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Dynamic Critical Phenomena in Magnetic Systems. I

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In order to deal with the *long-time behavior* of the time correlations of spins and take into account the *life-time effects* of all critical variables involved, we formulate a generalized continued fraction expansion of the time correlation functions. It is shown that, if the correlation length of spin fluctuations κ^{-1} and the wave-length of external disturbance κ^{-1} are very long, then the long range correlations of spin fluctuations involved yield the most dominant part in the limit of *long times* or *small frequencies.*

The asymptotic behavior of the most dominant part is determined by the equal-time correlations of long wave-length spin fluctuations which are treated with the static scaling laws. It is shown that, in the case of $\eta=0$, η being the parameter measuring the deviation of the spin pair correlation from the Ornstein-Zernike form, the dynamic scaling law proposed by Halperin and Hohenberg holds with the characteristic frequencies of the form $\kappa^{\theta}g(k/\kappa)$, where $\theta=5/2$ in ferromagnets and 3/2 in antiferromagnets. In the case of $\eta\neq 0$, however, the dynamic scaling law does not hold. In ferromagnets, this is due to a non-similarity between the longitudinal and transverse components in the ordered phase, and in antiferromagnets, this is due to a non-similarity between the critical slowing-down of the staggered polarization and the kinematical slowing-down of the small wave-number polarization. In ferromagnets in the paramagnetic region, however, there exists a characteristic frequency with $\theta=(5+\eta)/2$. These results are derived by first using the pair correlation approximation and then removing such an approximation.

§ 1. Introduction

In the vicinity of the critical point there appear enormous fluctuations of macroscopic scale. Critical phenomena,[1),2)] both static and dynamic, are believed to be due to these anomalous fluctuations. Dynamic critical phenomena which we know at present may be classified into the following types: (A) the critical scattering of light and neutron, (B) the critical slowing-down in return to equilibrium, (C) the anomalous increase in transport and relaxation coefficients and in their temperature derivatives, (D) the existence of diffuse oscillatory modes even in the magnetic disordered phases.

The fundamental processes underlying these critical phenomena would be (1) the critical fluctuation of critical variables involved, (2) their critical slowing-down in decay, (3) the kinematical slowing-down of conserved quantities involved, and (4) the memory effects. All the variables which show the critical fluctuation should show the critical slowing-down in decay, since a large fluctuation is difficult to occur unless its return to equilibrium is slow. Thus in the vicinity of the critical point, microscopic processes associated become very slow so that the microscopic time can be of the same order of magnitude as the macroscopic relaxation time. Then the memory effect becomes important, and could lead to a new type of motion which largely differs from the macroscopic motion. In the present paper, we attempt to formulate a systematic theory of dynamic critical phenomena, which enables us to study how these fundamental processes give rise to anomalous phenomena in the vicinity of the critical point.

The critical fluctuation of a usual critical variable is equivalent to the appearance of a long range correlation. The theory of the static scaling laws is based on the assumption of the existence of a unique correlation length ξ which becomes infinite at the critical point.[3] Let us take the Heisenberg model and denote the Fourier components of the spin operator by

$$S_{\boldsymbol{k}}^{\alpha}=\sum_{j=1}^{N} S_j^{\alpha} \exp[i\boldsymbol{k}\cdot\boldsymbol{r}_j],\ (\alpha=0,\ \pm), \tag{1·1}$$

where

$$S_j^0 \equiv S_j^z,\ S_j^{\pm} \equiv (S_j^x \pm iS_j^y)/\sqrt{2}\,. \tag{1·2}$$

Then the static scaling laws would imply in the case of isotropic ferromagnets that the pair correlation functions of spins are homogeneous functions of k and $\kappa(\equiv 1/\xi)$,

$$\langle S_{\boldsymbol{k}}^{\alpha} S_{\boldsymbol{k}}^{\alpha *}\rangle/N=\kappa^{-2+\eta} f_{\alpha}(k/\kappa), \tag{1·3}$$

if the values of k and κ are very small compared to the inverse range of interaction between spins.

If the system has an axial symmetry about the z axis, the linear dynamic responses of magnetization can be described in terms of the relaxation functions[4),5)]

$$(S_{\boldsymbol{k}}^{\alpha}(t),\ S_{\boldsymbol{k}}^{\alpha *}) \equiv \frac{1}{\beta}\int_0^{\beta} \langle \exp(\lambda \mathcal{H}) S_{\boldsymbol{k}}^{\alpha}(t) \exp(-\lambda \mathcal{H}) S_{\boldsymbol{k}}^{\alpha *}\rangle d\lambda, \tag{1·4}$$

where $\mathcal{H}$ is the hamiltonian of the system, and β the inverse temperature $1/k_B T$. The λ integral arises from the noncommutativity of $S_{\boldsymbol{k}}^{\alpha}$ and $\mathcal{H}$, which may be neglected for small values of k in the vicinity of the critical point. The dynamic scaling law, proposed by Ferrell et al.[6)] and by Halperin and Hohenberg,[7)] would amount to assuming that the time-correlation functions are functions of the form

$$(S_{\boldsymbol{k}}^{\alpha}(t),\ S_{\boldsymbol{k}}^{\alpha *})/N=\kappa^{-2+\eta} F_{\alpha}(z_k t,\ k/\kappa) \tag{1·5}$$

and that the characteristic imaginary frequency z_k is a homogeneous function of k and κ, being of the form

$$z_k=\kappa^{\theta} g_{\alpha}(k/\kappa)\,. \tag{1·6}$$

Equation (1·5) is a generalization of (1·3). It is the crucial point to assume that the critical index θ is constant irrespective of the lower or upper critical region, and of the transverse or longitudinal component.[7)] For example, it is assumed that the frequency and damping constant of spin waves have the same value of θ, and the diffusitivity of spins (the damping constant of the longitudinal component in the hydrodynamic regime) also has the identical value of θ. It is thus quite interesting to assume that the frequency spectrum and the damping constant should obey the same law with respect to the k and κ dependence. Such a law cannot be seen in the usual examples of collective motion, such as the spin waves in the magnon region (where κ represents the average wave number of thermal magnons). Thus the dynamic scaling law casts a challenging problem on the statistical mechanics of irreversible processes.

It would be the most fundamental problem to clarify whether and how the long range correlations of spin fluctuations involved determine dynamic critical phenomena as the most dominant part. Properties like the dynamic scaling law are believed to hold for the asymptotic behavior of such most dominant part. Such a separation of the most dominant part would be possible by taking into account the *life-time effects* of all critical variables involved, and thus by dealing with their *long time behavior* in the limit of long times or small frequencies. The moment method and its modifications which are short time expansions cannot deal with either of these two correctly. The simple continued fraction expansion cannot deal with the life-time effects correctly. Therefore we first develop a generalized continued fraction expansion of the time correlation functions with the aid of the theory of generalized Brownian motion presented by one of the authors.[8]

With the aid of this expansion we study the dynamic scaling law and collective motion in the vicinity of the critical point. Preliminary results have been reported elsewhere.[9] This formulation can be modified also to study anomalous transport phenomena, such as anomalous sound attenuation and ESR line width. Thus this is the first of a series of papers, presenting a systematic theory of dynamic critical phenomena.

§ 2. Generalized continued fractions

We consider the Heisenberg model whose hamiltonian is given by

$$\mathcal{H} = -\sum_{j\neq l}\sum J_{jl}[S_j^0 S_l^0 + (1-\lambda)(S_j^+ S_l^- + S_j^- S_l^+)] - \hbar\omega_0 \sum_j S_j^0 , \qquad (2\cdot1)$$

where ω_0 denotes the Zeeman frequency $g\mu_B H$, and λ is an anisotropy parameter, $\lambda=0$ leading to the isotropic case. Rewriting (2·1) in terms of the Fourier components and using the commutation relations

$$[S_k^+, S_{k'}^-] = S_{k+k'}^0, \quad [S_k^0, S_{k'}^\pm] = \pm S_{k+k'}^\pm , \qquad (2\cdot2)$$

we obtain

$$\begin{aligned} \dot{S}_k^0 &\equiv iLS_k^0 = [2(1-\lambda)/i\hbar N]\sum_q J_q^k S_q^+ S_{k-q}^- , \\ \dot{S}_k^\pm &\equiv iLS_k^\pm = \mp i\omega_0 S_k^\pm \pm (2/i\hbar N)\sum_q J_q^k(\lambda) S_q^0 S_{k-q}^\pm , \end{aligned} \qquad (2\cdot3)$$

where L is the Liouville operator, LF denoting the commutator $[\mathcal{H}, F]/\hbar$, and

$$J_{\boldsymbol{q}}^{\boldsymbol{k}} \equiv J_{\boldsymbol{q}}^{\boldsymbol{k}}(0), \quad J_{\boldsymbol{q}}^{\boldsymbol{k}}(\lambda) \equiv J(\boldsymbol{q}) - (1-\lambda) J(\boldsymbol{k}-\boldsymbol{q}),$$
$$J(\boldsymbol{q}) \equiv \sum_{j(\neq l)} J_{jl} \exp(i\boldsymbol{q}\cdot(\boldsymbol{r}_j - \boldsymbol{r}_l)) = J(-\boldsymbol{q}), \tag{2·4}$$

and the inversion symmetry of the crystal lattice has been assumed.

In order to study the time evolution of the relaxation functions, we define the normalized relaxation functions

$$\Xi_{\boldsymbol{k}}^{\alpha}(t) \equiv (S_{\boldsymbol{k}}^{\alpha}(t), S_{\boldsymbol{k}}^{\alpha *}) / (S_{\boldsymbol{k}}^{\alpha}, S_{\boldsymbol{k}}^{\alpha *}) \tag{2·5}$$

and introduce its Laplace transform

$$\Xi_{\boldsymbol{k}}^{\alpha}(z) \equiv \int_0^\infty \Xi_{\boldsymbol{k}}^{\alpha}(t) \exp(-zt)\, dt\,. \tag{2·6}$$

Then, using the theory of generalized Brownian motion,[8] we obtain

$$\Xi_{\boldsymbol{k}}^{\alpha}(z) = \frac{1}{z - i\omega_k^{\alpha} + \varphi_{\boldsymbol{k}}^{\alpha}(z)}, \tag{2·7}$$

where

$$\omega_k^{\alpha} \equiv (\dot{S}_{\boldsymbol{k}}^{\alpha}, S_{\boldsymbol{k}}^{\alpha *}) / i(S_{\boldsymbol{k}}^{\alpha}, S_{\boldsymbol{k}}^{\alpha *}), \tag{2·8}$$

$$\varphi_{\boldsymbol{k}}^{\alpha}(z) \equiv (f_{\boldsymbol{k}}^{\alpha}(z), f_{\boldsymbol{k}}^{\alpha *}) / (S_{\boldsymbol{k}}^{\alpha}, S_{\boldsymbol{k}}^{\alpha *}). \tag{2·9}$$

The random forces $f_{\boldsymbol{k}}^{\alpha}(t)$ are given by

$$f_{\boldsymbol{k}}^{\alpha}(t) = \exp[(1-\mathscr{P}_{\boldsymbol{k}}^{\alpha}) iLt](1-\mathscr{P}_{\boldsymbol{k}}^{\alpha}) \dot{S}_{\boldsymbol{k}}^{\alpha}, \tag{2·10}$$

where $\mathscr{P}_{\boldsymbol{k}}^{\alpha}$ is the projection operator onto $S_{\boldsymbol{k}}^{\alpha}$,

$$\mathscr{P}_{\boldsymbol{k}}^{\alpha} F \equiv [(F, S_{\boldsymbol{k}}^{\alpha *}) / (S_{\boldsymbol{k}}^{\alpha}, S_{\boldsymbol{k}}^{\alpha *})] S_{\boldsymbol{k}}^{\alpha}. \tag{2·11}$$

The time evolution of the random forces is governed by the operator $(1-\mathscr{P}_{\boldsymbol{k}}^{\alpha}) iL$ which differs from the mechanical one iL. This difference was the crucial point in the theory of generalized Brownian motion, and enabled us to define the correlation time τ of the random forces which distinctly differs from the macroscopic relaxation time $\tau_r \equiv 1/\mathrm{Re}\, \varphi_{\boldsymbol{k}}^{\alpha}(i\omega_{\boldsymbol{k}}^{\alpha})$.[8] It will turn out in our model (2·1) that if $k \ll \kappa$ and $\lambda \ll 1$, then $\tau \ll \tau_r$. Thus in the hydrodynamic regime where $|z|\tau \ll 1$, we have

$$\Xi_{\boldsymbol{k}}^{\alpha}(z) \simeq \frac{1}{z - z_k}, \tag{2·12}$$

$$z_k = i\omega_k^{\alpha} - \varphi_{\boldsymbol{k}}^{\alpha}(i\omega_k^{\alpha}). \tag{2·13}$$

The imaginary part of z_k gives us the frequency of the collective motion of spins, such as the spin wave frequency and the ESR resonance frequency, and the real part leads to their damping and the diffusivity of spins. In the very vicinity of the critical point, however, we could have $\tau \sim \tau_r$ if $k \gg \kappa$. Then the memory effect, namely, the z dependence of $\varphi_{\boldsymbol{k}}^{\alpha}(z)$ becomes important, and the approximation (2·12) would break down.

In order to determine whether the relaxation functions (2·7) satisfy the dynamic scaling law (1·5) or not, and also to study whether its characteristic imaginary frequency, for instance (2·13), has the form (1·6) or not, we have to go into the structure of the damping function $\varphi_k{}^\alpha(z)$ in more detail. Insertion of (2·3) into (2·10) leads to the form

$$f_k{}^\alpha(t)=\sum_q g_q{}^{\alpha k}A_q{}^{\alpha k}(t), \tag{2·14}$$

where

$$\left.\begin{aligned}A_q{}^{0k}&\equiv(1-\mathcal{P}_k{}^0)S_q{}^+S_{k-q}^-,\\ A_q{}^{\pm k}&\equiv(1-\mathcal{P}_k{}^\pm)S_q{}^0S_{k-q}^\pm,\end{aligned}\right\} \tag{2·15}$$

$$\left.\begin{aligned}g_q{}^{0k}&\equiv[2(1-\lambda)/i\hbar N]J_q{}^k,\\ g_q{}^{\pm k}&\equiv\pm(2/i\hbar N)J_q{}^k(\lambda).\end{aligned}\right\} \tag{2·16}$$

It should be noted that $A_q{}^{\alpha k}(t)$ is orthogonal to $S_k{}^\alpha$ and its time evolution is governed by the unusual operator $L_k{}^\alpha\equiv(1-\mathcal{P}_k{}^\alpha)L$. Inserting (2·14) into (2·9), however, we can write as

$$\varphi_k{}^\alpha(z)=\frac{1}{(S_k{}^\alpha, S_k{}^{\alpha *})}\sum_q\sum_{q'}g_q{}^{\alpha k}g_{q'}^{\alpha k*}\frac{(A_q{}^{\alpha k}, A_{q'}^{\alpha k*})}{z-i\omega_{qq'}^{\alpha k}+\varphi_{qq'}^{\alpha k}(z)} \tag{2·17}$$

with the aid of the following theorems.

[**Theorem 1**] Consider a quantity $A(t)$ whose equation of motion has the form

$$\frac{d}{dt}A(t)=i\mathcal{L}A(t), \tag{2·18}$$

where $\mathcal{L}$ is a linear operator. Then, for an arbitrary quantity B, we have

$$(A(z), B^*)/(A, B^*)=\frac{1}{z-i\omega_{AB}+\varphi_{AB}(z)}, \tag{2·19}$$

where

$$\omega_{AB}\equiv(\mathcal{L}A, B^*)/(A, B^*), \tag{2·20}$$

$$\varphi_{AB}(z)\equiv-(i\mathcal{L}f_A(z), B^*)/(A, B^*). \tag{2·21}$$

The random force $f_A(z)$ is defined by

$$f_A(t)\equiv\exp[(1-\mathcal{P}_A)i\mathcal{L}t](1-\mathcal{P}_A)i\mathcal{L}A \tag{2·22}$$

with the use of the projection operator $\mathcal{P}_A$,

$$\mathcal{P}_AF\equiv[(F, B^*)/(A, B^*)]A. \tag{2·23}$$

[**Theorem 2**] Define the hermite conjugate $\mathcal{L}'$ of $\mathcal{L}$ by

$$(f_A,\ [\mathcal{L}'B]^*) = (\mathcal{L}f_A,\ B^*). \tag{2·24}$$

Then, introducing the hermite conjugate of $\mathcal{P}_A$,

$$\mathcal{P}_B G \equiv [(G,\ A^*)/(B,\ A^*)]B, \tag{2·25}$$

we obtain

$$\varphi_{AB}(t) = (f_A(t), f_B{}^*)/(A,\ B^*), \tag{2·26}$$

$$= (f_A, f_B{}^*(-t))/(A,\ B^*), \tag{2·27}$$

where

$$f_B(t) \equiv \exp[(1-\mathcal{P}_B)i\mathcal{L}'t](1-\mathcal{P}_B)i\mathcal{L}'B. \tag{2·28}$$

[**Theorem 3**] In accordance with the propagators of $f_A(t)$ and $f_B(t)$, define the linear operators

$$\mathcal{L}_1 \equiv (1-\mathcal{P}_A)\mathcal{L}, \quad \mathcal{L}_1{}' \equiv (1-\mathcal{P}_B)\mathcal{L}'. \tag{2·29}$$

If $\mathcal{P}_A F = \mathcal{P}_B G = 0$, then we have

$$(\mathcal{L}_1 F,\ G^*) = (F,\ [\mathcal{L}_1{}'G]^*). \tag{2·30}$$

Namely, $\mathcal{L}_1$ and $\mathcal{L}_1{}'$ area hermite conjugate to each other.

These theorems are results of a straightforward generalization of the damping theory developed in the theory of generalized Brownian motion, and are proved in Appendix A. Equation (2·7) comes out from (2·19) by taking that $A=B=S_k{}^\alpha$ and $\mathcal{L}=\mathcal{L}'=L$. Since the evolution operator of $A_q{}^{\alpha k}(t)$,

$$L_k{}^\alpha \equiv (1-\mathcal{P}_k{}^\alpha)L, \tag{2·31}$$

is linear, (2·17) is derived by applying (2·19) and (2·26) to $(A_q{}^{\alpha k}(z),\ A_{q'}^{\alpha k*})$. Therefore, we have

$$\omega_{qq'}^{\alpha k} = (L_k{}^\alpha A_q{}^{\alpha k},\ A_{q'}^{\alpha k*})/(A_q{}^{\alpha k},\ A_{q'}^{\alpha k*}), \tag{2·32}$$

$$\varphi_{qq'}^{\alpha k}(z) = (f_q{}^{\alpha k}(z), f_{q'}^{\alpha k*})/(A_q{}^{\alpha k},\ A_{q'}^{\alpha k*}). \tag{2·33}$$

Equation (2·22) gives us

$$f_q{}^{\alpha k}(t) = \exp[iL_q{}^{\alpha k}t]iL_q{}^{\alpha k}A_q{}^{\alpha k}, \tag{2·34}$$

where

$$L_q{}^{\alpha k} \equiv (1-\mathcal{P}_q{}^{\alpha k})L_k{}^\alpha = (1-\mathcal{P}_q{}^{\alpha k}-\mathcal{P}_k{}^\alpha)L, \tag{2·35}$$

$$\mathcal{P}_q{}^{\alpha k}F \equiv [(F,\ A_{q'}^{\alpha k*})/(A_q{}^{\alpha k},\ A_{q'}^{\alpha k*})]A_q{}^{\alpha k}. \tag{2·36}$$

In deriving (2·35), we have used that $\mathcal{P}_q{}^{\alpha k}\mathcal{P}_k{}^\alpha = \mathcal{P}_k{}^\alpha\mathcal{P}_q{}^{\alpha k} = 0$ since $S_k{}^\alpha$ and $A_q{}^{\alpha k}$ are orthogonal to each other. Since $L_k{}^\alpha$ is hermitean in the subspace orthogonal to the vector $S_k{}^\alpha$,[8] we obtain from (2·28)

$$f_{q'}^{\alpha k} = (1-\mathcal{P}_{q'}^{\alpha k})iL_k{}^\alpha A_{q'}^{\alpha k}, \tag{2·37}$$

where

$$\mathcal{P}_{q'}^{\alpha k}G \equiv [(G, A_q^{\alpha k*})/(A_{q'}^{\alpha k}, A_q^{\alpha k*})]A_{q'}^{\alpha k}. \tag{2·38}$$

Thus the damping function $\varphi_k^\alpha(z)$ is written in terms of the microscopic variables $A_q^{\alpha k}(t)$.

In the vicinity of the critical point, the dynamic processes of the critical variables become slow (the critical slowing-down). Since $(S_k^\alpha, S_k^{\alpha *})$ and $(A_q^{\alpha k}, A_q^{\alpha k *})$ with small wave numbers become anomalously large in the vicinity of the ferromagnetic critical point, both S_k^α and $A_q^{\alpha k}$ with small wave numbers are the critical variables. As will be shown later, therefore, not only ω_k^α and $\varphi_k^\alpha(z)$ but also $\omega_{qq'}^{\alpha k}$ and $\varphi_{qq'}^{\alpha k}(z)$ become small in the vicinity of the ferromagnetic critical point when $\boldsymbol{k}$, $\boldsymbol{q}$ and $\boldsymbol{q}'$ are small wave numbers, thus representing the critical slowing down of S_k^α and $A_q^{\alpha k}$. Equation (2·17) will enable us to study how these anomalous fluctuations and dynamic processes of the microscopic variables $A_q^{\alpha k}(t)$ determine the critical behavior of the magnetization. For instance, the critical and kinematical slowing-down of $\omega_{qq'}^{\alpha k}$ and $\varphi_{qq'}^{\alpha k}(z)$, together with the anomalous increase of $(A_q^{\alpha k}, A_{q'}^{\alpha k*})$ with small wave numbers, will make the contributions of small wave number terms important in the sum of (2·17). Such a combined effect of the critical fluctuations and their dynamic properties was essential to understand the anomalous increase of the NMR line width. This is the crucial point in our theory, and removes a serious deficiency in the moment method[10] and its modifications[11),12] which cannot describe such a life-time effect.

The random forces $f_q^{\alpha k}$, (2·34), have the form

$$f_q^{\alpha k} = (1 - \mathcal{P}_q^{\alpha k} - \mathcal{P}_k^{\alpha}) iLA_q^{\alpha k}. \tag{2·39}$$

Insertion of (2·15) and (2·3), therefore, leads to

$$f_q^{\alpha k}(t) = \sum_{\mu=1}^{3} \sum_r g_{\mu r}^{\alpha k q} A_{\mu r}^{\alpha k q}(t). \tag{2·40}$$

Explicit expressions for $g_{\mu r}^{\alpha k q}$ and $A_{\mu r}^{\alpha k q}$ can be written down easily. For instance, since

$$iLA_q^{0k} = [\dot{S}_q^+ S_{k-q}^- + S_q^+ \dot{S}_{k-q}^-] - [(S_q^+ S_{k-q}^-, S_k^{0*})/(S_k^0, S_k^{0*})]\dot{S}_k^0, \tag{2·41}$$

we have

$$A_{1r}^{0kq} = (1 - \mathcal{P}_q^{0k} - \mathcal{P}_k^0) S_r^0 S_{q-r}^+ S_{k-q}^-,$$

$$A_{2r}^{0kq} = (1 - \mathcal{P}_q^{0k} - \mathcal{P}_k^0) S_q^+ S_r^0 S_{k-q-r}^-, \tag{2·42}$$

$$A_{3r}^{0kq} = [(S_q^+ S_{k-q}^-, S_k^{0*})/(S_k^0, S_k^{0*})](1 - \mathcal{P}_q^{0k} - \mathcal{P}_k^0) S_r^+ S_{k-r}^-,$$

$$g_{1r}^{0kq} = (2/i\hbar N) J_r^{q}(\lambda),$$

$$g_{2r}^{0kq} = -(2/i\hbar N) J_r^{k-q}(\lambda), \tag{2·43}$$

$$g_{3r}^{0kq} = -[2(1-\lambda)/i\hbar N] J_r^{k}.$$

Similarly, we have

$$A_{1r}^{\pm kq} = (1-\mathcal{P}_q{}^{\pm k} - \mathcal{P}_k{}^{\pm}) S_r{}^+ S_{q-r}^- S_{k-q}^{\pm},$$
$$A_{2r}^{\pm kq} = (1-\mathcal{P}_q{}^{\pm k} - \mathcal{P}_k{}^{\pm}) S_q{}^0 S_r{}^0 S_{k-q-r}^{\pm}, \tag{2·44}$$
$$A_{3r}^{\pm kq} = [(S_q{}^0 S_{k-q}^{\pm}, S_k{}^{\pm *})/(S_k{}^{\pm}, S_k{}^{\pm *})](1-\mathcal{P}_q{}^{\pm k} - \mathcal{P}_k{}^{\pm}) S_r{}^0 S_{k-r}^{\pm},$$
$$g_{1r}^{\pm kq} = [2(1-\lambda)/i\hbar N] J_r{}^q,$$
$$g_{2r}^{\pm kq} = \pm (2/i\hbar N) J_r{}^{k-q}(\lambda), \tag{2·45}$$
$$g_{3r}^{\pm kq} = \mp (2/i\hbar N) J_r{}^k(\lambda).$$

The random forces $f_{q'}^{\alpha k}$, (2·37), have the same form as (2·40). Thus, inserting (2·40) into (2·33), and then applying (2·19) we obtain

$$\varphi_{qq'}^{\alpha k}(z) = \frac{1}{(A_q{}^{\alpha k}, A_{q'}^{\alpha k *})} \sum_{\mu=1}^{3} \sum_{r} \sum_{\mu'=1}^{3} \sum_{r'} g_{\mu r}^{\alpha kq} g_{\mu' r'}^{\alpha kq' *} \times \frac{(A_{\mu r}^{\alpha kq}, A_{\mu' r'}^{\alpha kq' *})}{z - i\omega_{\mu r, \mu' r'}^{\alpha kqq'} + \varphi_{\mu r, \mu' r'}^{\alpha kqq'}(z)}. \tag{2·46}$$

This gives us the damping function $\varphi_{qq'}^{\alpha k}(z)$ in terms of the variables $A_{\mu r}^{\alpha kq}(t)$. The new damping function $\varphi_{\mu r, \mu' r'}^{\alpha kqq'}(z)$ also can be written down similarly with the aid of (2·19). Proceeding in this manner, we obtain a continued fraction expansion of the relaxation functions, which has the form

$$\Xi_k(z) = \cfrac{1}{z - i\omega_k + \sum_q \sum_{q'} \cfrac{M^{(1)}(q, q'; k)}{z - i\omega_{qq'}^{k} + \sum_{\mu, r} \sum_{\mu', r'} \cfrac{M_{\mu\mu'}^{(2)}(r, r'; q, q'; k)}{z - i\omega_{\mu r \mu' r'}^{kqq'} + \ddots}}}. \tag{2·47}$$

Explicit expressions for the numerators can be written down easily; for instance,

$$M^{(1)}(q, q'; k) = g_q{}^{\alpha k} g_{q'}^{\alpha k *} (A_q{}^{\alpha k}, A_{q'}^{\alpha k *})/(S_k{}^{\alpha}, S_k{}^{\alpha *}),$$
$$M_{\mu\mu'}^{(2)}(r, r'; q, q'; k) = g_{\mu r}^{\alpha kq} g_{\mu' r'}^{\alpha kq' *} (A_{\mu r}^{\alpha kq}, A_{\mu' r'}^{\alpha kq' *})/(A_q{}^{\alpha k}, A_{q'}^{\alpha k *}). \tag{2·48}$$

The n-th denominator consists of the double summation over wave vectors and of the double summation over $n(n+1)/2$ indices. Thus (2·47) is a generalization of the continued fraction expansion previously presented by one of the authors.[8] The continued fraction expansion (2·47) has the following properties: (A) Its coefficients, the frequencies ω's and the numerators M's, are determined entirely by the equal-time correlation functions of spins. (B) The long time behavior of $\Xi_k{}^{\alpha}(t)$ can be obtained by taking small values of $|z|$. For instance, the damping constants of spins in the hydrodynamic regime are given by $\varphi_k{}^{\alpha}(z=0+)$. (C) The life-time effects of all critical variables involved can be taken into account explicitly.

§ 3. Above the critical point

We consider the isotropic Heisenberg model ($\lambda=0$) above the critical temperature T_c in the absence of a magnetic field. Then the correlation functions of the odd numbers of spins vanish due to the time reversal symmetry. This leads to

$$A_{\boldsymbol{q}}{}^{0k}=S_{\boldsymbol{q}}{}^{+}S_{\boldsymbol{k}-\boldsymbol{q}}^{-}\,,\quad A_{\boldsymbol{q}}{}^{\pm k}=S_{\boldsymbol{q}}{}^{0}S_{\boldsymbol{k}-\boldsymbol{q}}^{\pm}\,, \tag{3·1}$$

$$\left.\begin{aligned}A_{1r}^{0kq}&=(1-\mathcal{P}_{\boldsymbol{k}}{}^{0})\,S_{\boldsymbol{r}}{}^{0}S_{\boldsymbol{q}-\boldsymbol{r}}^{+}S_{\boldsymbol{k}-\boldsymbol{q}}^{-}\,,\\ A_{2r}^{0kq}&=(1-\mathcal{P}_{\boldsymbol{k}}{}^{0})\,S_{\boldsymbol{q}}{}^{+}S_{\boldsymbol{r}}{}^{0}S_{\boldsymbol{k}-\boldsymbol{q}-\boldsymbol{r}}^{-}\,,\end{aligned}\right\} \tag{3·2}$$

$$A_{3r}^{0kq}\equiv 0\,. \tag{3·3}$$

Since the variables $S_{\boldsymbol{k}}{}^{\alpha}$, $A_{\boldsymbol{q}}{}^{\alpha k}$, $A_{\mu r}^{\alpha kq}$, $\cdots$ are thus odd or even with respect to the time reversal, we also have

$$\omega_{\boldsymbol{k}}{}^{\alpha}=\omega_{qq'}^{\alpha k}=\omega_{\mu r,\mu' r'}^{\alpha kqq'}=\cdots=0\,. \tag{3·4}$$

In the following we neglect the λ integral in (1·4), thus (F, G^*) agreeing with the correlation function $\langle FG^*\rangle$. The ordering of spins in the correlations is not important, since use of the commutation relations merely yields the correlation functions of odd numbers of spins which vanish identically. We employ the pair correlation approximation which replaces the static correlation functions by a product of pair correlation functions. Thus we have, for example,

$$\begin{aligned}(A_{\boldsymbol{q}}{}^{0k},\ A_{\boldsymbol{q}'}^{0k*})&\simeq\langle S_{\boldsymbol{q}}{}^{+}S_{\boldsymbol{k}-\boldsymbol{q}}^{-}S_{-\boldsymbol{k}+\boldsymbol{q}'}^{+}S_{-\boldsymbol{q}'}^{-}\rangle,\\ &\simeq\langle S_{\boldsymbol{q}}{}^{+}S_{-\boldsymbol{q}'}^{-}\rangle\langle S_{\boldsymbol{k}-\boldsymbol{q}}^{-}S_{-\boldsymbol{k}+\boldsymbol{q}'}^{+}\rangle\,,\\ &=N^{2}\delta_{\boldsymbol{q},\boldsymbol{q}'}(\boldsymbol{q})\,(\boldsymbol{k}-\boldsymbol{q}),\end{aligned} \tag{3·5}$$

where

$$(\boldsymbol{q})\equiv\langle S_{\boldsymbol{q}}{}^{\alpha}S_{\boldsymbol{q}}{}^{\alpha*}\rangle/N\,. \tag{3·6}$$

Since

$$\begin{aligned}&\mathcal{P}_{\boldsymbol{k}}{}^{0}S_{\boldsymbol{r}}{}^{0}S_{\boldsymbol{q}-\boldsymbol{r}}^{+}S_{\boldsymbol{k}-\boldsymbol{q}}^{-}\\ &\quad=[(S_{\boldsymbol{r}}{}^{0}S_{\boldsymbol{q}-\boldsymbol{r}}^{+}S_{\boldsymbol{k}-\boldsymbol{q}}^{-},\ S_{-\boldsymbol{k}}^{0})/(S_{\boldsymbol{k}}{}^{0},\ S_{-\boldsymbol{k}}^{0})]\,S_{\boldsymbol{k}}{}^{0},\\ &\quad\simeq\delta_{\boldsymbol{r},\boldsymbol{k}}\langle|S_{\boldsymbol{q}-\boldsymbol{r}}^{+}|^{2}\rangle S_{\boldsymbol{k}}{}^{0},\end{aligned} \tag{3·7}$$

we have

$$(A_{1r}^{0kq},\ A_{1r'}^{0kq*})\simeq\delta_{\boldsymbol{r},\boldsymbol{r}'}\langle|S_{\boldsymbol{r}}{}^{0}|^{2}\rangle\langle|S_{\boldsymbol{q}-\boldsymbol{r}}^{+}|^{2}\rangle\langle|S_{\boldsymbol{k}-\boldsymbol{q}}^{-}|^{2}\rangle\,. \tag{3·8}$$

Similarly we have

$$(A_{2r}^{0kq},\ A_{2r'}^{0kq*})\simeq\delta_{\boldsymbol{r},\boldsymbol{r}'}\langle|S_{\boldsymbol{q}}{}^{+}|^{2}\rangle\langle|S_{\boldsymbol{r}}{}^{0}|^{2}\rangle\langle|S_{\boldsymbol{k}-\boldsymbol{q}-\boldsymbol{r}}^{-}|^{2}\rangle, \tag{3·9}$$

$$(A_{1r}^{0kq},\ A_{2r'}^{0kq*})\simeq\delta_{\boldsymbol{r},\boldsymbol{r}'}\delta_{\boldsymbol{r},0}\langle|S_{\boldsymbol{r}}{}^{0}|^{2}\rangle\langle|S_{\boldsymbol{q}}{}^{+}|^{2}\rangle\langle|S_{\boldsymbol{k}-\boldsymbol{q}}^{-}|^{2}\rangle\,. \tag{3·10}$$

Equation (3·10) has two Kronecker δ's, thus giving only the contribution of the order of $0(1/N)$ to the sum of the second denominator of (2·47). Similarly, we can write the static correlations of higher order A's in terms of the pair correlation functions $(\boldsymbol{q})$. The projected parts onto lower order A's always yield negligible contributions. Since the variables A's were created from $S_{\boldsymbol{k}}^0$ by repeated use of the equations of motion (2·3), any of them consists of a cluster of spins which are linked by the exchange interaction. The static correlation of any of A's has only one term which contributes to the corresponding sum, and this term consists of a product of the pair correlations of spins between two clusters, e.g. as can be seen in (3·5). Thus writing the numerators of the continued fraction (2·47) in terms of the pair correlations, we obtain

$$\Xi_{\boldsymbol{k}}^0(z)=\cfrac{1}{z+\cfrac{1}{N}\sum_{\boldsymbol{q}}\cfrac{M(\boldsymbol{q},\boldsymbol{k})}{z+\cfrac{1}{N}\sum_{\mu=1}^{2}\sum_{\boldsymbol{r}}\cfrac{M_\mu(\boldsymbol{r},\boldsymbol{q},\boldsymbol{k})}{z+\cfrac{1}{N}\sum_{\nu=1}^{3}\sum_{\boldsymbol{s}}\cfrac{M_{\nu\mu}(\boldsymbol{s},\boldsymbol{r},\boldsymbol{q},\boldsymbol{k})}{z+\ddots}}}}, \tag{3·11}$$

where

$$M(\boldsymbol{q},\boldsymbol{k})=(2/\hbar)^2|J_{\boldsymbol{q}}^{\boldsymbol{k}}|^2(\boldsymbol{q})\,(\boldsymbol{k}-\boldsymbol{q})/(\boldsymbol{k}), \tag{3·12}$$

$$\left.\begin{aligned} M_1(\boldsymbol{r},\boldsymbol{q},\boldsymbol{k})&=M(\boldsymbol{r},\boldsymbol{q}),\\ M_2(\boldsymbol{r},\boldsymbol{q},\boldsymbol{k})&=M(\boldsymbol{r},\boldsymbol{k}-\boldsymbol{q}),\end{aligned}\right\} \tag{3·13}$$

$$\left.\begin{aligned} M_{11}(\boldsymbol{s},\boldsymbol{r},\boldsymbol{q},\boldsymbol{k})&=M(\boldsymbol{s},\boldsymbol{r}),\\ M_{21}(\boldsymbol{s},\boldsymbol{r},\boldsymbol{q},\boldsymbol{k})&=M(\boldsymbol{s},\boldsymbol{q}-\boldsymbol{r}),\\ M_{31}(\boldsymbol{s},\boldsymbol{r},\boldsymbol{q},\boldsymbol{k})&=M(\boldsymbol{s},\boldsymbol{k}-\boldsymbol{q}),\end{aligned}\right\} \tag{3·14}$$

$$\left.\begin{aligned} M_{12}(\boldsymbol{s},\boldsymbol{r},\boldsymbol{q},\boldsymbol{k})&=M(\boldsymbol{s},\boldsymbol{q}),\\ M_{22}(\boldsymbol{s},\boldsymbol{r},\boldsymbol{q},\boldsymbol{k})&=M(\boldsymbol{s},\boldsymbol{r}),\\ M_{32}(\boldsymbol{s},\boldsymbol{r},\boldsymbol{q},\boldsymbol{k})&=M(\boldsymbol{s},\boldsymbol{k}-\boldsymbol{q}-\boldsymbol{r}).\end{aligned}\right\} \tag{3·15}$$

The numerators M's all have the form

$$M(\boldsymbol{l}',\boldsymbol{l})=(2/\hbar)^2|J_{\boldsymbol{l}'}^{\boldsymbol{l}}|^2(\boldsymbol{l}')\,(\boldsymbol{l}-\boldsymbol{l}')/(\boldsymbol{l}), \tag{3·16}$$

where $\boldsymbol{l}'$ is the summation variable in the corresponding sum. Thus all M's have the similar structure in terms of the pair correlations. Such a similarity is basic, though not sufficient, for the validity of the dynamic scaling law. As can be seen in (2·17) and (2·46), the damping functions $\varphi(t)$ are decomposed into the components with two indices, e.g. $(\mu, \boldsymbol{r})$. The numerators M represent the magnitudes of such components at the initial time. Thus the above similarity means that the amplitudes of the components of the damping functions have the similar structure irrespective of their order.

Use of the pair correlation approximation could not be justified in the very vicinity of the critical point. In view of the formative stage of our theoretical understanding of dynamic critical phenomena, however, it should be useful to have a theory of anomalous relaxation which gives us the main physical features of the problem clearly as Landau's theory did in the static problem. Important results thus obtained are indeed confirmed without using the pair correlation approximation as will be shown in § 6.

§ 4. Ferromagnets above the critical point

We study here the continued fraction (3·11) in the case of isotropic Heisenberg ferromagnets with nearest neighbor interaction above the critical point.

Following the static scaling laws, we assume that if

$$q,\ \kappa \ll 1/b\,, \tag{4·1}$$

b being the nearest neighbor distance, then

$$(\boldsymbol{q}) \equiv \langle |S_{\boldsymbol{q}}^{\alpha}|^2\rangle / N = (\kappa R)^{-2+\eta} Q(q/\kappa)\,, \tag{4·2}$$

where R is a length of the order of b. Since (4·2) is a homogeneous function of q and κ, we should have

$$Q(s) \simeq 1 \quad \text{if} \quad s \ll 1\,, \tag{4·3}$$

$$Q(s) \simeq s^{-2+\eta} \quad \text{if} \quad s \gg 1\,. \tag{4·4}$$

The Ornstein-Zernike formula satisfies these assumptions with $\eta = 0$.

If $l,\ l' \ll b^{-1}$, then

$$J_{l'}^{l} \simeq c[l^2 - 2\boldsymbol{l}\cdot\boldsymbol{l}']\,, \tag{4·5}$$

where $c \equiv b^2 zJ/6 \simeq k_B T_c R^2/2$. Therefore, if all the wave numbers involved are much smaller than b^{-1}, (3·16) takes the form

$$M = \kappa^{2+\eta}(2c/\hbar)^2 R^{-2+\eta} Q(\boldsymbol{l}/\kappa,\ \boldsymbol{l}'/\kappa)\,, \tag{4·6}$$

where

$$Q(\boldsymbol{s},\ \boldsymbol{s}') \equiv [s^2 - 2\boldsymbol{s}\cdot\boldsymbol{s}']^2 Q(s') Q(|\boldsymbol{s}-\boldsymbol{s}'|)/Q(s)\,. \tag{4·7}$$

Now return to $\Xi_{\boldsymbol{k}}^0(z)$, (3·11). Let us consider a long wave length disturbance in the vicinity of the critical point such that k and κ are very small compared to b^{-1}. Then the imaginary frequency z of our interest stays in the neighborhood of origin in the complex z plane. Then the most important contribution in the sums of (3·11) will come from the terms with small wave numbers. First consider the sum $\sum_{\boldsymbol{q}}$. If q is small, then (3·12) and (3·13) lead to the factors

$$\frac{1}{R^4}\frac{1}{q^2+\kappa^2}\frac{1}{(\boldsymbol{k}-\boldsymbol{q})^2+\kappa^2}\,,\quad \frac{1}{z+[q^2(q^2+\kappa^2)+(\boldsymbol{k}-\boldsymbol{q})^2\{(\boldsymbol{k}-\boldsymbol{q})^2+\kappa^2\}]\phi(\boldsymbol{q},\boldsymbol{k};z)}\,, \tag{4·8}$$

where, for simplicity, the Ornstein-Zernike form has been assumed. Both of these factors become larger as q gets smaller. The first represents the critical fluctuation of the variable $A_{\boldsymbol{q}}^{0\boldsymbol{k}}$, and the second effect is due to its kinematical and critical slowing-down. Thus, though the state density and J_q^k yield a factor of q^4, the terms with small values of q give the most dominant contribution. The importance of the second factor of (4·8) should be noted. In the other sums we also have the same situation provided that the foregoing wave numbers and κ are small. Thus in all the sums the main contribution will come from the small wave number terms. Therefore, we may use (4·6) for all numerators.

Transforming the sums into the integrals and then changing the integration variables by

$$\frac{1}{N}\sum_q = \frac{\kappa^3}{(2\pi)^3\rho}\iiint_{-\infty}^{\infty} ds\,,$$

we thus obtain

$$\Xi_k^0(z) = \cfrac{1}{z+\Omega_\kappa^2\displaystyle\int\cfrac{m(s;\,k/\kappa)\,ds}{z+\Omega_\kappa^2\displaystyle\sum_{\mu=1}^{2}\int\cfrac{m_\mu(s',\,s;\,k/\kappa)\,ds'}{z+\Omega_\kappa^2\displaystyle\sum_{\mu'=1}^{3}\int\cfrac{m_{\mu'\mu}(s'',\,s',\,s;\,k/\kappa)\,ds''}{z+\ddots}}}}\,, \tag{4·9}$$

where

$$\Omega_\kappa^2 \equiv C^2\kappa^{5+\eta},\quad C \equiv \frac{k_B T_C}{\hbar\sqrt{8\rho R^3}}R^{(5+\eta)/2} \tag{4·10}$$

and $m(s;\,k/\kappa) = Q(k/\kappa,\,s)/\pi^3$. Equation (4·9) leads to a function of the form

$$\Xi_k^0(z) = G(z/\Omega_\kappa,\,k/\kappa)/\Omega_\kappa\,, \tag{4·11}$$

whose Laplace inversion gives a function of the form

$$\Xi_k^0(t) = F(\Omega_\kappa t,\,k/\kappa)\,. \tag{4·12}$$

The collective modes of spins are determined by the poles of (4·11) in the complex z plane, and thus their imaginary frequencies should have the k dependence of the form

$$z = \Omega_\kappa g(k/\kappa)\,. \tag{4·13}$$

Thus it is concluded that the dynamic scaling law holds and the critical exponent of the characteristic frequency is given by

$$\theta = (5+\eta)/2\,. \tag{4·14}$$

In the case of $\eta=0$ this result agrees with the previous theories.[7),11),12)]

So far we have not specified the relative magnitude of k and κ. The property of $\Xi_k^0(z)$, however, critically depends upon their relative magnitude. In the case of $k \ll \kappa$, we have

$$m(\boldsymbol{s};k/\kappa)=\left(\frac{k}{\kappa}\right)^2\left[\left(\frac{k}{\kappa}\right)-2\hat{\boldsymbol{k}}\cdot\boldsymbol{s}\right]^2 Q^2(s)/\pi^3, \qquad (4\cdot15)$$

$\hat{\boldsymbol{k}}$ denoting the unit vector directed along $\boldsymbol{k}$, and in the other numerators we can put $\boldsymbol{k}=\mathbf{0}$. Then (4·9) leads to the form

$$\Xi_{\boldsymbol{k}}{}^0(z)=\frac{1}{z+[k^2\kappa^{\eta+3}/\Omega_\kappa]D_F(z/\Omega_\kappa)}. \qquad (4\cdot16)$$

Thus in the limit of $k\ll\kappa$, neglecting the z dependence of D_F, we obtain

$$\Xi_{\boldsymbol{k}}{}^0(t)=\exp(-k^2\Lambda t), \qquad (4\cdot17)$$

where

$$\Lambda=\kappa^{(1+\eta)/2}D_F(0)/C. \qquad (4\cdot18)$$

This predicts that the spin diffusion constant Λ decreases as the temperature approaches the critical point T_c, being proportional to $(T-T_c)^\phi$, $\phi=(1+\eta)\nu/2$.

In the case of $k\gg\kappa$, the memory effect becomes important and the relaxation function will deviate from the simple decay (4·17) largely. In the limit of $k\gg\kappa$, (4·3) and (4·7) lead to

$$M=\Omega_k{}^2(8\rho/k^3)Q'(\boldsymbol{l}/k,\ \boldsymbol{l}'/k), \qquad (4\cdot19)$$

$$Q'(\boldsymbol{s},\ \boldsymbol{s}')\equiv[s^2-2\boldsymbol{s}\cdot\boldsymbol{s}']^2[s'|\boldsymbol{s}-\boldsymbol{s}'|/s]^{-2+\eta}. \qquad (4\cdot20)$$

Thus the numerators m in (4·9) take simpler forms which do not depend on either of k and κ. Calculation of the generalized continued fraction, however, is not simple. We will study this problem in a later communication.

§ 5. Antiferromagnets above the critical point

Let us consider an isotropic Heisenberg antiferromagnet whose sublattices alternate. The critical variable in this system is the staggered polarization $S_{\boldsymbol{K}}{}^\alpha$, where $\boldsymbol{K}$ is the half of the reciprocal lattice vector. Instead of (4·2), therefore, we assume that

$$(\boldsymbol{q}'+\boldsymbol{K})=(\kappa R)^{-2+\eta}Q(q'/\kappa), \qquad (5\cdot1)$$

where q', $\kappa\ll K$. The uniform polarization does not show a critical fluctuation.

Let us first study the relaxation function of the critical variable, $\Xi^0_{\boldsymbol{k}+\boldsymbol{K}}(z)$, $k\ll K$. This can be measured in the magnetic scattering of neutron by observing the scattered neutrons with the scattering vectors around $\boldsymbol{K}$. We begin to determine the wave number region which yields the most dominant part in the sums of (3·11). Shifting the wave vector $\boldsymbol{k}$ by $\boldsymbol{K}$, we find that the numerators M's take either of the following four types:

$$M(\boldsymbol{r},\ \boldsymbol{q})\sim|J_r{}^{\boldsymbol{q}}|^2(\boldsymbol{r})\ (\boldsymbol{q}-\boldsymbol{r})/(\boldsymbol{q}),$$

$$M(\boldsymbol{r}'+\boldsymbol{K},\ \boldsymbol{q})\sim|J^{\boldsymbol{q}}_{r'+K}|^2(\boldsymbol{r}'+\boldsymbol{K})\ (\boldsymbol{q}-\boldsymbol{r}'-\boldsymbol{K})/(\boldsymbol{q}), \qquad (5\cdot2)$$

$$M(\boldsymbol{r},\ \boldsymbol{q}'+\boldsymbol{K}) \sim |J_{\boldsymbol{r}}^{\boldsymbol{q}'+\boldsymbol{K}}|^2(\boldsymbol{r})\,(\boldsymbol{q}'-\boldsymbol{r}+\boldsymbol{K})/(\boldsymbol{q}'+\boldsymbol{K}),$$
$$M(\boldsymbol{r}'+\boldsymbol{K},\ \boldsymbol{q}'+\boldsymbol{K}) \sim |J_{\boldsymbol{r}'+\boldsymbol{K}}^{\boldsymbol{q}'+\boldsymbol{K}}|^2(\boldsymbol{r}'+\boldsymbol{K})\,(\boldsymbol{q}'-\boldsymbol{r}')/(\boldsymbol{q}'+\boldsymbol{K}),$$

where $\boldsymbol{r}$ and $\boldsymbol{r}'$ are the summation variables in the corresponding sum. If the wave numbers $\boldsymbol{q}$, $\boldsymbol{q}'$, $\boldsymbol{r}$, $\boldsymbol{r}'$ are small, then the pair correlations which do not have $\boldsymbol{K}$ are nearly constant and thus only the last three types have the pair correlations which show the critical fluctuation. Their kinematical properties are determined by

$$\begin{aligned} &J_{\boldsymbol{r}}^{\boldsymbol{q}} \cong -c'[q^2-2\boldsymbol{q}\cdot\boldsymbol{r}],\\ &J_{\boldsymbol{r}'+\boldsymbol{K}}^{\boldsymbol{q}} \cong c[q^2-2\boldsymbol{q}\cdot\boldsymbol{r}'],\\ &J_{\boldsymbol{r}}^{\boldsymbol{q}'+\boldsymbol{K}} \cong 2z_1J^{(1)},\\ &J_{\boldsymbol{r}'+\boldsymbol{K}}^{\boldsymbol{q}'+\boldsymbol{K}} \cong -2z_1J^{(1)}, \end{aligned} \tag{5·3}$$

where z_1 is the number of the nearest neighbors, $J^{(1)}$ their exchange interaction constant, and

$$\begin{aligned} &c = k_BT_cR^2/2,\quad R^2 = \frac{1}{4S(S+1)}\left[b_1^2\left(\frac{\Theta}{T_c}+1\right)-b_2^2\left(\frac{\Theta}{T_c}-1\right)\right],\\ &c' = [k_BT_c/8S(S+1)]\left[b_1^2\left(\frac{\Theta}{T_c}+1\right)+b_2^2\left(\frac{\Theta}{T_c}-1\right)\right], \end{aligned} \tag{5·4}$$

where Θ is the paramagnetic Curie temperature.

First consider the $\boldsymbol{q}$ sum. Its numerator $M(\boldsymbol{q},\ \boldsymbol{k}+\boldsymbol{K})$ takes the third type of (5·2) if $\boldsymbol{q}$ is small, and the fourth type if $\boldsymbol{q}$ is around $\boldsymbol{K}$. Thus, if $q \ll K$, then the summand takes the form

$$\frac{1}{(\boldsymbol{k}-\boldsymbol{q})^2+\kappa^2}\times\frac{1}{z+[q^2+\{(\boldsymbol{k}-\boldsymbol{q})^2+\kappa^2\}]\phi(\boldsymbol{q},\ \boldsymbol{k};\ z)},$$

where, for simplicity, the Ornstein-Zernike form has been assumed. If $\boldsymbol{q}=\boldsymbol{q}'+\boldsymbol{K}$, $q' \ll K$, then the summand takes the form

$$\frac{1}{q'^2+\kappa^2}\times\frac{1}{z+[\{q'^2+\kappa^2\}+(\boldsymbol{k}-\boldsymbol{q}')^2]\phi(\boldsymbol{q},\ \boldsymbol{k};\ z)}.$$

In the limit of the small values of k, κ and $|z|$, both of these diverge at the small value of q or q' as q^{-4} or q'^{-4}. Since the state density yields the factor q^2 or q'^2, these terms yield the divergence of q^{-2} or q'^{-2} in the $\boldsymbol{q}$ sum. It should be noted here that the divergence is entirely due to the kinematical slowing-down of the small wave number polarization involved and the critical slowing-down of the staggered polarization involved, both of which play the similar role. Thus the most dominant part arises from the region where $\boldsymbol{q}\sim\boldsymbol{0}$ and $\boldsymbol{q}\sim\boldsymbol{K}$. Even though the divergence is weaker than q^{-4} in the ferromagnetic case, we may replace the sum by this most dominant part, provided that k and

κ are very small.

Next consider the $\boldsymbol{r}$ sum which consists of the two terms with $M_1(\boldsymbol{r}, \boldsymbol{q}, \boldsymbol{k}+\boldsymbol{K})$ and $M_2(\boldsymbol{r}, \boldsymbol{q}, \boldsymbol{k}+\boldsymbol{K})$. The divergence which arises due to the slowing-down effect, is always of the order of r^{-2} or r'^{-2}. Since the first type of (5·2) yields the factor of r^2, the second r'^{-2}, the third r^{-2} and the fourth r'^{-2}, therefore, only the last three types yield the divergence. Thus, if the numerator is of the form $M(\boldsymbol{r}, \boldsymbol{q})$, $q \ll K$, then the most dominant part arises from $\boldsymbol{r} \sim \boldsymbol{K}$. If the numerator is of the form $M(\boldsymbol{r}, \boldsymbol{q}'+\boldsymbol{K})$, $q' \ll K$, however, the most divergent part arises from $\boldsymbol{r} \sim \boldsymbol{0}$ and $\boldsymbol{r} \sim \boldsymbol{K}$. Thus when $\boldsymbol{q} \sim \boldsymbol{0}$, the M_1 sum has the most dominant part at $\boldsymbol{r} \sim \boldsymbol{K}$, whereas the M_2 sum has it at both $\boldsymbol{r} \sim \boldsymbol{0}$ and $\boldsymbol{r} \sim \boldsymbol{K}$. When $\boldsymbol{q} \sim \boldsymbol{K}$, however, the M_1 sum has the most dominant part at $\boldsymbol{r} \sim \boldsymbol{0}$ and $\boldsymbol{r} \sim \boldsymbol{K}$, whereas the M_2 sum has it only at $\boldsymbol{r} \sim \boldsymbol{K}$.

We can make the same analysis in the other sums. Thus, in all of the sums of (3·11), we can find the most dominant parts whose divergence is of the order of r^{-2} or r'^{-2}. In the following we replace the sums by such most dominant parts. Then the numerators take either of the following three forms:

$$\begin{aligned} &M(\boldsymbol{r}'+\boldsymbol{K}, \boldsymbol{q}) \sim \kappa^{2\eta}\left[\left(\frac{q}{\kappa}\right)^2 - 2\left(\frac{\boldsymbol{q}}{\kappa}\right)\cdot\left(\frac{\boldsymbol{r}'}{\kappa}\right)\right]^2 Q\left(\frac{r'}{\kappa}\right) Q\left(\frac{|\boldsymbol{q}-\boldsymbol{r}'|}{\kappa}\right), \\ &M(\boldsymbol{r}, \boldsymbol{q}'+\boldsymbol{K}) \sim Q(|\boldsymbol{q}'-\boldsymbol{r}|/\kappa)/Q(q'/\kappa), \\ &M(\boldsymbol{r}'+\boldsymbol{K}, \boldsymbol{q}'+\boldsymbol{K}) \sim Q(r'/\kappa)/Q(q'/\kappa). \end{aligned} \tag{5·5}$$

Thus transforming the sums into the integrals and then rewriting in terms of the reduced wave vectors, we find that

$$\Xi^0_{\boldsymbol{k}+\boldsymbol{K}}(z) = \frac{1}{z+\kappa^3\left[\int \frac{m(\boldsymbol{q}; k/\kappa)\, d\boldsymbol{q}}{z+\varphi(\boldsymbol{q}; z)} + \int \frac{m'(\boldsymbol{q}'; k/\kappa)\, d\boldsymbol{q}'}{z+\varphi'(\boldsymbol{q}'; z)}\right]}, \tag{5·6}$$

where the unprimed quantities indicate the contribution from the small wave number region and the primed ones denote that from the $\boldsymbol{K}$ wave number region. The damping functions are given by

$$\begin{aligned} \varphi(\boldsymbol{q}; z) \equiv {} & \kappa^{3+2\eta} \frac{m_1^{(1)\prime}(\boldsymbol{r}', \boldsymbol{q}; k/\kappa)\, d\boldsymbol{r}'}{z+\varphi_1^{(1)\prime}(\boldsymbol{r}', \boldsymbol{q}; z)} \\ & + \kappa^3\left[\int \frac{m_2^{(1)}(\boldsymbol{r}, \boldsymbol{q}; k/\kappa)\, d\boldsymbol{r}}{z+\varphi_2^{(1)}(\boldsymbol{r}, \boldsymbol{q}; z)} + \int \frac{m_2^{(1)\prime}(\boldsymbol{r}', \boldsymbol{q}; k/\kappa)\, d\boldsymbol{r}'}{z+\varphi_2^{(1)\prime}(\boldsymbol{r}', \boldsymbol{q}; z)}\right], \end{aligned} \tag{5·7}$$

$$\begin{aligned} \varphi'(\boldsymbol{q}'; z) \equiv {} & \kappa^3\left[\int \frac{m_1^{(2)}(\boldsymbol{r}, \boldsymbol{q}'; k/\kappa)\, d\boldsymbol{r}}{z+\varphi_1^{(2)}(\boldsymbol{r}, \boldsymbol{q}'; z)} + \int \frac{m_1^{(2)\prime}(\boldsymbol{r}', \boldsymbol{q}'; k/\kappa)\, d\boldsymbol{r}'}{z+\varphi_1^{(2)\prime}(\boldsymbol{r}', \boldsymbol{q}'; z)}\right] \\ & + \kappa^{3+2\eta}\int \frac{m_2^{(2)\prime}(\boldsymbol{r}', \boldsymbol{q}'; k/\kappa)\, d\boldsymbol{r}'}{z+\varphi_2^{(2)\prime}(\boldsymbol{r}', \boldsymbol{q}'; z)}. \end{aligned} \tag{5·8}$$

The explicit expressions for the damping functions of (5·7) and (5·8) can be written down similarly, but they are omitted here. It is important, however, to note that all of them consist of both the κ^3 term and the $\kappa^{3+2\eta}$ term. The property of this continued fraction depends upon the value of η. In the case of $\eta=0$ it is clear that the dynamic scaling law holds and the critical exponent of the characteristic frequency is given by

$$\theta=3/2\,. \tag{5·9}$$

If $\eta>0$, then we can neglect the fractions which have $\kappa^{3+2\eta}$ in front. Consequently, in this case also the dynamic scaling law holds with the same critical exponent (5·9). Thus, including the case of $\eta\neq 0$, we have

$$\Xi^0_{k+K}(t)=F_1(t\kappa^{3/2},\ k/\kappa), \tag{5·10}$$

which agrees with the previous predictions.

Next we study the relaxation function of the small wave number polarization, $\Xi_k{}^0(z)$, $k\ll K$. In a similar manner, we can determine the wave number region which gives the dominant part in the sums of (3·11). The numerators of the most dominant contribution are of the either form of the three of (5·5). Thus we find that

$$\Xi_k{}^0(z)=\cfrac{1}{z+\kappa^{3+2\eta}\displaystyle\int\cfrac{m'(\boldsymbol{q}';\ k/\kappa)\,d\boldsymbol{q}'}{z+\kappa^3\displaystyle\sum_{\mu=1}^{2}\left[\int\frac{m_\mu(\boldsymbol{r},\ \boldsymbol{q}';\ k/\kappa)\,d\boldsymbol{r}}{z+\varphi_\mu(\boldsymbol{r},\ \boldsymbol{q}';\ z)}+\int\frac{m_\mu{}'(\boldsymbol{r}',\ \boldsymbol{q}';\ k/\kappa)\,d\boldsymbol{r}'}{z+\varphi_\mu{}'(\boldsymbol{r}',\ \boldsymbol{q}';\ z)}\right]}}\,. \tag{5·11}$$

The damping functions φ_μ and $\varphi_\mu{}'$ can be written down from (3·14) and (3·15). For example,

$$\varphi_1(\boldsymbol{r},\ \boldsymbol{q}';\ z)=\kappa^{3+2\eta}\int\frac{m_{11}{}'(\boldsymbol{s}',\ \boldsymbol{r},\ \boldsymbol{q}';\ k/\kappa)\,d\boldsymbol{s}'}{z+\varphi_{11}{}'(\boldsymbol{s}',\ \boldsymbol{r},\ \boldsymbol{q}';\ z)}$$
$$+\kappa^3\sum_{\nu=2}^{3}\left[\int\frac{m_{\nu1}(\boldsymbol{s},\ \boldsymbol{r},\ \boldsymbol{q}';\ k/\kappa)\,d\boldsymbol{s}}{z+\varphi_{\nu1}(\boldsymbol{s},\ \boldsymbol{r},\ \boldsymbol{q}';\ z)}+\int\frac{m'_{\nu1}(\boldsymbol{s}',\ \boldsymbol{r},\ \boldsymbol{q}';\ k/\kappa)\,d\boldsymbol{s}'}{z+\varphi'_{\nu1}(\boldsymbol{s}',\ \boldsymbol{r},\ \boldsymbol{q}';\ z)}\right]. \tag{5·12}$$

It is important to note that all of them consist of both the κ^3 term and the $\kappa^{3+2\eta}$ term. In the case of $\eta=0$, we thus obtain

$$\Xi_k{}^0(t)=F_2(t\kappa^{3/2},\ k/\kappa)\,. \tag{5·13}$$

In the case of $\eta\neq 0$, however, we can neglect those fractions below the third denominator which have $\kappa^{3+2\eta}$ in front, and obtain

$$\Xi_k{}^0(z)=\frac{1}{z+\kappa^{2\eta+3/2}g(z/\kappa^{3/2},\ k/\kappa)}\,, \tag{5·14}$$

where $g(x,\ y)$ is a function of two variables. In the limit of the small values of k and κ, neglecting the z dependence of g, we thus obtain

$$\Xi_k{}^0(t)=\exp(z_k t), \tag{5·15}$$

where

$$z_k = -\kappa^{2\eta+3/2} g(0, k/\kappa). \tag{5·16}$$

This has the critical exponent $\theta = 2\eta + 3/2$ which differs from (5·10). Namely, the dynamic scaling law proposed by Halperin and Hohenberg does not hold. This is due to the nonsimilarity between the first type and the last two of (5·5). Thus, in the case of $\eta \neq 0$, the memory effect does not appear irrespective of the relative magnitude of k and κ, which would mean that an oscillatory motion, such as diffuse oscillatory modes observed in $RbMnF_3$, is difficult to appear about the total polarization.

So far we have not specified the relative magnitude of k and κ. The property of the relaxation function, however, critically depends upon their relative magnitude. In the limit of $k \ll \kappa$, (5·5) leads to

$$M(\boldsymbol{q}' + \boldsymbol{K}, \boldsymbol{k}) \sim k^2 \kappa^{-2+2\eta} \left[\left(\frac{k}{\kappa} \right) - 2 \left(\frac{\boldsymbol{k}}{k} \right) \cdot \left(\frac{\boldsymbol{q}'}{\kappa} \right) \right]^2 Q^2 \left(\frac{q'}{\kappa} \right), \tag{5·17}$$

$$\begin{aligned} &M(\boldsymbol{q}, \boldsymbol{k} + \boldsymbol{K}) \sim Q(q/\kappa), \\ &M(\boldsymbol{q}' + \boldsymbol{K}, \boldsymbol{k} + \boldsymbol{K}) \sim Q(q'/\kappa). \end{aligned} \tag{5·18}$$

In the other numerators we can put $\boldsymbol{k} = \boldsymbol{0}$. Thus in the limit of $k \ll \kappa$, we obtain

$$\Xi_{\boldsymbol{k}}{}^0(z) = \frac{1}{z + [k^2 \kappa^{2\eta+1}/\kappa^{3/2}] D_A(z/\kappa^{3/2})}, \tag{5·19}$$

which leads to the diffusion of spins, similarly to (4·18), but with the diffusion constant

$$\Lambda = \kappa^{2\eta - 1/2} D_A(0). \tag{5·20}$$

If $\eta < 1/4$, then (5·20) shows the critical speeding-up predicted by Halperin and Hohenberg in the case of $\eta = 0$.

The relaxation of the staggered polarization is more complicated. In the limit of $k \ll \kappa$, (5·18) leads to

$$\Xi^0_{\boldsymbol{k}+\boldsymbol{K}}(z) = \frac{1}{z + \kappa^{3/2} f(z/\kappa^{3/2})}. \tag{5·21}$$

In this case, therefore, the memory effect is important and the relaxation should deviate from the simple decay. This result differs from the phenomenological theory which predicts the exponential decay in the vicinity of the critical point.[13] In the phenomenological theory, the short wave length terms were important, introducing a microscopic time distinctly smaller than the macroscopic relaxation time. However, if the dynamic scaling law should hold, then there exists only one time constant characterized by $\kappa^{3/2}$ in the case of the staggered polarization, thus yielding the memory effect. Therefore, a neutron scattering experiment determining whether the energy distribution of scattered neutrons is Lorentzian or not around the half of the reciprocal lattice vector will be very useful to study the validity of the dynamic scaling law. Thus if one finds that the energy distribution is Lorentzian in the region where $k \ll \kappa$, then the dynamic scaling law is to be disproved.

§ 6. Ferromagnets below the critical point

Below the critical point, the correlation functions of the odd numbers of spins do not vanish. Thus we should have the contribution from the frequency terms ω_k, $\omega_{qq'}^{k}$, $\cdots$ in the continued fraction (2·47). In the present section, we do not use the pair correlation approximation.

Let us first consider ω_k^{α}, (2·8). With the aid of the identity[14)]

$$([\mathcal{H}, F], G) = -k_B T \langle [F, G] \rangle , \tag{6·1}$$

this can be written as[5)]

$$\omega_k^{\alpha} = -\alpha\sigma [k_B T / \hbar (\boldsymbol{k})_{\alpha}], \tag{6·2}$$

where σ denotes the spontaneous polarization of spin for one spin $\langle S_0^0 \rangle / N$, and $(\boldsymbol{k})_{\alpha}$ is defined by

$$(\boldsymbol{k})_{\alpha} \equiv (S_{\boldsymbol{k}}^{\alpha}, S_{\boldsymbol{k}}^{\alpha *}) / N . \tag{6·3}$$

Following the static scaling laws, we have[3)]

$$\sigma \sim \kappa^{\beta/\nu}, \quad \beta/\nu = (d-2+\eta)/2 , \tag{6·4}$$

d being the dimensionality of the system, and we assume

$$(\boldsymbol{k})_{\alpha} = (\kappa R)^{-2+\eta} Q_{\alpha}(k/\kappa), \tag{6·5}$$

where $k, \kappa \ll R^{-1}$. Thus we find from (6·2) that ω_k^0 always vanishes and $\omega_k^{\pm}$ take the form

$$\omega_k^{\pm} = \kappa^{\lambda} g_{\pm}(k/\kappa), \quad \lambda \equiv (d+2-\eta)/2 . \tag{6·6}$$

The critical exponent λ with $d=3$ differs from (4·14) unless $\eta = 0$. As will be shown later, this means that the dynamic scaling laws do not hold below the critical point if $\eta \neq 0$.

It is interesting to determine the $\boldsymbol{k}$ dependence of $\omega_k^{\pm}$. In the limit of $\kappa \ll k$, we should have

$$Q_{\pm}(k/\kappa) \simeq (k/\kappa)^{-2+\eta}, \tag{6·7}$$

which yields

$$\omega_k^{\pm} \sim k^{2-\eta} \kappa^{(1+\eta)/2}. \tag{6·8}$$

This $\boldsymbol{k}$ dependence of the frequency spectrum differs from that of the usual spin wave frequency. In this region, however, the damping effect would prevail the frequency.

In the hydrodynamic regime where $k \ll \kappa$, we feel an uncertainty in determining $Q_{\pm}(k/\kappa)$. If we employ the Bogolyubov-Tyablikov approximation in the two-time Green's function method, then we obtain (6·7) with $\eta=0$ irrespective of the relative magnitude of k and κ.[15] Therefore, it is not unreasonable to assume (6·7) even in the hydrodynamic regime, thus yielding (6·8). This anomalous $\boldsymbol{k}$ dependence, however, cannot easily be understood. In order to have the normal $\boldsymbol{k}$ dependence

$$\omega_k{}^{\pm} = \mp Dk^2, \quad (\kappa \gg k), \tag{6·9}$$

we have to assume

$$\chi_k{}^{\pm} \sim k^{-2}\kappa^{\eta}, \quad (\kappa \gg k). \tag{6·10}$$

Then we have

$$D \sim \kappa^{(1-\eta)/2}. \tag{6·11}$$

It would be important to settle this problem.

The relations (6·2) and (6·6) can be extended to the higher order frequencies. The evolution operator of the n-th order A variable has the form

$$L_n = (1 - \sum_{i=0}^{n-1} \mathcal{P}_i) L\,, \quad (L_0 \equiv L), \tag{6·12}$$

where $\mathcal{P}_i$ is the projection operator onto the i-th order A variable A_i, $(A_0 \equiv S_{\boldsymbol{k}}{}^{\alpha})$. Since

$$A_n = (1 - \sum_{i=0}^{n-1} \mathcal{P}_i) A_n\,,$$

we thus obtain

$$(iL_n A_n, A_n{}'^*) = (iLA_n, A_n{}'^*), \tag{6·13}$$

which means that the n-th order frequency takes the form

$$\omega_n = -(k_B T/\hbar)\langle[A_n, A_n{}'^*]\rangle/(A_n, A_n{}'^*). \tag{6·14}$$

The static scaling laws imply that the multiple correlations of spin have the form[12)]

$$\langle S_{q_1}^{\alpha_1} S_{q_2}^{\alpha_2} \cdots S_{q_m}^{\alpha_m} \rangle = \kappa^{-m\lambda}\{(N\kappa^d) f_1 + \cdots + (N\kappa^d)^{[m/2]} f_{[m/2]}\}, \tag{6·15}$$

where $[m/2]=n$ if $m=2n-1$ or $2n$. The f_j is a function of (q_i/κ) with j Kronecker's delta corresponding to j conditions on the wave vectors, and has a definite value in the macroscopic limit ($N\to\infty$ with $\rho \equiv N/V =$ constant). The denominator of (6·14), $(A_n, A_n{}'^*)$, consists of the spin correlations of the $(2n+2)$-th and lower orders, and the numerator $\langle[A_n, A_n{}'^*]\rangle$ consists of the spin correlations of the $(2n+1)$-th and lower orders. Thus in the limit of the small values of κ, q_1, $\cdots$, q_n, k, we obtain

$$\omega_n = \kappa^{\lambda} g^{(n)}(q_n/\kappa, \cdots, q_1/\kappa, k/\kappa). \tag{6·16}$$

Equation (6·6) is a particular case of (6·16), corresponding to $n=0$.

The numerators of (2·47) can be studied similarly. The n-th order numerator has the form

$$M^{(n)}(\boldsymbol{q}_n, \boldsymbol{q}_n'; \cdots; \boldsymbol{q}_1, \boldsymbol{q}_1'; \boldsymbol{k}) = g_{\boldsymbol{q}_n} g_{\boldsymbol{q}_n'} (A_n, A_n'^*)/(A_{n-1}, A_{n-1}'^*). \tag{6·17}$$

Since $g_{\boldsymbol{q}} = N^{-1}\kappa^2 f(q/\kappa)$, therefore, we obtain

$$M^{(n)} = (N\kappa^d)^{-2}\kappa^{2\theta}[f_1' + \delta_{\boldsymbol{q}_n, \boldsymbol{q}_n'}(N\kappa^d) f_2'], \tag{6·18}$$

where

$$\theta = (2d + 4 - 2\lambda)/2 = (d + 2 + \eta)/2, \tag{6·19}$$

and f_1' and f_2' are definite functions of the reduced wave vectors.

As has been discussed before, the most dominant contribution in the continued fraction (2·47) arises from the small wave number terms due to the kinematical and critical slowing-down. Therefore we may use (6·16) and (6·18). Then the n-th denominator takes the form

$$z - i\kappa^\lambda g^{(n-1)} + \kappa^{2\theta} \sum_\nu \sum_{\nu'} \iint \frac{m_{\nu\nu'}^{(n)}(\boldsymbol{s}_n, \boldsymbol{s}_n'; \cdots; k/\kappa)\, d\boldsymbol{s}_n d\boldsymbol{s}_n'}{z - i\kappa^\lambda g^{(n)} + \ddots}. \tag{6·20}$$

This formulation is valid even for the paramagnetic region, where all the frequencies $\kappa^\lambda g^{(n)}$ vanish in the absence of magnetic field. Thus we can determine the validity of the dynamic scaling law proposed by Halperin and Hohenberg. If and only if $\eta = 0$, we have $\lambda = \theta$, (6·20) thus leading to the form

$$\Xi_{\boldsymbol{k}}^\alpha(t) = F_\alpha(t\kappa^\theta, k/\kappa) \tag{6·21}$$

with the critical index $\theta = (d+2)/2$, irrespective of the lower or upper critical region and of the transverse or longitudinal component. If $\eta \neq 0$, however, $\lambda \neq \theta$ and the dynamic scaling law in the above sense does not hold. As far as the paramagnetic region without magnetic field is concerned, however, the frequencies $\kappa^\lambda g^{(n)}$ vanish and the dynamic scaling law (6·21) holds with the critical index $\theta = (d + 2 + \eta)/2$, agreeing with (4·14).

These results differ from the other theories,[11),12)] which always lead to the frequency critical index $\lambda = (d + 2 - \eta)/2$. These theories, which ultimately assume short time expansions, do not deal with the long time behavior and thus cannot distinguish the essential difference between the frequencies and dampings in the macroscopic limit. It should be remembered that it was essential to extract the contribution from the long range correlations of spin fluctuations (or the correlations of small wave-number spin fluctuations) as the most dominant part whose asymptotic behavior can be treated with the static scaling laws, and this extraction became possible by dealing with the long time behavior in the macroscopic limit in order to take into account the life-time effects of the critical variables A_n in the vicinity of the critical point. Thus the difference between the frequency critical index λ and the damping critical index θ turns out to be meaningful.

In a similar manner, we can treat the antiferromagnetic case, which, however, will be discussed in a separate paper together with a study of the antiferromagnetic resonance line width.

§ 7. Concluding remarks

The first fundamental problem of the dynamic critical phenomena is to extract the most dominant part due to the critical fluctuation of the critical variables involved. In the static critical phenomena this seems to have been done successfully. In the relaxation and transport phenomena, the macroscopic motion dissipates its physical quantities to the microscopic thermal fluctuations. The dissipative processes depend on the microscopic structure of the system more sensitively than the static properties. Within the correct formalism of such irreversibility, we have to extract the anomalous part due to the critical fluctuations of macroscopic scale in the macroscopic limit.

To do this we employed the continued fraction expansion, which can be regarded as giving a general statement of the fluctuation-dissipation theorem. The most important properties of the continued fraction expansion are two-fold. (1) Its coefficients, the numerators M and the frequencies ω, are entirely determined by the static correlation functions. (2) The introduction of the irreversible character, namely, the analytic continuation of $\Xi(z)$ into the left half plane in the complex z plane can be done in a straightforward manner.[8),16)] This yielded a crucial difference from the moment method and its modifications.

All the fractions of the generalized continued fraction (2·47) have the form

$$\sum_{q}\sum_{q'} M_{\boldsymbol{q}\boldsymbol{q}'}\tau_{\boldsymbol{q}\boldsymbol{q}'}(z), \tag{7·1}$$

where $M_{\boldsymbol{q}\boldsymbol{q}'}$ and the real part of $\tau_{\boldsymbol{q}\boldsymbol{q}'}(z)$ represent something like the amplitude and the life time of the fluctuations of the corresponding A variables. As was discussed in (4·8) and below (5·4), if k, κ and $|z|$ are very small, then both of M and τ become anomalously large for the small values of $\boldsymbol{q}$ and $\boldsymbol{q}'$. Without the time factor $\tau_{\boldsymbol{q}\boldsymbol{q}'}(z)$, however, the anomaly would be weak and the sum would not have any dominant part. Due to the existence of this time factor, the sum can be approximated by the small wave number terms. For instance, let us take the isotropic Heisenberg ferromagnet above the critical point, and consider the damping function

$$\varphi_{\boldsymbol{k}}{}^{0}(z) = [N/(S_{\boldsymbol{k}}{}^{0}, S_{\boldsymbol{k}}{}^{0*})]L(z, \boldsymbol{k}), \tag{7·2}$$

where

$$L(z, \boldsymbol{k}) = \frac{1}{N}\int_0^\infty (f_{\boldsymbol{k}}{}^{0}(t), f_{\boldsymbol{k}}{}^{0*})\exp(-zt)\,dt, \tag{7·3}$$

$$= \frac{1}{N}\sum_{q}\sum_{q'} g_{\boldsymbol{q}}{}^{0k} g_{\boldsymbol{q}'}^{0k*}(A_{\boldsymbol{q}}{}^{0k}, A_{\boldsymbol{q}'}^{0k*})\tau_{\boldsymbol{q}\boldsymbol{q}'}(z). \tag{7·4}$$

Since $f_k{}^0=\dot{S}_k{}^0$, the static correlation of the random force is calculated to be

$$(f_k{}^0, f_k{}^{0*})/N = -(ik_BT/\hbar)\langle[S_k{}^0, \dot{S}^0_{-k}]\rangle ,$$
$$= (2J_0{}^k/3\hbar^2\beta J)\,\epsilon(T), \qquad (7\cdot5)$$

where we have assumed the nearest neighbor interaction and defined its average energy of one spin

$$\epsilon(T) \equiv 2J\langle \boldsymbol{S}_0\cdot\boldsymbol{S}_\delta\rangle . \qquad (7\cdot6)$$

Equation (7·5) does not show any anomalous increase. However, (7·4) does show the anomalous increase due to the anomalous increase of $\tau_{qq'}(z)$; from (6·15) and (6·20), we have

$$\tau_{qq'}(0) \sim \kappa^{-\theta}, \qquad (7\cdot7)$$

$$L(0, \boldsymbol{k}) \sim k^2\kappa^{-3(1-\eta)/2}. \qquad (7\cdot8)$$

A striking example of such a life-time effect can be seen also in the theory of the NMR line width near the critical point.[17),18)]

Thus the most dominant part was able to be extracted in the continued fraction expansion (2·47) in the long time limit. To study the most dominant part, we assumed the static scaling laws for the static correlation functions with small wave numbers. It was shown, however, that the dynamic scaling law does not hold in the ordered state if $\eta\neq0$. This was due to a difference between the frequencies and the damping functions. If $\eta\neq0$, then the dynamic scaling law in a wide sense did not hold also in the antiferromagnets above the critical point. This was due to a non-similarity between the kinematical slowing-down of the small wave number polarization and the critical slowing-down of the staggered polarization. As was shown in § 6, these conclusions were further confirmed without using the pair correlation approximation.

Appendix A

Derivation of the theorems quoted in § 2

Let us consider the time evolution of a dynamic quantity $A(t)$, starting from its equation of motion

$$dA(t)/dt = i\mathcal{L}A(t), \qquad (\mathrm{A}\cdot1)$$

where $\mathcal{L}$ is a linear operator. If A is a mechanical variable, then $\mathcal{L}$ is the Liouville operator L. If A is the random force of first or second order, then $\mathcal{L}$ is the linear operator defined by (2·31) or (2·35), respectively. The following formalism can be thus applied to any order of random force. Now, let us consider another dynamic quantity B and define the projection of a variable F onto the A axis through the variable B by

$$\mathcal{P}_AF = (F, B^*)\cdot(A, B^*)^{-1}\cdot A . \qquad (\mathrm{A}\cdot2)$$

This equation defines a linear projection operator $\mathcal{P}_A$, which satisfies the relation $(\mathcal{P}_A)^2=\mathcal{P}_A$, and the projection can be visualized geometrically by defining the Hilbert space of dynamic variables whose inner product of two variables F and G is given by (F, G^*). The damping theory developed in the previous paper[8] can be also applied to the present case by taking the projection operator (A·2). It would be instructive to quote here its mathematical structure. Let us separate $A(t)$ into the projective and vertical component with respect to the A axis;

$$A(t)=\Xi_{AB}(t)\cdot A+A'(t), \tag{A·3}$$

where

$$\Xi_{AB}(t)\equiv(A(t), B^*)\cdot(A, B^*)^{-1}, \tag{A·4}$$

$$A'(t)\equiv(1-\mathcal{P}_A)A(t). \tag{A·5}$$

From (A·1) we obtain an explicit expression for $A'(t)$ in the following manner. Operating $(1-\mathcal{P}_A)$ on (A·1) and using (A·3),

$$\frac{d}{dt}A'(t)-(1-\mathcal{P}_A)i\mathcal{L}A'(t)=\Xi_{AB}(t)\cdot f_A, \tag{A·6}$$

where

$$f_A\equiv(1-\mathcal{P}_A)i\mathcal{L}A. \tag{A·7}$$

This is integrated to yield

$$A'(t)=\int_0^t \Xi_{AB}(s)\cdot f_A(t-s)\,ds, \tag{A·8}$$

$$f_A(t)=\exp[(1-\mathcal{P}_A)i\mathcal{L}t](1-\mathcal{P}_A)i\mathcal{L}A. \tag{A·9}$$

Since $\mathcal{P}_A(1-\mathcal{P}_A)=0$, we have $\mathcal{P}_A f_A(t)=0$; namely, the random force $f_A(t)$ is orthogonal to the variable A. Differentiating (A·4) and then inserting (A·3) and (A·8), we obtain

$$\frac{d}{dt}\Xi_{AB}(t)=\Xi_{AB}(t)\cdot i\omega_{AB}-\int_0^t \Xi_{AB}(s)\cdot\varphi_{AB}(t-s)\,ds, \tag{A·10}$$

where

$$i\omega_{AB}\equiv(i\mathcal{L}A, B^*)\cdot(A, B^*)^{-1}, \tag{A·11}$$

$$\varphi_{AB}(t)\equiv-(i\mathcal{L}f_A(t), B^*)\cdot(A, B^*)^{-1}. \tag{A·12}$$

The Laplace transform of (A·10) thus leads to

$$\Xi_{AB}(z)=\frac{1}{z-i\omega_{AB}+\varphi_{AB}(z)}, \tag{A·13}$$

which agrees with (2·19). The foregoing treatment can be also applied to the many-variable case, where A and B are n-dimensional column matrices of independent variables. Then (A·2) denotes the projection into the n-dimensional subspace, and $\Xi_{AB}(t)$, ω_{AB}, and $\varphi_{AB}(t)$ are the square matrices, and the center dots denote the matrix multiplication.

Theorem 2: Let us introduce a projection operator $\mathcal{P}_B$ in the same way as $\mathcal{P}_A$;

$$\mathcal{P}_B G = (G, A^*) \cdot (B, A^*)^{-1} \cdot B, \tag{A·14}$$

which satisfies $(\mathcal{P}_B)^2 = \mathcal{P}_B$. Then we have

$$(\mathcal{P}_A F, G^*) = (F, [\mathcal{P}_B G]^*). \tag{A·15}$$

This equation means that $\mathcal{P}_A$ and $\mathcal{P}_B$ are hermitian conjugate to each other. We also introduce the hermitian conjugate propagator $\mathcal{L}'$ of $\mathcal{L}$ by

$$(\mathcal{L}F, G^*) = (F, [\mathcal{L}'G]^*). \tag{A·16}$$

Then the damping function (A·12) can be written as

$$\begin{aligned}(i\mathcal{L}f_A(t), B^*) &= -(f_A(t), [i\mathcal{L}'B]^*), \\ &= -(f_A(t), [(1-\mathcal{P}_B)i\mathcal{L}'B]^*),\end{aligned} \tag{A·17}$$

where use has been made of the identity $f_A(t) = (1-\mathcal{P}_A)f_A(t)$, which comes out of the relation $(1-\mathcal{P}_A)^2 = (1-\mathcal{P}_A)$. Substitution of (A·17) into (A·12) yields

$$\varphi_{AB}(t) = (f_A(t), f_B^*) \cdot (A, B^*)^{-1}, \tag{A·18}$$

where

$$f_B(t) \equiv \exp[(1-\mathcal{P}_B)i\mathcal{L}'t](1-\mathcal{P}_B)i\mathcal{L}'B. \tag{A·19}$$

Equation (A·18) agrees with (2·26). Since $\mathcal{P}_A f_A(t) = \mathcal{P}_B f_B(t) = 0$, (2·27) is readily obtained from the following theorem.

Theorem 3: Introducing the evolution operators $\mathcal{L}_1$ and $\mathcal{L}_1'$ by (2·29), and assuming that $\mathcal{P}_A F = \mathcal{P}_B G = 0$, we have

$$\begin{aligned}(\mathcal{L}_1 F, G^*) &= (\mathcal{L}F, G^*) = (F, [\mathcal{L}'G]^*) \\ &= (F, [(1-\mathcal{P}_B)\mathcal{L}'G]^*) = (F, [\mathcal{L}_1'G]^*),\end{aligned}$$

where (A·15) and (A·16) have been used. This is identical to (2·30).

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