High-temperature series expansion for an easy-axis ferromagnet with exchange and dipolar interactions

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High-temperature series expansions for the magnetic susceptibility and the specific heat have been developed for spin-one ferromagnetic systems with Heisenberg exchange interactions, magnetic dipole-dipole interactions, and single-ion easy-axis anisotropy. The results are valid for arbitrary strength of the single-ion anisotropy and therefore have broader application than Marquard's results. We have obtained the first four terms in each high-temperature series. Effects of the dipolar interactions and the uniaxial anisotropy on ferromagnetic ordering are also discussed.

I. INTRODUCTION

The problem of magnetic dipole-dipole interactions has had a long history. Various aspects have been discussed by many authors. The question of dipolar ferromagnetism at zero temperature has been summarized by Cohen and Keffer.[1] It was concluded that dipolar ferromagnetism cannot occur in the simple cubic lattice but is possible, at least metastable, in the face-centered cubic and the body-centered cubic lattices. In the presence of both exchange and dipole-dipole interactions, Holstein and Primakoff[2] have obtained the simple spin-wave energies at low temperatures and Marquard[3] has given the first four terms in the high-temperature series expansions for the susceptibility and the specific heat. There are also Green's-function works employing the random-phase approximation (RPA) type of decoupling[4] and the high-density diagrammatic expansion.[5] The Green's-function results generally contain the spin-wave findings but can be applied to a wider range of temperatures excluding, however, the neighborhood of the critical point. The calculations and results are also more involved. The hope for extension to higher orders is perhaps very slim. The high-temperature series expansion (HTSE) calculations, on the other hand, have been proved capable of extension to high orders.[6] While giving no information in the ordered regime, HTSE provides the most accurate description of the system in the paramagnetic phase and gives the best estimates of the critical temperature and the exponents characterizing the critical behavior.

Magnetic dipole-dipole interactions exist in all magnetic materials. For systems of high ordering temperature, exchange interactions dominate and the dipolar interaction can usually be ignored. On the other hand, if the ordering temperature is low (order of few Kelvin or lower), the dipolar interactions can contribute significantly to all the thermodynamic quantities measured. $GdCl_3$ is a well-known example.[7] $GdCl_3$ orders ferromagnetically at 2.2 °K. The Gd^{3+} ions are in orbital s states, thus the crystal-field anisotropy is negligible and the Hamiltonian consisting of the Heisenberg exchange interactions and the dipole-dipole interactions only can provide an accurate description of the system. Marquard[3] has obtained the HTSE for the specific heat and susceptibility for this compound and was able to deduce from the experimental data the nearest-neighbor and next-nearest-neighbor exchange interaction parameters. In a similar procedure, employing Marquard's high-temperature series, Wolf and collaborators[8] have been able to obtain the exchange interaction parameters of $Gd(OH)_3$ accurately. $Tb(OH)_3$ has also been treated.[9] While Tb^{3+} is not an s-state ion, the anisotropy of the system is so extreme that it behaves like an Ising system. Again Marquard's high-temperature series was used after an appropriate modification.

There are however numerous compounds in which the dipole-dipole interactions are comparable to the exchange interactions and the single-ion anisotropies lie between the limiting cases mentioned above. Recently, Friedberg and collaborators[10] have studied a series of uniaxial ferromagnetic Ni salts of the general formula $NiMF_6 \cdot 6H_2O$. All the compounds show significant dipolar interactions, and the single-ion anisotropy varies from a value comparable with the exchange interaction to a value much greater than the

latter. To analyze their heat capacity and susceptibility data, Friedberg and collaborators[10] adopted a theoretical model of the Oguchi type. While the fit was satisfactory, the dipolar interactions were left out of the calculations, and consequently the exchange interaction parameters so obtained contain the effects of the dipolar interactions to an unknown extent.

To construct a theory for systems where the dipolar interactions are important and where the single-ion anisotropy is allowed to take an arbitrary value is indeed the motivation of this work. Green's-function theory for magnetic systems with single-ion anisotropy can be formulated,[11] but perhaps will be rather involved in the presence of both dipolar interaction and the single-ion anisotropy (in addition to the exchange interactions). In view of the great success in employing the HTSE to deduce the interaction parameters for systems with vanishing single-ion anisotropy, we decided to obtain a high-temperature series which would permit the single-ion anisotropy to be of arbitrary strength.

The high-temperature series expansion technique for systems with only exchange interactions has been well developed.[6] Marquard[3] has extended the method to include the dipole-dipole interactions. In fact Marquard[3] has also considered the effects of single-ion anisotropy but his calculations are limited to systems with weak anisotropy (compared to the exchange interactions). To allow for arbitrary strength of the single-ion anisotropy, the single-ion potential must be treated exactly. The conventional HTSE technique[6] which would treat the single-ion potential in the expansion thus can not accomplish this goal. Very recently Wang and Lee[12] have shown a method to cope with this problem. They split the Hamiltonian into two parts. The single-ion terms which comprise the crystal-field potential and the mean-field potentials are treated exactly; the many-body diagrammatic expansion procedure is then invoked to account for the fluctuations from the mean field. The first five terms in the HTSE for a spin-one ferromagnet with anisotropic exchange interactions and an easy-axis single-ion anisotropy of arbitrary strength have been computed.[12]

In this paper we extend their calculations for the spin-one ferromagnet with easy-axis anisotropy to include the dipolar interactions. We have found the first four terms in the series expansions for the susceptibility and for the specific heat. Our results reduce to those of Marquard[3] in the limit of vanishing anisotropy for the spin-one case. It has not been possible to obtain the result for general spin. However the Ni-salts studied by Friedberg's group are spin-one systems[10] and in a subsequent paper application of the present results will be made to deduce the interaction parameters for these compounds. In this paper we shall concentrate on developing the theory and at the end give a general discussion of the effects of the dipolar interactions and the single-ion anisotropy.

In Sec. II we introduce the Hamiltonian which consists of the exchange interactions, the dipolar interactions and an easy-axis single-ion anisotropy potential. The HTSE for systems with single-ion anisotropy of arbitrary strength will be recapitulated in Sec. III before we present the HTSE results for the susceptibility and the specific heat of our system. In Sec. IV we discuss some general effects of the dipolar interactions and the anisotropy on the magnetic ordering.

II. HAMILTONIAN

For a spin-one ferromagnet with both exchange and dipolar interactions and an easy-axis anisotropy the Hamiltonian consists of three terms

$$H = H_{ex} + H_{dipolar} + H_{anisotropy} \ , \tag{2.1}$$

where

$$H_{ex} = - \sum_{l,m} J_{lm} \vec{S}_l \cdot \vec{S}_m \ , \tag{2.2}$$

$$H_{dipolar} = \frac{1}{2} \sum_{l,m} d_{lm} \left[\vec{S}_l \cdot \vec{S}_m - \frac{3(\vec{S}_l \cdot \vec{r}_{lm})(\vec{S}_m \cdot \vec{r}_{lm})}{r_{lm}^2} \right] \tag{2.3}$$

and

$$H_{anisotropy} = -D \sum_l (S_l^z)^2 \ . \tag{2.4}$$

In the equations above, $\vec{S}_l$ denotes the spin operator on the l th lattice site, S_l^z being the z component of $\vec{S}_l$. $\vec{r}_{lm}$ is the distance from the l th site to the mth site. The summation $\sum_l$ in Eq. (2.4) is over all sites in the crystal lattice, while the double summation $\sum_{l,m}$ in Eqs. (2.2) and (2.3) is restricted to exclude $l = m$. The parameter J_{lm} characterizes the strength of the Heisenberg exchange interaction between spins on sites l and m. The negative sign in Eq. (2.2) assures that for $J_{lm} > 0$ we have a ferromagnetic exchange interaction. The parameter characterizing the strength of the magnetic dipolar interaction is d_{lm} which is given by

$$d_{lm} = \frac{(g\mu_B)^2}{r_{lm}^3} \ , \tag{2.5}$$

where g is the Landé factor and μ_B the Bohr magneton. Thus the term H_{ex} represents the Heisenberg exchange interaction and the term $H_{dipolar}$ represents the interaction of magnetic dipoles in the lattice.

$H_{anisotropy}$ is a uniaxial single-ion anisotropy term arising from the effects of the crystalline electric field. The parameter D measures the strength of the anisotropy and the negative sign in Eq. (2.4) implies that for $D > 0$, the z axis is an easy axis for the magnetization.

The Hamiltonian of Eq. (2.1) can be separated into

a mean-field (single-ion) part H_0, and a fluctuation (two-body) part H_1. We write H

$$H = H_0 + H_1 + H_{\text{constant}} \quad , \tag{2.6}$$

where H_{constant} is a c-number term. To effect the separation of the Hamiltonian into these parts we replace S^z by $S^z - \langle S^z \rangle + \langle S^z \rangle$, where $\langle S^z \rangle$ is the expectation value of the operator S^z to be determined self-consistently later on. We also use the familiar identity

$$\vec{S}_l \cdot \vec{S}_m = S_l^z S_m^z + \tfrac{1}{2}(S_l^+ S_m^- + S_l^- S_m^+) \quad ,$$

where $S^{\pm} = S^x \pm iS^y$, S^x and S^y being the x and y components of $\vec{S}$, respectively. Thus we get the following:

$$H_0 = -D \sum_l (S_l^z)^2 - 2[J(0) + 2E(0)]\langle S^z \rangle \sum_l S_l^z \quad , \tag{2.7}$$

$$\begin{aligned} H_1 = &- \sum_{l,m} (J_{lm} - E_{lm}) S_l^+ S_m^- \\ &- \sum_{l,m} (J_{lm} + 2E_{lm})(S_l^z - \langle S^z \rangle)(S_m^z - \langle S^z \rangle) \\ &- \sum_{l,m} (F_{lm} S_l^+ S_m^z + \text{H.c.}) \\ &- \sum_{l,m} (B_{lm} S_l^+ S_m^+ + \text{H.c.}) \end{aligned} \tag{2.8}$$

and

$$H_{\text{constant}} = N[J(0) + 2E(0)]\langle S^z \rangle^2 \quad , \tag{2.9}$$

where

$$J(0) = \sum_l J_{lm} \quad , \tag{2.10}$$

$$E(0) = \sum_l E_{lm} \quad , \tag{2.11}$$

$$E_{lm} = -\tfrac{1}{4} d_{lm} \left[1 - 3 \left(\frac{r_{lm}^z}{r_{lm}} \right)^2 \right] \quad , \tag{2.12}$$

$$F_{lm} = \tfrac{3}{2} d_{lm} (r_{lm}^z r_{lm}^- / r_{lm}^2) \quad , \tag{2.13}$$

$$B_{lm} = \tfrac{3}{8} d_{lm} (r_{lm}^- / r_{lm})^2 \quad , \tag{2.14}$$

where r_{lm}^z is the z component of $\vec{r}_{lm}$, $r_{lm}^- = r_{lm}^x - i r_{lm}^y$, r_{lm}^x and r_{lm}^y being the x and y components of $\vec{r}_{lm}$, respectively, N is the total number of lattice sites, and H.c. means the Hermitian conjugate. In the nearest-neighbor interaction model for the Heisenberg exchange, $J(0)$ from Eq. (2.10) is simply Jz where z is the number of nearest neighbors. It is evident from

Eq. (2.8) that the presence of the dipolar interaction introduces new fluctuation terms, e.g., $S^- S^z$, $S^+ S^+$, etc., apart from the usual terms present due to the Heisenberg interaction, namely, $S^+ S^-$, and $(S^z - \langle S^z \rangle)(S^z - \langle S^z \rangle)$.

III. HIGH-TEMPERATURE SERIES EXPANSION (HTSE)

The method of obtaining a HTSE for the free-energy F and the susceptibility (χ) such that the single-ion anisotropy term is taken into account exactly was developed by Wang and Lee in Ref. 12. Here we apply the method to treat our Hamiltonian of Eq. (2.6). The single-ion part of the Hamiltonian, H_0, is given in Eq. (2.7). We add a Zeeman energy term to it to account for an applied external magnetic field along the axis of magnetic ordering. Thus

$$H_0 = -D \sum_l (S_l^z)^2 - h_m \sum_l S_l^z \quad , \tag{3.1}$$

where

$$h_m = 2[J(0) + 2E(0)]\langle S^z \rangle + g\mu_B h_{\text{ext}} \quad , \tag{3.2}$$

h_{ext} being the applied external magnetic field.

The eigenstates of H_0 for spin $S = 1$ are clearly the eigenstates of the operator S^z, namely, $|0\rangle$, $|+1\rangle$, and $|-1\rangle$, where the number n in $|n\rangle$ is the eigenvalue of S^z. Let the eigen-energies of these states be denoted by ϵ_0, ϵ_1, and $\epsilon_{\bar{1}}$, respectively, for $|0\rangle$, $|+1\rangle$, and $|-1\rangle$. We also add a c number D to H_0, to shift all the energies by an amount $+D$. Then

$$\begin{aligned} \epsilon_0 &= D \quad , \\ \epsilon_1 &= -h_m \quad , \\ \epsilon_{\bar{1}} &= +h_m \quad . \end{aligned} \tag{3.3}$$

The mean-field free energy per ion F_0 is thus given by

$$-\beta F_0 = \ln \sum_n \exp(-\beta \epsilon_n) - \beta[J(0) + 2E(0)]\langle S^z \rangle^2 \quad , \tag{3.4}$$

where $\beta = 1/k_B T$ (k_B being the Boltzmann constant and T the absolute temperature), and the sum over n consists of summing over the three states $|0\rangle$, $|+1\rangle$, and $|-1\rangle$.

The corrections to F_0 due to the fluctuation part H_1 can be obtained by using the standard finite-temperature perturbation theory for many-body systems.[13]

$$-\beta \Delta F = \sum_{n=1}^{\infty} \frac{(-1)^n}{n!} \int_0^{\beta} d\tau_1 \cdots \int_0^{\beta} d\tau_n \langle T_\tau H_1(\tau_1) H_1(\tau_2) \cdots H_1(\tau_n) \rangle_c \quad , \tag{3.5}$$

where H_1 has been written in the interaction representation with H_0 as the unperturbed Hamiltonian. The angular brackets denote the canonical thermal average over H_0. The subscript c denotes the cumulant part of the τ-ordered product, or, in the diagrammatic analysis, the contribution of only the connected diagrams. Equations (3.4) and (3.5) consist of a HTSE of the free energy in powers of $\beta[J(0)+2E(0)]$ from which other thermodynamic quantities can be obtained. For example, the zero-field susceptibility χ is the negative of the second derivative of F with respect to the external field, i.e., $\chi = -\partial^2 F/\partial h_{\text{ext}}^2$ and the specific heat $C_h = \beta^2[\partial^2(-\beta F)/\partial \beta^2]$.

In order to compute ΔF of Eq. (3.5) we need to evaluate the thermal averages of the τ-ordered products of spin operators. The diagrammatic method of Vaks, Larkin, and Pikin[14] is not applicable here, because the inclusion of the single-ion anisotropy term in H_0 destroys the simple τ dependence of the spin operators in the interaction representation which uses H_0 as the unperturbed Hamiltonian. However, a more general diagrammatic scheme has been developed by Yang and Wang,[11] which employs the standard basis operators $L_{mn} = |m\rangle \langle n|$. The operator L_{mn} transfers an ion from the state $|n\rangle$ to the state $|m\rangle$ and its simple τ dependence makes the diagrammatic analysis feasible.

The diagrams representing $-\beta\Delta F$ can be constructed with "semi-invariants" $\langle T_\tau S_i^\alpha S_i^\beta \cdots \rangle_c$ at a site and interaction lines connecting appropriate spin operators on different sites. The operators S_i^α, etc., that occur in the semi-invariants and the interaction lines connecting spin operators on different sites result from the terms in H_1, the fluctuation part of the Hamiltonian. These diagrams will be referred to as the main diagrams. For the Hamiltonian under consideration in this paper there are four second-order (in the interaction) and fourteen third-order main diagrams. These are shown in Fig. 1 where the semi-invariants $\langle T_\tau S_i^\alpha S_j^\beta \cdots S_l^\delta \rangle_c$ are represented by ovals, and the interactions are represented by wavy lines. It is worth noting that in the absence of the dipolar interaction only two second-order [(2,1) and (2,3)] and four third-order [(3,1), (3,2), (3,7), and (3,8)] main diagrams remain.

The evaluation of the semi-invariants (SI) is carried out using the standard basis operators, as discussed in Ref. 11. Thus an SI in the spin operators consists of a sum of SI in the standard basis opera-
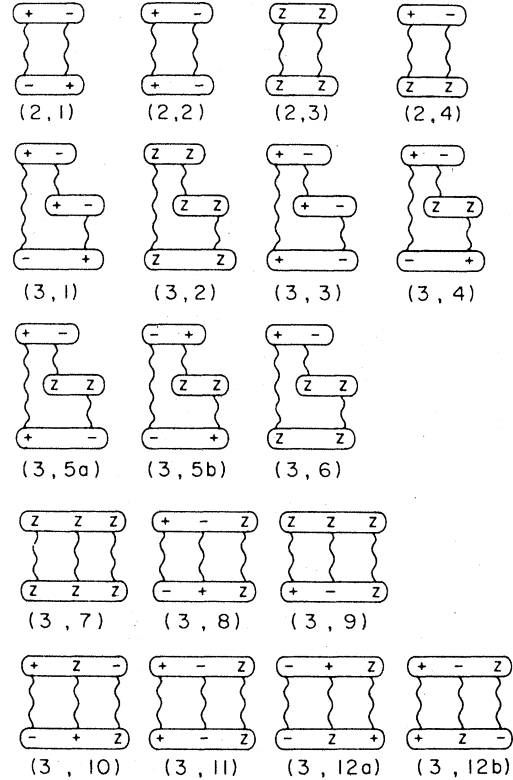

FIG. 1. Second- and third-order main diagrams for the corrections to the free energy due to the fluctuations. The spin operators S^z, S^+, and S^- are denoted by z, $+$, and $-$, respectively. The ovals indicate that the operators enclosed by them are on the same site. The wavy lines connecting operators on different sites are the appropriate interaction lines [See Eq. (2.8).]

tors. For our system we get

$$S^z = \sum_{m,n} \langle m|S^z|n\rangle L_{mn} = L_{11} - L_{\bar{1}\bar{1}} ,$$

$$S^+ = \sum_{m,n} \langle m|S^+|n\rangle L_{mn} = \sqrt{2}(L_{0\bar{1}} + L_{10}) , \qquad (3.6)$$

$$S^- = \sum_{m,n} \langle m|S^-|n\rangle L_{mn} = \sqrt{2}(L_{\bar{1}0} + L_{01}) ,$$

where

$$L_{mn} = |m\rangle \langle n| . \qquad (3.7)$$

As an illustration consider the evaluation of the SI $\langle T_\tau S^+(\tau) S^-(0)\rangle_c$. Substituting from Eq. (3.6) we obtain

$$\langle T_\tau S^+(\tau) S^-(0)\rangle_c = 2\langle T_\tau[L_{0\bar{1}}(\tau) + L_{10}(\tau)][L_{\bar{1}0}(0) + L_{01}(0)]\rangle_c$$

$$= 2[\langle T_\tau L_{0\bar{1}}(\tau) L_{\bar{1}0}(0)\rangle_c + \langle T_\tau L_{0\bar{1}}(\tau) L_{01}(0)\rangle_c + \langle T_\tau L_{10}(\tau) L_{\bar{1}0}(0)\rangle_c + \langle T_\tau L_{10}(\tau) L_{01}(0)\rangle_c]$$

$$= 2[D_{0\bar{1}}G_{\bar{1}0}^0(\tau) + 0 + 0 + D_{10}G_{01}^0(\tau)] , \qquad (3.8)$$

where

$$\langle T_\tau L_{mn}(\tau) L_{nm}(0) \rangle_c = D_{mn} G^0_{nm}(\tau)$$

$$= \begin{cases} D_m e^{\epsilon_{mn}\tau}, & \tau > 0 \\ D_n e^{\epsilon_{mn}\tau}, & \tau < 0 \end{cases} \qquad (3.9)$$

and

$$\epsilon_{mn} = \epsilon_m - \epsilon_n, \quad D_{mn} = D_m - D_n ,$$

$$D_m = e^{-\beta\epsilon_m} / \sum_n e^{-\beta\epsilon_n} .$$

The Green function $G^0_{nm}(\tau)$ can be represented by a line labeled by (nm) propagating from 0 to τ. D_{mn} is a weight factor associated with the SI $\langle T_\tau L_{mn}(\tau) L_{nm}(0) \rangle_c$. Thus the SI $\langle T_\tau S^+(\tau) S^-(0) \rangle_c$ consists of two nonzero SI's in the L operators, each of which is a Green function with a weight factor. Since each SI in spin operators consists of a sum of SI's in the L operators, each main diagram in the spin operators consists of a sum of subdiagrams in the L operators. The evaluation of the diagrams is carried out in Fourier space using the rules of Ref. 11. The spin and space degrees of freedom are decoupled. The spin degrees of freedom result in the Green function and weight factors, while the space degrees of freedom result in interaction lines. To evaluate a diagram in Fourier space one has to sum over internal frequencies and wave-vector variables. The former can be accomplished by using Poisson's summation formula, while the latter depends on the geometry of the lattice and the range of interactions.

A. Magnetic susceptibility

To facilitate the computation of the zero-field susceptibility, we follow the procedure of Rauchwarger

et al.[15] Namely, each of the quantities which enter the diagram such as ϵ_n and the weight factors is first expressed in a power series of h_m. The contribution of each free-energy diagram is then evaluated and also cast in the form of a power series of h_m. The zeroth-order coefficient corresponding to the zero-field free-energy contribution is retained for a later calculation of the specific heat. The first-order coefficient relating to the zero-field magnetization (in the disordered phase) should be identically zero and therefore serves as a check on the computation. The susceptibility is obtained from the second-order coefficient of the free energy (in powers of h_m). Indeed after multiplying by a factor 2β the coefficient gives

$$\chi_c = \beta \frac{\partial^2(-\beta F)}{\partial(\beta h_m)^2}$$

and the susceptibility χ is related to χ_c by

$$\chi^{-1} = \chi_c^{-1} - 2[J(0) + 2E(0)] .$$

We arrive at the following form for χ_c:

$$\chi_c/\chi_0 = 1 + \sum_{n,m} f(m,n)\,\sigma(m,n)\beta^m ,$$

where

$$\chi_0 = \frac{2\beta}{2+t}$$

and

$$t = \exp(-\beta D) .$$

χ_0 is easily recognized as the susceptibility in the absence of the exchange and dipolar interactions. While $f(m,n)$ is the contribution of the spin degrees of freedom from the diagram (m,n), $\sigma(m,n)$ is the contribution of the space degrees of freedom consisting of the lattice sums.

With the aid of a computer we obtain the following:

$$f(2,1) = \frac{1}{(2+t)^2}\left[4t(-2+t) + \frac{8}{\beta D}(1+t)(-1+t)\right] ,$$

$$f(2,2) = \frac{1}{(2+t)^2}\left[\frac{4t}{3}(-10+t) + \frac{8}{\beta D}(1+t+t^2) - \frac{8}{(\beta D)^2}(2+t) + \frac{4}{(\beta D)^3}(2+t)(1+t)(1-t)\right] ,$$

$$f(2,3) = \frac{4(-4+t)}{(2+t)^2} ,$$

$$f(2,4) = \frac{1}{(2+t)^2}\left[\frac{4}{\beta D}(-4+8t-t^2) - \frac{8}{(\beta D)^2}(2+t) + \frac{8}{(\beta D)^3}(1-t)(2+t)\right] ,$$

$$f(3,1) = \frac{1}{(2+t)^3}\left[4t(2+t^2) + \frac{12t}{\beta D}(-6+4t-t^2) + \frac{6}{(\beta D)^2}(1-t)(-4+3t+7t^2)\right] ,$$

$$f(3,2) = \frac{12(-4+t)}{(2+t)^3} ,$$

$$f(3,3) = \frac{1}{(2+t)^3}\left[\frac{4t}{3}(-14-14t+t^2)+\frac{4t}{3\beta D}(-14+64t-5t^2)+\frac{2}{(\beta D)^2}(4-11t-17t^3)\right.$$

$$\left.+\frac{8}{(\beta D)^3}(-4+t)(1-t)(2+t)+\frac{20}{(\beta D)^4}(1-t)^2(1+t)(2+t)\right],$$

$$f(3,4) = \frac{1}{(2+t)^3}\left[\frac{16}{(\beta D)^2}(1-t)(-4+11t-t^2)+\frac{64}{(\beta D)^3}(-1+t)(2+t)+\frac{64}{(\beta D)^4}(1-t)^2(2+t)\right],$$

$$f(3,5) = f(3,4),$$

$$f(3,6) = \frac{1}{(2+t)^3}\left[\frac{8}{\beta D}(-8+13t-2t^2)-\frac{16}{(\beta D)^2}(2+t)+\frac{16}{(\beta D)^3}(1-t)(2+t)\right],$$

$$f(3,7) = \frac{4(4-t)^2}{(2+t)^3},$$

$$f(3,8) = \frac{1}{(2+t)^3}\left[-16t^2+\frac{8}{\beta D}(4+t-2t^2)+\frac{4}{(\beta D)^2}(-1+t)(1+t)(2+t)\right],$$

$$f(3,9) = \frac{1}{(2+t)^3}\left[\frac{24t}{\beta D}(-4+t)+\frac{16}{(\beta D)^2}(4-t)(2+t)+\frac{16}{(\beta D)^3}(1-t)(-4+t)(2+t)\right],$$

$$f(3,10) = \frac{1}{(2+t)^3}\left[\frac{4t}{\beta D}(10-t)(2+t)+\frac{4}{(\beta D)^2}(-8-8t+36t^2+t^3)\right.$$

$$\left.+\frac{8}{(\beta D)^3}(2+t)(-2-10t+3t^2)+\frac{24}{(\beta D)^4}(1-t)(2+t)^2\right],$$

$$f(3,11) = \frac{1}{(2+t)^3}\left[\frac{16t}{3}(4-t)+\frac{8}{\beta D}(-4-t-t^2)+\frac{4}{(\beta D)^2}(3-3t-t^2)(2+t)\right.$$

$$\left.+\frac{4}{(\beta D)^3}(2+3t+4t^2)(2+t)+\frac{6}{(\beta D)^4}(-1+t)(1+t)(2+t)^2\right],$$

$$f(3,12) = \frac{1}{(2+t)^3}\left[\frac{4t}{3\beta D}(-10+t)(2+t)+\frac{4}{(\beta D)^2}(8-24t+24t^2+t^3)\right.$$

$$\left.+\frac{8}{(\beta D)^3}(4-6t+5t^2)(2+t)+\frac{8}{(\beta D)^4}(-1+t)(2+t)^2\right].\tag{3.10}$$

We note that the contributions of diagram $(3,5a)$ and $(3,5b)$ are summed to one term $f(3,5)$ and similarly diagrams $(3,12a)$ and $(3,12b)$ are summed to $f(3,12)$. Similarly the contributions of the space degrees of freedom for the main diagrams are as follows:

$$\sigma(2,1) = \frac{1}{2N}\sum_q[J(q)-E(q)]^2,\quad \sigma(2,2) = \frac{2}{N}\sum_q|B(q)|^2,\quad \sigma(2,3) = \frac{1}{N}\sum_q[J(q)+2E(q)]^2,$$

$$\sigma(2,4) = \frac{1}{N}\sum_q|F(q)|^2,\quad \sigma(3,1) = \frac{1}{3N}\sum_q[J(q)-E(q)]^3,\quad \sigma(3,2) = \frac{4}{3N}\sum_q[J(q)+2E(q)]^3,$$

$$\sigma(3,3) = \frac{4}{N}\sum_q[J(q)-E(q)]|B(q)|^2,\quad \sigma(3,4) = \frac{1}{N}\sum_q[J(q)-E(q)]|F(q)|^2,$$

$$\sigma(3,5) = \frac{1}{N}\sum_q[(F^*(q))^2B(q)+F^2(q)B^*(q)],\quad \sigma(3,6) = \frac{2}{N}\sum_q|F(q)|^2[J(q)+2E(q)],$$

$$\sigma(3,7) = \frac{2}{3N^2} \sum_{q_1} \sum_{q_2} [J(q_1) + 2E(q_1)] \, [J(q_2) + 2E(q_2)] \, [J(q_1 + q_2) + 2E(q_1 + q_2)] \quad,$$

$$\sigma(3,8) = \frac{1}{N^2} \sum_{q_1} \sum_{q_2} [J(q_1) - E(q_1)] \, [J(q_2) - E(q_2)] \, [J(q_1 + q_2) + 2E(q_1 + q_2)] \quad,$$

$$\sigma(3,9) = \frac{2}{N^2} \sum_{q_1} \sum_{q_2} [F(q_1) F^*(q_2)] \, [J(q_1 + q_2) + 2E(q_1 + q_2)] \quad,$$

$$\sigma(3,10) = \frac{1}{N^2} \sum_{q_1} \sum_{q_2} [J(q_1 + q_2) - E(q_1 + q_2)] \, [F(q_1) F^*(q_2)] \quad,$$

$$\sigma(3,11) = \frac{4}{N^2} \sum_{q_1} \sum_{q_2} [B(q_1) B^*(q_2)] \, [J(q_1 + q_2) + 2E(q_1 + q_2)] \quad,$$

$$\sigma(3,12) = \frac{1}{N^2} \sum_{q_1} \sum_{q_2} [\, F(q_1) F(q_2) B^*(q_1 + q_2) + F^*(q_1) F^*(q_2) B(q_1 + q_2)] \quad. \tag{3.11}$$

The quantities $J(q)$, $E(q)$, $B(q)$, and $F(q)$ are the Fourier transforms of the quantities (in real space) J_{lm}, E_{lm}, B_{lm}, and F_{lm}, respectively. For example,

$$J(q) = \sum_l J_{lm} \exp(i \, \vec{q} \cdot \vec{r}_{lm}) \quad. \tag{3.12}$$

The susceptibility χ can now be obtained as follows. Let

$$\chi_c = \chi_0 (1 + C_2 \beta^2 + C_3 \beta^3 + \cdots) \quad. \tag{3.13}$$

C_2 and C_3 are obtained from summing the results of the diagrammatic analysis, i.e.,

$$C_2 = \sum_{m-1}^{4} \sigma(2,m) f(2,m) \quad,$$
$$C_3 = \sum_{m-1}^{12} \sigma(3,m) f(3,m) \quad. \tag{3.14}$$

Thus from Eqs. (3.13) and (3.14) we obtain

$$\chi / \chi_0 = 1 + a_1 x + a_2 x^2 + a_3 x^3 + \cdots$$
$$= 1 + \sum_{n-1} a_n x^n \quad, \tag{3.15}$$

where $x = \beta[J(0) + 2E(0)]$ is the high-temperature

expansion parameter and

$$a_1 = \frac{4}{(2+t)} \quad,$$

$$a_2 = \frac{16}{(2+t)^2} + \frac{C_2}{[J(0) + 2E(0)]^2} \quad,$$

$$a_3 = \frac{64}{(2+t)^3} + \frac{8 C_2}{(2+t)[J(0) + 2E(0)]^2}$$
$$\quad + \frac{C_3}{[J(0) + 2E(0)]^3} \quad. \tag{3.16}$$

Since we have treated the single-ion anisotropy term exactly (by including it in the unperturbed Hamiltonian H_0), the resulting coefficients of the HTSE, a_n are exact functions of βD.

B. Specific heat

The high-temperature series for the specific heat which is exact to all orders of βD can be obtained from the free energy computed above. The zero-field specific heat is the simplest to obtain; one evaluates the free-energy diagram (which actually represents $-\beta F$) at $h_m = 0$ obtaining a function of βD, then takes the second-order derivative of the function with respect to βD, and finally multiplies the result by $(\beta D)^2$. Similar to the susceptibility series, the specific-heat series takes the form

$$C_{h-0} = C_0 + k_B \sum_{m,n} g(m,n) \sigma(m,n) \beta^m \quad, \tag{3.17}$$

where

$$C_0 = 2(2+t)^{-2} t (\beta D)^2 k_B \quad,$$

$$g(2,1) = 8(2+t)^{-4} [t(4 - 8t + t^2)(\beta D)^2 + 6t(-3 - t + t^2)(\beta D) + 6t(1+t)(2+t)] \quad,$$

$$g(2,2) = g(2,1) \quad,$$

$$g(2,3) = 8(2+t)^{-4} [2t(-1+t)(\beta D)^2 + 4t(2+t)(\beta D) + (2+t)^2] \quad,$$

$g(2,4) = 8(2+t)^{-4}[t(-8+12t-t^2)(\beta D) +2t(8+2t-t^2)]$,

$g(3,1) = 4(2+t)^{-5}[2t(4+2t-10t^2+t^3)(\beta D)^2 +6t(-4-42t+18t^2+t^3)(\beta D)$

$\qquad\qquad +3t(-26+119t+98t^2-11t^3) +54t(1-t)(1+t)(2+t)(1/\beta D)]$,

$g(3,2) = 24(2+t)^{-5}[t(-2+3t)(\beta D)^2 +6t(2+t)(\beta D) +2(2+t)^2]$,

$g(3,3) = g(3,1)$,

$g(3,4) = 32(2+t)^{-5}[t(-14+53t-22t^2+t^3) +2t(1-t)(14+5t-t^2)(1/\beta D)]$,

$g(3,5) = g(3,4)$,

$g(3,6) = 16(2+t)^{-5}[t(-10+23t-4t^2)(\beta D) +4t(10+t-2t^2) +2(1-t)(2+t)^2(1/\beta D)]$,

$g(3,7) = 0$,

$g(3,8) = 8(2+t)^{-4}[2t(1-t)(\beta D) +t(-9-7t+t^2) +2(-1+t)(1+t)(2+t)(1/\beta D)]$,

$g(3,9) = 0$,

$g(3,10) = 8(2+t)^{-4}[t(-4+8t-t^2)(\beta D) +t(8+12t-5t^2) +4t(1-t)(2+t)(1/\beta D)]$,

$g(3,11) = -g(3,8)$,

$g(3,12) = -g(3,10)$.

$$(3.18)$$

C_0 is obviously the mean-field result, and the other terms represent the correlations of the fluctuations which are ignored in the mean-field approximation. The lattice sums $\sigma(m,n)$ are the same as the ones listed for the susceptibility calculations.

IV. RESULTS FOR CUBIC LATTICES

As we emphasized in the beginning, the main purpose of this paper is to obtain the high-temperature series expansions applicable to systems in which the exchange interactions, the dipolar interactions, and the single-ion anisotropy can take arbitrary values. The application of the results to several Ni salts will be given in a subsequent paper. It is, however, instructive to demonstrate the application of the series to systems with lattices of simpler geometry, and consider some of the general effects of dipolar interactions and single-ion anisotropy on ferromagnetic ordering.

We focus on the three cubic lattices in this section. For simplicity, we also assume that the exchange interactions extend to the nearest-neighbor ions only. Thus $J(0) = Jz$ where z is the number of nearest-neighbor ions to a specific ion, and is 6, 8, 12 for the sc, bcc, and fcc lattices, respectively. The effective field due to the dipolar interaction $E(0)$ can be written as usual

$$E(0) = E'(0) + L , \qquad (4.1)$$

where $E'(0)$ is the contribution of the dipoles inside the Lorentz sphere. L comprises two terms, one from the inner surface of the Lorentz sphere yielding the well-known Lorentz factor and the other from the outer surface of the sample giving the demagnetizing field. Specifically

$$E'(0) = -\tfrac{1}{4}(g\mu_B)^2 \sum_l \{[r_{lm}^2 - 3(r_{lm}^z)^2]/r_{lm}^5\} \qquad (4.2)$$

and

$$L = \begin{cases} \left(\dfrac{\pi}{3} - \dfrac{N_D}{4}\right)(g\mu_B)^2/a^3, & \text{for sc lattice} \\[2ex] 2\left(\dfrac{\pi}{3} - \dfrac{N_D}{4}\right)(g\mu_B)^2/a^3, & \text{for bcc lattice} \\[2ex] 4\left(\dfrac{\pi}{3} - \dfrac{N_D}{4}\right)(g\mu_B)^2/a^3, & \text{for fcc lattice} . \end{cases} \qquad (4.3)$$

Here N_D is the sample shape-dependent demagnetizing factor which varies from $\tfrac{1}{3}(4\pi)$ for a spherical sample to zero for a needle-shaped sample. It can also be shown from symmetry that $E'(0) = 0$ for all cubic lattices. Now we define a dimensionless parameter

$$W = \frac{(g\mu_B)^2/a^3}{J} , \qquad (4.4)$$

where a is the lattice constant. W measures the

strength of the dipolar interaction relative to the exchange interaction. Then we can write

$$E(0) = \begin{cases} \dfrac{\pi}{3}JW, & \text{for sc lattice} \\[2ex] \dfrac{2\pi}{3}JW, & \text{for bcc lattice} \\[2ex] \dfrac{4\pi}{3}JW, & \text{for fcc lattice} \end{cases} \quad (4.5)$$

for a long thin cylinder or needle-shaped sample. The quantity $J(0) + 2E(0)$ is then

$$J(0) + 2E(0)$$

$$= \begin{cases} J\left[6 + \left(\dfrac{2\pi}{3}\right)W\right], & \text{for sc lattice} \\[2ex] J\left[8 + \left(\dfrac{4\pi}{3}\right)W\right], & \text{for bcc lattice} \\[2ex] J\left[12 + \left(\dfrac{8\pi}{3}\right)W\right], & \text{for fcc lattice} . \end{cases} \quad (4.6)$$

The lattice sums $\sigma(m,n)$ are evaluated numerically on a computer by summing over the lattice points in a given lattice. We list the results for the three cubic lattices in the Appendix.

With the above information in hand, the high-temperature series for the susceptibility and the specific heat are ready to be used. One of the important applications of the susceptibility series is to obtain an estimate of the critical temperature for the phase transition. Usually with five or more terms in the series the accuracy can be within a few percent of the exact value. Our series is rather short due to the complexity of the system. It is found, however, that at vanishing dipolar interaction,[12] the estimated values for the critical temperatures, using the first four terms in the series, all lie within 3% of the estimated values using the first five terms. The accuracy is best for the fcc lattice, as expected (within 1% of the values using five terms). We therefore tentatively assumed the validity of the estimate using the four-term series in the presence of the dipolar interaction. Quickly, however, we found that as the strength of the dipolar interaction relative to the exchange interaction, W, gets large ($W \gtrsim 20$), the results become questionable. This was apparent in the plot of the ratio a_n/a_{n-1} for the susceptibility series versus $1/n$. Instead of falling near a straight line the points become quite irregular for large W. Excluding situations where dipolar interactions dominate, we trust that the estimates from our series are generally acceptable.

We have chosen the following procedure to obtain the estimated value of the critical temperature T_c.

We first define $T_c^{(3)}$ as

$$\frac{k_B T_c^{(3)}}{[J(0) + 2E(0)]} = \frac{a_3}{a_2} . \quad (4.7)$$

Now[6]

$$\frac{a_n}{a_{n-1}} = \left[\frac{\gamma + n - 1}{n}\right]\frac{k_B T_c}{J(0) + 2E(0)} , \quad (4.8)$$

where γ is the critical exponent for the susceptibility. Our estimated T_c is therefore given by

$$\frac{k_B T_c}{J(0) + 2E(0)} = \frac{3}{\gamma + 2}\frac{k_B T_c^{(3)}}{J(0) + 2E(0)} . \quad (4.9)$$

For a fixed value of $D/[J(0) + 2E(0)]$, we solve Eq. (4.7) iteratively to obtain $T_c^{(3)}/[J(0) + 2E(0)]$. Then Eq. (4.9) gives the estimated value for T_c. We have taken $\gamma = 1.25$ in view of the fact that the system behaves Ising-like in the presence of uniaxial anisotropy.

The variations of $k_B T_c/[[J(0) + 2E(0)]$ for the three cubic lattices are plotted in Figs. 2–4, for various values of W. The mean-field theory predictions are also shown for comparison. We first note that including the correlations between fluctuations has greatly depressed the values of T_c from the mean-field values. The effect becomes even more important in the presence of dipolar interactions. This is contrary to the intuitive feeling that, because of the long-range nature of the dipolar interaction, it can be better approximated by a mean-field potential than can the exchange interactions which extend only to near-neighbor ions. In fact in the presence of dipolar interactions only, fluctuations can actually prevent ferromagnetic ordering from occurring even at zero temperature. The mean-field approximation, however, yields a nonzero ordering temperature. This is the situation with a simple cubic lattice as was also
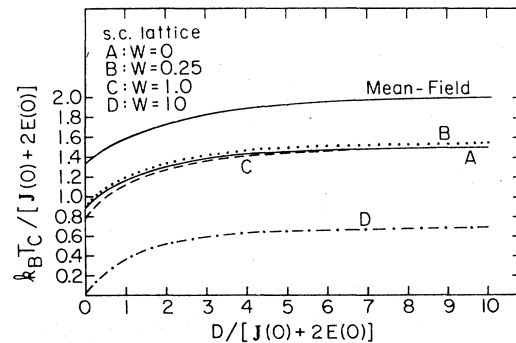

FIG. 2. Plot of $k_B T_c/[J(0) + 2E(0)]$ vs $D/[J(0) + 2E(0)]$ for some representative values of the parameter W, for a simple cubic lattice. The mean-field-theory result is shown for comparison with the HTSE results. These results are for a long needle-shaped sample.
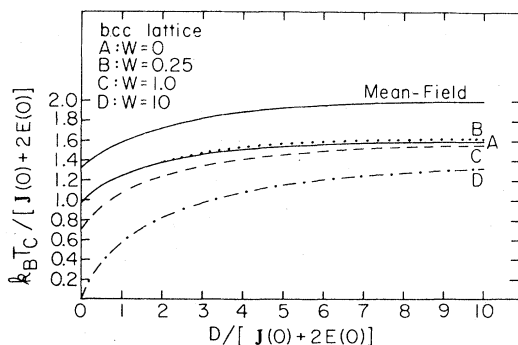
FIG. 3. Same as Fig. 2, except for a body-centered cubic lattice.

concluded by Cohen and Keffer.[1] Our results are consistent with their finding. As the relative strength of the dipolar interaction relative to the exchange interaction W increases, T_c is further depressed below the mean-field value. In fact T_c is driven to zero before one has pure dipolar interaction. This is a case not treated by Cohen and Keffer[1] but can be confirmed by extending their treatment to include the exchange interaction. Our results for the bcc and fcc also show no ferromagnetic ordering when dipolar interactions dominate, at least in the absence of the single-ion anisotropy $(D=0)$. This is in contradiction to the conclusion of Cohen and Keffer.[1] It is interesting to note that for a hexagonal lattice system without single-ion anisotropy as treated by Marquard,[3] the four term HTSE also gives no ferromagnetic ordering for pure dipolar interactions. But, as we pointed out earlier, for large $W(W \gtrsim 20)$ the estimate of T_c made from the short series is rather questionable.
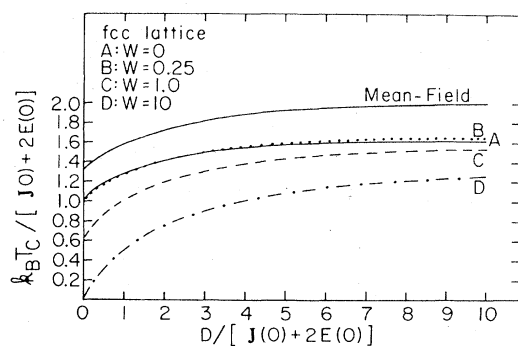

FIG. 4. Same as Fig. 2, except for a face-centered cubic lattice.

We should also note that for small enough amounts of dipolar interaction added to a system with predominant exchange interactions, $T_c/[J(0)+2E(0)]$ actually becomes larger than the value with pure exchange interaction. This is a general phenomenon and can be understood by noting that for small W, the effective field due to the dipolar interaction is linear in W, while the fluctuation effects which tend to suppress the ordering are of second order or higher.

Finally we note the effects of the easy-axis anisotropy. As expected, the anisotropy supports ordering and raises the ordering temperature. This is because increasing D raises the energy of the nonmagnetic $S^z=0$ state thus reducing its population. For large values of D the system approaches the Ising limit. More interesting, however, is the fact that for systems where no ferromagnetic ordering is allowed at $D=0$ because of dominant dipolar interactions, increasing the anisotropy can restore the ordering, obviously through suppressing transverse-type fluctuations. It is found that this is always possible for the bcc and fcc lattices but no so for the sc lattice. That is, if we examine the limiting case of $D \rightarrow \infty$, our estimated T_c for the fcc and bcc lattices are always finite and quite high even when the dipolar interactions dominate. On the other hand, for the sc lattice T_c is lowered to zero for large enough value of W even in this extreme anisotropic limit. Again we repeat that the conclusion should be received with reservation because the series is probably too short for treating cases with dominant dipolar interactions.

In conclusion, we have obtained the high-temperature series expansions for the susceptibility and the specific heat of systems in which the exchange interactions, the dipolar interactions and the easy-axis anisotropy can take arbitrary values. This will provide a theory for analyzing data on the Ni salts studied recently by Friedberg and collaborators,[10] for example. We have also given a general discussion on the effects of dipolar interactions and easy-axis anisotropy in a ferromagnet. While the reliability of the critical temperatures estimated for systems where the dipolar interactions dominate is still questionable, the general features, such as lower T_c for systems with higher fraction of dipolar interactions and the capability of stabilizing the magnetic ordering by an easy-axis anisotropy, should remain valid.

ACKNOWLEDGMENT

APPENDIX

Here we list the lattice sums $\sigma(m,n)$ for the three cubic lattices, namely, simple cubic, body-centered cubic, and face-centered cubic. They were obtained by carrying out the sums of Eq. (3.11) in real space using the computer.

Lattice sum	Simple cubic lattice	Body-centered cubic lattice	Face-centered cubic lattice
$\sigma(2,1)/J^2$	$3+0.4174\,W^2$	$4+0.4664\,W^2$	$6+2.1539\,W^2$
$\sigma(2,2)/J^2$	$1.4676\,W^2$	$4.0961\,W^2$	$16.6037\,W^2$
$\sigma(2,3)/J^2$	$6+3.3389\,W^2$	$8+3.7312\,W^2$	$12+17.2314\,W^2$
$\sigma(2,4)/J^2$	$0.8615\,W^2$	$10.7871\,W^2$	$40.5648\,W^2$
$\sigma(3,1)/J^3$	$1.4591\,W^2+0.1232\,W^3$	$5.9184\,W^2+0.1783\,W^3$	$16+31.532\,W^2+2.0576\,W^3$
$\sigma(3,2)/J^3$	$23.345\,W^2-3.9417\,W^3$	$94.6944\,W^2-5.7062\,W^3$	$64+504.512\,W^2-65.8432\,W^3$
$\sigma(3,3)/J^3$	$14.855\,W^2-1.3555\,W^3$	$19.125\,W^2-6.3079\,W^3$	$144.6872\,W^2-31.5864\,W^3$
$\sigma(3,4)/J^3$	$4.8431\,W^2+0.2469\,W^3$	$80.352\,W^2+4.7030\,W^3$	$470.7504\,W^2+13.0752\,W^3$
$\sigma(3,5)/J^3$	$-1.2498\,W^3$	$-21.6864\,W^3$	$-162.5184\,W^3$
$\sigma(3,6)/J^3$	$9.6862\,W^2-0.9876\,W^3$	$160.704\,W^2-18.8122\,W^3$	$941.5008\,W^2-52.3008\,W^3$
$\sigma(3,7)/J^3$	$4+6\,W^2+0.992\,W^3$	$\frac{16}{3}+1.0086\,W^3$	$8+24\,W^2-4.5406\,W^3$
$\sigma(3,8)/J^3$	$6-2.25\,W^2+0.372\,W^3$	$8+0.3783\,W^3$	$12-9\,W^2-1.7027\,W^3$
$\sigma(3,9)/J^3$	$0.1025\,W^3$	$18.963\,W^2+0.1244\,W^3$	$72\,W^2+51.3101\,W^3$
$\sigma(3,10)/J^3$	$-0.0256\,W^3$	$9.4815\,W^2-0.0311\,W^3$	$36\,W^2-12.8275\,W^3$
$\sigma(3,11)/J^3$	$2.25\,W^2-1.1672\,W^3$	$\frac{128}{27}\,W^2-1.1966\,W^3$	$27\,W^2-20.5459\,W^3$
$\sigma(3,12)/J^3$	$0.0923\,W^3$	$7.4218\,W^3$	$39.001\,W^3$

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