Extending the regime of linear response with synthetic forcings

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General setting

Notation

- The measure μ has associated density ψ
- $\mathcal{A}^\dagger$ denotes the L^2-adjoint of the operator $\mathcal{A}$, i.e.

$$\int (\mathcal{A}\varphi)\phi = \int \varphi(\mathcal{A}^\dagger \phi), \quad \forall \varphi, \phi \in C_0^\infty$$

- $\mathcal{A}^*$ denotes the $L^2(\psi)$-adjoint of the operator $\mathcal{A}$, i.e.

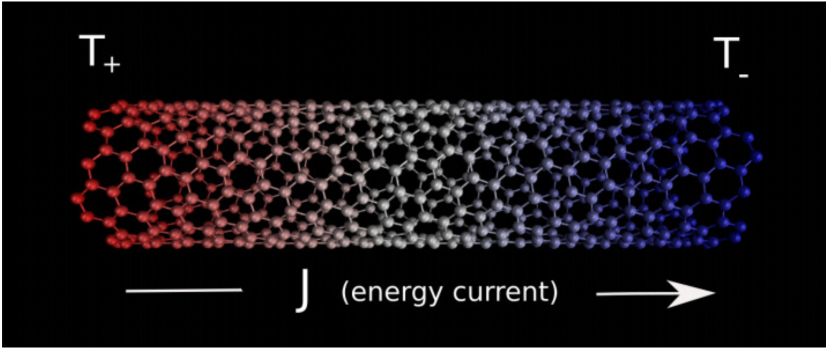
$$\int (\mathcal{A}\varphi)\phi\, \psi = \int \varphi(\mathcal{A}^* \phi)\, \psi, \quad \forall \varphi, \phi \in C_0^\infty$$

Physical context and motivation

Transport coefficients are **quantitative** estimates.

Example (thermal conductivity): Fourier's law

$$J = -\kappa \nabla T$$



Long computational times to estimate κ; can take up to weeks/months.

Nonequilibrium framework

Example: State-space $\mathcal{X} = \mathbb{T}^d$, periodic C^∞ potential V

Overdamped Langevin dynamics

$$dq_t = (-\nabla V(q_t) + \eta F(q_t))\, dt + \sqrt{\frac{2}{\beta}}\, dW_t$$

Generator $\mathcal{L}_\eta = \mathcal{L}_0 + \eta \widetilde{\mathcal{L}}_{\text{phys}}$, with

$$\mathcal{L}_0 = -\nabla V^T \nabla + \beta^{-1}\Delta, \qquad \widetilde{\mathcal{L}}_{\text{phys}} = F^T \nabla$$

Invariant probability measure ψ_η solves the Fokker–Planck

$$\mathcal{L}_\eta^\dagger \psi_\eta = 0, \qquad \psi_0(q) = \frac{1}{Z} e^{-\beta V(q)}$$

The perturbation $\eta F(q)$ induces a response $\mathbb{E}_\eta(R)$ for some observable $R(q)$. For small η, the response is linear in η.

Assumptions

1. Uniqueness of the invariant probability measure, trajectorial ergodicity.

$$\frac{1}{t}\int_0^t R(q_s^\eta)\,ds \xrightarrow[t\to+\infty]{\text{a.s.}} \mathbb{E}_\eta(R) := \int_{\mathcal{X}} R\,\psi_\eta$$

2. Lyapunov estimates

3. Stability of *regular* functions by inverse operators

4. Stability of *regular* functions by the perturbation operator

Regular: smooth functions that (and whose derivatives) grow at most polynomially

Expansion of the invariant measure

Although unique, ψ_η has closed form unknown

Perturbative regime:[1] Write ψ_η as $f_\eta \psi_0$. In powers of η,

$$f_\eta = \mathbf{1} + \eta \mathfrak{f}_1 + \eta^2 \mathfrak{f}_2 + \cdots$$
$$\psi_\eta = \psi_0 + \eta \overline{\psi}_1 + \eta^2 \overline{\psi}_2 + \cdots$$

Formal asymptotics on the Fokker–Planck $(\mathcal{L}_0 + \eta \widetilde{\mathcal{L}}_{\text{phys}})^\dagger \psi_\eta = 0$ leads to

$$-\mathcal{L}_0^* \mathfrak{f}_1 = \widetilde{\mathcal{L}}^*_{\text{phys}} \mathbf{1}, \qquad \mathfrak{f}_{n+1} = \left(-\mathcal{L}_0^*\right)^{-1} \widetilde{\mathcal{L}}^*_{\text{phys}} \mathfrak{f}_n$$

Remark: Converging infinite expansion requires some regularity results on $\widetilde{\mathcal{L}}_{\text{phys}}$ and $\mathcal{L}_0$.

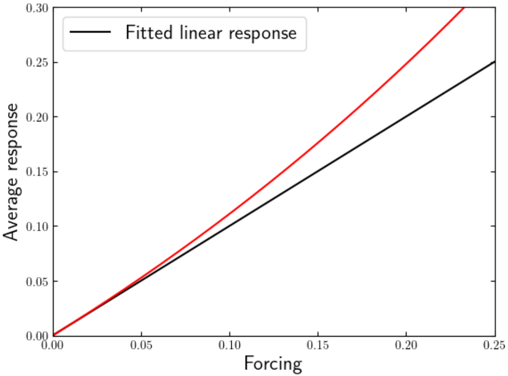
[1] T. Lelièvre and G. Stoltz, *Acta Numerica* **25**, (2016)

Linear response of nonequilibrium stochastic dynamics

Linear response (1)

Linear response is used to compute transport coefficients. It is defined as

$$\rho_1 = \lim_{\eta \to 0} \frac{\mathbb{E}_\eta(R) - \mathbb{E}_0(R)}{\eta} = \lim_{\eta \to 0} \frac{\mathbb{E}_\eta(R)}{\eta}$$



Example: For constant F and $R(q) = F^T \nabla V$, ρ_1 represents the mobility.

Linear response (2)

Since $\psi_\eta = f_\eta \psi_0$, write the linear response as

$$\rho_1 = \lim_{\eta \to 0} \frac{1}{\eta} \int_{\mathcal{X}} R\, \psi_\eta = \lim_{\eta \to 0} \frac{1}{\eta} \int_{\mathcal{X}} R f_\eta\, \psi_0 = \int_{\mathcal{X}} R \mathfrak{f}_1\, \psi_0$$

More generally, the n-th order response is given by

$$\rho_n = \int_{\mathcal{X}} R \mathfrak{f}_n\, \psi_0$$

Full response can be written as a polynomial in η

$$r(\eta) = \mathbb{E}_\eta(R) = \eta \rho_1 + \eta^2 \rho_2 + \eta^2 \rho_3 + \cdots$$

Linear response (3)

Estimator of linear response

$$\widehat{\rho}_{\eta,t} = \frac{1}{\eta t}\int_0^t R(q_s)\,ds \xrightarrow[t\to+\infty]{\text{a.s.}} \frac{1}{\eta}\int_{\mathcal{X}} R f_\eta\,\psi_0 = \rho_1 + O(\eta)$$

Main issue: Statistical error with asymptotic variance $O(\eta^{-2})$

Idea: Reduce variance by increasing η, but at which values of η does the nonlinear response develop?

Goal: Want to stay in the linear regime for η as large as possible.

Optimizing the perturbation
Synthetic forcings

Optimizing the perturbation

Idea: Introduce perturbation with generator $\widetilde{\mathcal{L}}_{\text{extra}}$, where

$$\widetilde{\mathcal{L}}^*_{\text{extra}}\mathbf{1} = 0$$

Resulting dynamics has generator

$$\mathcal{L}_\eta = \mathcal{L}_0 + \eta(\widetilde{\mathcal{L}}_{\text{phys}} + \alpha\widetilde{\mathcal{L}}_{\text{extra}})$$

The resulting perturbation is called a **synthetic forcing** [2].

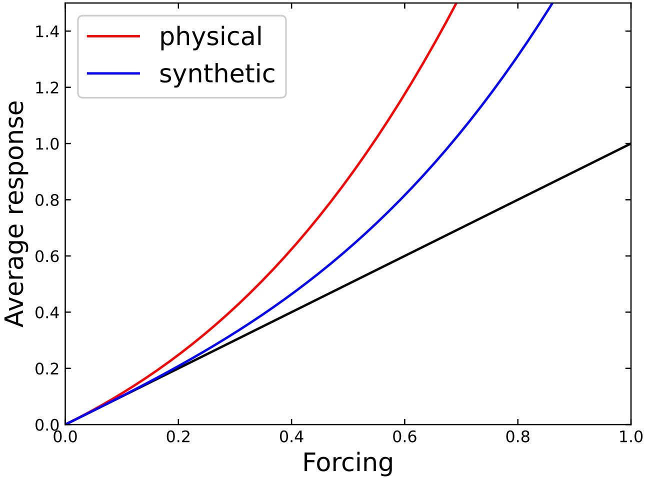
With the addition of the extra forcing, $\mathfrak{f}_1$ remains unchanged

$$\begin{aligned}\mathfrak{f}_1 &= -\mathcal{L}_0^{-1}(\widetilde{\mathcal{L}}_{\text{phys}} + \alpha\widetilde{\mathcal{L}}_{\text{extra}})^*\mathbf{1} \\ &= -\mathcal{L}_0^{-1}\widetilde{\mathcal{L}}^*_{\text{phys}}\mathbf{1}\end{aligned}$$

[2] Evans, Morriss, (2008)

Formulation of synthetic forcing

In theory, we can find some $\widetilde{\mathcal{L}}_{\text{extra}}$ that extends the linear regime



Classes of synthetic forcings

1. *First-order differential operators* $\widetilde{\mathcal{L}}_{\text{extra}} = G^T \nabla_x$, with $\operatorname{div}(G\psi_0) = 0$.

2. *Second-order differential operators* of the form $\widetilde{\mathcal{L}}_{\text{extra}} = -\partial_{x_i}^* \partial_{x_i}$. In this case, the operator is self-adjoint, *i.e.* $\widetilde{\mathcal{L}}_{\text{extra}} = \widetilde{\mathcal{L}}_{\text{extra}}^*$.

3. *First-order differential operators with nontrivial zero order parts*, such as $\widetilde{\mathcal{L}}_{\text{extra}} = \partial_{x_i}^* = \partial_{x_i} U - \partial_{x_i}$ for $\psi_0(x) = e^{-U(x)}$.

Examples of synthetic forcings (overdamped Langevin)

Example 1: Divergence-free vector field

$$\begin{cases} \widetilde{\mathcal{L}}_{\text{extra}} = G(q)^T \nabla, \quad \text{such that} \quad \operatorname{div}(G e^{-\beta V}) = 0 \\ dq_t = -\nabla V(q_t)\, dt + \eta F(q_t)\, dt + \eta \alpha G(q_t)\, dt + \sigma\, dW \end{cases}$$

Example 2: Modified fluctuation-dissipation

$$\begin{cases} \widetilde{\mathcal{L}}_{\text{extra}} = -\beta^{-1} \nabla^* \nabla = -\nabla V^T \nabla + \beta^{-1} \Delta \\ dq_t = -(1 + \alpha\eta) \nabla V(q_t)\, dt + \eta F(q_t)\, dt + \sqrt{\dfrac{2(1+\alpha\eta)}{\beta}} dW \end{cases}$$

Example 3: Feynman–Kac forcing

$$\begin{cases} \widetilde{\mathcal{L}}_{\text{extra}} = \xi(q)^T \nabla^* = -\xi^T \nabla + \xi^T \nabla V \\ dq_t = -\nabla V(q_t)\, dt + \eta F(q_t)\, dt - \eta \alpha \xi(q_t)\, dt + \sigma dW \\ \omega_t \propto \exp\left(\alpha\eta \displaystyle\int_0^t \xi^T V(q_s)\, ds \right) \end{cases}$$

Examples of synthetic forcings (Langevin dynamics)

Example 1: Divergence-free vector field $\widetilde{\mathcal{L}}_{\text{extra}} = G_1^T \nabla_q + G_2^T \nabla_p$

$$\begin{cases} dq_t = M^{-1} p_t \, dt + \alpha\eta G_1(q_t, p_t) \, dt, \\ dp_t = -\nabla V(q_t) \, dt + \eta(F(q_t) + \alpha G_2(q_t, p_t)) \, dt - \gamma M^{-1} p_t \, dt + \sqrt{\frac{2\gamma}{\beta}} \, dW_t. \end{cases}$$

Example 2: Modified fluctuation-dissipation $\widetilde{\mathcal{L}}_{\text{extra}} = -\beta^{-1} \nabla_q^* \nabla_q - \beta^{-1} \nabla_p^* \nabla_p$

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

Example 3: Feynman–Kac forcing $\widetilde{\mathcal{L}}_{\text{extra}} = \xi_1(p)^T \nabla_q^* + \xi_2(q)^T \nabla_p^*$

$$\begin{cases} dq_t = M^{-1} p_t \, dt - \alpha\eta \xi_1(p_t) \, dt, \\ dp_t = -\nabla V(q_t) \, dt + \eta(F(q_t) - \alpha \xi_2(q_t)) \, dt - \gamma M^{-1} p_t \, dt + \sqrt{\frac{2\gamma}{\beta}} \, dW_t, \end{cases}$$

Choosing magnitude of perturbation (1)

Recall: Response as a polynomial in η

$$r_\alpha(\eta) = \mathbb{E}_{\eta,\alpha}(R) = \eta\rho_1 + \eta^2\rho_2(\alpha) + \eta^2\rho_3(\alpha) + \cdots$$

Goal: Choose α s.t. the second-order response is killed.

With $\widetilde{\mathcal{L}}_{\text{extra}}$, $\mathfrak{f}_2$ is given by

$$\mathfrak{f}_2 = \mathcal{L}_0^{-1}(\widetilde{\mathcal{L}}_{\text{phys}} + \alpha\widetilde{\mathcal{L}}_{\text{extra}})^* \mathcal{L}_0^{-1} \widetilde{\mathcal{L}}^*_{\text{phys}} \mathbf{1}$$

Recall: Second-order response $\rho_2 = \int R\mathfrak{f}_2\,\psi_0$, so solve $\rho_2(\alpha^\star) = 0$

$$\rho_2 = \int_{\mathcal{X}} R\mathfrak{f}_{2,\text{phys}} + \alpha \int_{\mathcal{X}} R\mathfrak{f}_{2,\text{extra}} \implies \boxed{\alpha^* = \frac{\int R\mathfrak{f}_{2,\text{phys}}\psi_0}{\int R\mathfrak{f}_{2,\text{extra}}\psi_0}}$$

Choosing magnitude of perturbation (2)

Consider the relative error δ in the response relative to the linear response:

$$\delta_\alpha(\eta) = \left| \frac{r_\alpha(\eta) - \rho_1 \eta}{\rho_1 \eta} \right| .$$

Given some deviation ε, pick α such that

$$\alpha_\star(\varepsilon) = \arg\max_{\alpha \in \mathbb{R}} \eta_\alpha(\varepsilon), \qquad \eta_\alpha(\varepsilon) = \arg\min_{\eta \in \mathbb{R}} \left\{ |\eta| : \delta_\alpha(\eta) \geq \varepsilon \right\} .$$

In other words, stay within some neighborhood of the linear response for as long as possible.

Numerical illustration

Choosing the observable

Setting: Overdamped Langevin dynamics in 1D, potential $V(q) = \cos(2\pi q)$. We consider the observable

$$R(q) = (a\cos(2\pi q) + b\sin(2\pi q))\, e^{\beta V(q)}$$

where $a, b \in \mathbb{R}$.

- b is chosen such that $\rho_1 = 1$, i.e.,

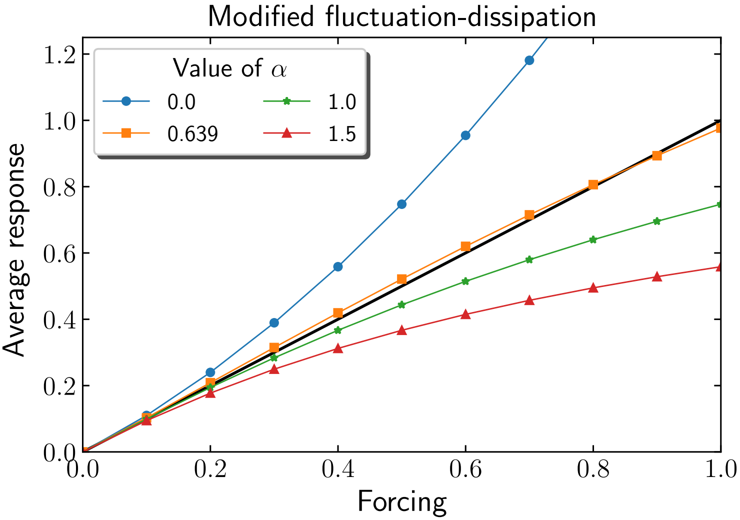
$$b = \left(Z^{-1} \int_{\mathcal{X}} \sin(2\pi q)\mathfrak{f}_1(q)\, dq \right)^{-1}, \quad Z = \int_{\mathcal{X}} e^{-\beta V(q)} dq$$

- Similarly, a can be chosen such that ρ_2 is arbitrarily large.

For nonsymmetric potentials, the above still gives $\rho_1, \rho_2 \sim O(1)$.

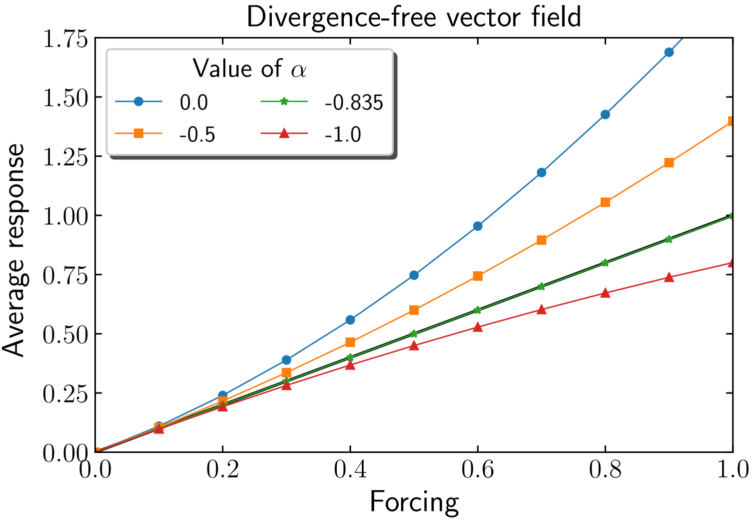
Numerical illustration: modified FD

$$\alpha^\star = 1.0, \qquad \alpha_\star(\varepsilon = 0.05) = 0.639$$



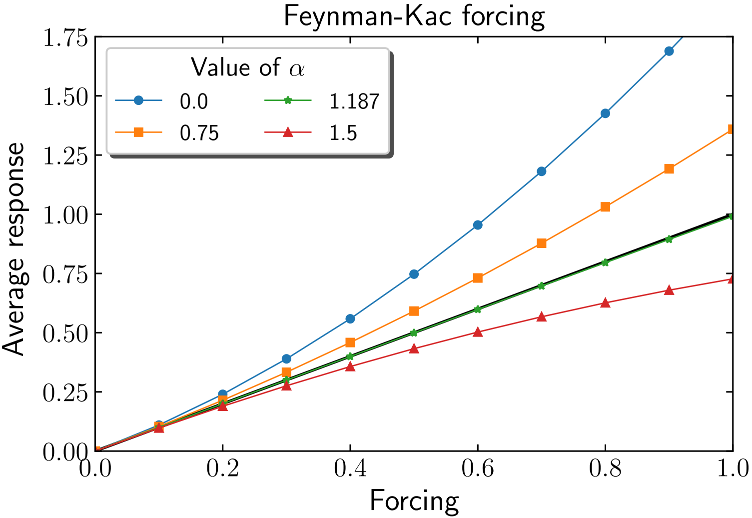
Numerical illustration: div-free vector field

$$\alpha^\star = -0.835$$

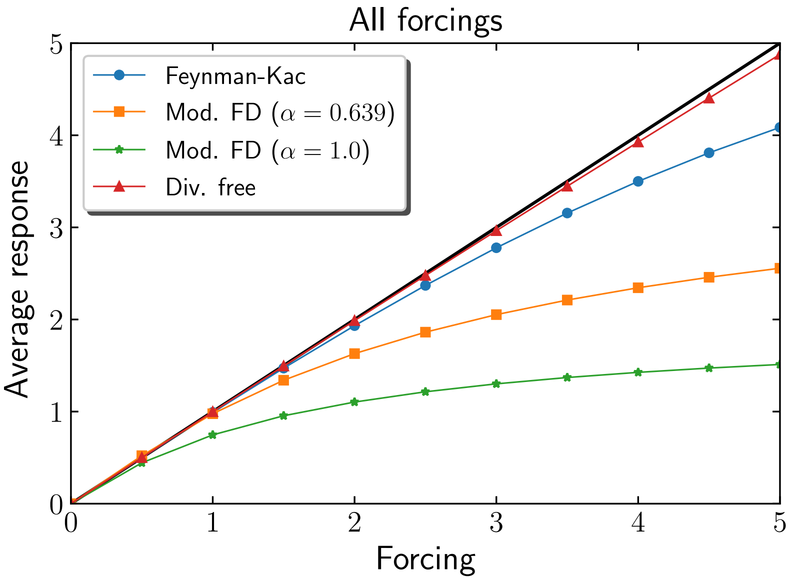


Numerical illustration: Feynman–Kac forcing

$$\alpha^\star = 1.187$$



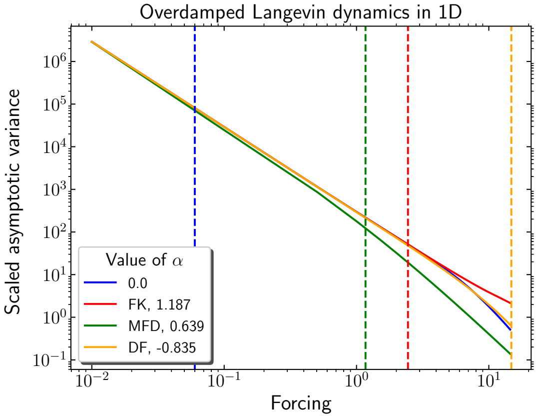
Numerical illustration: all forcings



Numerical illustration: variance reduction (1)

Asymptotic variance $\sigma^2_{R,\eta}$ same order as $\sigma^2_{R,0}$, up to small bias $O(\eta)$:

$$\frac{\sigma^2_{R,\eta}}{\eta^2} = \frac{\sigma^2_{R,0}}{\eta^2} + O\left(\frac{1}{\eta}\right)$$



Numerical illustration: variance reduction (2)

Define the gain as

$$\text{gain} = \left(\frac{\sigma^2_{R,\eta_0(\varepsilon)}}{\eta_0(\varepsilon)^2} \right) \left(\frac{\sigma^2_{R,\eta_\alpha(\varepsilon)}}{\eta_\alpha(\varepsilon)^2} \right)^{-1}.$$

Dynamics	**Extra forcing**				
	none	MFD	FK	DF (e^V)	DF ($A\nabla V$)
Ovd. 1D	1	6.56×10^2	1.58×10^3	1.28×10^5	-
Ovd. 2D	1	7.65×10^2	3.23×10^4	3.33×10^3	4.03×10^0
Lang. 1D	1	2.18×10^3	1.29×10^3	1.42×10^3	-

Future work and extensions

- **Promising method:** Can reduce variance by several orders of magnitude
- **Potential applications:** Lennard–Jones fluids (shear viscosity), systems of atom chains (thermal transport)
- **Extension to actual MD systems:** Transport coefficients are *intensive quantities*, i.e. not dependent on system size

  $\implies$ Notion of *preescreening*: two small simulations with $\alpha_1 \neq \alpha_2$, from which optimal α can be extrapolated