Matrix mean-field theory for the paramagnetic susceptibility of disordered magnets

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A matrix-based mean-field theory for the paramagnetic susceptibility of disordered Heisenberg and Ising magnets with conventional long-range order is outlined. The wave-vector-dependent static susceptibility is expressed in terms of the eigenvalues and eigenvectors of a symmetrized exchange matrix. The resulting expression is subsequently written as an integral involving a weighted eigenvalue distribution of the matrix. The conditions under which the distribution can be approximated by a single δ function are investigated, and a criterion is established for the validity of the δ-function approximation for the critical temperature. The criterion and the position of the δ function are written in terms of the moments of the distribution which are expressed as products of the elements of the exchange matrix. An alternative approach in which the susceptibility is expressed as the Laplace transform of a linear combination of functions obtained by integrating a set of coupled first-order differential equations is outlined. The theory is applied to a dilute ferromagnet with exponential exchange interactions. The limitations of the theory for strongly disordered systems are discussed, and it is pointed out that when all of the eigenvectors are localized, there is no phase transition in the matrix mean-field approximation. [S0163-1829(96)03421-2]

I. INTRODUCTION

For many years, mean-field theory has been a useful starting point for characterizing the behavior of magnetic materials. In addition, it has served as a point of reference in making comparisons with the predictions of more sophisticated theories for the behavior near magnetic phase transitions. In ferromagnets in which all spin sites are equivalent, the paramagnetic susceptibility in the mean-field approximation is inversely proportional to the difference between the ambient temperature and the critical temperature T_c with the latter being given by a sum involving the exchange integrals coupling the reference spin to its neighbors. In applying the mean-field approximation to systems where the translational symmetry is broken, what is done, typically, is to replace T_c with the configurational average of critical temperature for the corresponding ordered system. Thus, in the case of dilute systems, the ''naive'' mean-field critical temperature is identified with xT_{c0}, where x is the fraction of occupied sites and T_{c0} is the mean-field critical temperature for the fully occupied lattice.

The purpose of this paper is to develop a mean-field theory for the paramagnetic susceptibility of disordered magnets having conventional long-range magnetic order from a more fundamental point of view in which variations in the local fields from site to site are taken into account. In the analysis presented here, which is referred to as *matrix mean-field theory*, the wave-vector-dependent susceptibility is expressed in terms of the eigenvectors and eigenvalues of an exchange matrix.[1] A criterion is established for the validity of the naive mean-field approximation using the moments of a weighted density of eigenvalues of the exchange matrix, which, in turn, are expressed as configurational averages of products of exchange integrals. The theory is applied to a dilute Ising ferromagnet with an exponential exchange interaction. An alternative approach for calculating the susceptibility which replaces the diagonalization of the exchange matrix with the integration of a set of coupled first-order differential equations is outlined. Dynamics and other extensions of the theory as it applies to Ising systems are mentioned. It must be emphasized that the focus in this paper is on the treatment of compositional disorder *within the mean-field approximation*. No attempts are made to compare the results of mean-field treatments of the paramagnetic behavior of disordered magnets with results obtained from other approaches.

It should be noted that matrix mean-field concepts have had some use in the analysis of spin glasses.[2,3] To the best of our knowledge, our work is the first explicit application of the matrix mean-field approach to calculating the wave-vector-dependent paramagnetic susceptibility of disordered magnets with conventional long-range order.

II. CALCULATIONS

The starting point in the calculation is the set of mean-field equations for the thermal averages of the local spins s_i in the presence of a spatially varying field,

$$kTs_i/C_i^2 = g\mu H\,\exp(i\mathbf{q}\cdot\mathbf{r}_i) + \sum_j J_{ij}s_j, \quad i=1,\ldots,N, \tag{2.1}$$

where k is Boltzmann's constant, T is the temperature, g is the g factor, μ is the Bohr magneton, N is the number of spins, and J_{ij} is the exchange integral connecting spins i and j. $C_i=1$ for Ising systems and $C_i=[S_i(S_i+1)/3]^{1/2}$ for Heisenberg magnets ($S_i=1/2,1,\ldots$,etc.). As in the usual mean-field approach, these equations are valid for small fields and temperatures near and above T_c. Introducing the variable $v_i \equiv s_i/C_i$, Eq. (2.1) can be written in the form

$$kTv_i = g\mu HC_i\,\exp(i\mathbf{q}\cdot\mathbf{r}_i) + \sum_j A_{ij}v_j, \tag{2.2}$$

where A_{ij} denotes the symmetrized exchange matrix defined by

$$A_{ij}=C_iC_jJ_{ij}. \tag{2.3}$$

The solution to Eq. (2.2) can be written in terms of the eigenvalues Λ_v and the orthonormal eigenvectors, ϕ_{vi}, of the matrix A_{ij}. Introducing the transformations

$$\Theta_v=\sum_i \phi_{vi}v_i, \tag{2.4}$$

and

$$\begin{aligned} v_i&=\sum_v (\phi)^{-1}_{iv}\Theta_v,\\ &=\Sigma_v\phi^*_{vi}\Theta_v, \end{aligned} \tag{2.5}$$

one obtains the result

$$\Theta_v=g\mu H\sum_i \phi_{vi}C_i \exp(i\mathbf{q}\cdot\mathbf{r}_i)/(kT-\Lambda_v). \tag{2.6}$$

The magnetic moment $M_{\mathbf{q}}$ arising from the field $H\exp(i\mathbf{q}\cdot\mathbf{r}_i)$ is expressed as

$$M_{\mathbf{q}}=g\mu\sum_j s_j \exp(i\mathbf{q}\cdot\mathbf{r}_j). \tag{2.7}$$

Using (2.5) and (2.6) to rewrite Eq. (2.7) we obtain an expression for $M_{\mathbf{q}}$ which, when divided by H, yields a wave-vector-dependent susceptibility $X(\mathbf{q})$ of the form

$$X(\mathbf{q})=g^2\mu^2\sum_v \left|\sum_i \phi_{vi}C_i \exp(i\mathbf{q}\cdot\mathbf{r}_i)\right|^2\Big/(kT-\Lambda_v). \tag{2.8}$$

Equation (2.8) is the principal result of this section. It expresses the susceptibility in terms of the eigenvalues and eigenvectors which can be calculated by direct diagonalization of the matrix A_{ij} for a finite array of spins with, for example, periodic boundary conditions. Prior to analyzing Eq. (2.8) in detail, it is appropriate to verify that $X(\mathbf{q})$ reduces to standard results in various limits. In the high-temperature limit, the eigenvalues are all small in comparison with kT. Because of the orthonormal properties of the ϕ_{vi}, the susceptibility has the Curie form

$$X(\mathbf{q})=(g^2\mu^2/kT)\sum_i C_i^2. \tag{2.9}$$

The other case where a comparison can be made with exact results is a translationally invariant array of equivalent spins. In this situation, the eigenvectors are plane waves, $N^{-1/2}\exp(-i\mathbf{k}\cdot\mathbf{r}_j)$, where $\mathbf{k}$ is a vector in the Brillouin zone of the lattice of spins. The corresponding eigenvalue is $\Lambda_k=\Sigma_jA_{ij}\exp[i\mathbf{k}\cdot(\mathbf{r}_i-\mathbf{r}_j)]$. The resulting expression for the susceptibility takes the standard form

$$X(\mathbf{q})=g^2\mu^2C^2N/(kT-\Lambda_q), \tag{2.10}$$

where $C_i=C_j\equiv C$.

III. NAIVE MEAN-FIELD THEORY AND T_c

In discussing the relation between the results presented above and the ''naive'' mean-field theory mentioned in the Introduction, it is useful to introduce a normalized, weighted eigenvalue distribution $\rho(\mathbf{q},\lambda)$ by means of the equation

$$\rho(\mathbf{q},\lambda)=\sum_v \left|\sum_i \phi_{vi}C_i \exp(i\mathbf{q}\cdot\mathbf{r}_i)\right|^2\delta(\lambda-\Lambda_v)\Big/\sum_i C_i^2. \tag{3.1}$$

The susceptibility can then be written as

$$X(\mathbf{q})=g^2\mu^2\left(\sum_i C_i^2\right)\int_{-\infty}^{+\infty}d\lambda\rho(\mathbf{q},\lambda)/(kT-\lambda). \tag{3.2}$$

In what follows, the moments of $\rho(\mathbf{q},\lambda)$ play an important role. They are defined by

$$\begin{aligned} \langle\lambda^n\rangle_q&=\int_{-\infty}^{+\infty}d\lambda\ \lambda^n\rho(\mathbf{q},\lambda),\\ &=\sum_v \left|\sum_i \phi_{vi}C_i \exp(i\mathbf{q}\cdot\mathbf{r}_i)\right|^2\Lambda_v^n\Big/\sum_i C_i^2. \end{aligned} \tag{3.3}$$

In terms of these, the susceptibility can be written

$$X(\mathbf{q})=(g^2\mu^2/kT)\left(\sum_i C_i^2\right)\left[1+\sum_{n=1}^{\infty}\langle\lambda^n\rangle_q\Big/(kT)^n\right]. \tag{3.4}$$

Naive mean-field theory, as the term is used here, is equivalent to the assumption that for $\mathbf{q}$ in the neighborhood of $\mathbf{q}_0$, the wave vector characterizing the magnetically ordered state (i.e., $\Sigma_j s_j\exp(i\mathbf{q}_0\cdot\mathbf{r}_j)\neq 0$ for $H=0, T<T_c$), $\rho(\mathbf{q},\lambda)$ can be approximated by a single δ function; that is, one has $\rho(\mathbf{q},\lambda)\approx\delta(\lambda-\lambda_c(\mathbf{q}))$. If this is the case, then $X(\mathbf{q})\propto(kT-\lambda_c(\mathbf{q}))^{-1}$, and the critical temperature is identified with $\lambda_c(\mathbf{q}_0)/k$. For the δ-function approximation to be valid it is necessary that $\langle\lambda^n\rangle\approx\langle\lambda\rangle^n$ $(n>1)$, in which case kT_c is identified with the first moment $\langle\lambda\rangle_{q0}$. A simple criterion for the applicability of naive mean-field theory is obtained from the ratio of the square root of the variance to the mean: when $[\langle\lambda^2\rangle-\langle\lambda\rangle^2]^{1/2}/\langle\lambda\rangle<<1$, the width of the peak in the weighted density of states is small in comparison with its position so that a δ-function approximation is appropriate.

Using Eq. (3.3) and noting that Λ_v is an eigenvalue of the matrix $A_{ij}=C_iC_jJ_{ij}$, one can write the first and second moments in the form

$$\langle\lambda\rangle_q=\sum_{i,j} C_i^2C_j^2J_{ij}\exp(i\mathbf{q}\cdot\mathbf{r}_{ij})\Big/\sum_i C_i^2, \tag{3.5}$$

and

$$\langle\lambda^2\rangle_q=\sum_{i,j,k} C_i^2C_j^2C_k^2J_{ik}J_{jk}\exp(i\mathbf{q}\cdot\mathbf{r}_{ij})\Big/\sum_i C_i^2 \tag{3.6}$$

where $\mathbf{r}_{ij}=\mathbf{r}_i-\mathbf{r}_j$. From Eq. (3.5), it is evident that for $\mathbf{q}=\mathbf{q}_0$, $\langle\lambda\rangle_q$ is the configurational average of the mean-field expression for kT_c derived for the corresponding translationally invariant system.

The applicability of the single-δ-function approximation, and thus naive mean-field theory, can be inferred from the variance ratio calculated using Eqs. (3.5) and (3.6). As an example, we consider a lattice of identical spins where a fraction x of the sites are occupied. Assuming ferromagnetic nearest-neighbor interactions J, and z nearest neighbors, one readily obtains the results $\langle\lambda\rangle_0 = zxC^2J$ and $\langle\lambda^2\rangle_0 = C^4J^2[zx+z(z-1)x^2]$ from which one obtains $[\langle\lambda^2\rangle-\langle\lambda\rangle^2]^{1/2}/\langle\lambda\rangle=(1-x)^{1/2}/(zx)^{1/2}$. Thus, it is evident that the naive mean-field theory is appropriate when $x\approx 1$ or $z\gg 1$.

When the single-δ-function approximation is not valid, the question arises as to the ''critical temperature'' in the matrix version of mean-field theory. We define the critical temperature T_c to be the highest temperature at which $X(\mathbf{q}_0)$ diverges in the thermodynamic limit, which, from Eq. (2.8), is equivalent to the largest eigenvalue for which $|\Sigma_i \phi_{iv} C_i \exp(i\mathbf{q}_0\cdot\mathbf{r}_i)|^2/N$ is finite in the limit $N\to\infty$. This definition implies that the eigenstate in question is extended since localized states cannot give rise to a phase transition in the thermodynamic limit.[2,3] However, a more useful approach from the point of view of numerical studies of finite arrays of spins, where the thermodynamic limit is difficult to infer, is to identify the onset of a mean-field ''critical region'' with the upper cutoff of $\rho(\mathbf{q}_0,\lambda)$. For matrix mean-field theory to be consistent, the localized states with eigenvalues lying above the largest extended-state eigenvalue appearing in $X(\mathbf{q}_0)$ must make no contribution to the susceptibility in the thermodynamic limit.

IV. DIFFERENTIAL EQUATION APPROACH

The expression for the susceptibility given in Eq. (2.7) involves the eigenvectors and eigenvalues of the exchange matrix. Even with high-speed work stations, studies are limited to systems with $\approx 10^3$ spins. If one is interested only in $X(\mathbf{q})$, one can bypass the matrix calculation and evaluate the susceptibility directly by integrating a system of coupled first-order linear differential equations associated with the exchange matrix.

Introducing the set of functions $G_i^q(u)$ which obey the equations

$$dG_i^q(u)/du = \sum_j A_{ij} G_j^q(u), \quad u>0, \tag{4.1}$$

with the initial conditions

$$G_i^q(u=0+) = C_i \exp(i\mathbf{q}\cdot\mathbf{r}_i), \tag{4.2}$$

the susceptibility is given by the Laplace transform of a linear combination of the G_i. That is, one has

$$X(\mathbf{q}) = g^2\mu^2 \int_0^\infty du\ e^{-kTu} \sum_j C_j \exp(-i\mathbf{q}\cdot\mathbf{r}_j) G_j^q(u), \tag{4.3}$$

as can be verified by formally integrating Eq. (4.1) in terms of the exponential of the matrix A and subsequently transforming to a basis set involving the eigenvectors of A. In the differential equation approach, calculations can be carried out for systems of 10^4–10^5 spins without undue demands on computer time and memory.

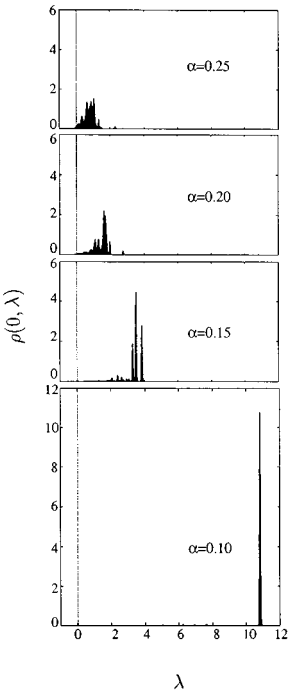

FIG. 1. Weighted density of states, $\rho(0,\lambda)$ vs λ for $\alpha=0.10$, 0.15, 0.20, and 0.25 for the model described in Sec. V. The data are from a single configuration of 1006 spins distributed at random on a fcc array of $4\times136\times136\times136$ sites (corresponding to a concentration $x=10^{-4}$) with periodic boundary conditions. The interaction was taken to be of the form $\exp[\alpha/\sqrt{2}]\exp[-\alpha r]$ with unit lattice constant.

V. MODEL CALCULATION

In this section, we present the results of a numerical study of the mean-field equations for a dilute ferromagnet ($\mathbf{q}_0=0$). The exchange interaction was of the form $\varepsilon(\alpha)\exp(-\alpha r)$, where ε was adjusted so that the interaction between nearest-neighbor spins was equal to one. The calculations were carried out for $N=1006$ spins randomly distributed on a face-centered-cubic lattice supercell with periodic boundary conditions and lattice constant equal to 1 so that $\varepsilon(\alpha)=\exp(\alpha/\sqrt{2})$. The concentration of spins was held fixed at $x=10^{-4}$ corresponding to a fcc supercell of $4\times136\times136\times136$ sites, and α was varied, $0.10\leqslant\alpha\leqslant0.25$. To avoid double counting of sites, the interaction was cut off at one-half of the fcc supercell lattice constant, i.e., at $r=136/2$. Eigenvalues and eigenvectors were calculated by standard methods.

Figure 1 shows the results for the weighted density of states [Eq. (3.1) with $\mathbf{q}=0$] for $\alpha=0.10$, 0.15, 0.20, and 0.25. In calculating the curves, the δ function in Eq. (3.1) has been approximated by a Gaussian, $(0.0025\pi)^{-1/2}\exp[-(\lambda-\Lambda_v)^2/0.0025]$. For $\alpha\leqslant0.10$, the weighted density of states can be approximated by a single δ function; with increasing α, the single-δ-function approximation breaks down, and the distribution becomes multipeaked. The explanation for this behavior can be found in the simple calculation outlined at the end of Sec. III. If one identifies α^{-1} with the range of the interaction, then the number of ''nearest neighbors'' scales as $x\alpha^{-3}$. Thus, for x fixed, increasing α amounts to reducing the number of nearest neighbors leading to an increase in the

ratio of the square root of the variance to the mean value of the distribution.

It is beyond the scope of this study to infer T_c from the eigenvalue criterion mentioned in Sec. III since that would require a detailed numerical study of the behavior of $|\Sigma_i \phi_{iv}|^2/N$ as $N \to \infty$. It can be stated, however, that for large α, matrix mean-field theory fails in its prediction for T_c. Studies of the localization of the eigenstates of a system with an interaction matrix identical to the one used here (apart from an overall sign) have shown that all of the states are localized for $\alpha \geq 0.23-0.25$.[4,5] Because extended states are necessary for phase transitions,[2,3] the conclusion is that there is no phase transition for large α in the matrix mean-field approximation. As a consequence, the theory does not reproduce the scaling behavior found in the extreme low-density limit, $\ln T_c = a - b\alpha(V/N)^{1/3}$, where a and b are material-dependent constants.[6]

VI. DISCUSSION

The purpose of this paper has been the systematic development of a matrix mean-field theory of the paramagnetic susceptibility in disordered magnets with conventional long-range order. The mean-field ''critical region'' was associated with the upper cutoff of $\rho(\mathbf{q}_0,\lambda)$, where $\mathbf{q}_0$ is the wave vector characterizing the magnetically ordered state. The critical temperature was identified with the divergence of the susceptibility $X(\mathbf{q}_0)$ or equivalently, with the largest (extended-state) eigenvalue for which $|\Sigma_i \phi_{iv} C_i \exp(i\mathbf{q}_0 \cdot \mathbf{r}_i)|^2/N$ is finite in the thermodynamic limit. It was pointed out that for the theory to be consistent, any localized states with eigenvalues lying above that eigenvalue must make no contribution to the susceptibility in the thermodynamic limit. The criterion based on the largest eigenvalue of the extended states which have a finite overlap with $\exp(i\mathbf{q}_0 \cdot \mathbf{r}_i)$ is similar to the criterion for the mean-field transition temperature in spin glasses, except that in the latter, it is the largest extended-state eigenvalue without reference to projection.[2,3,7] In situations where $\rho(\mathbf{q}_0,\lambda)$ could be approximated by a single δ function (i.e., naive mean-field theory is appropriate), T_c was given by the first moment of ρ, which is identified with a configurational average of the mean-field critical temperature in the corresponding system in the absence of disorder. A major limitation of the theory comes about because the interaction matrix will have only localized eigenvectors if the disorder is sufficiently strong. When this is the case, there is no phase transition in the matrix mean-field approximation.

Up to this point, the analysis has been of the static susceptibility. In the case of Ising systems, dynamical effects associated with the stochastic flipping of spins can be modeled in a straightforward way. The equation of motion for the spins takes the form

$$kT dv_i/dt = g\mu H \gamma C_i \exp(i\mathbf{q}\cdot\mathbf{r}_i)\exp(i\omega t) - \gamma\left(kTv_i - \sum_j A_{ij} v_j\right), \tag{6.1}$$

where γ is identified with the single-spin flipping rate characterizing the relaxation toward the local field. Assuming a harmonic time dependence, $\exp(i\omega t)$, one obtains a generalization of Eq. (2.6):

$$\Theta_\nu = g\mu H \sum_i \phi_{\nu i} \exp(i\mathbf{q}\cdot\mathbf{r}_i)/(kT + ikT\omega/\gamma - \Lambda_\nu), \tag{6.2}$$

which leads immediately to the complex wave-vector and frequency-dependent susceptibility

$$X(\mathbf{q},\omega) = g^2\mu^2 \sum_\nu \left|\sum_i \phi_{\nu i} \exp(i\mathbf{q}\cdot\mathbf{r}_i)\right|^2/(kT + ikT\omega/\gamma - \Lambda_\nu). \tag{6.3}$$

As a final comment, we note that in the case of Ising systems, one can improve upon the mean-field theory by using the random local-field approximation.[8–10] In this approach the exchange interaction in the matrix A_{ij} is replaced by an effective interaction $A_{\text{eff}\,ij}$. In the case of dilute systems with long-range interactions, the effective interaction matrix is defined by

$$A_{\text{eff}\,ij} = T^2 \int_0^\infty d\rho \exp[-F_1(\rho)]\sin(J_{ij}\rho)/\sinh(\pi\rho T/2), \tag{6.4}$$

where the function F_1 is given as

$$F_1(\rho) = (N/V)\int d\mathbf{r}[1-\cos(J(r)]. \tag{6.5}$$

Here N/V is the spin density and $J(r)$ the exchange interaction between spins separated by a distance r. Note that the effective interaction reduces to J_{ij} in the limit $T \to \infty$.

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