

Quantum Analysis and Nonequilibrium Response

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The quantum derivatives of e^{-A} , A^{-1} and $\log A$, which play a basic role in quantum statistical physics, are derived and their convergence is proven for an unbounded positive operator A in a Hilbert space. Using the quantum analysis based on these quantum derivatives, a basic equation for the entropy operator in nonequilibrium systems is derived, and Zubarev's theory is extended to infinite order with respect to a perturbation. Using the first-order term of this general perturbational expansion of the entropy operator, Kubo's linear response is rederived and expressed in terms of the inner derivation $\delta_{\mathcal{H}}$ for the relevant Hamiltonian \mathcal{H} . Some remarks on the conductivity $\sigma(\omega)$ are given.

§1. Introduction

Recently, the present author¹⁾⁻³⁾ proposed a new scheme of quantum calculus, the so-called quantum analysis. In this scheme, the derivative of an operator-valued function with respect to the relevant operator itself is expressed only in terms of the original operator and its inner derivation (i.e., a hyperoperator or superoperator), and an operator expansion formula is derived.

In the present paper, the quantum derivatives of e^{-A} , A^{-1} and $\log A$, which are basic operator functions in physics, are derived, and their convergence is proven in §2 for the unbounded positive operator A in a Hilbert space. (See also Appendices A and B.) Nonlinear responses in equilibrium are expressed in terms of quantum derivatives in §3. A basic equation for nonequilibrium systems is derived in §4 using quantum analysis. This derivation has the merit that it is valid even for an unbounded entropy operator. On the other hand, Zubarev's derivation is based on the power series expansion of the density matrix with respect to the entropy operator, and consequently it is restricted to a bounded entropy operator. Zubarev's theory⁴⁾ is extended to infinite order in §5. This gives a renormalized perturbation theory with respect to an external field. Kubo's formula of linear response^{5),6)} is then rederived and expressed in terms of an inner derivation in §6. Some remarks on the conductivity $\sigma(\omega)$ are given in §7. The entropy operator $\eta(t)$ in a dissipative system [namely $-\log \rho(t)$ for the density matrix $\rho(t)$] is expressed in a compact form using the inner derivation in Appendix C. This expression is convenient for studying quantum effects, because it is expressed only in terms of commutators.

§2. Quantum derivatives of e^{-A} , A^{-1} and $\log A$ and the convergence of the differential $df(A)$

In a previous paper,¹⁾ quantum analysis was formulated in a Banach space, namely for bounded operators. The term 'quantum analysis' refers to noncommutative differential calculus in terms of inner derivations, namely commutators. Formal expressions and several formulas of quantum analysis are derived in Ref. 1). In practical applications, for example, to quantum and statistical physics, we often must treat unbounded operators in a Hilbert space. As is well known, it is difficult to prove generally the convergence of such formal expressions for unbounded operators.^{7),8)} Fortunately, the density matrix ρ in statistical mechanics is a contraction operator when the relevant Hamiltonian \mathcal{H} is unbounded (even for a finite system) but positive definite (or bounded below). Furthermore, a perturbation may often be assumed to be bounded in statistical physics. (For example, a Zeeman energy is expressed by a bounded operator in a finite system, while the kinetic energy of an itinerant electron system is unbounded.) Hereafter we discuss the quantum calculus for these situations. Thus we study here the convergence of the Gâteaux differential

$$df(A) = \lim_{h \rightarrow 0} \frac{f(A + hdA) - f(A)}{h}, \quad (2.1)$$

where A is unbounded but $f(A)$ is bounded, and dA is an arbitrary bounded operator independent of A . To consider this situation is one of the key points for studying the convergence of Eq. (2.1). The quantum derivative $df(A)/dA$ is defined¹⁾ by

$$df(A) = \frac{df(A)}{dA} dA. \quad (2.2a)$$

Here $df(A)/dA$ is a hyperoperator which is a function of both A and the inner derivation δ_A defined by Eq. (2.7b). This property is crucial in quantum analysis.¹⁾ In fact, we have the formula¹⁾⁻³⁾

$$\frac{df(A)}{dA} = \frac{\delta_{f(A)}}{\delta_A} \quad (2.2b)$$

in a Banach space. Higher-order quantum derivatives will be discussed in §3.

i) Quantum derivatives of e^{-A} and A^{-1}

Here, we attempt to prove the convergence of Eq. (2.1) for two typical operator functions, $f(A) = e^{-A}$ and $f(A) = A^{-1}$, where A is a positive (but unbounded) operator. Clearly we have

$$\|e^{-A}\| < 1 \quad \text{and} \quad \|A^{-1}\| < \infty, \quad (2.3)$$

under the condition that $A \geq a > 0$ for a constant a .

First note that^{7),9)}

$$\frac{d}{dx} e^{-(A+xB)} = - \int_0^1 e^{-(1-s)(A+xB)} B e^{-s(A+xB)} ds. \quad (2.4a)$$

Integrating Eq. (2.4a) we obtain

$$e^{-(A+xB)} = e^{-A} - \int_0^x dt \int_0^1 ds e^{-(1-s)(A+tB)} B e^{-s(A+tB)}. \quad (2.4b)$$

Then we can prove the convergence

$$\lim_{h \rightarrow 0} \left\| (e^{-(A+hB)} - e^{-A}) / h - \int_0^1 e^{-(1-s)A} (-B) e^{-sA} ds \right\| = 0 \quad (2.5)$$

when A is a positive (but unbounded) operator and $B = dA$ is bounded, as is shown in detail in Appendix A. Thus we arrive at the differential

$$d(e^{-A}) = - \int_0^1 e^{-(1-s)A} (dA) e^{-sA} ds = -e^{-A} \Delta(A) dA, \quad (2.6a)$$

or the quantum derivative

$$\frac{de^{-A}}{dA} = -e^{-A} + \int_0^1 e^{-(1-s)A} \delta_{\exp(-sA)} ds = -e^{-A} \Delta(A). \quad (2.6b)$$

This is well defined for a positive operator A . Here, the hyperoperator $\Delta(A)$ is defined by¹⁾

$$\Delta(A) = \int_0^1 e^{t\delta_A} dt = \frac{e^{\delta_A} - 1}{\delta_A}, \quad (2.7a)$$

with the inner derivation δ_A defined by

$$\delta_A Q \equiv [A, Q] \equiv AQ - QA. \quad (2.7b)$$

The ratio of the hyperoperators $(e^{\delta_A} - 1)$ and δ_A is well defined, although δ_A^{-1} does not necessarily exist. The formula (2.6b) with Eq. (2.7a) will be used frequently later.

Concerning the convergence of the power series expansion of $e^{-A} \Delta(xA)$, we have the theorem.

Theorem 1: *The power series expansion of $e^{-A} \Delta(xA) dA$ with respect to x converges in the uniform norm topology for $A > 0$ and for $|x| < \alpha^{-1}$, where α is defined by the upper limit*

$$\alpha = \overline{\lim}_{n \rightarrow \infty} \|(A^{-1} \delta_A)^n dA\|^{\frac{1}{n}}. \quad (2.8)$$

The proof is easily given using the Stirling formula $n! \simeq n^n e^{-n}$ for large n and the following inequality.

Inequality: When $A > 0$, we have

$$\|e^{-A} \delta_A^n B\| \leq n^n e^{-n} \|(A^{-1} \delta_A)^n B\| \quad (2.9a)$$

for any positive integer n .

The proof of the above inequality is easily given using the inequalities

$$\|e^{-A} \delta_A^n B\| \leq \|e^{-A} A^n\| \cdot \|(A^{-1} \delta_A)^n B\| \quad (2.9b)$$

and

$$\| e^{-A}A^n \| \leq e^{-n}n^n. \tag{2.9c}$$

It should be noted here that δ_A and A commute.

Corollary: If $B^{1/k}$ is defined for any positive integer k and there exists the maximum $M \equiv \max_k \| A^{-1}B^{1/k}A \|$, then the power series expansion of $e^{-A}\Delta(xA)B$ with respect to x converges in the uniform norm topology for $A > 0$ and for $|x| < 1/(M+1)$.

Proof: First note that

$$(A^{-1}\delta_A)^n B = \sum_{k=0}^n (-1)^k \binom{n}{k} A^{-k} B A^k \tag{2.10a}$$

for any positive integer n . Then we have

$$\begin{aligned} \| (A^{-1}\delta_A)^n B \| &\leq \sum_{k=0}^n \binom{n}{k} \| A^{-k} B A^k \| \\ &\leq \sum_{k=0}^n \binom{n}{k} \| A^{-1} B^{1/k} A \|^k \leq (M+1)^n \end{aligned} \tag{2.10b}$$

under the conditions of the above corollary. Thus we obtain

$$\overline{\lim}_{n \rightarrow \infty} \| (A^{-1}\delta_A)^n B \|^{\frac{1}{n}} \leq M+1. \tag{2.10c}$$

Note that $\lim_{k \rightarrow \infty} \| A^{-1}B^{1/k}A \| = 1$. Then the maximum number M may exist when the deformation of $B^{1/k}$ by the transformation of an unbounded operator A is finite for all values of k .

Similarly we study the differential of the resolvent operator A^{-1} when $A \geq a > 0$. We easily obtain

$$\begin{aligned} \lim_{h \rightarrow 0} \left\| \left(\frac{1}{A+hB} - \frac{1}{A} \right) / h - \frac{1}{A} (-B) \frac{1}{A} \right\| \\ \leq \lim_{h \rightarrow 0} |h| \cdot \left\| \frac{1}{A} \right\|^2 \cdot \left\| \frac{1}{A+hB} \right\| \cdot \|B\|^2 = 0. \end{aligned} \tag{2.11a}$$

That is, we have

$$d \left(\frac{1}{A} \right) = -\frac{1}{A} (dA) \frac{1}{A} = (-A^{-2} + A^{-1}\delta_{A^{-1}})dA = -\frac{1}{A(A-\delta_A)}dA. \tag{2.11b}$$

This gives

$$\frac{d}{dA} \left(\frac{1}{A} \right) = -\frac{1}{A(A-\delta_A)}. \tag{2.11c}$$

This is also bounded when $A \geq a > 0$. Here we have used the relation¹⁾ $\delta_{A^{-1}} = A^{-1} - (A - \delta_A)^{-1}$.

ii) Quantum derivative of $\log A$

The above arguments can be extended to more general case in which $f(A)$ is also unbounded but $df(A)$ is bounded for bounded $B = dA$. A typical case is given by $f(A) = \log A$. The operator $\log(A + hB)$ is formally expressed by the following integral:

$$\begin{aligned} \log(A + hB) &= \int_0^\infty \left(\frac{1}{t+1} - \frac{1}{t+A+hB} \right) dt \\ &= \int_0^\infty \left(\frac{1}{t+1} - \frac{1}{t+A} \right) dt + h \int_0^\infty \frac{1}{t+A} B \frac{1}{t+A} dt \\ &\quad - h^2 \int_0^\infty \frac{1}{t+A} B \frac{1}{t+A} B \frac{1}{t+A+hB} dt. \end{aligned} \tag{2.12}$$

Then we have

$$\begin{aligned} &\left\| [\log(A + hB) - \log A] / h - \int_0^\infty \frac{1}{t+A} B \frac{1}{t+A} dt \right\| \\ &\leq |h| \cdot \|B\|^2 \int_0^\infty \left\| \frac{1}{t+A} \right\|^2 \cdot \left\| \frac{1}{t+A+hB} \right\| dt. \end{aligned} \tag{2.13}$$

Consequently we arrive at¹⁰⁾

$$d \log A = \int_0^\infty \frac{1}{t+A} (dA) \frac{1}{t+A} dt. \tag{2.14}$$

Clearly this is bounded when A is positive (i.e., $A \geq a > 0$) and dA is bounded. This is formally written as

$$\frac{d \log A}{dA} = \frac{1}{A} - \int_0^\infty \frac{1}{t+A} \delta_{(t+A)^{-1}} dt = -\delta_A^{-1} \log(1 - A^{-1} \delta_A). \tag{2.15}$$

The second expression of Eq. (2.15) gives the convergence of $df(A)/dA$.

iii) Convergence of $df(A)$ for an unbounded operator A and for the bounded differential dA .

In general, the derivative $df(A)/dA$ is formally given by the following formula.¹⁾

Formula 1: When $f(x)$ is an analytic function of x , we have

$$\frac{df(A)}{dA} = \frac{\delta_{f(A)}}{\delta_A} = \frac{f(A) - f(A - \delta_A)}{\delta_A} = \int_0^1 f^{(1)}(A - t\delta_A) dt. \tag{2.16}$$

Here $f^{(n)}(x)$ denotes the n -th derivative of $f(x)$. This is formally expanded as

$$\frac{df(A)}{dA} = f^{(1)}(A) - \frac{1}{2!} f^{(2)}(A) \delta_A + \dots + \frac{(-1)^n}{(n+1)!} f^{(n+1)}(A) \delta_A^n + \dots \tag{2.17}$$

Then, we have the following theorem.

Theorem 2: Let A be unbounded, and let $\{f^{(n)}(A)\}$ for $n = 0, 1, 2, \dots$ and dA be bounded. Then, the formal expansion (2.17) operating on dA converges to Eq. (2.16) in the uniform norm topology if

$$\alpha_1 \equiv \overline{\lim}_{n \rightarrow \infty} \left\| \frac{f^{(n+1)}(A)}{(n+1)!} \delta_A^n dA \right\|^{\frac{1}{n}} < 1. \tag{2.18a}$$

A proof of this theorem is easily obtained. Theorem 1 is a typical example of the above general theorem. This theorem can also be extended to higher-order derivatives (see §3 and Appendix B).

In the more general situation in which the operators $\{f^{(n+1)}(A)\delta_A^n dA\}$ are unbounded, the convergence proof of Eq. (2.17) can be studied using the strong norm convergence. Then, the condition (2.18a) is replaced by

$$\alpha'_1 \equiv \overline{\lim}_{n \rightarrow \infty} \left\| \frac{f^{(n+1)}(A)}{(n+1)!} \delta_A^n dA \psi \right\|^{\frac{1}{n}} < 1 \tag{2.18b}$$

for $\psi \in \mathcal{D}$ with some appropriate domain \mathcal{D} in Hilbert space.

§3. Higher-order quantum derivatives and nonlinear responses in equilibrium

The higher-order quantum derivative $d^n f(A)/dA^n$ is formally expressed¹⁾ by the multiple integral

$$\frac{d^n f(A)}{dA^n} = n! \int_0^1 dt_1 \int_0^{t_1} dt_2 \cdots \int_0^{t_{n-1}} dt_n f^{(n)}(A - t_1 \delta_1 - \cdots - t_n \delta_n), \tag{3.1}$$

where $f^{(n)}(x)$ denotes the n -th derivative of $f(x)$ and the inner derivation δ_j is defined by

$$\delta_j : dA \cdot dA \cdots dA = dA \cdot dA \cdots (\delta_A dA) \cdots dA. \tag{3.2}$$

Then we have the following operator Taylor expansion formula:^{1),2)}

$$f(A + xB) = \sum_{n=0}^{\infty} \frac{x^n}{n!} \frac{d^n f(A)}{dA^n} : B^n, \tag{3.3}$$

with the notation $B^n = B \cdots B$.

It is sometimes important to study nonlinear responses in condensed matter physics, as in spin glasses (in which only nonlinear susceptibilities diverge^{11),12)} at the transition point).

As is well known, an equilibrium system is described by the canonical density matrix

$$\rho = e^{-\beta(\mathcal{H} - HQ)} \tag{3.4}$$

for the Hamiltonian \mathcal{H} of the system in the presence of an external field H conjugate to a physical quantity Q . When Q does not commute with \mathcal{H} , nonlinear responses are described in terms of the canonical correlations of Q , namely by a multiple integral of the time correlation function of Q using the Feynman formula. They are now expressed as

$$\rho = \sum_{n=0}^{\infty} \frac{(-H)^n}{n!} \frac{d^n e^{-\beta\mathcal{H}}}{d\mathcal{H}^n} : \underbrace{Q \cdots Q}_n \tag{3.5}$$

in quantum analysis. Thus the n -th order nonlinear response is expressed by the n -th order quantum derivative of ρ . The above expression (3.1) of higher derivatives of $f(\mathcal{H}) = e^{-\beta\mathcal{H}}$ in terms of the inner derivation $\delta_{\mathcal{H}}$ is convenient for evaluating the required nonlinear responses explicitly, for example, using the high-temperature expansion method. The above static perturbational expansion with respect to the external field H can be extended to that of the nonequilibrium density matrix $\rho(t)$ given by a solution of the von Neumann equation (4.1).

§4. Basic equations in nonequilibrium systems

As is well known, the density matrix $\rho(t)$ in a nonequilibrium system satisfies the von Neumann equation

$$i\hbar \frac{d}{dt} \rho(t) = [\mathcal{H}(t), \rho(t)] = \delta_{\mathcal{H}(t)} \rho(t) \tag{4.1}$$

for the time-dependent Hamiltonian $\mathcal{H}(t)$ of the relevant system.

Now we attempt to find a solution of the exponential form

$$\rho(t) = e^{-\eta(t)}. \tag{4.2}$$

Concerning the “entropy operator” $\eta(t)$, we have the following formula which was pointed out by Zubarev.⁴⁾

Formula 2: The entropy operator $\eta(t)$ defined in Eq. (4.2) satisfies the equation

$$i\hbar \frac{d\eta(t)}{dt} = [\mathcal{H}(t), \eta(t)]. \tag{4.3}$$

This is a simple example of the following general formula.³⁾

Formula 3: Any operator-valued function $f(\rho(t))$ of the density matrix $\rho(t)$ satisfies the equation

$$i\hbar \frac{d}{dt} f(\rho(t)) = [\mathcal{H}(t), f(\rho(t))]. \tag{4.4}$$

It is instructive to give here a compact proof due to quantum analysis:

$$\begin{aligned} i\hbar \frac{d}{dt} f(\rho(t)) &= i\hbar \frac{df(\rho(t))}{d\rho(t)} \frac{d\rho(t)}{dt} = \frac{df(\rho(t))}{d\rho(t)} \delta_{\mathcal{H}(t)} \rho(t) \\ &= -\frac{df(\rho(t))}{d\rho(t)} \delta_{\rho(t)} \mathcal{H}(t) = -\delta_{f(\rho(t))} \mathcal{H}(t) = [\mathcal{H}(t), f(\rho(t))]. \end{aligned} \tag{4.5}$$

Here we have used Eq. (2.16). The above equation (4.3) is our starting point for deriving the renormalized expansion scheme (5.10).

§5. General perturbation theory on the entropy operator in nonequilibrium systems

We formulate here a general perturbation expansion of the entropy operator for the Hamiltonian $\mathcal{H}(t)$ taking the form

$$\mathcal{H}(t) = \mathcal{H} - AF(t), \tag{5.1}$$

with a time-dependent external force $F(t)$ (as in Kubo's linear response theory^{5),6}). Here A denotes an operator conjugate to the external force $F(t)$. Now, we define the correction term $\eta'(t)$ in

$$\eta(t) = \Phi + \beta\mathcal{H} + \eta'(t) \quad (5.2)$$

for $\eta(t) = -\log \rho(t)$, where $\beta = 1/k_B T$ and Φ is a normalization constant such that

$$e^\Phi = \text{Tr} e^{-\beta\mathcal{H} - \eta'(-\infty)}. \quad (5.3)$$

We then expand the correction term $\eta'(t)$ as

$$\eta'(t) = \sum_{n=1}^{\infty} \eta_n(t), \quad (5.4)$$

so that $\eta_n(t)$ is of n -th order in $F(t)$. This is a new type of renormalized perturbation theory for nonequilibrium systems, because even the first-order term $\eta_1(t)$ gives partially infinite-order terms in the density matrix $\rho(t)$. It is easily shown from Formula 2 that $\eta'(t)$ satisfies the inhomogeneous equation

$$\frac{d}{dt}\eta'(t) = \frac{1}{i\hbar}[\mathcal{H}(t), \eta'(t)] - \beta F(t)\dot{A} \quad (5.5)$$

with the initial condition $\eta'(-\infty) = 0$, which corresponds to the condition

$$\rho(-\infty) = \rho_{\text{eq}} = e^{-\beta\mathcal{H}} / \text{Tr} e^{-\beta\mathcal{H}}. \quad (5.6)$$

Here we have also used the relation

$$\dot{A} = \frac{1}{i\hbar}[A, \mathcal{H}] = \frac{1}{i\hbar}\delta_A \mathcal{H}. \quad (5.7)$$

Equation (5.5) is the basic formula derived here. A new aspect of this equation is that it has the temperature-dependent source term $-\beta F(t)\dot{A}$. Since $\mathcal{H}(t)$ contains an external force $F(t)$, Eq. (5.5) is nonlinear with respect to this force. The linearized equation is given by

$$\frac{d}{dt}\eta_1(t) = \frac{1}{i\hbar}[\mathcal{H}, \eta_1(t)] - \beta F(t)\dot{A}. \quad (5.8)$$

The solution of Eq. (5.8) with the initial condition $\eta_1(-\infty) = 0$ is obtained as

$$\eta_1(t) = -\beta \int_{-\infty}^t F(s) \exp\left(\frac{1}{i\hbar}(t-s)\delta_{\mathcal{H}}\right) \dot{A} ds = -\beta \int_{-\infty}^0 e^{\varepsilon s} F(t+s) \dot{A}(s) ds. \quad (5.9)$$

The adiabatic factor $e^{\varepsilon s}$ has been inserted to insure convergence.

The above first-order approximation $\{\rho_1(t) = \exp[-\Phi - \beta\mathcal{H} - \eta_1(t)]\}$ gives Zubarev's statistical operator⁴ when the Hamiltonian $\mathcal{H}(t)$ is given by $\mathcal{H}(t) = \mathcal{H} - AF(t)$. This first-order approximation, namely Zubarev's theory, is justified if the second-order term $\eta_2(t)$ is much smaller than $\eta_1(t)$.

For higher-order correction terms of $\eta'(t)$, we have the following.

Formula 4: The higher-order entropy operators $\{\eta_n(t)\}$ are given by

$$\begin{aligned} \eta_2(t) &= -\frac{\beta}{i\hbar} \int_{-\infty}^0 e^{\varepsilon s} ds F(t+s) \int_0^s F(t+s') [A(s'), \dot{A}(s)] ds' \\ &\dots\dots\dots \\ \eta_n(t) &= -\frac{\beta}{(i\hbar)^{n-1}} \int_{-\infty}^0 e^{\varepsilon s} ds F(t+s) \int_0^s dt_1 \int_0^{t_1} dt_2 \dots \int_0^{t_{n-2}} dt_{n-1} \\ &\quad \times F(t+t_1) \dots F(t+t_{n-1}) \delta_{A(t_1)} \delta_{A(t_2)} \dots \delta_{A(t_{n-1})} \dot{A}(s), \end{aligned} \quad (5.10)$$

with the hyperoperator $\delta_{A(t)}$ and with $\dot{A} = (i\hbar)^{-1} \delta_A \mathcal{H}$.

These formulas can be derived from Eq. (5.5). They will be useful in studying nonlinear responses, because they are renormalized perturbational expansions in contrast to the ordinary perturbational expansion⁵⁾ of the density matrix itself. In fact, even the above $\rho_1(t)$ includes terms up to infinite order in $F(t)$. Thus, our formulation (5.10) is a new useful result, compared with the ordinary expansion scheme of $\rho(t)$ itself. The quantum analysis of dissipative systems^{13),14)} will be presented in Appendix C, using ordered exponentials and free Lie elements.^{15),16)}

§6. Linear response in terms of the inner derivation

In this section we discuss linear response as an application of the general perturbation theory presented in the preceding section, and we express it in terms of the inner derivation $\delta_{\mathcal{H}}$ for the relevant Hamiltonian \mathcal{H} .

The density matrix $\rho(t)$ for the Hamiltonian $\mathcal{H}(t)$ in Eq (5.1) is given by

$$\begin{aligned} \rho &= e^{-\Phi - (\beta\mathcal{H} + \eta_1(t))} \\ &= e^{-\Phi} \left(e^{-\beta\mathcal{H}} + \frac{de^{-\beta\mathcal{H}}}{d(\beta\mathcal{H})} \eta_1(t) \right) \end{aligned} \quad (6.1)$$

up to first-order in external force $F(t)$. Here, $\eta_1(t)$ is given by Eq. (5.9). The quantum derivative $de^{-\beta\mathcal{H}}/d(\beta\mathcal{H})$ is expressed by

$$\frac{de^{-\beta\mathcal{H}}}{d(\beta\mathcal{H})} = -e^{-\beta\mathcal{H}} \Delta(\beta\mathcal{H}), \quad (6.2)$$

as is seen from Eq. (2.6b). Thus, the first-order term $\Delta\rho(t)$ is given by

$$\Delta\rho(t) = -e^{-\Phi} e^{-\beta\mathcal{H}} \Delta(\beta\mathcal{H}) \eta_1(t). \quad (6.3)$$

The average of the relevant current operator $J = \dot{A}$ is expressed as

$$\begin{aligned} \langle J \rangle_t &= \text{Tr} \Delta\rho(t) J = -\langle (\Delta(\beta\mathcal{H}) \eta_1(t)) J \rangle \\ &= \beta \int_{-\infty}^0 e^{\varepsilon s} F(t+s) \langle (\Delta(\beta\mathcal{H}) J(s)) J \rangle ds \\ &= \beta \int_0^{\infty} e^{-\varepsilon s} F(t-s) \langle (\Delta(\beta\mathcal{H}) J(-s)) J \rangle ds \\ &\equiv \text{Re}(\sigma(\omega) F e^{i\omega t}) \end{aligned} \quad (6.4)$$

under the assumption that $\text{Tr } J \exp(-\beta\mathcal{H}) = 0$ and $F(t) = F \cos(\omega t)$. Here $\langle \dots \rangle$ denotes the average with respect to the equilibrium density matrix, and the general conductivity $\sigma(\omega)$ is expressed as

$$\sigma(\omega) = \beta \int_0^\infty e^{-\varepsilon s - i\omega s} \langle (\Delta(\beta\mathcal{H})J)J(s) \rangle ds; \quad (6.5)$$

namely

$$\begin{aligned} \sigma(\omega) &= \beta \left\langle (\Delta(\beta\mathcal{H})J) \frac{1}{i\omega - (i/\hbar)\delta\mathcal{H}} J \right\rangle \\ &= \frac{\beta}{i\omega} \sum_{n=0}^\infty \left\langle (\Delta(\beta\mathcal{H})J) \left[\left(\frac{1}{\hbar\omega} \delta\mathcal{H} \right)^n J \right] \right\rangle \end{aligned} \quad (6.6)$$

for the Planck constant \hbar , using the hyperoperator $\Delta(A)$ defined by Eq. (2.7a).

It is also interesting to note that we have

$$\int_0^\beta e^{\lambda\mathcal{H}} J e^{-\lambda\mathcal{H}} d\lambda = \int_0^\beta e^{\lambda\delta\mathcal{H}} d\lambda J = \beta \Delta(\beta\mathcal{H})J \quad (6.7)$$

for the current operator J in our notation. This may be thought of as the 'dressed current operator', due to quantum fluctuation. Thus, Kubo's canonical correlation $\langle J : J(t) \rangle$ is expressed as

$$\langle J : J(t) \rangle \equiv \frac{1}{\beta} \int_0^\beta \langle e^{\lambda\mathcal{H}} J e^{-\lambda\mathcal{H}} J(t) \rangle d\lambda = \langle (\Delta(\beta\mathcal{H})J)J(t) \rangle. \quad (6.8)$$

Then, the Kubo formula for the frequency-dependent conductivity $\sigma(\omega)$ is expressed in the form

$$\sigma(\omega) = \beta \int_0^\infty \langle J : J(t) \rangle e^{-i\omega t} dt = \beta \left\langle (\Delta(\beta\mathcal{H})J) \frac{1}{i\omega - (i/\hbar)\delta\mathcal{H}} J \right\rangle. \quad (6.9)$$

In particular, we obtain

$$\sigma(\omega) \simeq \frac{\beta}{i\omega} \langle (\Delta(\beta\mathcal{H})J)J \rangle \quad (6.10)$$

for large ω . Some remarks on applications of Eqs. (6.6) and (6.9) will be given in the succeeding section.

The present derivation of the Kubo formula may be more transparent and the algebraic structure that $\sigma(\omega)$ is expressed only in terms of the commutators of \mathcal{H} and J (namely free Lie elements) is convenient in practical calculations, as will be shown elsewhere.

§7. Some remarks on the conductivity $\sigma(\omega)$

It is instructive to give some remarks on applications of the formulas (6.6) and (6.9) for the conductivity $\sigma(\omega)$.

When the current J is a constant of motion, $\sigma(0)$ is infinite,^{17),18)} as seen from (6.5). We consider the following more general situation that the current operator J contains some (not necessarily all) constants of motion $\{H_j\}$, that is,

$$J = \sum_j a_j H_j + J', \tag{7.1}$$

where J' is defined by the remaining part of J orthogonal to all the $\{H_j\}$; namely J' is off-diagonal with respect to energy¹⁸⁾ (i.e., $\langle m | J' | n \rangle = 0$ for $E_m = E_n$ with the energy eigenvalues $\{E_n\}$ of the Hamiltonian \mathcal{H} even in a degenerate case). Here, the coefficients $\{a_j\}$ in (7.1), namely the ergodicity constants, are given¹⁸⁾ by

$$a_j = \langle JH_j \rangle / \langle H_j^2 \rangle, \tag{7.2}$$

using the orthogonality condition

$$\langle H_j H_k \rangle = \langle H_j^2 \rangle \delta_{jk}. \tag{7.3}$$

Thus, the zero frequency (or static isolated) conductivity defined by

$$\sigma(0) = \beta \int_0^\infty \langle J : J(t) \rangle dt \tag{7.4}$$

is seen to diverge as

$$\sigma(0) = \beta \sum_j a_j^2 \int_0^\infty \langle H_j^2 \rangle dt + (\text{finite}) \rightarrow \infty, \tag{7.5}$$

when at least one of the ergodicity constants $\{a_j\}$ is non-vanishing. This remark is useful in practical applications^{6),19)} of the Kubo formula to some exactly soluble systems^{17),18)} with an infinite number of constants of motion.

§8. Concluding remarks

The quantum analysis introduced in previous papers¹⁾⁻³⁾ has been extended to the case of an unbounded operator A in a Hilbert space by restricting our consideration to the three typical operator functions e^{-A} , $1/A$ and $\log A$ under the situation that the differential dA is bounded. The proof is rather easy, but it is instructive for studying more difficult cases for unbounded operators.

Our new expressions of response functions in terms of the inner derivation $\delta_{\mathcal{H}}$ (or the dressed current operator $\Delta(\beta\mathcal{H})J$) are convenient for analytic and numerical calculations of these response functions. This result should be compared with the abstract operator representation of a KMS-state by Naudts, Verbeure and Weder²⁰⁾ in the more complicated situation of infinite systems.

The renormalized perturbation scheme of the density matrix $\rho(t)$ is one of the new results in the present paper. This is in sharp contrast to Kubo's well-known systematic expansion formula⁵⁾ of $\rho(t)$ itself, rather than $\log \rho(t)$.

It is also interesting to note that the quantum analysis is useful in expressing an exponential product of a dissipative density matrix in terms of a single exponential (namely the generalized BCH formula) composed only of commutators, as is exemplified in Appendix C.

Transport coefficients are also expressed in terms of commutators of the relevant current operators.

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Appendix A

— Convergence of Eq. (2.5) —

The convergence of Eq. (2.5) is shown as follows:

$$\begin{aligned}
 & \left\| (e^{-(A+hB)} - e^{-A}) / h - \int_0^1 e^{-(1-s)A} (-B) e^{-sA} ds \right\| \\
 & \leq \left\| \frac{1}{h} \int_0^h dt \int_0^1 ds \left[e^{-(1-s)(A+tB)} B \{ e^{-s(A+tB)} - e^{-sA} \} \right. \right. \\
 & \quad \left. \left. + \{ e^{-(1-s)(A+tB)} - e^{-(1-s)A} \} B e^{-sA} \right] \right\| \\
 & \leq \frac{\|h\| \cdot \|B\|^2}{2} \max_{|t| \leq |h|} \left[\int_0^1 ds \| e^{-(1-s)(A+tB)} \| \right. \\
 & \quad \times \int_0^s d\lambda \| e^{-(s-\lambda)(A+tB)} \| \cdot \| e^{-\lambda(A+tB)} \| \\
 & \quad \left. + \int_0^1 ds \int_0^{1-s} d\lambda \| e^{-(1-s-\lambda)(A+tB)} \| \cdot \| e^{-\lambda(A+tB)} e^{-sA} \| \right], \quad (\text{A.1})
 \end{aligned}$$

when A is a positive (but unbounded) operator and $B = dA$ is bounded. Therefore, we arrive at Eq. (2.5).

Appendix B

— Expansion Formulas and Convergence of Higher-Order Derivatives —

The n -th derivative of $f(A)$ is given by Eq. (3.1), namely by the following integral:¹⁾

$$\frac{d^n f(A)}{dA^n} = n! \int_0^1 dt_1 \cdots \int_0^{t_{n-1}} dt_n f^{(n)} \left(A - \sum_j^n t_j \delta_j \right). \quad (\text{B.1})$$

Here, δ_j is a hyperoperator defined by Eq. (3-2), namely by

$$\delta_j : (dA)^n = (dA)^{j-1}(\delta_A dA)(dA)^{n-j}. \tag{B-2}$$

This is also formally expanded as follows.

Formula A:

$$\frac{d^n f(A)}{dA^n} = \sum_{m=0}^{\infty} \frac{n!(-1)^m}{m!} f^{(n+m)}(A) \int_0^1 dt_1 \cdots \int_0^{t_{n-1}} dt_n \left(\sum_j^n t_j \delta_j \right)^m. \tag{B-3}$$

For example, the first derivative $df(A)/dA$ is given by Eq. (2-17), and

$$\begin{aligned} \frac{d^2 f(A)}{dA^2} &= \sum_{m=0}^{\infty} \frac{2!(-1)^m}{(m+2)!} f^{(m+2)}(A) \frac{1}{\delta_2} [(\delta_1 + \delta_2)^{m+1} - \delta_1^{m+1}], \\ \frac{d^3 f(A)}{dA^3} &= \sum_{m=0}^{\infty} \frac{3!(-1)^m}{(m+3)!} f^{(m+3)}(A) \left\{ \frac{\delta_1^{m+2}}{\delta_2(\delta_2 + \delta_3)} \right. \\ &\quad \left. - \frac{(\delta_1 + \delta_2)^{m+2}}{\delta_2 \delta_3} + \frac{(\delta_1 + \delta_2 + \delta_3)^{m+2}}{(\delta_2 + \delta_3)\delta_3} \right\}, \dots \end{aligned} \tag{B-4}$$

The expansion of

$$d^n f(A) \equiv \frac{d^n f(A)}{dA^n} : (dA)^n \tag{B-5}$$

converges in the uniform norm topology when

$$\alpha_n \equiv \lim_{m \rightarrow \infty} \left\| \frac{f^{(n+m)}(A)}{m!} \int_0^1 dt_1 \cdots \int_0^{t_{n-1}} dt_n \left(\sum_j^n t_j \delta_j \right)^m (dA)^n \right\|^{\frac{1}{m}} < 1. \tag{B-6}$$

Appendix C

— Quantum Analysis of Dissipative Density Matrices —

It is instructive to discuss first the following non-dissipative unitary case.

(i) Unitary case. Here we discuss the von Neumann equation

$$i\hbar \frac{d}{dt} \rho(t) = [\mathcal{H}(t), \rho(t)] = \delta_{\mathcal{H}(t)} \rho(t) \tag{C-1}$$

for the time-dependent Hamiltonian $\mathcal{H}(t)$ of the relevant system, as in (4-1). A formal solution of Eq. (C-1) is given by

$$\begin{aligned} \rho(t) &= \exp_+ \left(\frac{1}{i\hbar} \int_0^t \delta_{\mathcal{H}(s)} ds \right) \rho(0) \\ &= \exp_+ \left(\frac{1}{i\hbar} \int_0^t \mathcal{H}(s) ds \right) \rho(0) \exp_- \left(-\frac{1}{i\hbar} \int_0^t \mathcal{H}(s) ds \right). \end{aligned} \tag{C-2}$$

Here, we have used the following ordered exponentials:^{13),14)}

$$\exp_+ \int_0^t A(s) ds = 1 + \int_0^t A(s) ds + \cdots + \int_0^t dt_1 \cdots \int_0^{t_{n-1}} dt_n A(t_1) \cdots A(t_n) + \cdots \tag{C-3}$$

and

$$\exp_- \int_0^t A(s) ds = 1 + \int_0^t A(s) ds + \cdots + \int_0^t dt_1 \cdots \int_0^{t_{n-1}} dt_n A(t_n) \cdots A(t_1) + \cdots \quad (\text{C-4})$$

Clearly the ordered exponentials

$$\exp_+ \left(\frac{1}{i\hbar} \int_0^t \mathcal{H}(s) ds \right) \quad \text{and} \quad \exp_- \left(-\frac{1}{i\hbar} \int_0^t \mathcal{H}(s) ds \right) \quad (\text{C-5})$$

are both unitary and consequently are bounded when $\{\mathcal{H}(s)\}$ are self-adjoint. Thus, the arguments in §2 can also be applied to these ordered exponentials, namely operator functionals.²¹⁾ It is then shown that the operator functional derivation²¹⁾ for the variation $\delta\mathcal{H}(t_1)$

$$\begin{aligned} dF[\mathcal{H}(t)]_{t_1} &\equiv d \left[\exp_+ \left(\frac{1}{i\hbar} \int_0^t \mathcal{H}(s) ds \right) \right]_{t_1} \equiv \frac{\delta F[\mathcal{H}(t)]}{\delta \mathcal{H}(t_1)} \cdot \delta \mathcal{H}(t_1) \\ &= \exp_+ \left(\frac{1}{i\hbar} \int_{t_1}^t \mathcal{H}(s) ds \right) \delta \mathcal{H}(t_1) \exp_+ \left(\frac{1}{i\hbar} \int_0^{t_1} \mathcal{H}(s) ds \right) \end{aligned} \quad (\text{C-6})$$

is bounded when the elements of $\{\mathcal{H}(t)\}$ are self-adjoint and the elements of $\{\delta\mathcal{H}(t_1)\}$ are bounded. Similarly the operator functional derivation of $\exp_-[-\frac{1}{i\hbar} \int_0^t \mathcal{H}(s) ds]$ is bounded under the same conditions.

(ii) Dissipative case. We discuss here the unnormalized density operator $\hat{\rho}(t)$ of a dissipative system described by the master equation

$$\frac{d\hat{\rho}(t)}{dt} = \frac{1}{i\hbar} [\mathcal{H}, \hat{\rho}(t)] + \Lambda \hat{\rho}(t) + \hat{\rho}(t) \Lambda^\dagger. \quad (\text{C-7})$$

Here, Λ and Λ^\dagger denote some bounded operators expressing a dissipative effect. The Hamiltonian \mathcal{H} may be unbounded. The normalized density matrix $\rho(t)$ is given by $\rho(t) = N(t)\hat{\rho}(t)$ with $N(t)^{-1} = \text{Tr}\hat{\rho}(t)$. A formal solution of Eq. (C-7) is given as follows. First we put

$$\hat{\rho}(t) = \exp \left(\frac{t}{i\hbar} \mathcal{H} \right) f(t) \exp \left(-\frac{t}{i\hbar} \mathcal{H} \right). \quad (\text{C-8})$$

Then, Eq. (C-7) can be rewritten as

$$\frac{df(t)}{dt} = \Lambda_t f(t) + f(t) \Lambda_t^\dagger, \quad (\text{C-9})$$

where

$$\Lambda_t = \exp \left(-\frac{t}{i\hbar} \mathcal{H} \right) \Lambda \exp \left(\frac{t}{i\hbar} \mathcal{H} \right) \quad (\text{C-10})$$

and $\Lambda_t^\dagger = (\Lambda_t)^\dagger$. Next we put

$$f(t) = \exp_+ \left(-\int_0^t \Lambda_s^\dagger ds \right) g(t) \exp_- \left(\int_0^t \Lambda_s^\dagger ds \right) \quad (\text{C-11})$$

with $g(0) = f(0) = \hat{\rho}(0)$. Then Eq. (C-9) can again be rewritten as

$$\frac{dg(t)}{dt} = \mathcal{L}(t)g(t), \tag{C-12}$$

where

$$\mathcal{L}(t) = \exp_- \left(\int_0^t \Lambda_s^\dagger ds \right) (\Lambda_t + \Lambda_t^\dagger) \exp_+ \left(- \int_0^t \Lambda_s^\dagger ds \right). \tag{C-13}$$

A solution of Eq. (C-12) is given by

$$g(t) = \exp_+ \left(\int_0^t \mathcal{L} ds \right) g(0). \tag{C-14}$$

Thus we arrive at

$$\hat{\rho}(t) = \exp_+ \left(\int_0^t \mathcal{L}(s, t) ds \right) \hat{\rho}(t, 0), \tag{C-15}$$

where

$$\begin{aligned} \mathcal{L}(s, t) &= \exp \left(\frac{t}{i\hbar} \mathcal{H} \right) \exp_+ \left(- \int_0^t \Lambda_s^\dagger ds \right) \mathcal{L}(s) \\ &\quad \times \exp_- \left(\int_0^t \Lambda_s^\dagger ds \right) \exp \left(- \frac{t}{i\hbar} \mathcal{H} \right), \end{aligned} \tag{C-16}$$

and

$$\begin{aligned} \hat{\rho}(t, 0) &= \exp \left(\frac{t}{i\hbar} \mathcal{H} \right) \exp_+ \left(- \int_0^t \Lambda_s^\dagger ds \right) \hat{\rho}(0) \\ &\quad \times \exp_- \left(\int_0^t \Lambda_s^\dagger ds \right) \exp \left(- \frac{t}{i\hbar} \mathcal{H} \right). \end{aligned} \tag{C-17}$$

When both $\hat{\rho}(0)$ and Λ are bounded, $\hat{\rho}(t)$ is also bounded.

Now we put

$$\hat{\rho}(t, 0) = e^{-\eta(t, 0)}. \tag{C-18}$$

Then, we have

$$\hat{\rho}(t) = \exp_+ \left(\int_0^t \mathcal{L}(s, t) ds \right) \exp(-\eta(t, 0)). \tag{C-19}$$

Our purpose here is to find the logarithm of Eq. (C-19). For this, we put

$$\exp_+ \left(\int_0^x \mathcal{L}(s, t) ds \right) \exp(-\eta(t, 0)) = e^{\Phi(x)}. \tag{C-20}$$

Clearly we have $\Phi(0) = -\eta(t, 0)$. By differentiating Eq. (C-20) with respect to x , we obtain

$$e^{\Phi(x)} \Delta(-\Phi(x)) \frac{d\Phi(x)}{dx} = \mathcal{L}(x, t) e^{\Phi(x)}. \tag{C-21}$$

This is transformed into the equation

$$\begin{aligned} \frac{d\Phi(x)}{dx} &= \Delta^{-1}(-\Phi(x)) e^{-\delta_{\Phi(x)}} \mathcal{L}(x, t) \\ &= \Delta^{-1}(\Phi(x)) \mathcal{L}(x, t) \\ &= \frac{\delta_{\Phi(x)}}{e^{\delta_{\Phi(x)}} - 1} \mathcal{L}(x, t) = \frac{\log e^{\delta_{\Phi(x)}}}{e^{\delta_{\Phi(x)}} - 1} \mathcal{L}(x, t), \end{aligned} \tag{C-22}$$

using the identity $\delta_{\Phi} = \log e^{\delta_{\Phi}}$. Then we can apply Eq. (C-20) to Eq. (C-21). Thus we finally arrive at the following formula.

Formula B: The entropy operator $\hat{\eta}(t)$ of the system described by Eq. (C-7) is expressed in the form

$$\begin{aligned}\hat{\eta}(t) &= -\Phi(t) \equiv -\log \hat{\rho}(t) \\ &= \eta(t, 0) - \int_0^t dx \frac{\log[\exp_+(\int_0^x \delta_{\mathcal{L}(s,t)} ds) \exp(-\delta_{\eta(t,0)})]}{\exp_+(\int_0^x \delta_{\mathcal{L}(s,t)} ds) \exp(-\delta_{\eta(t,0)}) - 1} \mathcal{L}(x, t). \quad (\text{C-23})\end{aligned}$$

The final expression (C-23) is much more convenient than

$$\hat{\eta}(t) = -\log \left[\exp_+ \left(\int_0^t \mathcal{L}(s, t) ds \right) \exp(-\eta(t, 0)) \right], \quad (\text{C-24})$$

because Eq. (C-23) is expressed in terms of the commutators of $\{\mathcal{L}(s, t)\}$ and $\eta(t, 0)$, namely free Lie elements.¹⁵⁾

The present formulation can be easily extended to the following more general dissipative system:

$$\frac{d\hat{\rho}(t)}{dt} = \frac{1}{i\hbar} [\mathcal{H}(t), \hat{\rho}(t)] + \Lambda(t)\hat{\rho}(t) + \hat{\rho}(t)\Lambda^\dagger(t). \quad (\text{C-25})$$

The expression (C-23) is convenient for studying quantum effects¹⁶⁾ in nonequilibrium systems.

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