Understanding of Spin Glass from Susceptibility Measurement

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Abstract

Materials that are consist of disordered and frustrated magnetic spin alignment with complicated energy landscape are called spin glasses. This kind of magnetic material is first realized in susceptibility measurement. In this paper, I will discuss about three important magnetic behaviors observed in different susceptibility experiments and the unique properties we learn from these experiment. Then some theory models that used to describe spin glass will be mentioned as followed.

1 Introduction

In 1972, Cannella and Mydosh found sharp cusps in low frequency ac-susceptibility measurement respected to temperature of AuFe in low magnetic field environment [11]. Their experiments showed that the temperature at which cusps occurred depended on the concentration of Fe (see fig1). As the first evidence on the existence of a new kind of magnetic materials [1], these experiments attracted people's attention to study this kind of material both experimentally and theoretically for tens of years. This paper will focuses on this new type magnetic material which is now called spin glass. As an experimental student, I'd like to start with presenting experiments that show unique magnetic behavior of spin glasses: frequency-dependence ac-susceptibility, irreversible dc-susceptibility in zero-field cooling, and waiting-time dependence relaxation rate. Then I will use these experiment results to discuss about basic properties of spin glasses as well as Edwards and Anderson (EA) model and Sherrington-Kirkpatrick (SK) model that are used to help people understand spin glass state of materials.

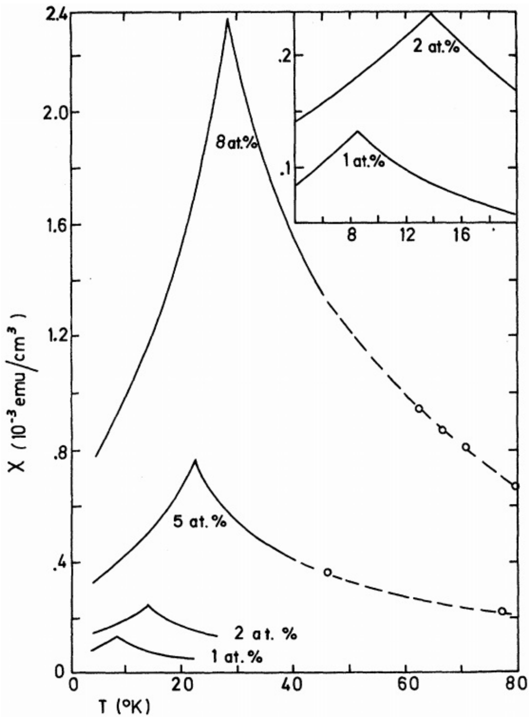

Figure 1: ac susceptibility have cusps with various of magnetic impurities concentration [11]

2 Susceptibility Experiments on Spin Glasses

There are many experiments have been conducted to characterize spin glass system. One of the earliest experiments, as we mentioned, is the susceptibility measurement. The susceptibility measurement not only helps people realize the existence of spin glass phase transition, but also help people to distinguish spin glasses from other magnetic materials.

One example will be the comparison between spin glasses and superparamagnets. One can think of superparmagnets as collections of magnetic nanoparticles and each particle can choose its magnetic moment alignment to be any direction as it want and can flip at a rate depending on temperature. Similar to paramagnet, superparamagnets have zero net magnetization. And it can be magnetized by an external magnetic field but with a much larger susceptibility than paramagnets. One way can people tell the difference between superparamagnets and spin glasses is by measuring the frequency dependence of the susceptibility cusps as shown in Mulder, Duyneveldt and Mydosh's paper [3].

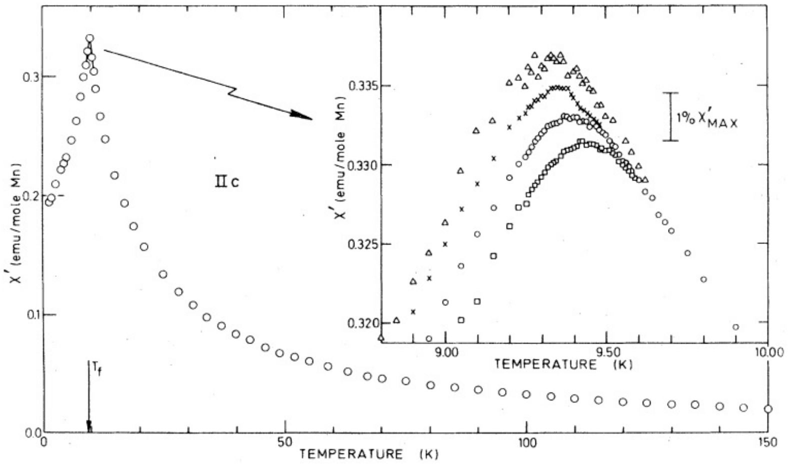

Figure 2: Zero-field susceptibility X' as a function of temperature. Measuring frequencies: □ 1.33 kHz, ○ 234 Hz; × 10.4Hz; and △ 2.6 Hz. [3]

In fig 2, the ac-susceptibility is measured on quenched CuMn, another example of spin glass material, with low Mn concentration. The main panel shows the real part of the ac-susceptibility as a function of temperature in a wide range and the insert panel is a closer look near the cusp. Here we can see that as frequency sweep from 1 Hz to 10 kHz, the $\Delta T_f/(T_f ln(f))$ shifts by about 0.005 where T_f is the freezing temperature of spins. In this paper they preformed the same frequency dependence measurement on high Mn concentrated CuMn alloy, but didn't observe similar result. On the other hand, similar experiments have been done for superparamagnets and temperature shift is about two orders larger than in spin glasses [2]. One should also notice that at high temperature regime (above T_f), the susceptibility curve fitted pretty good with a the Curie-Weiss Law:

$$\chi = C/(T - \Theta) \tag{1}$$

Where C and Θ are Curie constant and Curie temperature. This shows that after spin glass phase transition, CuMn becomes paramagnetic and the experimental result is reliable.

Another unique feature about spin glasses is that its irreversible magnetic behavior after non-zero field cooling. Nagata, Keesom, and Harrison reported the measurement dc-susceptibility on CuMn for two different processes: zero-field cooling (ZFC) and field cooling (FC), and observed different magnetic behavior [4] as shown in figure 3.

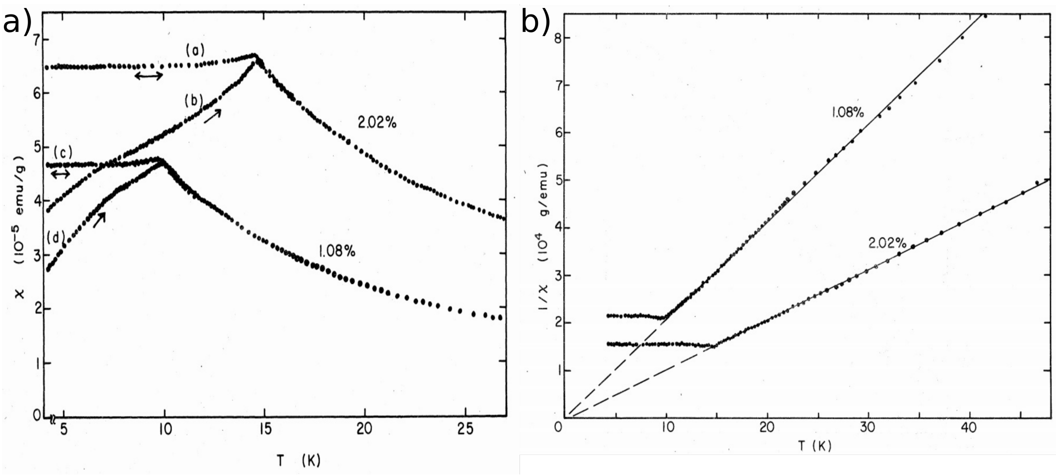

Figure 3: Left: dc-susceptibility measurement after field cooling (a and c) and zero field cooling (b and d). Right:Inverse susceptibility as a function of temperature.The dashed line showed the Curie-Weiss law prediction

Again, at high temperature we see a good agreement with the Curie law in the figure on the right. But at lower temperature region, its magnetic behavior depends on the cooling history of the sample: if the sample is cooled down to T_f without an external magnetic field, the measured susceptibility below T_f is irreversible; while cooling with a nonzero external magnetic field, the susceptibility measurement is reversible and almost independent of temperature. We will come back to this experiment later.

The last experiment that I'd like to discuss here is the susceptibility and its corresponding relaxation rate as a function of measuring time at a fixed temperature below T_f of quenched spin glasses reported by Sandlund and his coworkers [5]. The alloy of Cu with 10% Mn is quenched in zero-field-protocol and susceptibility is measurement in the enviornment of magnetic field aftering some waiting time.Here the waiting time is defined as the time gap between the moment at which samples is quenched down to designated temperature and the moment when data is collected. What surprises people is that the relaxation rate (the change of susceptibility respected to time) has waiting time dependence as shown in fig4. This waiting-time effect now is considered as one unique characteristic of spin glasses [1].

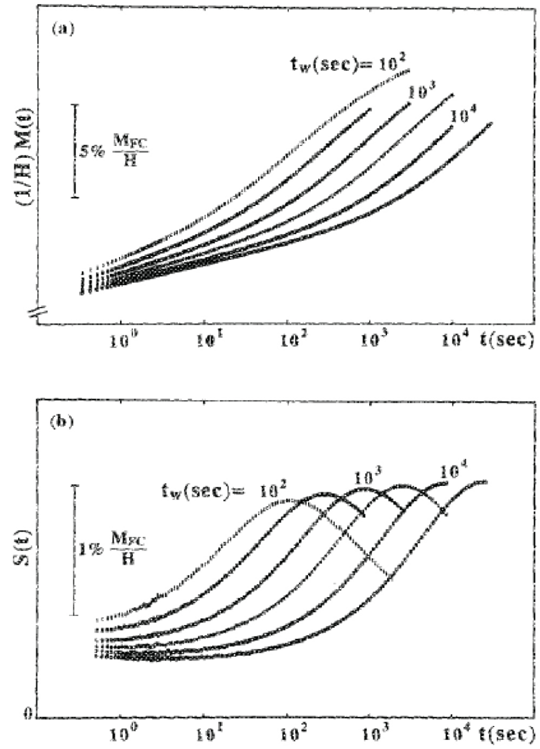

Figure 4: Zero-field-cooled susceptibility $(1/H)M(t)$ and the corresponding relaxation rate $S(t) = (l/H)\partial M/\partial In(t)$ of at different wait times $t_w = 10^2, 3x10^2, 10^3, 3x10^3, 10^4, 3x10^4$s plotted vs $ln(t)$

3 Basic Properties about Spin Glasses

What do we learn about spin glasses from these experiments that we discussed above? First of all, let's think of the materials. Both AuFe and CuMn are noble metals (Au,Cu, etc.) diluted with transition metals as Fe and Mn in here. These transition metal ions act like magnetic impurities in the alloys. At low concentration, these transition metal ions in the alloys' structure don't occupied periodic site. Instead, they distribute randomly, which provides the first important characteristic of spin glasses: randomness.

Indeed, the interaction between these magnetic impurities makes these special alloys become spin glass. But how can the magnetic impurities interact with each other in alloys? The answer is through conduction elections [6]. According to the Kondo effect, conducting electrons are scattered by magnetic impurities through exchange interaction, because of the spin of electrons and magnetic impurities. One electron can interact with many magnetic ions and effectively pass these interaction between different magnetic ions like a interaction bridge. This model is so-call RKKY interaction and has distance dependence as followed:

$$J(\mathbf{R}) = J_0 \frac{cos(2k_F\mathbf{R} + \psi_0)}{(k_F\mathbf{R})^3} \tag{2}$$

where J_0 and ψ_0 are constants, k_F is the Fermi wave number and $\mathbf{R}$ is the distance between two magnetic ions. One can plot the interaction (fig5)and see it alternates between position and negative: when the interaction is positive, two spins tend to be parallel to each other and form ferromagnetic bond; otherwise they tend to form antiferromagnetic bond and align in opposite direction. Since magnetic impurities or ions are randomly distributed, they can have either positive or negative. At low concentration, the distance between any pair ions are large so the positive and negative interactions have approximately equal strength as well as probability. As a result, the ferromagnetic and antiferromagnetic bonds are competing with each and system is frustrated. However, at high concentration, the antiferromagnetic interaction is smaller because the distance between ions will be suppressed and the system is no longer frustrated. This is a simply thought about why the frequency-dependence susceptibility cusp is not observed in high Mn concentrated CuMn alloy.

In terms of the energy landscape, both ferromagnets and antiferromagnets have long-range order ground state or a well-defined global minimum. On the other hand, the frustration of spin glasses introduce complexity into their

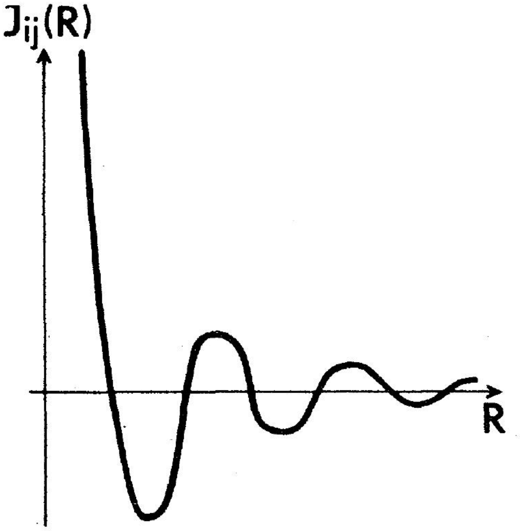

Figure 5: RKKY interaction as a function of distance [6]

energy landscapes [1]. At high temperature, the system is in paramagnetic phase and all spins are fluctuating; while at temperature below T_f, there are many metastable states with high energy barrier between them as drawn in figure 6. Let's think about how can this energy picture apply to previous experiment results. For the dc-susceptibility measurement, when there is not external field presented during cooling process, we may assume all energy valley to be similar with each other and the system can be tracked in either one. As heating up the sample again, it may not followed the same path to escape from the valley, thus the susceptibility is irreversible. However, if there is an external magnetic field while cooling, the energy landscape will be biased.Thus the system can only fall into the valley which has the lowest energy influenced by the magnetic field. For the waiting time effect, one explanation is that by manipulating the quench rate, the energy barrier heights are shuffled so they system need different amount to time to relax depending on how much you changed the barrier heights [8].

4 Theoretical Concepts for Spin Glasses

We shall now discuss about theory work about spin glasses. Experimentalists and theorists always work together to discover new physics. Sometime theorists propose a model and guide experimentalists to prove it, such as the

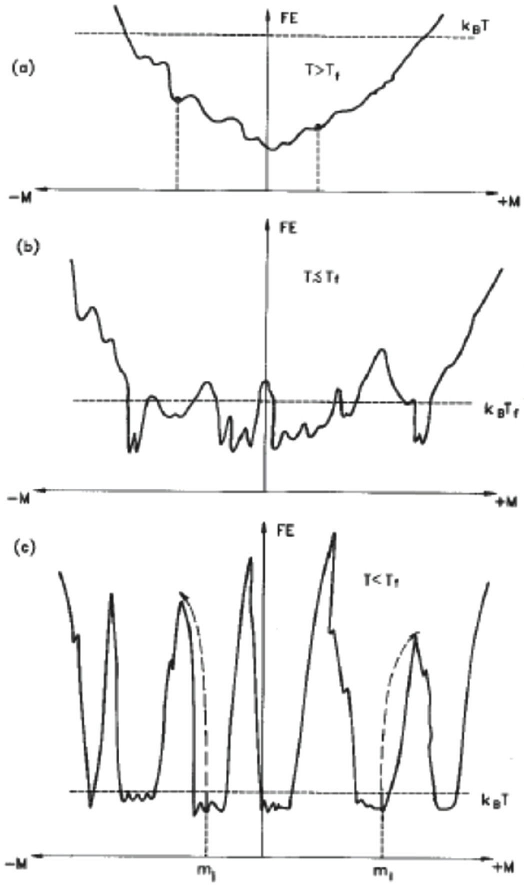

Figure 6: A cartoon of multi-valley landscape of spin glasses [1]

gravitational wave. Sometime experimental people observe some interesting phenomenon first, and theory people work to explain it later, such as superconductivity. Spin glass system is the second case. After the some early experiment results were published, theorists have propose several models in order to understand this new kind of magnetic phase. In here I will only discuss some early and simply example that closely related to the experiments that we have talk about.

Edwards and Anderson (EA) proposed one of the earliest model to describe spin glasses. They believed that spin glasses have a ground state in which all spins aligned in a definite directions even these directions might seem to be random [7]. Like ferromagnets and antiferromagnets both have a order parameters to describe their long-range ordered ground state, Edwards and Anderson defined an order parameter q_{EA} for spin glasses. They started from the Hamiltonian of an Ising model:

$$H = -\sum J_{ij} s_i s_j \tag{3}$$

Where J_{ij} is the RKKY interaction and $s_{i/j}$ is the spin moment. As we said above, in the limit of dilute magnetic ions, the interaction has about equal probability to be positive or negative. Thus $[J_{ij}]_{av} = 0$. At high temperature, $< s_i >= 0$ and $M \equiv \frac{1}{N}\sum_{i=1}^{N} < s_i >= 0$, while at temperature below T_f, the net magnetization $M = 0$ still holds but $< s_i > \quad = 0$ where $< \cdots >$ means average over all configurations. Thus the order parameter of spin glasses is defined as:

$$q_{EA} = \lim_{t\to\infty} \lim_{N\to\infty} [s_i(t_0) s_i(t_0 + t)] \tag{4}$$

The EA model explained the existence of cusp in the susceptibility measurement: at the freezing temperature, spins tend to get frozen along its preferred direction and thus they don't response to applied magnetic field like pure paramagnets or obey the Curie's law. The EA model also predicted that the susceptibility is quadratic in T at low temperature and approaches to non-zero value as temperature goes to 0. However, since this EA order parameter needs to average over all configuration, it will become zero unless the system is trapped in one energy valley in figure6. Therefore, this description is non-equilibrium and short-time. In order to find the long-time and equilibrium description, all valleys need to be scanned and the order parameter shouldn't go to zero when averaging over all valleys.

Followed from the EA model, Sherrington and Kirkpatrick (SK) solved this problem by proposed another so-called Ising mean-field model which took an infinite-range interaction between all spins into account [9]. In order to deal with infinite range, the SK model use a replica trick. The trick is to consider states with the same level of disorder to be replicas of each other. In order words, all energy valleys in fig6 are replicas and they all have the same order parameter. Thus averaging over all configurations will not result in zero order parameter. The order parameter of SK model have very good agreement of the dc-susceptibility experiments [4].

However, this SK model was shown to have negative entropy, thus unstable, by de Almeida and Thouless [1]. In this paper I will not go through details of de Almeida and Thouless is model. But soon later Parisi found the stable spin glass phase transition by breaking the replica symmetry of the SK model. A Previous student who took this course has written a term paper about replica symmetry breaking. Those of you who interested should take a look [10].

5 Conclusions

In conclusion, I present three susceptibility experiments on spin glasses: ac-susceptibility, frequency dependence dc-susceptibility, and waiting time dependence susceptibility relaxation rate. From these three experiments, I go through the basic properties of spin glasses: randomness and frustration. In the last section, I briefly talk about EA model and SK model as they both have good agreement with experimental results I discussed.

As you may have noticed, these experimental and theoretical works about conventional spin glasses were done more than 30 years ago. Nowadays there are still many scientists working on spin glasses system such as quantum spin glasses, which unfortunately have not be experimental realized yet, or other materials show glassy behavior: such as some high temperature superconductors like Cuprates. As a novice of this topic, my paper focuses on very basic and simple aspects of spin glasses. If you are interested about this topic and more recent study, I highly recommend you to read Mydosh's Reports on Progress in Physics published in 2015.

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