

## Two-Time Correlation Functions of a System Coupled to a Heat Bath with a Gaussian-Markoffian Interaction

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(Received November 14, 1988)

Two-time correlation functions of physical operators of a system interacting with a heat bath producing a Gaussian-Markoffian noise are treated with the use of Feynman-Vernon's influence functional formalism. The Laplace transforms of correlation functions are shown to be expressed in terms of continued fractions of relevant hyperoperators. The result is valid to all orders of the system-bath interaction. In the limitation of motional narrowing, it reduces to those obtained in conventional methods.

### §1. Introduction

In a previous paper,<sup>1)</sup> we considered a test system coupled to a bath system with linear interactions and derived a set of hierarchical equations for the evolution of its reduced density operator. The basic assumption is that the interaction acts on the test system as a noise which is essentially Gaussian and Markoffian. The Gaussian property is guaranteed for a bath consisting of harmonic oscillators and is also realized in more general classes of bath systems as long as the interaction consists of a large number of weak interactions. The Markoffian property assumes exponential relaxation of response of the bath when it acts with the test system. This assumption is not universal but is a reasonably good model for realistic systems. With the use of Feynman-Vernon's influence functional method, we have shown that the system-bath interaction is represented by a quantum Gaussian-Markoffian noise if the bath temperature is high enough so that the condition

$$\hbar\gamma/k_B T \ll 1 \quad (1.1)$$

is satisfied, where  $\gamma$  is the relaxation rate of the bath.

In the present paper we extend the previous treatment to correlation functions of physical operators of the test system and show that their Laplace transforms can be expressed in terms of continued fractions of relevant

hyperoperators. The results are valid to all orders of the system-bath interaction. In the lowest order, they reduce to the conventional results of the master equation approach which are commonly used in treating relaxation and fluctuation in dissipative systems. Our results are expected to be applicable to such problems in which the higher order effects of interactions become important. Some applications will be treated in forthcoming papers on the second and higher order optical processes.

In the next section we review the two-time correlation function. In §3 we evaluate the expression of §2 along the same line as the previous paper. In §4, the results are compared with those obtained with use of the projection operator method. It is shown that our results contain important features of nonperturbative interaction which have not been seen in conventional treatments. Section 5 is devoted to concluding remarks.

### §2. Two-Time Correlations

Let  $X$  and  $Y$  be physical operators of the test system  $\mathbf{A}$  which is coupled to the bath system  $\mathbf{B}$ . The Hamiltonian of the total system,  $\mathbf{A} + \mathbf{B}$ , is denoted by  $H(t)$ , which may depend on time  $t$ . We define the two-time correlation function of  $X$  and  $Y$  by

$$\begin{aligned} C_{YX}(t, t_0) &= \langle Y(t_0+t)X(t_0) \rangle \\ &= \text{tr} \{ \rho_{\text{tot}}(0) Y(t_0+t)X(t_0) \}, \quad (2.1) \end{aligned}$$

where  $Y(t)$  and  $X(t)$  are the Heisenberg operators,  $\rho_{\text{tot}}(t)$  is the density operator of the

total system at time  $t$  and  $\text{tr} \{ \}$  means the summation over the  $\mathbf{A} + \mathbf{B}$  states. The above expression is written as

$$C_{YX}(t, t_0) = \text{tr} \left\{ \rho_{\text{tot}}(0) \exp_{\rightarrow} \left[ \frac{i}{\hbar} \int_0^{t+t_0} H(t) dt \right] Y \exp_{\leftarrow} \left[ -\frac{i}{\hbar} \int_0^{t+t_0} H(t) dt \right] \right. \\ \left. \times \exp_{\rightarrow} \left[ \frac{i}{\hbar} \int_0^{t_0} H(t) dt \right] X \exp_{\leftarrow} \left[ -\frac{i}{\hbar} \int_0^{t_0} H(t) dt \right] \right\}, \quad (2.2)$$

where  $\exp_{\rightarrow} [ \ ]$  or  $\exp_{\leftarrow} [ \ ]$  means an exponential operator ordered in time as indicated by the arrow. Repeated use of the cyclic invariance of the trace operation leads to<sup>2)</sup>

$$C_{YX}(t, t_0) = \text{tr} \left\{ Y \exp_{\leftarrow} \left[ -\frac{i}{\hbar} \int_{t_0}^{t+t_0} H^{\times}(t) dt \right] \left\{ X \exp_{\leftarrow} \left[ -\frac{i}{\hbar} \int_0^{t_0} H^{\times}(t) dt \right] \rho_{\text{tot}}(0) \right\} \right\}, \quad (2.3)$$

where the commutator operation  $F^{\times}$  is introduced by the definition

$$F^{\times} G = FG - GF, \quad (2.4)$$

which gives

$$\exp_{\leftarrow} \left[ -i \int_0^{\tau} F^{\times}(t) dt \right] G = \exp_{\leftarrow} \left[ -i \int_0^{\tau} F(t) dt \right] G \exp_{\rightarrow} \left[ i \int_0^{\tau} F(t) dt \right], \quad (2.5)$$

for any two operators  $F$  and  $G$ .

The correlation functions provide us with important information on the system we observe. For example, the equilibrium power spectrum of  $X$  is evaluated as

$$I_{XX}(\omega) = \lim_{t_0 \rightarrow \infty} \int_{-\infty}^{\infty} dt e^{-i\omega t} C_{XX}(t, t_0) \\ = 2 \text{Re} \left( \lim_{s_0 \rightarrow 0} s_0 C_{XX}[s, s_0] \Big|_{s=i\omega} \right). \quad (2.6)$$

Here, the double Laplace transform is defined by

$$C_{YX}[s, s_0] = \int_0^{\infty} dt \int_0^{\infty} dt_0 e^{-st} e^{-s_0 t_0} C_{YX}(t, t_0), \quad (2.7)$$

and, for the time-independent Hamiltonian

$H$ , this is simply expressed as

$$C_{YX}[s, s_0] = \text{tr} \left\{ Y \frac{1}{s - (i\hbar)^{-1} H^{\times}} \right. \\ \left. \times \left( X \frac{1}{s_0 - (i\hbar)^{-1} H^{\times}} \rho_{\text{tot}}(0) \right) \right\}, \quad (2.8)$$

where the fractional operators indicate the corresponding inverse operators.

### §3. Hierarchical Equations for Correlation Functions

As was shown in the previous paper, the coherent state representation of the density operator of a system  $\mathbf{A}$  in the absence of its interaction with the bath is written as

$$\rho_A(t') = \iint N^{-1} d\phi_f^* d\phi_f \iint N^{-1} d\phi_f'^* d\phi_f' |\phi_f\rangle \rho_A(\phi_f^*, \phi_f'; t') \langle \phi_f' |, \quad (3.1)$$

with

$$\dot{\rho}'_A(\phi_f^*, \phi_f'; t') = \text{T} \left( \int D_f [\phi^*(\tau) \phi(\tau)] \int D_f' [\phi'^*(\tau) \phi'(\tau)] \exp [(i/\hbar) S_A(\phi^*, \phi; t', 0)] \right. \\ \left. \times \exp [-(i/\hbar) S_A^*(\phi'^*, \phi'; t', 0)] \right), \quad (3.2)$$

where  $\phi(t)$ ,  $\phi^*(t)$ , etc., are complex numbers for a Boson system and are Grassmann numbers

for a Fermion system in which case the symbol T indicates their proper ordering in time. The expression in eq. (3.1) is a path integral in which all possible paths start from an initial state  $\phi(0)=\phi_i$  and  $\phi'^*(0)=\phi'_i^*$  and end at  $t'$  at the states  $\phi^*(t')=\phi_i^*$  and  $\phi'(t')=\phi'_i$ . The action  $S_A(\phi^*, \phi; t', 0)$  is defined by

$$S_A(\phi^*, \phi; t', 0) \equiv \int_0^{t'} d\tau \left( i\hbar \dot{\phi}^*(\tau) \phi(\tau) - H_A(\phi^*(\tau), \phi(\tau)) \right), \quad (3.3)$$

where  $H_A$  is the Hamiltonian of **A**.

According to Feynman and Vernon,<sup>3)</sup> the effect of the interaction of **A** with the bath can be incorporated by introducing an influence functional into eq. (3.2), namely,

$$\begin{aligned} \rho'_A(\phi_i^*, \phi'_i; t') = & T \left( \int D_f[Q(\tau)] \int D_f[Q'(\tau)] \exp [(i/\hbar)S_A(Q; t', 0)] \right. \\ & \left. \times F(Q, Q'; t', 0) \exp [(-i/\hbar)S_A^*(Q'; t', 0)] \right), \end{aligned} \quad (3.4)$$

where we assume the influence functional to be of the form

$$\begin{aligned} F(Q, Q'; t', 0) = & \exp \left\{ (-i\Delta)^2 \int_0^{t'} d\tau' \int_0^{\tau'} d\tau e^{-\gamma(\tau'-\tau)} V^\times(Q, Q'; \tau) \right. \\ & \left. \times \left( V^\times(Q, Q'; \tau) - i \frac{\beta\hbar\gamma}{2} V^\circ(Q, Q'; \tau) \right) \right\}, \end{aligned} \quad (3.5)$$

in which

$$V^\circ(Q, Q'; \tau) \equiv V(Q(\tau)) + V(Q'(\tau)), \quad (3.6)$$

$$V^\times(Q, Q'; \tau) \equiv V(Q(\tau)) - V(Q'(\tau)). \quad (3.7)$$

In the above equations, we simply write  $Q(\tau)$  or  $Q'(\tau)$  for  $(\phi^*(\tau), \phi(\tau))$  or  $(\phi'^*(\tau), \phi'(\tau))$ . To derive this particular form, we assume that the bath is initially at a high temperature  $\hbar\beta\gamma \ll 1$  ( $\beta = 1/k_B T$ ) and shows the Debye relaxation with the damping constant  $\gamma$  for the external pulsvive excitation. The coupling strength with the test system is denoted by the dimensionless constant  $\Delta$ . The initial condition of the total system is assumed to be the factorized form:

$$\rho_{\text{tot}}(0) = \rho_A(0) \rho_B^e. \quad (3.8)$$

Here,  $\rho_A(0)$  is the initial state of the test system and  $\rho_B^e$  is the thermal equilibrium state of the bath with respect to its own Hamiltonian.

Consider the hierarchical density elements

$$\begin{aligned} \rho'_n(\phi_i^*, \phi'_i; t') = & T \left( \int D[Q(\tau)] \int D[Q'(\tau)] \right. \\ & \times \left\{ -i\Delta \int_0^{t'} d\tau e^{-\gamma(t'-\tau)} \left[ V^\times(Q, Q'; \tau) - i \frac{\beta\hbar\gamma}{2} V^\circ(Q, Q'; \tau) \right] \right\}^n \\ & \left. \times \exp [(i/\hbar)S_A(Q; t', 0)] F(Q, Q'; t) \exp [-(i/\hbar)S_A^*(Q'; t', 0)] \right), \end{aligned} \quad (3.9)$$

and corresponding operators  $\rho_n(t')$ . The first member of this equation has appeared in eq. (3.4) and, with this choice, the equation of motion for the test system is expressed in the hierarchical differential equations:

$$\frac{\partial}{\partial t'} \rho_n(t') = ((i\hbar)^{-1} H_A^\times - n\gamma) \rho_n(t') - i\Delta V^\times \rho_{n+1}(t') - ni\Delta \Theta \rho_{n-1}(t'). \quad (3.10)$$

Here, we set

$$\Theta \equiv V^\times - i \frac{\beta \hbar \gamma}{2} V^\circ, \quad (3.11)$$

with the notation of eq. (2.4) and

$$F^\circ G = FG + GF. \quad (3.12)$$

As seen from the definition, initial conditions of eq. (3.10) are given by

$$\rho_0(0) = \rho_A(0), \quad \rho_n(0) = 0. \quad (n \geq 1) \quad (3.13)$$

As was shown in ref. 1, the resolvent for the test system is then nonperturbatively evaluated in the continued fractional form:

$$\rho_0[s] = Z_0[s] \rho_A(0), \quad (3.14)$$

where

$$Z_0[s] = \frac{1}{s - (i\hbar)^{-1} H_A^\times + V^\times \frac{\Delta^2}{s + \gamma - (i\hbar)^{-1} H_A^\times + V^\times \frac{2\Delta^2}{s + 2\gamma - (i\hbar)^{-1} H_A^\times + \dots}} \Theta. \quad (3.15)$$

Now we wish to evaluate the time-dependent correlation functions of eq. (2.3). We suppose that the operators  $X$  and  $Y$  work only for the test system  $\mathbf{A}$ . Therefore, for example, a one-time correlation (or the expectation value) with the initial condition (3.8) is expressed by using the inverse Laplace transformation of  $\rho_0[s]$  as

$$\langle Y(t) \rangle = \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} ds e^{st} \text{tr}_A \{ Y \rho_0[s] \}, \quad (3.16)$$

where the constant  $c$  is chosen in such a way that all singular points of the integrand lie on the left side of the path and  $\text{tr}_A \{ \quad \}$  means the summation over the  $\mathbf{A}$  states. The evaluation of two-time correlations is more complicated than (3.16), though it can be done by considering the following hierarchical elements

$$\begin{aligned} \sigma'_n(\phi_f^*, \phi_f'; t'', t') = & \text{T} \left( \int D[Q(\tau)] \int D[Q'(\tau)] X(Q(t')) \right. \\ & \times \left\{ -i\Delta \int_0^{t''} d\tau e^{-\gamma(t''-\tau)} \left[ V^\times(Q, Q'; \tau) - i \frac{\beta \hbar \gamma}{2} V^\circ(Q, Q'; \tau) \right] \right\}^n \\ & \times \exp [(i/\hbar) S_A(Q; t'', 0)] F(Q, Q'; t) \exp [-(i/\hbar) S_A^*(Q'; t'', 0)] \Big), \quad (3.17) \end{aligned}$$

and corresponding operators  $\sigma_n(t'', t')$ . Using the first member of these operators, we write eq. (2.3) as

$$C_{YX}(t, t_0) = \text{tr}_A \{ Y \sigma_0(t+t_0, t_0) \}. \quad (3.18)$$

The Laplace transformation of this correlation with respect to  $t$  is performed by considering the time differentiation of  $\sigma_0(t'', t')$  with respect to  $t''$ . In the region  $t'' > t'$ , the same approach used in eq. (3.10) leads us to the following set of equations:

$$\frac{\partial}{\partial t''} \sigma_n(t'', t') = ((i\hbar)^{-1} H_A^\times - n\gamma) \sigma_n(t'', t') - i\Delta V^\times \sigma_{n+1}(t'', t') - ni\Delta \Theta \sigma_{n-1}(t'', t'). \quad (3.19)$$

These equations have the same form as eq. (3.10). However, from definition, the initial conditions of (3.19) are now given not by the element  $\rho_0(0)$  but by the set  $\{ \sigma_0(t_0, t_0), \sigma_1(t_0, t_0), \dots \}$ ,

where  $\sigma_n(t_0, t_0)$  is defined by eq. (3.17). Therefore, the Laplace transform of  $\sigma_0(t+t_0, t_0)$  with respect to  $t$  is now given by (see Appendix)

$$\sigma_0[s, t_0] = Z_0[s] \sigma_0(t_0, t_0) + \sum_{n=1}^{\infty} (-i\Delta)^n Z_0[s] \left( \prod_{k=1}^n (V^\times Z_k[s]) \right) \sigma_n(t_0, t_0), \quad (3.20)$$

where

$$Z_k[s] = \frac{1}{s + k\gamma - (i\hbar)^{-1}H_A^\times + V^\times \frac{(k+1)\Delta^2}{s + (k+1)\gamma - (i\hbar)^{-1}H_A^\times + V^\times \frac{(k+2)\Delta^2}{s + (k+2)\gamma - (i\hbar)^{-1}H_A^\times + \dots}} \Theta. \quad (3.21)$$

By using  $\rho_n(t_0)$ , the initial conditions  $\sigma_n(t_0, t_0)$  can be written as:

$$\sigma_n(t_0, t_0) = X\rho_n(t_0). \quad (3.22)$$

The Laplace transformation of  $\rho_n(t_0)$  is also given in the Appendix and the final expression of the two-time correlation function is then

$$C_{YX}[s, s_0] = \text{tr}_A \left\{ YZ_0[s] (XZ_0[s_0] \rho_A(0)) + \sum_{n=1}^{\infty} n! (i\Delta)^{2n} YZ_0[s] \times \left( \prod_{k=1}^n (V^\times Z_k[s]) \right) \left[ X \left( \prod_{j=1}^n (Z_{n-j+1}[s_0] \Theta) \right) Z_0[s_0] \rho_A(0) \right] \right\}. \quad (3.23)$$

#### §4. Nonperturbative Effects of Two-Time Correlations

In this section, we explore the nonperturbative features of eq. (3.23) and compare this equation with the results obtained from the projection operator method. Let us introduce the projection operator  $P$  and its complementary projection  $Q$  by

$$P = \rho_B^e \cdot \text{tr}_B \{, \quad (4.1)$$

and

$$Q = 1 - P = 1 - \rho_B^e \cdot \text{tr}_B \{, \quad (4.2)$$

where, as already shown in eq. (3.8),  $\rho_B^e$  represents the equilibrium density operator of the bath system. By inserting  $P$  and  $Q$  in eq. (2.8) on the left-hand side of  $X$ , we can decompose this equation into two components:

$$C_{YX}[s, s_0] = \text{tr} \left\{ Y \frac{1}{s - (i\hbar)^{-1}H^\times} P \left( X \frac{1}{s_0 - (i\hbar)^{-1}H^\times} \rho_{\text{tot}}(0) \right) \right\} + \text{tr} \left\{ Y \frac{1}{s - (i\hbar)^{-1}H^\times} Q \left( X \frac{1}{s_0 - (i\hbar)^{-1}H^\times} \rho_{\text{tot}}(0) \right) \right\}. \quad (4.3)$$

We assume that the initial condition of this equation is given by eq. (3.8). The first term of eq. (4.3) corresponds to that of eq. (3.23), since the first term of eq. (3.23) arises from the operator  $\sigma_0(t_0, t_0)$  of which the bath state at time  $t_0$  is in the equilibrium state  $\rho_B^e$ . Therefore, the second term of eq. (4.3) corresponds to the remaining terms of eq. (3.23). It should be mentioned that practical calculations of eq. (4.3) have been done by neglecting the second term, assuming that the bath state is always in its equilibrium state,  $\rho_B^e$ . As an example, consider an atomic two-level system linearly interacting with a vacuum radiation bath.<sup>4,5</sup> By eliminating the bath, the equation of motion for the two-level system (*master equation*) is written as

$$P\dot{\rho}_{\text{tot}}(t) = P\frac{-i}{\hbar} H^\times \rho_{\text{tot}}(t) \longrightarrow \dot{\rho}_A(t) = \frac{-i}{\hbar} H_A^\times \rho_A(t) - \Gamma\rho_A(t), \quad (4.4)$$

where  $H_A$  is the Hamiltonian of the two-level system and  $\Gamma$  is the hyperoperator expressed in the atomic creation and annihilation operators  $a^+$  and  $a$  as

$$-\Gamma\rho \equiv \kappa a\rho a^+ - \left(\frac{1}{2}\kappa + i\delta\right) a^+ a\rho - \left(\frac{1}{2}\kappa - i\delta\right) \rho a^+ a, \quad (4.5)$$

in which  $\kappa$  and  $\delta$  are the natural radiation damping and the Lamb shift, respectively. To derive this form, the resonant approximation was used and the weak and ultrashort correlated perturbation was assumed (*white-noise approximation*). Thus, the resolvent of this system is written as

$$\rho_A[s] = \frac{1}{s - (i\hbar)^{-1}H_A^\times + \Gamma} \rho_A(0). \quad (4.6)$$

Using this approach, the time evolution in two-time correlations is also described by eq. (4.4).<sup>6</sup> In our notation, this is expressed as

$$C_{YX}[s, s_0] = \text{tr}_A \left\{ Y \frac{1}{s - (i\hbar)^{-1}H_A^\times + \Gamma} \times \left( X \frac{1}{s_0 - (i\hbar)^{-1}H_A^\times + \Gamma} \rho_A(0) \right) \right\}, \quad (4.7)$$

which corresponds to the first term on the right-hand side of eq. (4.3) under the weak and the white-noise approximation. The second term of eq. (4.3) is neglected in this approach. This neglect is the result of using the weak and the white-noise bath such as the vacuum radiation field. The validity of eq. (4.7) is then determined from the strength of the interaction and the correlation time of the bath.

To view this point from our approach, we derive a corresponding equation from eq. (3.23). The effect of modulation in eq. (3.23) is generally characterized by the parameters  $\Delta$ ,  $\gamma$ , and  $\nu_A$ , where the last one is a characteristic frequency of the quantum level difference of the test system. If the condition

$$\Delta \ll \gamma, \nu_A \quad (4.8)$$

is satisfied, the higher order interactions can be neglected. Then, omitting the second term of eq. (3.23), we have

$$C_{YX}[s, s_0] = \text{tr}_A \left\{ Y \frac{1}{s - (i\hbar)^{-1}H_A^\times + \Gamma'} \times \left( X \frac{1}{s_0 - (i\hbar)^{-1}H_A^\times + \Gamma'} \rho_A(0) \right) \right\}, \quad (4.9)$$

where

$$\Gamma' = V^\times \frac{\Delta^2}{\gamma - (i\hbar)^{-1}H_A^\times} \Theta. \quad (4.10)$$

In the random modulation problem, the modulation condition which leads to the Markoffian character is said to be the motional narrowing limit and is given by

$$\nu_A \ll \gamma \ll 1/\beta\hbar. \quad (4.11)$$

In this case, eq. (4.10) is expressed as

$$\Gamma' \rightarrow \Gamma = \gamma' V^\times V^\times - i\delta V^\times V^\circ, \quad (4.12)$$

where

$$\gamma' = \Delta^2/\gamma, \quad (4.13)$$

and

$$i\delta = i\gamma'\hbar\beta\gamma/2. \quad (4.14)$$

Equation (4.9) with eq. (4.12) corresponds to eq. (4.7), but eq. (4.7) has been specified as the interaction of  $V = a + a^+$  and has been used as the resonant approximation. The validity of eq. (4.13) is now secured by inequalities (4.8) and (4.11). If these conditions are not fulfilled, the second term on the right-hand side of eq. (4.3) becomes quite important. We will discuss this point in forthcoming papers.

## §5. Conclusions

Using a specific model for the system, we have deduced the double Laplace transform of two-time correlations. The result is expressed in continued fractions in terms of hyperoperators. This result enables us to evaluate the effects of higher order interactions which are neglected in existing approaches. To observe a point more closely, we

introduced the projection operator and compared our results with those of the conventional master equation approach.

Finally, we should notice the relation between our results and those of the stochastic approach. As was discussed in ref. 1, our theory is connected with the stochastic Gaussian-Markoffian models. In the stochastic approach, some calculations have been carried out especially for the optical processes. It was shown that the strong random modulation gave rise to a mixture of the coherent and incoherent processes which was the result of the disturbance in the quantum coherence of the system by the random modulation.<sup>7-9)</sup> In the stochastic approach the second order emission spectrum from this mixture process was termed the broadened Rayleigh or the broad-

ened Raman line.<sup>10)</sup> However, the corresponding process has not been recognized in the dynamical approaches. Our results allow us to clarify this point more closely. We will show that the second term of eq. (3.23) gives rise to a mixture of the coherent and incoherent processes from the dynamical point of view. We will discuss the details in forthcoming paper.<sup>11)</sup>

**Acknowledgment**

One of the authors (Y.T.) is grateful to Dr. Hiroshi Takano for useful discussions.

**Appendix**

The Laplace transformation of eqs. (3.10) and (3.19) can be expressed in the matrix form. For example, eq. (3.19) is written as

$$\begin{bmatrix} \sigma_0[s, t_0] \\ \sigma_1[s, t_0] \\ \sigma_2[s, t_0] \\ \sigma_3[s, t_0] \\ \vdots \end{bmatrix} = \begin{bmatrix} s + \frac{i}{\hbar} H_A^\times & i\Delta V^\times & 0 & 0 & \cdots \\ i\Delta\Theta & s + \frac{i}{\hbar} H_A^\times + \gamma & i\Delta V^\times & 0 & \cdots \\ 0 & 2i\Delta\Theta & s + \frac{i}{\hbar} H_A^\times + 2\gamma & i\Delta V^\times & \\ 0 & 0 & 3i\Delta\Theta & \ddots & \ddots \\ \vdots & \vdots & \vdots & \ddots & \ddots \end{bmatrix}^{-1} \begin{bmatrix} \sigma_0(t_0, t_0) \\ \sigma_1(t_0, t_0) \\ \sigma_2(t_0, t_0) \\ \sigma_3(t_0, t_0) \\ \vdots \end{bmatrix} \tag{A.1}$$

We denote this equation by

$$\mathbf{G}[s] = \mathbf{R}[s] \mathbf{G}(t_0). \tag{A.2}$$

For any matrix  $\mathbf{D}_0$  with operators or matrix elements  $\mathbf{A}, \mathbf{B}, \mathbf{C}, \mathbf{D}_1$ , we have the following relation:

$$\mathbf{D}_0^{-1} = \left[ \begin{array}{c|c} \mathbf{A} & \mathbf{B} \\ \hline \mathbf{C} & \mathbf{D}_1 \end{array} \right]^{-1} = \left[ \begin{array}{c|c} \mathbf{Z} & -\mathbf{ZBD}_1^{-1} \\ \hline -\mathbf{D}_1^{-1}\mathbf{CZ} & \mathbf{D}_1^{-1} + \mathbf{D}_1^{-1}\mathbf{CZBD}_1^{-1} \end{array} \right], \tag{A.3}$$

where we put

$$\mathbf{Z} = (\mathbf{A} - \mathbf{BD}_1^{-1}\mathbf{C})^{-1}. \tag{A.4}$$

By using this relation, the element  $(\mathbf{R}[s])_{11}$  is easily evaluated as

$$(\mathbf{R}[s])_{11} = Z_0[s] = \frac{1}{s - (i\hbar)^{-1}H_A^\times + V^\times} \frac{\Delta^2}{s + \gamma - (i\hbar)^{-1}H_A^\times + V^\times} \frac{\Theta}{s + 2\gamma - (i\hbar)^{-1}H_A^\times + \cdots} \Theta \tag{A.5}$$

To evaluate the element  $\sigma_0[s, t_0]$ , we should evaluate  $(\mathbf{R}[s])_{1j}$ . Using eq. (A.3) and eq. (A.5), we may write

$$(\mathbf{D}_0^{-1})_{\mathbf{B}} = -\mathbf{ZBD}_1^{-1} = -Z_0[s] \times [i\Delta V^\times \ 0 \ 0 \ 0 \ \cdots] \times \mathbf{D}_1^{-1}, \tag{A.6}$$

where  $(\mathbf{D}_0)_B$  is the row vector with elements  $(\mathbf{R}[s])_{1j} (j > 1)$ . Then the element  $(\mathbf{R}[s])_{12}$  is evaluated as

$$(\mathbf{R}[s])_{12} = Z_0[s](-i\Delta V^\times)Z_1[s], \quad (\text{A} \cdot 7)$$

where  $Z_1[s]$  denotes the element  $(\mathbf{D}_1^{-1})_{11}$  and is similarly evaluated as eq. (A·5). By successive applications of eq. (A·3), we have  $(\mathbf{R}[s])_{1j}$  in the form:

$$\begin{aligned} (\mathbf{R}[s])_{1j} &= (-i\Delta)^{j-1} Z_0[s](V^\times Z_1[s])(V^\times Z_2[s]) \cdots (V^\times Z_{j-1}[s]) \\ &= (-i\Delta)^{j-1} Z_0[s] \prod_{k=1}^{j-1} (V^\times Z_k[s]), \quad (j \geq 2) \end{aligned} \quad (\text{A} \cdot 8)$$

where

$$Z_k[s] = \frac{1}{s + k\gamma - (i\hbar)^{-1}H_A^\times + V^\times \frac{(k+1)\Delta^2}{s + (k+1)\gamma - (i\hbar)^{-1}H_A^\times + V^\times \frac{(k+2)\Delta^2}{s + (k+2)\gamma - (i\hbar)^{-1}H_A^\times + \cdots}} \Theta. \quad (\text{A} \cdot 9)$$

Then the Laplace transform of  $t$  is given by

$$\sigma_0[s, t_0] = Z_0[s]\sigma_0(t_0, t_0) + \sum_{n=1}^{\infty} (-i\Delta)^n Z_0[s] \left( \prod_{k=1}^n (V^\times Z_k[s]) \right) \sigma_n(t_0, t_0). \quad (\text{A} \cdot 10)$$

Operators  $\rho_n[s]$  of eq. (3.22) are calculated by evaluating  $(\mathbf{R}[s])_{j1}$ . A similar method allows us to write these elements in the form:

$$\begin{aligned} (\mathbf{R}[s])_{j1} &= (j-1)!(-i\Delta)^{j-1} (Z_{j-1}[s]\Theta)(Z_{j-2}[s]\Theta) \cdots (Z_1[s]\Theta)Z_0[s] \\ &= (j-1)!(-i\Delta)^{j-1} \left( \prod_{k=1}^{j-1} (Z_{j-k}[s]\Theta) \right) Z_0[s]. \quad (j \geq 2) \end{aligned} \quad (\text{A} \cdot 11)$$

Therefore, we have

$$\begin{aligned} \rho_n[s] &= n!(-i\Delta)^n \left( \prod_{k=1}^n (Z_{n-k+1}[s]\Theta) \right) \\ &\quad \times Z_0[s]\rho_A(0). \end{aligned} \quad (\text{A} \cdot 12)$$

### References

- 1) Y. Tanimura and R. Kubo: J. Phys. Soc. Jpn. **58** (1989) 101.
- 2) T. Chikama, K. Saikawa and R. Kubo: J. Phys. Soc. Jpn. **53** (1984) 991.
- 3) R. P. Feynman and F. L. Vernon, Jr.: Ann. Phys. (USA) **24** (1963) 118.
- 4) B. R. Mollow and M. M. Miller: Ann. Phys. **52** (1969) 464.
- 5) G. S. Agarwal: *Quantum Optics*, Springer Tracts in Modern Physics 70 (Springer, 1974)
- 6) B. R. Mollow: Phys. Rev. **188** (1969) 1969.
- 7) T. Takagahara, E. Hanamura and R. Kubo: J. Phys. Soc. Jpn. **43** (1977) 802.
- 8) T. Takagahara, E. Hanamura and R. Kubo: J. Phys. Soc. Jpn. **43** (1977) 811.
- 9) H. Tsunetsugu and E. Hanamura: J. Phys. Soc. Jpn. **55** (1986) 3636.
- 10) Y. Tanimura, H. Takano and R. Kubo: J. Phys. Soc. Jpn. **55** (1986) 4550.
- 11) Y. Tanimura, T. Suzuki and R. Kubo: to be published in J. Phys. Soc. Jpn. **58** (1989) No. 5.