AC magnetic susceptibility

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(Date: September 12, 2009)

Abstract

This note is written for experimentalists of the condensed matter physics, who want to know the fundamental physics of the AC magnetic susceptibility. Experimentally, in recent days one can easily measure the AC magnetic susceptibility of various magnetic systems, using the SQUID magnetometer (such as the equipment from Quantum Design). However, it is sometime hard even for many professional researchers to figure out the physics of the magnetic behavior. We have been studying the magnetic phase transitions of low-dimensional magnetism, including spin glasses for many years. This note is based on our experience. In spite of many reviews on the AC magnetic susceptibility, it seems that there are few reviews on the physics of the AC magnetic susceptibility on the magnetic system such as random spin systems (spin glasses, superspin glasses, superparamagnets, quasi 2D ferromagnets, and so on). It is hoped that this note will be useful for experimentalists and students to understand the physics of magnetism.

In order to investigate the magnetism of the matters, it is important to observe directly the magnetic response of the magnetic systems by the application of external magnetic field. Immediately after the application of the magnetic field, the state of the system remains unchanged. After some characteristic times, the state of the system will reach a new state in thermal equilibrium, leading to the new magnetization. This is called as a magnetic relaxation phenomenon. In order to examine the mechanism, we need to know the magnetization in thermal equilibrium and also need to know the information concerning the relaxation process into the state in thermal equilibrium. The dynamic magnetization measurement is required for one to understand the time dependence of magnetic correlations. There are many methods for this purpose, including the inelastic magnetic neutron scattering, the relaxation rate measurement of the zero-field-cooled susceptibility, and the AC magnetic susceptibility. The most suitable dynamic methods should be chosen, depending on the properties of the systems (spin glasses, ferromagnetism, superspin glass, superparamagnetism), the condition of the observation, and the physical quantities derived. In this note, we present the physics on the AC magnetic susceptibility and the magnetic neutron scattering.

Contents

CONCLUSION
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1.   Simple theory of AC magnetic susceptibility

Suppose that the AC magnetization $m_{ac}(t)$ is generated in the magnetic system, when the AC magnetic field $h_{ac}(t)$ is applied to it. Here $m_{ac}(t)$ and $h_{ac}(t)$ are expressed in terms of the complex numbers as

$$h_{ac}(t) = \text{Re}[he^{i\omega t}]$$
$$m_{ac}(t) = \text{Re}[m(\omega)e^{i\omega t}] \tag{1.1}$$
$$\frac{dm_{ac}(t)}{dt} = \text{Re}[i\omega m(\omega)e^{i\omega t}],$$

where ω is the angular frequency, Re denotes the real part of the complex number, and $\chi(\omega)$ is the complex AC magnetic susceptibility. For convenience, we assume that h is real. The complex number $m(\omega)$ is related to h by the relation

$$m(\omega) = \chi(\omega)h = [\chi'(\omega) - i\chi''(\omega)]h, \tag{1.2}$$

where $\chi'(\omega)$ is called the dispersion and $\chi''(\omega)$ is called the absorption. Then the time dependence of $h_{ac}(t)$ and $m_{ac}(t)$ is expressed by

$$h_{ac}(t) = h\cos(\omega t), \tag{1.3}$$

and

$$\begin{aligned}
m_{ac}(t) &= \text{Re}[\{\chi'(\omega) - i\chi''(\omega)\}he^{i\omega t}] \\
&= \text{Re}[\{\chi'(\omega) - i\chi''(\omega)\}h\{\cos(\omega t) + i\cos(\omega t)\}] \\
&= \chi'(\omega)h\cos(\omega t) + \chi''(\omega)h\sin(\omega t) \\
&= \chi'(\omega)h\cos(\omega t) + \chi''(\omega)h\cos(\omega t - \frac{\pi}{2}),
\end{aligned} \tag{1.4}$$

where $\chi'(\omega)$ is also called the in-phase component and $\chi''(\omega)$ is called the out-of-phase component. The power absorbed by the system is given by

$$P_{avg} = \frac{1}{T}\int_0^T h_{ac}(t)\frac{dm_{ac}}{dt}dt$$

$$= \frac{1}{T}\int_0^T (\frac{he^{i\omega t} + h^* e^{-i\omega t}}{2})[\frac{i\omega m(\omega)e^{i\omega t} - i\omega m^*(\omega)e^{-i\omega t}}{2}]dt$$

$$= \frac{1}{4T}T[i\omega h^* m(\omega) - i\omega h m^*(\omega)]$$

$$= \frac{1}{4}2\,\mathrm{Re}[-i\omega h m^*(\omega)] \qquad\qquad (1.5)$$

$$= \frac{1}{2}\omega\,\mathrm{Re}[-ih(\chi' + i\chi'')h^*]$$

$$= \frac{1}{2}\omega|h|^2\,\mathrm{Re}[-i(\chi' + i\chi'')]$$

$$= \frac{1}{2}\omega|h|^2\chi''$$

where T is the period and is given by $T = 2\pi/\omega$. Then the power absorbed by the system is proportional to the absorption χ''.

((**Units**))

We have a comment on the cgs units of the AC magnetic susceptibility. The unit of $m(\omega)$ is emu (=erg/Gauss), where Gauss = Oe, while the unit of h is Oe. Then the unit of $\chi(\omega)$ is emu/Oe. Conventionally we use simply emu instead of emu/Oe as the unit of $\chi(\omega)$. When the weight of the system is given by the unit of g, the unit of the resultant susceptibility is given by emu/g. When the number of a specific magnetic atom is given by the unit of moles, the unit of the resultant susceptibility is emu/mole.

((**Experiment**))

We use the AC susceptibility option of MPMS SQUID magnetometer (Quantum Design) for the measurements of AC magnetic susceptibility, at the Department of Physics, SUNY at Binghamton. The frequency ranges between 0.01 Hz and 1000 Hz. When f = 0.01 Hz is used, it may take 2 hours for each measurement. The amplitude of the AC magnetic field (h) is between 1 mOe and 4.0 Oe. One can simultaneously and directly measure the in-phase component [h $\chi'(\omega)$] in the units of emu and the out-of-phase component [h $\chi''(\omega)$] in the units of emu. The remnant magnetic field is less than 1 mOe using the ultralow field capability option.

2. Debye relaxation with a single relaxation time[1-5]

We consider a Debye relaxation with a single relaxation time τ. We start with the Casimir-du Prè equations[4] given by

$$\chi(\omega) = \frac{\chi_T - \chi_S}{1 + i\omega\tau} + \chi_S \quad , \tag{2.1}$$

where

$$\chi(\omega = 0) = \chi_T - \chi_S + \chi_S = \chi_T = \chi_0$$
$$\chi(\omega = \infty) = \chi_S = \chi_\infty \tag{2.2}$$

Here $\chi_S (= \chi_\infty)$ is the adiabatic susceptibility and $\chi_T (= \chi_0)$ is the isothermal susceptibility. These susceptibilities are defined by

$$\chi_S = (\frac{\partial M}{\partial H})_S = \chi_\infty$$
$$\chi_T = (\frac{\partial M}{\partial H})_T = \chi_0 \tag{2.3}$$

where M is the DC magnetization of the system and H is the DC magnetic field. Then we have

$$\chi(\omega) = \frac{\chi_0 - \chi_\infty}{1 + i\omega\tau} + \chi_\infty$$
$$= \frac{\chi_0 - \chi_\infty}{1 + \omega^2\tau^2}(1 - i\omega\tau) + \chi_\infty \tag{2.4}$$
$$= \chi_\infty + \frac{\chi_0 - \chi_\infty}{1 + \omega^2\tau^2} - i\frac{(\chi_0 - \chi_\infty)\omega\tau}{1 + \omega^2\tau^2} \quad .$$

The dispersion and absorption are obtained as

$$\chi' = \chi_\infty + \frac{\chi_0 - \chi_\infty}{1 + \omega^2\tau^2}$$
$$\chi'' = \frac{(\chi_0 - \chi_\infty)\omega\tau}{1 + \omega^2\tau^2} \quad . \tag{2.5}$$

We assume that $\chi_\infty = 0$ (high frequency limit of susceptibility). Then we get

$$\frac{\chi'}{\chi_0} = \frac{1}{1 + \omega^2\tau^2}$$
$$\frac{\chi''}{\chi_0} = \frac{\omega\tau}{1 + \omega^2\tau^2} \quad . \tag{2.6}$$

We note that $\chi' = \chi_0$ and $\chi'' = 0$ in the limit of $\omega \to 0$. Figure shows the plots of χ'/χ_0 and χ''/χ_0 as a function of $\omega\tau$. At $\omega\tau = 1$, the absorption has a maximum, while the dispersion has an inflection point. The maximum of the absorption provides a method of determining an average relaxation time.
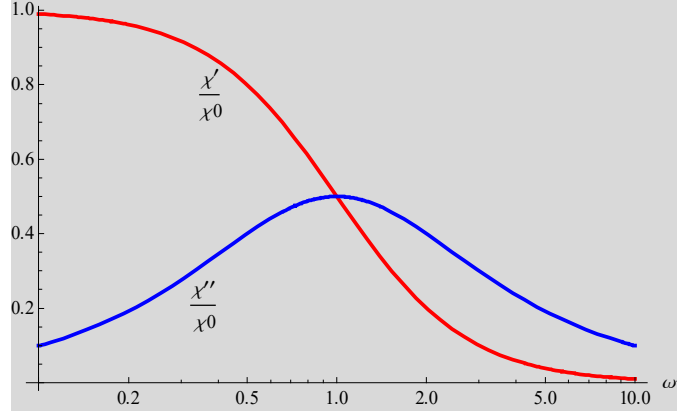
Fig.1 Plot of χ'/χ_0 and χ''/χ_0 as a function of x (= $\omega\tau$) for the Debye relaxation.

From the above equation, we find that χ'/χ_0 and χ''/χ_0 satisfy the equation

$$\left(\frac{\chi'}{\chi_0} - \frac{1}{2}\right)^2 + \left(\frac{\chi''}{\chi_0}\right)^2 = \left(\frac{1}{2}\right)^2 \tag{2.7}$$

.

The Argand diagram (the plot of real part and imaginary part of $\chi(\omega)$ in the x-y plane ($x = \chi'/\chi_0$ and $y = \chi''/\chi_0$,) shows a circle (radius $1/2$) centered at $(1/2, 0)$ in the plane of χ'/χ_0 vs χ''/χ_0. This plot is called a Cole-Cole plot. The deviation of the Cole-Cole plot from a semicircle suggests that the system does not follow the Debye relaxation. This indicates that there are more than two relaxation times in the system.
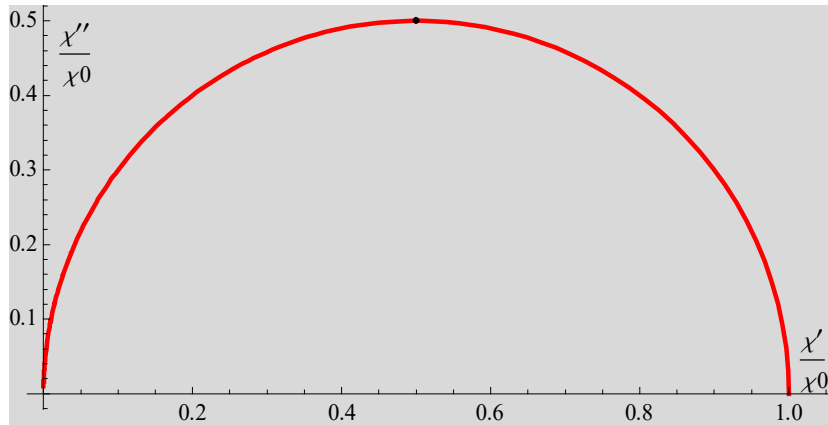

Fig.2 Cole-Cole plot of the real part χ'/χ_0 (= x) vs the imaginary part χ''/χ_0 (= y) for the Debye relaxation.

((Note))   **Definition**
(1)   **Argand diagram**
    A way of representing complex numbers as points on a coordinate plane, also known as the Argand plane or the complex plane, using the x-axis as the real axis and the y-axis as the

imaginary axis. It is named for the French amateur mathematician <u>Jean Robert Argand</u> (1768-1822) who described it in a paper in 1806.

(2)   Cole-Cole Plot

The Cole-Cole plot presents complex material parameters such as impedance or permittivity of dielectric materials as a locus in the Gaussian number plane as a function of frequency. The Cole-Cole diagram is named after two brothers, <u>Kenneth S. Cole and Robert H. Cole,</u> in 1931, the experimental investigations on the impedance of biological tissue conducted.

3.   Relaxation with a distribution of relaxation times

Suppose that there is a broad distribution of the relaxation time τ between τ_{min} and τ_{max}. In this case we need to modify the Casimir-du Prè equations.[4] The probability distribution of relaxation times $g(\tau)$ is supposed to be slowly varying in $\ln \tau$. Let the number of magnetic entities with relaxation times between $\ln \tau$ and $\ln \tau + d\ln \tau$ be $g(\tau)d\ln \tau$. According to Lundgren et al.(1981)[6] and Wenger (1986)[7] (developed for spin glasses) the dispersion χ' can be described by

$$\chi'(\omega) = \frac{1}{h} \int_{\tau_{min}}^{\tau_{max}} \frac{m_0(\tau)}{1+(\omega\tau)^2} g(\tau)d\ln \tau \qquad (3.1)$$

with the normalization condition

$$\int_{\tau_{min}}^{\tau_{max}} g(\tau)d\ln \tau = 1 \qquad (3.2)$$

where τ_{min} and τ_{max} are the minimum and maximum values of relaxation times in the system, and $m_0(\tau)$ is the magnetic moment of clusters. In the following we denote the system to be in the state when $\tau_{min} \ll \tau_m \ll \tau_{max}$, where $\tau_m = 1/\omega$.

$$\frac{\partial \chi'(\omega)}{\partial \ln \omega} = -\frac{1}{h} \int_{\tau_{min}}^{\tau_{max}} \frac{2m_0(\tau)\chi_0(\omega\tau)^2}{[1+(\omega\tau)^2]^2} g(\tau)d\ln \tau \qquad (3.3)$$

Suppose that $g(\tau)$ of $\ln \tau$ is approximately constant in a range $(\ln \tau_{min}, \ln \tau_{max})$. Then $g(\tau)$ in the integrant may be replaced by $g(\tau_m)$. Then we have

$$\begin{aligned}
\frac{\partial \chi'(\omega)}{\partial \ln \omega} &= -\frac{1}{h} \int_{\tau_{min}}^{\tau_{max}} \frac{2m_0(\tau)(\omega\tau)^2}{[1+(\omega\tau)^2]^2} g(\tau)d\ln \tau \\
&= -\frac{2}{h} g(\tau_m)m_0(\tau_m) \int_{\tau_{min}}^{\tau_{max}} \frac{(\omega\tau)^2}{[1+(\omega\tau)^2]^2} d\ln(\omega\tau)) \qquad (3.4) \\
&= -\frac{2}{h} g(\tau_m)m_0(\tau_m) \int_{\tau_{min}}^{\tau_{max}} \frac{(\omega\tau)}{[1+(\omega\tau)^2]^2} d(\omega\tau)
\end{aligned}$$

or

$$\frac{\partial \chi'(\omega)}{\partial \ln \omega} = \frac{1}{h} m_0(\tau_m) g(\tau_m) [\frac{1}{1+(\omega\tau_{max})^2} - \frac{1}{1+(\omega\tau_{min})^2}]$$

$$\approx -\frac{1}{h} m_0(\tau_m) g(\tau_m) \qquad (3.5)$$

The absorption is also evaluated as

$$\chi''(\omega) = \frac{1}{h} \int_{\tau_{min}}^{\tau_{max}} \frac{m_0(\tau)\omega\tau}{1+(\omega\tau)^2} g(\tau) d\ln\tau$$

$$= \frac{1}{h} m_0(\tau_m) g(\tau_m) \int_{\tau_{min}}^{\tau_{max}} \frac{\omega\tau}{1+(\omega\tau)^2} d\ln\tau \qquad (3.6)$$

$$= \frac{1}{h} m_0(\tau_m) g(\tau_m) \int_{\tau_{min}}^{\tau_{max}} \frac{\omega\tau}{1+(\omega\tau)^2} d\ln(\omega\tau),$$

or

$$\chi''(\omega) = \frac{1}{h} m_0(\tau_m) g(\tau_m) \int_{\tau_{min}}^{\tau_{max}} \frac{1}{1+(\omega\tau)^2} d(\omega\tau)$$

$$= \frac{1}{h} m_0(\tau_m) g(\tau_m) [\arctan(\omega\tau_{max}) - \arctan(\omega\tau_{min})] \qquad (3.7)$$

$$\approx \frac{\pi}{2} \frac{1}{h} m_0(\tau_m) g(\tau_m) .$$

Then we have a so-called the $\pi/2$ relation

$$\chi''(\omega) = -\frac{\pi}{2} \frac{\partial \chi'(\omega)}{\partial \ln \omega} . \qquad (3.8)$$

which is a very useful relation for the analysis of experimental data.

4.   Yoshimitsu-Matsubara method

It is known that for many dielectric substances, the presence of the distribution of relaxation times can be described by a fairly simple empirical law. We consider the case when the Cole-Cole plot is deviated from the circle. According to Yoshimitsu and Matsubara (1968),[8] the complex susceptibility may be described by

$$\chi(\omega,\tau) = \frac{\chi_0}{1+(i\omega\tau)^\beta}, \qquad (4.1)$$

where β is between 0 and 1. The real part (dispersion) is obtained as

$$x = \frac{\chi'(\omega,\tau)}{\chi_0} = \frac{1+(\omega\tau)^\beta \cos(\frac{\beta\pi}{2})}{1+(\omega\tau)^{2\beta} + 2(\omega\tau)^\beta \cos(\frac{\beta\pi}{2})} . \qquad (4.2)$$

The imaginary part (absorption) is obtained as

$$y = \frac{\chi''(\omega,\tau)}{\chi_0} = \frac{(\omega\tau)^\beta \sin(\frac{\beta\pi}{2})}{1+(\omega\tau)^{2\beta} + 2(\omega\tau)^\beta \cos(\frac{\beta\pi}{2})} \tag{4.3}$$

The susceptibility can be rewritten in a form given by

$$\chi(\omega,\tau) == \frac{\chi_0}{1+(i\omega\tau)^\beta} = \int \frac{\chi_0}{1+i\omega\tau_1} G(\tau_1) d\tau_1 \tag{4.4}$$

where $G(\tau_1)$ is the distribution function of the relaxation times. An explicit form of $G(\tau_1)$ is given by Yoshimitsu and Matsubara[8] as

$$y(x) = \tau G(\tau_1) = \frac{\sin(\beta\pi)}{\pi x[x^\beta + x^{-\beta} + 2\cos(\beta\pi)]}, \tag{4.5}$$

with $x = \tau_1/\tau$. Figures 3 and 4 show a plot of $y(x)$ as a function of x. The parameter β is a measure of the width of the distribution. For $\beta = 1$, $y(x)$ is a Dirac delta function and gives the Debye relaxation. As β is decreased, the peak of the distribution moves toward smaller x and finally $y(x)$ concentrates around $x = 0$ ($\beta \leq 1/2$). Figure 5 shows the Cole-Cole plot of x vs y, where $x = \chi'(\omega,\tau)/\chi_0$ and $y = \chi''(\omega,\tau)/\chi_0$. It is found that the circle with $\beta = 1$ is distorted as β is decreased.
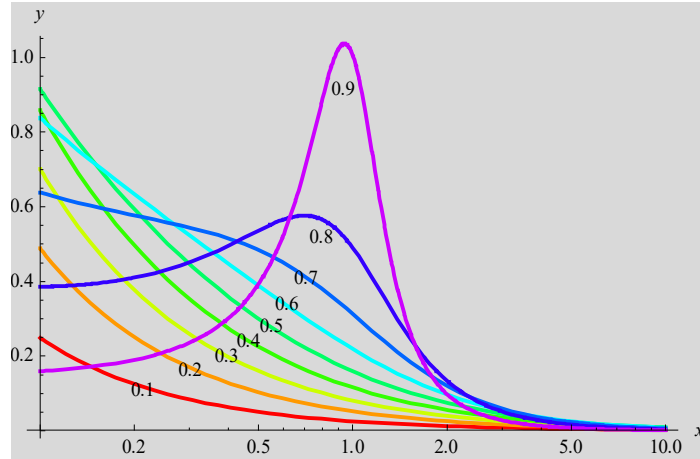

Fig.3 Plot of the distribution function of the relaxation, $y(x)$ as a function of x ($= \tau_1/\tau$) for $\beta = 0.1 - 0.9$.
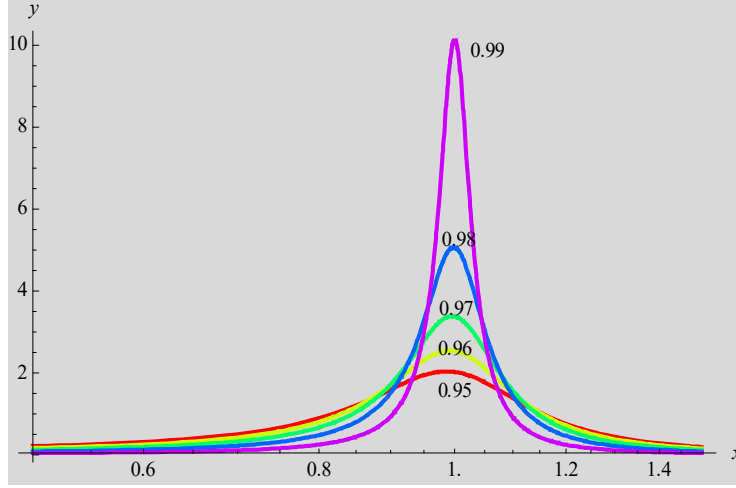
Fig.4 Plot of $y(x)$ as a function of x $(= \tau_1/\tau)$ for $\beta = 0.95 - 0.99$.
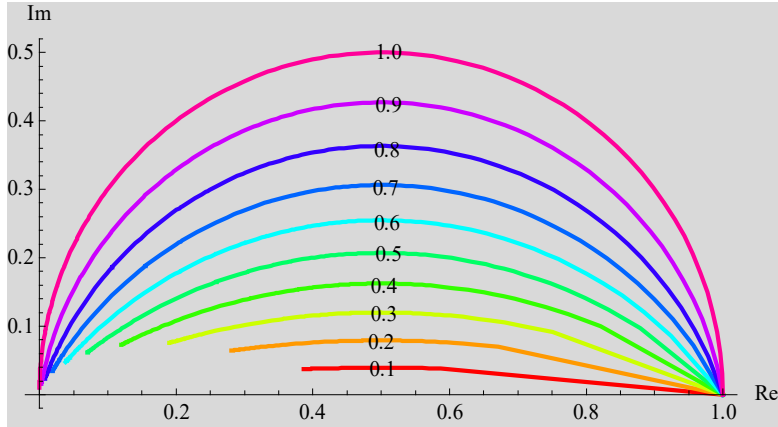

Fig.5 the Cole-Cole plot of x vs y, where $x = \chi'(\omega,\tau)/\chi_0$ and $y = \chi''(\omega,\tau)/\chi_0$. The pamater β is changed between 0.1 and 1.

5. Kramers-Kronig relation

The relation between the absorption and the dispersion is established though the Kramers-Kronig relation

$$\chi'(\omega) - \chi(\infty) = \frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{\chi''(\omega_1)}{\omega_1 - \omega} d\omega_1$$

$$\chi''(\omega) = -\frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{\chi'(\omega) - \chi(\infty)}{\omega_1 - \omega} d\omega_1 \quad ,$$

(5.1)

where P denotes the principal value of the integral. The Kramers–Kronig relations have a physical interpretation. The Kramers–Kronig relations imply that observing the dissipative response (out-of-phase) of a system is sufficient to determine its in-phase (reactive) response, and vice versa. The formulas above are not useful for reconstructing physical responses, as the integrals run from -∞ to ∞, implying we need to know the response at negative frequencies.

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Fortunately, in most systems, the positive frequency-response determines the negative-frequency response because of $\chi(-\omega) = \chi^*(\omega)$. This means that $\chi'(\omega)$ is an even function of frequency and $\chi''(\omega)$ is an odd function of ω. Using these properties, we have

$$\chi'(\omega) - \chi'(\omega = \infty) = \frac{2}{\pi} P \int_0^\infty \frac{\omega_1 \chi''(\omega_1)}{\omega_1^2 - \omega^2} d\omega_1$$

(5.2)

$$\chi''(\omega) = -\frac{2\omega}{\pi} P \int_0^\infty \frac{\chi'(\omega_1) - \chi(\infty)}{\omega_1^2 - \omega^2} d\omega_1$$

.

((**Example**)) We now apply the Kramers-Kronig relation to the case of the Debye relaxation with $\chi(\infty) = 0$. The dispersion can be derived from the absorption,

$$\chi'(\omega) = \frac{1}{\pi} P \int_{-\infty}^\infty \frac{\chi''(\omega_1)}{\omega_1 - \omega} d\omega_1 = \frac{1}{\pi} \chi_0 P \int_{-\infty}^\infty \frac{1}{(\omega_1 - \omega)} \frac{\omega_1 \tau}{1 + (\omega_1 \tau)^2} d\omega_1$$

(5.3)

.

We use the Residue theorem to solve this. To this end, we consider the contour integral around the closed loop C (semicircle with radius infinity) in the upper-half complex plane. C_1 is the semi circle (radius ε, in the limit of $\varepsilon \to 0$) centered at $z = \omega$ in the real axis. Then we have
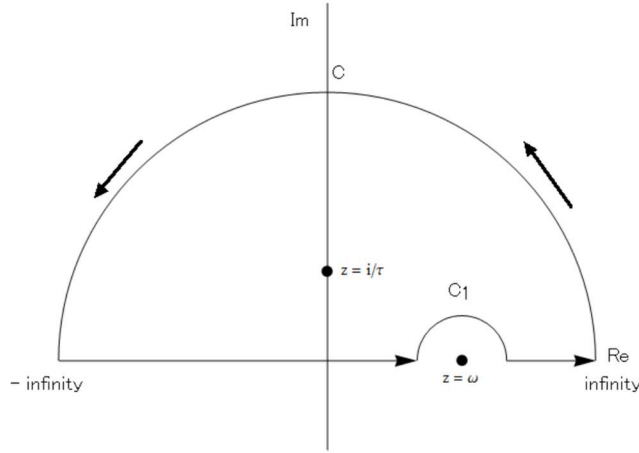

Fig.6 The path of the contour integral in the complex plane. The path consists of the three parts, the semicircle (counter clock-wise) with radius of ∞ centered at the origin, the semicircle (clockwise) with radius ε (in the limit of $\varepsilon \to 0$) centered at $z = \omega$, and the straight line of the real axis from $-\infty$ to $+\infty$, except for a region around the small semi circle C_1.

$$\oint_C \frac{1}{(z - \omega)} \frac{z\tau}{1 + (z\tau)^2} dz = P \int_{-\infty}^\infty \frac{1}{(\omega_1 - \omega)} \frac{\omega_1 \tau}{1 + (\omega_1 \tau)^2} d\omega_1 + \oint_{C_1} \frac{1}{(z - \omega)} \frac{z\tau}{1 + (z\tau)^2} dz$$

$$= 2\pi i \operatorname{Re} s[z = \frac{i}{\tau}]$$

,

(5.4)

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or

$$P\int_{-\infty}^{\infty}\frac{1}{(\omega_1-\omega)}\frac{\omega_1\tau}{1+(\omega_1\tau)^2}d\omega_1 = \pi i\,\mathrm{Re}\,s(z=\omega)+2\pi i\,\mathrm{Re}\,s[z=\frac{i}{\tau}] \tag{5.5}$$

$$= \frac{\pi}{1+\omega^2\tau^2}.$$

So we obtain the expression for $\chi'(\omega)$ as

$$\chi'(\omega) = \frac{\chi_0}{1+\omega^2\tau^2}. \tag{5.6}$$

Similarly, the absorption can be derived from the dispersion through the Kramers-Kronig relation

$$\chi''(\omega) = -\frac{1}{\pi}P\int_{-\infty}^{\infty}\frac{\chi'(\omega)}{\omega_1-\omega}d\omega_1 = -\frac{1}{\pi}\chi_0 P\int_{-\infty}^{\infty}\frac{1}{\omega_1-\omega}\frac{1}{1+(\omega_1\tau)^2}d\omega_1. \tag{5.7}$$

Using the Residue theorem for the contour integral around the path shown in Fig.6, we have

$$\oint_C\frac{1}{(z-\omega)}\frac{1}{1+(z\tau)^2}dz = P\int_{-\infty}^{\infty}\frac{1}{(\omega_1-\omega)}\frac{1}{1+(\omega_1\tau)^2}d\omega_1 + \pi\oint_{C1}\frac{1}{(z-\omega)}\frac{1}{1+(z\tau)^2}dz$$

$$= 2\pi i\,\mathrm{Re}\,s[z=\frac{i}{\tau}], \tag{5.8}$$

or

$$P\int_{-\infty}^{\infty}\frac{1}{(\omega_1-\omega)}\frac{1}{1+(\omega_1\tau)^2}d\omega_1 = \pi i\,\mathrm{Re}\,s[z=\frac{i}{\tau}]+2\pi i\,\mathrm{Re}\,s[z=\frac{i}{\tau}] \tag{5.9}$$

$$= -\frac{\pi\omega\tau}{1+(\omega\tau)^2},$$

Then we have

$$\chi''(\omega) = \frac{\chi_0(\omega\tau)}{1+\omega^2\tau^2}. \tag{5.10}$$

6. Magnetic correlation function[9-12]

The magnetic correlation function can be measured by inelastic neutron scattering measurement. It is closely related to the dynamic magnetic susceptibility through the fluctuation-dissipation theorem.

6.1 Definition of the magnetic correlation function

The magnetic correlation function is the time and spatial Fourier transform of spin-spin correlation $<\mathbf{S}_0^{\alpha}(t=0)\cdot\mathbf{S}_j^{\beta}(t)>$,

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$$S^{\alpha\beta}(\mathbf{Q},\omega) = \frac{1}{2\pi}\int dt e^{-i\omega t} <S^{\alpha}(-\mathbf{Q},0)\cdot S^{\beta}(\mathbf{Q},t)> \tag{6.1}$$

with

$$S^{\alpha}(\mathbf{Q},t) = \frac{1}{\sqrt{N}}\sum_{j}\exp(-i\mathbf{Q}\cdot\mathbf{R}_{j})\mathbf{S}_{j}^{\alpha}(t) \tag{6.2}$$

where $S_{j}^{\alpha}(t)$ ($\alpha = x, y, z$) is the localized spin at the site j ($=1, 2, \ldots N$) at the time t, N is the total number of localized spins in the system. We note that

$$S^{\alpha}(\mathbf{Q}=0,t) = \frac{1}{\sqrt{N}}\sum_{j}\mathbf{S}_{j}^{\alpha}(t) = \frac{1}{\sqrt{N}}\frac{1}{g\mu_{B}}\sum_{j}g\mu_{B}\mathbf{S}_{j}^{\alpha}(t) = \frac{1}{\sqrt{N}}\frac{1}{g\mu_{B}}M^{\alpha} \tag{6.3}$$

M^{α} is the total magnetization and is independent of time t (i.e., conserved quantity). Then the magnetic correlation function can be rewritten as

$$\begin{aligned}
S^{\alpha\beta}(\mathbf{Q},\omega) &= \frac{1}{2\pi}\int dt e^{-i\omega t} <S^{\alpha}(-\mathbf{Q},0)\cdot S^{\beta}(\mathbf{Q},t)> \\
&= \frac{1}{2\pi N}\sum_{j,k}\exp[i\mathbf{Q}\cdot(\mathbf{R}_{j}-\mathbf{R}_{k})]\int dt e^{-i\omega t} <\mathbf{S}_{j}^{\alpha}(t=0)\cdot\mathbf{S}_{k}^{\beta}(t)> \\
&= \frac{1}{2\pi}\sum_{j}\int dt e^{-i\omega t} e^{-i\mathbf{Q}\cdot\mathbf{R}_{j}} <\mathbf{S}_{0}^{\alpha}(t=0)\cdot\mathbf{S}_{j}^{\beta}(t)>
\end{aligned} \tag{6.4}$$

where we use

$$<\mathbf{S}_{j}^{\alpha}(t=0)\cdot\mathbf{S}_{k}^{\beta}(t)> = <\mathbf{S}_{0}^{\alpha}(t=0)\cdot\mathbf{S}_{k-j}^{\beta}(t)> \tag{6.5}$$

because of the translational symmetry in the system. In the inelastic magnetic neutron scattering measurements, the differential cross section to unpolarized neutrons is closely related to the magnetic correlation function,[9-12]

$$\begin{aligned}
\frac{d^{2}\sigma}{d\Omega dE_{f}} &\propto \frac{k_{f}}{k_{i}}|f_{M}(\mathbf{Q})|^{2}\sum_{\alpha,\beta}(\delta_{\alpha,\beta}-\hat{Q}_{\alpha}\hat{Q}_{\beta})\bar{S}^{\alpha\beta}(\mathbf{Q},\omega) \\
&= \frac{k_{f}}{k_{i}}|f_{M}(\mathbf{Q})|^{2}\sum_{\alpha,\beta}(\delta_{\alpha,\beta}-\hat{Q}_{\alpha}\hat{Q}_{\beta})\bar{S}^{\alpha\beta}(\mathbf{Q},\omega)
\end{aligned} \tag{6.6}$$

(Izuyama et al.), where $\mathbf{Q} = \mathbf{k}_{i} - \mathbf{k}_{f}$ is the scattering vector, and $\hbar\omega = E_{i} - E_{f}$. $\mathbf{k}_{i}$ and $\mathbf{k}_{f}$ are wave vectors of the incident neutron (initial state) and scattered neutron (final state), and E_{i} and E_{f} are the energies of incident and scattered neutrons, respectively. $f_{M}(\mathbf{Q})$ is the magnetic form factor. $\hat{Q}_{\alpha}$ is the α-component of the unit vector of $\mathbf{Q}$. $\bar{S}^{\alpha\beta}(\mathbf{Q},\omega)$ is the symmetrized correlation tensor defined by

$$\bar{S}^{\alpha\beta}(\mathbf{Q},\omega) = \frac{1}{2}[S^{\alpha\beta}(\mathbf{Q},\omega)+S^{\beta\alpha}(\mathbf{Q},\omega)] \tag{6.7}$$

6.2   Static correlation function

The integral of the magnetic correlation function over all frequencies ω keeping the scattering vector $\boldsymbol{Q}$ fixed, leads to the static correlation function;

$$
\begin{aligned}
\int d\omega S^{\alpha\beta}(\mathbf{Q},\omega) &= \frac{1}{2\pi}\int dt\int d\omega e^{-i\omega t} < S^{\alpha}(-\mathbf{Q},0)\cdot S^{\beta}(\mathbf{Q},t) > \\
&= \frac{1}{2\pi}2\pi\int dt\delta(t) < S^{\alpha}(-\mathbf{Q},0)\cdot S^{\beta}(\mathbf{Q},t) > \qquad (6.8) \\
&= < S^{\alpha}(-\mathbf{Q},0)\cdot S^{\beta}(\mathbf{Q},0) >
\end{aligned}
$$

This static correlation function is the Fourier transform of the spin correlation function over all the space, and is expressed by

$$
< S^{\alpha}(-\mathbf{Q},0)\cdot S^{\beta}(\mathbf{Q},0) >= N\sum_{j}\exp(-i\mathbf{Q}\cdot\mathbf{R}_{j})\left\langle S_{0}^{\alpha}(t=0)\cdot S_{j}^{\beta}(t=0)\right\rangle \qquad (6.9)
$$

using Eq.(6.2). Then Eq. (6.8) can be written as

$$
\int d\omega S^{\alpha\beta}(\mathbf{Q},\omega) = N\sum_{j}\exp(-i\mathbf{Q}\cdot\mathbf{R}_{j})\left\langle S_{0}^{\alpha}(t=0)\cdot S_{j}^{\beta}(t=0)\right\rangle \qquad (5.10)
$$

In a special case where $\boldsymbol{Q}=0$ and $\beta=\alpha$, Eq.(6.10) becomes

$$
\begin{aligned}
\int d\omega S^{\alpha\alpha}(\mathbf{Q}=0,\omega) &= < S^{\alpha}(-\mathbf{Q}=0,0)\cdot S^{\alpha}(\mathbf{Q}=0,0) > \\
&= \frac{1}{N}\frac{1}{g^{2}\mu_{B}^{2}}\left\langle\left(M^{\alpha}\right)^{2}\right\rangle \qquad (6.11) \\
&= \frac{1}{N}\frac{1}{g^{2}\mu_{B}^{2}}[\left\langle\left(M^{\alpha}-\left\langle M^{\alpha}\right\rangle\right)^{2}\right\rangle+\left\langle M^{\alpha}\right\rangle^{2}]
\end{aligned}
$$

using Eq.(6.3). Noting that the static susceptibility $\chi_{\alpha\alpha}^{(s)}$ and spontaneous magnetization M_{0} are defined as

$$
\chi_{\alpha\alpha}^{(s)} = \frac{1}{Nk_{B}T}\left\langle\left(M^{\alpha}-\left\langle M^{\alpha}\right\rangle\right)^{2}\right\rangle \qquad M_{0}=\left\langle M^{\alpha}\right\rangle, \qquad (6.12)
$$

then we obtain the expression,

$$
\int d\omega S^{\alpha\alpha}(\mathbf{Q}=0,\omega) = \frac{1}{N}\frac{1}{g^{2}\mu_{B}^{2}}[Nk_{B}T\chi_{\alpha\alpha}^{(s)}+M_{0}^{2}] \qquad (6.13)
$$

This is called the thermodynamic sum rule. The term in M_{0}^{2} gives a measure of the Bragg scattering, and the first term gives a measure for the static magnetic susceptibility.

7.   Fluctuation-dissipation theorem[13-16]

The general fluctuation-dissipation theorem tells us that the absorption of the dynamic complex susceptibility is closely related to the magnetic correlation function. Without proof we show that the symmetric magnetic correlation function is described by

$$\bar{S}^{\alpha\beta}(\mathbf{Q},\omega) = \frac{\hbar}{\pi} \frac{1}{g^2 \mu_B^2} \frac{1}{1 - e^{-\frac{\hbar\omega}{k_B T}}} \bar{\chi}_{\alpha\beta}''(\mathbf{Q},\omega)$$

$$\approx \frac{1}{\pi} \frac{1}{g^2 \mu_B^2} \frac{k_B T}{\omega} \bar{\chi}_{\alpha\beta}''(\mathbf{Q},\omega) \qquad (7.1)$$

where $\bar{\chi}_{\alpha\beta}''(\mathbf{Q},\omega)$ is the symmetric tensor of absorption, which is defined by (Izuyama et al. 1963)[15]

$$\bar{\chi}_{\alpha\beta}''(\mathbf{Q},\omega) = \frac{1}{2}[\chi_{\alpha\beta}''(\mathbf{Q},\omega) + \chi_{\beta\alpha}''(\mathbf{Q},\omega)] \qquad (7.2)$$

In Eq.(6.2), it is required that $\bar{\chi}_{\alpha\beta}''(\mathbf{Q},\omega)$ should be symmetric tensor. Then the magnetic correlation function is proportional to the absorption of the dynamic AC magnetic susceptibility $\bar{\chi}_{\alpha\beta}''(\mathbf{Q},\omega)$, which is dependent on $\mathbf{Q}$ and ω. Using the Kramers-Kronig relation, we have

$$\bar{\chi}_{\alpha\beta}'(\mathbf{Q},\omega) = \frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{\bar{\chi}_{\alpha\beta}''(\mathbf{Q},\omega_1)}{\omega_1 - \omega} d\omega_1 \qquad (7.3)$$

for the symmetric tensor $\bar{\chi}_{\alpha\beta}'(\mathbf{Q},\omega)$, which is defined by

$$\bar{\chi}_{\alpha\beta}'(\mathbf{Q},\omega) = \frac{1}{2}[\chi_{\alpha\beta}'(\mathbf{Q},\omega) + \chi_{\beta\alpha}'(\mathbf{Q},\omega)] \qquad (7.4)$$

Noting that

$$\bar{\chi}_{\alpha\alpha}'(\mathbf{Q},\omega) = \chi_{\alpha\alpha}'(\mathbf{Q},\omega)$$
$$\bar{\chi}_{\alpha\alpha}''(\mathbf{Q},\omega) = \chi_{\alpha\alpha}''(\mathbf{Q},\omega) \qquad (7.5)$$

we can calculate the value of $\chi_{\alpha\alpha}'(\mathbf{Q}=0,\omega=0)$ as follows.

$$\chi_{\alpha\alpha}'(\mathbf{Q}=0,\omega=0)$$

$$= \lim_{\mathbf{Q}\to 0} \int \frac{d\omega'}{\pi} \frac{\chi_{\alpha\alpha}''(\mathbf{Q},\omega')}{\omega'}$$

$$= \lim_{\mathbf{Q}\to 0} \int \frac{d\omega'}{\pi} \frac{\pi}{\hbar} g^2 \mu_B^2 \frac{(1 - e^{-\frac{\hbar\omega'}{k_B T}})}{\omega'} S^{\alpha\alpha}(\mathbf{Q},\omega')$$

$$= \frac{g^2 \mu_B^2}{k_B T} \lim_{\mathbf{Q}\to 0} \int d\omega' S^{\alpha\alpha}(\mathbf{Q},\omega') \qquad (7.6)$$

$$= \frac{g^2 \mu_B^2}{k_B T} [\frac{k_B T}{g^2 \mu_B^2} \chi_{\alpha\alpha}^{(s)}]]$$

$$= \chi_{\alpha\alpha}^{(s)}$$

for $\mathbf{Q}=0$ and $\omega=0$, where we assume that $<M^\alpha> = 0$ and $(1 - e^{-\frac{\hbar\omega'}{k_B T}})/\omega' \approx \hbar/(k_B T)$. This means that $\chi_{\alpha\alpha}'(\mathbf{Q}=0,\omega=0)$ coincides with the static susceptibility $\chi_{\alpha\alpha}^{(s)}$, as is predicted.

We note that the averaged power absorbed is related to $S^{\alpha\alpha}(\mathbf{Q},\omega)$ through

$$P_{avg}(\mathbf{Q},\omega) \propto \omega\chi_{\alpha\alpha}''(Q,\omega) = \frac{\pi g^2 \mu_B^2}{k_B T}\omega^2 S^{\alpha\alpha}(\mathbf{Q},\omega) \quad (7.7)$$

using the fluctuation-dissipation theorem.

8.   Relaxation rate of the ZFC susceptibility[17-19]

In order to appreciate the nature of the spin glass dynamics, a very wide time range has to be covered in experiments. For the conventional AC magnetic susceptibility measurement (AC SQUID measurements), the frequency ranges between f = 1 kHz and 0.01 Hz (typically) is used. Correspondingly, the time scale ranges between 10^{-3} and 10^2 sec. This is too short for the measurement of the relaxation phenomena in spin glasses. Here we show that the measurement of the time-dependent zero-field-cooled (ZFC) susceptibility allows one to probe the AC magnetic susceptibility of the system with relaxation time t ranging between t = 1 sec and 10^5 or 10^6 sec.

Recently the aging phenomena have been the subject of many experimental studies on slow dynamics in a variety of spin glass (SG) systems. Typically it can be observed in the evolution of a zero-field cooled (ZFC) magnetization $M_{ZFC}(t)$ with time t after the ZFC aging protocol for a wait time t_w. The detail of such an experiment is as follows. During the ZFC protocol, the system is quickly quenched from high temperatures well above the spin freezing temperature T_{SG} to a low temperature (T) below T_{SG} in the absence of external magnetic field. After the system is kept at T for a wait time t_w ($= 10^3$ sec - 10^6 sec), the magnetic field H (very small, typically 1 Oe) is applied at t =0. The ZFC susceptibility is measured at the same T as a function of t, where t is the observation time after the field application. The ZFC susceptibility χ_{ZFC} thus obtained is expressed by

$$\chi_{ZFC}(t,t_w) = M_{ZFC}(t,t_w)/H , \quad (8.1)$$

where $M_{ZFC}(t, t_w)$ is the ZFC magnetization. In the regime of linear response, these experiments reflect the time dependence of the dynamic spin correlation function. The fundamental relations are given by

$$\chi_{ZFC}(t,t_W) = \chi'(\omega = 1/t, t_w)$$
$$S_{ZFC}(t,t_W) = \frac{d\chi_{ZFC}(t)}{d\ln t} = \frac{2}{\pi}\chi''(\omega = 1/t, t_w) \quad (8.2)$$

with

$$t = t_m = 2\pi/\omega, \quad (8.3)$$

where S_{ZFC} is the relaxation rate and is related to the imaginary part of the complex susceptibility. There is a simple relation (so-called, $\pi/2$ relation) between $\chi'(\omega, t_w)$ and $\chi''(\omega, t_w)$,

$$\chi''(\omega, t_w) = -\frac{\pi}{2}\frac{\partial\chi'(\omega, t_w)}{\partial\ln\omega} . \quad (8.4)$$

9. Relaxation rate and the density of relaxation times[17-20]

An usual way to describe the slow relaxation of the ZFC magnetization is to postulate a statistical distribution of the relaxation times and to assume additive contributions. According to Lundgren et al,[17-19], Vincent,[20] and Suzuki and Suzuki,[21] the ZFC magnetization $M_{ZFC}(t_w,t)$ is described by a sum of exponential decay $\exp(-t/\tau)$ with the relaxation time τ multiplied by the density of relaxation times $g(t_w, \tau)$,

$$\frac{1}{H}[M_{ZFC}(t_w,t) - M_0] = q(t_w,t) = -\int_{\tau_0}^{\infty} g(t_w,\tau)\exp(-\frac{t}{\tau})d\tau \tag{9.1}$$

where H is the magnitude of an external magnetic field, M_0 is the ZFC magnetization at $t = 0$, and τ_0 is a microscopic relaxation time. The relaxation rate $S_{ZFC}(t_w, t)$ can be defined as

$$S_{ZFC}(t_w,t) = \frac{1}{H}\frac{dM_{ZFC}(t_w,t)}{d\ln t} = \frac{dq(t_w,t)}{d\ln t} = \int_{\tau_0}^{\infty} g(t_w,\tau)\frac{t}{\tau}\exp(-\frac{t}{\tau})d\tau \tag{9.2}$$

Here it is noted that a part of the integrand expressed by $f(x) = (1/x)\exp(-1/x)$ has a maximum at $x = 1.0$, where $x = \tau/t$. Using an assumption that $f(\tau/t)$ is approximated by a Dirac-delta function $[= \delta(t - \tau)]$, we get

$$S_{ZFC}(t_w,t) \approx \int_{\tau_0}^{\infty} g(t_w,\tau)\delta(t-\tau)d\tau = g(t_w,t) \tag{9.3}$$

Experimentally it is well known that $S_{ZFC}(t, t_w)$ has a relatively flat peak centered around $t = t_w$,. This implies that the density of the relaxation time $g(t_w, \tau)$ also exhibits a broad peak around $\tau = t_w$, because of $g(t_w, \tau) = S_{ZFC}(t_w, \tau)$. The aging behavior can be understood based on a phenomelogical domain model. In this picture, the aging involves the growth of the domain (denoted by R) during the ZFC protocol for a wait time t_w. The domain grows with time. The size of the domain R becomes equal to $R(t_w)$ after the wait time t_w. Through this process, only the relaxation time τ, which is nearly equal to t_w, can be selected since $g(t_w, \tau)$ has a broad peak around $\tau = t_w$. At $t = 0$ just after the ZFC protocol, a magnetic field is turned on. Then the ZFC magnetization is measured as a function of the observation time t. The size of the domain (R) remains constant $R(t_w)$ for $0 < t < t_w$. In contrast, the probing length scale (L) of the domain grows with the time t, starting from $t = 0$ in a similar way such that the domain (size R) grows for the wait time t_w during the ZFC protocol. The equilibrium dynamics is probed since $L < R(t_w)$ for $0 < t < t_w$, while the non-equilibrium dynamics is probed for $t > t_w$. The relaxation rate $S_{ZFC}(t_w, t)$ exhibits a peak around $t = t_w$, reflecting the crossover of the aging behavior between these two relaxation regimes. The density of relaxation time $g(t_w, \tau)$ marked with t_w is nearly equal to $S_{ZFC}(t_w, \tau)$. Here the label t_w is used for the notation of $M_{ZFC}(t_w, t)$ and $g(t_w, t)$, in order to emphasize that each relaxation rate $S_{ZFC}(t_w, t)$ $[(\approx g(t_w, t)]$ after the ZFC protocol for the wait time t_w represents the dominant feature of the aging dynamics for a specific domain which grows for the wait time t_w during the ZFC protocol. A set of data on $S_{ZFC}(t_w, t)$ as a function of t, for various t_w ($=10^2 - 10^5$ sec) provides an information on the snap shots of the aging behavior of domains whose size depends on the wait time. For short t_w, one can get the snap shot of the aging

behavior for small domains, while for long t_w, one can get the snap shot of the aging behavior for large domains. The relaxation mechanism for the small size domains is considered to be rather different from that for the large size domain. There is a crossover between the thermal-equilibrium dynamics inside the domains and the non-equilibrium dynamics in domain walls.

10.   Formulation by Komori, Yoshino, and Takayama[22]

The formula for the ZFC relaxation rate which are derived by Lundgren et al qualitatively, is derived theoretically by Komori and Takayama (2000).[22] The overview of the theory by them is as follows. One can see more detail of their paper. The autocorrelation function for the Ising spin glass system is defined by

$$C(t;t_w) = \overline{C_j(t;t_w)}, \tag{10.1}$$

with

$$C_j(t;t_w) = < S_j(t+t_w)S_j(t_w) >, \tag{10.2}$$

where $S_j(t)$ is the sign of the Ising spin at the site j at time t. The over-line denotes the average over sites and different realization of interactions. The bracket denotes the average over thermal noises. At time $t = 0$, the system is prepared in a random initial configuration and we let the system relax at T below T_{SG}. The absorption of the AC magnetic susceptibility is given by

$$\chi''(\omega;t_w) \approx \frac{\omega}{2T}\hat{C}(\omega;t_w), \tag{10.3}$$

where $\hat{C}(\omega;t_w)$ is the Fourier component of $C(t; t_w)$. The latter is estimated, to a good approximation, as

$$\hat{C}(\omega;t_w) \approx -\frac{\pi}{|\omega|}\frac{dC(t;t_w)}{d\ln t}|_{t=t_\omega}, \tag{10.4}$$

with $t_\omega = 2\pi/\omega$. Hence we obtain the expression for the absorption,

$$\chi''(\omega;t_w) \approx -\frac{\pi}{2T}\frac{\partial C(t;t_w)}{\partial \ln t}|_{t=t_\omega}, \tag{10.5}$$

The ZFC susceptibility is given by

$$\chi_{ZFC}(t;t_w) = \frac{1}{T}[1 - C(t;t_w)], \tag{10.6}$$

The ZFC relaxation rate is given by

$$S_{ZFC}(t=t_\omega;t_w) = \frac{\partial \chi_{ZFC}(t;t_w)}{\partial \ln t}|_{t=t_\omega} = \frac{2}{\pi}\chi''(\omega;t_w), \tag{10.7}$$

Here the relation in Eq.(10.7)

$$\chi''(\omega;t_w) \approx \frac{\pi}{2}\frac{\partial \chi_{ZFC}(t;t_w)}{\partial \ln t}|_{t=t_\omega}. \tag{10.8}$$

is equivalent to the $\pi/2$ rotation relation, since

$$\chi''(\omega;t_w) \approx -\frac{\pi}{2} \frac{\partial \chi_{ZFC}(t = t\omega;t_w)}{\partial \ln \omega} ,$$ (10.9)

and

$$\chi'(\omega;t_w) \approx \chi_{ZFC}(t = t_\omega;t_w) .$$ (10.10)

11. Experimental window

Any experiment has characteristic timescale (or characteristic frequency) associated with it. For a static magnetization measurement, for example, it is just the length of time over which the experiment is carried out (typically 10^2 sec).

(i) For the conventional AC magnetic susceptibility measurement (AC SQUID measurements), the time scale t_{obs} ranges between 10^{-3} and 10^2 sec.

$$S^{\alpha\alpha}(\mathbf{Q} = 0, t_{obs}) = < S^\alpha(-\mathbf{Q},0) \cdot S^\alpha(\mathbf{Q}, t_{obs}) > \propto \frac{1}{\omega} \chi_{\alpha\alpha}''(\mathbf{Q} = 0, \omega)|_{\omega=2\pi/t_{obs}} .$$ (11.1)

(ii) For the ZFC relaxation rate, the time scale t_{obs} ranges between 1 sec and 10^5 or 10^6 sec.

$$\chi''(\omega = 2\pi/t_{obs}, t_w) = \frac{\pi}{2} S_{ZFC}(t_{obs}, t_W) .$$ (11.2)

(iii) The characteristic time of a conventional neutron scattering experiments allows one to view process in magnetic systems on time scales t_{obs} between $10^{-11} - 10^{-13}$ sec. This is ideal for most magnetic systems, but in spin glasses it misses the most of the action, which lies at much lower frequencies.

$$S^{\alpha\alpha}(\mathbf{Q},\omega) \propto \frac{k_B T}{\omega} \chi_{\alpha\alpha}''(\mathbf{Q},\omega) ,$$ (11.3)

with $\omega = 2\pi/t_{obs}$.

(iv) For neutron spin echo experiments one can measures the evolution of the spin correlation function

$$< S^\alpha(-\mathbf{Q},0) \cdot S^\beta(\mathbf{Q}, t_{obs}) > ,$$ (11.4)

directly as a function of time, t rather than through its Fourier transform as in conventional neutron scattering. The time scale t_{obs} ranges between 10^{-12} and 10^{-8} sec.

12. Example of AC magnetic susceptibility

Here we show typical examples of the AC magnetic susceptibility (the absorption). These results are obtained by us using the MPMS SQUID magnetometer at the Department of Physics, State University of New York.

(i) Stage-2 CoCl₂ GIC (quasi 2D XY-like ferromagnet)[23]

Stage-2 CoCl₂ GIC (graphite intercalation compound) magnetically behaves like a quasi two dimensional XY-like ferromagnet. There is a very weak antiferromagnetic interaction between the adjacent intercalate layers. The system consists of the paramagnetic phase, 2D ferromagnetic phase (the intermediate phase), and spin glass phase (the low temperature phase).
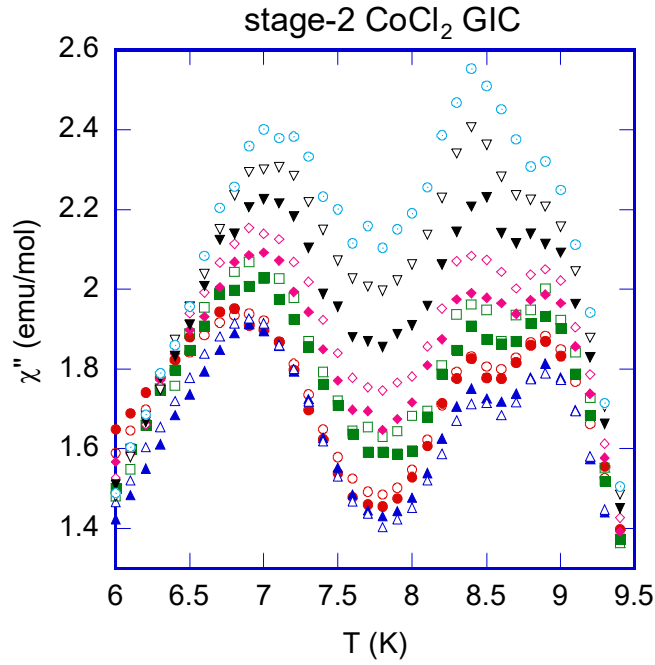
Fig.7 *T* dependence of the absorption $\chi''(\omega, T)$ of stage-2 $CoCl_2$ GIC at various frequencies: $f =$ 0. 1 – 1000 Hz . $h = 50$ mOe. $h \perp c$. The c axis is perpendicular to the basal plane of the system. $H = 0$ (no external DC magnetic field). Ref.[23].

(ii)   Spin glass phase in $Cu_{0.5}Co_{0.5}Cl_2$-$FeCl_3$ GBIC[24]

   The system $Cu_{0.5}Co_{0.5}Cl_2$-$FeCl_3$ GBIC (graphite bi-intercalation compound) is a typical spin glass system. It consists of the paramagnetic phase and the spin glass phase (low temperature phase).
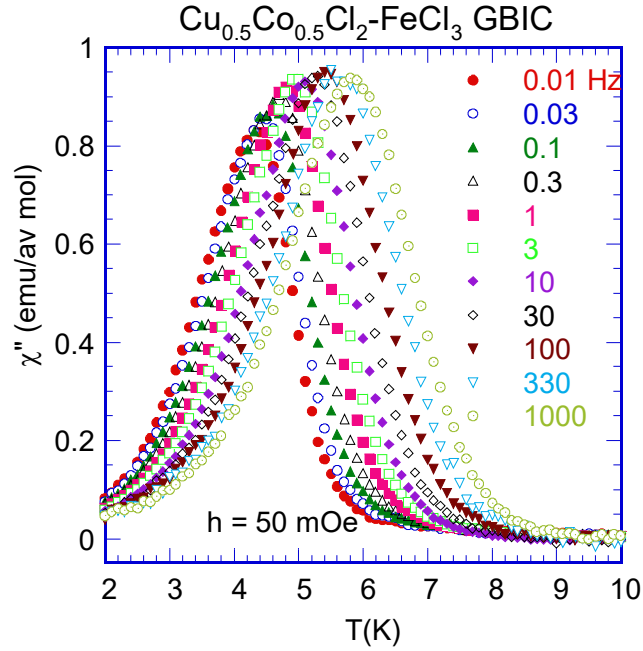
Fig.8 *T* dependence of the absorption $\chi''(\omega, T)$ of $Cu_{0.5}Co_{0.5}Cl_2$-$FeCl_3$ GBIC at various frequencies: $f = 0.01 - 1000$ Hz . $h = 50$ mOe. $h \perp c$. The *c* axis is perpendicular to the basal plane of the system. $H = 0$ (no external DC magnetic field). Ref.[24].

(iii)   Reentrant spin glass phase in stage-2 $Cu_{0.8}Co_{0.2}Cl_2$ GIC[25]

The system stage-2 $Cu_{0.8}Co_{0.2}Cl_2$ GIC is called a reentrant ferromagnet. It consists of the paramagnetic phase, the quasi 2D ferromagnetic phase (intermediate phase), and the reentrant spin glass phase (low temperature phase).
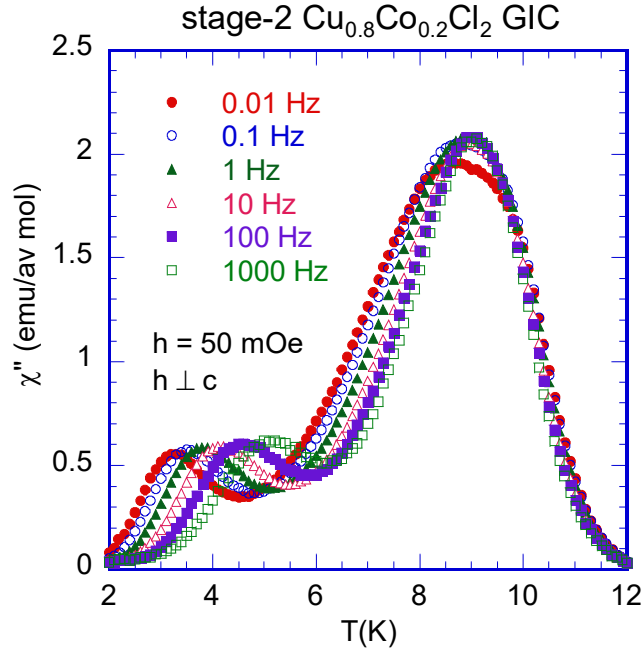.

Fig.9 *T* dependence of the absorption $\chi''(\omega, T)$ of stage-2 $Cu_{0.8}Co_{0.2}Cl_2$ GIC (graphite intercalation compound) at various frequencies: $f = 0.01 - 1000$ Hz . $h = 50$ mOe. $h \perp c$. The *c* axis is perpendicular to the basal plane of the system. $H = 0$ (no external DC magnetic field). Ref.[25].

(iv)   ZFC relaxation rate $S_{ZFC}(t, t_w)$ in spin glass $Cu_{0.5}Co_{0.5}$-FeCl$_3$ GBIC[26]

The *t* dependence of the ZFC susceptibility is measured. The time t = 0 is a time when the external DC magnetic field *H* (= 1 Oe) is applied after th ZFC protocol. The ZFC protocol consists of (i) quenching of the system from the high temperature well above the spin freezing temperature TSG to a temperature *T* below T_{SG}, (ii) annealing of the system at *T* for a wait time t_w (= 2.0 x 10^3 sec). The ZFC relaxation rate is defined by

$$S_{ZFC}(t) = \frac{d\chi_{ZFC}}{d \ln t}$$ (12.1)

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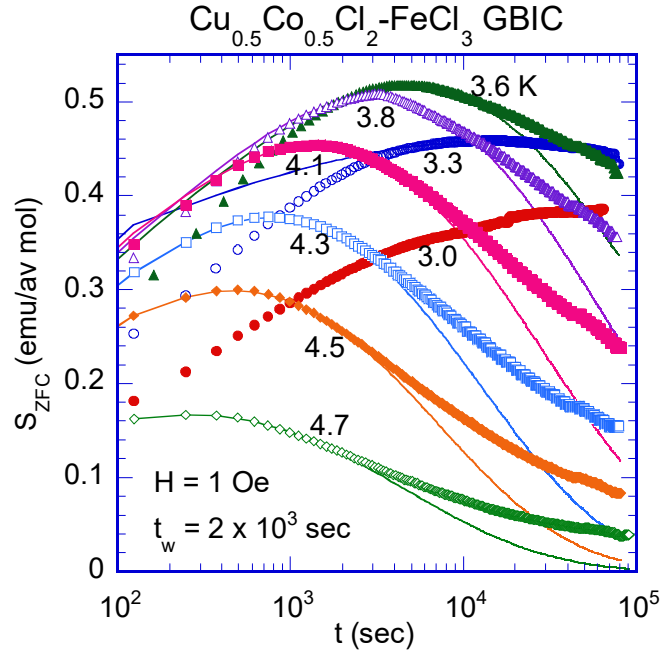**Fig.10** Time (*t*) dependence of the relaxation rate $S_{ZFC}(t)$ at various *T* for $Cu_{0.5}Co_{0.5}$-FeCl$_3$ GBIC (graphite bi-intercalation compound). $T = 3.0 - 4.7$ K. $H = 1$ Oe. $t_w = 2.0$ x 10^3 sec. The solid lines denote the least-squares fits to the stretched exponential relaxation form. Ref.[26].

(v)   Griffiths phase in Ni$_{0.8}$Mg$_{0.2}$(OH)$_2$[27]

The system Ni$_{0.8}$Mg$_{0.2}$(OH)$_2$ is a 3D Ising random spin system. It consists of the paramagnetic phase (high temperature phase), the 3D antiferromagnetic phase (the intermediate phase), and the reentrant spin glass phase (the low temperature phase). Figure 11 shows the *T* dependence of $\chi''(\omega, T, H)$. With increasing *H*, the heights of two peaks drastically increases. The peak in the high temperature side is associated with the occurrence of the Griffiths phase enhanced by the application of *H*. The peak in the low temperature side is associated with the 3D antiferromagnetic long range order.
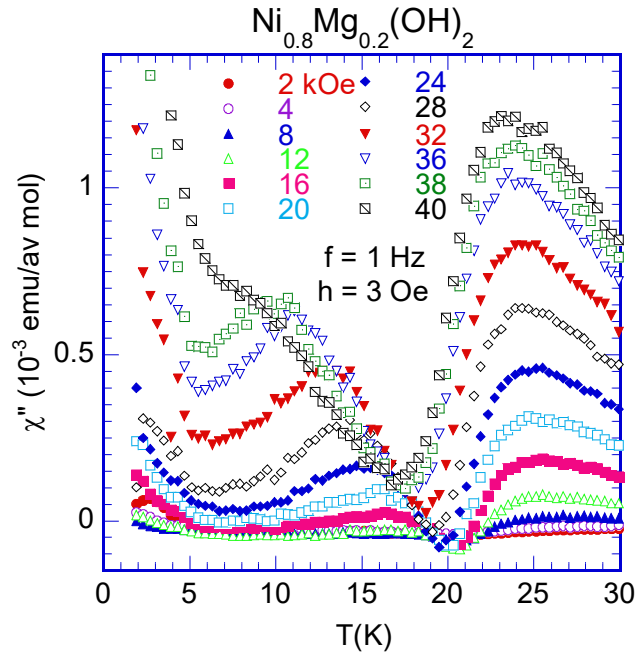
Fig.11 *T* dependence of the absorption $\chi''(\omega, T, H)$ of $Ni_{0.8}Mg_{0.2}(OH)_2$ in the presence of various DC magnetic field H (= 2 – 40 kOe). *f* = 1 Hz. *h* = 3 Oe. *T* = 2 – 30 K. Ref.[27].

(vi)   Supuspin glass Fe_3O_4 nanoparticles[28]

Fe_3O_4 nanoparticles show the superspin glass behavior. The ferromagnetic clusters play a role of single spin in the conventional spin glasses. The competing interactions among the ferromagnetic clusters) are mainly dipole-dipole interactions.
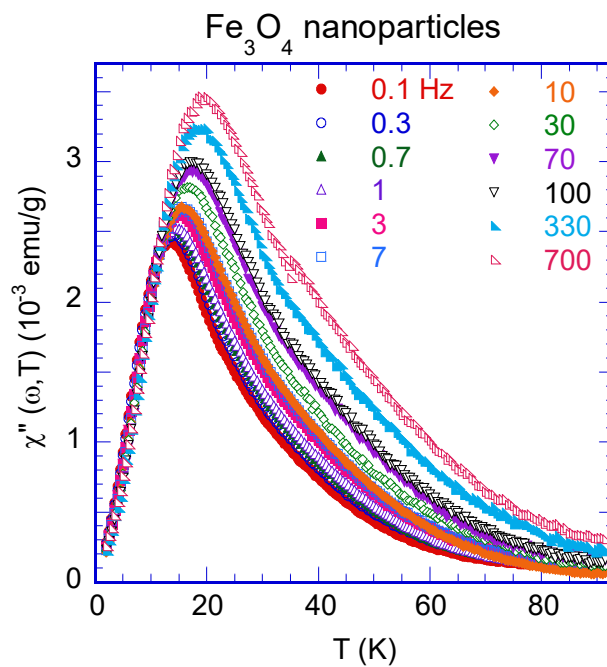
Fig.12 *T* dependence of the absorption $\chi''(\omega, T)$ of Fe_3O_4 nanoparticles at various frequencies: f = 0.1 – 1000 Hz. h = 0.5 Oe. T = 2 – 100 K. H = 0 (no external DC magnetic field). Ref.[28].

13.   Link

There are some useful links for the AC magnetic susceptibility including

(i)   Introduction to AC susceptibility (Quantum Design)
        http://www.qdusa.com/resources/pdf/1078-201.pdf
(ii)   Magnetic susceptibility Wikipedia
        http://en.wikipedia.org/wiki/Magnetic_susceptibility

CONCLUSION

We discussed the several topics of the AC magnetic susceptibility. The absorption of AC magnetic susceptibility [$\overline{\chi}_{\alpha\alpha}''(\mathbf{Q} = 0, \omega)$] is proportional to the magnetic correlation function $\overline{S}^{\alpha\alpha}(\mathbf{Q} = 0, \omega)$ through the Kramers-Kronig relation. So we can probe the relaxation process at the time scale of $t_m = 2\pi/\omega$. Using the conventional method as well as the method of ZFC relaxation rate, the time scale ranges between 10^{-3} sec and 10^6 sec (depending on the patience of experimentalists, including times they can use the equipments). One can examine the relaxation mechanism for even frustrated spin systems whose relaxation time is extremely large. We think that this note may be helpful for researchers and graduate students to understand the background of the physics in the AC magnetic susceptibility.

REFERENCES

1.   Y. Okuda, M. Matsuura, and T. Haseda, J. Phys. Soc. Jpn. **47**, 773 (1979).
2.   M. Matsuura, in *Magnetic measurements I*, edited by K. Kon and H. Yasuoka (Maruzen, Tokyo, 1999) pp. 89-124. [in Japanese].
3.   A.H. Morrish, *The Physical Principles of Magnetism*, John Wiley & Sons (New York, 1965).
4.   H.B.G. Casimir and F.K. du Prè, Physica **5**, 507 (1938).
5.   J.A. Mydosh, *Spin glasses; An experimental introduction* (Taylor & Francis, London, 1993).
6.   L. Lundgren, P. Svedlindh, O. Beckman, J. Mag. Mag. Mater. **25**, 33 (1981).
7.   L.E. Wenger, in *Lecture Notes in Physics*, 192 *Heisenberg Colloquium on Spin Glasses, Proceedings*, Heisenberg (Springer-Verlag, Berlin, 1983). pp.60 – 69.
8.   K. Yoshimitsu and T. Matsubara, Prog. Theor. Phys. Suppl. Extra number (1968). p.109.
9.   P.G. de Gennes, in *Magnetism III*, edited by G.T. Rado and H. Suhl (Academic Press, New York, 1963). pp.115 – 147.
10.   W. Marshall and R.D. Lowde, Reports on Progress in Physics **31**, 705 (1968).
11.   S.W. Lovesey, *Theory of Neutron Scattering from Condensed Matter*. Volume 2 (Oxford Science Publications, Clarendon Press, Oxford, 1984).

12. M.F. Collins, Magnetic Critical Scattering, (Oxford University Press, New York, 1989).

13. R. Kubo, J. Phys. Soc. Jpn. **12**, 570 (1957).

14. R. Kubo, Reports on Progress in Physics **29**, 255 (1966).

15. T. Izuyam, D.-J. Kim, and R. Kubo, J. Phys. Soc. Jpn. **18**, 1025 (1963).

16. D. Forster, *Hydrodynamic fluctuations, Broken Symmetry, and Correlation Functions* (Benjamin, New York, 1975).

17. L. Lundgren, *Relaxation in Complex Systems and Related Topics*, edited by I.A. Campbell and C. Giovannella, Experiments on spin glass dynamics (Plenum Press, New York, 1990). pp. 3-21.

18. L. Lundgren, P. Svedlindh, P. Nordblad, and O. Beckman, Phys. Rev. Lett. **51**, 911 (1983).

19. L. Lundgren, P. Nordblad, and P. Svedlindh, Phys. Rev. B **34**, 8164 (1986).

20. E. Vincent, *Lecture Notes in Physics* (Springer-Verlag, Berlin, 2007). Vol. 716, pp. 7-60.

21. I.S. Suzuki and M. Suzuki, Phys. Rev. B **78**, 214404 (2008).

22. T. Komori, H. Yoshino, and H. Takayama, J. Phys. Soc. Jpn. **69**, 1192 (2000).

23. M. Suzuki and I.S. Suzuki, Phys. Rev. B **58**, 840 (1998).

24. I.S. Suzuki and M. Suzuki, Phys. Rev. B **68**, 094424 (2003).

25. M. Suzuki and I.S. Suzuki, J. Phys. Cond. Matt. **18**, 10391 (2006).

26. M. Suzuki and I.S. Suzuki, Eur. Phys. J. B **41**, 457 (2004).

27. M. Suzuki, I. S. Suzuki, T.M. Onyango and T. Enoki, J. Phys. Soc. Jpn. **73**, 206 (2004).

28. M. Suzuki, S.I. Fullem, I. S. Suzuki, L. Wang and C.-J. Zhong, Phys. Rev. B **79**, 024418 (2009).