Institut des sciences
du calcul et des données
SORBONNE UNIVERSITÉ

MAESTRO

Learning the dynamics of systems with memory : Generalized Langevin equations

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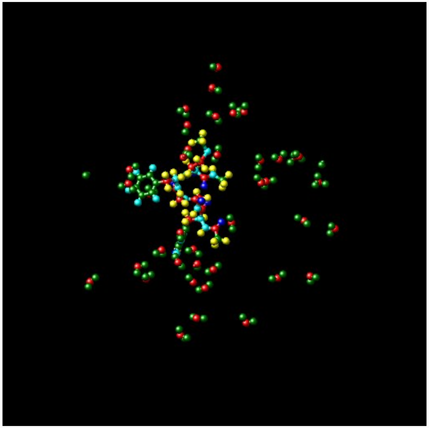
14 Juin 2022

Maestro (MAterials for Energy through STochastic sampling and high peRformance cOmputing), Institut des Sciences du Calcul et des Données

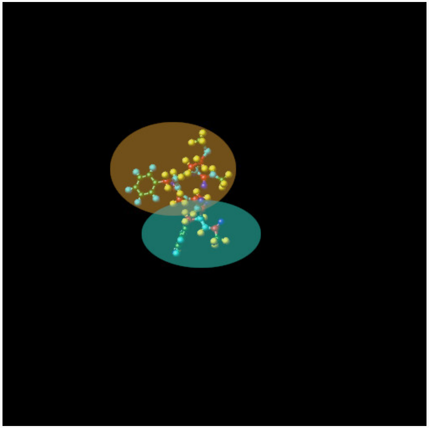
Dimensionality reduction

Tackle high dimensional configurational space with a low-dimensional projection.

- Choice of collective variables?
- *Effective dynamics?*



From all atoms simulations



To reduced dynamical models

- Speed up in computational times (multiscale dynamics)
- Better interpretability (less dimensions)

Maestro (MAterials for Energy through STochastic sampling and high peRformance cOmputing)

The MAESTRO project aims at a conceptual and operational breakthrough in computational materials science, particularly in the study of materials for energy applications and storage.

Collaboration between mathematicians, physicists and chemists @ ISCD, including Ludovic Goudenège, Pierre Monmarché, Fabio Pietrucci and Benjamin Rotenberg.

Prediciting thermodynamics (free energy) and kinetics (rates, timescales) properties of energy storage materials from numerical simulations.

Overview

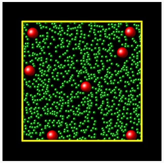
1) **The Generalized Langevin Equation (GLE): here comes the memory**

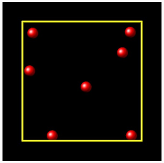
2) **Infering the dynamics?**

3) **Expectation maximization algorithm**

4) **Examples**

The Generalized Langevin Equation (GLE): here comes the memory



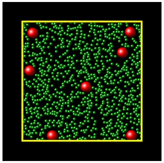


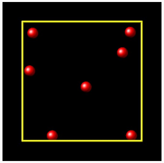
Dynamical (Hamilton) Equations:

$$\begin{cases} \dot{x}_{\text{red}} &= v_{\text{red}} \\ \dot{v}_{\text{red}} &= F_{r\to r}(x_{\text{red}}) + F_{g\to r}(x_{\text{red}}, x_{\text{green}}) \end{cases} \quad \begin{cases} \dot{x}_{\text{green}} &= v_{\text{green}} \\ \dot{v}_{\text{green}} &= F_{g\to g}(x_{\text{green}}) + F_{r\to g}(x_{\text{red}}, x_{\text{green}}) \end{cases}$$

Let's focus on red particles as collective variables.

Can we remove green particles variables? Yes, using Mori Zwanzig projection formalism.





Generalized Langevin equation[1]:

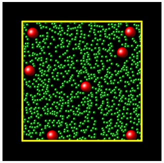
$$\dot{x}_{\mathrm{red}} = v_{\mathrm{red}}$$

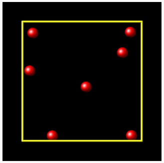
$$\dot{v}_{\mathrm{red}} = \widetilde{F}(x_{\mathrm{red}}) - \int_0^t K(\tau) v_{\mathrm{red}}(t-\tau)\mathrm{d}\tau + \xi(t)$$

The solvant is modeled via

- a non-Markovian memory (friction) kernel $K(\tau)$ (dissipation of energy)
- a diffusion term $\xi(t)$ (random noise coming from collision). This is a function of initial conditions

[1] Zwanzig, 2001





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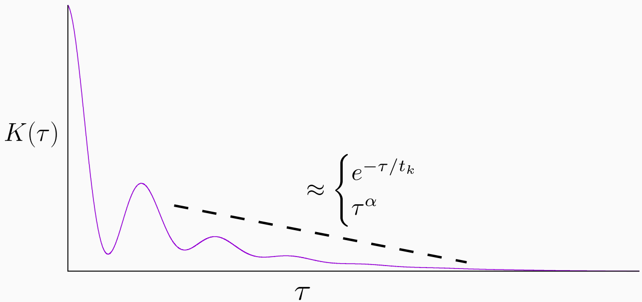
Position-dependent form of the Generalized Langevin Equations for Hamiltonian systems[2]:

$$\dot{v}(t) = \widetilde{F}(x(t)) - \int_0^t K(\tau, x(t-\tau)) v(t-\tau)\mathrm{d}\tau + \xi(t)$$

[1] Zwanzig, 2001 [2] Vroylandt and Monmarché 2022, arXiv:2201.02457

What are we modeling with this memory? Dynamical timescales of the rest of the system



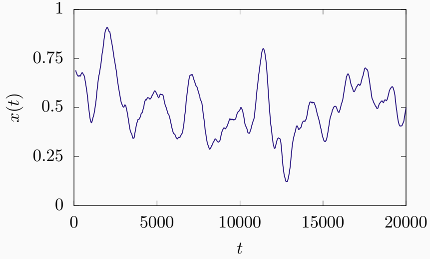
We can have "short" or "long" memory. We focus on "short" memory.
$K(\tau) = \delta(\tau) \Leftrightarrow$ Langevin description

Infering the dynamics?

The inference question

You are given:

Collective variable trajectories:



fit

Model: Generalized Langevin equation:

$$\dot{x} = v$$

$$\dot{v} = F(x) - \int_0^t K(\tau) v(t-\tau)\mathrm{d}\tau + \xi(t)$$

How do you infer the parameters in this equation from simulation data?

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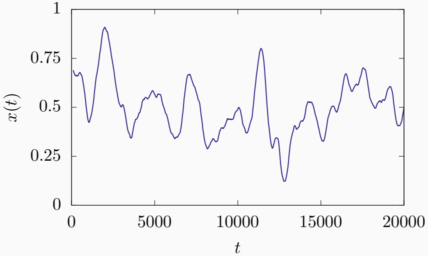

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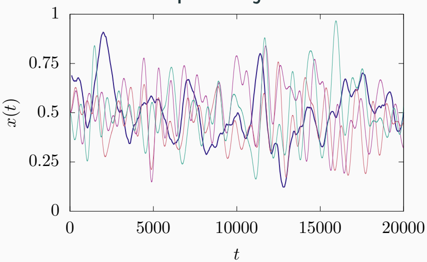
sample

New sample trajectories:



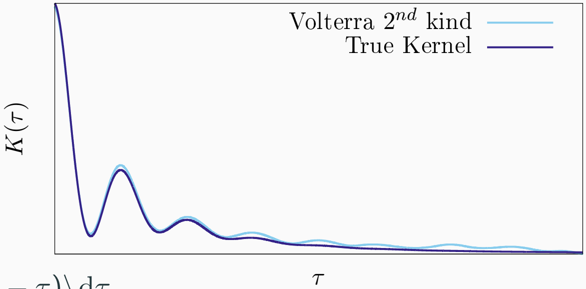
State of the art for memory fitting

Most of current approaches derive from GLE

$$\dot{v} = \widetilde{F}(x) - \int_0^t K(\tau)v(t-\tau)\mathrm{d}\tau + \xi(t)$$

a Volterra integral equation for the memory kernel

$$\left\langle v(0)\left(\dot{v}(t) - \widetilde{F}(x)\right)\right\rangle = -\int_0^t K(\tau)\left\langle v(0)v(t-\tau)\right\rangle \mathrm{d}\tau$$



That can be solved using

- Laplace transform[1]
- Integral discretization[2]
- ...

[a]Lei, Baker & Li, PNAS 2016, 113

[b]Daldrop, Kappler, Brünig, & Netz PNAS, 2018, 115

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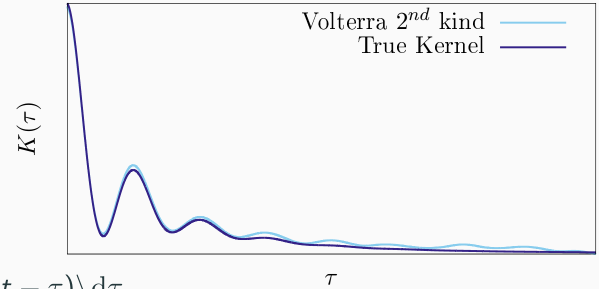

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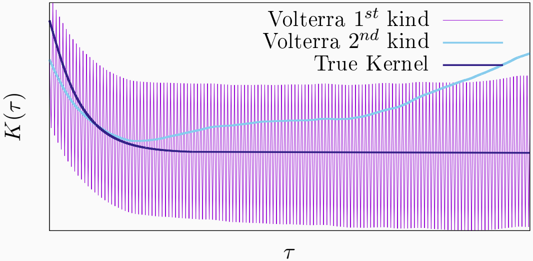
But

- This does not give a generative model
- Convergence issue for some cases



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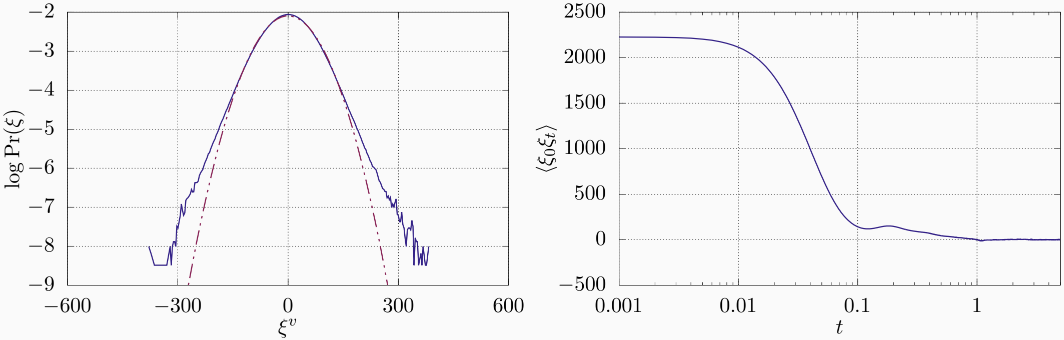
And the noise?

The noise is obtained from the inversion of

$$\dot{x} = v$$

$$\dot{v} = F(x) - \int_0^t K(\tau)v(t-\tau)\mathrm{d}\tau + \xi(t)$$

It exhibits non Gaussian tails and correlation in time



Auxiliary/hidden variables approach

Generalized Langevin equation:

$$\begin{cases} \dot{x} &= v \\ \dot{v} &= F(x) - \int_0^t K(\tau) v(t-\tau) \mathrm{d}\tau + \xi(t) \end{cases}$$

$$\begin{cases} \dot{\boldsymbol{x}} &= \boldsymbol{v} \\ \dot{\boldsymbol{v}} &= \boldsymbol{F}(\boldsymbol{x}) - \boldsymbol{A}_{vh}\, \boldsymbol{h} - \boldsymbol{A}_{vv} \boldsymbol{v} + \boldsymbol{\sigma}_v\, \boldsymbol{W}_v(t) \\ \dot{\boldsymbol{h}} &= -\boldsymbol{A}_{hh}\, \boldsymbol{h} - \boldsymbol{A}_{hv} \boldsymbol{v} + \boldsymbol{\sigma}_h\, \boldsymbol{W}_h(t) \end{cases}$$

Aiming to capture most important missed degrees of freedom.

Auxiliary/hidden variables approach

Generalized Langevin equation:

Hidden variable

$$\begin{cases} \dot{x} &= v \\ \dot{v} &= F(x) - \int_0^t K(\tau) v(t-\tau) \mathrm{d}\tau + \xi(t) \end{cases}$$

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Gaussian white noise

Auxiliary/hidden variables approach

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Aiming to capture most important missed degrees of freedom.

Equivalent equations?
Formal solution to last equation:

$$\boldsymbol{h}(t) = \int_0^t e^{-\boldsymbol{A}_{hh}\tau}\boldsymbol{A}_{hv}\boldsymbol{v}(t-\tau)\mathrm{d}\tau + \int_0^t e^{-\boldsymbol{A}_{hh}\tau}\boldsymbol{\sigma}_h\mathrm{d}\boldsymbol{W}_h(\tau)$$

Memory kernel is the sum of a Dirac and a Prony series:

$$K(\tau) = A_{vv}\delta(\tau) + \sum_i^{\dim h} c_i e^{-\lambda_i\tau}$$

Auxiliary/hidden variables approach

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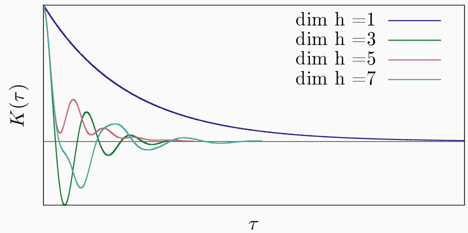
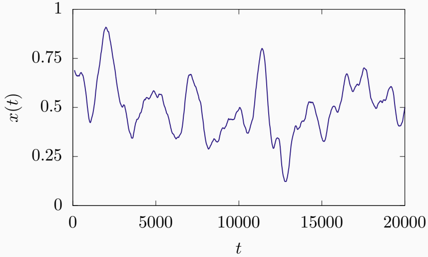


- GLE with memory kernel ⇒ Non-markovian but explicit
- Langevin equations with auxiliary variables ⇒ Markovian but hidden parts

The inference question (bis)

You are given:

Collective variable trajectories:



Model: Generalized Langevin equation with hidden variables

$$\begin{cases} \dot{\boldsymbol{x}} &= \boldsymbol{v} \\ \dot{\boldsymbol{v}} &= \boldsymbol{F}(\boldsymbol{x}) - \boldsymbol{A}_{vh}\boldsymbol{h} - \boldsymbol{A}_{vv}\boldsymbol{v} + \boldsymbol{\sigma}_v \boldsymbol{W}_v(t) \\ \dot{\boldsymbol{h}} &= -\boldsymbol{A}_{hh}\boldsymbol{h} - \boldsymbol{A}_{hv}\boldsymbol{v} + \boldsymbol{\sigma}_h \boldsymbol{W}_h(t) \end{cases}$$

How do you infer the parameters in this equation from simulation data?

How do you generate new trajectories from our inferred dynamics

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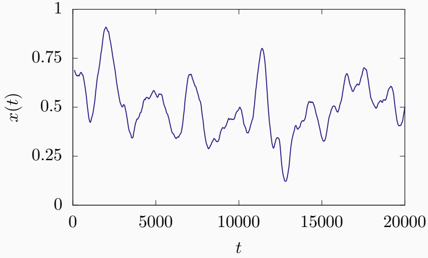

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How do you infer the parameters in this equation from simulation data?

Expectation maximization algorithm.

- Iterative algorithm to deal with hidden variables
- E-step: expectation (recovering hidden variables history)
- M-step: maximization of likelihood (recovering parameters)

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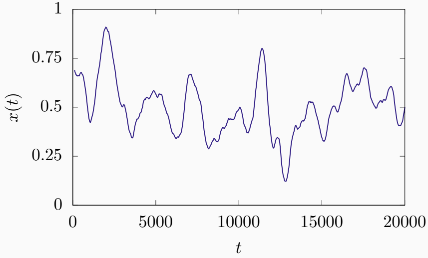

Model: Generalized Langevin equation with hidden variables

$$
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$$

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How do you generate new trajectories from our inferred dynamics

Once the parameters are fitted, this is a generative model.

Expectation maximization algorithm

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Discretizing the dynamics
We use a numerical scheme that discretizes the Langevin equation and a parametrization $\boldsymbol{\Theta} = \{F, \boldsymbol{A}, \boldsymbol{\sigma}\}$ of the (force, friction, diffusion) coefficients.

$$(\boldsymbol{X}_{t+\Delta t}, \boldsymbol{h}_{t+\Delta t}) = (\boldsymbol{X}_t, \boldsymbol{h}_t) + G_\Theta(\boldsymbol{X}_t, \boldsymbol{h}_t) + \boldsymbol{S}_{\boldsymbol{\Theta}} \boldsymbol{W}(t)$$

We separate between visible $\boldsymbol{X}_t$ and hidden $\boldsymbol{h}_t$ variables

Transition probability
Hence we have a *gaussian* transition probability

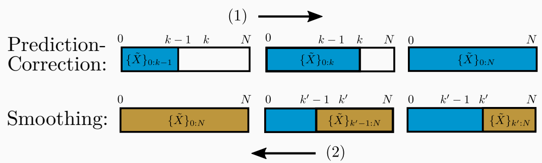
$$\mathbb{P}\left(\boldsymbol{X}_{t+\Delta t}, \boldsymbol{h}_{t+\Delta t} | \boldsymbol{X}_t, \boldsymbol{h}_t\right)_{\boldsymbol{\Theta}} = \mathcal{N}\left((\boldsymbol{X}_t, \boldsymbol{h}_t) + G_\Theta(\boldsymbol{X}_t, \boldsymbol{h}_t), \boldsymbol{S}_{\boldsymbol{\Theta}} \boldsymbol{S}_{\boldsymbol{\Theta}}^{\mathrm{T}}\right)$$

E-step: Reconstructing hidden variables

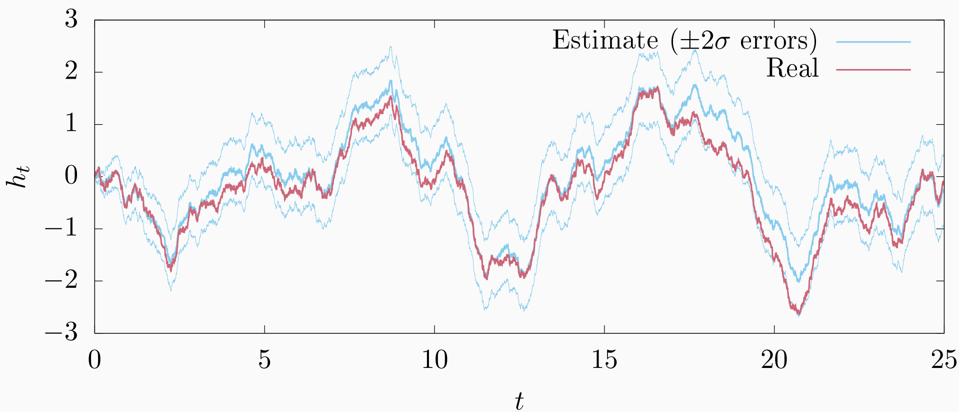
If you know the parameters: Markovianity of the system $\Rightarrow$ We use a predictor-corrector-smoother (or Kalman filer and Rauch smoother)

Using evolution equation

$$\begin{pmatrix} \boldsymbol{X}_{t+\Delta t} \\ \boldsymbol{h}_{t+\Delta t} \end{pmatrix} = \begin{pmatrix} \boldsymbol{X}_t \\ \boldsymbol{h}_t \end{pmatrix} + \boldsymbol{G}_{\Theta}(\boldsymbol{X}_t, \boldsymbol{h}_t) + \boldsymbol{S}_{\Theta} \boldsymbol{W}(t)$$



You get probabilistic reconstruction of the hidden variables $\mathbb{P}\left(\{\boldsymbol{h}_t\}_0^T | \{\boldsymbol{X}_t\}_0^T, \boldsymbol{\Theta}\right)$



Expectation-Maximization algorithm

The probability of a trajectory is

$$\mathbb{P}\left(\{\boldsymbol{X}_t, \boldsymbol{h}_t)\}_0^T | \boldsymbol{\Theta}\right) = \mathbb{P}\left((\boldsymbol{X}_0, \boldsymbol{h}_0)\right) \prod_{t=0}^{T} \mathbb{P}\left((\boldsymbol{X}_{t+\Delta t}, \boldsymbol{h}_{t+\Delta t} | (\boldsymbol{X}_t, \boldsymbol{h}_t)\right)_{\boldsymbol{\Theta}}$$

When dealing with hidden variables, we average over possible hidden variables values

$$\boldsymbol{\Theta}^* = \arg\max_{\boldsymbol{\Theta}} \int \mathrm{d}\{\boldsymbol{h}_t\}_0^T \mathbb{P}\left(\{\boldsymbol{h}_t\}_0^T | \{\boldsymbol{X}_t\}_0^T, \boldsymbol{\Theta}\right) \log \mathcal{L}(\{\boldsymbol{X}_t, \boldsymbol{h}_t\}_0^T; \boldsymbol{\Theta})$$

$$\boldsymbol{\Theta}^* = \arg\max_{\boldsymbol{\Theta}} F(\mathbb{P}\left(\{\boldsymbol{h}_t\}; \boldsymbol{\Theta}\right); \boldsymbol{\Theta})$$

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Algorithm - Iterative strategy

- Initialize parameters at some value $\boldsymbol{\Theta}_0$

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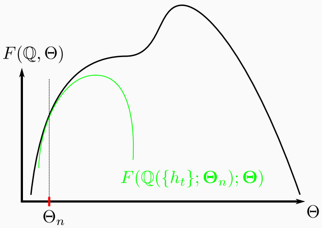
- E-step: Compute expectation of the log-likelihood over hidden variables $F(\mathbb{Q}\left(\{\boldsymbol{h}_t\}; \boldsymbol{\Theta}_n\right); \boldsymbol{\Theta})$



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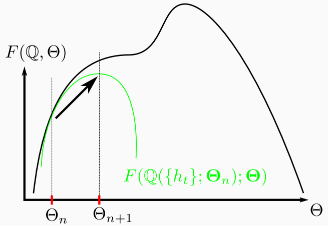
- E-step: Compute expectation of the log-likelihood over hidden variables $F(\mathbb{Q}\left(\{\boldsymbol{h}_t\}; \boldsymbol{\Theta}_n\right); \boldsymbol{\Theta})$
- M-step: Maximize the expectation of the log-likelihood



Expectation-Maximization algorithm

The probability of a trajectory is

$$\mathbb{P}\left(\{\boldsymbol{X}_t, \boldsymbol{h}_t)\}_0^T | \boldsymbol{\Theta}\right) = \mathbb{P}\left((\boldsymbol{X}_0, \boldsymbol{h}_0)\right) \prod_{t=0}^{T} \mathbb{P}\left((\boldsymbol{X}_{t+\Delta t}, \boldsymbol{h}_{t+\Delta t} | (\boldsymbol{X}_t, \boldsymbol{h}_t)\right)_{\boldsymbol{\Theta}}$$

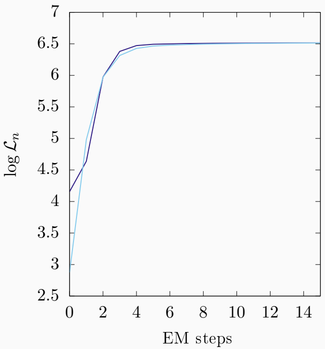
When dealing with hidden variables, we average over possible hidden variables values

$$\boldsymbol{\Theta}^* = \arg\max_{\boldsymbol{\Theta}} \int \mathrm{d}\{\boldsymbol{h}_t\}_0^T \mathbb{P}\left(\{\boldsymbol{h}_t\}_0^T | \{\boldsymbol{X}_t\}_0^T, \boldsymbol{\Theta}\right) \log \mathcal{L}(\{\boldsymbol{X}_t, \boldsymbol{h}_t\}_0^T; \boldsymbol{\Theta})$$

$$\boldsymbol{\Theta}^* = \arg\max_{\boldsymbol{\Theta}} F(\mathbb{P}(\{\boldsymbol{h}_t\}; \boldsymbol{\Theta}); \boldsymbol{\Theta})$$

Algorithm - Iterative strategy

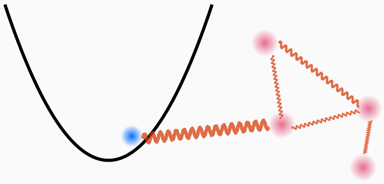
- Initialize parameters at some value $\boldsymbol{\Theta}_0$
- E-step: Compute expectation of the log-likelihood over hidden variables $F(\mathbb{Q}(\{\boldsymbol{h}_t\}; \boldsymbol{\Theta}_n); \boldsymbol{\Theta})$
- M-step: Maximize the expectation of the log-likelihood
- Iterate until convergence



Examples

Validation of the algorithm

- Quadratic potential
- 5 hidden dimensions
- Random value for the friction matrix A with an hierarchical structure.
- 20 trajectories of 25.10^3 timesteps.



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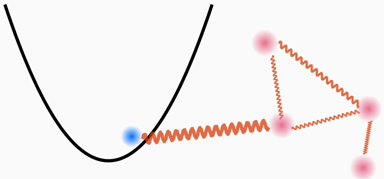
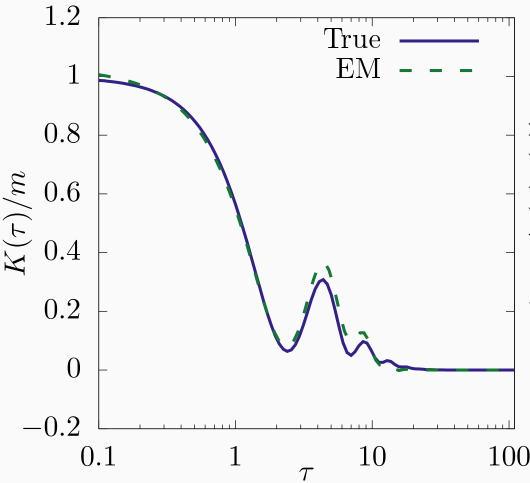
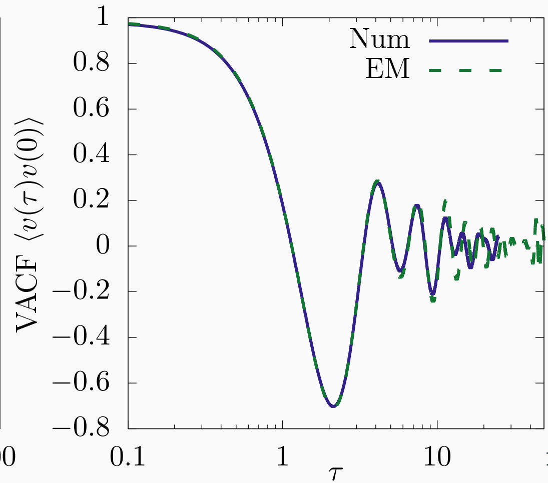
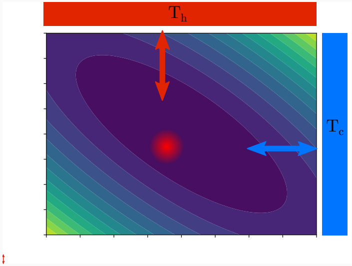






Non-equilibrium 2D setup[1]

- Quadratic potential with two different temperatures in x and y directions.
- 2 hidden dimensions
- Random value for the friction matrix A.
- 20 trajectories of 30.10^3 timesteps.



[1] Mancois, Marcos, Viot, and Wilkowski, 2018

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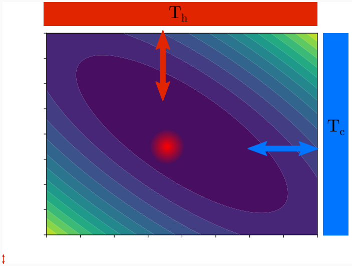

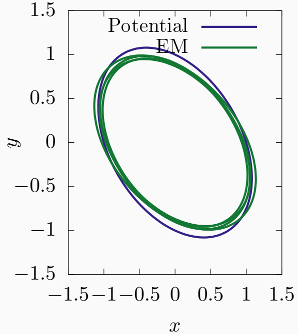

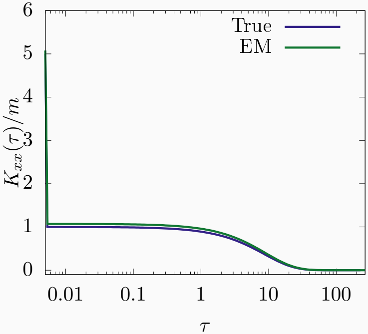

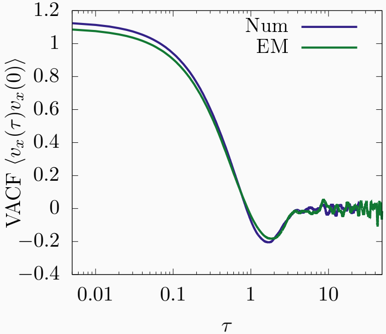

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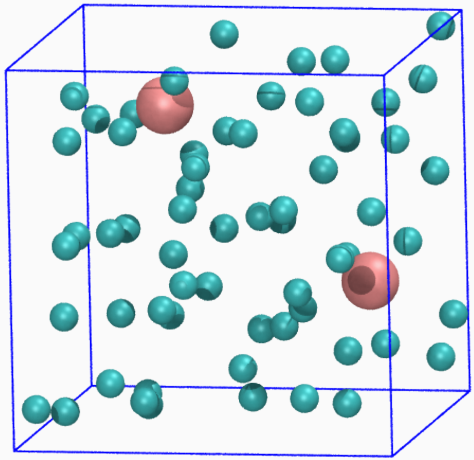
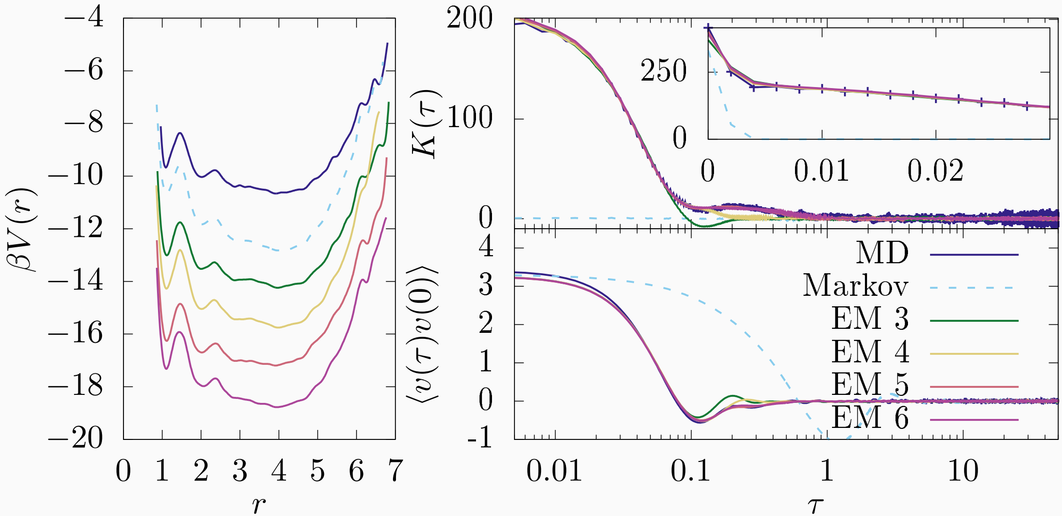
Lennard-Jones Dimer

A molecular dynamics example. We set up 512 LJ particles in a 3D box with periodic boundary conditions. 2 of the particles form a dimer.



Distance r between the two dimer particles as collective variable.

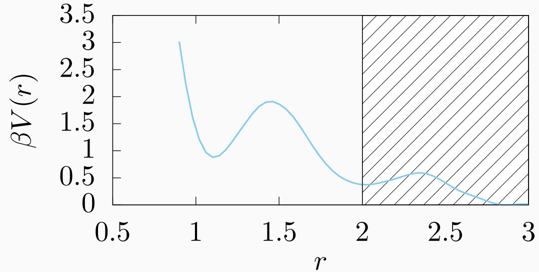
Lennard-Jones Dimer

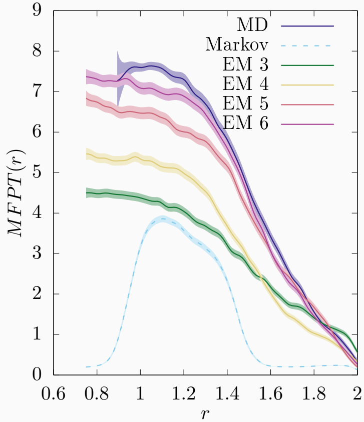


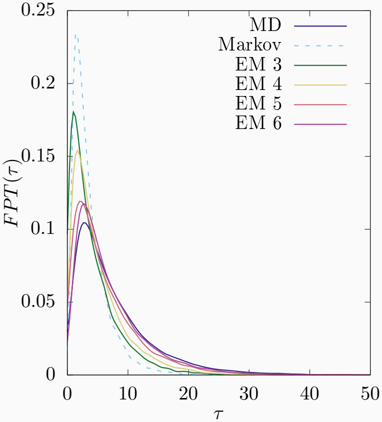
Generate 50 trajectories of 2×100000 timesteps from the fitted model.

We can change the timestep if wanted.

First passage time







Discussion

- This method fits the full statistical model for the dynamics (force + friction + noise).
- Generative model $\Rightarrow$ Possibility to use the inferred dynamics in future computations.
- Works for multidimensionnal and non equilibrium systems.
- Can be quite long to run...

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Thank you for your attention and questions?

Vroylandt, Hadrien, Ludovic Goudenège, Pierre Monmarché, Fabio Pietrucci, and Benjamin Rotenberg.*Likelihood-based non-Markovian models from molecular dynamics*. PNAS, 2022, 119

Hadrien Vroylandt and Pierre Monmarché. *Position-dependent memory kernel in generalized Langevin equations: theory and numerical estimation.* arXiv:2201.02457 (accepted in Journal of Chemical Physics)

Harmonic case

Let's assume that our green particles are harmonic particles, harmonically coupled to the red ones.

$$\begin{cases} \dot{x}_{\text{red}} &= v_{\text{red}} \\ \dot{v}_{\text{red}} &= F_{r\to r}(x_{\text{red}}) + \sum_j \gamma_j (x^j_{\text{green}} - \frac{\gamma_j}{\omega_j^2} x_{\text{red}}) \end{cases} \quad \begin{cases} \dot{x}^j_{\text{green}} &= v^j_{\text{green}} \\ \dot{v}^j_{\text{green}} &= -\omega_j^2 x^j_{\text{green}} + \gamma_j x_{\text{red}} \end{cases}$$

The motion of the green particles is easily solved (Second order differential equation + integration by part):

$$x^j_{\text{green}}(t) - \frac{\gamma_j}{\omega_j^2} x_{\text{red}}(t) = x^j_{\text{green}}(0) \cos \omega_j t - \gamma_j \int_0^t v_{\text{red}}(t-s) \frac{\cos \omega_j(s)}{\omega_j} \mathrm{d}s$$

This lead to the equation of motion for the red particules

$$\begin{cases} \dot{x}_{\text{red}} &= v_{\text{red}} \\ \dot{v}_{\text{red}} &= F_{r\to r}(x_{\text{red}}) - \int_0^t v_{\text{red}}(t-s) \sum_j \frac{\cos \omega_j(s)}{\omega_j} \mathrm{d}s + \sum_j x^j_{\text{green}}(0) \cos \omega_j t \end{cases}$$

Details: Derivation of GLE

Projection operator
For a given basis function family $\{b\}$, we define the projector as

$$\mathcal{P}_{\{b\}} F = B \cdot \langle B, B \rangle^{-1} \cdot \langle B, F \rangle$$

with scalar product $\langle f, g \rangle = \int \mathrm{d}\boldsymbol{X} \rho_{eq}(\boldsymbol{X}) f^{\mathrm{T}}(\boldsymbol{X}) g(\boldsymbol{X})$

The evolution equation of our observables are given by

Evolution equation

$$\frac{\partial \mathcal{O}(\boldsymbol{X}, t)}{\partial t} = \mathcal{L}\mathcal{O}(\boldsymbol{X}, t) = e^{t\mathcal{L}}\mathcal{L}\mathcal{O}(\boldsymbol{X}, 0) = e^{t\mathcal{L}}\mathcal{P}_{\{b\}}\mathcal{L}\mathcal{O}(\boldsymbol{X}, 0) + e^{t\mathcal{L}}(1 - \mathcal{P}_{\{b\}})\mathcal{L}\mathcal{O}(\boldsymbol{X}, 0)$$

Duhamel-Dyson identity:$e^{t\mathcal{L}} = e^{t(1-\mathcal{P}_{\{b\}})\mathcal{L}} + \int \mathrm{d}s e^{(t-s)\mathcal{L}} \mathcal{P}_{\{b\}} \mathcal{L} e^{s(1-\mathcal{P}_{\{b\}})\mathcal{L}}$

$$\begin{aligned} \frac{\partial \mathcal{O}(\boldsymbol{X}, t)}{\partial t} =& e^{t\mathcal{L}}\mathcal{P}_{\{b\}}\mathcal{L}\mathcal{O}(\boldsymbol{X}, 0) + \int \mathrm{d}s e^{(t-s)\mathcal{L}} \mathcal{P}_{\{b\}} \mathcal{L} e^{s(1-\mathcal{P}_{\{b\}})\mathcal{L}} (1 - \mathcal{P}_{\{b\}})\mathcal{L}\mathcal{O}(\boldsymbol{X}, 0) \\ &+ e^{t(1-\mathcal{P}_{\{b\}})\mathcal{L}}(1 - \mathcal{P}_{\{b\}})\mathcal{L}\mathcal{O}(\boldsymbol{X}, 0) \\ \frac{\partial \mathcal{O}(\boldsymbol{X}, t)}{\partial t} =& e^{t\mathcal{L}}\mathcal{P}_{\{b\}}\mathcal{L}\mathcal{O}(\boldsymbol{X}, 0) + \int \mathrm{d}s e^{(t-s)\mathcal{L}} \mathcal{P}_{\{b\}} \mathcal{L} F(\boldsymbol{X}, s) + F(\boldsymbol{X}, t) \end{aligned}$$

with $F(\boldsymbol{X}, t) = e^{t(1-\mathcal{P}_{\{b\}})\mathcal{L}}(1 - \mathcal{P}_{\{b\}})\mathcal{L}\mathcal{O}(\boldsymbol{X}, 0)$