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Derivation of the Linear and Nonlinear Non-Markov Fluctuation-Dissipation Relations of the First Kind for a Dynamical Model

R. L. Stratonovich[1]

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A dynamical system consisting of a subsystem having the variables $z = (q, p)$ and of another dynamical system (thermostat) is considered in the nonquantum case. Using a dynamical equation, it is shown that the linear and quadratic non-Markov fluctuation-dissipation relations (FDRs) of the first kind are valid in the first nonvanishing approximation in interaction constants. Applying these FDRs, one can determine the statistical properties of the fluctuations when the form of the nonlinear phenomenological equation is known. The non-Markov FDRs of the first kind are the direct generalization (to the inertial case) of the Markov FDRs that are the consequence of detailed balance.

KEY WORDS: Nonlinear systems with after-effect; fluctuation-dissipation theorem; reciprocal relations; interaction of dynamical systems; contact with thermostat.

1. INTRODUCTION

In his remarkable works[1] N. G. van Kampen first formulated the conditions of detailed balance for the Fokker–Plank equation. As is well known, detailed balance is conditioned by the time reversibility of the physical microprocesses, or, strictly speaking, by invariance of the equations describing these microprocesses with respect to the transformation $t \to -t$. Based on van Kampen's work[2] (see also ref. 3, pp. 160–165), the Onsager relations and the Markov fluctuation-dissipation theorem can be derived from detailed balance.

More than 10 years later I published work that may be regarded as a development and generalization of van Kampen's ideas to the case of the

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arbitrary (not only the Fokker–Planck) stationary Markov process of the nonlinear theory.

The concept of detailed balance also relates to the case of the arbitrary Markov process. If the basic equation is written as

$$\dot{p}(z) = \int L_{zz'} p(z')\, dz' \tag{1.1}$$

then the detailed balance takes the form

$$L_{zz'} p_0(z') = L_{\varepsilon z', \varepsilon z} p_0(z) \tag{1.2}$$

where $p_0(z)$ is the stationary probability density and $\varepsilon_\alpha = \pm 1$ are time signatures (z_α goes into εz_α when the transformation $t \to -t$ is made).

Equation (1.1) can be written in the form of the Kramers–Moyal expansion

$$\dot{p}(z) = \sum_{n=1}^{\infty} \sum_{\alpha_1, \ldots, \alpha_n} \frac{(-1)^n}{\alpha_1! \cdots \alpha_n!} \times \frac{\partial^n}{\partial z_{\alpha_1} \cdots \partial z_{\alpha_n}} [K_{\alpha_1 \cdots \alpha_n}(z)\, p(z)] \tag{1.3}$$

Here the summation over repeated subscripts is understood. It is expedient to introduce the images

$$\kappa_{\alpha_1 \cdots \alpha_n}(x) = \frac{\int K_{\alpha_1 \cdots \alpha_n}(z) \exp(\mu^{-1} z_\alpha x_\alpha)\, p_0(z)\, dz}{\int \exp(\mu^{-1} z_\alpha x_\alpha)\, p_0(z)\, dz} \tag{1.4}$$

of the functions $K_{\alpha_1 \cdots \alpha_n}$. For the moment the parameter μ is arbitrary. In addition we denote

$$l_{\alpha_1 \cdots \alpha_n, \beta_1 \cdots \beta_m} = \left[\frac{\partial^m \kappa_{\alpha_1 \cdots \alpha_n}(x)}{\partial x_{\beta_1} \cdots \partial x_{\beta_m}} \right]_{x=0} \tag{1.5}$$

In refs. 4 and 5 it has been shown that the detailed balance implies the exact linear and quadratic fluctuation-dissipation relations (FDRs)

$$\begin{aligned} l_{\beta,\alpha} &= \varepsilon_\alpha \varepsilon_\beta l_{\alpha,\beta} \\ l_{\alpha\beta} &= -\mu(l_{\alpha,\beta} + l_{\beta,\alpha}) \\ l_{\alpha\beta\gamma} &= \mu^2(1 - \varepsilon_\alpha \varepsilon_\beta \varepsilon_\gamma)(l_{\alpha,\beta\gamma} + l_{\beta,\alpha\gamma} + l_{\gamma,\alpha\beta}) \\ l_{\alpha\beta,\gamma} &= \mu(\varepsilon_\alpha \varepsilon_\beta \varepsilon_\gamma l_{\gamma,\alpha\beta} - l_{\alpha,\beta\gamma} - l_{\beta,\alpha\gamma}) \end{aligned} \tag{1.6}$$

The first of these relations is the Onsager relation and the second is the linear Markov fluctuation-dissipation theorem. The third and fourth relations are quadratic FDRs. The cubic FDRs having the form

$$\begin{aligned} c_{\alpha\beta,\gamma\delta} &= \varepsilon_\alpha \varepsilon_\beta \varepsilon_\gamma \varepsilon_\delta c_{\gamma\delta,\alpha\beta} \\ l_{\alpha\beta\mu\delta} &= \mu^3(1+\varepsilon_\alpha\varepsilon_\beta\varepsilon_\gamma\varepsilon_\delta)(c_{\alpha\beta,\gamma\delta}+c_{\alpha\gamma,\beta\delta}+c_{\beta\gamma,\alpha\delta}-l_{\alpha,\beta\gamma\delta} \\ &\quad - l_{\beta,\alpha\gamma\delta}-l_{\gamma,\alpha\beta\delta}-l_{\delta,\alpha\beta\gamma}) \\ l_{\alpha\beta\gamma,\delta} &= \mu^2(l_{\alpha,\beta\gamma\delta}+l_{\beta,\alpha\gamma\delta}+l_{\gamma,\alpha\beta\delta}+\varepsilon_\alpha\varepsilon_\beta\varepsilon_\gamma\varepsilon_\delta l_{\delta,\alpha\beta\gamma} \\ &\quad - c_{\alpha\beta,\gamma\delta}-c_{\alpha\gamma,\beta\delta}-c_{\beta\gamma,\alpha\delta}) \end{aligned} \tag{1.7}$$

where

$$c_{\alpha\beta,\gamma\delta} = \mu^{-1} l_{\alpha\beta,\gamma\delta} + l_{\alpha,\beta\gamma\delta} + l_{\beta,\alpha\gamma\delta} \tag{1.8}$$

were also derived in these works.

In many physical problems the stationary probability contains some small parameter μ as follows:

$$p_0(z) = \text{const} \cdot \exp[-\varphi(z)/\mu] \tag{1.9}$$

As an example, one can take the well-known equilibrium probability densities

$$\begin{aligned} p_0(z) &= \text{const} \cdot \exp[-F(z)/kT] \\ p_0(z) &= \text{const} \cdot \exp[S(z)/k] \\ p_0(z) &= \text{const} \cdot \exp[-E(z)/kT] \end{aligned} \tag{1.10}$$

where $F(z)$ is the free energy, $S(z)$ is the entropy, and $E(z)$ is the energy. In the fist case $\varphi(z) = F(z)$, $\mu = kT$; in the second case $\varphi(z) = -S(z)$, $\mu = k$; etc. The smallness of μ is provided by the smallness (from the macroscopic point of view) of the Boltzmann constant. Identifying the parameter μ in (1.4) with the small parameter μ in (1.9), we easily obtain that in the case (1.9), Eqs. (1.4) imply the asymptotic equations

$$\kappa_{\alpha_1 \cdots \alpha_n}(x) \approx K_{\alpha_1 \cdots \alpha_n}(z(x)) \tag{1.11}$$

where the dependence $z(x)$ is the inverse of the dependence

$$x_\alpha = \partial\varphi(z)/\partial z_\alpha \tag{1.12}$$

Thus, x_α can be interpreted as "forces" conjugate to z_α. From (1.11) it follows that

$$l_{\alpha\beta} = K_{\alpha\beta}(z(0)), \qquad l_{\alpha,\beta} = K_{\alpha,\gamma}\left(\frac{\partial z_\gamma}{\partial x_\beta}\right)_{x=0}$$
$$l_{\alpha\beta\gamma} = K_{\alpha\beta\gamma}(z(0)), \qquad l_{\alpha\beta,\gamma} = K_{\alpha\beta,\delta}\left(\frac{\partial z_\delta}{\partial x_\gamma}\right)_{x=0} \tag{1.13}$$

where

$$K_{\alpha_1\cdots\alpha_n,\beta_1\cdots\beta_m} = \left[\frac{\partial^m K_{\alpha_1\cdots\alpha_n}}{\partial z_{\beta_1}\cdots\partial z_{\beta_m}}\right]_{x=0} \tag{1.14}$$

Of course, when we use Eqs. (1.13), the FDRs (1.6) and (1.7) become asymptotic.

The Markov FDRs (1.6) and (1.7) can be generalized to the non-Markov case.[5] Suppose that the stochastic equation can contain inertia and that this equation is nonlinear, i.e.,

$$\dot{z}_\alpha(t) = -\int \Phi_{\alpha,\beta}(t,t')\,x_\beta(z(t'))\,dt' - \frac{1}{2}\iint \Phi_{\alpha,\beta\gamma}(t;t',t'')$$
$$\times\, x_\beta(z(t''))\,x_\gamma(z(t''))\,dt'\,dt'' - \cdots - \xi_\alpha(t) \tag{1.15}$$

where the dependence $x(z)$ is determined by formula (1.12) [if Eq. (1.9) is valid], and the noise $\xi_\alpha(t)$ ($\langle\xi_\alpha\rangle = 0$) is a functional of innovation noises $\zeta_\sigma(t)$ and of $x(z(t))$:

$$\xi_\alpha(t) = F_{\alpha t}[\zeta(t'), x(z(t'))] \tag{1.16}$$

It is convenient to write (1.15) in the shorter form

$$\dot{z}_1 = -\Phi_{1,2}x_2 - \tfrac{1}{2}\Phi_{1,23}x_2x_3 - \cdots - \xi_1 \tag{1.17}$$

Here the subscripts 1, 2,..., denote the pairs (α_1, t_1), (α_2, t_2),..., and the summation (and the integration with respect to time) over repeated subscripts is understood.

For the above case the linear FDRs have the form

$$\Phi_{12} \equiv \langle\xi_1\xi_2\rangle_{x\equiv0} = \mu(\Phi_{1,2} + \Phi_{2,1}) \tag{1.18a}$$

$$\Phi_{1,2}^{\text{t.c.}} = \Phi_{2,1} \tag{1.18b}$$

where t.c. denotes the time conjugation operation defined by

$$[f_{\alpha_1\cdots\alpha_n}(t_1,\dots,t_n)]^{\text{t.c.}} = \varepsilon_{\alpha_1}\cdots\varepsilon_{\alpha_n} f^*(-t_1,\dots,-t_n) \tag{1.19}$$

The quadratic non-Markov FDRs have the form

$$\begin{aligned} \Phi_{123} &= \mu^2(\Phi_{1,23} - \Phi_{1,23}^{\text{t.c.}} + \Phi_{2,13} - \Phi_{2,13}^{\text{t.c.}} + \Phi_{3,21} - \Phi_{3,21}^{\text{t.c.}}) \\ \Phi_{12,3} &= \mu(\Phi_{1,23} + \Phi_{2,13} - \Phi_{3,12}^{\text{t.c.}}) \end{aligned} \tag{1.20}$$

Here

$$\Phi_{123} = \langle \xi_1 \xi_2 \xi_3 \rangle_{x \equiv 0} \tag{1.21}$$

and $\Phi_{12,3}$ is determined by the expansion

$$\langle F_1[\zeta, x], F_2[\zeta, x] \rangle_x = \Phi_{12} + \Phi_{12,3} x_3 + \cdots \tag{1.22}$$

where $\langle a, b \rangle = \langle ab \rangle - \langle a \rangle \langle b \rangle$. In Eq. (1.22) the function $x(t)$ is assumed not as a random function, but as an argument function, i.e., it is fixed. In ref. 5, the FDRs (1.18) and (1.20) were called FDRs of the first kind. In addition, linear and nonlinear FDRs of other kinds are also valid. Here we will prove the validity of the FDRs (1.18) and (1.20) for a model in which z_α are the coordinates of a dynamical subsystem interacting with a thermostat.

2. THE DERIVATION OF THE STOCHASTIC EQUATION FOR THE COORDINATES OF THE DYNAMICAL SUBSYSTEM

We suppose that the subsystem has the Hamiltonian $H_1(q, p)$, and the thermostat has the Hamiltonian $H_2(Q, P)$. Denoting $z = (q, p)$ and $Y = (Q, P)$ and choosing the interaction Hamiltonian in the form $V = -\sum_\alpha \lambda_\alpha z_\alpha Y_{n_\alpha}$, we have the total Hamiltonian

$$H(z, Y) = H_1(z) + H_2(Y) - \sum_\alpha \lambda_\alpha z_\alpha Y_{n_\alpha} \tag{2.1}$$

Substituting (2.1) into the dynamical equation

$$\dot{z}_\alpha = -\{z_\alpha, H\} \tag{2.2}$$

we get

$$\dot{z}_\alpha = -\{z_\alpha, z_\beta\} \frac{\partial H_1}{\partial z_\beta} + \{z_\alpha, z_\beta\} \lambda_\beta Y_\beta \tag{2.3}$$

(the summation over β is understood). Here we have used the equation

$$\{z_\alpha, H_1(z)\} = \{z_\alpha, z_\beta\} \frac{\partial H_1}{\partial z_\beta} \tag{2.4}$$

whose validity can be verified by using the expansion of the Hamiltonian H_1 into its Taylor series. In Eqs. (2.3) and (2.4) the braces $\{\cdots\}$ denote the Poisson bracket that is the classical limit of the quantum Poisson bracket:

$$\{F, G\} = \lim_{\hbar \to 0} \frac{i}{\hbar} [F, G], \qquad [F, G] = FG - GF \tag{2.5}$$

By strength of (2.1), from the total Gibbs distribution $p_0(z, Y) = \text{const} \cdot \exp[-H(z, Y)/kT]$ we get, for small $\lambda = \{\lambda_\alpha\}$, the equilibrium probability density for the coordinates of the subsystem

$$p_0(z) = \text{const} \cdot \exp[-H_1(z)/kT] \tag{2.6}$$

Comparing (2.6) with (1.9), we see that in this case $\varphi(z) = H_1(z)$, $\mu = kT$, and formula (1.12) assumes the form

$$x_\alpha = \partial H_1(z)/\partial z_\alpha \tag{2.7}$$

Also denoting for brevity

$$c_{\alpha\beta} = \{z_\alpha, z_\beta\} \tag{2.8}$$

we rewrite (2.3) in the form

$$\dot{z}_\alpha = -c_{\alpha\beta} x_\beta + c_{\alpha\beta} \lambda_\beta Y_{n_\beta} \tag{2.9}$$

The coordinates of theermostat Y_{n_β} appearing in (2.9) are functionals of $x(t)$. The behavior of the thermostat with the interaction Hamiltonian $-\sum_\alpha \lambda_\alpha z_\alpha Y_{n_\alpha}$ can be identified with the behavior of the thermostat with the Hamiltonian

$$H(Y) = H_2(Y) - \sum_\alpha f_\alpha Y_{n_\alpha} \tag{2.10}$$

if we let $f_\alpha = \lambda_\alpha z_\alpha$. Let us expand Y_{n_α} into the functional Taylor series in f_α:

$$\begin{aligned} Y_{n_\alpha}(t_1) = {} & [Y_{n_\alpha}(t_1)]_{f \equiv 0} + \int [\delta Y_{n_\alpha}(t_1)/\delta f_\beta(t_2)]_{f \equiv 0} f_\beta(t_2)\, dt_2 \\ & + \frac{1}{2} \int [\delta^2 Y_{n_\alpha}(t_1)/\delta f_\beta(t_2)\, \delta f_\gamma(t_3)]_{f \equiv 0} \\ & \times f_\beta(t_2) f_\gamma(t_3)\, dt_2\, dt_3 + \cdots \end{aligned} \tag{2.11}$$

Taking into consideration the formula

$$\frac{\delta F(t)}{\delta f_\beta(t_1)} = \{F(t), Y_{n_\beta}(t_1)\}\, \eta(t - t_1) \tag{2.12}$$

$[2\eta(\tau)=1+\operatorname{sign}\tau]$, which is valid (for $\{F, Y\}=(i/\hbar)[F, Y]$) in the quantum case and also in the classical limit, we can obtain that the functional derivatives in (2.11) are expressed by the repeated Poisson brackets (or commutators)[6]

$$[\delta Y_{n_\alpha}(t_1)/\delta f_\beta(t_2)]_{f\equiv 0} \equiv v_{\alpha\beta}(t_1, t_2) = \{Y^0_{n_\alpha}(t_1), Y^0_{n_\beta}(t_2)\}\,\eta_{12}$$

$$\left[\frac{\delta^2 Y_{n_\alpha}(t_1)}{\delta f_\beta(t_2)\,\delta f_\gamma(t_3)}\right]_0 \equiv v_{\alpha\beta\gamma}(t_1, t_2, t_3)$$

$$= \{\{Y^0_{n_\alpha}(t_1), Y^0_{n_\beta}(t_2)\}, Y^0_{n_\gamma}(t_3)\}\,\eta_{123} + \{\{Y^0_{n_\alpha}(t_1), Y^0_{n_\gamma}(t_3)\}, Y^0_{n_\beta}(t_2)\}\,\eta_{132} \quad (2.13)$$

where $Y^0=[Y^0]_{f\equiv 0}$, $\eta_{12}=\eta(t_1-t_2)$, and $\eta_{123}=\eta_{12}\eta_{23}$. Considering Eqs. (2.11) and (2.13) and the equation $f_\alpha=\lambda_\alpha z_\alpha$, we reduce (2.9) to the form

$$\dot{z}_\alpha(t_1) = -c_{\alpha\beta}x_\beta + c_{\alpha\beta}\lambda_\beta Y^0_{n_\beta}(t_1) + c_{\alpha\beta}\lambda_\beta \int v_{\beta\gamma}(t_1, t_2)\,\lambda_\gamma \times z_\gamma(t_2)\,dt_2 + \frac{1}{2}c_{\alpha\beta}\lambda_\beta \int v_{\beta\gamma\delta}(t_1, t_2, t_3) \times \lambda_\gamma\lambda_\delta z_\gamma(t_2)\,z_\delta(t_3)\,dt_2\,dt_3 + \cdots \quad (2.14)$$

The thermostat variables Y^0 appearing in (2.14) fluctuate according to the law of thermal equilibrium. They can be regarded as innovation external random forces acting on the subsystem, i.e., Eq. (2.14) is the stochastic equation obtained in ref. 8.

3. PROOF OF LINEAR RELATIONS

Equation (2.14) is the specific form of Eq. (1.15). It remains to express z in terms of the forces x. In the linear approximation the terms nonlinear in x should be omitted. In addition, in the term linear in x the random function $v_{\beta\gamma}(t_1, t_2)$ should be replaced by its mean value $\langle v_{\beta\gamma}(t_1, t_2)\rangle$ since $(v-\langle v\rangle)\,z$ relates to the nonlinear approximation. Denoting $p_2=\partial/\partial t_2$, we have

$$\langle v_{\beta\gamma}(t_1, t_2)\rangle\,\lambda_\gamma z_\gamma(t_2)$$
$$= \langle v_{\beta\gamma}(t_1, t_2)\rangle\,\lambda_\gamma(1/p_2)\,\dot{z}_\gamma(t_2)$$
$$= \langle v_{\beta\gamma}(t_1, t_2)\rangle\,\lambda_\gamma(1/p_2)[-c_{\gamma\delta}x_\delta(t_2) + c_{\gamma\delta}\lambda_\delta Y^0_{n_\delta}(t_2) + \mathcal{O}(\lambda^2)] \quad (3.1)$$

[Eq. (2.14) is used]. Analogous transformations of the other terms can be performed. By virtue of (2.13) we get

$$\langle v_{\beta\gamma}(t_1, t_2)\rangle = U_{\beta\gamma}(t_1, t_2)\,\eta_{12} \tag{3.2}$$

where

$$U_{\beta\gamma}(t_1, t_2) = \langle \{ Y^0_{n\beta}(t_1), Y^0_{n_\gamma}(t_2)\} \rangle$$

Taking into account all that has been said above, we can write (2.14) in the form

$$\dot{z}_1 = -C_{12}x_2 - C_{13}\Lambda_{34}U_{45}\eta_{45}\Lambda_{56}C_{62}(1/p_2)\,x_2 - \cdots - \xi_1 \tag{3.3}$$

where

$$C_{12} = c_{\alpha_1\alpha_2}\delta(t_1 - t_2) \tag{3.4a}$$

$$\Lambda_{12} = \lambda_{\alpha_1}\delta_{\alpha_1\alpha_2}\delta(t_1 - t_2) \tag{3.4b}$$

$$\xi_1 = -C_{12}\Lambda_{23}Y^0_3 + \mathcal{O}(\lambda^3) \tag{3.4c}$$

Comparing (3.3) with (1.15) gives

$$\Phi_{1,2} = C_{12} + C_{13}\Lambda_{34}U_{45}\eta_{45}\Lambda_{56}C_{62}(1/p_2) + \mathcal{O}(\lambda^4) \tag{3.5}$$

In this case the linear FDR (1.18a) reads

$$\Phi_{12} = kT(\Phi_{1,2} + \Phi_{2,1}) \tag{3.6}$$

where $\Phi_{12} = \langle \xi_1\xi_2\rangle$. Using (3.4c) yields

$$\Phi_{12} = -C_{13}\Lambda_{34}\langle Y^0_4 Y^0_5\rangle\,\Lambda_{56}C_{62} + \mathcal{O}(\lambda^4) \tag{3.7}$$

(we have used that $C_{26} = -C_{62}$). The moment $\langle Y^0_4 Y^0_5\rangle$ is the equilibrium one, since the thermostat is in thermal equilibrium. It is known (see, for example, ref. 5, §16) that for equilibrium the moment formula

$$\langle Y^0_4 Y^0_5\rangle = \frac{E(p_4)}{E(p_4) - 1}\langle [Y^0_4, Y^0_5]\rangle \tag{3.8}$$

is valid in the quantum case, where $E(p) = \exp(i\hbar p/kT)$. Passing to the classical limit, we thus obtain

$$\langle Y^0_4\, Y^0_5\rangle = -\frac{kT}{p_4}U_{45} = \frac{kT}{p_5}U_{45} \tag{3.9}$$

Considering $\eta_{45}+\eta_{54}=1$, we can write formula (3.9) in the form

$$\langle Y_4^0 Y_5^0\rangle=\frac{kT}{p_5}U_{45}\eta_{45}-\frac{kT}{p_4}U_{45}\eta_{54}$$

$$=-kTU_{45}\eta_{45}\frac{1}{p_5}-kTU_{54}\eta_{54}\frac{1}{p_4} \tag{3.10}$$

since $U_{54}=-U_{45}$, $p_5^{-1}U_{45}=U_{45}(p_5^{-1})^T=-U_{45}p_5^{-1}$. Substituting (3.10) into (3.7) gives

$$\Phi_{12}=kT\left(C_{13}A_{34}U_{45}\eta_{45}A_{56}C_{62}\frac{1}{p_2}+C_{26}A_{65}U_{54}\eta_{54}A_{43}C_{31}\frac{1}{p_1}\right) \tag{3.11}$$

It is easy to verify that (3.11) is equal to the expression obtained by substituting (3.5) into the right-hand side of (3.6). Thus, the FDR (3.6) has been proved. In order to prove the second linear relation—reciprocal relation (1.18b)—one should take into consideration the equation

$$\langle Y_1^0 Y_2^0\cdots Y_m^0\rangle^{\text{t.c.}}=\langle Y_1^0 Y_2^0\cdots Y_m^0\rangle \tag{3.12}$$

which is satisfied [under the time-reversibility condition $H_2(Q,-P)=H_2(Q,P)$] by any equilibrium moment in the quantum case and also in the nonquantum case (see ref. 7 and also ref. 5). Equation (3.12) implies

$$\langle[Y_1^0, Y_2^0]\rangle^{\text{t.c.}}=\langle[Y_1^0, Y_2^0]\rangle \tag{3.13}$$

(the quantum case). Further, we have

$$\left(\frac{i}{\hbar}\langle[Y_1^0, Y_2^0]\rangle\right)^{\text{t.c.}}=-\frac{i}{\hbar}\langle[Y_1^0, Y_2^0]\rangle$$

i.e., the equation

$$\langle\{Y_1^0, Y_2^0\}\rangle^{\text{t.c.}}=-\langle\{Y_1^0, Y_2^0\}\rangle \tag{3.14}$$

is valid in the nonquantum case. Applying Eqs. (3.5) and (3.14) and the equations $\eta_{12}^{\text{t.c.}}=\eta_{21}$, $(p_2^{-1})^{\text{t.c.}}=-p_2^{-1}$, and $\delta(t_1-t_2)^{\text{t.c.}}=\delta(t_1-t_2)$, we find

$$\Phi_{1,2}^{\text{t.c.}}=\varepsilon_1 C_{12}\varepsilon_2+\varepsilon_1 C_{13}\varepsilon_3 A_{34}(U_{45}\eta_{45})^{\text{t.c.}}A_{56}\varepsilon_6 C_{62}\varepsilon_2\left(-\frac{1}{p_2}\right)$$

$$=C_{21}+C_{31}A_{34}(-U_{45})\,\eta_{54}A_{56}C_{26}\frac{1}{p_1}$$

$$=C_{21}+C_{26}A_{65}U_{54}\eta_{54}A_{43}C_{31}\frac{1}{p_1}$$

$$=\Phi_{2,1} \tag{3.15}$$

since $\varepsilon_\alpha c_{\alpha\beta}\varepsilon_\beta=c_{\beta\alpha}$ and $U_{45}=-U_{54}$. Equation (3.15) testifies to the validity of relation (1.18a) in the linear approximation.

4. QUADRATIC FDRs

In Eq. (2.14) let us consider the term quadratic in z. In this term, in the quadratic approximation, we can replace v_{123} by

$$\langle v_{123}\rangle = U_{123}\eta_{123} + U_{132}\eta_{132}$$

where $U_{123} = \langle\{\{Y_1^0, Y_2^0\}, Y_3^0\}\rangle$ [Eq. (2.13) has been used]. Transforming this term according to (3.1) and comparing the result with the appropriate term in (1.15), we get

$$\Phi_{1,23} = -C_{14}A_{45}(U_{567}\eta_{567} + U_{576}\eta_{576})\frac{1}{p_6 p_7}A_{68}A_{79}C_{82}C_{93} \tag{4.1}$$

Let us write this expression in the shorter form and perform some transformations

$$\begin{aligned}\Phi_{1,23} &= -C_1A_1(U_{123}\eta_{123} + U_{132}\eta_{132})\frac{1}{p_2p_3}A_2A_3C_2C_3\\ &= -C_1C_2^TC_3^TA_1A_2^TA_3^T\frac{1}{p_2p_3}(U_{123}\eta_{123} + U_{132}\eta_{132})\\ &= -C_1C_2C_3A_1A_2A_3\frac{1}{p_2p_3}(U_{123}\eta_{123} + U_{132}\eta_{132})\end{aligned} \tag{4.2}$$

We now find the time-conjugate function. In so doing, we use that (3.12) implies the formula

$$\langle[[Y_1^0, Y_2^0], Y_3^0]\rangle^{\text{t.c.}} = \langle[[Y_1^0, Y_2^0], Y_3^0]\rangle \tag{4.3}$$

analogous with (3.13), and therefore

$$U_{123}^{\text{t.c.}} = U_{123} \tag{4.4}$$

Using the equations $\eta_{123}^{\text{t.c.}} = \eta_{321}$, $\varepsilon C\varepsilon = -C$, and $p_2^{\text{t.c.}} = -p_2$, from (4.2) we get

$$\begin{aligned}\Phi_{1,23}^{\text{t.c.}} &= -\varepsilon_1C_1\varepsilon_1\varepsilon_2C_2\varepsilon_2\varepsilon_3C_3\varepsilon_3A_1A_2A_3\frac{1}{p_2p_3}(U_{123}\eta_{321} + U_{132}\eta_{231})\\ &= C_1C_2C_3A_1A_2A_3\frac{1}{p_2p_3}(U_{123}\eta_{321} + U_{132}\eta_{231})\end{aligned} \tag{4.5}$$

Now consider the triple moment

$$\Phi_{123} \equiv \langle\xi_1\xi_2\xi_3\rangle = -C_1C_2C_3A_1A_2A_3\langle Y_1^0Y_2^0Y_3^0\rangle \tag{4.6}$$

From the formula (ref. 5, §16)

$$\langle Y_1^0 Y_2^0 Y_3^0 \rangle = -\frac{E(p_1)}{E(p_1)-1}\left(\frac{1}{E(p_3)-1}\langle[[Y_1^0, Y_2^0], Y_3^0]\rangle + \frac{E(p_2)}{E(p_2)-1}\langle[[Y_1^0, Y_2^0], Y_3^0]\rangle\right) \tag{4.7}$$

which is valid for equilibrium moments in the quantum case, we obtain in the classical limit

$$\langle Y_1^0 Y_2^0 Y_3^0 \rangle = -(kT)^2\left(\frac{1}{p_1 p_3} U_{123} + \frac{1}{p_1 p_2} U_{132}\right) \tag{4.8}$$

Therefore Eq. (4.6) takes the form

$$(kT)^{-2}\Phi_{123} = C_1 C_2 C_3 A_1 A_2 A_3 \left(\frac{1}{p_1 p_3} U_{123} + \frac{1}{p_1 p_2} U_{132}\right) \tag{4.9}$$

To prove the relation

$$(kT)^{-2}\Phi_{123} = \Phi_{1,23} - \Phi_{1,23}^{\text{t.c.}} + \Phi_{2,13} - \Phi_{2,13}^{\text{t.c.}} + \Phi_{3,12} - \Phi_{3,12}^{\text{t.c.}} \tag{4.10}$$

we substitute (4.2) and (4.5) into the right-hand side of (4.10). This gives

$$\begin{aligned}
&\Phi_{1,23} + \Phi_{2,13} + \Phi_{3,12} - \text{t.c.} \\
&= C_1 C_2 C_3 A_1 A_2 A_3 \\
&\quad \times \left[\frac{1}{p_2 p_3} U_{123}(\eta_{123} + \eta_{321}) + \frac{1}{p_2 p_3} U_{132}(\eta_{132} + \eta_{231})\right. \\
&\quad + \frac{1}{p_1 p_3} U_{231}(\eta_{213} + \eta_{312}) + \frac{1}{p_1 p_3} U_{231}(\eta_{231} + \eta_{132}) \\
&\quad \left. + \frac{1}{p_1 p_2} U_{312}(\eta_{312} + \eta_{213}) + \frac{1}{p_1 p_2} U_{321}(\eta_{321} + \eta_{123})\right]
\end{aligned} \tag{4.11}$$

Due to the property $U_{klm} = -U_{lkm}$ of the Poisson brackets, from six functions obtained by permutation of the subscripts of U_{123} we obtain three independent functions, and due to the Jacobi identity

$$U_{123} + U_{231} + U_{312} = 0 \tag{4.12}$$

two independent functions remain. We take the functions appearing on the right-hand side of (4.9) as these independent functions. Passing to them in Eq. (4.11) gives

$$\begin{aligned}
&\Phi_{1,23}+\Phi_{2,13}+\Phi_{3,12}-\text{t.c.}\\
&\quad= C_1C_2C_3\Lambda_1\Lambda_2\Lambda_3\\
&\quad\times\left\{\left[-\left(\frac{1}{p_2p_3}+\frac{1}{p_1p_2}\right)(\eta_{123}+\eta_{321})\right.\right.\\
&\quad\left.+\frac{1}{p_1p_3}(\eta_{213}+\eta_{312}+\eta_{231}+\eta_{132})\right]U_{123}\\
&\quad+\left[-\left(\frac{1}{p_2p_3}+\frac{1}{p_1p_3}\right)(\eta_{231}+\eta_{132})\right.\\
&\quad\left.\left.+\frac{1}{p_1p_2}(\eta_{312}+\eta_{213}+\eta_{321}+\eta_{123})\right]U_{132}\right\}
\end{aligned}\tag{4.13}$$

By virtue of the equation

$$\left(\frac{1}{p_1p_2}+\frac{1}{p_1p_3}+\frac{1}{p_2p_3}\right)f_{klm}=0 \tag{4.14}$$

[which is valid in the stationary case when $(p_1+p_2+p_3)f_{klm}=0$] and by virtue of the obvious formula

$$\eta_{123}+\eta_{321}+\eta_{213}+\eta_{312}+\eta_{231}+\eta_{132}=1 \tag{4.15}$$

the expression on the right-hand side of (4.13) coincides with the expression on the right-hand side of (4.9), which proves the FDR (4.10).

We now pass to the other relation in (1.20), i.e., to the FDR

$$(kT)^{-1}\,\Phi_{12,3}=\Phi_{1,23}+\Phi_{2,13}-\Phi^{\text{t.c.}}_{3,12} \tag{4.16}$$

In proving this FDR, one should take into account the distinction between v_{12} and its mean value $\langle v_{12}\rangle$. Regarding the difference $v_{12}-\langle v_{12}\rangle$ as a random force ξ_1, from (2.14) we obtain (with appropriate accuracy) (1.15) if we now put

$$\begin{aligned}
\xi_1&=F_1[Y^0,x]\\
&=-C_1\Lambda_1Y_1^0+C_1\Lambda_1(v_{13}-\langle v_{13}\rangle)\frac{1}{p_3}\Lambda_3C_3x_3
\end{aligned}\tag{4.17}$$

which is more precise than (3.4c). Substituting (4.17) into (1.22) yields

$$\Phi_{12,3} = -C_1 C_2 A_1 A_2(\langle v_{13}, Y_2^0\rangle + \langle v_{23}, Y_1^0\rangle)\frac{1}{p_3} A_3 C_3$$

$$= -C_1 C_2 C_3 A_1 A_2 A_3 \frac{1}{p_3}(\langle v_{13}, Y_2^0\rangle + \langle v_{23}, Y_1^0\rangle) \qquad (4.18)$$

In the equilibrium case a formula of the type (3.9) is valid for any moments and cumulants. Therefore we have

$$\langle v_{13}, Y_2^0\rangle = \frac{kT}{p_2}\langle\{v_{13}, Y_2^0\}\rangle \qquad (4.19)$$

or

$$\langle v_{13}, Y_2^0\rangle = \frac{kT}{p_2}\langle\{\{Y_1^0, Y_3^0\}\eta_{13}, Y_2^0\}\rangle = \frac{kT}{p_2} U_{132}\eta_{13} \qquad (4.20)$$

if we use (2.13). Due to this, Eq. (4.18) assumes the form

$$(kT)^{-1}\Phi_{12,3} = -C_1 C_2 C_3 A_1 A_2 A_3\left(\frac{1}{p_2 p_3}\eta_{13} U_{132} + \frac{1}{p_1 p_3}\eta_{23} U_{231}\right) \qquad (4.21)$$

Substituting (4.2) and (4.5) into the right-hand side of (4.16) gives

$$\begin{aligned}
&\Phi_{1,23} + \Phi_{2,13} - \Phi_{3,12}^{\text{t.c.}} \\
&\quad = -C_1 C_2 C_3 A_1 A_2 A_3 \\
&\quad\quad \times\left(\frac{1}{p_2 p_3}\eta_{123} U_{123} + \frac{1}{p_2 p_3}\eta_{132} U_{132} + \frac{1}{p_1 p_3}\eta_{213} U_{213}\right. \\
&\quad\quad \left. + \frac{1}{p_1 p_3}\eta_{231} U_{231} + \frac{1}{p_1 p_2}\eta_{213} U_{312} + \frac{1}{p_1 p_2}\eta_{123} U_{321}\right)
\end{aligned} \qquad (4.22)$$

Using the above-mentioned properties of the function U_{123}, in (4.22) we perform the transformations such that the functions U_{132} and U_{231} appear in the right-hand side of (4.22). This gives

$$\begin{aligned}
&\Phi_{1,23} + \Phi_{2,13} - \Phi_{3,12}^{\text{t.c.}} \\
&\quad = -C_1 C_2 C_3 A_1 A_2 A_3 \\
&\quad\quad \times\left\{\left[\frac{1}{p_2 p_3}(\eta_{123} + \eta_{132}) - \left(\frac{1}{p_1 p_3} + \frac{1}{p_1 p_2}\right)\eta_{213}\right] U_{132}\right. \\
&\quad\quad \left. + \left[\frac{1}{p_1 p_3}(\eta_{213} + \eta_{231}) - \left(\frac{1}{p_2 p_3} + \frac{1}{p_1 p_2}\right)\eta_{123}\right] U_{231}\right\}
\end{aligned} \qquad (4.23)$$

Using (4.14) and the obvious equation

$$\eta_{123}+\eta_{132}+\eta_{213}=\eta_{13} \tag{4.24}$$

we easily verify that the expression on the right-hand side of (4.23) is equal to the expression on the right-hand side of (4.21). This proves the FDR (4.16).

The foregoing proof may have left the impression that the linear and quadratic FDRs are valid only when the inertial terms in (1.15) are small (which is connected with the smallness of λ). However, this is not so, since using the other methods of proof does not require this smallness. In ref. 5 the linear FDRs are proved by the projection operator method in phase space. The smallness of inertial terms is not assumed in this case. Other methods (nondynamical ones) of proving the linear and nonlinear FDRs of the first kind are also applied in ref. 5.

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