Magnetic order

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Part 2

1

- Interaction between pair of spins motivates the general form of the Heisenberg model:

$$H = -\sum_{ij} J_{ij}\mathbf{S_i}\cdot\mathbf{S_j}$$

- *Direct exchange*: important in many metals such as Fe, Co and Ni
- *Indirect exchange*: mediated through the conduction electrons (RKKY) - **metals**
- *Superexchange*: exchange interaction mediated by oxygen. This leads to a very long exchange path. Important in many magnetic oxides, e.g. MnO, La_2CuO_4. – **non-conducting oxides**

2

DE MAGNETE (1601)

"Thus do pretenders to science vainly and preposterously seek for remedies, ignorant of the true causes of things."

Lucas Gauricus thought that the lodestone belongs to the sign Virgo "and with a veil of mathematical erudition does he cover many similar disgraceful stupidities".

William Gilbert
1544-1603

Niels Bohr
(1885-1962)

Hendrika Johanna van Leeuwen
(1887-1974)

Bohr-van Leeuwen theorem

$$\mathbf{p} = m\mathbf{v} + q\mathbf{A}$$

$$\Longrightarrow Z = \int\int\cdots\int \exp(-\beta E(\{\mathbf{r}_i,\mathbf{p}_i\}))\, \mathrm{d}\mathbf{r}_1\cdots\mathrm{d}\mathbf{r}_N\, \mathrm{d}\mathbf{p}_1\cdots\mathrm{d}\mathbf{p}_N$$

In a magnetic field, we replace $\mathbf{p}_i$ by $\mathbf{p}_i - q\mathbf{A}$

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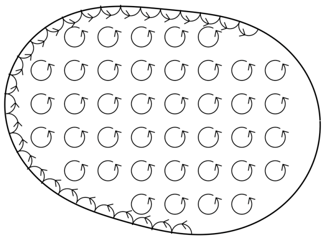
In a magnetic field, we replace $\mathbf{p}_i$ by $\mathbf{p}_i - q\mathbf{A}$

$$F = -k_{\mathrm{B}}T\log Z$$

$$\mathbf{M} = -\left(\frac{\partial F}{\partial B}\right)_T$$



Outline

Magnetic order

1. Paul spin operators
2. Two spins, three spins, four spins
3. Types of magnetic order

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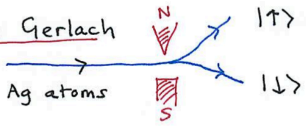
Stern Gerlach



2-state system

look for Hermitian operator as 2×2 matrix

$M = \begin{pmatrix} a & c-id \\ c+id & b \end{pmatrix}$ $a, b, c, d \in \mathbb{R}$

with eigenvalues ± 1.

$\therefore$ $\mathrm{Tr}\, M = \lambda_+ + \lambda_- = 0$ $\therefore$ $b = -a$

$\det M = \lambda_+ \lambda_- = -1$ $a^2 + c^2 + d^2 = 1$

$M = \begin{pmatrix} a & c-id \\ c+id & -a \end{pmatrix}$

Simplest $M = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$. Call this σ_z.

Eigenvectors $\begin{pmatrix} 1 \\ 0 \end{pmatrix}$ eigenvalue $+1$

$\begin{pmatrix} 0 \\ 1 \end{pmatrix}$ eigenvalue -1

General state: $|\psi\rangle = \alpha|\uparrow\rangle + \beta|\downarrow\rangle \equiv \begin{pmatrix} \alpha \\ \beta \end{pmatrix}$ ← spinor

Note $\langle\psi|\sigma_z|\psi\rangle = (\alpha^* \ \beta^*)\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}\begin{pmatrix} \alpha \\ \beta \end{pmatrix}$

$= |\alpha|^2 - |\beta|^2$

$$\hat{S}_z = \frac{\hbar}{2}\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.$$

Look for σ_x and σ_y. To be $\perp z$, need

$\langle\uparrow_z|\sigma_\perp|\uparrow_z\rangle = 0$
$\langle\downarrow_z|\sigma_\perp|\downarrow_z\rangle = 0$ $\Big\} \Rightarrow a = 0$

$\therefore$ $\sigma_\perp = \begin{pmatrix} 0 & c-id \\ c+id & 0 \end{pmatrix}$ with $c^2 + d^2 = 1$ i.e. $c + id = e^{i\phi}$

$\sigma_\perp = \begin{pmatrix} 0 & e^{-i\phi} \\ e^{i\phi} & 0 \end{pmatrix}$.

Choose $\phi = 0$ $\therefore$ $\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$. eigenvectors $\frac{1}{\sqrt{2}}\begin{pmatrix} 1 \\ 1 \end{pmatrix}, \frac{1}{\sqrt{2}}\begin{pmatrix} 1 \\ -1 \end{pmatrix}$

$$\hat{S}_x = \frac{\hbar}{2}\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \qquad \hat{S}_z = \frac{\hbar}{2}\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.$$

Construct σ_y from $\langle\uparrow_x|\sigma_y|\uparrow_x\rangle = 0$

$\therefore$ $\frac{1}{\sqrt{2}}(1 \ \ 1)\begin{pmatrix} 0 & e^{-i\phi} \\ e^{i\phi} & 0 \end{pmatrix}\frac{1}{\sqrt{2}}\begin{pmatrix} 1 \\ 1 \end{pmatrix} = 0$

$\therefore$ $\cos\phi = 0$ $\therefore$ $\phi = \frac{\pi}{2}$ $\sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$

eigenvectors $\frac{1}{\sqrt{2}}\begin{pmatrix} 1 \\ i \end{pmatrix}, \frac{1}{\sqrt{2}}\begin{pmatrix} 1 \\ -i \end{pmatrix}$.

$$\hat{\boldsymbol{S}} = \frac{\hbar}{2}\hat{\boldsymbol{\sigma}}$$

$$\hat{S}_x = \frac{\hbar}{2}\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \qquad \hat{S}_y = \frac{\hbar}{2}\begin{pmatrix} 0 & -\mathrm{i} \\ \mathrm{i} & 0 \end{pmatrix}, \qquad \hat{S}_z = \frac{\hbar}{2}\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.$$

- Spin operators: $\vec{S} = \frac{1}{2}\hbar\,\vec{\sigma}$
- Raising and lowering operators: $\sigma_\pm = \sigma_x \pm i\sigma_y$

$\sigma_+ = \begin{pmatrix} 0 & 2 \\ 0 & 0 \end{pmatrix}, \quad \sigma_- = \begin{pmatrix} 0 & 0 \\ 2 & 0 \end{pmatrix}.$

$$\hat{S}_z \,|S\, m_s\rangle = m_s \hbar\, |S\, m_s\rangle$$
$$\hat{S}_\pm \,|S\, m_s\rangle = \hbar \sqrt{S(S+1) - m_s(m_s \pm 1)}\; |S\, m_s \pm 1\rangle$$
$$\hat{S}^2 \,|S\, m_s\rangle = S(S+1)\hbar^2\, |S\, m_s\rangle$$

$$\vec{S}^a \cdot \vec{S}^b = S_x^a S_x^b + S_y^a S_y^b + S_z^a S_z^b$$

re-express in terms of raising and lowering operators:

$$\frac{1}{2}\left(S_+^a S_-^b + S_-^a S_+^b\right)$$

⇒ FLIP-FLOPS of antiparallel spins

$\uparrow_a \downarrow_b \leftrightarrow \downarrow_a \uparrow_b$

↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓
↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑
↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓ antiferromagnet
↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑
↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓↑↓

↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑
↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑
↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑ ferromagnet
↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑
↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑

17

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18

$\hat{\mathcal{H}} = A\hat{\mathbf{S}}^a \cdot \hat{\mathbf{S}}^b$

Two spins

↑↑ ferromagnetic

↓↓ ferromagnetic

↑↓ antiferromagnetic

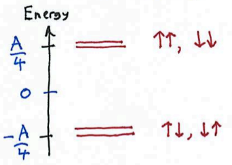
↓↑ antiferromagnetic

If $A > 0$, the antiferromagnetic state has lowest energy

19

Simple (wrong) picture

$$H = A\,\vec{S}^a \cdot \vec{S}^b$$
$$= A\,S_z^a S_z^b$$



$$\frac{1}{2}+\frac{1}{2}=0,1$$

$$\frac{1}{2}+\frac{1}{2}=0,1$$

$$D^{(\frac{1}{2})}\otimes D^{(\frac{1}{2})}=D^{(0)}\oplus D^{(1)}$$

2^2 = 1 + 3

singlet triplet

$\hat{\mathcal{H}} = A\hat{\mathbf{S}}^a \cdot \hat{\mathbf{S}}^b$

Two spins

$\uparrow\uparrow$ ferromagnetic

$\downarrow\downarrow$ ferromagnetic

$\uparrow\downarrow$ antiferromagnetic

$\downarrow\uparrow$ antiferromagnetic

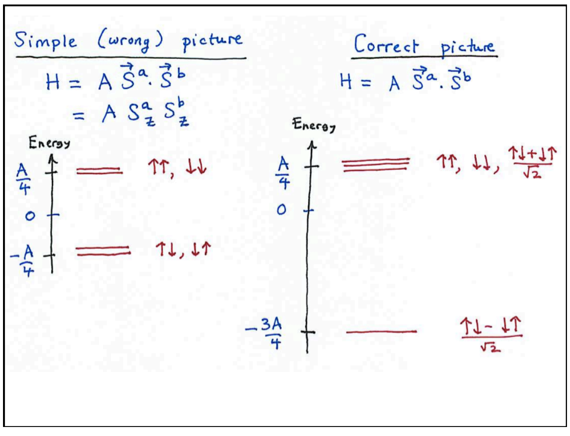
If $A > 0$, the antiferromagnetic state has lowest energy

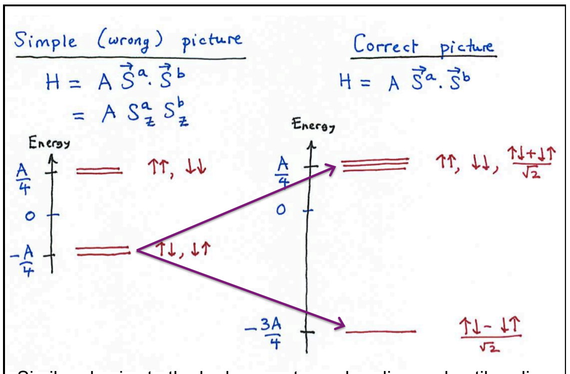
$\hat{\mathcal{H}} = A\hat{\mathbf{S}}^a \cdot \hat{\mathbf{S}}^b$ $\qquad \hat{\mathbf{S}}^{\text{tot}} = \hat{\mathbf{S}}^a + \hat{\mathbf{S}}^b$

$\Longrightarrow (\hat{\mathbf{S}}^{\text{tot}})^2 = (\hat{\mathbf{S}}^a)^2 + (\hat{\mathbf{S}}^b)^2 + 2\hat{\mathbf{S}}^a \cdot \hat{\mathbf{S}}^b$

$$\hat{\mathbf{S}}^a \cdot \hat{\mathbf{S}}^b = \begin{cases} \frac{1}{4} & \text{if} \quad s=1 \\ -\frac{3}{4} & \text{if} \quad s=0. \end{cases}$$

	Eigenstate	m_s	s	$\hat{\mathbf{S}}^a \cdot \hat{\mathbf{S}}^b$
Triplet: $E = A/4$	$\lvert\uparrow\uparrow\rangle$	1	1	$\frac{1}{4}$
	$\frac{\lvert\uparrow\downarrow\rangle+\lvert\downarrow\uparrow\rangle}{\sqrt{2}}$	0	1	$\frac{1}{4}$
	$\lvert\downarrow\downarrow\rangle$	-1	1	$\frac{1}{4}$
Singlet: $E = -3A/4$	$\frac{\lvert\uparrow\downarrow\rangle-\lvert\downarrow\uparrow\rangle}{\sqrt{2}}$	0	0	$-\frac{3}{4}$





Similar physics to the hydrogen atom – bonding and antibonding orbitals, with one *saving* energy and the other *costing* energy.

Spin waves

$$\mathcal{H} = -2\mathcal{J}\sum_i \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+1}$$

(Heisenberg ferromagnet J>0)

27

Spin waves

$$\mathcal{H} = -2\mathcal{J}\sum_i \left[\hat{S}_i^x \hat{S}_{i+1}^x + \hat{S}_i^y \hat{S}_{i+1}^y + \hat{S}_i^z \hat{S}_{i+1}^z\right]$$

(Heisenberg ferromagnet J>0)

28

Spin waves

$$\hat{\mathcal{H}} = -2\mathcal{J}\sum_i \left[\hat{S}_i^z \hat{S}_{i+1}^z + \frac{1}{2}(\hat{S}_i^+ \hat{S}_{i+1}^- + \hat{S}_i^- \hat{S}_{i+1}^+)\right]$$

(Heisenberg ferromagnet J>0)

29

Spin waves

$$\hat{\mathcal{H}} = -2\mathcal{J}\sum_i \left[\hat{S}_i^z \hat{S}_{i+1}^z + \frac{1}{2}(\hat{S}_i^+ \hat{S}_{i+1}^- + \hat{S}_i^- \hat{S}_{i+1}^+)\right]$$

Ground state $|\Phi\rangle$ ↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑

(Heisenberg ferromagnet J>0)

$$\hat{\mathcal{H}}|\Phi\rangle = -2NS^2\mathcal{J}|\Phi\rangle$$

30

Spin waves

$$\hat{\mathcal{H}} = -2\mathcal{J}\sum_i \left[\hat{S}_i^z \hat{S}_{i+1}^z + \frac{1}{2}(\hat{S}_i^+ \hat{S}_{i+1}^- + \hat{S}_i^- \hat{S}_{i+1}^+)\right]$$

Flipped spin $|j\rangle$ ↑↑↑↑↑↑↑↓↑↑↑↑↑↑↑↑↑↑↑↑

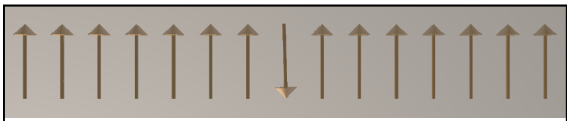
$$|j\rangle = \hat{S}_j^-|\Phi\rangle$$

$$\hat{\mathcal{H}}|j\rangle = 2\left[(-NS^2\mathcal{J} + 2S\mathcal{J})|j\rangle - S\mathcal{J}|j+1\rangle - S\mathcal{J}|j-1\rangle\right]$$

31

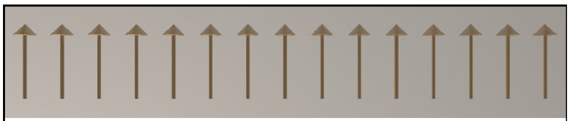
- Start with a ferromagnetic chain of spins

32

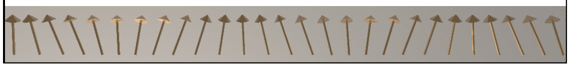


- Start with a ferromagnetic chain of spins
- Then reverse one spin

33



- Start with a ferromagnetic chain of spins
- Then reverse one spin
- The spins are exchange coupled and so the excitation becomes quickly delocalised on a time scale which scales with ħ/J



34

Spin waves

$$\hat{\mathcal{H}} = -2\mathcal{J}\sum_i \left[\hat{S}_i^z \hat{S}_{i+1}^z + \frac{1}{2}(\hat{S}_i^+ \hat{S}_{i+1}^- + \hat{S}_i^- \hat{S}_{i+1}^+)\right]$$

Flipped spin $|j\rangle$ ↑↑↑↑↑↑↑↓↑↑↑↑↑↑↑↑↑↑↑↑

$$|j\rangle = \hat{S}_j^- |\Phi\rangle$$

$$\hat{\mathcal{H}}|j\rangle = 2\left[(-NS^2\mathcal{J} + 2S\mathcal{J})|j\rangle - S\mathcal{J}|j+1\rangle - S\mathcal{J}|j-1\rangle\right]$$

35

Spin waves

$$\hat{\mathcal{H}} = -2\mathcal{J}\sum_i \left[\hat{S}_i^z \hat{S}_{i+1}^z + \frac{1}{2}(\hat{S}_i^+ \hat{S}_{i+1}^- + \hat{S}_i^- \hat{S}_{i+1}^+)\right]$$

Flipped spin $|j\rangle$ ↑↑↑↑↑↑↑↓↑↑↑↑↑↑↑↑↑↑↑↑

$$|j\rangle = \hat{S}_j^- |\Phi\rangle$$

$$\hat{\mathcal{H}}|j\rangle = 2\left[(-NS^2\mathcal{J} + 2S\mathcal{J})|j\rangle - S\mathcal{J}|j+1\rangle - S\mathcal{J}|j-1\rangle\right]$$

q-state $|q\rangle$ ↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑↑

smeared out single flipped spin

$$|q\rangle = \frac{1}{\sqrt{N}}\sum_j e^{iqR_j}|j\rangle$$

$$\hat{\mathcal{H}}|q\rangle = E(q)|q\rangle$$

$$E(q) = -2NS^2\mathcal{J} + 4\mathcal{J}S(1-\cos qa)$$

36

Number of thermally excited spin waves:

$$n_{\text{magnon}} = \int_0^\infty \frac{g(\omega)\,d\omega}{e^{\hbar\omega/k_B T} - 1}$$

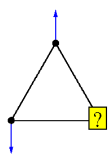
In three dimensions: $g(\omega) \propto \omega^{1/2}$

$$n_{\text{magnon}} \propto T^{3/2}$$

In one or two dimensions, the integral diverges.
Magnetism not stable in Heisenberg model in 1D or 2D
(Mermin-Wagner-Berenzinskii theorem,
due to also to Coleman)

37

Triangle of spins



AF interactions for n.n.

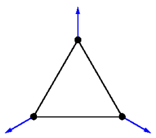
Magnetic frustration

38

Triangle of spins

Classical Heisenberg spins

120° state

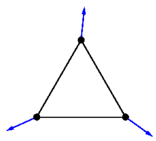


Two different chiralities

Triangle of spins

Classical Heisenberg spins

120° state



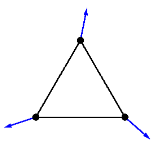
Two different chiralities

...and rotational symmetry

Triangle of spins

Classical Heisenberg spins

120° state



Two different chiralities

...and rotational symmetry

$$\frac{1}{2} + \frac{1}{2} = 0, 1$$

$$\frac{1}{2} + \frac{1}{2} + \frac{1}{2} = \frac{1}{2}, \frac{1}{2}, \frac{3}{2}$$

$$\frac{1}{2} + \frac{1}{2} = 0, 1$$

$$D^{(\frac{1}{2})} \otimes D^{(\frac{1}{2})} = D^{(0)} \oplus D^{(1)}$$

2^2 = 1 + 3

$$\frac{1}{2} + \frac{1}{2} + \frac{1}{2} = \frac{1}{2}, \frac{1}{2}, \frac{3}{2}$$

$$D^{(\frac{1}{2})} \otimes D^{(\frac{1}{2})} \otimes D^{(\frac{1}{2})} = 2D^{(\frac{1}{2})} \oplus D^{(\frac{3}{2})}$$

2^3 = 2 + 2 + 4

Triangle of spins

$$\sum_{\langle i,j \rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j = \frac{1}{2} \left(S_{\rm tot}(S_{\rm tot}+1) - 3s(s+1) \right)$$

$$S_{\rm tot} = \frac{3}{2} \implies \sum_{\langle i,j \rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j = +\frac{3}{4}$$ quartet

$$S_{\rm tot} = \frac{1}{2} \implies \sum_{\langle i,j \rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j = -\frac{3}{4}$$ 2 doublets

Triangle of spins

$$|\Psi_{M=3/2}\rangle = |\uparrow\uparrow\uparrow\rangle$$

$$|\Psi^{(k)}_{M=1/2}\rangle = \frac{1}{\sqrt{3}}\sum_{j=0}^{2} e^{2\pi ijk/3} C_3^j |\downarrow\uparrow\uparrow\rangle$$

$$|\Psi^{(k)}_{M=-1/2}\rangle = \frac{1}{\sqrt{3}}\sum_{j=0}^{2} e^{2\pi ijk/3} C_3^j |\uparrow\downarrow\downarrow\rangle$$

$$|\Psi_{M=-3/2}\rangle = |\downarrow\downarrow\downarrow\rangle$$

45

Triangle of spins

$$|\Psi_{M=3/2}\rangle = |\uparrow\uparrow\uparrow\rangle \qquad s=\tfrac{3}{2} \quad C_z|\Psi_{M=3/2}\rangle = 0$$

$$|\Psi^{(k)}_{M=1/2}\rangle = \frac{1}{\sqrt{3}}\sum_{j=0}^{2} e^{2\pi ijk/3} C_3^j |\downarrow\uparrow\uparrow\rangle$$

$k=0$ $\quad s=\tfrac{3}{2}$ $\quad C_z|\Psi^{(0)}_{M=1/2}\rangle = 0$

$k=1$ $\quad s=\tfrac{1}{2}$ $\quad C_z|\Psi^{(1)}_{M=1/2}\rangle = |\Psi^{(1)}_{M=1/2}\rangle$

$k=2$ $\quad C_z|\Psi^{(2)}_{M=1/2}\rangle = -|\Psi^{(2)}_{M=1/2}\rangle$

Chirality $\quad C_z = \frac{1}{4\sqrt{3}}\mathbf{S}_1\cdot(\mathbf{S}_2\times\mathbf{S}_3)$

see M. Trif et al. PRB **82**, 045429 (2010)

46

Tetrahedron of spins

$$D^{(1/2)}\otimes D^{(1/2)}\otimes D^{(1/2)}\otimes D^{(1/2)} = 2D^{(0)}\oplus 3D^{(1)}\oplus D^{(2)}$$

$$\sum_{\langle i,j\rangle}\hat{\mathbf{S}}_i\cdot\hat{\mathbf{S}}_j = \frac{1}{2}\left(S_{\rm tot}(S_{\rm tot}+1) - 4s(s+1)\right)$$

$$S_{\rm tot}=2 \implies \sum_{\langle i,j\rangle}\hat{\mathbf{S}}_i\cdot\hat{\mathbf{S}}_j = +\frac{3}{2}$$ pentet

$$S_{\rm tot}=1 \implies \sum_{\langle i,j\rangle}\hat{\mathbf{S}}_i\cdot\hat{\mathbf{S}}_j = -\frac{1}{2}$$ 3 triplets

$$S_{\rm tot}=0 \implies \sum_{\langle i,j\rangle}\hat{\mathbf{S}}_i\cdot\hat{\mathbf{S}}_j = -\frac{3}{2}$$ 2 singlets

47

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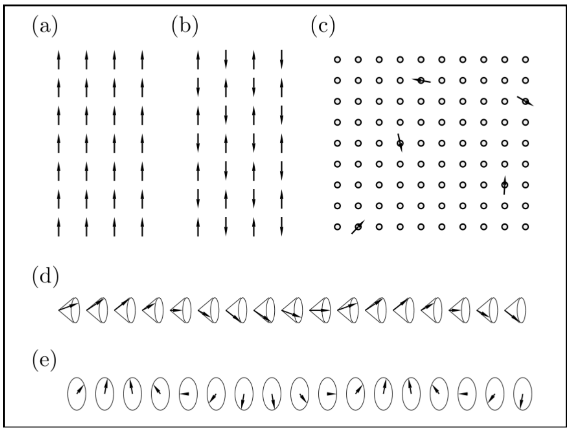
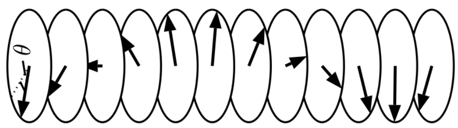
48





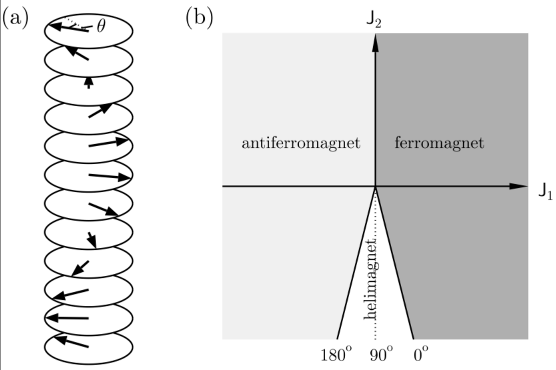
$E = -2NS^2(J_1\cos\theta + J_2\cos 2\theta)$

Minimize energy $\quad \frac{\partial E}{\partial\theta} = 0$

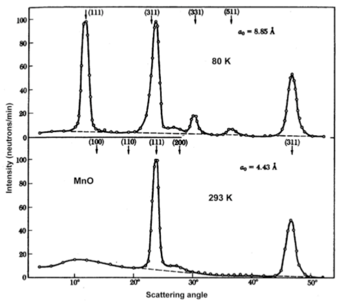
$\therefore\ (J_1 + 4J_2\cos\theta)\sin\theta = 0$

Solutions: $\theta = 0,\ \pi,\ \cos^{-1}\left(\frac{-J_1}{4J_2}\right)$

FM $\quad$ AFM $\quad$ helimagnetism



Magnetic neutron diffraction



MnO

C.G. Shull et al. (1951)

see also

A.L. Goodwin et al.
PRL **96,** 047209 (2006)

52

Heisenberg model:

$$H = -\sum_{ij} J_{ij} \mathbf{S_i} \cdot \mathbf{S_j}$$

The dot product only respects the **relative orientation** of spins, not the **absolute orientation** with respect to the lattice.

In the next lecture: effect of the **lattice**

- *Orbitals and the crystal field*
- *Magnetocrystalline anisotropy*
- *Magnetostriction and magnetoelasticity*

53