A mathematical introduction to steady-state nonequilibrium systems

Gabriel STOLTZ

gabriel.stoltz@enpc.fr

(CERMICS, Ecole des Ponts & MATHERIALS team, INRIA Paris)

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Outline

- **Definition and examples of nonequilibrium systems**
- **Computation of transport coefficients**
  - a survey of computational techniques
  - linear response theory
  - relationship with Green-Kubo formulas
- **Elements of numerical analysis**
  - estimation of biases due to timestep discretization
  - (largely) open issue: variance reduction

References

- **Some introductory references...**
  - L. Rey-Bellet, Open classical systems, *Lecture Notes in Mathematics*, **1881** (2006) 41–78
  - D. J. Evans and G. P. Morriss, *Statistical Mechanics of Nonequilibrium Liquids* (Cambridge University Press, 2008)
  - M. Tuckerman, *Statistical Mechanics: Theory and Molecular Simulation* (Oxford, 2010)
  - G. Stoltz, *Molecular Simulation: Nonequilibrium and Dynamical Problems*, Habilitation Thesis (2012) [Chapter 3]
  - T. Lelièvre and G. Stoltz, Partial differential equations and stochastic methods in molecular dynamics, *Acta Numerica* (2016)

- And many reviews on **specific topics**! For instance, thermal transport in one dimensional systems

Reference equilibrium dynamics

- Configuration $(q_t, p_t) \in \mathcal{E} = \mathbb{T}^d \times \mathbb{R}^d$
- Smooth periodic potential V

Langevin dynamics in a periodic potential

$$\begin{cases} \mathrm{d}q_t = M^{-1} p_t \, \mathrm{d}t, \\ \mathrm{d}p_t = -\nabla V(q_t) \, \mathrm{d}t - \gamma M^{-1} p_t \, \mathrm{d}t + \sqrt{\frac{2\gamma}{\beta}} \, \mathrm{d}W_t, \end{cases}$$

- Ergodic for the canonical measure with density $\psi_0(q,p) = Z_0^{-1} \mathrm{e}^{-\beta H(q,p)}$

$$\frac{1}{t} \int_0^t \varphi(q_s, p_s) \, ds \xrightarrow[t \to +\infty]{} \int_{\mathcal{E}} \varphi \, \psi_0 \qquad \text{a.s.}$$

- Exponential convergence of the law $\psi(t,q,p)$ (hypocoercivity, Lyapunov techniques, ...)

$$\|\psi(t) - \psi_0\|_E \leqslant C \, \mathrm{e}^{-\lambda t} \, \|\psi(0) - \psi_0\|_E$$

Definition and examples of nonequilibrium systems

Creating a particle flux

Langevin dynamics perturbed by a constant force term

$$\begin{cases} dq_t = M^{-1}p_t\,dt, \\ dp_t = (-\nabla V(q_t) + \eta F)\,dt - \gamma M^{-1}p_t\,dt + \sqrt{\frac{2\gamma}{\beta}}\,dW_t, \end{cases} \tag{1}$$

where

- $F \in \mathbb{R}^d$ with $|F| = 1$ is a given direction
- $\eta \in \mathbb{R}$ determines the strength of the external forcing

• Non-zero velocity in the direction F is expected in the steady-state

• F **does not derive from the gradient of a periodic function**

- of course, $F = -\nabla W_F(q)$ with $W_F(q) = -F^T q$
- ...but W_F is not periodic!

Creating an energy flux

Langevin dynamics with modified fluctuation

$$\begin{cases} \mathrm{d}q_t = M^{-1}p_t\,\mathrm{d}t, \\ \mathrm{d}p_t = -\nabla V(q_t)\,\mathrm{d}t - \gamma M^{-1}p_t\,\mathrm{d}t + \sqrt{2\gamma T_\eta(q)}\,\mathrm{d}W_t, \end{cases}$$

with non-negative temperature

$$T_\eta(q) = T_{\mathrm{ref}} + \eta\widetilde{T}(q)$$

Typically, $\widetilde{T}$ constant and positive on $\mathcal{D}_+ \subset \mathcal{D}$, and constant and negative on $\mathcal{D}_- \subset \mathcal{D}$

- Non-zero energy flux from $\mathcal{D}_+$ to $\mathcal{D}_-$ expected in the steady-state
- Simplified model of thermal transport (in 3D materials or atom chains)

SDEs: elements of analysis (1)

- Stochastic dynamics $dx_t = b(x_t)dt + \sigma(x_t)\, dW_t$
  - smooth drift and diffusion
  - configuration space $\mathcal{X}$, law $\psi(t,x)$ of x_t
  - unique invariant measure π
- Generator $\mathcal{L} = b \cdot \nabla + \frac{1}{2}\sigma\sigma^T : \nabla^2 = \sum_{i=1}^d b_i \partial_{x_i} + \sum_{i=1}^d \sum_{j=1}^d \left[\frac{\sigma\sigma^T}{2}\right]_{i,j} \partial_{x_i,x_j}$
- By definition, $\left.\frac{d}{dt}[\mathbb{E}^x(\varphi(x_t))]\right|_{t=0} = \mathcal{L}\varphi(x)$ (Itô formula)
- Analytic formulation of the time evolution $\frac{d}{dt}[\mathbb{E}^x(\varphi(x_t))] = \mathbb{E}^x(\mathcal{L}\varphi(x_t))$:

$$\frac{d}{dt}\left(\int_{\mathcal{X}} \varphi(x)\psi(t,x)\, dx\right) = \int_{\mathcal{X}} (\mathcal{L}\varphi)(x)\psi(t,x)\, dx$$

Understanding the Brownian motion and generators (1)

- Independant Gaussian increments whose variance is proportional to time

$$\forall\, 0 < t_0 \leqslant t_1 \leqslant \cdots \leqslant t_n, \qquad W_{t_{i+1}} - W_{t_i} \sim \mathcal{N}(0, t_{i+1} - t_i)$$

where the increments $W_{t_{i+1}} - W_{t_i}$ are independent

- $G \sim \mathcal{N}(m, \sigma^2)$ distributed according to the probability density

$$g(x) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(-\frac{(x-m)^2}{2\sigma^2}\right)$$

- The solution of $dq_t = \sigma dW_t$ can be thought of as the limit $\Delta t \to 0$

$$q^{n+1} = q^n + \sigma\sqrt{\Delta t}\, G^n, \qquad G^n \sim \mathcal{N}(0,1) \text{ i.i.d.}$$

where q^n is an approximation of $q_{n\Delta t}$

- Note that $q^n \sim \mathcal{N}(q^0, \sigma n \Delta t)$

- Multidimensional case: $W_t = (W_{1,t}, \ldots, W_{d,t})$ where W_i are independent

Understanding the Brownian motion and generators (2)

- Analytical study of the process: law $\psi(t,q)$ of the process at time t

$\rightarrow$ distribution of all possible realizations of q_t for

- a given initial distribution $\psi(0,q)$, *e.g.* δ_{q^0}
- and all realizations of the Brownian motion

Averages at time t

$$\mathbb{E}\Big(\varphi(q_t)\Big) = \int_{\mathcal{D}} \varphi(q)\,\psi(t,q)\,dq$$

- Partial differential equation governing the evolution of the law

Fokker-Planck equation

$$\partial_t \psi = \frac{\sigma^2}{2}\Delta\psi$$

Here, simple heat equation $\rightarrow$ "diffusive behavior"

Understanding the Brownian motion and generators (2)

- Proof: Taylor expansion, beware random terms of order $\sqrt{\Delta t}$

$$\varphi\left(q^{n+1}\right) = \varphi\left(q^n + \sigma\sqrt{\Delta t}\, G^n\right)$$
$$= \varphi\left(q^n\right) + \sigma\sqrt{\Delta t} G^n \cdot \nabla\varphi\left(q^n\right) + \frac{\sigma^2 \Delta t}{2}\left(G^n\right)^T \left(\nabla^2\varphi\left(q^n\right)\right) G^n + \mathrm{O}\left(\Delta t^{3/2}\right)$$

Taking expectations (Gaussian increments G^n independent from the current position q^n)

$$\mathbb{E}\left[\varphi\left(q^{n+1}\right)\right] = \mathbb{E}\left[\varphi\left(q^n\right) + \frac{\sigma^2 \Delta t}{2}\Delta\varphi\left(q^n\right)\right] + \mathrm{O}\left(\Delta t^{3/2}\right)$$

Therefore, $\mathbb{E}\left[\dfrac{\varphi\left(q^{n+1}\right) - \varphi\left(q^n\right)}{\Delta t} - \dfrac{\sigma^2}{2}\Delta\varphi\left(q^n\right)\right] \to 0$. On the other hand,

$$\mathbb{E}\left[\frac{\varphi\left(q^{n+1}\right) - \varphi\left(q^n\right)}{\Delta t}\right] \to \partial_t\Big(\mathbb{E}\left[\varphi(q_t)\right]\Big) = \int_{\mathcal{D}} \varphi(q)\partial_t\psi(t,q)\, dq.$$

This leads to

$$0 = \int_{\mathcal{D}} \varphi(q)\partial_t\psi(t,q)\, dq - \frac{\sigma^2}{2}\int_{\mathcal{D}} \Delta\varphi(q)\,\psi(t,q)\, dq = \int_{\mathcal{D}} \varphi(q)\left(\partial_t\psi(t,q) - \frac{\sigma^2}{2}\Delta\psi(t,q)\right) dq$$

This equality holds for all observables φ.

Understanding the Brownian motion and generators (4)

- State of the system $X \in \mathbb{R}^d$, m-dimensional Brownian motion, diffusion matrix $\sigma \in \mathbb{R}^{d\times m}$

$$dx_t = b(x_t)\,dt + \sigma(x_t)\,dW_t$$

to be thought of as the limit as $\Delta t \to 0$ of (X^n approximation of $X_{n\Delta t}$)

$$X^{n+1} = X^n + \Delta t\, b\left(X^n\right) + \sqrt{\Delta t}\,\sigma(X^n) G^n, \qquad G^n \sim \mathcal{N}\left(0, \mathrm{Id}_m\right)$$

- Generator

$$\mathcal{L} = b(x)\cdot\nabla + \frac{1}{2}\sigma\sigma^T(x) : \nabla^2 = \sum_{i=1}^{d} b_i(x)\partial_{x_i} + \frac{1}{2}\sum_{i,j=1}^{d}\left[\sigma\sigma^T(x)\right]_{i,j}\partial_{x_i}\partial_{x_j}$$

- Proceeding as before, it can be shown that

$$\frac{d}{dt}\Big(\mathbb{E}\left[\varphi(q_t)\right]\Big) = \int_{\mathcal{X}} \varphi\,\partial_t\psi = \mathbb{E}\Big[\left(\mathcal{L}\varphi\right)(x_t)\Big] = \int_{\mathcal{X}} \left(\mathcal{L}\varphi\right)\psi$$

SDEs: elements of analysis (2)

- Fokker-Planck equation $\partial_t \psi = \mathcal{L}^\dagger \psi$, with adjoint on $L^2(\mathcal{X})$:

$$\int_{\mathcal{X}} (\mathcal{L}\varphi)\,\phi = \int_{\mathcal{X}} \varphi\,\left(\mathcal{L}^\dagger \phi\right)$$

Invariance of measure π

Expressed as $\mathcal{L}^\dagger \pi = 0$ or $\quad \forall \varphi, \quad \displaystyle\int_{\mathcal{X}} \mathcal{L}\varphi\,d\pi = 0$

- Other framework: work in $L^2(\pi)$ and consider the adjoint $\mathcal{L}^*$ defined as

$$\int_{\mathcal{X}} (\mathcal{L}\varphi)\,\phi\,d\pi = \int_{\mathcal{X}} \varphi\,(\mathcal{L}^*\phi)\,d\pi$$

Invariance of measure π

Expressed as $\mathcal{L}^* \mathbf{1} = 0$ or $\quad \forall \varphi, \quad \displaystyle\int_{\mathcal{X}} \mathcal{L}\varphi\,d\pi = 0$

What is an equilibrium system?

Reversible dynamics: arrow of time cannot be read off trajectories

When $x_0 \sim \pi$, the law of the forward paths $(x_s)_{0 \leqslant s \leqslant t}$ is the same as the law of the backward paths $(x_{t-s})_{0 \leqslant s \leqslant t}$

- From an analytic viewpoint: self-adjointness of the generator on $L^2(\pi)$

$$\int_{\mathcal{X}} (\mathcal{L}f)\, g \, \mathrm{d}\pi = \int_{\mathcal{X}} f \, (\mathcal{L}g) \, \mathrm{d}\pi$$

- Example: overdamped Langevin dynamics $\mathrm{d}q_t = -\nabla V(q_t)\,\mathrm{d}t + \sqrt{\frac{2}{\beta}}\,\mathrm{d}W_t$

$$\pi(q) = Z^{-1}\mathrm{e}^{-\beta V(q)}, \qquad \int_{\mathcal{D}} (\mathcal{L}f)\, g\,\mathrm{d}\pi = -\frac{1}{\beta}\int_{\mathcal{D}} \nabla f \cdot \nabla g \,\mathrm{d}\pi$$

What is a nonequilibrium system?

Non-reversible dynamics

The time arrow can be read off the trajectories: the generator is not symmetric on $L^2(\pi)$

- Subtlety: reversibility may hold up to a one-to-one transformation
- For Langevin dynamics, reversibility up to momentum reversal

$$S(q,p) = (q,-p)$$

i.e. the law of the forward paths $(q_s,p_s)_{0 \leqslant s \leqslant t}$ is the same as the law of the backward paths with reverted momenta $(q_{t-s},-p_{t-s})_{0 \leqslant s \leqslant t}$

$$\int_{\mathcal{X}} (\mathcal{L}f)\, g \, \mathrm{d}\pi = \int_{\mathcal{X}} (f \circ S) \Big(\mathcal{L}(g \circ S) \Big) \, \mathrm{d}\pi$$

Stationary states (1)

- Stationary solutions of Fokker-Planck equation: $\mathcal{L}^\dagger \varphi = 0$ or

$$\forall \varphi, \qquad \int_{\mathcal{X}} \mathcal{L}\varphi \, d\pi = 0$$

- Usually not known for non-reversible dynamics... except in a perturbative framework

- Generic existence of long-range correlations[1]

- **Example:** 1D dynamics $dq_t = (-V'(q_t) + F)\, dt + \sqrt{2}\, dW_t$, invariant measure with density

$$\psi_F(q) = Z_F^{-1} \int_{\mathbb{T}} \mathrm{e}^{V(q+y)-V(q)-Fy}\, dy$$

Because of $F \neq 0$, a modification to V at a given point is felt everywhere!

[1] B. Derrida, J. L. Lebowitz and E. R. Speer, *J. Stat. Phys.* (2002)

Stationary states (2)

- For the model case: generator $\mathcal{L}_\eta = \mathcal{L}_0 + \eta\widetilde{\mathcal{L}}$ with

$$\widetilde{\mathcal{L}} = F \cdot \nabla_p, \qquad \mathcal{L}_0 = \mathcal{L}_{\mathrm{ham}} + \gamma \mathcal{L}_{\mathrm{OU}},$$

where $\mathcal{L}_{\mathrm{ham}} = p^T M^{-1} \nabla_q - \nabla V^T \nabla_p$ and $\mathcal{L}_{\mathrm{OU}} = -p^T M^{-1} \nabla_p + \dfrac{1}{\beta}\Delta_p$

- It can be shown that $\mathcal{L}_0^* = -\mathcal{L}_{\mathrm{ham}} + \gamma \mathcal{L}_{\mathrm{OU}}$
- Invariance of ψ_0 expressed as $\mathcal{L}_0^* \mathbf{1} = 0$
- Lyapunov functions $\mathcal{K}_n(q,p) = 1 + |p|^n$ for $n \geqslant 2$, with

$$\exists a_n > 0, b_n \in \mathbb{R}, \qquad \mathcal{L}_\eta \mathcal{K}_n \leqslant -a_n \mathcal{K}_n + b_n$$

- Weighted L^∞ norm defined as $\|\varphi\|_{L^\infty_{\mathcal{K}_n}} = \left\| \dfrac{\varphi}{\mathcal{K}_n} \right\|_{L^\infty}$
- Evolution semigroup $\left(\mathrm{e}^{t\mathcal{L}_\eta}\varphi\right)(q_0,p_0) = \mathbb{E}^{(q_0,p_0)}\left[\varphi(q_t,p_t)\right]$
- Lyapunov techniques to prove existence of invariant measure[2]

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[2] M. Hairer and J. Mattingly, *Progr. Probab.* (2011); Meyn and Tweedie (2009)

Stationary states (3)

Exponential convergence to equilibrium

Consider $\eta_* > 0$. For any $\eta \in [-\eta_*, \eta_*]$, the dynamics (1) admits a unique invariant probability measure with a C^∞ density $\psi_\eta(q,p)$ w.r.t. Lebesgue measure.

Moreover, for any $n \geqslant 2$, there exist $C_n, \lambda_n > 0$ (depending on η_*) such that, for any $\eta \in [-\eta_*, \eta_*]$ and for any $\varphi \in L^\infty_{\mathcal{K}_n}(\mathcal{E})$,

$$\forall t \geqslant 0, \qquad \left\| \mathrm{e}^{t\mathcal{L}_\eta}\varphi - \int_{\mathcal{E}} \varphi \, \psi_\eta \right\|_{L^\infty_{\mathcal{K}_n}} \leqslant C_n \mathrm{e}^{-\lambda_n t} \, \|\varphi\|_{L^\infty_{\mathcal{K}_n}}$$

- **Corollary:** $\mathcal{L}_\eta^{-1}$ is a bounded operator on

$$L^\infty_{\mathcal{K}_n,\eta}(\mathcal{E}) = \left\{ \varphi \in L^\infty_{\mathcal{K}_n}(\mathcal{E}) \,\middle|\, \int_{\mathcal{E}} \varphi \, \psi_\eta = 0 \right\}$$

and $\mathcal{L}_\eta = -\displaystyle\int_0^{+\infty} \mathrm{e}^{t\mathcal{L}_\eta} \, dt$ in $\mathcal{B}(L^\infty_{\mathcal{K}_n,\eta})$

Linear response theory and the computation of transport coefficients

Computation of transport properties: general classification

- There are three main types of techniques
  - Equilibrium techniques: Green-Kubo formula (autocorrelation)
  - Transient methods
  - Steady-state nonequilibrium techniques
    - boundary driven
    - bulk driven

- Definitions use analogy with macroscopic evolution equations

- **Examples of mathematical questions:**
  - (equilibrium) integrability of correlation functions
  - (steady-state nonequilibrium): existence and uniqueness of an invariant probability measure

Linear response of nonequilibrium dynamics (1)

- The force ηF induces a non-zero velocity in the direction F
- Encoded by $\mathbb{E}_\eta(R) = \displaystyle\int_{\mathcal{E}} R\,\psi_\eta$ with $R(q,p) = F^T M^{-1} p$

Definition of the mobility

$$\rho_F = \lim_{\eta \to 0} \frac{\mathbb{E}_\eta(R) - \mathbb{E}_0(R)}{\eta} = \lim_{\eta \to 0} \frac{\mathbb{E}_\eta(R)}{\eta}$$

- It is expected that $\psi_\eta = f_\eta \psi_0$ with $\psi_0(q,p) = Z^{-1} \mathrm{e}^{-\beta H(q,p)}$ and

$$f_\eta = \mathbf{1} + \eta \mathfrak{f}_1 + \mathrm{O}(\eta^2)$$

- In this case, $\rho_F = \displaystyle\int_{\mathcal{E}} R \mathfrak{f}_1 \psi_0$
- **Questions:** Can the expansion for f_η be made rigorous? What is $\mathfrak{f}_1$?

Linear response of nonequilibrium dynamics (2)

- Perturbative framework where $\mathcal{L}_0$ considered on $L^2(\psi_0)$ is the reference
- The invariance of ψ_η can be written as

$$\int_{\mathcal{E}} (\mathcal{L}_\eta \varphi) \psi_\eta = 0 = \int_{\mathcal{E}} (\mathcal{L}_\eta \varphi) f_\eta \psi_0$$

Fokker-Planck equation on $L^2(\psi_0)$

$$\mathcal{L}_\eta^* f_\eta = 0$$

- Formally, $\mathcal{L}_\eta^* f_\eta = (\mathcal{L}_0)^* \underbrace{\left(\mathrm{Id} + \widetilde{\mathcal{L}} \mathcal{L}_0^{-1}\right)^* f_\eta}_{=1?} = 0$
- To make the result precise, introduce $L_0^2(\psi_0) = \Pi_0 L^2(\psi_0)$ with

$$\Pi_0 f = f - \int_{\mathcal{E}} f \, \psi_0$$

Linear response of nonequilibrium dynamics (2)

Power expansion of the invariant measure

Spectral radius r of the bounded operator $(\widetilde{\mathcal{L}}\mathcal{L}_0^{-1})^* \in \mathcal{B}(L_0^2(\psi_0))$:

$$r = \lim_{n \to +\infty} \left\| \left[\left(\widetilde{\mathcal{L}}\mathcal{L}_0^{-1} \right)^* \right]^n \right\|^{1/n}.$$

Then, for $|\eta| < r^{-1}$, the unique invariant measure can be written as $\psi_\eta = f_\eta \psi_0$, where $f_\eta \in L^2(\psi_0)$ can be expanded as

$$f_\eta = \left(1 + \eta(\widetilde{\mathcal{L}}\mathcal{L}_0^{-1})^*\right)^{-1} \mathbf{1} = \left(1 + \sum_{n=1}^{+\infty} (-\eta)^n [(\widetilde{\mathcal{L}}\mathcal{L}_0^{-1})^*]^n \right) \mathbf{1}. \qquad (2)$$

- Note that $\displaystyle\int_{\mathcal{E}} \psi_\eta = 1$
- Linear response result: $\rho_F = -\displaystyle\int_{\mathcal{E}} R \left[(\widetilde{\mathcal{L}}\mathcal{L}_0^{-1})^* \mathbf{1} \right] \psi_0$

Elements of proof

- Since $\frac{\gamma}{\beta}\|\nabla_p \varphi\|^2_{L^2(\psi_0)} = -\langle \mathcal{L}_0 \varphi, \varphi \rangle_{L^2(\psi_0)}$, it follows that

$$\|\widetilde{\mathcal{L}}\varphi\|^2_{L^2(\psi_0)} \leqslant \|\nabla_p \varphi\|^2_{L^2(\psi_0)} \leqslant \frac{\beta}{\gamma}\|\mathcal{L}_0\varphi\|_{L^2(\psi_0)}\|\varphi\|_{L^2(\psi_0)}$$

- $\mathcal{L}_0^{-1}$ is a well defined bounded operator on $L^2_0(\psi_0)$ (hypocoercivity + hypoelliptic regularization)

$$\|\widetilde{\mathcal{L}}\mathcal{L}_0^{-1}\varphi\|^2_{L^2(\psi_0)} \leqslant \frac{\beta}{\gamma}\|\varphi\|_{L^2(\psi_0)}\|\mathcal{L}_0^{-1}\varphi\|_{L^2(\psi_0)}.$$

- $\Pi_0\widetilde{\mathcal{L}}\mathcal{L}_0^{-1}$ is bounded on $L^2_0(\psi_0)$, so $(\widetilde{\mathcal{L}}\mathcal{L}_0^{-1})^*\Pi_0 = (\widetilde{\mathcal{L}}\mathcal{L}_0^{-1})^*$ is also bounded on $L^2_0(\psi_0)$

- Invariance of f_η by $\mathcal{L}_\eta^* = \mathcal{L}^* + \eta\widetilde{\mathcal{L}}^*$

$$\mathcal{L}_\eta^* f_\eta = \mathcal{L}_0^*\left(1 + \eta(\widetilde{\mathcal{L}}\mathcal{L}_0^{-1})^*\right) f_\eta = \mathcal{L}_0^*\mathbf{1} = 0$$

- Prove that $f_\eta \geqslant 0$ (use some ergodicity result to show that $\psi_\eta = f_\eta\psi_0$)

Reformulation as integrated correlation functions

- Conjugate response $S = \widetilde{\mathcal{L}}^* \mathbf{1}$, equivalently $\displaystyle\int_{\mathcal{E}} \left(\widetilde{\mathcal{L}}\varphi\right) \psi_0 = \int_{\mathcal{E}} \varphi \, S \, \psi_0$

Green–Kubo formula

For any $R \in L^2_0(\psi_0)$,

$$\lim_{\eta \to 0} \frac{\mathbb{E}_\eta(R)}{\eta} = \int_0^{+\infty} \mathbb{E}_0\Big(R(q_t, p_t) S(q_0, p_0)\Big) dt,$$

where $\mathbb{E}_\eta$ is w.r.t. to $\psi_\eta(q,p)\, dq\, p$, while $\mathbb{E}_0$ is taken over initial conditions $(q_0, p_0) \sim \psi_0$ and over all realizations of the equilibrium dynamics.

- For the dynamics (1), it holds $S(q,p) = \beta R(q,p) = \beta F^T M^{-1} p$ so that

$$\rho_F = \lim_{\eta \to 0} \frac{\mathbb{E}_\eta(F \cdot M^{-1} p)}{\eta} = \beta \int_0^{+\infty} \mathbb{E}_0\Big((F \cdot M^{-1} p_t)(F \cdot M^{-1} p_0)\Big) dt$$

Elements of proof

- Proof based on the following equality on $\mathcal{B}(L_0^2(\psi_0))$

$$-\mathcal{L}_0^{-1} = \int_0^{+\infty} \mathrm{e}^{t\mathcal{L}_0} \, dt$$

- Then,

$$\begin{aligned}
\lim_{\eta \to 0} \frac{\mathbb{E}_\eta(R)}{\eta} &= -\int_{\mathcal{E}} R \left[(\widetilde{\mathcal{L}} \mathcal{L}_0^{-1})^* \mathbf{1} \right] \psi_0 = -\int_{\mathcal{E}} [\mathcal{L}_0^{-1} R][\widetilde{\mathcal{L}}^* \mathbf{1}] \, \psi_0 \\
&= \int_0^{+\infty} \left(\int_{\mathcal{E}} \left(\mathrm{e}^{t\mathcal{L}_0} R \right) \, S \, \psi_0 \right) dt \\
&= \int_0^{+\infty} \mathbb{E}\Big(R(q_t, p_t) S(q_0, p_0) \Big) dt
\end{aligned}$$

- Note also that S has average 0 w.r.t. invariant measure since

$$\int_{\mathcal{X}} S \, d\pi = \int_{\mathcal{X}} \widetilde{\mathcal{L}}^* \mathbf{1} \, d\pi = \int_{\mathcal{X}} \widetilde{\mathcal{L}} \mathbf{1} \, d\pi = 0$$

Generalization to other dynamics

- **Possible assumptions to justify the linear response**
  - existence of invariant measure with smooth density ψ_η
  - ergodicity $\displaystyle \frac{1}{t}\int_0^t \varphi(x_s)\,ds \xrightarrow[t\to+\infty]{} \int_{\mathcal{X}} \varphi\,\psi_\eta$
  - $\mathrm{Ker}(\mathcal{L}_0^*) = \mathbf{1}$ and $\mathcal{L}_0^*$ is invertible on $L_0^2(\psi_0)$
  - the perturbation $\widetilde{\mathcal{L}}$ is $\mathcal{L}_0$-bounded: there exist $a, b \geqslant 0$ such that

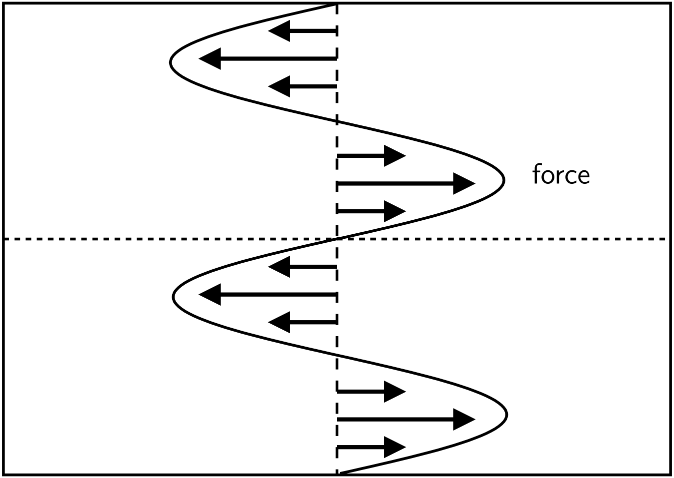
$$\|\widetilde{\mathcal{L}}\varphi\|_{L^2(\psi_0)} \leqslant a\|\mathcal{L}_0\varphi\|_{L^2(\psi_0)} + b\|\varphi\|_{L^2(\psi_0)}$$

- **When the perturbation is not sufficiently weak?** (thermal transport)
  - compute $\displaystyle \int_{\mathcal{X}} [(\mathcal{L}_0 + \eta\widetilde{\mathcal{L}})\varphi](1+\eta\mathfrak{f}_1)\psi_0 = \mathrm{O}(\eta^2)$
  - use a pseudo-inverse $Q_\eta = \Pi_0\mathcal{L}_0^{-1}\Pi_0 - \eta\Pi_0\mathcal{L}_0^{-1}\Pi_0\widetilde{\mathcal{L}}\Pi_0\mathcal{L}_0^{-1}\Pi_0$
  - allows to prove that $\displaystyle \int_{\mathcal{X}} \varphi\,\psi_\eta = \int_{\mathcal{X}} \varphi\,\psi_0 + \eta\int_{\mathcal{X}} \varphi\,\mathfrak{f}_1\,\psi_0 + \eta^2 r_{\varphi,\eta}$

Other examples

Shear viscosity in fluids (1)

2D system to simplify notation: $\mathcal{D} = (L_x\mathbb{T} \times L_y\mathbb{T})^N$



Shear viscosity in fluids (2)

- Add a smooth nongradient force in the x direction, depending on y

Langevin dynamics under flow

$$\begin{cases} dq_{i,t} = \dfrac{p_{i,t}}{m}\,dt, \\ dp_{xi,t} = -\nabla_{q_{xi}} V(q_t)\,dt + \eta F(q_{yi,t})\,dt - \gamma_x \dfrac{p_{xi,t}}{m}\,dt + \sqrt{\dfrac{2\gamma_x}{\beta}}\,dW_t^{xi}, \\ dp_{yi,t} = -\nabla_{q_{yi}} V(q_t)\,dt - \gamma_y \dfrac{p_{yi,t}}{m}\,dt + \sqrt{\dfrac{2\gamma_y}{\beta}}\,dW_t^{yi}, \end{cases}$$

- Existence/uniqueness of a smooth invariant measure provided $\gamma_x, \gamma_y > 0$
- The perturbation $\widetilde{\mathcal{L}} = \sum_{i=1}^{N} F(q_{y,i})\partial_{p_{x,i}}$ is $\mathcal{L}_0$-bounded
- Linear response: $\displaystyle \lim_{\eta \to 0} \frac{\langle \mathcal{L}_0 h \rangle_\eta}{\eta} = -\frac{\beta}{m} \left\langle h, \sum_{i=1}^{N} p_{xi} F(q_{yi}) \right\rangle_{L^2(\psi_0)}$

Shear viscosity in fluids (3)

- Average longitudinal velocity $u_x(Y) = \lim_{\varepsilon\to 0}\lim_{\eta\to 0} \frac{\langle U_x^\varepsilon(Y,\cdot)\rangle_\eta}{\eta}$ where

$$U_x^\varepsilon(Y,q,p) = \frac{L_y}{Nm}\sum_{i=1}^N p_{xi}\chi_\varepsilon\left(q_{yi}-Y\right)$$

- Average off-diagonal stress $\sigma_{xy}(Y) = \lim_{\varepsilon\to 0}\lim_{\eta\to 0} \frac{\langle\dots\rangle_\eta}{\eta}$, where ... =

$$\frac{1}{L_x}\left(\sum_{i=1}^N \frac{p_{xi}p_{yi}}{m}\chi_\varepsilon\left(q_{yi}-Y\right) - \sum_{1\leqslant i<j\leqslant N}\mathcal{V}'(|q_i-q_j|)\frac{q_{xi}-q_{xj}}{|q_i-q_j|}\int_{q_{yj}}^{q_{yi}}\chi_\varepsilon(s-Y)\,ds\right)$$

- Local conservation of momentum[3]: replace h by U_x^ε (with $\overline{\rho} = N/|\mathcal{D}|$)

$$\frac{d\sigma_{xy}(Y)}{dY} + \gamma_x\overline{\rho}u_x(Y) = \overline{\rho}F(Y)$$

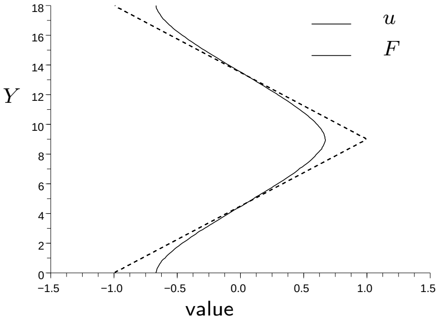
[3] Irving and Kirkwood, *J. Chem. Phys.* **18** (1950)

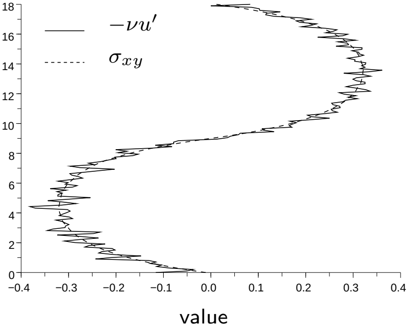
Shear viscosity in fluids (4)

- Definition $\sigma_{xy}(Y) := -\eta(Y)\dfrac{du_x(Y)}{dY}$, closure assumption $\eta(Y) = \eta > 0$

Velocity profile in Langevin dynamics under flow

$$-\eta u_x''(Y) + \gamma_x \overline{\rho} u_x(Y) = \overline{\rho} F(Y)$$





Thermal transport in one-dimensional chains (1)

- Atoms at positions $q_0, \ldots, q_N$ with $q_0 = 0$ fixed
- Hamiltonian $H(q,p) = \sum_{i=1}^{N} \frac{p_i^2}{2} + \sum_{i=1}^{N-1} v(q_{i+1} - q_i) + v(q_1)$

Hamiltonian dynamics with Langevin thermostats at the boundaries

$$\begin{cases} dq_i = p_i \, dt \\ dp_i = \Big(v'(q_{i+1} - q_i) - v'(q_i - q_{i-1})\Big) dt, \qquad i \neq 1, N \\ dp_1 = \Big(v'(q_2 - q_1) - v'(q_1)\Big) dt - \gamma p_1 \, dt + \sqrt{2\gamma(T + \Delta T)} \, dW_t^1 \\ dp_N = -v'(q_N - q_{N-1}) \, dt - \gamma p_N \, dt + \sqrt{2\gamma(T - \Delta T)} \, dW_t^N \end{cases}$$

- Perturbation $\widetilde{\mathcal{L}} = \gamma(\partial_{p_1}^2 - \partial_{p_N}^2)$ (not $\mathcal{L}_0$-bounded...)
- Proving the existence/uniqueness of the invariant measure already requires quite some work[4]

[4] P. Carmona, Stoch. Proc. Appl. (2007)

Thermal transport in one-dimensional chains (2)

- Response function: Total energy current

$$J = \frac{1}{N-1}\sum_{i=1}^{N-1} j_{i+1,i}, \qquad j_{i+1,i} = -v'(q_{i+1}-q_i)\frac{p_i+p_{i+1}}{2}$$

- Motivation: Local conservation of the energy (in the bulk)

$$\frac{d\varepsilon_i}{dt} = j_{i-1,i} - j_{i,i+1}, \qquad \varepsilon_i = \frac{p_i^2}{2} + \frac{1}{2}\Big(v(q_{i+1}-q_i) + v(q_i - q_{i-1})\Big)$$

- Definition of the thermal conductivity: linear response

$$\kappa_N = \lim_{\Delta T \to 0} \frac{\langle J \rangle_{\Delta T}}{\Delta T / N} = 2\beta^2 \frac{N}{N-1}\int_0^{+\infty} \sum_{i=1}^{N-1} \mathbb{E}\Big(j_{2,1}(q_t,p_t) j_{i+1,i}(q_0,p_0)\Big)\, dt$$

- Synthetic dynamics: fixed temperatures of the thermostats but external forcings $\to$ bulk driven dynamics with $\widetilde{\mathcal{L}}^* = -\widetilde{\mathcal{L}} + cJ$

Error estimates on the computation of transport coefficients

Reminder: Error estimates in Monte Carlo simulations

- General SDE $dx_t = b(x_t)\,dt + \sigma(x_t)\,dW_t$, invariant measure π
- Discretization $x^n \simeq x_{n\Delta t}$, invariant measure $\pi_{\Delta t}$. For instance,

$$x^{n+1} = x^n + \Delta t\, b(x^n) + \sqrt{\Delta t}\,\sigma(x^n)\,G^n, \qquad G^n \sim \mathcal{G}(0,\mathrm{Id}) \text{ i.i.d.}$$

- Ergodicity of the numerical scheme with invariant measure $\pi_{\Delta t}$

$$\frac{1}{N_{\mathrm{iter}}}\sum_{n=1}^{N_{\mathrm{iter}}} A(x^n) \xrightarrow[N_{\mathrm{iter}}\to+\infty]{} \int_{\mathcal{X}} A(x)\,\pi_{\Delta t}(dx)$$

Error estimates for finite trajectory averages

$$\widehat{A}_{N_{\mathrm{iter}}} = \frac{1}{N_{\mathrm{iter}}}\sum_{n=1}^{N_{\mathrm{iter}}} A(x^n) = \mathbb{E}_\pi(A) + \underbrace{C\Delta t^\alpha}_{\text{bias}} + \underbrace{\frac{\sigma_{A,\Delta t}}{\sqrt{N_{\mathrm{iter}}\Delta t}}\mathscr{G}}_{\text{statistical error}}$$

- Bias $\mathbb{E}_{\pi_{\Delta t}}(A) - \mathbb{E}_\pi(A) \longrightarrow$ **Focus today**

Weak type expansions

- Numerical scheme = Markov chain characterized by evolution operator

$$P_{\Delta t}\varphi(x) = \mathbb{E}\Big(\varphi\left(x^{n+1}\right)\Big| x^n = x\Big)$$

where (x^n) is an approximation of $(x_{n\Delta t})$

- (Infinitely) Many possibilities! Numerical analysis allows to discriminate
- Standard notions of error: fixed integration time $T < +\infty$
  - Strong error $\sup_{0 \leqslant n \leqslant T/\Delta t} \mathbb{E}|X^n - X_{n\Delta t}| \leqslant C\Delta t^p$
  - Weak error: $\sup_{0 \leqslant n \leqslant T/\Delta t} \Big|\mathbb{E}\left[\varphi\left(X^n\right)\right] - \mathbb{E}\left[\varphi\left(X_{n\Delta t}\right)\right]\Big| \leqslant C\Delta t^p$ (for any φ)

Δt-expansion of the evolution operator

$$P_{\Delta t}\varphi = \varphi + \Delta t\, \mathcal{A}_1\varphi + \Delta t^2 \mathcal{A}_2\varphi + \cdots + \Delta t^{p+1}\mathcal{A}_{p+1}\varphi + \Delta t^{p+2} r_{\varphi,\Delta t}$$

- Weak order p when $\mathcal{A}_k = \mathcal{L}^k/k!$ for $1 \leqslant k \leqslant p$

Example: Euler-Maruyama, weak order 1

- Scheme $x^{n+1} = \Phi_{\Delta t}(x^n, G^n) = x^n + \Delta t\, b(x^n) + \sqrt{\Delta t}\, \sigma(x^n)\, G^n$
- Note that $P_{\Delta t}\varphi(x) = \mathbb{E}_G\left[\varphi\big(\Phi_{\Delta t}(x,G)\big)\right]$
- Technical tool: Taylor expansion
$$\varphi(x+\delta) = \varphi(x) + \delta^T \nabla\varphi(x) + \frac{1}{2}\delta^T \nabla^2\varphi(x)\delta + \frac{1}{6}D^3\varphi(x) : \delta^{\otimes 3} + \dots$$
- Replace δ with $\sqrt{\Delta t}\,\sigma(x)\,G + \Delta t\, b(x)$ and gather in powers of Δt
$$\begin{aligned}\varphi\big(\Phi_{\Delta t}(x,G)\big) &= \varphi(x) + \sqrt{\Delta t}\,\sigma(x)\,G\cdot\nabla\varphi(x)\\ &\quad + \Delta t\left(\frac{\sigma(x)^2}{2}G^T\left[\nabla^2\varphi(x)\right]G + b(x)\cdot\nabla\varphi(x)\right) + \dots\end{aligned}$$
- Taking expectations w.r.t. G leads to
$$P_{\Delta t}\varphi(x) = \varphi(x) + \Delta t\underbrace{\left(\frac{\sigma(x)^2}{2}\Delta\varphi(x) + b(x)\cdot\nabla\varphi(x)\right)}_{=\mathcal{L}\varphi(x)} + \mathrm{O}(\Delta t^2)$$

Error estimates on the invariant measure (equilibrium)

- Assumptions on the operators in the weak-type expansion
  - invariance of π by $\mathcal{A}_k$ for $1 \leqslant k \leqslant p$, namely $\displaystyle\int_{\mathcal{X}} \mathcal{A}_k \varphi \, d\pi = 0$
  - $\displaystyle\int_{\mathcal{X}} \mathcal{A}_{p+1} \varphi \, d\pi = \int_{\mathcal{X}} g_{p+1} \varphi \, d\pi$ (*i.e.* $g_{p+1} = \mathcal{A}_{p+1}^* \mathbf{1}$)

Error estimates on $\pi_{\Delta t}$

$$\int_{\mathcal{X}} \varphi \, d\pi_{\Delta t} = \int_{\mathcal{X}} \varphi \Big(1 + \Delta t^p f_{p+1} \Big) d\pi + \Delta t^{p+1} R_{\varphi, \Delta t}$$

- In fact, $f_{p+1} = -\left(\mathcal{A}_1^*\right)^{-1} g_{p+1}$
  - when $\mathcal{A}_1 = \mathcal{L}$, the first order correction can be estimated by some integrated correlation function as $\displaystyle\int_0^{+\infty} \mathbb{E}\Big(\varphi(x_t) g_{p+1}(x_0)\Big) \, dt$
  - in general, first order term can be removed by Romberg extrapolation
- Error on invariant measure can be (much) smaller than the weak error

Sketch of proof (1)

Step 1: Establish the error estimate for $\varphi \in \mathrm{Ran}(P_{\Delta t} - \mathrm{Id})$

- Idea: $\pi_{\Delta t} = \pi(1 + \Delta t^p f_{p+1} + \dots)$
- by definition of $\pi_{\Delta t}$

$$\int_{\mathcal{X}} \left[\left(\frac{P_{\Delta t} - \mathrm{Id}}{\Delta t} \right) \psi \right] d\pi_{\Delta t} = 0$$

- compare to first order correction to the invariant measure

$$\begin{aligned} \int_{\mathcal{X}} \left[\left(\frac{P_{\Delta t} - \mathrm{Id}}{\Delta t} \right) \psi \right] (1 + \Delta t^p f_{p+1}) \, d\pi & \\ = \Delta t^p \int_{\mathcal{X}} \Big(\mathcal{A}_{p+1}\psi + (\mathcal{A}_1 \psi) f_{p+1} \Big) d\pi + \mathrm{O}\left(\Delta t^{p+1}\right) & \\ = \Delta t^p \int_{\mathcal{X}} \Big(g_{p+1} + \mathcal{A}_1^* f_{p+1} \Big) \psi \, d\pi + \mathrm{O}\left(\Delta t^{p+1}\right) & \end{aligned}$$

Suggests $f_{p+1} = -\left(\mathcal{A}_1^*\right)^{-1} g_{p+1}$

Step 2: Define an approximate inverse

- Issue: derivatives of $(\mathrm{Id} - P_{\Delta t})^{-1}\varphi$ are not controlled
- Consider $\left(\Pi \dfrac{P_{\Delta t} - \mathrm{Id}}{\Delta t} \Pi\right) Q_{\Delta t}\psi = \psi + \Delta t^{p+1} \widetilde{r}_{\psi,\Delta t}$ where

$$\Pi\varphi = \varphi - \int_{\mathcal{X}} \varphi \, d\pi$$

- Idea of the construction: truncate the formal series expression

$$(A + \Delta t\, B)^{-1} = A^{-1} - \Delta t\, A^{-1} B A^{-1} + \Delta t^2\, A^{-1} B A^{-1} B A^{-1} + \dots$$

Step 3: Conclusion

- Write the invariances with $\Pi \left(\dfrac{P_{\Delta t} - \mathrm{Id}}{\Delta t}\right) \Pi\psi$ instead of $\left(\dfrac{P_{\Delta t} - \mathrm{Id}}{\Delta t}\right)\psi$
- Replace ψ by $Q_{\Delta t}\varphi$, and gather in $R_{\varphi,\Delta t}$ all the higher order terms

Examples of splitting schemes for Langevin dynamics (1)

- Example: Langevin dynamics, discretized using a splitting strategy

$$A = M^{-1}p\cdot\nabla_q, \quad B_\eta = \Big(-\nabla V(q) + \eta\, F\Big)\cdot\nabla_p, \quad C = -M^{-1}p\cdot\nabla_p + \frac{1}{\beta}\Delta_p$$

- Note that $\mathcal{L}_\eta = A + B_\eta + \gamma C$
- Trotter splitting $\to$ weak order 1

$$P_{\Delta t}^{ZYX} = \mathrm{e}^{\Delta t Z}\mathrm{e}^{\Delta t Y}\mathrm{e}^{\Delta t X} = \mathrm{e}^{\Delta t \mathcal{L}} + \mathrm{O}(\Delta t^2)$$

- Strang splitting $\to$ weak order 2

$$P_{\Delta t}^{ZYXYZ} = \mathrm{e}^{\Delta t Z/2}\mathrm{e}^{\Delta t Y/2}\mathrm{e}^{\Delta t X}\mathrm{e}^{\Delta t Y/2}\mathrm{e}^{\Delta t Z/2} = \mathrm{e}^{\Delta t \mathcal{L}} + \mathrm{O}(\Delta t^3)$$

- Other category: Geometric Langevin[5] algorithms, *e.g.* $P_{\Delta t}^{\gamma C, A, B_\eta, A}$
$\to$ weak order 1 but measure preserved at order 2 in Δt

[5] N. Bou-Rabee and H. Owhadi, *SIAM J. Numer. Anal.* (2010)

Examples of splitting schemes for Langevin dynamics (2)

- $P_{\Delta t}^{B_\eta,A,\gamma C}$ corresponds to $\begin{cases} \widetilde{p}^{n+1} = p^n + \Big(- \nabla V(q^n) + \eta F \Big) \Delta t, \\ q^{n+1} = q^n + \Delta t \, M^{-1} \widetilde{p}^{n+1}, \\ p^{n+1} = \alpha_{\Delta t} \widetilde{p}^{n+1} + \sqrt{\dfrac{1-\alpha_{\Delta t}^2}{\beta} M} \, G^n \end{cases}$

where G^n are i.i.d. Gaussian and $\alpha_{\Delta t} = \exp(-\gamma M^{-1} \Delta t)$

- $P_{\Delta t}^{\gamma C, B_\eta, A, B_\eta, \gamma C}$ for $\begin{cases} \widetilde{p}^{n+1/2} = \alpha_{\Delta t/2} p^n + \sqrt{\dfrac{1-\alpha_{\Delta t}}{\beta} M} \, G^n, \\ p^{n+1/2} = \widetilde{p}^{n+1/2} + \dfrac{\Delta t}{2} \Big(- \nabla V(q^n) + \eta F \Big), \\ q^{n+1} = q^n + \Delta t \, M^{-1} p^{n+1/2}, \\ \widetilde{p}^{n+1} = p^{n+1/2} + \dfrac{\Delta t}{2} \Big(- \nabla V(q^{n+1}) + \eta F \Big), \\ p^{n+1} = \alpha_{\Delta t/2} \widetilde{p}^{n+1} + \sqrt{\dfrac{1-\alpha_{\Delta t}}{\beta} M} \, G^{n+1/2} \end{cases}$

Error estimates on linear response

Error estimates for nonequilibrium dynamics

There exists a function $f_{\alpha,1,\gamma} \in H^1(\mu)$ such that

$$\int_{\mathcal{E}} \psi \, d\mu_{\gamma,\eta,\Delta t} = \int_{\mathcal{E}} \psi \Big(1 + \eta f_{0,1,\gamma} + \Delta t^\alpha f_{\alpha,0,\gamma} + \eta \Delta t^\alpha f_{\alpha,1,\gamma} \Big) d\mu + r_{\psi,\gamma,\eta,\Delta t},$$

where the remainder is compatible with linear response

$$|r_{\psi,\gamma,\eta,\Delta t}| \leqslant K(\eta^2 + \Delta t^{\alpha+1}), \qquad |r_{\psi,\gamma,\eta,\Delta t} - r_{\psi,\gamma,0,\Delta t}| \leqslant K\eta(\eta + \Delta t^{\alpha+1})$$

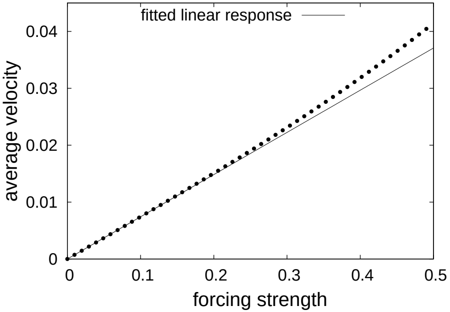
- Corollary: error estimates on the numerically computed mobility

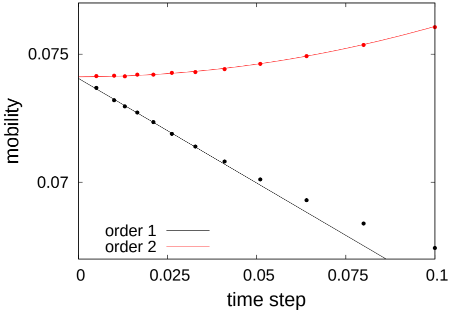
$$\rho_{F,\Delta t} = \lim_{\eta \to 0} \frac{1}{\eta} \left(\int_{\mathcal{E}} F^T M^{-1} p \, \mu_{\gamma,\eta,\Delta t}(dq\,dp) - \int_{\mathcal{E}} F^T M^{-1} p \, \mu_{\gamma,0,\Delta t}(dq\,dp) \right)$$
$$= \rho_F + \Delta t^\alpha \int_{\mathcal{E}} F^T M^{-1} p \, f_{\alpha,1,\gamma} \, d\mu + \Delta t^{\alpha+1} r_{\gamma,\Delta t}$$

- Results in the overdamped limit[6]

[6] B. Leimkuhler, C. Matthews and G. Stoltz, *IMA J. Numer. Anal.* (2015)

Numerical results





Left: Linear response of the average velocity as a function of η for the scheme associated with $P_{\Delta t}^{\gamma C, B_\eta, A, B_\eta, \gamma C}$ and $\Delta t = 0.01, \gamma = 1$.

Right: Scaling of the mobility $\nu_{F,\gamma,\Delta t}$ for the first order scheme $P_{\Delta t}^{A, B_\eta, \gamma C}$ and the second order scheme $P_{\Delta t}^{\gamma C, B_\eta, A, B_\eta, \gamma C}$.

Error estimates on Green-Kubo formulas (1)

- For methods of **weak order** 1, Riemman sum (ϕ, φ average 0 w.r.t. π)

$$\int_0^{+\infty} \mathbb{E}\Big(\phi(x_t)\varphi(x_0)\Big)dt = \Delta t \sum_{n=0}^{+\infty} \mathbb{E}_{\Delta t}\left(\Pi_{\Delta t}\phi\left(x^n\right)\varphi\left(x^0\right)\right) + \mathrm{O}(\Delta t)$$

$$\text{where } \Pi_{\Delta t}\phi = \phi - \int_{\mathcal{X}} \phi \, d\pi_{\Delta t}$$

- Correlation approximated in practice using K independent realizations

$$\mathbb{E}_{\Delta t}\left(\Pi_{\Delta t}\phi\left(x^n\right)\varphi\left(x^0\right)\right) \simeq \frac{1}{K}\sum_{m=1}^{K}\left(\phi(x^{n,k}) - \overline{\phi}^{n,K}\right)\left(\varphi(x^{n,k}) - \overline{\varphi}^{n,K}\right)$$

$$\text{where } \overline{\phi}^{n,K} = \frac{1}{K}\sum_{m=1}^{K}\phi(x^{n,k})$$

- For methods of **weak order** 2, trapezoidal rule

$$\int_0^{+\infty} \mathbb{E}\Big(\phi(x_t)\varphi(x_0)\Big)dt = \frac{\Delta t}{2}\mathbb{E}_{\Delta t}\left(\Pi_{\Delta t}\phi\left(x^0\right)\varphi\left(x^0\right)\right)$$

$$+ \Delta t \sum_{n=1}^{+\infty} \mathbb{E}_{\Delta t}\left(\Pi_{\Delta t}\phi\left(x^n\right)\varphi\left(x^0\right)\right) + \mathrm{O}(\Delta t^2)$$

Error estimates on Green-Kubo formulas (2)

- Error of order α on invariant measure: $\displaystyle \int_{\mathcal{X}} \psi \, d\pi_{\Delta t} = \int_{\mathcal{X}} \psi \, d\pi + \mathrm{O}(\Delta t^{\alpha})$
- Expansion of the evolution operator ($p+1 \geqslant \alpha$ and $\mathcal{A}_1 = \mathcal{L}$)

$$P_{\Delta t}\varphi = \varphi + \Delta t \, \mathcal{L}\varphi + \Delta t^2 \mathcal{A}_2 \varphi + \cdots + \Delta t^{p+1} \mathcal{A}_{p+1}\varphi + \Delta t^{p+2} r_{\varphi,\Delta t}$$

Ergodicity of the numerical scheme

$$\forall n \in \mathbb{N}, \qquad \|P_{\Delta t}^n\|_{\mathcal{B}(L^\infty_{\mathcal{K}_s,\Delta t})} \leqslant C_s \mathrm{e}^{-\lambda_s n \Delta t}$$

where $\mathcal{K}_s$ is a Lyapunov function ($1+|p|^{2s}$ for Langevin) and

$$L^\infty_{\mathcal{K}_s,\Delta t} = \left\{ \frac{\varphi}{\mathcal{K}_s} \in L^\infty(\mathcal{X}), \ \int_{\mathcal{X}} \varphi \, d\pi_{\Delta t} = 0 \right\}$$

- Proof: Lyapunov condition + uniform-in-Δt minorization condition[7]

[7] M. Hairer and J. Mattingly, *Progr. Probab.* (2011)

Error estimates on Green-Kubo formulas (3)

Error estimates on integrated correlation functions

Observables φ, ψ with average 0 w.r.t. invariant measure π

$$\int_0^{+\infty} \mathbb{E}\Big(\psi(x_t)\varphi(x_0)\Big)dt = \Delta t \sum_{n=0}^{+\infty} \mathbb{E}_{\Delta t}\left(\widetilde{\psi}_{\Delta t,\alpha}\left(x^n\right)\varphi\left(x^0\right)\right) + \Delta t^\alpha r_{\Delta t}^{\psi,\varphi},$$

where $\mathbb{E}_{\Delta t}$ denotes expectations w.r.t. initial conditions $x_0 \sim \pi_{\Delta t}$ and over all realizations of the Markov chain (x^n), and

$$\widetilde{\psi}_{\Delta t,\alpha} = \psi_{\Delta t,\alpha} - \int_{\mathcal{X}} \psi_{\Delta t,\alpha}\, d\pi_{\Delta t}$$

with $\psi_{\Delta t,\alpha} = \Big(\mathrm{Id} + \Delta t\, \mathcal{A}_2 \mathcal{L}^{-1} + \dots + \Delta t^{\alpha-1} \mathcal{A}_\alpha \mathcal{L}^{-1}\Big)\psi$

- Useful when $\mathcal{A}_k \mathcal{L}^{-1}$ can be computed, *e.g.* $\mathcal{A}_k = a_k \mathcal{L}^k$
- Reduces to trapezoidal rule for second order schemes

Sketch of proof (1)

- Define $\Pi_{\Delta t}\varphi = \varphi - \int_{\mathcal{X}} \varphi \, d\pi_{\Delta t}$
- Since $\mathcal{L}^{-1}\psi$ has average 0 w.r.t. π, introduce $\pi_{\Delta t}$ as

$$\int_{\mathcal{X}} \left(-\mathcal{L}^{-1}\psi\right) \varphi \, d\pi = \int_{\mathcal{X}} \left(-\mathcal{L}^{-1}\psi\right) \Pi_{\Delta t}\varphi \, d\pi = \int_{\mathcal{X}} \Pi_{\Delta t} \left(-\mathcal{L}^{-1}\psi\right) \Pi_{\Delta t}\varphi \, d\pi_{\Delta t} + \Delta t^{\alpha} r_{\Delta t}^{\psi,\varphi},$$

- Rewrite $-\Pi_{\Delta t}\mathcal{L}^{-1}$ in terms of $P_{\Delta t}$ as

$$\begin{aligned} -\Pi_{\Delta t}\mathcal{L}^{-1}\psi &= -\Pi_{\Delta t} \left(\Delta t \sum_{n=0}^{+\infty} P_{\Delta t}^n \right) \Pi_{\Delta t} \left(\frac{\mathrm{Id} - P_{\Delta t}}{\Delta t} \right) \mathcal{L}^{-1}\psi \\ &= \Delta t \left(\sum_{n=0}^{+\infty} \left[\Pi_{\Delta t} P_{\Delta t} \Pi_{\Delta t}\right]^n \right) \left(\mathcal{L} + \dots + \Delta t^{\alpha-1} S_{\alpha-1} + \Delta t^{\alpha} \widetilde{R}_{\alpha,\Delta t} \right) \mathcal{L}^{-1}\psi, \\ &= \Delta t \sum_{n=0}^{+\infty} \left[\Pi_{\Delta t} P_{\Delta t} \Pi_{\Delta t}\right]^n \widetilde{\psi}_{\Delta t,\alpha} + \Delta t^{\alpha} \left(\frac{\mathrm{Id} - P_{\Delta t}}{\Delta t} \right)^{-1} \Pi_{\Delta t} \widetilde{R}_{\alpha,\Delta t} \mathcal{L}^{-1}\psi. \end{aligned}$$

Sketch of proof (2)

- Uniform resolvent bounds $\left\| \left(\frac{\mathrm{Id} - P_{\Delta t}}{\Delta t} \right)^{-1} \right\|_{\mathcal{B}(L^\infty_{\mathcal{K}_s,\Delta t})} \leqslant \frac{C_s}{\lambda_s}$

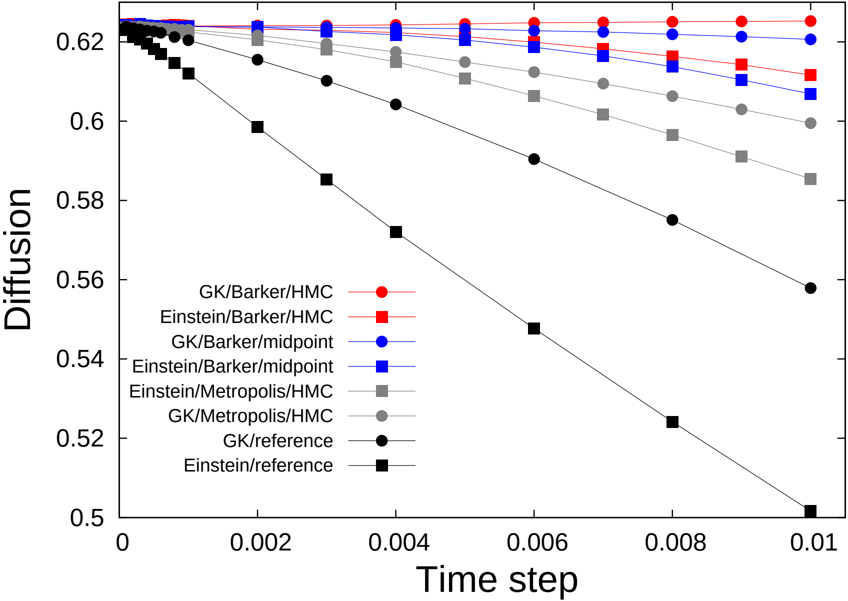
- Coming back to the initial equality,

$$\int_{\mathcal{X}} \left(-\mathcal{L}^{-1}\psi\right) \varphi \, d\pi = \Delta t \int_{\mathcal{X}} \sum_{n=0}^{+\infty} \left(\Pi_{\Delta t} P_{\Delta t}^n \widetilde{\psi}_{\Delta t,\alpha} \right) \left(\Pi_{\Delta t} \varphi \right) d\pi_{\Delta t} + \mathrm{O}\left(\Delta t^\alpha\right)$$

- Rewrite finally

$$\begin{aligned} \int_{\mathcal{X}} \sum_{n=0}^{+\infty} \left(\Pi_{\Delta t} P_{\Delta t}^n \widetilde{\psi}_{\Delta t,\alpha} \right) \left(\Pi_{\Delta t} \varphi \right) d\pi_{\Delta t} &= \int_{\mathcal{X}} \sum_{n=0}^{+\infty} \left(P_{\Delta t}^n \widetilde{\psi}_{\Delta t,\alpha} \right) \varphi \, d\pi_{\Delta t} \\ &= \sum_{n=0}^{+\infty} \mathbb{E}_{\Delta t} \left(\widetilde{\psi}_{\Delta t,\alpha}\left(q^n, p^n\right) \varphi\left(q^0, p^0\right) \right) \end{aligned}$$

Numerical results



Conclusion and perspectives

Main points: recall the outline!

- **Definition and examples of nonequilibrium systems**

- **Computation of transport coefficients**
  - a survey of computational techniques
  - linear response theory
  - relationship with Green-Kubo formulas

- **Elements of numerical analysis**
  - estimation of biases due to timestep discretization
  - (largely) open issue: variance reduction
  - (not discussed) use of non-reversible dynamics to enhance sampling

Variance reduction techniques?

- Importance sampling? Invariant probability measures ψ_∞, ψ_∞^A for

$$dq_t = b(q_t)\,dt + \sigma dW_t, \qquad dq_t = \Big(b(q_t) + \nabla A(q_t)\Big)dt + \sigma dW_t$$

In general $\psi_\infty^A \neq Z^{-1}\psi_\infty \mathrm{e}^A$ (consider $b(q) = F$ and $A = \widetilde{V}$)

- Stratification? (as in TI...) Consider $q \in \mathbb{T}^2$, $\psi_\infty = \mathbf{1}_{\mathbb{T}^2}$

$$\begin{cases} dq_t^1 = \partial_{q_2} U(q_t^1, q_t^2) + \sqrt{2}\,dW_t^1 \\ dq_t^2 = -\partial_{q_1} U(q_t^1, q_t^2) + \sqrt{2}\,dW_t^2 \end{cases}$$

Constraint $\xi(q) = q_2$, constrained dynamics

$$dq_t^1 = f(q_t^1)\,dt + \sqrt{2}\,dW_t^1, \qquad f(q^1) = \partial_{q_2} U(q^1, 0).$$

Then $\psi_\infty(q^1) = Z^{-1} \displaystyle\int_0^1 \mathrm{e}^{V(q^1+y) - V(q^1) - Fy}\,dy \neq \mathbf{1}_{\mathbb{T}}(q^1)$

where $F = \displaystyle\int_0^1 f$ and $V(q^1) = -\displaystyle\int_0^{q^1} (f(s) - F)\,ds$