Markovian Approximation and Linear Response Theory for Classical Open Systems

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Contents

- Consider the Langevin equation

$$\ddot{q} = -\nabla V(q) - \gamma \dot{q} + \sqrt{2\gamma\beta^{-1}}\, \dot{W}, \tag{1}$$

- with $V(q)$ being a confining potential (later on we will also consider periodic potentials), γ is the friction coefficient and β the inverse temperature.
- Write it as the first order system

$$dq_t = p_t\, dt, \tag{2a}$$

$$dp_t = -\nabla V(q_t)\, dt - \gamma p_t\, dt + \sqrt{2\gamma\beta^{-1}}\, dW_t. \tag{2b}$$

- The process $(q_t,\, p_t)$ is a Markov process on $\mathbb{R}^d \times \mathbb{R}^d$ (or $\mathbb{T}^d \times \mathbb{R}^d$ for periodic potentials) with generator

$$\mathcal{L} = p \cdot \nabla_q - \nabla_q V(q) \cdot \nabla_p + \gamma(-p \cdot \nabla_p + \beta^{-1} \Delta_p). \tag{3}$$

- The Fokker-Planck operator ($L^2(\mathbb{R}^{2d})-$adjoint of the generator) is

$$\mathcal{L}^* \cdot = -p \cdot \nabla_q \cdot + \nabla_q V \cdot \nabla_p \cdot + \gamma\big(\nabla_p (p \cdot) + \beta^{-1} \Delta_p \cdot \big) \tag{4}$$

- The law of the process at time t (distribution function) $\rho(q, p, t)$ is the solution of the Fokker-Planck equation

$$\frac{\partial \rho}{\partial t} = \mathcal{L}^* \rho, \quad \rho|_{t=0} = \rho_0. \tag{5}$$

- Introduce the notation $X_t^{q,p} := \big(q_t, p_t; q_0 = q, p_0 = p\big)$.
- The expectation of an observable

$$u(q,p,t) = \mathbb{E} f(X_t^{q,p}),$$

can be computed by solving the backward Kolmogorov equation

$$\frac{\partial u}{\partial t} = \mathcal{L} u, \quad u|_{t=0} = f(q,p). \tag{6}$$

- For sufficiently nice potentials the Langevin dynamics (2) is ergodic with respect to the Gibbs measure

$$\mu(dqdp) = \frac{1}{Z} e^{-\beta H(q,p)}\, dqdp =: \rho_\infty(q,p)\, dpdq, \tag{7a}$$

$$Z = \int_{\mathbb{R}^{2d}} e^{-\beta H(q,p)}\, dqdp. \tag{7b}$$

- The density ρ_∞ can be obtained as the solution of the stationary Fokker-Planck equation $\mathcal{L}^* \rho_\infty = 0$.
- Furthermore, we have exponentially fast convergence to equilibrium

$$\|\rho(t,\cdot) - \rho_\infty\| \leq C e^{-\lambda t}, \tag{8}$$

- for positive constants C, λ. See the lecture notes by F. Nier.

- Why use the Langevin equation?
  - As a thermostat, i.e. to sample from the distribution (7) and to also compute time dependent quantities such as correlation functions.
  - As a reduced description of a more complicated system. Think of the physical model of Brownian motion: colloid particle interacting with the molecules of the surrounding fluid. We obtain a closed (stochastic) equation for the dynamics of the Brownian particle by "integrating out" the fluid molecules.
- One of the goals of these lectures is to derive the Langevin equation from a more basic model ("first principles").
- We also want to give a miscroscopic definition of the friction coefficient γ.

- It is often useful (also in experimental set-ups) to probe a system which is at equilibrium by adding a weak external forcing.
- The perturbed Langevin dynamics (assumed to be at equilibrium at $t = 0$) is

$$\ddot{q}^{\epsilon} = -\nabla V(q^{\epsilon}) + \epsilon F(t) - \gamma \dot{q}^{\epsilon} + \sqrt{2\gamma\beta^{-1}}\,\dot{W}. \tag{9}$$

- We are interested in understanding the dynamics in the weak forcing regime $\epsilon \ll 1$. We will do this by developing **linear response theory** (first order perturbation theory).
- We will see that the response of the system to the external forcing is related to the fluctuations at equilibrium.
- A result of this analysis is the derivation of **Green-Kubo formulas** for transport coefficients. For the diffusion coefficient we have (Einstein relation/Green-Kubo)

$$D = \beta^{-1}\mu = \int_0^{+\infty} \langle p_t p_0 \rangle_{eq}\, dt, \tag{10}$$

- where $\mu = \lim_{F \to 0} \frac{V}{F}$, where V is the effective drift.

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DERIVATION OF THE LANGEVIN EQUATION

- We will consider the dynamics of a Brownian particle (”small system”) in contact with its environment (”heat bath”).
- We assume that the environment is at equilibrium at time $t = 0$.
- We assume that the dynamics of the full system is Hamiltonian:

$$H(Q, P; q, p) = H_{BP}(Q, P) + H(q, p) + \lambda H_I(Q, q), \tag{11}$$

- where Q, P are the position and momentum of the Brownian particle, q, p of the environment (they are actually fields) and λ controls the strength of the coupling between the Brownian particle and the environment.

- Let $\mathcal{H}$ denote the phase space of the full dynamics and $X := (P, Q, p, q)$. Our goal is to show that in some appropriate asymptotic limit we can obtain a closed equation for the dynamics of the Brownian particle.
- In other words, we need to find a projection $\mathcal{P} : \mathcal{H} \mapsto \mathbb{R}^{2n}$ so that $\mathcal{P}X$ satisfies an equation that is of the Langevin type (2).
- We can use projection operator techniques at the level of the Liouville equation

$$\frac{\partial \rho}{\partial t} = \{H, \rho\} \tag{12}$$

  associated to the Hamiltonian dynamics (11)–Mori-Zwanzig formalism.
- Projection operator techniques tend to give formal results. Additional approximation/asymptotic calculations are needed.

- We will consider a model for which we can obtain the Langevin equation (2) from the Hamiltonian dynamics (11) in a quite explicit way.
- We will make two basic assumptions:
  1. The coupling between the Brownian particle and the environment is linear.
  2. The Hamiltonian describing the environment is quadratic in positions (and momenta).
- In addition, the environment is much larger than the Brownian particle (i.e. infinite dimensional) and at equilibrium at time $t = 0$.

- We have 4 different levels of description:
  1. The full Hamiltonian dynamics (11).
  2. The generalized Langevin equation (GLE) that we will obtain after eliminating the heat bath variables:

$$\ddot{Q} = -\nabla V(Q) - \int_0^t \gamma(t-s)\dot{Q}(s)\,ds + F(t). \tag{13}$$

  3. The Langevin equation (1) that we will obtain in the rapid decorrelation limit
  4. The overdamped Langevin (Smoluchowski) dynamics that we obtain in the high friction limit

$$\dot{q} = -\nabla V(q) + \sqrt{2\beta^{-1}}\,\dot{W}. \tag{14}$$

- Each reduced level of description includes less information but is easier to analyze.
- For example, we can prove exponentially fast convergence to equilibrium for (1) and (14), something that is not known, without additional assumptions, for (11) and (13).

- The Hamiltonian of the Brownian particle (in one dimension, for simplicity) is described by the Hamiltonian (notice change of notation)

$$H_{BP} = \frac{1}{2}p^2 + V(q), \tag{15}$$

- where V is a confining potential.
- The environment is modeled as a linear the wave equation (with infinite energy):

$$\partial_t^2 \phi(t,x) = \partial_x^2 \phi(t,x). \tag{16}$$

- The Hamiltonian of this system (which is quadratic) is

$$\mathcal{H}_{HB}(\phi, \pi) = \int \left(|\partial_x \phi|^2 + |\pi(x)|^2 \right) dx. \tag{17}$$

- where $\pi(x)$ denotes the conjugate momentum field.
- The initial conditions are distributed according to the Gibbs measure (which in this case is a Gaussian measure) at inverse temperature β, which we formally write as

$$\text{"}\,\mu_\beta = Z^{-1} e^{-\beta \mathcal{H}_{HB}(\phi,\pi)}\, d\phi d\pi\text{"}. \tag{18}$$

- Under this assumption on the initial conditions, typical configurations of the heat bath have infinite energy. In this way, the environment can pump enough energy into the system so that non-trivial fluctuations emerge.
- The coupling between the particle and the field is linear:

$$H_I(Q, \phi) = q \int \partial_x \phi(x) \rho(x)\, dx, \tag{19}$$

- where The function $\rho(x)$ models the coupling between the particle and the field.
- The coupling (19) can be thought of as the first term in a Taylor series expansion of a nonlinear, nonlocal coupling (dipole coupling approximation).
- The Hamiltonian of the particle-field model is

$$H(Q, P, \phi, \pi) = H_{BP}(P, Q) + \mathcal{H}(\phi, \pi) + \lambda H_I(Q, \phi). \tag{20}$$

- Let us first consider a caricature of (20) where there is only one oscillator in the "environment":

$$H(Q,P,q,p) = \frac{P^2}{2} + V(Q) + \left[\left(\frac{p^2}{2} + \frac{1}{2}\omega^2 q^2 \right) - \lambda q Q \right], \tag{21}$$

- Hamilton's equations of motion are:

$$\begin{aligned} \ddot{Q} + V'(Q) &= \lambda q, &(22a)\\ \ddot{q} + \omega^2 (q - \lambda Q) &= 0. &(22b) \end{aligned}$$

- We can solve (22b) using the variation of constants formula. Set $z = (q\ p)^T$, $p = \dot{q}$. Eqn (22b) can be written as

$$\frac{dz}{dt} = Az + \lambda h(t), \tag{23}$$

- where

$$A = \begin{pmatrix} 0 & 1 \\ -\omega^2 & 0 \end{pmatrix} \quad \text{and} \quad h(t) = \begin{pmatrix} 0 \\ Q(t) \end{pmatrix}$$

- The solution of (23) is

$$z(t) = e^{At} z(0) + \lambda \int_0^t e^{A(t-s)} h(s)\, ds.$$

- We calculate

$$e^{At} = \cos(\omega t) I + \frac{1}{\omega} \sin(\omega t) A, \tag{24}$$

- Consequently: where I stands for the 2×2 identity matrix. From this we obtain

$$\begin{aligned} q(t) &= q(0)\cos(\omega t) + \frac{p(0)}{\omega}\sin(\omega t) \\ &\quad + \lambda \frac{1}{\omega} \int_0^t \sin(\omega(t-s)) Q(s)\, ds. \end{aligned} \tag{25}$$

- Now we substitute (25) into (22a) to obtain a closed equation that describes the dynamics of the Brownian particle:

$$\ddot{Q} = -V'(Q) - \lambda^2 \int_0^t D(t-s)Q(s)\, ds + \lambda F(t), \tag{26}$$

- where

$$D(t) = -\frac{1}{\omega} \sin(\omega t), \tag{27}$$

- and

$$F(t) = q(0)\cos(\omega t) + \frac{p(0)}{\omega} \sin(\omega t). \tag{28}$$

- Since $q(0)$ and $p(0)$ are Gaussian random variables with mean 0 and $\langle q(0)^2 \rangle = \beta^{-1}\omega^{-2}$, $\langle p(0)^2 \rangle = \beta^{-1}$, $\langle q(0)p(0) \rangle = 0$ we have

$$\langle F(t)F(s) \rangle = \beta^{-1}\omega^{-2} \cos(\omega(t-s)) =: \beta^{-1} C(t-s).$$

- Consequently,

$$D(t) = \dot{C}(t). \tag{29}$$

- This is a form of the **fluctuation-dissipation theorem.**

- We perform an integration by parts in (25):

$$
\begin{aligned}
q(t) &= \left(q(0) - \frac{\lambda}{\omega^2} Q(0) \right) \cos(\omega t) + \frac{p(0)}{\omega} \sin(\omega t) \\
&\quad + \lambda \frac{1}{\omega^2} Q(t) - \lambda \frac{1}{\omega^2} \int_0^t \cos(\omega(t-s)) \dot{Q}(s)\, ds.
\end{aligned}
$$

- We substitute this in equation (22a) to obtain the **Generalized Langevin Equation** (GLE)

$$
\ddot{Q} = -V_{\text{eff}}(Q) - \lambda^2 \int_0^t \gamma(t-s) \dot{Q}(s)\, ds + \lambda F(t), \tag{30}
$$

- where $V_{\text{eff}}(Q) = V(Q) - \frac{\lambda^2}{2\omega^2} Q^2$,

$$
\gamma(t) = \frac{1}{\omega^2} \cos(\omega t), \tag{31a}
$$

$$
F(t) = \left[\left(q(0) - \frac{\lambda}{\omega^2} Q(0) \right) \cos(\omega t) + \frac{p(0)}{\omega} \sin(\omega t) \right]. \tag{31b}
$$

- The same calculation can be done for an arbitrary number of harmonic oscillators in the environment.
- When writing the dynamics of the Brownian particle in the form (30) we need to introduce an effective potential.
- We have assumed that the initial conditions of the Brownian particle are deterministic, independent of the initial distribution of the environment. i.e. the environment is initially at equilibrium *in the absence* of the Brownian particle.
- We can also assume that the environment is initially in equilibrium *in the presence* of the distinguished particle, i.e. that the initial positions and momenta of the heat bath particles are distributed according to a Gibbs distribution, conditional on the knowledge of $\{Q_0, \, P_0\}$:

$$\mu_\beta(dpdq) = Z^{-1} e^{-\beta H_{\text{eff}}(q,p,Q)} \, dqdp, \tag{32}$$

- where

$$H_{\text{eff}}(q,p,Q) = \left[\frac{p^2}{2} + \frac{1}{2}\omega^2 \left(q - \frac{\lambda}{\omega^2} Q \right)^2 \right]. \tag{33}$$

- This assumption implies that

$$q(0) = \frac{\lambda}{\omega^2} Q_0 + \sqrt{\beta^{-1}\omega^{-2}}\,\xi, \quad p(0) = \sqrt{\beta^{-1}}\,\eta, \tag{34}$$

- where the ξ, η are independent $\mathcal{N}(0,1)$ random variables.
- This assumption ensures that the forcing term in (30) is mean zero.
- The fluctuation-dissipation theorem takes the form

$$\langle F(t)F(s)\rangle = \beta^{-1}\gamma(t-s). \tag{35}$$

- We can perform the same calculation for the coupled particle-field model (20). We obtain

$$\ddot{Q} = -V'(Q) - \int_0^t D(t-s)Q(s)\,ds + \langle \phi_0, e^{-\mathcal{L}t}\alpha \rangle, \tag{36}$$

- where

$$A = \begin{pmatrix} 0 & 1 \\ \partial_x^2 & 0 \end{pmatrix},$$

- $\langle f, h \rangle = \int (\partial_x f_1 \partial_x h_1 + f_2 h_2)\,dx$, $f = (f_1, f_2)$ and $\hat{\alpha}(k) = \big(-ik\hat{\rho}(k)/k^2, 0\big)$ in Fourier space.
- Furthermore

$$D(t) = \langle e^{\mathcal{L}t}\mathcal{L}\alpha, \alpha \rangle = \dot{C}(t).$$

- $C(t)$ is the covariance of the Gaussian noise process

$$F(t) = \langle \phi_0, e^{-\mathcal{L}t}\alpha \rangle.$$

- In particular:

$$\mathbb{E}(F(t)F(s)) = \beta^{-1}C(t-s) = \beta^{-1}\int |\hat{\rho}(k)|^2 e^{ikt}\,dk.$$

- The spectral density of the autocorrelation function of the noise process is the square of the Fourier transform of the density $\rho(x)$ which controls the coupling between the particle and the environment.
- The GLE (36) is equivalent to the original infinite dimensional Hamiltonian system with random initial conditions.
- Proving ergodicity, convergence to equilibrium etc for (36) is equivalent to proving ergodicity and convergence to equilibrium for the infinite dimensional Hamiltonian system.

- For general coupling functions $\rho(x)$ the GLE (36) describes a non-Markovian system. It can be represented as a Markovian system only if we add an infinite number of additional variables.
- However, for appropriate choices of the coupling function $\rho(x)$ the GLE (36) is equivalent to a Markovian process in a **finite dimensional** extended phase space.

Definition

We will say that a stochastic process X_t is *quasi-Markovian* if it can be represented as a Markovian stochastic process by adding a finite number of additional variables: There exists a stochastic process Y_t so that $\{X_t, \ Y_t\}$ is a Markov process.

Proposition

Assume that $p(k) = \sum_{m=1}^{M} c_m(-ik)^m$ is a polynomial with real coefficients and roots in the upper half plane then the Gaussian process with spectral density $|p(k)|^{-2}$ is the solution of the linear SDE

$$\left(p\left(-i\frac{d}{dt}\right) x_t \right) = \frac{dW_t}{dt}. \tag{37}$$

Proof.

The solution of (37) is

$$x_t = \int_{-\infty}^{t} k(t-s)\, dW(s), \quad k(t) = \frac{1}{\sqrt{2\pi}} \int e^{ikt} \frac{1}{p(k)}\, dk.$$

We compute

$$\mathbb{E}(x_t x_s) = \int e^{ik(t-s)} \frac{1}{|p(k)|^2}\, dk.$$

□

Example

Take $p(k) \sim (ik + \alpha)$. The spectral density is

$$|\hat{\rho}(k)|^2 = \frac{\alpha}{\pi} \frac{1}{\pi^2 + \alpha^2}.$$

The autocorrelation function is

$$C(t) = \frac{\alpha}{\pi} \int e^{ikt} \frac{1}{k^2 + \alpha^2} \, dk = e^{-|\alpha| t}.$$

The linear SDE is

$$dx_t = -\alpha x_t \, dt + \sqrt{2\alpha} dW_t.$$

- Consider the GLE

$$\ddot{Q} = -V'(Q) - \lambda^2 \int_0^t \gamma(t-s)\dot{Q}(s)\,ds + \lambda F(t), \tag{38}$$

- with $\langle F(t)F(s)\rangle = \beta^{-1}\gamma(t-s) = \beta^{-1}e^{-\alpha(t-s)}$.
- $F(t)$ is the stationary Ornstein-Uhlenbeck process:

$$\frac{dF}{dt} = -\alpha F + \sqrt{2\beta^{-1}\alpha}\frac{dW}{dt}, \quad \text{with } F(0) \sim \mathcal{N}(0, \beta^{-1}). \tag{39}$$

- We can rewrite (38) as a system of SDEs:

$$\begin{aligned}
\frac{dQ}{dt} &= P, \\
\frac{dP}{dt} &= -V'(Q) + \lambda Z, \\
\frac{dZ}{dt} &= -\alpha Z - \lambda P + \sqrt{2\alpha\beta^{-1}}\frac{dW}{dt},
\end{aligned}$$

where $Z(0) \sim \mathcal{N}(0, \beta^{-1})$.

- The process $\{Q(t),\, P(t),\, Z(t)\} \in \mathbb{R}^3$ is Markovian.
- It is a degenerate Markov process: noise acts directly only on one of the 3 degrees of freedom.
- The generator of this process is

$$\mathcal{L} = p\partial_q + \big(-V'(q) + \lambda z\big)\partial_p - \big(\alpha z + \lambda p\big)\partial_z + \alpha\beta^{-1}\partial_z^2.$$

- It is a hypoelliptic and hypocoercive operator.

- Rescale $\lambda \to \frac{\lambda}{\epsilon}$, $\alpha \to \frac{\alpha}{\epsilon^2}$
- Eqn. (40) becomes

$$
\begin{aligned}
dq^\epsilon &= p^\epsilon \, dt, & \text{(41a)}\\
dp^\epsilon &= -V'(q^\epsilon)\, dt + \frac{\lambda}{\epsilon} z^\epsilon \, dt, & \text{(41b)}\\
dz^\epsilon &= -\frac{\alpha}{\epsilon^2} z^\epsilon \, dt - \frac{\lambda}{\epsilon} p^\epsilon \, dt + \sqrt{\frac{2\alpha\beta^{-1}}{\epsilon^2}}\, dW, & \text{(41c)}
\end{aligned}
$$

- In the limit as $\epsilon \to 0$ we obtain the Langevin equation for $q^\epsilon(t)$, $p^\epsilon(t)$.

Proposition

Let $\{q^\epsilon(t), p^\epsilon(t), \mathbf{z}^\epsilon(t)\}$ *on* $\mathbb{R}^3$ *be the solution of* (41) *with* $V(q) \in C^\infty(\mathbb{T})$ *with stationary initial conditions. Then* $\{q^\epsilon(t), p^\epsilon(t)\}$ *converge weakly to the solution of the Langevin equation*

$$dq = p\,dt, \quad dp = -V'(q)\,dt - \gamma p\,dt + \sqrt{2\gamma\beta^{-1}}dW, \tag{42}$$

where the friction coefficient is given by the formula

$$\gamma = \frac{\lambda^2}{\alpha}. \tag{43}$$

- We have derived the (generalized) Langevin equation from a "particle + field" model where the field is at equilibrium at $t = 0$.
- The model that we can considered is very specific since the field and the coupling are linear.
- The GLE can be derived (at least formally) for more general Hamiltonian systems using the Mori-Zwanzig formalism (projection operator techniques for the Liouville equation).
- The Markovian approximation of the GLE leads to a finite dimensional hypoelliptic diffusion.
- Conjecture (F. Nier): Assume that

  $$\mathcal{L} = -\nabla V \cdot \nabla + \beta^{-1} \Delta$$

  has compact resolvent. Then so does

  $$\mathcal{L} == p \cdot \nabla_q - \nabla_q V \cdot \nabla_p + \gamma(-p \cdot \nabla_p + \beta^{-1} \Delta_p).$$

- Formulate a similar conjecture for all Markovian approximations of the GLE.

LINEAR RESPONSE THEORY

- Let X_t denote a stationary dynamical system with state space $\mathbb{X}$ and invariant measure $\mu(dx) = f_\infty(x)\,dx$.
- We probe the system by adding a time dependent forcing $\epsilon F(t)$ with $\epsilon \ll 1$ at time t_0.
- Goal: calculate the distribution function $f^\epsilon(x,t)$ of the perturbed systems X_t^ϵ, $\epsilon \ll 1$, in particular in the long time limit $t \to +\infty$.
- We can then calculate expectation value of observables as well as correlation functions.

- We assume that the distribution function $f^\epsilon(x,t)$ satisfies a linear kinetic equation e.g. the Liouville or the Fokker-Planck equation:

$$\frac{\partial f^\epsilon}{\partial t} = \mathcal{L}^{*\,\epsilon} f^\epsilon, \tag{44a}$$

$$f^\epsilon\Big|_{t=t_0} = f_\infty. \tag{44b}$$

- The choice of the initial conditions reflects the fact that at $t = t_0$ the system is at equilibrium.
- Since f_∞ is the unique equilibrium distribution, we have that

$$\mathcal{L}_0^* f_\infty = 0. \tag{45}$$

- The operator $\mathcal{L}^{*\,\epsilon}$ can be written in the form

$$\mathcal{L}^{*\,\epsilon} = \mathcal{L}_0^* + \epsilon \mathcal{L}_1^*, \tag{46}$$

- where cL_0^* denotes the Liouville or Fokker-Planck operator of the unperturbed system and $\mathcal{L}_1^*$ is related to the external forcing.
- We will assume that $\mathcal{L}_1$ is of the form

$$\mathcal{L}_1^* = F(t) \cdot D, \tag{47}$$

- where D is some linear (differential) operator.

Example

(A deterministic dynamical system). Let X_t be the solution of the differential equation

$$\frac{dX_t}{dt} = h(X_t), \tag{48}$$

on a (possibly compact) state space $\mathbb{X}$. We add a weak time dependent forcing to obtain the dynamics

$$\frac{dX_t}{dt} = h(X_t) + \epsilon F(t). \tag{49}$$

We assume that the unperturbed dynamics has a unique invariant distribution f_∞ which is the solution of the stationary Liouville equation

$$\nabla \cdot \Big(h(x) f_\infty \Big) = 0, \tag{50}$$

equipped with appropriate boundary conditions.

Example

The operator $\mathcal{L}^{*\,\epsilon}$ in (46) has the form

$$\mathcal{L}^{*\,\epsilon} \cdot = -\nabla \cdot \Big(h(x) \cdot \Big) - \epsilon F(t) \cdot \nabla \cdot .$$

In this example, the operator D in (47) is $D = -\nabla$.

- A particular case of a deterministic dynamical system of the form (48), and the most important in statistical mechanics, is that of an N-body Hamiltonian system.

Example

(A stochastic dynamical system). Let X_t be the solution of the stochastic differential equation

$$dX_t = h(X_t)\,dt + \sigma(X_t)\,dW_t, \tag{51}$$

on $\mathbb{R}^d$, where $\sigma(x)$ is a positive semidefinite matrix and where the Itô interpretation is used. We add a weak time dependent forcing to obtain the dynamics

$$dX_t = h(X_t)\,dt + \epsilon F(t)\,dt + \sigma(X_t)\,dW_t. \tag{52}$$

We assume that the unperturbed dynamics has a unique invariant distribution f_∞ which is the solution of the stationary Fokker-Planck equation

$$-\nabla \cdot \Big(h(x) f_\infty\Big) + \frac{1}{2} D^2 : \Big(\Sigma(x) f_\infty\Big) = 0, \tag{53}$$

where $\Sigma(x) = \sigma(x)\sigma^T(x)$.

Example

The operator $\mathcal{L}^{*\,\epsilon}$ in (46) has the form

$$\mathcal{L}^{*\,\epsilon}\cdot = -\nabla\cdot\Big(h(x)\,\cdot\Big) + \frac{1}{2}D^2 : \Big(\Sigma(x)\,\cdot\Big) - \epsilon F(t)\cdot\nabla\text{:}$$

As in the previous example, the operator D in (47) is $D = -\nabla$.

- A particular case of Example 5 is the Langevin equation:

$$\ddot{q} = -\nabla V(q) + \epsilon F(t) - \gamma\dot{q} + \sqrt{2\gamma\beta}\,\dot{W}. \tag{54}$$

- We can also add a forcing term to the noise in (51), i.e. we can consider a time-dependent temperature. For the Langevin equation the perturbed dynamics is

$$\ddot{q} = -\nabla V(q) + \epsilon F(t) - \gamma\dot{q} + \sqrt{2\gamma\beta^{-1}(1+\epsilon T(t))}\,\dot{W}, \tag{55}$$

  with $1+\epsilon T(t) > 0$. in this case the operator $\mathcal{L}_1^*$ is

$$\mathcal{L}_1^* = -F(t)\cdot\nabla_p + \gamma\beta^{-1}T(t)\Delta_p,$$

  where $p = \dot{q}$.

- We proceed with the analysis of (44). We look for a solution in the form of a power series expansion in ϵ:

$$f^{\epsilon} = f_0 + \epsilon f_1 + \dots .$$

- We substitute this into (44a) and use the initial condition (44b) to obtain the equations

$$\frac{\partial f_0}{\partial t} = \mathcal{L}_0^* f_0, \quad f_0\big|_{t=0} = f_\infty, \tag{56a}$$

$$\frac{\partial f_1}{\partial t} = \mathcal{L}_0^* f_1 + \mathcal{L}_1^* f_0, \quad f_1\big|_{t=0} = 0. \tag{56b}$$

- The only solution to (56a) is

$$f_0 = f_\infty .$$

- We use this into (56b) and use (47) to obtain

$$\frac{\partial f_1}{\partial t} = \mathcal{L}_0^* f_1 + F(t) \cdot D f_\infty, \quad f_1\big|_{t=0} = 0.$$

- We use the variation of constants formula to solve this equation:

$$f_1(t) = \int_{t_0}^{t} e^{\mathcal{L}_0^*(t-s)} F(s) \cdot Df_\infty \, ds. \tag{57}$$

- Now we can calculate the deviation in the expectation value of an observable due to the external forcing: Let $\langle \cdot \rangle_{eq}$ and $\langle \cdot \rangle$ denote the expectation value with respect to f_∞ and f^ϵ, respectively.
- Let $A(\cdot)$ be an observable and denote by $A(t)$ the deviation of its expectation value from equilibrium, to leading order:

$$\begin{aligned} A(t) &:= \langle A(X_t) \rangle - \langle A(X_t) \rangle_{eq} \\ &= \int A(x) \big(f^\epsilon(x,t) - f_{eq}(x) \big) \, dx \\ &= \epsilon \int A(x) \left(\int_{t_0}^{t} e^{\mathcal{L}_0^*(t-s)} F(s) \cdot Df_\infty \, ds \right) dx. \end{aligned}$$

- Assuming now that we can interchange the order of integration we can rewrite the above formula as

$$
\begin{aligned}
A(t) &= \epsilon \int A(x) \left(\int_{t_0}^{t} e^{\mathcal{L}_0^*(t-s)} F(s) \cdot Df_\infty \, ds \right) dx \\
&= \epsilon \int_{t_0}^{t} \left(\int A(x) e^{\mathcal{L}_0^*(t-s)} \cdot Df_\infty \, dx \right) ds \\
&=: \epsilon \int_{t_0}^{t} R_{\mathcal{L}_0, A}(t-s) F(s) \, ds, \qquad (58)
\end{aligned}
$$

- where we have defined the *response function*

$$
R_{\mathcal{L}_0, A}(t) = \int A(x) e^{\mathcal{L}_0^* t} \cdot Df_\infty \, dx \qquad (59)
$$

- We set now the lower limit of integration in (58) to be $t_0 = -\infty$ (define $R_{\mathcal{L}_0, A}(t)$ in (59) to be 0 for $t < 0$) and assume that we can extend the upper limit of integration to $+\infty$ to write

$$
A(t) = \epsilon \int_{-\infty}^{+\infty} R_{\mathcal{L}_0, A}(t-s) F(s) \, ds. \qquad (60)
$$

- As expected (since we have used linear perturbation theory), the deviation of the expectation value of an observable from its equilibrium value is a linear function of the forcing term.
- (60) has the form of the solution of a linear differential equation with $R_{\mathcal{L}_0,A}(t)$ playing the role of the Green's function. If we consider a delta-like forcing at $t = 0$, $F(t) = \delta(t)$, then the above formula gives
$$A(t) = \epsilon R_{\mathcal{L}_0,A}(t).$$
- Thus, the response function gives the deviation of the expectation value of an observable from equilibrium for a delta-like force.
- Consider a constant force that is exerted to the system at time $t = 0$, $F(t) = F\Theta(t)$ where $\Theta(t)$ denotes the Heaviside step function. For this forcing (58) becomes

$$A(t) = \epsilon F \int_0^t R_{\mathcal{L}_0,A}(t-s)\, ds. \tag{61}$$

- There is a close relation between the response function (59) and stationary autocorrelation functions.
- Let X_t be a stationary Markov process in $\mathbb{R}^d$ with generator $\mathcal{L}$ and invariant distribution f_∞.
- Let $A(\cdot)$ and $B(\cdot)$ be two observables.
- The stationary autocorrelation function $\langle A(X_t)B(X_0)\rangle_{eq}$ can be calculated as follows

$$
\begin{aligned}
\kappa_{A,B}(t) &:= \langle A(X_t)B(X_0)\rangle_{eq} \\
&= \int\int A(x)B(x_0)p(x,t|x_0,0)f_\infty(x_0)\,dxdx_0 \\
&= \int\int A(x)B(x_0)e^{\mathcal{L}^*t}\delta(x-x_0)f_\infty(x_0)\,dxdx_0 \\
&= \int\int e^{\mathcal{L}t}A(x)B(x_0)\delta(x-x_0)f_\infty(x_0)\,dxdx_0 \\
&= \int e^{\mathcal{L}t}A(x)B(x)f_\infty(x)\,dx,
\end{aligned}
$$

where $\mathcal{L}$ acts on functions of x.

- Thus we have established the formula

$$\kappa_{A,B}(t) = \left\langle S_t A(x), B(x) \right\rangle_{f_\infty}, \tag{62}$$

- where S_t denotes the semigroup generated by $\mathcal{L}$ and $\left\langle \cdot, \cdot \right\rangle_{f_\infty}$ denotes the L^2 inner product weighted by the invariant distribution of the diffusion process.
- Consider now the particular choice $B(x) = f_\infty^{-1} D f_\infty$. We combine (59) and (62) to deduce

$$\kappa_{A, f_\infty^{-1} D f_\infty}(t) = R_{\mathcal{L}_0, A}(t). \tag{63}$$

- This is a version of the **fluctuation-dissipation theorem**.

Example

- Consider the Langevin dynamics with an external forcing F.

$$dq = p\,dt, \quad dp = -V'(q)\,dt + F\,dt - \gamma p\,dt + \sqrt{2\gamma\beta^{-1}}\,dW_t.$$

- We have $D = -\partial_p$ and

$$B = f_\infty^{-1} D f_\infty = \beta p.$$

- We use (63) with $A = p$:

$$\beta \langle p(t) p(0) \rangle_{eq} = R_{\mathcal{L}_0, p}(t).$$

Example (continued)

- When the potential is harmonic, $V(q) = \frac{1}{2}\omega_0^2 q^2$, we can compute explicitly the response function and, consequently, the velocity autocorrelation function at equilibrium:

$$R_{q,\mathcal{L}}(t) = \frac{1}{\omega_1} e^{-\frac{\gamma t}{2}} \sin(\omega_1 t), \quad \omega_1 = \sqrt{\omega_0^2 - \frac{\gamma^2}{4}}$$

- and

$$R_{p,\mathcal{L}}(t) = e^{-\frac{\gamma t}{2}} \left(\cos(\omega_1 t) - \frac{\gamma}{2\omega_1} \sin(\omega_1 t) \right).$$

- Consequently:

$$\langle p(t) p(0) \rangle_{eq} = \beta^{-1} e^{-\frac{\gamma t}{2}} \left(\cos(\omega_1 t) - \frac{\gamma}{2\omega_1} \sin(\omega_1 t) \right).$$

- Similar calculations can be done for all linear SDEs.

Example

- Consider the Langevin dynamics with a perturbation in the temperature

$$dq = p\,dt, \quad dp = -V'(q)\,dt + F\,dt - \gamma p\,dt + \sqrt{2\gamma\beta^{-1}(1+F)}\,dW_t.$$

- We have $D = \gamma\beta^{-1}\partial_p^2$ and

$$B = f_\infty^{-1} D f_\infty = \gamma\beta(p^2 - \beta^{-1}).$$

- Let $H(p,q) = p^2/2 + V(q)$ denote the total energy. We have

$$f_\infty^{-1}\mathcal{L}_0^* H(p,q) f_\infty = \mathcal{L}_0 H(p,q) = \gamma(-p^2 + \beta^{-1}).$$

- Consequently (using (62)): $\kappa_{A, f_\infty^{-1} D f_\infty}(t) = -\beta \frac{d}{dt}\kappa_{A,H}(t)$.
- For $A = H$ we obtain $R_{H,\mathcal{L}}(t) = -\beta\frac{d}{dt}\langle H(t)H(0)\rangle_{eq}$.

- We calculate the long time limit of $A(t)$ when the external forcing is a step function.
- The following formal calculations can be justified e.g. for reversible diffusions using functional calculus.

$$
\begin{aligned}
\int_0^t R_{\mathcal{L},A}(t-s)\,ds &= \int_0^t \int A(x) e^{\mathcal{L}_0(t-s)} Df_\infty \,dx\,ds \\
&= \int \int_0^t \left(e^{\mathcal{L}(t-s)} A(x)\right) Df_\infty \,ds\,dx \\
&= \int \left(e^{\mathcal{L}_0 t} \int_0^t e^{\mathcal{L}(-s)}\,ds A(x)\right) Df_\infty \,dx \\
&= \int \left(e^{\mathcal{L}_0 t}(-\mathcal{L}_0)^{-1}\left(e^{\mathcal{L}_0(-t)} - I\right) A(x)\right) Df_\infty \,dx \\
&= \int \left(\left(I - e^{\mathcal{L}_0 t}\right)(-\mathcal{L})^{-1} A(x)\right) Df_\infty \,dx
\end{aligned}
$$

- Assuming now that $\lim_{t\to+\infty} e^{\mathcal{L}t} = 0$ (again, think of reversible diffusions) we have that

$$D := \lim_{t\to+\infty}\int_0^t R_{\mathcal{L},A}(t-s)\,ds = \int (-\mathcal{L})^{-1}A(x)Df_\infty\,dx.$$

- Using this in (61) and relabeling $\epsilon F \mapsto F$ we obtain

$$\lim_{F\to 0}\lim_{t\to+\infty}\frac{A(t)}{F} = \int (-\mathcal{L})^{-1}A(x)Df_\infty\,dx. \tag{64}$$

Remark

- *At least formally, we can interchange the order with which we take the limits in* (64).
- *Formulas of the form* (64) *enable us to calculate transport coefficients, such as the diffusion coefficient.*
- *We can rewrite the above formula in the form*

$$\lim_{F \to 0} \lim_{t \to +\infty} \frac{A(t)}{F} = \int \phi D f_{\infty} \, dx, \tag{65}$$

- *where* ϕ *is the solution of the Poisson equation (when* $A(x)$ *is mean zero)*

$$-\mathcal{L}\phi = A(x), \tag{66}$$

- *equipped with appropriate boundary conditions.*
- *This is precisely the formalism that we obtain using homogenization theory.*

- Consider the Langevin dynamics in a periodic or random potential.

$$dq_t = p_t \, dt, \quad dp_t = -\nabla V(q_t) \, dt - \gamma p_t \, dt + \sqrt{2\gamma\beta^{-1}} \, dW.$$

- From Einstein's formula (10) we have that the diffusion coefficient is related to the mobility according to

$$D = \beta^{-1} \lim_{F \to 0} \lim_{t \to +\infty} \frac{\langle p_t \rangle}{F}$$

- where we have used $\langle p_t \rangle_{eq} = 0$.
- We use now (65) with $A(t) = p_t, \ D = -\nabla_p, \ f_\infty = \frac{1}{Z} e^{-\beta H(q,p)}$ to obtain

$$D = \int \int \phi p f_\infty \, dp dq = \langle -\mathcal{L}\phi, \phi \rangle_{f_\infty}, \tag{67}$$

- which is the formula obtained from homogenization theory (e.g. Kipnis and Varadhan 1985, Rodenhausen 1989).

- Consider the unperturbed dynamics

$$dq = p\,dt, \quad dp = -V'(q)\,dt - \gamma p\,dt + \sqrt{2\gamma\beta^{-1}}\,dW, \tag{68}$$

- where V is a smooth periodic potential. Then the rescaled process

$$q^{\epsilon}(t) := \epsilon q(t/\epsilon^2),$$

- Converges to a Brownian motion with diffusion coefficient

$$D = \int\int ((-\mathcal{L})^{-1}p)pf_{\infty}\,dpdq. \tag{69}$$

- A similar result can be proved when V is a random potential.

- Consider now the perturbed dynamics

$$dq_F = p_F\, dt, \quad dp_F = -V'(q_F)\, dt + F\, dt - \gamma p_F\, dt + \sqrt{2\gamma\beta^{-1}}\, dW. \tag{70}$$

- At least for F sufficiently small, the dynamics is ergodic on $\mathbb{T} \times \mathbb{R}$ with a smooth invariant density $f_\infty^F(p,q)$ which is a differentiable function of F.
- The external forcing induces an effective drift

$$V_F = \int \int p f_\infty^F\, dpdq. \tag{71}$$

- The mobility is defined as

$$\mu := \frac{d}{dF} V_F \Big|_{F=0}. \tag{72}$$

Theorem (Rodenhausen J. Stat. Phys. 1989)

The mobility is well defined by (72) *and*

$$D = \beta^{-1}\mu. \tag{73}$$

Proof.

- Ergodic theory for hypoelliptic diffusions.
- Study of the Poisson and stationary Fokker-Planck equations.
- Girsanov's formula.

$\square$

- Upon combining (63) with (64) we obtain

$$D = \lim_{t \to +\infty} \int_0^t \kappa_{A, f_\infty^{-1} D f_\infty}(t - s)\, ds. \tag{74}$$

- Thus, a transport coefficient can be computed in terms of the time integral of an appropriate autocorrelation function.
- This is an example of the **Green-Kubo formula**

- We can obtain a more general form of the Green-Kubo formalism.
- We define the generalized drift and diffusion coefficients as follows :

$$V^f(x) = \lim_{h\to 0} \frac{1}{h}\mathbb{E}\Big(f(X_h) - f(X_0)\Big| X_0 = x\Big) = \mathcal{L}f \tag{75}$$

- and

$$\begin{aligned} D^{f,g}(x) \quad &:= \quad \lim_{h\to 0} \frac{1}{h}\mathbb{E}\Big((f(X_{t+h}) - f(X_t))((g(X_{t+h}) - g(X_t))\Big| X_t = x\Big) \\ &= \quad \mathcal{L}(fg)(x) - (g\mathcal{L}f)(x) - (f\mathcal{L}g)(x), \end{aligned} \tag{76}$$

- where f, g are smooth functions (in fact, all we need is $f,\, g \in D(\mathcal{L})$ and $fg \in D(\mathcal{L}_0)$).
- The equality in (75) follows from the definition of the generator of a diffusion process.
- We will prove the equality in (75).
- Sometimes $D^{f,g}(x)$ is called the *opérateur carré du champ* .

- To prove (76), notice first that, by stationarity, it is sufficient to prove it at $t = 0$. Furthermore

$$
\begin{aligned}
(f(X_h) - f(X_0))\,(g(X_h) - g(X_0)) \quad = \quad & (fg)(X_h) - (fg)(X_0) \\
& -f(X_0)(g(X_h) - g(X_0)) \\
& -g(X_0)(f(X_h) - f(X_0)).
\end{aligned}
$$

- Consequently:

$$
\begin{aligned}
D^{f,g}(x) \quad := \quad & \lim_{h\to 0} \frac{1}{h}\mathbb{E}\Big((f(X_h) - f(X_0))((g(X_h) - g(X_0))\Big| X_0 = x\Big) \\
= \quad & \lim_{h\to 0} \frac{1}{h}\mathbb{E}\Big((fg)(X_h) - (fg)(X_0)\Big| X_0 = x\Big) \\
& - \lim_{h\to 0} \frac{1}{h}\mathbb{E}\Big(g(X_h) - g(X_0)\Big| X_0 = x\Big) f(x) \\
& - \lim_{h\to 0} \frac{1}{h}\mathbb{E}\Big(f(X_h) - f(X_0)\Big| X_0 = x\Big) g(x) \\
= \quad & \mathcal{L}(fg)(x) - (g\mathcal{L}f)(x) - (f\mathcal{L}g)(x).
\end{aligned}
$$

We have the following result.

Theorem

(The Green-Kubo formula) *Let X_t be a stationary reversible diffusion process with state space $\mathbb{X}$, generator $\mathcal{L}$, invariant measure $\mu(dx)$ and let $V^f(x)$, $D^{f,g}(x)$. Then*

$$\frac{1}{2}\int D^{f,g}\mu(dx) = \int_0^\infty \mathbb{E}\Big(V^f(X_t)V^g(X_0)\Big)\,dt. \tag{77}$$

Proof.

- Let $(\cdot,\cdot)_\mu$ denote the inner product in $L^2(\mathbb{X},\mu)$. We note that

$$\frac{1}{2}\int D^{f,g}\mu(dx) = (-\mathcal{L}f, g)_\mu, \tag{78}$$

- In view of (78), formula (77) becomes

$$(-\mathcal{L}f, g)_\mu = \int_0^\infty \mathbb{E}\Big(V^f(X_t)V^g(X_0)\Big)\, dt. \tag{79}$$

- Now we use (62), together with the identity $\int_0^\infty e^{\mathcal{L}t}\cdot\, dt = (-\mathcal{L})^{-1}$ to obtain

$$\int_0^\infty \kappa_{A,B}(t)\, dt = ((-\mathcal{L})^{-1}A, B)_\mu.$$

- We set now $A = V^f = \mathcal{L}f$, $B = V^g = \mathcal{L}g$ in the above formula to deduce (79) from which (77) follows.

□

Remark

- *The above formal calculation can also be performed in the nonreversible case to obtain*

$$\frac{1}{2}\int D^{f,f}\mu(dx) = \int_0^\infty \mathbb{E}\Big(V^f(X_t)V^f(X_0)\Big)\,dt.$$

- *In the reversible case (i.e. $\mathcal{L}$ being a selfadjoint operator in $\mathcal{H} := L^2(\mathbb{X},\mu)$) the formal calculations can be justified using functional calculus.*
- *For the reversible diffusion process*

$$dX_t = -\nabla V(X_t)\,dt + \sqrt{2\beta^{-1}}\,dW_t$$

and under appropriate assumptions on the potential, the generator $\mathcal{L}$ has compact resolvent and its eigenfunctions form an orthonormal basis in $\mathcal{H}$. In this case the proof of (11) *becomes quite simple.*

- The spectral representation of $\mathcal{L}$ is

$$\mathcal{L} = \int_{-\infty}^{0} \lambda \, dE_\lambda,$$

- from which it follows that

$$e^{\mathcal{L}t} = \int_{-\infty}^{0} e^{\lambda t} \, dE_\lambda \quad \text{and} \quad e^{\mathcal{L}t}\mathcal{L} = \int_{-\infty}^{0} e^{\lambda t}\lambda.$$

- Remember that $V^f = \mathcal{L}f$ We calculate (with $S_t = e^{\mathcal{L}t}$)

$$\mathbb{E}[V^f(X_t)V^g(X_0)] = (S_t\mathcal{L}f, \mathcal{L}g)_\mu = \int_{-\infty}^{0} \lambda^2 e^{\lambda t} d(E_\lambda f, g)_\mu. \tag{80}$$

- From Fubini's theorem and Cauchy-Schwarz it follows that

$$\int_0^\infty \int_{-\infty}^{0} \lambda^2 e^{\lambda t} |d(E_\lambda f, g)_\mu| \, dt \leq D_\mathcal{L}(f)^{1/2} D_\mathcal{L}(g)^{1/2} < +\infty$$

- with $D_\mathcal{L}(f) := (-\mathcal{L}f, f)_\mu$.

- We use now (80) and Fubini's theorem to calculate

$$
\begin{aligned}
\int_0^{+\infty} \mathbb{E}[V^f(X_t) V^g(X_0)]\, dt &= \int_0^t (S_t \mathcal{L} f, \mathcal{L} g)_\mu \, dt \\
&= \int_0^{+\infty} \int_{-\infty}^0 \lambda^2 e^{\lambda t} d(E_\lambda f, g)_\mu \, dt \\
&= \int_{-\infty}^0 (-\lambda) d(E_\lambda f, g)_\mu \\
&= \left(\int_{-\infty}^0 (-\lambda) dE_\lambda f, \int_{-\infty}^0 dE_\lambda g \right)_\mu \\
&= (-\mathcal{L} f, g)_\mu = \frac{1}{2} \int D^{f,g}(x) \mu(dx).
\end{aligned}
$$

Example

- Consider the diffusion process

$$dX_t = b(X_t)\, dt + \sigma(X_t) \circ dW_t, \tag{81}$$

  where the noise is interpreted in the Stratonovich sense.
- The generator is

$$\mathcal{L}\cdot = b(x) \cdot \nabla + \frac{1}{2} \nabla \cdot \left(A(x) \nabla \cdot \right), \tag{82}$$

  where $A(x) = (\sigma \sigma^T)(x)$.
- We assume that the diffusion process has a unique invariant distribution which is the solution of the stationary Fokker-Planck equation

$$\mathcal{L}^* \rho = 0. \tag{83}$$

- When A is strictly positive definite ergodicity follows from PDE arguments.

Example (Continued)

- The stationary process X_t (i.e. $X_0 \sim \rho(x)dx$) is reversible provided that
$$b(x) = \frac{1}{2}A(x)\nabla \log \rho(x). \tag{84}$$
- Let $f = x_i$, $g = x_j$. We calculate
$$V^{x_i}(x) = \mathcal{L}x_i = b_i + \frac{1}{2}\partial_k A_{ik}, \; i = 1, \dots d. \tag{85}$$
- We use the detailed balance condition (84) and (78) to calculate
$$\begin{aligned} \frac{1}{2}\int D^{x_i,x_j}\mu(dx) &= = -\int \left(b_i(x) + \frac{1}{2}\partial_k A_{ik}(x)\right) x_j \rho(x)\, dx \\ &= -\frac{1}{2}\int \left(A_{ik}\partial_k \rho(x) + \partial_k A_{ik}(x)\rho(x)\right) x_j \, dx \\ &= \frac{1}{2}\int A_{ij}(x)\rho(x)\, dx. \end{aligned}$$

Example (Continued)

- The Green-Kubo formula (77) gives:

$$\frac{1}{2}\int A_{ij}(x)\rho(x)\,dx = \int_0^{+\infty} \mathbb{E}\Big(V^{x_i}(X_t)V^{x_j}(X_0)\Big)\,dt, \tag{86}$$

- where the drift $V^{x_i}(x)$ is given by (84).