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Derivation of the stationary generalized Langevin equation

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Abstract. The stationary generalized Langevin equation is derived from the Liouville equation. The 'random force' involved in the obtained equation satisfies the equation of motion for a dynamical variable. The fluctuation–dissipation theorems are derived and written for the present equation. The relationship with Mori's generalized Langevin equation is given.

1. Introduction

A Brownian particle suspended in a liquid moves under the influence of the collisions of the molecules constituting the liquid. The collisions are considered stochastic, but their effect reduces the velocity of the Brownian particle on the average. In writing the equation of motion for the Brownian particle one separates the friction term, leading to this average reduction of the velocity, from the remaining part which is stochastic. With this consideration, one is led to the Langevin equation

$$m\frac{\mathrm{d}}{\mathrm{d}t}u(t) = -\gamma u(t)+f(t). \tag{1.1}$$

The first term on the right-hand side represents the systematic reduction of velocity and the coefficient γ is called the friction constant. The second term is the remaining part which is stochastic. In treating this equation, the second term is usually treated as a random variable and hence it is called the random force.

When one is concerned with the behaviours of the particle in a long time scale, the random force in the Langevin equation may be assumed to have only an instantaneous correlation. In this case, γ has been shown to be connected with a correlation of $f(t)$ by the fluctuation–dissipation theorem (Kubo 1966). On the other hand, in the calculation of the short time behaviours, one can no longer neglect the correlation of the random force in the time of the order of a collision duration. In order to treat such a case, Kubo (1966) proposed the following generalization of (1.1):

$$m\frac{\mathrm{d}}{\mathrm{d}t}u(t) = -\int_{-\infty}^{t}\mathrm{d}t'\gamma(t-t')u(t')+f(t) \tag{1.2}$$

a memory effect is introduced in the friction term. Then Kubo suggested that the function $\gamma(t)$ must satisfy the relation:

$$\gamma(t)\langle u(0)^2\rangle = \langle f(t)f(0)\rangle. \tag{1.3}$$

Mori (1965) investigated the Liouville equations of motion for a number of dynamical variables, $X_1(t)$, $X_2(t)$, ..., and $X_n(t)$, and showed that those equations can be expressed as the following generalized Langevin equation:

$$\frac{\mathrm{d}}{\mathrm{d}t}X_j(t) = \sum_{k=1}^{n}\Omega_{jk}X_k(t)-\sum_{k=1}^{n}\int_{t_0}^{t}\mathrm{d}t'\Gamma_{jk}(t-t')X_k(t')+R_j^M(t). \tag{1.4}$$

He derived the second fluctuation–dissipation theorem for this equation

$$\sum_{k=1}^{n} \Gamma_{jk}(t) \langle X_k(0), X_l^*(0) \rangle = \langle R_j^M(t), R_l^{M*}(0) \rangle. \tag{1.5}$$

Mori's equation is applicable after an initial time t_0, and the random force occurring in the equation depends on the arbitrarily chosen initial time t_0. Since the physical quantity 'random force' must be defined in such a way that it is independent of such an arbitrary time, it is desired to split the random force of this nature out of $R_j^M(t)$. In fact equation (1.2) proposed by Kubo is stationary and $f(t)$ in it seems to have such a property.

In the present paper, we start from the Liouville equations for the variables $X_1(t)$, $X_2(t)$, ..., $X_n(t)$, and we show that such a generalization is possible. As a result, we obtain

$$\frac{\mathrm{d}}{\mathrm{d}t} X_j(t) = \sum_{k=1}^{n} \Omega_{jk} X_k(t) - \sum_{k=1}^{n} \int_{-\infty}^{t} \mathrm{d}t\, \Gamma_{jk}(t-t') X_k(t') + R_j(t). \tag{1.6}$$

Comparison of (1.4) with (1.6) shows that Mori's $R_j^M(t)$ involves an integral from $-\infty$ to t_0, in addition to the random force $R_j(t)$. It is also shown that the function $\Gamma_{jk}(t)$ is related with the random force $R_j(t)$ by the second fluctuation dissipation theorem:

$$\sum_{k=1}^{n} \Gamma_{jk}(t) \langle X_k(0), X_l^*(0) \rangle = \langle R_j(t), R_l^*(0) \rangle. \tag{1.7}$$

When one treats the set of equations (1.6), one may try to solve these equations by considering only the statistical properties of $R_j(t)$, without recourse to their equations of motion. Such is the standpoint usually adopted in treating the Langevin equation (1.1). Thus it would be natural to call the term $R_j(t)$ the random force.

The set of derived equations (1.6) may also be used in the calculation of the two-time correlation function. The correlation function $\langle X_j(t), X_k^*(0) \rangle$ appearing in the linear response theory is an example of such a function: in that case $\langle A, B^* \rangle$ is defined by

$$\langle A, B^* \rangle = \frac{1}{\beta} \int_0^{\beta} \mathrm{d}\lambda \langle A \exp(-\lambda H) B^* \exp(\lambda H) \rangle. \tag{1.8}$$

As is easily seen, the correlation function defined by (1.8) satisfies the stationariness condition

$$\langle A(t), B^*(t') \rangle = \langle A(t-t'), B^*(0) \rangle \tag{1.9}$$

and the symmetry property

$$\langle A, B^* \rangle^* = \langle B, A^* \rangle. \tag{1.10}$$

When we are interested in the calculation of the correlation function $\langle X_j(t), X_k^*(0) \rangle$, we set up the generalized Langevin equation with the aid of the definition of the correlation function $\langle A, B^* \rangle$. If we are interested in another type of correlation function different from (1.8) we are also able to derive another generalized Langevin equation, corresponding to the correlation function. In that case it would be sufficient to require the properties (1.9) and (1.10) for the correlation function.

The contents of this paper are as follows. The next section is devoted to the introduction of the notations convenient for the description in the subsequent sections. Under the assumption of the relations (1.9) and (1.10), the stationary generalized Langevin equation, (1.6) or (3.19), is derived in § 3. Then in § 4 a derivation is given of the second fluctuation–dissipation theorem (1.7). The first fluctuation–dissipation theorem which gives the time-dependent correlation function of the dynamical variables is given in § 5. An alternative derivation of the same result is given in the Appendix. The relationship of (1.6) with Mori's equation (1.4) is discussed in § 6. § 7 is for conclusions.

2. Notations

The correlation function $\langle A, B\rangle$ considered in this paper is a quantity which is defined as a function bilinear in the matrix elements $A_{\mu\nu}$ and $B_{\mu\nu}$. For simplicity of notations the set of matrix elements $A_{\mu\nu}$ is denoted either by $\langle A$ or by $A\rangle$. The correlation function $\langle A, B\rangle$ is then a bilinear form of $\langle A$ and $B\rangle$. According to the concept of tetradics (Zwanzig 1966 and Morita *et al.* 1970), $\langle A$ and $B\rangle$ are called the bra-vector and the ket-vector, respectively. Each of them is a vector representation of the operator A or B. From this point of view, a relation $A+B = C$ can be expressed as $\langle A+\langle B = \langle C$ or as $A\rangle + B\rangle = C\rangle$. In the same way, relation $xA = B$ can be expressed as $x\langle A = \langle Ax = \langle B$ or as $A\rangle x = xA\rangle = B\rangle$, where x is an arbitrary complex number.

In order to describe the time development of the system, we now introduce a linear operator L which is multiplied to the right of a bra-vector or to the left of a ket-vector. The operator L is defined by

$$\langle AL = -\frac{\mathrm{i}}{\hbar}\langle [A, H] \tag{2.1a}$$

and

$$LB\rangle = \frac{\mathrm{i}}{\hbar}[H, B]\rangle \tag{2.1b}$$

where H is the Hamiltonian of the system under consideration. The Hamiltonian H is assumed to be independent of time. The equation of motion for the Heisenberg operator $A(t) = \exp(\mathrm{i}Ht/\hbar)A\exp(-\mathrm{i}Ht/\hbar)$ can now be expressed in a simple form as follows:

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle A(t) = \langle A(t)L \tag{2.2a}$$

and

$$\frac{\mathrm{d}}{\mathrm{d}t}A(t)\rangle = LA(t)\rangle. \tag{2.2b}$$

As the solutions of these equations with initial value $A(0) = A$, the bra- and ket-vectors corresponding to the operator $A(t)$ are expressed as follows:

$$\langle A(t) = \langle A\exp(Lt) \tag{2.3a}$$

and

$$A(t)\rangle = \exp(Lt)A\rangle. \tag{2.3b}$$

We require the stationary property for the correlation function; that means the relation

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle A(t+\tau), B(t)\rangle = 0 \tag{2.4}$$

for an arbitrary value of τ and for an arbitrary pair of operators A and B. With the aid of equations (2.2*a*) and (2.2*b*), one sees that this equation is equivalent to

$$\langle A(t+\tau)L, B(t)\rangle + \langle A(t+\tau), LB(t)\rangle = 0. \tag{2.5}$$

By putting $t = \tau = 0$, one obtains

$$\langle AL, B\rangle = -\langle A, LB\rangle \tag{2.6}$$

in general. Namely the correlation function changes sign when the operator L is carried from the bra-vector to the ket-vector.

In the next place, we consider a number of dynamical variables X_j and their Hermitian conjugates $X_j^*(j = 1, 2, ..., n)$. According to the notations used in the matrix calculations† we denote a column matrix composed of a sequence of bra-

† Let us illustrate the situation when the quantities X_j and X_k^* are made to correspond to vectors a_j with components $a_{j1}, a_{j2}, \ldots$, and a_{jl} and b_k with components $b_{k1}, b_{k2}, \ldots$, and b_{kl} $(j, k = 1, 2, \ldots, n)$. In this case a bra-vector $(a_j$ represents a row matrix and a ket-vector $b_k)$ represents a column matrix:

$$(a_j = (a_{j1}a_{j2} \ldots a_{jl})$$

and

$$b_k) = \begin{pmatrix} b_{k1} \\ b_{k2} \\ \vdots \\ b_{kl} \end{pmatrix}.$$

In place of the correlation function $\langle X_j, X_k^*\rangle$ we consider a product of $(a_j$ and $b_k)$, namely

$$(a_j \,.\, b_k) = \sum_{s=1}^{l} a_{js}b_{ks}.$$

Similarly $\langle X$ and $X^*\rangle$ are replaced by $(a$ and $b)$ defined as follows:

$$(a = \begin{pmatrix} (a_1 \\ (a_2 \\ \vdots \\ (a_n \end{pmatrix} \quad \begin{pmatrix} a_{11} & a_{12} & \ldots & a_{1l} \\ a_{21} & a_{22} & \ldots & a_{2l} \\ \ldots & \ldots & \ldots & \ldots \\ a_{n1} & a_{n2} & \ldots & a_{nl} \end{pmatrix}$$

and

$$b) = (b_1)b_2)\ldots b_n)) = \begin{pmatrix} b_{11} & b_{21} & \ldots & b_{n1} \\ b_{12} & b_{22} & \ldots & b_{n2} \\ \ldots & \ldots & \ldots & \ldots \\ b_{1l} & b_{2l} & \ldots & b_{nl} \end{pmatrix}.$$

The role of the matrix $\langle X, X^*\rangle$ is played by the following matrix:

$$(a \,.\, b) = \begin{pmatrix} (a_1 \\ (a_2 \\ \vdots \\ (a_n \end{pmatrix} . (b_1)b_2)\ldots b_n)) = \begin{pmatrix} (a_1 \,.\, b_1) & (a_1 \,.\, b_2) & \ldots & (a_1 \,.\, b_n) \\ (a_2 \,.\, b_1) & (a_2 \,.\, b_2) & \ldots & (a_2 \,.\, b_n) \\ \ldots & \ldots & \ldots & \ldots \\ (a_n \,.\, b_1) & (a_n \,.\, b_2) & \ldots & (a_n \,.\, b_n) \end{pmatrix}.$$

vectors $\langle X_1$, $\langle X_2$, ..., and $\langle X_n$ by $\langle X$. Similarly, a row matrix composed of a sequence of ket-vectors $X_1{}^*\rangle$, $X_2{}^*\rangle$, ..., and $X_n{}^*\rangle$ is written as $X^*\rangle$. An $n \times n$ matrix whose jkth element is given by the correlation function $\langle X_j, X_k{}^*\rangle$ is expressed as $\langle X, X^*\rangle$. In the same way, $\langle A, X^*\rangle$ represents a row matrix whose elements are $\langle A, X_1{}^*\rangle$, $\langle A, X_2{}^*\rangle$, ... , and $\langle A, X_n{}^*\rangle$, and $\langle X, A\rangle$ a column matrix whose elements are $\langle X_1, A\rangle$, $\langle X_2, A\rangle$, ..., and $\langle X_n, A\rangle$.

If the determinant of the matrix $\langle X, X^*\rangle$ is not zero, we can introduce an operator P which is multiplied to the right of a bra-vector or to the left of a ket-vector. The operator P is defined by

$$\langle AP = \langle A, X^*\rangle \,.\, \langle X, X^*\rangle^{-1} \,.\, \langle X \tag{2.7a}$$

and

$$PB\rangle = X^*\rangle \,.\, \langle X, X^*\rangle^{-1} \,.\, \langle X, B\rangle \tag{2.7b}$$

where $\langle X, X^*\rangle^{-1}$ represents an inverse matrix of $\langle X, X^*\rangle$. The products of matrices appearing on the right-hand sides of the above equations are considered to be calculated by the usual rules on the matrix product. It is straightforward to show from the above definition, that $\langle AP^2 = \langle AP$ and $P^2A\rangle = PA\rangle$ for an arbitrary operator A. These relations mean that P is a projection operator.

If we substitute $\langle X_j$ and $X_k{}^*\rangle$, respectively, in place of $\langle A$ and $B\rangle$ of equations (2.7a) and (2.7b), we have

$$\langle X_j P = \langle X_j \qquad (j = 1, 2, ..., n) \tag{2.8a}$$

and

$$PX_k{}^*\rangle = X_k{}^*\rangle \qquad (k = 1, 2, ..., n). \tag{2.8b}$$

It will be natural to describe the above equations in terms of the matrices $\langle X$ and $X^*\rangle$ as follows:

$$\langle XP = \langle X \tag{2.9a}$$

and

$$PX^*\rangle = X^*\rangle. \tag{2.9b}$$

The projection operator P satisfies the following identity:

$$\langle AP, B\rangle = \langle A, PB\rangle \tag{2.10}$$

which is easily confirmed by using (2.7a) on the left-hand side and (2.7b) on the right-hand side.

In this place, we define a row matrix $\tilde{X}\rangle$, whose elements are $\tilde{X}_1\rangle$, $\tilde{X}_2\rangle$, ..., and $\tilde{X}_n\rangle$, by the relation.

$$\tilde{X}\rangle = X^*\rangle \,.\, \langle X, X^*\rangle^{-1}. \tag{2.11}$$

Then equations (2.7a) and (2.7b) are rewritten as follows:

$$\langle AP = \langle A, \tilde{X}\rangle \,.\, \langle X \tag{2.12a}$$

and

$$PB\rangle = \tilde{X}\rangle \,.\, \langle X, B\rangle. \tag{2.12b}$$

It is obvious from the definition (2.11) that

$$\langle X, \tilde{X}\rangle = 1. \tag{2.13}$$

The more explicit forms of (2.12a), (2.12b) and (2.13) are given by†

$$\langle AP = \sum_{j=1}^{n} \langle A, \tilde{X}_j \rangle \langle X_j \tag{2.14a}$$

$$PB \rangle = \sum_{k=1}^{n} \tilde{X}_k \rangle \langle X_k, B \rangle \tag{2.14b}$$

and

$$\langle X_j, \tilde{X}_k \rangle = \delta_{jk}. \tag{2.15}$$

(The notations introduced in the present section are summarized in table 1.)

Table 1. Summary of the notations

Notation	Operator
$\langle A$	A
$A^* \rangle$	A^*
$\langle AL$	$\frac{-\mathrm{i}}{\hbar} [A, H]$
$LA^* \rangle$	$\frac{\mathrm{i}}{\hbar} [H, A^*]$
$\langle AP$	$\sum_{k=1}^{n} \langle A, \tilde{X}_k \rangle X_k$
$PA^* \rangle$	$\sum_{k=1}^{n} \tilde{X}_k \langle X_k, A^* \rangle$

† The above statements will be understood if the example used in the previous footnote is considered. Since (a and b) are associated to $\langle X$ and $X^* \rangle$, the equations corresponding to (2.11), (2.14a), and (2.14b) are

$$\tilde{b}) = b) \,.\, (a \,.\, b)^{-1}$$

$$(rP = \sum_{j=1}^{n} (r \,.\, \tilde{b}_j)\, (a_j$$

and

$$Ps) = \sum_{k=1}^{n} \tilde{b}_k)\, (a_k \,.\, s).$$

The above relations show that the projection operator P in this example is formally written as

$$P = \sum_{j=1}^{n} \tilde{b}_j)\, (a_j.$$

In this case equation (2.15) becomes

$$(a_j \,.\, \tilde{b}_k) = \delta_{jk}.$$

It will be valuable to notice that P is a projection operator in the oblique coordinate system. P projects (r to the (hyper) plane spanned by (a_1, (a_2, ..., and (a_n and s) to the (hyper) plane spanned by $\tilde{b}_1$), $\tilde{b}_2$), ..., and $\tilde{b}_n$); cf. a footnote in a paper by Morita *et al.* (1970). As the latter (hyper) plane is spanned by b_1), b_2), ..., and b_n) one can say that P projects s) to the (hyper) plane spanned by the latter set $\{b_k)\}$.

Finally it is noticed that the projection operator P is used in the next section to divide a variable $\langle A$ into two parts as follows

$$\langle A = \langle AP + \langle A(1-P). \tag{2.16}$$

As is easily seen from (2.8*b*) and (2.10), the first term $\langle AP$ has the same correlation with $X_k^*\rangle$ as the left-hand side $\langle A$, i.e.,

$$\langle AP, X_k^*\rangle = \langle A, X_k^*\rangle. \tag{2.17}$$

$\langle AP$ is a linear combination of $\langle X_1$, $\langle X_2$, ..., and $\langle X_n$. The remaining term $\langle A(1-P)$ is the part which has no correlation with $X_k^*\rangle$.

3. Stationary generalized Langevin equation

The notations introduced in the previous section are used in this and the subsequent sections of this paper. The following arguments are based on the requirements (2.6) and (1.10) on the correlation functions: namely, the stationariness property

$$\langle AL, B\rangle = -\langle A, LB\rangle \tag{3.1}$$

and the symmetry property

$$\langle A, B^*\rangle^* = \langle B, A^*\rangle \tag{3.2}$$

where the asterisks denote the complex conjugate of a number or the Hermitian conjugate of an operator. If $\langle A$ in equation (3.1) is a column matrix composed of bra-vectors $\langle A_1$, $\langle A_2$, ..., and $\langle A_n$, $\langle AL$ means the column matrix which is composed of $\langle A_1L$, $\langle A_2L$, ..., and $\langle A_nL$. The situation is the same for $LB\rangle$ if $B\rangle$ is a row matrix. A similar property for $\langle AP$ and $PB\rangle$ follows from the definition of P.

We consider a number of the Heisenberg operators $X_1(t)$, $X_2(t)$, ..., and $X_n(t)$, which are expressed by bra-vectors:

$$\langle X_j(t) = \langle X_j \exp(Lt) \equiv \langle X_j T(t) \tag{3.3}$$

cf. (2.3*a*). We first take attention to the operator

$$T(t) = \exp(Lt). \tag{3.4}$$

Its time-derivative is expressed as

$$\frac{\mathrm{d}}{\mathrm{d}t} T(t) = LT(t). \tag{3.5}$$

With the aid of the projection operator P, we divide L into two parts, cf. (2.16):

$$\frac{\mathrm{d}}{\mathrm{d}t} T(t) = LPT(t) + L(1-P)T(t). \tag{3.6}$$

We solve this differential equation by treating the first term on the right-hand side as the inhomogeneous part, where an arbitrary time t_0 is chosen as the initial time. Then we have

$$T(t) = \exp\{L(1-P)(t-t_0)\}T(t_0) + \int_{t_0}^{t} \mathrm{d}t' \exp\{L(1-P)(t-t')\}LPT(t'). \tag{3.7}$$

Substituting this expression in the second term on the right hand side of (3.6), we obtain

$$\frac{\mathrm{d}}{\mathrm{d}t} T(t) = LPT(t) + L(1-P)\exp\{L(1-P)(t-t_0)\}T(t_0) + L(1-P)\int_{t_0}^{t} \mathrm{d}t' \exp\{L(1-P)(t-t')\}LPT(t'). \tag{3.8}$$

We take a time-derivative of (3.3), and substitute equations (3.8) and (2.14*a*). As a result, we obtain the following equation of motion for $X_j(t)$:

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle X_j(t) = \sum_{k=1}^{n} \Omega_{jk} \langle X_k(t) + \langle R_j^{(t-t_0)}(t) - \sum_{k=1}^{n} \int_{t_0}^{t} \mathrm{d}t' \Gamma_{jk}(t-t') \langle X_k(t') \tag{3.9}$$

where

$$\Omega_{jk} = \langle X_j L, \tilde{X}_k \rangle \tag{3.10}$$

$$\langle R_j^{(t-t_0)}(t) = \langle R_j^{(t-t_0)} \exp(Lt) \tag{3.11}$$

$$\langle R_j^{(t-t_0)} = \langle X_j L(1-P)\exp\{L(1-P)(t-t_0)\}\exp\{-L(t-t_0)\} \tag{3.12}$$

and

$$\Gamma_{jk}(t) = -\langle X_j L(1-P)\exp\{L(1-P)t\}L, \tilde{X}_k \rangle. \tag{3.13}$$

Multiplying $\langle X_k, X_l^* \rangle$ to (3.10) and (3.13) and summing over k, one obtains

$$\sum_{k=1}^{n} \Omega_{jk} \langle X_k, X_l^* \rangle = \langle X_j L, X_l^* \rangle \tag{3.14}$$

and

$$\sum_{k=1}^{n} \Gamma_{jk}(t) \langle X_k, X_l^* \rangle = -\langle X_j L(1-P)\exp\{L(1-P)t\}L, X_l^* \rangle \tag{3.15}$$

where use is made of the relation (2.8*b*).

Equation (3.9) is the equation of motion for the Heisenberg operator $X_j(t)$ and hence the right-hand side takes the same value independent of the arbitrary initial time t_0. We assume that $\Gamma_{jk}(t)$ decays to zero when t becomes large enough. If t_0 is chosen such that $t-t_0$ is large enough, then the time t_0 in the lower limit of the integral in the last term of (3.9) may be set equal to $-\infty$. Now all the terms other than the second term on the right-hand do not depend on t_0. Hence the second term also must be convergent. We write the limiting value of $R_j^{(t-t_0)}$ as R_j:

$$\langle R_j = \lim_{t\to\infty} \langle X_j L(1-P)\exp\{L(1-P)t\}\exp(-Lt). \tag{3.16a}$$

From table 1, the corresponding ket-vector is given by

$$R_k^* \rangle = \lim_{t\to\infty} \exp(-Lt)\exp\{(1-P)Lt\}(1-P)LX_k^* \rangle. \tag{3.16b}$$

As a consequence, one obtains

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle X_j(t) = \sum_{k=1}^{n} \Omega_{jk} \langle X_k(t) - \sum_{k=1}^{n} \int_{-\infty}^{t} \mathrm{d}t' \Gamma_{jk}(t-t') \langle X_k(t') + \langle R_j(t) \tag{3.17}$$

where Ω_{jk} and $\Gamma_{jk}(t)$ are given by (3.14) and (3.15), and $\langle R_j(t)$ by

$$\langle R_j(t) = \langle R_j \exp(Lt) \tag{3.18}$$

with (3.16).

Equation (3.17) is the stationary generalized Langevin equation, which we desired. It should be noticed that the random force $\langle R_j(t)$ in this equation is expressed as (3.18). It means that it is the Heisenberg operator corresponding to a dynamical variable R_j; Note the fact that the corresponding quantity in Mori's equation was not so.

In this place we regard $\langle X_j(t)$ and $\langle R_j(t)$ as the jth components of column vectors $\langle X(t)$ and $\langle R(t)$, respectively, and $X_j^*(t)\rangle$ and $R_j^*(t)\rangle$ as the jth components of row vectors $X^*(t)\rangle$ and $R^*(t)\rangle$, respectively. Ω_{jk} and $\Gamma_{jk}(t)$ are the jkth elements of matrices Ω and $\Gamma(t)$, respectively. Then (3.17) is written as

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle X(t) = \Omega \,.\, \langle X(t) - \int_{-\infty}^{t} \mathrm{d}t' \,\Gamma(t-t') \,.\, \langle X(t') + \langle R(t) \tag{3.19}$$

and the equations (3.14) and (3.15) are expressed as

$$\Omega \,.\, \langle X, X^* \rangle = \langle XL, X^* \rangle \tag{3.20}$$

$$\Gamma(t) \,.\, \langle X, X^* \rangle = -\langle XL(1-P) \exp\{L(1-P)t\}L, X^* \rangle. \tag{3.21}$$

4. Fluctuation–dissipation theorem

It is shown in this section that the 'friction function' $\Gamma_{jk}(t)$ is related with the correlation of random forces $R_j(t)$.

For this purpose, (3.15) is rewritten as follows with the aid of the properties (2.10) and (3.1):

$$\sum_{k=1}^{n} \Gamma_{jk}(t) \langle X_k, X_l^* \rangle = \langle X_j L(1-P) \exp\{(1-P)L(1-P)(t-t_0)\} \exp\{-L(t-t_0)\}$$
$$\times \exp(Lt), \exp(Lt_0) \exp\{-(1-P)L(1-P)t_0\}(1-P)LX_l^* \rangle. \tag{4.1}$$

This equation holds for an arbitrary value of t_0. If t_0 is tended to minus infinity, the factor before the comma converges to $\langle R_j(t)$ and the factor after that converges to $R_l^*\rangle$; cf. (3.16*a*) and (3.16*b*). As a result, one obtains

$$\sum_{k=1}^{n} \Gamma_{jk}(t) \langle X_k, X_l^* \rangle = \langle R_j(t), R_l^*(0) \rangle. \tag{4.2}$$

In the matrix notation, (4.2) is written as follows:

$$\Gamma(t) \,.\, \langle X, X^* \rangle = \langle R(t), R^*(0) \rangle. \tag{4.3}$$

As stressed at the end of the preceding section, $R_j(t)$ is the Heisenberg operator corresponding to the operator R_j. It follows from this fact that the correlation function $\langle R, R^* \rangle$ satisfies the following equation:

$$\langle \dot{R}, R^* \rangle^* = \langle R, \dot{R}^* \rangle = -\langle \dot{R}, R^* \rangle \tag{4.4}$$

where a dot on an operator denotes the time-derivative of the operator. Now the diagonal elements $\langle R_j, R_j^* \rangle$ are pure imaginary. The above arguments can be applied to classical systems. In that case, R_j, R_j^*, and $\langle \dot{R}_j, R_k^* \rangle$ are real for ordinary choices of variables X_j, and hence

$$\langle \dot{R}_j, R_j^* \rangle = 0 \qquad \text{(classical).} \tag{4.5}$$

5. Correlation functions of dynamical variables

We take a correlation of equation (3.17) with $X_l^*(0)$ and obtain

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle X_j(t), X_l^*(0) \rangle = \sum_{k=1}^{n} \Omega_{jk} \langle X_k(t), X_l^*(0) \rangle - \sum_{k=1}^{n} \int_{-\infty}^{t} \mathrm{d}t' \Gamma_{jk}(t-t') \langle X_k(t'), X_l^*(0) \rangle + \langle R_j(t), X_l^*(0) \rangle. \tag{5.1}$$

It is noticed that a part of the contribution of the second term on the right hand side cancels exactly with the last term:

$$\sum_{k=1}^{n} \int_{-\infty}^{0} \mathrm{d}t' \Gamma_{jk}(t-t') \langle X_k(t'), X_l^*(0) \rangle = \langle R_j(t), X_l^*(0) \rangle. \tag{5.2}$$

A proof is achieved by substituting the definition (3.13) of $\Gamma_{jk}(t)$ into the left hand side. Then one has

$$\begin{aligned}
&\sum_{k=1}^{n} \int_{-\infty}^{0} \mathrm{d}t' \Gamma_{jk}(t-t') \langle X_k(t'), X_l^*(0) \rangle \\
&= - \sum_{k=1}^{n} \int_{-\infty}^{0} \mathrm{d}t' \langle X_j L(1-P) \exp\{L(1-P)(t-t')\} L, \tilde{X}_k \rangle \\
&\quad \times \langle X_k \exp(Lt'), X_l^* \rangle \\
&= - \int_{-\infty}^{0} \mathrm{d}t' \langle X_j L(1-P) \exp\{L(1-P)(t-t')\} LP \exp(Lt'), X_l^* \rangle \\
&= - \langle X_j L(1-P) \exp\{L(1-P)(t-t')\} \exp(Lt'), X_l^* \rangle \Big|_{t'=-\infty}^{t'=0} \\
&= \langle R_j(t), X_l^*(0) \rangle - \langle R_j^{(t)}(t), X_l^*(0) \rangle
\end{aligned} \tag{5.3}$$

where use has been made of the definitions (3.11) and (3.18). As will be seen from (2.8*b*) and (2.10), the last term of the above equation vanishes, and hence we have proved (5.2).

By substituting (5.2) into (5.1), one obtains

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle X_j(t), X_l^*(0) \rangle = \sum_{k=1}^{n} \Omega_{jk} \langle X_k(t), X_l^*(0) \rangle - \sum_{k=1}^{n} \int_{0}^{t} \mathrm{d}t' \Gamma_{jk}(t-t') \langle X_k(t'), X_l^*(0) \rangle \tag{5.4}$$

or, in the matrix form,

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle X(t), X^*(0) \rangle = \Omega \,.\, \langle X(t), X^*(0) \rangle - \int_{0}^{t} \mathrm{d}t' \Gamma(t-t') \,.\, \langle X(t'), X^*(0) \rangle. \tag{5.5}$$

In this place we introduce the Laplace transform of $\langle X(t), X^*(0)\rangle$:

$$\langle X, X^* \rangle_p = \int_0^\infty \mathrm{d}t \exp(-pt) \langle X(t), X^*(0) \rangle. \tag{5.6}$$

Equation (5.5) is easily solved as follows:

$$\langle X(t), X^*(0) \rangle = \frac{1}{2\pi \mathrm{i}} \int_{-\mathrm{i}\infty}^{\mathrm{i}\infty} \mathrm{d}p \langle X, X^* \rangle_p \exp(pt) \tag{5.7}$$

$$\langle X, X^* \rangle_p = \frac{1}{p - \Omega + \Gamma_p} \cdot \langle X(0), X^*(0) \rangle \tag{5.8}$$

where Γ_p is the Laplace transform of $\Gamma(t)$.

In the above derivation of the result (5.7) with (5.8), we take advantage of the relation (5.2). If this relation is not used, the same result is attained by taking correlations of equation (3.17) with $X_l^*(0)\rangle$ and $R_l^*(0)\rangle$ and taking a Fourier transform, after using various relations between the correlation functions. The derivation is sketched in the Appendix.

6. Relation with Mori's generalized Langevin equation

The equation (3.9) becomes Mori's generalized Langevin equation when t_0 is set equal to zero. In this case we see from (2.8*b*) and (2.10) that

$$\langle R_j^{(t)}(t), X_l^*(0) \rangle = 0. \tag{6.1}$$

When we start, alternatively, with the stationary Langevin equation (3.17), it is supposed that Mori's equation is obtained from (3.17) simply by putting

$$\langle R_j^M(t) = \langle R_j(t) - \sum_{k=1}^{n} \int_{-\infty}^{0} \mathrm{d}t' \Gamma_{jk}(t-t') \langle X_k(t') \tag{6.2}$$

(cf. Kubo 1966). Then (3.17) reduces to

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle X_j(t) = \sum_{k=1}^{n} \Omega_{jk} \langle X_k(t) - \sum_{k=1}^{n} \int_0^t \mathrm{d}t' \Gamma_{jk}(t-t') \langle X_k(t') + \langle R_j^M(t). \tag{6.3}$$

Equation (5.2) shows that the correlation of $R_j^M(t)$ defined by (6.2) with $X_l^*(0)$ is zero:

$$\langle R_j^M(t), X_l^*(0) \rangle = 0. \tag{6.4}$$

This shows that equation (6.3) thus obtained is equivalent to Mori's equation. We expect from equations (6.1) and (6.4) that $R_j^M(t)$ is equivalent to $R_j^{(t)}(t)$; In fact, this is proved if all the terms in equation (5.3) are written without the factor $X_l^*\rangle$. It is well known that the fluctuation–dissipation theorem

$$\sum_{k=1}^{n} \Gamma_{jk}(t) \langle X_k, X_l^* \rangle = \langle R_j^M(t), R_l^{M*}(0) \rangle$$

follows from the equations (6.3), (6.4), (3.1) and (3.2); see, for example, Fukui and Morita (1970). When $R_j^M(t)$ is identified with $R_j^{(t)}(t)$, this relation is also derived from the definition of $\Gamma(t)$ as given by (3.15): namely

$$\begin{aligned} \sum_{k=1}^{n} \Gamma_{jk}(t) \langle X_k, X_l^* \rangle &= \langle X_j L(1-P) \exp\{L(1-P)t\}, (1-P)LX_l^* \rangle \\ &= \langle R_j^{(t)}(t), R_l^{(0)*}(0) \rangle. \end{aligned}$$

7. Conclusions

The equations of motion are written for the Heisenberg operators of a number of dynamical variables, $X_1(t)$, $X_2(t)$, ..., and $X_n(t)$. The equations are transformed by using a suitable definition of the correlation $\langle X_j(t), X_k^*(0)\rangle$. As a result, we obtain the stationary generalized Langevin equation:

$$\frac{\mathrm{d}}{\mathrm{d}t}X_j(t) = \sum_{k=1}^{n} \Omega_{jk}X_k(t) - \sum_{k=1}^{n}\int_{-\infty}^{t} \mathrm{d}t'\Gamma_{jk}(t-t')X_k(t')+R_j(t) \tag{7.1}$$

Ω_{jk} is expressed in terms of the static correlations of X_j and X_k^* (cf. (3.14) or (3.20)). The 'friction function' $\Gamma_{jk}(t)$ is defined by (3.13). $R_j(t)$, representing the 'random force', is found to be the Heisenberg operator for a variable R_j (cf. (3.18) and table 1).

In deriving (7.1), the correlation function $\langle A, B^*\rangle$ is assumed to satisfy the stationariness condition (1.9) and the symmetry property (1.10). We consider the case of linearly independent variables X_j for which the determinant of $\langle X_j(0), X_k^*(0)\rangle$ does not vanish. It is also assumed that the friction function $\Gamma_{jk}(t)$ vanishes sufficiently fast for large values of t.

The random force $R_j(t)$ is related with Mori's random force $R_j^M(t)$ by

$$R_j(t) = R_j^M(t)+ \sum_{k=1}^{n}\int_{-\infty}^{t_0} \mathrm{d}t'\Gamma_{jk}(t-t')X_k(t') \tag{7.2}$$

and satisfies the second fluctuation–dissipation theorem (4.2) or (4.3); thus these relations suggested by Kubo (1966) have been confirmed.

With the aid of the first fluctuation–dissipation theorem (5.8), the correlation function $\langle X_j(t), X_k^*(0)\rangle$ is calculated from $\Gamma_{jk}(t)$. It is assumed that $\Gamma_{jk}(t)$ decays fast. The second fluctuation–dissipation theorem shows that $\Gamma_{jk}(t)$ is determined from the correlation of the random force $\langle R_j(t), R_k^*(0)\rangle$ and that $\langle R_j(t), R_k^*(0)\rangle$ decays as fast as $\Gamma_{jk}(t)$. Thus one can evaluate the correlation function $\langle X_j(t), X_k^*(0)\rangle$ if the short-time behaviour of $\langle R_j(t), R_k^*(0)\rangle$ is known.

The left-hand side of the generalized Langevin equation (7.1), $\mathrm{d}X_j(t)/\mathrm{d}t$, is equal to the total force acting on the variable $X_j(t)$. The first two terms on the right-hand side are determined by the instantaneous values of $X_k(t)$ and their values within a short time before t. In addition to these systematically determined contributions, the total force usually involves another part that fluctuates in a random way. This randomly fluctuating part must, therefore, be included in the remaining term of (7.1), that is in $R_j(t)$. If some knowledge about the short-time behaviour of the random force is obtained from the physical consideration of the fluctuating behaviour of the total force, the stationary generalized Langevin equation (7.1) will be helpful in understanding the properties of many-particle systems.

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Appendix. Correlation functions of dynamical variables

In § 5, the correlation functions of the dynamical variables are shown to be solved with the aid of the Laplace transform. In this Appendix, we derive the same result without taking advantage of the relation (5.2).

Let us take the correlations of equation (3.19) with $R^*(0)\rangle$ and $X^*(0)\rangle$ and take a Fourier transform. The results are

$$[\mathrm{i}\omega - \Omega + \Gamma_{\mathrm{i}\omega}] \,.\, \langle X, R^* \rangle_\omega^{(F)} = \langle R, R^* \rangle_\omega^{(F)} \tag{A.1}$$

and

$$(\mathrm{i}\omega - \Omega + \Gamma_{\mathrm{i}\omega}) \,.\, \langle X, X^* \rangle_\omega^{(F)} = \langle R, X^* \rangle_\omega^{(F)} \tag{A.2}$$

where $\langle X, X^*\rangle_\omega^{(F)}$, $\langle R, R^*\rangle_\omega^{(F)}$, $\langle R, X^*\rangle_\omega^{(F)}$ and $\langle X, R^*\rangle_\omega^{(F)}$ are the Fourier transforms of $\langle X(t), X^*(0)\rangle$, $\langle R(t), R^*(0)\rangle$, $\langle R(t), X^*(0)\rangle$, and $\langle X(t), R^*(0)\rangle$, respectively: for example

$$\langle X, X^* \rangle_\omega^{(F)} = \int_{-\infty}^{\infty} \mathrm{d}t \exp(-\mathrm{i}\omega t) \langle X(t), X^*(0) \rangle \tag{A.3}$$

$\Gamma_{i\omega}$ being the value at $p = \mathrm{i}\omega$ of the Laplace transform Γ_p of $\Gamma(t)$. The Hermitian conjugate of equation (A.2) is

$$\langle X, X^* \rangle_\omega^{(F)} \,.\, [-\mathrm{i}\omega - \Omega^* + (\Gamma_{\mathrm{i}\omega})^*] = \langle X, R^* \rangle_\omega^{(F)}. \tag{A.4}$$

By eliminating $\langle X, R^*\rangle_\omega^{(F)}$ from (A.1) and (A.4), one obtains

$$\langle X, X^* \rangle_\omega^{(F)} = \frac{1}{\mathrm{i}\omega - \Omega + \Gamma_{\mathrm{i}\omega}} \,.\, \langle R, R^* \rangle_\omega^{(F)} \,.\, \frac{1}{-\mathrm{i}\omega - \Omega^* + (\Gamma_{\mathrm{i}\omega})^*}. \tag{A.5}$$

With the aid of the fluctuation–dissipation relation (4.3), the Fourier transform $\langle R, R^*\rangle_\omega^{(F)}$ is expressed as follows:

$$\begin{aligned}\langle R, R^* \rangle_\omega^{(F)} &= \Gamma_{\mathrm{i}\omega} \,.\, \langle X, X^* \rangle + \langle X, X^* \rangle \,.\, (\Gamma_{\mathrm{i}\omega})^* \\ &= (\mathrm{i}\omega - \Omega + \Gamma_{\mathrm{i}\omega}) \,.\, \langle X, X^* \rangle + \langle X, X^* \rangle \,.\, \{-\mathrm{i}\omega - \Omega^* + (\Gamma_{\mathrm{i}\omega})^*\}.\end{aligned} \tag{A.6}$$

(Note that $\Omega.\langle X, X^*\rangle + \langle X, X^*\rangle.\Omega^* = \langle XL, X^*\rangle + \langle X, LX^*\rangle = 0$). Substituting this equation into (A.5), one obtains

$$\langle X, X^* \rangle_\omega^{(F)} = \frac{1}{\mathrm{i}\omega - \Omega + \Gamma_{\mathrm{i}\omega}} \,.\, \langle X, X^* \rangle + \langle X, X^* \rangle \,.\, \frac{1}{-\mathrm{i}\omega - \Omega^* + (\Gamma_{\mathrm{i}\omega})^*}. \tag{A.7}$$

The two-time correlation function $\langle X(t), X^*(0)\rangle$ is evaluated by a Fourier inverse transform:

$$\begin{aligned}\langle X(t), X^*(0) \rangle &= \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}\omega \exp(\mathrm{i}\omega t) \langle X, X^* \rangle_\omega^{(F)} \\ &= \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}\omega \exp(\mathrm{i}\omega t) \frac{1}{\mathrm{i}\omega - \Omega + \Gamma_{\mathrm{i}\omega}} \,.\, \langle X, X^* \rangle \\ &\quad + \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}\omega \exp(\mathrm{i}\omega t) \langle X, X^* \rangle \,.\, \frac{1}{-\mathrm{i}\omega - \Omega^* + (\Gamma_{\mathrm{i}\omega})^*}.\end{aligned} \tag{A.8}$$

We can confirm the fact that the result obtained by (A.8) is consistent with the one given by (5.7) with (5.8). In doing so, we assume that the integral in (5.6) is absolutely convergent when $\mathrm{Re}p \geqslant 0$. This means $\langle X, X^*\rangle_p$ is regular on the right-hand side of the complex p-plane. As a result the first and the second terms of (A.7) are regular in the lower and the upper half planes, respectively, of the complex ω-plane. From this fact, we conclude that, when t is positive, the second integral of the right-hand side of (A.8) vanishes and the first one gives the identical result as

(5.7) with (5.8). This implies that the first term on the right-hand side of (A.7) is the Fourier transform of the positive part of the correlation function $\langle X(t), X^*(0)\rangle$. In the same way one can show that the second term is the Fourier transform of the negative part; it is easily seen also that this is a direct consequence of the identity

$$\langle X(t), X^*(0)\rangle^* = \langle X(-t), X^*(0)\rangle.$$

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