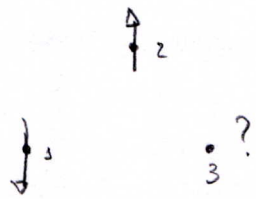
Magnetic frustration

Consider 3 spins in a triangle, interacting anti-ferromagnetically:



Suppose we minimize the 1-2 energy by making them anti-parallel. Then what should be the orientation of spin 3. If it is anti-parallel to 1 it will be parallel to two, and vice-versa. Thus, there is no way we can satisfy all 3 bonds.

This is a typical example of <u>magnetic frustration</u>, an idea which underlies many current topics of research, such as spin ices, spin glasses, spin liquids, high temperature superconductivity and so on.

The hallmark of magnetic frustration is a <u>macroscopic degeneracy</u> of the ground state. In these notes we will learn how to spot these frustrated configurations by making a classical analysis of the Heisenberg model.

These notes were based on a set of lecture notes made by Eric Andrade, from IFSC-USP. Thanks Eric.

Consider the Heisenberg model on an arbitrary lattice

$$\mathcal{H} = -\sum_{i,j} J_{ij}\, \$_i \cdot \$_j \tag{1}$$

where J_{ij} is the interaction between spins i and j (not necessarily nearest neighbors). Due to translation invariance, J_{ij} may depend only on the distance $R_i - R_j$ between the sites. Thus we write

$$\boxed{\mathcal{H} = -\sum_{i,j} J(R_i - R_j)\, \$_i \cdot \$_j} \tag{2}$$

Our goal is to find a method to determine the ground-state approximately for different lattices and interactions. We will do that by replacing the spin operators $\$_i$ with <u>classical vectors</u>. This will be reasonable when the magnitude S of the spin is large.

We can see that by looking at the commutation relations

$$[S^x, S^y] = i S^z \tag{3}$$

Dividing both sides by S^2 we get

$$\left[\frac{S^x}{S}, \frac{S^y}{S}\right] = i \frac{S^z}{S^2} \tag{4}$$

The commutation relations determine the degree of non-classicality of an algebra. In classical physics everything commutes. Looking at (4) we see that when $S \to \infty$ the operators S^x/S and S^y/S commute. Thus, when S is large, we may treat the $\$_i$ as classical vectors.

From quantum mechanics we know that the eigenvalues of $\$^2$ are $S(S+1) \simeq S^2$. Thus, each $\$_i$ will be a vector of magnitude S. Thus, we can parametrize a classical spin $\$$ by a point in a sphere of radius S

$$\begin{aligned} S^x &= S \sin\theta \cos\phi \\ S^y &= S \sin\theta \sin\phi \\ S^z &= S \cos\theta \end{aligned} \tag{5}$$

Now let us go back to the Heisenberg Hamiltonian (2). We define the <u>correlation function</u> as

$$\boxed{C(R_i - R_j) = \langle \$_i \cdot \$_j \rangle} \tag{6}$$

this measures the degree of statistical correlation (either quantum or classical) between spins i and j. If this function tends to a finite value when $R_i - R_j \to \infty$, it means the system has <u>long-range order</u>.

We now move to Fourier space by defining

$$\mathcal{S}_{\vec{q}} = \frac{1}{\sqrt{N}} \sum_i e^{-i \vec{q} \cdot \vec{R}_i} \mathcal{S}_i \tag{7}$$

$$\mathcal{S}_i = \frac{1}{\sqrt{N}} \sum_{\vec{q}} e^{i \vec{q} \cdot \vec{R}_i} \mathcal{S}_{\vec{q}} \tag{8}$$

In the last set of notes we saw that doing this for spin operators completely messed up the algebra. But now there is no algebra because everything is classical!

The Fourier transform of the correlation function is called the structure factors

$$\mathcal{H}(\vec{q}) = \frac{1}{N} \sum_{ij} e^{-i \vec{q} \cdot (\vec{R}_i - \vec{R}_j)} C(\vec{R}_i - \vec{R}_j) \tag{9}$$

Substituting (6) we get

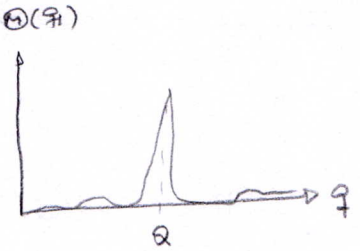
$$\mathcal{H}(\vec{q}) = \frac{1}{N} \sum_{ij} e^{-i \vec{q} \cdot (\vec{R}_i - \vec{R}_j)} \langle \mathcal{S}_i \cdot \mathcal{S}_j \rangle$$

$$= \left\langle \left[\frac{1}{\sqrt{N}} \sum_i e^{-i \vec{q} \cdot \vec{R}_i} \mathcal{S}_i \right] \cdot \left[\frac{1}{\sqrt{N}} \sum_j e^{i \vec{q} \cdot \vec{R}_j} \mathcal{S}_j \right] \right\rangle$$

Thus

$$\mathcal{H}(\vec{q}) = \langle \mathcal{S}_{\vec{q}} \cdot \mathcal{S}_{-\vec{q}} \rangle \tag{10}$$

The structure factor $\mathcal{S}(\vec{q})$ can be obtained experimentally using Neutron scatterings. Similarly to X-Ray diffraction, the structure factor presents sharp peaks at certain wave-vectors $\vec{Q}$



we will now show that these peaks are a signature of the ordered phase and tell us what is the ground-state configuration of the system.

To do that, we return to the Hamiltonian (2) and introduce the Fourier transform (3):

$$\mathcal{H} = -\sum_{ij} J(\vec{R}_i - \vec{R}_j)\, \frac{1}{N} \sum_{\vec{k},\vec{q}} e^{-i(\vec{q}\cdot\vec{R}_i + \vec{k}\cdot\vec{R}_j)}\, S_{\vec{q}} \cdot S_{\vec{k}}$$

this is a calculation we have already done 1000 times

$$\mathcal{H} = -\sum_{\vec{k},\vec{q}} S_{\vec{q}} \cdot S_{\vec{k}}\, \frac{1}{N} \sum_{i,j} J(\vec{R}_i - \vec{R}_j)\, e^{i(\vec{q}\cdot\vec{R}_i + \vec{k}\cdot\vec{R}_j)}$$

define $\vec{R}_\ell = \vec{R}_i - \vec{R}_j$

$$= -\sum_{\vec{k},\vec{q}} S_{\vec{q}} \cdot S_{\vec{k}} \sum_{\ell} J(\vec{R}_\ell)\, e^{i\vec{q}\cdot\vec{R}_\ell} \underbrace{\frac{1}{N} \sum_{j} e^{i(\vec{k}+\vec{q})\cdot\vec{R}_j}}_{\delta_{\vec{k},-\vec{q}}}$$

We define

$$J(\mathbf{q}_1) = \sum_i J(\mathbf{R}_i)\, e^{i\mathbf{k}\cdot\mathbf{R}_i} \tag{11}$$

then we get

$$\mathcal{H} = -\sum_{\mathbf{q}_1} J(\mathbf{q}_1)\, \$_{\mathbf{q}_1} \cdot \$_{-\mathbf{q}_1} \tag{12}$$

Notice how $J(\mathbf{q}_1)$ is exactly the <u>tight-binding dispersion relation</u>.

To find the ground-state of the Hamiltonian, we must now minimize (12) with respect to the variables $\$_{\mathbf{q}_1}$. This is a classical minimization. But it is subject to the constraint that

$$\$_i \cdot \$_i = S^2 \tag{13}$$

using Eq (8) we get

$$\$_i \cdot \$_i = \frac{1}{N} \sum_{\mathbf{q}_1, \mathbf{k}} \$_{\mathbf{q}_1} \cdot \$_{\mathbf{k}}\, e^{i(\mathbf{k}+\mathbf{q}_1)\cdot\mathbf{R}_n} \tag{14}$$

Thus, we have a quite messy minimization in our hands.

Working with the constraint (14) or (13) is quite difficult. Instead, we are going to first work with a less strict constraint and then we impose (13) at the end. The less strict constraint we impose is

$$\frac{1}{N}\sum_i \$_i \cdot \$_i = S^2 \tag{15}$$

then, summing (14) over i we get

$$\frac{1}{N}\sum_i \$_i \cdot \$_i = \frac{1}{N^2}\sum_{q_1,k} \$_{q_1}\cdot\$_k \sum_i e^{i(k+q_1)\cdot R_i}$$

$$= \frac{1}{N}\sum_{q_1} \$_{q_1}\cdot\$_{-q_1}$$

Thus we obtain the constraint

$$\frac{1}{N}\sum_{q_1} \$_{q_1}\cdot\$_{-q_1} = S^2 \tag{16}$$

There is one final piece of information. Going back to (7) or (8), we note that since $\$_i$ is a real vector,

$$\$^*_{q_1} = \$_{-q_1} \tag{17}$$

Thus, we may cast our problem as

$$\text{Minimize} \qquad \mathcal{H} = -\sum_{q} J(q)\,|S_q|^2$$

$$\text{subject to} \qquad \frac{1}{N}\sum_{q} |S_q|^2 = S^2 \tag{18}$$

To minimize $\mathcal{H}$ we should therefore choose those values of q which minimize $-J(q)$. Let us denote those values as Q. Thus, to make $\mathcal{H}$ as small as possible we choose

$$S_q = NS\,\delta_{q,Q} \tag{19}$$

This will satisfy the constraint and minimize $\mathcal{H}$.

The procedure outlined above is called the <u>Luttinger-Tisa</u> <u>method</u>. It tells you what is the configuration in Fourier space which gives the ground-state of the classical Heisenberg model. In a second we will see that magnetic frustration appears when there is a large (or infinite) number of Q vectors minimizing $-J(q)$.

As an example, consider a square lattice with nearest-neighbor interactions. Then, we know from tight-binding that

$$-J(q) = -2J[\cos q_x + \cos q_y] \tag{20}$$

where I set the lattice spacing to 1.

Now assume the interaction is ferromagnetic. That is $J > 0$. Then we make $-J(q)$ small by setting $q = (0,0)$. Thus

$$\boxed{J>0: \quad Q = (0,0)} \tag{21}$$

If $J<0$ (anti-ferromagnetic) then we minimize $-J(q)$ by putting $q = (\pi, \pi)$. Thus

$$\boxed{J<0: \quad Q = (\pi,\pi)} \tag{22}$$

But what is the meaning of these Q values? To understand that let us first look at the constraints (13) and (16). In (16) we get

$$\sum_q \$_q \cdot \$_{-q} = 2\,\$_Q \cdot \$_{-Q} = NS^2 \tag{23}$$

On the other hand, from Eq (8) we have

$$\$_i = \frac{1}{\sqrt{N}} \left[e^{iQ\cdot R_i} \$_Q + e^{-iQ\cdot R_i} \$_{-Q} \right] \tag{24}$$

thus Eq (13) becomes

$$\$_i \cdot \$_i = \frac{1}{N} \left\{ 2\,\$_Q \cdot \$_{-Q} + e^{2iQ\cdot R_i} \$_Q \cdot \$_Q + e^{-2iQ\cdot R_i} \$_{-Q} \cdot \$_{-Q} \right] \tag{25}$$

$$= S^2$$

To satisfy both (23) and (25) we therefore require that

$$\boxed{\$_Q \cdot \$_{-Q} = \frac{NS^2}{2} \qquad \$_Q \cdot \$_Q = 0} \tag{26}$$

Write $\$_Q = A_Q + i B_Q$. Then $\$_{-Q} = \$'_Q = A_Q - i B_Q$. Thus

$$\$_Q \cdot \$_{-Q} = A_Q^2 + B_Q^2 = \frac{NS^2}{2}$$

$$\$_Q \cdot \$_Q = A_Q^2 - B_Q^2 + 2i\, A_Q \cdot B_Q = 0$$

This therefore establishes that

$$\boxed{A_Q^2 = B_Q^2 = \frac{NS^2}{4} \qquad A_Q \cdot B_Q = 0} \tag{27}$$

the direction of A_Q and B_Q is arbitrary, as long as they are perpendicular to each other. This is a consequence of the fact that the Heisenberg interaction is isotropic.

For concreteness, let us then assume that $A_Q \propto \hat{x}$ and $B_Q \propto \hat{y}$.

Then Eq (24) becomes

$$S_i = \frac{S}{2}\left\{ e^{i Q \cdot R_i}(\hat{x} + i\hat{y}) + e^{-i Q \cdot R_i}(\hat{x} - i\hat{y}) \right\}$$

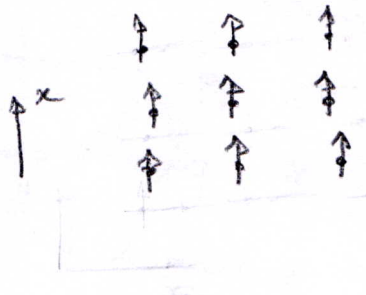
or

$$\boxed{\begin{aligned} S_i^x &= S \cos(Q \cdot R_i) \\ S_i^y &= S \sin(Q \cdot R_i) \\ S_i^z &= 0 \end{aligned}} \qquad (28)$$

This is a Helical spin configuration

Back to the square lattice, Eqs (21) and (22), if $Q = (0,0)$ then

$$S_i^x = S$$
$$S_i^y = 0$$
$$S_i^z = 0$$



This is the ferromagnetic configuration.

If $Q = (\pi, \pi)$ then, writting

$$R_i = n_1 \hat{x} + n_2 \hat{y}$$

we get

$$Q \cdot R_i = \pi (n_1 + n_2)$$

Thus

$$S_i^x = S \cos [\pi (n_1 + n_2)] = S (-1)^{n_1 + n_2}$$

$$S_i^y = 0$$

$$S_i^z = 0$$

This is the anti-ferromagnetic configuration.

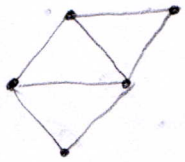
More general Heisenberg interactions will have more complex helical structures given by Eq (28) with different values of Q.

These Helical structures can be determined by neutron scattering, by identifying the peaks in the structure factor $\Theta(q_1)$, pages 4 and 5.

There are a bunch of materials in nature with these complex structures

Example: hexagonal lattice

The hexagonal lattice is a typical example of a frustrated system



In the problem set you compute the tight-binding dispersion relation for this lattice. It reads

$$-J(q_1) = -2J\left\{\cos q_x + 2\cos\frac{q_x}{2}\cos\frac{\sqrt{3}q_y}{2}\right\} \tag{29}$$

Suppose $J>0$ (FM). Then we minimize $-J(q_1)$ by setting $q_1 = (0,0)$. Thus, in the ferromagnetic case the situation is trivial. The situation becomes more interesting for the anti-ferromagnetic case, $J<0$. In this case, if we differentiate $J(q_1)$ with respect to q_x and q_y we get

$$\frac{\partial J(q_1)}{\partial q_x} = 2J\left\{-\sin q_x - \sin\frac{q_x}{2}\cos\frac{\sqrt{3}q_y}{2}\right\} = 0$$

$$\frac{\partial J(q_1)}{\partial q_y} = 2J\left\{-\frac{\sqrt{3}}{2}\cos\frac{q_x}{2}\sin\frac{\sqrt{3}q_y}{2}\right\} = 0$$

Thus we get the two equations

$$\sin q_x + \sin\frac{q_x}{2}\cos\frac{\sqrt{3}\,q_y}{2} = 0 \qquad (30a)$$

$$\cos\frac{q_x}{2}\sin\frac{\sqrt{3}\,q_y}{2} = 0 \qquad (30b)$$

Eq (30b) is satisfyed by $q_y = 0$. Thus Eq (30a) becomes

$$\sin q_x = -\sin\frac{q_x}{2}$$

this has the solution $q_x = 0$, which is the FM case, or the solution

$$q_x = \frac{4\pi}{3}$$

which is the one that interest us in the AFM case. Thus, we conclude that the configuration which minimizes the energy is

$$Q = \left(\frac{4\pi}{3}, 0\right) \qquad \text{(AFM case)} \qquad (31)$$

this is a <u>non-collinear</u> configuration. It is an ordered phase, but with the spins pointing in different directions, separated by 120°. If you plug this in (28) we get the pretty picture in the next page

Example: Kagomé lattice

Finally we discuss the Kagomé lattice. In the problem set you found that this lattice had 3 bands

$$-J_0(q_1) = 2J \qquad (32)$$

$$-J_\pm(q_1) = -2J\left[1 \pm \Delta q_1\right]$$

where

$$\Delta q_1 = \sqrt{3 + 2\cos q_x + 2\cos\left(\frac{\sqrt{3}}{2}q_y - \frac{q_x}{2}\right) + 2\cos\left(\frac{\sqrt{3}}{2}q_y + \frac{q_x}{2}\right)} \qquad (33)$$

If the model is FM the lowest energy comes by making $q_1 = (0,0)$ in $J_+(q_1)$, which gives $-J_+(0,0) = -8J$. Thus, again, nothing interesting in the FM case.

But in the AFM case the minimum occurs for the flat band $-J_0(q_1) = 2J$. Thus, we have here a <u>massive degeneracy</u>: literally every Q vector in the first BZ will have the same energy.

This is one of the signatures of magnetic frustration and it means that there is no ordered configuration. The Heisenberg in the Kagomé lattice is called a <u>spin liquid</u>