

## Magnetic Properties of Low Dimensional Spin Systems

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Using the method of the two-time Green's function, both static and dynamic properties of low dimensional spin systems with an anisotropic exchange interaction were investigated. It was shown that the magnetic behaviours of one- and two-dimensional spin systems are considerably different from those of the usual three-dimensional systems. That is, the transition temperature is lower than that expected from the magnitude of the coupling constant when an anisotropy is small enough. The short range order is developed more remarkably in the neighbourhood of the transition temperature. The damping constant increases in both ferro- and antiferromagnet with decreasing temperature owing to the anomalous growth of fluctuation. At the same time a broad shoulder is developed in the line shape, which may be considered as indicating quasi-collective modes of motion which persist in the paramagnetic phase.

### § 1. Introduction

Recently many experiments have revealed the magnetic properties of complex salts like  $\text{Cu}(\text{NH}_3)_4\text{SO}_4 \cdot \text{H}_2\text{O}$ ,<sup>1),2)</sup>  $\text{Cu}(\text{C}_6\text{H}_5\text{COO})_2 \cdot 3\text{H}_2\text{O}$ <sup>3)</sup> and  $\text{CoCl}_2 \cdot 2\text{H}_2\text{O}$ <sup>4)</sup>, etc., which are considered to have a rather strong magnetic coupling along a particular axis. Their magnetic behaviours have several features different from those of the usual three-dimensional spin systems. That is, the transition temperature is considerably lower than that expected from the magnitude of the coupling constant. Near the transition temperature the behaviours of susceptibility and specific heat deviate strongly from that in the simple molecular field theory. Anomalous relaxation phenomena are found in the vicinity of the transition point.

A number of theoretical works have been devoted to the study of static properties. However, only in the case of a plane and a linear Ising spin systems exact solutions have been found for the transition temperature, the specific heat,<sup>5)</sup> the spin polarization,<sup>6)</sup> and the magnetic susceptibility.<sup>7),8)</sup> On the other hand, on the Heisenberg spin system, only a linear chain system has been investigated extensively.<sup>9)~12)</sup> The dynamical properties of low dimensional spin system have scarcely been investigated theoretically.<sup>13),14)</sup>

The aim of this paper is to treat both static and dynamic properties and their relations in the low dimensional spin systems, and to look for the cause of difference between the lower dimensional systems and the three-dimensional one.

These problems are investigated here by using the Green's function method for two reasons. First, it allows us to treat both static and dynamic behaviours

in the same theoretical framework, as was shown in Tomita and Tanaka's paper.<sup>15),\*)</sup> Secondly, several papers have already succeeded in obtaining reasonable descriptions of the spin systems by means of this method.<sup>15)~19)</sup>

The response function of a transverse component is identical with the following Green's function<sup>16)</sup>

$$\begin{aligned} G^{+-}(\mathbf{r}_1|\mathbf{r}_2, t_1-t_2) &\equiv \Theta(t_1-t_2) \langle [S_{\mathbf{r}_1}^+(t_1), S_{\mathbf{r}_2}^-(t_2)] \rangle \\ &= \frac{1}{N^2} \sum_{\mathbf{q}_1} \sum_{\mathbf{q}_2} \int_{-\infty}^{\infty} G^{+-}(\mathbf{q}_1|\mathbf{q}_2; \omega) \exp\{i(\mathbf{q}_1\mathbf{r}_1 + \mathbf{q}_2\mathbf{r}_2)\} \exp\{-i\omega(t_1-t_2)\} d\omega, \end{aligned} \quad (1.1)$$

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

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

$\langle \dots \rangle$  is a canonical average and  $N$  indicates the total number of spins. Therefore the Green's function  $G^{+-}(\mathbf{q}_1|\mathbf{q}_2, \omega)$  is directly related to the generalized transverse susceptibility  $\chi^{+-}(\mathbf{q}, \omega)$ .

In § 2 the transverse static correlation functions are derived and the transition temperature, the paramagnetic polarization and the susceptibility are investigated both qualitatively and quantitatively. Also a discussion will be given of the degree of the short range order on the basis of the sum rule. Section 3 is devoted to the behaviour of the damping constant. In § 4, the line shape of the magnetic resonance absorption is studied. The approximations employed in this paper are examined in § 5.

## § 2. Static properties

### 2.1) Correlation functions

Let us consider a plane system of Heisenberg spins with  $S = \frac{1}{2}$ , which lies in the  $x$ - $z$  coordinate plane. The Hamiltonian is then given by

$$\begin{aligned} H = & - \sum_{\mathbf{r}} \omega_0(\mathbf{r}) S_{\mathbf{r}}^0 + \sum_{\mathbf{r} > \mathbf{s}} J(r_x - s_x) \delta_k(r_x - s_x) \{S_{\mathbf{r}}^0 S_{\mathbf{s}}^0 + \frac{1}{2}(1-\alpha)(S_{\mathbf{r}}^+ S_{\mathbf{s}}^- + S_{\mathbf{r}}^- S_{\mathbf{s}}^+)\} \\ & + \eta \sum_{\mathbf{r} > \mathbf{s}} J(r_x - s_x) \delta_k(r_x - s_x) \{S_{\mathbf{r}}^0 S_{\mathbf{s}}^0 + \frac{1}{2}(1-\alpha)(S_{\mathbf{r}}^+ S_{\mathbf{s}}^- + S_{\mathbf{r}}^- S_{\mathbf{s}}^+)\}, \end{aligned} \quad (2.1)$$

where the notation  $\delta_k(\mathbf{r})$  means Kronecker's  $\delta$  function. The first term stands for the Zeeman energy ( $\omega_0(\mathbf{r}) = \gamma H(\mathbf{r})$ ), and the second and the last terms are the anisotropic exchange energy of intra- and inter-spin chain, respectively. The exchange coupling  $J(x)$  is assumed to be nonvanishing only between the nearest neighbouring spins. The parameter  $\eta$  denotes the ratio of coupling constants along two coordinate directions; that is, for  $\eta = 0$ , the system is reduced to a

\*) Hereafter, this paper will be referred to as T-T. (See Appendix I.)

one-dimensional system along  $z$ -axis. The notation  $\alpha$  is a parameter which indicates the degree of anisotropy in the exchange energy; when  $\alpha=0$ , the interaction becomes isotropic, and when  $\alpha=1$ , the system is reduced to an Ising model.

Under the Hamiltonian Eq. (2.1), the equations of motion of the Fourier transformed Green's functions are given by

$$\begin{aligned} \omega G^{\pm\mp}(\mathbf{k}|\mathbf{k}'; \omega) &= \pm i2\langle S^0 \rangle \pm \sum_{\mathbf{q}} \omega_0(\mathbf{q}) G^{\pm\mp}(\mathbf{k}-\mathbf{q}|\mathbf{k}'; \omega) \\ &\pm \frac{1}{N} \sum_{\mathbf{q}} D(\mathbf{q}, \mathbf{k}-\mathbf{q}) G^{0\pm\mp}(\mathbf{k}-\mathbf{q}, \mathbf{q}|\mathbf{k}'; \omega), \end{aligned} \quad (2.2)$$

where

$$D(\mathbf{q}, \mathbf{k}-\mathbf{q}) = (1-\alpha)J(0) (\cos q_z + \eta \cos q_x) - J(0) \{ \cos(k_z - q_z) + \eta \cos(k_x - q_x) \}, \quad (2.3)$$

and the Fourier transform is defined by

$$A_x = \frac{1}{N} \sum_{\mathbf{q}} A(\mathbf{q}) \exp(-i\mathbf{q}\mathbf{x}). \quad (2.4)$$

The so-called random phase approximation is introduced to close the hierarchy of the Green's functions. The nonvanishing component of the polarization  $\sigma(\mathbf{q}) = N^{-1}\langle S^0(\mathbf{q}) \rangle$  is nothing but the one including a value of  $\mathbf{q}$  which is identical with that of the impressed static field, i.e.  $\mathbf{q}=\mathbf{0}$  for a uniform field  $\omega_0(\mathbf{0})$  and  $\mathbf{q}=\boldsymbol{\pi}$  for a fictitious staggered field  $\omega_0(\boldsymbol{\pi})$ .

The solution of Eq. (2.2) can easily be obtained as follows:<sup>15)</sup>

$$\begin{aligned} G^{+-}(\mathbf{k}|\mathbf{k}; \omega) &= i\sigma(\boldsymbol{\pi}) \frac{\Delta_0(\boldsymbol{\pi}, \mathbf{k}-\boldsymbol{\pi})}{\sqrt{\Delta_0(\mathbf{k}-\boldsymbol{\pi}, \boldsymbol{\pi}) \Delta_0(\mathbf{k}, \boldsymbol{\pi})}} \\ &\times \left[ \frac{1}{\omega - \sqrt{\Delta_0(\mathbf{k}-\boldsymbol{\pi}, \boldsymbol{\pi}) \Delta_0(\mathbf{k}, \boldsymbol{\pi})}} + \frac{1}{\omega + \sqrt{\Delta_0(\mathbf{k}-\boldsymbol{\pi}, \boldsymbol{\pi}) \Delta_0(\mathbf{k}, \boldsymbol{\pi})}} \right] N \\ &\text{for a staggered field, } (\mathbf{q}=\boldsymbol{\pi}) \end{aligned} \quad (2.5)$$

$$\begin{aligned} &= i \frac{2\sigma(\mathbf{0})}{\omega - \Delta_0(\mathbf{k}, \mathbf{0})} N \\ &\text{for a uniform field, } (\mathbf{q}=\mathbf{0}) \end{aligned} \quad (2.6)$$

where

$$\Delta_0(\mathbf{k}, \mathbf{q}) = \omega_0(\mathbf{q}) + \sigma(\mathbf{q}) D(\mathbf{k}, \mathbf{q}).$$

Transforming this solution into an ordinary correlation function the transverse static correlation function is found to be

$$\begin{aligned} \gamma_{\mathbf{k}, -\mathbf{k}}^{\pm-} &= \langle S_{\mathbf{k}}^+ S_{-\mathbf{k}}^- \rangle \\ &= \frac{N\sigma(\boldsymbol{\pi}) \Delta_0(\mathbf{k}-\boldsymbol{\pi}, \boldsymbol{\pi})}{\sqrt{\Delta_0(\mathbf{k}-\boldsymbol{\pi}, \boldsymbol{\pi}) \Delta_0(\mathbf{k}, \boldsymbol{\pi})}} \coth \frac{1}{2k_B T} \sqrt{\Delta_0(\mathbf{k}-\boldsymbol{\pi}, \boldsymbol{\pi}) \Delta_0(\mathbf{k}, \boldsymbol{\pi})} \end{aligned}$$

$$\text{for } \mathbf{q} = \boldsymbol{\pi}, \tag{2.7}$$

$$= 2N\sigma(\mathbf{0}) / \{1 - \exp(-\Delta_0(\mathbf{k}, \mathbf{0})/k_B T)\}$$

$$\text{for } \mathbf{q} = \mathbf{0}. \tag{2.8}$$

In the same way, the other transverse static correlation function can be easily obtained as

$$\gamma_{\mathbf{k}, -\mathbf{k}}^+, -\mathbf{k} = \gamma_{\mathbf{k}, -\mathbf{k}}^+, -\mathbf{k} \quad \text{for } \mathbf{q} = \boldsymbol{\pi}, \tag{2.9}$$

$$= \gamma_{\mathbf{k}, -\mathbf{k}}^+, -\mathbf{k} - 2\sigma(\mathbf{0})N \quad \text{for } \mathbf{q} = \mathbf{0}. \tag{2.10}$$

In the paramagnetic region  $\sigma(\mathbf{q})/k_B T$  is small enough compared with unity, so that

$$\gamma_{\mathbf{k}, -\mathbf{k}}^+, -\mathbf{k} = \gamma_{\mathbf{k}, -\mathbf{k}}^+, -\mathbf{k} = Nk_B T \frac{2\sigma(\mathbf{q})}{\Delta_0(\mathbf{k}, \mathbf{q})}, \tag{2.11}$$

under the external wavy field characterized by the vector  $\mathbf{q}$ .

### 2.2) Transition temperature

In order to get the transition temperature, the sum rule for  $S = \frac{1}{2}$ <sup>18)</sup>

$$\frac{1}{N} \sum_{\mathbf{k}} (\gamma_{\mathbf{k}, -\mathbf{k}}^+, -\mathbf{k} + \gamma_{\mathbf{k}, -\mathbf{k}}^-, +\mathbf{k}) = 1 \tag{2.12}$$

is now invoked. Inserting Eq. (2.11) into the above relation, the transition temperature  $T_C$  (or  $T_N$ ) is determined by requiring that the polarization  $\sigma(\mathbf{0})$  (or  $\sigma(\boldsymbol{\pi})$ ) is nonvanishing when the applied field  $\omega_0(\mathbf{0})$  (or  $\omega_0(\boldsymbol{\pi})$ ) on the ferromagnet (or antiferromagnet) is reduced to zero.

$$\begin{aligned} |J(0)| (1 + \eta) / 4k_B T_N &= |J(0)| (1 + \eta) / 4k_B T_C, \\ &= \frac{1}{N} \sum_{\mathbf{k}} \frac{1}{1 - ((1 - \alpha) / (1 + \eta)) (\cos k_z + \eta \cos k_x)}. \end{aligned} \tag{2.13}$$

If the summation in Eq. (2.13) may be replaced by a corresponding integration, the following formula is easily obtained,

$$|J(0)| (1 + \eta) / 4k_B T_C = \frac{\xi_1 - \xi_2}{\sqrt{(1 - \xi_1^2)g(\xi_1)}} \frac{1}{\nu} \frac{2}{\pi} F(\kappa), \tag{2.14}$$

where

$$\rho = (1 - \alpha) / (1 + \eta),$$

$$\xi_{1,2} = [\{1 - (1 - \eta^2)\rho^2\} \pm \sqrt{\{1 - (1 - \eta^2)\rho^2\}^2 - 4\eta^2\rho^2}] / 2\eta\rho,$$

$$g(\xi) = (\eta\rho\xi - 1)^2 - \rho^2,$$

$$\nu^2 = -g(\xi_2) / g(\xi_1), \quad \mu^2 = (1 - \xi_2^2) / (1 - \xi_1^2), \quad \kappa^2 = \mu^2 / \nu^2,$$

and  $F(\kappa)$  is the elliptic integral of the first kind.<sup>21a)</sup> This is a fundamental

relation between the transition temperature and the parameters  $\alpha$  and  $\eta$ .

Let us now look into several extreme cases with respect to the value of parameters. In the linear system ( $\eta=0$ ), Eq. (2.14) is rewritten as

$$|J(0)|/4k_B T_c = 1/\sqrt{1-(1-\alpha)^2}. \quad (2.15)$$

When the exchange interaction becomes isotropic, i.e.  $\alpha=0$ , this expression is consistent with the theorem that a linear spin system has no critical point.<sup>11)</sup> In the case of nonvanishing  $\alpha$ , however, according to the present formula, the linear spin system exhibits a transition point. The transition temperature is proportional to the square root of  $\alpha$  in the vicinity of  $\alpha=0$  and it is the same as that of the molecular field theory in the Ising limit  $\alpha=1$ .

In the planar system with  $\eta=1$ , the transition temperature is obtained in the form

$$|J(0)|/2k_B T_c = (4/(1-\alpha))\sqrt{\omega(\alpha)} \cdot (2/\pi)F(\omega(\alpha)), \quad (2.16)$$

where

$$\omega(\alpha) = (1 - \sqrt{1 - (1 - \alpha)^2}) / (1 + \sqrt{1 - (1 - \alpha)^2}).$$

The transition temperature tends to zero as the anisotropic parameter  $\alpha$  is decreased, and it vanishes for  $\alpha=0$ . It becomes

$$|J(0)|/2k_B T_c = 1 + \omega^2(\alpha)/4, \quad (2.17)$$

in the nearly Ising case ( $\alpha=1$ ).

The more quantitative relation between the transition temperature and parameters  $\alpha$  and  $\eta$  is shown in Fig. 1.

### 2.3) The spin polarization

The paramagnetic spin polarization is now discussed in the presence of a uniform external field. From Eqs. (2.11) and (2.12) the following equation can be obtained:

$$1/2 = \sigma(\mathbf{0}) \frac{1}{N} \sum_{\mathbf{k}} \coth \left[ \frac{|J(0)|}{2k_B T} (1+\eta) \left\{ \frac{\omega_0(\mathbf{0})}{|J(0)|(1+\eta)} \pm \sigma(\mathbf{0}) \left( 1 - \frac{1-\alpha}{1+\eta} (\cos k_z + \eta \cos k_x) \right) \right\} \right], \quad (2.18)$$

where the upper sign refers to a ferromagnet and the lower sign to an antiferromagnet. The argument of  $\coth$  is nothing but the product of  $(|J(0)|/2k_B T) \times (1+\eta)$  and the excitation energy from the ground state, which should not be negative as far as only stable solutions are concerned. Therefore the condition  $\sigma(\mathbf{0}) \leq (1/(2+\alpha))(\omega_0(\mathbf{0})/|J(0)|(1+\eta))$  is derived for the antiferromagnetic spin polarization,<sup>\*</sup> under which Eq. (2.18) determines the spin polarization uniquely

<sup>\*</sup>) In the ferromagnetic cases, the argument of  $\coth$  is always positive. Therefore there is no condition for spin polarization.

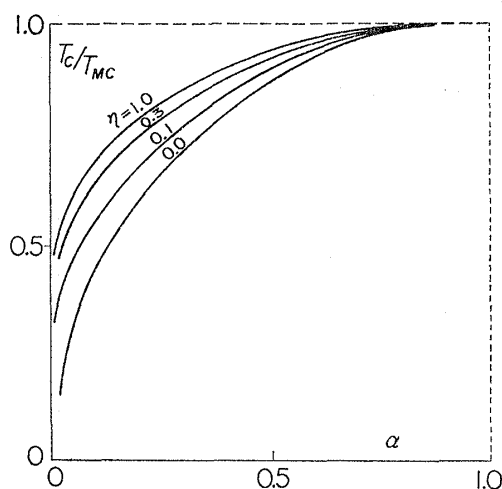


Fig. 1. The transition temperature as a function of  $\alpha$  and  $\eta$  in the plane Heisenberg magnet.  $T_{MC}$  denotes the transition temperature  $|J(0)|(1+\eta)/4k_B$ , given by the molecular field theory.

$$|J(0)|(1+\eta)/4k_B T$$

$$= \frac{1}{N} \sum_{\mathbf{k}} \frac{1}{1/|J(0)|(1+\eta)\chi(T) \pm \{1 - ((1-\alpha)/(1+\eta))(\cos k_x + \eta \cos k_y)\}}, \quad (2.20)$$

where the upper sign refers to ferromagnet and the lower sign to an antiferromagnet. A useful relation is derived from the above equation, i.e.

$$1/(|J(0)|(1+\eta)\chi_f(T)) = 1/(|J(0)|(1+\eta)\chi_a(T)) - 2, \quad (2.21)$$

where,  $\chi_f(T)$  and  $\chi_a(T)$  are the ferro- and antiferromagnetic susceptibilities, respectively.

Replacing the summation by the corresponding integration, we can easily calculate the right-hand side of Eq. (2.20)

$$|J(0)|(1+\eta)/4k_B T = \frac{\xi_1 - \xi_2}{\sqrt{(1-\xi_1^2)g(\xi_1)}} \frac{1}{\nu} \frac{2}{\pi} F(\kappa), \quad (2.22)$$

where  $\xi_1$ ,  $\xi_2$ , and  $g(\xi)$  are redefined as follows:

\*) For example, we investigate a linear spin system with  $\alpha=0$  at absolute 0°K. Replacing the summation in Eq. (2.18) by the corresponding integration, the following results are obtained.

A ferromagnetic linear chain system has no spontaneous spin polarization. On the other hand, for an antiferromagnet,  $\sigma(0)=1/2$  for  $\omega_0(0)/|J(0)| \geq 1$ , and  $\sigma(0) = (\omega_0(0)/|J(0)|)(1 + \sin \pi/4\sigma(0))$  for  $\omega_0(0)/|J(0)| < 1$ . There is an infinite series of solutions, namely, for  $\omega_0(0)=0$ , the relation  $\sigma(0) = 1/6n$  ( $n=1, 2, \dots, \infty$ ) satisfies Eq. (2.18), but from the above condition, only  $\sigma(0)=0$  must be regarded as a real solution.

as a function of temperature and field.\*)

Equation (2.18) must be solved numerically except a few limiting cases mentioned in the following. When  $\omega_0(\mathbf{0})/k_B T \gg 1$ , the spin polarization is generally given by

$$\sigma(\mathbf{0}) = \frac{1}{2} \tanh(\omega_0(\mathbf{0})/2k_B T), \quad (2.19)$$

which is exactly the same formula<sup>22)</sup> as that found for an independent spin system. In the opposite limit, i.e.  $\omega_0(\mathbf{0})/k_B T \ll 1$ , the spin polarization is proportional to  $\omega_0(\mathbf{0})/k_B T$ .

#### 2.4) The paramagnetic susceptibility

Using Eq. (2.18) and the relation  $\chi(T) = \lim_{\omega_0(\mathbf{0}) \rightarrow 0} (\sigma(\mathbf{0})/\omega_0(\mathbf{0}))$ , the paramagnetic susceptibility  $\chi(T)$  is determined from the following equation:

$$\xi_{1,2} = [K_0^2(T) - (1-\eta^2)\rho^2 \pm \sqrt{(K_0^2(T) - (1-\eta^2)\rho^2)^2 - 4\eta^2\rho^2 K_0(T)}] / 2\eta\rho K_0(T),$$

$$g(\xi) = (\eta\rho\xi - K_0(T))^2 - \rho^2$$

and

$$K_0(T) = 1 + 1/|J(0)|(1+\eta)\chi_f(T). \tag{2.23}$$

After a straightforward calculation the Curie-Weiss formula is found in the high temperature limit,

$$\chi_{a,f} = \frac{C}{T \pm \theta}, \tag{2.24}$$

where

$$C = 1/4k_B$$

and

$$\theta = |J(0)|(1+\eta)4k_B.$$

If we replace  $K_0(T)$  by unity, Eq. (2.22) coincides with Eq. (2.14) which determines the transition temperature. Therefore, at the transition point, the susceptibility for an antiferromagnet  $|J(0)|(1+\eta)\chi_a(T)$  is one half and for a ferromagnet  $\chi_f(T)$  is divergent. Near the critical point, the paramagnetic susceptibility decreases in proportion to  $(T-T_c)$ . Its coefficient is  $(4k_B/|J(0)|) \times \sqrt{1-(1-\alpha)^2}$  for both linear and planar systems.\*)

The  $\eta$  and the temperature dependences of the susceptibilities with various values of the anisotropic parameter are shown in Fig. 2 for the antiferromagnet.

It is interesting to compare the present results with those of the molecular

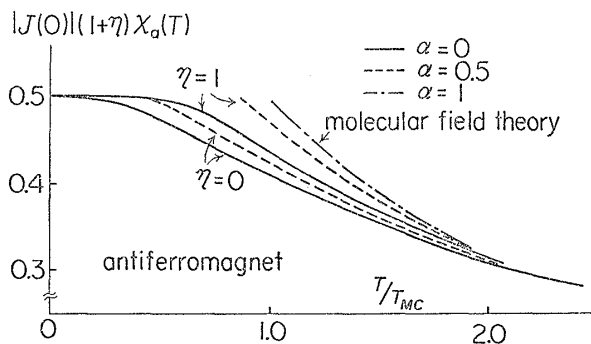


Fig. 2. Antiferromagnetic susceptibility versus the temperature  $T/T_{MC}$ . The solid lines represent the case of  $\alpha=0$  and the broken lines  $\alpha=0.5$ .

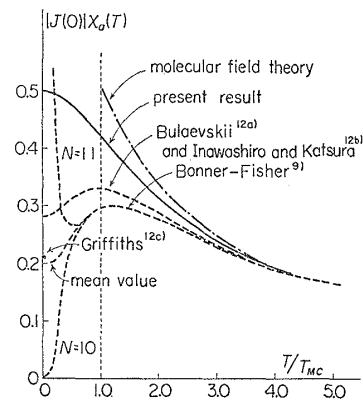


Fig. 3. Antiferromagnetic susceptibilities of an isotropic linear chain in various theories.

\*) If  $\alpha=0$ , the paramagnetic susceptibility decreases in proportion to  $(T-T_c)^2$ .

field theory and other previous theories.<sup>9),12a)~12c)</sup> As to the linear Heisenberg spin system this comparison is made in Fig. 3.

2.5) Short range order

Although there is no long range order in the paramagnetic region, the short range order is persisting near the transition temperature. The behaviour of this short range order is investigated on the basis of the sum rule indicating the total spin conservation Eq. (2.12).

For the sake of simplicity, let us confine ourselves to the case of isotropic Heisenberg ferromagnet ( $\alpha=0$ ) and  $\eta$  is zero or unity. Using the long wave approximation, Eq. (2.12) is rewritten as follows:

$$1 = \frac{4k_B T}{|J(0)|(1+\eta)} \frac{c(d)}{\pi^d} \int_0^\pi \frac{k^{d-1}}{K_0(T) - 1 + k^2/2d} dk, \tag{2.25}$$

$$= \frac{4k_B T}{|J(0)|(1+\eta)} \frac{c(d)}{\pi^d} \int_0^\pi n(k) dk,$$

where  $n(k)$  indicates the number density with the  $k$ -mode, 'd' the dimension of a system and  $c(d)$  the integration of the angular part.

At high temperature,  $n(k)$  slowly varies with  $k$ , while near the transition temperature, as shown in Fig. 4, it has a fairly sharp peak at a certain value of wave number denoted by  $k_M$ . When the temperature reaches the transition point, only the uniform mode may exist.

It is easy to calculate the value of  $k_M$  in one- and two-dimensional systems near the critical point, i.e.

$$k_M = \begin{cases} 0 & \text{for the one-dimension,} \\ \sim (T - T_C) & \text{for the two-dimension.}^*) \end{cases} \tag{2.26}$$

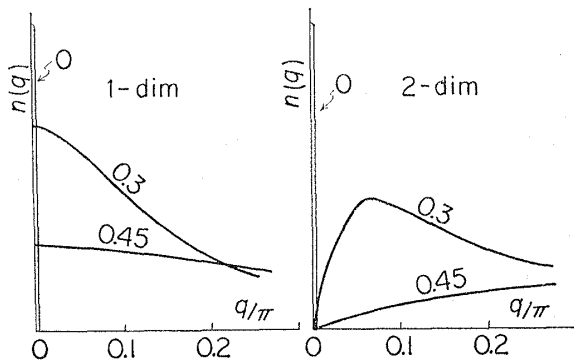


Fig. 4. The number density  $n(q)$  of spins with  $q$ -mode. The numerals on the curve indicate the temperature  $4k_B T/|J(0)|(1+\eta)$ . At the absolute zero degree only the uniform mode ( $q=0$ ) is allowed.

The reciprocal value of  $k_M$  may be a rough measure of the size of the short range order within which the spin arrangement is considered spatially uniform.<sup>14)</sup> In other words, the short range order is more developed when the temperature is lowered or the number of dimensionality is smaller. The same discussion can be applied to the antiferromagnet and their  $k_M$ 's are identical with those of ferromagnet if  $k$  is regarded as a deviation from its Bragg point.

\*) When  $\alpha \neq 0$ ,  $k_M \cong [(4/(1-\alpha)) \{ \alpha + (2k_B/|J(0)|) \sqrt{1-(1-\alpha)^2} (T-T_C) \}]^{1/2}$  for the two-dimension.



### § 3. Damping constant

Now, let us discuss the damping constant in one- and two-dimensional spin systems in the paramagnetic region. According to  $T$ - $T^{15)}$  (see Appendix I), the general expression of the damping constant is given by

$$\Gamma(\mathbf{k}, \omega) = \sqrt{\frac{\pi}{2}} \frac{1}{N} \sum_{\mathbf{q}} \left\{ \frac{a^{(2)}(\mathbf{k}, \mathbf{q})}{b(\mathbf{k}, \mathbf{q})} + \frac{c^{(3)}(\mathbf{k}, \mathbf{q}) (\omega - \Delta_0(\mathbf{q}, \mathbf{0}))}{b^{(2)}(\mathbf{k}, \mathbf{q})} \right\} \\ \times \exp \left\{ -\frac{(\omega - \Delta_0(\mathbf{q}, \mathbf{0}))^2}{2b^{(2)}(\mathbf{k}, \mathbf{q})} \right\}, \quad (3.1)$$

where the notations  $a^{(2)}(\mathbf{k}, \mathbf{q})$ ,  $b^{(2)}(\mathbf{k}, \mathbf{q})$  and  $c^{(3)}(\mathbf{k}, \mathbf{q})$  are the same as those listed in Table III of the paper  $T$ - $T$  except that  $D(\mathbf{q}, \mathbf{q}')$  should be replaced by Eq. (2.3) and  $C(\mathbf{q}, \mathbf{q}')$  should be defined by the following formula:

$$C(\mathbf{q}, \mathbf{q}') = \frac{1}{2}(1 - \alpha) J(0) \{ (\cos q_z + \eta \cos q_x) - (\cos q'_z + \eta \cos q'_x) \}. \quad (3.2)$$

On the right-hand side of Eq. (3.1), the term involving  $c^{(3)}(\mathbf{k}, \mathbf{q})$  is neglected, for the quantity  $c^{(3)}(\mathbf{k}, \mathbf{q})$  involves the three-spin correlation, which is much smaller than two-spin correlation at least in the paramagnetic phase. In the usual case the wavelength of an applied field is long enough compared with the sample dimension. Therefore, the special case  $\mathbf{k} = \mathbf{0}$  corresponds to the response of a system.

In the case of the plane spin systems with anisotropic exchange coupling, the damping constant of resonance absorption is rewritten in terms of the generalized susceptibilities<sup>17)</sup>

$$\Gamma(\mathbf{0}, 0) = \gamma_{\perp}(\mathbf{0}) \frac{1}{N} \sum_{\mathbf{q}} \varphi(\mathbf{q}), \quad (3.3)$$

where

$$\varphi(\mathbf{q}) = \sqrt{\frac{4k_B T}{|J(0)|(1+\eta)}} \left( \sqrt{\frac{\gamma_{\perp}(\mathbf{q})}{\gamma_{\parallel}(\mathbf{q})}} - \sqrt{\frac{\gamma_{\parallel}(\mathbf{q})}{\gamma_{\perp}(\mathbf{q})}} \right)^2 \left/ \left[ \gamma_{\perp}(\mathbf{q}) \frac{1}{N} \sum_{\mathbf{q}'} \left( \sqrt{\frac{\gamma_{\perp}(\mathbf{q}')}{\gamma_{\parallel}(\mathbf{q}-\mathbf{q}')}} \right. \right. \right. \\ \left. \left. \left. - \sqrt{\frac{\gamma_{\parallel}(\mathbf{q}-\mathbf{q}')}{\gamma_{\perp}(\mathbf{q}')}} \right)^2 + \gamma_{\parallel}(\mathbf{q}) \frac{1}{N} \sum_{\mathbf{q}'} \left( \sqrt{\frac{\gamma_{\perp}(\mathbf{q}')}{\gamma_{\parallel}(\mathbf{q}+\mathbf{q}')}} - \frac{\gamma_{\parallel}(\mathbf{q}+\mathbf{q}')}{\gamma_{\perp}(\mathbf{q}')} \right)^2 \right], \quad (3.4)$$

$$\gamma_{\perp}(\mathbf{q}) = K_0(T) \mp \frac{1-\alpha}{1+\eta} (\cos q_z + \eta \cos q_x)$$

and

$$\gamma_{\parallel}(\mathbf{q}) = K_0(T) \mp \frac{1}{1+\eta} (\cos q_z + \eta \cos q_x), \quad (3.5)$$

the upper sign refers to a ferromagnet and the lower one refers to an antiferromagnet. As was pointed out by Tomita,<sup>17)</sup> the damping constant consists of two factors, i.e. the configuration disparity  $\gamma_{\perp}(\mathbf{0})$  and the torque imbalance

$\sum_{\mathbf{q}} \varphi(\mathbf{q})$ . In the high temperature limit, the damping constant is of common to ferro- and antiferro magnetic spin systems, i.e.

$$\Gamma_{\infty}(\mathbf{0}, 0) = \sqrt{\frac{\pi}{2}} \frac{1}{N} \sum_{\mathbf{q}} \frac{D^2(\mathbf{q}, -\mathbf{q})}{\sqrt{\sum_{\mathbf{q}'} \{D^2(\mathbf{q}', \mathbf{q}-\mathbf{q}') + 4C^2(\mathbf{q}', \mathbf{q}-\mathbf{q}')\}}/N}. \quad (3.6)$$

This is essentially proportional to the square of anisotropic parameter. The temperature dependence of the damping constant will be discussed in more details in the following section.

3.1) *Linear spin systems* ( $\eta=0$ )

Replacing the summations in Eq. (3.3) by the corresponding integral, the damping constant is easily obtained as follows:

$$\begin{aligned} \Gamma(0, 0) &= \alpha |J(0)| \gamma_{\perp}(0) H(\lambda_1, \lambda_2; T) / (\sqrt{P(T)} \cdot \tau) \\ &\times \left[ \left\{ \frac{\alpha}{1-\alpha} \mp K_0(T) \left( \frac{1}{\delta_1^0} - \frac{1}{(1-\alpha)^2 \delta_1^{\alpha}} \right) \right\} \frac{2}{\pi} F(\kappa^2) \right. \\ &\left. + K_0(T) (\lambda_1 - \lambda_2) \left\{ \zeta_0 / (\delta_1^0)^2 \frac{2}{\pi} \Pi(\nu_0^2, \kappa^2) - \zeta_{\alpha} / \{(1-\alpha) \delta_1^{\alpha}\}^2 \frac{2}{\pi} \Pi(\nu_{\alpha}^2, \kappa^2) \right\} \right], \end{aligned} \quad (3.7)$$

where  $\Pi(\nu^2, \kappa^2)$  is the elliptic integral of the third kind,<sup>21a)</sup> and the upper sign refers to a ferromagnet and the lower ones to an antiferromagnet. The deriva-

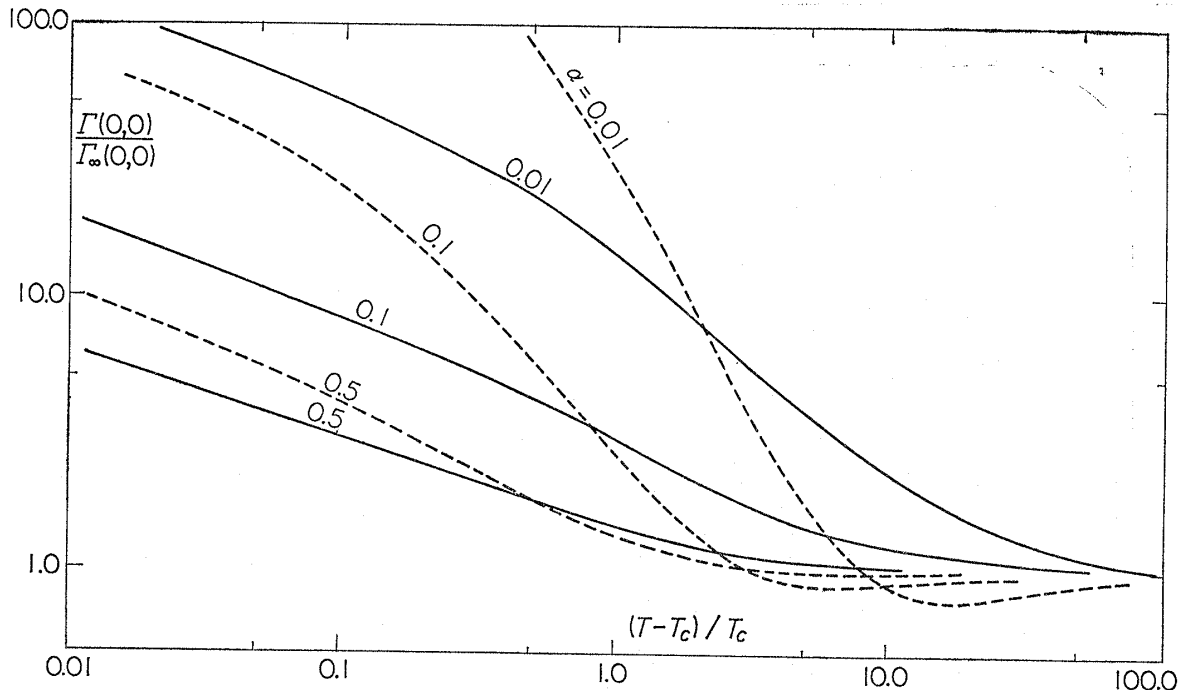


Fig. 5. The damping constant of a linear magnetic chain with various anisotropy. The notation  $\Gamma_{\infty}(0, 0)$  indicates that in the high temperature limit. The solid lines represent the ferromagnet and the broken lines the antiferromagnet,

tion of the above equation and various notations used in it are described in Appendix II. The temperature dependence of the damping constant for linear spin systems with various values of anisotropic parameter was computed by using Eq. (3.7) and the results are shown in Fig. 5. The normalized damping constant  $\Gamma(0, 0)/\Gamma_\infty(0, 0)$  of the ferromagnet grows monotonically with decreasing temperature, while in the antiferromagnet it has a minimum value in some cases and then becomes increasing toward the transition temperature. If the parameter  $\alpha$  approaches to unity, the difference between damping constants of ferromagnet and antiferromagnet disappears. In the neighbourhood of the transition temperature, the long wave approximation should be valid because the terms with the smaller  $q$  becomes dominant. In this approximation the damping constants are easily calculated as (see Appendix III)

$$\Gamma(0, 0) \cong \alpha |J(0)| / \sqrt[4]{\Delta T} \quad \text{for a ferromagnet}$$

and

$$(3.8)$$

$$\cong \sqrt{\alpha(2-\alpha)} |J(0)| / \sqrt[4]{\Delta T} \quad \text{for an antiferromagnet,}$$

where

$$\Delta T = (T - T_c) / T_c. \quad (3.9)$$

When the temperature approaches the critical point, the torque fluctuation increases in proportion to  $(T - T_c)^{-1/4}$  both in ferro- and antiferromagnets and  $\gamma_\perp(0)$  becomes  $\alpha$  for the former and  $2 - \alpha$  for the latter.

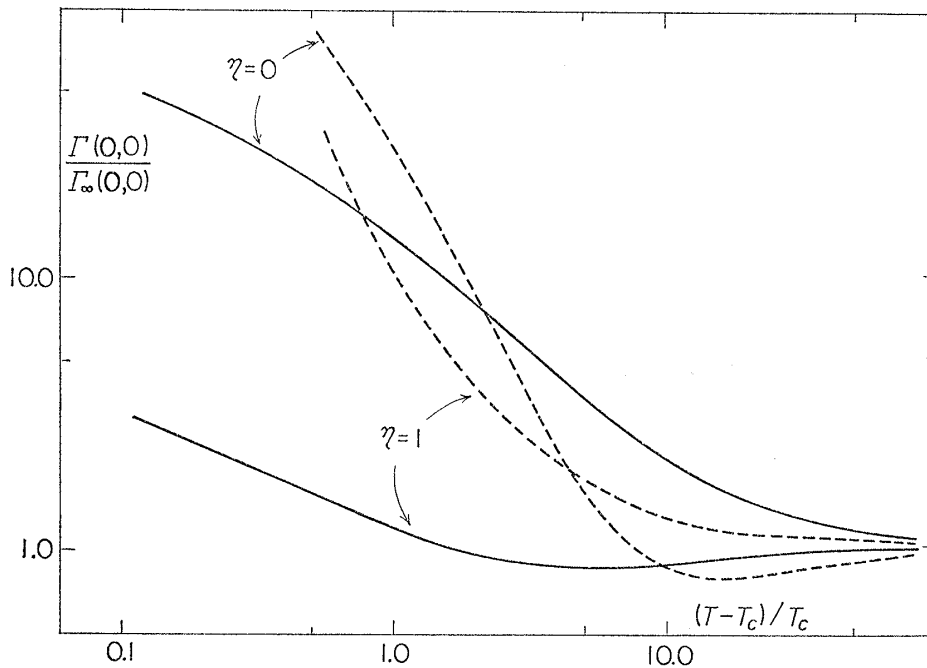


Fig. 6. The  $\eta$ -dependence of the damping constant. The solid lines represent the ferromagnet and broken lines the antiferromagnet. The numerals on the curve indicate the value of the parameter  $\eta$ .

3.2) Plane spin systems

Since the  $q$ -integration in Eq. (3.3) is analytically impossible in this case, it was computed numerically. There seems to be little difference in the  $\alpha$  dependence of the line width between the linear and planar spin systems. It is rather instructive to note how the line width is affected by the magnitude of the interaction between chains. The  $\eta$  dependence of the damping constant is shown in Fig. 6 for the system with  $\alpha=0.01$ .

As is easily seen, the characteristic behaviours of the line width of the linear chain are weakened by increasing  $\eta$ . That is, when  $\eta$  becomes unity, the damping constant of the ferromagnet shows the thermodynamical slowing down in the range  $T \cong 2T_c$  and then grows up below  $2T_c$ . The antiferromagnets have no dip at any temperature and increase monotonically with decreasing temperature as is well known in the three-dimensional case.<sup>17),23)</sup>

3.3) Physical situation

It is interesting to compare the present results with those of the three-dimensional spin systems. In the usual ferromagnet with a uniaxial anisotropy, the resonance line width shows thermodynamical slowing down except in the neighbourhood of the critical temperature while in the antiferromagnet it grows monotonically with decreasing temperature. In the one-dimensional systems with an anisotropic exchange, as was described in the previous sections, the line widths of both ferro- and antiferromagnet show different features. This is essentially due to the dimensionality of the system for the following two reasons. First, it is shown that the line width caused by the uniaxial anisotropy also has the same tendency as that caused by the anisotropic exchange, therefore difference does not seem to arise from the character of the interaction. Secondly, when the system is changed into a plane system from a linear chain, the damping constant approaches gradually to that of the three-dimensional case.

In order to show how the damping constant depends on temperature, the contribution of two factors; i.e. the configuration disparity  $\gamma_{\perp}(0)$  and the torque

imbalance  $\sum_q \varphi(q)/N$ , to the damping constant is investigated in the one-dimensional spin system with small  $\alpha (=0.01)$ .

In Fig. 7, their relations are shown. Although the quantity  $\gamma_{\perp}(0)$  is common to both magnets in the high temperature limit, it is reduced with decreasing temperature and has limiting values  $\alpha$  and  $2-\alpha$  for the ferromagnet and antiferromagnet, respectively, at the transition temperature. Therefore, the temperature dependence of configuration dispa-

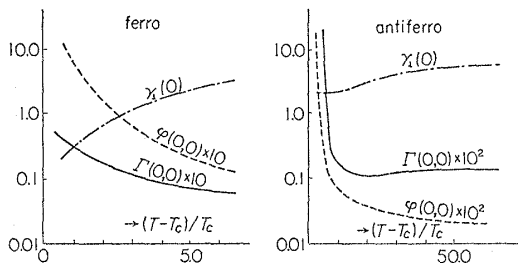


Fig. 7. The contribution of the configuration disparity  $\gamma_{\perp}(0)$  and the torque disparity  $\sum_q \varphi(q)/N$  to the damping constant of a linear chain with  $\alpha=0.01$ .

rity is more evident in the former case than in the latter unless  $\alpha$  is nearly equal to unity. On the other hand, the torque imbalance for both magnets increases rapidly with lowering temperature and the rate of change with temperature is larger than  $\gamma_{\perp}(0)$  in the neighbourhood of the transition temperature. Therefore, in one-dimensional system the line width increases with decreasing temperature. The thermodynamical slowing down can be seen only in the linear antiferromagnet with small  $\alpha$  above a certain temperature. In this temperature range the torque imbalance of an antiferromagnet is smaller than that of the ferromagnet while the quantities  $\gamma_{\perp}(0)$  are almost same value for both system, i.e. the torque imbalance plays a dominant role to show anomalous behaviours of the damping constants of the one dimensional spin system.

The remaining points are why the torque imbalance increases more rapidly with decreasing temperature and why it is larger in a ferromagnet than in an antiferromagnet. By using Eq. (2.26), torque imbalance will be rewritten in the following form,

$$\sqrt{\frac{4k_B T}{|J(0)|(1+\eta)}} \frac{1}{N} \sum_{\mathbf{q}} \varphi(\mathbf{q}) \rightarrow \frac{1}{\pi^a} \int (n(\mathbf{q})f(\mathbf{q})) (n(-\mathbf{q})f(-\mathbf{q})) d\mathbf{q},$$

where  $f(\mathbf{q})$  is considered to be the torque acted on the  $\mathbf{q}$ -mode of spin moment. In the case of a linear chain with the small  $\alpha$ ,  $f(q)$  is approximated from Eq. (3.4),

$$f(q) \cong \pm \alpha \cos q / \sqrt{\pm P(T) \cos^2 q - Q(T) \cos q + R(T)},$$

where the upper sign refer to the ferromagnet and the lower one to the antiferromagnet. The  $q$ -dependences of  $f(q)$  in ferro- and antiferromagnets are shown in Figs. 8. Near the transition temperature, their characters are quite different from each other, i.e.  $f(q)$  has a maximum at each Bragg point of the magnet; i.e. at  $q=0$  in the ferromagnet and  $q=\pi$  in the antiferromagnet, its value being

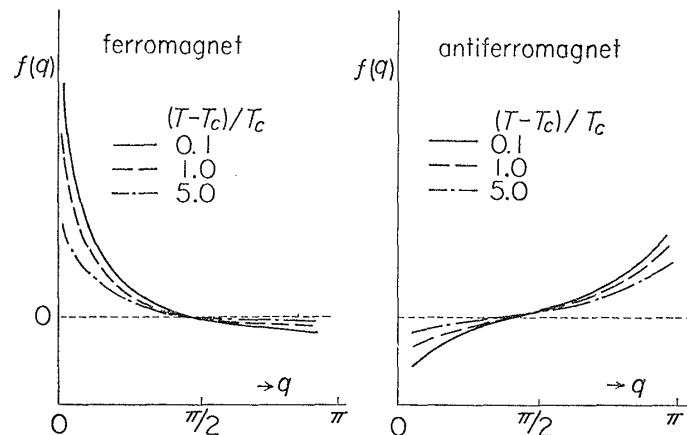


Fig. 8. The torque  $f(q)$  acted upon the spin moment with  $q$ -mode in the case of a linear chain with  $\alpha=0.01$ ,

smaller in the latter than in the former. This tendency may be attributed to the following facts: we are now concerned with the resonance line width under the constant external field ( $\omega_0(0) < |J(0)|$ ). In addition to the internal field due to the exchange coupling, the spin system is affected by this external one. In a ferromagnet, these two fields will cooperate to make the system uniform, while the opposite situation occurs in an antiferromagnet, namely the effect of the internal field makes the spin antiparallel and is reduced by the external one. Of course these characters will gradually decrease as the thermal agitation increases. Furthermore, it is easily seen that the number density  $n(q)$  for the ferro- and antiferromagnets reflect each other about  $q = \pi/2$ . Therefore the total torque  $(1/\pi) \int n(q) f(q) dq$  and its correlation are always larger in the ferromagnet than in the antiferromagnet and increase with decreasing temperature.

When the dimensionality of the system becomes larger, as is shown in Fig. 4, the  $q$ -value contributed dominantly to  $n(q)$  shifts to the larger (smaller)  $q$ -mode in the ferromagnet (antiferromagnet), and the short range order effect is reduced. Since the  $q$ -dependence of fluctuation has the same tendency as that of a linear chain, the important  $q$ -values in each of  $n(q)$  and  $f(q)$  are different and separated. The torque fluctuation is then reduced as compared with one-dimensional case. Moreover the thermodynamical slowing down in the plane antiferromagnetic damping constant is not remarkable because the quantity  $\gamma_{\perp}(0)$  varies more slowly with temperature than that of a linear chain. Under these circumstances, the characteristic behaviours of linear chain are made weakened.

Finally let us consider the  $\alpha$ -dependence of the damping constant. When  $\alpha$  increases, the torque  $f(q)$  becomes large while the number density  $n(q)$  has a more broadened shape in momentum space. Therefore the effect of the short range order is reduced. Consequently, although the damping constant itself becomes large with increasing  $\alpha$ , the rate of change of the damping constant  $\Gamma(\mathbf{0}, 0)/\Gamma_{\infty}(\mathbf{0}, 0)$  with temperature becomes gradually small and the difference between ferro- and antiferromagnet vanishes in the limit  $\alpha \rightarrow 1$ .

#### § 4. Line shape of resonance absorption

In order to see the dynamical behaviour in more detail, it is necessary to study the line shape of the resonance absorption as well as the damping constant. The line shape function is derived according to the paper T-T<sup>15)</sup>

$$I(\omega) = \frac{2\sigma(\mathbf{0})\Gamma(\mathbf{0}, \omega)}{[\omega - \Delta(\mathbf{0}, \omega)]^2 + [\Gamma(\mathbf{0}, \omega)]^2}, \quad (4.1)$$

where  $\Gamma(\mathbf{0}, \omega)$  and  $\Delta(\mathbf{0}, \omega)$  stand for the frequency dependent damping constant and its shift respectively. Namely

$$\Gamma(\mathbf{0}, \omega) = \sqrt{\frac{\pi}{2}} \frac{1}{N} \sum_{\mathbf{q}} \frac{a^{(2)}(\mathbf{0}, \mathbf{q})}{b(\mathbf{0}, \mathbf{q})} \exp(-\Omega^2/2b^2(\mathbf{0}, \mathbf{q})), \quad (4.2)$$

$$\Delta(\mathbf{0}, \omega) = \sqrt{2} \frac{1}{N} \sum_{\mathbf{q}} \frac{a^{(2)}(\mathbf{0}, \mathbf{q})}{b(\mathbf{0}, \mathbf{q})} \exp(-\Omega^2/2b^2(\mathbf{0}, \mathbf{q})) \int_0^{\Omega/\sqrt{2}b(\mathbf{0}, \mathbf{q})} \exp(y^2) dy \quad (4.3)$$

and

$$\Omega = \omega - \Delta(\mathbf{0}, 0). \quad (4.4)$$

The central intensity ( $\Omega=0$ ) is given by

$$I(\Delta) = \sigma(\mathbf{0})/\Gamma(\mathbf{0}, 0) \quad (4.5)$$

which is a measure of the line shape. The temperature dependence and  $\alpha$ - and  $\eta$ -dependence of  $I(0)$  are shown in Fig. 9 for the linear chain. The intensity curves have a maximum at a certain temperature. In the high temperature,  $\sigma(0)$  is an infinitesimal small and  $\Gamma(0, 0)$  becomes constant owing to the thermal agitation. Both the spin polarization  $\sigma(0)$  and the damping constant  $\Gamma(0, 0)$  increase with the decreasing temperature as was seen from previous sections. Near the transition temperature, the former has an upper limit (saturated polarization) while the latter diverges because of the critical fluctuation. Therefore there may be a possibility of the existence of a hump in the temperature dependence of the central intensity.

Next, the line shapes are studied by numerical analysis and are shown in Fig. 10 for the linear chain with various anisotropy. Paramagnetic resonance line shape has been conventionally considered to be a Lorentzian or Gaussian.<sup>24)</sup> The present result, however, shows that side peaks appear with decreasing temperature. It becomes more remarka-

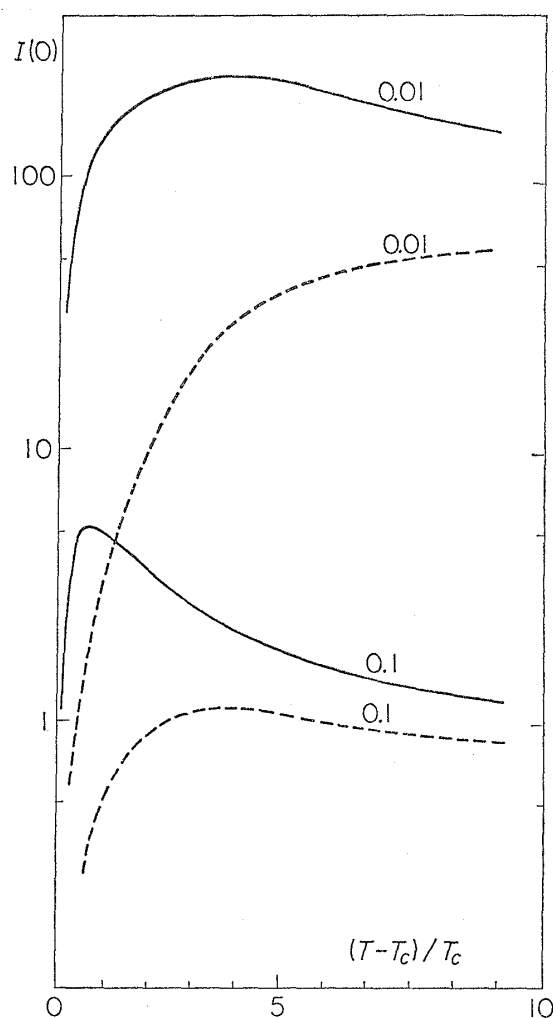


Fig. 9. The intensity of the magnetic resonance absorption for the linear chain with respect to the reduced temperature  $(T-T_c)/T_c$ . The numerals on the curve indicate the degree of anisotropy  $\alpha$ . The solid lines correspond to the ferromagnet and the broken lines to the antiferromagnet,

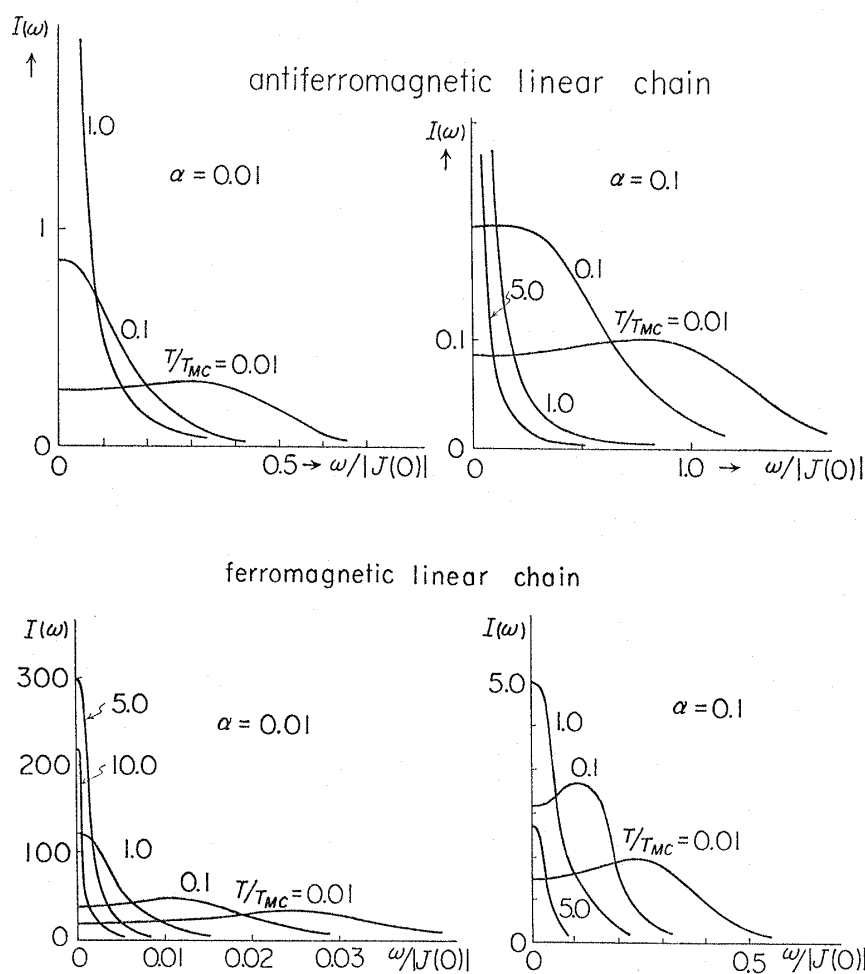


Fig. 10. The line shapes of the magnetic resonance absorption for the linear magnetic chains with various anisotropy  $\alpha$ .

ble with increase of anisotropy. Near the transition temperature, fluctuations are fairly developed while spins will arrange almost spatially uniform in the ferromagnet or alternatively in the antiferromagnet according to the anomalously developed short range order. In this situation the quasi-collective mode<sup>17)</sup> may be seen and this will correspond to the separate peaks.

Finally, the experimental data are examined. There are several samples which are considered to be nearly linear magnetic chain. The damping constant for  $\text{Cu}(\text{C}_6\text{H}_5\text{COO})_2 \cdot 3\text{H}_2\text{O}$ <sup>3)</sup> has a minimum at a certain temperature if the constant field is applied in a suitable direction. It grows monotonically in the antiferromagnet  $\text{Cu}(\text{NH}_3)_4\text{SO}_4 \cdot \text{H}_2\text{O}$ <sup>2)</sup> with decreasing temperature. These behaviours may be explained qualitatively within the present theory (see Figs. 5 and 6). A ferromagnetic chain system is found along  $c$ -axis in  $\text{CoCl}_2 \cdot 2\text{H}_2\text{O}$ . The concept of the cluster resonance<sup>4)</sup> for an Ising spin is introduced to explain the anomaly of the intensity for the resonance absorption. However, it seems more appro-



priate to treat this matter as a Heisenberg spin chain with a strong anisotropy. From our point of view, the facts that the absorption intensity decreases with lowering temperature and that double peaks appear in this substance may be due to the anomalous development of the short range order in this system.

### § 5. Discussion

Although it is an advantage of the Green's function method that both static and dynamic quantities can be treated within the same theoretical framework, several approximations have been invoked in practice. The most important approximations used in this paper are the following. First, higher order Green's functions were decoupled into a sum of products of lower order Green's functions and the static average. The truncation of higher order Green's function leads to the result that the local fluctuation of spins will be more or less smeared out. Secondly, in order to derive the damping constant, the torque correlation was assumed to be Gaussian in its time dependence. The assumption of the Gaussian decay will be reasonable in the high frequency limit and be enough to give an overall line spread. However, there is a possibility of improving this assumption when the line shape is under discussion. Thirdly, the static correlation functions, in terms of which the damping constant is expressed, were calculated at the lowest step of the equation of motion, though the third step is, at least, necessary in derivation of the general expression of the damping constant. That is, each quantity was obtained in the lowest approximation respectively. However, the present treatment may involve self-inconsistency between the static and dynamic quantities.

In spite of these approximations, the present treatment succeeded in explaining the unusual character of the lower dimensional spin systems; when the anisotropy is small the transition temperature becomes much lower than that given by molecular field theory. The effect of the short range order is the most remarkable in the linear chain. Owing to the anomalous growth of fluctuations the effect of the thermodynamical slowing down will be upset and the line width will increase in both ferro- and antiferromagnet with the lowering of temperature. Line shapes are also calculated, which have broad shoulders in the neighbourhood of the critical temperature. Their peak frequencies are determined by the anisotropy and the temperature as was described in the previous section. Therefore, their peaks may be concerned with the quasi-collective modes of spin motion which persists in the paramagnetic phase.<sup>17)</sup> These facts become evident in the study of inelastic neutron scattering.<sup>25)</sup>

In order to discuss quantitatively the anomalous behaviours of magnetic properties near the transition temperature, a more accurate treatment will be needed.

The numerical integration was performed by using the so-called Gaussian quadrature method.<sup>21b)</sup> The number of division is 96 within the range of the

wave vector  $[0, \pi]$ . It turned out that this number is large enough to be accurate when compared with analytical results in the case of a linear chain.

### Acknowledgements

The author would like to express her sincere gratitude to Professor K. Tomita for his suggestion of the problem and for his valuable advice and illuminating discussions. She also thanks the members of her laboratory for many critical discussions. Thanks are also due to Prof. S. Katsura for his helpful comments and valuable advice.

### Appendix I

#### *Summary of Tomita and Tanaka's theory<sup>15)</sup>*

In the derivation of the general expression of the damping constant, the following approximations were used in their paper.

(i) The higher order Green's functions which were generated in the equation of motion of the Green's function, were decomposed into those of the lower order ones, i.e.

$$G^{\mu_1\mu_0}(\mathbf{k}_1|\mathbf{k}_0; \omega) = G_c^{\mu_1\mu_2}(\mathbf{k}_1|\mathbf{k}_0; \omega),$$

$$G^{\mu_1\mu_2\mu_0}(\mathbf{k}_1, \mathbf{k}_2|\mathbf{k}_0; \omega) = \langle S_{\mathbf{k}_1}^{\mu_1} \rangle G_c^{\mu_2\mu_0}(\mathbf{k}_2|\mathbf{k}_0; \omega) + \langle S_{\mathbf{k}_2}^{\mu_2} \rangle G_c^{\mu_1\mu_0}(\mathbf{k}_1|\mathbf{k}_0; \omega) \\ + G_c^{\mu_1\mu_2\mu_0}(\mathbf{k}_1, \mathbf{k}_2|\mathbf{k}_0; \omega),$$

$$G^{\mu_1\mu_2\mu_3\mu_0}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3|\mathbf{k}_0; \omega) = \sum_{\text{cyclic}} \langle S_{\mathbf{k}_1}^{\mu_1} S_{\mathbf{k}_2}^{\mu_2} \rangle G_c^{\mu_3\mu_0}(\mathbf{k}_3|\mathbf{k}_0; \omega) \\ + \sum_{\text{cyclic}} \langle S_{\mathbf{k}_1}^{\mu_1} \rangle G_c^{\mu_2\mu_3\mu_0}(\mathbf{k}_2, \mathbf{k}_3|\mathbf{k}_0; \omega) + G_c^{\mu_1\mu_2\mu_3\mu_0}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3|\mathbf{k}_0; \omega),$$

where the cumulant-type Green's functions,  $G_c^{\mu_1\mu_2\mu_0}(\mathbf{k}_1, \mathbf{k}_2|\mathbf{k}_0; \omega)$ , etc., were introduced as a remainder of the decomposition process.

(ii) In the paramagnetic range, the hierarchy of the cumulant-type Green's functions must be accounted for at least the third step. At first step, a renormalized spectrum was found. A correction to spin wave spectrum was given at the next step. While it was shown not to be sufficient for the discussion of the damping in the paramagnetic range, the correct second moment of the distribution or the correct initial curvature of the higher order relaxation function is expressed at this stage. Therefore third step may be termed as a stochastic introduction of damping. Consequently, the relaxation function was assumed to be the Gaussian with the initial curvature which was evaluated at the second step.

(iii) After the above procedures, the damping constant was given in terms of the Fourier transforms of the spatial correlations. In order to obtain the damping constant definitely, the correlation functions were calculated by means of the lowest order approximation.

## Appendix II

## Derivation of Eq. (3.7)

Let us calculate the damping constant for the linear spin system by putting  $\eta=0$  in Eq. (3.3). In this case, the damping constant is given by

$$\begin{aligned} \Gamma(0, 0) &= \alpha |J(0)| \gamma_{\perp}(0) \sqrt{4k_B T/J(0)} \frac{1}{\pi} \int_0^{\pi} (1/\gamma_{\parallel}(q) - 1/\gamma_{\perp}(q)) \\ &\quad \times \frac{\cos q}{\sqrt{\pm P(T) \cos^2 q - Q(T) \cos q + R(T)}} dq, \\ &= \alpha |J(0)| \gamma_{\perp}(0) \sqrt{4k_B T/J(0)} I(T), \end{aligned} \quad (\text{II}\cdot 1)$$

where

$$\begin{aligned} I(T) &= \frac{1}{\pi} \int_0^{\pi} (1/\gamma_{\parallel}(q) - 1/\gamma_{\perp}(q)) \frac{\cos q}{\sqrt{\pm P(T) \cos^2 q - Q(T) \cos q + R(T)}} dq, \\ P(T) &= 3U_{\alpha}(T) + (1-\alpha)^2 U_0(T), \\ Q(T) &= \left\{ \left( \frac{1}{1-\alpha} + 2 \right) K_0(T) + (3-\alpha) \right\} U_{\alpha}(T) + (1-\alpha) (K_0(T) + 1) U_0(T), \\ R(T) &= K_0(T) (3U_{\alpha}(T) + U_0(T)), \\ U_{\alpha}(T) &= \frac{K_0(T)}{\sqrt{K_0(T) - (1-\alpha)^2}} - 1. \end{aligned} \quad (\text{II}\cdot 2)$$

Hereafter the upper sign refer to the ferromagnet and lower one to the antiferromagnet. If we set  $x = \cos q$ , from (II.1) we obtain

$$\begin{aligned} I(T) &= \frac{1}{\pi} \int_{-1}^1 dx \left\{ \frac{\alpha}{1-\alpha} \mp K_0(T) \left( \frac{1}{x \mp K_0(T)} - \frac{1}{(1-\alpha)^2 (x \mp K_0(T)/(1-\alpha))} \right) \right\} \\ &\quad \times \frac{1}{\sqrt{(1-x^2) (P(T)x^2 - Q(T)x + R(T))}}. \end{aligned} \quad (\text{II}\cdot 3)$$

After the following replacement  $x = (\lambda_1 t + \lambda_2)/(t+1)$ , it is found that

$$\begin{aligned} I(T) &= (\lambda_1 - \lambda_2) \left\{ \frac{\alpha}{1-\alpha} \mp K_0(T) \left( \frac{1}{\delta_1^0} \right. \right. \\ &\quad - \frac{1}{(1-\alpha)^2 \delta_1^1} \frac{1}{\pi} \int dt \frac{1}{\sqrt{(g(\lambda_1)t^2 + g(\lambda_2))(h(\lambda_1)t^2 + h(\lambda_2))}} \\ &\quad + K_0(T) (\lambda_1 - \lambda_2) \frac{1}{\pi} \int dt \left( \frac{\gamma_0}{(\delta_1^0)^2 (t^2 - \gamma_0^2)} - \frac{\gamma_{\alpha}}{\{(1-\alpha)\delta_1^{\alpha}\}^2 (t^2 - \gamma_{\alpha}^2)} \right) \\ &\quad \left. \left. \times \frac{1}{\sqrt{(g(\lambda_1)t^2 + g(\lambda_2))(h(\lambda_1)t^2 + h(\lambda_2))}} \right\}, \end{aligned} \quad (\text{II}\cdot 4)$$

where

$$\begin{aligned} \lambda_{1,2} &= \{R(T) + 1 \pm \sqrt{(R(T) + 1)^2 - Q^2(T)}\} / Q(T), \\ \delta_i^\alpha &= \lambda_i - K_0(T) / (1 - \alpha), \quad \gamma_\alpha = \delta_2^\alpha / \delta_1^\alpha, \\ g(\lambda) &= 1 - \lambda^2 \quad \text{and} \quad h(\lambda) = P(T)\lambda^2 - Q(T)\lambda + R(T). \end{aligned} \tag{II.5}$$

Using the following new variables:

$$\mu^2 = |g(\lambda_2) / g(\lambda_1)|, \quad \nu^2 = |h(\lambda_2) / h(\lambda_1)|,$$

the integrand of the first term of Eq. (II.4) is rewritten as

$$\begin{aligned} &\frac{1}{\sqrt{(g(\lambda_1)t^2 + g(\lambda_2))(h(\lambda_1)t^2 + h(\lambda_2))}} = \frac{1}{\sqrt{|g(\lambda_1)h(\lambda_1)|}} \\ &\times \begin{cases} \frac{1}{\sqrt{(\mu^2 - t^2)(t^2 + \nu^2)}}, & Q^2(T) \leq 4R(T), \\ \frac{1}{\sqrt{(\mu^2 - t^2)(t^2 - \nu^2)}}, & Q^2(T) > 4R(T), \end{cases} \end{aligned}$$

in the ferromagnet and

$$\frac{1}{\sqrt{(g(\lambda_1)t^2 + g(\lambda_2))(h(\lambda_1)t^2 + h(\lambda_2))}} = \frac{1}{\sqrt{|g(\lambda_1)h(\lambda_1)|}} \frac{1}{\sqrt{(t^2 - \mu^2)(t^2 - \nu^2)}}$$

in the antiferromagnet respectively. Therefore, the damping constants are represented by the elliptic integrals of the first and third kinds and can be easily calculated. Then Eq. (3.7) results where

$$H(\lambda_1, \lambda_2; T) = (\lambda_1 - \lambda_2) / \sqrt{|g(\lambda_1)h(\lambda_1)|},$$

and the following notations are used.

Table I.

	Ferromagnet for $4R(T) < Q(T)$ and Antiferromagnet	Ferromagnet for $4R(T) \geq Q(T)$
$\tau^2$	$\nu^2$	$\mu^2 + \nu^2$
$\kappa^2$	$\mu^2 / \nu^2$	$\mu^2 / (\mu^2 + \nu^2)$
$\zeta_\alpha$	$\gamma_\alpha / (\gamma_\alpha^2 - \mu^2)$	$1 / \gamma_\alpha$
$\nu_\alpha^2$	$\mu^2 / \gamma_\alpha^2$	$\mu^2 / (\gamma_\alpha^2 - \mu^2)$

### Appendix III

#### Derivation of Eq. (3.8)

In order to see the damping constant in the vicinity of the transition point, the long wave approximation is used. Then it is found that in the ferromagnet

$$\Gamma(0, 0) = \alpha |J(0)| \gamma_\perp(0) \frac{4k_B T / J(0)}{\sqrt{P(T) - Q(T) + R(T)}} \frac{1}{\pi}$$

$$\times \int_0^\pi dq \left\{ \frac{1}{A_0(T) + q^2} - \frac{1}{1 - \alpha} \frac{1}{A_\alpha(T) + q^2} \right\} \frac{2 - q^2}{\sqrt{1 - B(T)q^2}}, \quad (\text{III} \cdot 1)$$

where

$$A_\alpha(T) = 2(K_0(T) - (1 - \alpha)) / (1 - \alpha)$$

and

$$B(T) = (P(T) - Q(T)/2) / (P(T) - Q(T) + R(T)).$$

Since  $A_0(T)$  becomes extremely small in proportion to  $(T - T_c)$  near the transition temperature

$$P(T) \cong (1 - \alpha)^2 \frac{1}{\sqrt{\Delta T}}, \quad Q(T) \cong 2(1 - \alpha) \frac{1}{\sqrt{\Delta T}}$$

and

$$R(T) \cong \frac{1}{\sqrt{\Delta T}},$$

where

$$\Delta T = 2 \cdot (4k_B/J(0)) \cdot \sqrt{1 - (1 - \alpha)^2} (T - T_c).$$

Using above relations,  $\Gamma(0, 0)$  is easily calculated as follows:

$$\begin{aligned} \Gamma(0, 0) &= \alpha |J(0)|^4 \sqrt{\Delta T} \left[ \frac{1}{1 - \alpha} \frac{1}{\pi} \sqrt{\frac{\alpha}{1 - \alpha}} \sinh^{-1} \left( \sqrt{\frac{1 - \alpha}{\alpha}} \pi \right) + \frac{1}{\sqrt{\Delta T}} \right. \\ &\quad \left. - \frac{1}{\pi} \sqrt{\frac{1 - \alpha}{2\alpha}} \log \left| 1 + \frac{2\sqrt{2}[\pi^2 + \alpha/(1 - \alpha)]}{\pi - \sqrt{2}[\pi^2 + \alpha/(1 - \alpha)]} \right| \right] \sqrt{T_c} \\ &\cong \frac{\alpha}{2} |J(0)|^4 \frac{1}{\sqrt{\Delta T}} \sqrt{T_c}. \end{aligned} \quad (\text{III} \cdot 2)$$

In the antiferromagnet, we also find that

$$\begin{aligned} \Gamma(0, 0) &= \sqrt{\alpha(2 - \alpha)} |J(0)|^4 \sqrt{\Delta T} \left[ \frac{\alpha}{(1 - \alpha)^2} \sqrt{\alpha(2 - \alpha)} \frac{1}{\pi} \sinh^{-1} \left( \frac{1 - \alpha}{\sqrt{\alpha(2 - \alpha)}} \pi \right) \right. \\ &\quad \left. + \frac{1}{\sqrt{\Delta T}} + \sqrt{\frac{2}{1 - \alpha}} \frac{1}{\alpha^{3/2}} \tan^{-1} \left( \sqrt{\frac{1 - \alpha}{2\alpha}} \pi \sqrt{\frac{\alpha^2/(1 - \alpha)^2}{\pi^2 + \alpha(2 - \alpha)/(1 - \alpha)^2}} \right) \right] \\ &\cong \sqrt{\alpha(2 - \alpha)} |J(0)|^4 \frac{\sqrt{T_c}}{\sqrt{\Delta T}}. \end{aligned} \quad (\text{III} \cdot 3)$$

From Eqs. (III·2) and (III·3) the damping constants are divergent toward the transition temperature both in ferro and antiferromagnets.

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