

On the Green's Functions of the Heisenberg Spin Systems<sup>\*)</sup>Kyozi KAWASAKI<sup>\*\*)</sup> and Hazime MORI*Research Institute for Fundamental Physics  
Kyoto University, Kyoto*

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This paper is concerned with the study of the static spin pair correlation and the susceptibilities of ferromagnetics with spin 1/2 by the method of two-time Green's function of Bogolyubov and Tyablikov adopting the Heisenberg model. A marked difference is found in the behavior of these quantities for the systems with anisotropic and isotropic exchange interactions. In particular, for the case of the isotropic exchange interaction, the parallel susceptibility diverges below the Curie point down to the spin wave region. Consequently, the static spin pair correlation of the longitudinal component of spins shows the behavior with the distance which is different from that previously known. Due to the same cause, the susceptibility immediately above the Curie temperature deviates from the Curie-Weiss law.

## § 1. Introduction

The critical scattering of neutrons by ferro-, antiferro-, and ferrimagnetic substances near the transition points has been a subject of many recent theoretical and experimental investigations. The theoretical interest in this problem resides in the behavior of the spin pair correlations in the vicinity of the transition points, as first pointed out by van Hove.<sup>1)</sup> The spin pair correlation for the localized spin model is defined as

$$\langle S_m^\alpha(0) S_f^{-\alpha}(t) \rangle \equiv \frac{\text{Tr} S_m^\alpha e^{iHt/\hbar} S_f^{-\alpha} e^{-iHt/\hbar} e^{-\beta H}}{\text{Tr} e^{-\beta H}}, \quad (1.1)$$

where  $S_m^\alpha$  and  $S_f^{-\alpha}$  denote the spin operators at the lattice sites  $m$  and  $f$ , respectively, and  $H$  the system Hamiltonian. The following notations have been used:

$$S_m^0 \equiv S_m^z, \quad S_m^\pm \equiv S_m^x \pm iS_m^y. \quad (1.2)$$

The static spin pair correlations, which are defined by setting  $t$  equal to zero in (1.1), have since been investigated on the microscopic basis by de Gennes<sup>2)</sup> in the molecular field approximation, and by Elliott and Marshall<sup>3)</sup> in the Bethe-Peierls' approximation. The dynamical behavior of the spin pair correlations has been studied by de Gennes<sup>2)</sup> and by the present authors.<sup>4)</sup>

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<sup>\*\*)</sup> Present address: Department of Applied Physics, Faculty of Engineering, Nagoya University, Nagoya, Japan.

The present paper is concerned with the study of the static spin pair correlations, in particular, of the correlation of the longitudinal component, employing the method of Green's functions. This method, first introduced by T. Matsubara,<sup>5)</sup> has recently been successfully applied to various problems in statistical physics. In particular, Bogolyubov and Tyablikov<sup>6)</sup> introduced the two-time Green's functions and applied it to ferromagnetics, and derived an integral equation determining the magnetization, which is supposed to be valid in the entire region of temperatures. In the present paper we shall also employ the method of two-time Green's functions.

In § 2, we shall calculate the static correlation of the transverse components of spins. In § 3, we obtain an equation determining the magnetization as a function of the temperature and an external magnetic field. Using this equation we shall discuss the longitudinal susceptibility in some detail. It is shown that in the vicinity and below the Curie temperature, the longitudinal susceptibility shows a quite different behavior depending upon whether the exchange interaction is anisotropic or isotropic. In particular, for the isotropic exchange interaction and in the absence of an external field, the parallel susceptibility diverges below the Curie point, and does not follow the Curie-Weiss law in the immediate neighborhood of the Curie point. This particular feature is reflected in the static pair correlation of the longitudinal components of spins, which is treated in § 4. In Appendix B, we shall show that the correct temperature-dependent spin wave spectrum equivalent to Dyson's<sup>7)</sup> and Keffer and Loudon's<sup>8)</sup> theories is obtained in the second approximation of the Bogolyubov-Tyablikov approximation scheme.

### § 2. The two-time Green's function and the static correlation of perpendicular components of spins

Throughout this paper, we deal with the Heisenberg model of ferromagnetics with a uniaxial anisotropy and a homogeneous magnetic field  $H_e$  in the direction of anisotropy and shall write the Hamiltonian in the following form:

$$H = - \sum_{\substack{m, f \\ (m \neq f)}} [(1/2) K_{mf} (S_m^+ S_f^- + S_m^- S_f^+) + J_{mf} S_m^0 S_f^0] - \sum_m \omega^0 S_m^0, \quad (2.1)$$

where

$$\omega^0 = g \mu_B H_e.$$

$K_{mf}$  and  $J_{mf}$  denote the exchange interactions which may be anisotropic. For  $K_{mf} = J_{mf}$ , (2.1) reduces to the isotropic exchange interaction, whereas, for  $K_{mf} = 0$ , this is the Ising model Hamiltonian. The two-time Green's functions are defined as

$$G_{mf}^{ret}(t-t') \equiv \langle\langle S_m^+(t); S_f^-(t') \rangle\rangle_r \equiv -i\theta(t-t') \langle [S_m^+(t), S_f^-(t')] \rangle, \quad (2.2)$$

$$G_{mf}^{ad}(t-t') \equiv \langle\langle S_m^+(t); S_f^-(t') \rangle\rangle_a \equiv i\theta(t'-t) \langle [S_m^+(t), S_f^-(t')] \rangle, \quad (2.3)$$

where the step function  $\theta(t)$  is defined by

$$\theta(t) = \begin{cases} 1, & t > 0, \\ 0, & t < 0, \end{cases} \quad (2.4)$$

and

$$\langle A \rangle \equiv \frac{\text{Tr} A e^{-\beta H}}{\text{Tr} e^{-\beta H}}. \quad (2.5)$$

There exists an important property of these Green's functions due to Bogolyubov and Tyablikov.<sup>9)</sup> Let us introduce the Fourier transforms of these Green's functions with respect to time

$$G_{mf}^{\text{ret. or ad.}}(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} G_{mf}^{\text{ret. or ad.}}(t) e^{i\omega t} dt. \quad (2.6)$$

According to these authors, the left-hand side of this expression can be continued analytically in the complex  $\omega$ -plane to define a single analytic function  $G_{mf}(\omega)$  which has singularities on the real axis in this plane. With the use of this function, they derived an important relation called the spectral theorem connecting the Green's function and the correlation function. Namely,

$$\langle S_f^-(t') S_m^+(t) \rangle = i \int_{-\infty}^{\infty} \frac{e^{-i\omega(t-t')}}{e^{\beta\omega} - 1} [G_{mf}(\omega + i\varepsilon) - G_{mf}(\omega - i\varepsilon)] d\omega, \quad (2.7)$$

where  $\varepsilon$  is a small positive number. In particular, for  $t=t'$  this reduces to

$$\langle S_f^- S_m^+ \rangle = i \int_{-\infty}^{\infty} \frac{G_{mf}(\omega + i\varepsilon) - G_{mf}(\omega - i\varepsilon)}{e^{\beta\omega} - 1} d\omega. \quad (2.8)$$

As one can easily verify, both of the Green's functions (2.2) and (2.3), which we simply denote as  $G_{mf}(t-t')$ , satisfy the following equations of motion:

$$i \frac{d}{dt} G_{mf}(t-t') = \delta(t-t') \langle [S_m^+, S_f^-] \rangle + \omega_0 G_{mf}(t-t') - 2 \sum_q [K_{mq} \langle S_m^0(t) S_q^+(t); S_f^-(t') \rangle - J_{mq} \langle S_q^0(t) S_m^+(t); S_f^-(t') \rangle]. \quad (2.9)$$

In Bogolyubov and Tyablikov's approximation, we have

$$\left. \begin{aligned} \langle S_m^0(t) S_q^+(t); S_f^-(t') \rangle &\cong \sigma G_{qf}(t-t'), \\ \langle S_q^0(t) S_m^+(t); S_f^-(t') \rangle &\cong \sigma G_{mf}(t-t'), \quad \sigma \equiv \langle S_m^0 \rangle. \end{aligned} \right\} \quad (2.10)$$

Thus, Eq. (2.9) leads to

$$i \frac{d}{dt} G_{mf}(t-t') = 2\delta_{mf} \sigma \delta(t-t') + [\omega_0 + 2\sigma J(0)] G_{mf}(t-t') - 2\sigma \sum_q K_{mq} G_{qf}(t-t'), \quad (2.11)$$

where

$$J(0) \equiv \sum_f J_{mf}.$$

This equation can readily be solved if we introduce the Fourier transform  $G_q(\omega)$  of  $G_{mf}(t-t')$  with respect to the time and the lattice sites by the following :

$$G_{mf}(\omega) = \frac{1}{N} \sum_q e^{i(q, m-f)} G_q(\omega). \tag{2.12}$$

We thus obtain

$$G_q(\omega) = \frac{1}{\omega - \omega_0 - 2\sigma J_K(0, \mathbf{q})} \frac{\sigma}{\pi}, \tag{2.13}$$

where

$$J_K(\mathbf{q}, \mathbf{q}') \equiv J(\mathbf{q}) - K(\mathbf{q}'). \tag{2.14}$$

Here,  $J(\mathbf{q})$  and  $K(\mathbf{q}')$  stand for the Fourier components of  $J_{mf}$  and  $K_{mf}$  defined as

$$J_{mf} = \frac{1}{N} \sum_q e^{i(q, m-f)} J(\mathbf{q}), \quad K_{mf} = \frac{1}{N} \sum_q e^{i(q, m-f)} K(\mathbf{q}). \tag{2.15}$$

Use of (2.8) and (2.13) immediately yields

$$\langle S_f^- S_m^+ \rangle = \frac{2\sigma}{N} \sum_q \frac{1}{e^{\beta\omega_q} - 1} e^{i(q, m-f)}, \tag{2.16}$$

where

$$\omega_q \equiv \omega_0 + 2\sigma J_K(0, \mathbf{q}). \tag{2.17}$$

If we write the static pair correlation function of perpendicular components of spins as

$$\langle S_m^x S_f^x \rangle = \frac{1}{N} \sum_q \phi^{xx}(\mathbf{q}) e^{i(q, m-f)}, \text{ etc.}, \tag{2.18}$$

$\phi^{xx}(\mathbf{q})$ , etc., can be expressed by making use of (2.16) as

$$\phi^{xx}(\mathbf{q}) = \phi^{yy}(\mathbf{q}) = \frac{\sigma}{2} \frac{e^{\beta\omega_q} + 1}{e^{\beta\omega_q} - 1}. \tag{2.19}$$

Essentially the same result was obtained by a different method.\*)

\*) In our earlier work,<sup>4)</sup> (2.19) followed if we identify  $\omega_q$  as the frequency of the collective motion of the spin system, neglecting the shift and damping coming from the higher moments. The two approximations are, however, not equivalent. For instance, Bogolyubov and Tyablikov's approximation adopted in this paper failed to take into account the spin wave interactions correctly. See Appendix B.

In the low temperature limit, where  $\sigma$  reduces to  $S$ , the magnitude of the spin,  $\omega_q$  equals the spin wave frequency. In this case, the result (2.19) reduces to that obtained by van Hove.<sup>1)</sup> On the other hand, in the vicinity of the Curie point and for zero magnetic field, we must distinguish the two cases. Use of (2.17) and (2.19) lead to the following results.

(1) Above the Curie point:

$$\phi^{xx}(\mathbf{q}) = k_B T \left/ \left[ \frac{(g\mu_B)^2}{\chi_{11}} + 2J_K(0, \mathbf{q}) \right] \right., \quad (2.20)$$

where  $\chi_{11}$  is the parallel susceptibility for a single spin defined by

$$\chi_{11} = g\mu_B \frac{\sigma}{H_e} = (g\mu_B)^2 \frac{\sigma}{\omega_0}. \quad (2.21)$$

(2) Below the Curie point:

$$\phi^{xx}(\mathbf{q}) = \frac{k_B T}{2J_K(0, \mathbf{q})}. \quad (2.22)$$

For the isotropic exchange interaction  $J(\mathbf{q}) = K(\mathbf{q})$ , these are the familiar results obtained earlier.<sup>1),2)</sup> In particular, below the Curie point and for the small values of  $q$ ,  $J_J(0, \mathbf{q}) \cong J_0 a^2 q^2$ , where  $J_0$  is the magnitude of the exchange interaction and  $a$  the lattice constant. This means that the transverse components of spins have the spatial correlations of infinite range. For the anisotropic exchange interaction and below the Curie point, we can assume  $J(0) > K(0)$ , because otherwise the spin arrangements in which the spontaneous magnetization occurs in the  $z$ -direction would be unstable, as one can see from (2.17) that the spin wave frequency  $2SJ_K(0, \mathbf{q})$  becomes negative for certain values of  $q$ . Then, for the small values of  $q$ ,  $\phi^{xx}(\mathbf{q})$  takes the following form,

$$\phi^{xx}(\mathbf{q}) = \frac{k_B T}{2J_0 a^2} \frac{1}{q^2 + \kappa^2}, \quad (2.23)$$

where

$$\kappa^2 = \frac{J(0) - K(0)}{K_0 a^2}, \quad (2.24)$$

and we have put  $K(0) - K(\mathbf{q}) \cong K_0 a^2 q^2$ ,  $K_0$  being the magnitude of the exchange interaction of spins in the  $x, y$ -direction. This means that the anisotropy of the exchange interaction makes the range of correlation of the transverse components of spins, which is equal to  $1/\kappa$ , finite.

### § 3. The Magnetization and the parallel susceptibility

If we restrict ourselves to the case of spin 1/2, Bogolyubov and Tyablikov have shown that the method of the preceding section allows us to obtain a closed

equation for the average value of a spin  $\sigma$ . Setting  $f=m$  in (2.16) and noting the relation  $S_m^- S_m^+ = 1/2 - S_m^0$  valid for spin 1/2, (2.16) reduces to

$$\frac{1}{4\sigma} - \frac{1}{2} = \frac{1}{N} \sum_{\mathbf{q}} \frac{1}{\exp(\beta[\omega_0 + 2\sigma J_K(0, \mathbf{q})]) - 1} \tag{3.1}$$

This is the equation determining  $\sigma$  as a function of temperature and a magnetic field. For the case of the isotropic exchange interaction, the solution of this equation was discussed in detail by Tyablikov.<sup>9)</sup> Therefore, we shall not discuss this equation in general, except in the neighborhood of the Curie point, where there is a marked difference in the longitudinal susceptibility between the isotropic and anisotropic exchange interactions. Such a difference also exists in the parallel susceptibility in the entire region below the Curie point which is discussed below, and hence in the static correlation of the longitudinal components of the spins, which is the subject of the next section. Let us start with the parallel susceptibility above the Curie point.

The external field is set to zero in this section. Then setting  $\sigma = \chi' \omega_0$  in (3.1), where  $\chi'$  is connected to the parallel susceptibility  $\chi$  by  $\chi' = \chi / (g\mu_B)^2$ , and making  $\omega_0$  tend to zero, we obtain the equation for  $\chi'$

$$\frac{1}{4\chi'} = \frac{1}{\beta N} \sum_{\mathbf{q}} \frac{1}{1 + 2\chi' J_K(0, \mathbf{q})} \tag{3.2}$$

At the temperature defined by

$$\frac{\beta_c}{2} = \frac{1}{N} \sum_{\mathbf{q}} \frac{1}{J_K(0, \mathbf{q})} \tag{3.3}$$

$\chi'$  is seen to be infinite. Next we calculate the spontaneous magnetization  $\sigma_0$  below the Curie point, which is determined by the equation,

$$\frac{1}{4\sigma_0} - \frac{1}{2} = \frac{1}{N} \sum_{\mathbf{q}} \frac{1}{\exp(2\beta\sigma_0 J_K(0, \mathbf{q})) - 1} \tag{3.4}$$

In the vicinity of the Curie point, the right-hand side of this equation can be expanded as

$$\frac{1}{2\beta\sigma_0} \frac{1}{N} \sum_{\mathbf{q}} \frac{1}{J_K(0, \mathbf{q})} \left\{ 1 - \beta\sigma_0 J_K(0, \mathbf{q}) + \frac{1}{3} \beta^2 \sigma_0^2 J_K^2(0, \mathbf{q}) + \dots \right\}.$$

Applying (3.3) to the first term of this expression and considering the fact that

$$\frac{1}{N} \sum_{\mathbf{q}} 1 = 1,$$

Eq. (3.4) becomes

$$\frac{\beta - \beta_c}{2} = \frac{(\beta\sigma_0)^2}{3} \frac{1}{N} \sum_{\mathbf{q}} J_K(0, \mathbf{q}), \tag{3.5}$$

which expresses that the spontaneous magnetization  $\sigma_0$  is proportional to  $\sqrt{\beta - \beta_c}$

in the neighborhood of the temperature defined by  $T_c$ . These facts allow us to identify the temperature  $T_c = 1/k_B\beta_c$  with the Curie temperature.

In order to discuss the parallel susceptibility, we must distinguish the two cases with anisotropic and isotropic exchange interactions. The case with an anisotropic exchange interaction will be considered first. Below the Curie point, differentiating the both sides of (3.1) with respect to  $\omega_0$ , considering that  $\sigma$  is a function of  $\omega_0$ , setting  $\omega_0$  to zero, and defining  $\chi' \equiv \partial\sigma/\partial\omega_0|_{\omega_0=0}$ , we obtain

$$\chi' = \frac{\beta}{N} \sum_{\mathbf{q}} \frac{\exp(2\beta\sigma_0 J_K(0, \mathbf{q}))}{[\exp(2\beta\sigma_0 J_K(0, \mathbf{q})) - 1]^2} \left\{ \frac{1}{4\sigma_0^2} - \frac{2\beta}{N} \sum_{\mathbf{q}} \frac{J_K(0, \mathbf{q}) \exp(2\beta\sigma_0 J_K(0, \mathbf{q}))}{[\exp(2\beta\sigma_0 J_K(0, \mathbf{q})) - 1]^2} \right\}. \quad (3.6)$$

Near the Curie point, the expression

$$\frac{1}{N} \sum_{\mathbf{q}} \frac{J_K(0, \mathbf{q}) \exp(2\beta\sigma_0 J_K(0, \mathbf{q}))}{[\exp(2\beta\sigma_0 J_K(0, \mathbf{q})) - 1]^2}, \quad (3.7)$$

which appears in the denominator of (3.6), is expanded as

$$\frac{1}{4\sigma_0^2\beta^2} \frac{1}{N} \sum_{\mathbf{q}} \frac{1}{J_K(0, \mathbf{q})} \left[ 1 - \frac{1}{3}\beta^2\sigma_0^2 J_K^2(0, \mathbf{k}) + \dots \right].$$

Use of (3.3) and (3.5) simplifies the first two terms of this expression to the following:

$$\frac{2\beta_c - \beta}{8\sigma_0^2\beta^2}. \quad (3.8)$$

Substituting this into (3.6), and expanding the numerator and omitting the higher order terms in  $\beta - \beta_c$ , we finally arrive at the longitudinal susceptibility  $\chi'$  below and in the neighborhood of the Curie point,

$$\chi' = \frac{1}{2(\beta - \beta_c)} \frac{1}{N} \sum_{\mathbf{q}} \frac{1}{[J_K(0, \mathbf{q})]^2}, \quad (T < T_c). \quad (3.9)$$

The susceptibility above the Curie point is obtained more simply. Expanding (3.2) in  $1/\chi'$ , we have

$$\frac{1}{2} = \frac{1}{\beta N} \sum_{\mathbf{q}} \frac{1}{J_K(0, \mathbf{q})} - \frac{1}{2\beta N\chi'} \sum_{\mathbf{q}} \frac{1}{[J_K(0, \mathbf{q})]^2} + \dots \quad (3.10)$$

Use of (3.3) immediately yields

$$\chi' = \frac{1}{\beta_c - \beta} \frac{1}{N} \sum_{\mathbf{q}} \frac{1}{[J_K(0, \mathbf{q})]^2}, \quad (T > T_c). \quad (3.11)$$

Comparing (3.9) and (3.11), we conclude that the parallel susceptibility near the Curie point is inversely proportional to the temperature distance from the Curie point, the inverse proportionality constant above the Curie point being twice of that below. Such a feature is familiar in the behaviors of various thermodynamic quantities near the transition point of the second order transition.

In particular, for  $K(\mathbf{q})=0$ , that is, for the Ising system, the results obtained in this section reduce to those of the Weiss approximation. In fact Bogolyubov and Tyablikov's approximation is equivalent to the Weiss approximation for the Ising system. For the Ising system,  $\langle\langle S_m^0(t)S_j^+(t); S_j^-(t') \rangle\rangle$  in the right-hand side of (2.9) is missing. Thus the approximation (2.10) is equivalent to replacing the effect of the neighboring spins on the spin  $m$  by the average molecular field.

Let us now turn to the case of the isotropic exchange interaction. For the small value of  $q$ ,

$$J(0, \mathbf{q}) \equiv J_K(0, \mathbf{q}) \cong J_0 a^2 q^2, \tag{3.12}$$

where  $J_0$  is the magnitude of the exchange interaction. Thus, the inverse proportionality constants in (3.9) and (3.11) both diverge if we replace the sum over  $q$  by the integral. This means that the foregoing analysis cannot be directly applicable to this case. Let us first consider the parallel susceptibility above the Curie point. If we write  $\chi'$  as

$$\chi' = 1/2z, \tag{3.13}$$

the temperature-dependent parameter  $z$  is determined by the following equation, obtained by substituting (3.13) into (3.2),

$$\frac{\beta}{2} = \frac{1}{N} \sum_q \frac{1}{z + J(0, \mathbf{q})}, \tag{3.14}$$

where

$$J(\mathbf{q}, \mathbf{q}') \equiv J(\mathbf{q}) - J(\mathbf{q}').$$

The equations similar to (3.13) and (3.14) were obtained by Englert,<sup>10)</sup> who pointed out that they are equivalent to the results of the spherical model.<sup>11)</sup> At the Curie point, which is defined by (3.3), or in this case by

$$\frac{\beta_c}{2} = \frac{1}{N} \sum_q \frac{1}{J(0, \mathbf{q})}, \tag{3.15}$$

$z$  vanishes, as one expects. In order to study the behavior of  $z$  near the Curie point, subtract (3.14) from the both sides of (3.15), to obtain

$$\frac{\beta_c - \beta}{2} = \frac{z}{N} \sum_q \frac{1}{J(0, \mathbf{q}) [z + J(0, \mathbf{q})]} \tag{3.16}$$

$$= \frac{z}{2\pi^2 \rho} \int_0^{q_m} \frac{q^2}{J(0, \mathbf{q}) [z + J(0, \mathbf{q})]} dq, \tag{3.16'}$$

where  $\rho$  is the number of spins per unit volume,  $q_m$  the upper limit of the integration of the order  $1/a$ , and we have assumed that  $J(\mathbf{q})$  is a function of only the magnitude of  $q$ . The above integral diverges at the small values of  $q$



when  $z$  is zero. Therefore, in the vicinity of the Curie point, where  $z$  is very small, the main contribution to the integral comes from the small values of  $q$ . This permits us to use the approximation (3.12) in the integrand and extend the upper limit of the integration to infinity. Thus, the right-hand side of (3.16) becomes

$$\sqrt{z}/4\pi\rho J_0^{3/2} a^3.$$

The use of (3.13), then, yields

$$\chi' = \frac{1}{8\pi^2\rho^2 J_0^3 a^6} \frac{1}{(\beta_c - \beta)^2}. \quad (3.17)$$

On the other hand, at the high temperatures, where  $z$  is large, one easily obtains the following expression of  $z$  by expanding (3.16) in  $1/z$ ,

$$z = \frac{2}{\beta} - J(0) + \dots = \frac{2}{\beta} - \frac{2}{\beta_w} + \dots, \quad (3.18)$$

where  $\beta_w \equiv 2/J(0)$  corresponds to the Curie temperature in the Weiss approximation which is higher than the Curie temperature in our case. Substituting this into (3.13), we obtain the Curie-Weiss law for  $\chi'$ . We thus conclude that for the case of the isotropic exchange interaction in Bogolyubov and Tyablikov's approximation, the Curie-Weiss law with the Curie temperature in the Weiss approximation holds only at the very high temperatures and in the neighborhood of the Curie temperature the susceptibility grows with the law of the inverse square of the temperature distance from the Curie point.

In the Bethe-Peierls approximation, for both anisotropic and isotropic exchange interactions, the Curie temperature lies somewhat below that of the Weiss approximation, and the curve of  $1/\chi$  versus temperature becomes concave towards the top. This is also the general feature of the observed susceptibilities, and it is usually attributed to the effect of the short range order remaining above the Curie point. We want to point out that, as we have seen, the same tendency is produced by the quantum nature of the Heisenberg ferromagnet, which is absent in the Ising system.

#### § 4. The static correlations of the longitudinal components of spins

In the vicinity of the Curie temperature and above, the fluctuation of the longitudinal component of spins plays an important role along with those of the transverse components. The foregoing method, however, cannot be directly applied to obtain the static correlation of the longitudinal component. This is because the operators of the longitudinal components of spins commute with each other, and consequently the inhomogeneous term in the equation for Green's functions corresponding to (2.9) or (2.11) vanishes. It should in principle be possible to make a step further into the hierarchy of equations and start from

the simultaneous equations for the Green's functions involving three and four spins operators, but unfortunately such an approach has not been successful due to other mathematical difficulties.

Thus we have to look for the other approach. Let us suppose that a vanishingly small magnetic field  $h$  in the  $z$ -direction is applied to a particular spin, say, situated at the origin of the coordinates. The total Hamiltonian now becomes

$$\mathcal{H} = H - g\mu_B h S_0^z. \tag{4.1}$$

Then, the average value  $\sigma_m$  of the  $z$ -component of the spin situated at another site  $m$  would be, neglecting the higher order terms in  $h$ ,

$$\sigma_m \equiv \langle S_m^z \rangle = \sigma + g\mu_B h \beta \Phi_m, \tag{4.2}$$

where the first term  $\sigma$  gives the average in the absence of  $h$  and the second term expresses the effect of  $h$ . The quantity  $\Phi_m$  is defined by

$$\Phi_m \equiv \frac{\text{Tr} \int_0^\beta d\lambda e^{-\beta H} (S_m^z - \sigma) e^{-\lambda H} (S_0^z - \sigma) e^{\lambda H}}{\beta \text{Tr} e^{-\beta H}}. \tag{4.3}$$

This, in general, is not the same as the static correlation defined by

$$\langle (S_m^z - \sigma) (S_0^z - \sigma) \rangle = \frac{\text{Tr} e^{-\beta H} (S_m^z - \sigma) (S_0^z - \sigma)}{\text{Tr} e^{-\beta H}}. \tag{4.4}$$

In the following, we shall be interested in the Fourier transform of  $\Phi_m$  for the small wave vectors  $k$  defined by

$$\Phi(\mathbf{k}) = \sum_m \Phi_m e^{-i\mathbf{k}\cdot\mathbf{m}}. \tag{4.5}$$

For  $k=0$ , we have

$$\Phi(0) = \sum_m \langle (S_m^z - \sigma) (S_0^z - \sigma) \rangle. \tag{4.6}$$

This follows from the fact that the total spin operator commutes with the Hamiltonian. Thus we expect that for the small values of  $k$ ,  $\Phi(\mathbf{k})$  can be regarded as the Fourier transform of the static pair correlation function (4.4). Near the Curie point, the smallness of the spin diffusion constant further corroborates this approximation.<sup>2)</sup>

Thus, the problem reduces to the evaluation of  $\sigma_m$ . For this purpose the method of Bogolyubov and Tyablikov of the previous section is applicable with a suitable modification. Let us start from the equation corresponding to (2.9) for the Green's function  $G_{m\bar{f}}(t-t')$  defined in the same way, which is

$$i \frac{d}{dt} G_{m\bar{f}}(t-t') = \delta(t-t') \langle [S_m^+, S_{\bar{f}}^-] \rangle + [\omega_0 + g\mu_B h \delta_{m,0}] G_{m\bar{f}}(t-t') - 2 \sum_{\bar{g}} [K_{m\bar{g}} \langle \langle S_m^z(t) S_{\bar{g}}^+(t); S_{\bar{f}}^-(t') \rangle \rangle - J_{m\bar{g}} \langle \langle S_{\bar{g}}^z(t) S_m^+(t); S_{\bar{f}}^-(t') \rangle \rangle]. \tag{4.7}$$

Corresponding to (2.10) we make the following approximation,

$$\left. \begin{aligned} \langle S_m^0(t) S_g^+(t); S_f^-(t') \rangle &\cong \sigma_m G_{gf}(t-t'), \\ \langle S_g^0(t) S_m^+(t); S_f^-(t') \rangle &\cong \sigma_g G_{mf}(t-t'). \end{aligned} \right\} \quad (4.8)$$

Equations (4.7) and (4.8) with the relation  $\langle [S_m^+, S_f^-] \rangle = 2\sigma_m \delta_{mf}$  lead to

$$\begin{aligned} i \frac{d}{dt} G_{mf}(t-t') &= 2\sigma_m \delta_{mf} \delta(t-t') + [\omega_0 + 2\sum_g J_{mg} \sigma_g + g\mu_B h \delta_{m,0}] G_{mf}(t-t') \\ &\quad - 2\sum_g \sigma_m K_{mg} G_{gf}(t-t'). \end{aligned} \quad (4.9)$$

The Fourier transformation of this equation with respect to time yields

$$\begin{aligned} \omega G_{mf}(\omega) &= \frac{\sigma_m}{\pi} \delta_{mf} + [\omega_0 + 2\sum_g J_{mg} \sigma_g + g\mu_B h \delta_{m,0}] G_{mf}(\omega) \\ &\quad - 2\sum_g \sigma_m K_{mg} G_{gf}(\omega). \end{aligned} \quad (4.10)$$

This equation can be most conveniently handled by making use of the following matrix notations:

$$\left. \begin{aligned} [\mathbf{G}(\omega)]_{mf} &= G_{mf}(\omega), \quad [\boldsymbol{\sigma}]_{mf} = \sigma_m \delta_{mf}, \quad [\mathbf{J}]_{mf} = J_{mf}, \\ [\mathbf{K}]_{mf} &= K_{mf}, \quad [\mathbf{e}]_{mf} = e_m \delta_{mf}, \end{aligned} \right\} \quad (4.11)$$

where

$$e_m \equiv \omega_0 + 2\sum_g J_{mg} \sigma_g + g\mu_B h \delta_{m,0}. \quad (4.12)$$

Then, (4.10) reduces to the following simple form:

$$\omega \mathbf{G}(\omega) = \frac{\boldsymbol{\sigma}}{\pi} + [\mathbf{e} - 2\boldsymbol{\sigma} \cdot \mathbf{K}] \cdot \mathbf{G}(\omega). \quad (4.13)$$

Solving this with respect to  $\mathbf{G}(\omega)$ , we have

$$\mathbf{G}(\omega) = \frac{1}{\omega - \mathbf{e} + 2\boldsymbol{\sigma} \cdot \mathbf{K}} \cdot \frac{\boldsymbol{\sigma}}{\pi}. \quad (4.14)$$

The spectral theorem of Bogolyubov and Tyablikov, (2.8), can now be written as

$$\langle S_f^- S_m^+ \rangle = i \int_{-\infty}^{\infty} \frac{d\omega}{e^{\beta\omega} - 1} [\mathbf{G}(\omega + i\varepsilon) - \mathbf{G}(\omega - i\varepsilon)]_{mf}. \quad (4.15)$$

Use of (4.14) and the relation

$$\frac{1}{x + i\varepsilon} - \frac{1}{x - i\varepsilon} = -2\pi i \delta(x) \quad (4.16)$$

yields

$$\mathbf{G}(\omega + i\varepsilon) - \mathbf{G}(\omega - i\varepsilon) = -2i\delta[\mathbf{e} - 2\boldsymbol{\sigma} \cdot \mathbf{K} - \omega] \cdot \boldsymbol{\sigma}. \tag{4.17}$$

Substitution of this into (4.15) for  $f=m$  and the use of the relation  $\langle S_m^- S_m^+ \rangle = (1/2) - \sigma_m$  lead to

$$\frac{1}{4\sigma_m} - \frac{1}{2} = \left[ \frac{1}{e^{\beta[\mathbf{e} - 2\boldsymbol{\sigma} \cdot \mathbf{K}]} - 1} \right]_{mm}. \tag{4.18}$$

This is the equation which determines  $\sigma_m$  when a small magnetic field  $h$  is applied to the spin at the origin, and is a generalization of (3.1). We shall investigate the solution of this equation in a few cases of interest.

(1) The case with no uniform magnetization, that is, above the Curie point and in the absence of the uniform magnetic field. The fact that  $\sigma_m$  is a small quantity, proportional to  $h$ , permits us to expand (4.18) and obtain the following equation:

$$\frac{1}{4} = \sigma_m \left[ \frac{1}{\beta[\mathbf{e} - 2\boldsymbol{\sigma} \cdot \mathbf{K}]} \right]_{mm}.$$

This can further be transformed to yield

$$\frac{\beta}{4} = \left[ \left( 1 - 2 \frac{\boldsymbol{\sigma} \cdot \mathbf{K}}{e} \right)^{-1} \cdot \frac{\boldsymbol{\sigma}}{e} \right]_{mm}. \tag{4.19}$$

The important point to note here is that Eq. (4.19) looked upon as an equation determining  $\sigma_m/e_m$  does no longer involve  $h$ . In particular, as one easily verifies, the same equation (4.19) would come out when one applies the same small magnetic field to every spin of the system. This situation makes  $\sigma_m/e_m$  independent of  $m$  and equal to the value of  $\sigma_m/e_m$  when a small uniform magnetic field is applied to the system. Hence, considering the definition of  $e_m$ , (4.12), where  $\omega_0=0$ , we obtain

$$\frac{\sigma_m}{g\mu_B h \delta_{m,0} + 2 \sum_g J_{mg} \sigma_g} = \frac{\chi'}{1 + 2\chi' J(0)}; \tag{4.20}$$

where  $\chi'$  is the susceptibility. This, in turn, is an equation determining  $\sigma_m$ . If we remember the relation (4.2), this equation can be transformed to give the following equation for the static correlation  $\Phi_m$ :

$$\left[ \frac{1}{\chi'} + 2J(0) \right] \Phi_m - 2 \sum_g J_{mg} \Phi_g = \frac{1}{\beta} \delta_{m,0}. \tag{4.21}$$

If we introduce the Fourier transform of  $\Phi_m$ , corresponding to (2.18), by

$$\Phi_m = \frac{1}{N} \sum_q \Phi(\mathbf{q}) e^{i\mathbf{q} \cdot \mathbf{m}}, \tag{4.22}$$

(4.21) is easily solved to yield the following result:

$$\Phi(\mathbf{q}) = k_B T \left/ \left[ \frac{(g\mu_B)^2}{\chi_{11}} + 2J(0, \mathbf{q}) \right] \right. . \quad (4.23)$$

In contrast with the correlation of the transverse components, (2.20), the anisotropy of the exchange interaction, if it exists, appears only through the susceptibility. At the Curie point, where  $\chi_{11}$  tends to infinity, the range of the correlation of the longitudinal components becomes infinite, without being affected by the anisotropy. For the isotropic exchange interaction, the static correlations of the transverse and longitudinal components, (2.20) and (4.23), coincide as one expects. For the small values of  $q$ , with the use of (3.12), (4.23) can be expressed in the form

$$\Phi(\mathbf{q}) = \frac{1}{4r_1^2} \frac{1}{q^2 + \kappa_1^2}, \quad (4.24)$$

where van Hove's parameters<sup>1)</sup>  $r_1$  and  $\kappa_1$  are defined as

$$\begin{aligned} r_1^2 &= J_0 a^2 / 2k_B T, \\ \kappa_1^2 &= \frac{1}{r_1^2} \frac{\chi_0}{\chi_{11}}, \quad \chi_0 \equiv (g\mu_B)^2 / 4k_B T. \end{aligned} \quad (4.25)$$

(2) The case with a nonvanishing uniform magnetization. Because of the non-zero uniform magnetization  $\sigma$ , we put

$$\sigma_m = \sigma + \sigma_m'. \quad (4.26)$$

Then,  $\sigma_m'$  is a small quantity proportional to  $\hbar$ . Correspondingly, we shall put

$$e_m = e_0 + e_m', \quad (4.27)$$

where

$$\left. \begin{aligned} e_0 &\equiv \omega_0 + 2\sigma J(0), \\ e_m' &\equiv 2 \sum_g J_{mg} \sigma_g' + g\mu_B \hbar \delta_{m,0}, \end{aligned} \right\} \quad (4.28)$$

and introduce the matrices  $\sigma'$  and  $e'$  instead of  $\sigma$  and  $e$ , defined by

$$\left. \begin{aligned} [\sigma']_{mf} &= \sigma_m' \delta_{mf}, \\ [e']_{mf} &= e_m' \delta_{mf}. \end{aligned} \right\} \quad (4.29)$$

$e_m'$  is also small, being proportional to  $\hbar$ .

Equation (4.18) now becomes

$$\frac{1}{2} - \sigma - \sigma_m' = 2(\sigma + \sigma_m') \left[ \frac{1}{e^{\beta[e_0 - 2\sigma K] + \beta[e' - 2\sigma' \cdot K]} - 1} \right]_{mm}, \quad (4.30)$$

which determines  $\sigma_m'$ . This equation can be expanded in  $\hbar$ , if we use the following expansion formula for two matrices  $A$  and  $B$ ,  $B$  being assumed to be small compared with  $A$ ,

$$\frac{1}{e^{\beta(A+B)} - 1} \approx \frac{1}{e^{\beta A} - 1} - \frac{1}{e^{\beta B} - 1} \int_0^\beta d\lambda e^{(\beta-\lambda)A} B e^{\lambda A} \frac{1}{e^{\beta A} - 1}. \tag{4.31}$$

In (4.30), the terms independent of  $h$  cancel and equating the terms proportional to  $h$  yields the following equation,

$$\begin{aligned} \frac{\sigma_m'}{2} &= \frac{2\sigma^2}{N^2} \sum_{\mathbf{k}} \sum_{\mathbf{q}} \sum_f \frac{1}{\exp(\beta[\omega_0 + 2\sigma J_K(0, \mathbf{k})]) - 1} \\ &\quad \int_0^\beta d\lambda \exp((\beta - \lambda)[\omega_0 + 2\sigma J_K(0, \mathbf{k})]) \exp(i(\mathbf{k}, \mathbf{m} - \mathbf{f})) \\ &\quad \times [g\mu_B h \delta_{f,0} + 2\sum_g J_{fg} \sigma_g' - 2\sigma_f' K(\mathbf{q})] \exp(\lambda[\omega_0 + 2\sigma J_K(0, \mathbf{q})]) \\ &\quad \times \frac{1}{\exp(\beta[\omega_0 + 2\sigma J_K(0, \mathbf{q})]) - 1} \cdot \exp(-i(\mathbf{q}, \mathbf{m} - \mathbf{f})). \end{aligned} \tag{4.32}$$

This can be simplified, and remembering (4.2), we obtain

$$\frac{1}{2} \phi_m = \frac{2\sigma^2}{N^2} \sum_{\mathbf{k}} \sum_{\mathbf{q}} \sum_f F_K(\mathbf{k}, \mathbf{q}) [k_B T \delta_{f,0} + 2\sum_g J_{fg} \phi_g - 2K(\mathbf{q}) \phi_f] \exp(i(\mathbf{k} - \mathbf{q}, \mathbf{m} - \mathbf{f})) \tag{4.33}$$

where

$$F_K(\mathbf{k}, \mathbf{q}) \equiv \frac{1}{2\sigma K(\mathbf{k}, \mathbf{q})} \left[ \frac{1}{\exp(\beta[\omega_0 + 2\sigma J_K(0, \mathbf{k})]) - 1} - \frac{1}{\exp(\beta[\omega_0 + 2\sigma J_K(0, \mathbf{q})]) - 1} \right]. \tag{4.34}$$

This equation can be solved by the Fourier transformation with the result

$$\phi(\mathbf{q}) = \frac{k_B T \cdot (2\sigma^2/N) \cdot \sum_{\mathbf{k}} F_K(\mathbf{k}, \mathbf{k} - \mathbf{q})}{(1/2) - (4\sigma^2/N) \sum_{\mathbf{k}} F_K(\mathbf{k}, \mathbf{k} - \mathbf{q}) J_K(\mathbf{q}, \mathbf{k} - \mathbf{q})}. \tag{4.35}$$

We study the behavior of  $\phi(\mathbf{q})$  for the small values of  $q$ . As in § 3, the two cases must be distinguished according as whether the exchange interaction is anisotropic or isotropic. For the case with an anisotropic exchange interaction,  $\phi(0)$ , which is essentially the susceptibility, is finite except at the Curie point, thus enabling us to expand  $\phi(\mathbf{q})$  around  $q=0$ . After a somewhat lengthy calculation,  $\phi(\mathbf{q})$  can again be obtained in the form (4.24) with the following definitions of  $\kappa_1$  and  $r_1$ :

$$r_1^2 = \frac{J_0 a^2}{2k_B T} (1 + \hat{\epsilon}), \tag{4.36}$$

$$\kappa_1^2 = \frac{1}{r_1^2} \frac{\chi_0}{\chi_{||}},$$

where

$$\begin{aligned} \xi \equiv & \frac{(g\mu_B)^2}{2J_0\alpha^2\chi_{11}} \cdot \frac{1}{\sum_{\mathbf{q}} F_K(\mathbf{q}, \mathbf{q})} \cdot \sum_{\mathbf{q}} F_K(\mathbf{q}, \mathbf{q}) \left\{ \left[ \beta\sigma \left( 1 + \frac{2\chi_{11}}{(g\mu_B)^2} J_K(0, \mathbf{q}) \right) \right] \right. \\ & \times \frac{\exp(\varphi_q) + 1}{\exp(\varphi_q) - 1} - \frac{2\chi_{11}}{(g\mu_B)^2} \left. \right] \left( -\frac{K'(q)}{3q} + \frac{K''(q)}{6} \right) + \left[ \frac{2\chi_{11}}{(g\mu_B)^2} \frac{\exp(\varphi_q) + 1}{\exp(\varphi_q) - 1} \right. \\ & \left. - \beta\sigma \left( 1 + \frac{2\chi_{11}}{(g\mu_B)^2} J_K(0, \mathbf{q}) \right) \left( \frac{2}{3} + \frac{4\exp(\varphi_q)}{(\exp(\varphi_q) - 1)^2} \right) \right] \frac{\beta\sigma K'(q)^2}{3} \Big\}, \\ & \varphi_q \equiv \beta[\omega_0 + 2\sigma J_K(0, \mathbf{q})]. \quad (4.37) \end{aligned}$$

Above the Curie point and for the vanishingly small external field,  $\xi$  is shown to vanish, in accordance with our earlier result (4.24). In the vicinity of the Curie point, the use of (3.5) and (3.9) yields

$$\begin{aligned} \xi \equiv & \frac{\beta - \beta_c}{J_0\alpha^2} \cdot \frac{1}{N} \sum_{\mathbf{q}} \frac{1}{J_K(0, \mathbf{q})^2} \left\{ \left[ \frac{1}{J_K(0, \mathbf{q})} + \frac{J_K(0, \mathbf{q})}{2} \cdot \frac{\sum_{\mathbf{p}} \frac{1}{J_K(0, \mathbf{p})^2}}{\sum_{\mathbf{p}} J_K(0, \mathbf{p})} \right] \right. \\ & \left. \times \left[ -\frac{K'(q)}{3q} + \frac{K''(q)}{6} \right] - \frac{K'(q)^2}{3J_K(0, \mathbf{q})^2} \right\}, \quad (T < T_c). \quad (4.38) \end{aligned}$$

Thus the curve of  $r_1$  versus temperature has a kink at the Curie point, unless the factor multiplying  $\beta - \beta_c$  in the above expression vanishes.

Now, let us turn to the case of an isotropic exchange interaction. As we have seen in § 3, in the absence of an external magnetic field, the parallel susceptibility, hence  $\phi(\mathbf{q})$ , goes to infinity below the Curie point. Thus the expansion around  $q=0$  is impossible. This is also evident from the expression (4.35), because the numerator diverges for  $q=0$ . Therefore,  $q$  must always be kept finite during the calculation, which is performed in Appendix A. The result is

$$\phi(\mathbf{q}) = \frac{k_B T}{2J_0\alpha^2} \cdot \frac{1}{q(q+\delta)} \quad (4.39)$$

where

$$\delta \equiv \frac{4J_0\alpha^2\rho}{k_B T(1 - (d \ln \sigma / d \ln T))}. \quad (4.40)$$

Near the Curie point,

$$\delta \simeq \frac{8J_0\alpha^2\rho(T_c - T)}{k_B T_c^2}. \quad (4.41)$$

At the Curie point, (4.39) becomes

$$\phi(\mathbf{q}) = \frac{k_B T_c}{2J_0\alpha^2 q^2}. \quad (4.42)$$

This agrees with that obtained by approaching the Curie point from above that point. (4.39) yields the spatial dependence of the correlation function of the form

$$\Phi(R) \propto \begin{cases} 1/R, & T = T_c \\ 1/R^2, & T \neq T_c. \end{cases} \tag{4.43}$$

(4.39) and (4.43) are radically different from those obtained in the Weiss approximation, or those of an anisotropic exchange interaction. In the temperature region such that  $2J_0a^2q^2/k_B \ll T \ll T_c$ , we have

$$\begin{aligned} \delta &\approx \frac{4J_0a^2\rho}{k_B T}, \\ \chi_q &\approx (g\mu_B)^2 \beta \Phi(q), \end{aligned} \tag{4.44}$$

$\chi_q$  being the wave-vector-dependent susceptibility. In this case we further note that

$$q/\delta = qk_B T / 4J_0a^2\rho \sim aqT/T_c \ll 1. \tag{4.45}$$

Thus we obtain from (4.39), (4.44) and (4.45).

$$\chi_q \approx \frac{(g\mu_B)^2 k_B T}{8(J_0a^2)^2 \rho} \frac{1}{q}. \tag{4.46}$$

This is the result of the spin wave theory, (4.43) of reference 4). The peculiar behavior of the static correlation function and the susceptibility below the Curie point in this case is closely related to the similar features which appear in the spin wave region.<sup>12)</sup> The present calculation reveals that, in the Bogolyubov-Tyablikov approximation, such features persist right up to the Curie point, and give rise to the peculiar temperature dependence of the parallel susceptibility above the Curie point, Eq. (3.17). Thus we have three possibilities: (1) The spin wave theory and our calculation are essentially correct. (2) The spin wave theory is correct, but our calculation is not. That is, in the vicinity of the Curie point, the Weiss approximation or the Bethe-Peierls approximation is closer to the truth than the Bogolyubov-Tyablikov approximation. If this is the case, there must be a temperature below the Curie point, below which the parallel susceptibility goes to infinity, but above which it remains finite except at the Curie point. (3) Both the spin wave theory and the Bogolyubov-Tyablikov approximation are incorrect. That is, if we take the spin wave interaction correctly into account, the parallel susceptibility should remain finite even at the lowest temperatures.

At present, it is rather difficult to decide among these three. However, it is hard to imagine a second transition temperature below the Curie point, and the various evidences suggest that the spin wave theory should be essentially correct at low temperatures. Experimentally, it should be possible to decide among these by observing the critical scattering below the Curie point and the susceptibility just above the Curie point. But the small amount of anisotropy energy and external magnetic field would somewhat smear out the differences.



Anyway, more theoretical and experimental works are necessary to settle the question.

### § 5. Conclusion

In the previous sections, we have studied the susceptibility and the static pair correlation functions of the Heisenberg spin system with spin 1/2 by the method of two-time Green's function. A marked difference was revealed for the parallel susceptibility and the static pair correlation function of the longitudinal components of spins below and in the neighborhood of the Curie point for the systems with anisotropic and isotropic exchange interactions. Our results have more similarities to the spin wave results than those of the Weiss or Bethe-Peierls approximation. Although such a difference should show up in the susceptibility measurements and the critical scattering experiments, more theoretical and experimental studies will be necessary to decide the points. First of all it would be necessary to extend our method to higher magnitudes of spins<sup>13)</sup> and to take proper accounts of anisotropy energy, in order to compare our results with the experiments. It should be also interesting to apply the methods of diagrammatical expansion recently developed for this problem.<sup>14)</sup> Finally, we mention that the difference between the systems with anisotropic and isotropic exchange interactions has been noted in various places in the literatures.<sup>11), 15)</sup>

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### Appendix A

Here we shall evaluate  $\varphi(q)$  for the small values of  $q$  for the case of an isotropic exchange interaction. Let us start from the numerator (4.35),

$$C_1(q) \equiv \frac{1}{N} \sum_{\mathbf{k}} F_J(\mathbf{k}, \mathbf{k}-\mathbf{q}) = \frac{1}{\rho (2\pi)^3} \int d\mathbf{k} F_J(\mathbf{k}, \mathbf{k}-\mathbf{q}). \quad (\text{A}\cdot 1)$$

For small  $q$ , we have

$$F_J(\mathbf{k}, \mathbf{k}-\mathbf{q}) \cong \beta \frac{\exp(2\beta\sigma J(0, \mathbf{k}))}{[\exp(2\beta\sigma J(0, \mathbf{k})) - 1][\exp(2\beta\sigma J(0, \mathbf{k}-\mathbf{q})) - 1]}. \quad (\text{A}\cdot 2)$$

Because the integral (A.1) diverges at the small values of  $k$  for  $q=0$ , the main contribution to that integral for small  $q$  comes from the small values of  $k$ . This

allows us to further approximate (A·2) as follows :

$$F_J(\mathbf{k}, \mathbf{k}-\mathbf{q}) \simeq \frac{\beta}{(2\beta\sigma J_0 a^2)^2} \cdot \frac{1}{k^2(k^2-2kqx+q^2)}, \tag{A·3}$$

where  $x$  denotes the cosine of the angle between  $\mathbf{k}$  and  $\mathbf{q}$ . Substitution of this expression into (A·1) yields

$$\begin{aligned} C_1(q) &\simeq \frac{\beta}{\rho(4\pi\beta\sigma J_0 a^2)^2} \int_0^\infty dk \int_{-1}^1 dx \frac{1}{k^2-2kqx+q^2} \\ &= \frac{1}{16\pi^2\rho\beta(\sigma J_0 a^2)^2 q} \int_0^\infty \frac{dk}{k} \ln \left| \frac{k-q}{k+q} \right| \\ &= \frac{1}{32\rho\beta(\sigma J_0 a^2)^2} \cdot \frac{1}{q}. \end{aligned} \tag{A·4}$$

Next we calculate the function  $C_2(q)$  defined as

$$\begin{aligned} C_2(q) &\equiv \frac{1}{q} \cdot \frac{1}{N} \sum_{\mathbf{k}} [F_J(\mathbf{k}, \mathbf{k}-\mathbf{q})J_J(\mathbf{q}, \mathbf{k}-\mathbf{q}) - F_J(\mathbf{k}, \mathbf{k})J_J(0, \mathbf{k})] \\ &= \frac{1}{q} \cdot \frac{1}{\rho(2\pi)^3} \int d\mathbf{k} [F_J(\mathbf{k}, \mathbf{k}-\mathbf{q})J_J(\mathbf{q}, \mathbf{k}-\mathbf{q}) - F_J(\mathbf{k}, \mathbf{k})J_J(0, \mathbf{k})]. \end{aligned} \tag{A·5}$$

The use of (A·2) gives

$$\begin{aligned} C_2(q) &= \frac{\beta}{\rho(2\pi)^3} \int d\mathbf{k} \frac{\exp(2\beta J(0, \mathbf{k}))}{\exp(2\beta\sigma J(0, \mathbf{k})) - 1} \left\{ \frac{J(\mathbf{q}, \mathbf{k}-\mathbf{q})}{\exp(2\beta\sigma J(0, \mathbf{k}-\mathbf{q})) - 1} \right. \\ &\quad \left. - \frac{J(0, \mathbf{k})}{\exp(2\beta\sigma J(0, \mathbf{k})) - 1} \right\} / q. \end{aligned} \tag{A·6}$$

This integral again diverges at the small values of  $k$  for  $q=0$ , thus enabling us to make an approximation similar to (A·3), yielding

$$\begin{aligned} C_2(q) &= -\frac{\beta J_0 a^2}{\rho(4\pi\beta\sigma J_0 a^2)^2} \int_0^\infty dk \int_{-1}^1 dx \frac{q}{k^2-2kqx+q^2} \\ &= -\frac{1}{32\rho\beta\sigma^2 J_0 a^2}. \end{aligned} \tag{A·7}$$

Finally we evaluate the quantity  $C_3$  defined by

$$C_3 \equiv \frac{1}{N} \sum_{\mathbf{k}} F_J(\mathbf{k}, \mathbf{k}) J_J(0, \mathbf{k}) = \frac{1}{\rho(2\pi)^3} \int d\mathbf{k} F_J(\mathbf{k}, \mathbf{k}) J_J(0, \mathbf{k}). \tag{A·8}$$

This can be done most easily by differentiating the equation determining  $\sigma$ , (3·1), or in this case,

$$\frac{1}{4\sigma} - \frac{1}{2} = \frac{1}{\rho(2\pi)^3} \int d\mathbf{k} \frac{1}{\exp(2\beta\sigma J_J(0, \mathbf{k})) - 1} \quad (\text{A}\cdot 9)$$

with respect to  $\beta$ . This leads to

$$-\frac{1}{4\sigma^2} \frac{d\sigma}{d\beta} = -\frac{2}{\beta} \left( \sigma + \beta \frac{d\sigma}{d\beta} \right) \cdot \frac{1}{\rho(2\pi)^3} \cdot \int d\mathbf{k} F_J(\mathbf{k}, \mathbf{k}) J_J(\mathbf{k}, \mathbf{k}),$$

or

$$C_3 = \frac{1}{\rho(2\pi)^3} \int d\mathbf{k} F_J(\mathbf{k}, \mathbf{k}) J_J(\mathbf{k}, \mathbf{k}) = \frac{1}{8\sigma^2} \frac{d \ln \sigma / d \ln \beta}{1 + (d \ln \sigma / d \ln \beta)}. \quad (\text{A}\cdot 10)$$

In terms of  $C_1(q)$ ,  $C_2(q)$  and  $C_3$ ,  $\Phi(q)$  can be written as

$$\Phi(q) = \frac{2\sigma^2 k_B T C_1(q)}{(1/2) - 4\sigma^2 [C_3 + q C_2(q)]}. \quad (\text{A}\cdot 11)$$

The substitution of (A·4), (A·7) and (A·10) leads to (4·39) and (4·40).

### Appendix B

Sometimes, the method of two-time Green's function was questioned because in the lowest order approximation, it failed to reproduce the correct temperature-dependent spin wave spectrum equivalent to Dyson's theory.<sup>7)</sup> Here we shall show that, in the next approximation, the correct spin wave spectrum is obtained.

Consider the Green's function  $G_k(t-t')$  defined by

$$G_k(t-t') \equiv \langle\langle S_k^+(t); S_k^-(t') \rangle\rangle, \quad (\text{B}\cdot 1)$$

where

$$S_k^\alpha \equiv \sum_m e^{-ik \cdot m} S_m^\alpha, \quad \alpha = 0, \pm. \quad (\text{B}\cdot 2)$$

For the system with isotropic exchange interaction,  $G_k(t-t')$  satisfies the following equation of motion:

$$\begin{aligned} i \frac{d}{dt} G_k(t-t') &= 2N\sigma\delta(t-t') + \frac{2}{N} \sum_q J(\mathbf{q}, \mathbf{k}-\mathbf{q}) \langle\langle \{S_q^0(t), S_{k-q}^+(t)\}; S_{-k}^-(t') \rangle\rangle \\ &= 2N\sigma\delta(t-t') + 2\sigma J(0, \mathbf{k}) G_k(t-t') \\ &\quad + \frac{2}{N} \sum_q J(\mathbf{q}, \mathbf{k}-\mathbf{q}) \langle\langle \{S_q^0(t), S_{k-q}^+(t)\}; S_{-k}^-(t') \rangle\rangle, \end{aligned} \quad (\text{B}\cdot 3)$$

where  $\underline{A} \equiv A - \langle A \rangle$  for any operator  $A$  and  $\{A, B\} = (AB + BA)/2$ .

The Green's function appearing in the last term of (B·3) satisfies the equation of motion as follows:

$$i \frac{d}{dt} \langle\langle \{S_q^0(t), S_{k-q}^+(t)\}; S_{-k}^-(t') \rangle\rangle = \frac{1}{N} \sum_r J(\mathbf{r}, \mathbf{q}-\mathbf{r}) \langle\langle \{S_r^+(t), S_{q-r}^-(t)\}, S_{k-q}^+(t)\}; S_{-k}^-(t') \rangle\rangle;$$

$$S_{-k}^-(t') \rangle \rangle + \frac{2}{N} \sum_{\mathbf{r}} J(\mathbf{r}, \mathbf{k} - \mathbf{q} - \mathbf{r}) \langle \langle \{ \underline{S}_{\mathbf{q}}^0(t), \{ S_{\mathbf{r}}^0(t), S_{\mathbf{k}-\mathbf{q}-\mathbf{r}}^+(t) \}; S_{-k}^-(t') \} \rangle \rangle. \tag{B.4}$$

This equation can be reduced first by making the following approximation :

$$\langle \langle \{ \{ S_{\mathbf{r}}^+(t), S_{\mathbf{q}-\mathbf{r}}^-(t) \}, S_{\mathbf{k}-\mathbf{q}}^+(t) \}; S_{-k}^-(t') \} \rangle \rangle \cong \delta_{\mathbf{r}, \mathbf{q}-\mathbf{r}} \delta_{\mathbf{q}, 0} \langle \{ S_{\mathbf{r}}^+, S_{-\mathbf{r}}^- \} \rangle G_{\mathbf{k}}(t-t') \\ + \delta_{\mathbf{r}, \mathbf{k}} \langle \{ S_{\mathbf{k}-\mathbf{q}}^+, S_{-\mathbf{k}+\mathbf{q}}^- \} \rangle G_{\mathbf{k}}(t-t'). \tag{B.5}$$

The first term does not contribute to (B.4). Next, at the low temperature, the following approximation is permissible :

$$\langle \langle \{ \underline{S}_{\mathbf{q}}^0(t), \{ S_{\mathbf{r}}^0(t), S_{\mathbf{k}-\mathbf{q}-\mathbf{r}}^+(t) \}; S_{-k}^-(t') \} \rangle \rangle \cong \delta_{\mathbf{r}, 0} N \sigma \langle \langle \{ \underline{S}_{\mathbf{q}}^0(t), S_{\mathbf{k}-\mathbf{q}}^+(t) \}; S_{-k}^-(t') \} \rangle \rangle. \tag{B.6}$$

Substituting (B.5) and (B.6), Eq. (B.4) becomes

$$i \frac{d}{dt} \langle \langle \{ \underline{S}_{\mathbf{q}}^0(t), S_{\mathbf{k}-\mathbf{q}}^+(t) \}; S_{-k}^-(t') \} \rangle \rangle = \frac{1}{N} J(\mathbf{k}, \mathbf{q} - \mathbf{k}) \langle \{ S_{\mathbf{k}-\mathbf{q}}^+, S_{-\mathbf{k}+\mathbf{q}}^- \} \rangle G_{\mathbf{k}}(t-t') \\ + 2\sigma J(0, \mathbf{k} - \mathbf{q}) \langle \langle \{ \underline{S}_{\mathbf{q}}^0(t), S_{\mathbf{k}-\mathbf{q}}^+(t) \}; S_{-k}^-(t') \} \rangle \rangle. \tag{B.7}$$

Equations (B.3) and (B.7) constitute the simultaneous equations for determining the two kinds of Green's functions involved. These can be solved easily in terms of the Fourier transforms with respect to time. The solution for  $G_{\mathbf{k}}(\omega)$ , the Fourier transform of  $G_{\mathbf{k}}(t-t')$ , becomes

$$G_{\mathbf{k}}(\omega) = \frac{N\sigma}{\pi} \left/ \left\{ \omega - 2\sigma J(0, \mathbf{k}) - \frac{2}{N^2} \sum_{\mathbf{q}} \frac{J(\mathbf{q}, \mathbf{k} - \mathbf{q}) J(\mathbf{k}, \mathbf{q} - \mathbf{k}) \langle \{ S_{\mathbf{k}-\mathbf{q}}^+, S_{-\mathbf{k}+\mathbf{q}}^- \} \rangle}{\omega - 2\sigma J(0, \mathbf{k} - \mathbf{q})} \right\} \right. . \tag{B.8}$$

In order to obtain the pole of  $G_{\mathbf{k}}(\omega)$  in the low temperature, in the second term in the curly bracketed expression we replace  $\omega$  by  $2\sigma J(0, \mathbf{k})$  which is the pole in the lowest approximation, and further replace  $\sigma$  by  $S$ . Thus we obtain for the pole of  $G_{\mathbf{k}}(\omega)$ , the spin wave spectrum, the following expression :

$$\omega \cong 2\sigma J(0, \mathbf{k}) - \frac{1}{N^2 S} \sum_{\mathbf{q}} J(\mathbf{q}, \mathbf{k} - \mathbf{q}) \langle \{ S_{\mathbf{k}-\mathbf{q}}^+, S_{-\mathbf{k}+\mathbf{q}}^- \} \rangle. \tag{B.9}$$

Introducing the spin wave operators  $a_{\mathbf{k}}, a_{\mathbf{k}}^*$  by

$$S_{\mathbf{k}}^+ = \sqrt{2NS} a_{\mathbf{k}}, \quad S_{-\mathbf{k}}^- = \sqrt{2NS} a_{\mathbf{k}}^*, \tag{B.10}$$

(B.9) becomes

$$\omega \cong 2\sigma J(0, \mathbf{k}) - \frac{2}{N} \sum_{\mathbf{q}} J(\mathbf{q}, \mathbf{k} - \mathbf{q}) \langle \{ a_{\mathbf{k}-\mathbf{q}}, a_{\mathbf{k}-\mathbf{q}}^* \} \rangle \\ = 2\sigma J(0, \mathbf{k}) - \frac{2}{N} \sum_{\mathbf{q}} J(\mathbf{q}, \mathbf{k} - \mathbf{q}) \langle a_{\mathbf{k}-\mathbf{q}}^* a_{\mathbf{k}-\mathbf{q}} \rangle. \tag{B.11}$$

Here we used that  $\sum_{\mathbf{q}} J(\mathbf{q}, \mathbf{k} - \mathbf{q}) = 0$ .

This is the correct spin wave spectrum discussed by several authors.<sup>4), 8), 16)</sup>

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**Note added in proof:** We should like to correct misprints in reference 13), and add a comment on Tahir-Kheli and ter Haar's critique on this paper. (1) In Eqs. (20) and (21) of this paper,  $\langle s_m^+ s_q^- s_q^0 \rangle$  should read  $\langle s_m^+ \{s_q^-, s_q^0\} \rangle$ , where the curly brackets denote the symmetrized product. (2) In Phys. Rev. **127** (1962), 88, Tahir-Kheli and ter Haar discussed the above paper and concluded that the paramagnetic susceptibility calculated from (23) does not agree with the correct expression even at the order of  $(T_c/T)$  in the high temperature expansion, where  $T_c = 2\pi J_0 S(S+1)/3k_B$ . We examined this point and found that (23) leads to a result correct up to the order of  $(T_c/T)$  at variance with their conclusion.