STOCHASTIC NORTON DYNAMICS

Noé Blassel, Gabriel Stoltz

MATHERIALS Team, CERMICS, École des Ponts & Inria Paris

Introduction

Motivation: Estimate transport coefficients (mobility, thermal conductivity, shear viscosity).

Framework: Stochastic dynamics out of equilibrium.

Core idea: Make the flux the independent state variable instead of the forcing.

Hope: Derive estimators with better statistical properties (lower asymptotic variance).

General definition

Usual NEMD or "Thévenin" dynamics:

$$\mathrm{d}X_t^\eta = b(X_t^\eta)\,\mathrm{d}t + \sigma(X_t^\eta)\,\mathrm{d}W_t + \eta F(X_t^\eta)\,\mathrm{d}t,$$

Reference (equilibrium) dynamics: $\eta = 0$.

Norton dynamics:

$$\begin{cases} \mathrm{d}Y_t^r = b(Y_t^r)\,\mathrm{d}t + \sigma(Y_t^r)\,\mathrm{d}W_t + F(Y_t^r)\,\mathrm{d}\Lambda_t^r, \\ R(Y_t^r) = R(Y_0^r) = r. \end{cases}$$

Constrain the response flux R to be constant: dynamics on a submanifold of phase space.

Forcing process:

$$\mathrm{d}\Lambda_t^r = \lambda_t^r\,\mathrm{d}t + \mathrm{d}\widetilde{\Lambda}_t^r$$

with $\widetilde{\Lambda}^r$ a martingale.

Transport coefficients

Thévenin:

$$\rho_{F,R} = \lim_{\eta\to 0} \frac{\mathbb{E}_\eta\left[R\right]}{\eta}.$$

where $\mathbb{E}_\eta$ is expectation with respect to steady-state of Thévenin dynamics.

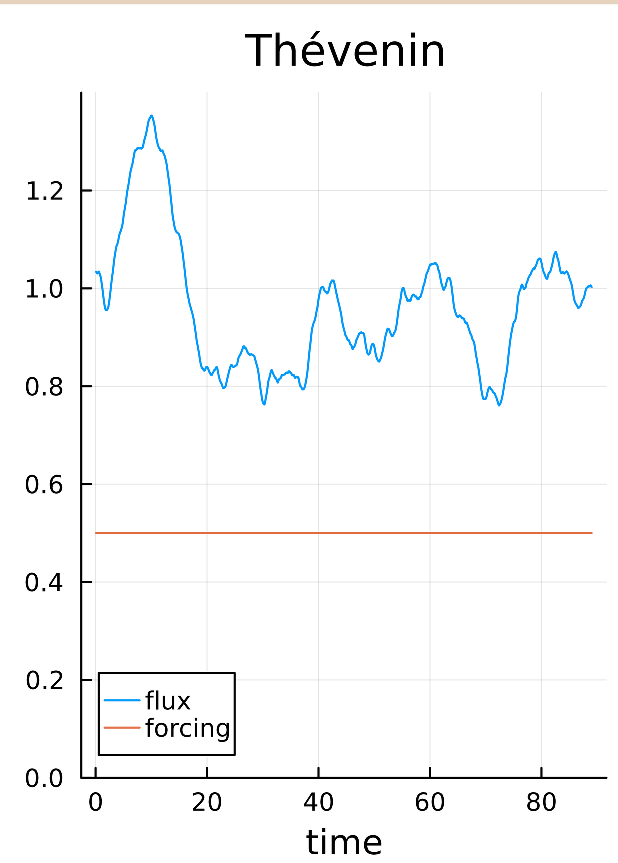
Norton

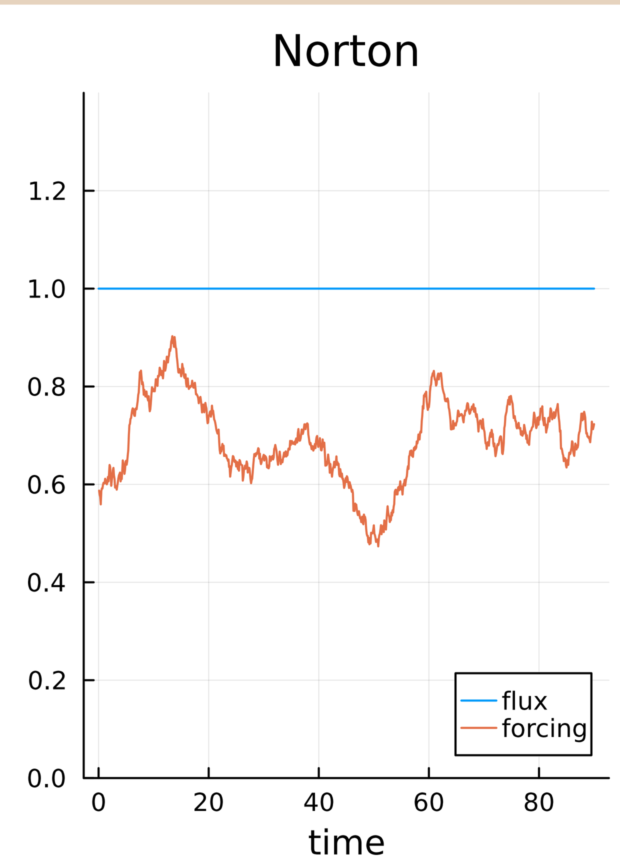
$$\widetilde{\rho}_{F,R} = \lim_{r\to 0} \frac{r}{\mathbb{E}_r^*\left[\lambda\right]},$$

where $\mathbb{E}_r^*$ is expectation with respect to steady-state of Norton dynamics, λ is the forcing.

In practice, these quantities are estimated through trajectory averages.

Illustration





Closed-form

The dynamics can be written

$$\mathrm{d}Y_t^r = \overline{P}_{F,\nabla R}(Y_t^r)\left[b(Y_t^r)\mathrm{d}t + \sigma(Y_t^r)\mathrm{d}W_t\right] - \frac{\left(\nabla^2 R : \Pi_{F,\nabla R,\sigma}\right)(Y_t^r)}{2\nabla R(Y_t^r)\cdot F(Y_t^r)} F(Y_t^r)\,\mathrm{d}t,$$

with $\overline{P}_{F,\nabla R}(y)$ a non-orthogonal projector, and

$$\Pi_{F,\nabla R,\sigma}(y) = \left[\overline{P}_{F,\nabla R}\sigma\sigma^\intercal \overline{P}_{F,\nabla R}^\intercal\right](y).$$

Non-martingale part of the forcing: $\lambda_t = \lambda(Y_t^r)$, with

$$\lambda(y) = \left[-\frac{1}{F\cdot\nabla R}\left(b\cdot\nabla R + \frac{1}{2}\nabla^2 R : \Pi_{F,\nabla R,\sigma}\right)\right](y).$$

Controllability condition: $F\cdot\nabla R \neq 0$.

Generalizations: Time-dependent flux $R(Y_t^r) = r_t$, multiple flux constraints $\mathbf{R}(Y_t^r) = \mathbf{r} \in \mathbb{R}^c$, also explicit.

Langevin setting

Take $R(q,p)$ of the form $G(q)\cdot p$, adapted for mobility and shear viscosity computations. **Norton dynamics:**

$$\begin{cases} \mathrm{d}q_t = M^{-1}p_t\,\mathrm{d}t, \\ \mathrm{d}p_t = \overline{P}_{F,G}(q_t)\left(-\nabla V(q_t)\,\mathrm{d}t - \gamma M^{-1}p_t\,\mathrm{d}t + \sqrt{\frac{2\gamma}{\beta}}\,\mathrm{d}W_t\right) + \frac{\nabla G(q_t)p_t\cdot M^{-1}p_t}{F(q_t)\cdot G(q_t)}F(q_t)\,\mathrm{d}t. \end{cases}$$

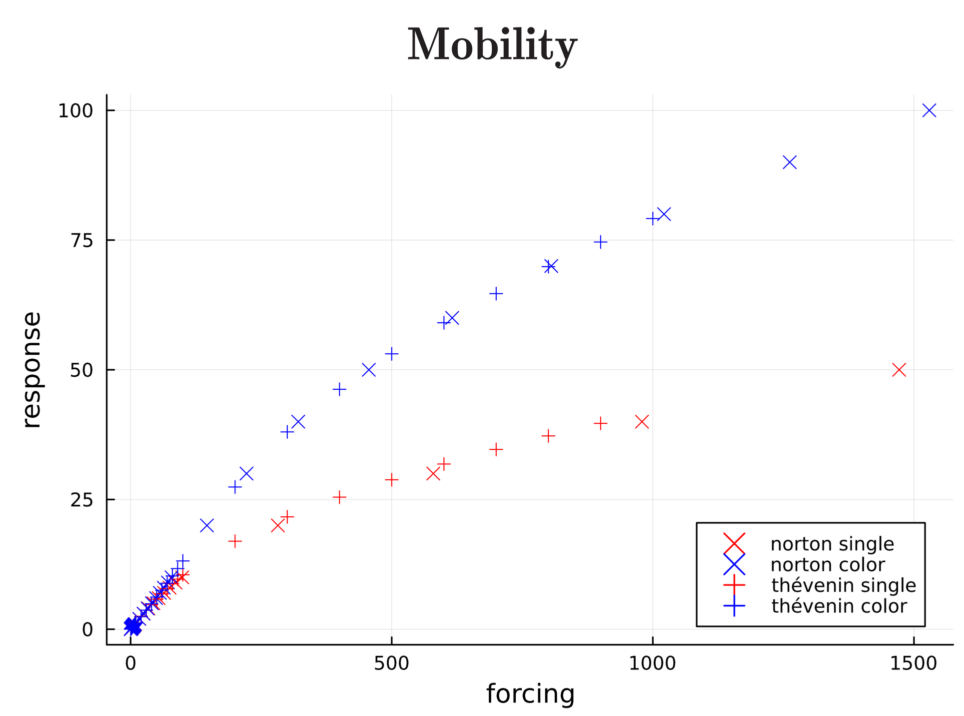
Mobility: $F\in\mathbb{R}^{dN}$, $G = M^{-1}F$.

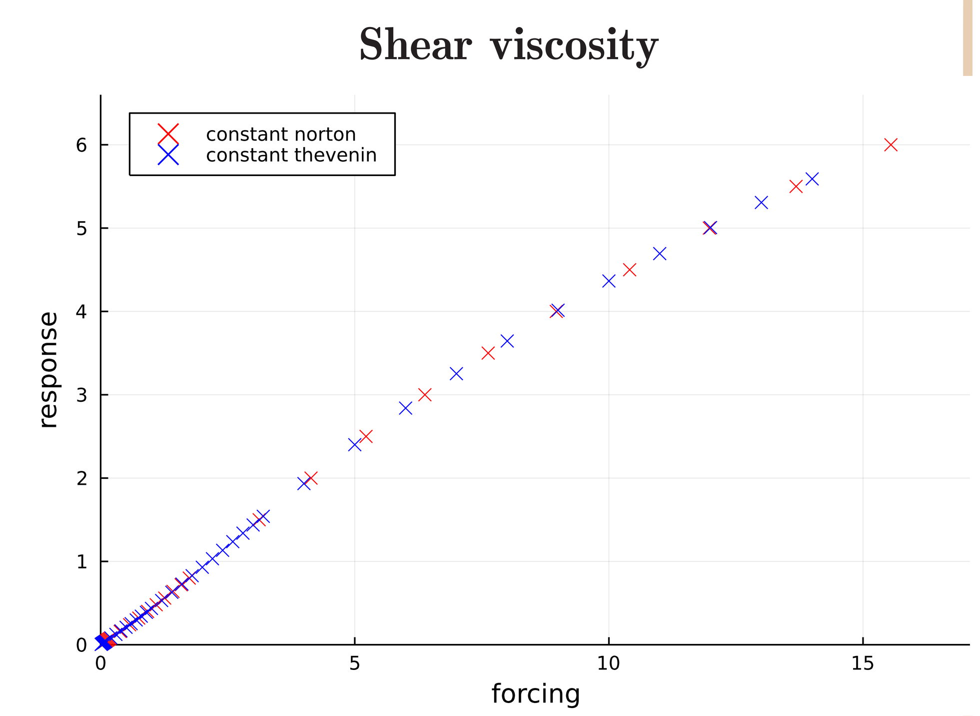
Shear-viscosity:

$$\forall\, 1\leq i\leq N,\ \forall\, 2\leq\alpha\leq d,\ F(q)_{i1} = f_y(q_{i2}),\ G(q)_{i1} = \frac{1}{m}\exp\left(\frac{2i\pi q_{i2}}{L}\right),\ F(q)_{i\alpha} = G(q)_{i\alpha} = 0.$$

This corresponds to a longitudinal force with a transverse intensity profile. The response is the Fourier coefficient of the transverse profile for the longitudinal velocity.

Equivalence of responses

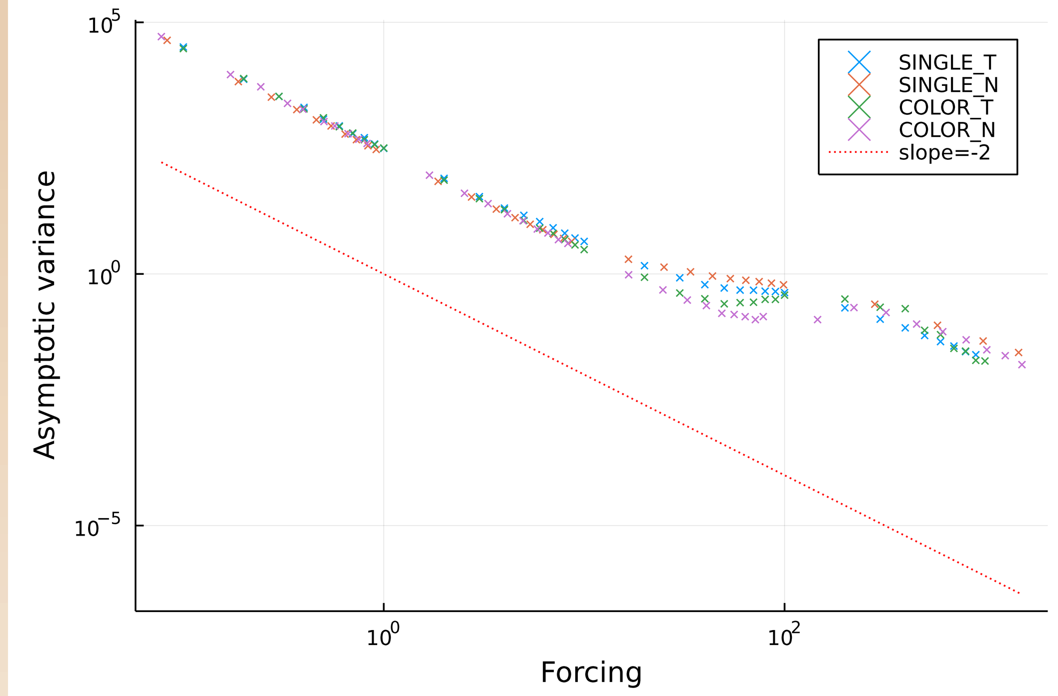




The Thévenin and Norton responses coïncide far outside of the linear response regime, and give the same transport coefficient. Computations were performed on a Lennard-Jones fluid of 1000 particles using a dedicated flux-preserving splitting implemented using the Julia package Molly.

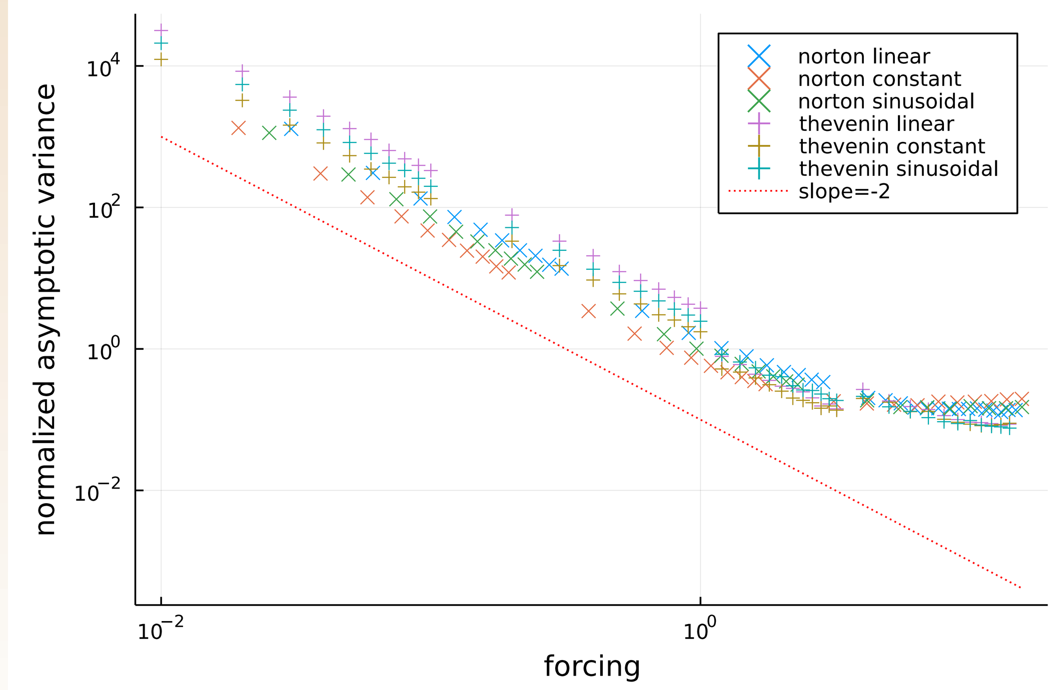
Asymptotic variances

Mobility



No gain in the case of mobility

Shear-Viscosity



Norton estimators systematically outperform their Thévenin counterparts.

Future questions

- Existence and ergodicity results for the solutions to the Norton dynamics.
- Equivalence of Thévenin and Norton ensembles.
- Linear response theory/ Green-Kubo relations for the Norton ensemble.

Bibliography

[1] Evans, Denis and Hoover, William G. and Failor, Bruce H. and Moran, Bill and Ladd, Anthony J.C. Nonequilibrium molecular dynamics via Gauss's principle of least constraint *Physical Review A*, 1983.

[2] Joubaud, Rémi and Stoltz, Gabriel Nonequilibrium Shear Viscosity Computations with Langevin Dynamics *Multiscale Modeling & Simulation*, 2016.

[3] Evans, Denis The equivalence of Norton and Thévenin ensembles *Molecular Physics*,1993.