇒ at H = 0 a first order phase transition : **M** changes from $\mathbf{n} M_s$ to $-\mathbf{n} M_s$

- **W.F. Brown** (1963): not at H = 0 but at $H = -{}_JH_c$ with

$${}_JH_c \geq H_A = 2K/(\mu_0 M)$$

⇒ for $-{}_JH_c < H < 0$ the magnetized state is metastable

- observed ${}_JH_c << H_A$ ⇒ *"Brown's paradox"* ⇒ ⇒

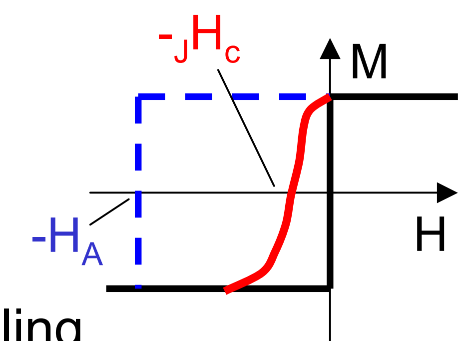
⇒ explained by imperfections in the material

⇒ supported by thermal fluctuations and quantum tunneling

⇒ soft magnetic materials : ${}_JH_c << M_s$

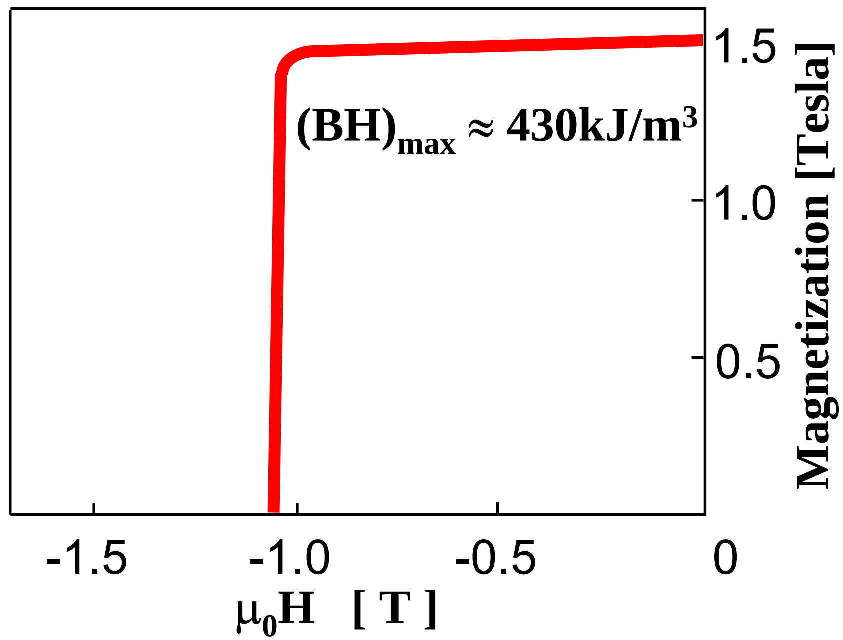
⇒ hard magnetic (or permanent magnet) materials : ${}_JH_c \geq M_s$

⇒ need of well defined microstructure ⇒ materials sciences

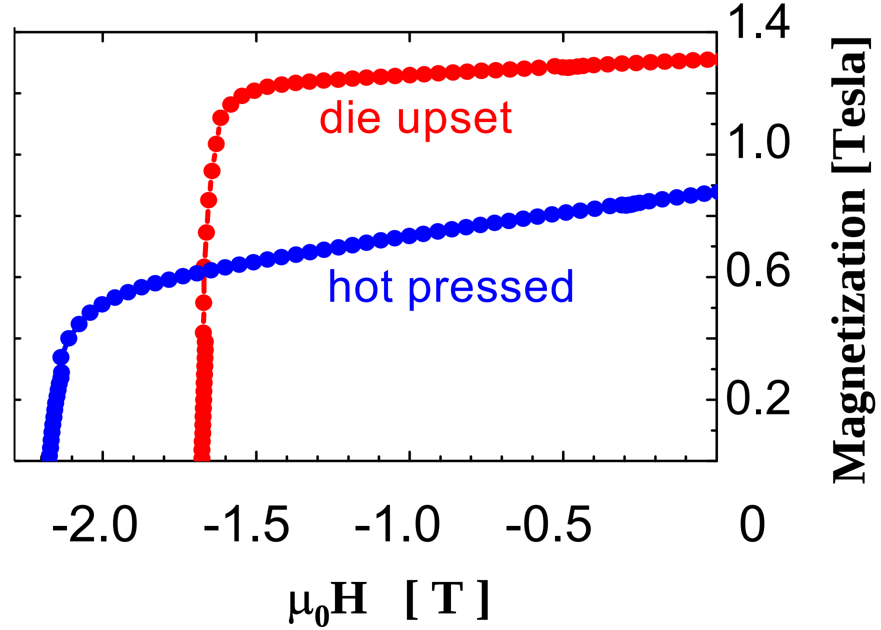


High-quality Nd-Fe-B permanent magnets

sintered



melt spun

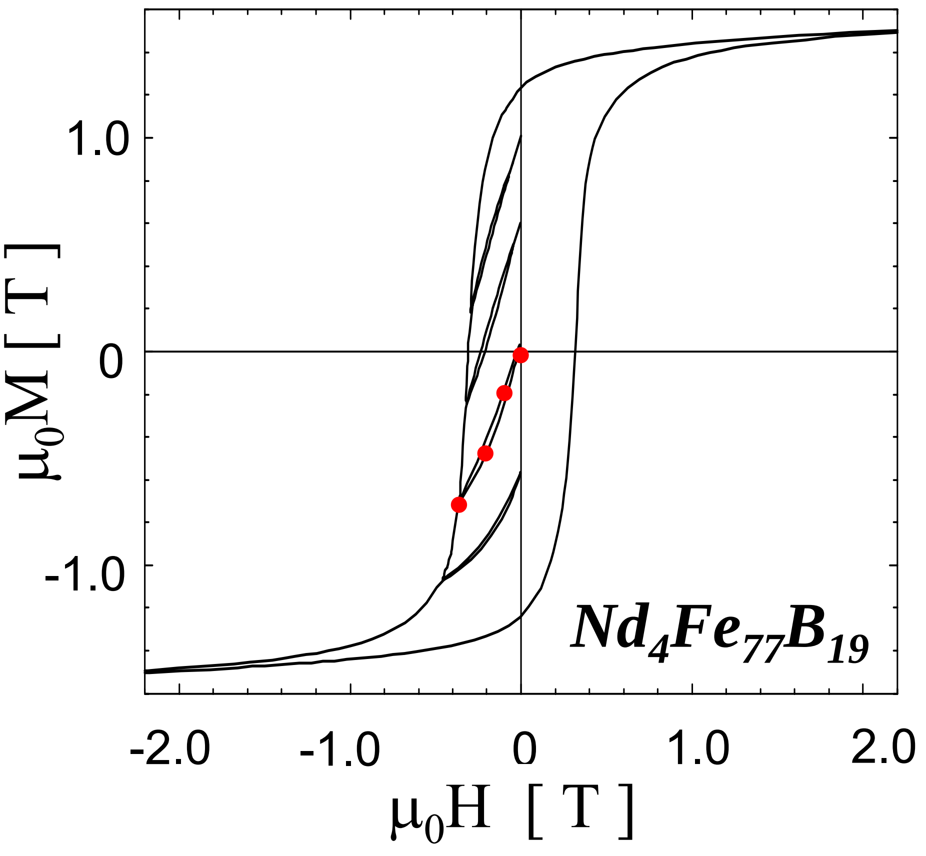


- $\mu_{0J}H_c \approx 1.1$ T ($\mu_0 H_A \approx 9$ T)
- $B_r \approx 1.5$ T $\mu_0 M_s \approx 1.6$ T
- coercivity is controlled by nucleation of reverse domains

Magnetic after effect (or viscosity)

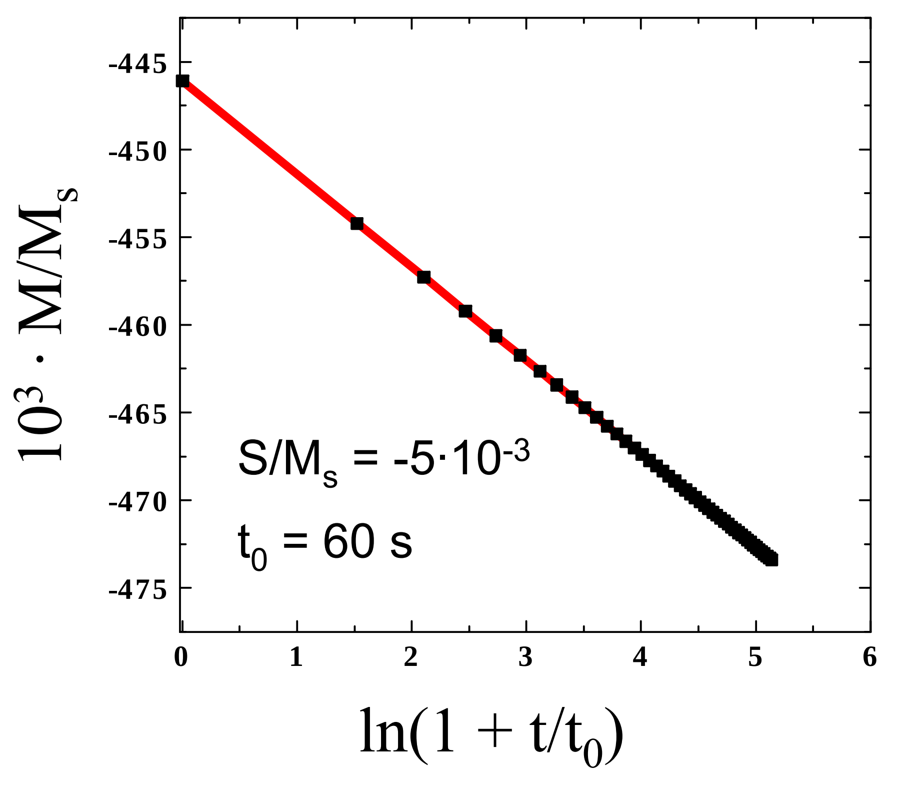
– a further consequence of metastability –

Hysteresis loop



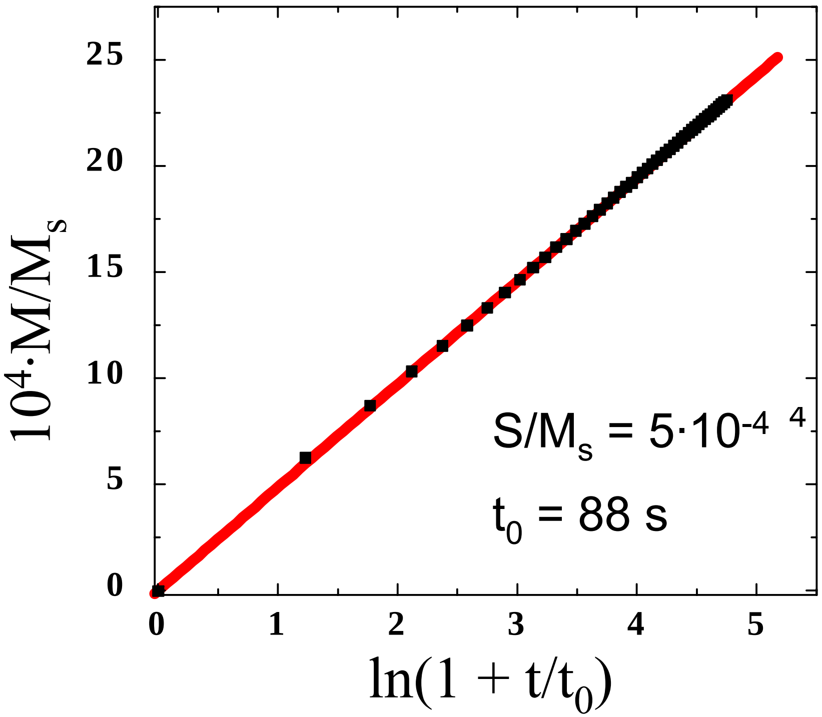
Viscosity experiment

$$M(t) = M(0) + S \ln(1 + t/t_0)$$

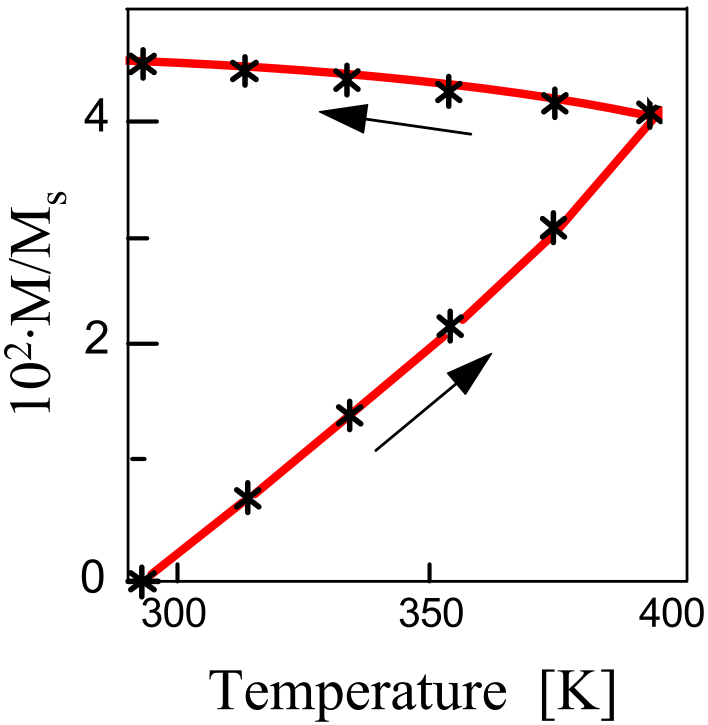


Remagnetization effects

– a further consequence of metastability –



• viscosity of opposite sign

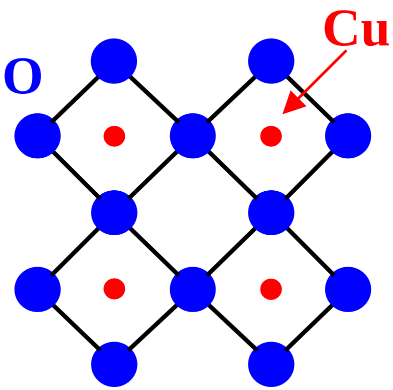


• thermal remagnetization

4-3 Magnetism in low dimensional systems

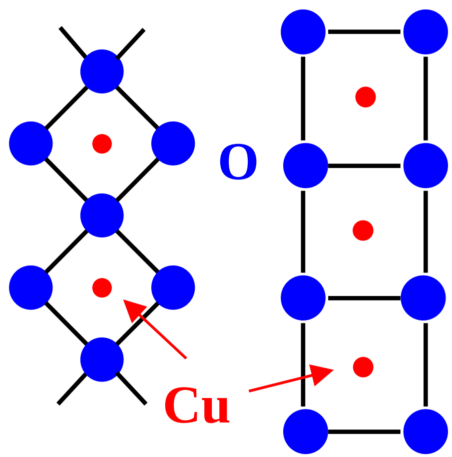
- examples: cuprates known from high-Tc superconductors with Cu^{++} ($S = \hbar/2$) magnetic moments

plane: d = 2



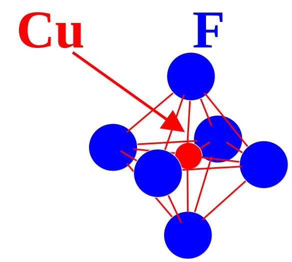
$LaCuO_4$

chains: d = 1



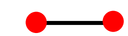
Sr_2CuO_3 Li_2CuO_2

isolated clusters: d = 0



Cs_3CuF_6

dimer: d = 0



- if the CEF and the **S** - **L**-interaction of such systems are neglected they will be described by

$$\mathcal{H}_{ex}(i,j) = -J\,\mathbf{s}_i\,\mathbf{s}_j$$

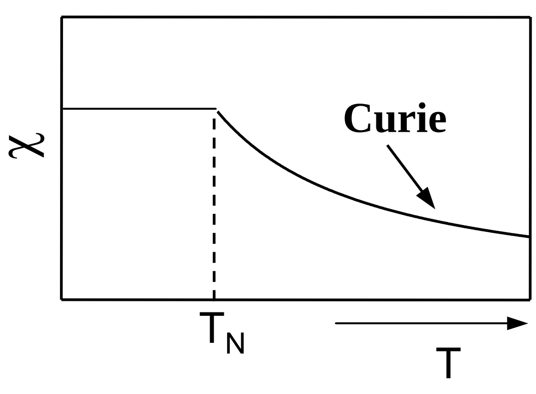
⇒ they are magnetically isotropic

⇒ nevertheless their behaviour is very different because thermal as well as quantum fluctuations make cooperative phenomena very sensitive to d

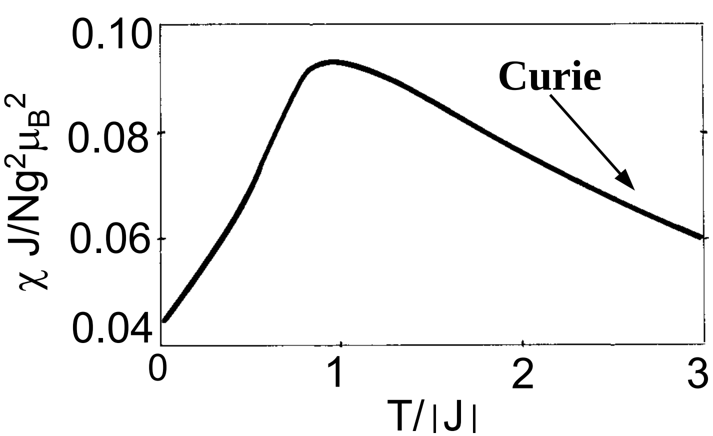
Spin 1/2 Heisenberg antiferromagnet on simple cubic lattices

$$H = -J \sum \vec{s}_i \vec{s}_j \qquad J < 0$$

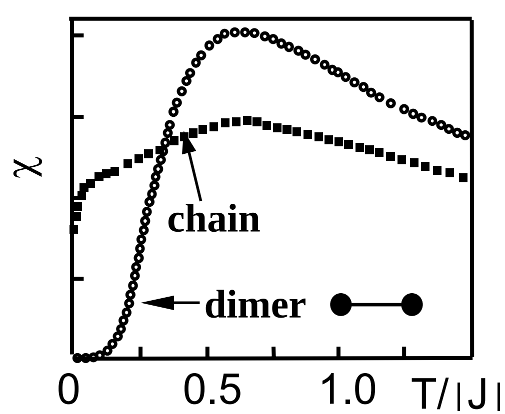
d	**ground state**	μ_{stagg}/μ_B	**T_N/| J |**	**T_{max}/| J|** in $\chi(T)$	**$\chi(T\to 0)$** $t \equiv T/\|J\|$
3	Néel AFM	0.85	0.93	–	≈ const.
2	Néel AFM	0.61	0	0.94	$0.04\cdot(1 + t)$
1	spin liquid	0	–	0.64	$\sim (1 + 0.5\cdot\ln(7.7/t))$
0 dimer	S = 0 singlet	0	–	0.62	$\sim e^{-1/t}/t$ (spin gap)



d = 3



d = 2



d = 1 and d = 0

Dimensionality crossover

- planes coupled by $J_\perp$ with $J_\perp < 0$, $| J_\perp | << | J |$ ($d = 2$) (C.M. Soukoulis et. al. 1991)

$$\Rightarrow \quad k_B T_N = \frac{4\pi J}{3\ln(32J/J_\perp)}$$

$\Rightarrow$ nevertheless at high temperatures it behaves as a $d = 2$ system

$\Rightarrow$ a maximum of χ at $k_B T_{max} \approx 0.94$ J.

example : **La_2CuO_4**: $k_B J \approx 1500$ K , $J_\perp/J \approx 10^{-5}$, resulting $T_N \approx 300$ K
$T_{max} \approx 1400$ K

5 - Phase transitions and magnetization processes

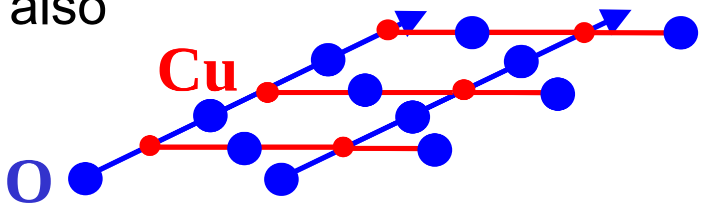
5-1 Types of magnetic order

- *ferromagnetic* ground state: in all systems with $J > 0$
  $\Rightarrow$ also in $d = 0$ clusters and other low-d- systems

- for $J < 0$ or mixed values $J \gtrless 0$ $\Rightarrow$ much more complicated

  - remember the dimer with $\mathcal{H} = -J\mathbf{s}_1\mathbf{s}_2$, $J < 0$, $s_1 = s_2 = 1/2$
  $\Rightarrow$ no antiferromagnet but a singlet state $\Psi_{\text{dim}} = (\psi_{1,+}\psi_{2,-} - \psi_{1,-}\psi_{2,+})/\sqrt{2}$
  $\Rightarrow$ no magnetic moments at the sites 1 and 2

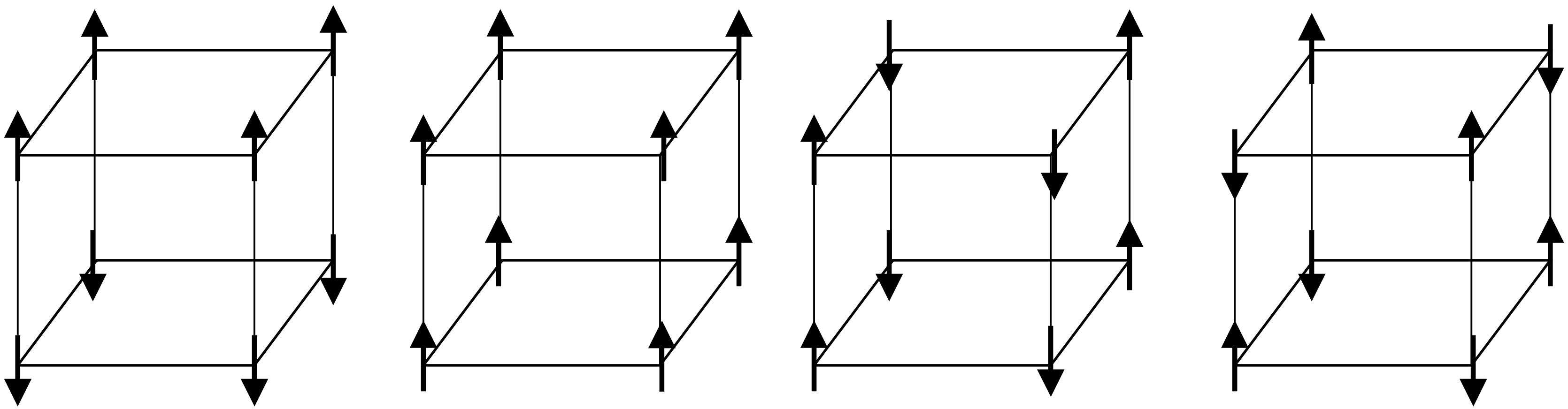
- non-magnetic ground states with a spin gap also in spin-ladder compounds e.g. in $SrCu_2O_3$



- antiferromagnetic order can only exist in infinitely large systems of sufficiently large dimensionality, $d \geq 2$ $\Rightarrow$ rich variety of different afm structures

- advanced technologies are needed
  neutron scattering, x-ray magnetic scattering, Mössbauer spectroscopy, nuclear magnetic resonance (NMR) and muon spin relaxation (μSR)

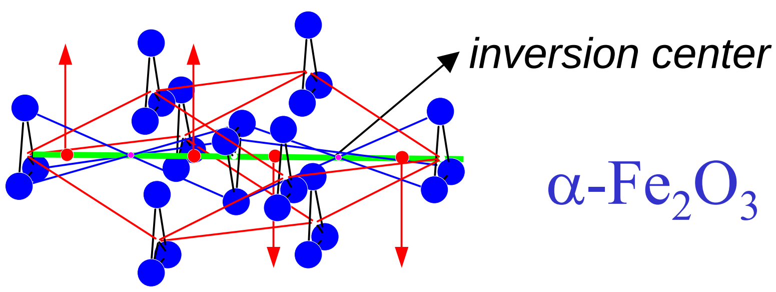
The mixed-valence perovskite $La_{1-x}Ca_xMnO_3$

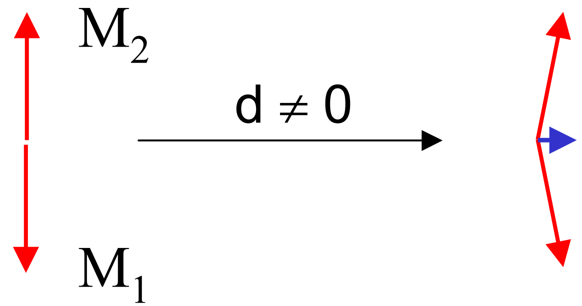


A	B	C	G
alternating fm planes	fm	alternating fm chains	"simple" afm
x = 0	x = 0.3	x = 0.5	x = 1

Types of magnetic ordering

- *weak ferromagnetism* for sufficiently low lattice symmetry the DM-interaction
  $\Rightarrow$ small net magnetization (as e.g. in α-Fe_2O_3, La_2CuO_4)





$\Rightarrow$ the center of inversion is not between the sites the afm sublattices $\Rightarrow$ WFM

- DM-interaction can also give rise to helical magnetization structures (e.g. MnSi)

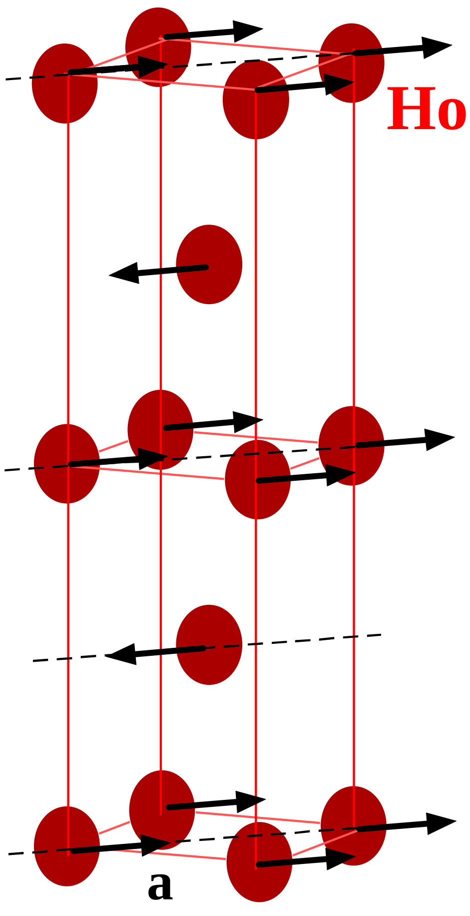
- *ferrimagnets:* a net magnetization arises because the magnetic moments are antiparallel and different in size  e.g. Fe_3O_4, $GdCo_5$

- *spin density waves and spiral structures* due to exchange between the local magnetic moments mediated by itinerant electrons (RKKY)

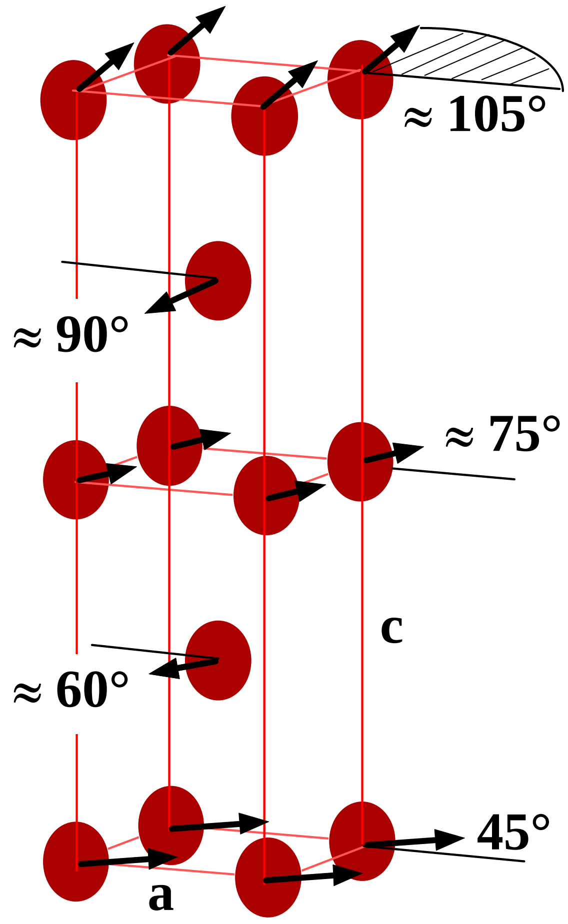
Magnetic structures in *$HoNi_2B_2C$*

determined by elastic neutron scattering

antiferromagnetic commensurate :



incommensurate c-axis modulated : (spiral structure) q = 0.916c*



Additionally there is an incommensurate a-axis modulated with q = 0.585a*

Magnetic structures

- *frustration* : if interactions (e.g. with next and overnext neighbours) are in competition

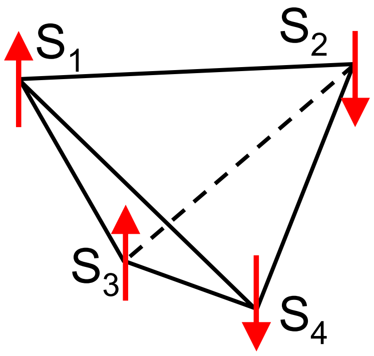
  ⇒ the system can form more than two magnetic sublattices

- in particular *geometrical frustration* (in e.g. the d = 3 pyrochlore lattice or the cubic Laves phase structure or the d = 2 Kagomé lattice

  ⇒ no long range order

  frustration in a tetrahedron (as in the pyrochlore structure) with afm coupled spins i (i =1 … 4):

  ⇒ the afm alignment cannot be satisfied, at the same time, for all spin pairs



- *spin glass states*: caused by frustration in the presence of disordered solid structures (as e.g. amorphous solids)

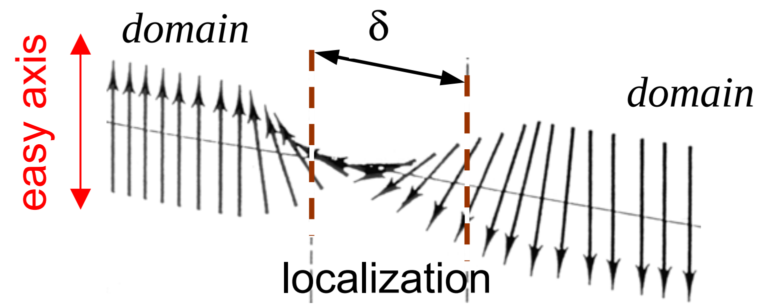
  ⇒ thermodynamic or kinetic "glass" state **?**

  ⇒ no equilibrium net magnetization

  ⇒ nevertheless strong hysteresis phenomena

5-2 Magnetic domains

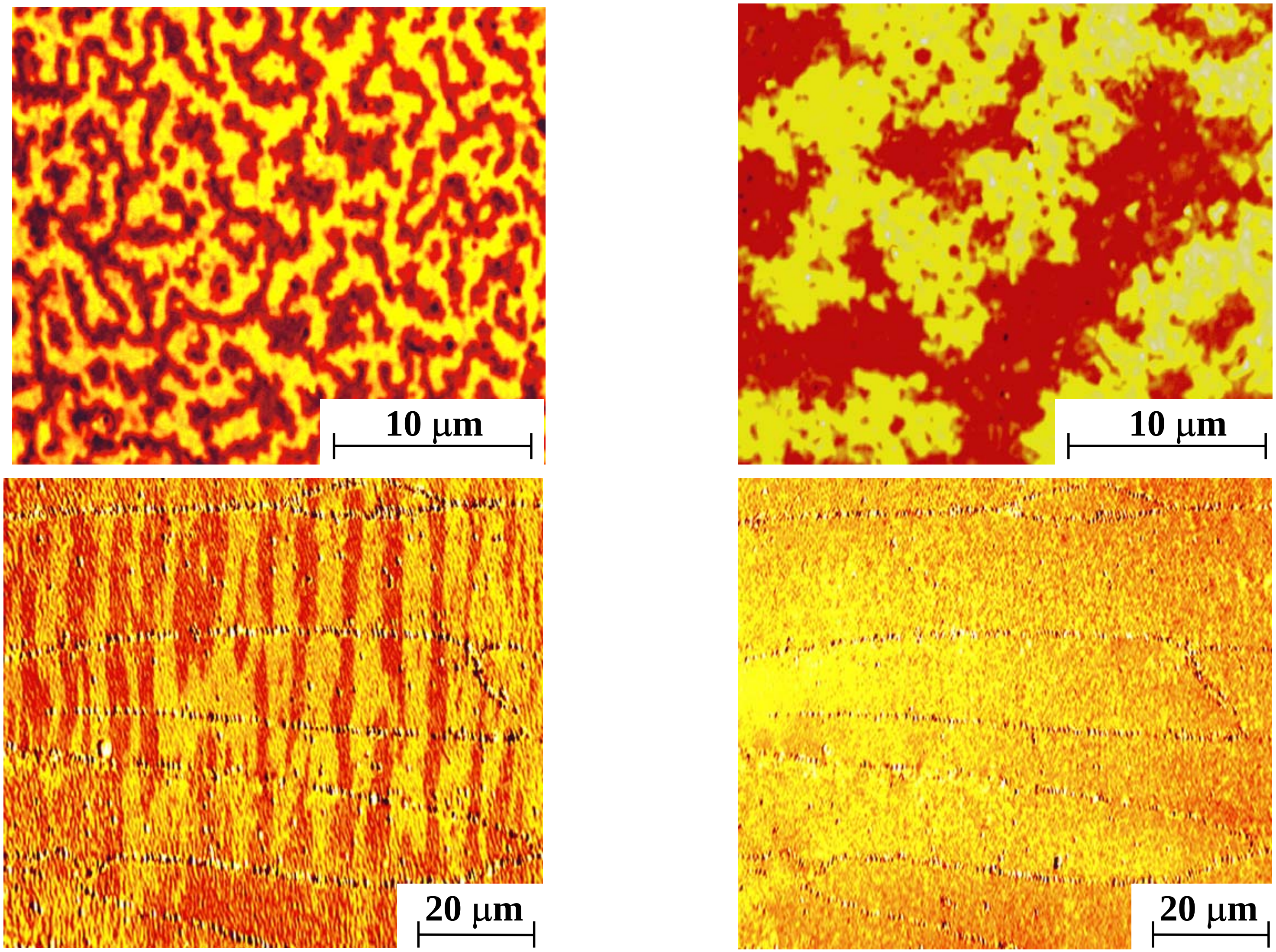
$$F = \int_V d\tau \left[A \frac{(\nabla \mathbf{M})^2}{M_s^2} - K \frac{(\mathbf{nM})^2}{M_s^2} - \frac{\mu_0}{2} \mathbf{MH'} \right]$$



- fixed boundary magnetic moment direction
  ⇒ due to the non-linearity of the problem a *domain wall* will be formed
  ⇒ *wall width* $\delta \sim \sqrt{A/K}$ ($Nd_2Fe_{14}B$: 3 nm)
  ⇒ *wall energy* per unit area $\gamma \sim \sqrt{AK}$
- magnetostatic selfenergy: $E_{dip} \sim VM^2$ reduced by spontaneous formation of domains
- *critical single-domain size* $D_c \sim \sqrt{AK} / \mu_0 M_s^2$ ($Nd_2Fe_{14}B$: 300 nm)
- Y. Imry and S.K. Ma (1975):
  in random anisotropy systems domain like structures
  ⇒ competition of exchange and random anisotropy
  ⇒ even without magnetostatic selfenergy
- *interaction domains* in fine-grained polycrystalline permanent magnets

Interaction domains in fine-grained materials

Hot-deformed melt-spun Nd-Fe-B : grain size ≈ 200 …300 nm ≤ D_c ≈ 400 nm



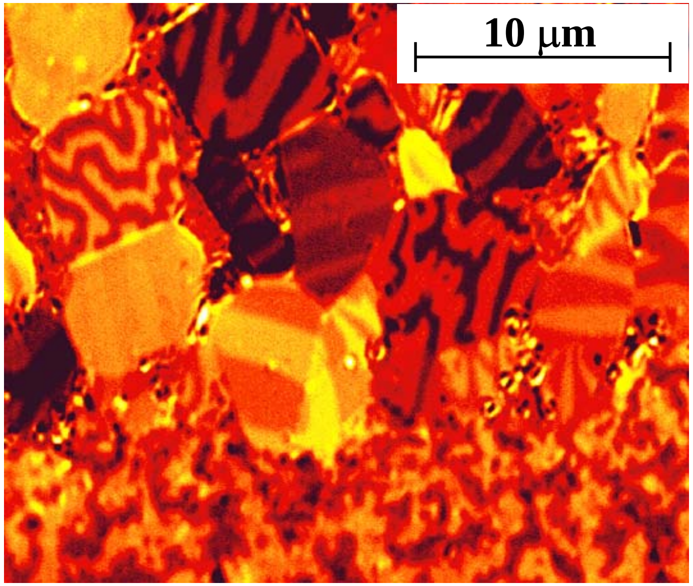
thermally demagnetized ***dc-field demagnetized***

– by Kerr micoscopy –

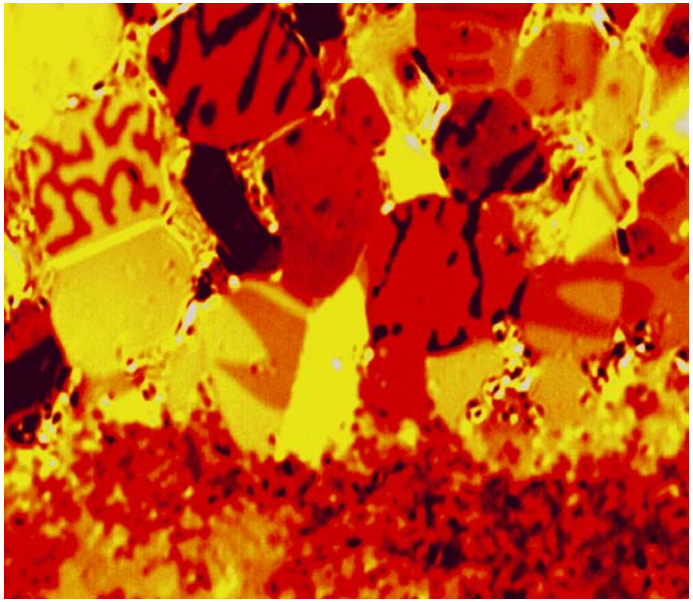
Classical and Interaction Domains

Hot-deformed melt-spun *Nd-Fe-B*

thermally demagnetized

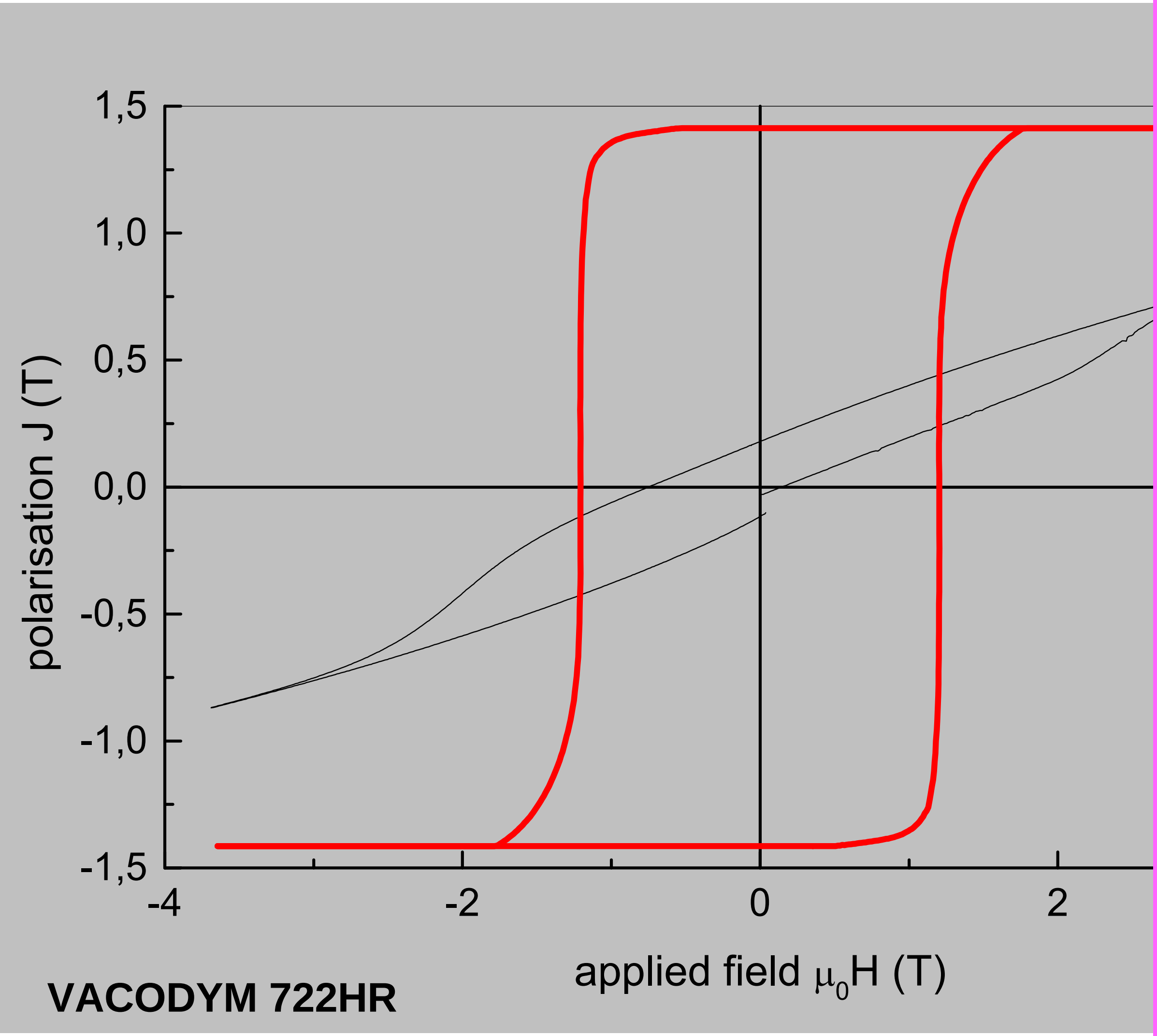


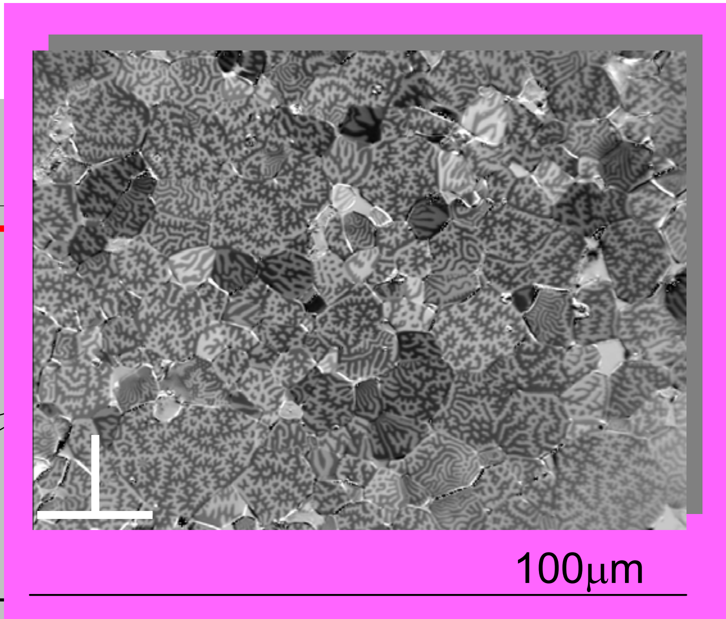
dc-field demagnetized

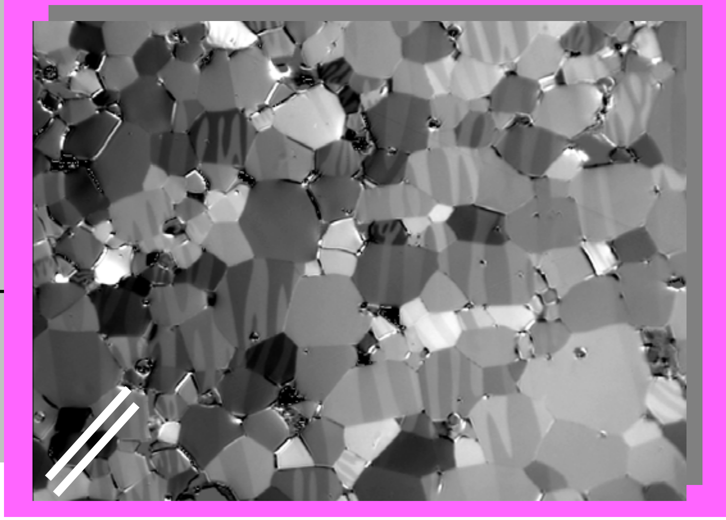


– by Kerr micoscopy –

High-quality sintered Nd-FeB magnet

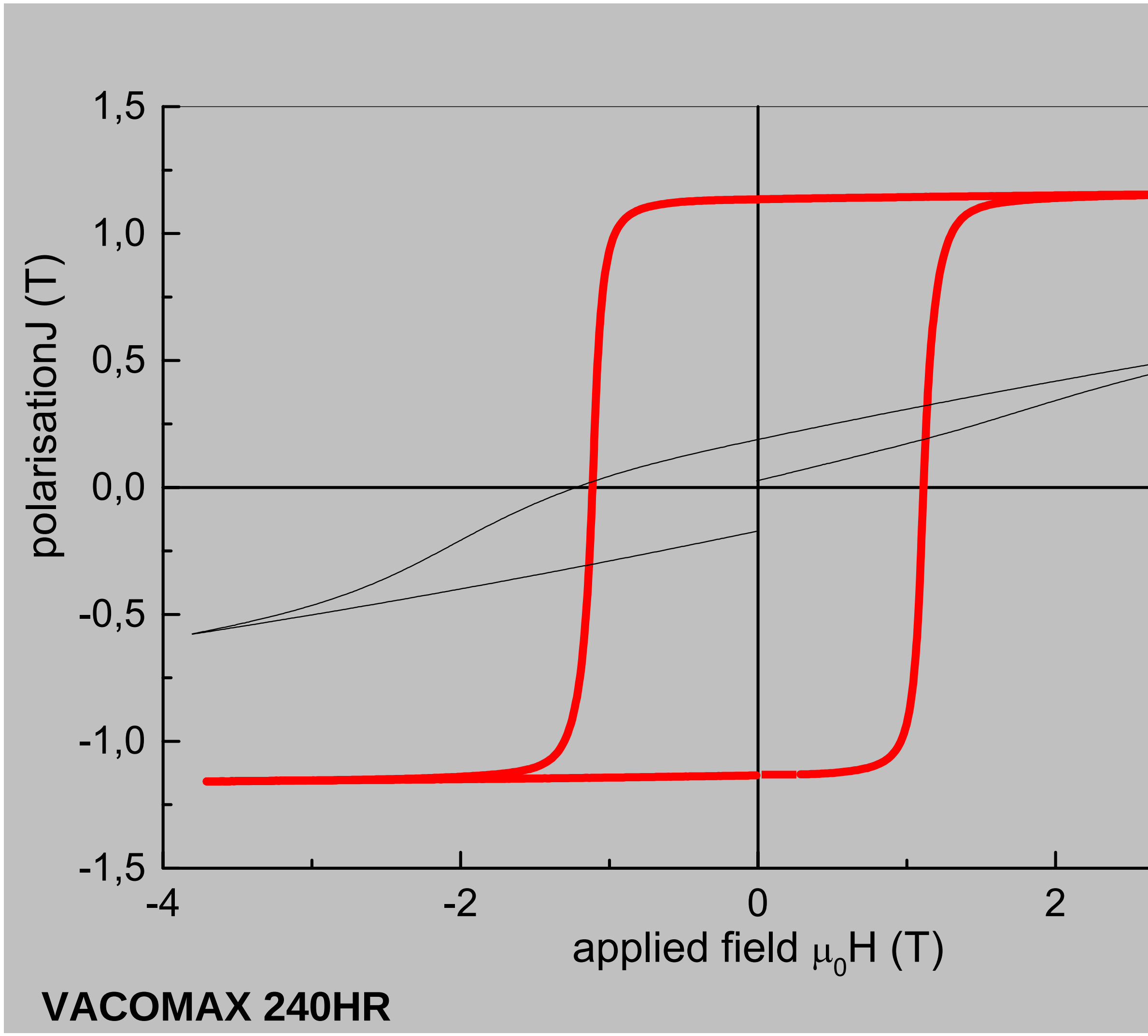


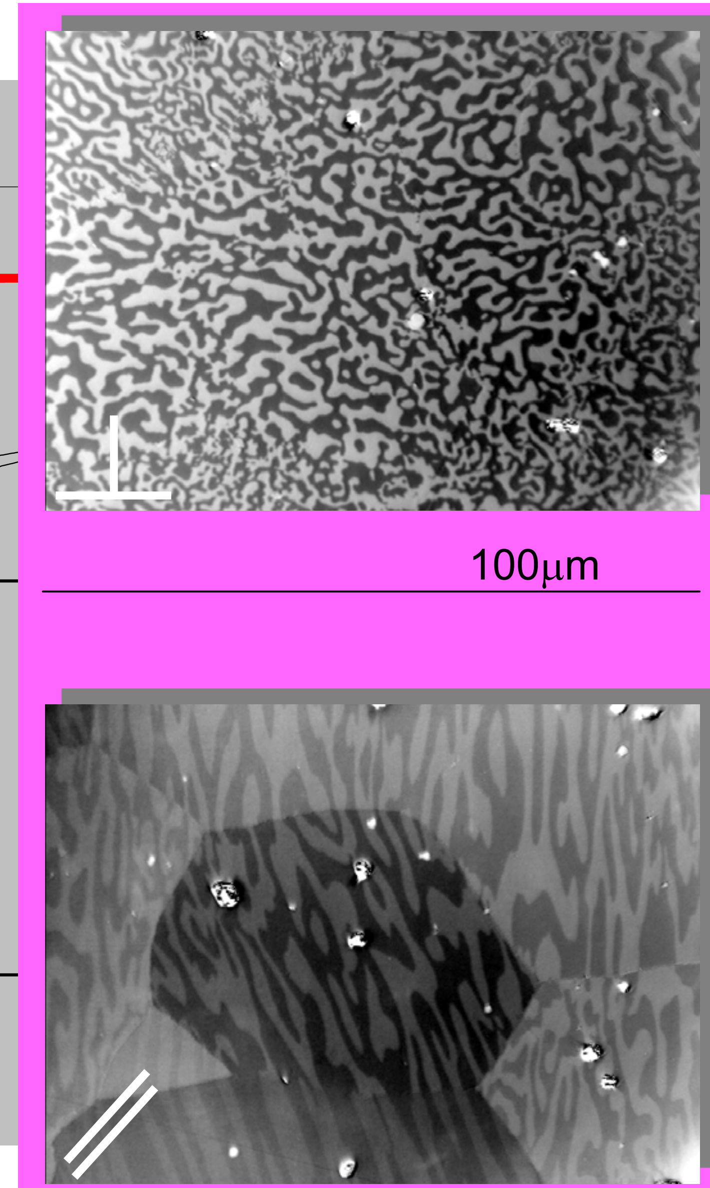




Kerr microscopy

High-quality Sm_2Co_{17} sintered magnet

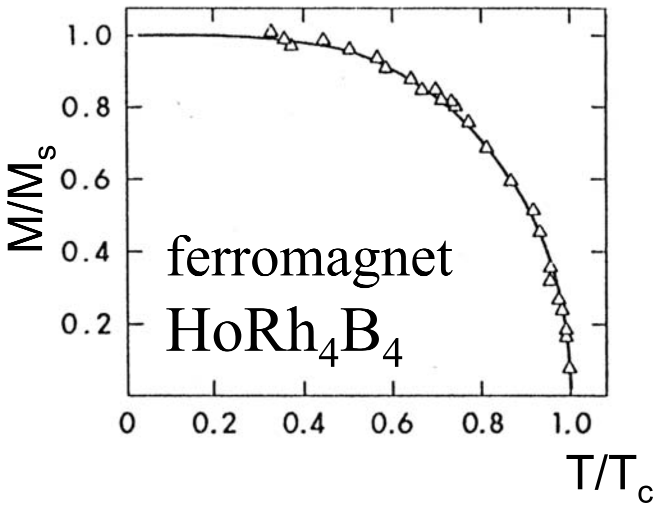




Kerr microscopy

5-3 Ordering and reorientation phase transitions

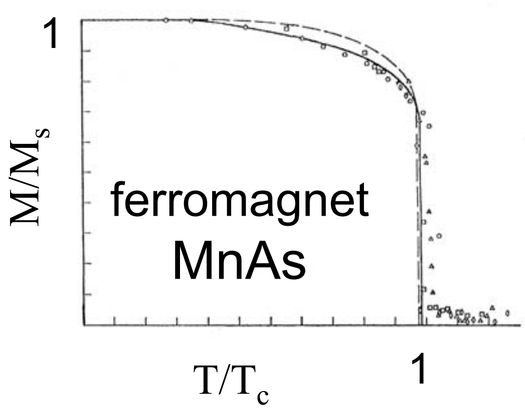
- a monotonic decrease of M(T)
  $\Rightarrow$ change at $T = T_c$ (or T_N) $\approx J/k_B$ from the paramagnetic state to fm or afm



- low temperature behaviour : $M/M_s = 1 - \alpha T^{3/2}$
  by spin waves
- ordering temperatures T_C and T_N
  by mean field approximations
  $M/M_s \approx (1 - T/T_c)^{1/2}$
- detailed behaviour near T_C or T_N
  by scaling and renormalization theory

- *second order phase transition* that separates
  states of different symmetry (L.D. Landau)

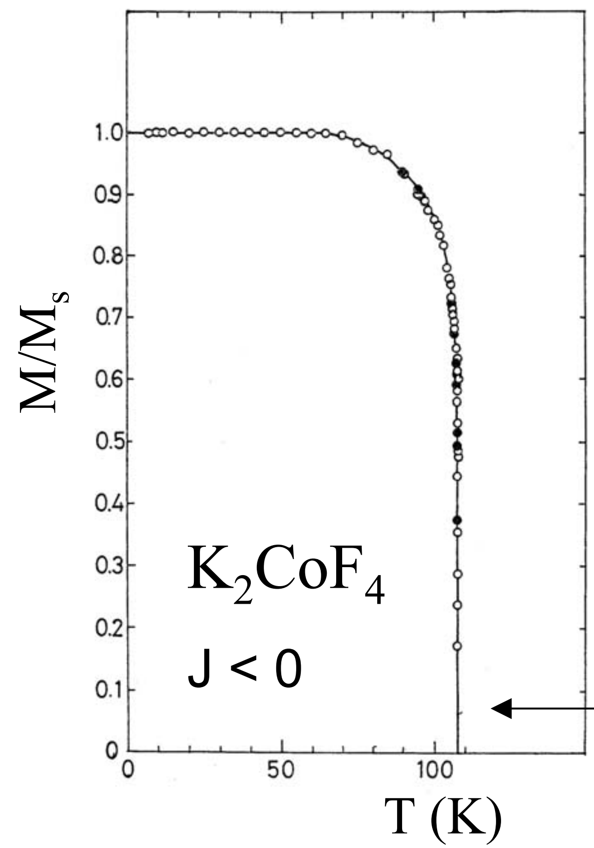
- magneto-elastic couplings $\Rightarrow$ *first order phase transition*
  e.g. MnAs, RCo_2 (R = Dy, Ho, Er) and $GdSi_2Ge_2$
  $\Rightarrow$ magnetic refrigeration



Magnetic ordering

- exact solution of the *d=2 Ising model* (L. Onsager 1944)

$$\mathcal{H} = -J \sum_{i,j} s_i^z s_j^z \qquad (s_i^z = \pm 1)$$



$k_B T_N = 2.27 J$

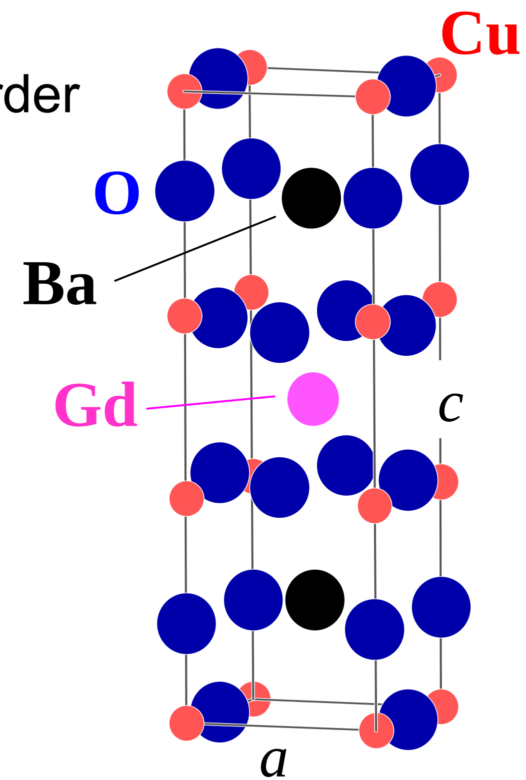
$M/M_s \approx (1 - T/T_N)^{1/8}$

$\Rightarrow$ strong influence of the dimensionality on the critical behaviour

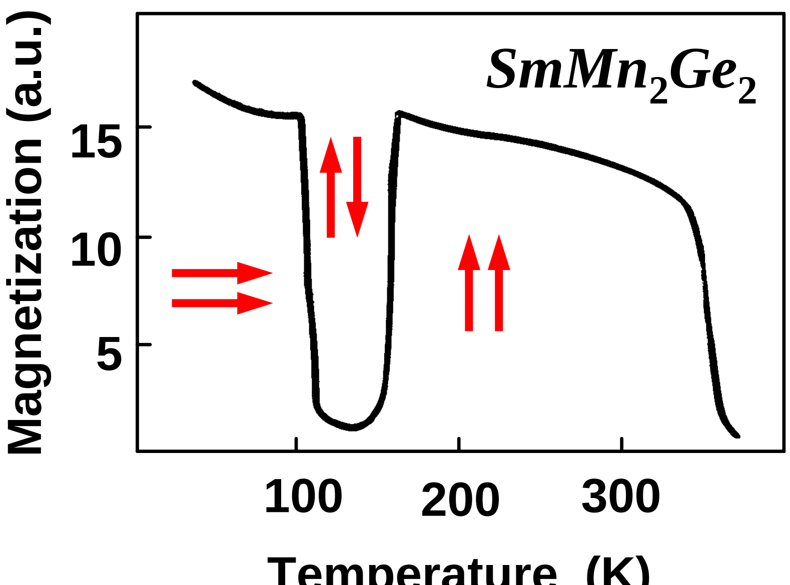
Magnetic ordering

- if the strength of the coupling between subsystems is moderate
  $\Rightarrow$ many different types of phase transitions

- e.g. : $GdBa_2Cu_3O_7$
  - *d* = 2 sublattice of Cu^{++} magnetic moments order antiferromagnetically at $T_N[Cu] \approx 95$ K
  - *d* ≈ 3 Gd-sublattice orders at $T_N[Gd] \approx 2.2$ K

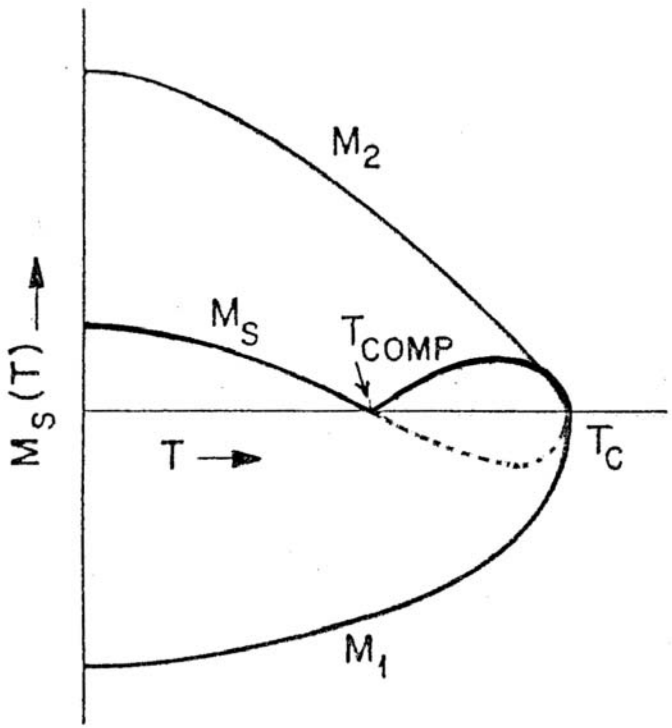


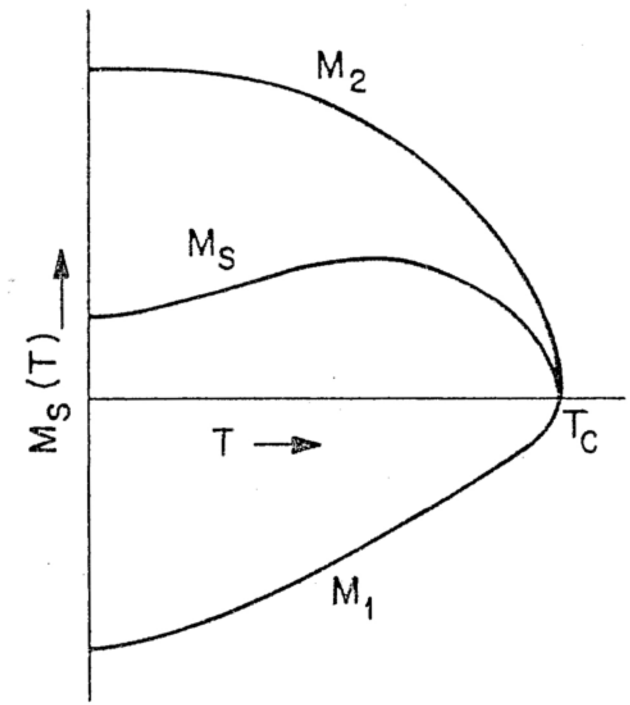
- reentrant ferromagnetism in $SmMn_2Ge_2$



Magnetic ordering

- *non-monotonic M(T) and compensation* of different contributions to M in ferrimagnets at $T = T_{comp}$





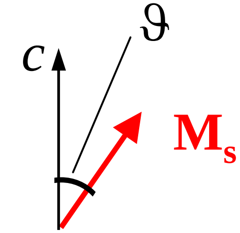
- e.g. $GdCo_4B$ $T_C = 505$ K and $T_{comp} = 410$ K

Magnetic ordering

- *spin reorientation transitions* :

  contributions of different subsystems to magnetic anisotropy are different

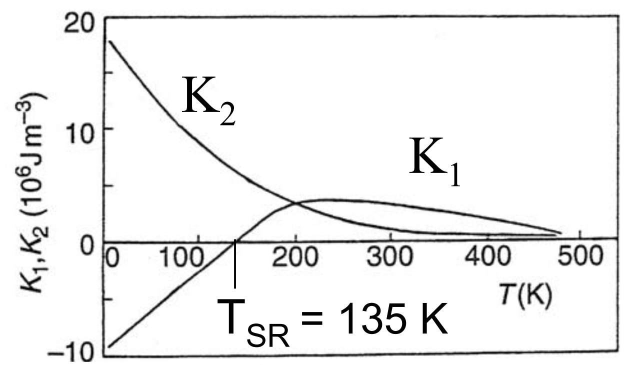
  example $Nd_2Fe_{14}B$:



$$E_A = \int_V d\tau [K_1 \sin^2\vartheta + K_2 \sin^4\vartheta + \ldots]$$

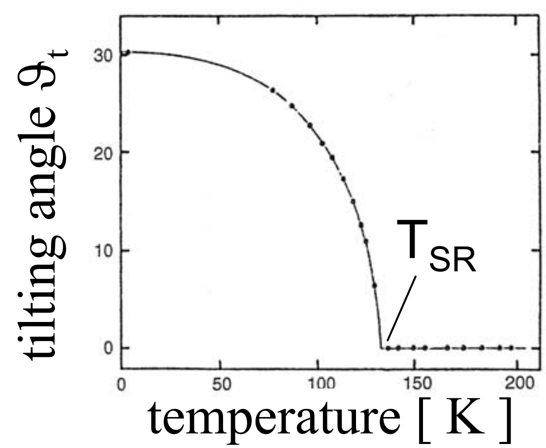
i.e. $K \rightarrow K_1$, …

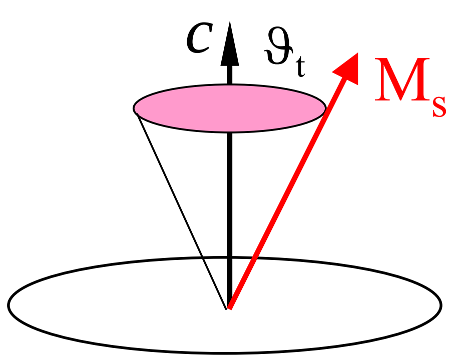
$K_1 = K_1[Nd] + K_1[Fe]$ ⇒ ⇒ ⇒



⇒ a transition at $T = T_{SR}$:

easy axis → easy cone

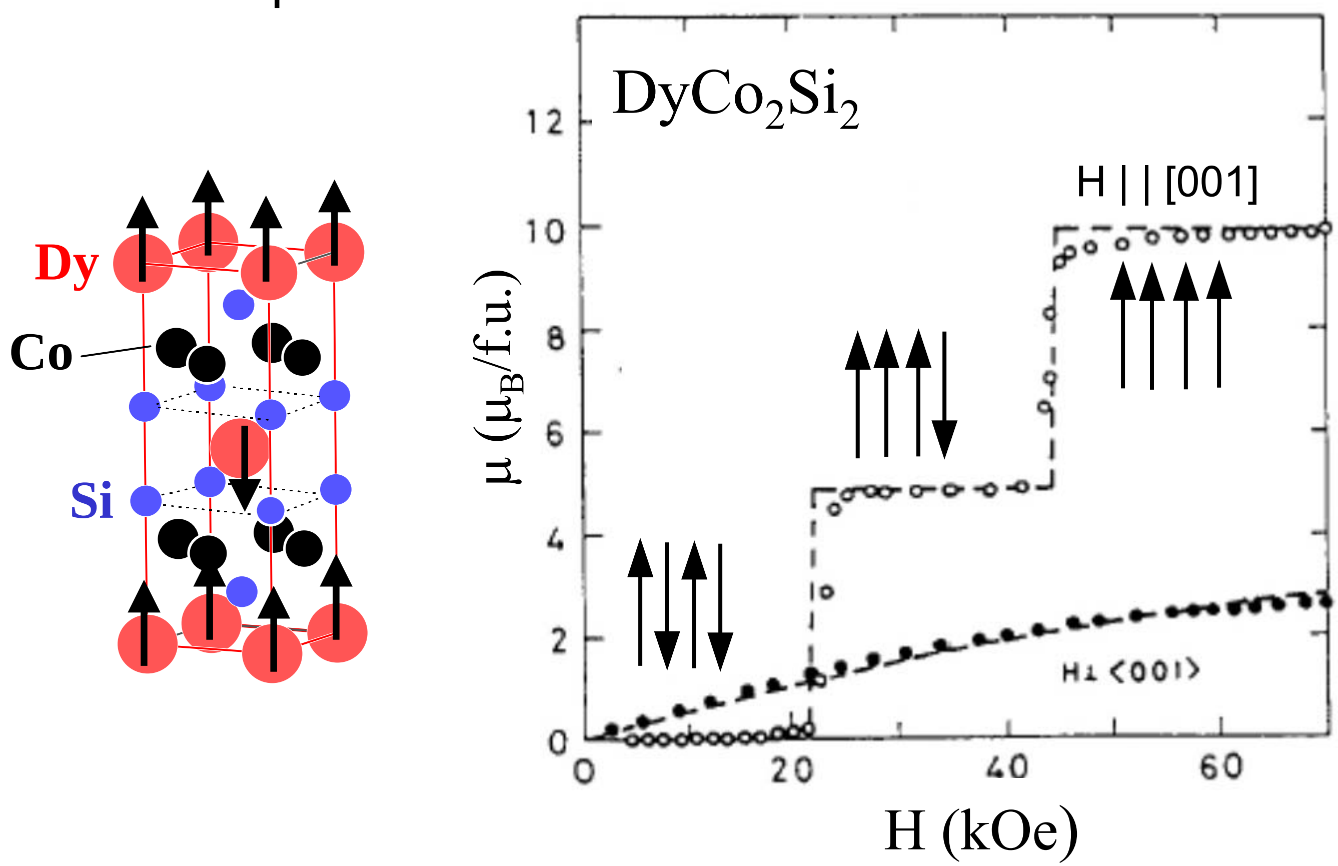




5-4 Metamagnetic transitions

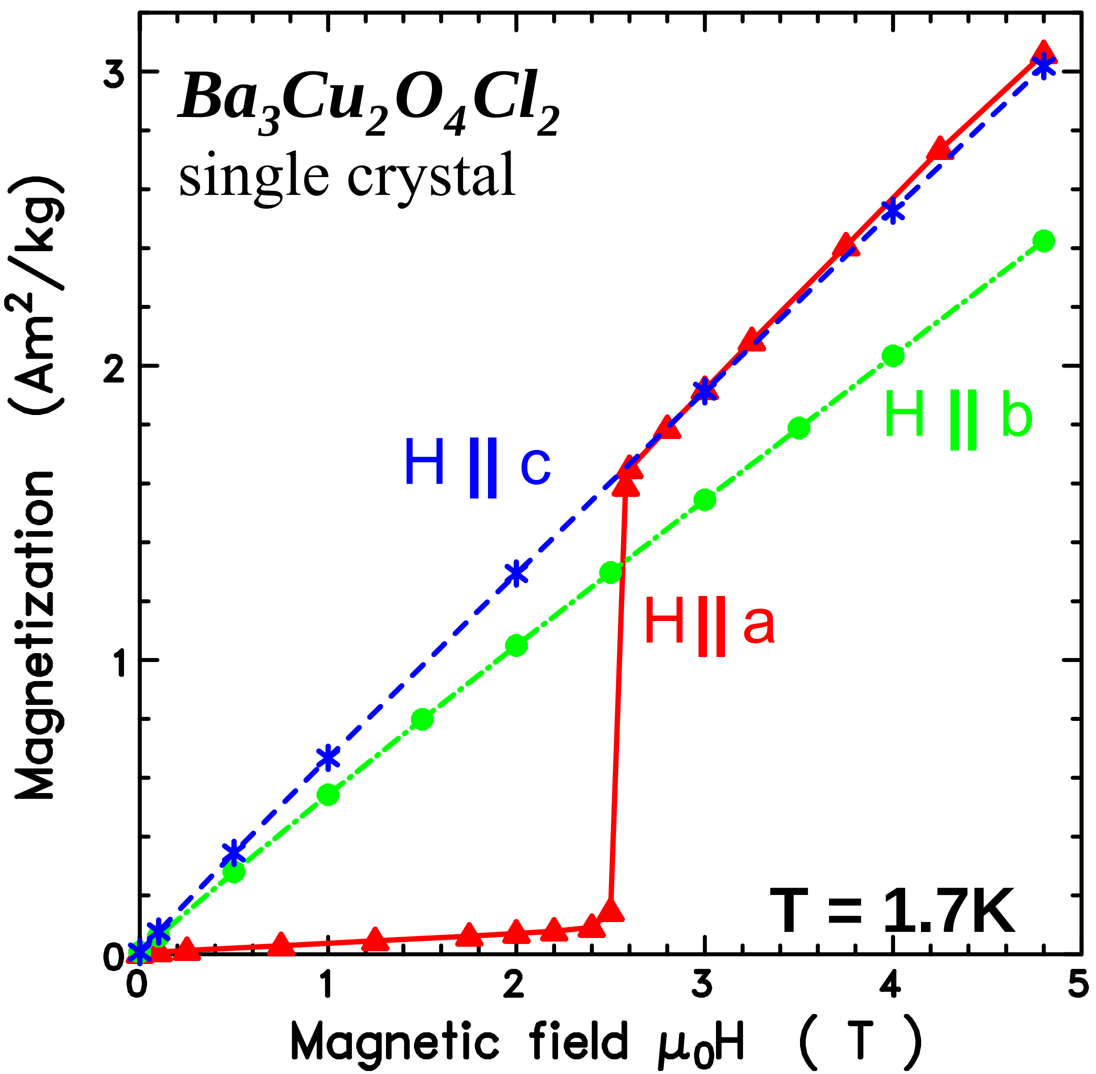
- field-induced magnetic phase transitions
- *spin-flip transition* 

  if afm exchange is relatively weak compared to the anisotropy field
- in $DyCo_2Si_2$ – A-type antiferromagnetic order with relatively weak interaction between the fm planes

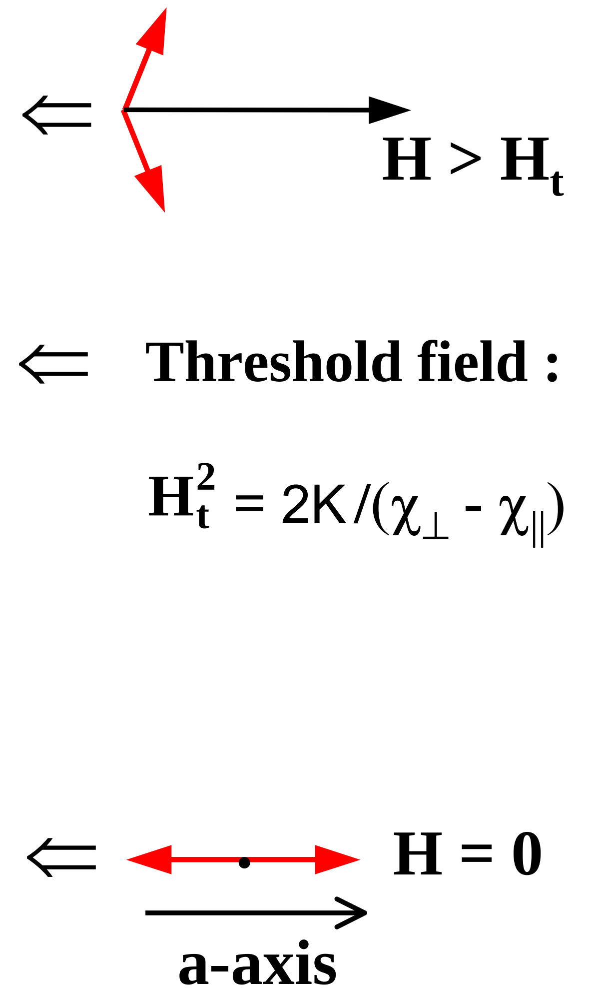


Metamagnetic transitions

- *spin flop transition*: If the exchange interaction in the antiferromagnet is relatively strong compared to the anisotropy field



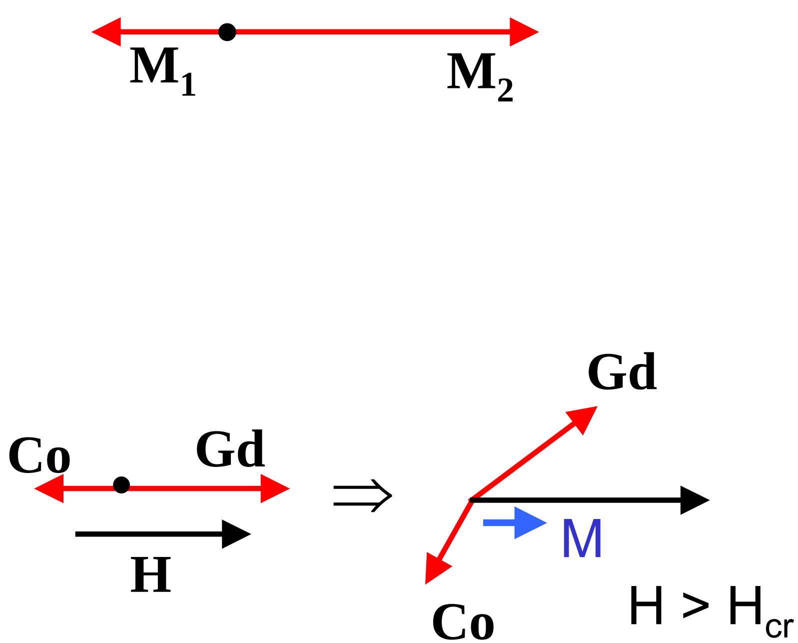
T < $T_N \approx$ 21K

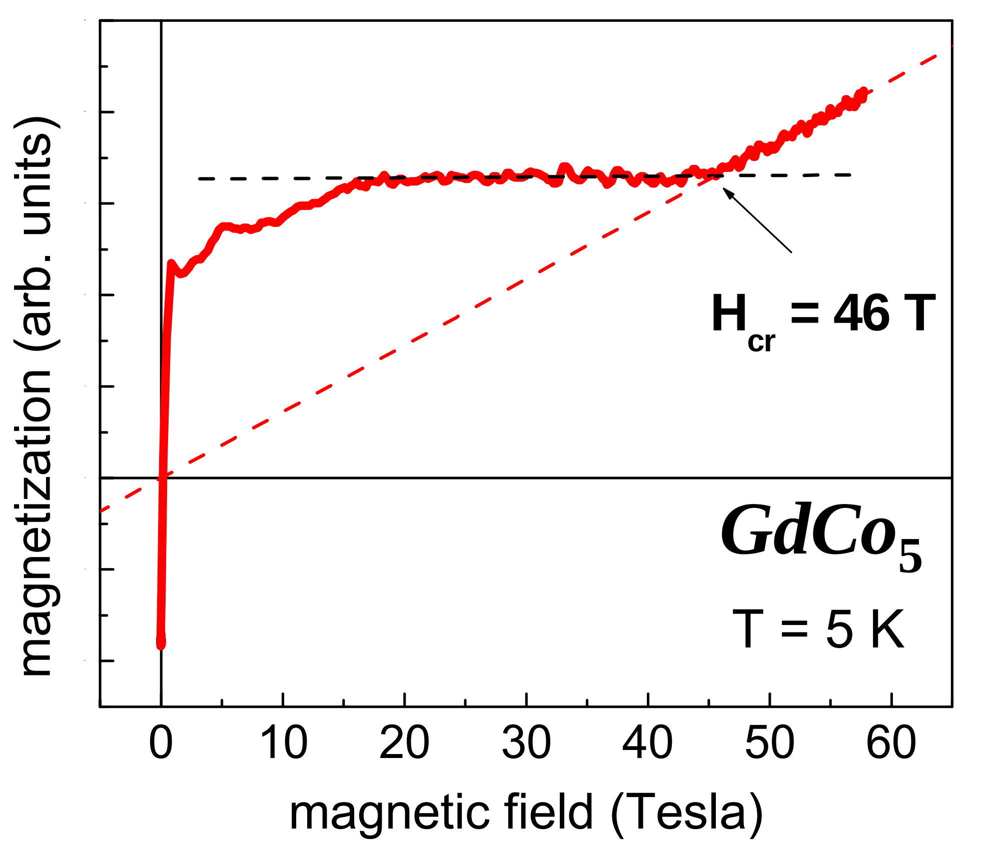


Metamagnetic transitions

- *spin canting in ferrimagnets*: a similar flopping phenomenon as the spin flop transition, examples $GdCo_5$, $DyCo_{12}B_6$
- second order phase transition (spin flop is first order !)
  - competition of exchange energy and field (Zeeman) energy

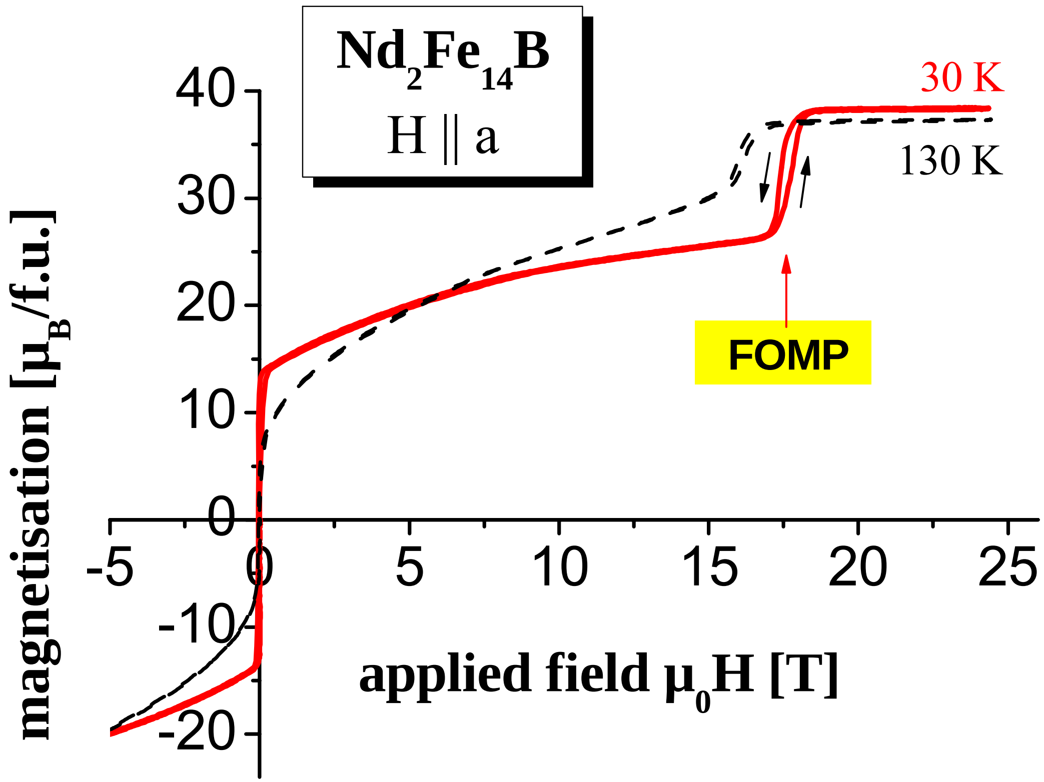
$$H_{cr} \sim |J||M_1 - M_2|$$





Metamagnetic transitions

- *First order magnetization processes* (FOMPs):
  ⇒ competition of magnetostatic (Zeeman) energy with the anisotropy energy of different orders



⇒ anisotropy in the tetragonal basal plane

Further forms of metamagnetic transitions

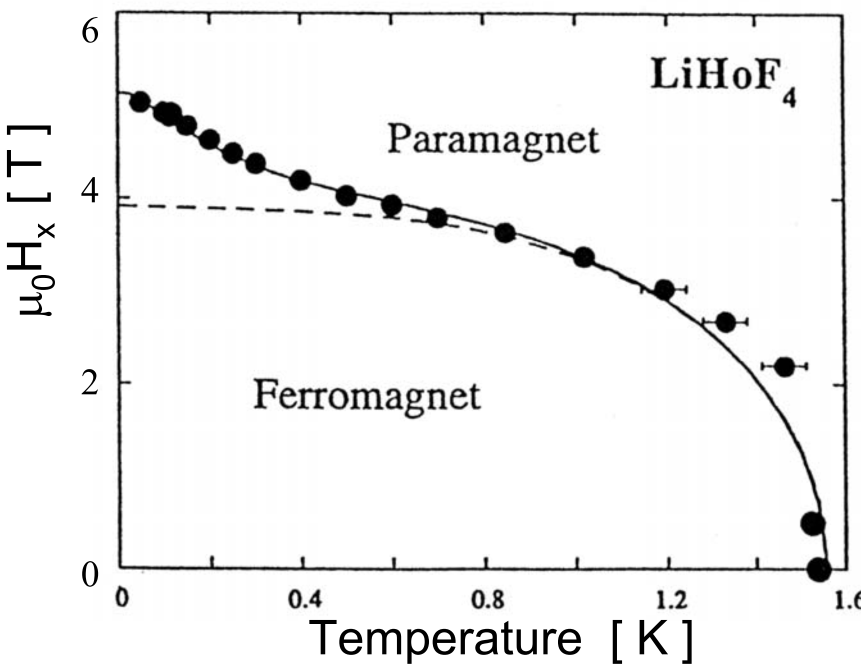
- *paramagnetic metamagnetism* in cases of non-Kramers ions
  e.g. TmSb, $PrNi_5$, Pr
  ⇒ from Van Vleck paramagnetism to Langevin-Curie paramagnetism

- *itinerant electron metamagnetism* (IEM): E.P. Wohlfarth and P. Rhodes (1962)
  ⇒ from Pauli paramagnetism to ferromagnetism, e.g. YCo_2, $LuCo_2$, $La(Fe,Si)_{13}$

5-5 Quantum phase transitions

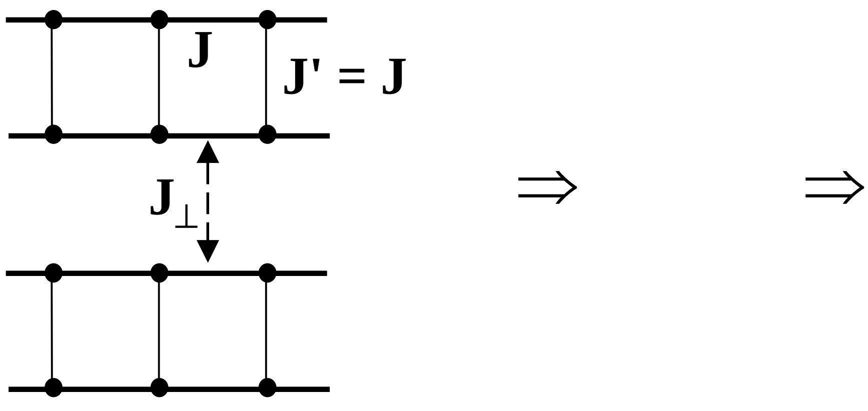
- phase transitions at zero temperature (first or second order)
- driven by quantum fluctuations instead of thermal fluctuations
  $\Rightarrow$ modification of scaling and renormalization
  $\Rightarrow$ consequences of the quantum critical behaviour at non-zero temperature **?**
- Ising ferromagnet $LiHoF_4$
  in a field applied perpendicularly to its magnetic axis
  it becomes a (quantum) paramagnet

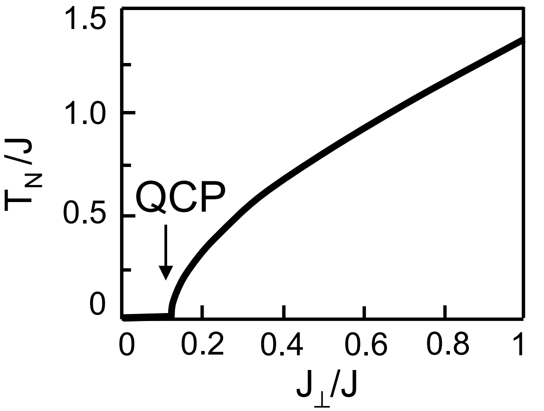
$$\mathcal{H} = -J \sum_{i,j} s_i^z s_j^z - g \mu_B H_x \sum_i s_i^x \qquad s_i^z = \pm 1/2 \;, \quad J > 0 \quad \Rightarrow \text{Ising ferromagnet}$$



Quantum phase transitions

- vanishing of the of the dimensionality crossover at a quantum critical points (QCP)
  - e.g. two-leg ladders show a spin gap similar as dimers
  - if such ladders are coupled to each other by a small $J_\perp$ the crossover to 3d behaviour at low temperatures occurs only above a critical value of $J_\perp/J$





- on the other hand, if *chains* are coupled by J' the spin gap Δ opens immediately upon the introduction of non-zero J'
  $\Rightarrow$ no QCP