Lecture 5.3

Antiferromagnetic and frustrated order

Here I address the ordering which happens when the interactions favor anti-alignment of the spins. Antiferromagnetic interactions predominate in Real local moment systems are mostly ionic insulators, in which the dominant interaction is superexchange which (as seen in Lec. 4.3 ) usually is antiferromagnetic. The spin Hamiltonian,

\[ H = -\frac{1}{2} \sum_{ij} J_{ij} \mathbf{s}_i \cdot \mathbf{s}_j \]  

for classical spins \( \mathbf{s}_i \) on some lattice. Note that \( J_{ij} \) are not limited to nearest neighbors, but are assumed (as in A& M eq. (33.4)) to have the symmetry of the lattice:

\[ J_{ij} = J_{ji} = J(\mathbf{R}_i - \mathbf{R}_j). \]

We can assume that the dominant \( J_{ij} \)'s are negative.

I’ll review the standard notion of a two-sublattice antiferromagnet, and discuss the use of neutron diffraction. However, more complicated ordering is both more interesting and more common. These include ferrimagnets and spin glasses. There are \textit{itinerant} antiferromagnets as well; this case (“spin density wave”) will be discussed in the next lecture.

All in all, apart from the ferrimagnets mentioned, antiferromagnets seem to have no practical uses. They are of interest because of the enormous variety of symmetries and phase diagrams, when one takes into account the competition with magnetic field and anisotropies. Quantum antiferromagnets (see Lec. 5.6 ) have been studied intensely for the past 10 years, at first because they are a possible mechanism of high-temperature superconductors, but also as realizations of exotic many-body correlations.

\[ ^1 \text{Antiferromagnetism, complex spin orderings, and spin glasses are covered in Lévy, 5.4-5.5.} \]

\[ ^2 \text{They are simpler experimentally than other systems since the microscopic Hamiltonian is relatively understood and has relatively few terms. On the theory side, the degrees of freedom (quantum spins) are discrete, and (unlike electrons) they don’t hop from site to site.} \]

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5.3 A Néel antiferromagnet

The simplest antiferromagnetic state is the Néel antiferromagnet, in which spin directions alternate. This occurs when the spins live on a bipartite lattice.\footnote{I believe bipartiteness was defined in Lec. 1.1 in connection with particle-hole symmetry in the hopping model bandstructure. A bipartite lattice can be (uniquely) divided into “even” and “odd” sublattices (equivalent by symmetry) such that the nearest neighbors of even sites are odd sites and vice versa. The coloring of the checkerboard demonstrates that the square lattice is bipartite. The 1D, square, bcc, and simple cubic lattices are bipartite, while the triangular and fcc lattices are not.}

From the viewpoint of the (classical) order and statistical mechanics, a Néel antiferromagnet is similar to an ferromagnet, since the broken symmetry is the same. Indeed, the transformation

\[ s'_i = \eta_i s_i \] \hspace{1cm} (5.3.3)

with \( \eta_i = \pm 1 \) depending whether the site is even or odd, maps the (classical) antiferromagnet into a (classical) ferromagnet. [That is, it changes the sign of the nearest-neighbor interaction \( J \), and it converts an alternating spin pattern to a uniform one.]

**Order parameter**

The antiferromagnet’s order parameter is called the staggered magnetization and the operator on site \( i \) is

\[ m_i = e^{iQ \cdot r_i} s_i \] \hspace{1cm} (5.3.4)

where \( Q \) is the ordering wavevector, and \( e^{iQ \cdot r_i} = \pm 1 \) usually. [This could be guessed from the ferromagnet’s order parameter, in light of the mapping (5.3.3).] [Note that the dagger in this notation does not indicate hermitian conjugate.] So, for example, for a 1D chain you would have the total staggered magnetization

\[ M = \sum_i (-1)^i s_i \] \hspace{1cm} (5.3.5)

The ordering temperature is called the Néel temperature \( T_N \). The mean-field theory is mathematically identical to that of a ferromagnet, after the substitution (5.3.3). The elastic theory looks the same, and there will be a macroscopic anisotropy arising in essentially the same fashion from the microscopic ones.

To theorists, the most significant difference is that in the antiferromagnet the order parameter operator does not commute with the exchange Hamiltonian (5.3.1). In the ferromagnet, the order parameter operator is the total spin which obviously commutes (that is the statement of spin conservation). As alluded to in connection with Goldstone modes (Lec. 1.4), this changes the dispersion law of the spin wave spectrum. (This will be worked out in Lec. 5.5 on spin waves [omitted.]) Another consequence will be that the antiferromagnet has intrinsic zero-point quantum spin fluctuations in its ground state, just as a crystal has intrinsic zero-point phonon fluctuations. (The ferromagnet has no zero-point fluctuations.) This second point will be discussed further in Lec. 5.6 on quantum antiferromagnets (omitted). In this section I stick to the classical limit.

**Elastic theory [incomplete]**

The elastic free energy for a ferromagnet looks like

\[ F_{el} = \int d^d r \left( -\frac{1}{2} \rho_s |\nabla m(r)|^2 \right), \] \hspace{1cm} (5.3.6)
5.3 B. FRUSTRATED ANTIFERROMAGNETIC ORDER

where \( \rho_s \) is the spin stiffness. Similarly, for an antiferromagnet it is

\[
F_{el} = \int d^d r \frac{1}{2} \rho_s |\nabla \mathbf{m}(r)|^2, \tag{5.3.7}
\]

**Susceptibility**

An antiferromagnet’s susceptibility \( \chi(T) \) behaves differently from a ferromagnet’s, because magnetization couples directly to magnetic field, but staggered magnetization doesn’t. The ferromagnetic \( \chi(T) \) diverges at \( T_c \), but the antiferromagnetic \( \chi(T) \) merely has a cusp (change of slope) at \( T_N \), as seen in Fig. 5.3.1. Also, below \( T_c \) the ferromagnetic \( \chi(T) \) is infinite (if we divide the magnetization by the field required to produce it), but the antiferromagnetic \( \chi(T) \) goes to a constant.

It’s easy to see why \( \chi(T = 0) \) is finite, and what the state of the antiferromagnet is in a field. \(^4\) If we rotate the afm configuration rigidly, we see we can gain nothing to first order in \( H \). \(^5\) The sublattices turn perpendicular to the external field \( H \), where each one feels the resultant of \( H \) and the exchange field (of order \( |J| \)) from the other sublattice, leading to an angular deviation of order \( H/|J| \) and thus a (finite) susceptibility of order \( 1/|J| \), with an energy gain of \( O(H^2/|J|) \). If we had instead perturbed around a state aligned with/against \( H \), there would be no way to gain this energy; thus \( \mathbf{M}^\perp \) turns normal to the field.

Why does \( \chi(T) \) have a maximum? It is true in mean-field theory, but it is harder to track down what it is supposed to do in a rigorous theory!

My best answer is to note an identity from stat mech that \( \chi_z \propto \langle \delta m_z^2 \rangle/T, \) the magnetization fluctuations (where \( \langle \ldots \rangle \) is thermal expectation here). The portion of spin that is free to fluctuation is roughly \( \sqrt{1 - |\mathbf{m}|^2} \). As temperature is lowered, the order parameter \( m^\perp \) increases and there are fewer such fluctuations. In this picture, \( \chi(T_N) - \chi(T) \propto |T - T_N|^{2\beta} \) where \( \beta \) is the order-parameter critical exponent, so it is not a simple linear cusp.

5.3 B Frustrated antiferromagnetic order

In fact, most lattices aren’t bipartite! Metals tend to be fcc, and the cation sublattice in the simplest ionic structures (zincblende or rock-salt) are fcc. The fcc lattice contains

\(^4\)This argument is very similar to Exercise [4.1.1?\] (classical antiferromagnetic pair), with each of those spins being replaced by an entire sublattice.

\(^5\) A very small particle, having (by chance) a significant imbalance of even and odd sites, will have a net real moment aligned with \( \mathbf{M}^\perp \), so \( \mathbf{M}^\perp \) might align with \( H \) in this case.
triangles which obviously prevent the even/odd labeling; there are also many hexagonal structures containing triangular layers.

Now we have “competing” interactions, or equivalently frustration. Frustration means that we cannot simultaneously have each coupling as best as it possibly can be. Antiferromagnetic triangles are frustrated since if \( s_1 \) is up, optimizing the \( J_{12} \) term tells \( s_1 \) to be down; but now the \( J_{12} \) and \( J_{23} \) terms give conflicting messages to \( s_3 \).

There is an enormous variety of frustrated structures. Frustration often leads to complicated, noncollinear magnetic structures. Their order parameters are more complicated, typically having components not only for the Cartesian component of spin, but also for the spin associated with different symmetry-related ordering vectors \( Q \).

Many of my research interests are in the ground-state order in frustrated spin systems; and, in particular, degeneracies that exist in a variety of frustrated antiferromagnets and how these are split by considering higher-order fluctuations. (Namely, the spin-wave zero-point energy is different around different ones of the classically degenerate ground states.)

**Frustrated spin ground states**

A general notion which helps is: perhaps you can write the Hamiltonian as a sum of terms \( \sum_k \mathcal{H}_k \) (each involving a local bunch of spins). For each term \( \mathcal{H}_k \), you find the ground state which minimizes its energy. If you find a state in which this is simultaneously true of all terms, it must be a ground state (and all g.s. must be of this form), since every term is as small as it can be. \(^6\) This idea can be applied to find the ground state of a triangular lattice (Fig. 5.3.2).

There is a systematic approach to finding the classical spin arrangement. It is based on Fourier analysis (the Luttinger-Tisza method). It tells you the ordering vector \( Q \) such that \( s(\mathbf{r}) \) can be written as a sum of \( e^{iQ\cdot\mathbf{r}} \). (In these complicated orderings, typically a linear combination of waves with different but symmetry-related \( Q \)). (But this method won’t work in complicated lattices such as garnets, and fails for most Ising cases.) See Sec. 5.3 Y.

**5.3 C Zoo of non-ferromagnetic states with spin order**

This section is just a guided tour through some interesting kinds of antiferromagnetic states. All of these states have well defined directions for each spin, so they constitute

\(^6\)It means, in some sense, the Hamiltonian appeared frustrated only so long as you demanded that \( \mathcal{H}_k \) be two spin clusters.
examples of spin order (as opposed to exotic quantum order we will meet in a later lecture.) All of the interesting states involve competing interactions, i.e. some kind of frustration.

**Helimagnets**

There are various arrangements in which the magnetic ordering vector $\mathbf{Q}$ is *incommensurate*. It typically happens when the nearest-neighbor exchange is ferro and the second-neighbor is antiferro. The spin directions twist through some angle as the lattice position is shifted in the $\mathbf{Q}$ direction, thus describing a spiral (helix), hence the name. There is also an exercise about this.

Note that since it arises from exchange interactions, the twist is equally likely to be in either sense: a symmetry breaking] determines it. Commensurate locking also occurs. (Compare Lec. 3.1 and Lec. 3.3 about incommensurate order.)

**Dipolar magnets**

Sometimes the exchange couplings are negligible compared to dipolar interactions. In this case, one uses the same Luttinger-Tisza approach to find the ground state (see Sec. 5.3 Y). A dipolar system is intrinsically frustrated, since the coupling between spins at $\mathbf{r}$ and $\mathbf{r}'$ has ferro or antiferro sign for the spin components parallel or perpendicular to $\mathbf{r}' - \mathbf{r}$. It turns out that the result depends on details of the lattice: I believe it is ferromagnetic on an fcc lattice, but antiferromagnetic on a diamond lattice.

UNDONE. Need to show the explicit form of $\tilde{J}$. Explain how this corresponds to large domains. Give experimental examples of dipolar ones.

**Degeneracies and selection**

In my own research, I’m interested in cases with nontrivial *degeneracies* in which there is a family of classical ground states, parametrized by one or more real numbers, which are *not* equivalent by any global symmetry. Normally that the spin wave spectra are different depending on which state you expand around, so adding up the zero-point energy of the spin waves suffices to lift the continuous degeneracy.

**Ferrimagnets**

Ferrimagnets are materials which have a net moment, but the spins are not all aligned. They have the same variety as antiferromagnets and are more naturally discussed here. The usual picture is two sublattices alternating up and down. This occurs when the sublattice of up spins is crystallographically inequivalent to that of the down spins. The order parameter is basically the moment, and macroscopically these are ferromagnets.

Ferrites are the prototypical ferrimagnet, namely Fe$_3$O$_4$. The Fe ions come in two different ionization states, with different spin lengths, and one of them with a big anisotropy. The high anisotropy is why they are used in magnetic recording media. (Ferrite was the basis of the first generation of solid-state computer fast memories, and I believe is still the material for recording tape.) Garnets are more complicated (but similar) structures. Being *insulators* (unlike iron and most other ferromagnets) they can be used for magnetooptic applications.

It is also possible to have a spontaneous symmetry breaking, as in some triangular Ising models, in which (say) two crystallographically equivalent sublattices point up and
Another scenario is to take an antiferromagnet (according to the exchange interactions) and turn on a DM antisymmetric exchange coupling. In certain crystal structures, rotating (say) the even sublattice clockwise and the odd one counterclockwise by an angle $\delta\theta$, it is found that $\delta\theta$ couples linearly to the DM term; hence a small such deviation appears, giving a net ferromagnetic moment. This is called “weak ferromagnetism” but it properly is just another variety of ferrimagnetism.

**Weak ferromagnetism**

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**Spin glasses**

[a section on these is planned but Omitted.]

### 5.3 X Experiment: neutron diffraction

If you could measure the *staggered susceptibility* $\chi^s$ which couples to $M^s$, then $\chi^s(T)$ would behave just like $\chi(T)$ in a ferromagnet. \footnote{It is possible to measure $\chi^s$ via neutron scattering: it is proportional to the structure factor $S(Q)$ at the ordering wavevector.} The order parameter of an antiferromagnet is not directly measured in the magnetic susceptibility (the net moment of the even sublattice, of course, cancels that of the odd one), so we must rely on neutron scattering. Returning to (5.3.4), notice $e^{2iQ \cdot r_i} = [(-1)^i = 1$ for every lattice site $r_i$. By the definition of Bravais lattice vectors, this says $2Q$ is one of them. Thus $Q$ is a *zone-boundary* wavevector, located somewhere on the Brillouin zone boundary. (Often at a corner – e.g. $(\pi/a, \pi/a)$ in the square lattice.) Thus the correlation functions satisfy

$$C^s(R) = e^{iQ \cdot R} C_s(R)$$  \hspace{1cm} (5.3.8)
where $C_s(R)$ is the ordinary spin correlation. The spin structure factor (at $T = 0$) is just

$$S(q) = \langle |\hat{s}(q)|^2 \rangle$$

(5.3.9)

where $\hat{s}(\cdot)$ is the Fourier transform. The neutron diffraction cross section is proportional to (5.3.9). It will be seen (by comparing (5.3.9) and (5.3.4)) that a Bragg peak appears at $q = Q$ and its intensity is proportional to $S(Q) = |M|^2$.  

Furthermore, since $S(q)$ is just the Fourier transform of the correlation function $C_s(R)$, it follows that $S(q + Q)$ is the Fourier transform of $C'_s(R)$. Thus, the long-distance correlations of antiferromagnetic order can be read from the diffuse diffraction around the Bragg point $Q$. In a typical material, this is convenient because the lattice constants are comparable to the wavelengths of thermal neutrons. Note too that neutrons have a form factor for scattering off electron moments – from the nonvanishing magnetic moment of the neutron and the standard electromagnetic coupling between them – which just happens to be about the same size as the form factor for scattering off the nuclei, which is due to the nuclear force. Thus the magnetic signal does not get lost in the noise from the nuclear signal. Also, the cross section in fact depends on the dot product of neutron spin and the local moment spin; so by using oriented single crystal samples and polarized neutron beams, you can obtain information about the actual alignment of the spins relative to real space.

Magnetic X-ray diffraction is currently being developed. The magnetic form factor is tiny compared to the atomic one, so this method is feasible only due to the high flux of synchrotron radiation.

**Magnetic unit cell**

We could define a magnetic unit cell as the Bravais lattice of the spin density. In an antiferromagnet, this cell is bigger than the unit cell of the lattice; its reciprocal lattice is correspondingly smaller. Since magnetic diffraction peaks occur at $Q$, this must also be a reciprocal lattice vector of the magnetic lattice.

### 5.3 Y Fourier approach to classical magnetic states

Mattis (*Theory of Magnetism, Vol. I*) calls this systematic approach the “method of Luttinger and Tisza.”

Say we are given the Hamiltonian (5.3.1) The problem is to find the state(s) minimizing (5.3.1) given the length constraint

$$|s_i|^2 = s^2 \quad \text{for all } i.$$  

(5.3.10)

A useful approach begins by Fourier transforming (5.3.1), into the form

$$\mathcal{H} = -\sum_q \mathcal{J}(q)|s_q|^2$$

(5.3.11)

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*8The order parameter and diffraction theory is very similar to that for a binary alloy, or for the lattice gas of Lec. 1.3 .

9Just as we saw in Lec. [1.3] that the nuclear Bragg diffraction intensities were simply the Fourier transform of the order-parameter correlation function.

10But neutron diffraction off nuclear spins – which has been done – is an impressive, difficult experiment since the signal is so small.

11Note – “magnetic space groups” have been defined, indeed more of them than of the nonmagnetic kind. They include operations which rotate the spins in spin space, combined with various symmetries of the positional degrees of freedom.*
where
\[ \tilde{J}(q) = \sum_{R} e^{-iq \cdot R} J(R). \] (5.3.12)

and the Fourier components of the spin density are \( \{s_q\} \equiv N^{-1/2} \sum e^{-i\vec{q} \cdot \vec{R}} s_i \). Now, the \( N \) length constraints (5.3.10) all look horribly complicated when expressed in terms of \( \{s_q\} \), but we can make one combination of them which is simple: Namely, eq. (5.3.10) implies \( \sum |s_i|^2 = s^2 N \) and so
\[ \sum_q |s_q|^2 = s^2 N \] (5.3.13)

Now, if there is a configuration of \( \{s_q\} \) which is the absolute minimum of energy (over all configurations satisfying (5.3.13)), and if this configuration happens to satisfy (5.3.10) as well, then this configuration is an absolute minimum over the configurations satisfying (5.3.10).

**Exercises**

**Ex. 5.3.1 Luttinger-Tisza**

Prove that the absolute minimum of energy, given (5.3.13), contains nonzero \( s_q \) only for \( q \in \{Q\} \), where \( \{Q\} \) is the set of vectors where \( \tilde{J}(q) \) attains its maximum.

To apply this, then, you must find the maximum of \( \tilde{J}(q) \), and must construct some linear combinations of the form
\[ s_i = \sum_Q s_Q e^{iQ \cdot R_i} \] (5.3.14)

with the conditions that \( s_i \) is real and has length \( s \) on every site. If you succeed, you know that all ground states must be of this form (so you know the ground state energy). Then, to find all possible ground states, you must find all ways of choosing the coefficients in (5.3.14) which satisfy the conditions.

**Ex. 5.3.2 1D chain and helimagnetic ground state**

In this exercise, the Luttinger-Tisza method is applied to a 1D chain with lattice constant \( a \). For each kind of order, draw two sketches one showing the form of \( \tilde{J}(q) \) as a function of \( q \), the other showing an example spin configuration.

(a). First, as a check do the trivial cases where you already know the answer:
(i). Ferromagnetic nearest neighbor coupling \( J_1 \).
(ii). Antiferromagnetic nearest neighbor coupling \(-|J_1|\).

In these cases the symmetry is labeled by the magnetization and staggered magnetization vectors, respectively. Either way, we have two continuous symmetry operations (One way to see this is that the space of possible orientations is labeled by two Euler angles.)

What is the ‘magnetic’ unit cell size?

(b). Now consider a second-neighbor \( J_2 \): first, what if \( J_1 = 0 \), and \( J_2 < 0 \)?
What is the ‘magnetic’ unit cell size in this case?

What is the order parameter that would label the states? How many continuous symmetries do we get in this case?
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(c). Now solve the case of general $J_1$ and $J_2$. Besides the phases found in (b) and (c), you should also find an incommensurate phase (called a “helimagnet” because the spins seem to spiral as you go along the line).

Draw a “phase diagram” of the $(J_1, J_2)$ plane indicating the portions where each type of order occurs.

(d). To the 1D Hamiltonian considered in (c), add a “biquadratic” term $\frac{1}{2} \sum_{ij} K(s_i \cdot s_j)^2$ (here $ij$ are nearest neighbors only). Out of the ground states found in (Ex. 5.3.2), which ones remain ground states? (HINT: The result depends on the sign of $K$.)

Remark: this models the zero-point energy of spin waves in the case of a degenerate ground state.

**Ex. 5.3.3 Spin order in 2D**

Continuation of (Ex. 5.3.2).

Consider a 2D square lattice with first and second neighbor couplings $J_1$ and $J_2$ (The second neighbor displacement is $[\pm a, \pm a]$).

(a). Write out $\tilde{J}(\mathbf{q})$. It will be convenient to express it as a simple polynomial in $c_x = \cos(q_x a)$ and $c_y = \cos(q_y a)$.

Find where the maximum of $\tilde{J}(\mathbf{q})$ occurs as a function of $(c_x, c_y)$, for $(c_x, c_y) \in [-1, 1]^2$.

For the square lattice, draw a “phase diagram” of the $(J_1, J_2)$ plane indicating the portions where each type of order occurs.

Show that there is never an incommensurate helimagnet phase.

Show that for $|J_1/J_2|$ nonzero but not too large, there is a phase in which the ground state still has extra continuous freedom.

(b). What Hamiltonian do you think would give an incommensurate phase? (You need only two or three nonzero kinds of $J(R)$.)