Method of Calculating High-Temperature Series Expansions Exact in the External Magnetic Field: Application to the Two-Spin Correlation Function and Magnetization of the Classical Heisenberg Magnet*

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A practical method of calculating high-temperature series expansions that are exact in the external magnetic field is presented. This method is illustrated by calculating the magnetization $\langle S^z \rangle$ and nearest-neighbor pair-correlation function $\langle \vec{S}_i \cdot \vec{S}_{i+1} \rangle$ for a classical Heisenberg ferromagnet on a fcc lattice. It is found that at $T \simeq 4T_C$, four terms in the expansion suffice to give information which is of value in interpreting data on EuO presented in the preceding paper. For $T \gg T_C$ and at low magnetic fields (i.e., $\langle S^z \rangle \ll S$), we find $\langle \vec{S}_i \cdot \vec{S}_{i+1} \rangle_H - \langle \vec{S}_i \cdot \vec{S}_{i+1} \rangle_0 = A \langle S^z \rangle^2$, where A is a coefficient of order unity which depends on T/T_C. The dependence of A on T/T_C is evaluated from the high-temperature series expansion and also by an alternative method.

I. INTRODUCTION

When a magnetic field H is applied to a Heisenberg ferromagnet at nonzero temperatures, both the magnetization $M(H, T)$ and the nearest-neighbor two-spin correlation function $\Gamma(H, T) \equiv \langle \vec{S}_i \cdot \vec{S}_{i+1} \rangle$ are expected to increase.[1] In this work we calculate the H dependence of $\Gamma(H, T)$ at temperatures well above the critical temperature, and we relate the H-induced increase in $\Gamma(H, T)$ to the increase in the magnetization $M(H, T)$. This work was motivated principally by the need to analyze electrical transport data in EuO,[2] for which a knowledge of the H dependence of $\Gamma(H, T)$ at $T \cong 4T_C$ was necessary.

In the paramagnetic limit of noninteracting spins $\langle \vec{S}_i \cdot \vec{S}_{i+1} \rangle = \langle S_i^z \rangle \langle S_{i+1}^z \rangle$, so that

$$\eta \equiv \Gamma(H, T)/\Gamma(\infty, T) \tag{1.1}$$

reduces simply to σ^2, where

$$\sigma \equiv M(H, T)/M(\infty, T) \tag{1.2}$$

is the reduced magnetization.

In the mean-field approximation to the Heisenberg model, the spins are treated as statistically independent so that $\eta = \sigma^2$ at *all* temperatures and fields.[3] On the other hand, in real systems, when $T \geqslant T_C$ and $H = 0$, $\sigma = 0$ but $\eta \neq 0$. The aim of the present work is to calculate the H dependence of η and σ at $T \cong 4T_C$ and to relate $\eta(H) - \eta(0)$ to $\sigma(H)$. The calculations of $\eta(H) - \eta(0)$ and σ were carried out only to an accuracy of several percent, but this accuracy was found to be sufficient for our purpose of analyzing the electrical transport data in EuO.[2]

The dependence of η upon both H and T has been calculated previously by Callen and Callen[4] using the two-spin Oguchi cluster approximation. However, this approximation has historically been less reliable than the method of series expansions in the variable J/kT, where J is the exchange parameter and k is the Boltzmann constant.[5] Accordingly, we have applied the latter approach. Previous series work has been limited either to the case of $H = 0$ or has been restricted to *double series* in J/kT and h,[5,6] where h is the magnetic field in units of energy. In this work we present what we believe to be the first high-temperature series expansions in J/kT that are exact in magnetic field. Our method requires considerable labor if higher orders in the expansion in (J/kT) are needed. However, in some cases meaningful results can be obtained by retaining only several terms in the expansion. In particular, the inclusion of only four terms in the expansion gives satisfactory results for the purpose of interpreting the data in Ref. 2.

In Sec. II we provide the general formalism for obtaining a series expansion which is exact in the magnetic field. The formalism described is actually more general than for the Heisenberg model, and is applicable for any Hamiltonian composed of two terms that commute[7] (e.g., exchange term and Zeeman term).

In Sec. III the calculation is carried out in detail for the classical approximation to the Heisenberg model, and in Sec. IV an alternate treatment for low magnetic fields is given.

II. GENERAL FORMALISM

A. Generalized Expansion

Consider a system governed by the Hamiltonian $\mathcal{H}$ of the form

$$\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2 , \tag{2.1}$$

where $\mathcal{H}_2$ is to be treated as a perturbation and[7]

$$[\mathcal{H}_1, \mathcal{H}_2] = 0. \tag{2.2}$$

The quantum-mechanical statistical average of an operator $\mathcal{A}$ is given by

$$\langle \mathcal{A} \rangle = \frac{\mathrm{Tr}\{\mathcal{A} e^{-\beta \mathcal{H}}\}}{\mathrm{Tr}\{e^{-\beta \mathcal{H}}\}}, \quad \beta = \frac{1}{kT} . \tag{2.3}$$

Let us now assume that $\langle \mathcal{A} \rangle$ can be expanded in the following way:

$$\langle \mathcal{A} \rangle = \sum_{m=0}^{\infty} \alpha_m(\mathcal{H}_2, \beta\mathcal{H}_1) \frac{(-1)^m}{m!} \beta^m, \tag{2.4}$$

where the $\alpha_m(\mathcal{H}_2, \beta\mathcal{H}_1)$, to be determined, satisfy the homogeneity relation

$$\alpha_m(\lambda\mathcal{H}_2, \beta\mathcal{H}_1) = \lambda^m \alpha_m(\mathcal{H}_2, \beta\mathcal{H}_1) . \tag{2.5}$$

Since, from (2.1) and (2.2),

$$e^{-\beta\mathcal{H}} = e^{-\beta\mathcal{H}_2} e^{-\beta\mathcal{H}_1}, \tag{2.6}$$

it follows that

$$\langle \mathcal{A} \rangle = \frac{\sum_{k=0}^{\infty}[(-1)^k/k!]\,\mathrm{Tr}\{\mathcal{A}\mathcal{H}_2^k e^{-\beta\mathcal{H}_1}\}\beta^k}{\sum_{l=0}^{\infty}[(-1)^l/l!]\,\mathrm{Tr}\{\mathcal{H}_2^l e^{-\beta\mathcal{H}_1}\}\beta^l} . \tag{2.7}$$

Equating the right-hand sides of (2.4) and (2.7), and multiplying through by the denominator of (2.7), we obtain

$$\sum_{k=0}^{\infty} \frac{(-1)^k}{k!} \beta^k \mathrm{Tr}\{\mathcal{A}\mathcal{H}_2^k e^{-\beta\mathcal{H}_1}\} = \sum_{l=0}^{\infty}\sum_{m=0}^{\infty} \frac{(-1)^{l+m}}{m!\,l!} \beta^{l+m} \alpha_m(\mathcal{H}_2, \beta\mathcal{H}_1) \times \mathrm{Tr}\{\mathcal{H}_2^l e^{-\beta\mathcal{H}_1}\}. \tag{2.8}$$

Equating terms of the same order in $\mathcal{H}_2$, we obtain

$$\mathrm{Tr}\{\mathcal{A}\mathcal{H}_2^k e^{-\beta\mathcal{H}_1}\} = \sum_{l=0}^{k} \binom{k}{l} \alpha_{k-l}(\mathcal{H}_2, \beta\mathcal{H}_1)\,\mathrm{Tr}\{\mathcal{H}_2^l e^{-\beta\mathcal{H}_1}\}. \tag{2.9}$$

We now introduce the quantities

$$\nu_k(\mathcal{A}) \equiv \frac{\mathrm{Tr}\{\mathcal{A}\mathcal{H}_2^k e^{-\beta\mathcal{H}_1}\}}{\mathrm{Tr}\{e^{-\beta\mathcal{H}_1}\}} \tag{2.10}$$

and

$$\mu_k \equiv \frac{\mathrm{Tr}\{\mathcal{H}_2^k e^{-\beta\mathcal{H}_1}\}}{\mathrm{Tr}\{e^{-\beta\mathcal{H}_1}\}} . \tag{2.11}$$

Then from (2.9)

$$\alpha_k(\mathcal{H}_2, \beta\mathcal{H}_1) = \nu_k(\mathcal{A}) - \sum_{l=1}^{k} \binom{k}{l} \alpha_{k-l}(\mathcal{H}_2, \beta\mathcal{H}_1)\,\mu_k . \tag{2.12}$$

Equation (2.12) gives a recursion relation for the α_k. The same relation has been obtained by Stanley and Kaplan[8] for the case $\mathcal{H}_1 = 0$.

B. Specialization to Heisenberg Model

For an isotropic Heisenberg ferromagnet in the presence of a magnetic field H, the Hamiltonian is given by equation (2.1) with

$$\mathcal{H}_1 = -g\mu_B H \sum_i S_i^z \tag{2.13}$$

and

$$\mathcal{H}_2 = -\frac{1}{2}\sum_{ij} J_{ij}\, \vec{S}_i \cdot \vec{S}_j, \tag{2.14}$$

where H is the magnetic field, μ_B is the Bohr magneton, g is the g factor, and $-J_{ij}$ is the exchange energy between parallel spins on sites i and j.

We will consider only nearest-neighbor interactions, i.e.,

$$J_{ij} = J \text{ for nearest neighbors}$$
$$= 0 \text{ otherwise.} \tag{2.15}$$

In this case

$$\mathcal{H}_2 = -J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j, \tag{2.16}$$

where $\sum_{\langle ij\rangle}$ is a summation over all *different* pairs of nearest neighbors.

C. Evaluation of the Expansion Coefficients

To obtain the expansion coefficients α_k we first evaluate μ_k and ν_k. From Eqs. (2.11) and (2.16)

$$\mu_l = (-J)^l \frac{\mathrm{Tr}\{(\sum_{\langle ij\rangle} \vec{S}_i \cdot \vec{S}_j)^l e^{-\beta\mathcal{H}_1}\}}{\mathrm{Tr}\{e^{-\beta\mathcal{H}_1}\}} . \tag{2.17}$$

With each pair of spins $\vec{S}_i$, $\vec{S}_j$ we can associate a line between sites i and j. Thus each term in μ_l of the form

$$(-J)^l \frac{\mathrm{Tr}\{(\vec{S}_i \cdot \vec{S}_j)\cdots(\vec{S}_m \cdot \vec{S}_n) e^{-\beta\mathcal{H}_1}\}}{\mathrm{Tr}\{e^{-\beta\mathcal{H}_1}\}}, \tag{2.18}$$

with l pairs in the trace, is associated with a diagram containing l lines. Denoting such a diagram by d, we have

$$\mu_l = (-J)^l \sum_d \mu_l(d), \tag{2.19}$$

where

$$\mu_l(d) = \sum_c \frac{\mathrm{Tr}\{(\vec{S}_i \cdot \vec{S}_j)\cdots(\vec{S}_m \cdot \vec{S}_n) e^{-\beta\mathcal{H}_1}\}}{\mathrm{Tr}\{e^{-\beta\mathcal{H}_1}\}}, \tag{2.20}$$

and the summation $\sum_c$ is over all the combinations of pairs $\langle ij \rangle$ that correspond to the given diagram.

In a similar fashion one can represent

$$\nu_k(\mathcal{A}) = (-J)^k \sum_{d^*} \nu_k^{\mathcal{A}}(d^*) . \tag{2.21}$$

The diagrams here are denoted by d^* since in addition to the lines for each pair of spins $\vec{S}_i$, $\vec{S}_j$ they contain an additional symbol corresponding to the operator $\mathcal{A}$; e.g., *wavy lines* for operators $\mathcal{A}$ that involve two sites (e.g., for $\vec{S}_1 \cdot \vec{S}_2$, the two-spin correlation function) and *crosses* for one-site operators (e.g., for S_1^z, the magnetization). Further, we will write $\nu_k(d^*)$ instead of $\nu_k^{\mathcal{A}}(d^*)$ for the sake of brevity.

Substituting (2.19) and (2.21) into (2.12) we have

$$\alpha_k(\mathcal{H}_2, \beta\mathcal{H}_1) = (-J)^k \sum_{d^*} \alpha_k(d^*) , \qquad (2.22)$$

where

$$\alpha_k(d^*) = \nu_k(d^*) - \sum_{l=1}^{k} \binom{k}{l} \sum_{d_1^*} \sum_{d_2} \alpha_{k-l}(d_1^*)\, \mu_l(d_2), \quad [d_1^* \cup d_2 = d^*] \qquad (2.23)$$

It can be proved by induction (Appendix A) that $\alpha_k(d^*) = 0$ for diagrams d^* such that there are two diagrams d_1^* and d_2 which satisfy both

$$d^* = d_1^* \cup d_2 \qquad (2.24)$$

and

$$\langle d^* \rangle = \langle d_1^* \rangle \langle d_2 \rangle . \qquad (2.25)$$

In particular $\alpha_k(d^*) = 0$ for all disconnected diagrams.

D. The Approximations

1. Classical Approximation

In computing $\langle \vec{S}_i \cdot \vec{S}_{i+1} \rangle$ and $\langle S_i^z \rangle$ we treat the spins as classical magnetic moments of strength $g\mu_B \hbar [S(S+1)]^{1/2}$. We feel that that this is quite a reasonable approximation for EuO, for which $S = \frac{7}{2}$.[8]

2. Third-order Truncation

The primary aim of the present work is to calculate the H dependence of $\langle \vec{S}_i \cdot \vec{S}_{i+1} \rangle$ and $\langle S_i^z \rangle$ at $T \cong 4T_C$. In evaluating $\langle \vec{S}_i \cdot \vec{S}_{i+1} \rangle$ and $\langle S_i^z \rangle$ from Eq. (2.4) one obtains a series whose successive terms involve successive powers of (T_C/T). To obtain an accuracy of several percent or better in $\langle \vec{S}_i \cdot \vec{S}_j \rangle$ or $\langle S_i^z \rangle$ at $T \geq 4T_C$, it is sufficient [cf. Figs. 1 and 2] to keep in the series (2.4) only terms up to order $(T_C/T)^3$.

III. RESULTS FOR $S = \infty$ HEISENBERG FERROMAGNET WITH fcc LATTICE

All calculations were carried out for a fcc lattice corresponding to the magnetic lattice of EuO.

The quantities calculated were

$$\sigma(H, T) = \langle S_i^z \rangle / [S(S+1)]^{1/2} , \qquad (3.1)$$

$$\eta(H, T) = \langle \vec{S}_i \cdot \vec{S}_{i+1} \rangle / S(S+1), \qquad (3.2)$$

and the factor A defined by the relation

$$\eta(H, T) - \eta(0, T) = A(H, T) \sigma^2(H, T) . \qquad (3.3)$$

The reader will note that at $T > T_C$, $A(H, T) \equiv 1$ for mean-field theory (m.f.t.). Thus the deviation of A from unity is a measure of departures from m.f.t.

Appendix B gives the formulas for (i) $\mu_k(d)$, (ii) $\nu_k(d^*)$ for $\mathcal{A} = \langle \vec{S}_i \cdot \vec{S}_{i+1} \rangle$, and (iii) $\nu_k(d^{**})$ for $\mathcal{A} = \langle S_i^z \rangle$.

The results of the calculations for EuO ($T_C = 69$ K) are shown in Figs. 1–3. Examination of Fig. 3 shows that the factor $A(H, T)$ is approximately H independent at low fields where $\sigma \ll 1$. In other words, at low fields $\eta(H) - \eta(0)$ is nearly proportional to $\sigma^2(H)$. At $T \gtrsim 4T_C$, the proportionality constant A is only slightly smaller than 1.

The typical errors shown in Figs. 1 and 2 represent the magnitude of the term of order $(T_C/T)^3$.

IV. ALTERNATIVE TREATMENTS FOR LOW FIELDS

The numerical calculations of the preceding section indicate that at low magnetic fields, where σ is small compared to unity, the field dependence of η is given by Eq. (3.3), where $A(H, T)$ is nearly independent of H but does depend on T/T_C. This result will now be rederived by two different methods. The first method is based on the fact that the exchange magnetostriction of a ferromagnet (i.e., the change in volume due to the exchange interaction) can be expressed: (i) as a function of η and, (ii) as a function of σ. By equating the two expressions one obtains a relation between $\eta(H) - \eta(0)$ and σ. This relation is equivalent to Eq. (3.3), and it also allows an evaluation of the parameter A as a function of T/T_C. In order to illustrate the procedure involved in this method of deriving Eq. (3.3), we first treat in Sec. IV A the case $T \gg T_C$. In Sec. IV B the procedure is extended to any T above

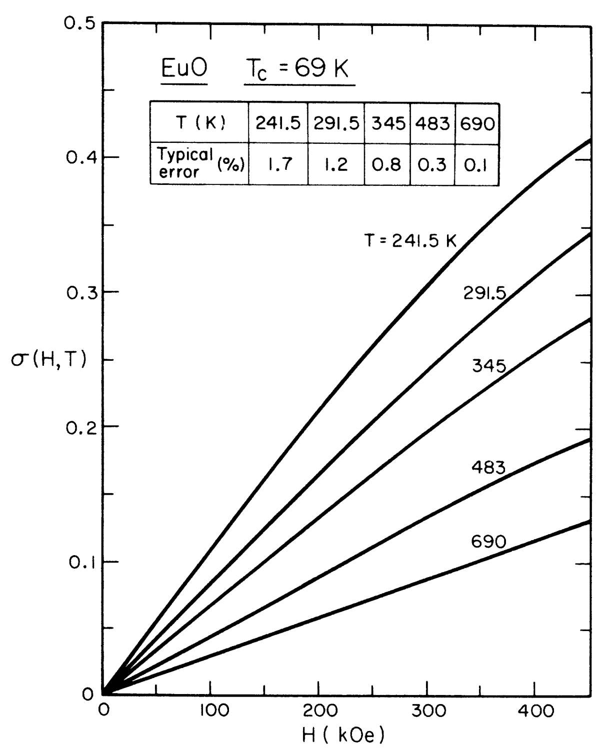

FIG. 1. Reduced magnetization $\sigma = \langle S^z \rangle / S$ vs magnetic field H for a fcc classical Heisenberg ferromagnet with $T_C = 69$ K.

T_C. A second method of deriving Eq. (3.3) from a purely thermodynamic argument is presented in Sec. IV C. Numerical results for A as a function of T/T_C are given in Sec. IV D.

A. $T \gg T_C$

We consider an isotropic ferromagnet with nearest-neighbor interaction only. At $T \gg T_C$, the magnetization *at low fields* is given by the Curie-Weiss law

$$M = CH/(T-\theta), \quad (4.1)$$

where

$$\theta = zJS(S+1)/3k \quad (4.2)$$

is the Curie-Weiss temperature, z is the number of nearest neighbors, and

$$C = Ng^2\mu_B^2 S(S+1)/3k. \quad (4.3)$$

Here N is the number of magnetic ions per cm^3.

When a magnetic field is applied, the volume V changes slightly. The change in V obeys one of Maxwell's relations

$$\left(\frac{\partial V}{\partial H}\right)_{P,T} = -\left(\frac{\partial M}{\partial P}\right)_{T,H}, \quad (4.4)$$

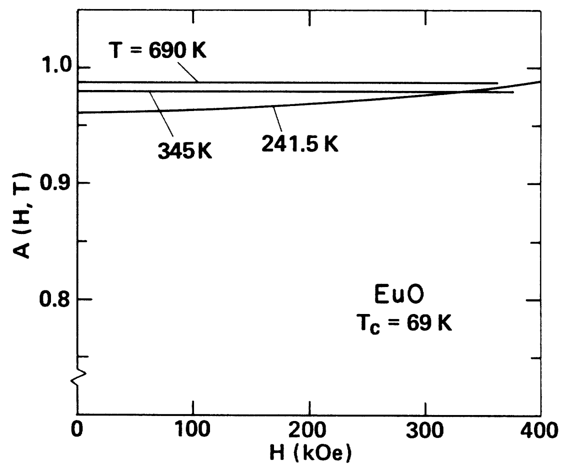

FIG. 3. Dependence of the parameter $A(H, T)$, defined by Eq. (3.3), on H at several values of T. The results are for a fcc classical Heisenberg ferromagnet with T_C = 69 K.

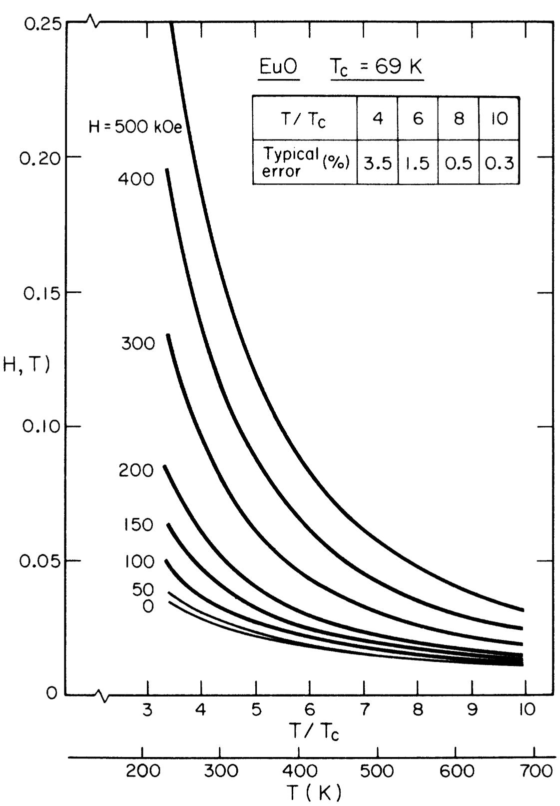

FIG. 2. Reduced two-spin correlation function $\eta = \langle \vec{S}_i \cdot \vec{S}_{i+1} \rangle / S^2$ vs T for various values of H. The results are for a fcc classical Heisenberg ferromagnet.

where P is the pressure. In order to evaluate the right-hand side of Eq. (4.4) we note that the exchange constant J depends on V. Since V depends on P, a change in P will result in a change in J. The change in J will result in a change of θ [see Eq. (4.2)], which will lead to a change in M [see Eq. (4.1)]. Mathematically,

$$\frac{\partial M}{\partial P} = \left(\frac{\partial M}{\partial \theta}\right)\left(\frac{\partial \theta}{\partial J}\right)\left(\frac{\partial J}{\partial V}\right)\left(\frac{\partial V}{\partial P}\right). \quad (4.5)$$

From Eqs. (4.1), (4.4), and (4.5)

$$\frac{\partial V}{\partial H} = -\frac{CH}{(T-\theta)^2}\left(\frac{\partial \theta}{\partial J}\right)\left(\frac{\partial J}{\partial V}\right)\left(\frac{\partial V}{\partial P}\right), \quad (4.6)$$

but

$$\left(\frac{\partial V}{\partial P}\right)_{T,H} = -VK_{T,H}, \quad (4.7)$$

where $K_{T,H}$ is the compressibility at constant T and H. Also, from Eqs. (4.2) and (4.3)

$$\frac{\partial \theta}{\partial J} = \frac{\theta}{J} = \frac{zC}{Ng^2\mu_B^2}. \quad (4.8)$$

Therefore,

$$\frac{\partial V}{\partial H} = \left(\frac{zVK_{T,H}}{Ng^2\mu_B^2}\right)\left(\frac{\partial J}{\partial V}\right)\frac{C^2H}{(T-\theta)^2}. \quad (4.9)$$

Integrating and using Eq. (4.1), one obtains the volume change at constant T and P

$$V(H) - V(0) = \left(\frac{zVK_{T,H}}{2Ng^2\mu_B^2}\right)\left(\frac{\partial J}{\partial V}\right)M^2. \quad (4.10)$$

But $M = Ng\mu_B\langle S^z\rangle$, so that

$$V(H) - V(0) = \tfrac{1}{2}NVzS^2K_{T,H}\left(\frac{\partial J}{\partial V}\right)\sigma^2, \quad (4.11)$$

where $\sigma = \langle s^z\rangle/S$ is the reduced magnetization.

Equation (4.11) relates the volume magnetostriction to σ. A second relation for $V(H) - V(0)$ in terms of η is obtained from the work of Callen and Callen,[4] who showed that the volume magnetostriction $\Delta V/V$ is proportional to η. The proportionality constant can be obtained from Eq. (21) of Argyle *et al.*,[9] viz.,

$$\frac{\Delta V}{V} = \tfrac{1}{2} N z K_{T,H}\left(\frac{\partial J}{\partial V}\right) \langle \vec{S}_i \cdot \vec{S}_{i+1}\rangle . \tag{4.12}$$

The same proportionality constant can also be calculated by considering the exchange magnetostriction ΔV at $T = H = P = 0$. Under these conditions ΔV is obtained by minimizing the energy

$$E = -\tfrac{1}{2} N z \langle \vec{S}_i \cdot \vec{S}_{i+1}\rangle \left[J(V_0) + \Delta V\left(\frac{\partial J}{\partial V}\right)\right] + \frac{(\Delta V)^2}{2K_{T,H} V} . \tag{4.13}$$

Minimizing Eq. (4.13) with respect to ΔV leads to Eq. (4.12).

From Eq. (4.12)

$$V(H) - V(0) = \tfrac{1}{2} N V z S^2 K_{T,H}\left(\frac{\partial J}{\partial V}\right)[\eta(H) - \eta(0)], \tag{4.14}$$

where $\eta = \langle \vec{S}_1 \cdot \vec{S}_{i+1}\rangle/S^2$. Equating Eqs. (4.11) and (4.14) gives

$$\eta(H) - \eta(0) = \sigma^2 . \tag{4.15}$$

Equation (4.15) is equivalent to Eq. (3.3) with $A = 1$. The result $A = 1$ in the limit $T \gg T_C$ is expected physically.

B. Any T Above T_C

At any T above T_C the magnetization at low fields ($\sigma \ll 1$) can be written

$$M = CH/(T - \theta^*) , \tag{4.16}$$

where

$$\theta^* = \theta f \tag{4.17}$$

is a new temperature which is equal to the Curie-Weiss temperature θ multiplied by a correction factor f. The factor f is a function of T/T_C (or T/J). In the limit $T/T_C \to \infty$, $f \to 1$.

Following the same procedure which was used to derive Eq. (4.11) one obtains

$$V(H) - V(0) = \tfrac{1}{2} N V z S^2 K_{T,H}\left(\frac{\partial J}{\partial V}\right) \sigma^2 f\left(1 + \frac{\partial \ln f}{\partial \ln J}\right) , \tag{4.18}$$

where the derivative $\partial \ln f/\partial \ln J$ is evaluated at constant T. Note that the right-hand side of Eq. (4.18) differs from that of Eq. (4.11) by the factor $f(1 + \partial \ln f/\partial \ln J)$. Since Eq. (4.14) is not restricted to $T \gg T_C$, a relation between η and σ can be obtained by equating the right-hand sides of Eqs. (4.18) and (4.14). This gives

$$\eta(H) - \eta(0) = A\sigma^2 , \tag{4.19}$$

where

$$A = f\left(1 + \frac{\partial \ln f}{\partial \ln J}\right). \tag{4.20}$$

C. Thermodynamic Treatment

Equation (4.20) can also be derived from purely thermodynamic arguments. We consider the energy U_A as defined by Kittel.[10] This energy is given by

$$U_A = \langle \mathcal{H}\rangle = \left\langle -J \sum_{\langle ij\rangle} \vec{S}_i \cdot \vec{S}_j - g\mu_B H \sum_i S_i^z \right\rangle, \tag{4.21}$$

or, since there are $\frac{1}{2}Nz$ independent pairs $\langle ij\rangle$ of nearest neighbors,

$$U_A = -\tfrac{1}{2} N z J \langle \vec{S}_i \cdot \vec{S}_{i+1}\rangle - MH. \tag{4.22}$$

Therefore,

$$dU_A = -\tfrac{1}{2} N z J\, d\langle \vec{S}_i \cdot \vec{S}_{i+1}\rangle - M dH - H dM . \tag{4.22}$$

But

$$dU_A = T ds - M dH, \tag{4.23}$$

where s is the entropy. Therefore,

$$T ds = -\tfrac{1}{2} N z J\, d\langle \vec{S}_i \cdot \vec{S}_{i+1}\rangle - H dM . \tag{4.24}$$

Hence

$$T\left(\frac{\partial s}{\partial H}\right)_T = -\tfrac{1}{2} N z J \left(\frac{\partial \langle \vec{S}_i \cdot \vec{S}_{i+1}\rangle}{\partial H}\right)_T - \left(\frac{CH}{T - \theta^*}\right), \tag{4.25}$$

where we used Eq. (4.16), which applies when $\sigma \ll 1$ and $T > T_C$. Using the Maxwell relation

$$\left(\frac{\partial M}{\partial T}\right)_H = \left(\frac{\partial s}{\partial H}\right)_T \tag{4.26}$$

and Eqs. (4.16), (4.17), and (4.25), one obtains after some algebra

$$\left(\frac{\partial \langle \vec{S}_i \cdot \vec{S}_{i+1}\rangle}{\partial H}\right)_T = \frac{2CH\theta^*}{NzJ(T - \theta^*)^2}\left(1 - \frac{\partial \ln f}{\partial \ln T}\right). \tag{4.27}$$

From Eqs. (4.8), (4.17), and (4.27),

$$\left(\frac{\partial \langle \vec{S}_i \cdot \vec{S}_{i+1}\rangle}{\partial H}\right)_T = \frac{2C^2 H f}{N^2 g^2 \mu_B^2 (T - \theta^*)^2}\left(1 - \frac{\partial \ln f}{\partial \ln T}\right) . \tag{4.28}$$

Integrating with respect to H and using Eq. (4.16)

$$\langle \vec{S}_i \cdot \vec{S}_{i+1}\rangle_H - \langle \vec{S}_i \cdot \vec{S}_{i+1}\rangle_0 = \frac{M^2 f}{N^2 g^2 \mu_B^2}\left(1 - \frac{\partial \ln f}{\partial \ln T}\right) \tag{4.29}$$

or

$$\eta(H) - \eta(0) = \sigma^2 f\left(1 - \frac{\partial \ln f}{\partial \ln T}\right). \tag{4.30}$$

Since f is a function of T/J,

$$\frac{\partial \ln f}{\partial \ln T} = \frac{-\partial \ln f}{\partial \ln J} , \tag{4.31}$$

so that

$$\eta(H)-\eta(0)=\sigma^2 f\left(1+\frac{\partial \ln f}{\partial \ln J}\right),$$

which is Eq. (4.20).

D. Numerical Results

Equation (4.20) shows that the coefficient A in Eqs. (3.3) or (4.19) is related to the factor f, which is a function of T/T_C (or T/J). The factor f can be evaluated if the zero-field susceptibility $\chi = C/(T-f\theta)$ is known. High-temperature series expansions for χ were given by several authors. In particular, expressions for a fcc lattice were given by Rushbrooke and Wood[11] and by Domb and Sykes[12] for an arbitrary S and up to order $(J/kT)^6$. Two additional terms, up to order $(J/kT)^8$, were given by Wood and Rushbrooke for the case $S=\infty$.[13]

Figures 4 and 5 show f and A as a function of T/T_C for a fcc lattice and $S=\infty$. These curves were calculated from the Wood-Rushbrooke series expansion for χ.[13] The solid curves were obtained using the expansion up to order $(J/kT)^8$, i.e., a nine-term expansion. To get an idea of the accuracy of these results, we have also calculated $f(T/T_C)$ and $A(T/T_C)$ from the expansion for χ, keeping terms only up to order $(J/kT)^6$ or $(J/kT)^4$. These are shown in Figs. 4 and 5 as a dashed, and a dashed-dot lines, respectively. It appears that the nine-term expansion for χ leads to values of A which are accurate to within ~1% at $T/T_C \gtrsim 4$. Figure 5 also shows some values for A which were obtained from the calculations in Sec. III.

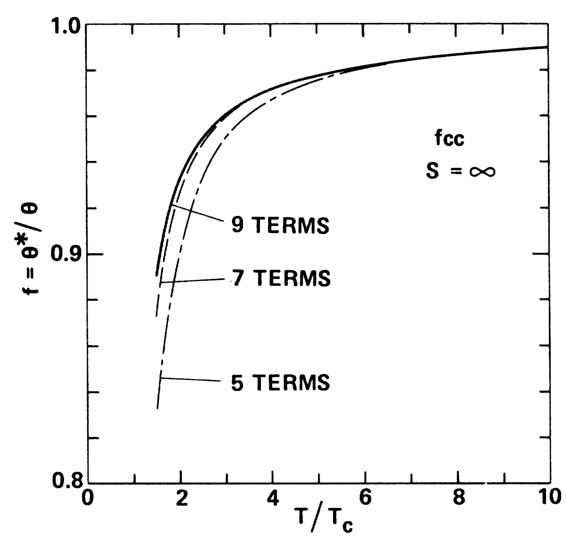

FIG. 4. T dependence of the factor f in Eq. (4.17). These results for a fcc classical Heisenberg ferromagnet were obtained using Wood-Rushbrooke's series expansion for χ. The various curves correspond to different truncations of this series.

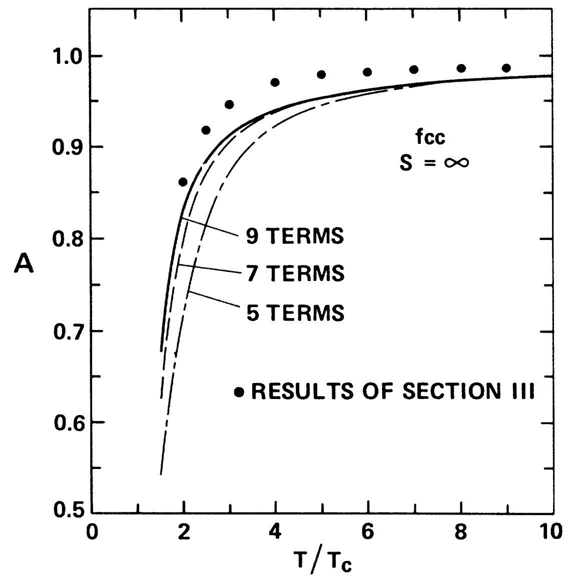

FIG. 5. T dependence of the parameter A in Eqs. (4.19) and (4.20). The various curves were obtained from the results in Fig. 4. The dots represent the numerical results of Sec. III for the parameter A at low fields (H = 50 kOe).

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APPENDIX A. PROOF OF "LINKED-CLUSTER" THEOREM

In the text it was stated that one can prove by induction from (3) that $\alpha_l(d^*)=0$ for diagrams d^* which may be partitioned $d^*=d_1^*\cup d_2$ such that $\langle d_1^*\rangle = \langle d_1^*\rangle\langle d_2\rangle$. This factorization occurs (even quantum mechanically) for all disconnected diagrams.

Proof: $\alpha_l(d^*)=0$ for $l=2$ by inspection. Assume, then, that $\alpha_l(d^*)=0$ for all $l\le\Lambda-1$, and consider Equation (2.23) for $l=\Lambda$:

$$\alpha_\Lambda(d^*)=\nu_\Lambda(d^*)-\sum_{k=1}^{\Lambda-1}\binom{\Lambda}{k}\sum_{d_a^*,d_b}\alpha_k(d_a^*)\,\mu_{\Lambda-k}(d_b), \tag{A1}$$

where $\sum_{d_a^*,d_b}$ denotes a summation on all partitions $d^*=d_a^*\cup d_b$ of d^* such that d_a^* has k lines and d_b has $\Lambda-k$ lines. Now write the summation $\sum_{k=1}^{\Lambda-1}(\cdots)$ in Equation (A1) as the sum of three terms:

$$\text{term I.} \equiv \sum_{k=1}^{l_1-1}(\cdots),$$

$$\text{term II.} \equiv (k=l_1\,\text{term}),$$

$$\text{term III.} \equiv \sum_{k=l_1+1}^{\Lambda-1}(\cdots),$$

where $l_1\equiv$ number of straight lines in d_1^*.

Now term III $=0$ since $\alpha_k(d_a^*)=0$ for $l_1+1 \leq k \leq \Lambda - 1$.

$$\text{term II} = \binom{\Lambda}{l_1} \alpha_{l_1}(d_1^*)\, \mu_{l_2}(d_2),$$

where $l_2 \equiv \Lambda - l_1 \equiv$ number of straight lines in d_2, since all other partitioning of d^* are such that $\alpha_k(d_a^*)=0$.

The only nonzero $\alpha_k(d_a^*)$ in term I are those for which all the lines of d_a^* are contained in d_1^*. But for these cases

$$\mu_{\Lambda-k}(d_b) = \binom{\Lambda-k}{l_2} \mu_{l_2}(d_2)\, \mu_{l_1-k}(d_c), \qquad \text{(A2)}$$

where d_c is such that $d_a^* \cup d_c = d_1^*$.

Observe now that

$$\binom{\Lambda}{k}\binom{\Lambda-k}{l_2} = \binom{\Lambda}{l_1}\binom{l_1}{k} \qquad \text{(A3)}$$

and also that

$$\nu_\Lambda(d) = \binom{\Lambda}{l_1} \nu_{l_1}(d_1^*)\, \mu_{l_2}(d_2). \qquad \text{(A4)}$$

Hence Equation (A1) may be written

$$\alpha_\Lambda(d^*) = \binom{\Lambda}{l_1} \mu_{l_2}(d_2)\{\cdots\}, \qquad \text{(A5)}$$

where

$$\{\cdots\} = \nu_{l_1}(d_1^*) - \alpha_{l_1}(d_1^*) - \sum_{k=1}^{l_1-1} \binom{l_1}{k} \sum_{d_a^*, d_c} \alpha_k(d_a^*)\, \mu_{l_1-k}(d_c) = 0. \qquad \text{(A6)}$$

Thus $\alpha_\Lambda(d^*)=0$ and the proof is complete.

APPENDIX B. EVALUATION OF EXPANSION COEFFICIENTS

In this appendix we give all the quantities necessary for the four-term expansion of η and σ. The diagrams are numbered according to Fig. 6. The numbers of diagrams d^* and d^{**} that can be placed on the fcc lattice is given in Table I.

To shorten the notation, let us define the following quantities:

$$X_i(z) \equiv \left(\frac{d^i}{dz^i} I(z)\right)\Big/ I(z), \quad i=1,2,3\cdots, \qquad \text{(B1)}$$

where

$$I(z) \equiv 4\pi \frac{\sinh z}{z} \quad \text{and} \quad z = \frac{g\mu_B \hbar [S(S+1)]^{1/2} H}{kT}. \qquad \text{(B2)}$$

Suppressing z in the following (for brevity), the coefficients in the expansion are given by

$$\mu_1(1.1) = X_1^2[S(S+1)],$$

$$\mu_2(2.1) = [X_2^2 + \tfrac{1}{2}(1-X_2)^2][S(S+1)]^2,$$

$$\mu_2(2.2) = 2X_1^2 X_2 [S(S+1)]^2,$$

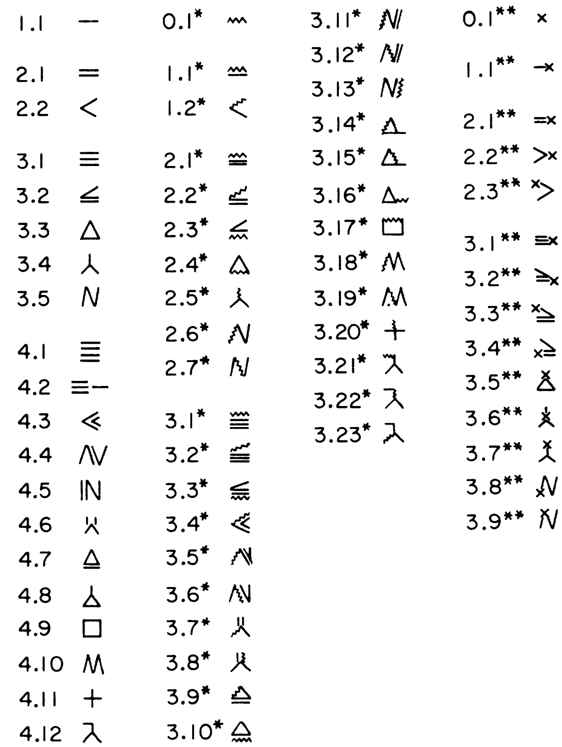

FIG. 6. Labeling of various graphs which appear in Appendix B.

$$\mu_3(3.1) = [X_3^2 + \tfrac{3}{2}(X_1-X_3)^2][S(S+1)]^3,$$

$$\mu_3(3.2) = 3X_1[X_2X_3 + \tfrac{1}{2}(1-X_2)(X_1-X_3)][S(S+1)]^3,$$

$$\mu_3(3.3) = 6[X_2^3 + \tfrac{1}{4}(1-X_2)^3][S(S+1)]^3,$$

$$\mu_3(3.4) = 6X_1^3 X_3 [S(S+1)]^3,$$

$$\mu_3(3.5) = 6X_1^2 X_2^2 [S(S+1)]^3,$$

$$\mu_4(4.1) = [X_4^2 + 3(X_2-X_4)^2 + \tfrac{3}{8}(1-2X_2+X_4)^2] \times [S(S+1)]^4,$$

$$\mu_4(4.2) = 4X_1[X_3X_4 + \tfrac{3}{2}(X_2-X_4)(X_1-X_3)] \times [S(S+1)]^4,$$

$$\mu_4(4.3) = 6[X_2^2X_4 + X_2(1-X_2)(X_2-X_4) + \tfrac{1}{4}(1-X_2)^2(1-2X_2+X_4)][S(S+1)]^4,$$

$$\mu_4(4.4) = 12X_1^2[X_3^2 + \tfrac{1}{2}(X_1-X_3)^2][S(S+1)]^4,$$

$$\mu_4(4.5) = 12X_1X_2[X_3X_2 + \tfrac{1}{2}(1-X_2)(X_1-X_3)] \times [S(S+1)]^4,$$

$$\mu_4(4.6) = 12X_1^2[X_2X_4 + \tfrac{1}{2}(X_2-X_4)(1-X_2)] \times [S(S+1)]^4,$$

$$\mu_4(4.7) = 12[X_2X_3^2 + \tfrac{1}{2}(X_1-X_3)^2][S(S+1)]^4,$$

$$\mu_4(4.8) = 24X_1[X_2^2X_3 + \tfrac{1}{4}(X_1-X_3)(1-X_2)^2] \times [S(S+1)]^4,$$

TABLE I. Numbers of diagrams d^* and d^{**} that can be placed on an fcc lattice.

Diagram	No.	Diagram	No.	Diagram	No.
0.1*	1	3.7*	110	0.1**	1
		3.8*	110		
1.1*	1	3.9*	4	1.1**	12
1.2*	22	3.10*	4		
		3.11*	234	2.1**	12
2.1*	1	3.12*	234	2.2**	66
2.2*	22	3.13*	234	2.3**	132
2.3*	22	3.14*	40		
2.4*	4	3.15*	80	3.1**	12
2.5*	110	3.16*	40	3.2**	66
2.6*	234	3.17*	22	3.3**	132
2.7*	234	3.18*	2490	3.4**	132
		3.19*	2450	3.5**	24
3.1*	1	3.20*	330	3.6**	220
3.2*	22	3.21*	1130	3.7**	660
3.3*	22	3.22*	1130	3.8**	1404
3.4*	22	3.23*	2300	3.9**	1404
3.5*	234				
3.6*	234				

$$\mu_4(4.9)=24[X_2^4+\tfrac{1}{8}(1-X_2)^4][S(S+1)]^4,$$
$$\mu_4(4.10)=24X_1^2X_2^3[S(S+1)]^4,$$
$$\mu_4(4.11)=24X_1^4X_4[S(S+1)]^4,$$
$$\mu_4(4.12)=24X_1^3X_2X_3[S(S+1)]^4.$$

The coefficients $\nu_k(d^*)$ for $\mathcal{A}=\langle\vec{S}_i\cdot\vec{S}_{i+1}\rangle$ are

$\nu_0(0.1^*)=\mu_1(1.1)$, $\nu_1(1.1^*)=\mu_2(2.1)$,

$\nu_1(1.2^*)=\tfrac{1}{2}\mu_2(2.2)$, $\nu_2(2.1^*)=\mu_3(3.1)$,

$\nu_2(2.2^*)=\tfrac{1}{3}\mu_3(3.2)$, $\nu_2(2.3^*)=\tfrac{2}{3}\mu_3(3.2)$,

$\nu_2(2.4^*)=\tfrac{1}{3}\mu_3(3.3)$, $\nu_2(2.5^*)=\tfrac{1}{3}\mu_3(3.4)$,

$\nu_2(2.6^*)=\tfrac{1}{3}\mu_3(3.5)$, $\nu_2(2.7^*)=\tfrac{1}{3}\mu_3(3.5)$,

$\nu_3(3.1^*)=\mu_4(4.1)$, $\nu_3(3.2^*)=\tfrac{1}{4}\mu(4.2)$,

$\nu_3(3.3^*)=\tfrac{3}{4}\mu_4(4.2)$, $\nu_3(3.4^*)=\tfrac{1}{2}\mu_4(4.3)$,

$\nu_3(3.5^*)=\tfrac{1}{4}\mu_4(4.4)$, $\nu_3(3.6^*)=\tfrac{1}{2}\mu_4(4.4)$,

$\nu_3(3.7^*)=\tfrac{1}{4}\mu_4(4.6)$, $\nu_3(3.8^*)=\tfrac{1}{4}\mu_4(4.6)$,

$\nu_3(3.9^*)=\tfrac{1}{4}\mu_4(4.7)$, $\nu_3(3.10^*)=\tfrac{1}{2}\mu_4(4.7)$,

$\nu_3(3.11^*)=\tfrac{1}{4}\mu_4(4.5)$, $\nu_3(3.12^*)=\tfrac{1}{4}\mu_4(4.5)$,

$\nu_3(3.13^*)=\tfrac{1}{2}\mu_4(4.5)$, $\nu_3(3.14^*)=\tfrac{1}{4}\mu_4(4.8)$,

$\nu_3(3.15^*)=\tfrac{1}{4}\mu_4(4.8)$, $\nu_3(3.16^*)=\tfrac{1}{4}\mu_4(4.8)$,

$\nu_3(3.17^*)=\tfrac{1}{4}\mu_4(4.9)$, $\nu_3(3.18^*)=\tfrac{1}{4}\mu(4.10)$,

$\nu_3(3.19^*)=\tfrac{1}{4}\mu_4(4.10)$, $\nu_3(3.20^*)=\tfrac{1}{4}\mu_4(4.11)$,

$\nu_3(3.21^*)=\tfrac{1}{4}\mu_4(4.12)$, $\nu_3(3.22^*)=\tfrac{1}{4}\mu_4(4.12)$,

$\nu_3(3.23^*)=\tfrac{1}{4}\mu_4(4.12)$.

The coefficients $\nu_k(d^{**})$ for $\mathcal{A}=\langle S_i^z\rangle$ are,

$$\nu_0(0.1^{**})=X_1[S(S+1)]^{1/2},$$
$$\nu_1(1.1^{**})=X_1X_2[S(S+1)]^{3/2},$$
$$\nu_2(2.1^{**})=[X_2X_3+\tfrac{1}{2}(1-X_2)(X_1-X_3)][S(S+1)]^{5/2},$$
$$\nu_2(2.2^{**})=2X_3X_1^2[S(S+1)]^{5/2},$$
$$\nu_2(2.3^{**})=2X_1X_2^2[S(S+1)]^{5/2},$$
$$\nu_3(3.1^{**})=[X_4X_3+\tfrac{3}{2}(X_1-X_3)(X_2-X_4)][S(S+1)]^{7/2},$$
$$\nu_3(3.2^{**})=3[X_4X_2X_1+\tfrac{1}{2}X_1(1-X_2)(X_2-X_4)]\times[S(S+1)]^{7/2},$$
$$\nu_3(3.3^{**})=3[X_3X_2^2+\tfrac{1}{2}X_2(1-X_2)(X_1-X_3)][S(S+1)]^{7/2},$$
$$\nu_3(3.4^{**})=3[X_1X_3^2+\tfrac{1}{2}X_1(X_1-X_3)^2][S(S+1)]^{7/2},$$
$$\nu_3(3.5^{**})=6[X_2^2X_3+\tfrac{1}{4}(1-X_2)^2(X_1-X_3)][S(S+1)]^{7/2},$$
$$\nu_3(3.6^{**})=6X_1^3X_4[S(S+1)]^{7/2},$$
$$\nu_3(3.7^{**})=6X_1^2X_2X_3[S(S+1)]^{7/2},$$
$$\nu_3(3.8^{**})=6X_1X_2^3[S(S+1)]^{7/2},$$
$$\nu_3(3.9^{**})=6X_1^2X_2X_3[S(S+1)]^{7/2}.$$

As an example of calculation of $\alpha_m(d^*)$ let us calculate $\alpha_3(3.3^*)$ and $\alpha_3(3.22^*)$

$$\alpha_3(3.3^*)=\nu_3(3.3^*)-\binom{3}{1}[\alpha_2(2.1^*)+d_2(2.2^*)]\,\mu_1(1.1)-\binom{3}{2}[\alpha_1(1.1^*)\,\mu_2(2.2)+\alpha_1(1.2^*)\,\mu_2(2.1)]-\binom{3}{3}\alpha_0(0.1^*)\,\mu_3(3.2) \quad \text{(B3)}$$

$$\alpha_3(3.22^*)=\nu_3(3.22^*)-\binom{3}{1}[\alpha_2(2.5^*)+2\alpha_2(2.7^*)]\,\mu_1(1.1)-\binom{3}{2}\alpha_1(1.2^*)\times[2\mu_1^2(1.1)+\mu_2(2.2)]-\binom{3}{3}\alpha_0(0.1^*)\,\mu_1(1.1)\,\mu_2(2.2). \quad \text{(B4)}$$

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[1]This statement is by no means rigorous, even for the ideal Heisenberg magnet. However, it is intuitively plausible and in accord with all of the calculations of this work.

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