Progress of Theoretical Physics, Vol. 34, No. 3, September 1965

A Continued-Fraction Representation of the Time-Correlation Functions

Hazime MORI

Research Institute for Fundamental Physics
Kyoto University, Kyoto

(Received April 28, 1965)

A continued-fraction expansion of the Laplace transform of the time-correlation functions is obtained, which enables us to express the generalized susceptibilities and the transport coefficients in terms of the static correlation functions of a set of quantities. This expansion has a different feature from the moment and cumulant expansions, and has a convenient form to introduce the long-time approximation as well as the short-time approximation. Its application to the anomalous relaxation and transport phenomena near the second-order phase transition points is discussed

An expansion formula is also obtained for the time evolution of dynamical quantities in order to describe the various modes of motion involved according to their characteristic time constants. These two expansions are closely related to the time-correlation function formalism of irreversible processes, and allow us to have physical intuition in calculating dissipative properties.

§ 1. Introduction

Dynamical properties of a macroscopic system can be expressed in terms of the time-correlation functions of appropriate physical variables. For instance, the electrical conductivity is the one-sided Fourier transform of the relaxation function of the electric current,[1] and the sound attenuation constant is the two-sided Fourier transform of the time-correlation function of the random force acting on the sound.[2] Therefore, it is important to develop a systematic method of investigating these time-correlation functions.

According to recent experiments, the relaxation and transport phenomena show interesting anomalous behaviors near the second-order phase transition points.[3] Near the critical points, the thermal fluctuations of some physical variables become anomalously large. On the basis of the fluctuation-dissipation theorem, therefore, we can expect that the dissipative properties also show anomalous behaviors near the critical points. From this point of view, we have studied the anomalous behavior of the damping constants of the inhomogeneous magnetization in ferromagnets[4] and of the line widths of the electron spin resonance in antiferromagnets.[5] These investigations were based on the expression for the damping constants of the magnetization in terms of the time-correlation function of the random torque acting on the spins, and the most important task was to extract some temperature-dependent factors which became anomalous near the critical points. In doing this we have assumed that the time-correlation function of the random torque decays in a Gaussian form. This assumption, however, is not valid when the temperature dependence of the static correlation function of the random torque is anomalous as in the case of the electron spin resonance in ferro- and antiferro-magnets. Therefore, it is quite desirable to extract the anomalous factors correctly without making any particular assumption of the decay form of the time-correlation function of the random torque. A similar situation also occurs when we wish to investigate anomalous transport phenomena such as the viscosity and sound attenuation in critical liquid solutions. The principal purpose of the present paper is therefore to develop such a scheme of extracting anomalous factors in the damping constants and the transport coefficients. This is done by generalizing a method of deriving a generalized Langevin equation of motion developed in a previous paper.[2)]

In § 2, we introduce an orthogonal set $\{f_j\}$, which plays an essential role in the present paper, and derive a new expansion formula for the description of the time evolution of dynamical quantities. The coefficients of this expansion are investigated in § 3, and we derive a hierarchy of equations relating a lower-order coefficient to a higher-order coefficient.

In § 4, a continued-fraction representation of the Laplace transform of the relaxation function is obtained, and its singularities in the complex z plane are investigated. As an application, we discuss the anomalous relaxation and transport phenomena near the critical points. Section 5 is devoted to some remarks, in which we discuss the Nyquist theorem on the voltage fluctuation in order to clarify the physical significance of the random forces. For illustration of the continued-fraction representation, we discuss simple examples for the long-time approximation in Appendix B.

§ 2. Expansion of $A(t)$ in terms of an orthogonal set

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

$$dA(t)/dt = iLA(t), \tag{2·1}$$

where L is the Liouville operator; namely, in the classical case, $iLA(t)$ represents the Poisson bracket of $A(t)$ with the Hamiltonian and, in the quantal case, the corresponding commutator. We assume that $A(t)$ denotes the deviation from its invariant part.

As in the previous paper,[2)] let us introduce a Hilbert space of dynamical variables whose invariant parts are set to be zero, and denote its inner product of two variables F and G by the parenthesis (F, G^*), where the asterisk denotes the Hermitian conjugate. The inner product is assumed to have the Liouville operator Hermitian;

$$(LF, G^*) = (F, [LG]^*). \tag{2·2}$$

Such an inner product is provided by

$$(F,\ G^*) \equiv \frac{1}{\beta}\int_0^\beta \langle \exp(\lambda\mathcal{H}) F \exp(-\lambda\mathcal{H}) G^* \rangle d\lambda, \tag{2·3}$$

where $\mathcal{H}$ is the Hamiltonian of the system and the angular brackets denote the average over the canonical ensemble with temperature $T=1/k_B\beta$, k_B being the Boltzmann constant. In the classical limit, Eq. (2·3) reduces to the correlation function $\langle FG^*\rangle$. The following formulation does not depend on the explicit form of the inner product. However, since the inner product (2·3) is most important from the physical point of view,[2] we adopt this definition (2·3) in the following. The quantity A, which denotes the value of $A(t)$ at time $t=0$, defines a vector in the Hilbert space. The projection of a vector G onto this A axis is given by

$$\mathcal{P}_0 G \equiv (G,\ A^*)\cdot(A,\ A^*)^{-1}\cdot A. \tag{2·4}$$

This equation defines a linear Hermitian operator $\mathcal{P}_0$, which is called the projection operator onto the A axis.

Now let us separate $A(t)$ into the projective and vertical components with respect to the A axis;

$$A(t) = \Xi_0(t)\cdot A + A'(t), \tag{2·5}$$

where

$$\Xi_0(t) \equiv (A(t),\ A^*)\cdot(A,\ A^*)^{-1}, \tag{2·6}$$

$$A'(t) \equiv (1-\mathcal{P}_0)A(t). \tag{2·7}$$

From Eq. (2·1) we obtain an explicit expression for $A'(t)$ in the following manner. Operating $(1-\mathcal{P}_0)$ on Eq. (2·1) and using Eq. (2·5),

$$dA'(t)/dt - iL_1 A'(t) = \Xi_0(t)\cdot f_1,$$

where

$$L_1 \equiv (1-\mathcal{P}_0)L, \tag{2·8}$$

$$f_1 \equiv iL_1 A. \tag{2·9}$$

This is integrated to yield

$$A'(t) = \int_0^t \Xi_0(s)\cdot f_1(t-s)\,ds, \tag{2·10}$$

where

$$f_1(t) \equiv \exp(iL_1 t) f_1. \tag{2·11}$$

Namely, the vertical component $A'(t)$ is the convolution of $\Xi_0(t)$ and $f_1(t)$. Therefore, taking its Laplace transform, we write

$$A(z)\equiv\int_0^\infty A(t)e^{-zt}\,dt=\Xi_0(z)\cdot[A+f_1(z)].\tag{2·12}$$

From Eq. (2·11) we have

$$(f_1(t),\ A^*)=0.\tag{2·13}$$

Namely, $f_1(t)$ always stays inside the hyperplane orthogonal to the A axis. Its evolution is governed by

$$df_1(t)/dt=iL_1\,f_1(t).\tag{2·14}$$

This equation has the same form as Eq. (2·1). We now proceed similarly to the foregoing procedure from Eq. (2·5) to Eq. (2·12). We first separate $f_1(t)$ into the projective and vertical components with respect to the vector f_1;

$$f_1(t)=\Xi_1(t)\cdot f_1+f_1'(t),\tag{2·15}$$

where

$$\Xi_1(t)\equiv(f_1(t),\ f_1^*)\cdot(f_1,f_1^*)^{-1},\tag{2·16}$$

$$f_1'(t)\equiv(1-\mathcal{P}_1)f_1(t),\tag{2·17}$$

where $\mathcal{P}_1$ is the projection operator onto the f_1 axis. Operating $(1-\mathcal{P}_1)$ on Eq. (2·14) and then integrating it, we finally obtain

$$f_1(z)=\Xi_1(z)\cdot[f_1+f_2(z)],\tag{2·18}$$

where

$$f_2(t)\equiv\exp(iL_2t)\,iL_2f_1,\tag{2·19}$$

$$L_2\equiv(1-\mathcal{P}_1)L_1.\tag{2·20}$$

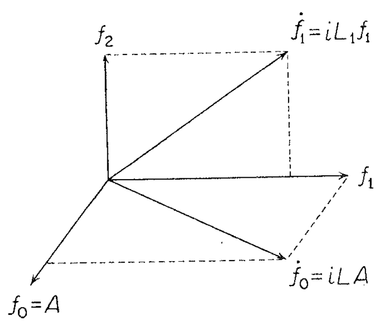

Fig. 1. Schematic relation between f_0, f_1 and f_2

The quantity f_2 is the vertical component of $\dot{f}_1$ with respect to the f_1 axis. Therefore, A, f_1 and f_2 are orthogonal to each other, as is shown in Fig. 1.

We next treat $f_2(t)$ similarly by introducing a new quantity $f_3(t)$. In this way we introduce a set of quantities $f_0(t)$, $f_1(t)$, $\cdots$ successively according to the equations

$$f_0(t)\equiv A(t),\tag{2·21}$$

$$f_j(t)\equiv\exp(iL_jt)\,iL_jf_{j-1},\quad(j\geq1),\tag{2·22}$$

where

$$L_j\equiv(1-\mathcal{P}_{j-1})L_{j-1},\quad L_0\equiv L,\tag{2·23}$$

where $\mathcal{P}_j$ is the projection operator onto the vector f_j. It should be noted that the time evolution of $f_j(t)$'s are governed by the propagators which are different from each other. Similarly to the derivation of Eq. (2·10) from Eq. (2·5), we obtain

$$f_j(t) = \Xi_j(t) \cdot f_j + \int_0^t \Xi_j(s) \cdot f_{j+1}(t-s)\, ds \tag{2·24}$$

where

$$\Xi_j(t) \equiv (f_j(t), f_j^*) \cdot (f_j, f_j^*)^{-1}. \tag{2·25}$$

The Laplace transform of Eq. (2·24) thus leads to

$$f_j(z) = \Xi_j(z) \cdot [f_j + f_{j+1}(z)]. \tag{2·26}$$

We can show from Eq. (2·22) that f_1 is orthogonal to f_0, f_2 is orthogonal to f_0 and f_1, and thus f_j is orthogonal to f_0, f_1, $\cdots f_{j-1}$, by observing that Eq. (2·23) can be written as

$$L_j = \prod_{i=0}^{j-1} (1-\mathscr{P}_i) L = [1 - \sum_{i=0}^{j-1} \mathscr{P}_i] L. \tag{2·27}$$

Therefore, the set $\{f_j\}$, $(j=0, 1, \cdots \infty)$, forms an orthogonal set. From Eq. (2·22) and Eq. (2·27) we also obtain

$$(f_j(t), f_i^*) = 0, \quad (i=0, 1, \cdots j-1). \tag{2·28}$$

Namely, $f_j(t)$ is always orthogonal to all the f_i's of lower indices.

Inserting Eq. (2·18) into Eq. (2·12) and then using Eq. (2·26) successively, we obtain the expansion of $A(z)$ in terms of the orthogonal set $\{f_j\}$; its inverse transform is

$$A(t) = \sum_{j=0}^{n-1} C_j(t) \cdot f_j + \int_0^t C_{n-1}(t-s) \cdot f_n(s)\, ds, \tag{2·29}$$

where the expansion coefficients are given by

$$C_j(z) = \Xi_0(z) \cdot \Xi_1(z) \cdots \Xi_j(z), \tag{2·30}$$

or

$$C_j(t) = \int_0^t dt_1 \Xi_0(t-t_1) \cdot \int_0^{t_1} dt_2 \Xi_1(t_1-t_2) \cdots$$

$$\times \int_0^{t_{j-1}} dt_j\, \Xi_{j-1}(t_{j-1}-t_j) \cdot \Xi_j(t_j). \tag{2·31}$$

Equation (2·5) together with Eq. (2·10) is one of the fundamental equations in the stochastic theory of generalized Brownian motion.[2),6)] Equation (2·29) is its generalization. It turns out from Eq. (2·28) that the linear sum of $C_j(t)$

in Eq. (2·29) describes the projection of $A(t)$ into the subspace spanned by $f_0, f_1, \cdots f_{n-1}$, namely, an average evolution of $A(t)$. Taking a larger n corresponds to a finer description of $A(t)$. The time-integral term describes a fluctuation from this average motion. The quantity $f_n(t)$ responsible for this fluctuation will be called the *n-th order random force* acting on the variable $A(t)$. It should be noted that the evolution of each $f_n(t)$ is governed by the different propagator. When $A(t)$ is the momentum of a Brownian particle suspended in a liquid, $f_1(t)$ is shown to represent the usual random force acting on the Brownian particle.[2)]

The foregoing treatment can be easily extended to the many-variable case. Then, A and A^* denote a n-dimensional column matrix of independent variables $A_1, \cdots A_n$, and its Hermitian row matrix, respectively, and Eq. (2·4) denotes the projection into the n-dimensional subspace spanned by the n variables.[2)] (A, A^*), $\Xi_j(t)$ and $C_j(t)$ are then square matrices, and the dots denote the matrix multiplication.

§ 3. A continued-fraction representation of $\Xi_j(z)$

Equation (2·24) relates $f_j(t)$ to $f_{j+1}(t)$. This leads to an equation relating $\Xi_j(t)$ to $\Xi_{j+1}(t)$, as will be shown in the following.

Let us denote the projection of $\dot{f}_j$ onto the f_j axis by $i\omega_j \cdot f_j$. Then we have

$$i\omega_j \equiv (\dot{f}_j, f_j^*) \cdot (f_j, f_j^*)^{-1}, \tag{3·1}$$

$$\dot{f}_j \equiv iL_j f_j = i\omega_j \cdot f_j + f_{j+1}. \tag{3·2a}$$

We thus obtain

$$df_j(t)/dt = i\omega_j \cdot f_j(t) + \exp(iL_j t) f_{j+1}. \tag{3·2b}$$

It will be shown in Appendix A that, if f and g are quantities orthogonal to the j-dimensional subspace spanned by the vectors $f_0, f_1, \cdots f_{j-1}$, then

$$(L_j f, g^*) = (f, [L_j g]^*). \tag{3·3}$$

This means that the propagator $\exp(iL_j t)$ is unitary inside the subspace orthogonal to the j-dimensional subspace defined above. Therefore, differentiating Eq. (2·25) and then inserting Eq. (3·2b), we obtain

$$d\Xi_j(t)/dt = i\omega_j \cdot \Xi_j(t) + (f_{j+1}, f_j(-t)^*) \cdot (f_j, f_j^*)^{-1}.$$

Inserting Eq. (2·24) into the second term and then using $\Xi_j(s)^* = \Xi_j(-s)$,*) we finally obtain

*) When A is a column matrix, it follows from Eq. (3·3) that

$$(f_j, f_j^*) \cdot \Xi_j(t)^* = \Xi_j(-t) \cdot (f_j, f_j^*).$$

This also leads to Eq. (3·4).

$$\frac{d}{dt}\Xi_j(t)=i\omega_j\cdot\Xi_j(t)-\int_0^t \Xi_{j+1}(t-s)\cdot\Delta_{j+1}^2\cdot\Xi_j(s)\,ds, \tag{3·4}$$

where

$$\Delta_j^2\equiv(f_j,f_j^*)\cdot(f_{j-1},f_{j-1}^*)^{-1}. \tag{3·5}$$

The Laplace transform of Eq. (3·4) leads to

$$\Xi_j(z)=\frac{1}{z-i\omega_j+\Xi_{j+1}(z)\cdot\Delta_{j+1}^2}. \tag{3·6}$$

This equation provides us with the hierarchy equations relating a Ξ function to a higher order Ξ function. In the one-variable case, applying Eq. (3·6) successively, we thus obtain

$$\Xi_j(z)=\cfrac{1}{z-i\omega_j+\cfrac{\Delta_{j+1}^2}{z-i\omega_{j+1}+\ddots\cfrac{\Delta_{n-1}^2}{z-i\omega_{n-1}+\Delta_n^2\Xi_n(z)}}} \tag{3·7}$$

where $j\leqq n-1$. If we let n tend to ∞, then we get an infinite continued fraction.[7]

Let us reduce Eq. (3·7) to the form

$$\Xi_j(z)=G_j^{(n)}(z)/F_j^{(n)}(z), \tag{3·8}$$

where $F_j^{(n)}(z)$ and $G_j^{(n)}(z)$ are such functions of z that, if we neglect the z dependence of $\Xi_n(z)$, then $F_j^{(n)}(z)$ is a polynomial of the order $(n-j)$ and $G_j^{(n)}(z)$ is a polynomial of the order $(n-j-1)$. From Eq. (3·6) we then have

$$G_j^{(n)}(z)=F_{j+1}^{(n)}(z), \tag{3·9}$$

$$F_j^{(n)}(z)=[z-i\omega_j]F_{j+1}^{(n)}(z)+\Delta_{j+1}^2F_{j+2}^{(n)}(z), \tag{3·10}$$

and

$$F_n^{(n)}(z)=1,\ F_{n-1}^{(n)}(z)=z-i\omega_{n-1}+\Delta_n^2\Xi_n(z). \tag{3·11}$$

Therefore, the expansion coefficients (2·30) take the form

$$C_j(z)=F_{j+1}^{(n)}(z)/F_0^{(n)}(z),\ (j\leqq n-1), \tag{3·12}$$

which leads to

$$A(z)=\frac{1}{F_0^{(n)}(z)}\Big[\sum_{j=0}^{n-1}F_{j+1}^{(n)}(z)f_j+f_n(z)\Big]. \tag{3·13}$$

This equation indicates that the time evolution of $A(t)$ is mainly determined by the zeros of $F_0^{(n)}(z)$ apart from the fluctuation part $f_n(z)$.

Finally let us derive an equation of motion for the n-th order random force $f_j(t)$. Inserting Eq. (3·6) into Eq. (2·26), we easily find

$$\frac{d}{dt}f_j(t)-i\omega_j\cdot f_j(t)+\int_0^t\varphi_j(t-s)\cdot f_j(s)\,ds=f_{j+1}(t), \qquad (3\cdot 14)$$

where

$$(f_{j+1}(t_1),\ f_{j+1}(t_2)^*)=\varphi_j(t_1-t_2)\cdot(f_j,f_j^*). \qquad (3\cdot 15)$$

If we take $j=0$, then Eq. (3·14) leads to a generalized Langevin equation of motion for $A(t)$ derived in a previous paper.[2] Therefore, we may visualize $f_{j+1}(t)$ as a generalized random force acting on $f_j(t)$.

§ 4. Singularities of the relaxation function $\Xi_0(z)$

According to Eq. (3·13) the time evolution of a dynamical quantity $A(t)$ is determined by the singularities of the Laplace transform of its relaxation function $\Xi_0(z)$ in the complex z plane. The admittance to an external perturbation can also be written in terms of the relaxation functions.[1] Thus the calculation of the relaxation functions is important for practical purposes as well as from the fundamental point of view. The continued-fraction representation (3·7) suggests a method of studying the relaxation functions. We now discuss this problem.

From Eq. (3·7) we obtain

$$\Xi_0(z)\equiv(A(z),\ A^*)/(A,\ A^*),$$
$$=\cfrac{1}{z-i\omega_0+\cfrac{\Delta_1^2}{z-i\omega_1+\ddots\cfrac{\Delta_{n-1}^2}{z-i\omega_{n-1}+\Delta_n^2\,\Xi_n(z)}}}. \qquad (4\cdot 1)$$

The parameters ω_j and Δ_j^2 are given by Eqs. (3·1) and (3·5) in terms of the static correlation functions of the quantities f_j's and $\dot{f}_j$'s. These quantities are defined by Eq. (2·22), and the use of Eq. (2·27) leads to

$$f_j=[1-\sum_{l=0}^{j-1}\mathcal{P}_l]\ iLf_{j-1}, \qquad (4\cdot 2a)$$

$$\dot{f}_j=[1-\sum_{l=0}^{j-1}\mathcal{P}_l]iLf_j. \qquad (4\cdot 2b)$$

These equations give explicit expressions for f_j and $\dot{f}_j$ in a straightforward manner; for instance,

$$f_0=A, \qquad (4\cdot 3)$$

$$f_1=\dot{A}-[(\dot{A},\ A^*)/(A,\ A^*)]A, \qquad (4\cdot 4)$$

$$f_2=\ddot{A}-[(\dot{A},\ A^*)/(A,\ A^*)+(\ddot{A},f_1^*)/(f_1,f_1^*)$$

$$-(\dot{A}, A^*)(\dot{A}, f_1^*)/(A, A^*)(f_1, f_1^*)]\dot{A}$$
$$-[(\ddot{A}, A^*)/(A, A^*)-(\dot{A}, A^*)^2/(A, A^*)^2$$
$$-(\dot{A}, A^*)(\ddot{A}, f_1^*)/(A, A^*)(f_1, f_1^*)$$
$$+(\dot{A}, A^*)^2(\dot{A}, f_1^*)/(A, A^*)^2(f_1, f_1^*)]A, \tag{4·5}$$

where $\dot{A}=iLA$. Thus Δ_j^2 and ω_j are related to the moments of the frequency distribution function of $\Xi_0(t)$,

$$\langle\omega^n\rangle\equiv\int_{-\infty}^{\infty}\omega^n P(\omega)\,d\omega=\frac{1}{i^n}\left[\frac{d^n\Xi_0(t)}{dt^n}\right]_{t=0}, \tag{4·6}$$

where

$$P(\omega)\equiv\frac{1}{\pi}\mathrm{Re}[\Xi_0(i\omega)]=\frac{1}{2\pi}\int_{-\infty}^{\infty}\Xi_0(t)\exp(-i\omega t)\,dt. \tag{4·7}$$

It turns out that Δ_j^2 is a function of the moments of lower order than $2j$, and ω_j is a function of those of lower order than $(2j+1)$.

When the first moment $\omega_0=\langle\omega\rangle$ is not zero, it is frequently more convenient to consider the function

$$\widetilde{\Xi}_j(z)\equiv\Xi_j(z+i\omega_0) \tag{4·8a}$$

and investigate its singularities around $z=0$. The inverse transform leads to

$$\Xi_0(t)=\exp(i\omega_0 t)\,\widetilde{\Xi}_0(t). \tag{4·8b}$$

If we take $n\to\infty$, then Eq. (4·1) leads to an infinite continued fraction. The time evolution of $\Xi_0(t)$ is determined by the singularities of this continued fraction in the complex z plane. The actual calculation of Δ_j^2 and ω_j of higher indices, however, is difficult except for simple systems. Useful approximations, however, will be obtained on the basis of this expression. If we go up to an appropriate $\Xi_n(z)$, then it frequently becomes possible to introduce either one of the following approximations for the $\Xi_n(z)$ or a combination of some of them:

(1) a long-time approximation
(2) a perturbation calculation
(3) a high or low temperature approximation
(4) a short-time approximation.

We now discuss these approximations.

The property of the infinite continued fraction depends upon what variable $A(t)$ and what region of value of z we are concerned with. When $\widetilde{A}(t)\equiv A(t)\exp(-i\omega_0 t)$ is a slowly-varying function of time in a certain time scale Δt, a $\widetilde{\Xi}_n(z)$ will be insensitive to z in a small region around the origin in the complex z plane. In such a case, we can neglect the z dependence of the $\widetilde{\Xi}_n(z)$;

$$\tilde{\Xi}_n(z) \cong \xi_n \equiv \int_0^\infty \Xi_n(t)\exp(-i\omega_0 t)\,dt. \tag{4·9}$$

Then from Eq. (3·8) we have

$$\tilde{\Xi}_0(z) \cong g_{n-1}(z)/g_n(z), \tag{4·10}$$

where $g_j(z)$ is a j-th order polynomial of z and is obtained from $F_{n-j}^{(n)}(z+i\omega_0)$ by inserting Eq. (4·9) into $\tilde{\Xi}_n(z)$ involved in $F_{n-j}^{(n)}$; namely,

$$g_j(z) = [z+i(\omega_0-\omega_{n-j})]g_{j-1}(z) + \Delta_{n-j+1}^2 g_{j-2}(z), \tag{4·11}$$

$$g_0(z)=1,\ g_1(z)=z+i(\omega_0-\omega_{n-1})+\lambda_{n-1}, \tag{4·12}$$

where we have introduced the quantities

$$\lambda_{j-1} \equiv \Delta_j^2\,\xi_j = \frac{\Delta_j^2}{i(\omega_0-\omega_j)+\lambda_j}. \tag{4·13}$$

If we neglect ω_0 in ξ_n, then our approximation is equivalent to neglecting the z dependence of $\Xi_n(z)$. It follows from Eqs. (4·11) and (4·12) that $g_j(z)$ and $g_{j-1}(z)$ do not have any common factor. Therefore the singularities of Eq. (4·10) are given by the zeros of $g_n(z)$. Denoting these zeros by $z_\alpha = i(\Omega_\alpha-\omega_0)-\gamma_\alpha$, ($\alpha=1, 2, \cdots n$), and taking the inverse Laplace transform of (4·10),

$$\Xi_0(t) \cong \sum_{\alpha=1}^n R_\alpha \exp[(i\Omega_\alpha-\gamma_\alpha)t], \tag{4·14}$$

where R_α is the residue of Eq. (4·10) at pole z_α. In this approximation, $C_j(z+i\omega_0)=g_{n-1-j}(z)/g_n(z)$, and Eq. (2·29) thus leads to

$$A(t) \cong \sum_{\alpha=1}^n \exp[(i\Omega_\alpha-\gamma_\alpha)t]$$

$$\times \{f^\alpha + C_{n-1}^\alpha \int_0^t \exp[-(i\Omega_\alpha-\gamma_\alpha)s]f_n(s)\,ds\}, \tag{4·15}$$

where

$$f^\alpha \equiv \sum_{j=0}^{n-1} C_j^\alpha f_j, \tag{4·16}$$

and C_j^α is the residue of $C_j(z+i\omega_0)$ at pole z_α.

The approximation (4·9) corresponds to the description of $\tilde{A}(t)$ in the time scale Δt distinctly larger than the decay time τ_n of $\Xi_n(t)$, and will be called the n-th order long-time approximation around $z=i\omega_0$. It thus turns out that (4·9) is valid if all of the n poles locate in the neighborhood of the center $z=i\omega_0$ of the semi-circle with the radius equal to $1/\tau_n$ in the left-half z plane. These poles represent slow processes, and the higher frequency components, which correspond to the remaining singularities of $\Xi_0(z)$, are represented by the n-th order random force $f_n(t)$ whose correlation time is much smaller than the relaxation times of the slow processes. If only $m(<n)$ poles locate near the center and the others are distributed away from them, then the m poles correctly represent m singularities of $\Xi_0(z)$ near the center, whereas the other $(n-m)$ poles are not insured to represent any singularities of $\Xi_0(z)$.

In Appendix B, we shall discuss simple examples for the long-time approximation. It will be concluded there in the case of $\omega_j=0$ that, if

$$\lambda_n \gg \lambda_{n-1}, \cdots \lambda_1, \lambda_0, \qquad (4\cdot17)$$

then $\Xi_0(z)$ has n poles in the neighborhood of the origin and these poles are determined from the n-th order long-time approximation around $z=0$.

In co-operative systems which undergo second-order phase transitions, the relaxation and transport phenomena also show anomalous properties near the critical points.[3] In treating these phenomena, it is essential to extract anomalous temperature-dependent factors correctly. Since the admittances and the kinetic coefficients are expressed in terms of the relaxation functions of appropriate variables, the expansion formula (4·1) will be useful for this problem. Near the critical points, some physical variables undergo critical fluctuations, giving rise to the anomalous increases of the specific heat and other compliance coefficients. These critical fluctuations are due to the fact that the static pair correlations of particles and spins become anomalously long-ranged near the critical points. The parameters ω_j and Δ_j^2 are written in terms of the static correlation functions of particles and spins. Thus some of ω_j and Δ_j^2 will have anomalous temperature dependence, leading to the anomalous relaxation and transport phenomena.

As an example let us consider the relaxation of the magnetization in ferro- and antiferro-magnets by taking as A the Fourier component of the longitudinal or transverse magnetization density with small wave number. Then Eq. (4·1) takes the form

$$\Xi_0(z) = \frac{1}{z - i\omega_0 + \lambda_0}, \qquad (4\cdot18)$$

where

$$\lambda_0 = \frac{\Delta_1^2}{i(\omega_0-\omega_1)+\lambda_1} = \frac{\Delta_1^2}{i(\omega_0-\omega_1)+\{\Delta_2^2/(i(\omega_0-\omega_2)+\lambda_2\}}. \qquad (4\cdot19)$$

Here we have used the first-order long-time approximation around $z=i\omega_0$. This is valid except for antiferromagnets below the Neél point. The real part of λ_0 gives the damping constant of the magnetization. First let us discuss the longitudinal magnetization with a small, but non-zero, wave number in ferromagnets. Then the anomalous temperature-dependence comes out from the factor (A, A^*) involved in Δ_1^2. Thus extracting this factor, we obtain

$$\lambda_0 \cong \frac{(A, A^*)_\infty}{(A, A^*)} \lambda_{0\infty} = \frac{[T\chi_k]_\infty}{T\chi_k} \lambda_{0\infty}, \tag{4·20}$$

where χ_k is the longitudinal magnetic susceptibility with wave number k. The subscripts ∞ mean to take their high temperature expression. When our interest lies in the phenomena below the Curie point, however, it is more convenient to take their low temperature expression on the basis of the spin wave calculation. The temperature dependence of Eq. (4·20) agrees with the previous result.[4] It should be noted here that, if we extract (f_1, f_1^*) also, then we get a more precise temperature dependence.

Next let us discuss the relaxation of the transverse uniform magnetization due to a small anisotropy energy in a weak magnetic field. Then, (A, A^*) and (f_1, f_1^*) have strong temperature dependence, and extracting these factors, we obtain

$$\lambda_0 \cong \frac{[T\chi_\perp]_\infty}{T\chi_\perp} \frac{(f_1, f_1^*)^2}{(f_1, f_1^*)_\infty^2} \lambda_{0\infty}, \tag{4·21}$$

where $\chi_\perp \equiv \beta(A, A^*)/2N$, N being the number of magnetic ions, is the transverse susceptibility. In antiferromagnets, the fluctuation of the random torque $(f_1, f_1^*)/N$ alone becomes anomalously large, thus leading to an anomalous increase of the damping constant λ_0 near the Neél point in agreement with experiments on the line width of the electron spin resonance.*) In ferromagnets, however, $\chi_\perp$ also becomes anomalously large, and its singularity is of higher order than that of $(f_1, f_1^*)/N$ in such a way that the damping constant λ_0 becomes anomalously small when the temperature goes down through the Curie point. This is in agreement with experiments on the ferromagnetic resonance. The details of these investigations will be reported in a forthcoming paper. It should be noted that we have extracted anomalous temperature-dependent factors without making any assumption about the decay form of the time-correlation function of the random torque $(f_1(t), f_1^*)$. This point is the most essential improvement of the previous theory,[4),5)] and enables us to investigate different kinds of phenomena, as will be shown in subsequent papers. In order to know the detailed form of λ_0 in terms of the interaction constants, however, we have to determine the decay form. This problem, however, is now not so difficult, since, after extracting the anomalous factors, we can employ a perturbation calculation in the spin wave region, and a short time approximation in the high temperature region.

§ 5. Some remarks

The time evolutions of the random forces $f_n(t)$, $n \geq 1$, are governed by the

*) The previous theory led to the power 3/2 instead of 2 in Eq. (4·21).[5] This deviation is due to the previous assumption of the Gaussian decay for the time-correlation of the random torque.

laws of motion different from the mechanical one. These evolutions, however, are physically meaningful, and, in fact, lower-order random forces are directly observable. As an example let us consider the thermal fluctuations of the electric current in a metal. It is well known that the conductivity tensor is given by[1)]

$$\sigma(\omega)=\beta\int_0^\infty (\boldsymbol{J}(t),\boldsymbol{J})\exp(-i\omega t)\,dt, \tag{5·1}$$

where $\boldsymbol{J}(t)$ is the electric current per unit volume, and its static correlation satisfies

$$\beta(J_\mu, J_\nu)=(ne^2/m)\delta_{\mu,\nu}, \tag{5·2}$$

where n is the number density of electrons. Therefore, taking $\boldsymbol{J}(t)$ as $A(t)$ and applying Eq. (3·6), we obtain

$$\sigma(\omega)=1/R(\omega), \tag{5·3}$$

$$R(\omega)=(m/ne^2)\,[i(\omega-\omega_0)+\varphi_0(i\omega)], \tag{5·4}$$

where

$$\varphi_0(i\omega)=\beta(m/ne^2)\int_0^\infty(\boldsymbol{f}_1(t),\boldsymbol{f}_1{}^*)\exp(-i\omega t)\,dt. \tag{5·5}$$

The $\boldsymbol{f}_1(t)$ is the random force responsible for the thermal fluctuation of the electric current, and from Eq. (3·14) we obtain

$$\frac{d}{dt}\boldsymbol{J}(t)-i\omega_0\cdot\boldsymbol{J}(t)+\int_0^\infty\varphi_0(s)\cdot\boldsymbol{J}(t-s)\,ds=\boldsymbol{f}_1(t) \tag{5·6}$$

for times t larger than the decay times of $\varphi_0(t)$. Therefore, taking the two-sided Fourier transform of Eq. (5·6), we obtain

$$R(\omega)\cdot\boldsymbol{J}(\omega)=\boldsymbol{V}(\omega), \tag{5·7}$$

where

$$\boldsymbol{V}(t)=(m/ne^2)\boldsymbol{f}_1(t). \tag{5·8}$$

The linear relation (5·7) shows that $\boldsymbol{V}(t)$ describes the time fluctuation of the voltage usually measured. This voltage fluctuation is related to the impedance $R(\omega)$ by Eqs. (5·4) and (5·5). Taking the real part of the diagonal element of Eq. (5·4), we have

$$\mathrm{Re}[R_{\mu\mu}(\omega)]=\frac{\beta}{2}\int_{-\infty}^{\infty}(V_\mu(t), V_\mu{}^*)\exp(-i\omega t)\,dt. \tag{5·9}$$

This leads to the Nyquist theorem on the voltage fluctuation.*) Thus Eqs. (5·7) and (5·8) provide us with a typical example showing that the random forces themselves have physical significance.

In the derivation of Eq. (5·7) it was essential to define the inner product by Eq. (2·3). This inner product satisfies the two requirements;

(a) The first term of Eq. (2·5), $\Xi_0(t)\cdot A$, describes the most probable path of the time evolution of $A(t)$ in the linear approximation.

(b) The second term, $A'(t)$, describes the linear response of $A(t)$ to its conjugate mechanical force $X(t)$ applied at time $t=0$ if one replaces the random force $f_1(t)$ in Eq. (2·10) by $X(t)$.

The requirement (a) was discussed in a previous paper.[2] Let us consider the external disturbance which produces the perturbation energy

$$\mathcal{H}' = -Q^*\cdot F(t), \quad \dot{Q}=A, \tag{5·10}$$

where $F(t)$ is a parameter and the dot of $\dot{Q}$ denotes its time rate with respect to the unperturbed Hamiltonian $\mathcal{H}$. Then the external force conjugate to A is defined by

$$X(t)\equiv(i/\hbar)\langle[\mathcal{H}', A]\rangle=\beta(A, A^*)\cdot F(t). \tag{5·11}$$

Then, with the aid of Kubo's theory of the linear response to mechanical disturbances,[1] we can easily show that the requirement (b) is also satisfied. Thus our generalized Langevin equation of motion, Eq. (3·10) of reference 2), or its integral representation (2·5) and (2·10) describes the mechanical and the thermal disturbances in a unified fashion.

The continued-fraction expansion (4·1) has a different feature from the moment expansion[9] and the cumulant expansion.[10] Namely, first, our expansion is entirely determined by the static correlation functions of f_j's, and $\dot{f}_j$'s. Thus the dissipative quantities associated are expressed in terms of the static correlation functions of particles and spins. As was discussed in § 4, this point is particularly important in studying the anomalous phenomena near the critical poins. The two-time Green's function method also has a similar feature[11]. Secondly, since Eq. (4·1) is exact for an arbitrary n, we can introduce a variety of approximations depending on the system concerned. Thus, for instance, we introduce a long-time approximation by neglecting the z dependence of an appropriate $\Xi_n(z)$, or a short-time approximation by treating the time dependence of an appropriate $\Xi_n(t)$ properly, or both at the same time. In this respect, our method differs from the two-time Green's function method, even though the latter

*) In the classical case, this is clear since the time integral is equal to the spectral density of the voltage fluctuation.[8] In the quantal case, this is also true in the isotropic case, since we obtain Re $[R(\omega)]=\pi\langle|V_\mu(\omega)|^2\rangle/E_\beta(\omega)$ by inserting Eq. (5·7) into Eq. (7·18) of reference 1). In a quantal anisotropic case, however, the situation becomes complicated and we may have a deviation from Nyquist's relation.

also produces a continued-fraction solution.[11),12)]

The quantity $\Xi_n(z)$ plays a similar role to the irreducible self-energy part of the thermal Green's function. In an infinite system, since $\Xi_n(t)$ decays in a finite time, $\Xi_n(z)$ is analytic in the right-half z plane. Therefore, the real and imaginary parts of $\Xi_n(i\omega)$ satisfy the Kramers-Kronig relations; for instance,

$$\mathrm{Im}[\Xi_n(i\omega)] = -\frac{1}{\pi}\int_{-\infty}^{\infty} \frac{\mathrm{Re}[\Xi_n(i\omega')]}{\omega-\omega'} d\omega', \tag{5·12}$$

where the principal part of the integral is to be taken. If A is a variable even or odd with respect to time reversal, then we have

$$[\omega_n]_{\boldsymbol{H}} = -[\omega_n]_{-\boldsymbol{H}}, \tag{5·13}$$

$$\mathrm{Im}[\Xi_n(i\omega)]_{\boldsymbol{H}} = -\mathrm{Im}[\Xi_n(-i\omega)]_{-\boldsymbol{H}}, \tag{5·14}$$

$$\mathrm{Re}[\Xi_n(i\omega)]_{\boldsymbol{H}} = \mathrm{Re}[\Xi_n(-i\omega)]_{-\boldsymbol{H}}, \tag{5·15}$$

where $-\boldsymbol{H}$ indicates the reversal of the external magnetic field $\boldsymbol{H}$. These time-reversal relations can be derived by extending a previous discussion of ω_0 and $\varphi_0(z)$.[2)] Their generalization to the many-variable case is obvious.

Acknowledgments

The author wishes to express his sincere gratitude to Professor R. Kubo for helpful discussions. This study was partially financed by the Scientific Research Fund of the Ministry of Education.

Appendix A

Derivation of Eq. (3·3)

Inserting Eq. (2·27) and then using that g is orthogonal to $\mathcal{P}_i Lf$,

$$(L_j f, g^*) = (Lf, g^*) = (f, [Lg]^*),$$

where Eq. (2·2) also has been employed. Since f is also orthogonal to $\mathcal{P}_i Lg$, this is equal to

$$= (f, [(1-\sum_{i=0}^{j-1}\mathcal{P}_i) Lg]^*).$$

Thus using Eq. (2·27) again, we obtain Eq. (3·3).

Appendix B

Simple examples for the long-time approximation

It would be instructive to discuss simple examples for the long-time approximation (4·9). For simplicity let us consider the case where

$$\omega_0 = \omega_1 = \cdots = 0.$$

Then it may be concluded from Eq. (4·13) that λ_j's are positive quantities. Introducing the third-order long-time approximation around $z=0$, we have

$$\Xi_0(z) \cong \frac{z^2 + \lambda_2 z + \Delta_2^2}{z^3 + \lambda_2 z^2 + (\Delta_1^2 + \Delta_2^2) z + \Delta_1^2 \lambda_2}. \tag{B·1}$$

This function has the three poles given by

$$\left.\begin{matrix} z_1 \\ z_2 \end{matrix}\right\} = -\frac{\lambda_2}{3} - \frac{1}{2}(u+v) \mp i\frac{\sqrt{3}}{2}(u-v), \tag{B·2}$$

$$z_3 = -\frac{\lambda_2}{3} + (u+v), \tag{B·3}$$

where

$$\left.\begin{matrix} u \\ v \end{matrix}\right\} = [B \pm \sqrt{R}]^{1/3}, \tag{B·4}$$

where

$$B = -\left(\frac{\lambda_2}{3}\right)^3 \left[1 - \frac{9\lambda_1}{2\lambda_2}\left(1 - \frac{2\lambda_0}{\lambda_2}\right)\right], \tag{B·5}$$

$$R = \lambda_1^2 \lambda_2 \left(\frac{\lambda_2}{3}\right)^3 \left[-\frac{1}{4} + \frac{\lambda_0}{\lambda_1} - \frac{2\lambda_0}{\lambda_2}\left(\frac{5}{2} - \frac{\lambda_0}{\lambda_2}\right) + \frac{\lambda_1}{\lambda_2}\left(1 + \frac{\lambda_0}{\lambda_2}\right)^3\right], \tag{B·6}$$

where λ_0 and λ_1 are given by Eq. (4·13) in terms of Δ_1^2, Δ_2^2 and λ_2. It follows from Eqs. (4·9) and (4·13) that $1/\lambda_j$ represents the mean decay time of $\Xi_j(t)$. Therefore, if $|z_1|$, $|z_2|$, $|z_3| \ll \lambda_2$, then these poles give the singularities of $\Xi_0(z)$ locating around the origin in the left-half z plane.

Now let us assume that

$$\lambda_2 \gg \lambda_1 = \Delta_2^2/\lambda_2, \text{ (i.e. } \lambda_2 \gg \Delta_2), \tag{B·7}$$

$$\lambda_2 \gg \lambda_0 = (\Delta_1/\Delta_2)^2 \lambda_2, \text{ (i.e. } \Delta_2 \gg \Delta_1). \tag{B·8}$$

Then Eqs. (B·5) and (B·6) lead to

$$B^{1/3} \cong -(\lambda_2/3) + (\lambda_1/2), \tag{B·9}$$

$$R \cong \lambda_2 (\lambda_2/3)^3 \Omega^2, \tag{B·10}$$

where

$$\Omega^2 = \lambda_1^2[(\lambda_0/\lambda_1) - (1/4)]. \tag{B·11}$$

Since $|B| \gg |\sqrt{R}|$, we thus obtain

$$z_1, z_2 \cong -(\lambda_1/2) \mp i\Omega, \tag{B·12}$$

$$z_3 \cong -\lambda_2 + \lambda_1. \tag{B·13}$$

The first two poles satisfy $|z_1|$, $|z_2| \ll \lambda_2$, and the third pole is well separated from these poles, as is shown in Fig. 2. In the time scale much larger than $1/\lambda_2$, therefore, we obtain

$$\Xi_0(t) \cong \exp(-\lambda_1 t/2)\left[\cos(\Omega t) + \frac{\lambda_1}{2\Omega}\sin(\Omega t)\right], \tag{B·14}$$

if $\lambda_1 < 4\lambda_0$, and

$$\Xi_0(t) \cong \frac{\lambda_1 - \gamma_1}{\gamma_2 - \gamma_1}\left[e^{-\gamma_1 t} - \frac{\lambda_1 - \gamma_2}{\lambda_1 - \gamma_1} e^{-\gamma_2 t}\right], \tag{B·15}$$

if $\lambda_1 > 4\lambda_0$, where

$$\gamma_1,\ \gamma_2 \equiv (\lambda_1/2)\{1 \mp [1-(4\lambda_0/\lambda_1)]^{1/2}\}. \tag{B·16}$$

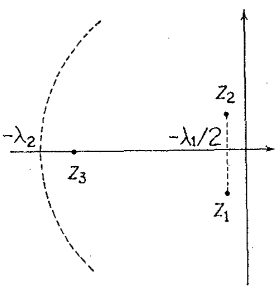

Fig. 2. $\lambda_2 \gg \lambda_1,\ \lambda_0$.

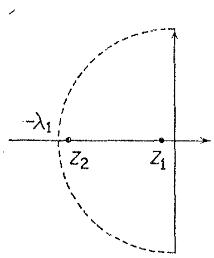

Fig. 3. $\lambda_1 \gg \lambda_0$.

Next let us assume that

$$\lambda_1 \gg \lambda_0 = \Delta_1^2/\lambda_1,\ \text{(i.e. } \lambda_1 \gg \Delta_1). \tag{B·17}$$

Then Eq. (B·16) reduces to

$$\gamma_1 \cong \lambda_0,\ \gamma_2 \cong \lambda_1 - \lambda_0. \tag{B·18}$$

In the time scale much larger than $1/\lambda_1$, therefore, we obtain

$$\Xi_0(t) \cong \exp(-\lambda_0\ t). \tag{B·19}$$

In this way, as λ_0/λ_1 reduces from a value larger than 1/4 to smaller values, the poles z_1 and z_2 change from Eq. (B·12) to Eq. (B·16) and finally to Eq. (B·18), as is shown in Figs. 2 and 3.

By extending the conditions (B·7—8) and (B·17), we may conclude that, if

$$\lambda_n \gg \lambda_{n-1},\ \cdots \lambda_1,\ \lambda_0, \tag{B·20}$$

then $\Xi_0(z)$ has n poles in the neighborhood of the origin inside the semi-circle with radius λ_n, and these poles are determined from the n-th order long-time approximation around $z=0$. A typical example satisfying Eq. (B·17) is the longitudinal magnetization with a small wave number in ferromagnets. The density fluctuation in a one-component system satisfies Eqs. (B·7—8) in the isothermal approximation. Therefore, in the neutron scattering by liquids and in the ultrasonic sound waves, the Brillouin doublet will undergo a similar transition to the change from Eq. (B·12) to Eq. (B·16) when the wave number or the frequency increases.

References

1) R. Kubo, J. Phys. Soc. Japan **12** (1957), 570.
2) H. Mori, Prog. Theor. Phys. **33** (1965), 423.
3) F. Johnson and A. Nethercot, Phys. Rev. **114** (1959), 705.
M. Ericson and B. Jacrot, J. Phys. Chem. Solids **13** (1960), 235.
M. Fixman, J. Chem. Phys. **36** (1962), 310.
A. Michels, J. Sengers and P. van der Gulik, Physica **28** (1962), 1216.
R. Hill and S. Ichiki, Phys. Rev. **128** (1962), 1140; **130** (1963), 150.
4) H. Mori and K. Kawasaki, Prog. Theor. Phys. **27** (1962), 529.
5) H. Mori and K. Kawasaki, Prog. Theor. Phys. **28** (1962), 971.
H. Mori, Prog. Theor. Phys. **30** (1963), 578.
6) S. Chandrasekhar, Rev. Mod. Phys. **15** (1943), 1.
7) H. Wall, *Continued Fractions* (D. Van Nostrand Company, Inc., Princeton, New Jersey, 1948).
8) L. Landau and E. Lifshitz, *Statistical Physics* (Pergamon Press, London-Paris, 1958), Chap. XII.
M. Wang and G. Uhlenbeck, Rev. Mod. Phys. **17** (1945), 323.
9) R. Kubo, in *Lectures in Theoretical Physics*, edited by W. Brittin and L. Dunham (Interscience Publishers, Inc., New York, 1959), Vol. 1.
10) R. Kubo, J. Phys. Soc. Japan **17** (1962), 1100.
11) K. Tomita and M. Tanaka, Prog. Theor. Phys. **29** (1963), 528.
12) T. Matsubara, Prog. Theor. Phys. **32** (1964), 50.