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Author(s)	SUZUKI, Masuo
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長谷川 洋

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Statistical Mechanics of
Non-Equilibrium Systems

— Extensive Property, Fluctuation and Nonlinear Response —

Masuo SUZUKI (Dept. of Physics, Univ. of Tokyo)

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1. Kubo's Ansatz: Extensive Property

$$P(X, t) = C \exp [\Omega \phi(x, t)] ,$$

for large Ω with $x = X/\Omega$; $X =$ macrovariable.

2. When X is a Markoffian macrovariable,

$$\frac{\partial P(x, t)}{\partial t} = \Gamma_M P(x, t) ; \Gamma_M = \text{time develop. op.}$$

evol. eq. for $y(t) =$ deterministic path :

$$\dot{y}(t) = C_1(y(t)) ; \quad C_n = n\text{-th moment,}$$

variance $\sigma(t)$:

$$\dot{\sigma}(t) = 2 C_1'(y(t)) \sigma(t) + C_2(y(t))$$

3. When X is a non-Markoffian macrovariable,

$$\frac{\partial P(\{\sigma_j\}; t)}{\partial t} = \Gamma P(\{\sigma_j\}; t),$$

$$P(X, t) = \sum^{\text{config.}} \delta(\mathbf{X}-X) P(\{\sigma_j\}; t)$$

4. For the macrovariable X in quantum systems,

$$\rho(X, t) = T_r \delta(\mathbf{X}-X) \rho(t) ; \quad i\hbar \frac{\partial \rho(t)}{\partial t} = [\mathcal{H}, \rho(t)]$$

Extensive Property:

$$\rho(X, t) = C \exp [\Omega \phi(x, t)]$$

5. Generating Function Formalism

$$\Psi(\lambda, t) = \begin{cases} T_r e^{\lambda X} \rho(t) \dots\dots \text{(quantal)} \\ \sum_{\text{config.}} e^{\lambda X} e^{t\Gamma} P_0 \dots\dots \text{(stochastic or classical)} \end{cases}$$

$$\rho(X, t) = \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} e^{-\lambda X} \Psi(\lambda, t) d\lambda$$

generalized thermodynamic potential

$$\mathcal{F}(\lambda, t) \equiv \log \Psi(\lambda, t) \text{ or } \Psi(\lambda, t) = e^{\mathcal{F}(\lambda, t)}$$

6. Assumption for the initial distribution:

$$\rho(0) = e^{\mathcal{H}^{(i)}} \text{ or } P_0 = e^{\mathcal{H}^{(i)}} \quad (\text{canonical})(\text{normalized})$$

7. Properties of $\Psi(\lambda, t)$ and $\mathcal{F}(\lambda, t)$ when $X, \mathcal{M}^{(i)}$ and \mathcal{H} are Hermitian:

- 1) $\Psi(\lambda, t) > 0$ and $\mathcal{F}(\lambda, t) = \text{real}$ for λ real.
- 2) When λ is real, Ψ and \mathcal{F} are convex functions of λ .
- 3) For complex λ (= not real), we have

$$|\Psi(\lambda, t)| < \Psi(\text{Re } \lambda, t)$$

8. If $\Psi(\lambda, t) = C_1 e^{\Omega \psi(\lambda, t)}$ (Extensive property),

$$\begin{aligned} \rho(X, t) &= \frac{C_1}{2\pi i} \int_{c-i\infty}^{c+i\infty} \exp[-\Omega\{\lambda x - \psi(\lambda, x)\}] d\lambda \\ &= C_2 \exp[\Omega\{\psi(\lambda_0, t) - \lambda_0 x\}] \end{aligned}$$

for large Ω , λ_0 being given by the solution

$$\frac{\partial \psi(\lambda, t)}{\partial \lambda} = x$$

9. "Local operator" $Q(\mathbf{r})$ is defined by a functional of field operators $\{\psi(\mathbf{r}')\}$; $\mathbf{r}' \in D(\mathbf{r}) \equiv$ circle domain of radius b .

10. General conditions for $X, \mathcal{M}^{(i)}$ and \mathcal{H} :

$$\mathbf{X} = \int \mathbf{X}(\mathbf{r}) d\mathbf{r}, \mathcal{M}^{(i)} = \int \mathcal{M}^{(i)}(\mathbf{r}) d\mathbf{r} \text{ and } \mathcal{H} = \int \mathcal{H}(\mathbf{r}) d\mathbf{r}.$$

11. Theorem for "extensive property" in quantal systems.

Theorem I (quantal) If the averages of local operators, $\langle \mathbf{X}(\mathbf{r}) \rangle^{(1)}$, $\langle \mathcal{M}^{(i)}(\mathbf{r}) \rangle^{(2)}$, and $\langle \mathcal{H}(\mathbf{r}) \rangle^{(3)}$ are bounded, then

$$\lim_{\Omega \rightarrow \infty} \psi_{\Omega}(\lambda, t) = \text{exist (uniformly convergent)}$$

for $|\lambda| < A$ (fixed) and t finite. Therefore $\Psi(\lambda, t)$ has the extensive property: $\Psi(\lambda, t) = C_1 \exp[\Omega \psi(\lambda, t)]$

and consequently so does $\rho(X, t)$.

12. Definition of the averages

$$\langle \mathbf{X}(\mathbf{r}) \rangle^{(1)}, \langle \mathcal{H}^{(i)}(\mathbf{r}) \rangle^{(2)} \text{ and } \langle \mathcal{H}(\mathbf{r}) \rangle^{(3)} :$$

First separate the system Ω into Ω_1 and Ω_2 :

$$\mathbf{X} = X_1 + X_2, \quad \mathcal{H}^{(i)} = \mathcal{H}_1^{(i)} + \mathcal{H}_2^{(i)} \text{ and } \mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2,$$

where $Q_j = \int_{\Omega_j} Q(\mathbf{r}) d\mathbf{r}$.

$$\text{Operational; } P_{(s,\mu)} Q \equiv e^{-s(P_1 + \mu P_2)} Q e^{s(P_1 + \mu P_2)},$$

$$\langle Q \rangle^{(j)} \equiv T_{\mathbf{r}} Q \rho_j / T_{\mathbf{r}} \rho_j,$$

where $\rho_j = \rho_j(s, \mu)$, $0 \leq s \leq 1$, $0 \leq \mu \leq 1$,

$$\begin{cases} \rho_1(s, \mu) \equiv \exp[\lambda(X_1 + \mu X_2)] \exp[\lambda X_{(s,\mu)} \mathcal{H}_{(s,\mu)}^{(i)}], \\ \rho_2(s, \mu) \equiv \exp[\mathcal{H}_1^{(i)} + \mu \mathcal{H}_2^{(i)}] \exp[\lambda \mathcal{H}_{(s,\mu)}^{(i)} (-it, 1) X_1] \\ \rho_3(s, \mu) \equiv \exp[\mathcal{H}_{(ist,\mu)}^{(i)} \mathcal{H}_1^{(i)}] \exp[\lambda \mathcal{H}_{(i(s-1)t,\mu)} X_1] \end{cases}$$

 13. Remarks on Theorem I

a) Ψ is a continuous function of λ .

b) "Bounded" $\Rightarrow |\langle Q(\mathbf{r}) \rangle^{(j)}| \leq c_j$ (indep. of Ω).

c) If $\{c_j\}$ are indep. of λ for $|\lambda| < A$, the limit $\psi_{\Omega}(\lambda, t) \Rightarrow$

$\psi(\lambda, t)$ is uniformly convergent in wider sense in the complex circle domain $|\lambda| < A$. Hence,

$$\lim_{\Omega \rightarrow \infty} \frac{d^p \psi_{\Omega}(\lambda, t)}{d\lambda^p} = \frac{d^p \psi(\lambda, t)}{d\lambda^p} \quad (\text{Weierstrass})$$

d) "bounded" for $\Omega_2/\Omega_1 = O(\Omega^{-1/d}) \Rightarrow \text{O.K.}$

e) "bounded" for $s = \mu = 1 \Rightarrow$ " " for $0 < s < 1$, $0 < \mu < 1$
(conjecture)

f) When $\mathbf{X}(\mathbf{r})$ commutes with $\mathbf{X}(\mathbf{r}')$, $\langle \mathbf{X}(\mathbf{r}) \rangle^{(1)}$ is bounded.

g) If $[\mathcal{H}^{(i)}(\mathbf{r}), \mathcal{H}^{(i)}(\mathbf{r}')] = 0 \Rightarrow \langle \mathcal{H}^{(i)}(\mathbf{r}) \rangle^{(1)}$ is bounded.

It is not easy to prove that $\langle \mathcal{H}(\mathbf{r}) \rangle^{(3)}$ is bounded when

$$[\mathcal{H}(\mathbf{r}), \mathcal{H}(\mathbf{r}')] = 0.$$

- h) Clearly, $\langle \mathcal{X}(\mathbf{r}) \rangle^{(3)}$ is bounded if $\mathcal{X}_2 \equiv 0$.
- i) Only the boundness of $\langle \mathbf{X}(r) \rangle^{(1)}$ etc. are assumed.
- j) $\lim_{\Omega \rightarrow \infty} =$ thermodynamic limit in van Hove's sense.
- k) Theorem I is extended to classical systems. (Classical Liouville op.).

14. Proof of Extensive Property:

Separation of the domain Ω

$$\Omega_n = \Omega_1 + \Omega_2$$

$n =$ large integer

$$\left\{ \begin{array}{l} \mathbf{X} = \mathbf{X}_1 + \mathbf{X}_2 \\ \mathcal{X}^{(i)} = \mathcal{X}_1^{(i)} + \mathcal{X}_2^{(i)} \\ \mathcal{X} = \mathcal{X}_1 + \mathcal{X}_2 \end{array} \right.$$

To prove Cauchy's condition on convergence.

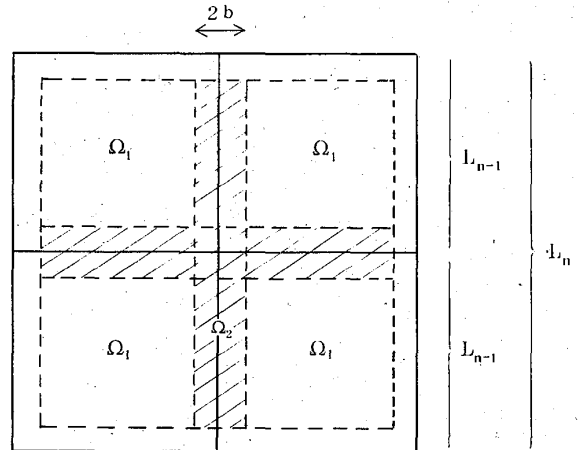


図1 Domains Ω_1 and Ω_2 with each margin of width b inside in it; Ω_2 is the shaded region.

$$|\psi_n(\lambda, t) - \psi_{n-1}(\lambda, t)| \leq 2^{-n} (Ac_1 + c_2 + 2tc_3)(2bd),$$

$d =$ dimension,

$$\therefore |\psi_{n+m}(\lambda, t) - \psi_n(\lambda, t)| \leq 2^{-n} (\quad \quad \quad),$$

Formula. $\therefore \lim_{n \rightarrow \infty} \psi_n(\lambda, t) = \text{exist} = \psi(\lambda, t)$

$$\frac{d}{dx} e^{A(x)} = \int_0^1 e^{(1-s)A(x)} A'(x) e^{sA(x)} ds = \int_0^1 e^{sA(x)} A'(x) e^{(1-s)A(x)} ds$$

15. Extensive property in stochastic models

$$\frac{\partial}{\partial t} P(\{\sigma_j\}, t) = \Gamma P(\{\sigma_j\}, t) ; \Gamma = \sum_j \Gamma_j$$

where $\sigma_j = \pm 1$ (spin system) and

$$\Gamma_j P(\{\sigma_j\}, t) = -W_j(\sigma_j) P(\dots, \sigma_j, \dots) + W_j(-\sigma_j) P(\dots, -\sigma_j, \dots)$$

16. Theorem II (stochastic).

If $P(st) \equiv e^{st(\Gamma_1 + \mu\Gamma_2)} \mathcal{A}(i) \in \mathcal{N}$

("normal") for any Γ_1 (and $\Gamma_2 = \Gamma - \Gamma_1$) and for $0 \leq s \leq 1$ and $0 \leq \mu \leq 1$, then

$\lim_{\Omega \rightarrow \infty} \psi_{\Omega}(\lambda, t) = \text{exist}$ (uniformly convergent)

$\Rightarrow \psi(\lambda, t)$ and $P(X, t)$ have extensive property.

17. Lemma 1. — If $f(\{\sigma_j\}) \leq g(\{\sigma_j\})$, then $e^{t\Gamma} f \leq e^{t\Gamma} g$ for $t \geq 0$.

Lemma 2. — If $f(\{\sigma_j\}) \in \mathcal{N}$ then $|\Gamma_2 f| \leq C_{\Omega_2} f$, where C_{Ω_2} is a constant dependent on Ω_2 ,

Def. $P \in \mathcal{N}$ ("normal") $\Leftrightarrow P(\dots, -\sigma_j, \dots, t) \leq C_3 P(\dots, \sigma_j, \dots)$,

where C_3 is a constant independent of Ω ,

18. Average motion, Fluctuation, and Nonlinear response

most probable path $y(t)$ for Ω large:

1) $x = y(t) + z, g(z, t) \equiv \phi(y+z, t) = \{\psi(\lambda(y, t), t) - y\lambda(y, t)\} - z\{\lambda(y, t) + [y - (\frac{\partial\psi}{\partial\lambda})_{x=y}](\frac{\partial\psi}{\partial\lambda})_{x=y}\} + \dots$,

$\Rightarrow y(t) = (\frac{\partial\psi}{\partial\lambda})_{\lambda=0} = \langle x \rangle_t; \langle X \rangle_t = \int X \rho(X, t) dX = T_{\Gamma} X \rho(t)$.

2) variance: $\sigma(t) = (\partial^2 \psi / \partial \lambda^2)_{\lambda=0} = \Omega \langle (x - \langle x \rangle_t)^2 \rangle_t$.

3) Nonlinear response: case (a); $\tilde{\mathcal{X}}^{(i)} = \mathcal{X}^{(i)} + KY$

$y(t) = \langle x \rangle_{t,k}^{(a)} \equiv \Omega^{-1} (\frac{\partial \tilde{\mathcal{X}}}{\partial \lambda})_{\lambda=0} = T_{\Gamma} x e^{\tilde{\mathcal{X}}^{(i)}(t)} / T_{\Gamma} e^{\mathcal{X}^{(i)}}$

Theorem IIIa. — The nonlinear response for case (a) is

expressed by the fluctuation in non-equilibrium as

$\Phi_{XY;Y}^{(a)} \equiv \frac{\partial y(t)}{\partial K} = \Omega^{-1} \int_0^1 d\mu \langle \delta X \delta Y(t; \mu) \rangle_{t,K}^{(a)} \equiv \Omega^{-1} \langle \delta X; \delta Y(t) \rangle_{t,K}^{(a)}$

for a quantal system, where

$$Y(t; \mu) = e^{\mu \tilde{\mathcal{X}}^{(i)}} Y(t) e^{-\mu \tilde{\mathcal{X}}^{(i)}} \quad \text{and} \quad Y(t) = e^{-it\mathcal{X}} Y e^{it\mathcal{X}}.$$

For stochastic systems, we have

$$\Phi_{XY; Y}^{(a)} \equiv \frac{\partial y(t)}{\partial K} = \Omega^{-1} \langle \delta X \delta Y(t) \rangle_{t, K}^{(a)};$$

$$Y(t) = (e^{t\Gamma} e^{\tilde{\mathcal{X}}^{(i)}})^{-1} e^{t\Gamma} (Y e^{\tilde{\mathcal{X}}^{(i)}}).$$

$$1) \langle \delta X; \delta Y(t) \rangle_{t, K}^{(a)} = \langle \delta Y(t); \delta X \rangle_{t, K}^{(a)},$$

$$2) \langle \delta X; \delta Y(t) \rangle_{t, K}^{(a)*} = \langle \delta Y(t); \delta X \rangle_{t, K}^{(a)},$$

$$3) \Phi_{XY; Y}^{(a)} = \text{real}.$$

For a general nonlinear disturbance of the form $\tilde{\mathcal{X}}^{(i)} = \mathcal{X}_K^{(i)}$,

$$\Phi_{XY; Y}^{(a)} \equiv \frac{\partial y(t)}{\partial K} = \langle \delta X; \delta \left(\frac{\partial}{\partial K} \mathcal{X}_K^{(i)}(t) \right) \rangle_{t, K}^{(a)}.$$

case (b) $\tilde{\mathcal{X}} = \mathcal{X} + KY$:

$$y(t) = \langle x \rangle_{t, K}^{(b)} \equiv \Omega^{-1} \left(\frac{\partial \mathcal{Z}}{\partial \lambda} \right)_{\lambda=0} = \text{Tr} x e^{-it\tilde{\mathcal{X}}} e^{\mathcal{X}^{(i)}} e^{it\tilde{\mathcal{X}}}$$

Theorem II|b.

$$\Phi_{XY; Y}^{(b)} \equiv \frac{\partial y(t)}{\partial K} = -\frac{i}{\Omega} \int_0^t \langle [\mathbf{X}, \mathbf{Y}(s)] \rangle_{t, K}^{(b)} ds,$$

for a quantal system where $Y(s) = \exp(-is\tilde{\mathcal{X}}) Y \exp(is\tilde{\mathcal{X}})$.

$$\Phi_{XY; Y}^{(b)} = \sum x e^{t\Gamma_K} \int_0^t ds e^{-s\Gamma_K} \left(\frac{d}{dK} \Gamma_K \right) e^{s\Gamma_K} e^{\mathcal{X}^{(i)}}$$

for a stochastic system, where $\Gamma_K \leftrightarrow \tilde{\mathcal{X}} = \mathcal{X} + KY$.

For a general nonlinear disturbance of the form $\tilde{\mathcal{X}} = \mathcal{X}_K$,

$$\Phi_{XY; Y}^{(b)} \equiv \frac{\partial}{\partial K} y(t) = -i \int_0^t \langle [x, \left(\frac{\partial}{\partial K} \mathcal{X}_K \right)(s)] \rangle_{t, K}^{(b)} ds.$$

19. Relation between $\phi(x, t)$ and cumulants

Theorem IV. — If $\Psi(\lambda, t)$ (and $\rho(x, t)$) has extensive property,

$\langle \mathbf{X}^n \rangle_{c,t}$ = extensive (i.e., $O(\Omega)$). Conversely, if $\langle \mathbf{X}^n \rangle_{c,t}$ are extensive, and

if $\overline{\lim}_{n \rightarrow \infty} (\langle \mathbf{X}^n \rangle_{c,t} / n!)^{1/n} = A_0^{-1} < \infty$, then $\Psi(\lambda, t)$ and $\rho(x, t)$ exist and have extensive property.

20. Theorem V. — The function $\phi(x, t)$ is expressed by the cumulants

$$\text{as } \phi(y+z, t) = - \sum_{n=2}^{\infty} \frac{z^n}{n!} \lim_{\Omega \rightarrow \infty} f_n (\langle \mathbf{X}^2 \rangle_{c,t} \Omega^{-1}, \dots, \langle \mathbf{X}^n \rangle_{c,t} \Omega^{-1}),$$

where f_n is defined by

$$(\psi_2^{-1} \partial / \partial \lambda)^{n-2} \psi_2^{-1} \equiv f_n (\psi_2, \psi_3, \dots, \psi_n); \psi_n \equiv \partial^n \psi / \partial \lambda^n,$$

and $\psi_2^{2n-3} f_n$ is a homogeneous polynomial of order $(n-2)$.

21. Explicit forms of $\{f_n\}$:

$$f_2 = \psi_2^{-1}, \quad f_3 = -\psi_3 \psi_2^{-3}, \quad f_4 = -(\psi_2 \psi_4 - 3\psi_2^2) \psi_2^{-5}, \quad \dots$$

22. Theorem VI. When $\phi(y+z, t) = \sum_{n=2}^{\infty} a_n z^n / n!$ is known, all

cumulants $\langle \mathbf{X}^n \rangle_{c,t}$ are obtained explicitly with use of the recursion formula;

$$\begin{aligned} \langle \mathbf{X}^n \rangle_{c,t} = & \Omega \{ a_n [\sigma(t)]^n + \\ & + [\sigma(t)]^{3-n} g_n (\langle \mathbf{X}^2 \rangle_{c,t} \Omega^{-1}, \dots, \langle \mathbf{X}^{n-1} \rangle_{c,t} \Omega^{-1}) \}, \end{aligned}$$

for $n \geq 3$, and for large Ω , where g_n is given by

$$g_n (\psi_2, \dots, \psi_{n-1}) = -\psi_2^{2n-3} f_n - \psi_2^{n-3} \psi_n.$$

†

23. Explicit forms of $\langle \mathbf{X}^n \rangle_{c,t}$ are

$$\langle \mathbf{X}^3 \rangle_{c,t} = \Omega a_3 [\sigma(t)]^3, \quad \langle \mathbf{X}^4 \rangle_{c,t} = \Omega \{ a_4 [\sigma(t)]^4 + 3a_3^2 [\sigma(t)]^5 \}, \dots$$

24. Composed macrovariable $f(x)$; $x = \mathbf{X}/\Omega$:

$$\langle f(x) \rangle = f(y(t)) + O(\Omega^{-1}),$$

and the variance $\sigma_f(t)$ is given by

$$\sigma_f(t) = \left(\frac{\partial f}{\partial x} \right)_{x=y(t)}^2 \sigma_x(t) + O(\Omega^{-1}),$$

25. Exactly soluble models:

Ex.1 Non-interacting system:

$$\mathcal{H}_{\text{NI}} = -J \sum_j \sigma_j^x, \quad X = \sum_{j=1}^{\Omega} \sigma_j^z \quad \text{and} \quad \mathcal{H}^{(i)} = h \sum_{j=1}^{\Omega} \sigma_j^z.$$

Solution: $\Psi(\lambda, t) = \exp [\Omega \psi(\lambda, t)]$ (exact for any Ω),

$$\psi(\lambda, t) = \log \{ \cosh \lambda + \sinh \lambda \tanh h \cos(2tJ/h) \},$$

$$\lambda(x, t) = \tanh^{-1} \left[\frac{x - a(t)}{1 - a(t)x} \right]; \quad a(t) = \tanh h \cos(2tJ/h),$$

$$\phi(x, t) = \log \{ a(t) \sinh \lambda(x, t) + \cosh \lambda(x, t) \} - x \lambda(x, t)$$

$$y(t) = a(t) \quad \text{and} \quad \text{variance} \quad \sigma(t) = 1 - y^2(t)$$

Ex.2 Kinetic Weiss Ising model (Kubo et al,

Ex.3 (Anisotropic) Weiss Heisenberg model

$$\mathcal{H} = \mathcal{H}(J_x, J_y, J_z; H) = -\Omega^{-1} \sum_{i,j} (J_x \sigma_i^x \sigma_j^x + J_y \sigma_i^y \sigma_j^y + J_z \sigma_i^z \sigma_j^z) - \mu H \sum_j \sigma_j^z,$$

$$X = \sum_j \sigma_j^z, \quad \text{and} \quad \mathcal{H}^{(i)} = -\beta \mathcal{H}(J_x^0, J_y^0, J_z^0; H_0)$$

Ex.4 Nonlinear relaxation in the linear kinetic Ising model.

Ex.5 Nonlinear relaxation in the linear anisotropic XY-model:

$$\mathcal{H} = \mathcal{H}(J_x, J_y; H) = -\sum_j^{\Omega} (J_x \sigma_j^x \sigma_{j+1}^x + J_y \sigma_j^y \sigma_{j+1}^y) - \mu H \sum_j \sigma_j^z,$$

$$X = \sum_j \sigma_j^z \quad (\text{or} \quad \sum_j \sigma_j^x \sigma_{j+1}^x \text{ etc.}), \quad \mathcal{H}^{(i)} = -\beta \mathcal{H}(J_x^0, J_y^0; H^0).$$

26. Applications: relaxation and fluctuation in superconductors.

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