

distribution. The rapid rise from the minimum for cesium suggests a rapid increase in the spectral distribution. Rubidium indicates a slower increase. Further analysis does not seem warranted in this paper. Also no procedure is apparent for examining the suggestion by Jones⁹ that the lattice may contribute a linear term to the heat capacity and it has been assumed that the linear heat capacity is associated with the electrons.

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Longitudinal Correlation Function of the Heisenberg Ferromagnet*

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The correlation function of the z components (parallel to the net magnetization) of two spins in a Heisenberg ferromagnet is computed by a Green function analysis. The results extend spin-wave theory to higher temperatures, although they are not satisfactory near the Curie temperature. The Fourier transform of the correlation function varies as $1/k$ for small k , with a more complicated behavior at large k .

1. INTRODUCTION

THE spin-wave theory of the Heisenberg ferromagnet, valid at low temperatures, previously has been extended to arbitrary temperature by the method of double-time, temperature-dependent Green functions.¹⁻⁴ This theory yields both the magnetization and the transverse correlation function $\langle S_m^\alpha S_p^\beta \rangle$, where $\mathbf{m}$ and $\mathbf{p}$ label lattice sites and α, β can be either x or y (the external magnetic field being parallel to the z axis). The longitudinal correlation function $\langle S_m^z S_p^z \rangle - \langle S^z \rangle^2$, which is somewhat more difficult to analyze, is the subject of this paper.

The longitudinal correlation function is of direct physical interest in several connections as, for example, the thermodynamic energy and specific heat, the magnetic scattering of neutrons, and the magnetic susceptibility.

Several studies of the correlation function have been carried out. Van Hove,⁵ by analogy with the classical, phenomenological, Ornstein-Zernicke⁶ theory of fluctuations, postulated that the longitudinal correlation function has the Yukawa form $e^{-\kappa r}/r$, both above and below the Curie temperature, and thereby analyzed

the critical scattering of neutrons. To discuss the same problem, De Gennes⁷ used a generalized (wavelength-dependent) molecular field model, and Elliott and Marshall⁸ employed a Bethe-Peierls-Weiss model; these models give the same functional dependence on distance as postulated by van Hove. However, at low temperatures the spin-wave theory yields a different form, the Fourier transform of the correlation function varying as $1/k$ rather than as $1/(k^2 + \kappa^2)$. Recently, Kawaski and Mori⁹ have given a very thorough investigation of the generalized susceptibility

$$\chi(k) = \mu^2 \beta \left[\beta^{-1} \int_0^\beta d\lambda \langle S_k^z S_{-k}^z(i\hbar\lambda) \rangle - \langle S_k^z \rangle \langle S_{-k}^z \rangle \right] \quad (1)$$

for the special case of spin $\frac{1}{2}$. Here S_k^z is the Fourier transform of S_m^z , $S_k^z(i\hbar\lambda)$ is the Heisenberg operator $\exp(-\lambda\mathcal{H}) S_k^z \exp(\lambda\mathcal{H})$, and $\beta = (k_B T)^{-1}$. Although $\chi(k)$ is not directly related to the correlation function for general k , in the limit of small k the quantity $\chi(k)/\mu^2 \beta$ approaches the Fourier transform of the correlation function. Kawasaki and Mori find the functional form $1/k(k + \kappa_1) \sim \chi(k)$ below T_c (with $\kappa_1 \rightarrow 0$ as $T \rightarrow T_c^-$), agreeing with spin-wave theory in the limit of low k , and they find $\chi(k) \sim 1/(k^2 + \kappa^2)$ above T_c .

By a Green function approach, we find the correlation function for arbitrary spin. In the limit of low k the

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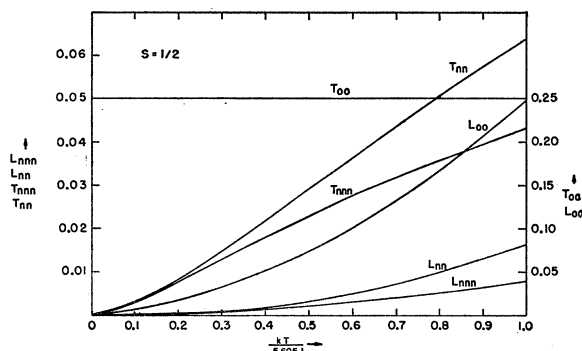


FIG. 1. Correlation functions for $S=\frac{1}{2}$, in the face-centered cubic lattice.

Fourier transform of the correlation function varies as $1/k$, agreeing precisely with spin-wave theory in the low-temperature region. The results of the calculation are shown in Figs. 1-4, in which we show the transverse correlation function for nearest-neighbor spins (T_{nn}) the transverse correlation function for next-nearest-neighbor spins (T_{nnn}), and the transverse self-correlation (T_{00}), as well as the corresponding longitudinal correlations (L_{nn} , L_{nnn} , and L_{00}), for spins $\frac{1}{2}$, $\frac{3}{2}$, $\frac{5}{2}$, and $\frac{7}{2}$, all in a face-centered cubic lattice. The magnetization as a function of temperature, as predicted by the Green function analysis of Ref. 3, is also shown in Fig. 5.

The decoupling approximation which we employ to terminate the hierarchy of Green function equations is that proposed by Callen.³ The range of validity of that approximation was clarified by a subsequent study of decoupling procedures by Tahir-Kheli.⁴ The Callen decoupling consists of a particular choice of a mass operator, whereas an attempt at a more rigorous formulation of the Green function theory by Wortis¹⁰ suggests that an additional inhomogeneous term must also appear. Tahir-Kheli inverted the problem to investigate the form of this inhomogeneous term and of the mass operator which would produce optimum agree-

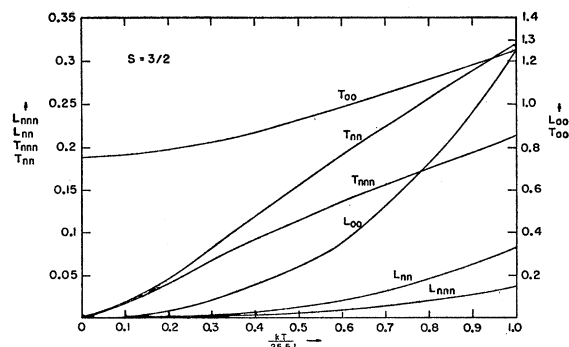


FIG. 2. Correlation functions for $S=\frac{3}{2}$, in the face-centered cubic lattice.

¹⁰ M. Wortis, Ph.D. thesis, Harvard University, 1963 (unpublished).

ment with those results known rigorously in the spin-wave region and in the high-temperature region by Opechowski series expansions and Padé extrapolations. His results indicate that the form of the mass operator assumed by Callen is correct, that the inhomogeneous term is large at all temperatures for spin $\frac{1}{2}$, but that for spin $\neq \frac{1}{2}$ the inhomogeneous term is small for temperatures appreciably lower than the Curie temperature. Hence the simple decoupling procedure used here is expected to yield a theory which, for $S \neq \frac{1}{2}$, extends the region of validity beyond the range of spin-wave theory. Our results correspondingly agree with spin-wave theory at low temperatures, but they become unreasonable in the vicinity of the Curie temperature, as expected. In particular the longitudinal and transverse correlation functions do not become isotropic at the Curie temperature in the absence of an applied field.

2. THE GREEN FUNCTION EQUATIONS

We consider a system described by an isotropic exchange interaction between localized spins, plus the

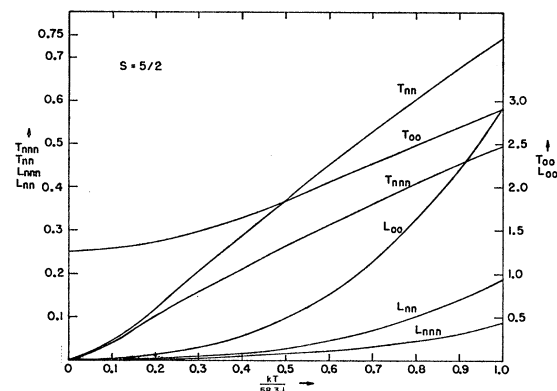


FIG. 3. Correlation functions for $S=\frac{5}{2}$, in the face-centered cubic lattice.

Zeeman interaction with an externally applied magnetic field H .

$$\mathcal{H} = - \sum_{\mathbf{m}, \mathbf{p}} J(\mathbf{m}-\mathbf{p}) \mathbf{S}_{\mathbf{m}} \cdot \mathbf{S}_{\mathbf{p}} - \mu H \sum_{\mathbf{p}} S_{\mathbf{p}}^z. \quad (2)$$

Here $\mathbf{m}$ and $\mathbf{p}$ label lattice sites, $J(\mathbf{m}-\mathbf{p})$ is a function only of the distance between sites $\mathbf{m}$ and $\mathbf{p}$, and $\mu \mathbf{S}$ is the magnetic moment of each site.

Although we are interested in the quantity $\langle S_{\mathbf{m}}^z S_{\mathbf{p}}^z \rangle$ it is convenient to consider the more general quantity

$$\Lambda^{(a)}(\mathbf{m}-\mathbf{p}) \equiv \langle S_{\mathbf{m}}^z \exp(a S_{\mathbf{p}}^z) \rangle. \quad (3)$$

By differentiation of $\Lambda^{(a)}$ with respect to the parameter a and then taking the limit $a \rightarrow 0$ we can obtain the desired quantity $\langle S_{\mathbf{m}}^z S_{\mathbf{p}}^z \rangle$. As shown in Ref. 3 the introduction of the parameter a facilitates the generalization of the Green function formalism to arbitrary spin.

In order to compute $\Lambda^{(a)}(\mathbf{m}-\mathbf{p})$ we introduce the double-time, temperature-dependent, retarded and advanced Green function:

$$G^{(a)}(\mathbf{g}, \mathbf{m}, \mathbf{p}; t-t') = \langle\langle S_{\mathbf{g}}^+(t); C^{(a)}(\mathbf{m}, \mathbf{p}; t') \rangle\rangle, \quad (4)$$

where

$$C^{(a)}(\mathbf{m}, \mathbf{p}; t') \equiv S_{\mathbf{m}}^z(t') \exp[as_{\mathbf{p}}^z(t')] S_{\mathbf{p}}^-(t'). \quad (5)$$

The Fourier transform $G_E^{(a)}(\mathbf{g}, \mathbf{m}, \mathbf{p})$, defined by

$$G_E^{(a)}(\mathbf{g}, \mathbf{m}, \mathbf{p}) = \frac{1}{2\pi} \int_{-\infty}^{\infty} G^{(a)}(\mathbf{g}, \mathbf{m}, \mathbf{p}; t-t') e^{iE(t-t')/\hbar} dt, \quad (6)$$

obeys³ the equation of motion

$$\begin{aligned} [E - \mu H] G_E^{(a)}(\mathbf{g}, \mathbf{m}, \mathbf{p}) &= -\frac{1}{2\pi} [\delta_{\mathbf{g}, \mathbf{p}} \mathcal{D}^{(a)} \Lambda^{(a)}(\mathbf{m}-\mathbf{g}) - \delta_{\mathbf{g}, \mathbf{m}} \langle S_{\mathbf{m}}^+ \exp(as_{\mathbf{p}}^z) S_{\mathbf{p}}^- \rangle] \\ &\quad + 2 \sum_{\mathbf{f}} J(\mathbf{g}-\mathbf{f}) \langle\langle S_{\mathbf{f}}^z(t) S_{\mathbf{g}}^+(t) - S_{\mathbf{g}}^z(t) S_{\mathbf{f}}^+(t); \\ &\quad \times C^{(a)}(\mathbf{m}, \mathbf{p}; t') \rangle\rangle_E, \quad (7) \end{aligned}$$

where

$$\mathcal{D}^{(a)} \equiv (e^{-a}-1)S(S+1) + (e^{-a}+1)\frac{d}{da} - (e^{-a}-1)\frac{d^2}{da^2}. \quad (8)$$

The higher-order Green functions in the last term of Eq. (7) are expressed in terms of lower-order Green functions by the decoupling recipe³

$$\begin{aligned} \langle\langle S_{\mathbf{m}}^3 S_{\mathbf{p}}^+; C \rangle\rangle &\xrightarrow{\mathbf{m} \neq \mathbf{p}} \langle S^z \rangle \langle\langle S_{\mathbf{p}}^+; C \rangle\rangle \\ &\quad - \frac{\langle S^z \rangle}{2S^2} \langle S_{\mathbf{m}}^- S_{\mathbf{p}}^+ \rangle \langle\langle S_{\mathbf{m}}^+; C \rangle\rangle, \quad (9) \end{aligned}$$

whence

$$\begin{aligned} [E - \mu H] G_E^{(a)}(\mathbf{g}, \mathbf{m}, \mathbf{p}) &= -\frac{1}{2\pi} [\delta_{\mathbf{g}, \mathbf{l}} \mathcal{D}^{(a)} \Lambda^{(a)}(\mathbf{m}-\mathbf{g}) - \delta_{\mathbf{g}, \mathbf{m}} \langle S_{\mathbf{m}}^+ \exp(as_{\mathbf{p}}^z) S_{\mathbf{p}}^- \rangle] \\ &\quad + 2 \langle S^z \rangle \sum_{\mathbf{f}} J(\mathbf{g}-\mathbf{f}) [G_E^{(a)}(\mathbf{g}, \mathbf{m}, \mathbf{p}) - G_E^{(a)}(\mathbf{f}, \mathbf{m}, \mathbf{p})] \\ &\quad - \frac{\langle S^z \rangle}{S^2} \sum_{\mathbf{f}} J(\mathbf{g}-\mathbf{f}) [\langle S_{\mathbf{f}}^- S_{\mathbf{g}}^+ \rangle G_E^{(a)}(\mathbf{f}, \mathbf{m}, \mathbf{p}) \\ &\quad - \langle S_{\mathbf{g}}^- S_{\mathbf{f}}^+ \rangle G_E^{(a)}(\mathbf{g}, \mathbf{m}, \mathbf{p})]. \quad (10) \end{aligned}$$

In order to solve Eq. (10) we define spatial Fourier transforms of all quantities:

$$J(\mathbf{k}) = J(-\mathbf{k}) \equiv \sum_{\mathbf{g}-\mathbf{f}} J(\mathbf{g}-\mathbf{f}) e^{i\mathbf{k} \cdot (\mathbf{g}-\mathbf{f})}, \quad (11)$$

$$\Lambda^{(a)}(\mathbf{k}) \equiv \sum_{\mathbf{m}-\mathbf{g}} \Lambda^{(a)}(\mathbf{m}-\mathbf{g}) e^{i\mathbf{k} \cdot (\mathbf{m}-\mathbf{g})}, \quad (12)$$

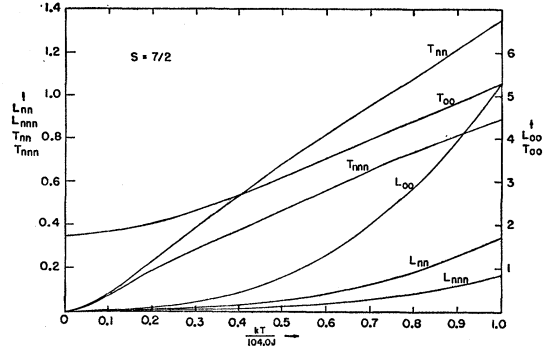


FIG. 4. Correlation functions for $S = \frac{7}{2}$, in the face-centered cubic lattice.

$$\begin{aligned} \langle S_{\mathbf{m}}^+ \exp(as_{\mathbf{p}}^z) S_{\mathbf{p}}^- \rangle_{\mathbf{k}} &\equiv \sum_{\mathbf{m}-\mathbf{p}} \langle S_{\mathbf{m}}^+ \exp(as_{\mathbf{p}}^z) S_{\mathbf{p}}^- \rangle e^{i\mathbf{k} \cdot (\mathbf{p}-\mathbf{m})}, \quad (13) \end{aligned}$$

$$\psi(\mathbf{k}, a) \equiv \sum_{\mathbf{m}-\mathbf{p}} \langle \exp(as_{\mathbf{p}}^z) S_{\mathbf{p}}^- S_{\mathbf{m}}^+ \rangle e^{i\mathbf{k} \cdot (\mathbf{p}-\mathbf{m})}, \quad (14)$$

and

$$G_E^{(a)}(\mathbf{k}_1, \mathbf{k}_2) \equiv \sum_{\mathbf{m}, \mathbf{p}} G_E^{(a)}(\mathbf{g}, \mathbf{m}, \mathbf{p}) e^{-i\mathbf{k}_1 \cdot (\mathbf{g}-\mathbf{m}) - i\mathbf{k}_2 \cdot (\mathbf{g}-\mathbf{p})}. \quad (15)$$

In terms of these quantities, the equation of motion (10) implies

$$[E - E(\mathbf{k}_1 + \mathbf{k}_2)] G_E^{(a)}(\mathbf{k}_1, \mathbf{k}_2) = \frac{1}{2\pi} [\mathcal{D}^{(a)} \Lambda^{(a)}(\mathbf{k}_1) - \langle S_{\mathbf{m}}^+ \exp(as_{\mathbf{p}}^z) S_{\mathbf{p}}^- \rangle_{\mathbf{k}_2}], \quad (16)$$

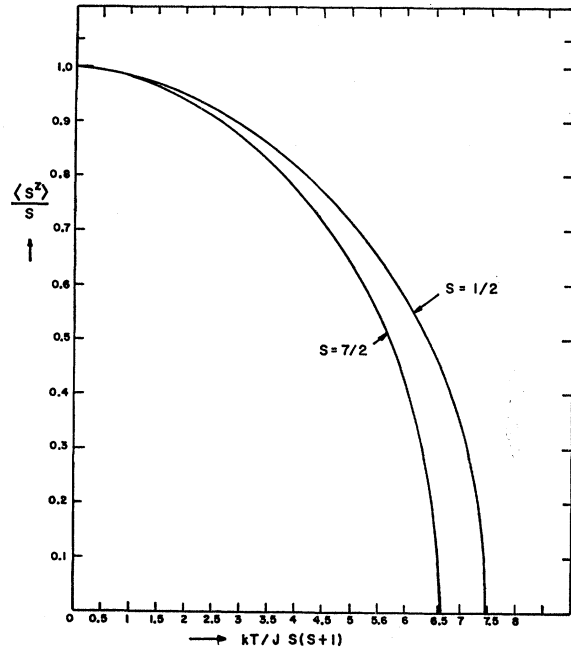


FIG. 5. Magnetization as a function of temperature, for various spin values, in the face-centered cubic lattice.

where

$$E(\mathbf{k}) = \mu H + 2\langle S^z \rangle J(0, \mathbf{k}) + \frac{\langle S^z \rangle}{NS^2} \sum_{\mathbf{k}'} J(\mathbf{k}', \mathbf{k}' - \mathbf{k}) \psi(\mathbf{k}', 0) \quad (17)$$

and

$$J(\mathbf{k}_1, \mathbf{k}_2) \equiv J(\mathbf{k}_1) - J(\mathbf{k}_2). \quad (18)$$

The quantities $\langle S_m^+ \exp(a S_p^z) S_p^- \rangle_{\mathbf{k}}$ and $\psi(\mathbf{k}, a)$ are related, for permuting the factors in the trace,

$$\begin{aligned} \langle S_m^+ \exp(a S_p^z) S_p^- \rangle_{\mathbf{k}} &= \langle \exp(a S_p^z) S_p^- \exp(-\beta \mathcal{H}) S_m^+ \exp(\beta \mathcal{H}) \rangle_{\mathbf{k}} \\ &= \langle \exp(a S_p^z) S_p^- S_m^+ (i\hbar\beta) \rangle_{\mathbf{k}}. \end{aligned} \quad (19)$$

However, if Eq. (26) of Ref. 3 is substituted into Eq. (4) of that reference, taking $t = i\hbar\beta$, we obtain from the last form of Eq. (19)

$$\langle S_m^+ \exp(a S_p^z) S_p^- \rangle_{\mathbf{k}} = e^{\beta E(\mathbf{k})} \psi(\mathbf{k}, a). \quad (20)$$

It was further shown in Ref. 3 that

$$\psi(\mathbf{k}, a) = \Theta(a) \phi(\mathbf{k}) \quad (21)$$

where

$$\phi(\mathbf{k}) \equiv [e^{\beta E(\mathbf{k})} - 1]^{-1}, \quad (22)$$

$$\Theta(a) \equiv \mathcal{D}^{(a)} \Omega(a), \quad (23)$$

$$\Omega(a) \equiv \langle e^{a S^z} \rangle. \quad (24)$$

Thus the equation for $G_E^{(a)}(k_1, k_2)$ can be rewritten

$$\begin{aligned} [E - E(\mathbf{k}_1 + \mathbf{k}_2)] G_E^{(a)}(\mathbf{k}_1, \mathbf{k}_2) &= -\frac{1}{2\pi} [\mathcal{D}^{(a)} \Lambda^{(a)}(\mathbf{k}_1) - \Theta(a) \phi(\mathbf{k}_2) e^{\beta E(\mathbf{k}_2)}] \end{aligned} \quad (25)$$

and, noting that $\Theta(a) = 2\langle S^z \rangle$, the equation for $E(k)$ simplifies to

$$E(\mathbf{k}) = \mu H + 2\langle S^z \rangle J(0, \mathbf{k}) + \frac{2\langle S^z \rangle^2}{NS^2} \sum_{\mathbf{k}'} \phi(\mathbf{k}') J(\mathbf{k}', \mathbf{k}' - \mathbf{k}). \quad (26)$$

The discontinuity of the Green function across the real axis (in the E plane) determines the correlation function $\langle S_m^z \exp(a S_p^z) S_p^- S_g^+ \rangle$, or its Fourier transform:

$$\begin{aligned} \mathcal{E}^{(a)}(\mathbf{k}_1, \mathbf{k}_2) &= \sum_{\mathbf{m}, \mathbf{p}} \langle S_m^z \exp(a S_p^z) S_p^- S_g^+ \rangle \\ &\quad \times e^{-i\mathbf{k}_1 \cdot (\mathbf{g} - \mathbf{m}) - i\mathbf{k}_2 \cdot (\mathbf{g} - \mathbf{p})}. \end{aligned} \quad (27)$$

The result follows directly from Eq. (25);

$$\mathcal{E}^{(a)}(\mathbf{k}_1, \mathbf{k}_2) = [\mathcal{D}^{(a)} \Lambda^{(a)}(\mathbf{k}_1) - \Theta(a) \phi(\mathbf{k}_2) e^{\beta E(\mathbf{k}_2)}] \times \phi(\mathbf{k}_1 + \mathbf{k}_2) \quad (28)$$

or

$$\begin{aligned} \langle S_m^z \exp(a S_p^z) S_p^- S_g^+ \rangle &= \frac{1}{N^2} \sum_{\mathbf{k}_1, \mathbf{k}_2} [\mathcal{D}^{(a)} \Lambda^{(a)}(\mathbf{k}_1) \\ &\quad - \Theta(a) \phi(\mathbf{k}_2) e^{\beta E(\mathbf{k}_2)}] \phi(\mathbf{k}_1 + \mathbf{k}_2) e^{i\mathbf{k}_1 \cdot (\mathbf{g} - \mathbf{m}) + i\mathbf{k}_2 \cdot (\mathbf{g} - \mathbf{p})}. \end{aligned} \quad (29)$$

We are interested in the special case $g = p$:

$$\begin{aligned} \langle S_m^z \exp(a S_p^z) S_p^- S_p^+ \rangle &= \frac{1}{N^2} \sum_{\mathbf{k}_1, \mathbf{k}_2} [\mathcal{D}^{(a)} \Lambda^{(a)}(\mathbf{k}_1) \\ &\quad - \Theta(a) \phi(\mathbf{k}_2) e^{\beta E(\mathbf{k}_2)}] \phi(\mathbf{k}_1 + \mathbf{k}_2) e^{i\mathbf{k}_1 \cdot (\mathbf{p} - \mathbf{m})}. \end{aligned} \quad (30)$$

In the first term in the summation we make the change of variables $\mathbf{k}_2' = \mathbf{k}_1 + \mathbf{k}_2$; in the second term we let $\mathbf{k}_1' = \mathbf{k}_1 + \mathbf{k}_2$; and in the left-hand member we use the identity

$$S_p^- S_p^+ = S(S+1) - S_p^z - (S_p^z)^2 \quad (31)$$

by means of which we can rewrite Eq. (30) in the form

$$\begin{aligned} \left[S(S+1) - \frac{d}{da} - \frac{d^2}{da^2} \right] \Lambda^{(a)}(\mathbf{m} - \mathbf{p}) &= \Phi \mathcal{D}^{(a)} \Lambda^{(a)}(\mathbf{m} - \mathbf{p}) - \Theta(a) [\Phi \delta_{\mathbf{m}, \mathbf{p}} + \phi^2(\mathbf{m} - \mathbf{p})], \end{aligned} \quad (32)$$

where

$$\Phi \equiv \frac{1}{N} \sum_{\mathbf{k}} \phi(\mathbf{k}) \quad (33)$$

and

$$\phi(\mathbf{m} - \mathbf{p}) \equiv \frac{1}{N} \sum_{\mathbf{k}} \phi(\mathbf{k}) e^{-i\mathbf{k} \cdot (\mathbf{m} - \mathbf{p})}. \quad (34)$$

Finally, by inserting the definition (8) for $\mathcal{D}(a)$, Eq. (32) takes the form

$$\begin{aligned} \left[\frac{d^2}{da^2} + \frac{(1+\Phi)e^a + \Phi}{(1+\Phi)e^a - \Phi} \frac{d}{da} - S(S+1) \right] \Lambda^{(a)}(\mathbf{m} - \mathbf{p}) &= \Theta(a) \frac{\Phi \delta_{\mathbf{m}, \mathbf{p}} + \phi^2(\mathbf{m} - \mathbf{p})}{1 - \Phi(e^{-a} - 1)}. \end{aligned} \quad (35)$$

The solution of this differential equation is

$$\begin{aligned} \Lambda^{(a)}(\mathbf{m} - \mathbf{p}) &= \langle S^z \rangle \Omega(a) + \left[\frac{d\Omega(a)}{da} - \langle S^z \rangle \Omega(a) \right] \\ &\quad \times \frac{\Phi \delta_{\mathbf{m}, \mathbf{p}} + \phi^2(\mathbf{m} - \mathbf{p})}{\Phi(1+\Phi)}. \end{aligned} \quad (36)$$

That this is, in fact, a solution can be corroborated by first noting that the first term of Eq. (36) is a solution of the homogeneous equation, as was demonstrated in Ref. 3. Introducing the second term of Eq. (36) into (35) and multiplying both sides by $[(1+\Phi)e^a - \Phi]$ we find a third-order differential equation for $\Omega(a)$ which is recognized as simply the derivative (d/da) of the homogeneous equation satisfied by $\Omega(a)$. We further note that the second term in Eq. (36) vanishes as $a \rightarrow 0$, so that $\Lambda^{(0)}(\mathbf{m} - \mathbf{p}) = \langle S^z \rangle$, as required by the definition (3). This, plus the boundary conditions imposed upon (and satisfied by) $\Omega(a)$ in Ref. 3, determine Eq. (36) as the appropriate solution of the differential Eq. (35).

We find $\langle S_m^z S_p^z \rangle$ by differentiation of $\Lambda^{(a)}(m-p)$:

$$\langle S_m^z S_p^z \rangle = \lim_{a \rightarrow 0} \frac{d}{da} \Lambda^{(a)}(m-p) = \langle S^z \rangle^2 + \frac{\langle (S^z)^2 \rangle - \langle S^z \rangle^2}{\Phi(1+\Phi)} \times [\Phi \delta_{m,p} + \phi^2(m-p)]. \quad (37)$$

In Ref. 3 it was shown that

$$\Omega(a) = \frac{\Phi^{2S+1} e^{-Sa} - (1+\Phi)^{2S+1} e^{(S+1)a}}{[\Phi^{2S+1} - (1+\Phi)^{2S+1}][\Phi(1+\Phi) - e^a]} \quad (38)$$

and

$$\langle S^z \rangle = \frac{d\Omega(0)}{da} = \frac{(S-\Phi)(1+\Phi)^{2S+1} + (S+1+\Phi)\Phi^{2S+1}}{(1+\Phi)^{2S+1} - \Phi^{2S+1}}, \quad (39)$$

whence

$$\langle (S^z)^2 \rangle = \frac{d^2\Omega(0)}{da^2} = S(S+1) - \langle S^z \rangle(1+2\Phi). \quad (40)$$

Equation (37) can then be written as

$$\langle S_m^z S_p^z \rangle - \langle S^z \rangle^2 = \left\{ 1 - \frac{(2S+1)^2 \Phi^{2S}(1+\Phi)^{2S}}{[(1+\Phi)^{2S+1} - \Phi^{2S+1}]^2} \right\} \times \{\Phi \delta_{m,p} + \phi^2(m-p)\}. \quad (41)$$

3. THE CORRELATION FUNCTION

To summarize, the longitudinal correlation function

$$\psi_z(m-p) \equiv \langle S_m^z S_p^z \rangle - \langle S^z \rangle^2 \quad (42)$$

is given by

$$\psi_z(m-p) = \psi_z^0 \frac{\Phi \delta_{m,p} + \phi^2(m-p)}{\Phi(1+\Phi)}, \quad (43)$$

where ψ_z^0 is the value of $\psi_z(m-m)$:

$$\psi_z^0 = \langle (S^z)^2 \rangle - \langle S^z \rangle^2 = \Phi(1+\Phi) - \frac{(2S+1)^2 \Phi^{2S+1} (1+\Phi)^{2S+1}}{[(1+\Phi)^{2S+1} - \Phi^{2S+1}]^2}. \quad (44)$$

The quantity Φ is the average value of the quasiboson occupation number $\phi(\mathbf{k})$:

$$\Phi = \frac{1}{N} \sum_{\mathbf{k}} \phi(\mathbf{k}) \quad (45)$$

$$\phi(\mathbf{k}) = [e^{\beta E(\mathbf{k})} - 1]^{-1} \quad (46)$$

and $\phi(m-p)$ is the Fourier transform of $\phi(\mathbf{k})$:

$$\phi(m-p) = \frac{1}{N} \sum_{\mathbf{k}} \phi(\mathbf{k}) e^{-i\mathbf{k} \cdot (m-p)} = \phi(p-m). \quad (47)$$

The identification of ψ_z^0 as the value of $\psi_z(m-m)$

follows from noting that $\phi(m-m) = \Phi$, so that the fraction in Eq. (43) reduces to unity when $p=m$. We also note that ψ_z^0 does reduce to $\langle (S^z)^2 \rangle - \langle S^z \rangle^2$ [as indicated in Eq. (44), following from Eq. (37) with $p=m$], as it should.

The Fourier transform of the correlation function

$$\psi_z(\mathbf{k}) = \sum_{m-p} \psi_z(m-p) e^{i\mathbf{k} \cdot (m-p)} \quad (47)$$

is then given by

$$\psi_z(\mathbf{k}) = \psi_z^0 [\Phi + \sum_{\mathbf{k}'} \phi(\mathbf{k}') \phi(\mathbf{k}-\mathbf{k}')]/\Phi(1+\Phi). \quad (48)$$

Combining these results with the transverse correlation function,³ we find the energy per ion as a function of Φ :

$$u = \langle \mathcal{H} \rangle / N = -\mu H \langle S^z \rangle - J(0) \langle S^z \rangle^2 - \frac{2}{N} \langle S^z \rangle \sum_{\mathbf{k}} J(\mathbf{k}) \phi(\mathbf{k}) - \frac{S(S+1) - \langle S^z \rangle(1+2\Phi + \langle S^z \rangle)}{\Phi(1+\Phi)} \frac{1}{N^2} \times \sum_{\mathbf{k}, \mathbf{k}'} \phi(\mathbf{k}) \phi(\mathbf{k}') J(\mathbf{k}-\mathbf{k}'), \quad (49)$$

where $\langle S^z \rangle$ is known in terms of Φ [see Eq. (39)] and where, of course, the specific heat c_p can be found by differentiation of u with respect to T .

4. LOW-TEMPERATURE BEHAVIOR

The function Φ has been analyzed in Ref. 3 and at low temperatures

$$\Phi = \tilde{\zeta}\left(\frac{3}{2}\right) \tau^{3/2} + \frac{3\pi}{4} \nu \tilde{\zeta}\left(\frac{5}{2}\right) \tau^{5/2} + \pi^2 \nu^2 \omega \tilde{\zeta}\left(\frac{7}{2}\right) \tau^{7/2} + (3/2S) \pi \nu \tilde{\zeta}\left(\frac{3}{2}\right) \tilde{\zeta}\left(\frac{5}{2}\right) \tau^4 + O(\tau^{9/2}), \quad (51)$$

where τ is of the order of ST/T_c , being given by the relation

$$\tau = [(4\pi/3)\beta J(0)S\nu]^{-1}. \quad (52)$$

ω and ν are constants defined in Ref. 3, and

$$\tilde{\zeta}(m) \equiv \sum_{n=1}^{\infty} n^{-m} \exp(-n\beta\mu H). \quad (53)$$

As $\Phi \ll 1$, we can expand Eq. (44) in powers of Φ :

$$\frac{\psi_z^0}{\Phi(1+\Phi)} = 1 - (2S+1)^2 \Phi^{2S} + (2S+1)^2 (2S+2) \Phi^{2S+1} + O(\Phi^{2S+3}). \quad (54)$$

The expansion of $\psi_z(m-p)$ therefore requires only the calculation of $\Phi(m-p)$ in a temperature series. By standard expansion methods as used in the theory of

spin waves, we find for the simple cubic lattice:

$$\begin{aligned} \phi(r) = & Z_{3/2}(0)\tau^{3/2} + \frac{\pi}{4}\tau^{5/2}[3Z_{5/2}(0) - Z_{5/2}(1) + \frac{1}{12}Z_{5/2}(2)] \\ & + \frac{\pi^2\nu^2}{16}\tau^{7/2}\left[\frac{33}{2}Z_{7/2}(0) - \frac{35}{3}Z_{7/2}(1) + \frac{11}{4}Z_{7/2}(2) \right. \\ & \left. - \frac{11}{60}Z_{7/2}(3) + \frac{1}{288}Z_{7/2}(4)\right] + Z_{3/2}(0)\xi\left(\frac{5}{2}\right)\frac{3\pi}{2S}\tau^4 \\ & + O(\tau^{9/2}Z_{9/2}(0)), \quad (55) \end{aligned}$$

where

$$\begin{aligned} Z_m(x) = & \left(\frac{4\pi}{f}\right)^x \sum_{n=1}^{\infty} n^{-m-x} \exp\left(-n\beta\mu H - \frac{r^2\pi\tau}{fn}\right) \tau^{x/2} x, \quad (56) \\ f = & 1 - \frac{\pi}{S}\xi\left(\frac{5}{2}\right)\tau^{5/2} - O(\tau^{7/2}), \quad (57) \end{aligned}$$

and where

$$ra = |\mathbf{m} - \mathbf{p}| \quad (58)$$

(a is the lattice constant).

The corresponding expressions for $\phi(\mathbf{m} - \mathbf{p})$ for other cubic lattices are similar in structure and will not be given here. In the limit of small r (i.e., $|\mathbf{m} - \mathbf{p}|$ of the order of a few lattice constants) the coefficients $Z_m(x)$ can be expanded in powers of τr^2 , giving

$$\begin{aligned} Z_m(x) = & \left(\frac{2\pi}{3f}\right)^x (\tau r^2)^x \left[\xi(m+x) - \frac{\pi}{f} \tau r^2 \xi(m+x+1) \right. \\ & \left. + \frac{1}{2}\left(\frac{\pi}{f}\right)^2 (\tau r^2)^2 \xi(m+x+2) + O(\tau^3 r^6) \right] \quad (59) \end{aligned}$$

and correspondingly

$$\begin{aligned} \psi_s(\mathbf{m} - \mathbf{p}) = & [1 - (2S+1)^2\Phi^{2S} + 2(S+1)(2S+1)^2\Phi^{2S+1} \\ & + O(\Phi^{2S+3})] \left[\Phi\delta_{m,p} + \left\{ \Phi - r^2 \left(\pi\xi\left(\frac{5}{2}\right)\tau^{7/2} \right. \right. \right. \\ & \left. \left. + \frac{7\pi^2}{4}\tau^{7/2}\xi\left(\frac{7}{2}\right) \right) + \frac{1}{2}r^4\pi^2\tau^{7/2}\xi\left(\frac{7}{2}\right) + \dots \right\} \right]. \quad (60) \end{aligned}$$

When $|\mathbf{m} - \mathbf{p}|$ is larger than a few lattice constants, the coefficient $Z_m(x)$ is best calculated by the Euler-Machlaurin sum formula. In the limit $\beta\mu H \ll 1$ and $r \gg 1$ the result is particularly simple:

$$Z_m(x) \simeq \left(\frac{4\pi}{f}\right)^x (\tau r^2)^x \pi^{1/2} (-1)^{m+x-3/2} \left(\frac{d}{d\alpha}\right)^{m+x-3/2} \frac{1}{\sqrt{\alpha}}, \quad (61)$$

where $\alpha \equiv r^2\pi\tau/f$, whence

$$\phi(\mathbf{m} - \mathbf{p}) \simeq \tau[r^{-1} + \frac{1}{4}r^{-3}], \quad (r \gg 1, \beta\mu H \ll 1). \quad (62)$$

5. CONCLUSION

In the homogeneous decoupling approximation here employed the longitudinal correlation function depends on the spatial separation of the spins through a single factor $\phi(\mathbf{m} - \mathbf{p})$. This factor is the Fourier transform of the quasi-spin-wave occupation number $[\exp(\beta E(\mathbf{k}) - 1)]^{-1}$. The transverse correlation function $\langle S_m^x S_p^x \rangle$ is also simply $\langle S^z \rangle \phi(\mathbf{m} - \mathbf{p}) + \frac{1}{2} \langle S^z \rangle \delta_{m,p}$.

The longitudinal correlation function reduces properly to $\langle (S^z)^2 \rangle - \langle S^z \rangle^2$ when $\mathbf{m} = \mathbf{p}$; its asymptotic limit for large $|\mathbf{m} - \mathbf{p}|$ is $\exp(-2\kappa \cdot (\mathbf{m} - \mathbf{p})) / |\mathbf{m} - \mathbf{p}|^2$ whereas that of the transverse correlation function is

$$\exp(-\kappa \cdot (\mathbf{m} - \mathbf{p})) / |\mathbf{m} - \mathbf{p}|.$$

Near the Curie temperature the decoupling approximation is inadequate, and we accordingly find that the longitudinal and transverse correlation functions do not become isotropic at T_c . Thus, from Eq. (41) we easily find

$$\frac{\langle S_m^z S_p^z \rangle}{\langle S_m^x S_p^x \rangle} = \frac{\langle S_m^z S_p^z \rangle}{\langle S^z \rangle \phi(\mathbf{m} - \mathbf{p})^T \rightarrow T_c} = \frac{\phi(\mathbf{m} - \mathbf{p})}{\Phi}. \quad (63)$$

Now $\phi(\mathbf{m} - \mathbf{p}) \rightarrow 0$ as $|\mathbf{m} - \mathbf{p}| \rightarrow \infty$, so that the correlation function becomes increasingly anisotropic for large distances, at T_c . Presumably the analysis is a reasonable approximation at temperatures above the spin wave region, but it is clearly not adequate near T_c .

At low temperatures the correlation function agrees with the first order Born approximation renormalization of spin wave theory, as given by Dyson,¹¹ for all spins other than $S = \frac{1}{2}$.

The transverse correlation functions (T_{00} , T_{nn} , and T_{nnn}) and longitudinal correlation functions (L_{00} , L_{nn} and L_{nnn}) corresponding to the self-correlation, nearest-neighbor spins, and next-nearest-neighbor spins, are shown for several spin values in a face-centered cubic lattice in Fig. 1-4. The magnetization as a function of temperature, predicted by the Green function analysis of Ref. 3, is also shown in Fig. 5.

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¹¹ F. J. Dyson, Phys. Rev. **102**, 1217, 1230 (1956).