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Asymmetric response and fluctuation in nonequilibrium steady state

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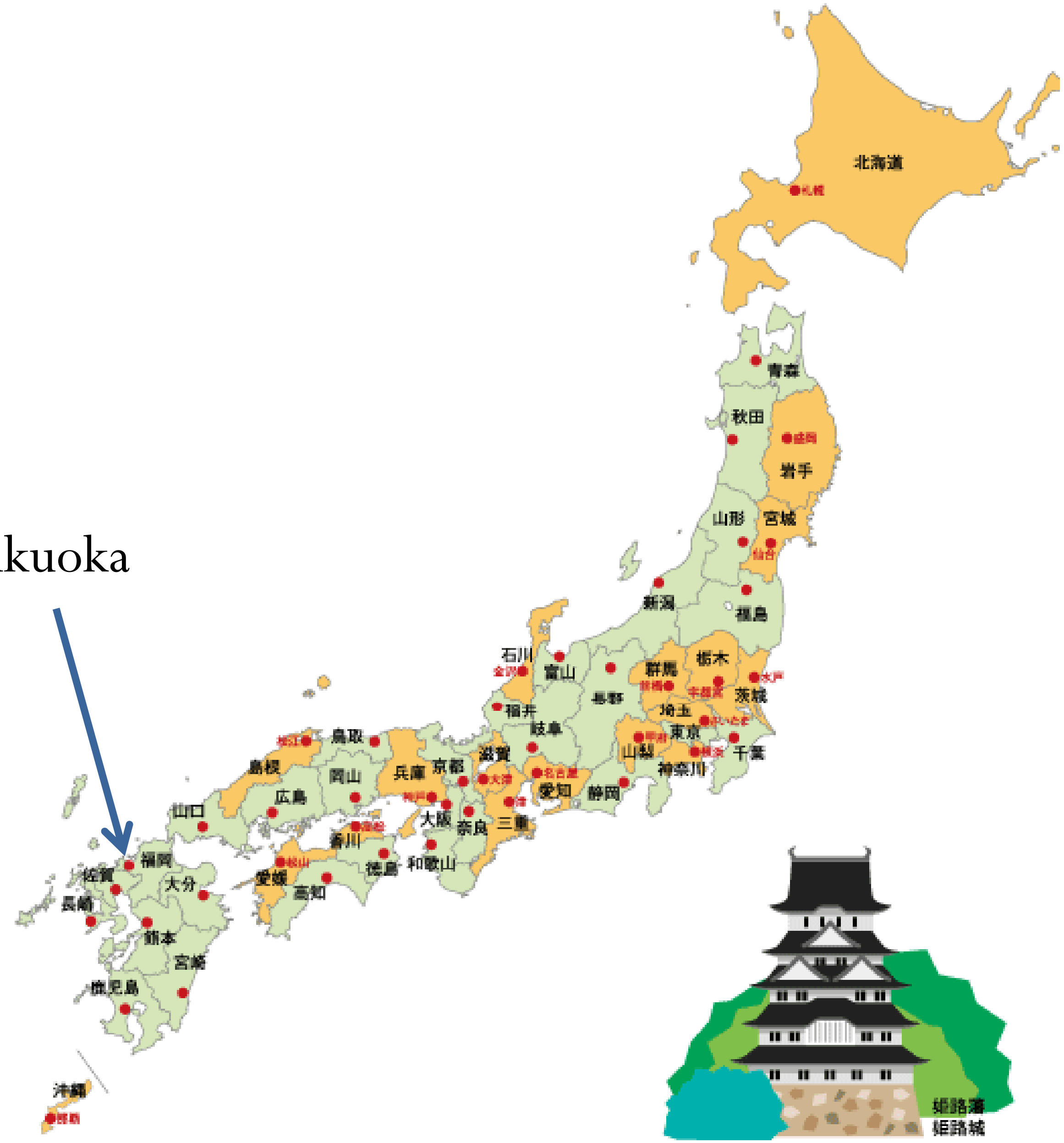
Fukuoka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徳島
和歌山
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高知
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佐賀
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姫路城

Background

Many systems described as a stochastic process

Virtually all systems treated in StatMech.

Secure motion + Fluctuations

Fluctuations reflect the system properties

(Co)valiance and Correlation function (2nd moment)

Background

Fluctuation-Dissipation Theorem (FDT)

Fluctuations in thermal equilibrium ~ the response property of the system

e.g., Einstein relation for a Brownian particle: $D=\mu T$

$C_{ij}(t)=T\chi_{ij}(|t|)$ and $C_{ij}(t)=C_{ij}(-t)$ $[=C_{ji}(t)]$: time reversal symmetry (and translational invariance)

→ $\chi_{ij}(t) = \chi_{ji}(t)$ $(t>0)$: Reciprocal relation (Onsager)

Deviations from FDT formula in Non-Equilibrium Steady State (NESS)

How to quantify?

various expressions seem to be possible …

How to quantify FRR in NESS

\# No detailed balance,
Broken time-reversal symmetry,
Reciprocal relation?

\# T_{eff} may be useful, but not always

\# Asymmetries are important in multi-freedom
and/or anisotropic systems
e.g., chemo-mechanical coupling in molecular motors

\# Nontrivial relations even in the Gaussian regime (linear)
→ many applications
(eg. confined colloids, polymers in flow, driven dissipative systems, etc.)

T_{eff} in molecular motors?

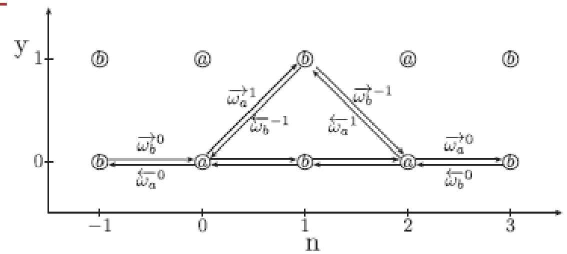
Response and diffusion matrix : λ and D (via FT)

$$\lambda_{11} \equiv \partial v / \partial f, \lambda_{12} \equiv \partial v / \partial \Delta\mu$$
$$\lambda_{21} \equiv \partial r / \partial f, \lambda_{22} \equiv \partial r / \partial \Delta\mu$$
$$2D_{ij} = \partial z_i \partial z_j \vartheta[0,0]$$

$$v = -\partial_{z_1} \vartheta[0,0]$$
$$r = -\partial_{z_2} \vartheta[0,0]$$

v: mean velocity, r: mean ATP consumption rate
f: external load, Δμ: chemical potential diff.

# of ATP consumed



cite (even: low E, odd: high E)

A.W. Lau et.al., PRL (07)

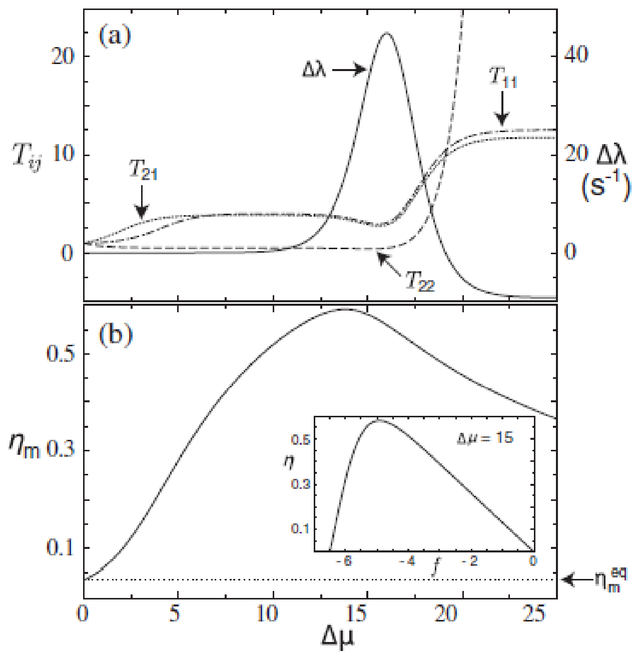
Near equilibrium

$$v = \lambda_{11}^0 f + \lambda_{12}^0 \Delta\mu \qquad \lambda_{ij}^0 = D_{ij}$$
$$r = \lambda_{21}^0 f + \lambda_{22}^0 \Delta\mu \qquad \lambda_{12}^0 = \lambda_{21}^0$$

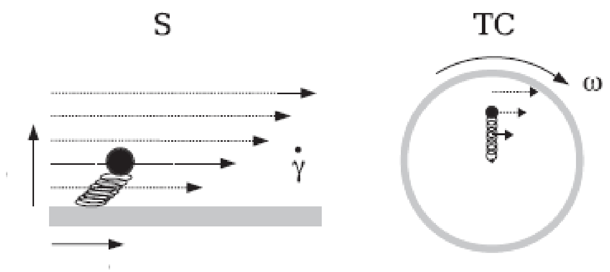
Far from equilibrium

$$T_{ij} \equiv D_{ij} / \lambda_{ij}$$
4 T_{eff} s

$$\Delta\lambda \equiv \lambda_{12} - \lambda_{21}$$
Asymmetrical response



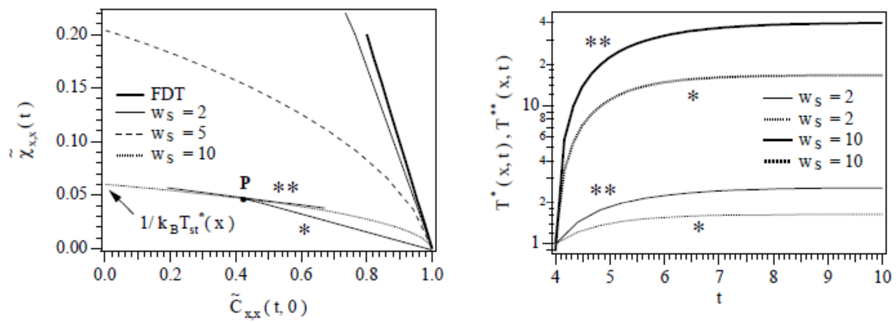
T_{eff} in confined driven colloids?



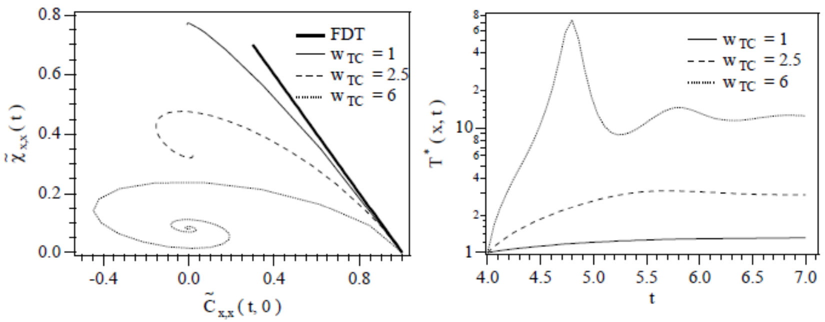
R. Mauri and D. Leporini, EPL (2006)

$$T^{*}(A,t) \equiv \frac{1-\tilde{C}_{A,A}(t)}{\tilde{R}_{A,A}(t)}, \qquad T^{**}(A,T) \equiv -\frac{\partial \tilde{C}_{A,A}(t)}{\partial \tilde{R}_{A,A}(t)}, \qquad \text{cf. } TR_{AB}(t) = C_{AB}(0) - C_{AB}(t) \text{ (equilibrium)}$$

$$R(t) = \int_0^t \chi(t')dt' : \text{integrated response, } \tilde{X}(t) = X(t)/C(0)$$



S-model



TC-model

$T^{**}(t) < 0$??

$T^*(\infty)$ $= T^*_{static}$: well-defined, but …

How to quantify FRR in NESS

\# No detailed balance,
Broken time-reversal symmetry,
Reciprocal relation?

\# T_{eff} may be useful, but not always

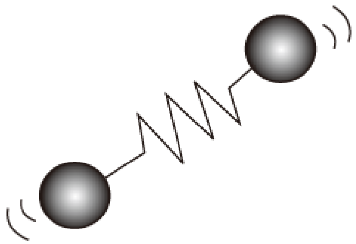
\# **Asymmetries** are important in multi-freedom
and/or anisotropic systems
e.g., chemo-mechanical coupling in molecular motors

\# Nontrivial relations even in the Gaussian regime (linear)
→ many applications (cf. Central limit theorem)
(eg. confined colloids, polymers in flow, driven dissipative systems, etc.)

→ General argument is possible

Simple model: Polymer in flow

Dumbbell model



$$\gamma\left(\frac{d\vec{x}(t)}{dt} - \mathbf{K}_2 \cdot \vec{x}(t)\right) = -\nabla U(x(t)) + \vec{\xi}(t)$$

$$U = \frac{1}{2}kx^2, \quad \langle \xi_\alpha(t)\xi_\beta(t')\rangle = 2\gamma T\delta_{\alpha\beta}\delta(t-t')$$

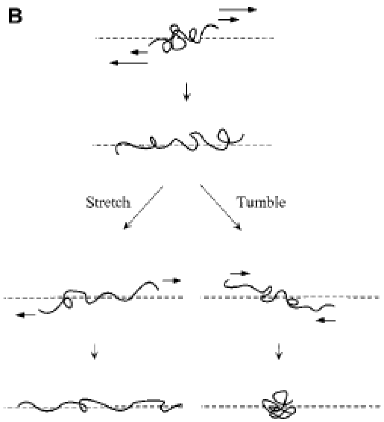
Velocity gradient tensor

$$\mathbf{K}_2 = \begin{pmatrix} 0 & \dot{\gamma} \\ 0 & 0 \end{pmatrix}$$ Simple shear

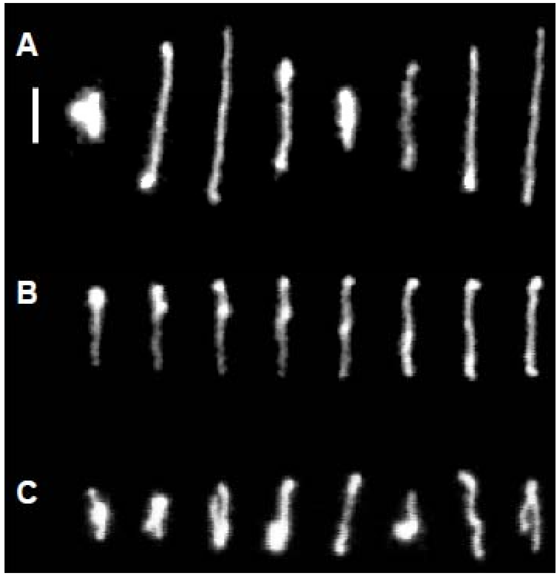
$$\tau_0 = \frac{\gamma}{k}$$:relaxation time

Single-Polymer Dynamics in Shear Flow

λ DNA in shear flow (Wi=19)



5μm

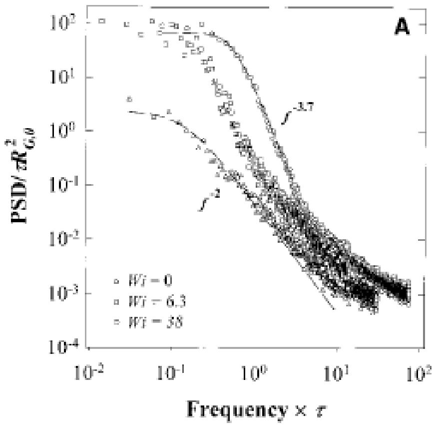


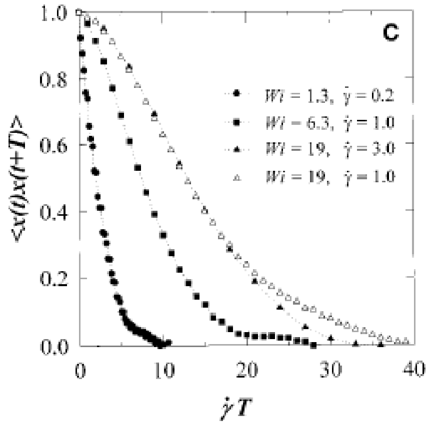
Δt= 6s

Δt= 0.84s

Δt= 6s

(A) Power spectrum and (C) auto-correlation of the extension fluctuation

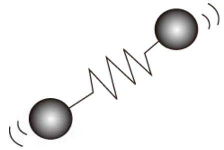




→ Rheology of dilute (and entangled, etc.) polymer solutions

D.E. Smith et. Al., Science (1999)

Dumbbell in thermal equilibrium



Correlation and response function

$$\mathbf{C}(t-t') \equiv \langle \delta\vec{x}(t)\delta\vec{x}(t') \rangle$$

$$\langle \vec{x}(t) \rangle_p - \langle \vec{x}(t) \rangle \equiv \int_{-\infty}^{\infty} \chi(t-t')\vec{f}_p(t')dt'$$

Integrated response

$$\mathbf{R}(t) \equiv \int_0^t ds \quad \chi(s)$$

$$\mathbf{C}(t) = \frac{T}{k} e^{-t/\tau_0} \quad \mathbf{I}$$

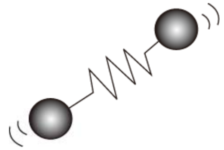
$$\mathbf{R}(t) = \frac{1}{k}\left(1 - e^{-t/\tau_0}\right) \quad \mathbf{I}$$

zz

FDT

$$T\mathbf{X}(t) = -\frac{d}{dt}\mathbf{C}(t) \Leftrightarrow T\mathbf{R}(t) = \mathbf{C}(0) - \mathbf{C}(t)$$

Dumbbell in thermal equilibrium



Correlation and response function

$$\mathsf{C}(t-t') \equiv \langle \delta\vec{x}(t)\delta\vec{x}(t') \rangle$$

$$\langle \vec{x}(t) \rangle_p - \langle \vec{x}(t) \rangle \equiv \int_{-\infty}^{\infty} \chi(t-t')\vec{f}_p(t')dt'$$

Power spectrum

$$\tilde{\mathsf{C}}(\omega) \equiv \int_{-\infty}^{\infty} \mathsf{C}(t)e^{i\omega t}dt$$

Complex admittance

$$\tilde{\chi}(\omega) \equiv \int_{0}^{\infty} \chi(t)e^{i\omega t}dt$$

$$\tilde{\mathsf{C}}(\omega) = \tilde{\mathsf{C}}'(\omega) = \frac{2\tau_0 T}{k\left[1+(\tau_0\omega)^2\right]}\begin{pmatrix}1 & 0\\ 0 & 1\end{pmatrix}$$

$$\tilde{\chi}'(\omega) = \frac{1}{k\left[1+(\tau_0\omega)^2\right]}\begin{pmatrix}1 & 0\\ 0 & 1\end{pmatrix}$$

$$\tilde{\chi}''(\omega) = \frac{\tau_0\omega}{k\left[1+(\tau_0\omega)^2\right]}\begin{pmatrix}1 & 0\\ 0 & 1\end{pmatrix}$$

FDT (1st kind)

$$\omega\tilde{C}(\omega)\Big|_{\alpha\alpha} = 2T\tilde{\chi}''(\omega)\Big|_{\alpha\alpha}$$

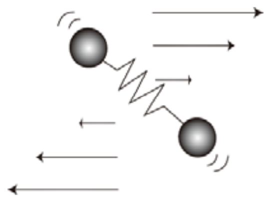
$$\Leftrightarrow \omega\tilde{\mathsf{C}}(\omega) = T\left\{\tilde{\chi}''(\omega) + \left[\tilde{\chi}''(\omega)\right]^T\right\}$$

Kramers-Kronig relation

$$\tilde{\chi}''(\omega) = \frac{1}{\pi}P\int_{-\infty}^{\infty}\frac{\tilde{\chi}'(\omega_1)}{\omega_1-\omega}d\omega_1$$

$$\Leftrightarrow \tilde{\chi}''(\omega) = \tau_0\omega\tilde{\chi}'(\omega)$$

Dumbbell in flow



$$\tau_0 = \gamma / k \,, W = \tau_0 \dot{\gamma}$$

Correlation

$$\mathbf{C}(t) = \frac{T}{k} e^{-t/\tau_0} \begin{pmatrix} 1 + \frac{W^2}{2}\left(1+\frac{t}{\tau_0}\right) & W\left(\frac{1}{2}+\frac{t}{\tau_0}\right) \\ \frac{W}{2} & 1 \end{pmatrix}$$

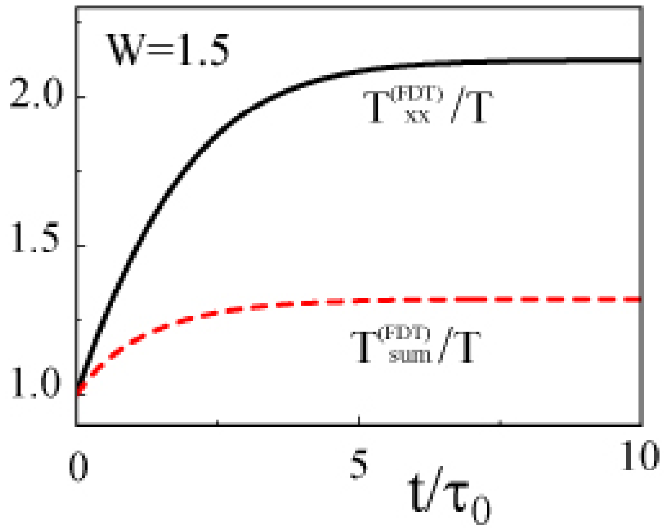
Response

$$\mathbf{R}(t) = \frac{1}{k} \begin{pmatrix} 1-e^{-t/\tau_0} & W\left(1-e^{-t/\tau_0}\left(1+\frac{t}{\tau_0}\right)\right) \\ 0 & 1-e^{-t/\tau_0} \end{pmatrix}$$

Break down of FDT

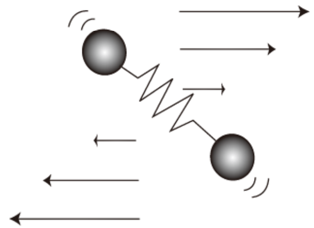
$$\mathbf{T}\mathbf{X}(t) \neq -\frac{d}{dt}\mathbf{C}(t) \Leftrightarrow \mathbf{T}\mathbf{R}(t) \neq \mathbf{C}(0) - \mathbf{C}(t)$$

FDT ratio (T_{eff}) $T_{ij}^{(FDT)}(t) \equiv \dfrac{C_{ij}(0) - C_{ij}(t)}{R_{ij}(t)}$



- Time dependent
- Approaching to static value after long enough time
- Static value $T_{(FDT)}(t \to \infty) > T$

Equality in matrix representation



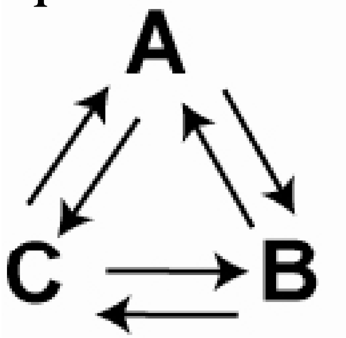
$$\chi_{ij}(t)\Theta_{jk} = -\frac{d}{dt}C_{ik}(t)$$

$$\Theta = T\begin{pmatrix} 1 & -W/2 \\ W/2 & 1 \end{pmatrix}$$

$W \equiv \tau_0 \dot{\gamma}$:Weissenberg number

Matrix representation

Equilibrium state



Detailed balance

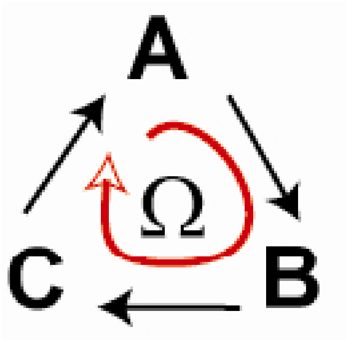
$p(\vec{x}) \sim e^{-U(\vec{x})/T}$: Canonical distribution

$$T\chi_{ij}(t) = -\frac{d}{dt} C_{ij}(t)$$

$$TR_{ij}(t) = C_{ij}(0) - C_{ij}(t)$$

$$R_{ij}(t) = \int_0^t ds \;\; \chi_{ij}(s)$$: integrated response

Non-equilibrium steady state



Circulating flux

$$\vec{j}(\vec{x}) = \mathbf{\Omega}\vec{\nabla} p(\vec{x}) \qquad \Omega_{ij} = -\Omega_{ji}$$

$$\vec{v}(\vec{x}) = \vec{j}(\vec{x}) / p(\vec{x}) = -\mathbf{\Omega}\vec{\nabla}\phi(\vec{x})$$

$p(\vec{x}) \sim e^{-\phi(\vec{x})}$: Steady dist. function

$\phi(\vec{x}) = \frac{1}{2}\sigma_{ij} x_i x_j$: Generalized potential (quadratic)

$$\chi_{ij}(t)\Theta_{jk} = -\frac{d}{dt} C_{ik}(t)$$

$$\Theta_{ij} = \left[\sigma_{ik}\tilde{\chi}_{kj}(0)\right]^{-1}$$

$\tilde{\chi}_{ij}(0) = \int_0^\infty dt \;\; \chi(t)$: Static susceptibility

Θ ??

General equation in Gaussian regime

$$\frac{\partial \vec{x}(t)}{\partial t} = -\mathsf{K}\vec{x}(t) + \vec{\xi}(t) + \vec{v}_p(t)$$

$$\vec{x} = \begin{pmatrix} x_1 \\ x_2 \\ \vdots \\ x_n \end{pmatrix}$$

$$\langle \xi_i(t) \rangle = 0, \quad \langle \xi_i(t)\xi_j(t') \rangle = 2D_{ij}\delta(t-t')$$

Regression matrix: $\mathsf{K} = \mathsf{K}^{(1)} + \mathsf{K}^{(2)}$

$K^{(1)}{}_{ij} x_j = L_{ij} \nabla_j V(\vec{x})$: conservative force

$K^{(2)}{}_{ij}$: non-conservative factor

Linear response analysis

$$\chi_{ij}(t)\Theta_{jk} = -\frac{d}{dt} C_{ik}(t)$$

$$\Theta_{ij} \equiv \left[\sigma_{ik}\tilde{\chi}_{kj}(0)\right]^{-1}$$

$$= L_{ik}^{-1}\left(D_{kj} + \Omega_{kj}\right)$$

$$\sigma_{ij}^{-1} = C_{ij}(0) = K_{ik}^{-1}(D_{kj} + \Omega_{kj})$$

$D_{ij} = TL_{ij}$

(FDT 2nd kind)

$$\Theta_{ij} = T(\delta_{ij} + L_{ik}^{-1}\Omega_{kj})$$

$$T\chi_{ij}(t) = -\frac{d}{dt} C_{ij}(t) - \chi_{ik}(t) L_{kl}^{-1} \Omega_{lj}$$

Deviation from FDT

Ω_{ij} : anti-symmetric part of Onsager coefficient
(irreversible circulation of fluctuation: K. Tomita&H. Tomita, PTP 1974)

Fourier space Expression

Power spectrum

$$\tilde{C}_{ij}(\omega\omega \equiv \int_{-\infty}^{\infty} dt\;\; C_{ij}(t)e^{i\omega\omega}$$

Complex admittance

$$\tilde{\chi}(\omega) \equiv \int_{0}^{\infty} dt\;\; \chi(t)e^{i\omega t}$$

Symmetric part

$$\Delta_{ij}^{(s)}\left[\chi''(\omega)\boldsymbol{\Theta}\right] = \omega\tilde{C}'_{ij}(\omega)$$

Anti-symmetric part

$$\Delta_{ij}^{(as)}\left[\tilde{\mathsf{X}}'(\omega)\boldsymbol{\Theta}\right] = \omega\tilde{C}''_{ji}(\omega)$$

$$\left(\begin{array}{l} \Delta_{ij}^{(s)}[\mathsf{A}] \equiv A_{ij} + A_{ji} \\ \Delta_{ij}^{(as)}[\mathsf{A}] \equiv A_{ij} - A_{ji} \end{array}\right)$$

$$D_{ij} = TL_{ij}$$

(FDT 2^{nd} kind) →

Symmetric part

$$T\Delta_{ij}^{(s)}\left[\tilde{\chi}''(\omega)\right] - \omega\tilde{C}'_{ij}(\omega) = -\Delta_{ij}^{(s)}\left[\tilde{\chi}''(\omega)\mathsf{L}^{-1}\boldsymbol{\Omega}\right]$$

Anti-symmetric part

$$T\Delta_{ij}^{(as)}\left[\tilde{\chi}'(\omega)\right] + \omega\tilde{C}''_{ij}(\omega) = -\Delta_{ij}^{(as)}\left[\tilde{\chi}'(\omega)\mathsf{L}^{-1}\boldsymbol{\Omega}\right]$$

Symmetric part: Ω=0 → L.H.S = 0

FDT 1^{st} kind

Anti-symmetric part: Ω=0 → both terms of L.H.S = 0

- time reversal symmetry
- symmetry of response, i.e., reciprocal relation

$$\tilde{C}''_{ij}(\omega) = \mathrm{Im}\left[\int_{0}^{\infty} dt\;\; [C_{ij}(t) - C_{ij}(-t)]e^{i\omega t}\right]$$

Anti-symmetric part of admittance

~ causality

A simple relation exists between real and imaginary parts of admittance for anti-symmetric part

cf. Kramers-Kronig relation

For a system with 2 gross variables for simplicity

$$\Delta_{12}^{(as)}\left[\tilde{\chi}'(\omega)\boldsymbol{\Theta}\right] = -\omega\tau_{12}\Delta_{12}^{(as)}\left[\tilde{\chi}''(\omega)\mathbf{L}^{-1}\mathbf{D}\right]: \ (\mathbf{K}_2 \neq 0)$$

$$\tau_{12} = \frac{2}{\mathrm{Tr}[\mathbf{K}]}$$

→

$$\tau_{12}\Delta_{12}^{(as)}\left[\tilde{\chi}''(\omega)\mathbf{L}^{-1}\mathbf{D}\right] = \tilde{C}_{12}''(\omega)$$

$$D_{ij} = TL_{ij}$$

(FDT 2nd kind)

→

$$T\tau_{12}\Delta_{12}^{(as)}\left[\tilde{\chi}''(\omega)\right] = \tilde{C}_{12}''(\omega)$$

cf. [equilibrium]

$$T\Delta_{12}^{(s)}\left[\tilde{X}''(\omega)\right] = \omega\tilde{C}_{12}(\omega)$$

A formula connecting asymmetries in fluctuation and response

Summary

Phys. Rev. E **77**, 050102(R) (2008)

\# Linear Response around NESS in general Gaussian systems

\# Departure from equilibrium formula through Ω

\# $\Omega \longleftrightarrow$ frequency moment of $^{(as)}[\chi''(w)] \longleftrightarrow$ house keeping heat

\# Connection between asymmetries in response and fluctuation

\# Applicable to nonlinear systems with normal fluctuations
(cf. system size expansion)