

# Chapter 8: Magnetism

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# 1 Introduction

Magnetism is one of the most interesting subjects in condensed matter physics. Magnetic effects are responsible for heavy fermion behavior, ferromagnetism, antiferromagnetism, ferrimagnetism and probably high temperature superconductivity.

Unlike our previous studies, most magnetic systems are not well described by simple models which ignore intersite correlations. The reason is simple: magnetism is inherently due to electronic correlations of moments on different sites. As we

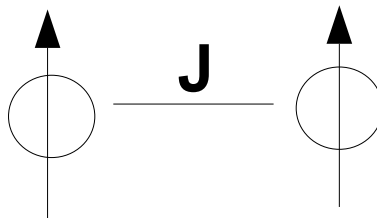


Figure 1: *Both moment formulation and the correlation between these moments ( $J$ ) are due to Coulombic effects*

will see, systems without these inter-spin correlations (or those without well defined moments to be correlated) have uninteresting and unimportant (energetically) magnetic properties.

## 1.1 The Relevance of Magnetostatics

Perhaps the term magnetism is a misnomer, or rather describes only the external probe ( $B$ ) which we use to study magnetic behavior. Magnetic effects are mainly due to electronic correlations, those mediated or due to Coulombic effects, and not due to magnetic correlations between moments (these are smaller by orders of  $v/c$ , ie., they are relativistic corrections). For example, consider the magnetic correlation between two moments separated by a couple of Angstroms.

$$U = \frac{1}{r^3}[\mathbf{m}_1 \cdot \mathbf{m}_2 - 3(\mathbf{m}_1 \cdot \mathbf{n})(\mathbf{m}_2 \cdot \mathbf{n})] \quad (1)$$

then,

$$U_{dipole-dipole} \approx \frac{m_1 m_2}{r^3} \quad (2)$$

If we let,

$$m_1 \sim m_2 \sim g\mu_B \sim \frac{e\hbar}{m}$$
$$r \approx 2 \text{ \AA}$$

then,

$$U \sim \frac{(g\mu_B)^2}{r^3} \sim \left(\frac{e^2}{\hbar c}\right)^2 \left(\frac{a_0}{r}\right)^3 \frac{e^2}{a_0} \sim 10^{-4} eV \quad (3)$$

Or roughly one degree Kelvin! Magnetic correlations due to magnetic interactions would be destroyed by thermal fluctuations at very low temperatures.

## 1.2 Non-interacting Magnetic Systems

We will define non-interacting magnetic systems as those for which the independent moments do not interact with each other, and only interact with the probing field. For the moment let's consider only the magnetic moments due to electrons (as we will see, since they can interact with each other, they are by far the most important moments in the system). They have a moment

$$\approx \mu_B(\mathbf{L} + 2\mathbf{S}) \quad (4)$$

The system energy will change by an amount

$$\Delta E \sim g\mu_B B(L + 2S) \sim \mu_B B, \quad \mu_B = \frac{e\hbar}{2m} \sim 5.8 \times 10^{-5} \frac{eV}{Te} \quad (5)$$

in an external field. The largest field which can regularly be produced in a lab is  $\sim 10Te$  ( $100Te$  or more can be produced

at LANL by blowing things up), thus

$$\Delta E \lesssim 10^{-3} eV \quad \text{or} \quad \sim k_B(10^0 K) \quad (6)$$

This is a very small energy. Thus magnetic effects are wiped out by thermal fluctuation for

$$k_B T > \mu_B B \quad (7)$$

at about 10 K! Thus experiments which measure the magnetism of non-interacting systems must be carried out at low temperatures. These experiments typically measure the susceptibility of the system with a Faraday balance or a magnetometer (SQUID).

For a collection of isolated moments (spin 1/2), the susceptibility may be calculated from the moment

$$\uparrow s = \frac{1}{2} \quad \langle m \rangle = g\mu_B \frac{1}{2} \frac{(e^{-\beta \frac{1}{2} g \mu_B B} - e^{\beta \frac{1}{2} g \mu_B B})}{(e^{-\beta \frac{1}{2} g \mu_B B} + e^{\beta \frac{1}{2} g \mu_B B})} \quad (8)$$

$$\simeq g \frac{\mu_B}{2} \tanh(\beta \mu_B B) \approx \mu_B \frac{\mu_B B}{T} \quad (g \sim 2)$$

$$\chi = \frac{\partial \langle m \rangle}{\partial B} \approx \frac{\mu_B^2}{k_B T} \quad (9)$$

Once again, the energy of the moment-field interaction is roughly

$$E \sim \frac{\mu_B^2}{T} B^2 \sim \frac{10^{-8} \left(\frac{eV}{T_e}\right)^2}{T \left(\frac{10^{-4} eV}{^\circ K}\right)^2} B^2, \quad (k = 1) \quad (10)$$

When  $E \sim T$ , thermal fluctuations destroy the orientation of the moments with the external field. If  $B \sim 10T_e$

$$E \sim \frac{100^\circ}{T} K \quad (11)$$

or  $E \sim T$  at  $10^\circ K$ ! However, we know that systems such as

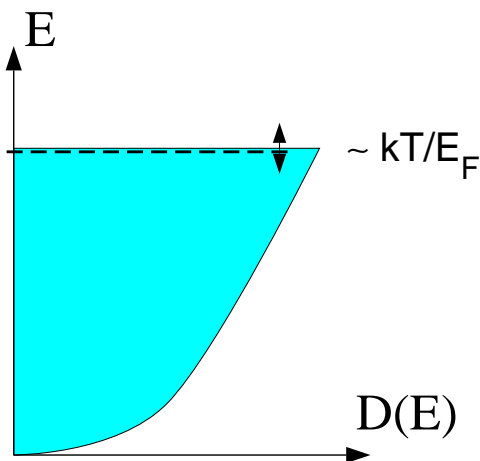


Figure 2: *In a metal, only the electrons near the Fermi surface, which are not paired into singlets, contribute to the bulk susceptibility  $\chi \sim \frac{kT}{E_F} \frac{\mu_B^2}{k_B T} \sim \mu_B^2 D(E_F)$*

iron exist for which a small field can induce a relatively large moment at room temperature. This is surprising since for a metal, or a free electron gas, the susceptibility is much smaller

than the free electron result, since only the spins near the Fermi surface can participate,  $\chi = \frac{\mu_B^2}{E_F}$ . Note that this is *even smaller* than the free electron result by a factor of  $\frac{k_B T}{E_F} \ll 1!$

## 2 Coulombic Correlation Effects

### 2.1 Moment Formation

Of course real materials are not composed of free isolated electrons. Nevertheless some insulators act almost as if they are composed of non-interacting atoms (ions) with moments given by Hund's rules: maximum  $S$  maximum  $L$  which leads to large atomic moments. Hund's rules reflect the atom's attempt to lower its Coulombic energy, see Fig. 3. By maximizing the total spin  $S$ , the spin part of the wavefunction becomes symmetric under electron exchange (i.e. for two electrons with  $s = \frac{1}{2}$   $\uparrow \downarrow$  the maximum value of total spin is  $S = 1$  with a wavefunction  $|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle$ ). Then, since the total wavefunction must be antisymmetric under exchange, the spatial part must be antisymmetric requiring it to have a node. The node keeps the



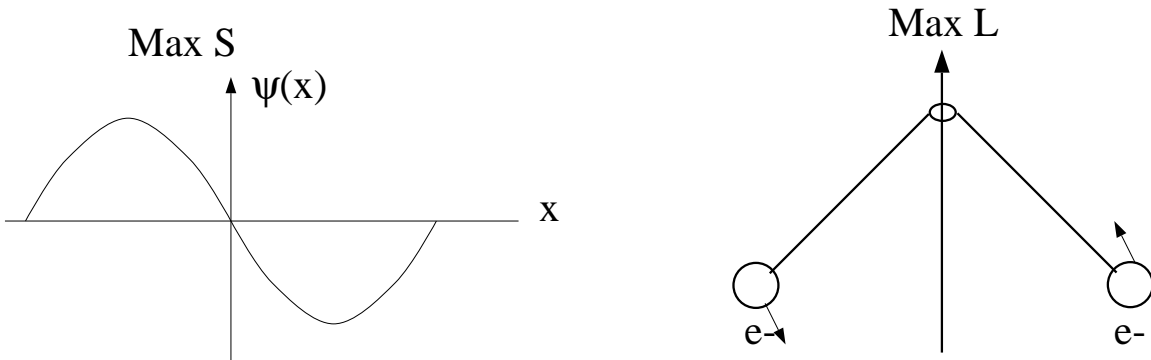


Figure 3: *Hunds rules, Maximize  $S$  and  $L$ , both result from minimizing the Coulomb energy.*

electrons apart, minimizing their Coulomb energy. The second Hund's rule is also due to Coulombic interactions. Maximizing  $L$  tends to keep the electrons apart, much like a centrifuge. (alternatively, the radial Schroedinger equation obtains an angular momentum barrier  $L(L + 1)/r^2$ ).

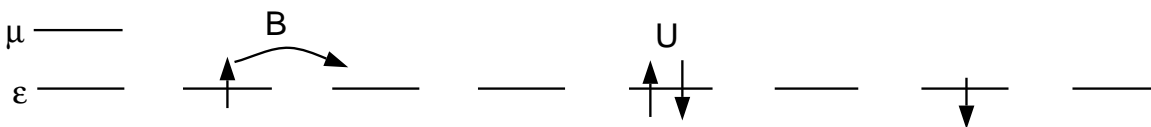


Figure 4: *A simple tight-binding model with a local Coulomb repulsion  $U$ . If  $U = 0$ , the rate that electrons hop on and off any site may be approximated using Fermi's golden rule  $\sim \pi|B|^2D(E_F) \sim \frac{1}{\Delta t}$ . Then by the uncertainty principal  $\Delta E\Delta t \sim \hbar$  so each site energy acquires an uncertainty or width  $\Delta E \sim \frac{\hbar}{\Delta t} \sim \pi|B|^2D(E_F) \equiv \Gamma$ . The sites will form moments (see Fig. 5) if  $\Gamma \gg U, |\epsilon|$*

To illustrate how band formation modifies this scenerio, lets

consider a simple tight binding model (See Fig. 4). By Fermi's golden rule, each level acquires a width (uncertainty in its energy)  $\Gamma = \pi B^2 D(E_F)$  and each level can be in one of the four states shown in Fig. 5. Clearly, the states  $\text{---}\bigcirc\text{---}$  and  $\text{---}\uparrow\downarrow\text{---}$

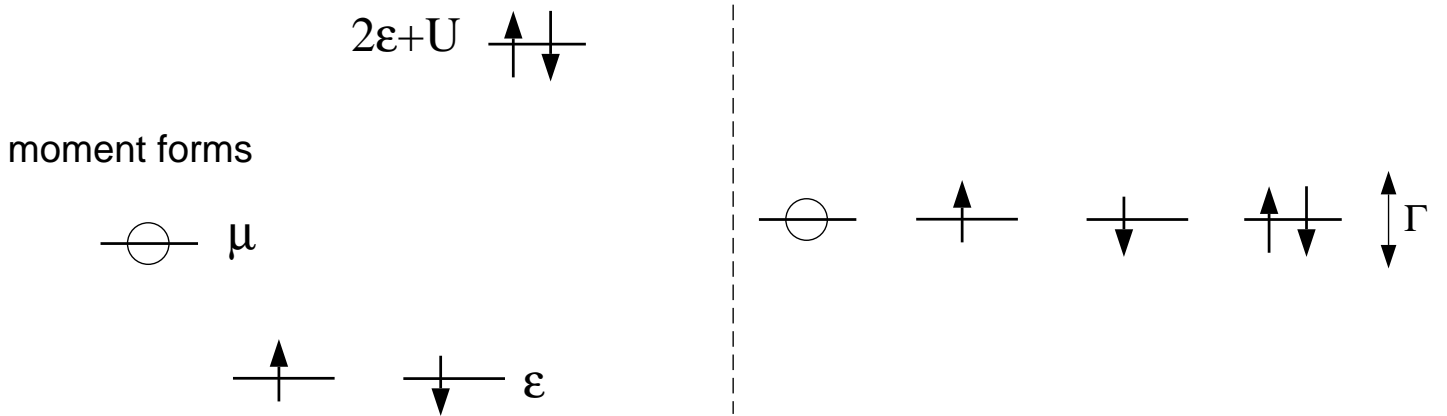


Figure 5: A moment forms on an orbital provided that  $\Gamma \gg U, |\epsilon|$ .  $U \sim \frac{e^2}{a} e^{-\frac{a}{r_{TF}}}$   $r_{TF}$  is small for a metal, large for an insulator

do not have a moment, and the states  $\text{---}\uparrow\text{---}$  and  $\text{---}\downarrow\text{---}$  do. If these states mix equally a moment will not form. The mixing between the states with moments is only through one of the other two states ( $\text{---}\bigcirc\text{---}$  or  $\text{---}\uparrow\downarrow\text{---}$ ) and may be suppressed, as can the occupancy of the moment-less states, by increasing the energy of the states without moments. Ie., a moment will form on each site if  $-\epsilon \gg \Gamma$  and  $\epsilon + U \gg \Gamma$ .

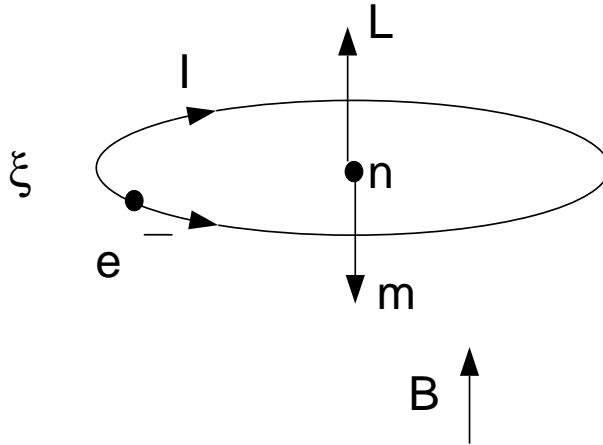


Figure 6: Here  $\xi = -\frac{1}{c} \frac{\partial \phi}{\partial t}$ ,  $m = -\mu_B L$  and  $\phi = B\pi a^2$

In this limit  $U \gg B$ , the system will act more like a system of free moments than a free electron gas. Thus, one might expect

$$\chi_{insulator} \gg \chi_{metal} \quad (12)$$

for noninteracting systems. However, this is not the case. The reason is that I have only told you half of the story. A real atom, or a system composed of such atoms, has a diamagnetic response due to the angular momentum  $\mathbf{L}$  of the rotating electrons. This effect is due to Lenz' Law. So that any introduced magnetic induction will induce an EMF and hence a current that opposes the electron current which reduces the moment.

$\Rightarrow$  diamagnetism with  $\chi \propto a^2$ . In the free electron limit (see J.M. Ziman, )

$$\chi = \mu_B^2 D(E_F) \left[ 1 - \frac{1}{3} \left( \frac{m}{m^*} \right)^2 \right] \quad (13)$$

$$\Delta E \sim 10^{-8} \left( \frac{eV}{T} \right)^2 a 10^{-1} eV^{-1} B^2 \sim 10^{-9} eV B^2 (T) \quad (14)$$

For insulators, often with  $\frac{m^*}{m} < 1$  the diamagnetism wins; whereas for metals, generally with  $\frac{m^*}{m} \geq 1$ , the Pauli paramagnetism wins.

## 2.2 Magnetism and Intersite Correlations

From both Hund's rules and a simple tight binding picture, we argued that moment formation in solids results from local Coulomb correlations between electrons. We also saw that a collection of such isolated moments is rather boring since all magnetic behavior is washed out by thermal fluctuations at very low temperatures. Consider once again an isolated moment of magnitude  $m\mu_B$  in an external field.

$$\chi \approx \frac{(m\mu_B)^2}{k_B T} \quad (15)$$

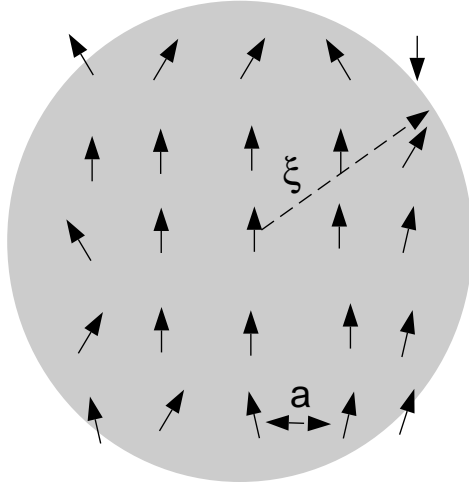


Figure 7: *If the magnetic moments in a small volume are correlated, then the magnetic susceptibility is strongly enhanced.*

$$E \approx \frac{(m\mu_B B)^2}{k_B T} \quad (16)$$

For magnetism to be significant at room temperature ( $300K$ ) we must increase the energy of our system in a field. This may be accomplished by increasing the effective moment  $m$  by correlating adjacent moments. If the range of this correlation is  $\xi$ , so that roughly  $\frac{4\pi\xi^3}{3a^3}$  moments are correlated, then let  $\frac{\xi}{a} = 3$  so that  $\sim 10^2$  moments are correlated and  $m \sim 10^2$ . This increases  $E$  by about  $10^4$ , so that  $E \sim k_b T$  at  $T \sim 10^3 K$ . The observed (measured) susceptibility also then increases by about  $10^4$ , all by only correlating moments in a range of 3 lattice

spacings.

Clearly correlations between adjacent spins can make magnetism in materials relevant. Such correlations are due to *electronic* effects and are hence usually short ranged due to electronic (Thomas-Fermi) screening. If we consider two  $s = \frac{1}{2}$  spins,  $\uparrow_1 \downarrow_2$ , then the correlation is usually parameterized by the Heisenberg exchange Hamiltonian, or

$$H = -2J\sigma_1 \cdot \sigma_2 \quad (17)$$

where  $J$  is the exchange splitting between the singlet and triplet energies.

$$\left\{ \begin{array}{l} |\uparrow \uparrow\rangle \\ |\uparrow \uparrow\rangle + |\downarrow \uparrow\rangle \\ |\downarrow \downarrow\rangle \end{array} \right\} E_t \quad (18)$$

$$\{|\uparrow \downarrow\rangle - |\downarrow \uparrow\rangle\} E_s \quad (19)$$

$$E_t - E_s = -J \quad (20)$$

The trick then is to calculate  $J$ !

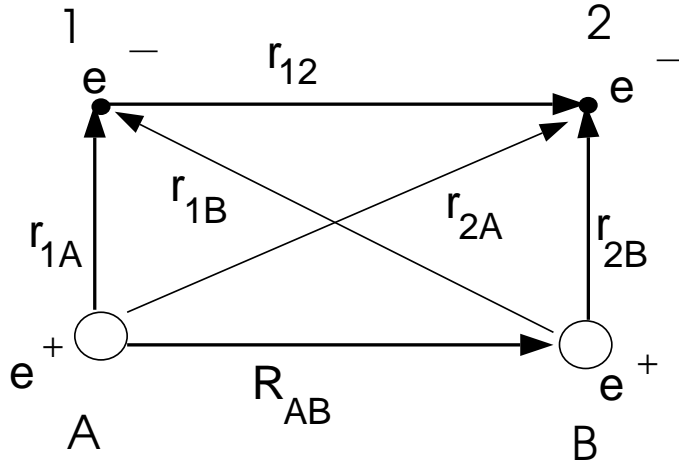


Figure 8: Geometry of two electrons, 1 and 2, bound to two ions A and B.

### 2.2.1 The Exchange Interaction Between Localized Spins

Imagine that we have two hydrogen atoms A and B which localize two electrons 1 and 2. As these two electrons approach, their spins will become correlated.

$$H = H_1 + H_2 + H_{12} \quad (21)$$

$$H_1 = -\frac{\hbar^2}{2m} \nabla^2 - \frac{e^2}{r_{1A}} - \frac{e^2}{r_{1B}} \quad (22)$$

$$H_{12} = \frac{e^2}{r_{12}} + \frac{e^2}{R_{AB}} \quad (23)$$

As we did in Chap. 1 to describe binding, we will use the atomic wave functions to approximate the molecular wavefunction  $\psi_{12}$ .

$$\psi_{12} = (\phi_A(1) + \phi_B(1)) (\phi_A(2) + \phi_B(2)) \otimes \text{spin part}$$

$$\begin{aligned}
&= (\phi_A(1)\phi_A(2) + \phi_B(1)\phi_B(2) + \phi_A(1)\phi_B(2) + \phi_A(2)\phi_B(1)) \\
&\quad \otimes \text{spin part} \tag{24}
\end{aligned}$$

If  $\frac{e^2}{r_{12}}$  is strong (it is) then the first two *states with both electrons on the same ion are suppressed*, especially if the ions are far apart. Thus we neglect them, and make the Heitler-London approximation; for example

$$\psi_{12} \simeq (\phi_A(1)\phi_B(2) + \phi_B(1)\phi_A(2)) \otimes \text{spin singlet} \tag{25}$$

The spatial wave function is symmetric, and thus appropriate for the spin singlet state since the total electronic wave function must be antisymmetric. For the symmetric spin triplet states, the electronic wave function is

$$\psi_{12} = (\phi_A(1)\phi_B(2) - \phi_B(1)\phi_A(2)) \otimes \text{spin triplet} \tag{26}$$

or

$$\psi_{12} = \phi_A(1)\phi_B(2) \pm \phi_B(1)\phi_A(2) \otimes \text{spin part} \tag{27}$$

The energy of these states may then be calculated by evaluating  $\frac{\langle \psi_{12} | H | \psi_{12} \rangle}{\langle \psi_{12} | \psi_{12} \rangle}$ .

$$E = \frac{\langle \psi_{12} | H | \psi_{12} \rangle}{\langle \psi_{12} | \psi_{12} \rangle} = 2E_I + \frac{C \pm A}{1 \pm S}, \quad + \text{ singlet } , - \text{ triplet} \tag{28}$$



where

$$E_I = \int d^3r_1 \phi_A^*(1) \left\{ -\frac{\hbar^2}{2m} \nabla_1^2 - \frac{e^2}{r_{1A}} \right\} \phi_A(1) < 0 \quad (29)$$

the Coulomb integral

$$C = e^2 \int d^3r_1 d^3r_2 \left\{ \frac{1}{R_{AB}} + \frac{1}{r_{12}} - \frac{1}{r_{2A}} - \frac{1}{r_{1B}} \right\} |\phi_A(1)|^2 |\phi_B(2)|^2 < 0 \quad (30)$$

the exchange integral

$$A = e^2 \int d^3r_1 d^3r_2 \left\{ \frac{1}{R_{AB}} + \frac{1}{r_{12}} - \frac{1}{r_{2A}} - \frac{1}{r_{1B}} \right\} \phi_A^*(1) \phi_A(2) \phi_B(1) \phi_B^*(2) \quad (31)$$

and finally, the overlap integral is

$$S = \int d^3r_1 d^3r_2 \phi_A^*(1) \phi_A(2) \phi_B(1) \phi_B^*(2) \quad (0 < S < 1) \quad (32)$$

All  $E_I, C, A, S \in \mathfrak{R}$ . So

$$\begin{aligned} -J &= E_t - E_s = 2E_I + \frac{C - A}{1 - S} - \left\{ 2E_I + \frac{C + A}{1 + S} \right\} \\ -J &= \frac{C - A}{1 - S} - \frac{C + A}{1 + S} > 0 \\ J &= 2 \frac{A - SC}{1 - S^2} < 0 \end{aligned} \quad (33)$$

where the inequality follows since the last two terms in the  $\{ \}$  dominate the integral for  $A$  and in the Heitler-London approx-

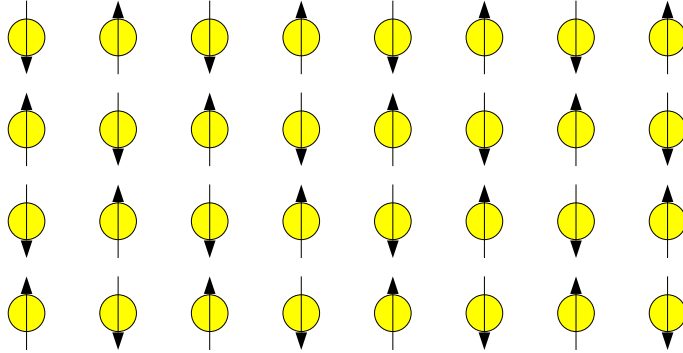


Figure 9:

imation  $S \ll 1$ . Or, for the effective Hamiltonian.

$$H = -2J \sigma_1 \cdot \sigma_2, \quad J < 0 \quad (34)$$

Clearly this favors an antiparallel or *antiferromagnetic* alignment of the spins (See Fig. 9) since then (classically)  $\sigma_1 \cdot \sigma_2 < 0$  and  $E < 0$ , so minimizing the energy. This type of interaction is clearly appropriate for insulators which may be approximate as a collection of isolated atoms. Indeed antiferromagnets are generally insulators for this and other reasons.

$$H = -2J \sum_{\langle ij \rangle} \sigma_i \cdot \sigma_j, \quad J < 0 \quad (35)$$

### 2.2.2 Exchange Interaction for Delocalized Spins

Ferromagnetism, where adjacent spins tend to align forming a bulk magnetic moment, is most often seen in conducting metals such as Fe. As we will see in this section, the Pauli principle, the Coulomb interactions, and the itinerancy of free (metallic) electrons favors a ferromagnetic ( $J > 0$ ) exchange interaction.

Consider two like-spin (triplet) free electrons in a volume  $V$  (See Fig. 10). If we describe the spatial part of their wave

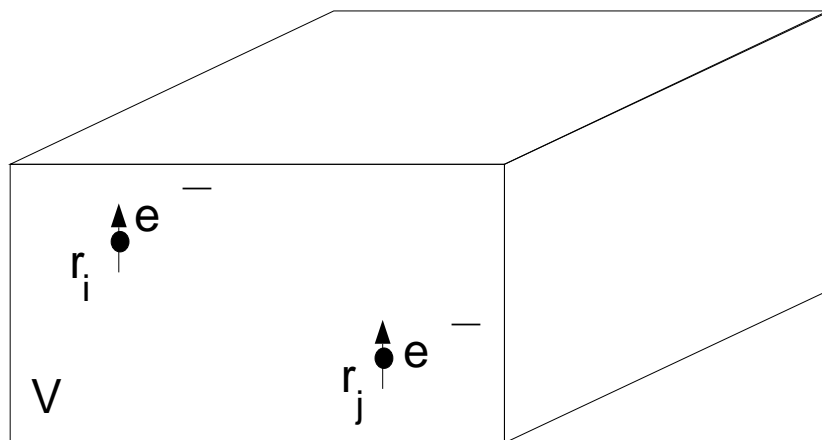


Figure 10:  $|\uparrow \uparrow\rangle$  triplet-symmetric

function with plane waves, then

$$\begin{aligned} \psi_{ij} &= \frac{1}{\sqrt{2V}} \left\{ e^{ik_i \cdot r_i} e^{ik_j \cdot r_j} - e^{ik_i \cdot r_j} e^{ik_j \cdot r_i} \right\} \\ &= \frac{1}{\sqrt{2V}} e^{ik_i \cdot r_i} e^{ik_j \cdot r_j} \left\{ 1 - e^{i(k_i - k_j) \cdot (r_i - r_j)} \right\} \end{aligned} \quad (36)$$

The probability that the electrons are in volumes  $d^3r_i$  and  $d^3r_j$  is

$$|\psi_{ij}|^2 d^3r_i d^3r_j = \frac{1}{V^2} \{1 - \cos [(k_i - k_j) \cdot (r_i - r_j)]\} d^3r_i d^3r_j \quad (37)$$

As required by the Pauli principle, this probability vanishes when  $r_i = r_j$ . This would not be the case for electrons in the singlet spin state (if the coulomb interaction continues to be ignored). Thus there is a hole, called the “exchange hole”, in the probability density for  $r_i \approx r_j$  for triplet spin electrons, but not singlet spin ones.

Now consider the effects of the electron-ion and the electron-electron coulomb interactions (See Figure 11). If one electron comes near an ion, it will screen the potential of that ion seen by other electrons; thereby raising their energy. Thus the effect of allowing electrons to approach each other, is to increase the electron-ion coulomb energy, and of course the electron-electron Coulomb energy. Thus, anything which keeps them apart without an energy cost, like the exchange hole for triplet spin electrons, will reduce their energy. As a result, like-spin

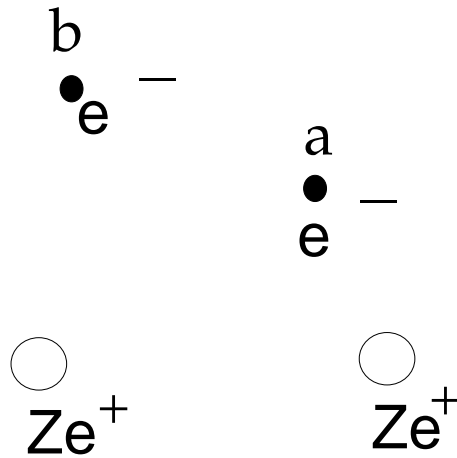


Figure 11: *Electron a screens the potential seen by electron b, raising its energy. Anything which keeps pairs of electrons apart, but costs no energy like the exchange hole for the electronic triplet, will lower the energy of the system. Thus, triplet formation is favored thermodynamically.*

electrons have lower energy and are thermodynamically favored  
 $\Rightarrow$  Ferromagnetism.

To determine the range of this FM exchange interaction, we must average the effect over the Fermi sea. If one of the electrons is fixed at the origin (See Figure 12), then the probability that a second is located a distance  $\mathbf{r}$  away, in a volume element  $d^3r$  is

$$P_{\uparrow\uparrow}(r)d^3r = n_{\uparrow}d^3r \underbrace{\overline{(1 - \cos[(k_i - k_j) \cdot r])}}_{\text{Fermi sea average}} \quad (38)$$

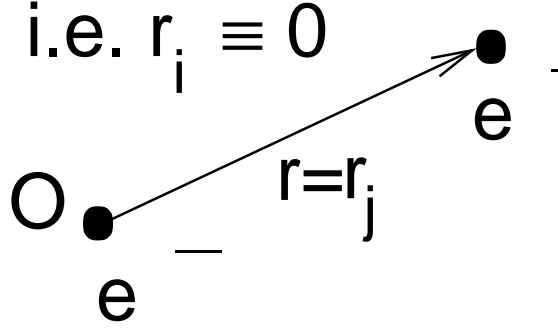


Figure 12: Geometry to calculate the exchange interaction.

$$n_{\uparrow} = \frac{1}{2}n = \frac{1}{2} \frac{\# \text{ electrons}}{\text{volume}} \quad (39)$$

In terms of an electronic charge density, this is

$$\begin{aligned} \rho_{ex}(r) &= \frac{en}{2} \overline{(1 - \cos [(k_i - k_j) \cdot r])} \\ &= \frac{en}{2} \left\{ 1 - \frac{1}{\left(\frac{4}{3}k_F^3\right)^2} \int_0^{k_F} d^3k_i d^3k_j \frac{1}{2} \left( e^{i(k_i - k_j) \cdot r} + e^{-i(k_i - k_j) \cdot r} \right) \right\} \\ &= \frac{en}{2} \left\{ 1 - \left(\frac{4}{3}k_F^3\right)^{-2} \int_0^{k_F} d^3k_i e^{ik_i \cdot r} \int_0^{k_F} d^3k_j e^{ik_j \cdot r} \right\} \\ &= \frac{en}{2} \left\{ 1 - 9 \frac{(\sin k_F r - k_F r \cos k_F r)^2}{(k_F r)^6} \right\} \end{aligned} \quad (40)$$

Note that both of the exponential terms in the second line are the same, since we integrate over all  $k_i \rightarrow -k_i$  &  $k_j \rightarrow -k_j$ . Since we have only been considering Pauli-principle effects, the electronic density of spin down electrons remains unchanged. Thus, the total charge density around the up spin electron fixed

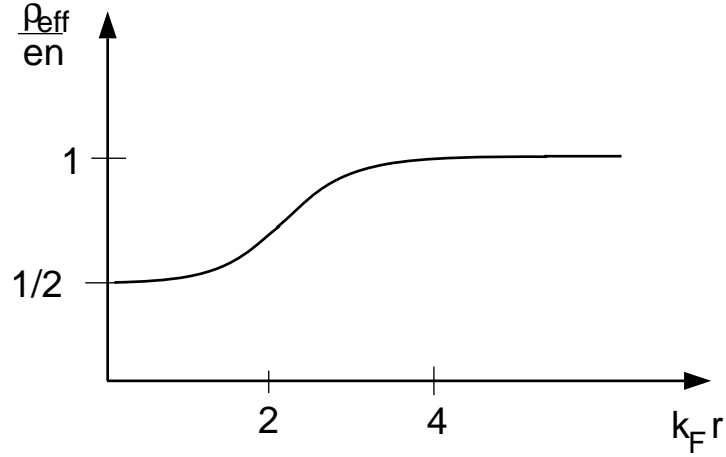


Figure 13: *Electron density near an electron fixed at the origin. Coulomb effects would reduce the density for small  $r$  further, but would not significantly effect the size of the exchange hole or the range of the corresponding potential, both  $\sim 1/k_F$ .*

at the origin is

$$\rho_{eff}(r) = en \left\{ 1 - \frac{9 (\sin k_F r - k_F r \cos k_F r)^2}{2 (k_F r)^6} \right\} \quad (41)$$

The size of the exchange hole, and the range of the corresponding ferromagnetic exchange potential, is  $\sim \frac{1}{k_F} \sim a$  which is rather short.

### 3 Band Model of Ferromagnetism

Due to the short range of this potential, its Fourier transform is essentially flat in  $k$ . This fact may be used to construct a band

theory of FM where the mean effect of a spin-up electron is to lower the energy of all other band states of spin up electrons by a small amount, independent of  $k$ .

$$E_{\uparrow}(k) = E(k) - \frac{IN_{\uparrow}}{N}; \quad I \lesssim 1eV \quad (42)$$

Likewise for spin down

$$E_{\downarrow}(k) = E(k) - \frac{IN_{\downarrow}}{N}. \quad (43)$$

Where  $I$ , the stoner parameter, quantifies the exchange hole energy. The relative spin occupation  $R$  is related to the bulk moment

$$R = \frac{(N_{\uparrow} - N_{\downarrow})}{N}, \quad M = \mu_B \left( \frac{N}{V} \right) R \quad (44)$$

Then

$$E_{\sigma}(k) = E(k) - \frac{I(N_{\uparrow} + N_{\downarrow})}{2N} - \frac{\sigma IR}{2}, \quad (\sigma = \pm) \quad (45)$$

$$\equiv \tilde{E}(k) - \frac{\sigma IR}{2}. \quad (46)$$

If  $R$  is finite and real, then we have ferromagnetism.

$$R = \frac{1}{N} \sum_k \frac{1}{\exp \left\{ (\tilde{E}(k) - IR/2 - E_F)/k_B T \right\} + 1} - \frac{1}{\exp \left\{ (\tilde{E}(k) + IR/2 - E_F)/k_B T \right\} + 1} \quad (47)$$



For small  $R$ , we may expand around  $\tilde{E}(k) = E_F$ .

$$f(x - a) - f(x + a) = -2af' - \frac{2}{3!}a^3 f''' \quad (48)$$

All derivatives will be evaluated at  $\tilde{E}(k) = E_F$ , so  $f' < 0$  and

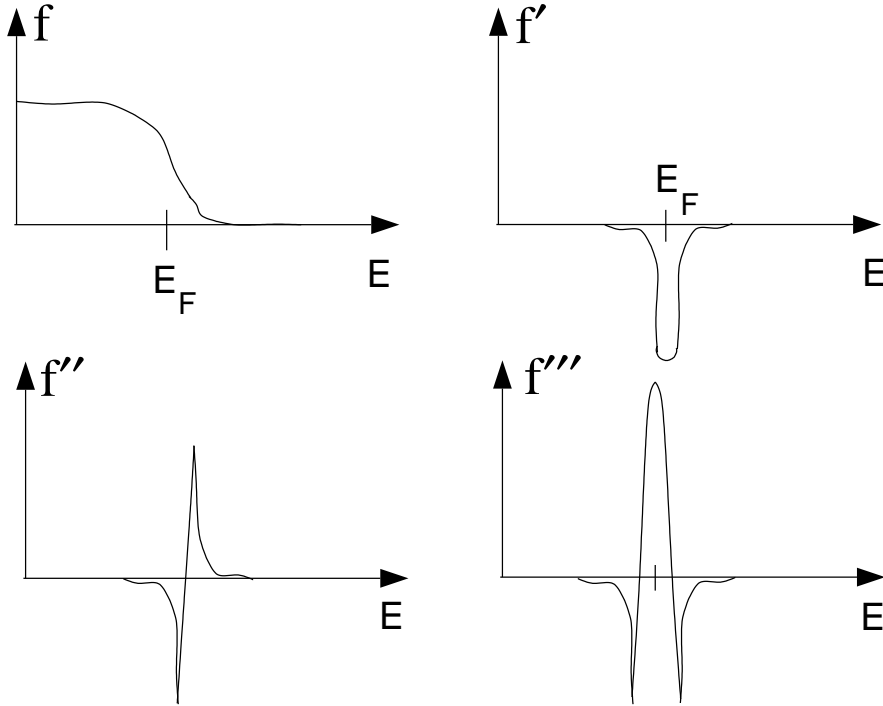


Figure 14:

$f''' > 0$ . Thus,

$$R = -2 \frac{IR}{2} \frac{1}{N} \sum_k \left. \frac{\partial f}{\partial \tilde{E}(k)} \right|_{E_F} - \frac{2}{6} \left( \frac{IR}{2} \right)^3 \frac{1}{N} \sum_k \left. \frac{\partial^3 f}{\partial \tilde{E}^3(k)} \right|_{E_F} \quad (49)$$

This is a quadratic equation in  $R$

$$-1 - \frac{I}{N} \sum_k \left. \frac{\partial f}{\partial E(k)} \right|_{E_F} = \frac{1}{24} I^3 R^2 \frac{1}{N} \sum_k \left. \frac{\partial^3 f}{\partial E^3(k)} \right|_{E_F} \quad (50)$$

which has a real solution iff

$$-1 - \frac{I}{N} \sum_k \left. \frac{\partial f}{\partial E(k)} \right|_{E_F} > 0 \quad (51)$$

Or, the derivative of the Fermi function summed over the BZ must be enough to overcome the -1 and produce a positive result. Clearly this is most likely to happen at  $T = 0$ , where  $\left. \frac{\partial f}{\partial E(k)} \right|_{E_F} \rightarrow -\delta(\tilde{E} - E_F)$

$$T = 0, \quad -\frac{1}{N} \sum_k \frac{\partial f}{\partial E_k} = \int d\tilde{E} \frac{V}{2N} D(\tilde{E}) \delta(\tilde{E} - E_F) = \frac{V}{2N} D(E_F) = \tilde{D}(E_F) \quad (52)$$

So, the condition for FM at  $T = 0$  is  $I\tilde{D}(E_F) > 1$ . This is known as the Stoner criterion.  $I$  is essentially flat as a function

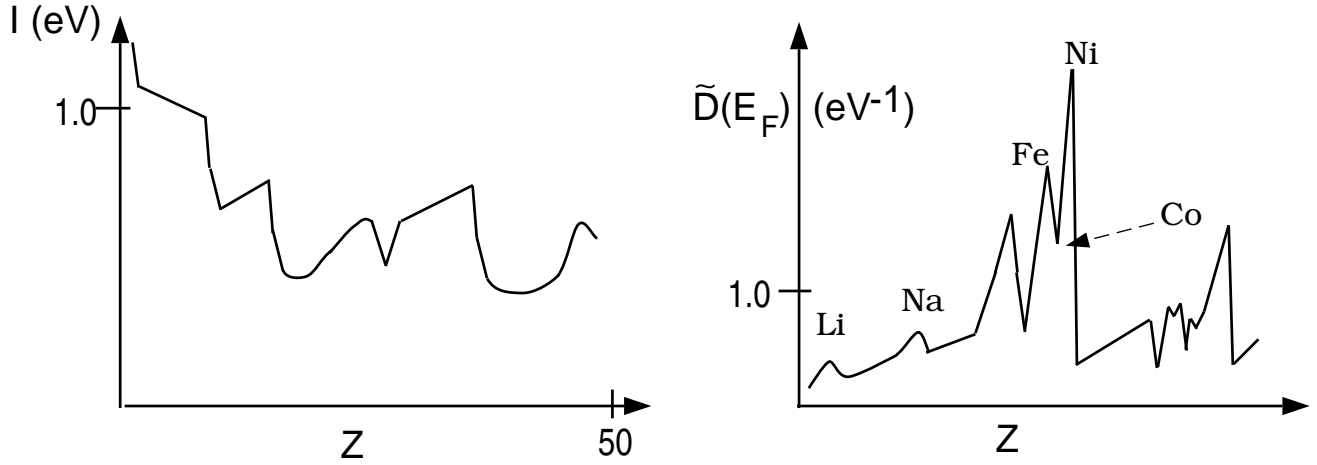


Figure 15:

of the atomic number, thus materials such as Fe, Co, & Ni with a large  $\tilde{D}(E_F)$  are favored to be FM.

### 3.1 Enhancement of $\chi$

Even those systems without a FM ground state have their susceptibility strongly enhanced by this mechanism. Let us reconsider the effect of an external field ( $gS = 1$ ) on the band energies.

$$E_\sigma(k) = E(k) - \frac{In_\sigma}{N} - \mu_B\sigma B \quad (53)$$

Then

$$\begin{aligned} R &= -\frac{1}{N} \sum_k \frac{\partial f}{\partial E_k} (IR + 2\mu_B B) \\ &= \tilde{D}(E_F) (IR + 2\mu_B B) \end{aligned} \quad (54)$$

or as  $M = \mu_B \frac{N}{V} R$ , we get

$$M = 2\mu_B^2 \frac{N}{V} \frac{\tilde{D}(E_F)}{1 - I\tilde{D}(E_F)} B \quad (55)$$

or

$$\chi = \frac{\partial M}{\partial B} = \frac{\chi_0}{1 - I\tilde{D}(E_F)} \quad (56)$$

$$\chi_0 = 2\mu_B^2 \frac{N}{V} \tilde{D}(E_F) \quad (57)$$

$$= \mu_B^2 D(E_F) \quad (58)$$

Thus, when  $I\tilde{D}(E_F) \lesssim 1$ , the susceptibility can be considerably enhanced over the non-interacting result  $\chi_0$ . However, this approximation usually overestimates  $\chi$  since it neglects diamagnetic contributions, and spin fluctuations (at  $T \neq 0$ ). As we will see, the latter especially are important for estimating  $T_c$ .

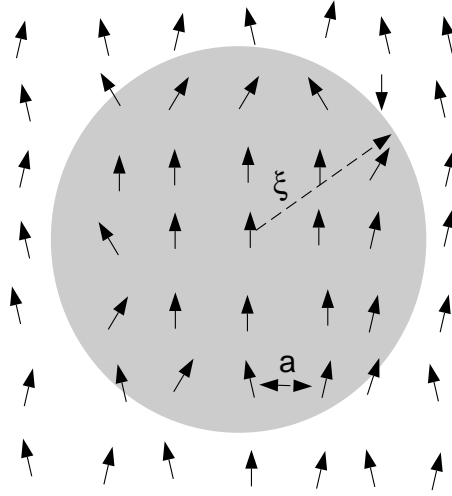


Figure 16: *Spin fluctuations can reduce the total moment within the correlated region, and even reduce  $\xi$  itself. Both effects lead to a reduction in the bulk susceptibility  $\chi \sim \text{moment}^2$*

### 3.2 Finite T Behavior of a Band Ferromagnet

In principle, one could start from an ab-initio calculation of the electronic band structure of  $E(k)$  and  $I$ , such as Ni, and

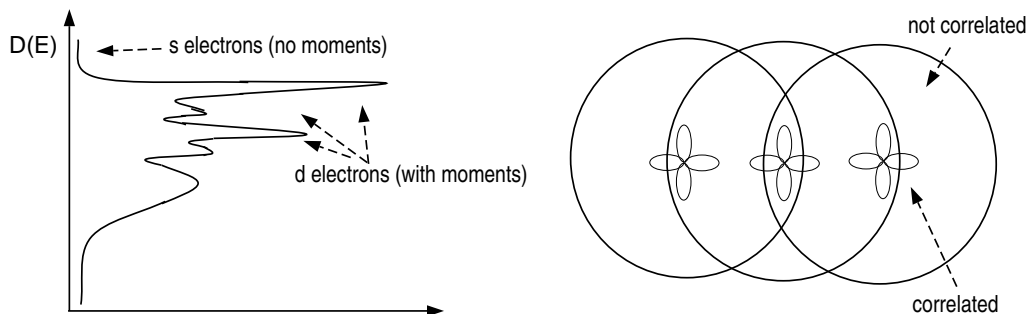


Figure 17: *In metallic Ni, the d-orbitals are compact and hybridize weakly due to low overlap with the s-orbitals (due to symmetry) and with each other (due to low overlap). Thus, moments tend to form on the d-orbitals and they contribute narrow features in the electronic density of states. The s-orbitals hybridize strongly and form a broad metallic band.*

calculate the temperature dependence of  $R$  (and hence the magnetization) using

$$R = \frac{1}{N} \sum_k f\left(\tilde{E}_k - \frac{IR}{2} - \mu_B B_0 - E_F\right) - f\left(\tilde{E}_k + \frac{IR}{2} + \mu_B B_0 - E_F\right) \quad (59)$$

with  $f(x) = \frac{1}{e^{\beta x} + 1}$ . However, this would be pointless since all of the approximations made to this point have destroyed the *quantitative* validity of the calculation. However, it still retains

a *qualitative* use. For Ni, we can do this by approximating the very narrow d-electron feature in  $D(E)$  as a  $\delta$  function and performing the integral. *However*, only the d-electrons have a strong exchange splitting  $I$  and hence only they will tend to contribute to the magnetization. *Thus our  $\tilde{D}(E)$  should reflect only the d-electron contribution*, we will accommodate this by setting

$$\tilde{D}(E) \approx C\delta(E - E_F), \quad (C < 1) \quad (60)$$

$C$ , an unknown constant, will be determined by the  $T = 0$  behavior. Then

$$R = C \left\{ f\left(-\frac{IR}{2} - \mu_B B_0\right) - f\left(\frac{IR}{2} + \mu_B B_0\right) \right\} \quad (61)$$

Let  $\frac{R}{C} \equiv \tilde{R}$  and  $T_c = \frac{IC}{4k_B}$ , then if  $B_0 = 0$

$$\tilde{R} = \frac{1}{\exp\left(\frac{-2\tilde{R}T_c}{T}\right) + 1} - \frac{1}{\exp\left(\frac{2\tilde{R}T_c}{T}\right) + 1} = \tanh \frac{\tilde{R}T_c}{T} \quad (62)$$

If  $T = 0$ , then  $\tilde{R} = 1 = \frac{R}{C} = \frac{1}{C} \frac{n_\uparrow - n_\downarrow}{N}$ . For Ni, the measured ground state magnetization per Ni atom is  $\frac{\mu_{eff}}{\mu_B} = 0.54 = \frac{n_\uparrow - n_\downarrow}{N}$ . Therefore,  $C = 0.54 = \frac{\mu_{eff}}{\mu_B}$ .

For small  $x$ ,  $\tanh x \simeq x - \frac{1}{3}x^3$ , and for large  $x$

$$\begin{aligned} \tanh x &= \frac{\sinh x}{\cosh x} = \frac{e^x - e^{-x}}{e^x + e^{-x}} = \frac{1 - e^{-2x}}{1 + e^{-2x}} \\ &= (1 - e^{-2x})(1 - e^{-2x}) \simeq 1 - 2e^{-2x} \end{aligned} \quad (63)$$

Thus,

$$\tilde{R} = 1 - 2e^{-\frac{2T_c}{T}}, \quad \text{for } T \ll T_c \quad (64)$$

$$\tilde{R} = \sqrt{3}\left(1 - \frac{T}{T_c}\right)^{\frac{1}{2}}, \quad \text{for small } \tilde{R} \text{ or } T \lesssim T_c \quad (65)$$

However, neither of the formulas is verified by experiment. The

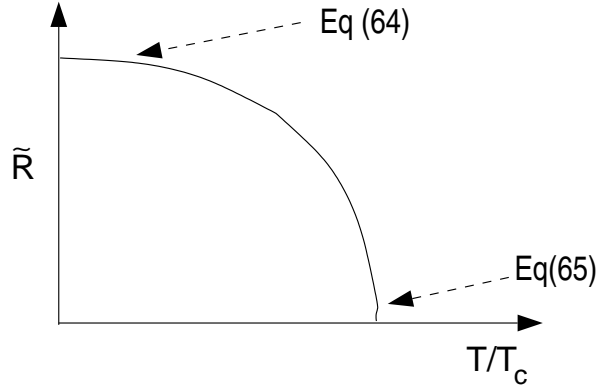


Figure 18:

critical exponent  $\beta = \frac{1}{2}$  in Eq. 65 is found to be reduced to  $\approx \frac{1}{3}$ , and Eq. 64 loses its exponential form, in real systems. Using more realistic  $\tilde{D}(E)$  or values of  $I$  will not correct these problems. Clearly something fundamental is missing from this

model (spin waves).

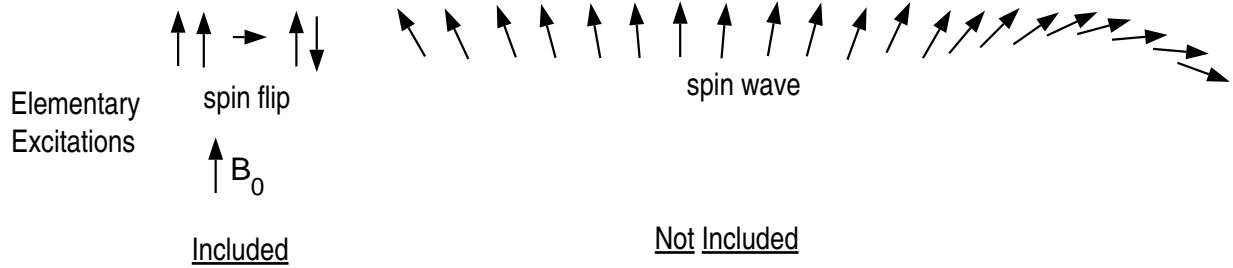


Figure 19: *Local spin-flip excitations, left, due to thermal fluctuations are properly treated by mean-field like theories such as the one discussed in Secs. 3 and 4. However, non-local spin fluctuations due to intersite correlations between the spins are neglected in mean-field theories. These low-energy excitations can fundamentally change the nature of the transition.*

### 3.2.1 Effect of B

If there is an external field  $B_0 \neq 0$ , then

$$\tilde{R} = \tanh \left\{ \frac{\tilde{R}T_c + \mu_B B_0 / 2k_B}{T} \right\} \quad (66)$$

Or for small  $R$  and  $B_0$ , (or rather, large  $T \gg T_c$ .)

$$\tilde{R} = \frac{\mu_B}{2kT} B_0 + \frac{T_c}{T} \tilde{R} \Rightarrow \tilde{R} = \frac{\mu_B}{2k} \frac{1}{T - T_c} B_0 \quad (67)$$

Thus since  $M = \frac{\mu_B N}{V} R = \frac{C \mu_B N}{V} \tilde{R} \Rightarrow \chi = \frac{\partial M}{\partial B_0} = \frac{C \mu_B^2}{2kV} \frac{N}{T - T_c}$ .

This form for  $\chi$

$$\chi = \frac{\text{Const}}{T - T_c} \quad (68)$$



is called the Curie-Weiss form which is qualitatively satisfied for  $T \gg T_c$ ; however, the values of Const and  $T_c$  predicted by band structure are inaccurate. Again, this is due to the neglect of low-energy excitations.

## 4 Mean-Field Theory of Magnetism

### 4.1 Ferromagnetism for localized electrons (MFT)

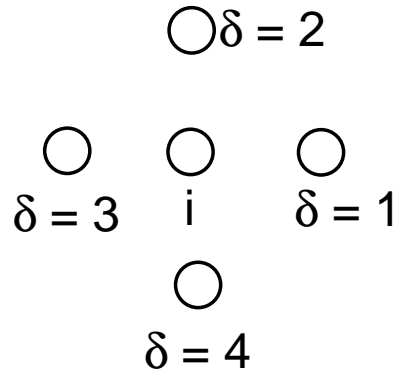


Figure 20: *Terms in the Heisenberg Hamiltonian  $H = -\sum_{i\delta} J_{i\delta} S_i \cdot S_{i\delta} - g\mu_B B_0 \sum_i S_i$ . Here  $i$  refers to the sites and  $\delta$  refers to the neighbors of site  $i$ .*

Some of the rare earth metals or ionic materials with valence d or f electrons are both ferromagnetic and have largely localized electrons for which the band theory of FM is inappropriate (A good example is  $\text{CeSi}_{2-x}$ , with  $x > 0.2$ ). As we have seen,

systems with localized spins are described by the Heisenberg Hamiltonian.

$$H = - \sum_{i\delta} J_{i\delta} S_i \cdot S_{i\delta} - g\mu_B B_0 \sum_i S_i \quad (69)$$

In general, this Hamiltonian has no solution, and we must resort to (further) approximation. In this case, we will approximate the field (exchange plus external magnetic) felt by each spin as the average field due to the neighbors of that spin and the external field. (See Fig. 21.) Then

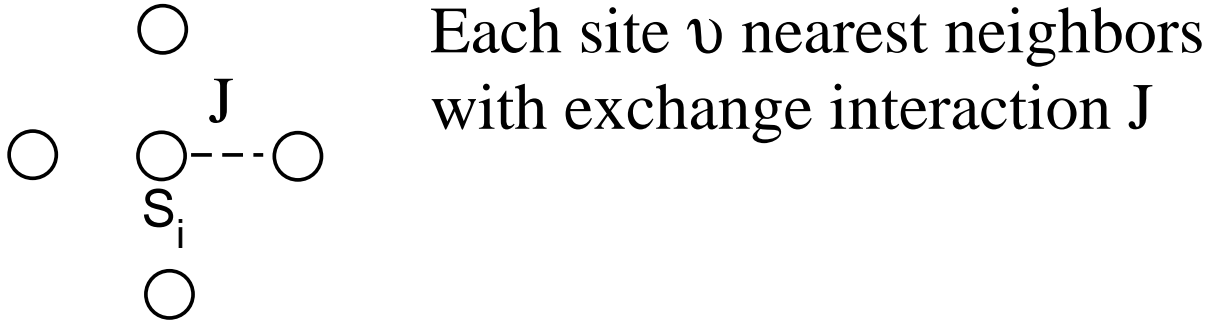


Figure 21: *The mean or average field felt by a spin  $S_i$  at site  $i$ , due to both its neighbors and the external magnetic field, is  $\frac{1}{g\mu_B} \langle \sum_{\delta} J_{i\delta} S_{i\delta} \rangle + B_0 = B_{ieff}$ . Where  $\langle \sum_{\delta} J_{i\delta} S_{i\delta} \rangle$  is the internal field, due to the neighbors of site  $i$ .*

$$H \approx - \sum_i g\mu_B B_{ieff} \cdot S_i = - \sum_i S_i \cdot \left\{ \sum_{\delta} J_{i\delta} \langle S_{i\delta} \rangle + g\mu_B B_0 \right\} \quad (70)$$

If  $J_{i\delta} = J$  is a constant (independent of  $i$  and  $\delta$ ) describing the

exchange between the spin at site  $i$  and its  $\nu$  nearest neighbors, then

$$B_{eff} = \frac{J \sum_{i\delta} \langle S_{i\delta} \rangle + g\mu_B B_0}{g\mu_B} = \frac{J\nu}{g\mu_B} \langle S \rangle + B_0 \quad (71)$$

$$M = g\mu_B \frac{N}{V} \langle S \rangle; \quad \nu = \#nn. \quad (72)$$

For a homogeneous, ordered system,

$$B_{eff} = \frac{V}{Ng^2\mu_B^2} \nu JM + B_0 = B_{MF} + B_0 \quad (73)$$

and

$$H \approx -g\mu_B B_{eff} \cdot \sum_i S_i \quad (74)$$

ie., a system of independent spins in a field  $B_{eff}$ . The probability that a particular spin is up, is then

$$P_{\uparrow} \propto e^{-\beta(-g\mu_B B_{eff} \frac{1}{2})} \quad (75)$$

and

$$P_{\downarrow} \propto e^{-\beta(+g\mu_B B_{eff} \frac{1}{2})} \quad (76)$$

so, on average

$$\frac{N_{\downarrow}}{N_{\uparrow}} = e^{-\beta g\mu_B B_{eff}} \quad (77)$$

and, since  $N_{\uparrow} + N_{\downarrow} = N$

$$M = \frac{1}{2}g\mu_B \frac{N_{\uparrow} - N_{\downarrow}}{V} = \frac{1}{2}g\mu_B \frac{N}{V} \tanh\left(\frac{\beta}{2}g\mu_B B_{eff}\right) \quad (78)$$

Since  $\tanh$  is odd and  $B_{eff} \propto M$ , this will only have non-trivial solutions if  $J > 0$  (if  $B_0 = 0$ ). If we identify

$$M_s = \frac{N}{V} \frac{g\mu_B}{2}; \quad T_c = \frac{1}{4} \nu \frac{J}{k} \quad (79)$$

$$\frac{M}{M_s} = \tanh\left(\frac{T_c}{T} \frac{M}{M_s}\right) \quad (80)$$

and again for  $T = 0$ ,  $M(T = 0) = M_s$ , and for  $T \lesssim T_c$

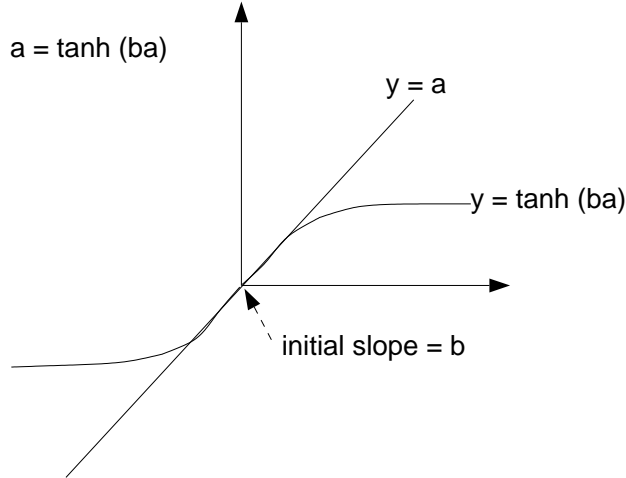


Figure 22: *Equations of the form  $a = \tanh(ba)$ , i.e. Eq. 80, have nontrivial solutions ( $a \neq 0$ ) solutions for all  $b > 1$ .*

$$\frac{M}{M_s} \simeq \sqrt{3} \left(1 - \frac{T}{T_c}\right)^{\frac{1}{2}} \quad (81)$$

Again, we get the same (wrong) exponent  $\beta = \frac{1}{2}$ .

When is this approximation good? When each spin really feels an "average" field. Suppose we have an ordered solid, so that

$$B_{MF} = \frac{J\nu}{2g\mu_B} = B_{real} \quad (82)$$

Now, consider one spin flip excitation adjacent to site  $i$  only,

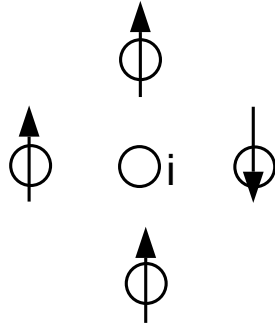


Figure 23: *The flip of a single spin adjacent to site  $i$  makes a significant change in the effective exchange field, felt by spin  $S_i$ , if the site has few nearest neighbors.*

Fig. 23.

If there are an infinite # of spins then  $B_{MF}$  remains unchanged but for  $\nu < \infty$

$$B_{real} = \frac{J\nu}{2g\mu_B} \frac{\nu - 2}{\nu} \neq B_{MF} = \frac{J\nu}{2g\mu_B}. \quad (83)$$

Clearly, for this approximation to remain valid, we need  $B_{real} =$

$B_{MF}$ , which will only happen if  $\frac{\nu-2}{\nu} = 1$  or  $\nu \gg 2$ . The more nearest neighbors to each spin, the better MFT is! (This remains true even when we consider other lower energy excitations, other than a local spin flip, such as spin waves).

## 4.2 Mean-Field Theory of Antiferromagnets

Oxides of Fe Co Ni and of course Cu often display antiferromagnetic coupling between the transition-metal d orbitals. Lets assume we have such a magnetic system on a bipartite lattice composed of two inter-penetrating sublattices, like bcc. We consider the magnetization of each lattice separately: For

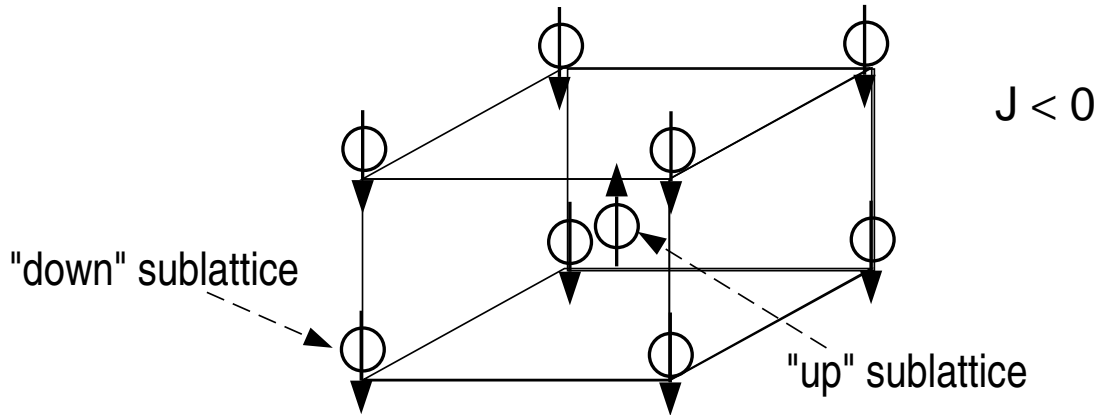


Figure 24: *Antiferromagnetism (the Neel state) on a bcc lattice is composed of two interpenetrating sc sublattices lattices.*

example, the central site shown in Fig. 24 feels a mean field from the  $\nu = 8$  near-neighbor spins on the “down” sublattice.

so

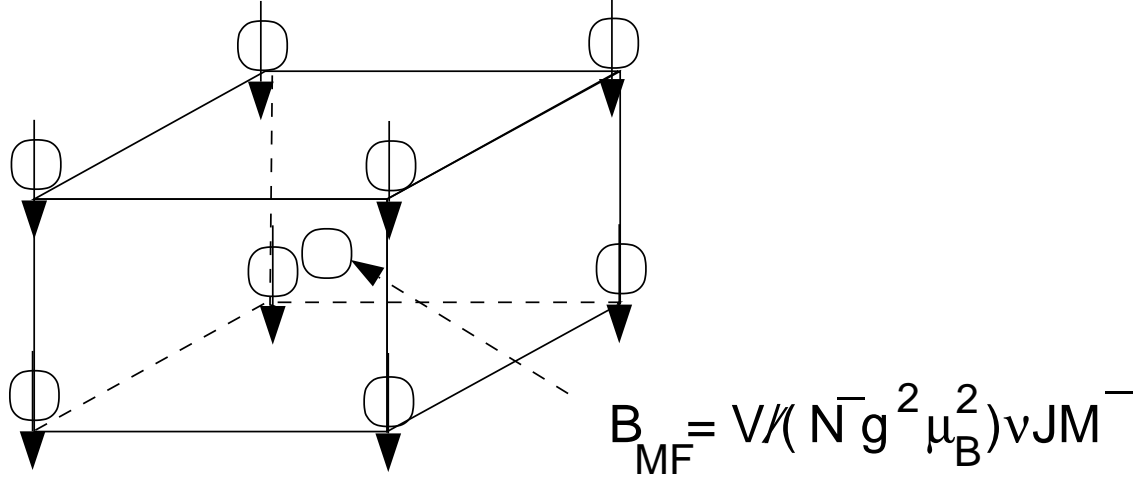


Figure 25:

$$M^+ = \frac{1}{2} g \mu_B \frac{N^+}{V} \tanh \left\{ \frac{g \mu_B}{2kT} \frac{V}{N^- g^2 \mu_B^2} \nu J M^- \right\} \quad (84)$$

$$M^- = (+ \leftrightarrow -) \dots \quad (85)$$

where  $M^+$  is the magnetization of the up sublattice. These equations have the same form as that for the ferromagnetic case!

We can make a closer analogy by realizing that  $N^+ = N^-$  and  $M^+ = -M^-$ , so that

$$M^+ = \frac{1}{2} g \mu_B \frac{N^+}{V} \tanh \left\{ -\frac{V \nu J M^+}{2kT N^+ g \mu_B} \right\}, \quad J < 0 \quad (86)$$

$$M^- = -M^+ \quad (87)$$

Again, these equations will saturate at

$$M_s^+ = -M_s^- = \frac{1}{2}g\mu_B \frac{N^+}{V} \quad (88)$$

so

$$\frac{M^+}{M_s^+} = \tanh \left\{ \frac{T_N M^+}{T M_s^+} \right\} \quad (89)$$

where  $T_N = -\frac{1}{4} \frac{\nu J}{k_B}$

Now consider the effect of a *small external field*  $B_0$ . This will yield a small increase or decrease in each sublattice's magnetization  $\Delta M^\pm$ .

$$\begin{aligned} M^+ + \Delta M^+ &= \frac{1}{2}g\mu_B \frac{N^+}{V} \tanh \left\{ \frac{g\mu_B}{2kT} \left[ B_0 + \frac{V\nu J}{N^- g^2 \mu_B^2} (M^- + \Delta M^-) \right] \right\} \\ M^- + \Delta M^- &= (+ \leftrightarrow -) \dots \end{aligned} \quad (90)$$

Or, since  $\frac{d}{dx} \tanh x = \frac{1}{\cosh^2 x}$ , then  $\left\{ \Delta M = \frac{\partial M}{\partial B_{eff}} \Delta B_{eff} \right\}$ .

$$\Delta M = \Delta M^+ + \Delta M^- = \frac{1}{2}g\mu_B \frac{N^+}{V} \frac{g\mu_B}{2kT} \frac{1}{\cosh^2 x} \left[ B_0 + \frac{V\nu J}{2N^- g^2 \mu_B^2} \Delta M \right] \quad (91)$$

where  $x = \frac{T_N M^+}{T M_s^+}$ . For  $T > T_N$ ,  $M^+ = 0$  and so  $x = 0$ , and

$$\Delta M = 2 \frac{g^2 \mu_B^2 N}{8V k_B T} \left[ B_0 - \frac{4k_B T_N V}{N g^2 \mu_B^2} \Delta M \right] \quad (92)$$

$$T \Delta M = \frac{g^2 \mu_B^2 N}{4V k_B} B_0 - \Delta M T_N \quad (93)$$



$$\Delta M = \frac{g^2 \mu_B^2 N}{4V k_B (T + T_N)} B_0 \quad (94)$$

$$\chi = \frac{g^2 \mu_B^2 N}{4V k_B (T + T_N)} \quad (95)$$

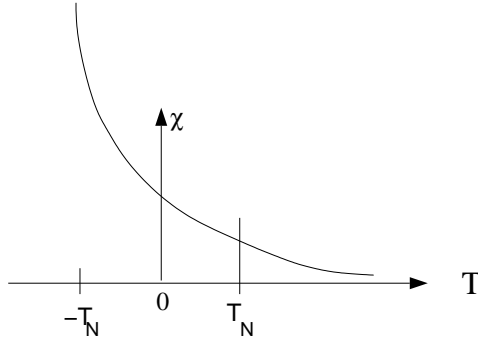


Figure 26: Sketch of  $\chi = \text{Const}/(T + T_N)$ . Unlike the ferromagnetic case, the bulk susceptibility  $\chi$  does not diverge at the transition. However, as we will see, this equation only applies for the paramagnetic state ( $T > T_N$ ), and even here, there are important corrections.

Below the transition,  $T < T_N$ , the susceptibility displays different behaviors depending upon the orientation of the applied field. For  $T \ll T_N$  and a small  $B_0$  **parallel to the axis** of the sublattice magnetization, we can approximate  $M^+(T) \approx M_s^+$  and  $x \approx \frac{T_N}{T}$  in Eq. 91

$$\chi \simeq \frac{g^2 \mu_B^2 N}{4V k_B} \frac{1}{T \cosh^2\left(\frac{T_N}{T}\right) + T_N} \quad (96)$$

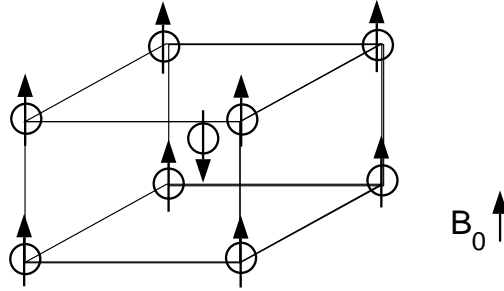


Figure 27: When  $T \ll T_N$ , a weak field applied parallel to the sublattice magnetization axis only weakly perturbs the spins. Here  $M^+(T) \approx M_s^+$  and  $x \approx \frac{T_N}{T}$

$$\chi \simeq \frac{g^2 \mu_B^2 N}{4V k_B} e^{-2\frac{T_N}{T}} \quad (97)$$

Now consider the case where  $B_0$  is perpendicular to the magnetic axis. The external field will cause each spin to rotate a

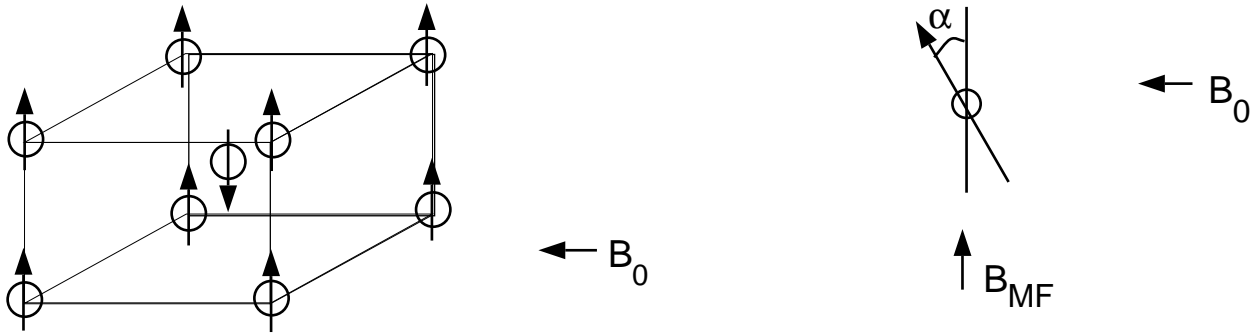


Figure 28: When  $T \ll T_N$ , a weak field  $B_0$  applied perpendicular to the sublattice magnetization, can still cause a rotation of each spin by an angle proportional to  $B_0/B_{MF}$ .

small angle  $\alpha$ . (See Fig. 28) The energy of each spin in this

external field and the mean field  $\propto \nu \frac{J}{g\mu_B}$  to the first order in  $B_0$  is

$$E = -\frac{1}{2}g\mu_B B_0 \sin \alpha + \frac{1}{2}\nu J \cos \alpha \quad (98)$$

Equilibrium is obtained when  $\frac{\partial E}{\partial \alpha} = 0$ . Since  $B_0$  is taken as small,  $\alpha \ll 1$ .

$$E \sim -\frac{1}{2}g\mu_B B_0 \alpha + \frac{1}{2}\nu J \left(1 - \frac{1}{2}\alpha^2\right) \quad (99)$$

or

$$\frac{\partial E}{\partial \alpha} = 0 = \frac{1}{2}g\mu_B B_0 + \frac{1}{2}\nu \alpha \Rightarrow \alpha = -\frac{g\mu_B B_0}{\nu J} \quad (100)$$

The induced magnetization is then

$$\Delta M = \frac{1}{2} \frac{g\mu_B B_0}{V} \alpha = -\frac{g^2 \mu_B^2 N B_0}{2\nu J V} \quad (101)$$

so

$$\chi_{\perp} = \frac{g^2 \mu_B^2 N}{2\nu |J| V} = \text{constant} \quad (102)$$

Of course, in general, in a powdered sample, the susceptibility will reflect an average of the two forms, see for example Fig. 30.

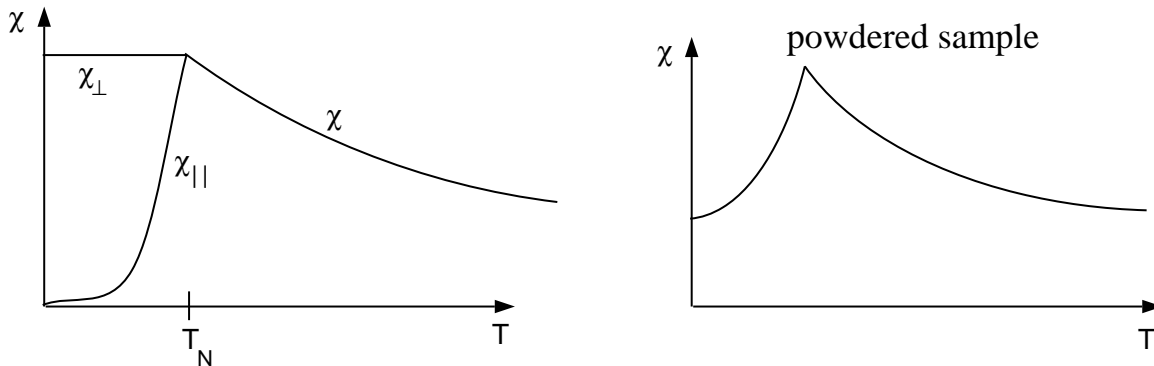


Figure 29: *Below the Neel transition, the lattice responds very differently to a field applied parallel or perpendicular to the sublattice magnetization. However, in a powdered sample, or for a field applied in an arbitrary direction, the susceptibility looks something like the sketch on the right.*

## 5 Spin Waves

We have discussed the failings of our mean-field approaches to magnetism in terms of their inability to account for low-energy processes, such as the flipping of spins. ( $S^\alpha \rightarrow -S^\alpha$ ) However, we have yet to discuss the lowest energy spin flip processes which are spin waves.

We will approach spin-waves two ways. First following Ibach and Luth we will determine a spin wave in a ferromagnet. Second, we will argue that they should be quantized and then introduce a (canonical) transformation to a Boson representa-

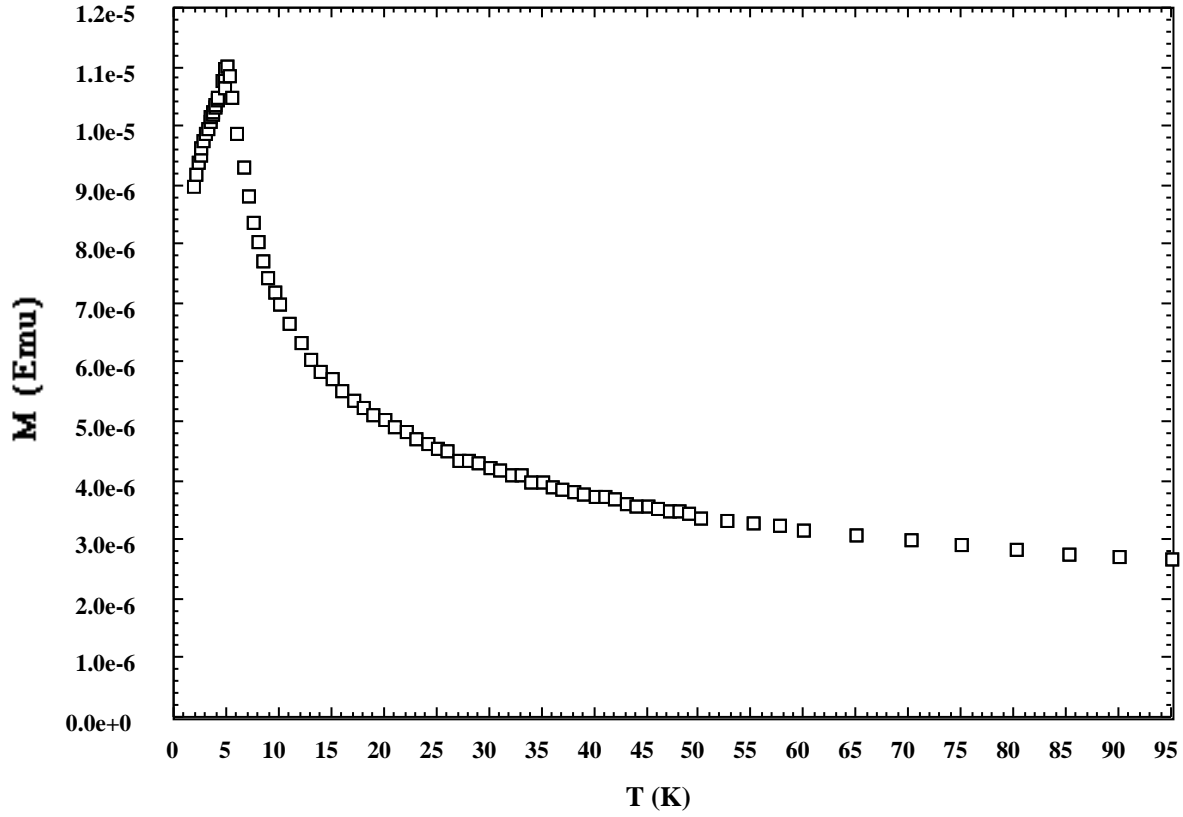


Figure 30: *High temperature superconductor Y123 with 25% Fe substituted on Cu, courtesy W. Joiner, data from a SQUID magnetometer.*

tion.

Consider a ferromagnetic Heisenberg Model

$$H = -J \sum_{i\delta} S_i \cdot S_{i+\delta} \quad (103)$$

where  $S_i = \hat{\mathbf{x}}S_i^x + \hat{\mathbf{y}}S_i^y + \hat{\mathbf{z}}S_i^z$ . If we define  $|\alpha\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$  (i.e.

$|\uparrow\rangle\rangle, \beta = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$  (i.e.  $|\downarrow\rangle\rangle$ ) so that

$$S^z \begin{pmatrix} 0 \\ 1 \end{pmatrix} = -\frac{1}{2} \begin{pmatrix} 0 \\ 1 \end{pmatrix} \dots \quad (104)$$

and

$$S^x = \frac{1}{2} \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \quad S^y = \frac{1}{2} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad S^z = \frac{1}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (105)$$

$$[S^\alpha, S^\beta] = i\epsilon_{\alpha\beta\gamma} S^\gamma \quad (106)$$

It is often convenient to introduce spin lowering and raising operators  $S^-$  and  $S^+$ .

$$S^+ = S^x + iS^y = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \quad [S^z, S^\pm] = \pm S^{\pm} \quad (107)$$

$$S^- = S^x - iS^y = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \quad [S^2, S^\pm] = 0 \quad (108)$$

$$S^+ \begin{pmatrix} 0 \\ 1 \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \end{pmatrix}, S^- \begin{pmatrix} 0 \\ 1 \end{pmatrix} = 0 \dots \quad (109)$$

They allow us to rewrite  $H$  as

$$H = -J \sum_{i\delta} S_i^z S_{i+\delta}^z + \frac{1}{2} (S_i^+ S_{i+\delta}^- + S_i^- S_{i+\delta}^+) \quad (110)$$

Since  $J > 0$ , the ground state is composed of all spins oriented, for example

$$|0\rangle = \prod_i |\alpha\rangle_i \quad (\text{i.e. all up}) \quad (111)$$

This is an eigenstate of  $H$ , since

$$S_i^+ S_{i+\delta}^- |0\rangle = 0 \quad (112)$$

and

$$S_i^z S_{i+\delta}^z |0\rangle = \frac{1}{4} |0\rangle \quad (113)$$

so

$$H |0\rangle = -\frac{1}{4} J \nu N |0\rangle \equiv E_0 |0\rangle \quad (114)$$

where  $N$  is the number of spins each with  $\nu$  nearest neighbors.

Now consider a spin-flip excitation. (See Fig. 31)

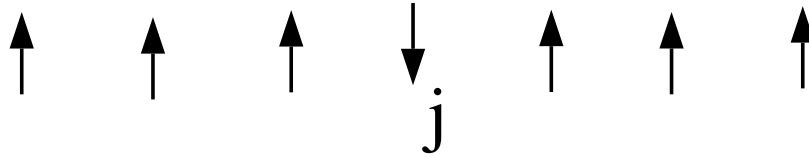


Figure 31: A single local spin-flip excitation of a ferromagnetic system. The resulting state is not an eigenstate of the Heisenberg Hamiltonian.

$$|\downarrow_j\rangle = S_j^- \prod_n |\alpha\rangle_n \quad (115)$$

This is *not* an eigenstate since the Hamiltonian operator  $S_j^+ S_{j+\delta}^-$  will move the flipped spin to an adjacent site, and hence create another state.

However, if we delocalize this spin-flip excitation, then we can create a lower energy excitation (due to the non-linear nature of the inter-spin potential) which is an eigenstate. Consider the

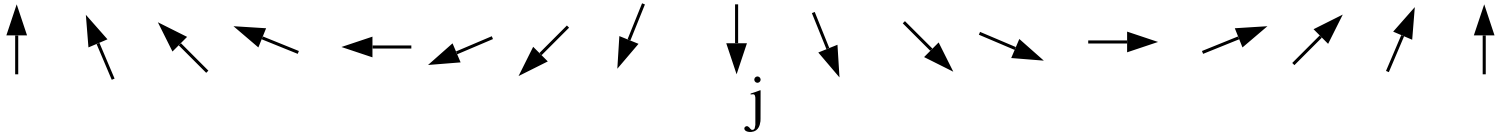


Figure 32: *If we spread out the spin-flip over a wider region then we can create a lower energy excitation. A spin-wave is the completely delocalized analog of this with one net spin flip.*

state

$$|\mathbf{k}\rangle = \frac{1}{\sqrt{N}} \sum_j e^{i\mathbf{k}\cdot\mathbf{r}_j} |\downarrow_j\rangle . \quad (116)$$

It *is* an eigenstate. Consider:

$$H |\mathbf{k}\rangle = \frac{1}{\sqrt{N}} \sum_j e^{i\mathbf{k}\cdot\mathbf{r}_j} \left\{ -\frac{1}{4}\nu J(N-2) |\downarrow_j\rangle + \frac{1}{2}\nu J |\downarrow_j\rangle - \frac{1}{2}J \sum_{\delta} (|\downarrow_{j+\delta}\rangle + |\downarrow_{j-\delta}\rangle) \right\} \quad (117)$$

where the sum in the last two terms on the right is over the near-



neighbors  $\delta$  to site  $j$ . The last two terms may be rewritten:

$$\frac{1}{\sqrt{N}} \sum_j e^{i\mathbf{k}\cdot\mathbf{r}_j} |\downarrow_{j+\delta}\rangle = 1\sqrt{N} \sum_m e^{i\mathbf{k}\cdot(\mathbf{r}_m-\mathbf{r}_\delta)} |\downarrow_m\rangle \quad (118)$$

where

$$\mathbf{r}_{j+\delta} = \mathbf{r}_j + \mathbf{r}_\delta = \mathbf{r}_m \quad (119)$$

so

$$H |\mathbf{k}\rangle = \left\{ -\frac{1}{4}\nu JN + \nu J - \frac{1}{2}J \sum_\delta (e^{i\mathbf{k}\cdot\mathbf{r}_\delta} + e^{-i\mathbf{k}\cdot\mathbf{r}_\delta}) \right\} \frac{1}{\sqrt{N}} \sum_j e^{i\mathbf{k}\cdot\mathbf{r}_j} |\downarrow_j\rangle \quad (120)$$

Thus  $|\mathbf{k}\rangle$  is an eigenstate with an eigenvalue

$$E = E_0 + J\nu \left\{ 1 - \frac{1}{\nu} \sum_\delta \cos \mathbf{k} \cdot \mathbf{r}_\delta \right\} \quad (121)$$

Apparently the energy of the excitation described by  $|\mathbf{k}\rangle$  vanishes as  $\mathbf{k} \rightarrow 0$ .

What is  $|\mathbf{k}\rangle$ ? First, consider

$$\begin{aligned} S^z |\mathbf{k}\rangle &= \sum_i S_i^z |\mathbf{k}\rangle = \sum_i S_i^z \frac{1}{\sqrt{N}} \sum_j e^{i\mathbf{k}\cdot\mathbf{r}_j} |\downarrow_j\rangle \\ &= \frac{1}{\sqrt{N}} \sum_j e^{i\mathbf{k}\cdot\mathbf{r}_j} \sum_i S_i^z |\downarrow_j\rangle = (SN - 1) |\mathbf{k}\rangle \end{aligned} \quad (122)$$

I.e., it is an excitation of the ground state with one spin flipped. Apparently, since  $E_{k=0} = E_0$ , the energy to flip a spin in this way vanishes as  $k \rightarrow 0$ .

## 5.1 Quantization of Ferromagnetic Spin Waves

In the ground state all of the spins are up. If we flip a spin, using a spin-wave excitation, then

$$S^z |\mathbf{k}\rangle = (SN - 1) |\mathbf{k}\rangle, \quad S^z |0\rangle = SN |0\rangle \quad (123)$$

If we add another spin wave, then  $\langle S^z \rangle = (SN - 2)$ . For spin  $\frac{1}{2}$ ,  $\langle S^z \rangle = \frac{N}{2} - n$ , where  $n$  is the number of the spin waves. Since  $S^z$  is quantized, so must be the number of spin waves in each mode. Thus, we may describe spin waves using Boson creation and annihilation operators  $a^\dagger$  and  $a$ . By specifying the number  $n_k$  excitations in each mode  $k$ , the corresponding excited spin state can be described by a Boson state vector  $|n_1 n_2 \dots n_N\rangle$

We can introduce creation and annihilation operators to describe the spin excitations *on each site*. Suppose, in the ground state, the spin is saturated in the state  $S^z = S$ , then  $n = 0$ . If  $S^z = S - 1$ , then  $n = 1$ , and so on. Apparently

$$\begin{aligned} S_i^z &= S - a_i^\dagger a_i \\ S_i^+ &\propto a_i \\ S_i^- &\propto a_i^\dagger \end{aligned} \quad (124)$$

| $S^z$          | $n$ |
|----------------|-----|
| $\frac{3}{2}$  | 0   |
| $\frac{1}{2}$  | 1   |
| $-\frac{1}{2}$ | 2   |
| $-\frac{3}{2}$ | 3   |

Table 1: *The correspondence between  $S^z$  and the number of spin-wave excitations on a site with  $S = 3/2$ .*

If these excitations are Boselike, then

$$[a_i, a_i^\dagger] = 1 \quad (125)$$

$$a_i |n\rangle = \sqrt{n_i} |n - 1\rangle \quad (126)$$

$$a_i^\dagger |n\rangle = \sqrt{n_i + 1} |n + 1\rangle \quad (127)$$

This transformation is faithful (canonical) and will maintain the dynamical properties of the system (given by  $\frac{\partial}{\partial t}\theta = i\hbar [H, \theta]$ ) if it preserves the commutator algebra

$$[S_i^+, S_i^-] = 2S_i^z, \quad [S_i^-, S_i^z] = 2S_i^-, \quad [S_i^+, S_i^z] = -2S_i^+ \quad (128)$$

consider

$$\{S^+ S^- - S^- S^+\} |n\rangle = 2S^z |n\rangle = 2(S - n) |n\rangle \quad (129)$$

If  $S^+ = a$ , and  $S^- = a^\dagger$ , then the left-hand side of the above equation would be  $\{(n+1) - n\} |n\rangle = |n\rangle \neq 2(S - n) |n\rangle$ .

In order to maintain the commutators, we need

$$S^+ = \sqrt{2S - n} a \quad S^- = a^\dagger \sqrt{2S - n} \quad (130)$$

Then

$$\begin{aligned} [S^+, S^-] |n\rangle &= S^+ S^- |n\rangle - S^- S^+ |n\rangle \\ &= \sqrt{2S - a^\dagger a} a a^\dagger \sqrt{2S - a^\dagger a} |n\rangle - a^\dagger (2S - a^\dagger a) a |n\rangle \\ &= (2S - n)(n+1) |n\rangle - n(2S - (n-1)) |n\rangle \\ &= (2Sn + 2S - n^2 - n - 2Sn + n^2 - n) |n\rangle \\ &= 2(S - n) |n\rangle \end{aligned} \quad (131)$$

You can check that this transformation preserves the other commutators. Of course, we need one other constraint, since  $-S \leq S^z \leq S$ , we also must have

$$n \leq 2S \quad (132)$$

This transformation

$$S_i^+ = \sqrt{2S - a_i^\dagger a_i} a_i \quad S_i^- = a_i^\dagger \sqrt{2S - a_i^\dagger a_i} \quad S_i^z = S - a_i^\dagger a_i \quad (133)$$

is called the *Holstein Primakoff transformation*.

If we Fourier transform these operators,

$$a_i^\dagger = \frac{1}{\sqrt{N}} \sum_k e^{i\mathbf{k}\cdot\mathbf{R}_i} a_k^\dagger; \quad a_i = \frac{1}{\sqrt{N}} \sum_k e^{-i\mathbf{k}\cdot\mathbf{R}_i} a_k \quad (134)$$

then (since the Fourier transform is *unitary*) these new operators satisfy the same commutation relations

$$[a_k, a_{k'}^\dagger] = \delta_{kk'} \quad [a_k^\dagger, a_{k'}^\dagger] = [a_k, a_{k'}] = 0 \quad (135)$$

To convert the Hamiltonian into this form, assume the number of magnons in each mode is small and expand

$$\begin{aligned} S_i^+ &= \sqrt{2S - n_i} a_i \simeq \sqrt{2S} \left(1 - \frac{n_i}{4S}\right) a_i \\ &\simeq \left\{ \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{R}_i} a_{\mathbf{k}} - \frac{1}{4SN^{\frac{3}{2}}} \sum_{\mathbf{k}\mathbf{p}\mathbf{q}} e^{i(\mathbf{p}+\mathbf{q}-\mathbf{k})\cdot\mathbf{R}_i} a_{\mathbf{k}}^\dagger a_{\mathbf{p}} a_{\mathbf{q}} \right\} \end{aligned} \quad (136)$$

Of course this is only exact for  $n_i \ll 2S$ , i.e. for low  $T$  where there are few spin excitations, and large  $S$  (the classical spin limit).

In this limit

$$S_i^+ \simeq \sqrt{\frac{2S}{N}} \sum_k e^{i\mathbf{k}\cdot\mathbf{R}_i} a_{\mathbf{k}} \quad (137)$$

$$S_i^- \simeq \sqrt{\frac{2S}{N}} \sum_k e^{-i\mathbf{k}\cdot\mathbf{R}_i} a_{\mathbf{k}}^\dagger \quad (138)$$

$$S_i^z = S - \frac{1}{N} \sum_{kk'} e^{i(\mathbf{k}-\mathbf{k}') \cdot \mathbf{R}_i} a_k^\dagger a_{k'}, \quad (139)$$

the Hamiltonian

$$H = -J \sum_{i\delta} \left\{ S_i^z S_{i+\delta}^z + \frac{1}{2} (S_i^+ S_{i+\delta}^- + S_i^- S_{i+\delta}^+) \right\} \quad (140)$$

may be approximated as

$$H \simeq -NJ\nu S^2 + 2J\nu S \sum_k a_k^\dagger a_k - 2J\nu S \sum_k \left( \frac{1}{\nu} \sum_\delta e^{i\mathbf{k} \cdot \mathbf{R}_\delta} \right) a_k^\dagger a_k + O(a_k^4) \quad (141)$$

$$H \simeq E_0 + \sum_k 2J\nu S(1 - \gamma_k) a_k^\dagger a_k + O(a_k^4) \quad (142)$$

where  $\gamma_k = \frac{1}{\nu} \sum_\delta e^{i\mathbf{k} \cdot \mathbf{R}_\delta}$ . This is the Hamiltonian of a collection of

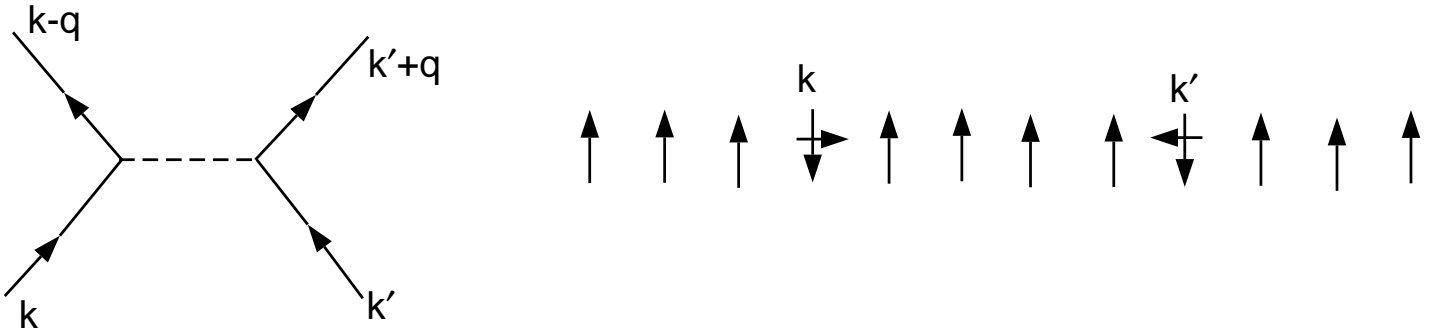


Figure 33: *The fourth order correction to Eq. 142 corresponds to interactions between the spin waves, giving them a finite lifetime*

harmonic oscillators plus some other term of order  $O(a_k^4)$  which

corresponds to interactions between the spin waves. These interactions are a result of our definition of a spin-wave as an itinerant spin flip in an otherwise perfect ferromagnet. Once we have one magnon, another cannot be created in a “perfect” ferromagnetic background.

Clearly if the number of such excitations is small ( $T$  small) and  $S$  is large, then our approximation should be valid. Furthermore, since these are the lowest energy excitations of our spin system, they should *dominate* the low- $T$  thermodynamic properties of the system such as the specific heat and the magnetization. Consider

$$\langle E \rangle = \sum_k \frac{\hbar\omega_k}{e^{\beta\hbar\omega_k} - 1}. \quad (143)$$

For small  $k$ ,  $\hbar\omega_k = 2J\nu S(1 - \gamma_k) = 2J\nu Sk^2$  on a cubic lattice. Then let's assume that the  $k$ -space is isotropic, so that  $d^3k \sim k^2 dk$ , then

$$\gamma_k = \frac{2}{\nu}(\cos k_x + \cos k_y + \dots) = 1 - \frac{k^2}{\nu} \quad (144)$$

and

$$\langle E \rangle \approx \sum_k \frac{2J\nu Sk^2}{e^{\beta 2J\nu Sk^2} - 1} \propto \int_0^\infty \frac{k^4 dk}{e^{\beta\alpha k^2} - 1} \quad (145)$$

$$x = \beta\alpha k^2 \quad k = \left(\frac{x}{\beta\alpha}\right)^{\frac{1}{2}} \quad dk = \frac{1}{2} \left(\frac{1}{\beta\alpha}\right)^{\frac{1}{2}} x^{-\frac{1}{2}} dx \quad (146)$$

so that

$$\langle E \rangle \propto \beta^{-2} \beta^{-1/2} \int_0^\infty dx \frac{x^{3/2}}{e^x - 1} \propto T^{5/2} \quad (147)$$

Thus, the specific heat at constant volume  $C_V \propto T^{3/2}$ , which is in agreement with experiment.

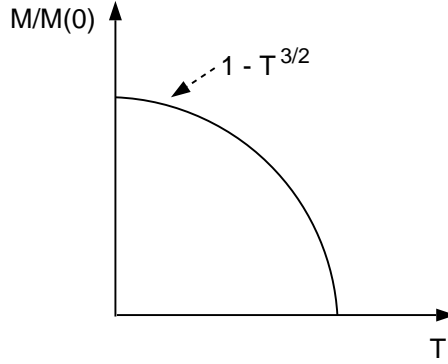


Figure 34: *The magnetization in a ferromagnet versus temperature. At low temperatures, the spin waves reduce the magnetization by a factor proportional to  $T^{3/2}$ , which dominates the reduction due to local spin fluctuations, derived from our mean-field theory. This result is also consistent with experiment.*

If we increase the temperature from zero, then the change in the magnetization is proportional to the number of magnons generated

$$M(0) - M(T) = \left\langle \sum_k n_k \right\rangle \frac{g\mu_B}{V} \quad (148)$$



since each magnon corresponds to spin flip. Thus

$$M(T) - M(0) \sim - \int \frac{k^2 dk}{e^{\beta \alpha k^2} - 1} \sim T^{\frac{3}{2}} \quad (149)$$

which clearly dominates the exponential form found in MFT ( $1 - 2e^{-\frac{2T_c}{T}}$ ). This is also consistent with experiment!

## 5.2 Antiferromagnetic Spin Waves

Since the antiferromagnetic ground state is unknown, the spin wave theory will perturb around the Neel mean-field state in which there are both a spin up and down sublattices. Spin

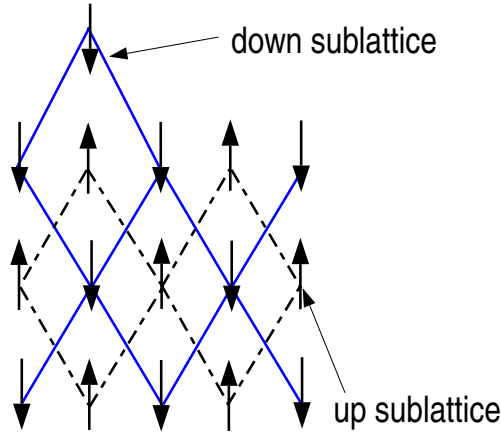


Figure 35: *To formulate an antiferromagnetic spin-wave theory, we once again consider a bipartite lattice, which may be decomposed into interpenetrating spin up and spin down sublattices.*

operators can then be written in terms of the Boson creation

and annihilation operators as before

$$\begin{array}{ll}
\text{“up” sublattice} & \text{“down” sublattice} \\
S_i^z = S - n_i & S_i^z = -S + n_i \quad (150) \\
S_i^+ = (S_i^-)^+ = \sqrt{2S} f_i(S) a_i & S_i^+ = (S_i^-)^+ = \sqrt{2S} a_i^\dagger f_i(S)
\end{array}$$

where

$$f_i(S) = \sqrt{1 - \frac{n_i}{2S}} \quad \text{and} \quad n_i = a_i^\dagger a_i \quad (151)$$

Again this transformation is exact (canonical) within the manifold of allowed states

$$0 \leq n_i \leq 2S \Leftrightarrow -S \leq S_z \leq S. \quad (152)$$

The Hamiltonian

$$H = -J \sum_{i\delta} S_i^z S_{i+\delta}^z = \frac{1}{2} (S_i^+ S_{i+\delta}^- + S_i^- S_{i+\delta}^+) \quad (153)$$

may be rewritten in terms of Boson operators as

$$\begin{aligned}
H &= +JS^2 N\nu + J \sum_{i\delta} a_i^\dagger a_i a_{i+\delta}^\dagger a_{i+\delta} \\
&- JS \sum_{i\delta} \left\{ a_i^\dagger a_i + a_{i+\delta}^\dagger a_{i+\delta} \right. \\
&+ \left. f_i(S) a_i f_{i+\delta}(S) a_{i+\delta} + a_i^\dagger f_i(S) a_{i+\delta}^\dagger f_{i+\delta}(S) \right\} \quad (154)
\end{aligned}$$

Once again, we will expand

$$f_i(S) = \sqrt{1 - \frac{n_i}{2S}} = 1 - \frac{n_i}{4S} - \frac{n_i^2}{32S^2} - \dots \quad (155)$$

and include terms in  $H$  only to  $\mathcal{O}(a^2)$

$$H \simeq JS^2 N \nu - JS \sum_{i\delta} \left\{ a_i^\dagger a_i + a_{i+\delta}^\dagger a_{i+\delta} + a_i a_{i+\delta} + a_i^\dagger a_{i+\delta}^\dagger \right\} \quad (156)$$

This Hamiltonian may be diagonalized using a Fourier transform

$$a_i = \frac{1}{\sqrt{N}} \sum_k e^{-i\mathbf{k} \cdot \mathbf{R}_i} a_k \quad (157)$$

and the Bogoliubov transform

$$a_k = \alpha_k \cosh u_k - \alpha_{-k}^\dagger \sinh u_k \quad (158)$$

$$a_k^\dagger = \alpha_k^\dagger \cosh u_k - \alpha_{-k} \sinh u_k \quad (159)$$

$$\tanh 2u_k = -\gamma_k \quad (160)$$

$$\gamma_k = \frac{1}{\nu} \sum_\delta e^{i\mathbf{k} \cdot \mathbf{R}_\delta} \quad (161)$$

Here the  $\alpha_k$  are also Boson operators  $[\alpha_k, \alpha_k^\dagger] = 1$ . To see if this transform is canonical, we must ensure that the commutators are preserved.

$$1 = [a_k, a_{k'}^\dagger] = [\alpha_k C_k - \alpha_{-k}^\dagger S_k, \alpha_{k'}^\dagger C_{k'} - \alpha_{-k'} S_{k'}]$$

$$\begin{aligned}
&= \left\{ C_k^2 [\alpha_k, \alpha_k^\dagger] + S_k^2 [\alpha_{-k}^\dagger, \alpha_{-k}] \right\} \delta_{kk'} \quad (162) \\
&= \left\{ C_k^2 - S_k^2 \right\} \delta_{kk'} = \delta_{kk'}
\end{aligned}$$

where  $C_k$  ( $S_k$ ) is shorthand for  $\cosh u_k$  ( $\sinh u_k$ ). You should check that the other relations,  $[a_k, a_{k'}] = [a_k^\dagger, a_{k'}^\dagger] = 0$ , are preserved.

After this transformation,

$$H \approx JN\nu S(S+1) + \sum_k \hbar\omega_k \left( \alpha_k^\dagger \alpha_k + \frac{1}{2} \right) + O(a^4) \quad (163)$$

where  $\hbar\omega_k = -2JS\nu\sqrt{1-\gamma_k^2}$ . Notice that for small  $k$ ,  $\hbar\omega_k \sim \sqrt{2}JS\nu k \equiv Ck$  ( $C = -\sqrt{2}JS\nu$  is the spin-wave velocity).

The ground state energy of this system (no magnons), is

$$E_0 = JN\nu S(S+1) - JS\nu \sum_k \sqrt{1-\gamma_k^2} \quad (164)$$

If  $\gamma_k = 0$ , then each spin decouples from the fluctuations of its neighbors and  $E_0 = JN\nu S^2$  ( $J < 0$ ) which is the energy of the Neel state. However, since  $\gamma_k \neq 0$ , the ground state energy  $E_0 < E_N$ . Thus the ground state is *not the Neel state*, and is thus *not composed* of perfectly antiparallel aligned spins. Each sublattice has a small amount of disorder  $\propto \langle n_i \rangle + 1/2$  in its spin alignment.

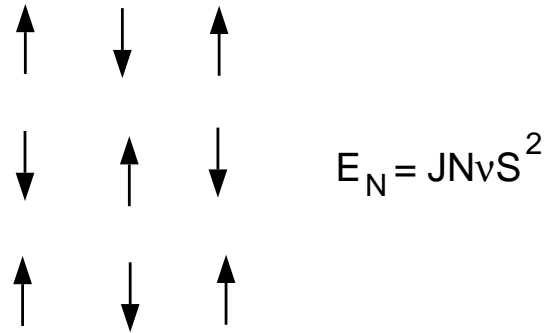


Figure 36: *The Neel state of an antiferromagnetic lattice. Due to zero point motion, this is not the ground state of the Heisenberg Hamiltonian when  $J < 0$  and  $S$  is finite.*

The linear dispersion of the antiferromagnet means that its bulk thermodynamic properties will emulate those of a phonon lattice. For example

$$\begin{aligned}
 \langle E \rangle &\simeq \sum_k \frac{\hbar\omega_k}{e^{\beta\hbar\omega_k} - 1} \simeq \sum_k \frac{\alpha k}{e^{\beta\alpha k} - 1} \\
 &\sim \int_0^\infty \frac{\alpha k^3 dk}{e^{\beta\alpha k} - 1} \\
 \langle E \rangle &\sim T^4 \int_0^\infty \frac{x^3 dx}{e^x - 1} \tag{165}
 \end{aligned}$$

$$C = \frac{\partial \langle E \rangle}{\partial T} \sim T^3 \quad \text{like phonons!} \tag{166}$$

Which means that a calorimeter experiment cannot distinguish phonon and magnon excitations of an antiferromagnet.

Therefore, perhaps the most distinctive experiment one may

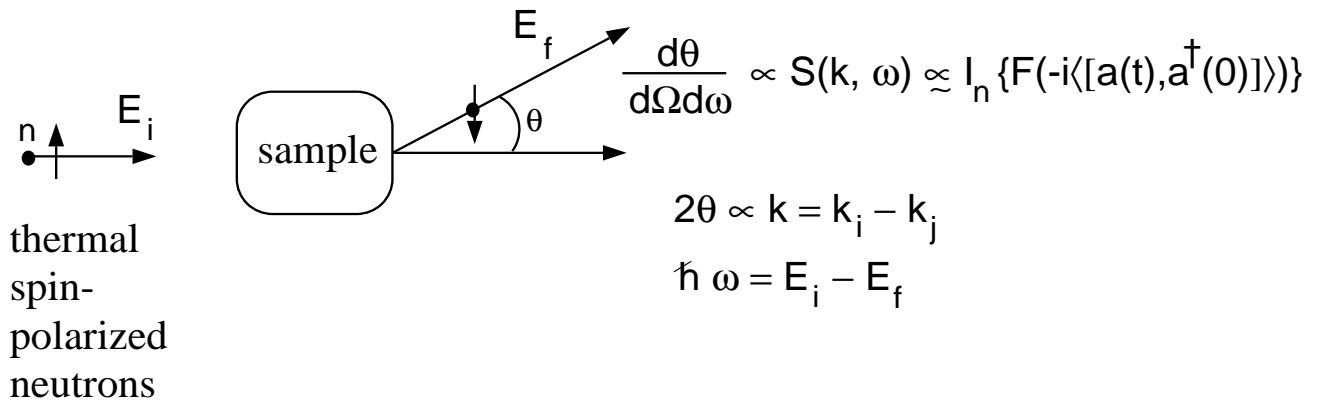


Figure 37: *Polarized neutrons are used for two reasons. First if we look at only spin flip events, then we can discriminate between phonon and magnon contributions to  $S(k, \omega)$ . Second the dispersion may be anisotropic, so excitations with orthogonal polarizations may disperse differently.*

perform on an antiferromagnet is inelastic neutron scattering. If spin-polarized neutrons are scattered from a sample, then only those with flipped spins have created a magnon. If the neutron creates a phonon, then its spin remains unchanged. The time of flight of the neutron allows us to determine the energy loss or gain of the neutron. Thus, if we plot the differential cross section of neutrons with flipped spins, we learn about magnon dispersion and lifetime. Notice that the peak in  $S(k, \omega)$  has a width. This is not just due to the instrumental resolution of the experiment; rather it also reflects the fact that magnons have a

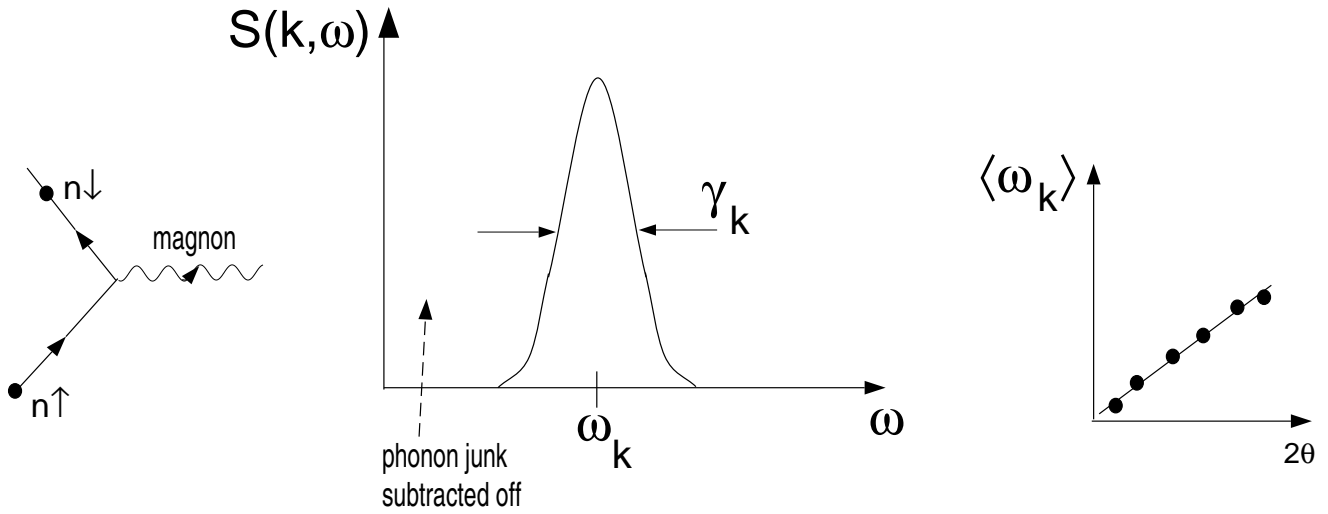


Figure 38: *Sketch of neutron structure factor from scattering off of a magnetic system. The spin-wave peak is centered on the magnon dispersion. It has a width due to the finite lifetime of magnon excitations.*

finite life time  $\delta t$ , which broadens their neutron signature by  $\gamma_k$ .

$$\gamma_k \delta t \sim \hbar \quad \delta t \sim \frac{1}{\gamma_k} \quad (167)$$

However in the quadratic spin wave approximation the lifetime of the modes  $\hbar\omega_k$  is infinite. It is the neglected terms in  $H$ , of order  $O(a^4)$  and higher which give the magnons a finite lifetime.

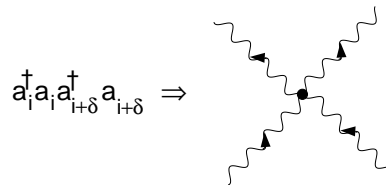


Figure 39: