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THERMODYNAMICS AND CORRELATION FUNCTIONS OF A HEISENBERG ANTIFERROMAGNET

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The diagram method proposed for a ferromagnet[1] is extended to include the case of an antiferromagnet, for arbitrary values of the magnetic field and of the temperature. The corrections to the magnetization, susceptibility, and the spin-wave spectrum at low temperature are obtained. It is shown that the correction to the spin-wave spectrum is proportional in first approximation to $\alpha(T/I_0S)^4$, where $\alpha \sim 1$ is a parameter. The corrections to the ground-state energy are calculated.

INTRODUCTION

ANTIFERROMAGNETS at sufficiently low temperatures exhibit a magnetic ordering whereby the spins of the neighboring atoms are antiparallel. Such an orientation is due to the exchange interaction between the electrons (direct or indirect), and the exchange integral should be negative. The exchange interaction causes the spins of the electrons to exchange places. This is of no significance in the case when the spins are oriented in parallel[1], but for the antiferromagnetic order it causes the nonstationary character of the state in which the magnetic moments of the neighboring atoms are opposite in direction and their magnitude is equal to S. Therefore an exact solution, describing the ground state of the antiferromagnet, was obtained only for the one-dimensional case[2]. In the three-dimensional case, the energy of the ground state and the thermodynamic quantities were calculated predominantly by perturbation-theory methods. However, the application of ordinary perturbation theory to spin systems encounters a number of difficulties, since the spin operators satisfy neither the Fermi nor the Bose commutation relations. The commutators of the spin operators are not c-numbers, and therefore the usual Wick method[3] cannot be applied to these operators.

To get around these difficulties, some workers change over from spin operators to Bose operators with the aid of the Holstein-Primakoff transformation[4], and then employ ordinary perturbation theory[5,6]. However, such an approach can lead to unphysical states. Another method, frequently used in the theory of antiferromagnetism[7], is the method of time-dependent Green's functions. But the decoupling of the equations for the Green's functions is not sufficiently well founded, and therefore the results obtained by this method are only qualitative in character.

In a number of papers[1,8-14] an attempt was made to extend the ordinary perturbation theory to the spin operators. The perturbation theory was applied either directly to the spin operators[1,9,11,12] or else fermions or coupled boson pairs[8,10,13,14] were introduced, automatically taking into account the kinematic interaction. These methods yielded corrections to the energy of the ground state and the spin-wave spectrum of the antiferromagnets at a temperature T = 0.

In the present paper, by generalizing the method proposed in[1] (see also[15]), we investigate the thermodynamic properties and the correlation functions of antiferromagnets. We note that such calculations were carried out in the molecular-field approximation in[15], where the authors confined themselves to temperature corrections of first approximation to the thermodynamic quantities (magnetic moment, specific heat, etc.).

In the present paper we develop a diagram technique for the calculation of the higher-order corrections to the spin-wave spectrum and to the thermodynamic quantities. We find that at T = 0 the second-order correction to the sublattice magnetization vanishes, and the region of applicability of the obtained results is indicated.

1. FORMULATION OF PROBLEM AND GENERAL ANALYSIS

We consider an antiferromagnet whose anisotropy is equal to zero and in which exchange interaction I exists only between two sublattices. The Hamiltonian of the system under consideration is given by

$$\mathscr{H} = \sum_{\mathbf{rr}'} I(\mathbf{r}-\mathbf{r}')\,\mathbf{S}_{1\mathbf{r}}\mathbf{S}_{2\mathbf{r}'} - \mu H \sum_{\mathbf{r}} (S_{1\mathbf{r}}^z + S_{2\mathbf{r}}^z), \qquad (1.1)$$

where $\mathbf{S}_j$ is the spin operator of the j-th sublattice, μ is the Bohr magneton, and H is the constant external magnetic field.

In order to calculate the spin-wave spectrum and the thermodynamic quantities, we investigate the free energy of the system F:

$$-\beta F = \frac{1}{N}\ln \operatorname{Sp}\exp(-\beta\mathscr{H}), \qquad (1.2)$$

where $\beta = 1/T$, N is the number of cells in the crystal, and the correlation functions $K_{jk}(\mathbf{k}, i\omega_n)$[1] are given by:

$$K_{j_1j_2}(\mathbf{k}, i\omega_n)$$

$$= \frac{1}{2\beta}\sum_{\mathbf{r}_2} e^{i\mathbf{k}(\mathbf{r}_1-\mathbf{r}_2)} \int_{-\beta}^{\beta} dt e^{i\omega_n t} \langle \hat{T}(\mathbf{S}_{j_1\mathbf{r}_1}(t) - \langle \mathbf{S}_{j_1}\rangle)(\mathbf{S}_{j_2\mathbf{r}_2}(0) - \langle \mathbf{S}_{j_2}\rangle)\rangle, \qquad (1.3)$$

where $i\omega_n = 2i\pi nT$ are the imaginary frequencies of the temperature technique[16], j_1 and j_2 are the indices of the

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sublattices, $S(t) = e^{i\mathscr{H}t}Se^{-i\mathscr{H}t}$, and $\hat{T}$ is the T-ordering symbol.

To calculate these quantities, we generalize the method proposed in[1]. We represent the spin operator of the j-th sublattice in the form

$$\mathbf{S}_j = \mathbf{e}_{j\xi}S_{j\xi} + \mathbf{e}_{j\eta}S_{j\eta} + \mathbf{e}_{j\zeta}S_{j\zeta}, \tag{1.4}$$

where the unit vectors are chosen as follows:

$$\begin{aligned} \mathbf{e}_{j\xi} &= \mathbf{e}_x, \\ \mathbf{e}_{j\eta} &= \cos\vartheta\mathbf{e}_y + (-1)^j \sin\vartheta\mathbf{e}_z, \\ \mathbf{e}_{j\zeta} &= (-1)^{j+1}\sin\vartheta\mathbf{e}_y + \cos\vartheta\mathbf{e}_z. \end{aligned} \tag{1.5}$$

Here the angle ϑ is a parameter yet to be determined.

We separate the Hamiltonian of the system into two parts:

$$\mathscr{H} = \mathscr{H}_0 + \mathscr{H}_1, \tag{1.6}$$

where

$$\begin{aligned} \mathscr{H}_0 = -N\langle S^\zeta\rangle^2 I_0\cos 2\vartheta + \sum_{\mathbf{r}}(I_0\cos 2\vartheta\langle S^\zeta\rangle - \mu H\cos\vartheta)(S_{1\mathbf{r}}^\zeta + S_{2\mathbf{r}}^\zeta) \\ - \sum_{\mathbf{r}}\sin\vartheta(2I_0\langle S^\zeta\rangle\cos\vartheta - \mu H)(S_{1\mathbf{r}}^\eta - S_{2\mathbf{r}}^\eta) \end{aligned} \tag{1.7}$$

is the single-particle Hamiltonian, corresponding to the molecular-field approximation, and

$$\mathscr{H}_1 = \sum_{\mathbf{rr}'} I(\mathbf{r}-\mathbf{r}')(\mathbf{S}_{1\mathbf{r}} - \langle\mathbf{S}_1\rangle)(\mathbf{S}_{2\mathbf{r}'} - \langle\mathbf{S}_2\rangle) \tag{1.8}$$

is a small perturbation to the system. Here $\langle S^\zeta\rangle$, like the angle ϑ, is a still undetermined parameter. It is convenient to determine $\langle S^\zeta\rangle$ and ϑ from the minimum of the free energy, and then $\langle S^\zeta\rangle$ denotes the average spin, and ϑ—the angle between the vector of the average spin of the j-th sublattice $\langle S_j\rangle$ and the magnetic field H.

We can determine here the values of $\langle S_j^\zeta\rangle$ and ϑ either in the molecular-field approximation and fix their values, or else find these parameters from the considered free-energy approximation. We shall henceforth employ the first variant. The free energy in the molecular-field approximation is given by[15]

$$\begin{aligned} -\beta F^{(0)} = \frac{(y-\beta\mu H\cos\vartheta)^2}{\beta I_0\cos 2\vartheta} + 2\ln\frac{\operatorname{sh}(S+{}^1/_2)y}{\operatorname{sh}{}^1/_2 y} \\ + \beta\frac{b(y)}{y}\sin^2\vartheta(2I_0\langle S^\zeta\rangle\cos\vartheta - \mu H)^2, \end{aligned} \tag{1.9}$$

where

$$y = \beta(-I_0\langle S^\zeta\rangle\cos 2\vartheta + \mu H\cos\vartheta). \tag{1.10}$$

In formula (1.9) we retain terms that are quadratic in the quantity $\beta\sin\vartheta(2I_0\langle S^\zeta\rangle\cos\vartheta - \mu H)$. From the condition (1.10) for the minimum of the free energy we can obtain the average spin $\langle S^\zeta\rangle$ and the angle ϑ[15]:

$$\langle S^\zeta\rangle = -\frac{y-\beta\mu H\cos\vartheta}{\beta I_0\cos 2\vartheta} = b(y), \tag{1.11}$$

$$\cos\vartheta = \mu H / 2I_0 b(y), \tag{1.12}$$

where $b(y) = (S + 1/2)\coth(S + 1/2)y - (1/2)\coth(y/2)$ is the Brillouin function.

The correlation function $K_{j_1j_2}(\mathbf{k}, i\omega_n)$ satisfies the Dyson equation:

$$K_{j_1j_2}(\mathbf{k}, i\omega_n) = \Sigma_{j_1j_2}(\mathbf{k}, i\omega_n) - \Sigma_{j_1j_3}(\mathbf{k}, i\omega_n)(\beta I_\mathbf{k})_{j_3j_4}K_{j_4j_2}(\mathbf{k}, i\omega_n) \tag{1.13}$$

(repeated indices mean summation). If we represent the function $K_{j_1j_2}(\mathbf{k}, i\omega_n)$ in the form of a sum of all possible diagrams, starting with the sublattice line j_1 and ending with the line j_2, then $\Sigma_{j_1j_2}(\mathbf{k}, i\omega_n)$ denotes the sum of all the irreducible diagrams with initial line j_1 and final line j_2. From the system (1.13) we can determine the correlation functions

$$\begin{aligned} K_{11}(\mathbf{k}, i\omega_n) &= \frac{\Sigma_{11}(\mathbf{k}, i\omega_n)}{1+\beta I_\mathbf{k}(\Sigma_{12}+\Sigma_{21}) + (\beta I_\mathbf{k})^2(\Sigma_{12}\Sigma_{21} - \Sigma_{11}\Sigma_{22})}, \\ K_{12}(\mathbf{k}, i\omega_n) &= \frac{\Sigma_{12} + \beta I_\mathbf{k}(\Sigma_{12}\Sigma_{21} - \Sigma_{11}\Sigma_{22})}{1+\beta I_\mathbf{k}(\Sigma_{12}+\Sigma_{21}) + (\beta I_\mathbf{k})^2(\Sigma_{12}\Sigma_{21} - \Sigma_{11}\Sigma_{22})}. \end{aligned} \tag{1.14}$$

Let us calculate now the quantities $\Sigma_{j_1j_2}(\mathbf{k}, i\omega_n)$. In our case the diagrams for the mass operators $\Sigma_{j_1j_2}(\mathbf{k}, i\omega_n)$ are made up of individual single-term blocks interconnected by the effective interaction. The Fourier component of an n-th order block of this type, i.e., one having n interaction lines, can be written in the form[1]

$$\begin{aligned} \hat{\Gamma}_{jn}(\omega_1, \omega_2, \ldots, \omega_n) = \frac{1}{\beta^n}\int_0 dt_1\,dt_2\ldots dt_n\Big[\langle\hat{T}\mathbf{S}_j(t_1)\mathbf{S}_j(t_2)\ldots\mathbf{S}_j(t_n)\rangle_0 \\ - \sum_{m_1+m_2+\ldots+m_k=n}\hat{\Gamma}_{jm_1}\hat{\Gamma}_{jm_2}\ldots\hat{\Gamma}_{jm_k}\Big], \end{aligned} \tag{1.15}$$

where the mean value $\langle\ldots\rangle_0$ is taken with the density matrix $\rho_0 = \exp(-\beta\mathscr{H}_0)/\operatorname{Sp}\exp(-\beta\mathscr{H}_0)$. To calculate the single-cell blocks we can use the diagram technique described in[1]. Here the blocks are expressed in terms of the Green's functions, which are determined in the following manner:

$$\begin{aligned} G(t_1-t_2) &= \frac{1}{b}\langle\hat{T}S^+(t_1)S^-(t_2)\rangle_0, \\ G(\omega_n) &\equiv G_n = 1/(y - i\beta\omega_n), \\ S^\pm &= \frac{1}{\sqrt{2}}(S_x \pm iS_y). \end{aligned} \tag{1.16}$$

We shall represent the block of n-th order by a point with n outgoing lines, each of which is set in correspondence to a definite frequency and momentum, with the momenta and the frequencies conserved in each block. The blocks of the first and second sublattices are shown respectively by solid and dashed lines. The junction point of the solid and dashed lines (Fig. 1a) is set in correspondence with the effective interaction tensor $\hat{\Lambda}_{12\mathbf{q}}(\omega_1, -\omega_1)$:

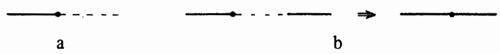


FIG. 1

$$\hat{\Lambda}_{12\mathbf{q}}(\omega_1, -\omega_1) = \frac{-\beta I_\mathbf{q}}{1-(\beta I_\mathbf{q})^2\hat{\Gamma}_2(\omega_1, -\omega_1)\hat{\Gamma}_1(\omega_1, -\omega_1)}. \tag{1.17}$$

The point joining two solid lines (Fig. 1b) corresponds to the effective interaction $\hat{\Lambda}_{11\mathbf{q}}(\omega_1, -\omega_1)$:

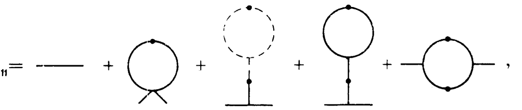
$$\hat{\Lambda}_{11\mathbf{q}}(\omega_1, -\omega_1) = -\hat{\Lambda}_{12\mathbf{q}}(\omega_1, -\omega_1)\beta I_\mathbf{q}\hat{\Gamma}_2(\omega_1, -\omega_1). \tag{1.18}$$

The tensors $\hat{\Lambda}_{21\mathbf{q}}(\omega_1, -\omega_1)$ and $\hat{\Lambda}_{22\mathbf{q}}(\omega_1, -\omega_1)$ are defined analogously. Since the minus sign is introduced in the definition of the tensor $\hat{\Lambda}_{12\mathbf{q}}(\omega_1, -\omega_1)$, all the diagrams are taken with a plus sign. Summation is carried out over all internal frequencies and momenta.

Since one summation over the momentum results in an expression proportional to $1/r_0^3$, where r_0 is the average interaction radius, it follows that the m-th order correction in the expansion in $1/r_0^3$ is represented by the aggregate of all possible connected diagrams containing m closed loops.

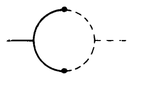
Thus, the operators Σ_{11} and Σ_{12} can be represented, accurate to $1/r_0^3$, by the following diagrams:

$\Sigma_{11}=$



(1.19)

$\Sigma_{12}=$



(1.20)

From the diagram (1.20) we see that Σ_{12} is proportional to $1/r_0^3$. Therefore the product $\Sigma_{12}\Sigma_{21}$ in formulas (1.14) should be left out in the first approximation in the parameter $1/r_0^3$.

The foregoing diagram technique is valid for all values of the temperature and of the magnetic field. For simplicity we shall henceforth consider an antiferromagnet in the absence of a magnetic field at low temperatures. We note that the operators $\Sigma_{j_1 j_2}$ can be written in this case with the aid of the diagrams proposed in[10] for absolute zero temperature. It turns out that this method is a particular case of the method developed here for the low-temperature interval.

2. SPECTRUM OF HEISENBERG ANTIFERROMAGNET

At low temperatures we can put in the expressions for the single-cell blocks b(y) = S and b′(y) = 0. Then the nonzero components of the blocks of second, third and fourth order are given by

$$\begin{aligned}
&\Gamma^{+-}(\omega_1, \omega_2) = SG(\omega_1)\delta(\omega_1+\omega_2),\\
&\Gamma^{+-\zeta}(\omega_1, \omega_2, \omega_3) = -SG(\omega_1)G(-\omega_2)\delta(\omega_1+\omega_2+\omega_3),\\
&\Gamma^{++--}(\omega_1, \omega_2, \omega_3, \omega_4) = -SG(\omega_1)G(\omega_2)(G(-\omega_3)\\
&\quad + G(-\omega_4))\delta(\omega_1+\omega_2+\omega_3+\omega_4),\\
&\Gamma^{+-\zeta\zeta}(\omega_1, \omega_2, \omega_3, \omega_4) = SG(\omega_1)G(-\omega_2)(G(\omega_1+\omega_3)\\
&\quad + G(\omega_1+\omega_4))\delta(\omega_1+\omega_2+\omega_3+\omega_4).
\end{aligned} \tag{2.1}$$

In the absence of an external magnetic field H, the effective-interaction tensors (1.17) and (1.18) can be written in the form

$$\hat{\Lambda}_{12\mathbf{q}}(\omega_1, -\omega_1) = -\beta I_{\mathbf{q}}\left[\frac{\mathbf{e}_2^+\mathbf{e}_1^+ + \mathbf{e}_2^-\mathbf{e}_1^-}{1-(S\beta I_{\mathbf{q}})^2 G(\omega_1)G(-\omega_1)} - \mathbf{e}_2^\zeta\mathbf{e}_1^\zeta\right], \tag{2.2}$$

$$\hat{\Lambda}_{11\mathbf{q}}(\omega_1, -\omega_1) = \frac{S(\beta I_{\mathbf{q}})^2}{1-(S\beta I_{\mathbf{q}})^2 G(\omega_1)G(-\omega_1)} \times [G(\omega_1)\mathbf{e}_2^-\mathbf{e}_2^+ + G(-\omega_1)\mathbf{e}_2^+\mathbf{e}_2^-],$$

where the cyclic unit vectors are given by the expressions

$$\mathbf{e}_j^\pm = \frac{1}{\sqrt{2}}(\mathbf{e}_j^\xi \pm i\mathbf{e}_j^\eta); \tag{2.3}$$

Here the index j = 1, 2 is the number of the sublattice.

Using diagrams (1.19) and (1.20), we can obtain analytic expressions for the operators $\Sigma_{j_1 j_2}(\mathbf{k}, i\omega_n)$:

$$\begin{aligned}
\Sigma_{11} = {} & \mathbf{e}_1^-\mathbf{e}_1^+\left\{SG_n + G_n\sum_{\mathbf{q}}\left[\frac{n_{\mathbf{q}}+{}^1/_2}{\beta\varepsilon_{\mathbf{q}}}(\beta^2\varepsilon_{\mathbf{q}}{}^2G_n - y) - \frac{1}{2}(yG_n - 1)\right]\right\}\\
& + \mathbf{e}_1^+\mathbf{e}_1^-\left\{SG_{-n} + G_{-n}\sum_{\mathbf{q}}\left[\frac{n_{\mathbf{q}}+{}^1/_2}{\beta\varepsilon_{\mathbf{q}}}(\beta^2\varepsilon_{\mathbf{q}}{}^2G_{-n} - y) - \frac{1}{2}(yG_{-n} - 1)\right]\right\}\\
& + \mathbf{e}_1^\zeta\mathbf{e}_1^\zeta\Sigma_{11}{}^{\zeta\zeta},
\end{aligned} \tag{2.4}$$

$$\Sigma_{12} = -S^2G_nG_{-n}\sum_{\mathbf{q}}\frac{n_{\mathbf{q}}+{}^1/_2}{\beta\varepsilon_{\mathbf{q}}}\beta I_{\mathbf{q}}\,\beta I_{\mathbf{k}-\mathbf{q}}(\mathbf{e}_1^+\mathbf{e}_2^+ + \mathbf{e}_1^-\mathbf{e}_2^-) + \Sigma_{12}{}^{\zeta\zeta}\mathbf{e}_1^\zeta\mathbf{e}_2^\zeta. \tag{2.5}$$

Here $\epsilon_q = S(I_0^2 - I_q^2)^{1/2}$ is the frequency of the spin waves in the zeroth approximation[15], and $n_q = (e^{\beta\epsilon_q} - 1)^{-1}$ is the Bose distribution function. In formulas (2.4) and (2.5) we have left out the terms containing the exponentially small quantity $n_y = (e^y - 1)^{-1}$, and did not write out the explicit expressions for the components $\Sigma^{\zeta\zeta}_{j_1 j_2}$, which give an exponentially small contribution to the spin-wave spectrum. By analytic continuation $i\omega_n \to \omega$ from the solution of the equation

$$\det\{1 + \beta I_{\mathbf{k}}[\Sigma_{12}(\mathbf{k},\omega) + \Sigma_{21}(\mathbf{k},\omega)] - (\beta I_{\mathbf{k}})^2\Sigma_{11}(\mathbf{k},\omega)\Sigma_{22}(\mathbf{k},\omega)\} = 0, \tag{2.6}$$

we obtain the spin-wave spectrum

$$\omega_{\mathbf{k}} = \varepsilon_{\mathbf{k}} + \omega_{\mathbf{k}}{}',$$

$$\omega_{\mathbf{k}}{}' = S^2\sum_{\mathbf{q}}\frac{n_{\mathbf{q}}+{}^1/_2}{\varepsilon_{\mathbf{k}}\varepsilon_{\mathbf{q}}}[I_0(I_{\mathbf{k}}{}^2 + I_{\mathbf{q}}{}^2) - I_{\mathbf{k}}I_{\mathbf{q}}I_{\mathbf{k}-\mathbf{q}} - I_0{}^3] + \frac{\varepsilon_{\mathbf{k}}}{2S}. \tag{2.7}$$

Recognizing that in the case of the interaction with the nearest neighbors[10] we have

$$\sum_{\mathbf{q}} I_{\mathbf{k}-\mathbf{q}}f(\mathbf{q}) = \sum_{\mathbf{q}}\frac{I_{\mathbf{k}}I_{\mathbf{q}}}{I_0}f(q),$$

if f(q) depends only on the modulus |**q**|, we obtain for the spin-wave spectrum

$$\omega_{\mathbf{k}} = \varepsilon_{\mathbf{k}}(1 + \Delta/2S), \tag{2.8}$$

where

$$\Delta = \sum_{\mathbf{q}}\left[1 - \left(1 - \left(\frac{I_{\mathbf{q}}}{I_0}\right)^2\right)^{1/2}\right] - 2\sum_{\mathbf{q}} n(\varepsilon_q)\left(1 - \left(\frac{I_{\mathbf{q}}}{I_0}\right)^2\right)^{1/2}. \tag{2.9}$$

Expanding Δ in powers of the temperature, we get

$$\Delta = \sum_{\mathbf{q}}\left[1 - \left(1 - \left(\frac{I_{\mathbf{q}}}{I_0}\right)^2\right)^{1/2}\right] - \frac{6\zeta(4)}{\pi^2\eta^3}\left(\frac{T}{I_0S}\right)^4, \tag{2.10}$$

where $\eta = 3^{-1/2}2^{-1/3}$ for a primitive cubic lattice and $\eta = 1/2$ for a body centered lattice. From (2.8) and (2.9) it follows that the temperature correction to the spectrum of the spin waves is proportional in this approximation to T^4. Such a dependence of ω_k on T agrees with the result obtained in[17] by another method.

In deriving (2.9) and (2.8) we have neglected the exponentially small terms containing b′(y) and n(y), so that these expressions can be used at the temperatures

$$T \ll I_0S \sim T_N/S.$$

It is easy to show that, just as in the case of ferromagnets, for large spins S expressions (2.8) and (2.9) are suitable in a wider temperature interval, particularly when $T_N/S \leq T \ll T_N$ (T_N—Neel temperature). Neglecting the exponentially small terms of order $\exp(-Sy) \sim \exp(-T_N/T)$, we can put here $b(y) = S - n_y$. In the limiting case $I_0S \ll T \ll I_0S^2$, accurate to terms proportional to T/I_0S^2, we get

$$\omega_{\mathbf{k}} = \varepsilon_{\mathbf{k}}(1 + 1/2S - T/I_0S^2). \tag{2.11}$$

It turns out here that the renormalization factor coincides exactly with the factor obtained for ferromagnets.

3. FREE ENERGY OF THE HEISENBERG ANTIFERROMAGNET

To calculate the corrections to the free energy, we shall use the diagram technique developed in Sec. 1. The corrections to $F^{(0)}$ will correspond here to diagrams analogous to the diagram of Fig. 1, but with closed ends.

The first correction $F^{(1)}$ is represented by the following diagrams (Fig. 2):

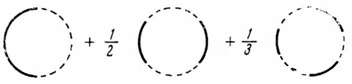


FIG. 2

Here each diagram containing 2n interaction points is proceeded by a factor 1/n, owing to the fact that cyclic permutation of the operators $\mathscr{H}_1(t)$ under the averaging sign does not change the diagram.

The second correction $F^{(2)}$ corresponds to the diagrams of Fig. 3. At low temperatures and H = 0, the diagrams e, e, g, and h make an exponentially small contribution and can be omitted.

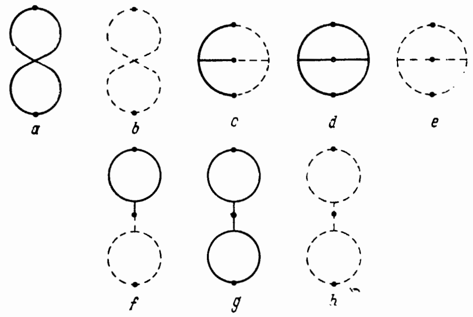


FIG. 3

The foregoing diagrams correspond to the following analytic expressions for the corrections to the free energy F:

$$\beta F^{(1)} = 2 \sum_{\mathbf{q},\,\omega_1} [\ln(\beta\varepsilon_{\mathbf{q}} - i\beta\omega_1) - \ln(y - i\beta\omega_1)], \tag{3.1}$$

$$\beta F^{(2)} = -\frac{1}{\beta S^2 I_0}\left[\sum_{\mathbf{q}}\left(n_{\mathbf{q}}\beta\varepsilon_{\mathbf{q}} - \frac{\beta^2 S^2 I_{\mathbf{q}}^2}{2(y+\beta\varepsilon_{\mathbf{q}})}\right)\right]^2. \tag{3.2}$$

A more general expression for the first correction to the free energy $F^{(1)}$ was obtained in[15], where $F^{(1)}$ should contain a factor 1/2. Using (3.1) and (3.2), we obtain the following value for the internal energy $E = E^{(0)} + E^{(1)} + E^{(2)} + \ldots$:

$$E^{(0)} = -S^2 I_0,$$
$$E^{(1)} = 2\sum_{\mathbf{q}}\left[n_{\mathbf{q}}\varepsilon_{\mathbf{q}} + \frac{1}{2}(\varepsilon_{\mathbf{q}} - SI_0)\right],$$
$$E^{(2)} = -\frac{1}{S^2 I_0}\left\{\left[\sum_{\mathbf{q}}\left(n_{\mathbf{q}}\varepsilon_{\mathbf{q}} - \frac{1}{2}(I_0 S - \varepsilon_{\mathbf{q}})\right)\right]^2 - 2\beta\sum_{\mathbf{q}\mathbf{p}}\left[n_{\mathbf{q}}\varepsilon_{\mathbf{q}} - \frac{1}{2}(I_0 S - \varepsilon_{\mathbf{q}})\right] n_{\mathbf{p}}(n_{\mathbf{p}}+1)\varepsilon_{\mathbf{p}}^2\right\}. \tag{3.3}$$

It follows therefore that

$$E = -I_0 S^2\left[1 + \frac{1}{2S}\sum_{\mathbf{q}}\left(1 - \frac{\varepsilon_{\mathbf{q}}}{I_0 S}\right)\right]^2 + 2I_0 S\frac{3\zeta(4)}{\pi^2\eta^3}\left[1 - \frac{3}{2S}\sum_{\mathbf{q}}\left(1 - \frac{\varepsilon_{\mathbf{q}}}{I_0 S}\right)\right]\left(\frac{T}{I_0 S}\right)^4 + \ldots. \tag{3.4}$$

Since the temperature correction to the energy is proportional to T^4, we get for the specific heat $C \sim T^3$.

The corrections to the average spin $\langle S^\zeta\rangle$ can be calculated from the minimum of the free energy. For the first-approximation correction we get

$$\langle S^\zeta\rangle^{(1)} = -\sum_{\mathbf{q}}\left[\frac{n_{\mathbf{q}}}{\beta\varepsilon_{\mathbf{q}}}y + \frac{1}{2}\left(\frac{y}{\beta\varepsilon_{\mathbf{q}}} - 1\right)\right] \tag{3.5}$$

or

$$\langle S^\zeta\rangle^{(0)} + \langle S^\zeta\rangle^{(1)} = S - \frac{1}{2}\sum_{\mathbf{q}}\left(\frac{I_0 S}{\varepsilon_{\mathbf{q}}} - 1\right) - \frac{\zeta(2)}{2\pi^2\eta^3}\left(\frac{T}{I_0 S}\right)^2. \tag{3.6}$$

Taking into account the equality $I_0 S = 3T_N/(S+1)$, we see from (3.6) that the temperature correction to $\langle S^\zeta\rangle$ coincides with that given in[15].

In the calculation of the corrections to the average value of the spin $\langle S^\zeta\rangle$ in the second approximation, we get

$$\langle S^\zeta\rangle^{(2)} = -\frac{1}{S}\sum_{\mathbf{q}} n_{\mathbf{q}}(n_{\mathbf{q}}+1)\sum_{\mathbf{p}}\left[n_{\mathbf{p}}\beta\varepsilon_{\mathbf{p}} - \frac{\beta}{2}(I_0 S - \varepsilon_{\mathbf{p}})\right]. \tag{3.7}$$

We see from this that when T = 0 the second correction vanishes, and the correction in (3.6), which is proportional to T^2, is multiplied by the coefficient $(1 - S^{-1}\Delta|_{T=0})$.

Expression (3.5) is valid not only for the temperatures $T \ll T_N/S$, but also for large spins in the limiting case $T_N/S \ll T \ll T_N$:

$$\langle S^\zeta\rangle = S\left(1 - \frac{1}{2S} - \sum_{\mathbf{q}}\frac{1}{1 - I_{\mathbf{q}}^2/I_0^2}\frac{T}{I_0 S^2} + \ldots\right). \tag{3.8}$$

From the experiments performed in[8] (known to the authors) on the determination of the magnetization of the sublattices at T = 0, we cannot draw any final conclusion that the theoretical and experimental data agree or disagree. It turns out for example, that in the substances MnF_2, $KMnF_3$ (Jones and Jefferts, Montgomery et al.) they observed a smaller deviation of the average spin from the nominal value than that obtained in the theory in first approximation[5], but in K_2MnF_4 (Loopstra et al.) the experimental and the theoretical results[5] are in agreement.

The result obtained in the present paper for the average spin in second approximation does not affect the foregoing arguments concerning the determination of the sublattice magnetization at T = 0.

It seems that a better agreement between the experimental and the theoretical results on the magnetization at T = 0 can be sought not in an improved approximation for the average spin, but in an analysis of other types of interactions, such as the dipole-dipole interaction, allowance for the anisotropy and interactions with more remote neighbors.

Finally, using the expressions for the correlation functions, let us calculate the statistical magnetic susceptibility[15]

$$\chi_\perp = 2\frac{\beta\mu^2}{v_0}[K_{11}^{+-}(\mathbf{k}=0, \omega=0) + K_{12}^{+-}(\mathbf{k}=0, \omega=0)]. \tag{3.9}$$

In the first approximation we have

$$\chi_\perp = \frac{\mu^2}{v_0}\frac{\langle S^\zeta\rangle}{I_0 S}\frac{1}{1+\Delta/2S}, \tag{3.10}$$

where Δ is determined by (2.9). Substituting $\langle S^\zeta\rangle$ in (3.7), we get

$$\chi_\perp = \frac{\mu^2}{I_0 v_0}\left[1 - \frac{\zeta(2)}{2\pi^2\eta^3 S}\left(\frac{T}{I_0 S}\right)^2 - \frac{1}{2I_0}\sum_{\mathbf{q}}\frac{I_{\mathbf{q}}^2}{\varepsilon_{\mathbf{q}}}\right]. \tag{3.10$'$}$$

Similar calculations for the static magnetic susceptibility $\chi_{\parallel}$ yield

$$\chi_{\parallel} = 2\frac{\beta\mu^2}{v_0}[K_{11}^{\zeta\zeta}(\mathbf{k}=0,\ \omega=0) + K_{12}^{\zeta\zeta}(\mathbf{k}=0,\ \omega=0)]$$
$$= \frac{2\beta\mu^2}{v_0}\sum_{\mathbf{q}} n_{\mathbf{q}}(n_{\mathbf{q}}+1), \tag{3.11}$$

or

$$\chi_{\parallel} = \frac{\mu^2}{S I_0 v_0}\frac{2\zeta(2)}{\pi^2\eta^3}\left(\frac{T}{I_0 S}\right)^2$$

The expressions for the magnetic susceptibility can be obtained also from the free energy. Allowance for the first correction of $\beta F^{(2)}$ leads to a multiplication of the terms in (3.11), proportional to T^2, by a coefficient

$$\left(1 - \frac{3\Delta}{2S}\bigg|_{T=0}\right)$$

We note that similar second-order corrections to $\chi_\perp$, $\chi_{\parallel}$, and $\langle S^\zeta\rangle$ are obtained also in the case when the renormalized spectrum (2.8) is substituted in the expression for the first order corrections.

CONCLUSION

The ground state of antiferromagnets and their behavior at low temperatures was considered by many authors[5-8,10,13,14]. Oguchi[5] and Liu[6] used the Holstein-Primakoff formalism to calculate the corrections to the spin-wave spectrum, to the internal energy of the system, and to the static magnetic susceptibility, and to the sublattice magnetization at low temperatures. However, as is well known, an investigation of antiferromagnets at high temperatures cannot be carried out with the aid of the Holstein-Primakoff method, and the limits of its applicability at low temperatures are unknown. Other authors (see, for example,[10]) generalize Wick's theorem to the spin operators by various methods.

An advantage of our method is its applicability for all temperatures and magnetic fields. In addition, it turns out that the method of Wang and Callen[10] is a particular case of our method, with the theorem on the breakdown of the T-product of spin operators, formulated but not proved by Wang and Callen, being readily proved by our method. The region of applicability of the formulas obtained by us can be pointed out accurately. In antiferromagnets, as well as in ferromagnets, the obtained expression for ω_k and $\langle S^\zeta\rangle$ are applicable also in the interval $T_N/S \leq T \ll T_N$ (for large values of the spins S). In the low-temperature region we obtain agreement with the results of[5,6].

In[7], to calculate the thermodynamic quantities and the spectrum of the spin waves, the method of decoupling the equations for the Green's functions was used. However, as is well known, such a decoupling does not have sufficient justification and, as seen from an examination of these investigations, leads to internally inconsistent results.

The value obtained for the magnetic susceptibility $\chi_{\parallel}$ (Hewson and Ter Haar) coincides with the value of $\chi_{\parallel}$ (see (3.11)) obtained by us in the first approximation. The values of the magnetic susceptibilities ($\chi_{\parallel}$ and $\chi_\perp$) (Lee and Liu) and also $\chi_\perp$ (Hewson and Ter Haar, Lines) coincide approximately with the values of $\chi_{\parallel}$ and $\chi_\perp$ (see (3.11) and (3.10)) obtained in the present paper, i.e., in the expansion in the parameter 1/S the first approximation coincides and the second approximation does not coincide—the factor $(1 - (3\Delta/2S)|_{T=0})$ in the term $(T/I_0S)^2$ differs from the factor in the corresponding term in the investigations listed above.

In addition, the additional terms obtained by lines for $\chi_{\parallel}$, of the order of

$$(2S+1)^2\,\delta^{2S}/2^{2S}, \qquad \delta = \sum_{\mathbf{q}}\left(\frac{I_0 S}{\varepsilon_{\mathbf{q}}} - 1\right),$$

are in error. The quadratic temperature correction to the spin-wave spectrum, calculated by Lee and Liu[7], is incorrect (compare with (2.8) and (2.10)), in view of the fact that in the decoupling of the equation for the Green's function the energy of the spin waves is always proportional to the magnetization.

In the diagram method proposed by us, no such difficulties arise.

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[1] V. G. Vaks, A. I. Larkin, and S. A. Pikin, Zh. Eksp. Teor. Fiz. 53, 281 (1967) [Sov. Phys.-JETP **26**, 188 (1968)].

[2] H. Bethe, Z. Physik **71**, 205 (1931); L. Hulthen, Arkiv Mat. Astron. Fysik, 26A No. 1 (1938).

[3] G. S. Wick, Phys. Rev. **80**, 268 (1950).

[4] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).

[5] T. Oguchi, Phys. Rev. **117**, 117 (1960).

[6] S. H. Liu, Phys. Rev. **142**, 267 (1966).

[7] M. E. Lines, Phys. Rev. **135A**, 1336 (1964); **139A**, 1304 (1965); A. C. Hewson and D. ter Haar, Physica **30**, 890 (1964); K. H. Lee and S. H. Liu, Phys. Rev. **159**, 390 (1967).

[8] H. L. Davis, Phys. Rev. **120**, 789 (1960).

[9] R. B. Stinchcombe, G. Horwitz, F. Englert, and R. Brout, Phys. Rev. **130**, 155 (1963).

[10] Y. L. Wang, S. Shtrikman, and H. Callen, Phys. Rev. **148**, 419 (1966).

[11] B. Giovannini, Scientific Papers of the College of General Education, University of Tokyo, 15, 49 (1965); B. Giovannini and S. Koide, Progr. Theor. Phys. **34**, 705 (1965).

[12] S. Doniach, Phys. Rev. **144**, 382 (1966).

[13] T. Arai and B. Goodman, Phys. Rev. **155**, 514 (1967).

[14] R. P. Kenan, Phys. Rev. **159**, 430 (1967).

[15] M. A. Savchenko and V. V. Tarasenko, Fiz. Tverd. Tela 9, 3284 (1967) [Sov. Phys.-Solid State 9, 2584 (1967)].

[16] A. A. Abrikosov, L. P. Gor'kov, and I. E. Dzyaloshinskiĭ, Metody kvantovoi teorii polya v statisticheskoi fizike, Fizmatgiz, 1962 [Quantum Field Theoretical Methods in Statistical Physics, Pergamon, 1965].

[17] M. Bloch, J. Appl. Phys. **34**, 1151 (1963).

[18] E. D. Jones and K. B. Jefferts, Phys. Rev. **135A**, 1277 (1964); H. Montgomery, D. T. Teaney, and W. A. Walsh, Jr., Phys. Rev. **128**, 80 (1962); B. O. Loopstra, B. Van Laar, and D. I. Breed, Phys. Lett. **26A**, 526 (1968).

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