Dynamic specific heat of frustrated Ising spin rings

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Recibido el 29 de noviembre de 2001; aceptado el 22 de noviembre de 2002

The dynamic specific heat **C(**ω) is calculated exactly for rings of six coupled Ising spins within Glauber dynamics. We used the response of the internal energy to small temperature oscillations to find **C(**ω). The spin glass **(SG)** and disordered ferromagnetic **(DFM)** rings showed here have four energy minima and thus four diverging relaxation times in the time evolution of magnetization and three such times in the evolution of energy. The properties of the real and imaginary parts of dynamic specific heat are investigated for different temperatures and frequencies. The dynamic susceptibility is affected by the longest relaxing mode while the dynamic specific heat is not. Our results confirm that **C(**ω) is sensitive only to rapidly relaxing processes for ferromagnetic **(FM)** and anti-ferromagnetic **(AFM)** cases.

Keywords: Relaxation times; frequency; specific heat; spin glass; disordered ferromagnet.

Hemos calculado minuciosamente el calor dinámico especifico **C(**ω**)** de círculos que contienen seis *Ising spins* conectados, utilizando la dinámica de Glauber. Hemos utilizado la influencia de la energía interna sobre las oscilaciones de temperatura para calcular el **C(**ω**)**. Los círculos de cristal revuelto y ferromagnetizados desorganizados han demostrado que tienen cuatro niveles de energía, y por eso tienen cuatro tiempos de divergencia durante la evolución de la magnetización. Sin embargo, tienen tres tiempos de divergencia durante la evolución de energía. Hemos examinado las características de las partes reales e imaginadas de la **C(**ω**)** en distintas temperaturas y frecuencias. La susceptabilidad dinámica se influye por el tiempo de divergencia más largo, mientras que la **C(**ω**)** no se influye por el mismo. Los resultados de nuestra investigación aseguran que la **C(**ω**)** es sensible solamente a los procesos de relajamiento rápidos para los casos de ferromagneticos **(FM)** y antiferromagneticos **(AFM)**.

Descriptores: Tiempos de relajación; calor específico; círculo de cristal revuelto; ferromagnetos desorganizados.

PACS: 75.10.Nr; 76.20 + q

1. Introduction

The specific heat yields information about the excitations of a spin glass. It exhibits rather a broad peak at temperature exceeding the freezing temperature *(T_f)* by about 20% . For $T < T_f$, it shows a linear temperature dependence, as observed in *CuMn, AuFe, EuSrS* and other spin glasses [1-3]. There is no sharp cusp or singularity in the specific heat of random systems. Unusual behaviour has been also observed in many dynamic properties (very slow relaxation to equilibrium, the aging effect etc....). The heat capacity and magnetic susceptibility of barium holmium, barium erbium floride and barium thulium floride were measured by the magnetic resonance and optical absorption spectroscopy [4]. It is not easy to determine the magnetic specific heat *(C_M)* accurately, since it is only a small fraction of the total specific heat which consists of vibrational, electronic and magnetic parts, and the subtraction of the low temperature electronic contribution is especially difficult [5].

The entropy difference up to a given temperature can be found by using the specific heat. Al-Wahsh H. *et al.* [6] calculated exactly the specific heat *C(T)* for finite *N* Ising spins with antiferromagnetic couplings. They obtained round peaks for *C(T)* versus *T* and found that in the thermodynamic limit *C(T)* $\rightarrow$ *0*. The dynamic specific heat for glasses and other systems with many modes of configurational relaxation becomes an important tool to study the spectrum of relaxation times due to the even-spin correlations. On the other hand, the dynamic susceptibility is interesting for investigating the relaxation times corresponding to the odd-spin correlations. Recently Tien and others [7] measured the specific heat of Pr_2 $CuGe_6$ in an adiabatic calorimeter by a modified heat-pulse method. They found that it exhibits a short-range spin glass-like order at low temperature.

Specific heat studies of magnetic materials are useful for several reasons as characterization of the low temperature properities in zero magnetic field, the nature of the magnetic transition and determination of the magnetic entropy evolved. It is also helpful to know the temperature dependence of the specific heat of any material used for magnetooptic recording when heating it from room temperature to above the magnetic ordering temperature. Microcalorimeters have been used to measure the temperature dependence of the specific heat C_p*(T)* of amorphus R_x Fe_{100-x} *(R = Gd, Tb)* thin films prepared by both sputtering and e-beam coevaporation [8]. All samples show a relatively sharp peak in C_p*(T)* at the Curie temperature T_c determined by magnetization measurements.

The study of the effect of disorder on the critical behaviour of magnetic systems has been the subject of a great amount of investigations during the last few decades, both theoretically and experimentally [9]. From the theoreti-

cal point of view, it is known that Ising-like models are well suitable to describe the critical behaviour of insulating anisotropic magnetic systems [10]. Disorder effects play a particular role in one-dimensional systems since any small deviations from regularity destabilize the pure phases. Real experimental systems naturally contain impurities and other types of disorder. Therefore, it is very important to understand the influence of disorder on the properties of such systems in order to interpret experimental results. This is one of the very important reasons why in recent years random spin systems have been investigated intensively.

In recent years much interest has been given to the magnetism of systems with a small number of atoms, such as small magnetic clusters and thin films [11]. The magnetic properties depend on several geometric features which in turn depend on the conditions during the growth. Thus the connection between the experimental and theoretical results requires a detailed study of the dependence of physical quantities on the geometry of the systems. Some specific properties have already been considered in theoretical works, such as the size and lattice structure of small clusters [12]. It thus seems interesting to study well defined microscopic models and to determine properties of their relaxational spectra. The simplest of such models are small clusters of up to seven Ising spins which undergo the Glauber dynamics [13]. The dynamics of such systems are discussed in detail in Ref. 14 but they didn't study the dynamic specific heat and the disordered ferromagnetic systems.

The simplest models with this feature built-in are chains or , if the periodic boundary conditions are imposed, rings of the Ising spins coupled via the Gaussian interactions. The properties of such rings have been studied in detail by Reger and Binder [15] where they derived the configurationally averaged dynamic susceptibility. However, a discussion of the ω-dependent dynamic specific heat has not been within their focus. The model discussed here displayed a zero-temperature phase transition. It can be analyzed exactly and it allows the study of several modes of behaviour depending on the choice of the exchange couplings *J*. These choices will be summarized in Sec. 2. We shall focus our attention on the ferromagnetic *(FM)*, antiferromagnetic *(AFM)*, spin glass *(SG)* and disordered ferromagnetic *(DFM)* systems. We outline the equations of motion for the spin correlations using single spin-flip Glauber dynamics. A discussion on the spectrum of relaxation times will be given in Sec. 3. In Sec. 4 we explain the notion of the dynamic specific heat and derive expressions for its real and imaginary parts for a system with coordination number of 2. It was known that the dynamic susceptiblity is given in terms of odd-spin correlations but (in this work) we show that the dynamic specific heat couples to the time evolution of even-spin correlations. The dynamic specific heat allows us to investigate a different subset of the relaxational spectrum than the one derived through the magnetization. Finally, we analyze the dynamic specific heat in Sec. 5 for the models mentioned here .

2. The model and equations of motion

The energy of an Ising system of N spins *($S_i = \pm 1$)* could be described by the Hamiltonian

$$H = -\sum_{i=1}^{N} J_{i,i+1} S_i S_{i+1}, \tag{1}$$

where $J_{i,i+1}$'s are the exchange couplings restricted only to the nearest neighbouring interactions. We impose the periodic boundary conditions $S_{N+1} = S_1$. We study 6-spins arranged in Fig. 1 and investigate various special configurations of the following exchange couplings.

(i) *SG:* As an example, we take $J_{i,i+1}$'s as shown in Fig. 1. The system has four non-trivial local energy minima.

(ii) *DFM:* The exchange couplings are taken to be the absolute values of the exchange couplings shown in Fig. 1. There are four non-trivial energy minima in this systems.

For *FM* and *AFM* cases, the system has only one non-trivial energy minimum of energy -6.

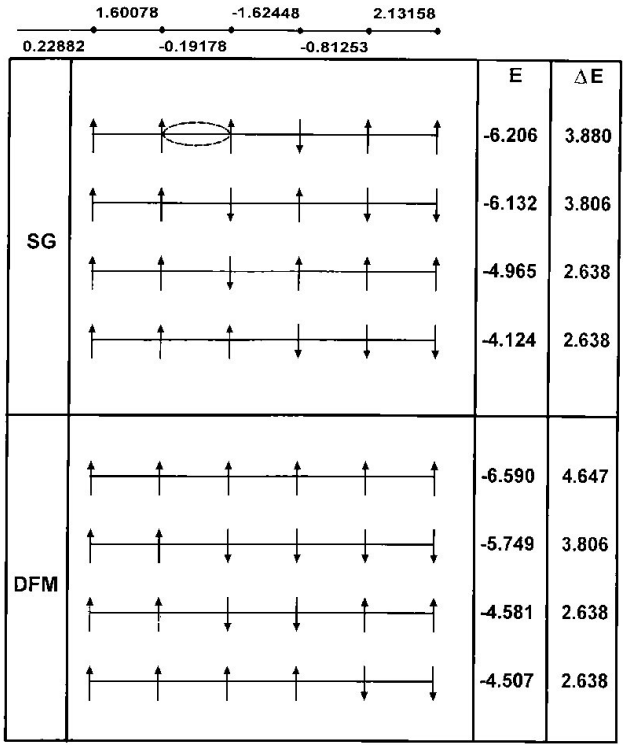

FIGURE 1. The six-spin model considered in this paper. The numbers generated from a Gaussian distribution of a unit dispersion indicate values of the exchange couplings. Periodic boundary conditions are assumed. The arrows indicate spin configurations in the four local energy minima found in this model for *SG* and *DFM* cases. The corresponding energies *(E)* and reversal energies *(ΔE)* are written to the right.

Owing to the master equation of Glauber [13] with detailed balance condition, the transition probability $W_i(S_i)$ for flipping a spin at site *i* is given by

$$W_i(S_i) = (1/2\tau_0)\left[1 - S_i \tanh \beta h_i\right]. \quad (2)$$

In this equation $h_i = J_{i,i+1}\, S_{i+1} + J_{i,i-1} S_{i-1}$ is the local exchange field acting on S_i and τ_0 is a microscopic flipping time, and $\beta = 1/k_B T$, where T is the temperature and k_B is Boltzmann's constant. For one-dimensional systems with the coordination number 2, we have

$$\tanh \beta\, h_i = \gamma_{i,i+1} S_{i+1} + \gamma_{i,i-1} S_{i-1} \quad (3)$$

The coefficients $\gamma_{i,i\pm1}$ depend on temperature and all of the exchange couplings associated with this site. Then

$$\gamma_{i,i\pm1} = \frac{1}{2}\big(\tanh\beta\,(J_{i,i+1} + J_{i,i-1}) \pm \tanh\beta\,(J_{i,i+1} - J_{i,i-1})\big). \quad (4)$$

In the six-spin problem considered here there are twelve coefficients. The linear terms in (3), when used in the master equation, couple the single-spin correlations to single-spin correlations. The triple-spin correlations couple to single- and triple-spin correlations and so on. Similarly, the two-spin correlations couple to the two-spin correlations and the four-spin ones, these in turn to the six-spin terms. If the system is finite then it can be solved exactly. For systems with a four-fold coordination, Eq. (3) should contain triple-order terms as well.

In a system consisting of N spins there are altogether $n = 2^N - 1$ correlations. There are $n_0 = 2^{N-1}$ odd and $n_0 - 1$ even correlations. It is convenient to form a vector $\vec{V}$ out of these 63 correlations by placing first the single-spin correlations, then triple, etc., until the first n_0 components are filled. After that we place two-spin correlations, four-spin correlations, and so on. Later on, it will turn out to be useful to arrange the two-spin correlations in such a way that the components $n_0 + 1$ through $n_0 + 2N$ correspond to the correlations between the neighbouring spins.

Regarding Refs. 13 and 14 in the absence of the external magnetic field, the equations of motion can be written in the form

$$\tau_0 \frac{\partial}{\partial t}\vec{V}(T,t) = \hat{M}(T)\,\vec{V}(T,t) + \vec{L}(T), \quad (5)$$

where

$$\hat{M}(T) = \begin{bmatrix} A & \hat{0} \\ \hat{0} & B \end{bmatrix} \quad (6)$$

and

$$\hat{L}(T) = \begin{bmatrix} \vec{o} \\ L' \end{bmatrix}, \quad (7)$$

where $\hat{0}$ and $\vec{o}$ denote the zero matrix and zero vector, respectively. The matrix A is of $n_0 x\, n_0$ and it couples the odd-spin correlations among themselves. The matrix B couples the even-spin correlations. The inhomogeneous term L affects the latter.

3. The relaxation times

The relaxation times τ_v are obtained as the inverse of the eigenvalues of the matrix $-(1/\tau_0)M$. Out of the n_0 relaxation times derived from the odd submatrix A there are some which diverge when T approaches zero. The number of diverging relaxation times is equal to the number of non-trivial energy minima [14] (by non-trivial we mean that we don't consider a state obtained by reversing all of the spins as a different minimum). The "even" submatrix B has been analyzed and found that; in the limit of low T the "even" spectrum coincides with the "odd" one (see Fig. 2 at $T = 0.4$) while the longest relaxation times, being related to the reversal of the ground state, belongs only to the odd part. Figure 2 shows how the long relaxation times for *SG* and *DFM* cases compare to the remaining times at several values of T. The spectrum of τ_0 is presented in the logarithmic scale.

For both *SG* and *DFM* systems shown, in Fig. 1 there are four local energy minima and thus four diverging relaxation times in the odd part denoted by τ_1 which is the longest and then τ_3 , τ_5 and τ_7. Meanwhile there are three diverging relaxation times in the even part taken as τ_2 , τ_4 and τ_6. The temperature variation of the relaxation times in the odd and even parts are shown in Fig. 3.

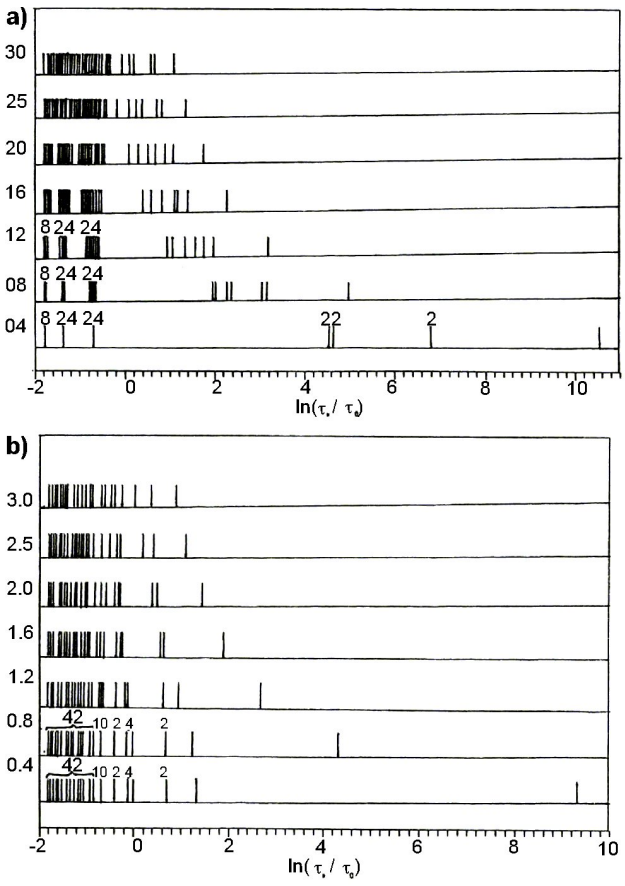

FIGURE 2. The spectra of relaxation times of *SG* and *DFM* systems shown in Fig.1 for different temperatures *(T's)* at the left.

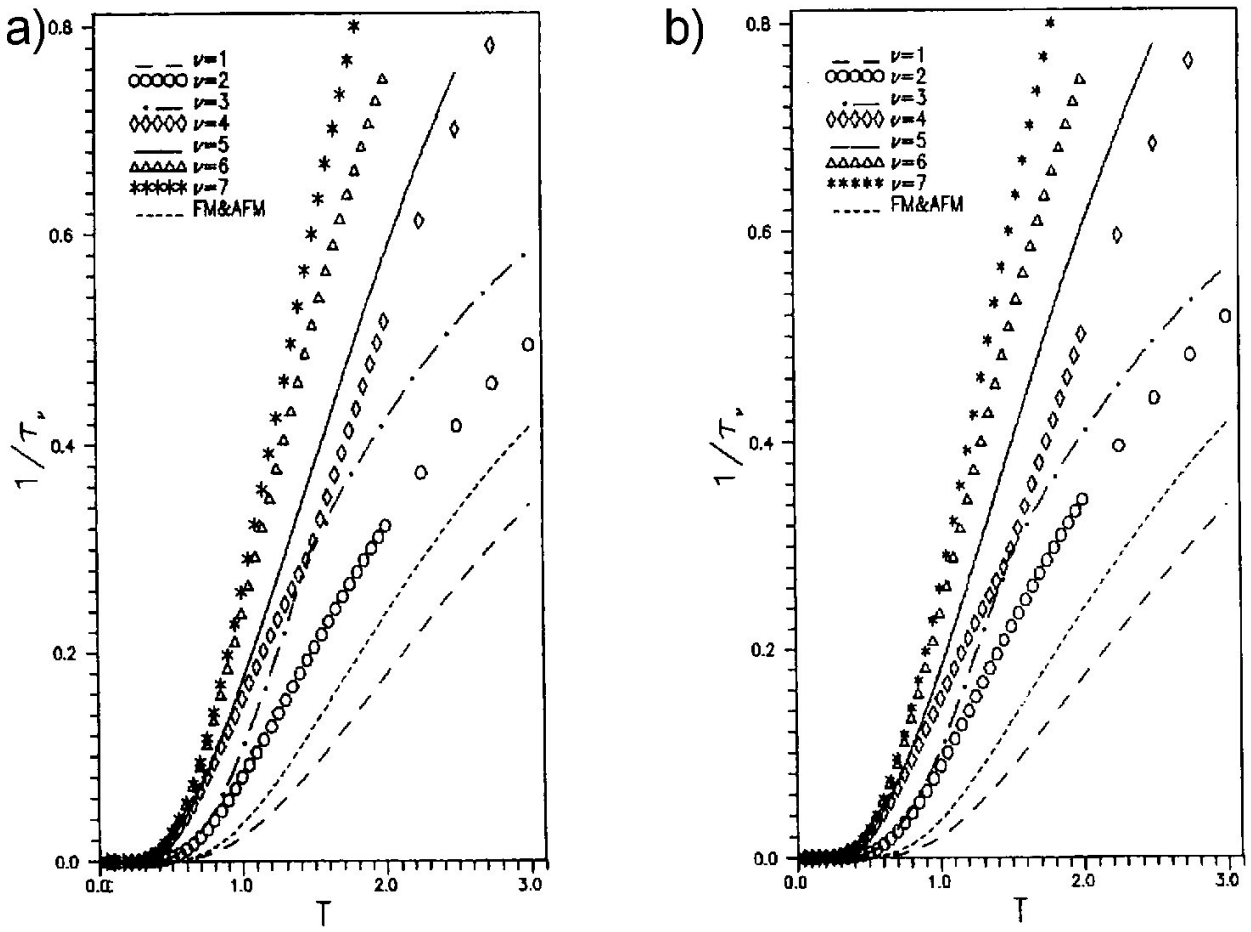

FIGURE 3. The inverse of the divergent relaxation times in units of τ for *SG* and *DFM* systems shown in Fig. 1 vs. temperature (k_BT is measured in terms of *J*). The longest diverging relaxation times for *FM* and *AFM* systems are also apperead.

For uniform ferromagnets and antiferromagnets, in which the ground state is the only local energy minimum, there is only one diverging relaxation time as in Fig. 3. The even part of the spectrum does not correspond to any long-lasting processes.

Disordered ferromagnetic rings have as many local energy minima as the "spin glassy" rings. The two systems have also identical relaxational spectra. This, however, is the peculiarity of the one-dimensional physics. It has also been found that the spectrum of the coupled even-spin correlations at low *T* coincides with the "odd" spectrum [16]. The only exception is that the longest time occurs exclusively in the odd part. The longest time is related to the reversal of the true ground state.

4. Dynamic specific heat

The temperature oscillations was caused by an oscillatory heat as used in some experiments. For this reason, one can define the same specific heat by inverting the situation.

Consider a system of *N* Ising spins in a heat bath and let the temperature changes periodically with time as follows:

$$T(t) = T + \delta T(t), \tag{8}$$

where

$$\delta T(t) = \delta T \sin(\omega t). \tag{9}$$

It is clear that in the absence of a static magnetic field, the even correlations only are affected by the temperature oscillations. This property could be altered if a static magnetic field is present. The perturbation (8) appears in a phase-shifted oscillatory evolution of the internal energy. The average of Hamiltonian, *i.e.*, internal energy, of our investigated system has the form

$$\langle H\rangle = -\sum_{i=1}^{N} J_{i,,i+1} \langle S_i(t) S_{i+1}(t)\rangle \tag{10}$$

The energy depends on time then once the transients die out this dependence becomes oscillatory. The average is made with respect to an equilibrium ensemble.

Following some calculations the oscillatory part, $\delta\langle H\rangle$, of this Hamiltonian can be represented as

$$\delta\langle H\rangle = -\sum_{i=1}^{N} J_{i,,i+1} \sum_{i=1}^{n} U_{\upsilon}^{n_0+i} N_{\upsilon}(\omega) (\sin(\omega t) - \omega\tau_{\upsilon}\cos(\omega t))\,\delta T, \tag{11}$$

where

$$N_\upsilon(\omega) = \frac{\tau_\upsilon/\tau_0}{1+\omega^2\tau_\upsilon^2}\left[\lambda_\upsilon + \sum_{\upsilon'=1}^{n} \mu_{\upsilon\upsilon'} L_{\upsilon'}\tau_{\upsilon'}/\tau_0\right], \quad (12)$$

where

$$L_{\upsilon'} = u_{\upsilon'}\vec{L}(T), \lambda_\upsilon = u_\upsilon\vec{\lambda}(T) \text{ and } \mu_{\upsilon\upsilon'} = u_\upsilon\hat{\mu}(T)U_\upsilon.$$

$\vec{u}$ is the left eigenvector of $\hat{M}(T)$ and U_υ^i is the $i^{\underline{th}}$ component of $\upsilon^{\underline{th}}$ right eigenvectors of $\hat{M}(T)$. n_0 is the number of odd spin correlations, $n_0 = 32$ for our six-spin cluster.

Once the transients die out the real *C'(*ω) and imaginary *C"(*ω) parts of the dynamic specific heat may be defined as follows:

$$\delta\langle H\rangle/N = \delta\mathrm{T}\big(C'(\omega)\sin(\omega\mathrm{t}) - C''(\omega)\cos(\omega\mathrm{t})\big). \quad (13)$$

Comparing Eqs. (17) and (18), one can write

$$C'(\omega) = -\frac{1}{N}\sum_{i=1}^{N}\sum_{\upsilon=1}^{n} J_{i,i+1}U_\upsilon^{n_0+i}N_\upsilon(\omega) \quad (14)$$

and

$$C''(\omega) = -\frac{\omega}{N}\sum_{i=1}^{N}\sum_{\upsilon=1}^{n} J_{i,i+1}U_\upsilon^{n_0+i}N_\upsilon(\omega)\tau_\upsilon \quad (15)$$

The real part of dynamical specific heat, *C'(*ω), approaches the equilibrium, *dc* specific heat *(C(0))* in the limit of $\omega \to 0$. The imaginary part, *C"(*ω), is a measure of the fluctuations in energy and will disappear in the *dc* limit *i.e.*, *C"(0) = 0*.

5. Results and discussion

Considering the 6-spin models shown in Fig.1 there are, altogether, 31 even correlations. By simple numerical routines, we are able to determine all the necessary eigenvectors. The properties of the dynamic specific heat for various temperatures and frequencies, will be discussed as follows:

5.1. Temperature dependence of the dynamic specific heat

(i) *For SG;* Figure 4 shows the variation of *C'(*ω) and *C"(*ω) as a function of *T* for several values of ω. At high temperature, *C'(*ω) essentially follows the equilibrium specific heat *(C_{eq})*. The longer the period of oscillations *(ω^{-1})*, the higher the maximum. However ω, the maxima are very rounded peaks. A similar situation is encountered for the real part, χ*'(*ω), of the dynamic susceptibility (see Ref. 17). In three dimensional systems the maximum in C_{eq} should also be at higher *T*'s than the cusp in $\chi(\omega)$ as found in real systems.

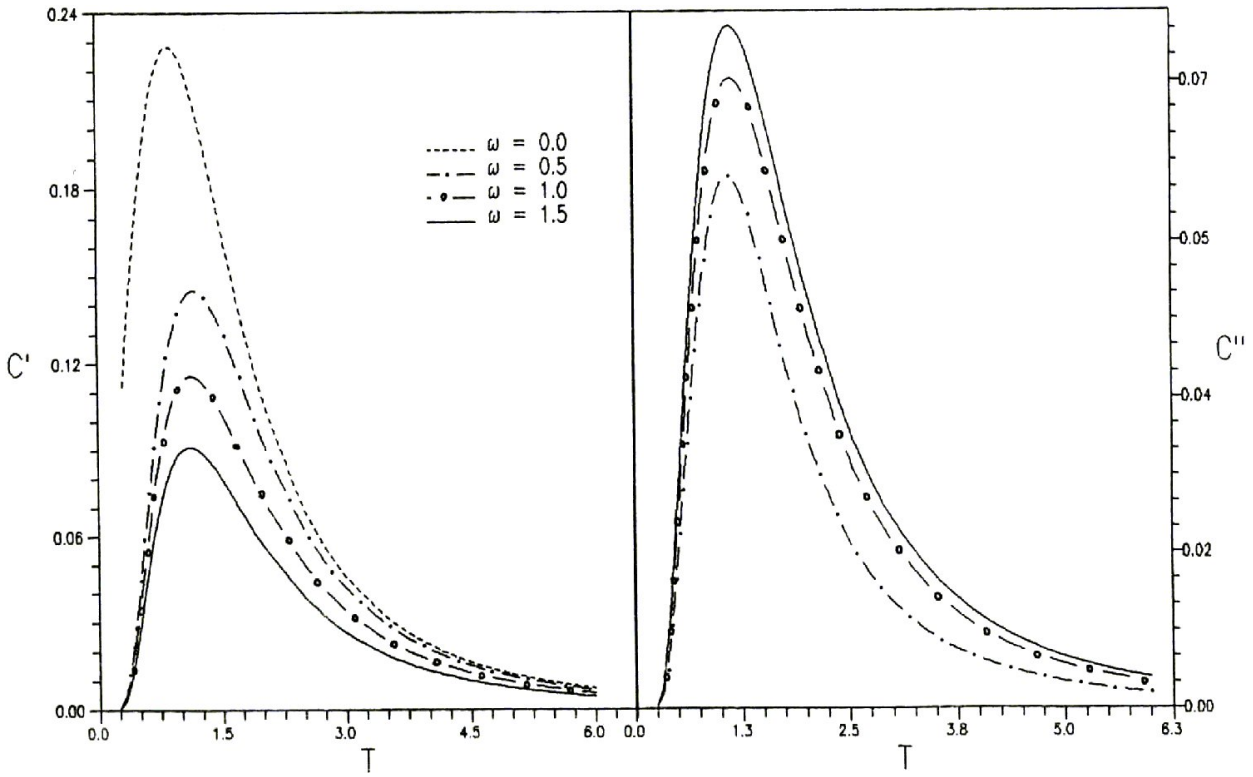

FIGURE 4. The real *C'(*ω*)* and imaginary *C"(*ω*)* parts of dynamic specific heat in units of *k* /spin, for *SG* system shown in Fig. 1 as a function of *T* at different ω (k_BT is measured in terms of *J*).

The plots of $C''(\omega)$ versus T resemble those of $C'(\omega)$ except that the zero frequency limit corresponds to $C''(\omega) = 0$. Then the smaller the value of ω, the smaller and narrower the maximum. The positions of maxima move towards $T = 0$ on decreasing ω. It is also found that, at $\omega = 0.01\tau_0^{-1}$ the maximum of $C''(\omega)$ is greater and narrower than that at $\omega = 0.1\tau_0^{-1}$. For $\omega = 0.01\tau_0^{-1}$, $0.1\tau_0^{-1}$ the maximum does not locate at the position for $\omega = 1.5\tau_0^{-1}$, because of the big difference between $\omega = 0.01\tau_0^{-1}$ and $\omega = 1.5\tau_0^{-1}$.

(ii) *For DFM;* The plots of $C'(\omega)$ and $C''(\omega)$ versus T (see Fig. 5) are similar to those of *SG* except for the following: the maximum of $C'(\omega)$ at $\omega = 0$ is not quite rounded for the *DFM*. In addition the highest maximum of $C''(\omega)$, for *DFM* is found at $\omega = 0.01\tau_0^{-1}$.

For *FM* and *AFM*, as examples for ordered systems, we found that the T-dependence of $C'(\omega)$ is qualitatively the same as for the spin-glassy couplings and disordered ferromagnet. While in the case of $C''(\omega)$ versus T, the less the ω, the greater the maximum as shown in Fig. 5. For very small ω (*e.g.*, $\omega = 0.01\tau_0^{-1}$) the greater the ω, the greater the maximum.

5.2. Frequency dependence of the dynamic specific heat

Figures 6 and 7 show $C'(\omega)$ and $C''(\omega)$ versus $log_{10}\,\omega$. It's known that the plot of $C'(\omega)$ should consist of a sequence of plateaus, whereas the plot of $C''(\omega)$ should have a sequence of corresponding maxima. The number of plateaus is equal to the number of maxima which is also equal to the number of divergent relaxation times due to the even-spin correlations. The two parts of dynamic specific heat are connected via the Kramers-Kronig relations.

(i) *For SG;* The $log_{10}\,\omega$ -dependence of $C'(\omega)$ and $C''(\omega)$ for several values of T is shown in Figs. 6 and 7. At low temperatures (*e.g.* $T = 0.25$ and 0.3) $C'(\omega)$ essentially displays three plateaus corresponding to $\omega^{-1} = \tau_2$, τ_4 and τ_6 due to even-spin correlations. These three plateaus are clear at $T = 0.25$ and the two upper ones overlap at $T = 0.3$ to merge in one plateau gradually at $T = 0.4$. The third (down) plateau becomes very much obvious at $T = 0.3, 0.4, 0.5, 0.55$ and 0.6. Finally, we got only one plateau at $T = 2.0$ (*i.e.*, high T). We can also see that both of the length of the most upper plateau and its height increase with increasing temperature.

Figure 7 depicts the plot of C'' against $log_{10}\,\omega$ where three clear maxima at $T = 0.25$ corresponding to the divergent relaxation times due to even-spin correlations as stated above in C' are noticed. The first, on the left side, maximum merges with the second to become only one, then two distinguished maxima will appear at $T = 0.4$. These two maxima increase and are shifted to the right side with increasing of T. At $T = 2.0$, C'' has only one maximum corresponding to the one plateau appeared in $C'(\omega)$ versus $log_{10}\,\omega$ (see Fig. 7).

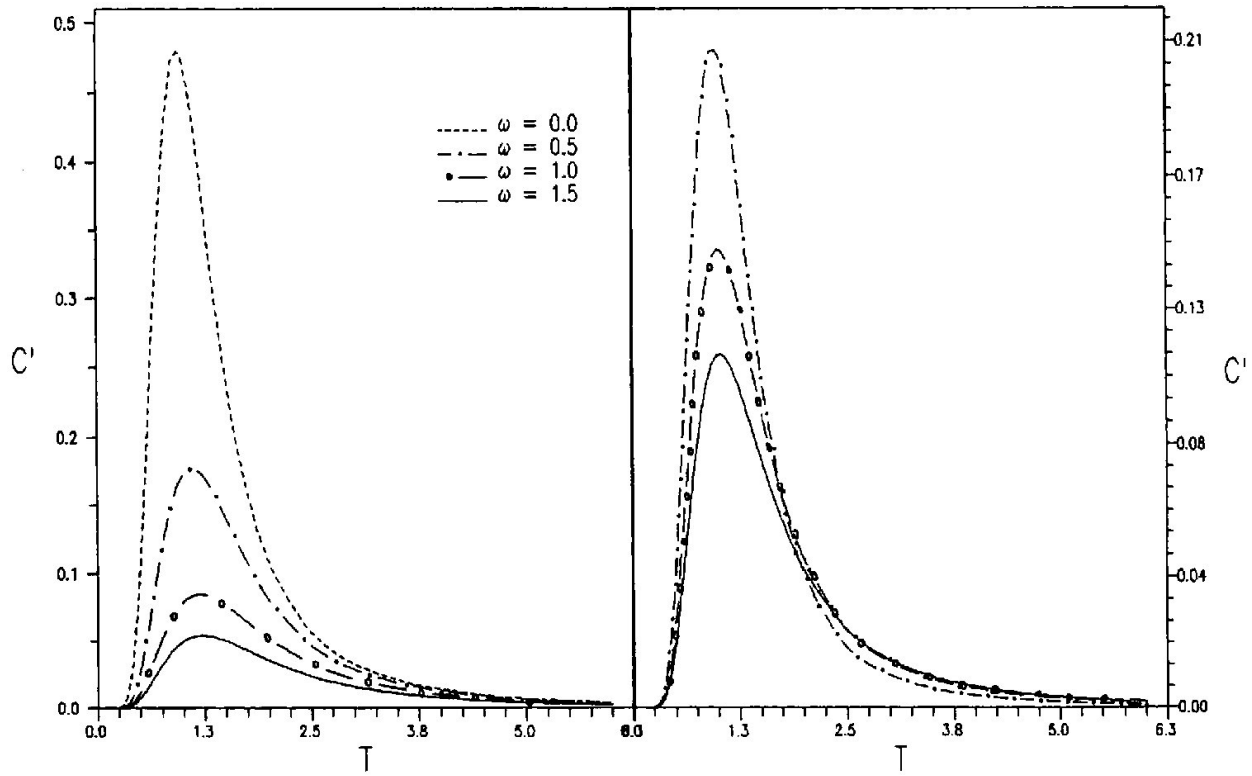

FIGURE 5. The same as in Fig.4 but for the two ordered systems; *FM* and *AFM* which are coincident.

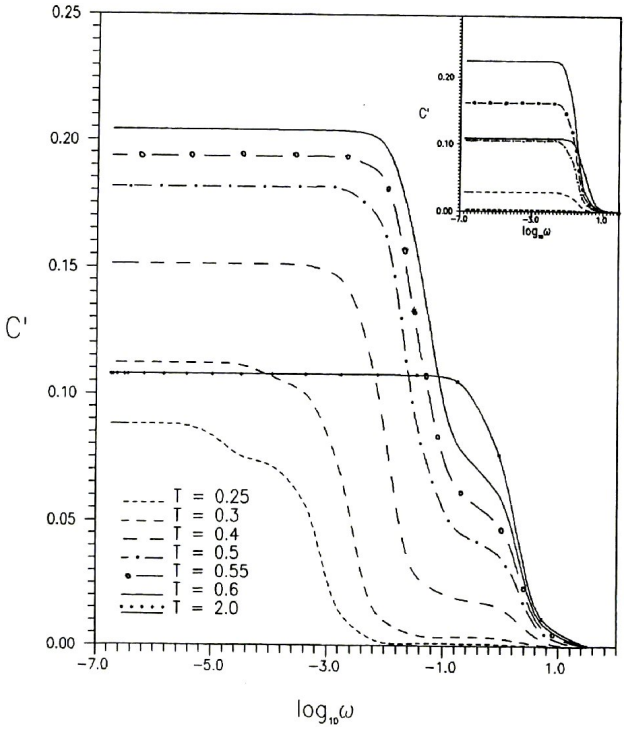

FIGURE 6. The *C'(ω)* part of dynamic specific heat versus log_{10} ω for *SG* system shown in Fig. 1 at different *T*. (ω is measured in terms of τ). The enclosed figure is the same but for *FM* and *AFM* systems.

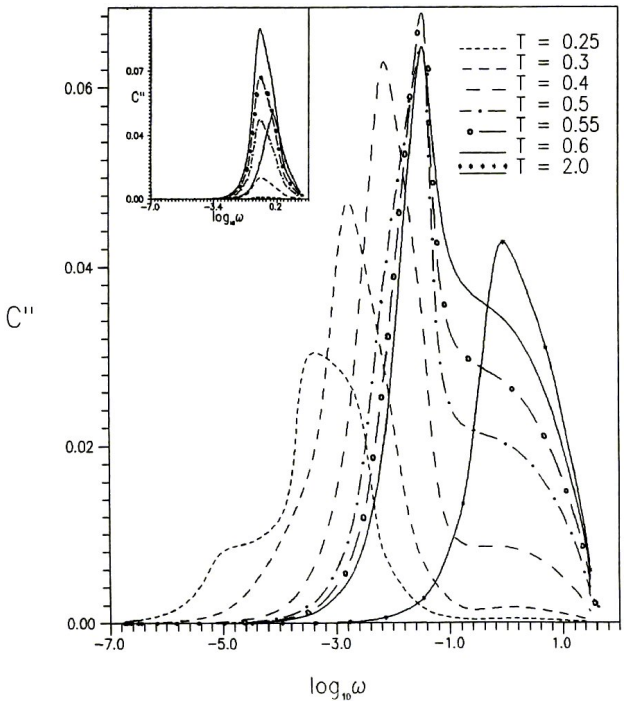

FIGURE 7. The imaginary *C"(ω)* part of dynamic specific heat versus log_{10} ω for *SG* system shown in Fig. 1 at different *T*. (ω is measured in terms of τ). The enclosed figure is the same but for *FM* and *AFM* systems.

For *FM* and *AFM* cases, the log_{10} ω-dependence of *C'(ω)* and *C"(ω)* has only one plateau and also only one maximum as plotted in Figs. 6 and 7. Each of these two ordered systems has only one divergent relaxation time (see Fig. 2).

(ii) *For DFM;* The plots of *C'(ω)* and *C"(ω)* versus $log_{10}\omega$ are found to be qualitatively the same as for the spin-glassy couplings as shown in Fig. 8. But the three plateaus in *C'(ω)* and the corresponding three maxima in *C"(ω)* are not sufficiently clear. The randomness in *DFM (i.e.,* $|J_{ij}|$*)* is less than that in *SG* therefore we might expect three obvious plateaus and maxima as long as $T < 0.25$. In the case of χ*"(ω)* as in Ref. 17, we did not find any clear evidence for maxima at τ_3 and τ_5 either. These results indicate that the *SG* and *DFM* systems exhibit a phase transition from disordered state into an ordered one.

Figure 9 shows the Cole-Cole plots on which *C"(ω)* is drawn as a function of *C'(ω)* at *T = 0.4, 0.6* and *2.0* for *SG*

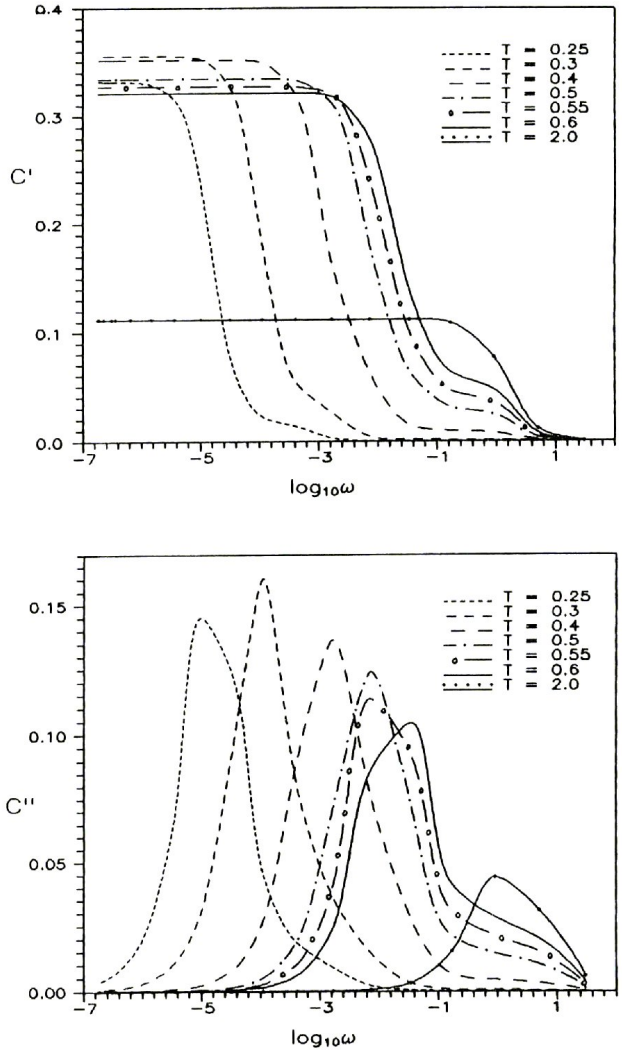

FIGURE 8. The same as in Figs. 6 and 7 but for *DFM* system.

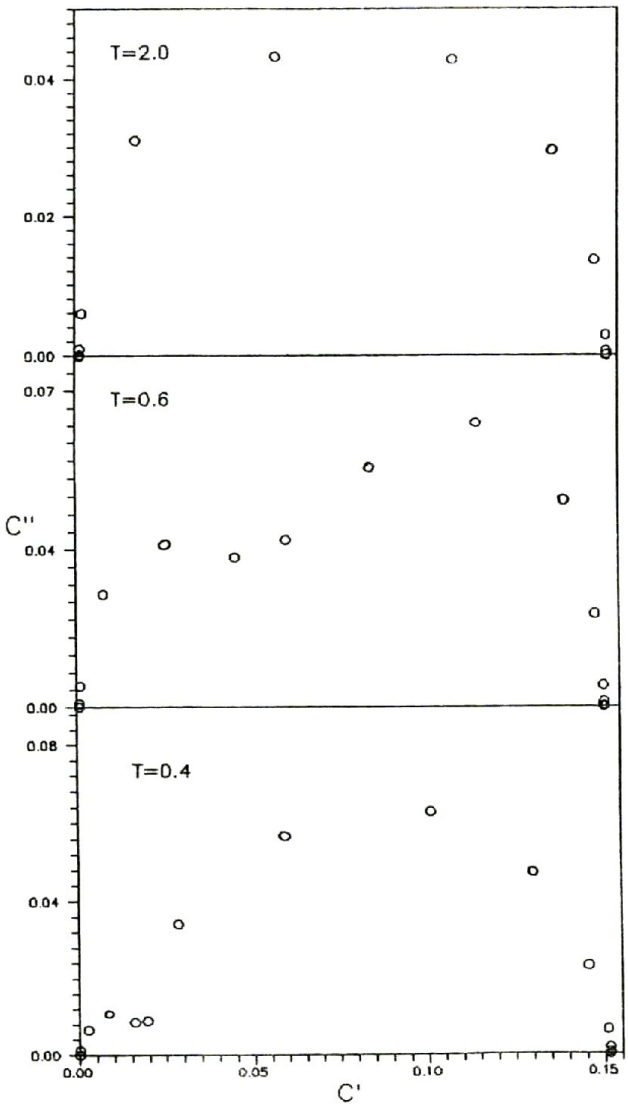

FIGURE 9. The Cole-Cole plot (*i.e. C'(ω)* vs *C"(ω)*) for *SG* and *DFM* systems at different temperatures.

and *DFM* rings shown in Fig. 1. At *T = 0.4*, the plot consists of two overlapping semicircles, indicating the time separation of modes. The left circle is very small compared with the right one. On increasing *T* beyond *0.6*, the left circle increases gradually then these two semicircles merge together to be only one at *T = 2.0*. At very low temperature, we got three overlapping semicircles but we can not draw the third one (lies on the left side). Its values are too small to be visible with respect to others.

We investigated many other different samples for both *SG* and *DFM* systems and we obtained the same features and properties like what we have here. More studies for spin systems might motivate experimentalists to undertake measurements of $C(\omega)$ for spin glasses and disordered ferromagnets. The *DFM* rings are very much similar to *SG* ones and the difference turns to be quantitative only.

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