PHYSICAL REVIEW VOLUME 159, NUMBER 2 10 JULY 1967

Green's-Function Method for Antiferromagnetism*

K. H. LEE AND S. H. LIU

Institute for Atomic Research and Department of Physics, Iowa State University, Ames, Iowa

(Received 15 February 1967)

The statistical mechanics of simple antiferromagnets is studied over the entire temperature range by methods similar to those of Callen and Liu for ferromagnets. It is shown that the energy spectrum and the sublattice magnetization as functions of temperature are the solutions of a set of coupled equations. The equations are solved numerically for $S=\frac{1}{2}$, and the results compare very well with existing theories in both high- and low-temperature limits. The Néel temperature is also calculated for general spin value. Compared with other theoretical estimates, the Néel temperature obtained this way is appreciably higher for $S=\frac{1}{2}$ but approaches agreement in the large-spin limit. The transverse correlation functions of two spins and the correlation length for short-range order above the transition temperature are also calculated. The longitudinal correlation functions of two spins are calculated by extending Liu's method for ferromagnetism to antiferromagnetism. A calculation is carried out for $S=\frac{1}{2}$ using the Callen decoupling scheme for three-spin Green's functions. It is shown that the longitudinal correlation functions are related to the first-order response of the system to a space- and time-varying field. Thus, to calculate the longitudinal correlation functions, a perturbation calculation to first order is necessary and Callen's original decoupling scheme for the three-spin Green's functions has to be extended for Green's functions with perturbation. A satisfactory extension of the Callen decoupling scheme to the first-order equation of motion for Green's functions is found. The extended decoupling scheme leads to a number of desirable results: Rotational invariance of the correlation functions and susceptibilities at and above the Néel temperature and the validity of the sum rule for spin operators over the same temperature range. The parallel susceptibility is also calculated, as well as the internal energy and the specific heat above the Néel temperature. The ground-state energy turns out to be slightly lower than the values predicted by other theories. Numerical calculations of parallel and perpendicular susceptibilities for $S=\frac{1}{2}$ are carried out and the results are presented.

INTRODUCTION

A STATISTICAL theory for antiferromagnetism usually starts from a specific molecular model, and does not inquire very much into its justification. One frequently used model is the Heisenberg model, which consists of a regular array of localized spins that are coupled together by exchange interaction. It is generally hoped that the statistical theory of the model will explain the observed behavior of antiferromagnetic substances, at least semiquantitatively.

The crudest approximation for the solution of the model is provided by the so-called molecular-field theory.[1] This theory gives several successful qualitative predictions, including the existence of the transition temperature, specific-heat anomaly, etc., but it has many weak points, such as the failure of the analysis at very low temperatures and the lack of short-range order above the transition temperature. The more refined Bethe-Peierls-Weiss approximation was first applied to antiferromagnetism by Li,[2] and this method gives short-range order above the Néel temperature but again fails at very low temperatures. General experience with cooperative phenomena suggests two ways of making better approximations: the high-temperature approximation and the low-temperature approximation. The $1/T$ expansion method developed by Opechowski[3] is an analog of the Bethe-Kirkwood expansion method for a regular assembly and belongs to the first category. The most important method for low temperatures is the spin-wave theory.[4] Both approximations rely on series expansions and they suffer from the drawback that their applicability is limited to a certain range of temperature where a small number of terms is sufficient. They have not been able to discuss the existence of phase transitions since it has not been possible to determine the general term in any of these expansions.

Tyablikov[5] first employed the technique of double-time temperature-dependent Green's functions to Heisenberg ferromagnet with spin $\frac{1}{2}$. Extension of the theory to higher spin has been achieved by Tahir-Kheli and ter Haar[6] and Callen.[7] The application of the theory to Heisenberg antiferromagnet has been studied by Pu,[8] Oguchi and Honma,[9] Lines,[10] Anderson and Callen,[11] and Lines and Jones.[12] The most important feature of the Green's function theory is that it treats the entire temperature range and agrees with the

---

* Work was performed in the Ames Laboratory of the U. S. Atomic Energy Commission. Contribution No. 2044.

[1] L. Néel, Ann. Phys. (Paris) **18**, 5 (1932); F. Bitter, Phys. Rev. 54, (1938); J. H. Van Vleck, J. Chem. Phys. **9**, 85 (1941).

[2] Y. Y. Li, Phys. Rev. **84**, 72 (1951).

[3] W. Opechowski, Physica **4**, 181 (1937).

[4] H. Bethe, Z. Physik **71**, 205 (1931); G. Heller and H. A. Kramers, Proc. Roy. Acad. Amsterdam **37**, 378 (1934); L. Hulthén, Proc. Amsterdam Acad. Sci. **39**, 190 (1936); P. W. (1952).
Anderson, Phys. Rev. **86**, 694 (1952); R. Kubo, *ibid.* **87**, 568

[5] S. V. Tyablikov, Ukr. Mat. Zh. **11**, 287 (1959).

[6] R. Tahir-Kheli and D. ter Haar, Phys. Rev. **127**, 88 (1962).

[7] H. B. Callen, Phys. Rev. **130**, 890 (1963).

[8] F.-C. Pu, Dokl. Akad. Nauk SSSR **130**, 1244 (1960); **131**, 546 (1960) [English transls.: Soviet Phys.—Doklady **5**, 128 (1960); **5**, 321 (1960)].

[9] T. Oguchi and A. Honma, J. Appl. Phys. **34**, 1153 (1963).

[10] M. E. Lines, Phys. Rev. **131**, 540 (1963); **135**, A1336 (1964); **139**, A1304 (1965).

[11] F. B. Anderson and H. B. Callen, Phys. Rev. **136**, A1068 (1964).

[12] M. E. Lines and E. D. Jones, Phys. Rev. **139**, A1313 (1965); **141**, 525 (1966).

spin-wave theory at very low temperatures and with statistical theory at very high temperatures. Thus there is hope that the result may not be too far from the correct implication of the Heisenberg model over the entire temperature range. The exact equation of motion for the Green's function involves higher-order Green's functions and must be linearized by some approximation so that it can be solved for the Green's function. The random-phase approximation is the simplest and most popular decoupling scheme for this purpose, but it results in a disagreement with the low-temperature theory.[7,10] Callen[7] and Anderson and Callen[11] proposed a more desirable decoupling scheme from heuristic physical grounds.

Although much progress has been made in the direction of Green's function theory, it has not been possible to make a complete study of the Heisenberg model in this way because an essential quantity, the longitudinal or zz correlation function of two spins, does not follow directly from the theory, z being the direction of preferred antiferromagnetic spin alignment. For the Heisenberg ferromagnet, this problem was solved by Liu[13] for general spin values using the random-phase approximation.

The present work treats the statistical mechanics of cubic antiferromagnets with isotropic, nearest-neighbor, Heisenberg interactions by use of Green's-function technique. In the first part of the paper, the energy spectrum, sublattice magnetization, and Néel temperature are studied for the general spin value in close analogy with Callen's work for ferromagnet.[7] In addition to these, the transverse correlation functions, perpendicular susceptibility, and correlation length for short-range order above the transition temperature are also calculated. In the later part, Liu's work for ferromagnet[13] is extended to antiferromagnet and an explicit calculation is carried out for $S=\frac{1}{2}$. The longitudinal correlation functions, parallel susceptibility, internal energy, and specific heat are calculated and discussed there.

THE GREEN'S-FUNCTION EQUATIONS

For a system with time-independent Hamiltonian H, the temperature-dependent causal Green's function involving two Bose operators A and B, is defined by

$$G_{AB}(t)=\langle\langle A(t)B(0)\rangle\rangle=-i\langle TA(t)B(0)\rangle, \tag{1}$$

where $A(t)$ is the Heisenberg operator at time t; T is the time-ordering operator of Dyson, which is defined in the usual way so that

$$TA(t)B(0)=\theta(t)A(t)B(0)+\theta(-t)B(0)A(t), \tag{2}$$

$\theta(t)$ being unity for positive t and zero for negative t; and the single bracket denotes an average with respect to the canonical density matrix of the system. The equation of motion for $G_{AB}(t)$ is derived easily from its definition and the equation of motion for $A(t)$ and it reads[14]

$$i(d/dt)G_{AB}(t)=\delta(t)\langle[A,B]\rangle-i\langle T[A(t),H(t)]B(0)\rangle. \tag{3}$$

One can define the Green's function for imaginary time argument τ by an analytic continuation which is equivalent to the replacement of t by $-i\tau$ and $G_{AB}(t)$ by $-iG_{AB}(\tau)$.[15] Then the equation of motion for $G_{AB}(\tau)$ reads

$$(d/d\tau)G_{AB}(\tau)=\delta(\tau)\langle[A,B]\rangle+\langle T[H(\tau),A(\tau)]B\rangle. \tag{4}$$

The model of the present problem is taken as a cubic, two-sublattice antiferromagnet with nearest-neighbor interaction. The Hamiltonian has the form

$$H=J\sum_i\sum_\delta \mathbf{S}_{1i}\cdot\mathbf{S}_{2,i+\delta}+J\sum_j\sum_\delta \mathbf{S}_{1,j+\delta}\cdot\mathbf{S}_{2j}, \tag{5}$$

where $1i$ refers to the lattice site i in the sublattice 1; and the index for the nearest neighbor of i is $i+\delta$. It is assumed that the spontaneous magnetization of the sublattice 1 is in the $+z$ direction and that of the sublattice 2 in the $-z$ direction. Following Callen,[7] the Green's function to be used in this section is defined by

$$\begin{aligned}G_{\lambda i,\mu j}{}^a(\tau)&=\langle\langle S_{\lambda i}{}^+;\,S_{\mu j}{}^-\exp(aS_{\mu j}{}^z)\rangle\rangle\\&=\langle TS_{\lambda i}{}^+(\tau)S_{\mu j}{}^-\exp(aS_{\mu j}{}^z)\rangle,\end{aligned} \tag{6}$$

where a in the exponent is a parameter and $S_{\lambda i}{}^+(\tau)$ is the imaginary time version of the Heisenberg operator $S_{\lambda i}{}^+(t)$ and λi and μj refer to lattice sites. The equation of motion for the Green's function can be written as

$$\begin{aligned}(d/d\tau)G_{\lambda i,\mu j}{}^a(\tau)&=\delta(\tau)\delta_{\lambda\mu}\delta_{ij}\theta_\lambda(a)\\&+2J\sum_\delta[\langle TS_{\lambda i}{}^+(\tau)S_{\nu,i+\delta}{}^z(\tau)S_{\mu j}{}^-\exp(aS_{\mu j}{}^z)\rangle\\&-\langle TS_{\lambda i}{}^z(\tau)S_{\nu,i+\delta}{}^+(\tau)S_{\mu j}{}^-\exp(aS_{\mu j}{}^z)\rangle],\quad \nu\neq\lambda\end{aligned} \tag{7}$$

where

$$\theta_\lambda(a)=\langle[S_{\lambda i}{}^+,\,S_{\lambda i}{}^-\exp(aS_{\lambda i}{}^z)]\rangle, \tag{8}$$

and it is independent of i because of translational invariance.

In order to solve Eq. (7) for $G_{\lambda i,\mu j}{}^a(\tau)$ it must be linearized. The simplest approximation is the random-phase approximation (RPA), in which the fluctuations of $S_{\lambda i}{}^z$ are ignored and the operator $S_{\lambda i}{}^z$ is replaced by its average value $\langle S_{\lambda i}{}^z\rangle$. To take into account the fluctuations of $S_{\lambda i}{}^z$ around its average, Callen[7] sug-

---

[13] S. H. Liu, Phys. Rev. **139**, A1522 (1965).

[14] D. N. Zubarev, Usp. Fiz. Nauk **71**, 71 (1960) [English transl.: Soviet Phys.—Usp. **3**, 320 (1960)].

[15] See for, example, L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (W. A. Benjamin, Inc., New York, 1962), Chap. 1.

gested a decoupling scheme for ferromagnet of the form

$$\langle\langle S_i^z S_j^+; B\rangle\rangle_{i\neq j}\rightarrow\langle S_i^z\rangle\langle\langle S_j^+; B\rangle\rangle - b\langle S_i^- S_j^+\rangle\langle\langle S_i^+; B\rangle\rangle, \quad (9)$$

and he chose b to be $\langle S_i^z\rangle/2S^2$ from physical grounds. If one tries to extend this decoupling scheme to an antiferromagnet, an ambiguity arises from the fact that there are two sublattices. If one writes $1i$ for i and $2j$ for j in Eq. (9), then there are two possible choices of b: $\langle S_{1i}^z\rangle/2S^2$ and $-\langle S_{2j}^z\rangle/2S^2$. Anderson and Callen[11] claim that the first choice results in an internal inconsistency. But without an external field, which is the case treated by the present work, there seems to be no internal inconsistency. We carried out calculations based on both, but only the latter is reported here. It turns out that the latter gives not only simpler algebra in perturbation calculation, but also the following *a posteriori* advantages over the first: continuity of a number of physical quantities at the Néel temperature, rotational invariance of correlation functions and susceptibilities at and above the Néel temperature, and the validity of the sum rule for spin operators over the same temperature range. After decoupling, Eq. (7) leads to

$$(d/d\tau)G_{\lambda i,\mu j}{}^a(\tau)=\delta(\tau)\delta_{\lambda\mu}\delta_{ij}\theta_\lambda(a) -2J\sum_\delta\{m_\lambda[1-\psi_{\nu(i+\delta),\lambda i}(0)/2S^2]G_{\nu(i+\delta),\mu j}{}^a(\tau) -m_\nu[1-\psi_{\lambda i,\nu(i+\delta)}(0)/2S^2]G_{\lambda i,\mu j}{}^a(\tau)\}, \qquad \nu\neq\lambda, \quad (10)$$

where

$$\psi_{\lambda i,\mu j}(a)=\langle S_{\lambda i}^-[\exp(aS_{\lambda i}^z)]S_{\mu j}^+\rangle, \quad (11)$$

and m_λ is written for $\langle S_{\lambda i}^z\rangle$.

Green's functions can be Fourier analyzed:

$$G_{\lambda i,\mu j}{}^a(\tau)=(2/N\beta)\sum_{\mathbf{k}}\sum_m G_{\lambda\mu}{}^a(\mathbf{k},m) \times\exp[i\mathbf{k}\cdot(\mathbf{R}_{\lambda i}-\mathbf{R}_{\mu j})-i\omega_m\tau],$$

$$\psi_{\lambda i,\mu j}(a)=(2/N)\sum_{\mathbf{k}}\psi_{\lambda\mu}(\mathbf{k},a)\exp[-i\mathbf{k}\cdot(\mathbf{R}_{\lambda i}-\mathbf{R}_{\mu j})], \quad (12)$$

where $\frac{1}{2}N$ is the number of magnetic ions in each sublattice, the sum over $\mathbf{k}$ runs over $\frac{1}{2}N$ points of the first zone in $\mathbf{k}$ space, m is an integer, $\omega_m=2\pi m/\beta$, and $\beta=(k_BT)^{-1}$. The equation for $G_{\lambda\mu}{}^a(\mathbf{k},m)$ is easily found to be

$$-i\omega_m G_{\lambda\mu}{}^a(\mathbf{k},m)=\delta_{\lambda\mu}\theta_\lambda(a)+2m_\nu J(0)[1-(NS^2)^{-1}\sum_{\mathbf{k}'}\gamma(\mathbf{k}')\psi_{\lambda\nu}(\mathbf{k}',0)]G_{\lambda\mu}{}^a(\mathbf{k},m) -2m_\lambda J(\mathbf{k})[1-(NS^2)^{-1}\sum_{\mathbf{k}'}\gamma(\mathbf{k}')\psi_{\nu\lambda}(\mathbf{k}',0)]G_{\nu\mu}{}^a(\mathbf{k},m), \qquad \nu\neq\lambda \quad (13)$$

where

$$J(\mathbf{k})=zJ\gamma(\mathbf{k})=J\sum_\delta\exp(i\mathbf{k}\cdot\boldsymbol{\delta}), \quad (14)$$

z being the number of nearest neighbors and $\boldsymbol{\delta}$ being a nearest-neighbor vector. If one defines $A_\lambda(\mathbf{k})$ by

$$A_\lambda(\mathbf{k})=2m_\lambda J(\mathbf{k})[1-(NS^2)^{-1}\sum_{\mathbf{k}'}\gamma(\mathbf{k}')\psi_{\nu\lambda}(\mathbf{k}',0)], \qquad \nu\neq\lambda \quad (15)$$

then from the symmetry of two sublattices one can show that

$$A(\mathbf{k})\equiv A_1(\mathbf{k})=\gamma(\mathbf{k})A(0)=-A_2(\mathbf{k}). \quad (16)$$

The solution of Eq. (13) is

$$G_{11}{}^a(\mathbf{k},m)=\tfrac{1}{2}[\theta_1(a)]\{[1+A(0)/\omega_{\mathbf{k}}](\omega_{\mathbf{k}}-i\omega_m)^{-1}+[1-A(0)/\omega_{\mathbf{k}}](-\omega_{\mathbf{k}}-i\omega_m)^{-1}\},$$

$$G_{12}{}^a(\mathbf{k},m)=[\theta_2(a)A(\mathbf{k})/2\omega_{\mathbf{k}}][(\omega_{\mathbf{k}}-i\omega_m)^{-1}-(-\omega_{\mathbf{k}}-i\omega_m)^{-1}],$$

$$G_{21}{}^a(\mathbf{k},m)=-[\theta_1(a)A(\mathbf{k})/2\omega_{\mathbf{k}}][(\omega_{\mathbf{k}}-i\omega_m)^{-1}-(-\omega_{\mathbf{k}}-i\omega_m)^{-1}],$$

$$G_{22}{}^a(\mathbf{k},m)=\tfrac{1}{2}[\theta_2(a)]\{[1-A(0)/\omega_{\mathbf{k}}](\omega_{\mathbf{k}}-i\omega_m)^{-1}+[1+A(0)/\omega_{\mathbf{k}}](-\omega_{\mathbf{k}}-i\omega_m)^{-1}\}, \quad (17)$$

with

$$\omega_{\mathbf{k}}=[A^2(0)-A^2(\mathbf{k})]^{1/2}=A(0)[1-\gamma^2(\mathbf{k})]^{1/2}. \quad (18)$$

If one uses the standard relationship

$$\beta^{-1}\sum_m\exp(-i\omega_m\tau)/(\pm\omega_{\mathbf{k}}-i\omega_m)=\exp(\mp\omega_{\mathbf{k}}\tau)/[1-\exp(\mp\beta\omega_{\mathbf{k}})], \quad (19)$$

Eq. (17) leads to

$$G_{11}{}^{a}(\mathbf{k},\tau)=\tfrac{1}{2}[\theta_1(a)]\left\{\left[1+\frac{A(0)}{\omega_{\mathrm{k}}}\right]\frac{\exp(-\omega_{\mathrm{k}}\tau)}{1-\exp(-\beta\omega_{\mathrm{k}})}+\left[1-\frac{A(0)}{\omega_{\mathrm{k}}}\right]\frac{\exp(\omega_{\mathrm{k}}\tau)}{1-\exp(\beta\omega_{\mathrm{k}})}\right\},$$

$$G_{12}{}^{a}(\mathbf{k},\tau)=\frac{\theta_2(a)A(\mathbf{k})}{2\omega_{\mathrm{k}}}\left[\frac{\exp(-\omega_{\mathrm{k}}\tau)}{1-\exp(-\beta\omega_{\mathrm{k}})}-\frac{\exp(\omega_{\mathrm{k}}\tau)}{1-\exp(\beta\omega_{\mathrm{k}})}\right],$$

$$G_{21}{}^{a}(\mathbf{k},\tau)=-\frac{\theta_1(a)A(\mathbf{k})}{2\omega_{\mathrm{k}}}\left[\frac{\exp(-\omega_{\mathrm{k}}\tau)}{1-\exp(-\beta\omega_{\mathrm{k}})}-\frac{\exp(\omega_{\mathrm{k}}\tau)}{1-\exp(\beta\omega_{\mathrm{k}})}\right],$$

$$G_{22}{}^{a}(\mathbf{k},\tau)=\tfrac{1}{2}[\theta_2(a)]\left\{\left[1-\frac{A(0)}{\omega_{\mathrm{k}}}\right]\frac{\exp(-\omega_{\mathrm{k}}\tau)}{1-\exp(-\beta\omega_{\mathrm{k}})}+\left[1+\frac{A(0)}{\omega_{\mathrm{k}}}\right]\frac{\exp(\omega_{\mathrm{k}}\tau)}{1-\exp(\beta\omega_{\mathrm{k}})}\right\}. \qquad (20)$$

It is evident that one can write

$$\psi_{\lambda\mu}(\mathbf{k},a)=G_{\mu\lambda}{}^{a}(\mathbf{k},\tau=0^-)=\theta_\lambda(a)\phi_{\lambda\mu}(\mathbf{k}), \qquad (21)$$

where

$$\phi_{11}(\mathbf{k})=\tfrac{1}{2}[-1+A(0)\omega_{\mathrm{k}}^{-1}\coth\tfrac{1}{2}\beta\omega_{\mathrm{k}}],$$
$$\phi_{21}(\mathbf{k})=-\phi_{12}(\mathbf{k})=\tfrac{1}{2}A(\mathbf{k})\omega_{\mathrm{k}}^{-1}\coth\tfrac{1}{2}\beta\omega_{\mathrm{k}},$$
$$\phi_{22}(\mathbf{k})=\tfrac{1}{2}[-1-A(0)\omega_{\mathrm{k}}^{-1}\coth\tfrac{1}{2}\beta\omega_{\mathrm{k}}]. \qquad (22)$$

One also observes that

$$\langle G_{\lambda\lambda}{}^{0}(\mathbf{k},\tau=0^{\mp})\rangle_{\mathrm{k}}\equiv(2/N)\sum_{\mathrm{k}}G_{\lambda\lambda}{}^{0}(\mathbf{k},\tau=0^{\mp})$$
$$=S(S+1)-\langle(S_{\lambda i}{}^{z})^2\rangle\mp m_\lambda$$
$$=\tfrac{1}{2}\mp m_\lambda \quad\text{for}\quad S=\tfrac{1}{2}. \qquad (23)$$

Thus, for $S=\frac{1}{2}$,

$$(\tfrac{1}{2}N)\langle G_{11}{}^{0}(\mathbf{k},0^-)\rangle_{\mathrm{k}} \quad\text{and}\quad (\tfrac{1}{2}N)\langle G_{22}{}^{0}(\mathbf{k},0^+)\rangle_{\mathrm{k}}$$

can be interpreted as the total number of spin reversals in each sublattice. Then $G_{11}{}^{0}(\mathbf{k},0^-)$ and $G_{22}{}^{0}(\mathbf{k},0^+)$ are the occupation numbers of elementary excitation of wave vector **k** in each sublattice. For higher spin this interpretation is not so evident because of the term $\langle(S_{\lambda i}{}^{z})^2\rangle$. From symmetry one expects occupation numbers to be equal in pairs corresponding to the two sublattices, and in fact this is the case as one can easily see from Eq. (20).

Equations (8), (15), (18), (21), and (22) are the basic set of coupled equations for the theory. If one introduces a quantity

$$\Omega_\lambda(a)=\langle\exp(aS_{\lambda i}{}^{z})\rangle, \qquad (24)$$

and exploits the functional dependence of $\Omega_\lambda(a)$, $\theta_\lambda(a)$, and $\langle\psi_{\lambda\lambda}(\mathbf{k},a)\rangle_{\mathrm{k}}$ on the parameter a, then one finds that the formal relationship between these three quantities is exactly the same as that of ferromagnetic case as worked out by Callen.[7] For example, the sublattice magnetization m_λ is given by

$$m_\lambda=[(d/da)\Omega_\lambda(a)]_{a=0}$$
$$=\frac{(S-\phi_{\lambda\lambda})(1+\phi_{\lambda\lambda})^{2S+1}+(S+1+\phi_{\lambda\lambda})(\phi_{\lambda\lambda})^{2S+1}}{(1+\phi_{\lambda\lambda})^{2S+1}-(\phi_{\lambda\lambda})^{2S+1}} \qquad (25)$$

with

$$\phi_{\lambda\lambda}=\langle\phi_{\lambda\lambda}(\mathbf{k})\rangle_{\mathrm{k}}=(2/N)\sum_{\mathrm{k}}\phi_{\lambda\lambda}(\mathbf{k}). \qquad (26)$$

From Eq. (22) one observes that

$$\phi_{11}+\phi_{22}=-1,$$

and substituting this into Eq. (25) one finds $m_2=-m_1$ as expected from symmetry of two sublattices.

For $S=\frac{1}{2}$ it is not necessary to go through the above procedure with parameter a since Eq. (21) and the relation

$$\theta_1(0)=2m_1=-2m_2=-\theta_2(0) \qquad (27)$$

determine $\psi_{\lambda\lambda}(\mathbf{k},0)$ as a function of m_λ and Eq. (23) provides a requirement of self-consistency which determines m_λ.

ENERGY SPECTRUM, SUBLATTICE MAGNETIZATION, TRANSVERSE CORRELATION FUNCTIONS, AND NÉEL TEMPERATURE

If one introduces the Fourier transform of spin operator defined by

$$\mathbf{S}_{\lambda,\mathrm{k}}=\sum_i\mathbf{S}_{\lambda i}\exp(-i\mathbf{k}\cdot\mathbf{R}_{\lambda i}), \qquad (28)$$

then from Eq. (22) and the definitions for the involved quantities one easily gets

$$\langle S_{2,-\mathrm{k}}{}^{+}S_{2,\mathrm{k}}{}^{-}\rangle=\langle S_{1,-\mathrm{k}}{}^{-}S_{1,\mathrm{k}}{}^{+}\rangle$$
$$=\tfrac{1}{2}mN[A(0)\omega_{\mathrm{k}}^{-1}\coth\tfrac{1}{2}\beta\omega_{\mathrm{k}}-1],$$
$$\langle S_{2,-\mathrm{k}}{}^{+}S_{1,\mathrm{k}}{}^{-}\rangle=\langle S_{1,-\mathrm{k}}{}^{+}S_{2,\mathrm{k}}{}^{-}\rangle$$
$$=\langle S_{1,-\mathrm{k}}{}^{-}S_{2,\mathrm{k}}{}^{+}\rangle=\langle S_{2,-\mathrm{k}}{}^{-}S_{1,\mathrm{k}}{}^{+}\rangle$$
$$=-\tfrac{1}{2}mNA(\mathbf{k})\omega_{\mathrm{k}}^{-1}\coth\tfrac{1}{2}\beta\omega_{\mathrm{k}},$$
$$\langle S_{1,-\mathrm{k}}{}^{+}S_{1,\mathrm{k}}{}^{-}\rangle=\langle S_{2,-\mathrm{k}}{}^{-}S_{2,\mathrm{k}}{}^{+}\rangle$$
$$=\tfrac{1}{2}mN[A(0)\omega_{\mathrm{k}}^{-1}\coth\tfrac{1}{2}\beta\omega_{\mathrm{k}}+1], \qquad (29)$$

with m written for m_1. Then the transverse correlation function of two spins or $\langle S_{\lambda i}{}^{-}S_{\mu j}{}^{+}\rangle$ is given by the inverse Fourier transform of Eq. (29). The energy

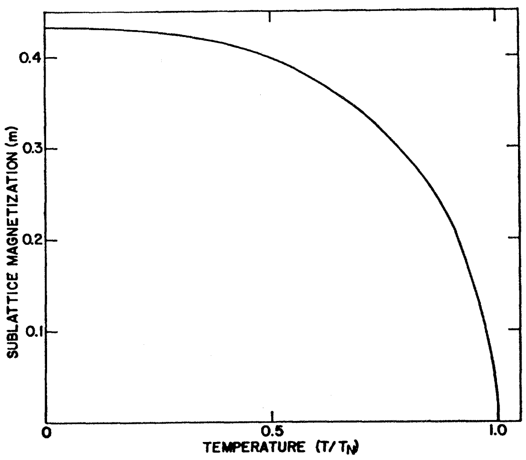

FIG. 1. Sublattice magnetization per spin for a simple cubic antiferromagnet ($S=\frac{1}{2}$).

spectrum in Eq. (18) turns out to be

$$\omega_{\mathrm{k}}=2SzJR[1-\gamma^2(\mathbf{k})]^{1/2}, \tag{30}$$

where

$$R=\frac{m}{S}\left[1+\frac{m}{NS^2}\sum_{\mathrm{k}}\frac{\gamma^2(\mathbf{k})}{[1-\gamma^2(\mathbf{k})]^{1/2}}\coth\tfrac{1}{2}\beta\omega_{\mathrm{k}}\right] \tag{31}$$

is the renormalization factor for energy spectrum. $R=m/S$ is the result of decoupling by RPA while $R=1$ corresponds to noninteracting spin-wave theory.

Thus, for arbitrary spin S, temperature T, and nearest-neighbor interaction J, one may determine the energy spectrum ω_{k}, the sublattice magnetization per spin m, and the transverse correlation function of two spins completely via a set of coupled equations: Eqs. (25), (29), (30), and (31). For $S=\frac{1}{2}$, numerical calculations of m and R have been carried out and the results for simple cubic (sc) structure are shown in Figs. 1 and 2. The results for bcc are very similar.

By a procedure similar to that for the ferromagnet,[7] the transition temperature can be determined and the discussion of properties at the transition temperature and very low temperatures is possible.[11] For example, the Néel temperature at which m vanishes is determined by

$$(k_BT_N/2zJS)$$
$$=[(S+1)/3I_0][1+(S+1)(I_0-1)/3SI_0], \tag{32}$$

with

$$I_0=2N^{-1}\sum_{\mathrm{k}}[1-\gamma^2(\mathbf{k})]^{-1}$$
$$\approx 1.516 \text{ for sc; } 1.393 \text{ for bcc.} \tag{33}$$

For the right-hand side of Eq. (32) the molecular-field theory gives $(S+1)/3$ while the RPA result[10] is $(S+1)/3I_0$. At very low temperatures it is possible to expand m and R in powers of T, and one finds that the deviation between the present theory and the interacting spin-wave theory[16,17] appears in T^2 terms and becomes very small as S increases. Thus, it turns out that the agreement between two theories becomes better with increasing S.

PERPENDICULAR SUSCEPTIBILITY

Using the Kubo method for the linear-response function,[18] one can write the perpendicular susceptibility of the system in terms of spin-correlation functions as

$$\chi_{\perp}=(g\mu_B)^2\int_0^{\beta}\langle T\sum_{\lambda i}S_{\lambda i}^x(\tau)\sum_{\mu j}S_{\mu j}^x(0)\rangle d\tau, \tag{34}$$

where g is the Landé g factor and μ_B is the Bohr magneton. If one uses Eq. (29), Eq. (34) leads to

$$\chi_{\perp}=mN(g\mu_B)^2/(4zJSR), \tag{35}$$

which decreases slightly as the temperature rises from 0 to T_N because of the factor m/R. This is also the case in the interacting spin-wave theory,[17] while a constant value is predicted by the molecular-field theory, the noninteracting spin-wave theory, and the Green's-function theory with RPA.[10] If the above susceptibility is expanded in powers of T at very low temperatures, the temperature-independent term turns out to be the same as that of interacting spin-wave theory while the T^2 term gives small deviation proportional to the $1/S$. A numerical calculation result for (35) is presented in Fig. 3.

ANALYSIS ABOVE THE NÉEL TEMPERATURE

In a real antiferromagnet, there still exists short-range order even above the Néel temperature. In the absence of an external field, a proper way of treating the

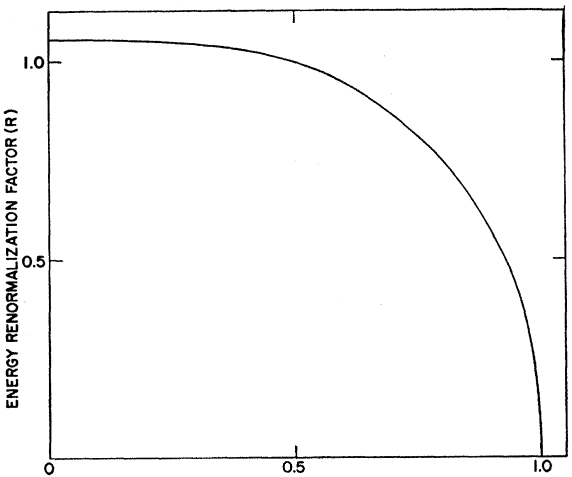

FIG. 2. Energy-spectrum renormalization factor for a simple cubic antiferromagnet ($S=\frac{1}{2}$).

---

[16] T. Oguchi, Phys. Rev. **117**, 117 (1960).
[17] S. H. Liu, Phys. Rev. **142**, 267 (1966).
[18] R. Kubo, J. Phys. Soc. Japan **12**, 570 (1957).

problem is to introduce a fictitious field H_f pointing in the $+z$ direction at the sites of sublattice 1 and in the $-z$ direction at those of sublattice 2 and to take the vanishing H_f limit at the end of calculation.[19] To this end the Hamiltonian

$$H=J\sum_i\sum_\delta \mathbf{S}_{1i}\cdot\mathbf{S}_{2,1+\delta}+J\sum_j\sum_\delta \mathbf{S}_{1,j+\delta}\cdot\mathbf{S}_{2j} -h\sum_i S_{1i}^z+h\sum_j S_{2j}^z, \quad (36)$$

with $h=g\mu_BH_f$, is used, and the vanishing h limit is taken after the calculation is carried out. The symmetry between two sublattices still exists and hence the relation (16) holds. The results of explicit calculation are the same as those of the Hamiltonian (5) except for the replacements: $A(0)\rightarrow h+A(0)$ and

$$\omega_\mathbf{k}=[A^2(0)-A^2(\mathbf{k})]^{1/2}\rightarrow\omega_\mathbf{k}=\{[h+A(0)]^2-A^2(\mathbf{k})\}^{1/2}.$$

To investigate the properties above the Néel point, it is convenient to introduce an order parameter x and the quantity A defined by

$$x=\lim_{h\to 0}(m/h) \quad\text{and}\quad A=\lim_{h\to 0}[A(0)/m], \quad (37)$$

since $m\rightarrow 0$ in the limit of $h\rightarrow 0$ above the Néel point. The quantity x is related with the correlation length between two spins and approaches infinity as the temperature is lowered to T_N as shown in the following. Above T_N one obtains

$$A=2zJ\left[1+\frac{2x^2A}{NS^2}\sum_\mathbf{k}\frac{k_BT\gamma^2(\mathbf{k})}{(1+xA)^2-[xA\gamma(\mathbf{k})]^2}\right], \quad (38)$$

$$\phi\equiv\phi_{11}\rightarrow\frac{2}{hN}\sum_\mathbf{k}\frac{k_BT(1+xA)}{(1+xA)^2-[xA\gamma(\mathbf{k})]^2}, \quad (39)$$

so ϕ diverges as $h\rightarrow 0$. In the same limit m can be expanded in powers of ϕ^{-1}, and from the first term of this expansion and Eq. (39) one obtains the equation for x

$$\frac{S(S+1)}{3x}=\frac{2k_BT}{N}\sum_\mathbf{k}\frac{(1+xA)}{(1+xA)^2-[xA\gamma(\mathbf{k})]^2}. \quad (40)$$

Putting $x\rightarrow\infty$ in Eqs. (38) and (40) and solving them, one finds the temperature at which x diverges:

$$(k_BT_\infty/2zJS)=[(S+1)/3I_0][1+(S+1)(I_0-1)/3SI_0], \quad (41)$$

the right-hand side of which is nothing but that of Eq. (32). Thus the temperature at which x diverges is the Néel temperature. At very high temperatures, it is possible to write

$$x=S(S+1)/3k_BT+O(T^{-2}). \quad (42)$$

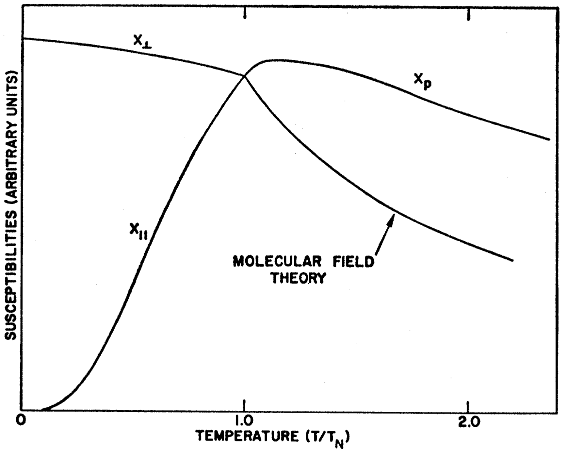

FIG. 3. Susceptibilities for a simple cubic antiferromagnet ($S=\frac{1}{2}$).

THE CORRELATION LENGTH

Above T_N the transverse correlation functions are found to be

$$\langle S_{1,-\mathbf{k}}^-S_{1,\mathbf{k}}^+\rangle=\langle S_{2,-\mathbf{k}}^-S_{2,\mathbf{k}}^+\rangle=\frac{Nk_BTx(1+xA)}{(1+xA)^2-[xA\gamma(\mathbf{k})]^2},$$

$$\langle S_{1,-\mathbf{k}}^-S_{2,\mathbf{k}}^+\rangle=\langle S_{2,-\mathbf{k}}^-S_{1,\mathbf{k}}^+\rangle=\frac{-Nk_BTx^2A\gamma(\mathbf{k})}{(1+xA)^2-[xA\gamma(\mathbf{k})]^2}, \quad (43)$$

and the perpendicular susceptibility is given by

$$\chi_\perp=xN(g\mu_B)^2/(1+2xA). \quad (44)$$

By putting $x\rightarrow\infty$ one easily sees that the above correlation functions and susceptibility are continuous at the Néel point with the lower-temperature values given in Eqs. (29) and (35).

When x is large and k is small, it follows from Eq. (43) that

$$\langle S_{\lambda,-\mathbf{k}}^-S_{\lambda,\mathbf{k}}^+\rangle\approx Nk_BT/A\eta^2(k^2+K^2), \quad (45)$$

where

$$[1-\gamma^2(\mathbf{k})]^{1/2}=\eta k+O(k^2), \quad (46)$$

$$K^2=2/xA\eta^2, \quad (47)$$

η being a constant depending on the lattice structure. Hence, for large $|\mathbf{R}_{\lambda i}-\mathbf{R}_{\lambda j}|$ one finds that

$$\begin{aligned}\langle S_{\lambda i}^-S_{\lambda j}^+\rangle&=(2/N)^2\sum_\mathbf{k}\langle S_{\lambda,-\mathbf{k}}^-S_{\lambda,\mathbf{k}}^+\rangle\\ &\quad\times\exp[-i\mathbf{k}\cdot(\mathbf{R}_{\lambda i}-\mathbf{R}_{\lambda j})]\\ &\approx(k_BT/2\pi\eta^2A)\\ &\quad\times\exp[-K(|\mathbf{R}_{\lambda i}-\mathbf{R}_{\lambda j}|)]/|\mathbf{R}_{\lambda i}-\mathbf{R}_{\lambda j}|.\end{aligned} \quad (48)$$

Thus the quantity K^{-1} can be interpreted as a correlation length between two spins, and the square root of

---

[19] J. Van Kranendonk and J. H. Van Vleck, Rev. Mod. Phys. **30**, 1 (1958).

the order parameter x is proportional to the correlation length in the long-wavelength limit. As the temperature is lowered to T_N the correlation length becomes infinite, i.e., long-range ordering commences at T_N. At very high temperatures the correlation length decreases as $T^{-1/2}$. Equation (48) is very similar to the ferromagnetic result.[20]

LONGITUDINAL CORRELATION FUNCTIONS

In this section Liu's work for ferromagnet[13] is extended to antiferromagnet. For this purpose the following perturbed Hamiltonian is used:

$$H_\sigma' = H - fS_{\sigma,-\mathbf{q}}{}^z \exp(-i\omega t + \epsilon t), \tag{49}$$

where H is the unperturbed Hamiltonian (5); f is a small parameter proportional to the amplitude of a fictitious ac field which is applied to the sublattice σ; $S_{\sigma,-\mathbf{q}}{}^z$ is a Fourier transform of $S_{\sigma i}{}^z$; and $\epsilon = 0^+$. It is to be noted that there are four longitudinal two-spin correlation functions for an antiferromagnet in contrast with one for a ferromagnet, and to calculate those four it is necessary to use two different perturbation terms in the Hamiltonian so that each choice gives two correlation functions. By definition,

$$\langle S_{\mu j}{}^z \rangle'^\sigma = \mathrm{Tr}(\rho_\sigma' S_{\mu j}{}^z), \tag{50}$$

where σ is the perturbation index and ρ_σ' is the density operator under perturbation. It can be shown that[18]

$$\langle S_{\mu j}{}^z \rangle'^\sigma = m_\mu + f\Sigma_{\mu j}{}^\sigma(\omega) \exp(-i\omega t + \epsilon t) + O(f^2), \tag{51}$$

with

$$\Sigma_{\mu j}{}^\sigma(\omega) = -i\int_{-\infty}^{0} \langle [S_{\sigma,-\mathbf{q}}{}^z(t'), S_{\mu j}{}^z(0)] \rangle \times \exp(-i\omega t' + \epsilon t')\, dt'. \tag{52}$$

It will be convenient to define the Fourier component of $\Sigma_{\mu j}{}^\sigma(\omega)$:

$$\Sigma_\mu{}^\sigma(\mathbf{q}, \omega) = \Sigma_j \Sigma_{\mu j}{}^\sigma(\omega) \exp(-i\mathbf{q}\cdot\mathbf{R}_{\mu j}).$$

The quantity $\Sigma_\mu{}^\sigma(\mathbf{q}, \omega)$ has the same expression as $\Sigma_{\mu j}{}^\sigma(\omega)$ except for the replacement of $S_{\mu j}{}^z(0)$ by $S_{\mu,\mathbf{q}}{}^z(0)$.

The two-spin correlation functions in Eq. (52) have the spectral representations

$$\langle S_{\lambda,-\mathbf{q}}{}^z(t) S_{\mu,\mathbf{q}}{}^z(0) \rangle = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} J_{\lambda\mu}{}^z(\mathbf{q}, \omega) \exp(-i\omega t), \tag{53}$$

where

$$J_{\lambda\mu}{}^z(\mathbf{q}, \omega) = 2\pi \sum_{m,n} \rho_m \langle m | S_{\lambda,-\mathbf{q}}{}^z | n \rangle \times \langle n | S_{\mu,\mathbf{q}}{}^z | m \rangle \delta(E_m - E_n + \omega), \tag{54}$$

and the sum is taken over the complete set of eigenstates of the unperturbed Hamiltonian H. If one defines a new quantity $S_\mu{}^\lambda(\mathbf{q}, n)$ by

$$S_\mu{}^\lambda(\mathbf{q}, n) = \int_0^\beta \langle T S_{\lambda,-\mathbf{q}}{}^z(\tau) S_{\mu,\mathbf{q}}{}^z(0) \rangle \exp(i\omega_n \tau)\, d\tau, \tag{55}$$

it is easy to show that[13]

$$\Sigma_\mu{}^\lambda(\mathbf{q}, \omega) = S_\mu{}^\lambda(\mathbf{q}, n = i\omega - \epsilon), \tag{56}$$

$$J_{\lambda\mu}{}^z(\mathbf{q}, \omega) = \{i/[\exp(-\beta\omega) - 1]\} \times [S_\mu{}^\lambda(\mathbf{q}, -i\omega + \epsilon) - S_\mu{}^\lambda(\mathbf{q}, -i\omega - \epsilon)]. \tag{57}$$

Putting this result into Eq. (53), one obtains the dynamical zz correlation function.

In the following, the calculation is carried out for $S = \frac{1}{2}$. The general S problem is in principle the same as the spin-$\frac{1}{2}$ case except for much more complex algebra. Since H' is time-dependent, one must be more careful in defining Green's functions. The system is unperturbed at $t = -\infty$, and the subsequent motion is described by the time-development operator $U(t, -\infty)$. The Green's function under perturbation can be defined in terms of U's, and its analytic continuation to imaginary-time domain is possible.[13] As already mentioned, for the spin-$\frac{1}{2}$ case it suffices to use a Green's function with $a = 0$. In the imaginary-time formalism, the Green's function to be used in this section is

$$G_{\lambda i,\mu j}{}'^\sigma(\tau) = \langle T S_{\lambda i}{}^+(\tau) S_{\mu j}{}^-(0) \rangle'^\sigma, \tag{58}$$

where the average is taken with the perturbed density matrix ρ_σ'. The equation of motion for G' is found to be

$$(d/d\tau) G_{\lambda i,\mu j}{}'^\sigma(\tau) = 2\delta(\tau)\delta_{\lambda\mu}\delta_{ij}\langle S_{\lambda i}{}^z \rangle'^\sigma + \langle T[H_\sigma'(\tau), S_{\lambda i}{}^+(\tau)] S_{\mu j}{}^-(0) \rangle, \tag{59}$$

where the first term of the right-hand side originated from the relationship $\theta_\lambda{}'^\sigma(0) = \langle [S_{\lambda i}{}^+, S_{\lambda i}{}^-] \rangle'^\sigma = 2\langle S_{\lambda i}{}^z \rangle'^\sigma$. This is the essential simplification of the algebra for the spin-$\frac{1}{2}$ case, and for general S the quantity $\theta_\lambda{}'^\sigma(a)$ should be carried through the rest of the calculation.

To calculate the longitudinal correlation function, one needs to calculate the first-order quantity $S_\mu{}^\lambda(\mathbf{q}, n)$ in the presence of perturbation. Callen's decoupling scheme for the unperturbed three-spin Green's function must accordingly be extended for the perturbed Green's function. That is, if one makes expansions

$$G_{\lambda i,\mu j}{}'^\sigma(\tau) = G_{\lambda i,\mu j}(\tau) + fG_{\lambda i,\mu j}{}^{(1)\sigma}(\tau) + O(f^2),$$

$$\langle S_{\lambda i}(\tau) \rangle'^\sigma = m_\lambda + fS_{\lambda i}{}^\sigma(n) \exp(-i\omega_n \tau) + O(f^2), \tag{60}$$

where $S_{\lambda i}{}^\sigma(n)$ is the imaginary-time representation of $\Sigma_{\lambda i}{}^\sigma(\omega)$, and tries to solve the equations for the first-order quantities, there arises the problem of how to take the first-order quantity in small parameter f from the last term of Eq. (9). Since the introduction of this term contains a high degree of arbitrariness, one may have some freedom in choosing the best way to approximate its first-order response. The choice made in this

---

[20] L. Van Hove, Phys. Rev. **95**, 1382 (1954).

work is

$$[\langle S_{\mu j}^z\rangle\langle S_{\lambda i}^- S_{\mu j}^+\rangle\langle\langle S_{\lambda i}^+;B\rangle\rangle]^{(1)}$$
$$\approx[\langle S_{\mu j}^z\rangle^{(1)}\langle\langle S_{\lambda i}^+;B\rangle\rangle+\langle S_{\mu j}^z\rangle\langle\langle S_{\lambda i}^+;B\rangle\rangle^{(1)}]\langle S_{\lambda i}^- S_{\mu j}^+\rangle. \tag{61}$$

The ultimate justification of this choice lies in the fact that the resulting zz correlation functions satisfy a number of physical properties a correct theory requires.

The equations for zeroth-order quantities have already been solved in the preceding sections. After decoupling by the extended Callen scheme, the first-order equation for G turns out to be

$$\begin{aligned}(d/d\tau)G_{\lambda i,\mu j}{}^{(1)\sigma}(\tau)&=2\delta(\tau)\delta_{\lambda\mu}\delta_{ij}S_{\lambda i}{}^{\sigma}(n)\exp(-i\omega_n\tau)-\delta_{\lambda\sigma}G_{\lambda i,\mu j}(\tau)\exp(-i\mathbf{q}\cdot\mathbf{R}_{\lambda i}-i\omega_n\tau)\\&\quad-2J\sum_{\delta}\{S_{\lambda i}{}^{\sigma}(n)[1-2\psi_{\nu(i+\delta),\lambda i}]G_{\nu(i+\delta),\mu j}(\tau)-S_{\nu(i+\delta)}{}^{\sigma}(n)[1-2\psi_{\lambda i,\nu(i+\delta)}]G_{\lambda i,\mu j}(\tau)\}\exp(-i\omega_n\tau)\\&\quad-2J\sum_{\delta}\{m_\lambda[1-2\psi_{\nu(i+\delta),\lambda i}]G_{\nu(i+\delta),\mu j}{}^{(1)\sigma}(\tau)-m_\nu[1-2\psi_{\lambda i,\nu(i+\delta)}]G_{\lambda i,\mu j}{}^{(1)\sigma}(\tau)\},\qquad\nu\neq\lambda.\end{aligned}\tag{62}$$

The first-order quantities can be Fourier analyzed:

$$\begin{aligned}S_{\lambda i}{}^{\sigma}(n)&=(2/N)\sum_{\mathbf{k}}S_{\lambda}{}^{\sigma}(\mathbf{k},n)\exp(i\mathbf{k}\cdot\mathbf{R}_{\lambda i}),\\G_{\lambda i,\mu j}{}^{(1)\sigma}(\tau)&=(2/N)^2\beta^{-1}\sum_{\mathbf{k}_1,\mathbf{k}_2,m}G_{\lambda\mu}{}^{(1)\sigma}(\mathbf{k}_1,\mathbf{k}_2,m)\exp(i\mathbf{k}_1\cdot\mathbf{R}_{\lambda i}-i\mathbf{k}_2\cdot\mathbf{R}_{\mu j}-i\omega_m\tau).\end{aligned}\tag{63}$$

After the Fourier transform, Eq. (62) can be written as

$$\begin{aligned}-[A_\nu(0)+i\omega_m]G_{\lambda\mu}{}^{(1)\sigma}(\mathbf{k}_1,\mathbf{k}_1-\mathbf{q},m)+A_\lambda(\mathbf{k}_1)G_{\nu\mu}{}^{(1)\sigma}(\mathbf{k}_1,\mathbf{k}_1-\mathbf{q},m)&=2\delta_{\lambda\mu}S_\lambda{}^\sigma(\mathbf{q},n)-(N/2)\delta_{\lambda\sigma}G_{\lambda\mu}(\mathbf{k}_1-\mathbf{q},m-n)\\&\quad+B_\nu{}^\sigma(0)G_{\lambda\mu}(\mathbf{k}_1-\mathbf{q},m-n)-B_\lambda{}^\sigma(\mathbf{k}_1)G_{\nu\mu}(\mathbf{k}_1-\mathbf{q},m-n),\qquad\nu\neq\lambda\end{aligned}\tag{64}$$

where

$$B_\lambda{}^\sigma(\mathbf{k})=S_\lambda{}^\sigma(\mathbf{q},n)A_\lambda(\mathbf{k}-\mathbf{q})/m_\lambda,\tag{65}$$

and the linear response to the perturbation expressed by $S_\lambda{}^\sigma(\mathbf{k},n)=\delta_{\mathbf{k},\mathbf{q}}S_\lambda{}^\sigma(\mathbf{q},n)$ was taken into account. It is straightforward to write down the solution of the above equation in terms of the zeroth-order Green's functions and $S_\lambda{}^\sigma(\mathbf{q},n)$'s.

To calculate $S_\lambda{}^\sigma(\mathbf{q},n)$ one uses the self-consistency condition for Green's functions which is a relation connecting $S_\lambda{}^\sigma(\mathbf{q},n)$ and $G_{\lambda\lambda}{}^{(1)\sigma}(\mathbf{k}_1,\mathbf{k}_1-\mathbf{q},\tau=0^-)$. From the definition (58) it follows that

$$G_{\lambda i,\lambda i}{}'^{\sigma}(0^-)=\tfrac{1}{2}-m_\lambda-fS_{\lambda i}{}^\sigma(n)+O(f^2).\tag{66}$$

Equating the first-order terms of Eqs. (60) and (66) and making Fourier summation, one gets

$$-S_\lambda{}^\sigma(\mathbf{q},n)=(2/N)\sum_{\mathbf{k}}G_{\lambda\lambda}{}^{(1)}(\mathbf{k},\mathbf{k}-\mathbf{q},0^-).\tag{67}$$

By use of the solution of Eq. (64), Eq. (67) can be written explicitly as follows:

$$\begin{aligned}-S_1{}^\sigma(\mathbf{q},n)&=(2/N)S_1{}^\sigma(\mathbf{q},n)\sum_{\mathbf{k}}[A_1(0)\omega_{\mathbf{k}}{}^{-1}\coth\tfrac{1}{2}\beta\omega_{\mathbf{k}}-1]+C_0{}^\sigma+C_1S_1{}^\sigma(\mathbf{q},n)+C_2S_2{}^\sigma(\mathbf{q},n),\\-S_2{}^\sigma(\mathbf{q},n)&=(2/N)S_2{}^\sigma(\mathbf{q},n)\sum_{\mathbf{k}}[A_2(0)\omega_{\mathbf{k}}{}^{-1}\coth\tfrac{1}{2}\beta\omega_{\mathbf{k}}-1]+D_0{}^\sigma+D_1S_1{}^\sigma(\mathbf{q},n)+D_2S_2{}^\sigma(\mathbf{q},n),\end{aligned}\tag{68}$$

where

$$\begin{aligned}C_0{}^\sigma&=-m_1\sum_{\mathbf{k}}(2\omega_{\mathbf{k}}\omega_{\mathbf{k}-\mathbf{q}})^{-1}\sum_{\lambda\mu}N_{\lambda\mu}\{\delta_{\sigma1}[A_1(0)+\omega_{\mathbf{k}}{}^\lambda][A_1(0)+\omega_{\mathbf{k}-\mathbf{q}}{}^\mu]+\delta_{\sigma2}A_1(\mathbf{k})A_2(\mathbf{k}-\mathbf{q})\},\\C_1&=N^{-1}\sum_{\mathbf{k}}(\omega_{\mathbf{k}}\omega_{\mathbf{k}-\mathbf{q}})^{-1}\sum_{\lambda\mu}N_{\lambda\mu}\{A_1(\mathbf{k})A_2(\mathbf{k}-\mathbf{q})A_1(\mathbf{q})-[A_1(0)+\omega_{\mathbf{k}}{}^\lambda]A_2(\mathbf{k}-\mathbf{q})A_1(\mathbf{k}-\mathbf{q})\},\\C_2&=N^{-1}\sum_{\mathbf{k}}(\omega_{\mathbf{k}}\omega_{\mathbf{k}-\mathbf{q}})^{-1}\sum_{\lambda\mu}N_{\lambda\mu}\{-[A_1(0)+\omega_{\mathbf{k}}{}^\lambda][A_1(0)+\omega_{\mathbf{k}-\mathbf{q}}{}^\mu]A_2(\mathbf{q})+A_1(\mathbf{k})[A_1(0)+\omega_{\mathbf{k}-\mathbf{q}}{}^\mu]A_2(\mathbf{k}-\mathbf{q})\},\\D_0{}^\sigma&=-m_2\sum_{\mathbf{k}}(2\omega_{\mathbf{k}}\omega_{\mathbf{k}-\mathbf{q}})^{-1}\sum_{\lambda\mu}N_{\lambda\mu}\{\delta_{\sigma1}A_2(\mathbf{k})A_1(\mathbf{k}-\mathbf{q})+\delta_{\sigma2}[A_2(0)+\omega_{\mathbf{k}}{}^\lambda][A_2(0)+\omega_{\mathbf{k}-\mathbf{q}}{}^\mu]\},\\D_1&=N^{-1}\sum_{\mathbf{k}}(\omega_{\mathbf{k}}\omega_{\mathbf{k}-\mathbf{q}})^{-1}\sum_{\lambda\mu}N_{\lambda\mu}\{-[A_2(0)+\omega_{\mathbf{k}}{}^\lambda][A_2(0)+\omega_{\mathbf{k}-\mathbf{q}}{}^\mu]A_1(\mathbf{q})+A_2(\mathbf{k})[A_2(0)+\omega_{\mathbf{k}-\mathbf{q}}{}^\mu]A_1(\mathbf{k}-\mathbf{q})\},\\D_2&=N^{-1}\sum_{\mathbf{k}}(\omega_{\mathbf{k}}\omega_{\mathbf{k}-\mathbf{q}})^{-1}\sum_{\lambda\mu}N_{\lambda\mu}\{A_2(\mathbf{k})A_1(\mathbf{k}-\mathbf{q})A_2(\mathbf{q})-[A_2(0)+\omega_{\mathbf{k}}{}^\lambda]A_1(\mathbf{k}-\mathbf{q})A_2(\mathbf{k}-\mathbf{q})\},\end{aligned}\tag{69}$$

$$\begin{aligned}N_{11}=N_{22}{}^*&=[N(\mathbf{k})-N(\mathbf{k}-\mathbf{q})]/(-\omega_{\mathbf{k}}+\omega_{\mathbf{k}-\mathbf{q}}-i\omega_n),\\N_{12}=N_{21}{}^*&=[N(\mathbf{k})+N(\mathbf{k}-\mathbf{q})+1]/(\omega_{\mathbf{k}}+\omega_{\mathbf{k}-\mathbf{q}}-i\omega_n),\end{aligned}\tag{70}$$

$$N(\mathbf{k})=[\exp(\beta\omega_{\mathbf{k}})-1]^{-1},\tag{71}$$

and it is to be understood that $\omega_{\mathbf{k}}^{\lambda}$ means $\omega_{\mathbf{k}}$ for $\lambda=1$ and $-\omega_{\mathbf{k}}$ for $\lambda=2$. By use of the relation

$$(2m_{\lambda})^{-1}=2N^{-1}\sum_{\mathbf{k}}A_{\lambda}(0)\,\omega_{\mathbf{k}}^{-1}\coth\tfrac{1}{2}\beta\omega_{\mathbf{k}} \qquad (72)$$

which follows from Eq. (25) for $S=\frac{1}{2}$, Eq. (68) can be solved for $S_1{}^{\sigma}(\mathbf{q},n)$ and $S_2{}^{\sigma}(\mathbf{q},n)$:

$$\begin{aligned}S_1{}^{\sigma}(\mathbf{q},n)&=-2m\Delta^{-1}[C_0{}^{\sigma}-2m(C_0{}^{\sigma}D_2-D_0{}^{\sigma}C_2)],\\ S_2{}^{\sigma}(\mathbf{q},n)&=2m\Delta^{-1}[D_0{}^{\sigma}+2m(D_0{}^{\sigma}C_1-C_0{}^{\sigma}D_1)],\end{aligned} \qquad (73)$$

with

$$\Delta=1+2m(C_1-D_2)-(2m)^2(C_1D_2-C_2D_1). \qquad (74)$$

Putting these results into Eqs. (53) and (57), one achieves the task of calculating the dynamical *zz* correlation function of two spins.

To get the longitudinal correlation functions for the temperature above the Néel point, one uses the same procedure as that of the fifth section and the result turns out to be

$$\begin{aligned}S_1{}^1(\mathbf{q},n)=S_2{}^2(\mathbf{q},n)&=\tfrac{1}{2}N\frac{x(1+xA)}{(1+xA)^2-[xA\gamma(\mathbf{q})]^2}\delta_{n0},\\ S_1{}^2(\mathbf{q},n)=S_1{}^2(\mathbf{q},n)&=\tfrac{1}{2}N\frac{-x^2A\gamma(\mathbf{q})}{(1+xA)^2-[xA\gamma(\mathbf{q})]^2}\delta_{n0}.\end{aligned} \qquad (75)$$

When continued to the real-frequency domain, the Kronecker δ becomes $\beta^{-1}\delta(\omega)$. Hence, one gets the dynamical *zz* correlation functions of two spins

$$\begin{aligned}\langle S_{1,-\mathbf{q}}{}^z(t)\,S_{1,\mathbf{q}}{}^z(0)\rangle&=\langle S_{2,-\mathbf{q}}{}^z(t)\,S_{2,\mathbf{q}}{}^z(0)\rangle\\ &=\frac{N}{2\beta}\frac{x(1+xA)}{(1+xA)^2-[xA\gamma(\mathbf{q})]^2},\\ \langle S_{1,-\mathbf{q}}{}^z(t)\,S_{2,\mathbf{q}}{}^z(0)\rangle&=\langle S_{2,-\mathbf{q}}{}^z(t)\,S_{1,\mathbf{q}}{}^z(0)\rangle\\ &=\frac{N}{2\beta}\frac{-x^2A\gamma(\mathbf{q})}{(1+xA)^2-[xA\gamma(\mathbf{q})]^2}.\end{aligned} \qquad (76)$$

These results together with Eq. (43) show that *xx*, *yy*, and *zz* correlation functions are all equal above the Néel point as expected from the fact that all spin directions must be equivalent when $h\to 0$ in the paramagnetic region.

One expects the correlation functions to be continuous at the Néel temperature. By putting $x\to\infty$ in the expressions for correlation functions of the paramagnetic phase and comparing the result with the lower-temperature phase result, one easily sees that they are continuous at T_N.

There is a sum rule for the static correlation functions of spins, namely

$$(2/N)\sum_{\mathbf{k}}\langle \mathbf{S}_{\lambda,-\mathbf{k}}\cdot\mathbf{S}_{\lambda,\mathbf{k}}\rangle=\tfrac{1}{2}NS(S+1). \qquad (77)$$

If one uses Eqs. (43) and (76) and the paramagnetic version of the relation (72)

$$\tfrac{1}{2}=\frac{4}{N\beta}\sum_{\mathbf{k}}\frac{x(1+xA)}{(1+xA)^2-[xA\gamma(\mathbf{k})]^2}, \qquad (78)$$

the left-hand side of Eq. (77) becomes equal to $\frac{1}{2}N(\frac{3}{4})$ and thus the sum rule checks above T_N. But below T_N there seems to be no easy way of checking the sum rule because of complicated algebra. Even in the spin-wave region, the *zz* correlation functions are so complicated that it is not feasible to try to check the sum rule. Perhaps one should not even expect to do so because the expressions for the correlation functions are only approximate.

PARALLEL SUSCEPTIBILITY

The parallel susceptibility can be calculated as the ratio of the total magnetization to the applied field which is parallel to the spin alignment direction. The Hamiltonian to be used for this purpose is, in imaginary-time formalism,

$$H'=H-f(S_{1,\mathbf{0}}{}^z+S_{2,\mathbf{0}}{}^z). \qquad (79)$$

Then the parallel susceptibility is given by

$$\begin{aligned}\chi_{||}&=(g\mu_B)^2\sum_{\lambda i}\langle S_{\lambda i}{}^z\rangle'/f\\ &=(g\mu_B)^2(\langle S_{1,\mathbf{0}}{}^z\rangle'+\langle S_{2,\mathbf{0}}{}^z\rangle')/f,\end{aligned} \qquad (80)$$

the primes meaning averages under the perturbation. Similar calculation as that of deriving Eq. (51) gives

$$\begin{aligned}\langle S_{1,\mathbf{0}}{}^z\rangle'&=\tfrac{1}{2}Nm_1+fS_1{}^1(\mathbf{0},0)+fS_1{}^2(\mathbf{0},0)+O(f^2),\\ \langle S_{2,\mathbf{0}}{}^z\rangle'&=\tfrac{1}{2}Nm_2+fS_2{}^1(\mathbf{0},0)+fS_2{}^2(\mathbf{0},0)+O(f^2).\end{aligned} \qquad (81)$$

Then, taking into account the relationships

$$S_2{}^2(-\mathbf{q},-n)=S_1{}^1(\mathbf{q},n) \quad\text{and}\quad S_2{}^1(-\mathbf{q},-n)=S_1{}^2(\mathbf{q},n)$$

which can be proved by direct substitution in Eq. (73), one gets the parallel susceptibility

$$\begin{aligned}\chi_{||}&=2(g\mu_B)^2\lim_{\mathbf{q}\to 0}[S_1{}^1(\mathbf{q},0)+S_1{}^2(\mathbf{q},0)]\\ &=2\beta(2mg\mu_B)^2\left\{\sum_{\mathbf{k}}\frac{\exp(\beta\omega_{\mathbf{k}})}{[\exp(\beta\omega_{\mathbf{k}})-1]^2}\right\}\\ &\qquad\left\{1+8N^{-1}\beta mA(0)\sum_{\mathbf{k}}\frac{\exp(\beta\omega_{\mathbf{k}})}{[\exp(\beta\omega_{\mathbf{k}})-1]^2}\right\}^{-1}.\end{aligned} \qquad (82)$$

If one uses the RPA which is equivalent to putting $2mzJ$ for $A(0)$, the modified result agrees with that of Lines.[10] It is to be noted that the second term of the denominator is absent in the nonlinear spin-wave theory,[17] and the spin-wave theory does not give the factor $(2m)^2$ in the numerator. The extra term in the denominator vanishes at $T=0$ and is equal to unity at T_N, while $(2m)^2$ varies from $(2m_0)^2$ to zero so that the effect of these two terms compensate to some extent. At very low temperatures m varies slightly with temperature and the new term in the denominator is vanishingly small, so one expects very good agreement of the above result with that of the spin-wave theory.

It is easy to show that the parallel susceptibility in Eq. (82) vanishes as $T\to 0^+$ and becomes equal to the value of perpendicular susceptibility at T_N, which is given in Eq. (35), as $T\to T_N^-$. One can also show that the paramagnetic temperature version of Eq. (82) leads to Eq. (44). Numerical calculations of parallel and perpendicular susceptibilities for $S=\frac{1}{2}$ have been carried out and the results are shown in Fig. 3.

ENERGY AND SPECIFIC HEAT

For the Heisenberg antiferromagnet with nearest-neighbor interactions described by the Hamiltonian (5), the internal energy is given by

$$E=\langle H\rangle=4N^{-1}zJ\sum_{\mathbf{q}}\gamma(\mathbf{q})\langle \mathbf{S}_{1,-\mathbf{q}}\cdot\mathbf{S}_{2,\mathbf{q}}\rangle, \tag{83}$$

so the internal energy can be computed if the static correlation functions of spins are known, and differentiating the result with respect to temperature one obtains the specific heat due to spin disorder. Although expressions for all the correlation functions have already been obtained for $S=\frac{1}{2}$, the study of the problem for general temperature is rather hard because of the complex algebra involved. In the following, the problem only in paramagnetic region and ground state is studied.

In the paramagnetic temperature region when $h\to 0$, it can be verified that

$$E=-\frac{6zJ}{A\beta}\sum_{\mathbf{q}}\frac{[xA\gamma(\mathbf{q})]^2}{(1+xA)^2-[xA\gamma(\mathbf{q})]^2}. \tag{84}$$

Putting $x\to\infty$ in Eq. (84) and using Eq. (78) in the same limit, one obtains the internal energy at T_N

$$\begin{aligned}E_N&=-\tfrac{1}{4}NzJ[3(I_0-1)/I_0]\\&\approx-\tfrac{1}{4}NzJ(1.021)\text{ for sc, }-\tfrac{1}{4}NzJ(0.846)\text{ for bcc.}\end{aligned} \tag{85}$$

The specific heat is given by

$$\begin{aligned}C=dE/dT=\tfrac{3}{8}N\{[\beta(1+xA)+4x]I_1-8x(1+xA)^2I_2\}\\ \times\{8\beta^{-1}x^3A[I_1-2(1+xA)^2I_2]I_3-A^{-1}\\ \times[(2+xA)I_1-2(1+xA)^2I_2]\}^{-1},\end{aligned} \tag{86}$$

where

$$\begin{aligned}I_1&=2N^{-1}\sum_{\mathbf{q}}\{(1+xA)^2-[xA\gamma(\mathbf{q})]^2\}^{-1},\\ I_2&=2N^{-1}\sum_{\mathbf{q}}\{(1+xA)^2-[xA\gamma(\mathbf{q})]^2\}^{-2},\\ I_3&=2N^{-1}\sum_{\mathbf{q}}\gamma^2(\mathbf{q})\{(1+xA)^2-[xA\gamma(\mathbf{q})]^2\}^{-1},\end{aligned} \tag{87}$$

and the Boltzmann constant k_B is set equal to unity for convenience. At very high temperatures one gets

$$C=\tfrac{3}{4}(zJ/T)^2\sum_{\mathbf{q}}\gamma^2(\mathbf{q})+O(T^{-3}). \tag{88}$$

At this point, it is to be noticed that all the correlation functions are calculated for $\mathbf{q}\neq 0$, and the ground-state energy consists of three parts: the contributions from $\mathbf{q}=0$, from the transverse correlation function $(\mathbf{q}\neq 0)$, and from the longitudinal correlation function $(\mathbf{q}\neq 0)$; so one can write

$$E_0=E^0+E^t+E^l. \tag{89}$$

Then from Eqs. (83) and (29) it follows that

$$E^0=-\tfrac{1}{4}NzJ(2m_0)^2, \tag{90}$$

$$E^t=-\tfrac{1}{4}NzJ(2m_0)[2(c+c')], \tag{91}$$

where

$$\begin{aligned}c&=2N^{-1}\sum_{\mathbf{k}}\{1-[1-\gamma^2(\mathbf{k})]^{1/2}\}\\&\approx 0.097\text{ for sc, }0.073\text{ for bcc,}\end{aligned} \tag{92}$$

$$\begin{aligned}c'&=2N^{-1}\sum_{\mathbf{k}}\{[1-\gamma^2(\mathbf{k})]^{-1/2}-1\}\\&\approx 0.156\text{ for sc, }0.150\text{ for bcc.}\end{aligned} \tag{93}$$

One needs the explicit expression for $\langle S_{1,-\mathbf{q}}{}^zS_{2,\mathbf{q}}{}^z\rangle$, and it is given by the inverse transform of Eq. (55)

$$\langle S_{1,-\mathbf{q}}{}^zS_{2,\mathbf{q}}{}^z\rangle=\beta^{-1}\sum_n S_2{}^1(\mathbf{q},n)\exp(-i\omega_n\epsilon), \tag{94}$$

with $S_1{}^2(\mathbf{q},n)$ given by (73). If one keeps only the first term $D_0{}^\sigma$ in the numerator of (73) and approximates Δ by unity, one gets

$$\begin{aligned}E^l\approx\frac{4zJm_0{}^2}{\beta N}\sum_{n,\mathbf{q},\mathbf{k}}\frac{\gamma(\mathbf{q})}{\omega_{\mathbf{k}}\omega_{\mathbf{k}-\mathbf{q}}}\\ \times\sum_{\lambda\mu}N_{\lambda\mu}[A(\mathbf{k})A(\mathbf{k}-\mathbf{q})]\exp(-i\omega_n\epsilon).\end{aligned} \tag{95}$$

If one sums over n and takes the limit of $T\to 0^+$, Eq. (95) leads to

$$E^l\approx-\tfrac{1}{4}NzJ(2m_0)^2(c+c')^2. \tag{96}$$

Since Eq. (72) leads to

$$m_0=[2(1+c')]^{-1} \tag{97}$$

as $T\to 0^+$, one obtains the ground-state energy of an antiferromagnet to the above-mentioned approximation

$$E_0\approx-\tfrac{1}{4}NzJ(1.234)\text{ for sc, }-\tfrac{1}{4}NzJ(1.173)\text{ for bcc.} \tag{98}$$

The value for sc agrees with the result obtained by Boon, but the present result is a few percent lower than the values given by other theories.[21] If one includes the next terms in the numerator and denominator of (73), the resulting correction for the ground-state energy is estimated to be less than 1%. It seems that a calculation using the entire expression of Eq. (73) would be very difficult.

---

[21] P. W. Anderson, Ref. 4; R. Kubo, Ref. 4; W. Marshall, Proc. Roy. Soc. (London) **A232**, 48 (1955); J. C. Fisher, J. Phys. Chem. Solids **10**, 44 (1959); H. L. Davis, Phys. Rev. **120**, 789 (1960); M. H. Boon, Nuovo Cimento **21**, 885 (1961); T. Oguchi, J. Phys. Chem. Solids **24**, 1049 (1963); S. H. Liu, Ref. 17; T. Arai and B. Goodman, Phys. Rev. **155**, 514 (1967).