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Entropy Production and Its Positivity in Nonlinear Response Theory of Quantum Dynamical Systems

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A formulation of *entropy production* is given with the aid of relative entropy in the nonlinear response theory of a quantum dynamical system. It allows a natural interpretation of the quantity in terms of familiar thermodynamic notions, such as force and response current conjugate to it, without sacrificing the full nonlinearity in the perturbing force. For the understanding of *dissipativity* as *positive* entropy production, the stationarity of states and coarse graining of time scale turn out to be essential, which are implemented by some time averaging procedures involving *almost periodic* external forces. Finally, it is shown that the obtained result reduces, in the linear response regime, to the power dissipation appearing in the well-known fluctuation-dissipation relation.

KEY WORDS: Entropy production; relative entropy; nonlinear response theory; almost periodic perturbation.

1. INTRODUCTION

A statistical mechanical description of irreversible processes was formulated by Kubo in 1957,[(1)] which goes by the name of *linear response theory*.[(2)] This was motivated in part by the prior work of Callen and Welton,[(3)] who proposed a quantum mechanical perturbative calculation with the external forces exerted on a dissipative system. They pointed out a general relationship between the power dissipation induced by the perturbation and the average of a squared fluctuation of the current of the system in thermal equilibrium. This is a prototype of the fluctuation-dissipation

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theorem, which was discussed most comprehensively by Kubo subsequently.

A characteristic feature of the theory was that it abandoned the use of kinetic equations by adopting the formalism of Hamiltonian dynamics. Thus, this has sometimes led to a question about the origin of irreversibility in the description, as exemplified by van Kampen's criticism[4]: He argued that the macroscopically observable linearity of irreversible phenomena must be entirely different from the linearity in the logic of the linear response theory and that the irreversibility in the latter description was provided virtually through the procedure of the linearity approximation in it. However, Kubo *et al.*[2] replied to this criticism by claiming the general legitimacy of interchanging the two procedures in computing transport coefficients, namely, *stochastization* (randomization) and *linearization*: Both linear response theory and kinetic-equation theory have a wide range of common applicability that should stem from this legitimacy.

Although nowadays there is no doubt about the validity and utility of the linear response theory, the above-mentioned issue does not seem to be settled yet. This motivates us to seek a better understanding of the problem. The point of investigation can be specified by the following two questions:

1. Would the response theory, extended beyond the linear regime, still be valid for discussing nonequilibrium dissipative phenomena?
2. How can the dissipation be understood in such a response theory from the standard thermodynamic viewpoint?

To examine the above questions in more detail, we investigate the dynamical change of a quantum statistical state by adopting the framework of C^*-dynamical systems[5] with time-inhomogeneous perturbations. More explicitly, we ask how a state φ of a C^*-algebra $\mathfrak{A}$ changes when it is driven by a time-dependent external force $\mathbf{X}(t)$ in the perturbing Hamiltonian

$$\begin{aligned} H_1(t) &= -\mathbf{A}\cdot\mathbf{X}(t) \\ \mathbf{A} &= (A_1, A_2, ..., A_r), \qquad A_i \in \mathfrak{A} \end{aligned} \tag{1.1}$$

As is the case in the Kubo formalism, the state φ is assumed to be a canonical equilibrium state ω_β with temperature β^{-1} at an initial time t_0 (which is later shifted away to $-\infty$ by some limiting procedures). A major problem is then to clarify how far from the initial state ω_β the time-dependent state φ_t evolves and whether the "distance" of φ_t from ω_β increases or decreases. Since the usual von Neumann entropy of the total system does not change in the "unitary" time evolution of a dynamical system, it cannot be used for describing the dissipativity in the present context. What we are going to apply to this question is the notion of *relative entropy*[6,7] $S(\psi \mid \varphi)$ of two states ψ and φ. When the states are represented by density matrices (in a Hilbert space of an irreducible representation of the observable algebra) in such a way that

$$\varphi(A) = \operatorname{tr} \rho_\varphi A \tag{1.2}$$

the relative entropy can be expressed as

$$S(\psi \mid \varphi) = \operatorname{tr} \rho_\psi (\log \rho_\psi - \log \rho_\varphi) \tag{1.3}$$

[The above notation for $S(\psi \mid \varphi)$ conforms to Ref. 7, but not to Ref. 6.] Under the same circumstances, the equilibrium density matrix is given by

$$\rho_{\omega_\beta} (\equiv \rho_\beta) = e^{-\beta(H - F_\beta)}, \qquad e^{-\beta F_\beta} = \operatorname{tr} e^{-\beta H} \tag{1.4}$$

in terms of the system Hamiltonian H, i.e., the unperturbed Hamiltonian. The expression (1.4) is valid, however, only for those Hamiltonian systems with *discrete spectrum*. For the study of irreversible processes it is necessary to adopt a more general class of states and dynamics that admit a *continuous* energy spectrum. Hence, we replace the formulation (1.2)–(1.4) in terms of density matrices by that of the GNS representation associated with the KMS state of the observable algebra. With this understanding, the results to be given in the subsequent sections are outlined as follows: Since the time evolution of observables in $\mathfrak{A}$ involves time-dependent forces $\mathbf{X}(t)$, the family of time evolution mappings on $\mathfrak{A}$ does not form a one-parameter group $\{\alpha_t : t \in \mathbb{R}\}$, but forms a family of two-time indexed automorphisms, each member of which we denote by $\alpha_{s \to t}$ $(s, t \in \mathbb{R})$. It operates on an observable A as well as on a state φ satisfying the following relations:

$$\alpha_{s \to t}(A_s) = A_t, \qquad \varphi_t = \varphi_s \circ \alpha_{s \to t} \tag{1.5}$$

$$\varphi_s(A_t) = \varphi_t(A_s) \tag{1.6}$$

$$\alpha_{r \to s} \circ \alpha_{s \to t} = \alpha_{r \to t} \qquad \text{(chain rule)} \tag{1.7}$$

$$\alpha_{s \to s} = \mathrm{Id}_{\mathfrak{A}} \ (\equiv \text{identity mapping on } \mathfrak{A}) \tag{1.8}$$

From (1.7) and (1.8) it follows that $\alpha_{t \to s} = \alpha_{s \to t}^{-1}$. Accordingly, our main result in Section 3 can be expressed as

$$\begin{aligned} S(\varphi_t \mid \omega_\beta) &= \beta \int_{t_0}^{t} \varphi_\tau(\mathbf{J}_A) \cdot \mathbf{X}(\tau)\, d\tau \\ &= \beta \int_{t_0}^{t} \omega_\beta \circ \alpha_{t_0 \to \tau}(\mathbf{J}_A) \cdot \mathbf{X}(\tau)\, d\tau \end{aligned} \tag{1.9}$$

where

$$\mathbf{J}_A = i[H, \mathbf{A}] \tag{1.10}$$

is the current associated with the observable **A**. By interpreting $\omega_\beta(\alpha_{t_0 \to \tau}(\mathbf{J}_A))$ as the conjugate flux to the external force **X**, the integrand of (1.9) can be identified with the thermodynamic notion of *entropy production*.[(8)] This is an analogue of the entropy production discussed in Ref. 9 in the context of an open system described by a dynamical semigroup of completely positive maps.

However, the entropy production[4] $P(t, t_0) \equiv \beta\varphi_t(\mathbf{J}_A) \cdot \mathbf{X}(t)$ $[= (d/dt) S(\varphi_t \mid \omega_\beta)]$ is generally a time-dependent quantity depending on both the present time t and the initial time t_0, and its positivity characterizing the dissipativity of the system cannot be seen directly. In the linear response regime where the **X** dependence of the time development $\alpha_{t \to t_0}$ is retained up to first order, the positivity is ensured by the additional ingredient of the Kubo formalism, namely, the limiting procedure $t_0 \to -\infty$ letting the external disturbance set in at the infinite past. By this procedure, the stationarity of the state φ_t at t is realized automatically if the external force is periodic in time. Beyond the linear regime, however, this stationarity is not evident even in the limit $t_0 \to -\infty$, and considerations about the mechanism of randomizing the fluctuation of the force are needed for the definiteness of the entropy production. In Section 4, we consider a broad class of external forces, namely those expressed by *almost periodic functions* of time, and establish the following results:

1. If the external force $\mathbf{X}(t)$ is almost periodic, so is the entropy production

$$P(t + t_0, t_0) = \omega_\beta(\alpha_{t_0 \to t + t_0}(\mathbf{J}_A)) \cdot \mathbf{X}(t)$$

as a function of t_0 with t fixed, $t_0 \mapsto P(t + t_0, t_0)$. Therefore, its average over t_0 exists uniquely:

$$P(t) \equiv \lim_{T_0 \to \infty} \frac{1}{T_0} \int_{-T_0}^{0} dt_0 \, P(t + t_0, t_0) \tag{1.11}$$

[4] The apparent sign difference of $P(t, t_0)$ from $\sigma = -(d/dt) S(\rho_t \mid \rho_\beta)|_{t=0}$ of Ref. 9 is simply due to the difference in the treated situations: Here in the response theory we are considering an "uphill" process toward a final nonequilibrium state starting from an initial equilibrium state ω_β, whereas Ref. 9 treats a "downhill" process with a nonequilibrium state ρ as its starting point whose final destination will be the equilibrium state ρ_β.

2. If the long-time average $\bar{P}$ of $P(t)$ exists, then it represents the nonnegative entropy production for a stationary state:

$$\bar{P} \equiv \lim_{T \to \infty} \frac{1}{T} \int_0^T dt\, P(t) \geqslant 0 \tag{1.12}$$

In short, the relative entropy of a driven system may increase or decrease, but *it increases on the average.*

In Section 5 we restate the result in the form of a nonlinear response version of the fluctuation-dissipation theorem. It will also be explicitly shown that the mean entropy production so defined reduces, in the linear response regime, to the power dissipation in the Kubo formula.

2. PERTURBED DYNAMICS AND THE RELATIVE ENTROPY FORMULA. I. THE CASE OF THE DENSITY-MATRIX STATE

In order to illustrate the essence of the story, this section is devoted to a discussion of the relative entropy formula in terms of the familiar density matrix. We then proceed in the next section to remove its limitation by adopting a general framework which involves some mathematical terminology and techniques. We hope that such a presentation will make our derivation of the formula expository enough without losing its mathematical rigor.

Let us consider the Schrödinger equation for a quantum system with a Hamiltonian H, a self-adjoint operator acting on a Hilbert space $\mathfrak{H}_0$. When an external perturbation of the form (1.1) is switched on to the system, its time evolution will be governed by a unitary operator $\mathcal{U}(t, s)$ on $\mathfrak{H}_0$, defined as the solution to the equation of motion with an initial value at $t = s$ as follows:

$$\begin{aligned} \frac{\partial}{\partial t} \mathcal{U}(t, s) &= -i[H - \mathbf{A} \cdot \mathbf{X}(t)]\, \mathcal{U}(t, s) \\ \mathcal{U}(t = s, s) &= \mathbb{1} \end{aligned} \tag{2.1}$$

The unperturbed Hamiltonian H (unbounded, in general) can be eliminated from this equation by taking the interaction representation: Define another unitary operator $U(t, s)$ through

$$\mathcal{U}(t, s) = e^{-itH} U(t, s)\, e^{isH} \tag{2.2}$$

Then, we obtain

$$\frac{\partial}{\partial t} U(t, s) = i\mathbf{A}_t \cdot \mathbf{X}(t)\, U(t, s) \tag{2.3}$$

$$U(t = s, s) = \mathbb{1} \tag{2.3a}$$

and also

$$\frac{\partial}{\partial s} U(t, s) = -U(t, s)\, i\mathbf{A}_s \cdot \mathbf{X}(s) \tag{2.4}$$

$$U(t, s = t) = \mathbb{1} \tag{2.4a}$$

where $\mathbf{A}_s$ is defined by

$$\mathbf{A}_s \equiv e^{isH} \mathbf{A} e^{-isH} \tag{2.5}$$

The equation of motion for a density matrix ρ of the system is given as usual by

$$d\rho/dt = i[\rho, H - \mathbf{A} \cdot \mathbf{X}(t)]; \qquad \rho(t = t_0) = \rho_0 \tag{2.6}$$

whose solution is given by

$$\begin{aligned} \rho(t) &= \mathscr{U}(t, t_0)\, \rho_0 \mathscr{U}(t, t_0)^* \\ &= e^{-itH} U(t, t_0)\, e^{it_0 H} \rho_0 e^{-it_0 H} U(t, t_0)^*\, e^{itH} \end{aligned} \tag{2.7}$$

If ρ_0 is the canonical equilibrium state ρ_β of the unperturbed Hamiltonian H at the temperature β^{-1} given by (1.4), then, due to the stationarity of ρ_β, (2.7) reduces to

$$\rho(t) = e^{-itH} U(t, t_0)\, \rho_\beta U(t, t_0)^*\, e^{itH} \tag{2.8}$$

Let us put this density matrix $\rho(t)$ in the form

$$\rho(t) = e^{-\beta[H - F(t)]} \tag{2.9}$$

with an *operator-valued free energy* $F(t) = F(t)^*$. The operator nature of $F(t)$ is due to the deviation of $\rho(t)$ from the equilibrium, as can be seen in the following. From Eq. (2.8) together with (2.9), we obtain

$$\begin{aligned} F(t) &= \beta^{-1} e^{-itH} U(t, t_0) \log \rho_\beta\, U(t, t_0)^*\, e^{itH} + H \\ &= e^{-itH} U(t, t_0)[F_\beta + U(t, t_0)^*\, HU(t, t_0) - H] \\ &\quad \times U(t, t_0)^*\, e^{itH} \end{aligned} \tag{2.10}$$

Note that $[\cdots]$ in the above expression is given by

$$\begin{aligned} &F_\beta + \int_{t_0}^{t} d\tau \frac{d}{d\tau} [U(\tau, t_0)^*\, HU(\tau, t_0)] \\ &\quad = F_\beta + \int_{t_0}^{t} d\tau\, U(\tau, t_0)^* [iH, \mathbf{A}_\tau \cdot \mathbf{X}(\tau)]\, U(\tau, t_0) \\ &\quad = F_\beta + \int_{t_0}^{t} d\tau\, e^{it_0 H} \alpha_{t_0 \to \tau}(\mathbf{J}_A)\, e^{-it_0 H} \cdot \mathbf{X}(\tau) \end{aligned}$$

with

$$\alpha_{t_0 \to \tau}(\mathbf{J}_A) = \mathscr{U}(\tau, t_0)^* [iH, A] \, \mathscr{U}(\tau, t_0)$$
$$= e^{-it_0 H} U(\tau, t_0)^* \, e^{i\tau H} \mathbf{J}_A e^{-i\tau H} U(\tau, t_0) \, e^{it_0 H} \tag{2.11}$$

Thus, we obtain

$$\log \rho(t) - \log \rho_\beta$$
$$= \beta \mathscr{U}(t, t_0) \left[\int_{t_0}^{t} \alpha_{t_0 \to \tau}(\mathbf{J}_A) \cdot \mathbf{X}(\tau) \, d\tau \right] \mathscr{U}(t, t_0)^* \tag{2.12}$$

and hence

$$S(\varphi_{\rho(t)} \,|\, \omega_\beta) = \operatorname{tr} \rho(t) \, [\log \rho(t) - \log \rho_\beta]$$
$$= \beta \int_{t_0}^{t} \operatorname{tr}[\rho_\beta \alpha_{t_0 \to \tau}(\mathbf{J}_A)] \cdot \mathbf{X}(\tau) \, d\tau$$
$$= \beta \int_{t_0}^{t} \operatorname{tr}[\rho(\tau) \, \mathbf{J}_A] \cdot \mathbf{X}(\tau) \, d\tau \tag{2.13}$$

This is just the desired result in its density matrix version.[10]

As remarked in Section 1, however, the validity of the above version is restricted to the case where the Hamiltonian H has discrete spectra only [otherwise, the density matrices ρ_β and $\rho(t)$ do not belong to the trace-class operators in the Hilbert space $\mathfrak{H}_0$]. To get rid of this limitation, one should adopt the general algebraic formulation of quantum dynamical systems[5] where the notion of the modular automorphism group associated with a KMS state plays an important role. This is the subject of the next section.

3. PERTURBED DYNAMICS AND THE RELATIVE ENTROPY FORMULA. II. THE GENERAL CASE

Let us take up a C^*-dynamical system consisting of a C^*-algebra $\mathfrak{A}$ of observables and of its one-parameter automorphism group $\{\alpha_t : t \in \mathbb{R}\}$ describing the time development of the system. In this setting, a state means an expectation functional in general, pure or mixed, which is formulated mathematically as a positive linear functional on $\mathfrak{A}$. In order to extend the notion of equilibrium Gibbs ensemble to a general infinite system where the familiar formula (1.4) is not valid, Haag *et al.*[11] extracted its essence in the form of the *KMS condition*[1,5,11]

$$\omega_\beta(A \alpha_t(B)) = \omega_\beta(\alpha_{t - i\beta}(B) \, A) \tag{3.1}$$

The state ω_β satisfying (3.1) for an arbitrary pair of observables A and B (precisely speaking, for those belonging to the dense subalgebra $\mathfrak{A}_\alpha$ of entire elements for the automorphism group $\{\alpha_t\}$) is called a *KMS state* of the C^*-dynamical system $(\mathfrak{A}, \alpha_t)$ at the inverse temperature β, or a *β-KMS state* for short. While this is a direct consequence of (1.4) following from the cyclic invariance of the trace and $\alpha_t(B) = e^{itH}Be^{-itH}$, the condition (3.1) itself is free from the restrictions inherent in the trace formula (1.4). The profound meaning behind this simple condition has been brought to light from both the physical and mathematical sides; for instance, in discussions about reciprocity,[1] the fluctuation-dissipation theorem,[1,2] and the stability of the equilibrium state,[5] through its relation to the Hawking–Unruh effect, and, above all, in its essential connection to the Tomita–Takesaki theory for von Neumann algebras.[5] With regard to the last-named context, some points relevant to our discussion will be briefly explained in what follows.

Starting from a β-KMS state ω_β defined above, we now consider external perturbations on it and investigate the problem of how and to what extent the state changes. For this purpose, we need some standard machinery of quantum theory, such as a Hilbert space, operators on it, and so on. This is supplied by the GNS representation $(\mathfrak{H}, \pi, \Omega, U_t)$ associated with the KMS state ω_β, where $\pi: \mathfrak{A} \to B(\mathfrak{H})$ is a representation of the C^*-algebra $\mathfrak{A}$ in a Hilbert space $\mathfrak{H}$ with a cyclic vector $\Omega \in \mathfrak{H}$, i.e.,

$$\overline{\pi(\mathfrak{A})\,\Omega} = \mathfrak{H} \tag{3.2}$$

satisfying

$$\omega_\beta(A) = \langle \Omega, \pi(A)\,\Omega \rangle \qquad \text{for} \quad \forall A \in \mathfrak{A} \tag{3.3}$$

$\{U_t : t \in \mathbb{R}\}$ is a one-parameter unitary group in $\mathfrak{H}$ implementing the (unperturbed) dynamical automorphism group $\{\alpha_t\}$:

$$\pi(\alpha_t(A)) = U_t \pi(A)\, U_t^*; \qquad U_t \Omega = \Omega \tag{3.4}$$

The special situation arising from the KMS condition (3.1) is the *separating* property of the cyclic vector Ω for the von Neumann algebra $\mathfrak{M} \equiv \pi(\mathfrak{A})''$ [= the closure of $\pi(\mathfrak{A})$ with respect to the weak operator topology in $B(\mathfrak{H})$],

$$x\Omega = 0 \ \text{ for } x \in \mathfrak{M} \Rightarrow x = 0 \tag{3.5}$$

which is equivalent to the cyclicity of Ω for $\mathfrak{M}'$ ($\equiv$ the commutant of $\mathfrak{M} \equiv \{y \in B(\mathfrak{H});\ [x, y] = 0 \text{ for } \forall x \in \mathfrak{M}\}$):

$$\overline{\mathfrak{M}'\Omega} = \mathfrak{H} \tag{3.6}$$

On the basis of (3.2) and (3.6), the following two (closable) antilinear operators S and F are defined in $\mathfrak{H}$ by the equations

$$Sx\Omega = x^*\Omega \qquad x \in \mathfrak{M} \tag{3.7}$$

$$Fx'\Omega = x'^*\Omega \qquad x' \in \mathfrak{M}' \tag{3.8}$$

Through the polar decomposition of (the closures $\bar{S}$ and $\bar{F}$ of) S and F,

$$\bar{S} = J\Delta^{1/2}\,(=F^*), \qquad \bar{F} = \Delta^{1/2}J\,(=S^*) \tag{3.9}$$

a strictly positive self-adjoint operator Δ $(\equiv S^*\bar{S} > 0)$ and an involutive antiunitary operator J (i.e., $J^2 = \mathbb{1}$, $J^{-1} = J^*$) are defined on $\mathfrak{H}$ and are called, respectively, the *modular operator* and the *modular conjugation operator*. The important properties about Δ and J are

$$\text{(i)}\quad \Delta^{it}\mathfrak{M}\Delta^{-it} = \mathfrak{M}; \qquad \text{(ii)}\quad J\mathfrak{M}J = \mathfrak{M}' \tag{3.10}$$

The property (i) allows us to define an automorphism group $\{\sigma_t : t \in \mathbb{R}\}$ on $\mathfrak{M}$, called the *modular automorphism group*, by

$$\mathfrak{M} \ni x \mapsto \sigma_t(x) = \Delta^{it}x\Delta^{-it} \in \mathfrak{M} \tag{3.11}$$

Using the property (ii), (3.7), and (3.9), one can show that this automorphism group satisfies the KMS condition with $\beta = -1$ for the extension $\hat{\omega}_\beta$ of ω_β to the von Neumann algebra $\mathfrak{M}$ $[=\pi(\mathfrak{A})'']$:

$$\hat{\omega}_\beta(x\sigma_t(y)) = \hat{\omega}_\beta(\sigma_{t+i}(y)\,x) \tag{3.12}$$

where

$$\hat{\omega}_\beta(x) \equiv \langle \Omega, x\Omega \rangle \qquad \text{for} \quad x \in \mathfrak{M} \tag{3.13}$$

Since the automorphism group satisfying the KMS condition for a given state is proved to be unique, we obtain

$$\hat{\alpha}_t(x) \equiv U_t x U_t^* = \sigma_{-t/\beta}(x) = \Delta^{-it/\beta}x\Delta^{it/\beta} \tag{3.14}$$

for $x \in \mathfrak{M}$. Therefore, the operator H_β defined by

$$H_\beta \equiv -\beta^{-1}\log\Delta \qquad \text{or} \qquad \Delta = e^{-\beta H_\beta} \tag{3.15}$$

can be thought of as a Hamiltonian in some extended sense.[5] From this,

[5] Due to $JH_\beta J = -H_\beta$ following from (3.7)–(3.9), H_β has positive and negative spectrum symmetrically. For the Hamiltonian system with discrete spectrum as in Section 2, the GNS representation is given by the left multiplication of operators in $\mathfrak{H}_0$ on the Hilbert–Schmidt class operators $\mathfrak{H} = \{\sigma \in B(\mathfrak{H}_0);\ \mathrm{tr}\,|\sigma|^2 < \infty\}$: $\pi(A)\,\sigma = A\sigma\,(\sigma \in \mathfrak{H})$; $\Omega = \rho_\beta = e^{-\beta H}/\mathrm{tr}\,e^{-\beta H}$. Here J and Δ are given, respectively, by $J\sigma = \sigma^*$ and $\Delta\sigma = e^{-\beta H}\sigma e^{\beta H}$, which implies $H_\beta = H - JHJ$. For the general infinite systems where the "genuine" Hamiltonian H is *ill-defined* in the GNS representation space $\mathfrak{H}$ of a KMS state, this decomposition becomes meaningless, while H_β itself survives taking the role of a Hamiltonian.

one can reasonably expect that the role of the equilibrium density matrix (1.4) will be played by the modular operator Δ.

Equipped with the above machinery, we extend the density-matrix version of the results in Section 2 to the general case in the following way. Since the von Neumann algebra $\mathfrak{M} = \pi(\mathfrak{A})''$ is the completion of $\pi(\mathfrak{A})$ with respect to the weak operator topology, any (Hermitian) operator in $\mathfrak{M}$ can be regarded as a generalized observable obtained from those belonging to the original observable algebra $\mathfrak{A}$ through some limiting procedure. Furthermore, $\mathfrak{A}$ is faithfully embedded in $\mathfrak{M}$ by the GNS representation π due to the faithfulness of the KMS state ω_β:

$$\pi(A) = 0 \rightarrow \|\pi(A)\,\Omega\|^2 = \omega_\beta(A^*A) = 0 \rightarrow A = 0$$

Therefore, *unless* the external force is so strong that the perturbed state φ_t gives rise to the GNS representation π_{φ_t} *disjoint* from our starting one π, our physical system can be treated in the fixed Hilbert space $\mathfrak{H}$ by regarding the von Neumann algebra $\mathfrak{M}$ on $\mathfrak{H}$ as the algebra of observables.[6] Thus, we replace notions such as the dynamics α_t and the KMS state ω_β formulated in the general context of C^*-algebra $\mathfrak{A}$ with those,

$$\hat{\alpha}_t(\cdot) = U_t(\cdot)\,U_t^* = e^{itH_\beta}(\cdot)\,e^{-itH_\beta}$$

$\hat{\omega}_\beta(\cdot) = \langle \Omega, \cdot\, \Omega \rangle$, etc., extended to the von Neumann algebra $\mathfrak{M}$. With this understanding, we can neglect the distinctions between α_t and $\hat{\alpha}_t$, between ω_β and $\hat{\omega}_\beta$, and between A $(\in \mathfrak{A})$ and $\pi(A)$ $(\in \mathfrak{M})$. Therefore, the caret and π are omitted henceforth.

Now, the perturbation of the dynamics α_t by the external force (1.1) with each A_i belonging to $\mathfrak{M}$ can be formulated by the differential equation governing the time-inhomogeneous dynamics $\alpha_{s\rightarrow t}$,

$$\frac{d}{dt}\alpha_{s\rightarrow t}(B) = \alpha_{s\rightarrow t}(i[H_\beta - \mathbf{A}\cdot\mathbf{X}(t), B]) \tag{3.16}$$

where B should belong to the domain $D(\delta) \subset \mathfrak{M}$ of the derivation

$$\delta \equiv \frac{d}{d}\alpha_t\Big|_{t=0} = i[H_\beta, \cdot\,]$$

[6] It is important to note that the von Neumann algebra $\mathfrak{M}$ of observables is a *proper subalgebra* of the algebra $B(\mathfrak{H})$ of all bounded operators in $\mathfrak{H}$, as is easily seen by observing that, if every operator in $B(\mathfrak{H})$ were an observable of the system, ω_β should necessarily be a *pure* state, in contradiction to the mixture character of Gibbs states.

As long as each $X_i(t)$ is a bounded continuous function on $\mathbb{R}$, the solution $\alpha_{s\to t}$ of (3.16) with the initial condition $\alpha_{s\to t=s}=\mathrm{Id}_{\mathfrak{M}}$ is uniquely determined and is given by

$$\alpha_{s\to t}(B)=\alpha_{-s}[U(t,s)^*\,\alpha_t(B)\,U(t,s)] \tag{3.17}$$

with

$$U(t,s)\equiv\mathbb{1}+\sum_{n=1}^{\infty}i^n\int_s^t dt_1\cdots\int_s^{t_{n-1}}dt_n\alpha_{t_1}(\mathbf{A})\cdot\mathbf{X}(t_1)\cdots\alpha_{t_n}(\mathbf{A})\cdot\mathbf{X}(t_n)$$
$$\left(\equiv T\exp\left[i\int_s^t d\tau\,\alpha_\tau(\mathbf{A})\cdot\mathbf{X}(\tau)\right]\right) \tag{3.18}$$

The series on the right-hand side of (3.18) converges uniformly in every finite time interval of s and t with respect to the uniform operator topology of $\mathfrak{M}$, and hence also to the strong and weak operator topologies. Thus, we have

$$U(t,s)\in\mathfrak{M} \tag{3.19}$$

Aside from the difference between H in Section 2 and H_β here (in the sense of footnote 5), the results (3.17) and (3.18) become formally identical to those in Section 2 by the following identification:

$$\alpha_{s\to t}(A)=\mathscr{U}(t,s)^*\,A\mathscr{U}(t,s) \tag{3.20}$$

$$\mathscr{U}(t,s)\equiv e^{-itH_\beta}U(t,s)\,e^{isH_\beta} \tag{3.21}$$

Thus, the chain rule for $\mathscr{U}(t,s)$,

$$\mathscr{U}(t,s)\,\mathscr{U}(s,r)=\mathscr{U}(t,r);\qquad \mathscr{U}(t,s)^{-1}=\mathscr{U}(s,t) \tag{3.21}$$

and the one [Eq. (1.7)] for $\alpha_{s\to t}$ follows from that for $U(t,s)$. Although

$$\alpha_t(\mathfrak{M})=e^{itH_\beta}\mathfrak{M}e^{-itH_\beta}=\Delta^{-it/\beta}\mathfrak{M}\Delta^{it/\beta}=\mathfrak{M}$$

and also

$$\alpha_{s\to t}(\mathfrak{M})=\mathscr{U}(t,s)^*\,\mathfrak{M}\mathscr{U}(t,s)=\mathfrak{M}$$

the unitary operator $\exp(itH_\beta)$ itself does not belong to $\mathfrak{M}$, nor does $\mathscr{U}(t,s)$. In any case, the time evolution of the state φ_t starting from $\varphi_{t_0}=\omega_\beta$ can now be given by

$$\begin{aligned}\varphi_t&=\varphi_{t_0}\circ\alpha_{t_0\to t}\\&=\omega_\beta(\alpha_{t_0}^{-1}(U(t,t_0)^*\,\alpha_t(\cdot)\,U(t,t_0)))\\&=\langle e^{-itH_\beta}U(t,t_0)\,\Omega,(\cdots)\,e^{-itH_\beta}U(t,t_0)\,\Omega\rangle\\&=\langle\Phi_t,(\cdots)\,\Phi_t\rangle\end{aligned} \tag{3.22}$$

with

$$\Phi_t \equiv e^{-itH_\beta} U(t, t_0)\, \Omega = \alpha_{-t}(U(t, t_0))\, \Omega \tag{3.23}$$

To get the expression for the relative entropy $S(\varphi_t \mid \omega_\beta)$ corresponding to (2.13), its definition (1.3) should be extended into a form independent of density matrices. Such a generalization has been achieved by Araki[6] in the context of von Neumann algebras by virtue of the notion of relative modular operator: If Ψ and Φ are both cyclic and separating vectors belonging to *one and the same natural positive cone*,[12,13],7 then the relative entropy $S(\psi \mid \varphi)$ between the two states $\psi(\cdot) \equiv \langle \Psi, \cdot\, \Psi \rangle$ and $\varphi(\cdot) \equiv \langle \Phi, \cdot\, \Phi \rangle$ is given by

$$S(\psi \mid \varphi) = \langle \Psi, \log \Delta_{\Psi,\Phi} \Psi \rangle \geqslant 0 \tag{3.24}$$

which is always nonnegative and vanishes if and only if $\psi = \varphi$. The relative modular operator $\Delta_{\Psi,\Phi}$ is defined, similarly to the definition (3.7)–(3.9) of the modular operator Δ, through the polar decomposition of $S_{\Psi,\Phi}$ and $F_{\Psi,\Phi}$ defined by

$$S_{\Psi,\Phi} x\Phi = x^* \Psi \qquad (x \in \mathfrak{M}) \tag{3.25}$$

$$F_{\Psi,\Phi} x'\Phi = x'^* \Psi \qquad (x' \in \mathfrak{M}') \tag{3.26}$$

$$\Delta_{\Psi,\Phi} \equiv S^*_{\Psi,\Phi} \bar{S}_{\Psi,\Phi} \qquad [= (F^*_{\Phi,\Psi} \bar{F}_{\Phi,\Psi})^{-1}] \tag{3.27}$$

$$\bar{S}_{\Psi,\Phi} = J_{\Psi,\Phi} \Delta^{1/2}_{\Psi,\Phi} \qquad (= F^*_{\Psi,\Phi}) \tag{3.28}$$

The definitions of S, F, J, and Δ in (3.7)–(3.9) are special cases of the above ones, $\Delta_{\Psi,\Psi} \equiv \Delta_\Psi$ and $J_{\Psi,\Psi} \equiv J_\Psi$, etc., for $\Psi = \Phi$: $\Delta = \Delta_\Omega$, $J = J_\Omega$, etc. Thus, what is needed now is the expression for $\Delta_{\Phi'_t,\Omega}$ in terms of the external force $\mathbf{X}$, where Φ'_t is the representative vector of the state φ_t determined uniquely by the requirement that it should belong to the same natural positive cone

$$\mathscr{P}_\Omega \equiv \overline{\{xJ_\Omega xJ_\Omega \Omega;\, x \in \mathfrak{M}\}}$$

that Ω does. In view of (3.23) with $\alpha_{-t}(U(t, t_0)) \equiv u \in \mathfrak{M}$ due to (3.19) and of $j_\Omega(u) \equiv J_\Omega u J_\Omega \in \mathfrak{M}'$, the vector Φ'_t is given by

$$\Phi'_t \equiv J_\Omega u J_\Omega \Phi_t = u j_\Omega(u)\, \Omega \tag{3.29}$$

7 A natural positive cone $\mathscr{P} = \mathscr{P}_\Omega$ associated with Ω is defined by $\mathscr{P} \equiv \overline{\{xJxJ\Omega;\, x \in \mathfrak{M}\}} = \overline{\Delta^{1/4}\mathfrak{M}_+\Omega}$ and satisfies the following properties: (1) $\Delta^{it}\mathscr{P} = \mathscr{P}$ for $\forall t \in \mathbf{R}$, (2) $f(\log \Delta)\, \mathscr{P} \subset \mathscr{P}$ for $\forall f$: positive-definite function, (3) $J\xi = \xi$ for $\forall \xi \in \mathscr{P}$, (4) $xJxJ\mathscr{P} \subset \mathscr{P}$ for $\forall x \in \mathscr{P}$, (5) $\langle \xi, \eta \rangle \geqslant 0$ for $\forall \xi \in \mathscr{P} \leftrightarrow \eta \in \mathscr{P}$, (6) for any *normal* (i.e., σ-weakly continuous) state φ of $\mathfrak{M}$, there exists a unique $\xi \in \mathscr{P}$ satisfying $\varphi = \langle \xi, \cdot\, \xi \rangle$. If $\xi \in \mathfrak{H}$ is another cyclic and separating vector, the definitions of J, Δ, and $\mathscr{P}$ can be repeated with Ω replaced by ξ, the results of which are denoted by J_ξ, Δ_ξ, and $\mathscr{P}_\xi$. Then, we have (7) $\xi \in \mathscr{P}_\Omega \leftrightarrow \mathscr{P}_\xi = \mathscr{P}_\Omega$ and (8) for any cyclic and separating vectors ξ and η, there exist unitary elements $U_{\xi,\eta} \in \mathfrak{M}$ and $U'_{\xi,\eta} \in \mathfrak{M}'$ such that $\mathscr{P}_\xi = U_{\xi,\eta} \mathscr{P}_\eta = U'_{\xi,\eta} \mathscr{P}_\eta$.

Then the relative modular operator $\Delta_{\Phi'_t,\Omega}$ is determined as follows. The definition (3.25) for $S_{\Phi'_t,\Omega}$ combined with (3.29) yields

$$\begin{aligned} S_{\Phi'_t,\Omega} x\Omega &= x^*\Phi'_t = x^* u j_\Omega(u)\,\Omega = j_\Omega(u)(u^*x)^*\,\Omega \\ &= j_\Omega(u)\,S_\Omega u^* x\Omega = [j_\Omega(u)\,S_\Omega u^*]\,x\Omega \end{aligned} \tag{3.30}$$

for any $x \in \mathfrak{M}$. Due to the cyclicity of Ω for $\mathfrak{M}$, this implies

$$\bar{S}_{\Phi'_t,\Omega} = j_\Omega(u)\,\bar{S}_\Omega u^* \tag{3.31}$$

and hence (3.27) gives

$$\Delta_{\Phi'_t,\Omega} = u\Delta_\Omega u^* = e^{-\beta u H_\beta u^*} \tag{3.32}$$

Thus, we obtain

$$\begin{aligned} S(\varphi_t \,|\, \omega_\beta) &= \langle \Phi'_t,\, -\beta u H_\beta u^* \Phi'_t \rangle \\ &= -\beta \langle u j_\Omega(u)\,\Omega,\, u H_\beta u^* u j_\Omega(u)\,\Omega \rangle \\ &= -\beta \langle \Omega,\, J_\Omega u^* J_\Omega H_\beta J_\Omega u J_\Omega \Omega \rangle \\ &= \beta \langle \Omega,\, u^* H_\beta u\Omega \rangle \\ &= \beta \langle \Phi_t,\, H_\beta \Phi_t \rangle \end{aligned} \tag{3.33}$$

Subtracting $\beta\langle \Omega,\ H_\beta \Omega\rangle = 0$ from the above, we arrive at the result generalizing (2.13):

$$\begin{aligned} S(\varphi_t \,|\, \omega_\beta) &= \beta(\langle \Omega,\, u^* H_\beta u\Omega \rangle - \langle \Omega,\, H_\beta \Omega \rangle) \\ &= \beta \langle \Omega,\, U(\tau, t_0)^*\, e^{i\tau H_\beta} H_\beta e^{-i\tau H_\beta} U(\tau, t_0)\,\Omega \rangle \big|_{\tau = t_0}^{\tau = t} \\ &= \beta \int_{t_0}^{t} d\tau \frac{d}{d\tau} \langle \Omega,\, U(\tau, t_0)^*\, H_\beta U(\tau, t_0)\,\Omega \rangle \\ &= \beta \int_{t_0}^{t} d\tau \langle \Omega,\, U(\tau, t_0)^*\, [iH_\beta, \alpha_\tau(\mathbf{A}) \cdot \mathbf{X}(\tau)]\, U(\tau, t_0)\,\Omega \rangle \\ &= \beta \int_{t_0}^{t} d\tau \langle \Omega,\, \alpha_{-t_0}(U(\tau, t_0)^*\, \alpha_\tau(\mathbf{J}_A)\, U(\tau, t_0))\,\Omega \rangle \cdot \mathbf{X}(\tau) \\ &= \beta \int_{t_0}^{t} d\tau\, \varphi_\tau(\mathbf{J}_A) \cdot \mathbf{X}(\tau) \end{aligned} \tag{3.34}$$

$$P(t) = \frac{d}{dt} S(\varphi_t \,|\, \omega_\beta) = \beta \varphi_t(\mathbf{J}_A) \cdot \mathbf{X}(t) \tag{3.35}$$

which is valid if each A_i belongs to $\mathrm{Dom}([iH_\beta, \cdot\,])$.

Finally, it may be instructive to note the relation among the "operator-valued free energy" $F(t)$, $\Delta_{\Phi'_t,\Omega}$ $(=\Delta_{\Phi_t,\Omega})$, and the modular operator $\Delta_{\Phi'_t}$ $[=j_\Omega(u^*)\,\Delta_{\Phi_t} j_\Omega(u)]$ associated with the state φ_t. According to Araki,[6] a state ψ^h obtained through a perturbation of a state ψ by a *relative Hamiltonian* $h \in \mathfrak{M}$ is defined to be the state whose *Connes cocycle* relative to ψ is given by

$$\begin{aligned}(D\psi^h\colon D\psi)_t &\equiv \Delta^{it}_{\Psi(h),\Psi}\Delta^{-it}_{\Psi}\\ &= \sum_{n=0}^{\infty} i^n \int_0^t dt_1 \cdots \int_0^{t_{n-1}} dt_n\, \sigma^{\psi}_{t_n}(h)\cdots\sigma^{\psi}_{t_1}(h)\\ &= \bar{T}\exp\left[i\int_0^t ds\, \sigma^{\psi}_s(h)\right]\\ &= \exp[it(\log\Delta_\Psi + h)]\exp(-it\log\Delta_\Psi)\end{aligned} \tag{3.36}$$

Here, $\mathbf{\Psi}$ and $\mathbf{\Psi}(h)$ are, respectively, representative vectors of ψ and ψ^h. This implies that the relative modular operator $\Delta_{\Psi(h),\Psi}$ is given by

$$\log\Delta_{\Psi(h),\Psi} = \log\Delta_\Psi + h \tag{3.37}$$

and according also to Refs. 12 and 13, that the cyclic and separating vector $\mathbf{\Psi}(h)$ belonging to the natural positive cone $\mathscr{P}_\Psi$ of $\mathbf{\Psi}$ is uniquely determined as

$$\mathbf{\Psi}(h) = \Delta^{1/2}_{\Psi(h),\Psi}\mathbf{\Psi} = \{\exp[(\log\Delta_\Psi + h)/2]\}\,\mathbf{\Psi} \tag{3.38}$$

The modular operator $\Delta_{\Psi(h)}$ associated with this vector is given by

$$\begin{aligned}\log\Delta_{\Psi(h)} &= \log\Delta_\Psi + h - J_\Psi h J_\Psi\\ &= \log\Delta_{\Psi(h),\Psi} - J_\Psi h J_\Psi\end{aligned} \tag{3.39}$$

The relative entropy $S(\psi^h\,|\,\psi)$ is now given by

$$S(\psi^h\,|\,\psi) = \psi^h(h) \tag{3.40}$$

Applying these results to our case with the identifications

$$\psi = \omega_\beta, \qquad \mathbf{\Psi} = \Omega; \qquad \psi^h = \varphi_t, \qquad \mathbf{\Psi}(h) = \Phi'_t \tag{3.41}$$

we get

$$\begin{aligned}h &= \log\Delta_{\Phi'_t,\Omega} - \log\Delta_\Omega\\ &= -\beta\alpha_{-t}(U(t,t_0))\,H_\beta\,\alpha_{-t}(U(t,t_0))^* + \beta H_\beta\\ &= \beta e^{-itH_\beta}U(t,t_0)[U(t,t_0)^*\,H_\beta U(t,t_0) - H_\beta]\,U(t,t_0)^*\,e^{itH_\beta}\\ &= \beta\alpha^{-1}_{t_0\to t}\left\{\int_{t_0}^t d\tau\,\alpha_{t_0\to\tau}([iH_\beta, \mathbf{A}\cdot\mathbf{X}(\tau)])\right\}\end{aligned} \tag{3.42}$$

If the A_i belong to Dom$[iH_\beta, \cdot]$ (and the X_i are L^1-functions), h belongs to $\mathfrak{M}$. By comparing (3.42) with (2.12), we can identify the relative Hamiltonian h in our case with the ⟦difference of⟧ operator-valued free energy $F(t)$ ⟦and the c-number equilibrium free energy F_β, if it is well-defined even for a general infinite system⟧ multiplied by β:

$$h = \beta F(t) \; [\![-\beta F_\beta]\!] \tag{3.43}$$

$$\Delta_{\Phi'_t, \Omega} = \exp\{-\beta(H_\beta - F(t) \; [\![+ F_\beta]\!])\} \tag{3.44}$$

The modular operator $\Delta_{\Phi'_t}$ associated with Φ'_t is given by

$$\Delta_{\Phi'_t} = \exp\{-\beta[H_\beta - F(t) + J_\Omega F(t) J_\Omega]\} \tag{3.45}$$

Substituting (3.42) into (3.40), we obtain the same expression for the relative entropy $S(\varphi_t | \omega_\beta)$ as (3.34), as should be the case.

4. ALMOST PERIODIC PERTURBATIONS

An almost periodic (a.p.) function f on the time axis $\mathbb{R}$ is a continuous function admitting a uniformly convergent series expansion

$$f(t) = \lim_{N \to \infty} \sum_{n=1}^{N} a_n e^{i\omega_n t} \tag{4.1}$$

in terms of a countable set of frequencies $\{\omega_n\}_{n=1}^{\infty}$. The set AP of a.p. functions is a commutative C^*-algebra such that the following properties are satisfied:

(i) AP is closed under the algebraic operations of addition $(+)$, multiplication $(\cdot)$, and complex conjugation $(*)$, defined as usual by

$$(f_1 + f_2)(t) = f_1(t) + f_2(t), \qquad (f_1 \cdot f_2)(t) = f_1(t) \cdot f_2(t)$$
$$f^*(t) = \overline{f(t)}$$

(ii) AP is complete with respect to its uniform topology, i.e., the limit of a uniformly convergent sequence of a.p. functions is also a.p.

(iii) AP is invariant under the translations and the inversion in $\mathbb{R}$ defined by $f_s(t) \equiv f(t-s)$, $\check{f}(t) \equiv f(-t)$.

(iv) For $\forall f \in AP$, the mean value,

$$\bar{f} \equiv \lim_{T \to \infty} \frac{1}{T} \int_0^T f(t)\, dt$$

exists, and $\bar{f} = 0$ for $f \geqslant 0$ implies $f \equiv 0$.

Currently the interest in almost periodic functions has revived, especially in connection with the problem of disordered states.[14] Here, we use this notion to control the fluctuating behavior of the entropy production driven by that of the external force. The stationary-state aspects should emerge from this fluctuation through the time average and/or limiting procedure. Now, we denote by $P(t, t_0)$ the entropy production defined as the time derivative of the relative entropy $S(\varphi_t | \varphi_{t_0} = \omega_\beta)$, namely,

$$P(t, t_0) = \beta\varphi_t(\mathbf{J}_A) \cdot \mathbf{X}(t) = \beta\omega_\beta(\alpha_{t_0 \to t}(\mathbf{J}_A)) \cdot \mathbf{X}(t) \tag{4.2}$$

Instead of taking directly the limit $t_0 \to -\infty$ of *infinite past*, we consider the averages of the function $P_{t_0}(t) \equiv P(t + t_0, t_0)$, first over the initial time t_0,[8] and then over the final time t, through which the stationarity as well as the positivity of the entropy production is attained. The precise results can be stated as follows:

I. When the external force $\mathbf{X}(t)$ is a.p., the function $P_{t_0}(t)$ is also a.p. with respect to t_0, as a consequence of which its average over t_0 exists uniquely:

$$\bar{P}(t) \equiv \lim_{T_0 \to \infty} \frac{1}{T_0} \int_{-T_0}^{0} dt_0\, P(t + t_0, t_0) \tag{4.3}$$

II. Although the long-time average of $\bar{P}(t)$ is in general not guaranteed to exist uniquely, the uniform boundedness of $P(t, t_0)$

$$|P(t, t_0)| \leqslant \sum_i \|J_A^i\| \ \|X^i\|_\infty \tag{4.4}$$

assures the existence of a sequence $(T_n)_{n=1}^{\infty}$ tending to ∞, $T_n \to \infty$, such that the sequence

$$\left[\frac{1}{T_n} \int_0^{T_n} dt\, \bar{P}(t)\right]_{n=1}^{\infty}$$

converges to a limiting value $\bar{P}$, which is *nonnegative*:

$$\begin{aligned} \bar{P} &= \lim_{n \to \infty} \frac{1}{T_n} \int_0^{T_n} dt\, \bar{P}(t) \\ &= \lim_{n \to \infty} \frac{1}{T_n} \lim_{T_0 \to \infty} \frac{1}{T_0} \int_{-T_0}^{0} dt_0\, S(\varphi_{T_n + t_0} | \varphi_{t_0} = \omega_\beta) \geqslant 0 \end{aligned} \tag{4.5}$$

[8] If the limit at $t_0 \to -\infty$ exists, then the initial-time average $\lim_{T_0 \to \infty} (1/T_0) \int_{-T_0}^{0} dt$ also exists, and is equal to the former, but the converse is not true. The authors owe to Prof. J. Bellissard the precise formulation of this idea to replace the infinite-past limit by the initial-time average. The problem of controlling the fial-time average is under investigation in collaboration with him.

Each such limit $\bar{P}$ represents the nonnegative entropy production in one of the (possibly many) stationary states characterizing a dissipation of the system.

Now, Bochner's theorem[9] to characterize the a.p. function is our basic tool for establishing the above result, and it can be stated as follows: A bounded, continuous function $f(t)$ defined on $\mathbb{R}$ is a.p. if and only if the family $\mathscr{F} \equiv \{f_\lambda; f_\lambda(t) \equiv f(t-\lambda), \lambda \in \mathbb{R}\}$ of translates of f is *precompact* in the uniform topology defined by the sup-norm $\|f\|_\infty \equiv \sup_{t \in \mathbb{R}} |f(t)|$, namely, any sequence $\{f_{\lambda_n}\}_n \subset \mathscr{F}$ contains a convergent (i.e., Cauchy) subsequence with respect to $\|\cdot\|_\infty$. Denoting the entropy production by $P(t, t_0; \mathbf{X})$ to make explicit its functional dependence on the force $\mathbf{X}(t)$, we use this theorem to deduce the almost periodicity of the function $t_0 \mapsto P(t+t_0, t_0; \mathbf{X})$ in two steps:

1. *Covariance property of* $P(t, t_0; \mathbf{X})$: The effect of the simultaneous time shift in the initial and final times is absorbed in the time shift $\mathbf{X}(t) \to \mathbf{X}_\lambda(t) = \mathbf{X}(t-\lambda)$ of the external force $\mathbf{X}$:

$$P(t-\lambda, t_0-\lambda; \mathbf{X}) = P(t, t_0; \mathbf{X}_\lambda) \tag{4.6}$$

2. *Uniform continuity of* $P(t, t_0; \mathbf{X})$ in $\mathbf{X}$ with respect to the uniform topology [for each fixed (t, t_0)].

It is then easy to verify our claim, almost periodicity of the function $t_0 \mapsto P(t+t_0, t_0; \mathbf{X})$ with t fixed, in view of 1 and 2, together with the following two general results:

(a) $\lambda \mapsto \mathbf{X}_\lambda$ is continuous in the uniform topology if and only if $\mathbf{X}$ is uniformly continuous, which is the case for an almost periodic $\mathbf{X}$.

(b) A uniformly continuous mapping maps a precompact set into another precompact set.[(16)]

Thus, what remains now is to prove statements 1 and 2.

Step 1. From Section 3, we obtain

$$\alpha_{t_0 \to t}(\mathbf{J}_A) = \alpha_{-t_0}(U(t, t_0)^* \, \alpha_t(\mathbf{J}_A) \, U(t, t_0)) \tag{4.7}$$

with

$$U(t, t_0) = T \exp\left[i \int_{t_0}^{t} d\tau \, \alpha_\tau(\mathbf{A}) \cdot \mathbf{X}(\tau) \right] \tag{4.8}$$

[9] For a brief survey see, e.g., Appendix 1 of Ref. 15.

This satisfies

$$U(t-\lambda, t_0-\lambda) = T\exp\left[i\int_{t_0-\lambda}^{t-\lambda} d\tau\, \alpha_\tau(\mathbf{A})\cdot\mathbf{X}(\tau)\right]$$

$$= T\exp\left[i\int_{t_0}^{t} d\tau\, \alpha_{\tau-\lambda}(\mathbf{A})\cdot\mathbf{X}(\tau-\lambda)\right]$$

$$= \alpha_{-\lambda}\left\{T\exp\left[i\int_{t_0}^{t} d\tau\, \alpha_\tau(\mathbf{A})\cdot\mathbf{X}_\lambda(\tau)\right]\right\} \tag{4.9}$$

Thus, $U(t, t_0)$ with its $\mathbf{X}$ dependence made explicit as $U(t, t_0; \mathbf{X})$ satisfies the following covariance condition:

$$U(t-\lambda, t_0-\lambda; \mathbf{X}) = \alpha_{-\lambda}(U(t, t_0; \mathbf{X}_\lambda)) \tag{4.10}$$

Therefore, we obtain

$$\alpha_{t_0-\lambda\to t-\lambda}(\mathbf{J}_A) = \alpha_{-t_0}(U(t, t_0; \mathbf{X}_\lambda)^*\, \alpha_t(\mathbf{J}_A)\, U(t, t_0; \mathbf{X}_\lambda)) \tag{4.11}$$

and hence [(4.6)]

$$P(t-\lambda, t_0-\lambda; \mathbf{X}) = P(t, t_0; \mathbf{X}_\lambda)$$

Step 2. Since the unitary operator $U(t, t_0; \mathbf{X}) = U(t, t_0)$ defined by (4.8) satisfies the equation

$$U(t, t_0; \mathbf{X}) = \mathbb{1} + \int_{t_0}^{t} d\tau\, i\alpha_\tau(\mathbf{A})\cdot\mathbf{X}(\tau)\, U(\tau, t_0; \mathbf{X}) \tag{4.12}$$

we obtain

$$\|U(t, t_0; \mathbf{X}) - U(t, t_0; \mathbf{Y})\|$$

$$= \left\|\int_{t_0}^{t} d\tau\, \alpha_\tau(\mathbf{A})\cdot\{\mathbf{X}(\tau)\, U(\tau, t_0; \mathbf{X}) - \mathbf{Y}(\tau)\, U(\tau, t_0; \mathbf{Y})\}\right\|$$

$$\leqslant \int_{t_0}^{t} d\tau \left\{L\, \|U(\tau, t_0; \mathbf{X}) - U(\tau, t_0; \mathbf{Y})\| + M\sum_i \|X^i - Y^i\|_\infty\right\} \tag{4.13}$$

where $M \equiv \max_i \|A_i\|$ and $L \equiv M\sum_i \|X_i\|_\infty$. Applying Gronwall's lemma "solving" the inequality

$$0 \leqslant p(t) \leqslant \int_{t_0}^{t} d\tau [Lp(\tau) + q(\tau)] \qquad (\forall t \geqslant t_0)$$

with $L \geqslant 0$ and $q(t) \geqslant 0$ in favor of $p(t)$ as

$$p(t) \leqslant \int_{t_0}^{t} d\tau\, q(\tau)\, e^{L(t-\tau)}$$

we obtain from (4.13)

$$\|U(t, t_0; \mathbf{X}) - U(t, t_0; \mathbf{Y})\| \leqslant \frac{M}{L}(e^{L|t-t_0|} - 1) \sum_i \|X^i - Y^i\|_\infty \quad (4.14)$$

This asserts the uniform continuity of $U(t, t_0; \mathbf{X})$ in $\mathbf{X}$ with respect to $\|\cdot\|_\infty$. The desired conclusion, uniform continuity of $P(t, t_0; \mathbf{X})$ in $\mathbf{X}$, follows from this result together with the formula

$$P(t, t_0; \mathbf{X}) = \beta \langle U(t, t_0; \mathbf{X})\,\Omega, \alpha_t(\mathbf{J}_A)\, U(t, t_0; \mathbf{X})\,\Omega \rangle \cdot \mathbf{X}(t) \quad (4.15)$$

5. RESULTS AND DISCUSSION

We have formulated the mean entropy production $\bar{P}$ as the time average of $P(t+t_0, t_0)$ over the initial and final times t_0 and t:

$$\begin{aligned}\bar{P} &= \lim_{n\to\infty} \frac{1}{T_n} \int_0^{T_n} dt \lim_{T_0\to\infty} \frac{1}{T_0} \int_{-T_0}^{0} dt_0\, P(t+t_0, t_0) \\ &= \lim_{n\to\infty} \frac{1}{T_n} \lim_{T_0\to\infty} \frac{1}{T_0} \int_{-T_0}^{0} dt_0\, S(\varphi_{T_n+t_0} \mid \varphi_{t_0} = \omega_\beta) \geqslant 0 \end{aligned} \quad (5.1)$$

The positivity of this expression stems from the fact that $\bar{P}$ is essentially the relative entropy per unit time of the stationary state realized through the averaging over the initial time. The limit of the second average over t, however, may not be unique in general, reflecting the possible occurrence of multiple stationary states in the nonlinear response. Furthermore, one should be able to distinguish between a true nonequilibrium stationary state with $\bar{P} > 0$ and a *quasi*equilibrium state with $\bar{P} = 0$, different from the starting KMS state. These problems will be important issues for future investigation. Assuming that a limit of such an average exists, we get another expression for $\bar{P}$,

$$\bar{P} = \beta \sum_{n=0}^{\infty} \hat{\mathbf{J}}_A^*(\omega_n) \cdot \hat{\mathbf{X}}(\omega_n) \quad (5.2)$$

where $\hat{\mathbf{X}}(\omega_n)$ is the Fourier coefficient of the almost periodic $\mathbf{X}(t)$,

$$\mathbf{X}(t) = \sum_{n=0}^{\infty} \hat{\mathbf{X}}(\omega_n)\, e^{i\omega_n t} \quad (5.3)$$

and $\hat{\mathbf{J}}_A(\omega_n)$ is defined by

$$\begin{aligned}\hat{\mathbf{J}}_A(\omega_n) = \lim_{T\to\infty} \lim_{T_0\to\infty} \frac{1}{T} \int_0^T dt \frac{1}{T_0} \int_{-T_0}^{0} dt_0 \\ \times e^{-i\omega_n(t+t_0)} \omega_\beta(\alpha_{t_0 \to t+t_0}(\mathbf{J}_A)) \end{aligned} \quad (5.4)$$

This result retains a full nonlinearity in the force X and still allows a physically meaningful interpretation of "entropy production" familiar in the context of thermodynamics.[17]

A formal structure of the relationship between *dissipation* and *fluctuation* in a quantum dynamical system, well formulated at least in the linear regime,[1–3] can be seen to hold in the present framework as follows. The rate at which the mechanical work is done on the system by the external force is equal to the rate of energy change in the system,[10]

$$\frac{d}{dt}\langle \Phi_t, H_\beta \Phi_t \rangle = \mathbf{X}(t) \cdot \frac{d}{dt}\varphi_t(\mathbf{A}) \tag{5.5}$$

which directly follows from (3.33)–(3.35) and from

$$\begin{aligned} \mathbf{X}(t) \cdot \frac{d}{dt}\varphi_t(\mathbf{A}) &= i\varphi_t\left(\left[H_\beta - \sum_k A_k X_k, \sum_j A_j X_j\right]\right) \\ &= \varphi_t(\mathbf{J}_A) \cdot \mathbf{X}(t) \end{aligned} \tag{5.6}$$

Noting that the effect of the external force is to reduce the entropy of the system (to produce more order in the system), we can interpret

$$-\beta \frac{d}{dt}\langle \Phi_t, H_\beta \Phi_t \rangle = -\beta \frac{d}{dt}\langle \Phi_t, (\text{system Hamiltonian})\, \Phi_t \rangle$$

[10] As noted in footnote 5, H_β cannot be directly identified with the energy of the system, but $\langle \Phi_t, H_\beta \Phi_t \rangle$ and its time derivative $(d/dt)\langle \Phi_t, H_\beta \Phi_t \rangle$ can be interpreted properly as the energy gain of the system due to the work done by the external force and the rate of the energy change in the system, respectively. This can be easily seen in the case of the density-matrix formalism with $H_\beta = H - JHJ$ valid:

$$\begin{aligned} &\langle \Phi_t, H_\beta \Phi_t \rangle \\ &\quad = \langle \Omega, \alpha_{-t}(U(t, t_0))^*(H - JHJ)\, \alpha_{-t}(U(t, t_0))\, \Omega \rangle \\ &\quad = \langle \Phi_t, H\Phi_t \rangle - \langle \Omega, H\Omega \rangle = \Delta E; \end{aligned}$$

$$\frac{d}{dt}\langle \Phi_t, H_\beta \Phi_t \rangle = \frac{d}{dt}\langle \Phi_t, H\Phi_t \rangle$$

In this context, it is interesting to note that the *positivity* of this energy gain, $\Delta E = (1/\beta)\, S(\varphi_t | \omega_\beta) \geqslant 0$, can also be understood as the *passivity*[18] of the KMS state ω_β, because (3.34) can be rewritten as

$$\begin{aligned} \langle \Phi_t, H_\beta \Phi_t \rangle &= (1/\beta)\, S(\varphi_t | \omega_\beta) \\ &= \langle \Omega, (U(t, t_0)^*\, H_\beta U(t, t_0) - H_\beta)\, \Omega \rangle \\ &= \omega_\beta(i\delta(U(t, t_0)^*)\, U(t, t_0)) \geqslant 0 \end{aligned}$$

with $U(t, t_0) \in \mathscr{U}(\mathfrak{M})$ and because the KMS condition is equivalent to the inequality[5]

$$-i\beta\omega(A^*\delta(A)) \geqslant \omega(A^*A) \log[\omega(A^*A)/\omega(AA^*)]$$

(= the rate of energy dissipation divided by kT)[10] as an entropy change due to the contact of the system with the environment and we denote it as $[(d/dt)\,S]_{\text{ext}}$ (extrinsic entropy production),

$$\left(\frac{d}{dt}S\right)_{\text{ext}} \equiv -\beta \frac{d}{dt}\langle \Phi_t, H_\beta \Phi_t \rangle \tag{5.7}$$

On the other hand, the intrinsic entropy production $[(d/dt)\,S]_{\text{int}}$, intrinsic to the system, can be identified with the relative entropy production so far discussed:

$$\left(\frac{d}{dt}S\right)_{\text{int}} \equiv \frac{d}{dt}S(\varphi_t \,|\, \omega_\beta) \tag{5.8}$$

Then, (5.5) can be put into the form

$$\left(\frac{d}{dt}S\right)_{\text{ext}} + \left(\frac{d}{dt}S\right)_{\text{int}} = 0 \tag{5.9}$$

which is also equivalent to the equality

$$\varphi_t([iH_\beta, \mathbf{A}]) = \varphi_t([iF(t), \mathbf{A}]) \tag{5.10}$$

In the conventional formulation in terms of density matrices, the origin of this equality is easily understood as the invariance of the system entropy (-tr $\rho \log \rho$) under the unitary evolution. In the general case, it derives from the equality

$$\log \Delta_{\Phi'_t} \Phi'_t = -\beta[H_\beta - F(t) + J_\Omega F(t)\, J_\Omega]\, \Phi'_t = 0 \tag{5.11}$$

and from $J_\Omega F(t)\, J_\Omega \in \mathfrak{M}'$. In view of the expression for the operator-valued free energy $F(t)$ [(3.42) and (3.43)], the relation (5.10) can be further rewritten as

$$\begin{aligned}\langle \mathbf{J}_A \rangle(t) &\equiv \varphi_t([iH_\beta, \mathbf{A}]) \\ &= \varphi_t\left(\left[i\int_{t_0}^{t} d\tau\, \alpha_{t\to\tau}(\mathbf{J}_A)\cdot \mathbf{X}(\tau), \mathbf{A}\right]\right)\Bigg|_{t_0\to -\infty} \\ &= \lim_{\varepsilon\to +0} \int_{-\infty}^{t} d\tau\, e^{\varepsilon(\tau - t)}\omega_\beta([i\alpha_{-\infty\to\tau}(\mathbf{J}_A)\cdot \mathbf{X}(\tau), \alpha_{-\infty\to t}(\mathbf{A})])\end{aligned} \tag{5.12}$$

In the final expression here, we have conformed to the conventional scheme of the "adiabatic switching" procedure, taking the infinite-past limit $t_0 \to -\infty$ first in the presence of a convergence factor $e^{\varepsilon(\tau - t)}$ in $\mathbf{X}$, and then removing it through the limit of $\varepsilon \to +0$. Although the latter limit is difficult to control, in contrast to the initial-time averaging, this scheme makes it easy to see that (5.12) gives a generalization of the Kubo formula in Ref. 1. Namely, it reproduces, in the leading order of $\mathbf{X}$, the following well-known formula in linear response theory:

$$\begin{aligned} \langle J_A^i \rangle(t) &= \lim_{\varepsilon \to +0} \int_{-\infty}^{t} d\tau\, e^{\varepsilon(\tau - t)} \sum_j \omega_\beta([i\alpha_{\tau - t}(J_A^j), A^i])\, X^j(\tau) \\ &= \lim_{\varepsilon \to +0} \int_0^\infty d\tau\, e^{-\varepsilon\tau} \int_0^\beta d\lambda \sum_j \omega_\beta(\alpha_{\tau - i\lambda}(J_A^i)\, J_A^j)\, X^j(t - \tau) \end{aligned} \tag{5.13}$$

The dissipative aspect in this context has been exhibited in Ref. 1 as the positivity of the transport coefficient matrix, by relating the kernel functions in this integral representation (5.13) to the current–current correlation functions [see (5.15) below]. From the present standpoint of general nonlinear response theory, it should be attributed to the positivity of the mean entropy production discussed so far. By combining the latter with the equalities (5.10) and (5.12), we obtain

$$\begin{aligned} \bar{P} &= \beta\overline{\langle \mathbf{J}_A \rangle(t) \cdot \mathbf{X}(t)} \\ &= \beta\overline{(d/dt)\langle \Phi_t, H_\beta \Phi_t \rangle} \\ &= \beta\overline{\int_{-\infty}^{t} d\tau\, \omega_\beta([i\alpha_{-\infty \to \tau}(\mathbf{J}_A) \cdot \mathbf{X}(\tau), \alpha_{-\infty \to t}(\mathbf{J}_A) \cdot \mathbf{X}(t)])} \geqslant 0 \end{aligned} \tag{5.14}$$

where the bar denotes the long-time average over t. As a formula describing the general relationship between the dissipation of energy and the current fluctuations, this can properly be taken as a nonlinear generalization of the fluctuation-dissipation theorem. From this viewpoint, it would be instructive to see to what form (5.14) reduces in the linear-response regime. Substituting (5.13) into it, we obtain

$$\begin{aligned} \bar{P} &= \beta \lim_{T \to \infty} \frac{1}{T} \int_0^T dt \int_0^\infty d\tau \int_0^\beta d\lambda \\ &\quad \times \sum_{i,j} \omega_\beta(\alpha_{\tau - i\lambda}(J_A^i)\, J_A^j)\, X^j(t - \tau)\, X^i(t) \\ &= \beta \int_0^\infty d\tau \int_0^\beta d\lambda \sum_{i,j} \omega_\beta(\alpha_{\tau - i\lambda}(J_A^i)\, J_A^j) \\ &\quad \times \lim_{T \to \infty} \frac{1}{T} \int_0^T dt\, X^i(t - \tau)\, X^j(t) \\ &= \beta \sum_{n=0}^{\infty} \omega_n^{-1} \tanh \frac{\beta\omega_n}{2} \sum_{i,j} X^i(\omega_n)^*\, L_{ij}(\omega_n)\, X^j(\omega_n) \end{aligned} \tag{5.15}$$

where the sum over frequencies $\{\omega_n\}$ is taken only for those with $\omega_n \geqslant 0$ in view of the reality condition of $\mathbf{X}(t)$: $\mathbf{X}(\omega_n)^* = \mathbf{X}(-\omega_n)$. The coefficient matrix $\{L_{ij}(\omega_n)\}$ defined by

$$L_{ij}(\omega_n) \equiv \int_{-\infty}^{\infty} d\tau\, e^{i\omega_n \tau} \omega_\beta(\{\alpha_\tau(J^i), J^j\}) \\ = L_{ji}(\omega_n)^* = L_{ij}(-\omega_n)^*$$

is easily seen to be positive, and hence the expression (5.15) just agrees with the *power dissipation* (divided by kT)[1] for a linear dissipative quantum system.

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