Progress of Theoretical Physics, Vol. 27, No. 3, March 1962

Theory of Dynamical Behaviors of Ferromagnetic Spins*)

Hazime MORI and Kyozi KAWASAKI

Research Institute for Fundamental Physics, Kyoto University, Kyoto

(Received November 6, 1961)

The dynamical behavior of ferromagnetic spins is studied on the basis of the statistical mechanics of irreversible processes. A macroscopic equation determining the change in time of an inhomogeneous magnetization is derived with explicit expressions for the frequency spectrum and damping constant. With the use of the general expressions thus obtained, the following problems are discussed on the basis of the Heisenberg model of ferromagnetic spins: (1) the pair correlation of spins, (2) the magnetic scattering cross section of neutrons, (3) the frequency spectrum and the damping constant of spin waves, (4) the damping of the longitudinal spin component above and below the Curie point.

In the low temperature limit, a straightforward reduction of our expressions leads to the spin wave frequency and damping equivalent to Dyson's theory of spin wave interactions. A general expression is given for van Hove's parameters describing the asymptotic behavior of the spin pair correlation at large distances.

It is shown that the longitudinal spin damping in the low temperature limit exhibits a variety of k dependences, depending upon the relative magnitudes of the spin wave energy Dk^2, the effective magnetic field H coming from other than the exchange interaction and the thermal energy k_BT; in particular, for $g\mu_BH \ll Dk^2 \ll k_BT$, the longitudinal damping is proportional to k^2, namely, the change in time of the longitudinal spin density obeys the diffusion equation. In the vicinity of the Curie point and in the paramagnetic region, the longitudinal damping is shown to obey the diffusion equation. The diffusion constant thus obtained vanishes at the Curie point and is in good agreement with the values observed by Ericson and Jacrot for iron above the Curie point.

§ 1. Introduction

Since the discovery of the critical phenomena in the magnetic scattering of neutrons experimentally by Palevsky and Hughes[1] and by Squires,[2] and theoretically by van Hove,[3] a number of neutron scattering experiments have been performed with the purpose of investigating the dynamical behavior of ferromagnetic spins. When approaching the Curie point from either side, there occurs a strong increase in the intensity of the diffuse peaks in the neighborhood of the magnetic Bragg reflections. This was explained by van Hove as being due to the strong increase in the fluctuations of the magnetization in this temperature region. The theoretical exposition in the critical region, where the

---

*) The main results of this paper were reported in a talk with the title of "Theory of Spin Diffusion" at the International Conference on Magnetism and Crystallography held in Kyoto, September 25-30, 1961.

spin wave approximation is not valid, has been worked out in more detail by Elliott and Marshall[4] and by de Gennes.[5]

Recent neutron scattering experiments by Riste and his collaborators[6] revealed the temperature dependence of the spin wave frequency and damping constant. The theory of spin wave interactions was established in the low temperature limit by Dyson.[7] The following studies of the spin wave frequency by Bogolyubov and Tjablikov,[8] by Keffer and Loudon,[9] and by Brout and Englert[10] yield the temperature dependence of the effective exchange coupling consistent with the experiments. Dyson's theory, however, does not tell us how to obtain the damping constant of the longitudinal spin component, although his theory gives the dynamical interaction between spin waves so that one can calculate the spin wave damping by applying the kinetic method with appropriate assumptions.

In fact, the damping of the longitudinal component of an inhomogeneous magnetization due to the exchange interaction is a typical example which is out of the usual kinetic treatment. The damping constant for iron above the Curie point has been determined by Ericson and Jacrot[11] by observing the inelastic magnetic scattering of neutrons, which shows that the damping obeys the diffusion equation and the diffusion constant vanishes at the Curie point. Prior to this experiment, van Hove proposed to describe the inelastic part of the critical scattering of neutrons by a ferromagnetic above the Curie point by introducing the concept of spin diffusion, and argued that the diffusion constant would be zero at the Curie point as a result of the thermodynamic braking of fluctuations and would increase linearly with temperature in the vicinity of this point. Recently de Gennes[5],[12] extended van Hove's theory to give a concise theory of critical scattering, and obtained an expression for the diffusion constant which is in agreement with the experiment. His treatment, however, is still phenomenological and is based on several nuclear assumptions, and a microscopic basis of his theory remain to be clarified.

Thus, in the theoretical investigation of the spin damping due to the exchange interaction, we only have Dyson's work on the transverse damping in the low temperature limit, and we know little about the longitudinal damping except the phenomenological investigation above the Curie point.

The dynamical property of the magnetic system is disclosed by stimulating the system with external disturbances such as neutron beams and magnetic fields and by investigating the response to them. The response can be expressed in terms of the time-dependent correlation function of the magnetization[3] or the relaxation function $\chi_k(t)$;[13]

$$\chi_k(t) = \frac{1}{N}\int_0^\beta d\lambda \langle e^{\lambda H} \boldsymbol{M}_k(t)\, e^{-\lambda H} \boldsymbol{M}_k{}^* \rangle - \frac{\beta}{N} \langle \boldsymbol{M}_k \rangle \langle \boldsymbol{M}_k{}^* \rangle, \tag{1·1}$$

where the angular brackets mean the average with the canonical ensemble

$\exp(-\beta H)/\mathrm{Tr}\exp(-\beta H)$ and $\boldsymbol{M}_k$ denotes the Fourier component of the magnetization with wave vector $\boldsymbol{k}$. N is the total number of magnetic spins concerned. Thus the neutron scattering cross section and the magnetic resonance absorption are written in terms of the relaxation function, and the problem is reduced to the investigation of the relaxation function.

There is another concept in the description of the dynamical behavior of ferro- and antiferromagnetic spin systems. That is the collective motion of spins, which is one of the most important properties of the ferro- and antiferromagnetic interactions. Far below the transition temperature, this is the spin wave motion representing the collective precession of spins. One may imagine some analogy with the collective motion of fluids described by the hydrodynamical equations. Suppose a ferromagnetic spin system as being a spin fluid. Then one may expect a collective motion even above the Curie point corresponding to the diffusion of particles in fluids determining the temporal development of an inhomogeneous density of particles. In fact de Gennes took this way of looking at the problem and formulated the theory of spin diffusion abovementioned. As will be shown later, the collective motion of spin systems can be described in terms of the relaxation function $\chi_k(t)$ with the aid of a general property of the ferro- and antiferromagnetic interactions, and thus the neutron scattering cross section can be expressed in terms of the collective motion.

Thus the neutron scattering experiment is the most direct way of investigating the collective motion of macroscopic systems. This is not restricted to the magnetic systems. For instance, we know that the neutron scattering experiment is now revealing interesting properties of the collective motion of liquid helium II. One of the exciting problems of this aspect is the determination of the frequency spectrum and damping constant of the collective motion below and above the transition point. However, the foregoing works by Dyson, by Bogolyubov and Tjablikov, by van Hove, and by de Gennes, although very important and very elaborate, are still not enough to meet these situations.

The principal purpose of the present paper is, therefore, to formulate the collective motion of ferromagnetic spins on the basis of rigorous principles. This will be done by formulating the frequency spectrum and the damping constant for the motion of an inhomogeneous magnetization with the aid of the statistical mechanics of irreversible processes.[13),14)] Explicit calculation of the general expressions thus obtained will be carried out in the low temperature limit, in the vicinity of the Curie point, and in the paramagnetic region by employing the Heisenberg model of ferromagnetic spins. A preliminary formulation of the problem has been reported elsewhere.[15)]

In § 2, where a collective description of the ferromagnetic spins is presented, we emphasize that, due to the fact that the exchange interaction which is much stronger than any other interaction commutes with the total spin, the Fourier components of the magnetization density form a set of state variables complete enough to describe macroscopic magnetic disturbances of the system. This guarantees the existence of the collective motion of ferromagnetic spins. The situation is quite the same as in the case of the hydrodynamical motion of fluids, in which case the conservation laws of particle density, momentum density, and Hamiltonian density form the basis for the statistical-mechanical foundation of the hydrodynamical description.[14] Thus we derive a macroscopic equation of motion for an inhomogeneous magnetization with general expressions for the frequency spectrum and damping constant. Two different ways for the derivation which are equivalent to each other will be given; one goes along with the theory of transport in fluids developed by one of the authors,[14] and the other corresponds to a generalization of Kubo-Tomita's theory[16] of magnetic resonance absorption.

The frequency spectrum thus obtained consists of two parts: The first part is the first moment $\langle\omega\rangle_k$ of the frequency distribution $\Xi_k(\omega)$ defined by

$$\Xi_k(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}dt\,e^{-it\omega}\chi_k(t)/\chi_k(0), \tag{1·2}$$

$$\langle\omega^n\rangle_k=\int_{-\infty}^{\infty}d\omega\,\omega^n\,\Xi_k(\omega)=\frac{1}{i^n}\left[\frac{d^n}{dt^n}\chi_k(t)/\chi_k(0)\right]_{t=0}, \tag{1·3}$$

and the second is the contribution from the higher moments. The frequency spectrum vanishes above the Curie point in the absence of external field. Far below the Curie point the first moment yields a good approximation to the frequency spectrum.

In § 3, we investigate the first moment frequency spectrum and the associated quantities. It is shown that a straightforward reduction of the first moment frequency spectrum in the low temperature limit leads to the equation obtained, by Keffer and Loudon,[9),10] and shown to reproduce Dyson's theory of ferromagnetism.

§ 4 is concerned with the investigation of the static pair correlation of spins and the neutron scattering cross section, where we obtain a general expression for van Hove's parameters describing the asymptotic behavior of the spin pair correlation, and determine an exact form for the scattering cross section in terms of the collective description.

The damping constant and shift of frequency are investigated in §§ 5, 6, and 7. It is shown that a straightforward reduction of our expression for the transverse component of the damping constant in the low temperature limit leads to the spin wave damping which equals one half of the transition rate of the number of spin waves obtained from Dyson's dynamical interaction by using the kinetic treatment with Schlömann's assumption.[17] Thus we show that our expressions for the frequency spectrum and transverse damping constant agree, in the low temperature limit, with Dyson's theory of spin wave interactions, just by starting from the original Hamiltonian with the Heisenberg exchange interaction and setting up the Heisenberg equation of motion for the spin operator. As will be shown in Appendix B, this is a direct consequence of the fact that the change in time of the transverse spin operator can be written, in the spin wave region, simply in terms of Dyson's dynamical interaction.

The longitudinal component of the damping constant is calculated in the last half of § 6 and in § 7. The longitudinal damping has a quite different feature from the transverse one below the Curie point. It is shown that the longitudinal damping in the low temperature limit exhibits a variety of k dependences, depending upon the relative magnitudes of the spin wave energy Dk^2, the effective magnetic field H coming from other than the exchange interaction and the thermal energy k_BT; in particular, for $g\mu_BH \ll Dk^2 \ll k_BT$, it obeys the diffusion equation. In the vicinity of the Curie point and in the paramagnetic region, the longitudinal damping is shown to obey the diffusion equation. The diffusion constant thus obtained vanishes at the Curie point, being proportional to the temperature distance from this point, and is in good agreement with the values observed by Ericson and Jacrot for iron above the Curie point.

The last section is devoted to a brief summary and some remarks.

§ 2. Collective motion of ferromagnetic spins

Let us suppose that the system consists of N ferromagnetic spins in a volume V and the magnetization density $\boldsymbol{M}(\boldsymbol{r})$ deviates from an equilibrium value $\overline{\boldsymbol{M}}/V$. In this section, we shall formulate the macroscopic equations of motion for $\boldsymbol{M}(\boldsymbol{r})$ which determine how the inhomogeneous density becomes uniform and which thus describe the spin waves with damping and the spin diffusion. This will be done with the aid of the statistical mechanics of irreversible processes and in the limitation of the linear theory.

Let us define the Fourier components of the magnetization

$$M_k{}^\alpha = \int d\boldsymbol{r}\, e^{i\boldsymbol{k}\cdot\boldsymbol{r}} M^\alpha(\boldsymbol{r}), \quad (\alpha=0,\ \pm), \tag{2·1}$$

where $M^0(\boldsymbol{r})$ denotes the z component of the magnetization density $M_z(\boldsymbol{r})$ and $M^\pm(\boldsymbol{r}) = M_x(\boldsymbol{r}) \pm iM_y(\boldsymbol{r})$. With the aid of the density matrix of the system $\rho(t)$, we have

$$M_k{}^\alpha(t) = -g\mu_B \operatorname{Tr}\rho(t)\, S_k{}^\alpha, \tag{2·2}$$

$$= -g\mu_B \operatorname{Tr}\rho(0)\, S_k{}^\alpha(t), \tag{2·3}$$

where S_k^α is the Fourier component of the α component of the spin operator.

If the ferromagnetic spins are distributed on periodic lattice points and the lattice point f has the spin S_f, then we have

$$S_k^{\alpha}=\sum_f e^{ik\cdot r_f}S_f^{\alpha}=(S_{-k}^{-\alpha})^*, \tag{2·4}$$

where the position vector of the lattice point f has been denoted by $\boldsymbol{r}_f$, and the asterisk * means the Hermitean conjugate. If the spins are not localized, for instance, in accordance with the band model of ferromagnetic spins in metals, then we should replace the summation $\sum_f$ by the integration $\int d\boldsymbol{r}$, taking the spin density operator $S^{\alpha}(\boldsymbol{r})$ instead of S_f^{α}.

We assume that a constant magnetic field H_c is present in the negative z direction. Then the total Hamiltonian of the system takes the form

$$H=\omega_0 S_0^0+H_0, \tag{2·5}$$

where S_0^0 is the z component of the total spin and the Zeeman frequency is denoted by $\omega_0(=-g\mu_B H_c)$. Use of (2·3) and the Heisenberg equation of motion for $S_k^{\alpha}(t)$ leads to

$$\frac{d}{dt}M_k^{\alpha}(t)=i\alpha\omega_0 M_k^{\alpha}(t)-g\mu_B \operatorname{Tr}\rho(t)\,\dot{S}_k^{\alpha}, \tag{2·6}$$

where

$$\dot{S}_k^{\alpha}\equiv i[H_0, S_k^{\alpha}], \tag{2·7}$$

which represents the change in time due to the interactions between spins. The unit has been chosen so that $\hbar=1$. Our problem is now to express the second term of (2·6) in terms of the magnetization $\{M_k^{\alpha}(t)\}$ and thus to obtain a set of equations determining the temporal development of the magnetization. It will be shown that this comes out possible with the aid of a general property of the ferromagnetic system.

In ferromagnetics without external field, the exchange interaction, which is much stronger than any other interactions, commutes with the total spin so that the total magnetization is, in a very good approximation, conserved. This fact endows the macroscopic treatment of ferromagnetics with greater physical significance. Namely, due to this fact, the magnetization can be regarded as an independent variable. Thus, in the macroscopic treatment of the critical magnetic scattering of neutrons by ferromagnetics, van Hove divided a ferromagnetic into a cubic array of identical small cells, but of macroscopic size, and regarded the magnetizations of the cells as the macroscopic state variables, to follow Ornstein-Zernike's theory of the anomalous fluctuation of density in liquids or dense gases near the critical point.

It thus turns out that the Fourier components of the magnetization density form a set of macroscopic state variables complete enough to describe macroscopic magnetic disturbances. In the statistical-mechanical description of macroscopic states, it is convenient to introduce a canonical ensemble, which, in the present case, is the local equilibrium ensemble[14)]

$$\rho_t = Z^{-1} \exp\{-\beta[H + g\mu_B \sum_q \boldsymbol{h}_q \cdot \boldsymbol{S}_q^*]\}, \tag{2·8}$$

where Z is the normalization constant, and $\boldsymbol{h}_k$ represents a thermodynamic magnetic field which arises as a result of the fact that the system deviates from equilibrium, having the non-equilibrium magnetization $\boldsymbol{M}_k(t)$. These parameters depend on time t and are determined by the requirements

$$\mathrm{Tr}\,\rho_t = \mathrm{Tr}\,\rho(t) = 1, \tag{2·9}$$

$$\mathrm{Tr}\,\rho_t \boldsymbol{S}_k = \mathrm{Tr}\,\rho(t)\,\boldsymbol{S}_k = -\boldsymbol{M}_k(t)/g\mu_B. \tag{2·10}$$

Equation (2·10) is required so that ρ_t gives the same macroscopic state as the density matrix $\rho(t)$. Expanding (2·8) in terms of $\boldsymbol{h}_q$ and retaining only the first order term, we obtain

$$\rho_t = \rho\left[1 - g\mu_B \sum_q \boldsymbol{h}_q \cdot \int_0^\beta d\lambda\, e^{\lambda H}(\boldsymbol{S}_q^* - \langle \boldsymbol{S}_q^* \rangle)\, e^{-\lambda H}\right], \tag{2·11}$$

where

$$\langle A \rangle = \mathrm{Tr}\,\rho A, \tag{2·12}$$

$$\rho = e^{-\beta H}/\mathrm{Tr}\, e^{-\beta H}, \tag{2·13}$$

where ρ denotes the equilibrium ensemble with the temperature $T = 1/k_B\beta$, k_B being the Boltzmann constant. Insertion of (2·11) into (2·10) leads to*)

$$\boldsymbol{M}_k(t) - \bar{\boldsymbol{M}}\delta_{k,0} = N\chi_k \cdot \boldsymbol{h}_k(t), \tag{2·14}$$

where χ_k is a generalized susceptibility tensor

$$\chi_k = \frac{1}{N}(g\mu_B)^2 (\boldsymbol{S}_k, \boldsymbol{S}_k^*), \tag{2·15}$$

where we have defined the bracket notation by

$$(A, B) = \int_0^\beta d\lambda \langle e^{\lambda H} A e^{-\lambda H} B \rangle - \beta \langle A \rangle \langle B \rangle, \tag{2·16}$$

$$= (B, A) = (A^*, B^*)^*. \tag{2·17}$$

In the limit of $\boldsymbol{k} = \boldsymbol{0}$, χ_k agrees with the well-known expression for the susceptibility tensor for a homogeneous magnetic field. If a constant inhomogeneous magnetic field equal to $\boldsymbol{h}(\boldsymbol{r}) = \sum_q e^{-i\boldsymbol{q}\cdot\boldsymbol{r}} \boldsymbol{h}_q$ were applied, the system would be, in final equilibrium state, described by the local equilibrium ensemble (2·8) to

*) Having reached the final equilibrium described by (2·13), the system is homogeneous so that the Fourier components of quantities $A(\boldsymbol{r})$ and $B(\boldsymbol{r})$ satisfy, in the limit of $V \to \infty$,

$$(A_k, B_{k'}) = \sum_{\boldsymbol{r},\boldsymbol{r}'} e^{i\boldsymbol{k}\cdot\boldsymbol{r} + i\boldsymbol{k}'\cdot\boldsymbol{r}'} (A(\boldsymbol{0}), B(\boldsymbol{r}' - \boldsymbol{r})),$$

$$= \delta_{k', -k} (A_k, B_{-k}).$$

have an inhomogeneous magnetization density equal to $\boldsymbol{M}_k(t)$. Thus (2·15) turns out to be a straightforward generalization of the usual susceptibility to an inhomogeneous case. When the vector components $\boldsymbol{M}_k=(M_k^0, M_k^+, M_k^-)$ are used, the following should be taken as the corresponding components of $\boldsymbol{h}_k$ and χ_k:

$$h_k{}^0=h_{zk}, \quad h_k{}^{\pm}=\frac{1}{2}(h_{xk}\pm ih_{yk}), \tag{2·18}$$

$$(\chi_k)_{\alpha\gamma}=\frac{1}{N}(g\mu_B)^2(S_k{}^\alpha, S_k^{\gamma *}), \quad (\alpha, \gamma=0, +, -). \tag{2·19}$$

Equation (2·14) thus determines the thermodynamic magnetic field $\boldsymbol{h}_k(t)$ in terms of the inhomogeneous magnetization $\boldsymbol{M}_k(t)$.

Equation (2·8) describes an inhomogeneous precession of spins. It is a useful approximation to assume that

$$\rho(t)\approx\rho_t, \tag{2·20}$$

which, on inserting (2·11) into (2·6), leads to

$$\frac{d}{dt}\boldsymbol{M}_k(t)\approx i[\hat{\omega}_0+\hat{\omega}_k]\cdot\boldsymbol{M}_k(t), \tag{2·21}$$

where

$$i\hat{\omega}_k=(\dot{\boldsymbol{S}}_k, \boldsymbol{S}_k{}^*)\cdot(\boldsymbol{S}_k, \boldsymbol{S}_k{}^*)^{-1}, \tag{2·22}$$

$$\hat{\omega}_0=(\alpha\omega_0\delta_{\alpha,\gamma}), \tag{2·23}$$

where the second factor of (2·22) denotes the inverse matrix of $(\boldsymbol{S}_k, \boldsymbol{S}_k^*)$. Equation (2·22) determines the frequency spectrum of the inhomogeneous precession in the first approximation and turns out to equal the first moment $\langle\omega\rangle_k$ of the frequency distribution defined by (1·2) and (1·3). If we neglect the dipolar interaction between spins, the spin system has the axial symmetry about the z axis; $[H, S_0{}^0]=0$. Then the susceptibility and frequency tensors become diagonal and the diagonal elements of the frequency take the form

$$\omega_k{}^\alpha=(\dot{S}_k{}^\alpha, S_k^{\alpha *})/i(S_k{}^\alpha, S_k^{\alpha *}), \quad (\alpha=0, \pm), \tag{2·24}$$

which are real. It will be shown later that a direct calculation of the first moment frequency (2·24) in the spin wave region leads to an expression for the spin wave frequency spectrum which has been obtained, by Keffer and Loudon,[9] and shown to reproduce Dyson's theory of ferromagnetism.[7] It will also be shown that (2·24) leads to a general relation between the spin pair correlation and the frequency spectrum, which, above the Curie point, determines the asymptotic behavior of the spin pair correlation discussed first by van Hove[3] in the theory of critical scattering of neutrons.

The characteristic time τ_k, in which the Fourier component of the magnetization density $\boldsymbol{M}_k(t)$ changes by an appreciable amount due to the exchange interaction, becomes longer as the wave number k gets smaller, and becomes infinity if k goes to zero. Thus the characteristic time τ_k is very long compared to the microscopic time $\tau_0=\hbar/J$, J being the magnitude of the exchange interaction. There is an important physical situation that the ferromagnetic spins in a uniform small volume element, say, around the position $\boldsymbol{r}$, attain, in the short time interval τ_0, approximate internal equilibrium to have $\boldsymbol{M}(\boldsymbol{r})$ as a local average value of the magnetization.*) This situation quite resembles the local equilibrium of mass elements in non-equilibrium fluids.[14] Thus the local equili brium ensemble ρ_t turns out to yield a good approximation of $\rho(t)$. The deviation of the density matrix from ρ_t is denoted by $\rho'(t)$:

$$\rho(t)=\rho_t+\rho'(t), \qquad (2\cdot25)$$

$$\mathrm{Tr}\,\rho'(t)=\mathrm{Tr}\,\rho'(t)\,\boldsymbol{S}_k=0. \qquad (2\cdot26)$$

The deviation expresses the microscopic process and leads to the damping of the magnetization and a higher moment contribution to the frequency spectum.

We are now in a position to determine the damping effects for the temporal development of the magnetization $\boldsymbol{M}_k(t)$. The determination of $\rho'(t)$ suffices for this purpose, which, however, will be given in Appendix A. Instead, we take another way of looking at the problem, which corresponds to a generalization of Kubo-Tomita's method[16] for dealing with the magnetic resonance absorption.

Suppose a system whose density matrix at time $t=0$ is given by $(2\cdot8)$ with a value $\boldsymbol{h}_{0q}$ for $\boldsymbol{h}_q$. Then the temporal development of the magnetization thereafter can be described by

$$\boldsymbol{M}_k(t)=\overline{\boldsymbol{M}}\delta_{k,0}+N\chi_k(t)\cdot\boldsymbol{h}_{0k}, \qquad (2\cdot27)$$

where $\chi_k(t)$ is the dynamical susceptibility tensor

$$\chi_k(t)=\frac{1}{N}(g\mu_B)^2(\boldsymbol{S}_k(t),\boldsymbol{S}_k{}^*), \qquad (2\cdot28)$$

which, at $t=0$, agrees with $(2\cdot15)$. The temporal development of the magnetization density is, except the initial transient period of the order of magnitude of τ_0, independent of the way of the initial preparation of the system as far as the initial value of the magnetization density is fixed to be the same.[14] Therefore, we can employ $(2\cdot27)$ to determine macroscopic equations of motion for the magnetization density $\boldsymbol{M}_k(t)$ by investigating the asymptotic behavior of the relaxation function $\chi_k(t)$.

---

*) This situation has also been explicitly recognized in the theory of collective motion of spins in ferro- and antiferromagnets by J. Kanamori and M. Tachiki.[18] Their expression for the frequency spectrum, however, differs from the present results $(2\cdot24)$ and becomes divergent in the vicinity of the Curie point. This is because of the fact that their description of the collective precession is incomplete. The difference, however, does not seem to be serious in the low temperature limit.

Let us define the generating function

$$\Xi_k(t) = (\boldsymbol{S}_k(t), \boldsymbol{S}_k^*) \cdot (\boldsymbol{S}_k, \boldsymbol{S}_k^*)^{-1}, \tag{2·29}$$

in terms of which we can write as

$$\chi_k(t) = \Xi_k(t) \cdot \chi_k(0), \tag{2·30}$$

$$\boldsymbol{M}_k(t) - \overline{\boldsymbol{M}}\delta_{k,0} = \Xi_k(t) \cdot [\boldsymbol{M}_k(0) - \overline{\boldsymbol{M}}\delta_{k,0}], \quad (t \gg \tau_0). \tag{2·31}$$

Expansion of (2·29) in powers of time leads to

$$\begin{aligned}\Xi_k(t) &= \sum_{n=0}^{\infty} \frac{t^n}{n!} \left(\left[\frac{d^n}{dt^n} \boldsymbol{S}_k(t) \right]_{t=0}, \boldsymbol{S}_k^* \right) \cdot (\boldsymbol{S}_k, \boldsymbol{S}_k^*)^{-1}, \\ &= 1 + it(\hat{\omega}_0 + \hat{\omega}_k) \\ &\quad - \frac{t^2}{2} ([H, \boldsymbol{S}_k], [\boldsymbol{S}_k^*, H]) \cdot (\boldsymbol{S}_k, \boldsymbol{S}_k^*)^{-1} + \cdots, \end{aligned} \tag{2·32}$$

where $\hat{\omega}_0$ and $\hat{\omega}_k$ are the frequency tensors defined by (2·22) and (2·23). The simplest approximation is to take

$$\Xi_k(t) \approx \exp[it(\hat{\omega}_0 + \hat{\omega}_k)]. \tag{2·33}$$

This approximation is equivalent to (2·20) and (2·21), and expresses the collective motion of spins without the damping effect.

To proceed in obtaining the damping effect due to the exchange interaction between spins, we simplify the problem by neglecting the dipolar interaction. Then the generating function (2·29) forms a diagonal matrix with the following diagonal elements:

$$\Xi_k^{\alpha}(t) = (S_k^{\alpha}(t), S_k^{\alpha*}) / (S_k^{\alpha}, S_k^{\alpha*}), \quad (\alpha = 0, \pm). \tag{2·34}$$

It is convenient to rewrite, with the use of the frequency spectrum (2·24), as

$$\Xi_k^{\alpha}(t) = e^{it\bar{\omega}_k^{\alpha}} (\widetilde{S}_k^{\alpha}(t), S_k^{\alpha*}) / (S_k^{\alpha}, S_k^{\alpha*}), \tag{2·35}$$

where

$$\bar{\omega}_k^{\alpha} = \alpha\omega_0 + \omega_k^{\alpha}, \tag{2·36}$$

$$\widetilde{S}_k^{\alpha}(t) = e^{-it\bar{\omega}_k^{\alpha}} S_k^{\alpha}(t). \tag{2·37}$$

The quantity $\widetilde{S}_k^{\alpha}(t)$ is something similar to the interaction representation in the perturbation theory, and represents the motion due to the irregular fluctuation of spins around the collective oscillation given by (2·33). Since

$$\begin{aligned}\frac{d}{dt} \widetilde{S}_k^{\alpha}(t) &= -i\bar{\omega}_k^{\alpha} \widetilde{S}_k^{\alpha}(t) + i[H, \widetilde{S}_k^{\alpha}(t)], &(2·38)\\ &= -i\omega_k^{\alpha} \widetilde{S}_k^{\alpha}(t) + i[H_0, \widetilde{S}_k^{\alpha}(t)], &(2·39)\end{aligned}$$

we define

$$f(t)=(\widetilde{S}_k^\alpha(t),\ S_k^{\alpha *})/(S_k^\alpha,\ S_k^{\alpha *}),\tag{2·40}$$

which satisfies

$$\left[\frac{d}{dt}f(t)\right]_{t=0}=0,\tag{2·41}$$

$$f(t)=1+\int_0^t ds\,(t-s)\,\frac{d^2}{ds^2}f(s).\tag{2·42}$$

Since

$$\left(-i\bar{\omega}_k^\alpha\frac{d}{dt}\widetilde{S}_k^\alpha(t)+i\left[H,\ \frac{d}{dt}\widetilde{S}_k^\alpha(t)\right],\ S_k^{\alpha *}\right)$$

$$=\left(\frac{d}{dt}\widetilde{S}_k^\alpha(t),\ -i\bar{\omega}_k^\alpha S_k^{\alpha *}-i[H,\ S_k^{\alpha *}]\right),\tag{2·43}$$

the integrand in (2·42) can be written as

$$\frac{d^2}{dt^2}f(t)=-e^{-it\bar{\omega}_k^\alpha}(I_k^\alpha(t),\ I_k^{\alpha *})/(S_k^\alpha,\ S_k^{\alpha *}),\tag{2·44}$$

where

$$I_k^\alpha(t)=i[H,\ S_k^\alpha(t)]-i\bar{\omega}_k^\alpha S_k^\alpha(t),\tag{2·45}$$

$$=\dot{S}_k^\alpha(t)-i\omega_k^\alpha S_k^\alpha(t).\tag{2·46}$$

The first term of (2·45) represents the total torque acting on the respective spins and the second is the torque corresponding to the precession (2·33). Thus the quantity I_k^α expresses the fluctuation of the torque around the precessional motion represented by (2·21), satisfying $\mathrm{Tr}\,\rho_t I_k^\alpha=0$- The damping of the spin motion comes from this fluctuation of the torque.

Now it is assumed that the correlation function (2·44) vanishes in a time interval τ_c much shorter than the decay time τ_k of the generating function (2·40). In ferromagnetic substances, as has been discussed before, the decay time τ_k becomes longer as the wave number k gets smaller, whereas the correlation time τ_c is almost constant which is of the order of magnitude of $\tau_0\equiv\hbar/J$.[14] Thus the assumption is satisfied at least in the case of small wave numbers. Taking time intervals satisfying $\tau_k\gg t\gg\tau_0$, we write (2·42) as

$$f(t)=1+t\int_0^{\tau_c}ds\ \frac{d^2}{ds^2}f(s)+\cdots,$$

$$\cong\exp\left[t\int_0^{\tau_c}ds\ \frac{d^2}{ds^2}f(s)\right].\tag{2·47}$$

By inserting this into (2·35) we obtain the following asymptotic equation for the generating function:

$$\Xi_k^\alpha(t) = \exp[it(\alpha\omega_0 + \omega_k^\alpha) - t\Gamma_k^\alpha], \quad (t \gg \tau_0), \tag{2·48}$$

where

$$\Gamma_k^\alpha = \int_0^\infty dt\, e^{-it(\alpha\omega_0+\omega_k^\alpha)} (I_k^\alpha(t), I_k^{\alpha *}) / (S_k^\alpha, S_k^{\alpha *}), \tag{2·49}$$

where the time integration has been extended to infinity, which is guaranteed by the fact that the decay time τ_k becomes infinity if k goes to zero. Equations (2·31) and (2·48) are combined to give

$$\frac{d}{dt} M_k^\alpha(t) = [i(\alpha\omega_0 + \omega_k^\alpha) - \Gamma_k^\alpha] M_k^\alpha(t). \tag{2·50}$$

This provides us with the macroscopic equation of motion for the magnetization density $M_k^\alpha(t)$. The real part of Γ_k^α yields the damping effects, which lead to the damping of spin waves and the spin diffusion.

The imaginary part of Γ_k^α, which we denote by $-\Delta\omega_k^\alpha$, leads to a shift of the frequency, coming from the fluctuation of spins around the collective oscillation represented by the first moment of (1·3). Thus the shift of frequency $\Delta\omega_k^\alpha$ corresponds to the contribution from the higher moments to the frequency spectrum.

§ 3. First moment frequency spectrum

In the present paper, we are mainly concerned with the exchange interaction between spins, neglecting the dipolar interaction. Then the first moment frequency spectrum is given by (2·24). In this section, we shall investigate the properties of this frequency spectrum.

With the aid of the identity[19)]

$$([H, A], B) = -\langle [A, B] \rangle, \tag{3·1}$$

Eq. (2·24) can be transformed to be

$$\begin{aligned} \omega_k^\alpha &= -\alpha\omega_0 + ([H, S_k^\alpha], S_k^{\alpha *}) / (S_k^\alpha, S_k^{\alpha *}), \\ &= -\alpha\omega_0 - 2\alpha\sigma N / (S_k^\alpha, S_k^{\alpha *}), \end{aligned} \tag{3·2}$$

where use has been made of the fact that

$$\left.\begin{aligned} [S_k^+, S_{k'}^-] &= 2S_{k+k'}^0, \\ [S_k^0, S_{k'}^\pm] &= \pm S_{k+k'}^\pm, \end{aligned}\right\} \tag{3·3}$$

and

$$[\langle S_k^0 \rangle]_{k=0} \equiv \sigma N, \quad \langle S_k^\pm \rangle = 0. \tag{3·4}$$

Thus it turns out that

$$\omega_k^0 = 0, \quad \omega_k^\pm = (\omega_k^\pm)^* = -\omega_{-k}^\mp. \tag{3·5}$$

Above the Curie temperature, the equilibrium magnetization σ (for one spin) vanishes in the absence of external magnetic field so that $\omega_k^{\pm}=0$ from (3·2).

If the system has the inversion symmetry with respect to each lattice point, we have $(A(\mathbf{0}), B(\mathbf{r}))=(A(\mathbf{0}), B(-\mathbf{r}))$ so that

$$\chi_k{}^{\alpha}\equiv\frac{1}{N}(g\mu_B)^2(S_k{}^{\alpha}, S_k^{\alpha *})=\chi_{-k}^{\alpha}=\chi_k^{-\alpha}, \tag{3·6}$$

$$\omega_k{}^{\pm}=\omega_{-k}^{\pm}=\mp\omega_k, \quad \omega_k\equiv\omega_k{}^{-}. \tag{3·7}$$

Equation (3·2) gives the perpendicular susceptibility in terms of the frequency spectrum:

$$\chi_{\perp k}\equiv\frac{1}{2}\chi_k{}^{+}=(g\mu_B)^2\frac{\sigma}{-\omega_0+\omega_k}, \tag{3·8}$$

$$=\frac{(g\mu_B)^2}{(g\mu_B)^2/\chi_{\perp}+\omega_k/\sigma}, \tag{3·9}$$

where $\chi_{\perp}\equiv-(g\mu_B)^2\sigma/\omega_0$, which represents the perpendicular susceptibility for one spin for a homogeneous magnetic field.

Now we assume the Heisenberg model of spins with the exchange interaction J_{fg} or $J(\mathbf{q})$, to calculate the frequency spectrum. The Hamiltonian takes the form

$$H_0=-\sum_{f\neq g}\sum J_{fg}\mathbf{S}_f\cdot\mathbf{S}_g, \tag{3·10}$$

$$=-\frac{1}{N}\sum_q J(\mathbf{q})\,\mathbf{S}_q\cdot\mathbf{S}_q{}^*, \tag{3·11}$$

which leads to

$$\begin{cases}\dot{S}_k{}^0=-iN^{-1}\sum_q J(\mathbf{q}, \mathbf{k}-\mathbf{q})\,S_q{}^+S_{k-q}^-, & (3·12)\\ \dot{S}_k{}^{\pm}=\mp 2iN^{-1}\sum_q J(\mathbf{q}, \mathbf{k}-\mathbf{q})\,S_q{}^0 S_{k-q}^{\pm}, & (3·13)\end{cases}$$

where

$$J(\mathbf{q})\equiv\sum_{f(\neq g)}e^{i\mathbf{q}\cdot(\mathbf{r}_f-\mathbf{r}_g)}J_{fg}=J(-\mathbf{q})=J(\mathbf{q})^*, \tag{3·14}$$

$$J(\mathbf{q}, \mathbf{q}')\equiv J(\mathbf{q})-J(\mathbf{q}'), \tag{3·15}$$

where the inversion symmetry with respect to each lattice point has been assumed to obtain the second equation of (3·14).

Let us rewrite (3·13) as

$$\dot{S}_k{}^{\pm}=\mp 2i\sigma J(\mathbf{0}, \mathbf{k})\,S_k{}^{\pm}\mp 2iN^{-1}\sum_q J(\mathbf{q}, \mathbf{k}-\mathbf{q})[S_q{}^0-\langle S_q{}^0\rangle]S_{k-q}^{\pm}. \tag{3·16}$$

The second term expresses the torque arising from the fluctuation of the z component around the equilibrium value. The simplest approximation is to neglect this fluctuation term, which leads, from (2·24), to

$$\omega_k\cong 2\sigma J(\mathbf{0}, \mathbf{k}). \tag{3·17}$$

Bogolyubov and Tjablikov[8] investigated this approximation, employing the two-time Green's function method, and worked out a theory of ferromagnetism which is approximately valid at all temperatures. In the low temperature limit, however, their result disagrees with Dyson's theory of spin waves. This disagreement has been shown, by Keffer and Loudon, to be removed by taking into account the fact that a spin wave, being generated in a region of long wavelength spin waves, which are already present, feels the exchange field obtained not from the equilibrium magnetization $\langle \mathbf{S}_q \rangle$ but from the nonequilibrium magnetization moving along with the existing background spin waves.

To see the effect due to this situation in the low temperature limit, we calculate the contribution of the secod term of (3·16), using the spin wave approximation

$$S_q^+ \cong \sqrt{2NS}\, a_q, \tag{3·18}$$

$$S_q^0 = NS\,\delta_{q,0} - \sum_p a_p^* a_{p+q}, \tag{3·19}$$

where

$$[a_q, a_{q'}^*] = \delta_{q,q'}, \tag{3·20}$$

and S denotes the magnitude of the spin. Since

$$\dot{S}_k^- = 2iSJ(\mathbf{0}, \boldsymbol{k})\, S_k^- - (2i/N) \sum_q J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q}) \sum_p a_p^* a_{p+q} S_{k-q}^-, \tag{3·21}$$

$$\omega_k = 2SJ(\mathbf{0}, \boldsymbol{k}) - (2/N) \sum_q J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q}) \left(\sum_p a_p^* a_{p+q} S_{k-q}^-, S_{-k}^+\right) / (S_k^-, S_{-k}^+), \tag{3·22}$$

we obtain, in the lowest order approximation,

$$\omega_k \cong 2SJ(\mathbf{0}, \boldsymbol{k}) - (2/N) \sum_q \sum_p J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q}) (a_p^* a_{p+q} a_{q-k}^*, a_{-k}) / (a_{-k}^*, a_{-k}). \tag{3·23}$$

For the free spin waves, it is easily shown that

$$(a_{-k}^*, a_{-k}) = 1/\omega_k^0, \tag{3·24}$$

$$(a_p^* a_{p+q} a_{q-k}^*, a_{-k}) = \langle a_p^* a_{p+q} a_{q-k}^* a_{-k} \rangle (e^{\beta\omega_k^0} - 1)/\omega_k^0, \tag{3·25}$$

$$\langle a_p^* a_{p+q} a_{q-k}^* a_{-k} \rangle = n_p n_{-k} \delta_{q,0} + n_{-k}(1 + n_{q-k})\, \delta_{p,-k}, \tag{3·26}$$

where $\omega_k^0 = 2SJ(\mathbf{0}, \boldsymbol{k})$ and

$$n_q = \langle a_q^* a_q \rangle = 1/\{\exp[2\beta SJ(\mathbf{0}, \boldsymbol{q})] - 1\}. \tag{3·27}$$

Inserting these equations into (3·23), we thus obtain

$$\omega_k \cong 2SJ(\mathbf{0}, \boldsymbol{k}) + (2/N) \sum_q n_q [J(\boldsymbol{q}, \boldsymbol{k}+\boldsymbol{q}) - J(\mathbf{0}, \boldsymbol{k})]. \tag{3·28}$$

Taking the cubic lattices and employing the relation

$$\nabla_k^2 J(\boldsymbol{k}) = -b^2 J(\boldsymbol{k}), \tag{3·29}$$

where b is the nearest neighbor distance, we arrive at

$$\omega_k \cong 2\left(1-\frac{\epsilon}{2S}\right) SJ(\mathbf{0}, \boldsymbol{k}), \tag{3·30}$$

where we have used the long wavelength approximation for the background spin waves represented by n_q, and defined the parameter

$$\begin{aligned}\epsilon &= (b^2/3N) \textstyle\sum_q n_q q^2, \\ &= 2\pi b^2 \left(\frac{V}{N}\right) \zeta\left(\frac{5}{2}\right)\left(\frac{k_B T}{4\pi D}\right)^{5/2},\end{aligned} \tag{3·31}$$

where

$$D = 2S \lim_{q\to 0} J(\mathbf{0}, \boldsymbol{q})/q^2 = 2Sa^2 J, \quad (6a^2 = zb^2), \tag{3·32}$$

J being the magnitude of the exchange interaction, and a the lattice constant. Equation (3·30) agrees with Keffer and Loudon's expression,[9] and thus turns out to reproduce Dyson's theory of spin waves, which gives the spontaneous magnetization of a ferromagnetic correctly up to the lowest order correction arising from the exchange interaction of spin waves and having the temperature dependence of T^4.

§ 4. Spin pair correlation and scattering cross section

The spin pair correlation function, defined by

$$\phi_k^{\alpha}(t) = \langle \{S_k^{\alpha}(t), S_k^{\alpha *}\} \rangle - \langle S_k^{\alpha} \rangle \langle S_k^{\alpha *} \rangle, \quad (\alpha = 0, \pm), \tag{4·1}$$

the curly brackets denoting the symmetrized product $\{A, B\} = (AB+BA)/2$, plays an important role in various aspects of the theoretical investigation of ferromagnetics. In this section we shall discuss some properties of the spin pair correlation and the magnetic scattering cross section of neutrons.

With the aid of the fluctuation-dissipation theorem,[19] the spin pair correla tion function is related to the dynamical susceptibility in the following form:

$$\phi_k^{\alpha}(\omega) = E_\beta(\omega)\, (S_k^{\alpha}, S_k^{\alpha *})\, \Xi_k^{\alpha}(\omega), \tag{4·2}$$

where

$$E_\beta(\omega) = \frac{\hbar\omega}{2} \frac{e^{\beta\hbar\omega}+1}{e^{\beta\hbar\omega}-1} = E_\beta(|\omega|), \tag{4·3}$$

$$= \begin{cases} k_B T, & \hbar\omega \ll k_B T, \\ \hbar\omega/2, & \hbar\omega \gg k_B T, \end{cases} \tag{4·4}$$

which equals the average energy of a harmonic oscillator with the frequency ω at the temperature $T = 1/k_B\beta$. $\phi(\omega)$ and $\Xi(\omega)$ denote the Fourier components defined as

$$\phi(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}dt\, e^{-it\omega}\phi(t). \tag{4·5}$$

From the expression for the generating function (2·48), we obtain

$$\Xi_k{}^\alpha(\omega)=\frac{1}{2\pi}\int_0^\infty dt[e^{-it(\omega-\Omega_k{}^\alpha)}+e^{it(\omega-\Omega_k{}^\alpha)}]e^{-t\gamma_k{}^\alpha},$$

$$=\frac{1}{\pi}\frac{\gamma_k{}^\alpha}{(\gamma_k{}^\alpha)^2+(\omega-\Omega_k{}^\alpha)^2}, \tag{4·6}$$

where

$$\Omega_k{}^\alpha\equiv\alpha\omega_0+\omega_k{}^\alpha-\mathrm{Im}\,(\Gamma_k{}^\alpha), \tag{4·7}$$

$$\gamma_k{}^\alpha\equiv\mathrm{Re}\,(\Gamma_k{}^\alpha)>0. \tag{4·8}$$

The real part of Γ_k^α, namely γ_k^α, represents the damping constant, and will be shown later to be positive as it should be. It is a useful approximation, at least for long wavelength components, to take the damping constant to be zero. Then

$$\Xi_k{}^\alpha(\omega)\doteqdot\delta(\omega-\Omega_k{}^\alpha), \tag{4·9}$$

which is combined with (4·2) to yield

$$\phi_k{}^\alpha(t)\doteqdot E_\beta(\Omega_k{}^\alpha)(S_k{}^\alpha,S_k^{\alpha *})\,e^{it\Omega_k{}^\alpha},$$

$$\doteqdot E_\beta(\Omega_k{}^\alpha)(S_k{}^\alpha(t),S_k^{\alpha *}), \tag{4·10}$$

which provides us with a simple, though approximate, relation between the spin pair correlation and the susceptibility.

Application of (3·2) and (4·10) leads to

$$\phi_k{}^0=\langle S_k{}^0 S_k^{0*}\rangle\doteqdot k_BT(S_k{}^0,S_k^{0*}), \tag{4·11}$$

$$\phi_k{}^\pm=\langle\{S_k{}^\pm,S_k^{\pm *}\}\rangle\doteqdot E_\beta(\Omega_k{}^\pm)\frac{2N\sigma}{-\omega_0\mp\omega_k{}^\pm}. \tag{4·12}$$

It is sometimes convenient to use the unsymmetrized correlation function which can be obtained from

$$\langle S_k{}^\pm S_k^{\pm *}\rangle=\phi_k{}^\pm\pm N\sigma,\quad (k\neq 0), \tag{4·13}$$

$$=\begin{Bmatrix}2NS\\0\end{Bmatrix},\quad \text{as}\quad T\to 0. \tag{4·14}$$

With the aid of the inversion symmetry, (4·12) is rewritten as

$$\phi_k{}^\pm\doteqdot N\sigma\frac{e^{\beta(-\omega_0+\omega_k)}+1}{e^{\beta(-\omega_0+\omega_k)}-1}, \tag{4·15}$$

which is valid at all temperatures.

Above the Curie temperature T_c, (4·15) leads, in the absence of external field, to

$$\phi_k{}^0 = \frac{1}{2}\phi_k{}^{\pm} \doteq \frac{Nk_BT}{(g\mu_B)^2/\chi + (\omega_k/\sigma)_{\sigma=0}}, \quad (T>T_c), \tag{4·16}$$

which has the same form as (3·9), as it should. This equation provides us with a general expression for the static spin pair correlation. To see the asymptotic behavior of the pair correlation at very large distances, we define

$$r_1{}^2 = \frac{S(S+1)}{3k_BT}\lim_{q\to 0}\frac{\omega_q}{\sigma q^2}, \tag{4·17}$$

where the limit of $q\to 0$ always exists since ω_q should vanish and should become an even function of q, as $q\to 0$, due to the property of the exchange interaction and the symmetry of the system. Introducing the parameter κ_1 by

$$(\kappa_1 r_1)^2 = \chi_0/\chi, \quad \chi_0 = (g\mu_B)^2 S(S+1)/3k_BT, \tag{4·18}$$

Eq. (4·16) can be transformed to be

$$\phi_q{}^0 = \frac{NS(S+1)}{3r_1{}^2}\frac{1}{\kappa_1{}^2+q^2}, \quad \text{as} \quad q\to 0, \tag{4·19}$$

which is equivalent to the following expression for the spatial correlation:

$$\begin{aligned}\phi^0(\boldsymbol{R}) &= \langle S^0(\boldsymbol{r})\, S^0(\boldsymbol{r}+\boldsymbol{R})\rangle, \\ &= \left(\frac{V}{N}\right)\frac{S(S+1)}{3}\frac{1}{4\pi r_1{}^2}\frac{e^{-\kappa_1 R}}{R}, \quad \text{as} \quad R\to\infty,\end{aligned} \tag{4·20}$$

where $S^0(\boldsymbol{r})$ denotes the z component of a spin (or the spin density) at the position $\boldsymbol{r}$. Equation (4·20) justifies van Hove's phenomenological investigation of the asymptotic behavior of the spin pair correlation, and (4·17) and (4·18) determine van Hove's parameters.[3]

Assuming the Heisenberg model of spins and applying the Bogolyubov-Tjablikov approximation (3·17) to (4·16) and (4·17), we obtain

$$\phi_k{}^0 \simeq Nk_BT\Big/\left[\frac{(g\mu_B)^2}{\chi} + 2J(\boldsymbol{0},\boldsymbol{k})\right], \tag{4·21}$$

$$r_1{}^2 \simeq 2a^2 JS(S+1)/3k_BT, \tag{4·22}$$

which agree with de Gennes' result obtained by using the Weiss approximation.[5] Use of the Weiss approximation

$$\chi/\chi_0 = T/(T-T_c), \quad T_c = 2zJS(S+1)/3k_B, \tag{4·23}$$

yields from (4·18) and (4·22)

$$\kappa_1{}^2 \simeq (z/a^2)(T-T_c)/T_c, \quad r_1{}^2 \simeq (a^2/z)\,T_c/T, \tag{4·24}$$

z being the number of the nearest neighbor ions.

Below the Curie temperature T_c, (4·15) leads, in the absence of external field, to

$$\phi_k^{\pm} \doteqdot N\sigma \frac{e^{\beta\omega_k}+1}{e^{\beta\omega_k}-1}, \quad (T<T_c), \tag{4·25}$$

$$=N\sigma, \quad \text{as} \quad T\to 0. \tag{4·26}$$

In the limit of $k\to 0$, we obtain

$$\phi_k^{\pm} \doteqdot [2NS(S+1)/3r_1^2]\frac{1}{k^2}, \quad \text{as} \quad k\to 0, \tag{4·27}$$

where r_1^2 is defined by (4·17). In the vicinity of the Curie point, since σ and ω_k are small, (4·25) leads to

$$\phi_k^{\pm} \doteqdot 2Nk_BT_c/\left(\frac{\omega_k}{\sigma}\right) \simeq \frac{Nk_BT_c}{J(\mathbf{0}, \boldsymbol{k})}, \quad \frac{\sigma}{S} \ll S+1, \tag{4·28}$$

where the approximation (3·17) for the localized model has been used. It should be noted from (4·27) that the van Hove parameter κ_1 identically vanishes for the $\pm$ components. This is a result of the fact that the perpendicular susceptibility $\chi_{\perp}$ becomes infinity below the Curie temperature. This disagrees with Elliott and Marshall's result[4] and supports de Gennes' investigation.[5]

The spin correlation of the z component below the Curie point cannot be obtained from the present method. If we adopt the Weiss approximation, we obtain

$$(S_k^0, S_k^{0*}) = [N/(g\mu_B)^2]\chi_{\parallel}, \quad \text{as} \quad k\to 0, \tag{4·29}$$

where

$$\chi_{\parallel} = N^{-1}(\partial \bar{M}^0/(\partial H_c)_{H_c\to 0}, \tag{4·30}$$

$$\simeq \chi_0 T/2(T_c-T), \quad T\approx T_c. \tag{4·31}$$

It has been shown by several authors[4),5)] that, in the Weiss approximation, the spin correlation of the z component takes the same form as (4·21) :

$$\phi_k^0 \simeq Nk_BT/\left[\frac{(g\mu_B)^2}{\chi_{\parallel}} + 2J(\mathbf{0}, \boldsymbol{k})\right], \quad (T<T_c), \tag{4·32}$$

whence the parameter r_1 is given also by (4·22). In the vicinity of the Curie point, (4·31) and (4·22) are inserted into (4·18) to lead to

$$\kappa_1^2 \simeq 2(z/a^2)(T_c-T)/T_c, \quad T\approx T_c. \tag{4·33}$$

If the result of the Weiss approximation is correct, it may be supposed that, except in the vicinity of the Curie point, the magnetization $\bar{M}^0$ changes only very slightly with the magnetic field so that $\chi_{\parallel}$ and ϕ_0^0 may be very small, vanishing at $T=0$.

On the other hand, it is well known that, in the spin wave region, one cannot define the parallel susceptibility $\chi_{\parallel}$ in the absence of external field.[20] More precisely, in the presence of an external field H in the z direction, the additional magnetization is proportional to $\sqrt{H}$. We study this situation by calculating χ_k^0 with the use of the spin wave approximation (3·19). In this case, χ_k^0 is expressed as

$$\chi_k{}^0=(g\mu_B)^2 N^{-1}\sum_q\sum_p (a_{q+k}^* a_q,\, a_p{}^* a_{p+k}). \tag{4·34}$$

For $k\neq 0$, we obtain

$$(a_{q+k}^* a_q,\, a_p{}^* a_{p+k})=\delta_{q,p}\frac{e^{(\omega_{q+k}-\omega_q)}-1}{\omega_{q+k}-\omega_q}N_{q+k}(N_q+1), \tag{4·35}$$

where

$$\begin{aligned} &N_q=1/[\exp(\beta\Omega_q)-1],\\ &\Omega_q=\omega_q+g\mu_B H,\quad \omega_q=Dq^2, \end{aligned} \tag{4·36}$$

D being defined by (3·32). For the small values of k which satisfy the condition $Dk^2\ll k_BT$ and for those values of q such that $Dq^2\sim k_BT$ which give the main contribution to the sum (4·34), we have

$$\omega_{q+k}-\omega_q\ll k_BT.$$

Equation (4·35) for the small values of k thus reduces to

$$(a_{q+k}^* a_q,\, a_p{}^* a_{p+k})\cong\delta_{q,p}\beta N_{q+k}(N_q+1), \tag{4·37}$$

which leads to

$$\chi_k{}^0=(g\mu_B)^2\beta N^{-1}\sum_q N_{k+q}\beta(N_q+1). \tag{4·38}$$

For $H=0$ and $k=0$, the summation in this expression diverges at $q=0$. In the case in which Dk^2, $g\mu_BH\ll k_BT$, we can approximate the summation to yield the following expression for χ_k^0:

$$\chi_k{}^0=\frac{(g\mu_B)^2 k_BT}{8\pi^2 D^2\rho k}f\left(\frac{g\mu_BH}{Dk^2}\right),\quad \rho\equiv\frac{N}{V}, \tag{4·39}$$

where

$$f(x)=\int_0^\infty\frac{t}{t^2+x}\ln\frac{(t+1)^2+x}{(t-1)^2+x}dt, \tag{4·40}$$

$$=\begin{cases}\pi/\sqrt{x}, & x\gg 1,\\ \pi^2, & x\ll 1.\end{cases} \tag{4·41}$$

We consider the two limiting cases of this expression:

(1) $Dk^2 \ll g\mu_B H \ll k_B T$;

$$\chi_k{}^0 = \frac{(g\mu_B)^{3/2} k_B T}{8\pi D^{3/2} \rho} \frac{1}{\sqrt{H}}. \tag{4·42}$$

This is the uniform case considered by Holstein and Primakoff.[20]

(2) $g\mu_B H \ll Dk^2 \ll k_B T$;

$$\chi_k{}^0 = \frac{(g\mu_B)^2 k_B T}{8D^2 \rho} \frac{1}{k}. \tag{4·43}$$

As we shall see in § 6, the behavior of χ_k^0 for the small values of k is essential for determining the k dependence of the damping of the z spin component at very low temperatures.

Before concluding the discussion about the longitudinal correlation, we add a few words about what happens if we adopt the Bogolyubov-Tyablikov approximation. χ_k^0 for the spin one half case was calculated by the authors[21] which is, in the absence of magnetic field and for small values of k, expressed as

$$\chi_k{}^0 = \frac{(g\mu_B)^2}{2Ja^2} \frac{1}{k(k+\delta)}, \quad (T < T_c), \tag{4·44}$$

where δ is a positive constant vanishing at the Curie point. In the spin wave region, where $k+\delta$ is replaced by the value of δ evaluated at 0°K, this expression is shown to agree with the spin wave result (4·43). However, it should be noted that this anomalous correlation in the entire temperature region below the Curie point is still open to question, and, moreover, this anomaly is eliminated by introducing the small amount of anisotropy energy or the constant external field.

Next we shall express the magnetic scattering cross section of neutrons by a ferromagnetic in terms of the frequency spectrum $\omega_k^{\pm}$ and the damping constants γ_k^{α}, to show clearly that the collective motion of the system described by the macroscopic equations (2·48) or (2·50) is the collective excitation observed by the neutron scattering experiment. Employing the formula for the differential scattering cross section given by van Hove,[3] we have

$$\frac{d^2\sigma}{d\Omega\, d\omega} = A(\boldsymbol{K}, \boldsymbol{K}_0) \sum_{\mu,\nu} (\delta_{\mu,\nu} - q_\mu q_\nu / q^2) \frac{1}{2\pi} \int_{-\infty}^{\infty} dt\, e^{-it\omega} \langle S_{\mu q}^* S_{\nu q}(t) \rangle, \tag{4·45}$$

where

$$A(\boldsymbol{K}, \boldsymbol{K}_0) \equiv \left(\frac{2g_0 e^2}{m_0 c^2} \right)^2 \frac{K}{K_0} |F(\boldsymbol{q})|^2, \tag{4·46}$$

and $\boldsymbol{K}_0$ and $\boldsymbol{K} = \boldsymbol{K}_0 - \boldsymbol{q}$ are the initial and final wave vectors of the neutron, whereas $\hbar\omega = \hbar^2 (K_0^2 - K^2)/2m$, m being the neutron mass. The indices $\mu, \nu = x, y, z$ refer to rectangular coordinates in space. In the following, we consider the fluctuation part which is obtained by replacing $S_{\mu q}$ in (4·45) by the fluctuation part $S'_{\mu q} = S_{\mu q} - \langle S_{\mu q} \rangle$. Since

$$\langle S_{\mu q}^{\prime *} S_{\nu q}(t)\rangle = \langle \{S_{\mu q}^{\prime *}, S_{\nu q}(t)\}\rangle + \frac{\hbar}{2i}(S_{\mu q}^{*}, \dot{S}_{\nu q}(t)), \tag{4·47}$$

we have

$$\lim_{\varepsilon\to 0+}\int_{-\infty}^{\infty} dt\, e^{-it\omega-\varepsilon|t|}\langle S_{\mu q}^{\prime *} S_{\nu q}(t)\rangle = \left[E_\beta(\omega)+\frac{\hbar\omega}{2}\right]\int_{-\infty}^{\infty} dt\, e^{-it\omega}(S_{\mu q}^{*}, S_{\nu q}(t)). \tag{4·48}$$

Therefore, in terms of the dynamical susceptibility, we obtain

$$\left(\frac{d^2\sigma}{d\Omega\, d\omega}\right)_{fluct.} = A(\boldsymbol{K}, \boldsymbol{K}_0)\left[\frac{\omega}{2}+E_\beta(\omega)\right]\frac{N}{(g\mu_B)^2}\left\{\left(1-\frac{q_z^2}{q^2}\right)\chi_q^{0}(\omega)\right.$$
$$\left. +\frac{1}{2}\left(1-\frac{q_x^2+q_y^2}{2q^2}\right)[\chi_q^{+}(\omega)+\chi_q^{-}(\omega)]\right\}. \tag{4·49}$$

Use of (4·6) thus leads to

$$\left(\frac{d^2\sigma}{d\Omega\, d\omega}\right)_{fluct.} = A(\boldsymbol{K}, \boldsymbol{K}_0)\left[\frac{\omega}{2}+E_\beta(\omega)\right]\frac{N}{\pi(g\mu_B)^2}\left\{\left(1-\frac{q_z^2}{q^2}\right)\chi_q^{0}\frac{\gamma_q^{0}}{\omega^2+(\gamma_q^{0})^2}\right.$$
$$\left. +\frac{1}{4}\left(1+\frac{q_z^2}{q^2}\right)\chi_q^{+}\left[\frac{\gamma_q^{+}}{(\omega-\Omega_q^{+})^2+(\gamma_q^{+})^2}+\frac{\gamma_q^{-}}{(\omega-\Omega_q^{-})^2+(\gamma_q^{-})^2}\right]\right\}, \tag{4·50}$$

which is valid at all temperatures and holds irrespective of the specialized models of the ferromagnetic spins.

Far below the Curie temperature, the spin correlation of z component χ_q^0 may become small and the damping factor γ_q^α may be neglected for long wave-length components. Thus if one neglects the longitudinal component, (4·50) takes the form

$$\left(\frac{d^2\sigma}{d\Omega\, d\omega}\right)_{fluct.} \simeq A(\boldsymbol{K}, \boldsymbol{K}_0)\frac{1}{2}\left(1+\frac{q_z^2}{q^2}\right)N\sigma\left[(N_q+1)\,\delta(\omega-\Omega_q)+N_q\,\delta(\omega+\Omega_q)\right], \tag{4·51}$$

where it has been used that $\Omega_q=-\omega_0+\omega_q\geqq 0$,

$$E_\beta(\Omega_q)=\Omega_q[1/2+N_q],\quad N_q=1/[\exp(\beta\Omega_q)-1]. \tag{4·52}$$

Equation (4·51) expresses that the scattered neutrons create and absorb the collective modes described by the macroscopic equation (2·21) or (2·33), whose energy spectrum is given by (2·24). It was shown in § 3 that these collective modes turn out to be the usual spin wave excitations at very low temperatures.

Above the Curie point, we have $\Omega_q^{\pm}=0$, $\gamma_q\equiv\gamma_q^0=\gamma_q^{\pm}$, $\chi_q^0=(1/2)\chi_q^{\pm}$ in the absence of external field so that, employing (3·9) and (4·4),

$$\left(\frac{d^2\sigma}{d\Omega\, d\omega}\right)_{fluct.} = A(\boldsymbol{K}, \boldsymbol{K}_0)\frac{2Nk_BT}{(g\mu_B)^2/\chi+(\omega_q/\sigma)_{\sigma=0}}\frac{1}{\pi}\frac{\gamma_q}{\omega^2+\gamma_q^2}, \tag{4·53}$$

where we have required that $\hbar\omega\ll k_BT$. With the assumption of the diffusion type damping for γ_q, (4·53) becomes identical with that obtained by van Hove[3] with the aid of a phenomenological investigation of the correlation of spins. Very recently, Ericson and Jacrot have performed a neutron scattering measurment which serves for a determination of the damping constant γ_q for iron. This problem will be discussed in § 7 on the basis of our theoretical results.

§ 5. Damping and shift

In the previous sections, we were mainly concerned with the frequency spectrum ω_k^α, ($\alpha=0, \pm$), and with the general properties of the collective motions of the system described by (2·48) and (2·50). In the present section, we shall investigate the damping constants γ_k^α and the frequency shift $\Delta\omega_k^\alpha$, which are defined by the real and imaginary parts of Γ_k^α, respectively:

$$\Gamma_k{}^\alpha = -i\Delta\omega_k{}^\alpha + \gamma_k{}^\alpha, \tag{5·1}$$

$$= (\Gamma_{-k}^{-\alpha})^*, \tag{5·2}$$

where the second equation can be obtained by using (3·5).

Since $I(t) = e^{itH} I e^{-itH}$, $(A(t), B) = (A, B(-t))$, it turns out from (2·17) that

$$\gamma_k{}^\alpha = \frac{1}{2(S_k{}^\alpha, S_k^{\alpha*})} \int_{-\infty}^{\infty} dt\, e^{-it(\alpha\omega_0+\omega_k{}^\alpha)} (I_k{}^\alpha(t), I_k^{\alpha*}), \tag{5·3}$$

$$\Delta\omega_k{}^\alpha = \frac{i}{2(S_k{}^\alpha, S_k^{\alpha*})} \int_{-\infty}^{\infty} dt\, \mathrm{Sgn}(t)\, e^{-it(\alpha\omega_0+\omega_k{}^\alpha)} (I_k{}^\alpha(t), I_k^{\alpha*}), \tag{5·4}$$

where

$$I_k{}^\alpha = \dot{S}_k{}^\alpha - i\omega_k{}^\alpha S_k{}^\alpha = (I_{-k}^{-\alpha})^*. \tag{5·5}$$

Applying the fluctuation-dissipation theorem,[19] we obtain

$$\gamma_k{}^\alpha = \frac{1}{2(S_k{}^\alpha, S_k^{\alpha*})} \frac{1}{E_\beta(\alpha\omega_0+\omega_k{}^\alpha)} \int_{-\infty}^{\infty} dt\, e^{-it(\alpha\omega_0+\omega_k{}^\alpha)} \langle \{I_k{}^\alpha(t), I_k^{\alpha*}\} \rangle, \tag{5·6}$$

$$\Delta\omega_k{}^\alpha = \frac{-1}{(S_k{}^\alpha, S_k^{\alpha*})} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{\mathcal{P}}{\omega-(\alpha\omega_0+\omega_k{}^\alpha)} \frac{1}{E_\beta(\omega)} \int_{-\infty}^{\infty} dt\, e^{-it\omega} \langle \{I_k{}^\alpha(t), I_k^{\alpha*}\} \rangle, \tag{5·7}$$

where, in deriving the second equation, use has been made of

$$\frac{i}{2} \int_{-\infty}^{\infty} dt\, \mathrm{Sgn}(t)\, e^{-it\omega} = \frac{\mathcal{P}}{\omega}. \tag{5·8}$$

Since, as can be seen easily by writing down in the representation diagonalizing the total Hamiltonian H,

$$(A, A^*) \geqq 0, \quad \int_{-\infty}^{\infty} dt\, e^{-it\omega} (A(t), A^*) \geqq 0, \tag{5·9}$$

we obtain

$$\gamma_k{}^\alpha \geqq 0, \tag{5·10}$$

which guarantees that γ_k^α represent the damping constants for the temporal development of the magnetization.

According to the requirement of time reversibility, the total Hamiltonian $H(\omega_0)$ and the dynamical motion of the system are invariant with respect to the simultaneous change of signs of the time t and the external magnetic field ω_0, whereas the spin operator changes the sign: namely, denoting the time reversal operator by K,[22)]

$$K^{-1} H(\omega_0) K = H(-\omega_0), \tag{5·11}$$

$$K^{-1} iK = -i, \tag{5·12}$$

$$K^{-1} S_k{}^\alpha K = -S_{-k}^{-\alpha} = S_k^{\alpha *}, \quad (\alpha = 0, \pm). \tag{5·13}$$

And, if the average value of a quantity F is real,

$$\mathrm{Tr}\, \rho(\omega_0) F = \mathrm{Tr}\, \rho(-\omega_0) K^{-1} F K. \tag{5·14}$$

Therefore, it follows that

$$\chi_k{}^\alpha(\sigma, \omega_0) = \chi_k{}^\alpha(-\sigma, -\omega_0), \tag{5·15}$$

$$\omega_k{}^\alpha(\sigma, \omega_0) = -\omega_k{}^\alpha(-\sigma, -\omega_0), \tag{5·16}$$

and

$$\Delta\omega_k{}^\alpha(\sigma, \omega_0) = -\Delta\omega_k{}^\alpha(-\sigma, -\omega_0), \tag{5·17}$$

$$\gamma_k{}^\alpha(\sigma, \omega_0) = \gamma_k{}^\alpha(-\sigma, -\omega_0), \tag{5·18}$$

where we have used that $K^{-1} I_k^\alpha(\omega_0) K = [I_k^\alpha(-\omega_0)]^*$.

If the system has the inversion symmetry with respect to each lattice point, we have

$$\Delta\omega_k{}^\pm = -\Delta\omega_{-k}^\mp = -\Delta\omega_k{}^\mp, \quad \Delta\omega_k{}^0 = 0, \tag{5·19}$$

$$\gamma_k{}^\alpha = \gamma_{-k}^{-\alpha} = \gamma_k^{-\alpha}. \tag{5·20}$$

Especially above the Curie temperature, we have, in the absence of external magnetic field,

$$\Delta\omega_k{}^\pm = 0, \quad \gamma_k{}^\pm = \gamma_k{}^0, \tag{5·21}$$

where the second equation can be derived by writing down $S_k^\pm$ in terms of S_{xk} and S_{yk}, and assuming the equivalence of the x, y and z directions.

In the following sections, we shall calculate the damping constants γ_k^α and the frequency shift $\Delta\omega_k^\alpha$, and discuss the spin wave damping and the spin diffusion.

§ 6. Damping constants in the spin wave region

In the present section, we shall calculate the three components of the damping, $\gamma_k^{\pm}$ and γ_k^0, due to the exchange interaction, employing the spin wave approximation. In terms of the spin wave terminology, $\gamma_k^{\pm}$ represent the amplitude damping, while γ_k^0 corresponds to the damping of the spin wave density.

The problem of spin wave damping was discussed by many authors,[17),23)] mostly starting from the kinetic theory and the spin wave interaction of the Dyson type. In the kinetic theoretical treatments, one calculates the rate of decrease of the number of spin waves due to the interaction, and identifies the half of this rate with the amplitude damping constant. However, the validity of this procedure is not always obvious. Recently, Akhiezer, Bar'yakhtar and Peletminskii[24)] formulated the amplitude damping starting from Kubo's general theory of linear response[19)] and employing the quantum field theoretic techniques. Their method, although elegant, is rather complicated. We shall see that the same result is obtained as a simple application of the present general method. In the last half part, the calculation of γ_k^0 will be carried out and it will be seen that the damping of the z component is quite different from the amplitude damping.

As was shown in § 2, the damping constants are expressed in terms of the time correlation of the torques I_k^{α} arising from the irregular fluctuation of spins around the collective oscillation described by (2·33). At low temperatures, this collective oscillation turned out to be the spin wave excitation and the frequency spectrum was given by (3·28). We are now in a position to calculate the damping effect on this oscillation due to the exchange interaction. Insertion of (3·28) and (3·13) into (5·5) leads to

$$I_k^+ = -2iN^{-1}\sum_q J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q})[S_q^0 - NS\delta_{q,0}]S_{k-q}^+ \\ +2iN^{-1}\sum_q [J(\boldsymbol{q}, \boldsymbol{k}+\boldsymbol{q}) - J(\boldsymbol{0}, \boldsymbol{k})]n_q S_k^+, \tag{6·1}$$

which, by using the spin wave expansion (3·18) and (3·19), and retaining up to the third order, becomes

$$I_k^+ = 2i\sqrt{2S/N}\left\{\sum_q\sum_r J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q})\, a_r^* a_{r+q} a_{k-q} \right.\\ \left. - \sum_q [J(\boldsymbol{0}, \boldsymbol{k}) - J(\boldsymbol{q}, \boldsymbol{k}+\boldsymbol{q})] n_q a_k\right\}. \tag{6·2}$$

It is convenient to put this expression in a slightly different form:

$$I_k^+ = 2i\sqrt{2S/N}\sum_q\sum_r J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q})\,\{a_r^* a_{r+q} a_{k-q} - \langle a_r^* a_{r+q}\rangle a_{k-q} \\ - \langle a_r^* a_{k-q}\rangle a_{r+q}\}, \tag{6·3}$$

$$= 2i\sqrt{2S/N}\sum_q\sum_r J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q})\,\{a_r^* a_{r+q} a_{k-q} - (\text{RPA})\}. \tag{6·4}$$

The last two terms of (6·3) are the operators obtained by applying the random phase approximation to the foregoing operator, whence we have denoted them by (RPA). In order to evaluate (2·49), we neglect the interaction between the spin waves in the temporal development of $I_k^\alpha(t)$ and in the canonical ensemble ρ, following the spirit of perturbational treatment. This allows us to obtain the following: defining $\omega_k \equiv \omega_k^- = -\omega_k^+$,

$$\int_0^\infty dt\, e^{it\omega_k - \varepsilon t} I_k^+(t)$$

$$\cong -2\sqrt{2S/N} \sum_q \sum_r J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q}) \left\{ \frac{1}{\omega_k + \omega_r - \omega_{r+q} - \omega_{k-q} + i\varepsilon} a_r^* a_{r+q} a_{k-q} - (\text{RPA}) \right\}, \tag{6·5}$$

$$\int_0^\beta d\lambda\, e^{\lambda H} I_k^{+*} e^{-\lambda H}$$

$$\cong -2i\sqrt{2S/N} \sum_q \sum_r J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q}) \left\{ \frac{e^{\beta(\omega_{k-q} + \omega_{r+q} - \omega_r)} - 1}{\omega_{k-q} + \omega_{r+q} - \omega_r} a_{k-q}^* a_{r+q}^* a_r - (\text{RPA}) \right\}, \tag{6·6}$$

where ε is a small positive number which is taken to be zero after calculation. From (3·9), it follows that

$$(S_k^+, S_k^{+*}) = 2N\sigma/\omega_k \cong 2NS/\omega_k. \tag{6·7}$$

Insertion of (6·5), (6·6) and (6·7) into (2·49) immediately leads to

$$\Gamma_k^+ = (4\omega_k/N^2) \sum_{q,r} \sum_{q',r'} J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q})\, J(\boldsymbol{q}', \boldsymbol{k}-\boldsymbol{q}') \frac{e^{\beta(\omega_{k-q'} + \omega_{r'+q'} - \omega_{r'})} - 1}{\omega_{k-q'} + \omega_{r'+q'} - \omega_{r'}}$$

$$\times \frac{1}{i(\omega_{r+q} + \omega_{k-q} - \omega_k - \omega_r) + \varepsilon} \langle a_{k-q'}^* a_{r'+q'}^* a_{r'} | a_r^* a_{r+q} a_{k-q} \rangle_c. \tag{6·8}$$

The symbol $\langle\, | \,\rangle_c$ means that any operator on either side of the center line should be paired with one in another side of the line to evaluate the ensemble average. Other terms exactly cancel with those arising from (RPA) in (6·5) and (6·6). This expectation value is easily shown to be

$$\langle a_{k-q'}^* a_{r'+q'}^* a_{r'} | a_r^* a_{r+q} a_{k-q} \rangle_c = (N_r + 1) N_{r+q} N_{k-q} \delta_{r',r} (\delta_{q',q} + \delta_{q',k-q-r}), \tag{6·9}$$

where N_q is the Bose distribution defined by (4·52). Thus, (6·8) turns out to be

$$\Gamma_k^+ = (4\omega_k/N^2) \sum_{q,r} J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q}) [J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q}) + J(\boldsymbol{k}-\boldsymbol{q}-\boldsymbol{r}, \boldsymbol{q}+\boldsymbol{r})]$$

$$\times \frac{e^{\beta(\omega_{r+q} + \omega_{k-q} - \omega_r)} - 1}{\omega_{r+q} + \omega_{k-q} - \omega_r} \frac{1}{i(\omega_{r+q} + \omega_{k-q} - \omega_k - \omega_r) + \varepsilon} (N_r + 1) N_{r+q} N_{k-q}. \tag{6·10}$$

Now we take the real part of this. Using the relation

$$\frac{1}{ix+\varepsilon}=\pi\delta(x)-i\frac{\mathcal{P}}{x}, \quad (\varepsilon\to 0), \tag{6·11}$$

we thus arrive at the following expression for the amplitude damping constant:

$$\gamma_k^+=(4\pi/N^2)\sum_{q,r}J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})[J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})+J(\boldsymbol{k}-\boldsymbol{q}-\boldsymbol{r},\boldsymbol{q}+\boldsymbol{r})]$$

$$\times\delta(\omega_{r+q}+\omega_{k-q}-\omega_k-\omega_r)\frac{(N_r+1)N_{r+q}N_{k-q}}{N_k} \tag{6·12}$$

$$=(2\pi/N^2)\sum_{q,r}[J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})+J(\boldsymbol{k}-\boldsymbol{q}-\boldsymbol{r},\boldsymbol{q}+\boldsymbol{r})]^2$$

$$\times\delta(\omega_{r+q}+\omega_{k-q}-\omega_k-\omega_r)\frac{(N_r+1)N_{r+q}N_{k-q}}{N_k}, \tag{6·13}$$

where the second equation has been obtained by rearranging the terms by changing the summation variable $\boldsymbol{q}$ to $\boldsymbol{k}-\boldsymbol{q}-\boldsymbol{r}$ and adding the resulting expression to (6·12).

As one can verify directly, the following identity holds when the condition $\omega_k+\omega_r=\omega_{r+q}+\omega_{k-q}$ is satisfied:

$$\frac{(N_r+1)N_{r+q}N_{k-q}}{N_k}=N_r(N_{r+q}+1)(N_{k-q}+1)-(N_r+1)N_{r+q}N_{k-q}. \tag{6·14}$$

Application of this identity to (6·13) leads to the form to be compared with the kinetic theoretical result. Namely,

$$\gamma_k^+=(2\pi/N^2)\sum_{q,r}[J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})+J(\boldsymbol{k}-\boldsymbol{q}-\boldsymbol{r},\boldsymbol{q}+\boldsymbol{r})]^2\delta(\omega_k+\omega_r-\omega_{r+q}-\omega_{k-q})$$

$$\times\left\{N_r(N_{r+q}+1)(N_{k-q}+1)-(N_r+1)N_{r+q}N_{k-q}\right\}. \tag{6·15}$$

This is just one half of the rate of decrease of the number of spin waves with the wave vector $\boldsymbol{k}$ obtained from the interaction

$$-(1/2N)\sum_k\sum_r\sum_q[J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})+J(\boldsymbol{k}-\boldsymbol{q}-\boldsymbol{r},\boldsymbol{q}+\boldsymbol{r})]a_{r+q}^*a_{k-q}^*a_k a_r, \tag{6·16}$$

by using the kinetic treatment with Schlömann's assumption.[17] This is nothing but Dyson's dynamical interaction. Thus, we have shown that our method gives not only the correct spin wave spectrum, but also the correct transition rate equivalent to Dyson's, by working directly with the original Hamiltonian.

As aforementioned, the same result as ours was obtained by I. A. Akhiezer et al.[24] However, they started from the Hamiltonian expressed in terms of the spin wave operators, which consists of the free spin wave Hamiltonian and Dyson's dynamical interaction, ignoring the kinematical interaction completely. Strictly speaking, one has to justify this Hamiltonian as has been done by Dyson,[7] Oguchi[25] and others.[26] On the other hand, we started from the original Hamiltonian with the exchange interaction, and, only after obtaining the general expression for the damping constant, we used the spin wave approximation. The spin wave interaction has come out automatically from the original equation of motion for the spin operators. As will be shown in Appendix B, this is a direct consequence of the fact that the change in time of the transverse spin operator can be written, in the spin wave region, simply in terms of Dyson's dynamical interaction.

Next we calculate the damping of the z component γ_k^0. Since $\omega_k^0=0$, the z component of the torque (5·5) takes the form

$$I_k{}^0=\dot{S}_k{}^0=-iN^{-1}\sum_q J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q})\, S_q{}^+ S_{k-q}^- . \qquad (6\cdot17)$$

Using the spin wave approximation (3·18) and retaining only the lowest order term, we obtain

$$I_k{}^0=-2iS\sum_q J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q})\, a_q a_{q-k}^* . \qquad (6\cdot18)$$

In order to evaluate (5·6) in the case of $\alpha=0$, we neglect the interaction between the spin waves in the temporal development of $I_k^0(t)$ and in the ensemble average. Insertion of (6·18) into (5·6) thus leads to

$$\gamma_k{}^0=\frac{(g\mu_B)^2}{Nk_BT\chi_k{}^0}(2S)^2\pi\sum_{q,q'} J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q})\, J(\boldsymbol{q}', \boldsymbol{k}-\boldsymbol{q}')\,\delta(\omega_q-\omega_{q-k})$$
$$\times\langle\{a_q a_{q-k}^*, a_{q'-k} a_{q'}^*\}\rangle . \qquad (6\cdot19)$$

It is easily shown that

$$\langle\{a_q a_{q-k}^*, a_{q'-k} a_{q'}^*\}\rangle=\frac{1}{2}\delta_{q',q}[(N_q+1)N_{q-k}+N_q(N_{q-k}+1)], \quad (k\neq 0). \qquad (6\cdot20)$$

Thus we arrive at the following expression for the damping of the longitudinal spin component:

$$\gamma_k{}^0=\frac{4\pi S^2(g\mu_B)^2}{Nk_BT\chi_k{}^0}\sum_q [J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q})]^2\delta(\omega_q-\omega_{k-q})\, N_q(N_{k-q}+1), \qquad (6\cdot21)$$

where we have rearranged the term arising from the first term of (6·20) by changing the summation variable $\boldsymbol{q}$ to $\boldsymbol{k}-\boldsymbol{q}$ and applying the relations $\omega_q=\omega_{-q}$ and $J(\boldsymbol{q}, \boldsymbol{q}')=-J(\boldsymbol{q}', \boldsymbol{q})$. It may be observed that (6·21) is quite different from (6·13) and is out of the usual kinetic theoretical understanding.

To evaluate (6·21) explicitly, we take the long wavelength approximation

$$\omega_q\simeq Dq^2, \quad J(\boldsymbol{q}, \boldsymbol{0})\simeq -Cq^2, \qquad (6\cdot22)$$

where

$$C=a^2J, \quad D=2Sa^2J\left(1-\frac{\epsilon}{2S}\right). \qquad (6\cdot23)$$

Since $J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q})=-2C\boldsymbol{k}\cdot\boldsymbol{q}$ as $k\to 0$, (6·21) leads, for the long wavelength components, to

$$\gamma_k^0 = \frac{4\pi S^2 (g\mu_B)^2}{Nk_B T\chi_k^0} \frac{4C^2}{D} \sum_q (\boldsymbol{k}\cdot\boldsymbol{q})^2 \delta(k^2 - 2\boldsymbol{k}\cdot\boldsymbol{q}) N_q (N_{k-q}+1). \quad (6\cdot24)$$

The summation is easily evaluated to yield

$$\gamma_k^0 = \frac{16\pi S^2 C^2 (g\mu_B)^2}{NDk_B T\chi_k^0} k \frac{V}{(2\pi)^3} 2\pi \int_{k/2}^{\infty} dq \int_0^1 dx\, \delta(k-2qx)\, q^4 x^2 N_q (N_q+1), \quad (6\cdot25)$$

$$= \alpha \frac{(g\mu_B)^2}{\chi_k^0} \frac{k^3}{\exp[\beta(g\mu_B H + Dk^2/4)] - 1}, \quad (6\cdot26)$$

where

$$\alpha = \frac{V}{N} \cdot \frac{S^2 C^2}{4\pi D^2} \simeq \frac{1}{16\pi} \frac{V}{N}, \quad (6\cdot27)$$

and H is the sum of the external field present and the effective field due to the anisotropy energy and the dipolar interaction. Equation (6·26) reveals a complicated behavior of the damping constant γ_k^0 at low temperatures depending on the relative magnitudes of the various quantities involved. For instance, for $Dk^2 \gg g\mu_B H \gg k_B T$, use of (4·34) and (4·35) leads to

$$\chi_k^0 \simeq \frac{2S(g\mu_B)^2}{Dk^2} \left(1 - \frac{\sigma}{S}\right), \quad (6\cdot28)$$

which is inserted into (6·26) to yield

$$\gamma_k^0 = \alpha \left(1 - \frac{\sigma}{S}\right) \frac{D}{2S} k^5 \exp\left(-\frac{Dk^2}{4k_B T}\right). \quad (6\cdot29)$$

For the opposite case, where Dk^2, $g\mu_B H \ll k_B T$, use of (6·26) and (4·39) yields

$$\gamma_k^0 = \frac{\pi}{2} D^2 \frac{k^4}{[Dk^2/4 + g\mu_B H] f(g\mu_B H/Dk^2)}. \quad (6\cdot30)$$

In particular, for $Dk^2 \ll g\mu_B H$, this becomes

$$\gamma_k^0 = \frac{1}{2} \frac{D^{3/2}}{(g\mu_B H)^{1/2}} k^3, \quad (6\cdot31)$$

and, for $Dk^2 \gg g\mu_B H$, we obtain

$$\gamma_k^0 = (2/\pi) Dk^2. \quad (6\cdot32)$$

Equation (6·32) shows that, in the spin wave region, the longitudinal damping obeys the diffusion equation,

$$\frac{d}{dt} M_k^0 = -k^2 \Lambda M_k^0, \quad (6\cdot33)$$

with the diffusion constant

$$\Lambda = (4S/\pi) a^2 (J/\hbar), \quad (6\cdot34)$$

if and only if the following condition is satisfied:

$$ak \ll 1, \quad g\mu_B H \ll Dk^2 \ll k_B T. \tag{6·35}$$

It is interesting to note that the diffusion constant obtained in the spin wave region is of the same order of magnitude as that obtained in the high temperature limit, (7·17).

§ 7. Spin diffusion

In the preceding section, we calculated the spin wave damping and the damping of the z spin component at very low temperatures. As the temperature goes up, the spin waves lose the parts of good normal modes, vanishing at the Curie temperature, whereas the contribution of the z component becomes predominant. Above the Curie temperature, there is no collective oscillation and the motion of the magnetization is determined entirely by the damping constant γ_k^0. In this section we shall discuss this longitudinal damping constant in the vicinity of the Curie point and in the high temperature limit, using the localized model of ferromagnetic spins.

As was shown in the preceding section, the longitudinal damping constant has a quite different feature from the amplitude damping, and cannot be treated with the usual kinetic method even in the spin wave region. Thus this phenomenon provides us with an example of the so-called non-Boltzmann transport processes. Although a great deal of knowledge has been accumulated about the fundamental aspect of the usual processes,[27] we know little about the non-Boltzmann processes. This very often makes it difficult to do a theoretical investigation.

In fact, it is difficult to perform the exact integration of the time correlation functions of the torques, (5·3), in the vicinity of the Curie point and in the paramagnetic region. In parallel with the treatment of the exchange narrowing in paramagnetic resonance absorption,[16] we therefore assume a Gaussian decay for the function

$$f(t) = e^{-it[\omega_k{}^\alpha + \alpha\omega_0]} (I_k{}^\alpha(t), I_k^{\alpha *}) / (I_k{}^\alpha, I_k^{\alpha *}), \tag{7·1}$$

in the following form:

$$f(t) = e^{it\nu_{k\alpha}} e^{-t^2 g_{k\alpha}{}^2}. \tag{7·2}$$

Then use of (5·3) and (5·4) leads to

$$\gamma_k{}^\alpha = \frac{(I_k{}^\alpha, I_k^{\alpha *})}{(S_k{}^\alpha, S_k^{\alpha *})} \frac{\sqrt{\pi}}{2g_{k\alpha}} e^{-(\nu_{k\alpha}/2g_{k\alpha})^2}, \tag{7·3}$$

$$\Delta\omega_k{}^\alpha = -\frac{(I_k{}^\alpha, I_k^{\alpha *})}{(S_k{}^\alpha, S_k^{\alpha *})} \frac{1}{g_{k\alpha}} e^{-(\nu_{k\alpha}/2g_{k\alpha})^2} \int_0^{(\nu_{k\alpha}/2g_{k\alpha})} dx\, e^{-x^2}. \tag{7·4}$$

The quantity $\tau_c = \sqrt{\pi}/2g_{k\alpha}$ represents the correlation time of the torques $I_k^\alpha(t)$. The parameters $g_{k\alpha}$ and $\nu_{k\alpha}$ are determined by

$$\nu_{k\alpha} + \omega_k{}^\alpha + \alpha\omega_0 = ([H, I_k{}^\alpha], I_k^{\alpha *}) / (I_k{}^\alpha, I_k^{\alpha *}), \tag{7·5}$$

$$2g_{k\alpha}^2 = -(\nu_{k\alpha} + \omega_k{}^\alpha + \alpha\omega_0)^2 + ([H, I_k{}^\alpha], [I_k^{\alpha *}, H]) / (I_k{}^\alpha, I_k^{\alpha *}). \tag{7·6}$$

It can be shown, in parallel with the properties of $\Delta\omega_k^\alpha$ and γ_k^α, that $\nu_{k0} = 0$ and, above the Curie point and in the absence of external field, $\nu_{k\pm} = 0$, $g_{k\mp} = g_{k0}$.

Now we calculate the longitudinal damping constant

$$\gamma_k{}^0 = \frac{(I_k{}^0, I_k^{0*})}{(S_k{}^0, S_k^{0*})} \frac{\sqrt{\pi}}{2g_{k0}}, \quad g_{k0}^2 = \frac{(\dot{I}_k{}^0, \dot{I}_k^{0*})}{2(I_k{}^0, I_k^{0*})}. \tag{7·7}$$

Using (3·1) and (6·17), we obtain

$$\begin{aligned}(I_k{}^0, I_k^{0*}) &= -i\langle [S_k{}^0, I_k^{0*}] \rangle, \\ &= 2N^{-1} \sum_q J(\boldsymbol{q}, \boldsymbol{k}+\boldsymbol{q}) \langle \{S_q{}^+, S_q^{+*}\} \rangle,\end{aligned} \tag{7·8}$$

where use has been made of the relations $\Sigma_q J(\boldsymbol{q}, \boldsymbol{k}+\boldsymbol{q}) = 0$ and $\langle \{S_q^+, S_q^{+*}\} \rangle = \langle \{S_{-q}^+, S_{-q}^{+*}\} \rangle$. In the following we consider the case of no external magnetic field. In the vicinity of the Curie temperature and in the paramagnetic region, the spin pair correlation is written, from (4·21) and (4·28), as

$$\langle \{S_q{}^+, S_q^{+*}\} \rangle \simeq \frac{2Nk_B T}{(g\mu_B)^2/\chi_\perp + 2J(\boldsymbol{0}, \boldsymbol{q})}, \tag{7·9}$$

where the perpendicular susceptibility $\chi_\perp$ is infinity below the Curie temperature; above the Curie point, it is given by (4·23) in the Weiss approximation. In the cubic lattices, the Fourier component of the exchange interaction $J(\boldsymbol{q})$ is even with respect to each component of the wave vector $\boldsymbol{q}$, and has a form equivalent in the three directions. Then, taking a small wave vector $\boldsymbol{k}$, we obtain

$$(I_k{}^0, I_k^{0*}) = k^2 (b^2/3N) \sum_q J(\boldsymbol{q}) \langle \{S_q{}^+, S_q^{+*}\} \rangle, \tag{7·10}$$

where the relation (3·29) has been used.

In the high temperature limit, insertion of (4·23) into (7·9) leads to

$$\langle \{S_q{}^+, S_q^{+*}\} \rangle = (2N/3) S(S+1) \left[1 + \frac{2S(S+1)}{3k_B T} J(\boldsymbol{q}) + \cdots\right], \tag{7·11}$$

where the term with $J(\boldsymbol{0}) = zJ$ exactly cancels with that arising from the T_c term in the expression for χ_0/χ. The Weiss approximation yields the rigorous result up to the second term in the high temperature expansion (7·11). Inserting (7·11) into (7·10) and using $\Sigma_q [J(\boldsymbol{q})]^2 = NzJ$, we obtain

$$(I_k{}^0, I_k^{0*}) = k^2 b^2 (N/k_B T)(4/27) zS^2(S+1)^2 J^2, \tag{7·12}$$

where the first term of (7·11) does not contribute since $\Sigma_q J(\boldsymbol{q})=0$. The first term of (7·11) leads to

$$(S_k^0, S_k^{0*}) = (N/3k_BT)\, S(S+1). \tag{7·13}$$

It will be shown in Appendix C that a tedious but elementary calculation yields

$$g_{k0}^2 = (8z\xi/3)\, S(S+1)\, J^2, \tag{7·14}$$

where

$$\xi = 1 - \frac{39}{5z^2}\left[1 + \frac{3}{26S(S+1)}\right]. \tag{7·15}$$

The quantity ξ is close to unity, and $\xi=0.87$ in the case of $z=8$ and $S=1$. Thus, combining (7·12), (7·13) and (7·14) with (7·7), we arrive at

$$\gamma_k^0 = k^2 \Lambda, \tag{7·16}$$

where

$$\Lambda = (J/\hbar)\, b^2 \sqrt{(\pi z/54\xi)\, S(S+1)}, \quad T \gg T_c, \tag{7·17}$$

where we have inserted the Planck constant $\hbar$ properly. Thus the temporal development of an inhomogeneous magnetization follows the diffusion equation; inserting (7·16) into (2·50),

$$\frac{d}{dt} M_k^0 = -k^2 \Lambda M_k^0,$$

or, in the spatial representation,

$$\frac{d}{dt} M^0(\boldsymbol{r}) = \Lambda \nabla^2 M^0(\boldsymbol{r}). \tag{7·18}$$

The correlation time of the irregular fluctuation of the torque $I_k^0(t)$ is given by $\tau_c = \sqrt{\pi}/2g_0 \approx \hbar/J$. This may be regarded as being of the order of magnitude of the time in which a localized spin moves to a neighboring lattice site by the flip-up-and-down motion due to the exchange interaction J. Then the diffusion constant is estimated to be

$$\Lambda = (\Delta x)^2/\Delta t \approx b^2/\tau_c \approx (J/\hbar)\, b^2, \tag{7·19}$$

which is consistent with (7·17).

De Gennes[12] treated the same problem by employing the moment method and assuming a truncated Lorentzian distribution for the spectrum of the time correlation of the spin operator. And he calculated the diffusion constant for the simple cubic lattice, obtaining the value which is, in the case of $S=1$, about 0.70 times as large as ours.

Next we consider the temperature dependence of the diffusion constant in the vicinity of the Curie point. As has been pointed out by van Hove,[3] there is a large spontaneous fluctuation of the magnetization near the Curie point.

Due to this critical fluctuation, there appears an anomaly in the dynamical behavior of the ferromagnetic systems. Van Hove and de Gennes[5] discussed this problem by phenomenological treatments, showing that the diffusion constant vanishes at the Curie point due to the critical fluctuation. The longitudinal susceptibility (S_k^0, S_k^{0*}) is determined from (3·9) above the Curie point, and from (4·29), (4·32) and (4·11) below the Curie point. Thus use of (4·17), (4·18) and (4·33) leads, for a small wave number k, to

$$\frac{1}{(S_k^0, S_k^{0*})} = \frac{3k_BTr_1^2}{NS(S+1)}(\kappa_1^2+k^2), \tag{7·20}$$

$$= \frac{3k_BT}{NS(S+1)}\frac{\chi_0}{\chi_{\parallel}} \quad \text{as} \quad k \to 0, \tag{7·21}$$

where the parallel susceptibility $\chi_{\parallel}$ is given by (4·23) and (4·31) in the Weiss approximation. Therefore the correlation of the spin operator S_k^0, (S_k^0, S_k^{0*}), has a singularity at the Curie point, (7·21) vanishing at this point. The correlation of the torque $I_k^0 = \dot{S}_k^0$, $(\dot{S}_k^0, \dot{S}_k^{0*})$, will, however, be shown to have no singularity, which corresponds to the fact that it expresses a microscopic motion and is not affected seriously by the change of the long range order. The correlation time $\tau_c = \sqrt{\pi}/2g_0$ of the torque is determined by the correlation of the higher derivative of the spin operator, $(\ddot{S}_k^0, \ddot{S}_k^{0*})$, and may, therefore, be regarded as having no singularity either. Thus, it can be seen from (7·7) that the damping constant γ_k^0 vanishes at the Curie point as a result of the singularity of the susceptibility $\chi_{\parallel}/\chi_0$. It should be noted, however, that there is an additional nonvanishing term arising from the k^4 term. From (7·7), (7·10) and (7·20), we write as

$$\gamma_k^0 = k^2\Lambda + k^4\Psi + \cdots. \tag{7·22}$$

Then the diffusion constant Λ vanishes at the Curie point as discussed above, whereas Ψ remains finite at that point.

To see this situation in detail, we calculate (7·7). Insertion of (7·9) into (7·10) leads to

$$(I_k^0, I_k^{0*}) = k^2(Nb^2k_BT/3)[-1+(1+\delta)I_\delta], \tag{7·23}$$

where

$$I_\delta = \frac{1}{N}\sum_q \frac{1}{(1+\delta)-[J(\boldsymbol{q})/zJ]}, \tag{7·24}$$

$$\delta = (1/2zJ)(g\mu_B)^2/\chi_\perp. \tag{7·25}$$

For the simple cubic lattice, (7·24) takes the form

$$I_\delta = \frac{1}{(2\pi)^3}\iiint_{-\pi}^{\pi} dx\,dy\,dz \frac{1}{(1+\delta)-(\cos x+\cos y+\cos z)/3}. \tag{7·26}$$

At the Curie point, $\delta=0$ so that[28)]

$$I_\delta=\frac{3}{(2\pi)^3}\iiint_{-\pi}^{\pi}\frac{dx\,dy\,dz}{3-(\cos x+\cos y+\cos z)},$$

$$=3\times 0.50546\simeq 1.52. \tag{7·27}$$

For the body centered cubic lattice, (7·24) leads to

$$I_\delta\doteqdot\frac{1}{(2\pi)^3}\iiint_{-\pi}^{\pi}\frac{dx\,dy\,dz}{(1+\delta)-\cos x\cos y\cos z}, \tag{7·28}$$

$$=1.39320, \quad \text{as} \quad \delta=0. \tag{7·29}$$

For the face centered cubic lattice, we obtain $I_0\doteqdot 3\times 0.44822$. Thus it turns out that the correlation of the torques acting on the respective spins has a definite value at the Curie point, which is in contrast with the correlation of the spin operator. Inserting (7·21) and (7·23) into (7·7), we thus arrive at

$$\Lambda=\frac{b^2(k_BT)^2}{S(S+1)}\frac{\chi_0}{\chi_\parallel}[(1+\delta)I_\delta-1]\frac{\sqrt{\pi}}{2g_{c0}}, \tag{7·30}$$

which is valid in the vicinity of the Curie point and in the paramagnetic region. The assumption made in the derivation of (7·30) is the use of the Bogolyubov-Tjablikov approximation (3·17) and the Gaussian decay (7·2).

As has been mentioned in the above, the correlation time of the torque may be regarded as being nearly constant in the temperature region of interest. Thus, using the classical value for g_{00} given by (7·14), we obtain the following expression for the spin diffusion constant in the vicinity of the Curie point:

$$\Lambda=\frac{T\chi_0}{\chi_\parallel}\frac{b^2k_B{}^2T_c(I_0-1)}{2\hbar J}\sqrt{\frac{3\pi}{8z\xi S^3(S+1)^3}}, \tag{7·31}$$

where

$$\frac{T\chi_0}{\chi_\parallel}=\begin{cases}(T-T_c), & T>T_c,\\ 2(T_c-T), & T<T_c,\end{cases} \tag{7·32}$$

in the Weiss approximation and we have inserted the Planck constant $\hbar$ properly. It should be noted here that (7·31) is valid also below the Curie point. Use of the Weiss approximation for T_c, (4·23), reduces this to the following form:

$$\Lambda\simeq(\chi_0/\chi_\parallel)\,z\,(I_0-1)\,\Lambda_\infty, \tag{7·33}$$

where Λ_∞ denotes the classical expression (7·17).

The k^4 term of the longitudinal damping constant is obtained from (7·20) and (7·23), and turns out to be, at the Curie point,

$$\Psi=a^4(k_BT_c/\hbar)(I_0-1)\sqrt{3\pi/2z\xi S(S+1)}, \tag{7·34}$$

where (4·22) has been used. Thus it follows that the diffusion constant Λ vanishes at the Curie point, being proportional to the temperature distance from T_c in the vicinity of that point, while the k^4 term Ψ remains finite, having the value given by (7·34).

We compare the above results with de Gennes' theory[5),12)] and Ericson-Jacrot's experiment on the spin diffusion constant of iron.[11)] De Gennes looks upon a ferromagnetic spin system as being a fluid and introduces a spin current obeying the equation of continuity. Using a phenomenological consideration of the spin flow, he obtained the following expression for the spin diffusion constant:

$$\Lambda = (\chi_0/\chi_{\parallel})\, D, \tag{7·35}$$

where D was assumed to be nearly constant and was determined, by taking the classical limit, to be

$$D = 0.19\,(J/\hbar)\, b^2 \sqrt{zS(S+1)} = 0.79\sqrt{\xi}\,\Lambda_\infty. \tag{7·36}$$

Equation (7·35) is justified by (7·30), and the explicit expression for D is easily obtained. It is not easy, however, to see whether D is nearly constant or not. If we apply this assumption to (7·33), then we get $z(I_0-1)=1$. Apart from a numerical factor, this is consistent with (7·27) and (7·29) which satisfy $I_0-1 \doteqdot 3.1/z$.

Recently Ericson and Jacrot observed the critical magnetic scattering of neutrons by iron above the Curie point, and determined the spin diffusion constant, obtaining

$$\Lambda_{\text{obs}} = 1.8\times10^{-5}\,(T-T_c)\ \text{cm}^2/\text{sec}. \tag{7·37}$$

Equation (7·31) leads, for iron, to

$$\Lambda = 1.3\times10^{-5}\left(\frac{T\chi_0}{\chi_{\parallel}}\right)\ \text{cm}^2/\text{sec}, \tag{7·38}$$

where we have used the following values for the constants involved:

$$z=8,\quad S=1,\quad a=2.86\,\text{Å},$$
$$T_c=1043°\text{K},\quad J=200\,k_B. \tag{7·39}$$

If we employ the Weiss approximation (7·33), we obtain the numerical factor 2.6 instead of 1.3 in (7·38). Thus we find an excellent agreement between our theory and the experiment.

We shall estimate the magnitude of the k^4 term in (7·22). Equation (7·34) leads, for iron at the Curie point, to

$$k^4\Psi \simeq 3\times10^{14}\,(ak)^4\ \text{sec.}^{-1} \tag{7·40}$$

On the other hand, (7·31) yields $k^2\Lambda \simeq (ak)^2\times10^{12}$ for $T=T_c+50°$, which leads to

$$k^4\Psi/k^2\Lambda \simeq 3\times10^2\,(ak)^2. \tag{7·41}$$

Therefore, in order that the k^4 term can be neglected so that the diffusion equation is valid for the spin damping γ_k^0 for iron around the temperature $T=T_c+50°$, the wave number k should satisfy

$$k^{-1} \gg 17a. \tag{7·42}$$

Recently Riste[29] observed the spin wave damping in Fe_3O_4 and showed the interesting behavior that it goes up as the temperature rises and reaches a value of the order of magnitude of $10^{12}\,\text{sec}^{-1}$ at the Curie point. At the Curie point, the spin wave damping becomes identical with γ_k^0, being given by $k^4\Psi$. Therefore it is interesting to note that his value is of the same order of magnitude as (7·40) with $k^{-1}\sim 4a$. We will investigate this problem in a separate paper.

§ 8. Summary and some remarks

In our formulation of the quantum-statistical theory of dynamical behavior of ferromagnetic spins, our particular intention was to establish the macroscopic equation of motion for an inhomogeneous magnetization and to formulate the frequency spectrum and the damping constant for the collective motion of ferromagnetic spins. This was done with the aid of the statistical mechanics of irreversible processes. A simple reduction of the general expressions for those quantities, (2·22) and (2·49), in the low temperature limit led to the exact results for the spin wave frequency and damping equivalent to Dyson's theory of spin wave interactions. The general expression for the relaxation function $\chi_k(t)$ was also used to determine the asymptotic behavior of the pair correlation of spins and to obtain an exact expression for the neutron scattering cross section.

It was especially delightful that the present theory made it possible to obtain the longitudinal damping constant which is out of the usual kinetic treatment. Employing the Heisenberg model of ferromagnetic spins, we calculated the longitudinal damping constant in the low temperature limit, in the vicinity of the Curie point, and in the paramagnetic region, and showed that the motion of the longitudinal component of an inhomogeneous magnetization obeys the diffusion equation. The diffusion constant thus obtained was investigated carefully in the vicinity of the Curie point and was shown to vanish at this point.

The longitudinal damping in the low temperature limit was shown, in particular, to have a variety of k dependences, depending on the relative magnitudes of the spin wave energy Dk^2, the effective magnetic field (due to the external field, the anisotropy energy and the dipolar interaction), and the temperature. This was critically related to the k dependence of the longitudinal susceptibility. Equations (4·42), (4·43), (6·28), (6·31), (6·32), and (6·29) determine all the quantities involved in the longitudinal component of the magnetic scattering cross section of neutrons (4·50) in those various cases. It should be very interesting to make an experimental investigation on this problem.

The dynamical behavior of ferromagnetic spins is a typical example of the collective motion of the macroscopic body. Thus we hope that the present method provides us with a powerful means to deal with dynamical behaviors of collective motions such as antiferromagnetic resonance absorption and transport processes in liquid helium II. The essential point underlying the present method is the fact that the dynamical variables describing the collective motion are the approximate constants of motion. Thus, for instance, in the collective description of antiferromagnetic substances, the sublattice magnetizations $\boldsymbol{M}^A$ and $\boldsymbol{M}^B$ serve to provide us with such dynamical variables. Then the canonical ensemble for the collective description of non-equilibrium states takes the form

$$\rho_t = Z^{-1} \exp\{-\beta[H - \sum_q (\boldsymbol{h}_q{}^A \cdot \boldsymbol{M}_q^{A*} + \boldsymbol{h}_q{}^B \cdot \boldsymbol{M}_q^{B*})]\},$$

where $\boldsymbol{M}_q^A$ and $\boldsymbol{M}_q^B$ are the Fourier components of the non-uniform sublattice magnetizations, and $\boldsymbol{h}_q^A$ and $\boldsymbol{h}_q^B$ represent thermodynamic magnetic fields whose concept was introduced and discussed in § 2. Thus we have extended the present theory to the antiferromagnetic case, which will be discussed in a subsequent paper.

Acknowledgments

The authors wish to express their sincere gratitude to Professor R. Kubo and to Professor T. Matsubara for their enlightening discussions and continued encouragement. It is also a pleasure to thank Dr. P. G. de Gennes for his stimulating discussions. This work was carried out as one of the projects on "Dynamical Problems in Statistical Physics" supported by the Research Institute for Fundamental Physics, Kyoto University. This study was partially financed by the Scientific Research Fund of the Ministry of Education.

Appendix A

Deviation Density Matrix $\rho'(t)$

Here we shall outline the determination of the deviation density matrix $\rho'(t)$, (2·25), by making use of the statistical-mechanical theory of transport in fluids developed by one of the authors.[14] Most of the physical ideas underlying the theory have been described in the text and used to obtain the explicit expression (2·48) for the generating function.

For simplicity, we consider the exchange interaction between spins, neglecting the dipolar interaction. The extension to a general case can be made in a straightforward manner.

By using (2·21), (2·46), and (2·26), (2·6) is written as

$$\frac{d}{dt} M_k{}^\alpha(t) = i(\alpha\omega_0 + \omega_k{}^\alpha) M_k{}^\alpha(t) - g\mu_B \operatorname{Tr} \rho'(t) I_k{}^\alpha. \tag{A·1}$$

As has been emphasized in the previous paper,[14] the time rate of macroscopic conservative quantities, $\dot{F}$, consists of two parts: $\dot{F}=Q+I$, such that Q is an observable satisfying the characteristic relation of macroscopic quantities, $\mathrm{Tr}\,\rho'(t)Q=0$, and I is an operator describing microscopic processes and satisfying $\mathrm{Tr}\,\rho_t I=0$. In the present case $Q=i\omega_k^\alpha S_k^\alpha$ and $I=I_k^\alpha$. Thus the fluctuating part of the torque I_k^α represents the microscopic process which gives rise to the damping of the magnetization density.

As was mentioned right after (2·28), the temporal development of the system is, except the initial transient period of the order of magnitude of τ_0, independent of the way of the initial preparation of the system as far as the initial values of the macroscopic state variables are fixed to be the same. Therefore the density matrix $\rho(t)$ can be determined by considering an auxiliary system which has started from ρ_t at time t. Namely, considering time intervals, τ, satisfying the relation $\tau_0 \ll \tau \ll \tau_r$, where τ_r is the time in which the macroscopic state changes only by an appreciable amount, we obtain

$$\rho(t) \doteqdot \exp(-i\tau H)\,\rho_t \exp(i\tau H), \tag{A·2}$$

$$=\rho\left[1-g\mu_B\sum_\alpha\sum_q h_q^\alpha\int_0^\beta d\lambda\, e^{\lambda H}(S_q^{\alpha*}(-\tau)-\langle S_q^{\alpha*}\rangle)\,e^{-\lambda H}\right]. \tag{A·3}$$

Use of (2·37), (2·39), and (2·46) leads to

$$S_q^\alpha(t)=e^{it\bar{\omega}_q^\alpha}\left[S_q^\alpha+\int_0^t ds\, e^{-is\bar{\omega}_q^\alpha} I_q^\alpha(s)\right], \tag{A·4}$$

which yields

$$S_q^{\alpha*}(-\tau)\simeq S_q^{\alpha*}-\int_0^\tau ds\, e^{-is\bar{\omega}_q^\alpha} I_q^{\alpha*}(-s), \tag{A·5}$$

since $\tau\bar{\omega}_q^\alpha \simeq 0$ from the definition of τ. Insertion of this into (A·3) leads to

$$\rho(t)=\rho_t+\rho'(t), \tag{A·6}$$

$$\rho'(t)=g\mu_B\rho\sum_\alpha\sum_q h_q^\alpha\int_0^\tau ds\exp(-is\bar{\omega}_q^\alpha)\int_0^\beta d\lambda\, e^{\lambda H} I_q^{\alpha*}(-s)\,e^{-\lambda H}. \tag{A·7}$$

This provides us with an explicit expression for the deviation density matrix $\rho'(t)$. Inserting this into (A·1) and rewriting the thermodynamic magnetic field $h_q^\alpha(t)$ in terms of the inhomogeneous magnetization with the aid of (2·14) and (2·15), we immediately arrive at the macroscopic equation of motion for the magnetization density, (2·50).

Appendix B

On Dyson's Dynamical Interaction

In this appendix, we shall first derive Dyson's dynamical interaction, starting from the exact equation of motion for the spin operators and introducing the spin wave approximation properly.

The equation of motion for S^-_{-k}, (3·13), is written as

$$\dot{S}^-_{-k}=2iN^{-1}\sum_{q}J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})\,S^0_{-q}S^-_{-k+q}. \tag{B·1}$$

Use of (3·19) yields

$$\dot{S}^-_{-k}=2iSJ(\mathbf{0},\boldsymbol{k})\,S^-_{-k}-2iN^{-1}\sum_{q}\sum_{r}J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})\,a^*_{q+r}a_rS^-_{-k+q}, \tag{B·2}$$

or

$$\left[\frac{d}{dt}-2iSJ(\mathbf{0},\boldsymbol{k})\right]S^-_{-k}=-2iN^{-1}\sum_{q}\sum_{r}J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})\,a^*_{q+r}a_rS^-_{-k+q}. \tag{B·3}$$

The both sides of this equation being a small quantity, we can make the spin wave approximation. Namely,

$$\begin{aligned}\left[\frac{d}{dt}-2iSJ(\mathbf{0},\boldsymbol{k})\right]a_k^*&=-2iN^{-1}\sum_{q}\sum_{r}J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})\,a^*_{q+r}a_ra^*_{k-q},\\&=-2iN^{-1}\sum_{q}\sum_{r}J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})\,a^*_{q+r}a^*_{k-q}a_r,\end{aligned} \tag{B·4}$$

where the identity $\sum_q J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})=0$ has been used. Changing the summation index $\boldsymbol{q}$ to $\boldsymbol{k}-\boldsymbol{q}-\boldsymbol{r}$ in the right-hand side of this expression, adding together, and dividing by two, we obtain

$$\left[\frac{d}{dt}-2iSJ(\mathbf{0},\boldsymbol{k})\right]a_k^*=-\frac{i}{N}\sum_{q}\sum_{r}\left[J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})+J(\boldsymbol{k}-\boldsymbol{q}-\boldsymbol{r},\boldsymbol{q}+\boldsymbol{r})\right]a^*_{q+r}a^*_{k-q}a_r. \tag{B·5}$$

If one writes the right-hand side as $i[H'_{\text{eff}},a_k^*]$, one obtains

$$H'_{eff}=-\frac{1}{2N}\sum_{k}\sum_{q}\sum_{r}\left[J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})+J(\boldsymbol{k}-\boldsymbol{q}-\boldsymbol{r},\boldsymbol{q}+\boldsymbol{r})\right]a^*_{q+r}a^*_{k-q}a_ka_r. \tag{B·6}$$

This is equal to Dyson's dynamical interaction (6·16).

Now, if we apply a random phase approximation to the right-hand side of (B·5), that is,

$$\begin{aligned}a^*_{q+r}a^*_{k-q}a_r&\simeq\langle a^*_{q+r}a_r\rangle a^*_{k-q}+\langle a^*_{k-q}a_r\rangle a^*_{q+r},\\&=n_r(\delta_{q,0}+\delta_{k-q,r})\,a_k^*,\end{aligned} \tag{B·7}$$

Eq. (B·5) becomes

$$\left[\frac{d}{dt}-2iSJ(\mathbf{0},\boldsymbol{k})\right]a_k^*=-2iN^{-1}\sum_{r}\left[J(\mathbf{0},\boldsymbol{k})+J(\boldsymbol{k}-\boldsymbol{r},\boldsymbol{r})\right]n_ra_k^*. \tag{B·8}$$

Defining the spin wave frequency by

$$\frac{d}{dt}a_k^*=i\omega_ka_k^*, \tag{B·9}$$

we thus obtain

$$\omega_k = 2\sigma J(\mathbf{0}, \mathbf{k}) + \frac{2}{N} \sum_q J(\mathbf{q}, \mathbf{k}-\mathbf{q})\, n_q. \tag{B·10}$$

This is equal to the spin wave frequency discussed recently by Brout and Englert.[10)]

Next we shall derive a kinetic equation for N_k from the interaction Hamiltonian (B·6). In parallel with Schlömann's theory of parallel pumping,[17)] suppose that N_k deviates from its equilibrium value, other N's being equal to their equilibrium values which we denote by N^0's. The transition probability that a magnon with the wave vector $\mathbf{k}$ collides with another magnon with the wave vector $\mathbf{r}$ to yield two magnons with the wave vectors $\mathbf{k}-\mathbf{q}$ and $\mathbf{q}+\mathbf{r}$ respectively, is, according to the ordinary time-dependent perturbation theory,

$$\frac{8\pi}{N^2}[J(\mathbf{q}, \mathbf{k}-\mathbf{q}) + J(\mathbf{k}-\mathbf{q}-\mathbf{r}, \mathbf{q}+\mathbf{r})]^2 \delta(\omega_k + \omega_r - \omega_{k-q} - \omega_{q+r})$$
$$\times N_k N_r^0 (N_{k-q}^0 + 1)(N_{q+r}^0 + 1), \tag{B·11}$$

where the matrix element of the interaction Hamiltonian has been multiplied by four, because there are four identical terms in the interaction Hamiltonian resulting from the interchange of $\mathbf{k}$ and $\mathbf{r}$, as well as of $\mathbf{q}+\mathbf{r}$ and $\mathbf{k}-\mathbf{q}$.

In order to obtain the total probability for a magnon with the wave vector $\mathbf{k}$ to collide with any one of the existing thermal magnons, one has to sum this expression over $\mathbf{r}$ and $\mathbf{q}$, and to divide by two not to count the final states twice. The result is

$$w_k^{(1)} = \frac{4\pi}{N^2} \sum_q \sum_r [J(\mathbf{q}, \mathbf{k}-\mathbf{q}) + J(\mathbf{k}-\mathbf{q}-\mathbf{r}, \mathbf{q}+\mathbf{r})]^2 \delta(\omega_k + \omega_r - \omega_{k-q} - \omega_{q+r})$$
$$\times N_k N_r^0 (N_{k-q}^0 + 1)(N_{q+r}^0 + 1). \tag{B·12}$$

The transition probability of the inverse process, in which any two of the thermal magnons collide with each other to produce a magnon with the wave vector $\mathbf{k}$ and another magnon with an arbitrary wave vector, can be obtained in exactly the same manner; namely,

$$w_k^{(2)} = \frac{4\pi}{N^2} \sum_q \sum_r [J(\mathbf{q}, \mathbf{k}-\mathbf{q}) + J(\mathbf{k}-\mathbf{q}-\mathbf{r}, \mathbf{q}+\mathbf{r})]^2 \delta(\omega_k + \omega_r - \omega_{k-q} - \omega_{q+r})$$
$$\times (N_k + 1)(N_r^0 + 1) N_{k-q}^0 N_{q+r}^0. \tag{B·13}$$

The average rate of decrease N_k is evidently the difference of these two probabilities, that is,

$$\frac{d}{dt} N_k = -w_k^{(1)} + w_k^{(2)}. \tag{B·14}$$

If we consider the fact that for $N_k = N_k^0$ there should be no net change of N_k, (B·14) can be brought to the following form:

$$\frac{d}{dt} N_k = -2\gamma_k (N_k - N_k^0), \tag{B·15}$$

with the same expression for γ_k as given by (6·15) of the text.

Appendix C

Evaluation of g_{k0} in the High Temperature Limit

Here we shall outline the calculation of the constant g_{k0} defined by (7·7). This quantity, expressed in terms of the spin operators by using the equations of motion, involves the correlations of six spin operators. Even after reducing with the help of the identity (3·1), four spin operators are involved. Thus the evaluation of this quantity at the temperatures close to or lower than the Curie point with any rigor is a prohibittingly difficult task. Even the calculation in the pair approximation has not been successful. Therefore, we evaluated g_{k0} in the high temperature limit.

According to the result of § 7, the only quantity we have to evaluate is $(\dot{I}_k^0, \dot{I}_k^{0*})$. First, note that at the high temperature limit we have

$$(\dot{I}_k^0, \dot{I}_k^{0*}) = \frac{1}{k_B T} \langle \{\dot{I}_k^0, \dot{I}_k^{0*}\} \rangle. \tag{C·1}$$

Use of the equation of motion yields

$$\begin{aligned}
\langle \{\dot{I}_k^0, \dot{I}_k^{0*}\} \rangle = &\frac{1}{N^2} \sum_q \sum_p J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q}) J(\boldsymbol{p}, -\boldsymbol{k}-\boldsymbol{p}) \\
&\times \frac{4}{N^2} \sum_r \sum_{r'} \Big[J(\boldsymbol{r}, \boldsymbol{q}-\boldsymbol{r}) J(\boldsymbol{r}', \boldsymbol{p}-\boldsymbol{r}') \langle \{\{S_r^0 S_{q-r}^+, S_{-q}^-\}, \{S_{r'}^0 S_{p-r'}^+, S_{-p}^-\}\} \rangle \\
&- J(\boldsymbol{r}, \boldsymbol{q}-\boldsymbol{r}) J(\boldsymbol{r}', -\boldsymbol{p}-\boldsymbol{r}') \langle \{\{S_r^0 S_{q-r}^+, S_{-q}^-\}, \{S_p^+, S_{r'}^0 S_{-p-r'}^-\}\} \rangle \\
&- J(\boldsymbol{r}, -\boldsymbol{q}-\boldsymbol{r}) J(\boldsymbol{r}', \boldsymbol{p}-\boldsymbol{r}') \langle \{\{S_{r'}^0 S_{p-r'}^+, S_{-p}^-\}, \{S_q^+, S_r^0 S_{-q-r}^-\}\} \rangle \\
&+ J(\boldsymbol{r}, -\boldsymbol{q}-\boldsymbol{r}) J(\boldsymbol{r}', -\boldsymbol{p}-\boldsymbol{r}') \langle \{\{S_q^+, S_r^0 S_{-q-r}^-\}, \{S_p^+, S_{r'}^0 S_{-p-r'}^-\}\} \rangle \Big].
\end{aligned} \tag{C·2}$$

At the high temperature limit, the ensemble averages of the product of the spin operators on the right-hand side are replaced by their traces divided by $2S+1$. After a tedious but elementary calculation, (C·2) reduces to the following:

$$\begin{aligned}
&\frac{8}{N} \sum_p \sum_q J(\boldsymbol{q}, \boldsymbol{k}-\boldsymbol{q}) J(\boldsymbol{p}, \boldsymbol{k}-\boldsymbol{p}) \Big[\{\delta_{p,q} \sum_r [J(\boldsymbol{r}, \boldsymbol{q}-\boldsymbol{r})]^2 - J(\boldsymbol{q}-\boldsymbol{p}, \boldsymbol{p}) J(\boldsymbol{p}-\boldsymbol{q}, \boldsymbol{q})\} \\
&\quad \times \langle 0^2 \rangle \langle \{+, -\} \rangle^2 + \frac{1}{N} \sum_r J(\boldsymbol{r}, \boldsymbol{q}-\boldsymbol{r}) J(\boldsymbol{r}, \boldsymbol{p}-\boldsymbol{r}) [\langle 0^2 \rangle \\
&\quad \times \{\langle \{+, -\}^2 \rangle - 3\langle \{+, -\} \rangle^2\} + \langle \{+, -\} \rangle \langle \{0+, 0-\} \rangle]
\end{aligned}$$

$$-\frac{1}{N}\sum_{r} J(\boldsymbol{r},\boldsymbol{q}-\boldsymbol{r})\,J(\boldsymbol{r},\boldsymbol{p}+\boldsymbol{r})\langle 0^2\rangle\Big[\langle\{+,-\}^2\rangle-2\langle\{+,-\}\rangle^2\Big]\Big], \qquad (\mathrm{C}\cdot 3)$$

where we have used the following abbreviations:

$$\langle 0^2\rangle \equiv \frac{\mathrm{Tr}\,(S_m^0)^2}{\mathrm{Tr}\,1}, \quad \langle\{+,-\}^2\rangle \equiv \frac{\mathrm{Tr}\,\{S_m^+, S_m^-\}^2}{\mathrm{Tr}\,1}, \quad \text{and so on.}$$

For the small values of k, we have

$$\frac{1}{N^2}\sum_q\sum_p J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})\,J(\boldsymbol{p},\boldsymbol{k}-\boldsymbol{p})\,J(\boldsymbol{q}-\boldsymbol{p},\boldsymbol{p})\,J(\boldsymbol{p}-\boldsymbol{q},\boldsymbol{q})=0,$$

$$\frac{1}{N^3}\sum_p\sum_q\sum_r J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})\,J(\boldsymbol{p},\boldsymbol{k}-\boldsymbol{p})\,J(\boldsymbol{r},\boldsymbol{q}-\boldsymbol{r})\,J(\boldsymbol{r},\boldsymbol{p}+\boldsymbol{r})=-2J^4(kb)^2,$$

$$\frac{1}{N^3}\sum_p\sum_q\sum_r J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})\,J(\boldsymbol{p},\boldsymbol{k}-\boldsymbol{p})\,J(\boldsymbol{r},\boldsymbol{q}-\boldsymbol{r})\,J(\boldsymbol{r},\boldsymbol{p}-\boldsymbol{r})=2J^4(kb)^2,$$

$$\frac{1}{N^2}\sum_q\sum_r [J(\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q})\,J(\boldsymbol{r},\boldsymbol{q}-\boldsymbol{r})]^2=\frac{2}{3}z^2J^4(kb)^2. \qquad (\mathrm{C}\cdot 4)$$

We thus obtain

$$(\dot{I}_k^0, \dot{I}_k^{0*})=\frac{1}{k_BT}\frac{32N}{9}\left\{\frac{2z^2S^2(S+1)^2}{9}-\frac{26S^2(S+1)^2}{15}-\frac{S(S+1)}{5}\right\}$$
$$\times S(S+1)\,J^4(kb)^2. \qquad (\mathrm{C}\cdot 5)$$

This result together with (7·7) and (7·12) yields the following value for g_{k0}^2:

$$g_{k0}^2=\frac{8}{3}z\xi S(S+1)\,J^2, \qquad (\mathrm{C}\cdot 6)$$

where

$$\xi=1-\frac{39}{5z^2}\left[1+\frac{3}{26S(S+1)}\right].$$

In particular, for $z=6$ and $S=1$, we have

$$g_{k0}^2=24.7\,J^2. \qquad (\mathrm{C}\cdot 7)$$

This value is slightly smaller than that obtained by de Gennes,[12] which is, using his notation,

$$g_{k0}^2=\frac{\langle\omega^4\rangle_k}{2\langle\omega^2\rangle_k}=27\,J^2. \qquad (\mathrm{C}\cdot 8)$$

References

1) H. Palevsky and D. J. Hughes, Phys. Rev. **92** (1953), 202 (L).
2) G. L. Squires, Proc. Phys. Soc. (London) **A 67** (1954), 248.
3) L. van Hove, Phys. Rev. **95** (1954), 249, 1374.

4) R. J. Elliott and W. Marshall, Rev. Mod. Phys. **30** (1958), 75.
5) P. G. de Gennes, Report to the C. E. A. de Saclay, No. 929 (1959).
6) T. Riste, K. Blinowski and J. Janik, J. Phys. Chem. Solids **9** (1959), 153.
T. Riste, J. Phys. Chem Solids **17** (1961), 308.
T. Riste, and A. Wanic, J. Phys. Chem. Solids **17** (1961), 318.
7) F. Dyson, Phys. Rev. **102** (1956), 1217.
8) N. Bogolyubov and S. Tjablikov̌, DAN. USSR **126** (1959), 53.
9) F. Keffer and R. Loudon, J. Appl. Phys. **32** (1961), 2S.
10) R. Brout and F. Englert, Bull. Am. Phys. Soc. **6** (1961), 55.
11) M. Ericson and S. Jacrot, J. Phys. Chem. Solids **13** (1960), 235.
12) P. G. de Gennes, J. Phys. Chem. Solids **4** (1958), 223.
P. G. de Gennes and J. Villain, J. Phys. Chem. Solids **13** (1960), 10.
13) R. Kubo, *Some Aspects of the Statistical-Mechanical Theory of Irreversible Processes*, Lectures in Theoretical Physics Vol. 1, (Interscience Publishers, Inc., New York, 1959).
14) H. Mori, Phys. Rev. **112** (1958), 1829; **115** (1959), 298.
15) H. Mori and K. Kawasaki, Prog. Theor. Phys. **25** (1961), 723, 866.
16) R. Kubo and K. Tomita, J. Phys. Soc. Japan **9** (1954), 888.
17) E. Schlömann, Phys. Rev. **121** (1961), 1312.
18) J. Kanamori and M. Tachiki, Abstract for the Int. Conf. on Magn. and Cryst. Kyoto (1961).
19) R. Kubo, J. Phys. Soc. Japan **12** (1957), 570.
20) T. Holstein and H. Primakoff, Phys. Rev. **58** (1940), 177.
21) K. Kawasaki and H. Mori, Prog. Theor. Phys. **25** (1961), 1043.
22) R. G. Sachs, *Nuclear Theory* (Addison-Wesley Publishing Co., Cambridge, Mass., 1953).
23) A. I. Akhiezer, V. G. Bar'yakhtar and I. M. Kaganov, Usp. Fiz. Nauk SSSR, **71** (1960), 533; Soviet Physics Uspekhi **3** (1961), 567.
24) I. A. Akhiezer, V. G. Bar'yakhtar and S. V. Peletminskii, J. Exptl. Theoret. Phys. (USSR) **40** (1961), 365; Soviet Physics JETP **13** (1961), 249.
25) T. Oguchi, Phys. Rev. **117** (1960), 117.
26) T. Mannari, Prog. Theor. Phys. **19** (1958), 201.
T. Morita, Prog. Theor. Phys. **20** (1958), 614 and 728.
27) L. van Hove, Physica **21** (1955), 517; **23** (1957), 441.
W. Kohn and J. M. Luttinger, Phys. Rev. **108** (1957), 590.
J. M. Luttinger and W. Kohn, Phys. Rev. **109** (1958), 1892.
H. Mori, Phys. Rev. **111** (1958), 694.
G. V. Chester and A. Thellung, Proc. Phys. Soc. **73** (1959), 745.
28) G. N. Watson, Quart. J. Math. **10** (1939), 266.
29) T. Riste, Abstract for the Int. Conf. on Magn. and Cryst. Kyoto (1961).