Progress of Theoretical Physics, Vol. 28, No. 6, December 1962

Antiferromagnetic Resonance Absorption

Hazime MORI and Kyozi KAWASAKI*)

Research Institute for Fundamental Physics, Kyoto University, Kyoto

(Received June 21, 1962)

The line widths of antiferromagnetic resonance absorption in the vicinity of the Neél point are investigated by the use of a method of relaxation function similar to that employed by the present authors in studying the dynamical behavior of ferromagnetic spins.

Above the Neél point, the line width of the high frequency mode is shown to increase rapidly when the temperature approaches the Neél point, being proportional to $(T-T_N)^{-3/4}$ in the vicinity of this point. This anomalous increase is due to the critical fluctuation of spins and is in agreement with the observation on MnF_2. Below the Neél point, the situation is more complicated and the effects of the anomalous fluctuation upon the line widths are discussed.

§ 1. Introduction

In a previous paper,[1] hereafter referred to as I, we have presented a theory for dealing with the dynamical behavior of ferromagnetic spins and calculated the damping of the longitudinal spin component above and below the Curie point, The purpose of this paper is to extend this theory to systems with more than one sublattice, in order to investigate the problem of antiferromagnetic resonance absorption in the vicinity of the Neél point.[2),3)]

The interesting aspect of this problem is that in the vicinity of the Neél point, a certain type of motion of spins slows down due to the enormous thermodynamic fluctuations associated with this point, which reveals itself through a rapid increase of the line width of the high frequency resonance mode at the Neél point. The same types of phenomena are the vanishing of the spin diffusion constant in ferromagnetics at the Curie point,[1] and the anomalous increase of the NMR line width[4] near the transition points of ferro-, antiferro-, and ferrimagnetics.

In §§ 2 and 3, we shall discuss the collective motion of antiferromagnetic spins and its damping according to a theory of collective motion at finite temperatures described elsewhere,[5] introducing the normalized relaxation matrix. The frequency matrix, which determines the frequency spectrum of the collective motion, is defined in terms of the first moment of this matrix. In § 3, the frequency matrix is diagonalized by introducing a transformation matrix, in

*) Present address: Department of Applied Physics, Faculty of Engineering, Nagoya University, Nagoya, Japan.

order to define the normal modes of the collective motion as linear combinations of the sublattice magnetization operators. The transformed normalized relaxation matrix describes the temporal development of the normal modes. The expressions for the damping constants in terms of the correlation of torques are also obtained here.

Section 4 deals with the AFMR. The present approach is a refinement of the previous formulation of the same problem reported elsewhere.[6] As was shown there, in the Weiss approximation, the resonance frequencies thus obtained agree with Nagamiya and Kittel's formula.[7] Assuming the Gaussian decay for the time dependence of the correlation function of torques, the damping constants are expressed in terms of the 1st, 2nd, 3rd, and 4th moments of the normal modes. Section 5 is concerned with the line width in the vicinity of and above the Neél point. Due to an anomalous increase of the second moment near the Neél point, the width of the high frequency mode is shown to increase rapidly as this point is approached, in agreement with the line width observed by Hutchison and Stout.[2] In § 6, we discuss the line width below the Neél temperature, and it is pointed out that the anomalous thermodynamic fluctuation does not appear in the low frequency mode. In § 7, we attempt to give a simple reason for the effects of the anomalous thermodynamic fluctuation upon the resonance line widths.

§ 2. Collective motion of antiferro- and ferrimagnetic spins

The treatment of the collective motion of ferromagnetic spins, described in I, can be extended to the cases in which the system is divided into more than one sublattices. Consider the system of unit volume containing N magnetic ions, and introduce the Fourier transform of the magnetization belonging to each sublattice as

$$[M_q^\alpha]_j = \sum_f \exp(i\boldsymbol{q}\cdot\boldsymbol{r}_f)\,[M_f^\alpha]_j, \quad (\alpha = +,-,0), \tag{2·1}$$

where j denotes the j-th sublattice and the summation runs over the lattice points pertaining to the j-th sublattice. It is more convenient to introduce the vector $M_q{}^\alpha$ whose j-th component is $[M_q{}^\alpha]_j$. We assume that the macroscopic state of the system is specified by giving the average value of sublattice magnetizations $M_q{}^\alpha$ with values of q very much smaller than the reciprocal of the lattice constant. In the ferromagnetic case, this assumption was based on the fact that $M_q{}^\alpha$ with a small value of q is an approximate constant of motion since the total magnetization commutes with the exchange interaction Hamiltonian. On the contrary, in the present case, each sublattice magnetization does not commute with the total exchange interaction Hamiltonian, and our assumption needs further justification. It should be remembered, however, that, according to P.W. Anderson's estimate,[8] the time necessary for the switching over of the sublattices is extremely long—of the order of a year. A recent attempt[9] to detect this switching over has not been successful. Thus, by a generalization of the previous prescription, the temporal development of the average sublattice magnetization density $\langle M_q{}^\alpha\rangle(t)$ is described by the normalized relaxation matrix $\Xi_q{}^\alpha(t)$ in the following manner[1),5)]:

$$\langle M_q^\alpha\rangle(t) = \Xi_q^\alpha(t)\cdot\langle M_q^\alpha\rangle(0), \tag{2·2}$$

where

$$\Xi_q^\alpha(t) = (M_q^\alpha(t), M_q^{\alpha *})\cdot(M_q^\alpha, M_q^{\alpha *})^{-1}, \tag{2·3}$$

where we have introduced the notation

$$(A, B) \equiv \int_0^\beta d\lambda\langle\exp(\lambda H)A\exp(-\lambda H)B\rangle, \tag{2·4}$$

the angular brackets denoting the ensemble average. As in I, the frequency matrix is obtained as

$$i\hat{\omega}_q^\alpha = \dot{\Xi}_q^\alpha(0) = (\dot{M}_q^\alpha, M_q^{\alpha *})\cdot(M_q^\alpha, M_q^{\alpha *})^{-1}. \tag{2·5}$$

For later convenience, we derive here a simple relation between conjugate frequency matrices. Indicating the transposed matrix by a superscript T, we have

$$\begin{aligned}(\hat{\omega}_{-q}^{-\alpha})^T &= (1/i)\,(M_{-q}^{-\alpha *}, M_{-q}^{-\alpha})^{-1}\cdot(M_{-q}^{-\alpha *}, \dot{M}_{-q}^{-\alpha}),\\ &= -(M_q^\alpha, M_q^{\alpha *})^{-1}\cdot\hat{\omega}_q^\alpha\cdot(M_q^\alpha, M_q^{\alpha *}).\end{aligned} \tag{2·6}$$

In a similar manner, it follows that $(\hat{\omega}_q{}^\alpha)^* = -(\hat{\omega}_{-q}^{-\alpha})^T$. We note also that, in the simplest approximation, the normalized relaxation matrix (2·3) takes the form

$$\Xi_q^\alpha(t) \approx \exp[it\hat{\omega}_q^\alpha]. \tag{2·7}$$

This equation describes the collective precession of antiferromagnetic spins without damping.

§ 3. Normal modes and their damping

In order to obtain the frequency spectrum of the collective motion, it is necessary to diagonalize the frequency matrix (2·5). Since the collective motions with different α's and $\boldsymbol{q}$'s are disconnected from each other for the systems having the translation invariance and strong exchange interactions, the diagonalization can be performed separately for each value of α and $\boldsymbol{q}$. Denoting the transformation matrix for the diagonalization by U_q^α, and the diagonalized frequency matrix by $\hat{\Omega}_q{}^\alpha$, we have

$$\hat{\Omega}_q^\alpha = U_q^\alpha\cdot\hat{\omega}_q^\alpha\cdot(U_q^\alpha)^{-1}. \tag{3·1}$$

The corresponding normal modes of the collective motion, which we denote by $\mathcal{M}_q{}^\alpha$, then become

$$\mathcal{M}_q^\alpha = U_q^\alpha \cdot M_q^\alpha, \tag{3·2}$$

in terms of which (3·1) is written as

$$i\hat{\Omega}_q^\alpha = (\dot{\mathcal{M}}_q^\alpha, \mathcal{M}_q^{\alpha *}) \cdot (\mathcal{M}_q^\alpha, \mathcal{M}_q^{\alpha *})^{-1}, \tag{3·3}$$

which can be obtained by inserting the factor $(U_q{}^\alpha)^* \cdot [(U_q{}^\alpha)^*]^{-1} = 1$ between the two matrices in (2·5). Since $\hat{\Omega}_{-q}^{-\alpha} = -\hat{\Omega}_q{}^\alpha$ due to the time reversal property, we obtain

$$\hat{\Omega}_q^\alpha = (\mathcal{M}_q^\alpha, \mathcal{M}_q^{\alpha *})^{-1} \cdot \hat{\Omega}_q^\alpha \cdot (\mathcal{M}_q^\alpha, \mathcal{M}_q^{\alpha *}),$$

by using the relation obtained by replacing $M_q{}^\alpha$ in (2·6) by $\mathcal{M}_q{}^\alpha$. In other words,

$$(\mathcal{M}_q^\alpha, \mathcal{M}_q^{\alpha *}) \cdot \hat{\Omega}_q^\alpha = \hat{\Omega}_q^\alpha \cdot (\mathcal{M}_q^\alpha, \mathcal{M}_q^{\alpha *}). \tag{3·4}$$

This equation means that if the frequency spectrum given by $\hat{\Omega}_q{}^\alpha$ is non-degenerate, the matrix $(\mathcal{M}_q{}^\alpha, \mathcal{M}_q{}^{\alpha *})$ and, by (3·3), the matrix $(\dot{\mathcal{M}}_q{}^\alpha, \mathcal{M}_q{}^{\alpha *})$ should be both diagonal. The elements of $\hat{\Omega}_q{}^\alpha$ give the temperature-dependent frequency spectrum of the collective motion. In the low temperature limit, this frequency spectrum coincides with the spin wave spectrum, and, for $q=0$ and $\alpha = +, -$, the elements of $\hat{\Omega}_0{}^\alpha$ give the antiferro- and ferrimagnetic resonance frequencies. If we adopt the Weiss approximation, these frequencies agree with those given by Nagamiya and Kittel.[6]

The temporal development of the normal modes $\langle \mathcal{M}_q{}^\alpha \rangle (t)$ is described by the following relaxation matrix:

$$\tilde{\Xi}_q^\alpha(t) \equiv U_q^\alpha \cdot \Xi_q^\alpha(t) \cdot [U_q^\alpha]^{-1} = (\mathcal{M}_q^\alpha(t), \mathcal{M}_q^{\alpha *}) \cdot (\mathcal{M}_q^\alpha, \mathcal{M}_q^{\alpha *})^{-1}. \tag{3·5}$$

In order to obtain the shift and damping of the collective motion arising from the interaction between different normal modes, it is convenient to make use of the following identity[5]:

$$\tilde{\Xi}_q^\alpha(t) = \exp(it\hat{\Omega}_q^\alpha) \cdot [1 - \int_0^t ds \exp(-is\hat{\Omega}_q^\alpha) \cdot \hat{\Gamma}_{sq}^\alpha \cdot \exp(is\hat{\Omega}_q^\alpha)], \tag{3·6}$$

where

$$\hat{\Gamma}_{sq}^\alpha = \int_0^s d\tau (\mathcal{K}_q^\alpha(\tau), \mathcal{K}_q^{\alpha *}) \cdot (\mathcal{M}_q^\alpha, \mathcal{M}_q^{\alpha *})^{-1} \cdot \exp(-i\tau\hat{\Omega}_q^\alpha), \tag{3·7}$$

$$\mathcal{K}_q^\alpha \equiv \left[\frac{d}{dt} \exp(-it\hat{\Omega}_q^\alpha) \cdot \mathcal{M}_q^\alpha(t) \right]_{t=0} = \dot{\mathcal{M}}_q^\alpha - i\hat{\Omega}_q^\alpha \cdot \mathcal{M}_q^\alpha. \tag{3·8}$$

This identity can be verified using the formula

$$(\mathcal{K}_q^\alpha,\ \mathcal{M}_q^{\alpha *})=0,$$

which follows from (3·3); $\mathcal{K}_q{}^\alpha$ represents a kind of random force acting on the normal mode $\mathcal{M}_q{}^\alpha$ due to the interactions between the normal modes. Therefore, $\hat{\Gamma}_s$ expresses the effects of these interactions on the motion of the normal modes. The fundamental assumption of the statistical mechanics of irreversible processes is that the relaxation function of this random force,

$$(\mathcal{K}_q^\alpha(s),\ \mathcal{K}_q^{\alpha *}),$$

vanishes for time intervals s larger than a microscopic correlation time τ_c which characterises a microscopic process occurring in the system. If the normal modes $\mathcal{M}_q{}^\alpha$ turn out to provide a good description of the motion of the system, the effect of the second term in the square brackets of (3·6) should be small. In this case, $\tilde{\Xi}_q{}^\alpha$ takes the following approximate form[5]:

$$\tilde{\Xi}_q^\alpha(t)\approx\exp_{(+)}\{\int_0^t ds[i\hat{\Omega}_q^\alpha-\hat{\Gamma}_{sq}^\alpha]\}, \tag{3·9}$$

where $\exp_{(+)}$ denotes the ordered exponential (time ordering from the right). Let us consider the two extreme cases of this expression.

(i) *Lorentzian limit:* τ_c is very small compared with the times which we are interested in. This applies to the central part of the resonance line shape, and to the case of extreme narrowing. Then, in the most part of the dcmain of integration, $\hat{\Gamma}_{sq}{}^\alpha$ is equal to the following asymptotic limit:

$$\hat{\Gamma}_q^\alpha\equiv\lim_{s\to\infty}\hat{\Gamma}_{sq}^\alpha=\int_0^\infty d\tau(\mathcal{K}_q^\alpha(\tau),\ \mathcal{K}_q^{\alpha *})\cdot(\mathcal{M}_q^\alpha,\ \mathcal{M}_q^{\alpha *})^{-1}\cdot\exp(-i\tau\hat{\Omega}_q^\alpha). \tag{3·10}$$

And (3·9) becomes

$$\tilde{\Xi}_q^\alpha(t)=\exp[(i\hat{\Omega}_q^\alpha-\hat{\Gamma}_q^\alpha)t]. \tag{3·11}$$

The macroscopic equation of motion of the normal modes is then expressed as

$$\frac{d}{dt}\langle\mathcal{M}_q^\alpha\rangle(t)=[i\hat{\Omega}_q^\alpha-\hat{\Gamma}_q^\alpha]\cdot\langle\mathcal{M}_q^\alpha\rangle(t). \tag{3·12}$$

Here we should note that the frequency given by $\hat{\Omega}_q{}^\alpha$ need not be small compared to $\tau_c{}^{-1}$ for the discussion of this paragraph to be valid.

(ii) *Gaussian limit:* Here we are interested in the times very small compared with τ_c and with the periods of the collective oscillations. This applies to the part at the far wings of the resonance line shape, and to the case of very little narrowing. In this case, in (3·7) it is possible to neglect $\tau\hat{\Omega}_q{}^\alpha$ and to replace $(\mathcal{K}_q{}^\alpha(\tau),\ \mathcal{K}_q{}^{\alpha *})$ by $(\mathcal{K}_q{}^\alpha,\ \mathcal{K}_q{}^{\alpha *})$, thus yielding

$$\hat{\Gamma}_{sq}^\alpha\approx s(\mathcal{K}_q^\alpha,\ \mathcal{K}_q^{\alpha *})\cdot(\mathcal{M}_q^\alpha,\ \mathcal{M}_q^{\alpha *})^{-1}. \tag{3·13}$$

Thus we obtain*)

$$\tilde{\Xi}_q^\alpha(t) \approx \exp(it\hat{\Omega}_q^\alpha) \cdot \exp\left[-\frac{t^2}{2}(\mathcal{K}_q^\alpha, \mathcal{K}_q^{\alpha*}) \cdot (\mathcal{M}_q^\alpha, \mathcal{M}_q^{\alpha*})^{-1}\right]. \quad (3\cdot14)$$

In the following sections, we shall be mainly interested in the Lorentzian limit (3·11) which applies to AFMR except at the immediate vicinity of the Neél point.

§ 4. Antiferromagnetic resonance absorption

As an application of the foregoing theory, we shall consider, in this and the following sections, the line width of the antiferromagnetic resonance absorption. The frequency was investigated elsewhere.[6)]

First we shall determine the normal modes. Denoting the two sublattices by A and B, the frequency matrices of the uniform mode $q=0$ is, from (2·5), given by

$$\left.\begin{aligned} \hat{\omega}^+ &= \begin{bmatrix} -a & -c \\ -b & -d \end{bmatrix}, \\ \hat{\omega}^- &= \begin{bmatrix} a & c^* \\ b^* & d \end{bmatrix}, \end{aligned}\right\} \quad (4\cdot1)$$

where we have introduced the notation,

$$\begin{aligned} a &= -2g\mu_B\langle M_A{}^0\rangle (M_B{}^+, M_B{}^-)/\mathcal{D}, \\ b &= 2g\mu_B\langle M_B{}^0\rangle (M_B{}^+, M_A{}^-)/\mathcal{D}, \\ c &= 2g\mu_B\langle M_A{}^0\rangle (M_A{}^+, M_B{}^-)/\mathcal{D}, \quad (4\cdot2) \\ d &= -2g\mu_B\langle M_B{}^0\rangle (M_A{}^+, M_A{}^-)/\mathcal{D}, \\ \mathcal{D} &\equiv (M_A{}^+, M_A{}^-)(M_B{}^+, M_B{}^-) - (M_B{}^+, M_A{}^-)(M_A{}^+, M_B{}^-), \end{aligned}$$

and we have used that

$$(\dot{M}_X{}^+, M_Y{}^-) = \delta_{XY} 2ig\mu_B\langle M_X{}^0\rangle, \qquad X, Y \equiv A \text{ or } B, \quad (4\cdot3)$$

which is derived by using Kubo's formula valid for any operators $\mathfrak{A}$ and $\mathfrak{B}$,

$$(i\dot{\mathfrak{A}}, \mathfrak{B}) = \langle[\mathfrak{A}, \mathfrak{B}]\rangle. \quad (4\cdot4)$$

*) Note the following formula for the ordered exponential:

$$\exp_{(+)}\left[\int_0^t ds\{A(s)+B(s)\}\right] = \exp_{(+)}\left[\int_0^t dsA(s)\right]$$
$$\cdot\exp_{(+)}\left\{-\int_0^t ds \exp_{(+)}\left[\int_s^0 ds' A(s')\right] B(s) \exp_{(+)}\left[\int_0^s ds' A(s')\right]\right\}.$$

The two eigenvalues of $\hat{\omega}^+$, or the resonance frequencies of the two modes, denoted by Ω_1 and Ω_2 are easily obtained as

$$\left.\begin{aligned}\Omega_1 &= -\frac{a+d}{2}+\frac{1}{2}\sqrt{(a-d)^2+4bc},\\ \Omega_2 &= -\frac{a+d}{2}-\frac{1}{2}\sqrt{(a-d)^2+4bc}.\end{aligned}\right\} \quad (4\cdot5)$$

Those of $\hat{\omega}^-$ are the same as (4·5) except for the sign. The corresponding transformation matrices U^+ and U^-, which diagonalize $\hat{\omega}^+$ and $\hat{\omega}^-$, respectively, are obtained as

$$\left.\begin{aligned}U^+ &= \begin{bmatrix} -bx^+ & (a+\Omega_1)\,x^+ \\ -by^+ & (a+\Omega_2)\,y^+ \end{bmatrix},\\ U^- &= \begin{bmatrix} (a+\Omega_2)\,x^- & cx^- \\ (a+\Omega_1)\,y^- & cy^- \end{bmatrix},\end{aligned}\right\} \quad (4\cdot6)$$

where x^+, x^-, y^+ and y^- are the numbers which are chosen to satisfy the relations

$$\left.\begin{aligned}x^+x^- &= \frac{-(a+\Omega_2)\,(M_B{}^+,\ M_A{}^-)+b(M_A{}^+,\ M_A{}^-)}{(\Omega_1-\Omega_2)\,\mathcal{D}bc^*},\\ y^+y^- &= \frac{(a+\Omega_1)\,(M_B{}^+,\ M_A{}^-)-b(M_A{}^+,\ M_A{}^-)}{(\Omega_1-\Omega_2)\,\mathcal{D}bc^*}.\end{aligned}\right\} \quad (4\cdot7)$$

It was shown[6] that, in the Weiss approximation, the resonance frequencies given by (4·5) lead to those given by Nagamiya and Kittel,[7]

$$\left.\begin{aligned}\Omega_1/g\mu_B &= \sqrt{(2A+\alpha)\,\alpha M^2+\left(\frac{A\chi_{//}H}{2}\right)^2}+\left(1-\frac{A-\alpha}{2}\chi_{//}\right)H,\\ -\Omega_2/g\mu_B &= \sqrt{(2A+\alpha)\,\alpha M^2+\left(\frac{A\chi_{//}H}{2}\right)^2}-\left(1-\frac{A-\alpha}{2}\chi_{//}\right)H,\end{aligned}\right\} \quad (4\cdot8)$$

where A is the exchange field constant, α the anisotropy field constant,*) M the spontaneous magnetization of each sublattice in the absence of an external field, $\chi_{//}$ the parallel susceptibility and H the external magnetic field. Thus we see that the mode 1 and mode 2 correspond to the high frequency and the low frequency mode, respectively.

The damping constants of the two normal modes denoted by γ_1 and γ_2 are derived from (3·10): noting that the matrix $(\mathcal{M}^\alpha,\ \mathcal{M}^{\alpha *})$ is diagonal,

---

*) α is related to the constant K of the usual definition by $\alpha=K/M^2$.

$$\gamma_j = \mathrm{Re}\int_0^\infty ds\, (\mathcal{K}_j^+(s),\ \mathcal{K}_j^-)\ \exp(-is\Omega_j)/(\mathcal{M}_j^+,\ \mathcal{M}_j^-),\qquad (j=1,\ 2). \tag{4·9}$$

In the low temperature region, (4·9) should be able to be evaluated by employing the perturbation theory, as we have shown in I, and yield the damping constants of the spin waves with zero wave vector. These damping constants have been recently calculated by Russian authors.[10] Here we shall study the damping constants at higher temperatures, putting emphasis on their behavior near the Neél temperature. As in I, we assume the time dependence of the correlation function $(\mathcal{K}_j^+(s),\ \mathcal{K}_j^-)$ of the following form:

$$(\mathcal{K}_j^+(s),\ \mathcal{K}_j^-) = (\mathcal{K}_j^+,\ \mathcal{K}_j^-)\exp(i\nu_j s - g_j^2 s^2), \tag{4·10}$$

where ν_j and g_j are determined by the first and the second moments of this correlation function as follows:

$$\nu_j = \frac{1}{i}\frac{(\dot{\mathcal{K}}_j^+,\ \mathcal{K}_j^-)}{(\mathcal{K}_j^+,\ \mathcal{K}_j^-)},\quad g_j^2 = -\frac{1}{2}\nu_j^2 + \frac{1}{2}\frac{(\dot{\mathcal{K}}_j^+,\ \dot{\mathcal{K}}_j^-)}{(\mathcal{K}_j^+,\ \mathcal{K}_j^-)}. \tag{4·11}$$

Then, (4·9) gives

$$\gamma_j = \frac{\sqrt{\pi} f_j}{2g_j}\cdot\exp\left[-\frac{(\nu_j-\Omega_j)^2}{4g_j^2}\right], \tag{4·12}$$

where

$$f_j \equiv (\mathcal{K}_j^+,\ \mathcal{K}_j^-)/(\mathcal{M}_j^+,\ \mathcal{M}_j^-). \tag{4·13}$$

The approximation (4·10) and (4·11) is certainly a very crude one. However, this type of approximation is the only feasible one we have. Nevertheless, we might hope that main qualitative features of the damping constant are still given by (4·12). Thus our problem is reduced to the calculation of the various moments of the relaxation function $(\mathcal{M}_j^+(s),\ \mathcal{M}_j^-)$.

Using the relation,

$$\mathcal{M}^\pm = U^\pm\cdot M^\pm, \tag{4·14}$$

with $U^\pm$ given by (4·6), these moments are expressed in the following form: writing $\mathfrak{A}_{(u)} \equiv [(d^u/dt^u)\mathfrak{A}(t)]_{t=0}$,

$$(\mathcal{M}_{j(u)}^+,\ \mathcal{M}_{j(v)}^-) = A_j\{p_j(T_{(u)}^+,\ T_{(v)}^-) + q_j(T'^+_{(u)},\ T'^-_{(v)}) + r_j(T'^+_{(u)},\ T_{(v)}^-) + s_j(T_{(u)}^+,\ T'^-_{(v)})\},\qquad (j=1,\ 2), \tag{4·15}$$

where A_j are unimportant numerical constants, and T^α and T'^α $(\alpha=+,\ -)$ are the sums and the differences of the sublattice magnetizations given by

$$\left.\begin{aligned} T^\alpha &\equiv M_A^\alpha + M_B^\alpha, \\ T'^\alpha &\equiv M_A^\alpha - M_B^\alpha. \end{aligned}\right\} \tag{4·16}$$

The constants p_j, q_j, r_j and s_j are defined in terms of the quantities given by (4·2) and (4·5) as

$$\left.\begin{aligned}
p_1 &\equiv -a-\Omega_2+\frac{c^*}{b}(a+\Omega_1)-c-c^*,\\
q_1 &\equiv -a-\Omega_2+\frac{c^*}{b}(a+\Omega_1)+c+c^*,\\
r_1 &\equiv -a-\Omega_2-\frac{c^*}{b}(a+\Omega_1)+c-c^*,\\
s_1 &\equiv -a-\Omega_2-\frac{c^*}{b}(a+\Omega_1)-c+c^*,\\
p_2 &\equiv -a-\Omega_1+\frac{c^*}{b}(a+\Omega_2)-c-c^*,\\
q_2 &\equiv -a-\Omega_1+\frac{c^*}{b}(a+\Omega_2)+c+c^*,\\
r_2 &\equiv -a-\Omega_1-\frac{c^*}{b}(a+\Omega_2)+c-c^*,\\
s_2 &\equiv -a-\Omega_1-\frac{c^*}{b}(a+\Omega_2)-c+c^*.
\end{aligned}\right\}\quad(4\cdot17)$$

In the Weiss approximation, (4·17) reduces to the following:

$$\left.\begin{aligned}
p_1/g\mu_B &= 2\sqrt{(2A+\alpha)\alpha M^2+\left(\frac{A\chi_{//}H}{2}\right)^2}+A(M+\chi_{//}H),\\
q_1/g\mu_B &= 2\sqrt{(2A+\alpha)\alpha M^2+\left(\frac{A\chi_{//}H}{2}\right)^2}-A(M+\chi_{//}H),\\
r_1/g\mu_B &= s_1/g\mu_B=(A+\alpha)M,\\
p_2 &= -q_1,\ q_2=-p_1,\ r_2=s_2=r_1=s_1.
\end{aligned}\right\}\quad(4\cdot18)$$

In particular, above the Neél point, we obtain

$$\left.\begin{aligned}
p_1/g\mu_B &= -q_2/g\mu_B=2A\chi_{//}H,\\
p_2 &= q_1=r_1=s_1=0.
\end{aligned}\right\}\quad(4\cdot19)$$

§ 5. Line width in the vicinity of the Neél Point —above the Neél point—

First we shall consider the line width above the Neél point. The normal modes then reduce to the following: aside from numerical factors,

$$\left.\begin{aligned}\mathcal{M}_1^\alpha &= M_A^\alpha + M_B^\alpha, \\ \mathcal{M}_2^\alpha &= M_A^\alpha - M_B^\alpha.\end{aligned}\right\} \quad (T \geqq T_N) \tag{5·1}$$

Since the two sublattices A and B are indistinguishable above the Neél point, the mode 2 is not excited by a microwave, and shall be omitted here. We are concerned with the mode 1, which is the paramagnetic resonance. The resonance frequency of the mode 1 is, according to (4·8), equal to $g\mu_B H$ with the neglect of a small term involving the anisotropy field constant. As we shall show later at almost all temperatures except the immediate neighborhood of the Neél point, the following conditions are satisfied:

$$|\nu_1 - \Omega_1| \ll 2g_1, \qquad f_1 \ll \frac{4}{\pi} g_1^2. \tag{5·2a \& b}$$

Then, the damping constant (4·12) becomes

$$\gamma_1 = \frac{\sqrt{\pi} f_1}{2g_1}. \tag{5·3}$$

The latter condition of (5·2) is then equivalent to the condition

$$\gamma_1 \ll g_1, \tag{5·4}$$

which is also the condition that the line shape be Lorentzian with the width given by

$$\Delta H_1 = \gamma_1 / \frac{\partial \Omega_1}{\partial H} = \gamma_1 / g\mu_B. \tag{5·5}$$

In order to see the temperature dependence of the line width in the vicinity of the Neél point, it is a good approximation to replace the relaxation function appearing in the expressions for f_1 and g_1 by β times the corresponding correlation functions properly symmetrized. Thus we obtain

$$f_1 = \frac{\beta}{2\chi_\perp} \langle \{\mathcal{K}_1^+, \mathcal{K}_1^-\} \rangle, \tag{5·6}$$

$$g_1^2 = g_{1\infty}^2 \left(\frac{g_1^2}{g_{1\infty}^2} \right), \tag{5·7}$$

$$\frac{g_1^2}{g_{1\infty}^2} = \frac{\langle \{\dot{\mathcal{K}}_1^+, \dot{\mathcal{K}}_1^-\} \rangle}{\langle \{\dot{\mathcal{K}}_1^+, \dot{\mathcal{K}}_1^-\} \rangle_\infty} \, \frac{\langle \{\mathcal{K}_1^+, \mathcal{K}_1^-\} \rangle_\infty}{\langle \{\mathcal{K}_1^+, \mathcal{K}_1^-\} \rangle}, \tag{5·8}$$

where $\chi_\perp$ is the perpendicular susceptibility given by

$$\chi_\perp \equiv \frac{1}{2} (M_A^+ + M_B^+, \, M_A^- + M_B^-), \tag{5·9}$$

and $g_{1\infty}$ denotes the value of g_1 in the high temperature limit and $\langle \ldots \rangle_\infty$ indicates the correlation function evaluated in this limit. For our purpose, it is also permissible to neglect the temperature dependence of $\langle\{\dot{\mathcal{K}}_1^+, \dot{\mathcal{K}}_1^-\}\rangle$. That this approximation does not affect the temperature dependence of the width near the Neél point is seen as follows. Replacing $\langle\{\dot{\mathcal{K}}_1^+, \dot{\mathcal{K}}_1^-\}\rangle$ by $\beta^{-1}(\dot{\mathcal{K}}_1^+, \dot{\mathcal{K}}_1^-)$ and using (4·4), we have

$$\langle\{\dot{\mathcal{K}}_1^+, \dot{\mathcal{K}}_1^-\}\rangle = \frac{1}{i\beta}\langle[\mathcal{K}_1^+, \dot{\mathcal{K}}_1^-]\rangle.$$

Each of the operators $\mathcal{K}_1^+$ and $\dot{\mathcal{K}}_1^-$ is the sum of the products of the spin operators which are spatially very close to each other. Their commutator has, therefore, the same property. The static correlation function of spins situated very close to each other varies slowly with the temperature near the Neél point and, hence, its temperature dependence can be ignored in the first approximation. Thus, g_1^2 becomes

$$g_1^2 \approx g_{1\infty}^2 \frac{\langle\{\mathcal{K}_1^+, \mathcal{K}_1^-\}\rangle_\infty}{\langle\{\mathcal{K}_1^+, \mathcal{K}_1^-\}\rangle}. \tag{5·10}$$

Substituting (5·6) and (5·10) into (5·3), we thus obtain

$$\gamma_1 = \frac{\sqrt{\pi}\,\beta}{4\chi_\perp g_{1\infty}[\langle\{\mathcal{K}_1^+, \mathcal{K}_1^-\}\rangle_\infty]^{1/2}}[\langle\{\mathcal{K}_1^+, \mathcal{K}_1^-\}\rangle]^{3/2}. \tag{5·11}$$

Now we adopt the Heisenberg model for describing the antiferromagnetics with a uniaxial anisotropy and an external magnetic field, both of which are in the z-direction. The system Hamiltonian is then written as

$$\mathcal{H} = -\sum_{m,f} J_{mf}\,\mathbf{S}_m\cdot\mathbf{S}_f - D\sum_m \mathbf{S}_m^{0\,2} + g\mu_B H\sum_m S_m^0, \tag{5·12}$$

where the summations run over all the lattice sites. The equations of motion for $\mathcal{M}_1^\pm$, then, become

$$\dot{\mathcal{M}}_1^\pm = \pm 2ig\mu_B D\sum_m\{S_m^\pm, S_m^0\} \mp i(g\mu_B)^2 H\sum_m S_m^\pm. \tag{5·13}$$

The calculation of $\langle\{\mathcal{K}_1^+, \mathcal{K}_1^-\}\rangle$ can be performed most easily by replacing this again by the relaxation function divided by β:

$$\begin{aligned}\langle\{\mathcal{K}_1^+, \mathcal{K}_1^-\}\rangle &\approx \frac{1}{\beta}(\dot{\mathcal{M}}_1^+ - i\Omega_1\mathcal{M}_1^+, \dot{\mathcal{M}}_1^- + i\Omega_1\mathcal{M}_1^-)\\ &= \frac{1}{\beta}\left\{\frac{1}{i}\langle[\mathcal{M}_1^+, \dot{\mathcal{M}}_1^-]\rangle + 2\Omega_1\langle[\mathcal{M}_1^+, \mathcal{M}_1^-]\rangle + \Omega_1^2(\mathcal{M}_1^+, \mathcal{M}_1^-)\right\}\\ &= \frac{(g\mu_B)^2}{\beta}[2\chi_{//}H^2 + 2D\sum\langle 3S_m^{0\,2} - S(S+1)\rangle - 4\chi_{//}H^2 + 2\chi_\perp H^2]\\ &\approx \frac{2(g\mu_B)^2}{\beta}D\sum_m\langle 3S_m^{0\,2} - S(S+1)\rangle.\end{aligned} \tag{5·14}$$

Here, we have used that $\chi_{//}\approx\chi_\perp$ near the Neél point. The quantity $\langle 3S_m^{0\,2} - S(S+1)\rangle$

appearing in (5·14) vanishes in the absence of an anisotropy or an external field above the Neél point. Therefore, in the presence of a small amount of anisotropy or external field, it is necessary to expand the density matrix with respect to these small quantities to obtain the first nonvanishing contributions. The contribution from the external field vanishes because this involves odd powers of spin operators. Thus we are left with

$$\langle 3S_m^{0\,2}-S(S+1)\rangle \approx \frac{D}{3}\sum_f (3S_m^{0\,2}-S(S+1),\ 3S_f^{0\,2}-S(S+1)).$$

Therefore, (5·14) becomes

$$\langle\{\mathcal{K}_1^+,\ \mathcal{K}_1^-\}\rangle \approx \frac{2(g\mu_B D)^2}{3\beta}\sum_m\sum_f (3S_m^{0\,2}-S(S+1),\ 3S_f^{0\,2}-S(S+1)), \tag{5·15}$$

$$=\frac{6(g\mu_B)^2}{\beta}(\mathcal{H}_{\rm anis}-\langle\mathcal{H}_{\rm anis}\rangle,\ \mathcal{H}_{\rm anis}-\langle\mathcal{H}_{\rm anis}\rangle), \tag{5·16}$$

where $\mathcal{H}_{\rm anis}$ is the anisotropy energy operator given by

$$\mathcal{H}_{\rm anis}=-D\sum_m S_m^{0\,2}. \tag{5·17}$$

Thus, we see that the second moment of the resonance line is essentially given by the fluctuation of the anisotropy energy, which is in agreement with the physical interpretation of Ohlmann.[3]

Now we evaluate the double sum in (5·15), which can be written, again replacing it by β times the correlation function, as

$$\beta N\sum_f\langle\{3S_m^{0\,2}-S(S+1),\ 3S_f^{0\,2}-S(S+1)\}\rangle, \tag{5·18}$$

where N is the number of magnetic lattice sites per unit volume. This involves the correlation of four spin operators. We approximate it by replacing it with the product of pair correlation functions of four spin operators. The terms involving $\langle S_m^{0\,2}\rangle$ or $\langle S_f^{0\,2}\rangle$ cancel with those of $S(S+1)$. Then (5·18) reduces to

$$18\beta N\sum_f\langle S_m^0\ S_f^0\rangle^2. \tag{5·19}$$

The spin pair correlation function $\langle S_m^0\ S_f^0\rangle$ of the antiferromagnet at a large distance R_{mf} is evaluated to be given by[11]

$$\langle S_m^0\ S_f^0\rangle=\epsilon_m\epsilon_f\frac{S(S+1)}{12\pi r_1^2 N}\ \frac{1}{R_{mf}}\exp(-\kappa_1 R_{mf}), \tag{5·20}$$

where

$$\epsilon_m=\begin{cases}+1 & \text{for } m \text{ on the sublattice } A,\\ -1 & \text{for } m \text{ on the sublattice } B,\end{cases} \tag{5·21}$$

and r_1 and κ_1 are Van Hove's parameters of the neutron scattering. In the vicinity of the Neél temperature, r_1 is a constant, whereas κ_1 vanishes in the Weiss approximation according to the formula

$$(\kappa_1 r_1)^2 = \frac{T-T_N}{T} \equiv \delta. \tag{5·22}$$

Thus, we obtain from (5·15), (5·18), (5·19), (5·20) and (5·22),

$$\langle\{\mathcal{K}_1^+,\ \mathcal{K}_1^-\}\rangle = \frac{[g\mu_B DS(S+1)]^2}{6\pi r_1^3\ \delta^{1/2}}. \tag{5·23}$$

Therefore, from (5·10) we see that in the vicinity of the Neél point, g_1^2 vanishes as $\delta^{1/2}$. This means that as we approach the Neél point, the correlation of torque decays more and more slowly. Although this conclusion was drawn with the assumption of Gaussian decay, it is natural to expect that this feature qualitatively holds in general.

It is appropriate here to remark that the singular behavior of (5·23) at the Neél point is the genuine singularity, not masked by the presence of the anisotropy or the external field. This is inferred from the fact that this comes from the fluctuations of the z component of spins and $\langle(M_A^z - M_B^z)^2\rangle$ shows the true singularity at the Neél point. This is the characteristic feature of antiferromagnets in contrast with ferromagnets. On the other hand, the fluctuations of the transverse components of spins do not show such a singularity in the presence of the anisotropy energy or the external field. $g_{1\infty}$ as well as $\langle\{\mathcal{K}_1^+,\ \mathcal{K}_1^-\}\rangle_\infty$ are evaluated by a straight-forward calculation, and we quote the results for b.c.c. lattice:

$$\langle\{\mathcal{K}_1^+,\ \mathcal{K}_1^-\}\rangle_\infty = \frac{2}{5}N(Dg\mu_B)^2 S(S+1)[4S(S+1)-3], \tag{5·24}$$

$$g_{1\infty}^2 = \frac{74}{9}\,\frac{S(S+1)[S(S+1)-(47/111)]}{4S(S+1)-3}[z_1J_1^2 + z_2J_2^2], \tag{5·25}$$

where J_1 and J_2 are the magnitudes of the exchange interactions between the first and the second neighbors, and z_1 and z_2 are the numbers of the first and the second neighbor atoms.

Substituting (5·23) to (5·25) into (5·11), we finally deduce the following expression for the damping constant near the Neél point:

$$\gamma_1 = \frac{\beta[Dg\mu_B S(S+1)]^2}{2N^{1/2}\chi_\perp r_1^{9/2}} \cdot \left[\frac{5}{222\times(4\pi)^3\times[S(S+1)-(47/111)][z_1J_1^2+z_2J_2^2]}\right]^{1/2} \cdot \delta^{-3/4}. \tag{5·26}$$

For numerical calculation, it is more convenient to express (5·26) in terms of the exchange fields and the anisotropy field at the zero temperature which are related to the constants J_1, J_2 and D by

$$H_E=\frac{z_1J_1S}{g\mu_B},\ H_E'=\frac{z_2J_2S}{g\mu_B},\ H_A=\frac{2DS}{g\mu_B}. \tag{5·27}$$

The line width ΔH then becomes

$$\Delta H=\frac{3S(S+1)}{4}\left[\frac{5}{222\times(4\pi)^3\times[S(S+1)-(47/111)]}\right]^{1/2}$$
$$\times\frac{H_A^2/H_E}{(1-\theta)(Nr_1^3)^{3/2}[1/z_1+\theta^2/z_2]^{1/2}}\delta^{-3/4}, \tag{5·28}$$

where $\theta\equiv H_E'/H_E=(\Theta-T_N)/(\Theta+T_N)$, Θ being the paramagnetic Curie temperature.

In the Weiss approximation, de Gennes has shown that[11)]

$$r_1^2=\frac{1}{12}\left[\left(1+\frac{\Theta}{T_N}\right)b_1^2+\left(1-\frac{\Theta}{T_N}\right)b_2^2\right], \tag{5·29}$$

where b_1 and b_2 are the distances between the first and the second neighbors, respectively. As a numerical example, we consider MnF_2, for which the values of the various quantities are taken as follows:[2),7)]

$$H_A=8{,}800\text{ oe},\ H_E=556{,}000\text{ oe},\ b_1=3.82\text{ Å},$$
$$b_2=4.35\text{ Å},\ T_N=67°\text{K},\ \Theta=113°\text{K}, \tag{5·30}$$

where b_2 is the average value of the second neighbor distances. Substituting these values into (5·29) and (5·28), the width becomes

$$\Delta H=62\times\delta^{-3/4}\text{ oe}. \tag{5·31}$$

The line width above the Neél point has been measured by Hutchison and Stout,[2)] and shows the rapid increase near the Neél point in qualitative agreement with (5·31). The theoretical value (5·31) is, however, a few times smaller than the observed one, but it is no wonder considering the crude approximations we had to make. Before concluding this section, we examine the condition (5·2) which limits the validity of our result. By a similar analysis to that which has been done in the earlier part of this section, we find that ν_1 is of the order of $g\mu_BH\cdot\delta^{1/2}$, and is negligible compared with $\Omega_1\approx g\mu_BH$. On the other hand, by (5·10), (5·23), (5·24), (5·25) and (5·27), we have $g_1\sim g\mu_BH_E\delta^{1/4}$. Thus, the condition (5·2a) is satisfied as long as $\delta^{1/4}>H/H_E$. That is, in the example of MnF_2, for $H\sim5{,}000$ oe, the exponential factor in (4·12) can be omitted except within 10^{-6}°K of the Neél point. Therefore, this condition can be ignored completely. Turning to the condition (5·2b), γ_1 can be estimated from (5·28) as $\gamma_1\sim g\mu_B\cdot(H_A^2/H_E)\cdot\delta^{-3/4}$. Thus this condition becomes

$$H_A^2/H_E\cdot\delta^{-3/4}<H_E\cdot\delta^{1/4}.$$

This may be written as

$$\delta>(H_A/H_E)^2.$$

In other words, the line shape is well approximated by the Lorentzian curve except within 4×10^{-2} °K of the Neél point. Within this temperature region, the discussion of § 4 shows that the narrowing becomes ineffective and the line shape changes to be Gaussian.

§ 6. Line width below the Neél point

Below the Neél point, the full complexity of the two normal modes of § 4 must be taken into account because the sharp rise of the spontaneous magnetizations below the Neél point affects both the nature of the normal modes and the various static correlations even just below the Neél point. And so far we have not been able to obtain a clear analysis of the line width in this region. However, a qualitative study similar to the one discussed earlier has shown that a genuine thermodynamical singularity does not appear in the mode 2, although for the mode 1 we expect a similar singularity to that obtained above the Neél point. Johnson and Nethercot[2] observed that the line width of the mode 2 below the Neél point increases enormously as we approach the Neél point. If our analysis is not wrong, the observed increase must be attributed to other causes. One of such causes may be a field dependence of the resonance frequency. The line width are related to the damping constants by the formula

$$\Delta H_j=\gamma_j\Big/\left|\frac{\partial\Omega_j}{\partial H}\right|,\quad (j=1,\ 2). \tag{6·1}$$

Using (4·8), we obtain

$$\left.\begin{aligned}\left|\frac{1}{g\mu_B}\cdot\frac{\partial\Omega_1}{\partial H}\right|&=1-\frac{A-\alpha}{2}\chi_{//}+\frac{(A\chi_{//}/2)^2H}{\sqrt{(2A+\alpha)\alpha M^2+(A\chi_{//}H/2)^2}},\\ \left|\frac{1}{g\mu_B}\cdot\frac{\partial\Omega_2}{\partial H}\right|&=1-\frac{A-\alpha}{2}\chi_{//}-\frac{(A\chi_{//}/2)^2H}{\sqrt{(2A+\alpha)\alpha M^2+(A\chi_{//}H/2)^2}}.\end{aligned}\right\} \tag{6·2}$$

In the low temperatures, where M is large and $A\chi_{//}$ is small, both quantities in the above expressions are of the order of unity. At the Neél point where $M=0$ and $A\chi_{//}=1$, (6·2) reduces to

$$\left.\begin{aligned}\left|\frac{1}{g\mu_B}\ \frac{\partial\Omega_1}{\partial H}\right|&=1+\frac{\alpha}{2A},\\ \left|\frac{1}{g\mu_B}\ \frac{\partial\Omega_2}{\partial H}\right|&=\frac{\alpha}{2A}.\end{aligned}\right\} \tag{6·3}$$

Thus for the small anisotropy constant α such that $\alpha/2A\ll1$, the line width of the mode 2 is expected to increase enormously near the Neél point, even if other factors remain constant. However, the numerical computation has shown that such an increase appears only within an extremely small region (within a few thousandths of a degree) around the Neél point due to the smallness of the external fields compared with the exchange field and completely fails to explain the observed increase. It seems that a more elaborate analysis along the line of § 4 is needed to clarify the widths below the Neél point.

§ 7. Discussion

In the foregoing two sections we have studied the behaviors of line widths of the two normal modes in the vicinity of the Neél point, and have seen that the singular thermodynamic fluctuation connected with the antiferromagnetic transition reveals itself only in the high frequency mode 1. Let us consider why this is so. For this purpose, the sublattice magnetizations for the two

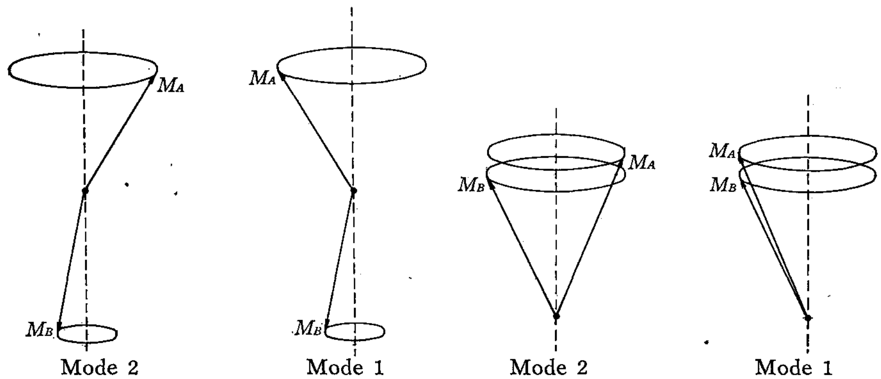

Fig. 1. Normal modes for T well below T_N. Fig. 2. Normal modes near T_N.

normal modes in a rotating coordinate system are shown in Fig. 1 and Fig. 2. Figure 1 shows the magnetizations well below the Neél point whereas Fig. 2 gives them just below the Neél point. As we have discussed in § 2, if we neglect the damping and the shift, the state of the system is described by the local equilibrium density matrix. The local equilibrium is a kind of equilibrium with certain constraints. In this sense, the states of the system depicted in Fig. 1 and Fig. 2 may be looked upon as a kind of equilibrium state. As we approach the Neél temperature, the two sublattice magnetizations of the mode 1 get closer and closer to each other, and finally they coincide at the Neél point, whereas in the mode 2, they always remain separated from each other as we go through the Neél point. Thus we expect that in the mode 1, as we approach the Neél point, an enormous amount of fluctuations of the sublattice magnetizations should appear, just as in the ordinary antiferromagnetic transition. In the mode 2, on the other hand, this cannot happen because the two sublattice magnetizations are always separated by a macroscopic amount. According to the fluctuation-dissipation theorem, the dissipation such as the resonance line width is always associated with the fluctuation. Thus the above consideration explains why the genuine thermodynamic singularity appears only in the width of the mode 1.

Acknowledgments

The authors wish to express their sincere gratitude to Professor T. Matsubara and Professor R. Kubo for their enlightening discussions and continued encouragement. This work was partially financed by the Scientific Research Fund of the Ministry of Education.

References

1) H. Mori and K. Kawasaki, Prog. Theor. Phys. **27** (1962), 529.
2) F. M. Johnson and A. H. Nethercot, Phys. Rev. **114** (1959), 705.
3) R. C. Ohlmann, Dissertation, Physics Department, University of California, Berkeley (June, 1960).
4) T. Moriya, Prog. Theor. Phys. **28** (1962), 371.
5) H. Mori, Prog. Theor. Phys. **28** (1962), 763.
6) H. Mori, K. Kawasaki and K. Tani, Prog. Theor. Phys. **27** (1962), 230.
7) T. Nagamiya, K. Yosida and R. Kubo, Advances in Phys. **4** (1955), 1.
8) P. W. Anderson, **83** (1951), 1260.
9) W. J. O'Sullivan, W. A. Robinson and W. W. Simmons, Phys. Rev. **124** (1961), 1317.
10) G. I. Urushadze, JETP **39** (1960), 680; Soviet Phys. JETP, **12** (1961), 476.
V. N. Genkin and V. M. Fain, JETP **41** (1961), 1522.
11) P. G. de Gennes, Report to CEA de Saclay, No. 929, (1959).