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Nonequilibrium Thermodynamics of Ising Magnets

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1. Introduction

Real magnets and Ising models have provided a rich and productive field for the interaction between theory and experiment over the past 86 years (Ising, 1925). In order to identify the real magnets with a simple microscopic Hamiltonian, one needs to understand the behaviour of individual magnetic ions in crystalline environment (Wolf, 2000). Spin-1/2 Ising model and its variants such as Blume-Capel, Blume-Emery-Griffiths and mixed spin models were regarded as theoretical simplifications, designed to model the essential aspects of cooperative systems without detailed correspondence to specific materials. The similarities and differences between theoretical Ising models and a number of real magnetic materials were widely reviewed by many authors. The early experiments were focused on identifying Ising-like materials and characterizing the parameters of the microscopic Hamiltonian. Various approximate calculations were then compared with thermodynamic measurements. Although both the theoretical and experimental studies concerning Ising-like systems have concentrated on static properties, very little has been said about its dynamic characteristics.

Lyakhimets (Lyakhimets, 1992) has used a phenomenological description to study the magnetic dissipation in crystalline magnets with induced magnetic anisotropy. In his study, the components of the second-order tensor which describes the induced anisotropy of the magnet were taken as thermodynamic variables and the nonequilibrium linear Onsager thermodynamics was formulated for the system. Such an approach reflects all symmetry characteristics of the relaxation problem. The relaxation parameters and their angular dependencies were formulated for spin waves and moving domain walls with the help of the dissipation function. The implications of nonequilibrium thermodynamics were also considered for magnetic insulators, including paramagnets, uniform and nonuniform ferromagnets (Saslow & Rivkin, 2008). Their work was concentrated on two topics in the damping of insulating ferromagnets, both studied with the methods of irreversible thermodynamics: (a) damping in uniform ferromagnets, where two forms of phenomenological damping were commonly employed, (b) damping in non-uniform insulating ferromagnets, which become relevant for non-monodomain nanomagnets. Using the essential idea behind nonequilibrium thermodynamics, the long time dynamics of these systems close to equilibrium was well defined by a set of linear kinetic equations for the magnetization of insulating paramagnets (and for ferromagnets). The dissipative properties

of these equations were characterized by a matrix of rate coefficients in the linear relationship of fluxes to appropriate thermodynamic forces.

Investigation of the relaxation dynamics of magnetic order in Ising magnets under the effect of oscillating fields is now an active research area in which one can treat the sound propagation as well as magnetic relaxation. In most classes of magnets, a very important role is played by the order parameter relaxation time and it is crucial parameter determining the sound dynamics as well as dynamic susceptibility. As a phenomenological theory, nonequilibrium thermodynamics deals with approach of systems toward steady states and examines relaxation phenomena during the approach to equilibrium. The theory also encompasses detailed studies of the stability of systems far from equilibrium, including oscillating systems. In this context, the notion of nonequilibrium phase transitions is gaining importance as a unifying theoretical concept.

In this article, we will focus on a general theory of Ising magnets based on nonequilibrium thermodynamic. The basics of nonequilibrium thermodynamics is reviewed and the time-reversal signature of thermodynamic variables with their sources and fluxes are discussed in Section 2. Section 3 then considers Ising spin models describing statics of ferromagnetic and antiferromagnetic orders in magnets. Section 4 contains a detailed description of the kinetic model based on coupled linear equations of motion for the order parameter(s). The effect of the relaxation process on critical dynamics of sound propagation and dynamic response magnetization is investigated in Section 5. Comparison with experiments is made and reasons for formulating a phenomenological theory of relaxation problem are given in Section 6. Finally, the open questions and future prospects in this field are outlined.

2. Basics of nonequilibrium thermodynamics

Nonequilibrium thermodynamics (NT), a scientific discipline of 20th century, was invented in an effort to rationalize the behavior of irreversible processes. The NT is a vast field of scientific endeavour with roots in physics and chemistry. It was developed in the wake of the great success of certain symmetry relations, known as Onsager reciprocal relations in the phenomenological laws. These symmetry relations between irreversible phenomena have found a wide field of application in all branches of the physical science and engineering, and more recently in a number of interdisciplinary fields, including environmental research and, most notably, the biological sciences. Above applications can be classified according to their tensorial character. First one has scalar phenomena. These include chemical reactions and structural relaxation phenomena. Onsager relations are of help in this case, in solving the set of ordinary differential equations which describe the simultaneous relaxation of a great number of variables. Second group of phenomena is formed by vectorial processes, such as diffusion, heat conduction and their cross effects (e.g. thermal diffusion). Viscous phenomena and theory of sound propagation have been consistently developed within the framework of nonequilibrium thermodynamics.

Before introducing the notion of nonequilibrium thermodynamics we shall first summarize briefly the linear and nonlinear laws between thermodynamic fluxes and forces. A key concept when describing an irreversible process is the macroscopic state parameter of an adiabatically isolated system. These parameters are denoted by A_i . At equilibrium the state parameters have values A_i^0 , while an arbitrary state which is near or far from the equilibrium may be specified by the deviations α_i from the equilibrium state:

$$\alpha_i = A_i - A_i^0. \quad (1)$$

It is known empirically that the irreversible flows, time derivatives of deviations ($J_i = \dot{\alpha}_i$), are linear functions of the thermodynamic forces (X_i)

$$J_i = \sum_j L_{ij} X_j, \quad (2)$$

where the quantities L_{ij} are called the phenomenological coefficients and the Eqs. (2) are referred to as the phenomenological equations. The coefficients L_{ij} obey either Onsager's reciprocal relations $L_{ij} = L_{ji}$ or Casimir's one $L_{ij} = -L_{ji}$. These relations, also known as Onsager-Casimir reciprocal relations (Onsager, 1931; Casimir, 1945; De Groot, 1963), express an important consequence of microscopic time-reversal invariance for the relaxation of macroscopic quantities in the linear regime close to thermodynamic equilibrium. The proof of these relations involves the assumption that the correlation functions for the thermal fluctuations of macroscopic quantities decay according to the macroscopic relaxation equation.

It is well known that the entropy of an isolated system reaches its maximum value at equilibrium: so that any fluctuation of the thermodynamic parameters results with a decrease in the entropy. In response to such a fluctuation, entropy-producing irreversible process spontaneously drive the system back to equilibrium. Consequently, the state of equilibrium is stable to any perturbation that reduces the entropy. In contrast, one can state that if the fluctuations are growing, the system is not in equilibrium. The fluctuations in temperature, volume, magnetization, quadrupole moment, etc. are quantified by their magnitude such as δT , δV , δM and δQ the entropy of a magnetic system is a function of these parameters in general one can expand the entropy as power series in terms of these parameters:

$$S = S_{eq} + \delta S + \frac{1}{2} \delta^2 S + O(\delta^3 S), \quad (3)$$

In this expansion, the second term represents the *first-order* terms containing δT , δV , δM , δQ , etc., the third term indicates the *second-order* terms containing $(\delta T)^2$, $(\delta V)^2$, $(\delta M)^2$, $(\delta Q)^2$, etc., and so on. On the other hand, since the entropy is maximum, the *first-order* terms vanishes whereas the leading contribution to the increment of the entropy originates from the *second-order* term $\delta^2 S$ (Kondepudi & Prigogine, 2005).

The thermodynamic forces in Eqs. (2) are the intensive variables conjugate to the variables α_i :

$$X_i = \left. \frac{\partial S}{\partial \alpha_i} \right|_{\alpha_i}, \quad (4)$$

where S is the entropy of the system described by the fundamental relation $S = S(\alpha_1, \dots, \alpha_n)$. The Eqs. (2) could be thought of as arising from a Taylor-series expansion of the fluxes in

terms of the forces. Such a Taylor series will only exist if the flux is an analytic function of the forces at $X = 0$:

$$J_i(X) = J_i(0) + \sum_j \left. \frac{\partial J_i}{\partial X_j} \right|_{X=0} X_j + \sum_{j,k} \frac{1}{2!} \left. \frac{\partial^2 J_i}{\partial X_j \partial X_k} \right|_{X=0} X_j X_k + O(X^3). \quad (5)$$

Clearly the first term in Eq. (5) is zero as the fluxes vanish when the thermodynamic forces are zero. The term which is linear in the forces is evidently derivable, at least formally, from the equilibrium properties of the system as the functional derivative of the fluxes with respect to the forces computed at equilibrium, $X = 0$. The quadratic term is related to what are known as the nonlinear contributions to the linear theory of irreversible thermodynamics. In general, Eq. (5) may be written as nonlinear functions of the forces in the expanded form

$$J_i(X) = \sum_j L_{ij} X_j + \sum_{j,k} M_{ijk} X_j X_k + \sum_{j,k,l} N_{ijkl} X_j X_k X_l + \dots, \quad (6)$$

where the coefficients defined by

$$L_{ij} = \left. \frac{\partial J_i}{\partial X_j} \right|_{X=0}, \quad M_{ijk} = \frac{1}{2} \left. \frac{\partial^2 J_i}{\partial X_j \partial X_k} \right|_{X=0}, \quad N_{ijkl} = \frac{1}{6} \left. \frac{\partial^3 J_i}{\partial X_j \partial X_k \partial X_l} \right|_{X=0}. \quad (7)$$

Here the coefficients L_{ij} are the cross coefficients which are scalar in character. The second order coefficients M_{ijk} are vectorial. The third order coefficients N_{ijkl} are again scalar. Within the linear range, there is a lot of experimental evidence of Onsager relation.

In the nonlinear thermodynamic theory, a nonlinear generalization of Onsager's reciprocal relations was obtained using statistical methods (Hurley & Garrod, 1982). Later, the same generalization was also proved with pure macroscopic methods (Verhas, 1983). The proof of the generalization is based on mathematical facts. None of these generalizations are of general validity. The principle of macroscopic reversibility proposed by Meixner gives a good insight to the structure of the Onsager-Casimir reciprocal relations and says that the entropy production density is invariant under time inversion if it is quadratic function of independent variables. Demanding its validity to higher order leads to conflict only with the rules of the chemical reactions (Meixner, 1972).

3. Ising model and equilibrium properties based on the mean field approximation

In this section, we consider the Ising model on a regular lattice where each interior site has the same number of nearest-neighbour sites. This is called the coordination number of the lattice and will be denoted by z . We assume that, in the thermodynamic limit, boundary sites can be disregarded and that, with N sites, the number of nearest-neighbour site pairs is $Nz/2$. The standard Hamiltonian for the the simplest Ising model is given by

$$H = -J \sum_{\langle ij \rangle} s_i s_j - h \sum_i s_i,$$

with

$$s_i = \pm 1, \quad (8)$$

where h is the external magnetic field at the site i and the summation $\sum_{\langle ij \rangle}$ is performed for nearest-neighbour sites. J is the exchange interaction between neighbouring sites $\langle ij \rangle$. Two distinctive cases corresponding to different signs of intersite interaction is considered, i.e., $J < 0$ (ferromagnetic coupling) and $J > 0$ (antiferromagnetic coupling). On the other hand, Eq. (8) may be extended by allowing values $s = 0, \pm 1, \pm 2, \dots, \pm S$ for the variables. It is then possible to consider higher order interactions such as $K \sum_{\langle ij \rangle} s_i^2 s_j^2$ or a chemical potential such as $\Delta \sum_i s_i^2$. These generalizations are regarded as extensions of the Blume-Emery-Griffiths model (BEG) (Blume *et al.*, 1971). Recently, there have been many theoretical studies of mixed spin Ising systems. These are of interest because they have less translational symmetry than their single-spin counterparts since they consist of two interpenetrating inequivalent sublattices. The latter property is very important to study a certain type of ferrimagnetism, namely molecular-based magnetic materials which are of current interest (Kaneyoshi & Nakamura, 1998).

For sake of the brevity, here we will focus on the equilibrium properties of the $S = 1/2$ case which is described by the Hamiltonian given in Eq. (8). The Gibbs free energy depends on the three extensive variables ψ , N , and V . Here ψ , N , and V are magnetization per site, the total number of Ising spins and the volume of the lattice, respectively. Using the definition of the entropy the configurational Gibbs free energy in the Curie-Weiss approximation G ($G = E - TS - h\psi$) is obtained

$$G(V(a), \psi, h, T) = G_0(V, T) - \frac{1}{2} NJz\psi^2 + NkT \left[\left(\frac{1+\psi}{2} \right) \ln \left(\frac{1+\psi}{2} \right) + \left(\frac{1-\psi}{2} \right) \ln \left(\frac{1-\psi}{2} \right) \right] - h\psi, \quad (9)$$

where a , k , T are the lattice constant, the Boltzmann factor, the absolute temperature, respectively. $G_0(V, T)$ is the lattice free energy which is independent of spin configuration. One can see that G is an even function of ψ . Thus the second derivative of G with respect to ψ is

$$\frac{\partial^2 G}{\partial \psi^2} = -NJz + \frac{NkT}{1-\psi^2}, \quad (10)$$

and we define the critical temperature T_C by

$$T_C = Jz. \quad (11)$$

From Eq. (10), it is seen that the G vs. ψ curve is convex downwards for all ψ in the range $(-1, +1)$ for $T > T_C$, as shown in Figure 1. At $T = T_C$ the curvature changes sign to becomes convex upwards for $T < T_C$. The magnetic field h is conjugate to magnetization density ψ and from the fundamental relations of the thermodynamics one can write the following expression

$$h = \frac{\partial G}{\partial \psi} = -NJz\psi + \frac{1}{2}NkT \ln \frac{1+\psi}{1-\psi}. \quad (12)$$

To find the magnetization we solve Eq. (12) for ψ and obtain the equation of state (or self-consistent equation):

$$\psi = \tanh(\beta(Jz\psi + h)). \quad (13)$$

Now, using the definition

$$\chi(T) = \left. \frac{\partial \psi}{\partial h} \right|_{h \rightarrow 0}, \quad (14)$$

one obtains the following expression for the susceptibility

$$\chi(T) = \frac{1 - \tanh^2(z\psi/T + h)}{T \left\{ 1 - \frac{(1 - \tanh^2(z\psi/T + h))z}{T} \right\}}. \quad (15)$$

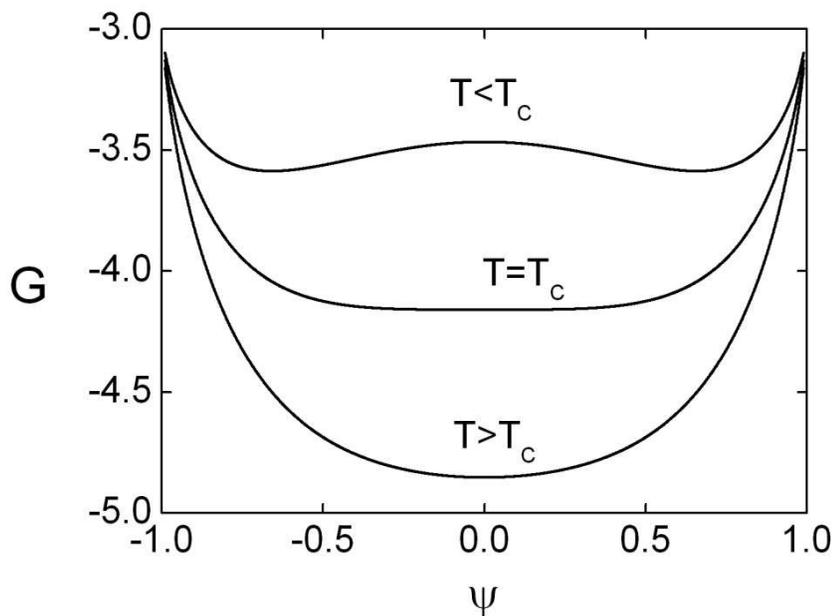


Fig. 1. Free energy-magnetization isotherms ($z = 6$)

Among the physical systems which undergo phase transitions, the most interesting class is the ferromagnet-paramagnet transitions in simple magnets. The free energy in such systems is nonanalytical function of its arguments. This is a manifestation of very strong fluctuations of quantity called order parameter. Phase transformations in ferromagnets are the continuous phase transitions which show no latent heat, seen in Figure 2. On the other hand, many physical quantities such as specific heat and static susceptibility diverge to infinity or tend to zero when approaching the critical temperature T_C . The behaviour of the static

susceptibility of an Ising ferromagnet on a simple cubic lattice ($z = 6$) in the neighborhood of the critical point is shown in Figure 3. One can see that the static susceptibility diverges at the critical point on both sides of the critical region (Lavis & Bell, 1998).

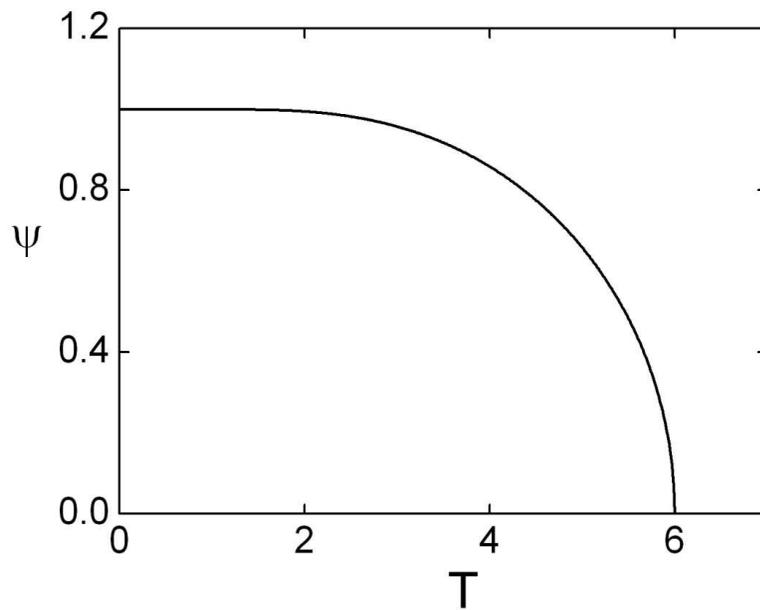


Fig. 2. The spontaneous magnetization plotted against temperature ($z = 6$)

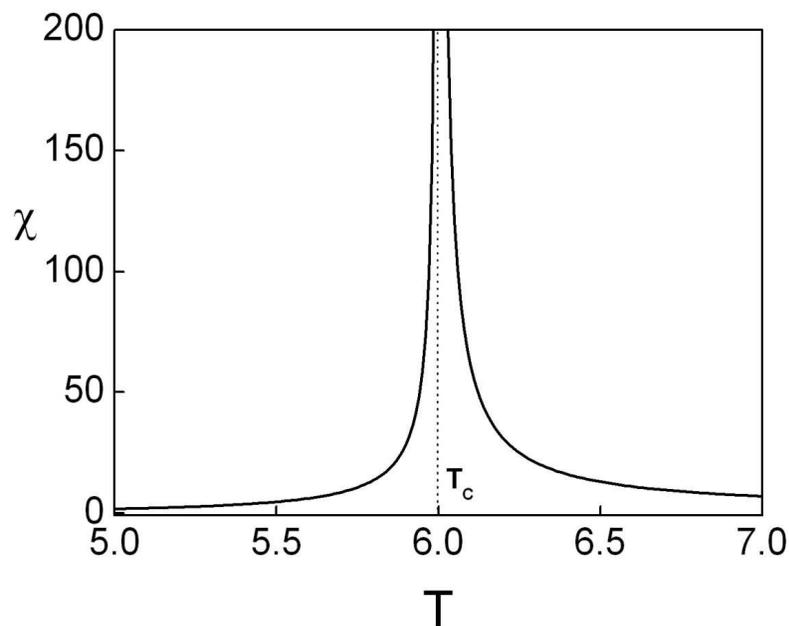


Fig. 3. The temperature dependence of the static susceptibility for a cubic lattice ($z = 6$)

4. Thermodynamic description of the kinetic model

In this section, a molecular-field approximation for the magnetic Gibbs free-energy production is used and a generalized force and a current are defined within the irreversible

thermodynamics. Then the kinetic equation for the magnetization is obtained within linear response theory. Finally, the temperature dependence of the relaxation time in the neighborhood of the phase-transition points is derived by solving the kinetic equation of the magnetization. For a simple kinetic model of Ising magnets, we first define the time-dependent long-range order parameter $\psi(t)$ (or magnetization), describing the ferromagnetic ordering, as the thermodynamic variable. In the nonequilibrium theory of the Ising system, the relaxation towards equilibrium is described the equation

$$\dot{\psi} = -\frac{\psi - \bar{\psi}}{\tau}, \quad (16)$$

where τ is the relaxation time characterizing the rate at which the magnetization ψ approaches the equilibrium ($\bar{\psi}$). Eq. (16) is the simplest equation of irreversible thermodynamics (De Groot & Mazur, 1962) and can also be written in the form

$$\dot{\psi} = LX, \quad (17)$$

where L is the rate constant (or kinetic coefficient) and X is the thermodynamic force which causes the current $\dot{\psi}$. In Eq. (17) X is found from the derivative of mean-field Gibbs energy production (ΔG) with respect to deviation of magnetization from the equilibrium:

$$X = \frac{d(\Delta G)}{d(\psi - \bar{\psi})}, \quad (18)$$

with

$$\Delta G = \frac{1}{2} \left[A(\psi - \bar{\psi})^2 + 2B(\psi - \bar{\psi})(h - \bar{h}) + C(h - \bar{h})^2 + D(\psi - \bar{\psi})(a - \bar{a}) + E(h - \bar{h})(a - \bar{a}) + F(a - \bar{a})^2 + G'(h - \bar{h}) \right], \quad (19)$$

In Eq. (19), the coefficients are called as Gibbs production coefficients:

$$A = \left(\frac{\partial^2 G}{\partial \psi^2} \right)_{eq} = -\frac{N(-Jz + Jz\bar{\psi}^2 + kT)}{-1 + \bar{\psi}^2}, \quad (20)$$

$$B = \left(\frac{\partial^2 G}{\partial \psi \partial h} \right)_{eq} = -1, \quad (21)$$

$$C = \left(\frac{\partial^2 G}{\partial h^2} \right)_{eq} = 0, \quad (22)$$

$$D = \left(\frac{\partial^2 G}{\partial \psi \partial a} \right)_{eq} = -Nz\bar{\psi} \left(\frac{\partial J}{\partial a} \right)_{eq}, \quad (23)$$

$$E = \left(\frac{\partial^2 G}{\partial h \partial a} \right)_{eq} = 0, \quad (24)$$

$$F = \left(\frac{\partial^2 G}{\partial a^2} \right)_{eq} = \left(\frac{\partial^2 G_0}{\partial a^2} \right)_{eq} - \frac{1}{2} Nz \bar{\psi}^2 \left(\frac{\partial^2 J}{\partial a^2} \right)_{eq}, \quad G' = \left(\frac{\partial G}{\partial h} \right)_{eq} = -\bar{\psi}. \quad (25)$$

The rate (or kinetic) equation is obtained using Eqs. (18)-(25) in the relaxation equation (Eq. (17)):

$$\dot{\psi} = LA(\psi - \bar{\psi}) + LB(h - \bar{h}) + DL(a - \bar{a}). \quad (26)$$

In order to find the relaxation time (τ) for the single relaxation process, one considers the rate equation when there is no external stimulation, i.e., $h = \bar{h}$, $a = \bar{a}$. Eq. (26) then becomes

$$\dot{\psi} = LA(\psi - \bar{\psi}). \quad (27)$$

Assuming a solution of the form $\psi - \bar{\psi} \approx \exp(-t/\tau)$ for Eq. (27), one obtains

$$\frac{1}{\tau} = -LA. \quad (28)$$

Using Eq. (20) yields

$$\tau = -\frac{1 - \bar{\psi}^2}{NL(-Jz + Jz\bar{\psi}^2 + kT)}. \quad (29)$$

The behaviour of the relaxation time near the phase-transition points can be derived analytically from the critical exponents. It is a well-known fact that various thermodynamic functions represents singular behavior as one approaches the critical point. Therefore, it is convenient to introduce an expansion parameter, which is a measure of the distance from the critical point ($\varepsilon = T - T_c$). Here T_c is the critical temperature given by Eq. (11). In the neighborhood of the transition point the relaxation time of the Ising model can be written in the form,

$$\tau(\varepsilon) = -\frac{1 - (\bar{\psi}(\varepsilon))^2}{NL(-Jz + Jz(\bar{\psi}(\varepsilon))^2 + k(\varepsilon + T_c))}. \quad (30)$$

In the vicinity of the second-order transition the magnetization vanishes at T_c as

$$\bar{\psi}(\varepsilon) = (\varepsilon)^{1/2}. \quad (31)$$

The critical exponent for the function $\tau(\varepsilon)$ is defined as

$$\lambda = \lim_{\varepsilon \rightarrow 0} \frac{\ln \tau(\varepsilon)}{\ln \varepsilon}. \quad (32)$$

This description is valid for all values of λ , where the negative value corresponds to the divergence of the variable $\tau(\varepsilon)$ as ε goes to zero, positive value corresponding to relaxation time that approaches zero, and the zero value corresponding to logarithmic divergence, jump singularity or a cusp (the relaxation time is finite at the critical point but one of its derivative diverges (Reichl, 1998)). On the other hand, in order to distinguish a cusp from a logarithmic divergence, another type of critical exponent, λ' , is introduced. To find the exponent λ' that describes the singular parts of τ with a cusplike singularity, we first find the smallest integer m for which the derivative $\tau^{(m)} = (\partial^m \tau / \partial \varepsilon^m)$ diverge as $\varepsilon \rightarrow 0$:

$$\lambda' = m + \lim_{\varepsilon \rightarrow 0} \frac{\ln \tau(\varepsilon)}{\ln \varepsilon}. \quad (33)$$

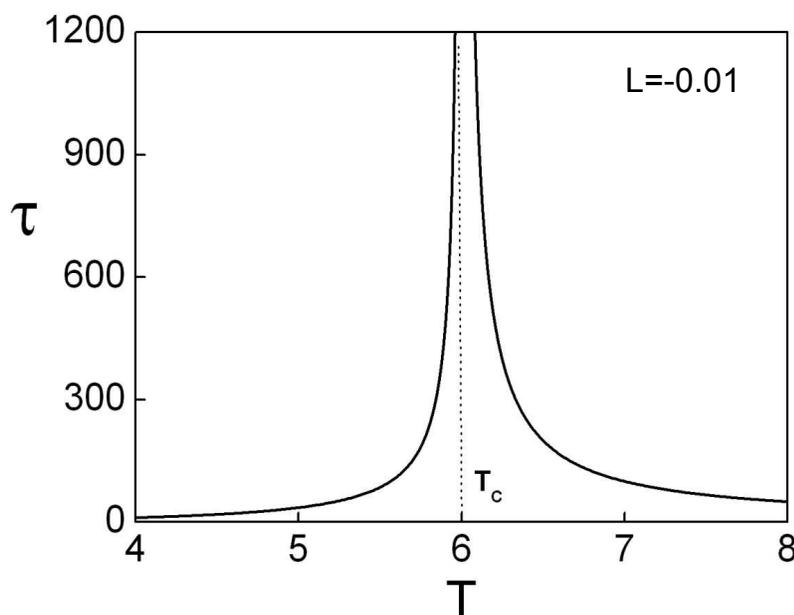


Fig. 4. Relaxation time vs temperature in the neighbourhood of critical point

The behavior of the relaxation time τ as a function of temperature is given in Figure 4. One can see from Figure 4 that τ grows rapidly with increasing temperature and diverges as the temperature approaches the second-order phase-transition point. In accordance with this behavior, the critical exponent of τ is found to be $\lambda = -1.0$. On the other hand, the scaling form of the relaxation time reads $\tau \approx \xi^z \approx |T - T_c|^{\nu z}$, where ξ , ν and z are the correlation length, critical exponent for ξ and dynamical critical exponent, respectively (Ray *et al.*, 1989). According to mean-field calculations, the dynamic critical exponent of the Ising model is $z = 2$ at the critical point. In addition to studies on Blume-Capel model which undergoes first-order phase transitions and represents rich variety of phase diagrams has revealed the fact that the dynamical critical exponent is also $z = 2$ at the critical endpoint, and double critical endpoints as well as tricritical point, whereas $z = 0$ for first-order critical transition points (Gulpinar & İyikanat, 2011). We should note that the analysis used in this article is identical to Landau-Ginzburg kinetic theory of phase transitions of a spatially

homogenous system. As is discussed extensively by Landau and Lifshitz (Landau & Lifshitz, 1981), in the case of spatially inhomogeneous medium where $\psi - \bar{\psi} = \partial\psi(t, r)$, the Landau-Ginzburg kinetic theory of critical phenomena reveals the fact that the relaxation time becomes finite for $T = T_c$ for components with $q \neq 0$. Here q is the Fourier transform of the spatial variable r . On the other hand, the renormalization-group formalism has proved to be very useful in calculating not only the static behavior but also the dynamic scaling. By making use of this method, Halperin *et al.* (Halperin *et al.*, 1974) found the critical-point singularity of the linear dynamic response of various models. The linear response theory, however, describes the reaction of a system to an infinitesimal external disturbance, while in experiments and computer simulations it is often much easier to deal with nonlinear-response situations, since it is much easier to investigate the response of the system to finite changes in the thermodynamic variables. A natural question is whether the critical-point singularity of the linear and nonlinear responses is the same. The answer is yes for ergodic systems, which reach equilibrium independently of the initial conditions (Racz, 1976). The assumption that the initial and intermediate stages of the relaxation do not affect the divergence of the relaxation time (motivated by the observation that the critical fluctuations appear only very close to equilibrium) led to the expectation that in ergodic systems τ^{nl} and τ^l diverge with same critical exponent. This view seemed to be supported by Monte Carlo calculations (Stoll *et al.*, 1973) and high-temperature series expansion of the two-dimensional one-spin flip kinetic Ising model. Later, Koch *et al.* (Koch *et al.*, 1996) presented field-theoretic arguments by making use of the Langevin equation for the one-component field $\phi(r, r)$ as well as numerical studies of finite-size effects on the exponential relaxation times τ_1 and τ_2 of the order parameter and the square of the order parameter near the critical point of three-dimensional Ising-like systems.

For the ferromagnetic interaction, a short range order parameter as well as the long range order is introduced (Tanaka *et al.*, 1962; Barry, 1966) while there are two long range sublattice magnetic orders and a short range order in the Ising antiferromagnets (Barry & Harrington, 1971). Similarly the number of thermodynamic variables (order parameters) also increases when the higher order interactions are considered (Erdem & Keskin, 2001; Gülpınar *et al.*, 2007; Canko & Keskin, 2010). For a general formulation of Ising spin kinetics with a multiple number of spin orderings (ψ_i), the Gibbs free energy production is written as

$$\Delta G = \frac{1}{2} \left\{ \sum_{i,j=1}^n \beta_{ij} (\psi_i - \bar{\psi}_i) (\psi_j - \bar{\psi}_j) + 2 \sum_{i=1}^n \left[\sum_{k=1}^m \left[\gamma_{ik} (\psi_i - \bar{\psi}_i) (h_k - \bar{h}_k) + \phi_k (h_k - \bar{h}_k)^2 + \eta_k (h_k - \bar{h}_k) (a - \bar{a}) \right] + \mu_i (\psi_i - \bar{\psi}_i) (a - \bar{a}) + \sigma (a - \bar{a})^2 \right] \right\}, \quad (34)$$

where the coefficients are defined as

$$\beta_{ij} = \left(\frac{\partial^2 G}{\partial \psi_i \partial \psi_j} \right)_{eq}, \quad \gamma_{ik} = \left(\frac{\partial^2 G}{\partial \psi_i \partial h_k} \right)_{eq}, \quad \phi_k = \left(\frac{\partial^2 G}{\partial h_k^2} \right)_{eq},$$

$$\eta_k = \left(\frac{\partial^2 G}{\partial h_k \partial a} \right)_{eq}, \quad \mu_i = \left(\frac{\partial^2 G}{\partial \psi_i \partial a} \right)_{eq}, \quad \sigma = \left(\frac{\partial^2 G}{\partial a^2} \right)_{eq}. \quad (35)$$

Then a set of linear rate equations may be written in terms of a matrix of phenomenological coefficients which satisfy the Onsager relation (Onsager, 1931):

$$\begin{bmatrix} \dot{\psi}_1 \\ \vdots \\ \dot{\psi}_n \end{bmatrix} = \begin{bmatrix} L_{11} & \dots & L_{1n} \\ \vdots & \ddots & \vdots \\ L_{n1} & \dots & L_{nn} \end{bmatrix} \begin{bmatrix} X_1 \\ \vdots \\ X_n \end{bmatrix}, \quad (36)$$

where the generalized forces are

$$X_j = \frac{\partial(\Delta G)}{\partial(\psi_j - \bar{\psi}_j)} = \sum_{i,j=1}^n \beta_{ij}(\psi_i - \bar{\psi}_i) + \sum_{i=1}^n \left[\mu_i(a - \bar{a}) + \sum_{k=1}^m \gamma_{ik}(h_k - \bar{h}_k) \right]. \quad (37)$$

The matrix equation given by Eq. (36) can be written in component form using Eq. (37), namely a set of n coupled, linear inhomogenous first-order rate equations. Embedding this relation into Eq. (36) one obtains the following matrix equation for the fluxes:

$$\hat{\psi} = \hat{L}\hat{\beta}\partial\hat{\psi} + \hat{L}\hat{\gamma}\hat{h} + \hat{L}a\hat{\mu}, \quad (38)$$

where the matrixes are defined by

$$\hat{\beta} = \begin{bmatrix} \beta_{11} & \dots & \beta_{1n} \\ \vdots & \ddots & \vdots \\ \beta_{n1} & \dots & \beta_{nn} \end{bmatrix}, \quad \hat{\gamma} = \begin{bmatrix} \gamma_{11} & \dots & \gamma_{1m} \\ \vdots & \ddots & \vdots \\ \gamma_{n1} & \dots & \gamma_{nm} \end{bmatrix}, \quad \hat{L} = \begin{bmatrix} L_{11} & \dots & L_{1n} \\ \vdots & \ddots & \vdots \\ L_{n1} & \dots & L_{nn} \end{bmatrix},$$

$$\hat{\psi} = \begin{bmatrix} \psi_1 \\ \vdots \\ \psi_n \end{bmatrix}, \quad \partial\hat{\psi} = \begin{bmatrix} \psi_1 - \bar{\psi}_1 \\ \vdots \\ \psi_n - \bar{\psi}_n \end{bmatrix}, \quad \hat{\mu} = \begin{bmatrix} \mu_1 \\ \vdots \\ \mu_n \end{bmatrix}, \quad \hat{h} = \begin{bmatrix} h_1 - \bar{h}_1 \\ \vdots \\ h_m - \bar{h}_m \end{bmatrix} \quad (39)$$

Since the phenomenological coefficients L_{ij} in matrix \hat{L} obey one of the reciprocal relations $L_{ij} = \pm L_{ji}$ according to microscopic time-reversal invariance of relaxing macroscopic quantities $\psi_i(t)$, the matrix may be symmetric or antisymmetric. In order to obtain the relaxation times, one considers the corresponding inhomogenous equations (Eq. (38)) resulting when the external fields are equal to their equilibrium values, i.e., $h_k = \bar{h}_k$ for $k = 1, \dots, m$ and $a = \bar{a}$. In the neighbourhood of the equilibrium states, solutions of the form

$\psi_i - \bar{\psi}_i \approx \exp(-t / \tau_i)$ are assumed for the linearized kinetic equations and approaches of the order parameters $\psi_i(t)$ to their equilibrium values are described by a set of characteristic times, also called relaxation times τ_i . To find each time (τ_i) one must solve the secular equation. Critical exponents (λ_i and λ_i' , $i=1, \dots, n$) for the functions $\tau_i(\varepsilon)$ are also calculated using Eqs. (32) and (33) to see the divergences, jumps, cusps etc. for the relaxation times $\tau_i(\varepsilon)$ at the transition points.

5. Critical behaviours of sound propagation and dynamic magnetic response

In this section, we will discuss the effect of the relaxation process on critical dynamics of sound propagation and dynamic response magnetization for the Ising magnets with single order parameter (ψ). Firstly we study the case in which the lattice is under the effect of a sound wave. Then the sound velocity and sound attenuation coefficient of the system are derived using the phenomenological formulation based on the method of thermodynamics of irreversible processes. The behaviors of these quantities near the phase transition temperatures are analyzed. Secondly, we consider case where the spin system is stimulated by a small uniform external magnetic field oscillating at an angular frequency. We examine the temperature variations of the non-equilibrium susceptibility of the system near the critical point. For this aim, we have made use of the free energy production and the kinetic equation describing the time dependency of the magnetization which are obtained in the previous section. In order to obtain dynamic magnetic response of the Ising system, the stationary solution of the kinetic equation in the existence of sinusoidal external magnetic field is performed. In addition, the static and dynamical mean field critical exponents are calculated in order to formulate the critical behavior of the magnetic response of a magnetic system.

In order to obtain the critical sound propagation of an Ising system we focus on the case in which the lattice is stimulated by the sound wave of frequency ω for the case $h = \bar{h}$. In the steady state, all quantities will oscillate with the same frequency ω and one can find a steady solution of the kinetic equation given by Eq. (26) with an oscillating external force $a - \bar{a} = a_1 e^{i\omega t}$. Assuming the form of solution $\psi(t) - \bar{\psi} = \psi_1 e^{i\omega t}$ and introducing this expression into Eq. (26), one obtains the following inhomogenous equation for ψ_1

$$i\omega \psi_1 e^{i\omega t} = LA\psi_1 e^{i\omega t} + LD a_1 e^{i\omega t} \quad (40)$$

Solving Eq. (40) for ψ_1 / a_1 gives

$$\frac{\psi_1}{a_1} = \frac{LD}{i\omega - LA} = \frac{\tau LD}{1 + i\omega\tau} \quad (41)$$

The response in the pressure ($p - \bar{p}$) is obtained by differentiating the minimum work with respect to $(V - \bar{V})$ and using Eqs. (9) and (19)

$$p - \bar{p} = \frac{\partial \Delta G}{\partial (V - \bar{V})} = -\frac{\bar{a}}{3\bar{V}} \frac{\partial \Delta G}{\partial (a - \bar{a})} \quad (42)$$

then

$$p - \bar{p} = -\frac{\bar{a}}{3\bar{V}} [D(\psi - \bar{\psi}) + F(a - \bar{a})] . \quad (43)$$

Finally, the derivative of the pressure with respect to volume gives

$$\left(\frac{\partial p}{\partial V}\right)_{sound} = \left(\frac{\bar{a}}{3\bar{V}}\right)^2 \left[F + D \frac{\psi_1}{a_1} \right] . \quad (44)$$

Here F and D are given by Eqs. (23) and (25). Introducing the relation (41) and the density $\rho = M / \bar{V}$ into Eq. (44) one obtains

$$\left(\frac{\partial p}{\partial \rho}\right)_{sound} = \frac{\bar{a}^2}{9M} \left[\left(\frac{\partial^2 G_0}{\partial a^2}\right)_{eq} - \frac{1}{2} Nz \bar{\psi}^2 \left(\frac{\partial^2 J}{\partial a^2}\right)_{eq} + \frac{\tau LD^2}{1 + i\omega\tau} \right] . \quad (45)$$

From the real and imaginary parts of Eq. (45) one obtains the velocity of sound and attenuation coefficient for a single relaxational process as

$$c(\omega, T) = \text{Re}[c_{sound}] = c_0 \left[1 - \frac{Nz\bar{a}^2\bar{\psi}^2}{18Mc_0^2} \left(\frac{\partial^2 J}{\partial a^2}\right) + LD^2 \frac{\tau}{1 + \omega^2\tau^2} \right] , \quad (46)$$

$$\alpha(\omega, T) = \text{Im}\left[-\frac{\omega}{c_{sound}}\right] = LD^2 \frac{\omega^2\tau^2}{1 + \omega^2\tau^2} , \quad (47)$$

where c_0 is the velocity of sound at very high frequencies or at very high temperatures and $c_{sound} = (\partial p / \partial \rho)^{1/2}$ is the a complex expression for sound velocity. We perform some calculations for the frequency and temperature dependencies of $c(\omega, T)$ and $\alpha(\omega, T)$. Figures (5) and (6) show these dependencies. From the linear coupling of a sound wave with the order parameter fluctuations $(\psi - \bar{\psi})$ in the Ising system, the dispersion which is relative sound velocity change displays a frequency-dependent velocity or dispersion minimum (Figure 5) while the attenuation exhibits a frequency-dependent broad peak (Figure 6) in the ordered phase. Calculations of $c(T)$ and $\alpha(T)$ for the simple Ising spin system reveals the same features as in real magnets, i.e. the shifts of the velocity minima and attenuation maxima to lower temperatures with increasing frequency are seen. The velocity minima at each frequency occur at temperatures lower than the corresponding attenuation maxima observed for the same parameters used. The notions of minimum in sound velocity and maximum in attenuation go back to Landau and Khalatnikov (Landau & Khalatnikov, 1954; Landau & Khalatnikov, 1965) who study a more general question of energy dissipation mechanism due to order parameter relaxation. Their idea was based on the slow relaxation of the order parameter. During this relaxation it allows internal irreversible processes to be switch on so as to restore local equilibrium; this increases the entropy and involves energy dissipation in the system. In the critical region, behaviours of both quantities are verified analytically from definition of critical exponents given in Eq. (32) for the functions $c(\varepsilon)$ and

$\alpha(\varepsilon)$. It is found that the dispersion just below the critical temperature is expressed as $c(\varepsilon) \propto \varepsilon^0$ while the attenuation goes to zero as $\alpha(\varepsilon) \propto \varepsilon$.

In the presence of many thermodynamic variables for more complex Ising-type magnets, there exist more than one relaxational process with relaxational times (τ_i). Contribution of these processes to the sound propagation were treated in more recent works using the above technique in the general phenomenological formulation given in the previous section. Dispersion relation and attenuation coefficient for the sound waves of frequency ω were derived for several models with an Ising-type Hamiltonian (Keskin & Erdem, 2003; Erdem & Keskin, 2003; Gulpinar, 2008; Albayrak & Cengiz, 2011). In these works, various mechanisms of the sound propagation in Ising-type magnets were given and origin of the critical attenuation with its exponent was discussed.

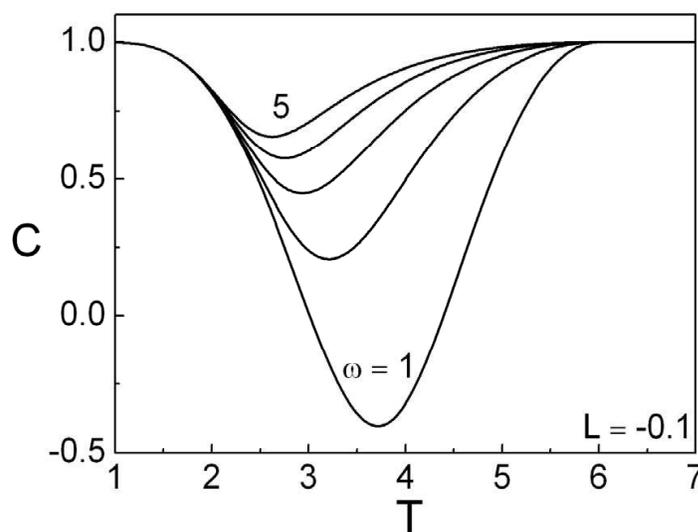


Fig. 5. Sound dispersion $c(T)$ at different frequencies ω for $L = 10$

Similarly, theoretical investigation of dynamic magnetic response of the Ising systems has been the subject of interest for quite a long time. In 1966, Barry has studied spin-1/2 Ising ferromagnet by a method combining statistical theory of phase transitions and irreversible thermodynamics (Barry, 1966). Using the same method, Barry and Harrington has focused on the theory of relaxation phenomena in an Ising antiferromagnet and obtained the temperature and frequency dependencies of the magnetic dispersion and absorption factor in the neighborhood of the Neel transition temperature (Barry & Harrington, 1971). Erdem investigated dynamic magnetic response of the spin-1 Ising system with dipolar and quadrupolar orders (Erdem, 2008). In this study, expressions for the real and imaginary parts of the complex susceptibility were found using the same phenomenological approach proposed by Barry. Erdem has also obtained the frequency dependence of the complex susceptibility for the same system (Erdem, 2009). In Ising spin systems mentioned above, there exist two or three relaxing quantities which cause two or three relaxation contributions to the dynamic magnetic susceptibility. Therefore, as in the sound dynamics case, a general formulation (section 4) is followed for the derivation of susceptibility expressions. In the

following, we use, for simplicity, the theory of relaxation with a single characteristic time to obtain an explicit form of complex susceptibility.

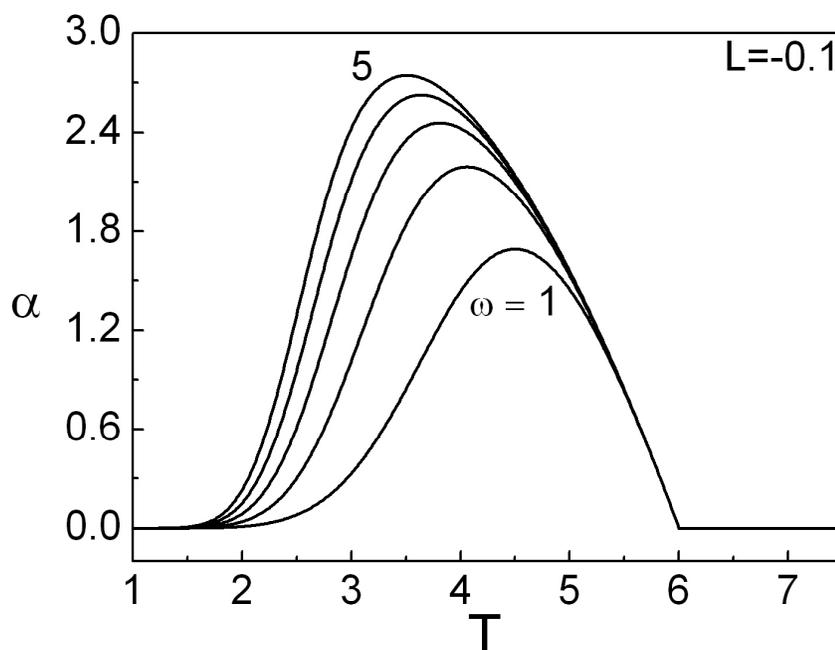


Fig. 6. Sound attenuation $\alpha(T)$ at different frequencies ω for $L = 10$

If the spin system described by Eq. (8) is stimulated by a time dependent magnetic field $h(t) = h_1 e^{i\omega t}$ oscillating at an angular frequency ω , the order parameter of the system will oscillate near the equilibrium state at this same angular frequency at the stationary state:

$$\psi(t) - \bar{\psi} = \psi_1 e^{i\omega t}, \quad (48)$$

If this equation is substituted into the kinetic equation Eq. (17) we find following form:

$$i\omega \psi_1 e^{i\omega t} = LA\psi_1 e^{i\omega t} + LBh_1 e^{i\omega t}. \quad (49)$$

Solving Eq. (49) for ψ_1 / h_1 gives

$$\frac{\psi_1}{h_1} = \frac{LB}{i\omega - LA} \quad (50)$$

Eq. (50) is needed to calculate the complex initial susceptibility $\chi(\omega)$. The Ising system induced magnetization (total induced magnetic moment per unit volume) is given by

$$\psi(t) - \psi_\infty = \text{Re}(\psi_1 e^{i\omega t}), \quad (51)$$

where ψ_∞ is the magnetization induced by a magnetic field oscillating at ω . Also, by definition, the expression for $\chi(\omega)$ may be written

$$\psi(t) - \psi_\infty = \text{Re} \left[\chi(\omega) h_1 e^{i\omega t} \right], \quad (52)$$

where $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$ is the complex susceptibility whose real and imaginary parts are called as magnetic dispersion and absorption factors respectively. Comparing Eqs. (38) to Eq. (40) one may write

$$\chi(\omega) = \frac{\psi_1}{h_1}. \quad (53)$$

Finally the magnetic dispersion and absorption factors become

$$\chi'(\omega) = \frac{AL^2}{A^2L^2 + \omega^2} = LB \frac{\tau}{1 + \omega^2\tau^2}. \quad (54)$$

$$\chi''(\omega) = \frac{L\omega}{A^2L^2 + \omega^2} = LB \frac{\tau^2\omega}{1 + \omega^2\tau^2}. \quad (55)$$

In Figures 7 and 8 we illustrate the temperature variations of the magnetic dispersion and absorption factor in the low frequency limit $\omega\tau \ll 1$. These plots illustrate that both $\chi'(\omega)$ and $\chi''(\omega)$ increase rapidly with temperature and tend to infinity near the phase transition temperature. The divergence of $\chi'(\omega)$ does not depend on the frequency while the divergence of $\chi''(\omega)$ depends on ω and gets pushed away from the critical point as ω increases. When compared with the static limit ($\omega \rightarrow 0$) mentioned in section 3, a good agreement is achieved. Above critical behaviours of both components for the regime $\omega\tau \ll 1$ may be verified by calculating the critical exponents for the functions $\chi'(\varepsilon)$ and $\chi''(\varepsilon)$ using Eq. (32). Results of calculation indicates that $\chi'(\varepsilon)$ and $\chi''(\varepsilon)$ behave as ε^{-1} and ε^{-2} , respectively.

Finally the high frequency behavior ($\omega\tau \gg 1$) of the magnetic dispersion and absorption factor are given in Figures 9 and 10. The real part $\chi'(\omega)$ has two frequency-dependent local maxima in the ordered and disordered phase regions. When the frequency increases, the maximum observed in the ferromagnetic region decreases and shifts to lower temperatures. The peak observed in the paramagnetic region also decreases but shifts to higher temperatures. On the other hand, the imaginary part $\chi''(\omega)$ shows frequency-dependent maxima at the ferromagnetic-paramagnetic phase transition point. Again, from Eq. (32), one can show that the real part converges to zero ($\chi'(\varepsilon) \propto \varepsilon$) and the imaginary part displays a peak at the transition ($\chi''(\omega) \propto \varepsilon^0$) as $\varepsilon \rightarrow 0$.

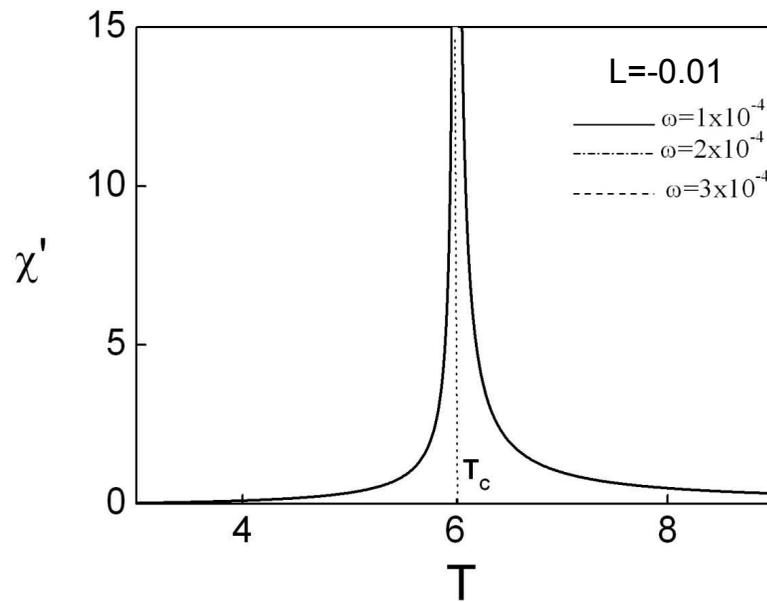


Fig. 7. Magnetic dispersion $\chi'(\omega)$ vs temperature for the low frequency limit ($\omega\tau \ll 1$ in the neighbourhood of critical point

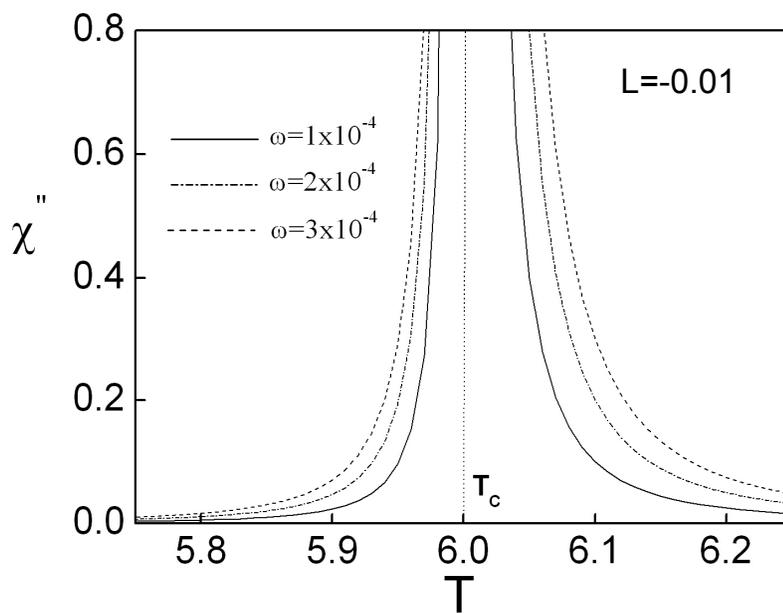


Fig. 8. Same as Figure 7 but for the magnetic absorption factor $\chi''(\omega)$

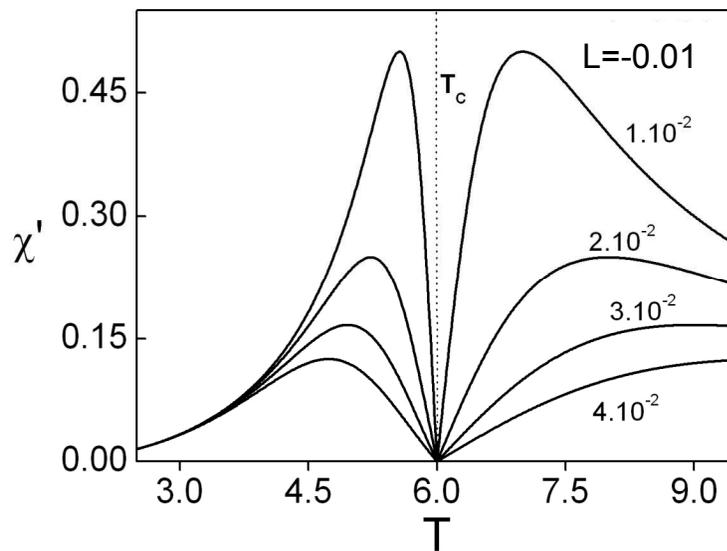


Fig. 9. Magnetic dispersion $\chi'(\omega)$ vs temperature for the high frequency limit ($\omega\tau \gg 1$) in the neighbourhood of critical point

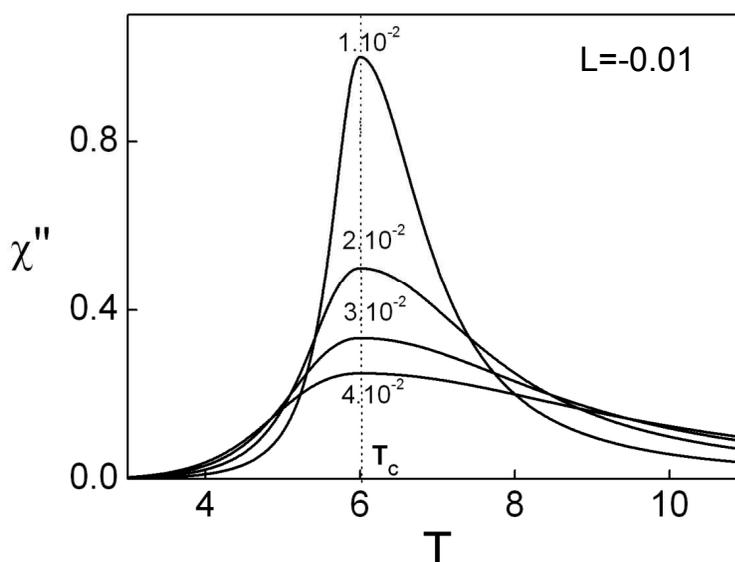


Fig. 10. Same as Figure 9 but for the magnetic absorption factor $\chi''(\omega)$

6. Comparison of theory with experiments

The diverging behavior of the relaxation time and corresponding slowing down of the dynamics of a system in the neighborhood of phase transitions has been a subject of experimental research for quite a long time. In 1958, Chase (Chase, 1958) reported that liquid helium exhibits a temperature dependence of the relaxation time consistent with the scaling relation $(T - T_c)^{-1}$. Later Naya and Sakai (Naya & Sakai, 1976) presented an analysis of the critical dynamics of the polyorientational phase transition, which is an extension of the statistical equilibrium theory in random phase approximation. In addition, Schuller and

Gray (Schuller & Gray, 1976) have shown that the relaxation time of the superconducting order parameter diverges close to the transition temperature, in accordance with the theoretical prediction of several authors (Lucas & Stephen, 1967; Schmid & Schon, 1975). Recently, Sperkach et al. (Sperkach et al, 2001) measured the temperature dependence of acoustical relaxation times in the vicinity of a nematic-isotropic phase-transition point in 5CB liquid crystal. Comparing Figures 2(a) and Fig. 5 of their work one can observe the similarity between the temperature-dependent behavior of the low-frequency relaxation time of the 5CB liquid crystal and the Blume-Capel model with random single-ion anisotropy (Gulpinar & İyikanat, 2011). Moreover, very recently, Ahart et al. (Ahart et al., 2009) reported that a critical slowing down of the central peak. These results indicate that the relaxation time of the order parameter for an Ising magnet diverges near the critical point, which corresponds to a familiar critical slowing down.

It is well known fact that measurements of sound propagation are considered useful in investigating the dynamics of magnetic phase transitions and therefore many experimental and theoretical studies have been carried out. Various aspects of ultrasonic attenuation in magnetic insulators (Lüthi & Pollina, 1969; Moran & Lüthi, 1971) and in magnetic metals (Lüthi et al., 1970; Maekawa & Tachiki, 1978) have been studied. In these works, the transition temperature was associated with the experimentally determined peaks whose maximum shift towards the lower temperatures as the sound frequency increases. Similarly, acoustic studies, especially those of dispersion, have also been made on several magnetic systems such as transition metals (Golding & Barmatz, 1969), ferromagnetic insulators (Bennett, 1969) and antiferromagnetic semiconductors (Walter, 1967). It was found that the critical changes in sound velocity show a uniform behaviour for all substances studied, namely, a frequency-independent and weak temperature-dependent effect. It was also found that, in the ordered phase, the minima of the sound velocity shifted to lower temperatures with increasing frequency (Moran & Lüthi, 1971).

Dynamic response of a spin system to a time-varying magnetic field is an important subject to probe all magnetic systems. It is also called AC or dynamic susceptibility for the magnetization. The dynamic susceptibility is commonly used to determine the electrical properties of superconductors (Kılıç et al, 2004) and magnetic properties of some spin systems such as spin glasses (Körtzler & Eiselt, 1979), cobalt-based alloys (Durin et al., 1991), molecule-based magnets (Girtu, 2002), magnetic fluids (Fannin et al., 2005) and nanoparticles (Van Raap et al., 2005). The dynamic magnetic response of these materials and the development of methods for its modification are important for their potential applications. For example, cores made of cobalt-based alloys in low signal detectors of gravitational physics contribute as a noise source with a spectral density proportional to the ac susceptibility of the alloy. The knowledge of dynamic susceptibility for nanocomposite particles is very important for the design of magneto-optical devices.

7. Conclusion

In this chapter, we have discussed a simple kinetic formulation of Ising magnets based on nonequilibrium thermodynamics. We start with the simplest relaxation equation of the irreversible thermodynamics with a characteristic time and mention a general formulation based on the research results in the literature for some well known dynamic problems with more than one relaxational processes. Recent theoretical findings provide a more precise

description for the experimental acoustic studies and magnetic relaxation measurements in real magnets.

The kinetic formulation with single relaxation process and its generalization for more coupled irreversible phenomena strongly depend on a statistical equilibrium description of free energy and its properties near the phase transition. The effective field theories of equilibrium statistical mechanics, such as the molecular mean-field approximation is used as this century-old description of free energy. However, because of its limitations, such as neglecting fluctuation correlations near the critical point and low temperature quantum excitations, these theories are invaluable tools in studies of magnetic phase transitions. To improve the methodology and results of mean-field analysis of order parameter relaxation, the equilibrium free energy should be obtained using more a reliable theory including correlations. This was recently given on the Bethe lattice using some recursion relations. The first major application of Bethe-type free energy for the relaxation process was on dipolar and quadrupolar interactions to study sound attenuation problem (Albayrak & Cengiz, 2011).

Bethe lattice treatment of phenomenological relaxation problem mentioned above has also some limitations. It predicts a transition temperature higher than that of a bravais lattice. Also, predicting the critical exponents is not reliable. Therefore, one must consider the relaxation problem on the real lattices using more reliable equilibrium theories to get a much clear relaxation picture. In particular, renormalization group theory of relaxational sound dynamics and dynamic response would be of importance in future.

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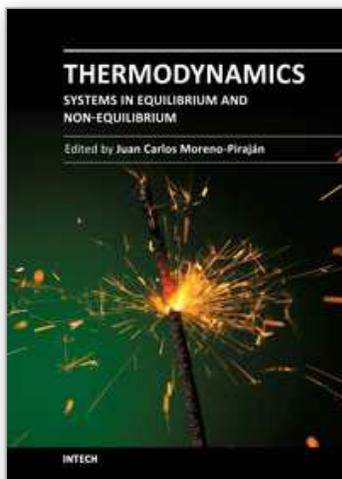
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