Critical Behavior of AC Antiferromagnetic and Ferromagnetic Susceptibilities of a Spin-1/2 Metamagnetic Ising System

Gul Gulpinar a , Erol Vatansever a

Dokuz Eylul University, Department of Physics, 35160-Buca, Izmir, Turkey

Abstract

In this study, the temperature variations of the equilibrium and the non-equilibrium antiferromagnetic and ferromagnetic susceptibilities of a metamagnetic system are examined near the critical point. The kinetic equations describing the time dependencies of the total and staggered magnetizations are derived by utilizing linear response theory. In order to obtain dynamic magnetic relaxation behavior of the system, the stationary solutions of the kinetic equations in existence of sinusoidal staggered and physical external magnetic fields are performed. In addition, the static and dynamical mean field critical exponents are calculated in order to formulate the critical behavior of antiferromagnetic and ferromagnetic magnetic response of a metamagnetic system. Finally, a comparison of the findings of this study with previous theoretical and experimental studies is represented and it is shown that a good agreement is found with our results.

Key words: Irreversible Thermodynamics, Staggered Magnetic Susceptibility, Direct Magnetic Susceptibility, Metamagnetism, Mean Field Dynamic Critical Exponents.

1 Introduction

Investigation of the AC or dynamic susceptibilities are the most preferred techniques to study Magnetic relaxation (MR) which is related to magnetic

* corresponding author.

Email address: gul.gulpinar@deu.edu.tr (Gul Gulpinar a ).
hysteresis and present in all stages of the development of ordered phase is an important approach to probe magnetic systems. They are obtained from the dynamic response of the system to time-dependent magnetic field and now commonly used to investigate the magnetic properties of high-$T_c$ systems [1], cobalt-based alloys [2], nanoparticles [3], spin glasses [4], molecule-based magnets [5], and magnetic fluids [6]. Theoretical investigation of dynamic magnetic response of the Ising models has been an subject of interest for quite a long time: In 1966, Barry has studied spin-1/2 Ising model by a method combining statistical theory of phase transitions and irreversible thermodynamics [7]. Using the same method, Barry and Harrington has focused on the theory of relaxation phenomena in an antiferromagnet [8]. In addition, they obtained the temperature and frequency dependencies of the magnetic absorption and dispersion factor in the neighborhood of the critical temperature. On the other hand, Suzuki and Kubo has obtained the time dependent susceptibility of the kinetic Ising model [9]. Acharyya and Chakrabarti presented the real and imaginary parts of magnetic susceptibility near the order-disorder transition point of a spin-1/2 Ising system in the presence of a periodically varying external field by using Monte-Carlo simulations [10]. On the other hand, The dynamic magnetic response of the materials and the development of methods for its modification are important for their potential applications: 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 [2]; MR effects in nanocomposite particles are used in design of magneto-optical devices [3]; the superposition principle for the imaginary part of complex magnetic susceptibility of composite magnetic fluids is crucial for the design of absorbers and microwave attenuators which are based on ferromagnetic resonance absorption of the electromagnetic field [6].

Recently, Erdem investigated the magnetic relaxation in a spin-1 Ising model near the second-order phase transition point. In this study, time derivatives of the dipolar and quadrupolar order parameters are treated as fluxes conjugate to their appropriate generalized forces in the sense of irreversible thermodynamics [11]. In addition, Erdem has obtained the frequency dependence of the complex susceptibility for the same system [12]. Though the MR has been subject of many theoretical and experimental investigations mentioned above, there has been no study to investigate the behavior of ac susceptibility in the metamagnetic Ising system.

Metamagnets, systems in which antiferromagnetic and ferromagnetic interactions exist simultaneously, are of great interest because it is possible to induce novel kinds of critical behavior by forcing competition between these couplings, in particular by applying a magnetic field [13]. The metamagnetic model that has in-plane ferromagnetic coupling and antiferromagnetic coupling between adjacent planes, and the next nearest neighbor (nnn) spin-1/2 Ising model with antiferromagnetic nearest-neighbour ($nn$) and ferromagnetic
nnn interactions are the theoretical Hamiltonian models showing a similar kind of behavior. These models have been investigated by Monte-Carlo simulations [14], as well as High-temperature series expansion calculations [15].

\[ \text{FeBr}_2 \text{ and FeCl}_2 \text{ are typical Ising type metamagnets [13]. In these structures, in the antiferromagnetic phase when the iron ions in the triangular layers order ferromagnetically, a layer with a negative sign follows a layer with a positive sign. Due to this fact, the external field acts differently on the oppositely oriented layers which leads to different ordered states and associates a sequence of phase transitions as a function of these two interaction strengths.} \]

A Monte-Carlo simulation has been performed on a realistic model of \( \text{FeCl}_2 \) under an external magnetic field [16], in addition this typical metamagnet has also been treated by a high-density expansion method on a two-sublattice collinear Heisenberg Ising model with three- and four-ion anisotropy [17,18]. Dynamic properties of these systems have been investigated by using three-spin flip dynamics [19], Kawasaki dynamics [20], Glauber dynamics [21], and dynamic Monte Carlo renormalization group method [22]. In our recent works we have calculated the kinetic phase diagrams of the system under an oscillating field [23,24]. Moreover, we have presented an investigation of the relaxation dynamics of iron group dihalides and studied the field and temperature dependence of the relaxation times via the phenomenological kinetic coefficients [25]. Recently Gulpinar and Karaaslan performed an investigation of the relaxation times with taking in account the interference between the relaxation processes of antiferromagnetic and ferromagnetic order parameters [26].

The purpose of this paper is to study the dynamical magnetic response properties of the spin-1/2 Ising model in the presence of oscillating external magnetic field. On the other hand, to the best our knowledge there is no study in the literature which represents the mean field calculations for the static staggered and direct susceptibilities of a metamagnetic Ising system. Since then we have given the derivation of the static magnetic response functions and their temperature variations near the critical point in Sec.2. Then, in Sec.3 we derived the temperature and frequency dependent dynamic staggered and direct susceptibilities by a similar method previously used to study relaxation dynamics and sound propagation [27] and investigate the behaviors of magnetic dispersion and absorption factors near the second order phase transition temperature. The method used in this paper provides information about the dynamical critical properties based on the phenomenological kinetic coefficients of the phenomenological rate equations, which governs the magnetization relaxation, is due to an "ad hoc" spin-lattice coupling.
2 Derivation of Static Staggered Magnetic and Magnetic Susceptibilities of Spin-1/2 Metamagnetic Ising Model

In order to obtain static staggered magnetic susceptibility one should introduce a staggered external field \( B_s \) to the system \([28]\) whereas magnetic susceptibility is the response of the total magnetization to a physical external field \( B \). Since then one should add two different magnetic fields to the Hamiltonian of the spin-1/2 Metamagnetic Ising Model:

\[
\hat{H} = -J \sum_{nn} S_i S_j + J' \sum_{nnn} S_i S_j - \mu B \sum_{i=1}^{N} S_i - \mu B_s \left( \sum_{1}^{N} S_i - \sum_{2}^{N} S_j \right).
\] (1)

\( J, J', \mu, B \) and \( B_s \) are ferromagnetic and antiferromagnetic exchange interaction constants, spin magnetic moment, external magnetic field and external staggered field respectively. By making use of mean field approximation free energy of the system can be obtained as following:

\[
G(m_t, m_s, T, B, B_s) = \frac{1}{2} J N (m_t^2 - m_s^2) - \frac{1}{2} J' N (m_s^2 + m_t^2)
\]

\[-\frac{1}{2} k_B T N (4 \ln(2) - (1 + m_s + m_t) \ln(1 + m_s + m_t))
\]

\[-(1 - m_s - m_t) \ln(1 - m_s - m_t) - (1 + m_s + m_t) \ln(1 - m_s + m_t) \ln(1 + m_t - m_s) - (1 - m_t + m_s) \ln(1 - m_t + m_s))
\]

\[-N g \mu_B B m_t - N g \mu_B m_s B_s,
\] (2)

where \( k_B, g, \mu_B, N \) are the Boltzmann’s constant, the spin factor, the Bohr magneton and the total number of metamagnetic Ising spins respectively. For a simple cubic lattice in which intralayer interactions are ferromagnetic and interlayer interactions are antiferromagnetic \( z = 2 \) and \( z' = 4 \). The equilibrium conditions, \( \left( \frac{\partial G}{\partial m_t} = 0, \frac{\partial G}{\partial m_s} = 0 \right) \) result in the following transcendental equations:

\[
m_t = \frac{\sinh(2B_m T)}{\cosh(2B_m T) + \cosh(2B_m T)},
\]

\[
m_s = \frac{\sinh(2B_m T)}{\cosh(2B_m T) + \cosh(2B_m T)},
\] (3)

Here, \( f = J - J' \) and \( t = J + J' \). These equations may be solved without difficulty by an iterative procedure, i.e. Newton Raphson method. The equilibrium solutions should correspond to extremum of the free energy so that one has to determine the solution that minimizes \( G \) \([28]\). Since the solution of these equations are discussed in Ref. \([25]\) extensively, we shall only give a brief summary here as follows: Topology of the metamagnetic Ising model
phase diagram depends on the value of the ratio of the exchange interactions 
\( \eta = \frac{J}{J'} \): (i) If \( \eta > 0.6 \), the transitions between the anti-ferromagnetic and 
paramagnetic phases are of first order at low temperatures and strong fields 
while it is of second order at higher temperatures. The two types of transi-
tions are connected by a tricritical point. (ii) For \( 0 < \eta < 0.6 \), the tricritical 
point decomposes into a critical end point (CEP) and a double critical end 
point (DCP) with a line of first order transitions in between, separating two 
anti-ferromagnetic phases \([28,29]\), see Fig.1(b)-(c) in Ref.[25]. By definition 
staggered magnetic susceptibility is,

\[
\chi_{s,\text{static}} = \lim_{B_s \to 0} \frac{\partial m_s}{\partial B_s}, \quad (B = B_0)
\]

and total (direct) magnetic susceptibility can be expressed as,

\[
\chi_{t,\text{static}} = \lim_{B_s \to 0} \frac{\partial m_t}{\partial B}, \quad (B = B_0).
\]

If one uses the equations of state given in Eq.(3), the static staggered magnetic 
and magnetic susceptibilities can be found as,

\[
\chi_{s,\text{static}} = \lim_{B_s \to 0} \left( \frac{a_{21}c_1 - a_{11}c_2}{a_{22}a_{11} - a_{21}a_{12}} \right),
\]

\[
\chi_{t,\text{static}} = \lim_{B_s \to 0} \left( \frac{b_{22}d_1 - b_{12}d_2}{b_{11}b_{22} - b_{12}b_{21}} \right),
\]

Where, \( a_{11}, a_{12}, a_{21}, a_{22}, c_1, c_2 \), and \( b_{11}, b_{12}, b_{21}, b_{22}, d_1, d_2 \) are given in Appendix-A.

Fig.1(a)-(b) represents the critical behavior of static staggered magnetic 
susceptibility \( (\chi_s) \) and direct magnetic susceptibility. The arrows illustrate 
the phase transition temperature \( T_C(B) \) for \( B = 1.0 \). The staggered magnetic 
susceptibility \( (\chi_s) \) increases rapidly with increasing temperature and diverges 
as the temperature approaches to the second order phase transition point on 
either side, as seen in Fig.1(a). On the other hand, the magnetic suscepti-
bility \( (\chi_t) \) also increases rapidly when the temperature is raised but makes 
finite jump discontinuity at the second order phase transition temperature, 
which is illustrated in Fig.1(b). These findings are in accordance with the re-
results given in Refs.\([30,31]\) obtained within effective field approximation for an 
diluted metamagnetic Ising Model. We should also note that Barry and Harr-
ington have given an detailed analysis for the static total susceptibility of an 
antiferromagnetic Ising system in the Bethe-Takagi approximation which has 
the same result of the constant-coupling approximation of Kasteleyn and Van 
Kranendock applied to Ising antiferromagnetism \([32]\). In both studies an finite 
jump discontinuity is observed at the critical points which is in accordance 
with our results. On the other hand, a infinite-slope singularity is found by
exact series expansions [33]. One might also emphasize that the mean field
approximation only barely fails to exhibit a maximum in the static direct sus-
ceptibility above the $T_N$, that implies a need for a method with takes into spin
correlations [34].

3 Derivation of Non-equilibrium Staggered Magnetic and Mag-
netic Susceptibilities of Spin-1/2 Metamagnetic Ising Model

In order to study the relaxation processes in the anhydrous iron
group dihalides, within the spin-$\frac{1}{2}$ metamagnetic Ising model, one assumes a
small deviation in the value of the applied external magnetic field. This fact
removes the system slightly from its equilibrium state, and one can investigate
how rapidly the metamagnetic system relaxes back to its equilibrium state.
On the other hand, it is well known that the metamagnetic - paramagnetic
phase transition lines occur at places which are away from the $B = 0$ axis
[28]. Consequently, the magnetic Gibbs free energy production ($\Delta G$) due to
the deviations in the applied magnetic fields ($\delta B = B - B_0$, $\delta B_s = B_s - B_{s0}$)
can be expressed as:

$$ G(m_t, m_s, T, B, B_s) = G^0(m_{t0}, m_{s0}, T, B_0, B_{s0}) + \Delta G. $$ (7)

Where $G(m_t, m_s, T, B)$ is the free energy in the neighborhood of equilibrium,
and $G^0$ is the equilibrium Gibbs free energy and $\Delta G$ is the production of the
Gibbs energy due to the variance of the external field, and is given following
form:

$$ \Delta G = a(B - B_0) + b(B_s - B_{s0}) + \frac{1}{2}c(m_t - m_{t0})^2 + \frac{1}{2}d(m_s - m_{s0})^2 $$

$$ + \frac{1}{2}e(B - B_0)^2 + \frac{1}{2}f(B_s - B_{s0})^2 + g(m_t - m_{t0})(m_s - m_{s0}) $$

$$ + h(m_t - m_{t0})(B - B_0) + i(m_t - m_{t0})(B_s - B_{s0}) $$

$$ + j(m_s - m_{s0})(B - B_0) + k(m_s - m_{s0})(B_s - B_{s0}) $$ (8)

The coefficients $a - k$ are given in Appendix-B.

In the sense of Onsager’s theory of irreversible thermodynamics, the time
derivatives of the antiferromagnetic and ferromagnetic order parameters are
treated as generalized fluxes conjugate to their appropriate generalized forces.
One obtains the generalized forces \((X_{mt}, X_{ms})\) conjugate to the currents \((\dot{m}_t, \dot{m}_s)\) respectively, by differentiating \(\Delta G\) with respect to \((m_t - m_{t0}), (m_s - m_{s0})\):

\[
X_{mt} = \frac{\partial \Delta G}{\partial (m_t - m_{t0})} = c(m_t - m_{t0}) + g(m_s - m_{s0}) + h(B - B_0) + i(B_s - B_{s0}),
\]

\[
X_{ms} = \frac{\partial \Delta G}{\partial (m_s - m_{s0})} = d(m_s - m_{s0}) + g(m_t - m_{t0}) + j(H - H_0) + k(B_s - B_{s0}).
\]

The linear relations between the currents and forces may be written in terms of a matrix of phenomenological rate coefficients and since both \(m_t\) and \(m_s\) are odd variables under time inversion this matrix should be symmetric [35]:

\[
\begin{bmatrix}
\dot{m}_t \\
\dot{m}_s
\end{bmatrix} =
\begin{bmatrix}
\gamma_{mt} & \gamma_m \\
\gamma_m & \gamma_{ms}
\end{bmatrix}
\begin{bmatrix}
X_{mt} \\
X_{ms}
\end{bmatrix}.
\]

Consequently, this matrix equation can be written in component form using equations [9], namely a set of two coupled, linear inhomogenous first order rate equations,

\[
\dot{m}_t = (\gamma_{mt}c + \gamma g)(m_t - m_{t0}) + (\gamma_{ms}g + \gamma d)(m_s - m_{s0})
+ (\gamma_{mt}h + \gamma j)(B - B_0) + (\gamma_{ms}i + \gamma k)(B_s - B_{s0}),
\]

\[
\dot{m}_s = (\gamma c + \gamma_{ms}g)(m_t - m_{t0}) + (\gamma g + \gamma_{ms}d)(m_s - m_{s0})
+ (\gamma h + \gamma_{ms}j)(B - B_0) + (\gamma i + \gamma_{ms}k)(B_s - B_{s0}).
\]

In order to find the relaxation times, one considers the corresponding homogeneous equations resulting when there is no external stimulation, namely \(B = B_0, B_s = B_{s0}\). Eqs. [11] then become

\[
\begin{bmatrix}
\dot{m}_t \\
\dot{m}_s
\end{bmatrix} =
\begin{bmatrix}
\gamma_{mt}c + \gamma g & \gamma_{mt}g + \gamma d \\
\gamma_{ms}c + \gamma_{ms}g & \gamma_{ms}g + \gamma_{ms}d
\end{bmatrix}
\begin{bmatrix}
m_t - m_{t0} \\
ms - ms_{s0}
\end{bmatrix},
\]

In iron group dihalides and all metamagnetic spin systems, there exists two order parameters, total and staggered magnetization \((m_t\) and \(m_s)\) which characterize the magnetic behavior of the system. Further, one can see from Eq. [2] that, antiferromagnetic and ferromagnetic order are coupled to each
other. Due to this fact, there is an interference between the ferromagnetic and antiferromagnetic relaxation processes. In the Onsager’s theory of irreversible thermodynamics, the effect of cross effects between these two relaxation processes in the iron group dihalides is embedded in the kinetic rate coefficient $\gamma$. Further, as it is discussed extensively by Barry and Harrington in Ref.\[34\], other operator quantities should be added to the Ising Hamiltonian which do not commute with $\sigma_z$ thereby introducing transitions within the spin system permitting, consequently, longitudinal relaxation. It is well known that, longitudinal relaxation is closely related to spin-lattice relaxation time so that these added operator quantities should contain some kind of spin-lattice coupling, e.g. the frequently designated $T_1$ appearing in the longitudinal Bloch equation representation of spin-lattice relaxation in solids \[34\].

The kinetic equations of motions given by Eqs.(11) can be solved by assuming a form for the solution ($m_t - m_{t0}, m_s - m_{s0} \approx e^{-\frac{t}{\tau}}$) by making use of the secular equation given below Thus, one obtains the following secular equation:

$$\begin{vmatrix}
\frac{1}{\tau} + \gamma_m a - \gamma b & \gamma c - \gamma_m b \\
\gamma a - \gamma_m b & \frac{1}{\tau} + \gamma_m c - \gamma b
\end{vmatrix} = 0,$$

(13)

the resulting relaxation times can be found as:

$$\frac{1}{\tau_1} = -\gamma g - \frac{1}{2}\gamma_m d - \frac{1}{2}\gamma_m c + \frac{1}{2}\Delta^{1/2}$$

(14)

$$\frac{1}{\tau_2} = -\gamma g - \frac{1}{2}\gamma_m d - \frac{1}{2}\gamma_m c - \frac{1}{2}\Delta^{1/2}$$

where $\Delta$ is

$$\Delta = \left(4\gamma_m d\gamma g + 4\gamma g\gamma_m c + \gamma_m^2 d^2 - 2\gamma_m c\gamma_m d + \gamma_m^2 c^2 + 4\gamma_m g^2 \gamma_m + 4\gamma^2 dc\right).$$

(15)

Temperature dependencies of the relaxation times for non zero external magnetic field in the metamagnetic (antiferromagnetic) and paramagnetic phases for the cases ($\gamma_m \gamma_{ms} - \gamma^2 \approx \gamma_m \gamma_{ms}$) and ($\gamma^2 \geq \gamma_m \gamma_{ms}$) were discussed in Refs. \[25,26\].

3.1 Derivation of Kinetic Equations Leading to AC Total Susceptibility

If one stimulates the metamagnetic Ising system by magnetic field $B$ oscillating at an angular frequency $\omega$, all quantities will oscillate near the equilibrium state at this same angular frequency:

$$m_t - m_{t0} = m_{t1} e^{i\omega t}, \quad m_s - m_{s0} = m_{s1} e^{i\omega t}, \quad B - B_0 = B_1 e^{i\omega t}.$$  

(16)
Substituting these equations into the kinetic equations given by Eqs. (11) for $B_s = B_{s0}$ we find following form:

\[
\begin{align*}
(-I\omega + \gamma_{m_c} + \gamma g) m_{t1} + (\gamma_{m_c} + \gamma d) m_{s1} &= -\gamma_{m_c} h B_1, \\
(\gamma c + \gamma_{m_s} g) m_{t1} + (-I\omega + \gamma g + \gamma_{m_s} d) m_{s1} &= -\gamma h B_1.
\end{align*}
\]

(17)

After some algebra, we can find the matrix form of $\frac{m_{t1}}{B_1}$:

\[
\frac{m_{t1}}{B_1} = \begin{vmatrix}
-\gamma_{m_c} h & \gamma_{m_c} + \gamma d \\
-\gamma h & -I\omega + \gamma g + \gamma_{m_s} d \\
-I\omega + \gamma_{m_c} + \gamma g & \gamma_{m_s} + \gamma d \\
\gamma c + \gamma_{m_s} g & -I\omega + \gamma g + \gamma_{m_s} d
\end{vmatrix}.
\]

(18)

The determinant in the denominator of Eq. (20) is the same as the secular determinant used to calculate the reciprocal relaxation times given by Eq. (13) except for the replacement of $\frac{1}{\tau}$ by $i\omega$. Hence Eq. (20) may be written as

\[
\frac{m_{t1}}{B_1} = -h \left( \gamma_{m_c} (-I\omega) + d (\gamma_{m_c} \gamma_{m_s} - \Gamma^2) \right) \frac{1}{(-I\omega - \tau_1^{-1}) (-I\omega - \tau_2^{-1})}.
\]

(19)

Eq. (19) can be used in order to calculate the complex total magnetic susceptibility $\chi_t(\omega)$. This may be seen as follows: The induced magnetization (total induced magnetic moment per unit volume) of the spin-$\frac{1}{2}$ Ising model with $nn$ and $nnn$ interactions is given by

\[
m_t - m_{t\infty} = Re(m_{t1} e^{I\omega t}),
\]

(20)

where $m_{t\infty}$ is the magnetization induced by a magnetic field oscillating at $\omega$. Also, by definition, the expression for $\chi_t(\omega)$ may be written

\[
m_t - m_{t\infty} = Re \left( \chi_t(\omega) B_1 e^{I\omega t} \right),
\]

(21)

where

\[
\chi_t(\omega) = \chi'_t(\omega) - i\chi''_t(\omega)
\]

(22)

here $\chi'_m(\omega)$, $\chi''_m(\omega)$ are the magnetic dispersion and absorption factor, respectively. Comparing Eqs. (21) and (22), one may write

\[
\chi_t(\omega) = \frac{m_{t1}}{B_1},
\]

(23)
and

$$\chi'(\omega) = \mathcal{H} \frac{(\tau_1 \tau_2)}{\tau_2 - \tau_1} \left( \frac{\gamma_m + \tau_1 d(\gamma_m \gamma_m - \Gamma^2)}{1 + \omega^2 \tau_1^2} - \frac{\gamma_m + \tau_2 d(\gamma_m \gamma_m - \Gamma^2)}{1 + \omega^2 \tau_2^2} \right),$$

(24)

$$\chi''(\omega) = \mathcal{H} \frac{(\tau_1 \tau_2)}{\tau_2 - \tau_1} \left( \frac{\omega \tau_1 (\gamma_m + \tau_1 d(\gamma_m \gamma_m - \Gamma^2))}{1 + \omega^2 \tau_1^2} - \frac{\omega \tau_2 (\gamma_m + \tau_2 d(\gamma_m \gamma_m - \Gamma^2))}{1 + \omega^2 \tau_2^2} \right).$$

(25)

### 3.2 Derivation of Kinetic Equations Leading to Dynamical Staggered Magnetic Susceptibility

Now we assume that the system is stimulated by a staggered magnetic field $B_s$ oscillating at an angular frequency $\omega$, all quantities will oscillate near the equilibrium state at this angular frequency:

$$m_t - m_{t0} = m_t e^{i\omega t}, \quad m_s - m_{s0} = m_s e^{i\omega t}, \quad B_s - B_{s0} = B_s e^{i\omega t}, \quad (26)$$

embedding Eqs.(26) into the kinetic equations given by Eqs.(11) for $B = B_0$ one obtains

$$(-I\omega + \gamma_m c + \gamma g) m_{t1} + (\gamma_m g + \gamma d) m_{s1} = -\gamma k B_{s1},$$

$$\gamma c + \gamma_m g) m_{t1} + (-I\omega + \gamma g + \gamma_m d) m_{s1} = -\gamma_m k B_{s1}. \quad (27)$$

solving the Eq(27) yields the following matrix form of $\frac{m_{s1}}{B_{s1}}$

$$\frac{m_{s1}}{B_{s1}} = \begin{vmatrix}
-I\omega + \gamma_m c + \gamma g & -\gamma k \\
\gamma c + \gamma_m g & -\gamma_m k \\
-I\omega + \gamma_m c + \gamma g & \gamma_m g + \gamma d \\
\gamma c + \gamma_m g & -I\omega + \gamma g + \gamma_m d
\end{vmatrix}. \quad (28)$$

Following the same analysis given in Sec.3.1 one obtains

$$\frac{m_{s1}}{B_{s1}} = -\frac{k (\gamma_m (-I\omega) + c(\gamma_m \gamma_m - \Gamma^2))}{(-I\omega - \tau_1^{-1})(-I\omega - \tau_2^{-1})}, \quad (29)$$

10
The induced staggered magnetization due to the effect of the oscillating staggered magnetic field is

\[ m_s - m_{s\infty} = \text{Re}(m_{s1} e^{I\omega t}). \]  

(30)

where \( m_{s\infty} \) is the staggered magnetization induced by a staggered field oscillating at infinite frequency. Also, by definition the expression for the complex staggered susceptibility reads

\[ m_s - m_{s\infty} = \text{Re}(\chi_s(\omega)B_{s1} e^{I\omega t}). \]  

(31)

By making use of Eqs.(28) and (31) one obtains

\[ \chi_s(\omega) = \frac{m_{s1}}{B_{s1}}, \]  

(32)

finally, by using the relation \( \chi_s(\omega) = \chi_s'(\omega) - i\chi_s''(\omega) \) we find

\[ \chi_s'(\omega) = k \frac{(\tau_1 \tau_2)}{\tau_2 - \tau_1} \left( \frac{\gamma_{m1} + \tau_1 c(\gamma_{m1} \gamma_{m2} - \Gamma^2)}{1 + \omega^2 \tau_1^2} \right), \]  

\[ \chi_s''(\omega) = k \frac{(\tau_1 \tau_2)}{\tau_2 - \tau_1} \left( \frac{\omega \tau_1 (\gamma_m + \tau_1 c(\gamma_{m1} \gamma_{m2} - \Gamma^2)}{1 + \omega^2 \tau_1^2} \right), \]  

(33)

(34)

here \( \chi_s'(\omega), \chi_s''(\omega) \) are the magnetic dispersion and absorption factors respectively.

4 Results

Fig.2(a)-(b) shows the critical behavior of staggered magnetic dispersion, \( \chi_s'(\omega) \) and direct magnetic dispersion \( \chi_t'(\omega) \) with varying temperature in the low frequency region \( \omega \tau_2 \ll 1 \). One can observe from Fig.2(a) that real part of the staggered ac susceptibility \( \chi_s'(\omega) \) rises rapidly with increasing temperature and tends to infinity near the second order phase transition. In accordance with the expectations the staggered dispersion factor converges to static staggered susceptibility for \( \omega \tau_2 \ll 1 \) (compare Fig.1(a) and Fig.2(a)). Fig2.(b) represents the temperature variation of the direct magnetic dispersion factor in the neighborhood of \( T_N \) for \( B = 1.0 \) and \( \eta = 2.0 \). The real part of direct complex susceptibility increases slowly with changing temperature and makes a cusp behavior near the second order phase transition. In addition, \( \chi_t'(\omega) \) converges to static direct susceptibility in the low frequency region (compare Fig.2(b) and Fig.1(b)). Moreover the behavior of \( \chi_s'(\omega) \) and \( \chi_t'(\omega) \) are independent of the frequency. This behavior is in accordance with results obtained for spin-1 Ising model [11].
Fig. 3(a)-(b) illustrates temperature variations of staggered magnetic dispersion and direct magnetic dispersion for the high frequency region at several values of $\omega$ in the neighborhood second phase transition point. $\chi_s'(\omega)$ has a local maximum before the second order phase transition point and shows a local minimum near the critical point in the high frequency region. In these figures the number accompanying each curve denotes the value of $\omega$ and the vertical arrows refer to the critical temperature for $B = B_0$ and $\eta = 2.0$. The direct dispersion factor exhibits a maxima for $T \to T_N$ whose amplitude increases with rising frequency. This result is in parallel with findings for the total magnetic dispersion factor for the antiferromagnetic Ising model [34].

In Fig. 4(a)-(b) temperature variations of staggered magnetic absorption and direct magnetic absorption are given for the low frequency region at several values of $\omega$. $\chi''_s$ exhibits a divergence, while $\chi''_t$ makes a local maximum near the critical point. Fig. 5(a)-(b) shows temperature variations of staggered magnetic absorption and direct magnetic absorption for the high frequency region at several values of $\omega$ in the neighborhood second phase transition point. $\chi''_s$ exhibits a local maximum at the phase transition point and amplitude of the maximum changes with frequency (See Fig. 5(a)). On the other hand, $\chi''_t$ makes local minimum at the phase transition point, but it exhibits a local maximum before the critical temperature (See Fig. 5(b)). The divergencies of $\chi'_s(\omega)$ and $\chi''_s(\omega)$ for $\omega \tau_2 \ll 1$ on both sides of the critical point are characterized by the critical exponents. Thus, we may assume that, for temperatures smaller than the transition temperatures, $\chi'_s(\omega)$ and $\chi''_s(\omega)$ follow laws of the form $\chi'_s(\omega) \sim |T - T_N|^{\gamma'_ac}$ and $\chi''_s(\omega) \sim |T - T_N|^{\gamma''_ac}$, respectively. Here $\gamma'_ac$ and $\gamma''_ac$ are the dynamic critical exponents of the staggered magnetic dispersion and absorption factors. In order to calculate the mean field values of $\gamma'_ac$ and $\gamma''_ac$, we have sketched the the plots of $\chi'_s(\omega)$ versus $\log(1 - T/T_N)$ and $\chi''_s(\omega)$ versus $\log(1 - T/T_N)$ which are represented in Fig. 6(a). We have found only one linear part on these $\log - \log$ plots for $\chi'_s(\omega)$ and $\chi''_s(\omega)$, i.e., one values of $\gamma'_ac$ and $\gamma''_ac$, respectively. For each $\omega$ value the slope of the line is equal to $-1$ for $\gamma'_ac$; and $-2$ for $\gamma''_ac$. These values are in accordance with the diverging behavior of the magnetic dispersion and absorption factor near the second order phase transition temperature for $B = B_0$ and $B_s = B_{s0} = 0$. It is important to note that these findings are agreement with the critical exponents found for the magnetic spin-1 Ising model [11]. Further these results are in well agreement with the MR studies of an AB type Ising model for ferromagnetic Ising antiferromagnetism [34] and Ising ferromagnet [36]. In addition, Fig. 6(b) and (c) show logarithmic plot of staggered magnetic dispersion and staggered magnetic absorption versus reduced temperature $(1 - T/T_C)$ for $T < T_C$ at several values in the low frequency region and high frequency region. Finally, Table-1 represents mean field critical exponents for staggered magnetic dispersion, staggered magnetic absorption, direct magnetic dispersion and direct magnetic absorption in the low and high frequency regions.
5 Concluding Remarks

Within the mean field approximation, we have analyzed steady state solutions of the spin-$\frac{1}{2}$ metamagnetic Ising model under a time-dependent oscillating external physical and staggered magnetic fields. In this paper, the formulation is based on a method which combines the equilibrium statistical theory of critical phenomena with the theory of irreversible thermodynamics. It is assumed that the amplitude of both the physical and staggered fields are so small that we made use of linear response theory in studying the magnetic relaxation processes in a metamagnetic system. In this system there exists two coupled relaxation processes which correspond to the relaxation of ferromagnetic and antiferromagnetic order parameters. We have shown that these processes are characterized by two distinct relaxation times ($\tau_1$ and $\tau_2$). $\tau_2$ is the dominant relaxation time which characterizes the critical slowing down of the staggered magnetization and therefore the temperature variation of the second relaxation time determines the separation of the so-called low- and high-frequency regions. We should note that similar behavior has been observed in the investigation of the antiferromagnetic Ising model with the same method [34]. Since $\frac{1}{\tau_2} \to 0$ as $T \to T_N$ and keeping the frequency $\omega$ fixed, we observed the low-frequency behaviors followed by the high-frequency behaviors for the dynamic susceptibilities. One can see from Figs.2(a) and 3(a) that the slope of the staggered magnetic dispersion curve changes in sign as $T \to T_N$ (positive slope for the low-frequency region, negative slope for the high-frequency region). Similar behavior exists also in the high and low frequency regions for total magnetic dispersion factor. Finally, it should be emphasized that we have assumed in this study the rate coefficients have negligible temperature dependence. The validity of this assumption should be testified either by experiments or a more powerful theory such as path probability method. Kikuchi has represented an investigation of Order-disorder configurational relaxation on a bcc AB-type lattice and showed that The diagonal Onsager coefficients tend to finite values whereas the off-diagonal coefficient which characterizes the interference between coupled irreversible processes [37] tends to vanish as temperature approaches critical temperature [38].

6 Acknowledgements

This work was supported by the Scientific and Technological Research Council of Turkey (TUBITAK), Grant No. 109T721. In addition authors thank A.N. Berker for valuable discussions, Sabanci University and Massachusetts Institute of Technology.
The list of coefficients in Eqs. (35):

\[ a_{11} = 1 + \frac{2 \cosh\left(\frac{B-m_f}{k_B T}\right) f}{k_B T \left( \cosh\left(\frac{B-m_f}{k_B T}\right) + \cosh\left(\frac{B_s+m_t}{k_B T}\right) \right)} \frac{2 \sinh\left(\frac{B-m_f}{k_B T}\right)^2}{k_B T \left( \cosh\left(\frac{B-m_f}{k_B T}\right) + \cosh\left(\frac{B_s+m_t}{k_B T}\right) \right)^2}, \]

\[ a_{12} = \frac{2 \sinh\left(\frac{B-m_f}{k_B T}\right) \sinh\left(\frac{2 B_s+m_t}{k_B T}\right) t}{k_B T \left( \cosh\left(\frac{B-m_f}{k_B T}\right) + \cosh\left(\frac{B_s+m_t}{k_B T}\right) \right)^2}, \]

\[ a_{21} = -\frac{2 \sinh\left(\frac{2 B_s+m_t}{k_B T}\right) \sinh\left(\frac{B-m_f}{k_B T}\right) f}{k_B T \left( \cosh\left(\frac{B-m_f}{k_B T}\right) + \cosh\left(\frac{2 B_s+m_t}{k_B T}\right) \right)^2}, \]

\[ a_{22} = 1 - \frac{2 \cosh\left(\frac{2 B_s+m_t}{k_B T}\right) t}{k_B T \left( \cosh\left(\frac{B-m_f}{k_B T}\right) + \cosh\left(\frac{2 B_s+m_t}{k_B T}\right) \right)^2} + \frac{2 \sinh\left(\frac{2 B_s+m_t}{k_B T}\right)^2}{k_B T \left( \cosh\left(\frac{B-m_f}{k_B T}\right) + \cosh\left(\frac{2 B_s+m_t}{k_B T}\right) \right)^2}, \]

\[ c_1 = -\frac{2 \sinh\left(\frac{2 B-m_f}{k_B T}\right) \sinh\left(\frac{2 B_s+m_t}{k_B T}\right)}{k_B T \left( \cosh\left(\frac{B-m_f}{k_B T}\right) + \cosh\left(\frac{2 B_s+m_t}{k_B T}\right) \right)^2}, \]

\[ c_2 = \frac{2 \cosh\left(\frac{2 B_s+m_t}{k_B T}\right)}{k_B T \left( \cosh\left(\frac{B-m_f}{k_B T}\right) + \cosh\left(\frac{2 B_s+m_t}{k_B T}\right) \right)^2} - \frac{2 \sinh\left(\frac{2 B_s+m_t}{k_B T}\right)^2}{k_B T \left( \cosh\left(\frac{B-m_f}{k_B T}\right) + \cosh\left(\frac{2 B_s+m_t}{k_B T}\right) \right)^2}, \]

\[ b_{11} = 1 + \frac{2 \cosh\left(\frac{B-m_f}{k_B T}\right) f}{k_B T \left( \cosh\left(\frac{B-m_f}{k_B T}\right) + \cosh\left(\frac{2 B_s+m_t}{k_B T}\right) \right)} - \frac{2 \sinh\left(\frac{2 B-m_f}{k_B T}\right)^2}{k_B T \left( \cosh\left(\frac{B-m_f}{k_B T}\right) + \cosh\left(\frac{2 B_s+m_t}{k_B T}\right) \right)^2}, \]

\[ b_{12} = \frac{2 \sinh\left(\frac{2 B-m_f}{k_B T}\right) \sinh\left(\frac{2 B_s+m_t}{k_B T}\right) t}{k_B T \left( \cosh\left(\frac{B-m_f}{k_B T}\right) + \cosh\left(\frac{2 B_s+m_t}{k_B T}\right) \right)^2}, \]

\[ b_{21} = -\frac{2 \sinh\left(\frac{2 B_s+m_t}{k_B T}\right) \sinh\left(\frac{B-m_f}{k_B T}\right) f}{k_B T \left( \cosh\left(\frac{B-m_f}{k_B T}\right) + \cosh\left(\frac{2 B_s+m_t}{k_B T}\right) \right)^2}, \]

\[ (36) \]
\[ b_{22} = 1 - \frac{2 \cosh\left(\frac{B_s + m_4 t}{k_B T}\right)t}{k_B T \left(\cosh\left(\frac{B - m_1 f}{k_B T}\right) + \cosh\left(\frac{2 B_s + m_4 t}{k_B T}\right)\right)} + \frac{2 \sinh\left(\frac{2 B_s + m_4 t}{k_B T}\right)^2 t}{k_B T} \]

\[ d_1 = \frac{2 \cosh\left(\frac{2 B - m_1 f}{k_B T}\right)}{k_B T \left(\cosh\left(\frac{B - m_1 f}{k_B T}\right) + \cosh\left(\frac{2 B_s + m_4 t}{k_B T}\right)\right)} - \frac{2 \sinh\left(\frac{2 B - m_1 f}{k_B T}\right)^2}{k_B T \left(\cosh\left(\frac{B - m_1 f}{k_B T}\right) + \cosh\left(\frac{2 B_s + m_4 t}{k_B T}\right)\right)^2} \]

\[ d_2 = -\frac{2 \sinh\left(\frac{B_s + m_4 t}{k_B T}\right) \sinh\left(\frac{2 B - m_1 f}{k_B T}\right)}{\left(\cosh\left(\frac{B - m_1 f}{k_B T}\right) + \cosh\left(\frac{2 B_s + m_4 t}{k_B T}\right)\right)^2 k_B T} \]

(37)
8 Appendix-B

The list of coefficients in Eq.(8):

\[ a = \left( \frac{\partial G}{\partial B} \right)_{eq} = -N\mu m_t \]

\[ b = \left( \frac{\partial G}{\partial B_s} \right)_{eq} = -N\mu m_s \]

\[ c = \left( \frac{\partial^2 G}{\partial m_t^2} \right)_{eq}, \]

\[ = N \left( J - J' - \frac{1}{4} k_B T \left( -\frac{1}{1 + m_s + m_t} - \frac{1}{1 - m_s - m_t} - \frac{1}{1 + m_t - m_s} - \frac{1}{1 - m_t + m_s} \right) \right) \]

\[ d = \left( \frac{\partial^2 G}{\partial m_s^2} \right)_{eq}, \]

\[ = -N \left( J + J' + \frac{1}{4} k_B T \left( -\frac{1}{1 + m_s + m_t} - \frac{1}{1 - m_s - m_t} - \frac{1}{1 + m_t - m_s} - \frac{1}{1 - m_t + m_s} \right) \right) \]

\[ e = \left( \frac{\partial^2 G}{\partial B^2} \right)_{eq} = 0, \]

\[ f = \left( \frac{\partial^2 G}{\partial B_s^2} \right)_{eq} = 0, \]

\[ g = \left( \frac{\partial^2 G}{\partial m_t \partial m_s} \right)_{eq}, \]

\[ = -\frac{1}{4} k_B T N \left( -\frac{1}{1 + m_s + m_t} - \frac{1}{1 - m_s - m_t} + \frac{1}{1 + m_t - m_s} + \frac{1}{1 - m_t + m_s} \right), \]

\[ h = \left( \frac{\partial^2 G}{\partial B \partial m_t} \right)_{eq} = -N\mu, \]

\[ i = \left( \frac{\partial^2 G}{\partial m_t \partial B_s} \right)_{eq} = 0, \]

\[ j = \left( \frac{\partial^2 G}{\partial B \partial m_s} \right)_{eq} = 0. \]
References

[1] P. E. Engelstad and K. Yamada, Phys. Rev. B 52 (1995) 13029.

[2] G. Durin, M. Bonaldi, M. Cerdonio, R. Tommasini, S. Vitale, J. Magn. Magn. Mater. 101 (1991) 89.

[3] M.B.F. van Raap, F.H. Sanchez, C.E.R. Torres, L. Casas, A. Roig, E. Molins, J. Phys.: Condens. Matter 17 (2005) 6519.

[4] J. Ktzler, G. Eiselt, J. Phys. C 12 (1979) L469.

[5] M.A. Girtu, J. Optoelect. Adv. Mater. 4 (2002) 85.

[6] P.C. Fannin, C.N. Marin, I. Malaescu, A.T. Giannitsis, J. Magn. Magn. Mater. 289 (2005) 78.

[7] J.H. Barry, J. Chem. Phys. 45 (1966) 4172.

[8] J.H. Barry, D.A. Harrington, Phys. Rev. B 4 (1971) 3068.

[9] M. Suzuki and R. Kubo, J. Phys. Soc. Jpn. 24 (1968) 51.

[10] M. Acharyya, B.K. Chakrabarti, Phys. Rev. B 52 (1995) 6550.

[11] R.Erdem, J.Magn. Magn. Mater. 320 (2008) 2273.

[12] R.Erdem, J.Magn. Magn. Mater. 321 (2009) 2592.

[13] E.Stryjewski, N. Giordano, Adv. Phys. 26 (1977) 487.

[14] D.P. Landau, Phys. Rev. Lett. 28 (1972) 449; B.L. Arora, D.P. Landau, AIP Conf. Proc. 10 (1973) 870.

[15] F. Harbus, H.E. Stanley, Phys. Rev. B 8 (1973) 1156; F. Harbus, H.E. Stanley, Phys. Rev. B 8 (1973) 1141.

[16] L. Hernandez, H.T. Diep, D. Bertrand, Phys. Rev. B 47 (1993) 2602.

[17] Z. Onyszkiewicz, A. Wierzbicki, Physica B 151 (1988) 462.

[18] Z. Onyszkiewicz, A. Wierzbicki, Physica B 151 (1988) 475.

[19] D. Das, M. Barma, Physica A 270 (1999) 245.

[20] P. S. Sahni, J. D. Gunton, S. L. Katz, R. H. Timpe, Phys. Rev. B 25 (1982) 389.

[21] Z. R. Yang, Phys. Rev. B 46 (1992) 11578.

[22] E.Oguz, J. Phy. A 21 (1988) 2799.

[23] G.Gulpinar, D.Demirhan, F. Buyukkilic, Physica A 383 (2007) 372.

[24] G.Gulpinar, D.Demirhan, F. Buyukkilic, Phys. Lett. A 373 (2009) 511.
[25] G. Gulpinar, D. Demirhan, F. Buyukkilic, Phys. Rev. E 75 (2007) 021104.
[26] G. Gulpinar, Y. Karaaslan, Phys. Lett. A, 375 (2011) 978983.
[27] G. Gulpinar, Phys. Lett. A 372 (2008) 98.
[28] J. M. Kincaid and E. G. D. Cohen, Phys. Reports 22 (1975) 57.
[29] W. Selke, Z. Phys. B 101 (1996) 145.
[30] M. Zukovic, A. Bobak and T. Idogaki, J. Magn. Magn. Mater. 188 (1998) 52.
[31] M. Zukovic, A. Bobak and T. Idogaki, J. Magn. Magn. Mater. 192 (1999) 363.
[32] P.W. Kasteleyn, Physica 22 (1956) 387.
[33] M.E. Fisher and M.F. Sykes, Physica 28 (1962) 919; 28, 939 (1962).
[34] J. H. Barry, D. A. Harrington, Phys. Rev. B 45 (1971) 3068.
[35] S.R. Groot and P. Mazur, Nonequilibrium Thermodynamics, Amsterdam, North Holland Pub. Co. (1961).
[36] J.H. Barry, J. Chem. Phys. 45 (1966) 4172.
[37] L. Onsager, Phys. Rev. 37, (1931) 405; 37 (1931) 2265.
[38] R. Kikuchi, Ann. Phys. 10 (1960) 127; (1963) Hughes Research Rep. No. 271.
Fig. 1. Constant field (B) cross-sections of the staggered magnetic susceptibility (a) and direct susceptibility (b) in the neighborhood of critical point while $B = 1.0$ and $\eta = \frac{zJ}{z'f'}$. 

Fig. 2. Temperature variations of staggered magnetic dispersion (a) and direct magnetic dispersion (b) for the low frequency region at several values of $\omega$ in the neighborhood second phase transition point.
Fig. 3. Temperature variations of staggered magnetic dispersion (a) and direct magnetic dispersion (b) for the high frequency region at several values of $\omega$ in the neighborhood second phase transition point. The number accompanying each curve denotes the frequency.

Fig. 4. Temperature variations of staggered magnetic absorption (a) and direct magnetic absorption (b) for the low frequency region at several values of $\omega$ in the neighborhood second phase transition point.
Fig. 5. Temperature variations of staggered magnetic absorption (a) and direct magnetic absorption (b) for the high frequency region at several values of $\omega$ in the neighborhood second phase transition point. The number accompanying each curve denotes the frequency.
Fig. 6. Logarithmic plot of staggered magnetic dispersion and staggered magnetic absorption vs. reduced temperature \((1 - T/T_C)\) for \(T < T_C\) at several values in the low frequency region (a) and high frequency region (b). Dotted lines are for \(\chi_s'\) and solid lines for \(\chi_s''\).
Table 1
Mean field critical exponents for staggered magnetic dispersion, staggered magnetic absorption, direct magnetic dispersion and direct magnetic absorption in the low and high frequency regions.

| Quantity | Frequency Range         | Exponent Value | Critical Singularity |
|----------|-------------------------|----------------|----------------------|
| $\chi_s'(\omega)$ | $2.10^{-7} - 1.10^{-6}$ (low freq.) | -1             | divergence           |
| $\chi_s''(\omega)$ | $2.10^{-7} - 1.10^{-6}$ (low freq.) | -2             | divergence           |
| $\chi_s'(\omega)$ | $0.2 - 1.0$ (high freq.)   | 0              | cusp                 |
| $\chi_s''(\omega)$ | $0.2 - 1.0$ (high freq.)   | +1             | cusp                 |
| $\chi_t'(\omega)$ | 0.2 (high freq.)        | 0.02           | cusp                 |
| $\chi_t'(\omega)$ | 0.4 - 1.0 (high freq.)   | 0              | cusp                 |
| $\chi_t''(\omega)$ | 0.2 - 1.0 (high freq.)   | 0              | cusp                 |