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Statistical Mechanics of Non-Equilibrium Systems

— Extensive Property, Fluctuation and Nonlinear Response —

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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,

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

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

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

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,

$$\rho(X,t) = T_r\,\delta(\mathbf{X}-X)\,\rho(t) \; ; \; ih\frac{\partial \rho(t)}{\partial t} = [\mathscr{H}, \rho(t)]$$

Extensive Property:

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

5. Generating Function Formalism

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

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

generalized thermodynamic potential

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

6. Assumption for the initial distribution:

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

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

1) $\Psi(\lambda, t) > 0$ and $\mathcal{F}(\lambda, t) =$ 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

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

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

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

$$= C_2 \exp[\Omega\{\psi(\lambda_0, t) - \lambda_0 x\}]$$

for large Ω, λ_0 being given by the solution

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

9. "Local operator" $Q(r)$ 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, $\mathcal{H}^{(i)}$ and $\mathcal{H}$:

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

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

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

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

for $|\lambda| < \Lambda$ (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

$<\mathbf{X}(\mathbf{r})>^{(1)}$, $<\mathscr{H}^{(i)}(\mathbf{r})>^{(2)}$ and $<\mathscr{H}(\mathbf{r})>^{(3)}$:

First separate the system Ω into Ω_1 and Ω_2 :

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

where $Q_j = \int_{\Omega_j} Q(r)\,dr$.

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

$$<Q>^{(j)} \equiv T_r\, Q\, \rho_j / T_r\, \rho_j ,$$

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

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

13. Remarks on Theorem I

a) Ψ is a continuous function of λ。

b) "Bounded" $\Longrightarrow$ $|<Q(\mathbf{r})>^{(j)}| \leqq c_j$ (indep. of Ω).

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

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

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

d) "bounded" for $\Omega_2/\Omega_1 = O(\Omega^{-1/d}) \Rightarrow$ 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 $\mathbf{X}(r')$, $<\mathbf{X}(\mathbf{r})>^{(1)}$ is bounded.

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

It is not easy to prove that $<\mathscr{H}(\mathbf{r})>^{(3)}$ is bounded when $[\mathscr{H}(\mathbf{r}), \mathscr{H}(\mathbf{r}')] = 0$。

h) Clearly, $<\mathcal{H}(\mathbf{r})>^{(3)}$ is bounded if $\mathcal{H}_2 \equiv 0$.

i) Only the boundness of $<\mathbf{X}(r)>^{(1)}$ etc. are assumed.

j) $\mathrm{lnm}_{\Omega\to\infty}$ = thermodynamic limit in van Hove's sense.

k) Theorem I is extended to classical systems. (Classical Liouville op.).

14: Proof of Extensive Porperty: Separation of the domain Ω

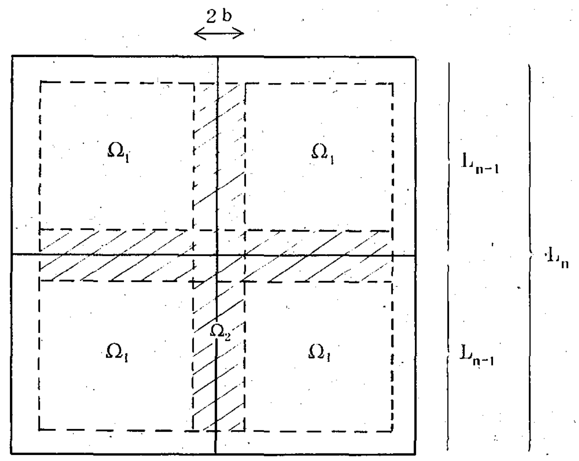

図 1 Domains Ω_1 and Ω_2 with each margin of width b inside in it; Ω_2 is the shaded region.

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

n = large integer

$$\begin{cases} \mathbf{X} = \mathbf{X}_1 + \mathbf{X}_2 \\ \mathcal{H}^{(i)} = \mathcal{H}_1^{(i)} + \mathcal{H}_2^{(i)} \\ \mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2 \end{cases}$$

To prove Cauchy's condition on convergence.

$$|\psi_n(\lambda,t) - \psi_{n-1}(\lambda,t)| \leq 2^{-n}(\Lambda c_1 + c_2 + 2tc_3)(2bd),$$

d = dimension,

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

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

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

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

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

16. Theorem II (stochastic).

If $P(st) \equiv e^{st(\Gamma_1+\mu\Gamma_2)} e^{\mathscr{H}(i)} \in \mathscr{N}$ ("normal") for any Γ_1 (and $\Gamma_2 = \Gamma - \Gamma_1$) and for $0 \leqq s \leqq 1$ and $0 \leqq \mu \leqq 1$, then

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

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

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

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

Def. $P \in \mathscr{N}$ ("normal") $\Longleftrightarrow P(\cdots, -\sigma_j, \cdots, t) \leqq C_3 P(\cdots, \sigma_j, \cdots)$, 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) $x = y(t) + z$, $g(z,t) \equiv \phi(y+z, t) = \{\psi(\lambda(y,t), t) - y\lambda(y,t)\}$

$$-z\{\lambda(y,t) + [y - (\frac{\partial\psi}{\partial\lambda})_{x=y}](\frac{\partial\psi}{\partial\lambda})_{x=y}\} + \cdots\cdots,$$

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

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) ; $\tilde{\mathscr{H}}^{(i)} = \mathscr{H}^{(i)} + KY$

$$y(t) = \langle x\rangle^{(a)}_{t,k} \equiv \Omega^{-1}(\frac{\partial\mathscr{F}}{\partial\lambda})_{\lambda=0} = T_r\, x\, e^{\tilde{\mathscr{H}}^{(i)}}(t) / T_r\, e^{\mathscr{H}^{(i)}}$$

Theorem IIIa. — The nonlinear response for case (a) is expressed by the fluctuation in non-equilibrium as

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

for a quantal system, where

$$Y(t;\mu) = e^{\mu\widetilde{\mathscr{H}}^{(i)}}\, Y(t)\, e^{-\mu\widetilde{\mathscr{H}}^{(i)}} \quad \text{and} \quad Y(t) = e^{-it\mathscr{H}}\, Y\, e^{it\mathscr{H}}.$$

For stochastic systems, we have

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

$$Y(t) = \left(e^{t\Gamma} e^{\widetilde{\mathscr{H}}^{(i)}}\right)^{-1} e^{t\Gamma}\left(Y e^{\widetilde{\mathscr{H}}^{(i)}}\right).$$

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

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

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

For a general nonlinear disturbance of the form $\widetilde{\mathscr{H}}^{(i)} = \mathscr{H}^{(i)}_K$,

$$\Phi^{(a)}_{XY;Y} \equiv \frac{\partial y(t)}{\partial K} = \langle \delta X; \delta\left(\frac{\partial}{\partial K}\mathscr{H}^{(i)}_K(t)\right) \rangle^{(a)}_{t,K}.$$

case (b) $\widetilde{\mathscr{H}} = \mathscr{H} + K\mathbf{Y}$:

$$y(t) = \langle x \rangle^{(b)}_{t,K} \equiv \Omega^{-1} \left(\frac{\partial \mathscr{F}}{\partial \lambda}\right)_{\lambda=0} = \mathrm{Tr}\ x\, e^{-it\widetilde{\mathscr{H}}}\, e^{\mathscr{H}^{(i)}}\, e^{it\widetilde{\mathscr{H}}}$$

Theorem IIIb.

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

for a quantal system where $Y(s) = \exp(-is\widetilde{\mathscr{H}})\, Y \exp(is\widetilde{\mathscr{H}})$.

$$\Phi^{(b)}_{XY;Y} = \Sigma\, x\, e^{t\Gamma_K} \int_0^1 ds\, e^{-s\Gamma_K} \left(\frac{d}{dK}\Gamma_K\right) e^{s\Gamma_K}\, e^{\mathscr{H}(i)}$$

for a stochastic system, where $\Gamma_K \longleftrightarrow \widetilde{\mathscr{H}} = \mathscr{H} + KY$.

For a general nonlinear disturbance of the form $\widetilde{\mathscr{H}} = \mathscr{H}_K$,

$$\Phi^{(b)}_{XY;Y} \equiv \frac{\partial}{\partial K}\, y(t) = -i \int_0^t \langle [x, \left(\frac{\partial}{\partial K}\mathscr{H}_K\right)(s)] \rangle^{(b)}_{t,K}\, ds.$$

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

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

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

if $\overline{\lim_{n\to\infty}} (\langle X^n\rangle_{c,t}/n!)^{1/n} = \Lambda_0^{-1} < \infty$, 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 as $\phi(y+z,t) = -\sum_{n=2}^{\infty} \frac{z^n}{n!} \lim_{\Omega\to\infty} f_n(\langle X^2\rangle_{c,t}\Omega^{-1}, \cdots, \langle X^n\rangle_{c,t}\Omega^{-1})$, where f_n is defhned by

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

and ψ_2^{2n-3} f_n is a homogeneous polynomial of order $(n-2)$.

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

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

22. Thorem VI. When $\phi(y+z,t) = \sum_{n=2}^{\infty} a_n z^n/n!$ is known, all cumulants $\langle X^n\rangle_{c,t}$ are obtained explicitly with use of the recursion formula;

$$\langle X^n\rangle_{c,t} = \Omega\{a_n[\sigma(t)]^n +$$

$$+[\sigma(t)]^{3-n} g_n(\langle X^2\rangle_{c,t}\Omega^{-1},\cdots;\langle X^{n-1}\rangle_{c,t}\Omega^{-1})\},$$

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

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

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

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

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

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

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

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

25. Exactly soluble models:

Ex.1 Non-interacting system:

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

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

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

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

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

$y(t) = a(t)$ and variance $\sigma(t) = 1 - y^2(t)$

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

Ex.3 (Anisotropic) Weiss Heisenberg model

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

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

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

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

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

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

26. Applications: relaxation and fluctuation in superconductors.

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