Lecture 14: Probes of quantum antiferromagnetism

In this lecture, our main goal is to understand the physics behind some standard probes of antiferromagnetic Mott insulators. It is important to discuss this since, insulators, unlike good or bad metals or superconductors, cannot be studied by applying voltages and measuring currents.

We begin by quickly reviewing the interpretation of standard thermodynamic measurements like specific heat and susceptibility before focussing on two techniques: Nuclear Magnetic Resonance (NMR) and Inelastic Neutron Scattering (INS). Given any material, the most basic thermodynamic measurement (which is not necessarily the easiest to make with good accuracy) is a measurement of its specific heat (heat capacity C divided by volume L^d) as a function of temperature. To interpret the results of such a measurement, we note that

$$C = T\frac{dS}{dT} \tag{1}$$

Thus, a measurement of $C(T)$ immediately gives us information about the entropy of the system by integrating $C(T)/T$ up (numerically) from the lowest temperatures at which measurements are possible. It is important to go to low temperature because the integration constant can then be fixed by using the Third Law of Thermodynamics, which tells us that

$$S(T \to 0) \to 0 \, . \tag{2}$$

However, one needs to remember that the magnetic system we are studying may contribute only a small fraction of the total C—the rest could very well be a crystal effect, *i.e.* contributed by the phonon modes of vibration of the crystal. Therefore, ideally, one should compare the heat capacity of the magnetic material with that of a homologous material with very similar crystal structure and magnetic ion replaced by a non-magnetic ion, and use this comparison to "subtract out" the phonon contribution.

Let us say we have done this, and we obtain a magnetic contribution to the specific heat that is a power law T^p at low temperature. What does this tell us? Well, it basically says that the magnet has low energy bosonic gapless modes. To see this, let us take a gas of free bosons with dispersion

$$\epsilon(k) \sim k^\alpha \tag{3}$$

and work out the temperature dependence of the specific heat. To do this we remember that

$$\frac{E}{L^d} = \int \frac{d^d q}{(2\pi)^d} \frac{\epsilon(q)}{e^{\beta\epsilon(q)} - 1} \tag{4}$$

Here, we have assumed that these bosons can be created and destroyed, like phonons in a crystal, so that their number is not fixed, and have therefore used the Bose-Einstein distribution without a chemical potential. At very low temperature, the integral is dominated by q near zero, and has the following temperature dependence:

$$\frac{E}{L^d} \sim T^{\frac{d}{\alpha}+1} \tag{5}$$

Therefore, the specific heat has the temperature dependence

$$\frac{C}{L^d} \sim T^{\frac{d}{\alpha}} \tag{6}$$

Thus, the specific heat is a power law controlled by the spatial dimensionality and the power α. For instance, a two dimensional system with lineary dispersing gapless excitations will have a T^2 specific heat. In the magnetic system, one obvious candidate for such bosons are the Goldstone modes we introduced earlier in this course. Therefore, one very plausible interpretation of such power law specific heat is that it is the result of spontaneous breaking of a continuous symmetry in the ground state, and the concomitant existence of gapless Goldstone bosons.

On the other hand, if the magnetic specific heat dies away exponentially at low temperature, *i.e.* if

$$\frac{C}{L^d} \sim \exp(-\Delta/T) \, , \tag{7}$$

then this signals the lack of low energy excitations below an energy gap of Δ. In other words, the ground state is separated from all other excited states by a gap. This is typically a signature of a phase in which long-range antiferromagnetic order has been destroyed by quantum fluctuations, and we will see examples of this later in these lectures.

Measurements of the thermodynamic suscepbitility, defined as the ratio of the induced magnetization per unit volume M/L^d in response to a very small external magnetic field B, *i.e.*

$$\chi_{tot} = \frac{1}{L^d} \lim_{B\to 0} \frac{M}{B} \, , \tag{8}$$

can also be interpreted in a similar way. If the suscepbilitity decays exponentially below a temperature scale of Δ, this signals the fact that the ground state carries no spin, and further, that there are no spin-carrying magnetic excitations below an energy gap of Δ. On the other hand, a power law susceptibility implies the presence of gapless spinful excitations.

A more detailed probe of the system is provided by the response of the system to an A.C. (oscillatory) magnetic field that oscillates with frequency ω in time and wavevector $\vec{q}$ in space, for instance

$$\vec{B}(\vec{r},t) \;=\; \hat{z}B\cos(\vec{q}\cdot\vec{r}-\omega t) \tag{9}$$

If B is small enough, such a perturbation leads to a linear response in the system magnetization

$$\vec{M}(\vec{r},t) \;\sim\; \hat{z}M_1\cos(\vec{q}\cdot\vec{r}-\omega t)+\hat{z}M_2\sin(\vec{q}\cdot\vec{r}-\omega t) \tag{10}$$

From our earlier lectures on linear response theory, we know that M_1 and M_2 can be interpreted in terms of the real and the imaginary parts of the corresponding linear response function $\chi(\vec{q},\omega)$ in the following way:

$$\begin{aligned}\chi'_{zz}(\vec{q},\omega) &\sim \frac{1}{L^d}\lim_{B\to 0}\frac{M_1}{B}\\ \chi''_{zz}(\vec{q},\omega) &\sim \frac{1}{L^d}\lim_{B\to 0}\frac{M_2}{B}\end{aligned} \tag{11}$$

To interpret the measured real and imaginary parts $\chi'_{zz}(\vec{q},\omega)$ and $\chi''_{zz}(\vec{q},\omega)$, we can now go back to their spectral representations derived in the earlier lectures on linear response theory:

$$\begin{aligned}\chi''_{zz}(\vec{q},\omega) &= -\frac{i\pi}{Z}\frac{1}{L^d}\sum_{mn}|\langle n|S^z(\vec{q})|m\rangle|^2 e^{-\beta E_n}\left(\delta(\omega-E_{mn})-\delta(\omega+E_{mn})\right)\\ \chi'_{zz}(\vec{q},\omega) &= \Pr\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\chi''_{zz}(\omega')}{\omega'-\omega}\end{aligned} \tag{12}$$

This is written in terms of exact eigenstates $|m\rangle$ with eigenenergies E_m and the matrix elements are those of the operator

$$S^z(\vec{q}) \;\equiv\; \sum_j \exp(i\vec{q}\cdot\vec{r}_j)S^z(\vec{r}_j) \tag{13}$$

which represents the Fourier component of the spin density at wavevector $\vec{q}$.

Thus, a measurement of the frequency, wavevector and temperature dependence of the dynamic susceptibility provides, through its imaginary part, pretty detailed information on the spectrum of spin-carrying excitations that are connected to the ground state by the action of $\vec{S}(\vec{q})$. Compared to specific heat and thermodynamic susceptibility measurements, this is a more detailed way of probing the low-lying spectrum.

In this respect, NMR and inelastic neutron scattering provide even more detailed information, and we will now go through a "theorist's caricature" of these important measurements and their interpretation. Let's first discuss NMR measurements: In the conceptually simplest setting of so-called "continuous wave" or CW measurements of the so-called "Knight-shift", NMR experiments basically have the following set-up: A constant uniform external magnetic field B is applied along the $\hat{z}$ direction. This results in a polarization of the nuclear spins along the $\hat{z}$ direction. To a first approximation, one can ignore the dipolar interactions between nuclear spins, and treat each nuclear spin as a free moment polarized by an external field. Now, one applies a perturbing magnetic field δB_{xy} in the xy plane at frequency ω. At a conceptual level, it is simplest to imagine sweeping this frequency ω (although in actual practice, it is quite common to sweep the magnitude of the static field B).

Now, when the frequency ω equals the resonance frequency $\omega_{\rm res}$, energy $\hbar\omega_{\rm res}$ is absorbed from the the source of the oscillatory perturbing field δB_{xy} and the nuclear spins make a transition that changes the value of the m_z quantum number by 1. Clearly, this resonance frequency is determined by the the Zeeman splitting of the nuclear spin levels as

$$\hbar\omega_{\rm res} \;\; = \;\; g_N \mu_N B + \Delta B_{\rm ex} \tag{14}$$

where g_N is the nuclear gyromagnetic ratio, μ_N is the nuclear magneton, and $\Delta B_{\rm ex}$ represents the magnetic field created at the site of the nuclear spins by the nearby electronic spins (*i.e.* the magnetic moments of the localized electrons of our antiferromagnetic Mott insulator) which are *hyperfile coupled* to the nuclear spins by the hyperfine interaction familiar from your Atomic Physics lectures

$$\Delta B_{\rm ex} \;\; = \;\; A_{\rm hyperfine} \langle S^z \rangle_{\rm loc} \tag{15}$$

where $\langle S^z \rangle_{\rm loc}$ represents the local equilibrium spin polarization of the electronic magnetic moments in response to the static external field B.

Since g_N, μ_N, and $A_{\rm hyperfine}$ are known from other measurements, measurements of the "Knight shift" of the resonance frequency $\omega_{\rm res}$ away from its "bare" value $g_N\mu_N B$ allows us to probe the local magnetization of the electronic spin system *at a nuclear site* as a function of B. In other words, one obtains the local magnetic susceptibility $\chi_{\rm loc}$ at a given site by such Knight shift measurements:

$$\chi_{\rm loc} \;=\; \lim_{B\to 0} \frac{\Delta B_{\rm ex}}{A_{\rm hyperfine} B} \tag{16}$$

Thus, NMR can provide information of the local susceptibility of the system. This is particularly useful in cases where the system is inhomogenous, and the local environment of each nuclear spin is therefore different. In such cases, the resonance line broadens due to so-called "inhomogenous broadening", which can basically be thought of as representing a histogram of local susceptibilities of the electronic spins in various parts of the inhomogenous sample.

There is another quantity that is readily accessed in NMR measurements, and this is the relaxation rate conventionally denoted by $1/T_1$. To understand what this quantity is, it is again useful to consider a "theorist's caricature" of the corresponding measurement protocol: One again starts with a steady external field $B\hat{z}$ that polarizes the nuclear spins to lie along the $\hat{z}$ axis. Then, one applies a so-called π pulse, which is pulse of a field in the xy plane of exactly the right duration to *rotate* all the nuclear spins to the $-\hat{z}$ direction. In other words, we *prepare* the nuclear spins in a excited state which costs Zeeman energy $g_N\mu_N B = \hbar\omega_N$ for each isolated nuclear spin (apart from the Zeeman energy contribution from the exchange field, which we ignore since it will only contribute at higher orders in the $A_{\rm hyperfine}$ to the rate calculation we do below).

Now, we simply watch the relaxation of these excited spins back to their ground state by monitoring the *recovery* of the polarization along the $+\hat{z}$ direction. This recovery is characterized by a rate $1/T_1$ which is directly measured by fitting the nuclear polarization as a function of time.

With this background, let us now ask: What does this rate measure? To answer this, let us work out the Fermi's Golden rule formula for this rate. In order to do this, we must remember that that transitions from this excited state to the ground state of say the nuclear spin $\vec{I}(\vec{r}_0)$ of the magnetic ion at

$\vec{r}_0$ must be caused by the hyperfine coupling

$$H_{\rm hyperfine} \;=\; A_{\rm hyperfine} \sum_j \vec{I}(\vec{r}_j) \cdot \vec{S}(\vec{r}_j) \tag{17}$$

that couples each nuclear spin to the corresponding ionic (electronic) spin. Since each such transition decreases the nuclear Zeeman energy by $\hbar\omega_N$, it must *increase* the electronic energy by the same amount. Furthermore, in this idealized example with a rotationally invariant hyperfine coupling, the z components of the coupling term cannot cause any transitions since they commute with the nuclear Hamiltonian.

Putting all this together, we obtain the Fermi's Golden Rule expression for the transition rate of the nuclear spin $\vec{I}(\vec{r}_0)$:

$$\frac{1}{T_1} \;=\; \frac{2\pi |A_{\rm hyperfine}|^2}{Z} \sum_{nm} e^{-\beta E_n} |\langle m|S^-(\vec{r}_0)|n\rangle|^2 \delta(\omega_N - (E_m - E_n)) \tag{18}$$

where $S^- = S^x - iS^y$, the $|m\rangle$ denote exact eigenstates of the electronic spin system with eigenenergies E_m, and we have gone back to setting $\hbar = 1$.

We can rewrite this expression in terms of the $+-$ component of the Fourier transform of the local spin density correlation function to wavevector and frequency space, *i.e.* in terms of the $+-$ component of the dynamic structure factor of the spins, which is defined as in our lecture on the fluctuation-dissipation theorem:

$$S^{+-}(\vec{q},\omega) \;=\; \frac{2\pi}{Z} \sum_{nm} e^{-\beta E_n} |\langle m|S^-(\vec{q})|n\rangle|^2 \delta(\omega - (E_m - E_n)) \tag{19}$$

where $S^-(\vec{q}) \equiv \sum_j \exp(i\vec{q}\cdot\vec{r}_j) S^-(\vec{r}_j)$. Clearly, the required formula is

$$\frac{1}{T_1} \;=\; |A_{\rm hyperfine}|^2 \int \frac{d^d q}{(2\pi)^d} S^{+-}(\vec{q},\omega_N) \tag{20}$$

Thus, the measured NMR relaxation rate $1/T_1$ directly probes the momentum integral (over the full Brillouin zone) of the dynamic spin structure factor at frequency $\omega = \omega_N$, the nuclear Larmor frequency (in this simple case of an isotropic hyperfine Hamiltonian, it probes the $+-$ component of the structure factor, but more generally, there can be a contribution from the zz component of the structure factor as well).

Finally, let us quickly understand how inelastic neutron scattering can serve as a probe of magnetism. The basic idea is quite simple: A beam of neutrons (from a reactor typically) with a definite energy E (how such a mono-energetic beam is created is a subject for another course on experimental techniques) is directed to the sample-holding station in which a sample is placed in equilibrium with a heat-bath at temperature T (cryostat). These neutrons scatter off the sample, since they interact both with the nuclei (mainly via short-range nuclear forces) as well as with the electronic magnetic moments in the sample via the magnetic dipole interaction between the magnetic dipole moment of the neutron and the magnetic dipole moment of the electronic spins.

There are experimental techniques that allow one to focus exclusively on the magnetic part of the neutron scattering (in effect by "subtracting out" the scattering from the nuclei). By Fermi's Golden rule, the magnetically scattered intensity for neutrons with momentum transfer $\hbar\vec{q}$ and energy transfer $\hbar\omega$ is proportional to the modulus square of the matrix element of the magnetic dipole interaction between the initial and final states. Using initial and final plane-wave states for the neutron beam, and a formal expansion in terms of the exact eigenstates of the electronic spin system for the final states of the electronic system (the initial state is assumed to be an equilibrium state), and the form of the magnetic dipole interaction, this modulus square of the transition matrix element can be related to the equilibrium dynamic structure factor of the electronic spin system at wavevector q and frequency ω (in a manner completely analogous to our earlier Fermi's Golden Rule analysis of $1/T_1$).

Thus, a measurement of the inelastic neutron scattering intensity $I(\vec{q},\omega)$ over a range of energy transfers and momentum transfers provides a sort of map of the frequency and wavevector dependence of the dynamic structure factor:

$$I(\vec{q},\omega) \quad \propto \quad \sum_{\alpha\beta}(\delta_{\alpha\beta} - \hat{q}_\alpha\hat{q}_\beta)S^{\alpha\beta}(\vec{q},\omega)\ . \tag{21}$$

Here, $\hat{q}_\alpha = q_\alpha/|\vec{q}|$, and this prefactor's dependence on the direction of the momentum transfer is completely dictated by the form of the magnetic dipolar interaction.