JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN VOL. 17, SUPPLEMENT B-I, 1962
PROCEEDINGS OF INTERNATIONAL CONFERENCE ON MAGNETISM AND CRYSTALLOGRAPHY, 1961, VOL. I

Theory of Spin Diffusion

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The dynamical behavior of the 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 an explicit expression for the frequency spectrum and damping constant. With the use of the general expression thus obtained, the following problems are discussed on the basis of the Heisenberg model; (1) the pair correlation of spins, (2) the frequency spectrum and damping constant of the spin waves, (3) the damping of the z component of the magnetization.

In the spin wave region, a straightforward reduction of our expression leads to the spin wave frequency and damping equivalent to Dyson's theory of spin waves. It is shown that, in the vicinity of the Curie point and in the paramagnetic region, the motion of the z component of the magnetization obeys the diffusion equation. The diffusion constant thus obtained vanishes at the Curie point and is in good agreement with the experimental values observed by Ericson and Jacrot for iron above the Curie point.

As reviewed by Dr. Lowde and reported by Dr. Riste and by Dr. Jacrot in the Joint session, the neutron scattering experiment reveals interesting dynamical behaviors of the ferromagnetic spins. For instance, it turned out that the spin diffusion constant of iron vanishes at the Curie point.[1] Recently Dr. de Gennes extended van Hove's theory[2] to give an exposition of this phenomenon.[3] His treatment, however, is still phenomenological and a microscopic basis of his theory remains to be elucidated.

On the other hand, the theory of spin waves, including the spin wave damping, was established by Dyson.[4] However, it is not clear how to obtain the damping constant of the longitudinal component, even if we know the dynamical interaction between spin waves.

In this talk, therefore, I would like to discuss the damping of the longitudinal component of an inhomogeneous magnetization, and also some related topics, in the spin wave region, in the vicinity of the Curie point and in the paramagnetic region, making use of the statistical mechanics of irreversible processes.[5,6]

The dynamical behavior of the ferromagnetic spins is described by the dynamical susceptibility given by[5]

$$\chi_k^{\alpha}(t)=(g\mu_B)^2\,(S_k^{\alpha}(t),\,S_k^{\alpha *})\,,\ (\alpha=0,\pm),\tag{1}$$

where S_k^{α} is the Fourier component of the α spin, $\alpha=0$ denoting the z component of the spin operator. The bracket notation above is defined by

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

$$\langle G\rangle=\mathrm{Tr}\,G\,e^{-\beta H}/\mathrm{Tr}\,e^{-\beta H},\tag{3}$$

where H is the total Hamiltonian which consists of the Zeeman energy (with the Zeeman frequency ω_0) for a constant field in the z direction and the exchange interaction H_0 between spins.

Now we define the generating function Ξ by taking the dynamical susceptibility divided by its initial value:

$$\Xi_k^{\alpha}(t)=(S_k^{\alpha}(t),\,S_k^{\alpha *})/(S_k^{\alpha},\,S_k^{\alpha *}),\tag{4}$$

$$=1+it(\alpha\omega_0+\omega_k^{\alpha})+O(t^2).\tag{5}$$

Equation (5) is the expansion in powers of t for a small time interval t, and the frequency ω_k^{α} is given by

$$\omega_k^{\alpha}=(\dot{S}_k^{\alpha},\,S_k^{\alpha *})/i(S_k^{\alpha},\,S_k^{\alpha *}),\tag{6}$$

where

$$\dot{S}_k^{\alpha}=i\,[H_0,\,S_k^{\alpha}].\tag{7}$$

The Fourier components of the magnetization density form a set of macroscopic state variables complete enough to describe macroscopic magnetic disturbances. This situation leads to the following:[6]

$$\Xi_k^{\alpha}(t)=\exp\,[it\,(\alpha\omega_0+\omega_k^{\alpha})-t\,\Gamma_k^{\alpha}],\tag{8}$$

where

$$\Gamma_k{}^\alpha=\int_0^\infty dt\exp(-it(\alpha\omega_0+\omega_k{}^\alpha))\times(I_k{}^\alpha(t),I_k{}^{\alpha*})/(S_k{}^\alpha,S_k{}^{\alpha*}) \quad (9)$$

where

$$I_k{}^\alpha=\dot{S}_k{}^\alpha-i\omega_k{}^\alpha S_k{}^\alpha. \quad (10)$$

Equation (8) is the asymptotic equation which is valid for times t larger than the correlation time τ_c of the fluctuating torque $I_k{}^\alpha$. The correlation time τ_c is of the order of magnitude of $\hbar/J$, J being the magnitude of the exchange interaction. The quantity $\omega_k{}^\alpha$ given by (6) expresses the frequency spectrum of the collective oscillation of the spins. The imaginary part of $\Gamma_k{}^\alpha$ yields the shift of the frequency and becomes appreciable in the vicinity of the Curie point. The real part of $\Gamma_k{}^\alpha$ leads to the damping of the collective oscillation.

Denoting by $J(q)$ the Fourier component of the Heisenberg exchange interaction and introducing the notation,

$$J(q,q')=J(q)-J(q'),\ J(q)=J\Sigma_f\exp(iq\cdot r_f), \quad (11)$$

we can show easily that (6) leads, with a simple approximation, to

$$\omega_k{}^\pm=\mp\omega_k, \quad (12)$$

$$\omega_k\cong2\sigma J(0,k),\ \sigma=\langle S_0{}^0\rangle/N, \quad (13)$$

σ being the equilibrium value of the z spin component per ion, and, with a higher approximation in the spin wave region, to

$$\omega_k\cong2SJ(0,k)+(2/N)\sum_q n_q[J(q,k+q)-J(0,k)], \quad (14)$$

where

$$n_q=1/\exp[2\beta SJ(0,q)]-1, \quad (15)$$

and S is the magnitude of the spin. Equation (13) agrees with Bogolyubov and Tjablikov's,[7] and (14) agrees with the one obtained by Keffer and Loudon,[8] and shown to reproduce Dyson's theory of ferromagnetism.[4]

The present expression for the frequency spectrum is valid in the vicinity of the Curie point, and also serves to determine the static correlation of spins even above the Curie point. A simple transformation of (6) leads to

$$\chi_k{}^\pm/(g\mu_B)^2\equiv(S_k{}^\pm,S_k{}^{\pm*})=\frac{2N\sigma}{-\omega_0+\omega_k}. \quad (16)$$

Using the fluctuation-dissipation theorem[5] and neglecting the damping effect,

$$\langle\{S_k{}^\pm,S_k{}^{\pm*}\}\rangle\doteq N\sigma\coth[\beta(-\omega_0+\omega_k)/2], \quad (17)$$

where $\{A,B\}=(AB+BA)/2$. Above the Curie point, (17) leads to

$$\langle\{S_k{}^0,S_k{}^{0*}\}\rangle\doteq\frac{Nk_BT}{\dfrac{(g\mu_B)^2}{\chi}+\left(\dfrac{\omega_k}{\sigma}\right)_{\omega_0=0}},\ (T>T_c), \quad (18)$$

χ being the susceptibility. Since $(\omega_k/\sigma)\propto k^2$ as $k\to0$, (18) leads to a general form for the asymptotic behavior of the spin pair correlation, determining van Hove's parameters[2] as

$$r_1{}^2=\frac{S(S+1)}{3k_BT}\lim_{q\to0}\frac{\omega_q}{\sigma q^2},\ (\kappa_1 r_1)^2=\chi_0/\chi, \quad (19)$$

where

$$\chi_0\equiv(g\mu_B)^2S(S+1)/3k_BT. \quad (20)$$

Below the Curie point, (17) leads to

$$\langle\{S_k{}^\pm,S_k{}^{\pm*}\}\rangle\doteq N\sigma\coth[\beta\omega_k/2], \quad (21)$$

$$\to\frac{NS(S+1)}{3r_1{}^2}\frac{1}{k^2}\ \text{as}\ k\to0. \quad (22)$$

It should be noted that the above equations from (16) to (22) are independent of the specialized models of the ferromagnetic spins.

Next we calculate the damping constants from (9). In the spin wave region, the time integration can be easily carried out to yield

$$\gamma_k{}^+\equiv Re(\Gamma_k{}^+)=\left(\frac{2\pi}{N^2}\right)\sum_{q,r}[J(q,k-q)+J(k-q-r,q+r)]^2\,\delta(\omega_k+\omega_r-\omega_{r+q}-\omega_{k-q})\times\{n_r(n_{r+q}+1)(n_{k-q}+1)-(n_r+1)n_{r+q}n_{k-q}\}, \quad (23)$$

$$\gamma_k{}^0=\frac{4\pi S^2(g\mu_B)^2}{k_BT\chi_k{}^0}\sum_q[J(q,k-q)]^2\,\delta(\omega_q-\omega_{k-q})\times n_q(n_{k-q}+1), \quad (24)$$

$$=\frac{V}{16\pi N}\frac{N(g\mu_B)^2}{\chi_k{}^0}\frac{k^3}{\exp[(\beta/4)Dk^2]-1} \quad (25)$$

where $\omega_q=Dq^2$. The spin wave damping $\gamma_k{}^+$ is just one half of the transition rate of the number of spin waves k obtained from Dyson's dynamical interaction by using the kinetic treatment with Schlömann's assumption.[9] As you can see, the damping of the z spin $\gamma_k{}^0$ has a quite different feature from the spin wave damping (23), and is out of the usual kinetic theoretical understanding.

In the vicinity of the Curie point and in the paramagnetic region, it is very difficult to carry out the integration of the time correlation exactly. So we assume the Gaussian decay,

$$(I_k{}^0(t),I_k{}^{0*})\propto\exp(-t^2g_k{}^2), \quad (26)$$

in parallel to the treatment of the exchange narrowing in paramagnetic resonance absorption. Then the z damping constant takes the form,

$$\gamma_k{}^0=\frac{(I_k{}^0, I_k{}^{0*})}{(S_k{}^0, S_k{}^{0*})}\frac{\sqrt{\pi}}{2g_k},\ g_k=\sqrt{\frac{(\dot{I}_k{}^0, \dot{I}_k{}^{0*})}{2(I_k{}^0, I_k{}^{0*})}}.\quad (27)$$

The bracket SS means the longitudinal static susceptibility and has a singularity at the Curie point:

$$(S_k{}^0, S_k{}^{0*})=\frac{NS(S+1)}{3k_BT}\frac{\chi_{/\!/}}{\chi_0}$$

$$\propto\begin{cases}\dfrac{1}{(T-T_c)},\ T>T_c,\\[2ex] \dfrac{1}{2(T_c-T)},\ T<T_c.\end{cases}\quad (28)$$

The bracket II means the static correlation of the torque, and has no singularity at the Curie point, being given by

$$(I_k{}^0, I_k{}^{0*})=(k^2/3)Nb^2k_BT_c(3.1/z)\ \text{at}\ T=T_c.\quad (29)$$

This implies that the correlation of the torque expresses a microscopic motion and is not affected seriously by the change of the long range order. The quantity g means the inverse of the correlation time of the torque and may be regarded to have no singularity either, being, in the classical limit, given by

$$g_k=(J/\hbar)\sqrt{8z\xi S(S+1)/3}\ ,\quad (30)$$

$$\xi\equiv1-(39/5z^2)\,[1+3/26S(S+1)]\ .\quad (31)$$

Therefore, the damping constant vanishes at the Curie point as a result of the singularity of the susceptibility. This agrees with the phenomenological investigation by van Hove and de Gennes.

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

$$\Lambda_{\text{obs}}=1.8\times10^{-5}(T-T_c)\ \text{cm}^2/\text{sec}.\quad (32)$$

Inserting (28), (29) and (30) into (27), we obtain, for iron above the Curie point,

$$\Lambda_{\text{theor}}=1.3\times10^{-5}\,(T-T_c)\ \text{cm}^2/\text{sec.}\ ,\quad (33)$$

where we have assumed $J=200\,k_B$ for the exchange interaction. This is in good agreement with (32).

The dynamical behavior of the ferromagnetic spins is a typical example of the macroscopic body. I hope the present method provides us with a powerful means to treat the dynamical behavior of various collective modes, such as excitations in liquid He II.

References

1 M. Ericson and B. Jacrot: J. Phys. Chem. Solids **13** (1960) 235.

2 L. van Hove: Phys. Rev. **95** (1954) 1374.

3 P. G. de Gennes: Report to the C. E. A. de Saclay No. 929 (1959).

4 F. Dyson: Phys. Rev. **102** (1956) 1217.

5 R. Kubo: Some Aspects of the Statistical-Mechanical Theory of Irreversible Processes, Lectures in Theoretical Physics Vol. 1, (Interscience Publishers, Inc., New York, 1959).

6 H. Mori: Phys. Rev. **112** (1958) 1829; **115** (1959) 298.

7 N. Bogolyubov and S. Tjablikov: D.A.N. USSR **126** (1959) 53.

8 F. Keffer and R. Loudon: J. Appl, Phys. **32** (1961) 2S.

9 E. Schlömann: Phys. Rev. **121** (1961) 1312.

DISCUSSION

R. Kubo: Is it necessary to take the shift term into account in your calculation in order to get Dyson's result for the ferromagnet spin wave system? Or, the first approximation of the frequency is enough?

H. Mori: The shift of the spin wave frequency coming from the imaginary part of $\Gamma_k{}^\alpha$ is not necessary to obtain Dyson's result, but becomes appreciable near T_c.