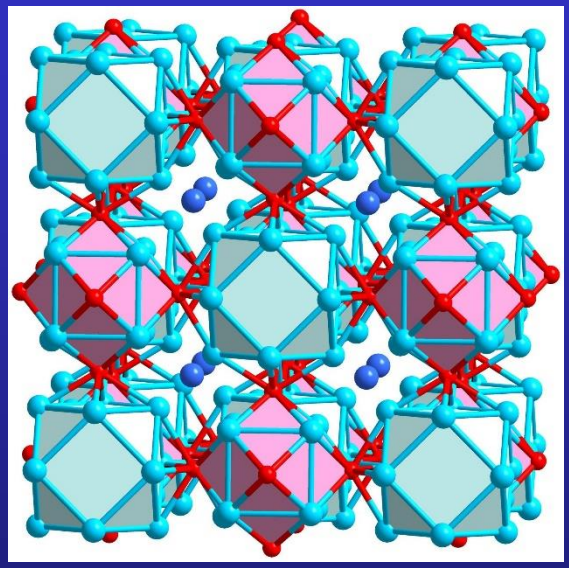
UNIVERSITY OF Southampton
School of Physics and Astronomy

Crystalline Solids

PHYS3004

Tim Freegarde



Crystalline Solids

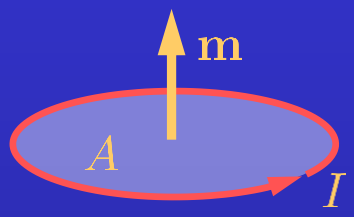
STRUCTURAL PROPERTIES	molecular bonds and crystal binding
	crystal structures and lattices
	reciprocal lattices
CLASSICAL PROPERTIES	x-ray, neutron & electron scattering and diffraction
	thermodynamics & transport
ELECTRONIC PROPERTIES	electron distributions
	band theory
	semiconductors
MAGNETIC PROPERTIES	magnetic interactions, ordering and domains

Physical principles

MAGNETIC MOMENTS

- magnetic dipole moment $\mathbf{m}$ may arise from circulating currents

$$\mathbf{m} = \frac{1}{2}\int \mathbf{r} \times \mathbf{j}\,\mathrm{d}V = I\,\mathbf{A}$$



- in magnetic flux density $\mathbf{B}$, magnetic dipole moment $\mathbf{m}$
  - experiences torque $\tau = \mathbf{m} \times \mathbf{B}$
  - has energy (Hamiltonian) $\mathcal{H} = -\mathbf{m} \cdot \mathbf{B}$
- bulk magnetization $\mathbf{M} = n\,\mathbf{m}$ is magnetic moment per unit volume (n dipoles/unit volume)

ELECTROMAGNETISM

- definition of susceptibility $\mathbf{M} = \chi\mathbf{H}$ and differential susceptibility $\chi' = \dfrac{\mathrm{d}\mathbf{M}}{\mathrm{d}\mathbf{H}}$

  and magnetic field strength $\mathbf{B} = \mu_0\mu_r\mathbf{H} = \mu_0(\mathbf{H}+\mathbf{M})$

CRYSTALLINE MAGNETISM

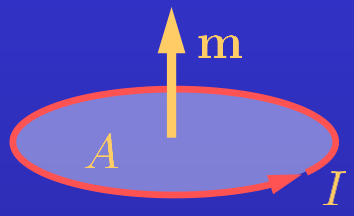
- dominated by electron orbital and spin angular momenta; nuclear contributions small
- arrangements determined by Coulomb interactions and Pauli exclusion principle

Physical principles

MAGNETIC MOMENTS

- magnetic dipole moment $\mathbf{m}$ may arise from circulating currents

$$\mathbf{m} = \frac{1}{2}\int \mathbf{r}\times\mathbf{j}\,\mathrm{d}V = I\,\mathbf{A}$$



- hence is related to angular momentum $\mathbf{L}_{\mathrm{class}}$ of particle with charge $\mathbf{q}$ and mass $\mathbf{m_q}$

$$\mathbf{m} = \frac{1}{2}\int \mathbf{r}\times q\mathbf{v}\,\mathrm{d}V = \frac{q}{2m_q}\mathbf{L}_{\mathrm{class}}$$

- so with quantum mechanical angular momentum $\hbar\mathbf{L}$,

$$\mathbf{m} = -\frac{e\hbar}{2m_e}\mathbf{L} = -\mu_{\mathrm{B}}\mathbf{L} \qquad \text{where Bohr magneton } \mu_{\mathrm{B}} \equiv \frac{e\hbar}{2m_e}$$

- with spin angular momentum $\hbar\mathbf{S}$, relation is modified by gyromagnetic ratio $g \approx 2.0023$

$$\mathbf{m} = -g\mu_{\mathrm{B}}\mathbf{S}$$

so in general, $\mathbf{m} = -\mu_{\mathrm{B}}(\mathbf{L} + g\mathbf{S})$

Caution: common to use $\mathbf{S}$ for $\mathbf{J}$

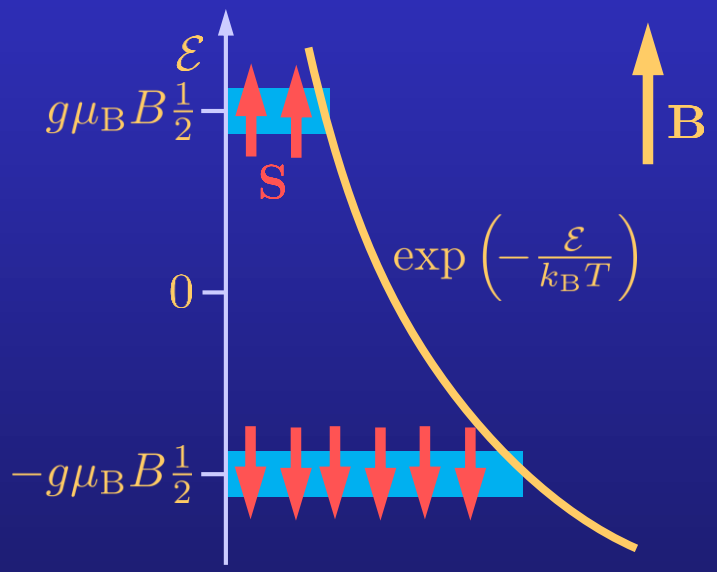
or $m_z = -\tilde{g}\mu_{\mathrm{B}}J_z$ where Landé *g*-factor $\tilde{g} = \frac{1}{2}(g+1) + \frac{1}{2}(g-1)\left[\frac{S(S+1)-L(L+1)}{J(J+1)}\right]$

Types of magnetism

external field		internal field
single atom	multiple atoms	
Curie-Langevin paramagnetism $\mathcal{H} = g\mu_B \mathbf{B}\cdot\mathbf{S}$ $\chi > 0$ Boltzmann	Pauli paramagnetism $\mathcal{H} = g\mu_B \mathbf{B}\cdot\mathbf{S}$ $\chi > 0$ Fermi-Dirac	ferromagnetism  $J_{ij} > 0$
Larmor diamagnetism $\mathcal{H} = \frac{e^2}{2m}\frac{1}{4}\lvert\mathbf{B}\times\mathbf{r}\rvert^2$ $\chi < 0$ $\mathbf{S} = 0$	Landau diamagnetism $\chi_{\text{Landau}} = -\frac{1}{3}\chi_{\text{Pauli}}$ $\mathbf{S} = 0$	ferrimagnetism  dissimilar spins in unit cell
		antiferromagnetism $\chi = 0$  $J_{ij} < 0$

Curie-Langevin paramagnetism

- single/isolated atoms in external field
  → magnetic+non-magnetic compounds
- Boltzmann distribution
- Hamiltonian $\mathcal{H} = g\mu_B \mathbf{B} \cdot \mathbf{S}$



- remember that $\mathbf{m}$ is antiparallel to $\mathbf{S}$
- $g \approx 2$ so $g|S| \approx 1$

Single spin-$\frac{1}{2}$

$H = g\mu_B \underline{B}\cdot\underline{S}$ where $\underline{\mu_e} = -g\mu_B \underline{S}$ $\mu_B = \frac{e\hbar}{2m}$

⇒ at temperature T, mean magnetic moment per electron will be

$$\langle\mu\rangle = \mu_B \frac{\left(-\exp\frac{-\mu_B B}{kT} + \exp\frac{\mu_B B}{kT}\right)}{\exp\frac{-\mu_B B}{kT} + \exp\frac{\mu_B B}{kT}}$$

$$= \mu_B \tanh\frac{\mu_B B}{kT}$$

$$\Rightarrow \chi = \frac{\partial M}{\partial H} = \frac{\partial}{\partial B}\mu_0 n\mu_B \tanh\frac{\mu_B B}{kT}$$

CURIE LAW $= \frac{n\mu_0\mu_B^2}{kT}$ for $\mu_B B \ll kT$

General form for angular momentum $\underline{S}$

$$\langle\mu\rangle = \mu_B \frac{\sum_{S_z} -S\exp\frac{-\mu_B B S_z}{kT}}{\sum_{S_z}\exp\frac{-\mu_B B S_z}{kT}}$$

Check for spin $S = \frac{1}{2}$:

$$\Rightarrow \chi = \frac{n\mu_0\mu_B^2}{kT}\frac{\tilde{g}^2 S(S+1)}{3} \qquad \left(\frac{2^2\,\frac{1}{2}\,\frac{3}{2}}{3} = 1\right)$$

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- single/isolated atoms in external field
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- Hamiltonian $\mathcal{H} = g\mu_B \mathbf{B}\cdot\mathbf{S}$

$$\chi = \frac{n\mu_0\mu_B^2}{k_{\mathbf{B}}T} \rightarrow \frac{n\mu_0\mu_B^2}{k_{\mathbf{B}}T}\frac{\tilde{g}^2 S(S+1)}{3}$$

CURIE LAW

- susceptibility varies inversely with T

- magnetization parallel to applied field: $\chi > 0$
- saturation at high B as permanent dipoles all aligned

- Landé g-factor

$$\tilde{g} = \frac{1}{2}(g+1) + \frac{1}{2}(g-1)\left[\frac{S(S+1)-L(L+1)}{J(J+1)}\right]$$

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$$= \frac{n\mu_0\mu_B^2}{kT} \text{ for } \mu_B B \ll kT$$

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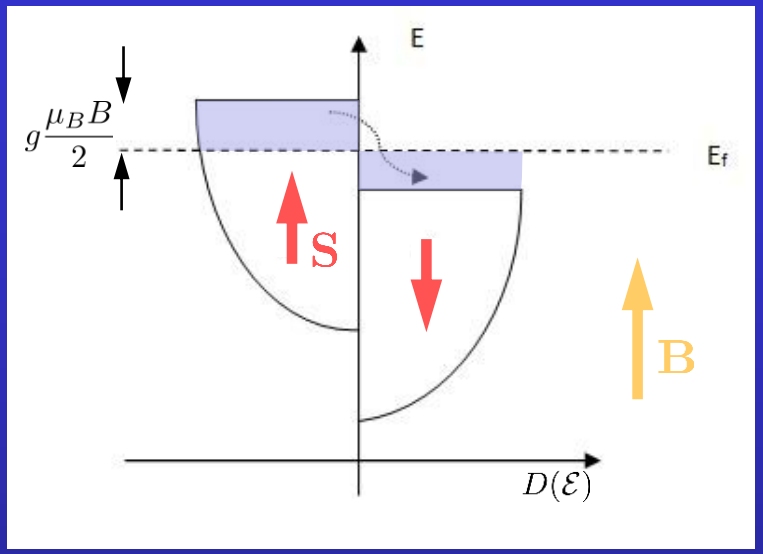
Pauli paramagnetism

- ensemble of atoms in external field
- requires overlapping electron wavefunctions & incompletely full bands → metals
- Fermi-Dirac distribution
- electrons move between spin states to equilibrate chemical potential

$$\chi = \mu_0 \mu_B^2 \, D(E_F)$$

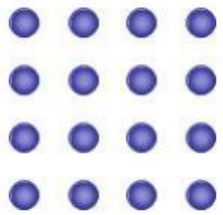
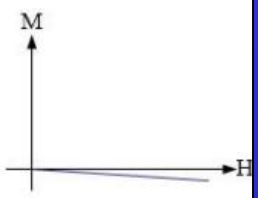
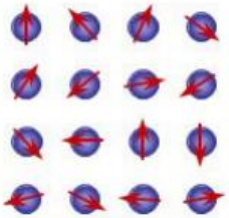
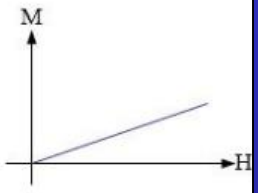
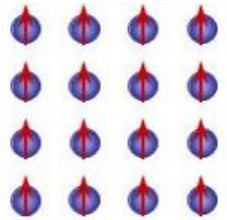
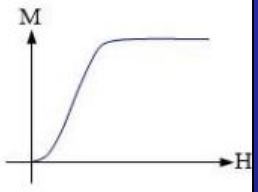
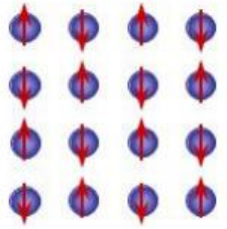
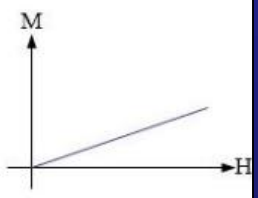
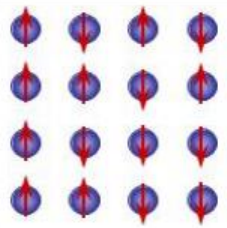
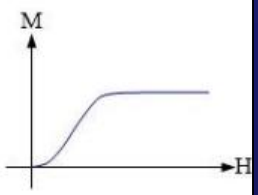
DENSITY OF STATES

- susceptibility smaller than typical paramagnetic susceptibilities by factor $\sim k_B T / E_F$
- magnetization parallel to applied field: $\chi > 0$
- remember that $\mathbf{m}$ is antiparallel to $\mathbf{S}$



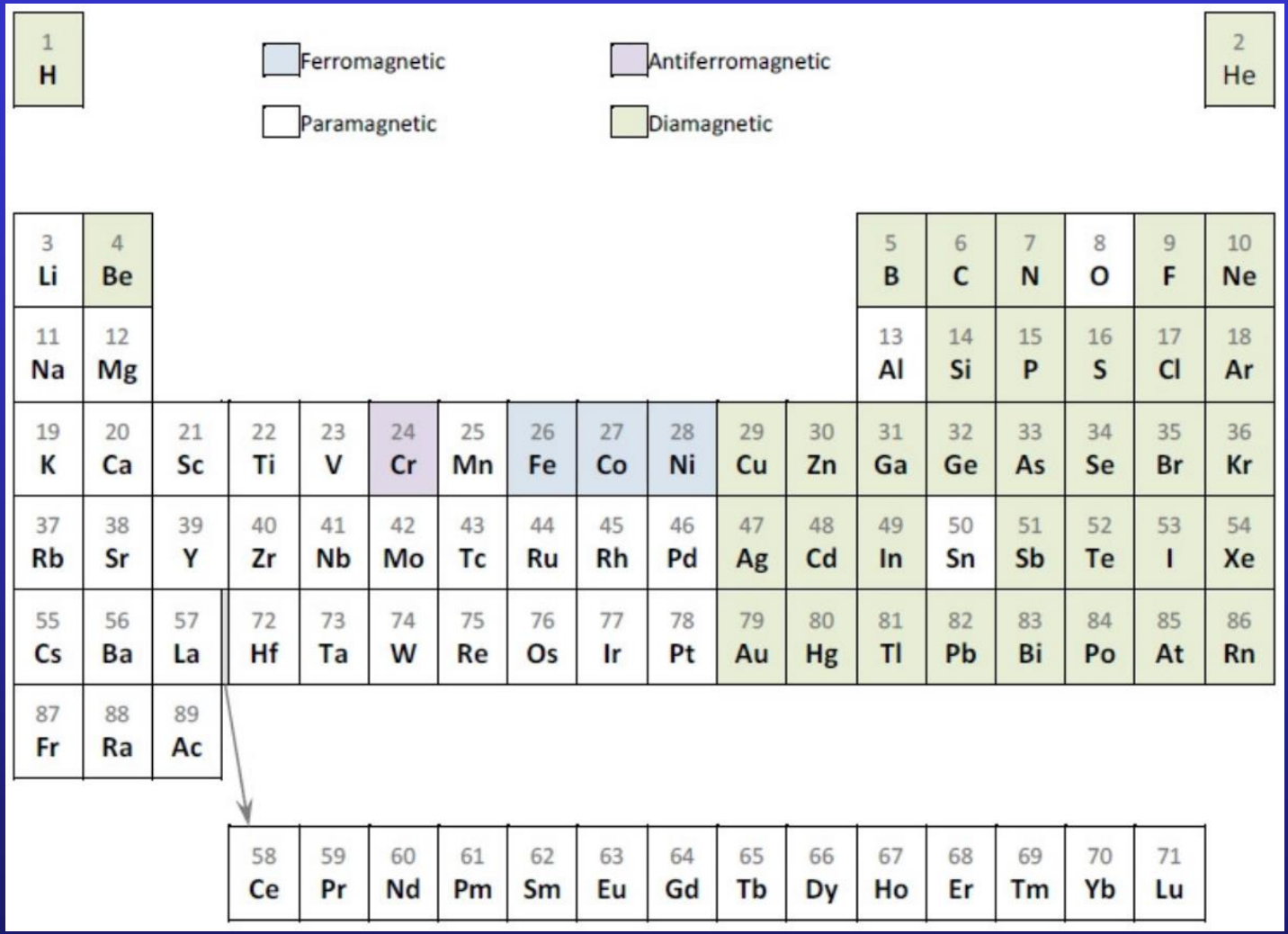
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Magnetic materials

Type	Example	Atomic/Magnetic Behaviour		
Diamagnetism	Inert gases; many metals eg Au, Cu, Hg; non-metallic elements e.g. B, Si, P, S; many ions e.g. Na^+, Cl^- & their salts; diatomic molecules e.g. H_2, N_2; H_2O; most organic compounds	Atoms have no magnetic moment. Susceptibility is small & negative, -10^{-6} to -10^{-5}		
Paramagnetism	Some metals, e.g. Al; some diatomic gases, e.g. O_2, NO; ions of transition metals and rare earth metals, and their salts; rare earth oxides.	Atoms have randomly oriented magnetic moments. Susceptibility is small & positive, $+10^{-5}$ to $+10^{-3}$		
Ferromagnetism	Transition metals Fe, H, Co, Ni; rare earths with $64 \leq Z \leq 69$; alloys of ferromagnetic elements; some alloys of Mn, e.g. MnBi, Cu_2MnAl.	Atoms have parallel aligned magnetic moments. Susceptibility is large (below T_C)		
Antiferromagnetism	Transition metals Mn, Cr & many of their compound, e.g. MnO, CoO, NiO, Cr_2O_3, MnS, MnSe, $CuCl_2$.	Atoms have anti-parallel aligned magnetic moments. Susceptibility is small & positive, $+10^{-5}$ to $+10^{-3}$		
Ferrimagnetism	Fe_3O_4 (magnetite); γ-Fe_2O_3 (maghemite); mixed oxides of iron and other elements such as Sr ferrite.	Atoms have mixed parallel and anti-parallel aligned magnetic moments. Susceptibility is large (below T_C)		

www.birmingham.ac.uk/Documents/college-eps/metallurgy/research/Magnetic-Materials-Background/Magnetic-Materials-Background-4-Classification-of-Magnetic-Materials.pdf

Magnetic elements



https://www.birmingham.ac.uk/Documents/college-eps/metallurgy/research/Magnetic-Materials-Background/Magnetic-Materials-Background-4-Classification-of-Magnetic-Materials.pdf

- remember that many magnetic materials are compounds or alloys

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		antiferromagnetism $\chi = 0$  $J_{ij} < 0$

Larmor diamagnetism

- single atom in external field
- exhibited by all materials, but masked by paramagnetism or ferro/ferrimagnetism unless angular momentum $\mathbf{S} = 0$

$$\Delta E = \frac{e^2}{8m}\langle|\mathbf{B}\times\mathbf{r}|\rangle^2$$

$$\chi = -\frac{n\mu_0 e^2}{6m}\langle r^2\rangle$$

- material response resembles Lenz's law
- magnetization antiparallel to applied field: $\chi < 0$

With a magnetic field $\underline{B} = \nabla \times \underline{A}$,

$$H = \frac{(\underline{p} + e\underline{A})^2}{2m} + g\mu_B \underline{B}.\underline{S} + V(r)$$

$$\Rightarrow \text{if } S=0,\ H = H_0 + \frac{e^2}{2m}\frac{1}{4}|\underline{B}\times\underline{r}|^2$$

$$\Rightarrow \Delta E = \frac{e^2}{8m}\langle|\underline{B}\times\underline{r}|\rangle^2$$

$$= \frac{e^2 B_z^2}{8m}\langle x^2+y^2\rangle = \frac{e^2 B_z^2}{8m}\frac{2}{3}\langle r^2\rangle$$

Now $\Delta E = -\frac{1}{2}\underline{M}.\underline{B} = -\frac{1}{2}\chi\underline{H}.\underline{B} = \frac{-1}{2\mu_0}B^2\chi$

$$\Rightarrow \chi = -n\frac{\mu_0 e^2}{6m}\langle r^2\rangle$$ for electron density n

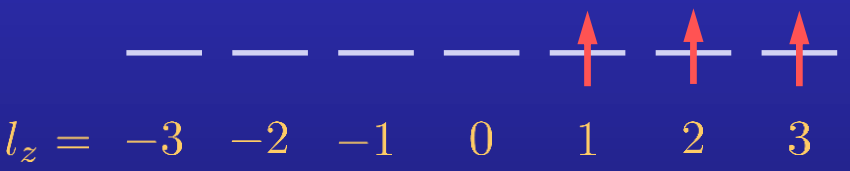
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Hund's rules

- lowest energy term has
  - maximum multiplicity – spins align
  - largest orbital angular momentum
  - anti-/aligned spin and orbital angular momenta
- Pauli exclusion principle separates repulsive electrons / nuclear screening
- highest $\mathbf{L}$ states have lowest Coulomb interactions
- lowest spin-orbit dipole-dipole interaction energy $J = |\mathbf{L} \pm \mathbf{S}|$

- eg Praseodymium Pr $[\mathrm{Xe}].4f^3.6s^2$



- bonding concerned Coulomb energy
  - of single-electron states
  - anti-/symmetry with respect to position
  - neglected electron-electron interaction
- Hund's rules concern
  - symmetry wrt exchange of 2 or more electrons
  - electron-electron & electron-nucleus interactions

Exchange energy

- consider two states A and B
- place one electron in each state
- label states $|12\rangle$
- states coupled where they overlap
- Eigenstates will be superpositions

$$|\text{singlet}\rangle = (|AB\rangle + |BA\rangle)/\sqrt{2}$$
$$|\text{triplet}\rangle = (|AB\rangle - |BA\rangle)/\sqrt{2}$$

- labels reflect total spin of states with overall anti-symmetry
- general representation of alignment in terms of angular momenta
- Ising model when anisotropy confines angular momentum orientation

Let $V(r_1, r_2)$ represent the Coulomb interaction between electrons

Energies will be

$$2E_{singlet} = (\langle AB| + \langle BA|)\, V\, (|AB\rangle + |BA\rangle)$$
$$= \langle AB|V|AB\rangle + \langle BA|V|BA\rangle + \langle AB|V|BA\rangle + \langle BA|V|AB\rangle$$
$$= \langle AB|V|AB\rangle + \langle BA|V|BA\rangle + 2\,\mathrm{Re}\langle AB|V|BA\rangle$$

$$2E_{triplet} = (\langle AB| - \langle BA|)\, V\, (|AB\rangle - |BA\rangle)$$
$$= \langle AB|V|AB\rangle + \langle BA|V|BA\rangle - \langle AB|V|BA\rangle - \langle BA|V|AB\rangle$$
$$= \langle AB|V|AB\rangle + \langle BA|V|BA\rangle - 2\,\mathrm{Re}\langle AB|V|BA\rangle$$

$$\Rightarrow E_{singlet} = E_1 + J \qquad E_1 = \langle AB|V|AB\rangle \quad \uparrow\downarrow \; S=0$$
$$E_{triplet} = E_1 - J \quad \text{where} \quad J = \mathrm{Re}\langle AB|V|BA\rangle \quad \uparrow\uparrow \; S=1$$

$$E = E_1 - 4J\, \underbrace{\underline{S}_1 \cdot \underline{S}_2}$$

conventionally refer to total angular momenta, not just spin

Magnetic interactions

EXTERNAL MAGNETIC FIELD

- paramagnetic interaction with 'free' magnetic dipoles
- diamagnetic interaction with induced magnetic dipoles

INTERNAL MAGNETIC FIELD

- exchange interaction between magnetic dipoles
- exchange term J_{ij} may be +ve or -ve

MEAN FIELD THEORY

- combined exchange interaction represented as magnetic interaction with effective mean field

Add magnetic field $\underline{B} = \nabla \times \underline{A}$:

$$H = \frac{(\underline{p} + e\underline{A})^2}{2m} + g\mu_B \underline{B} \cdot \underline{S} + V(r)$$

$$= H_0 + \mu_B \underline{B} \cdot (\underline{l} + g\underline{s}) + \frac{e^2}{2m} \frac{1}{4} |\underline{B} \times \underline{r}|^2$$

PARAMAGNETIC DIAMAGNETIC

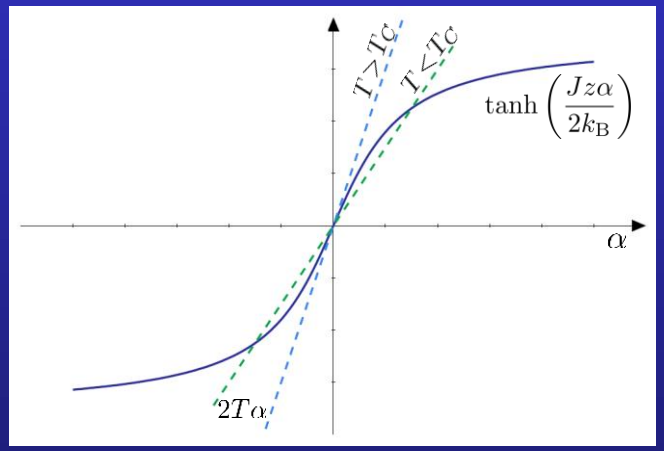
$$H = -\frac{1}{2} \sum_{i,j} \underline{S}_i \cdot \underline{S}_j J_{ij}$$

$$H_i = -\underline{S}_i \cdot \sum_j J_{ij} \underline{S}_j$$

Ferromagnetism & Curie-Weiss law

FERROMAGNETISM

- solve self-consistency equation – eg graphically for $B = 0$:
  - set $\alpha \equiv \langle S \rangle / T$



- magnetization without presence of external field if

$$T < T_C \approx \frac{Jz}{4k_B}$$

CURIE TEMPERATURE

- exchange interactions between magnetic atoms in lattice

Consider mean-field interaction:

$$H = -\tfrac{1}{2}\sum_{ij} J_{ij}\,\underline{S}_i \cdot \underline{S}_j + g\mu_B \underline{B} \cdot \sum_i \underline{S}_i$$

$$= \sum_i \underline{S}_i \cdot \left\{ -\tfrac{1}{2}\sum_j J_{ij}\,\underline{S}_j + g\mu_B \underline{B} \right\}$$

$$= \sum_i H_i$$

where $H_i = \underline{S}_i \cdot \left\{ -\tfrac{1}{2}\sum_j J_{ij}\,\underline{S}_j + g\mu_B \underline{B} \right\}$

$$= \underline{S}_i \cdot g\mu_B \left\{ \underline{B} - \frac{1}{2g\mu_B}\sum_j J_{ij}\,\underline{S}_j \right\} = \underline{S}_i \cdot g\mu_B \underline{B}_{eff}$$

For spin-$\tfrac{1}{2}$ Ising model, we follow the Curie-Langevin calculation

$$\Rightarrow \quad \langle s \rangle = -\tfrac{1}{2}\tanh\frac{g\mu_B B_{eff}}{2kT}$$

where if J_{ij} is constant for z nearest neighbours,

$$g\mu_B B_{eff} = g\mu_B B - Jz\langle s \rangle$$

$$\Rightarrow \quad \boxed{\langle s \rangle = -\tfrac{1}{2}\tanh\frac{g\mu_B B - Jz\langle s \rangle}{2kT}}$$

The magnetization is then

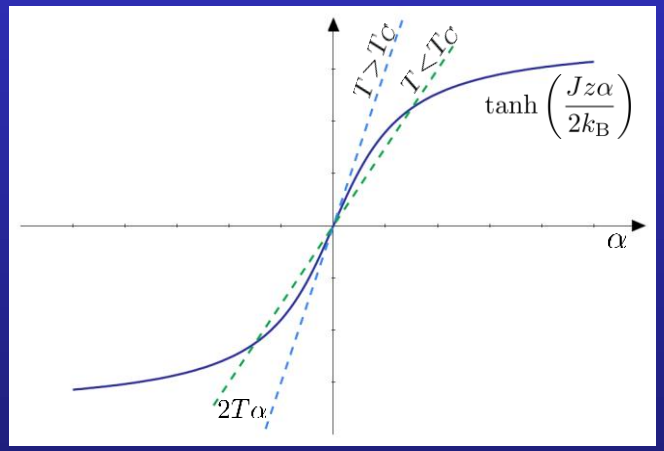
$$M = -ng\mu_B \langle s \rangle$$

Ferromagnetism & Curie-Weiss law

CURIE-WEISS LAW

- paramagnetic behaviour if $T > T_C$

$$\chi = \frac{\frac{1}{4} g^2 \mu_B^2 \mu_0 n}{k(T - T_C)}$$



- magnetization without presence of external field if

$$T < T_C \approx \frac{Jz}{4k_B}$$

CURIE TEMPERATURE

- exchange interactions between magnetic atoms in lattice

For small fields and $T > T_C$,

$$\langle s \rangle \approx -\frac{1}{2} \frac{g\mu_B B - Jz\langle s \rangle}{2kT}$$

$$\Rightarrow \langle s \rangle \left(1 - \frac{Jz}{4kT}\right) = \frac{-g\mu_B B}{4kT}$$

$$\Rightarrow \langle s \rangle = \frac{-g\mu_B B}{4kT\left(1 - Jz/4kT\right)} = \frac{-g\mu_B B/4}{k\left(T - \frac{Jz}{4k}\right)} = \frac{-g\mu_B B/4}{k(T - T_C)}$$

Hence the susceptibility will be

$$\chi = \mu_0 \frac{\partial M}{\partial B} = \mu_0 \frac{\partial}{\partial B}\left(-g\mu_B n \langle s \rangle\right)$$

$$= -g\mu_B \mu_0 n \frac{\partial \langle s \rangle}{\partial B}$$

$$\Rightarrow \chi = \frac{\frac{1}{4} g^2 \mu_B^2 \mu_0 n}{k(T - T_C)}$$

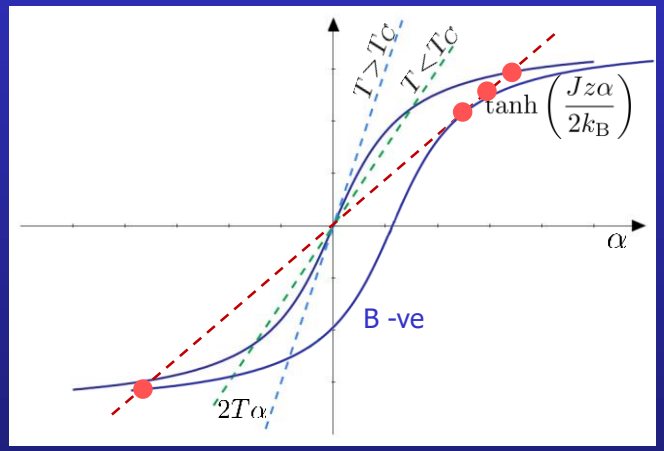
paramagnetic susceptibility

CURIE-WEISS LAW

Ferromagnetism Ising model: coercive field

FERROMAGNETISM

- solve self-consistency equation – eg graphically for $B = 0$:
  - set $\alpha \equiv \langle S \rangle / T$



- magnetization without presence of external field if

$$T < T_C \approx \frac{Jz}{4k_B}$$

CURIE TEMPERATURE

- exchange interactions between magnetic atoms in lattice

Write $\alpha = \frac{M}{ng\mu_B T}$

$\Rightarrow 2\alpha T = \frac{2}{ng\mu_B} M = \tanh\left(\frac{Jz}{2k}\alpha + \frac{g\mu_B B}{2kT}\right)$

$F_1(\alpha) = F_2(\alpha + aB)$

For spin-$\frac{1}{2}$ Ising model, we follow the Curie-Langevin calculation

$\Rightarrow \langle S \rangle = -\frac{1}{2}\tanh\frac{g\mu_B B_{eff}}{2kT}$

where if B_{ij} is constant for z nearest neighbours,

$g\mu_B B_{eff} = g\mu_B B - Jz\langle S \rangle$

$\Rightarrow \langle S \rangle = -\frac{1}{2}\tanh\frac{g\mu_B B - Jz\langle S \rangle}{2kT}$

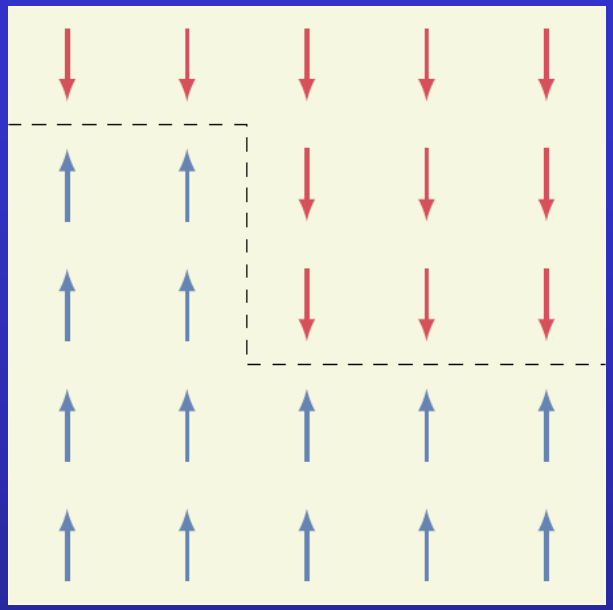
The magnetization is then

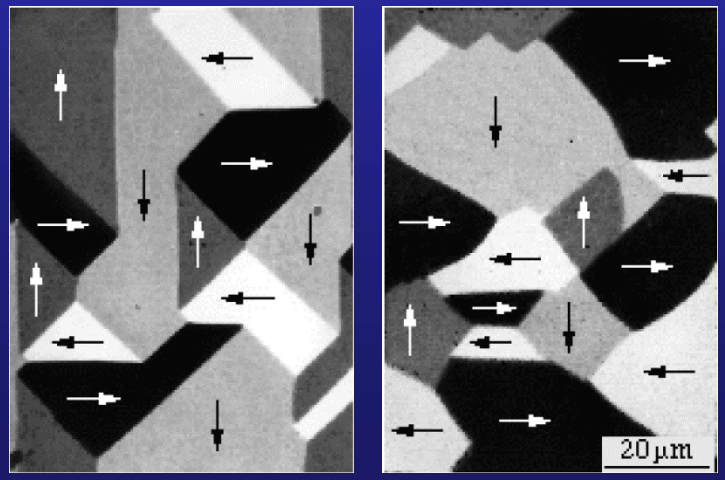
$M = -ng\mu_B \langle S \rangle$

Ferromagnetism: domains

FERROMAGNETISM

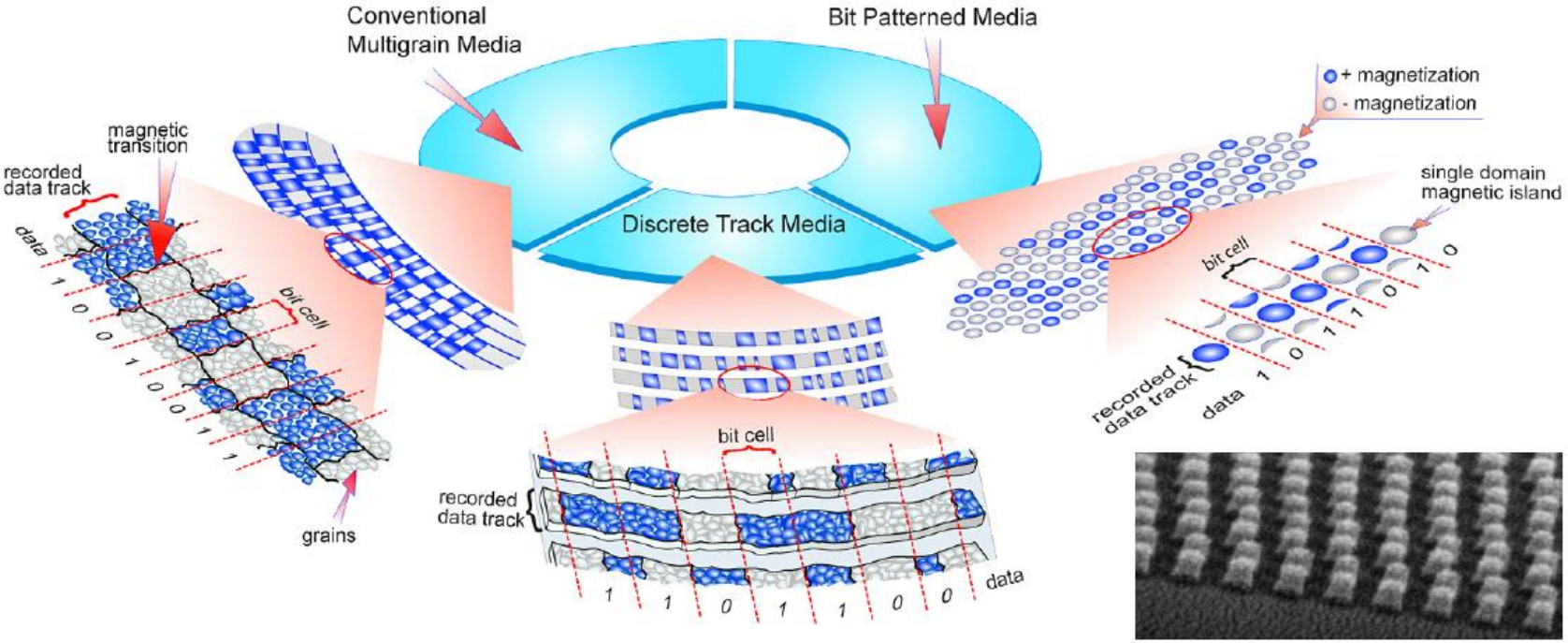
- aligned magnetic moments create a B-field outside the material
- energy of B-field scales with volume
- domains reduce overall magnetic moment
- domains energetically favoured when dimensions >~100 nm
- domain boundaries can be 'pinned' to dislocations and defects





Si-Fe magnetic domains

Magnetic data storage



- Continuous granular recording layer
- Multiple grains per bit
- Boundaries between bits determined by grains
- Thermal stability unit: 1 grain (~ 6 nm)
- Reaching its limits

- PMR media, with patterned tracks.
- Multiple grains per bit
- Eliminates track edge noise
- Thermal stability unit: 1 grain (~ 6 nm)
- Modest areal density gain.

- Each island is a single domain
- Bit locations determined by lithography
- Thermal stability unit: 1 island (~15 nm)
- Substantial areal density gain should be feasible

Suggested workplan

- work through derivations of exchange, paramagnetic and diamagnetic Hamiltonians
- work through derivations of Curie's law and Curie temperature
- read about magnetic domains and hysteresis

Ashcroft & Mermin	ch 31-33
Blundell & Blundell	sect 17.3-4
Girvin & Yang	sects 17.1-17.5
Hook & Hall	ch 7,8
Ibach & Lüth	ch 8
Kittel	ch 11,12
Myers	ch 11
Rosenberg	ch 11,12
Simon	ch 19-22
Snoke	sects 17.1-17.4

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Crystalline Solids

PHYS3004

Tim Freegarde

