Parareal algorithms for molecular dynamics simulations

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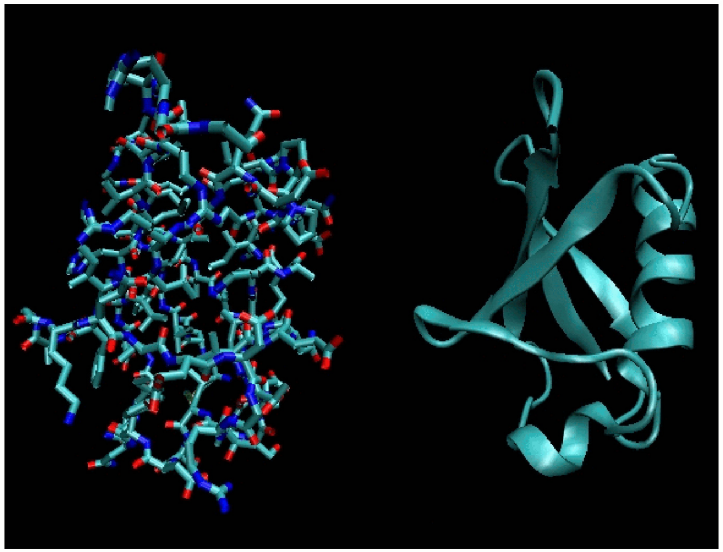
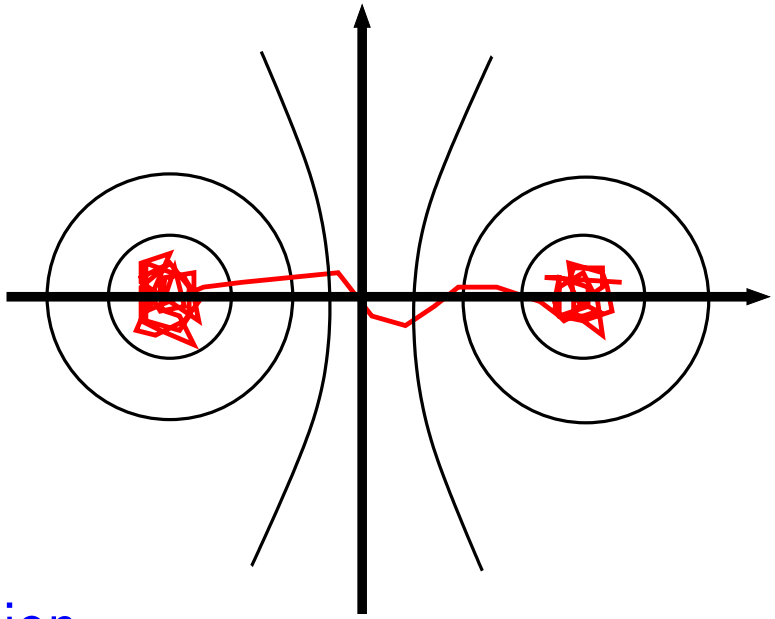
Joint with T. Lelièvre (ENPC), O. Gorynina (ENPC, now in Davos), D. Perez (LANL) and U. Sharma (Berlin)

11th conference on Parallel-in-Time integration, CIRM, 11-15 July 2022

Motivation

This work is motivated by molecular simulation, where we often have to simulate long trajectories of complex, metastable systems.





Typical dynamics: the Langevin equation

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

where V is the potential energy, $\gamma > 0$ a damping parameter, β^{-1} the temperature and $W(t)$ a Brownian motion.

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- It turns out that this algorithm is not stable for MD problems when the time horizon is too large
- We therefore introduce an adaptive parareal algorithm, which performs simulations on shorter time slabs and paste them together, thereby allowing for a significant CPU gain

F.L., T. Lelièvre and U. Sharma, *An adaptive parareal algorithm: application to the simulation of molecular dynamics trajectories*, SIAM Journal on Scientific Computing 2022

O. Gorynina, F.L., T. Lelièvre and D. Perez, *Long-time simulation of diffusing self-interstitial atoms using the parareal algorithm*, in preparation

Parallel in time algorithm for ODEs

The parareal algorithm is based upon two integrators to propagate the system over a time ΔT:

- a fine, accurate integrator $\mathcal{F}_{\Delta T}$
- a cheap coarse integrator $\mathcal{C}_{\Delta T}$

Algorithm:

- Initialization: coarse propagation that yields $\left\{x_n^{k=0}\right\}_n$:

$$\forall n, \quad x_{n+1}^{k=0} = \mathcal{C}_{\Delta T}(x_n^{k=0})$$

- Iterate over $k \geq 0$:
  - compute jumps (in parallel):

$$J_n^k = \mathcal{F}_{\Delta T}(x_n^k) - \mathcal{C}_{\Delta T}(x_n^k)$$

  - sequential update to obtain $\left\{x_n^{k+1}\right\}_n$:

$$\forall n, \quad x_{n+1}^{k+1} = \mathcal{C}_{\Delta T}(x_n^{k+1}) + J_n^k$$

Parareal algorithm for MD simulations

- In MD, we often run simulations with time steps just below the stability limit (this often provides sufficient accuracy on the quantities of interest): no room for choosing a much larger time step in $\mathcal{C}_{\Delta T}$ than in $\mathcal{F}_{\Delta T}$

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- We thus turn to a paradigm where $\mathcal{C}_{\Delta T}$ integrates a simpler dynamics than $\mathcal{F}_{\Delta T}$ (say with the same time step):
  - $\mathcal{F}_{\Delta T}$ integrates the original Langevin dynamics (with $V_f \equiv V$)

$$dq = p\,dt, \qquad dp = -\nabla V_f(q)\,dt - \gamma\,p\,dt + \sqrt{2\gamma\beta^{-1}}\,dW$$

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- Identical Gaussian increments for $\mathcal{F}_{\Delta T}$ and $\mathcal{C}_{\Delta T}$ and over all parareal iterations (to ensure as best as possible trajectorial convergence)

Similar paradigm (in terms of V_f vs V_c) in [Baffico et al, PRE 2002]

Simulations on toy model problems

Two model problems

- A quadratic model in 1D:

$$V_f(q) = \frac{q^2}{2}, \qquad V_c(q) = \omega \frac{q^2}{2} \quad \text{for some } \omega > 0$$

Simple enough to be amenable to theoretical analysis, and exhibits the same issues as those appearing with more complex models.

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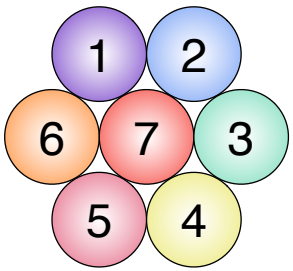
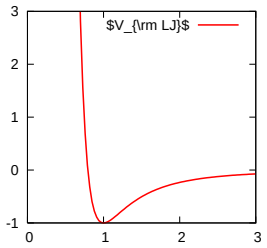
- A slightly less simple model: a 7-atom Lennard-Jones cluster in 2D:

$$V_f(q) = \frac{1}{2} \sum_{i,j \in \{1,\dots,7\},\ i \neq j} \phi_f(|q^i - q^j|), \qquad \phi_f(r) = r^{-12} - 2\, r^{-6}$$

$V_c \equiv$ harm. approx. of V_f at the global minimum (the initial condition is chosen in the corresponding well).





Convergence criteria

- Relative error between consecutive parareal trajectories:

$$E(k,N) = \frac{\sum_{n=1}^{N} |q_n^k - q_n^{k-1}|}{\sum_{n=1}^{N} |q_n^{k-1}|}.$$

- We stop the algorithm at the first parareal iteration $\overline{k}$ for which

$$E(\overline{k},N) < \delta_{\text{conv}} = 10^{-5}$$

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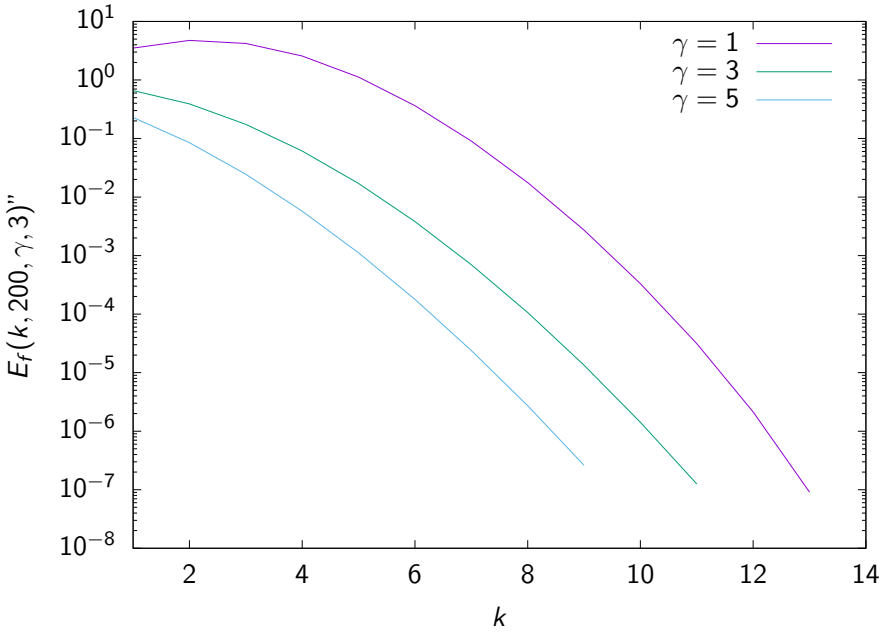
- The gain is then

$$\text{gain} = \frac{N}{\overline{k}} = \frac{\#\text{ fine propagations for a sequential algorithm}}{\#\text{ fine propagations for the parareal algorithm}}$$

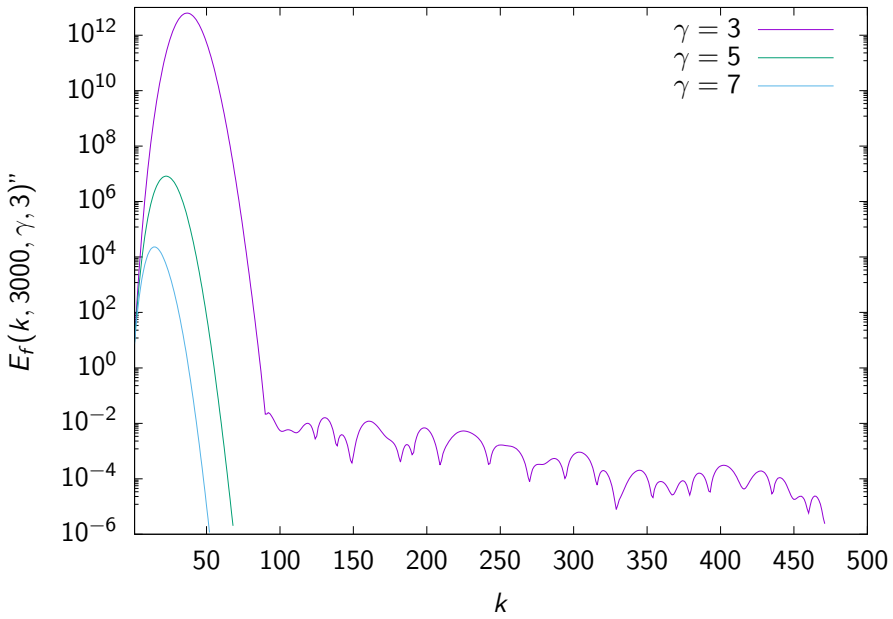
$E_f(k,N) =$ relative error with respect to the reference trajectory

Instability at large times

Plot of $E_f(k, N)$ as a function of k:



$N = 200$

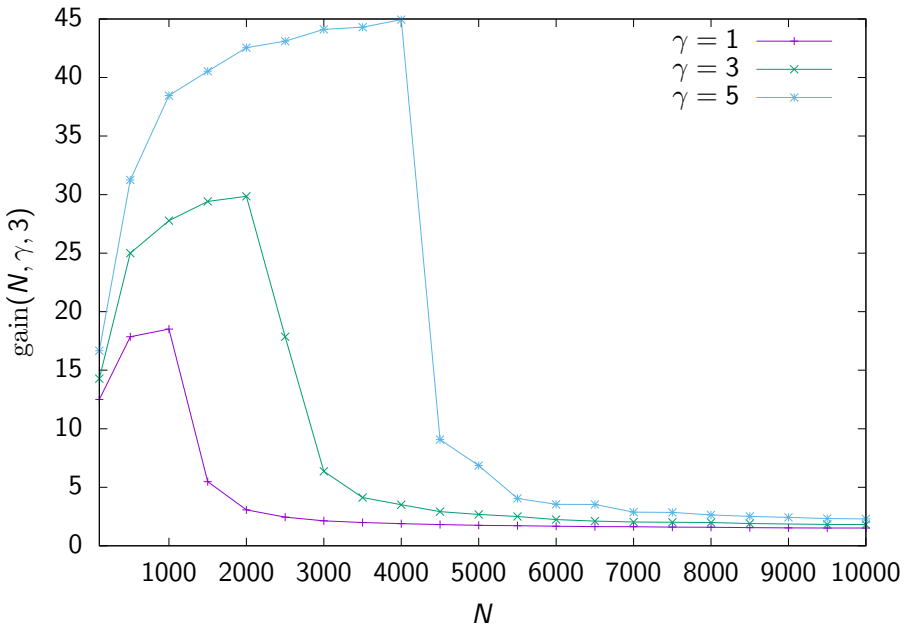


$N = 3000$

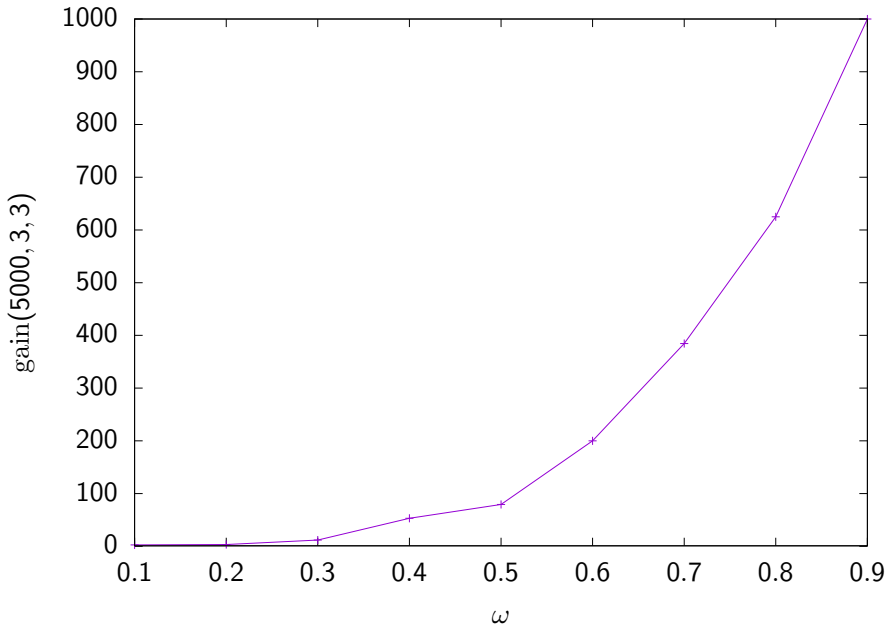
- For small N, fast convergence of the parareal iterations (gain ≈ 10)
- For large N, the error increases to large values when k increases (because the trajectory goes far away), before eventually converging

Harmonic model, $\omega = 0.1$, $\Delta T = 0.05$, $\beta = 3$

Gain as a function of N and ω



Gain as a function of N ($\omega = 0.1$)



Gain as a function of ω ($\gamma = 3$, $N = 5000$)

- Fixed coarse model: the gain decreases to 1 when N increases
- Fixed N: the gain decreases when the coarse model differs too much from the fine one

Harmonic model, $\Delta T = 0.05$, $\beta = 3$

Theoretical analysis on a toy problem

- Reference problem: $V_f(x) = x^2/2$ and

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- Coarse model: $V_c(x) = \omega\, x^2/2$ and

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- We have a complete understanding (see [SISC 2022]) of the error as a function of the trajectory length N and of $y = \dfrac{\mathcal{F}_{\Delta T}}{\mathcal{C}_{\Delta T}} - 1$, which quantifies how much the coarse and fine models differ.

A typical result

$$R_{n,k} = \text{relative error at time } n \text{ and iteration } \# \ k$$

In the case $y = \frac{\mathcal{F}_{\Delta T}}{\mathcal{C}_{\Delta T}} - 1 > 0$, then

$$0 < R_{n,k} \leq 1 \qquad \text{and} \qquad k \mapsto R_{n,k} \text{ is decreasing}$$

- If y is such that $y \leq c/n$ (the coarse model is very close to the fine; the longer the trajectory, the closer the models should be), then

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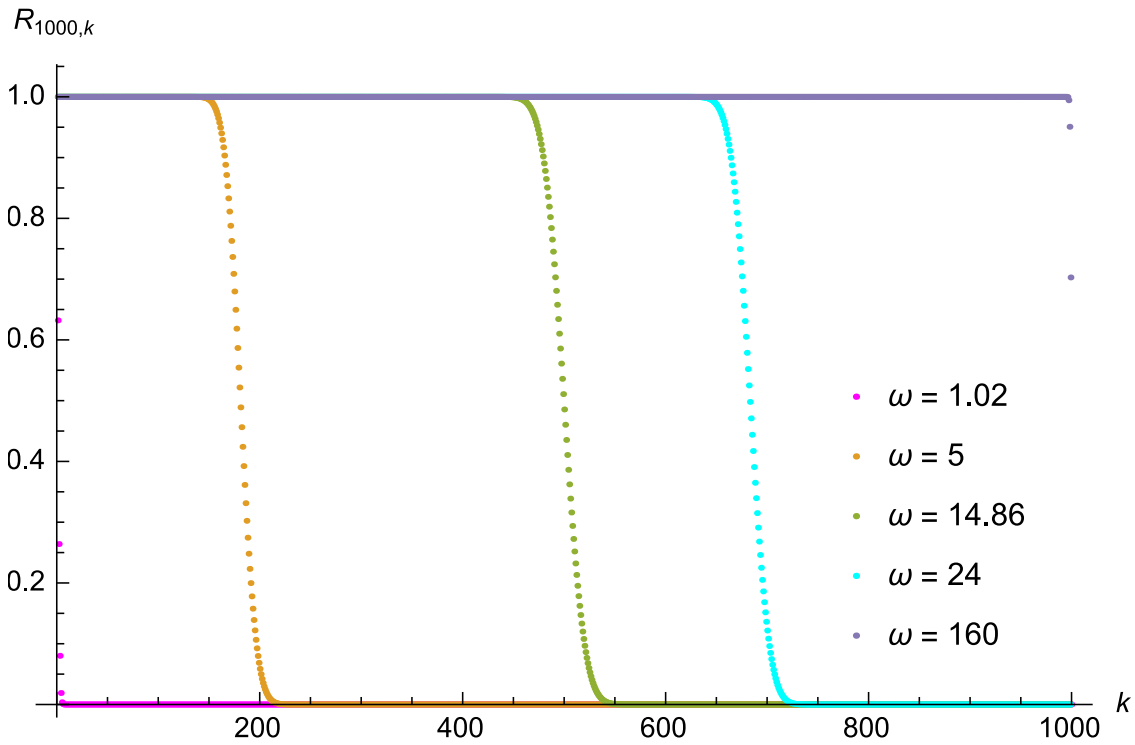
- If y is such that $y \geq c\, n$ (the coarse model is very different from the fine), then

$$k \mapsto R_{n,k} \quad \text{is concave}$$

- If y is in-between, then there exists $\overline{p} \approx \dfrac{n|y|}{1+|y|} \in [1, n]$ such that

$$k \mapsto R_{n,k} \text{ is concave for } 1 \leq k \leq \overline{p} \text{ and convex for } \overline{p} \leq k \leq n$$

Numerical illustration



Plot of $k \mapsto R_{n,k}$ for $n = 1000$

The error (as a function of k) is

- convex for $\omega = 1.02$ (excellent convergence)
- concave for $\omega = 160$ (error close to 100% for almost all k . . .)
- concave then convex for in-between ω (infl. point depends on y & n)

This analysis on an oversimplified model is illustrative of the general situation.

Adaptive algorithm

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Relative error $E \equiv$ relative error between consecutive trajectories

- On the time-slab $[0, N\Delta T]$, we run the parareal algorithm until E is
  - either smaller than the convergence threshold δ_{conv}
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- In the blow-up case, for the parareal iteration k_{cur}, we find the first time iteration $1 + \widetilde{m}_1 \leq N$ for which E exceeds δ_{expl}, and we shorten the slab to $[0, \widetilde{m}_1 \Delta T]$.
- We then proceed with the parareal iterations on the slab $[0, \widetilde{m}_1 \Delta T]$, that we possibly further shorten, until the relative error (on $[0, \widetilde{m}_1 \Delta T]$) is smaller than δ_{conv}. No need to redo the first k_{cur} iterations!

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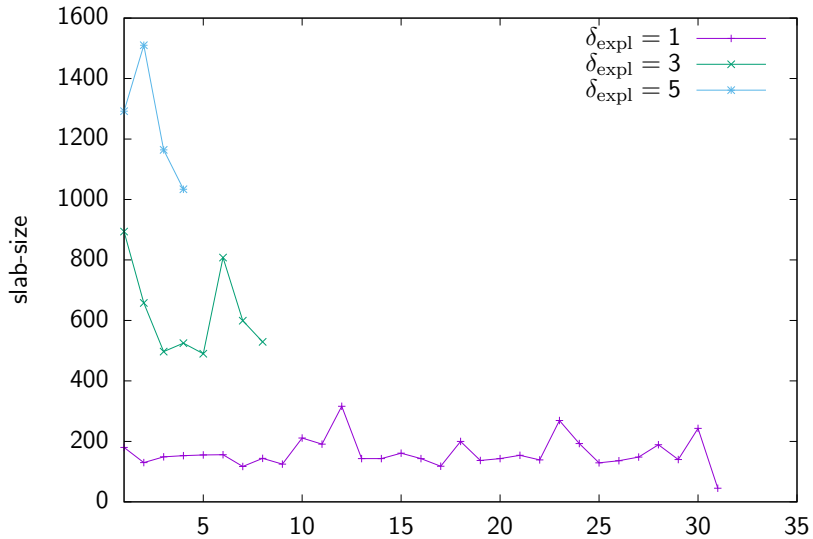
- Once we have converged on $[0, \widetilde{m}_1 \Delta T]$, we proceed with the next part of the time range and define the new (tentative) time-slab as $[\widetilde{m}_1 \Delta T, N \Delta T]$.

Explosion threshold

The slab sizes are such that $E \leq \delta_{\text{expl}}$:

- if δ_{expl} is chosen large, the adaptive criterion is never triggered: vanilla parareal
- if δ_{expl} is chosen small, the slabs are short: no parallelism anymore
- the optimal choice of δ_{expl} is somewhere in-between

List of the sizes of the time-slabs found by the algorithm:



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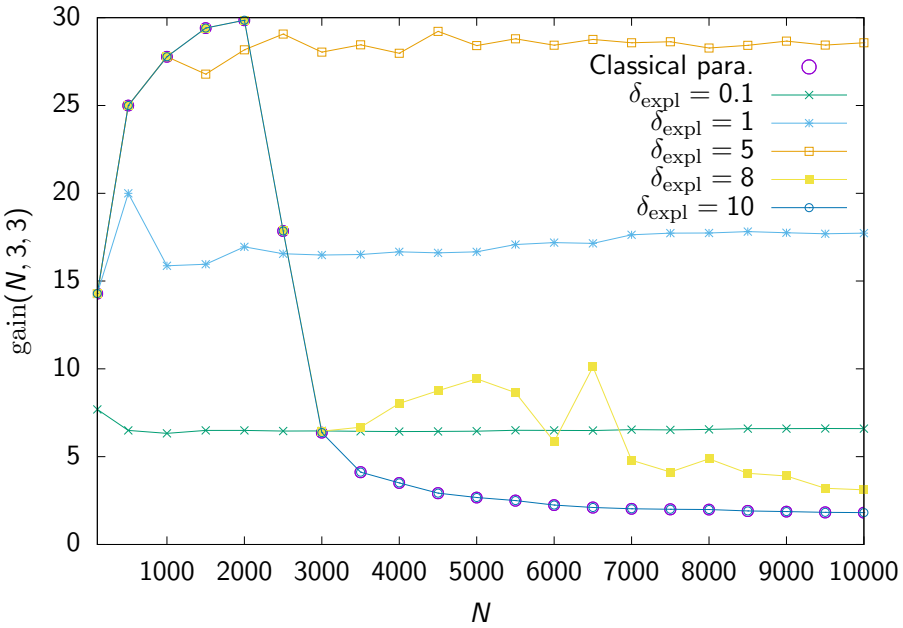
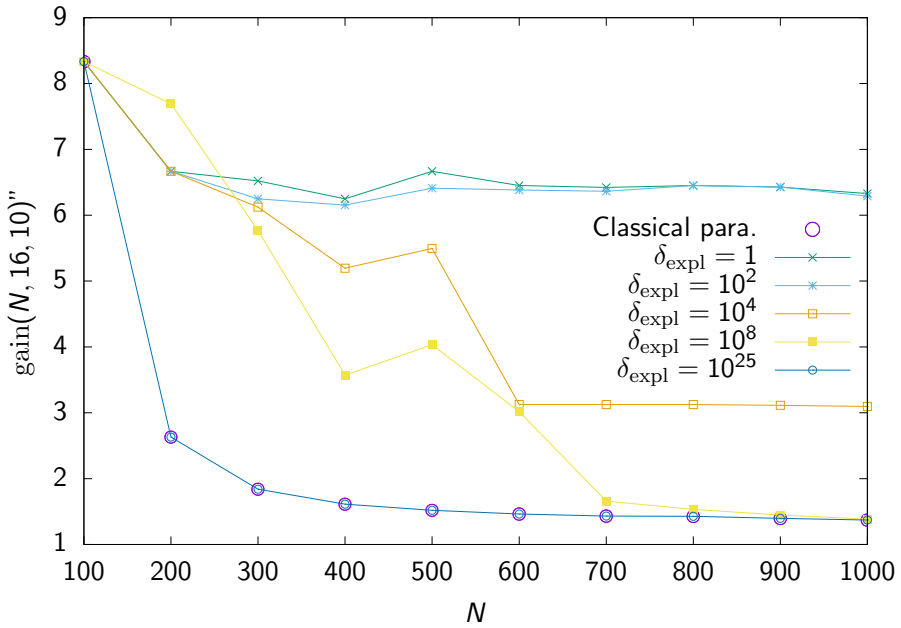
Gain



Har-1d ($\omega = 0.1$)



LJ7-2d

- For moderate values of δ_{expl}, the gain seems independent of N
- For large N, the adaptive algorithm always outperforms the classical version (gain ≈ 30 for Har-1d, gain ≈ 7 for LJ7-2d)

Conclusions on this part

- In the long time limit, the trajectories provided by the classical parareal algorithm are far away from the reference trajectories
- The adaptive algorithm always outperforms the classical version

F.L., T. Lelièvre and U. Sharma, SIAM J. Scientific Computing 2022

Support of ANR (through project CINE-PARA) and of EuroHPC (through project TIME-X) are gratefully acknowledged

Applications to the simulation of self-interstitial atoms in tungsten

Joint work with O. Gorynina, T. Lelièvre and D. Perez

Our aim

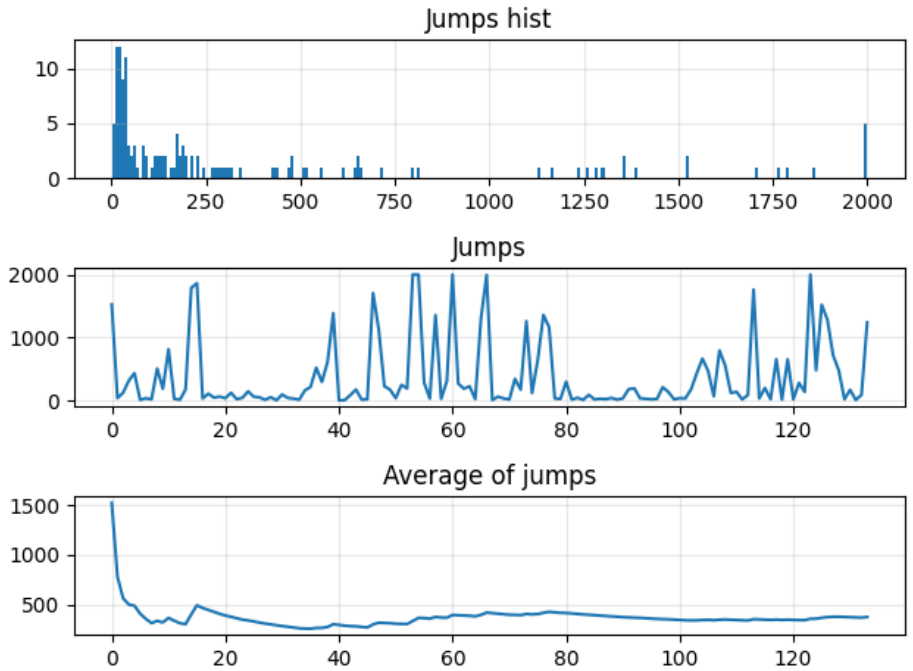
- We consider a periodic lattice of 128 tungsten atoms and add a defect in the lattice in the form of a self-interstitial atom (SIA)
- For the chosen physical parameters,
  - the system is metastable
  - within affordable trajectories, several jumps of the SIA are observed
- Quantity of interest: distribution of the residence times $\equiv$ time spent by the SIA in a given well before jumping into another well (monitor "statistical accuracy")
- Potentials:
  - fine (reference) potential V_f: SNAP $\equiv$ an expensive empirical potential adjusted on quantum mechanics (DFT) results
  - coarse potential V_c: EAM $\equiv$ a cheap empirical potential
- Simulations performed with LAMMPS, a widespread MD code

Reference results

History of residence times on a single trajectory:

$$[122, 23, 27, 476, 14, 32, 560, 245] \quad \times \delta t$$

Distribution of residence times (25 trajectories of 2000 time steps):



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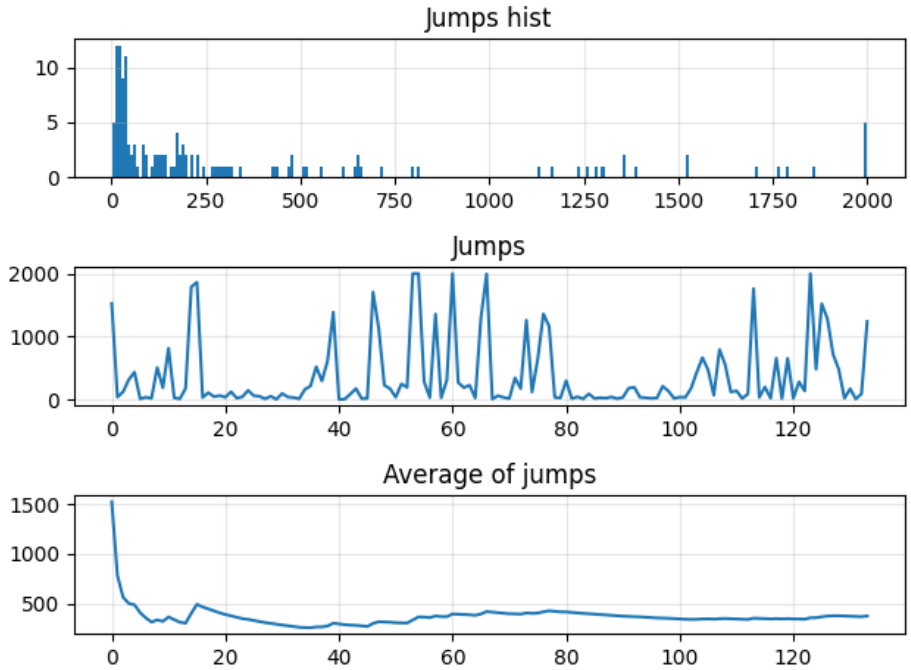
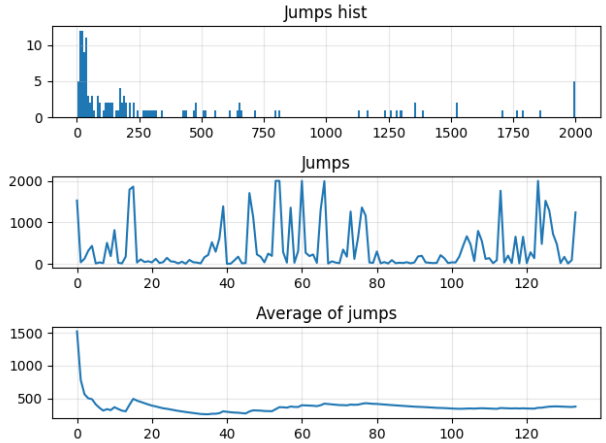


Mean residence time: $T_{\text{mean}} = 373 \times \delta t$, confidence interval $[278; 469] \times \delta t$

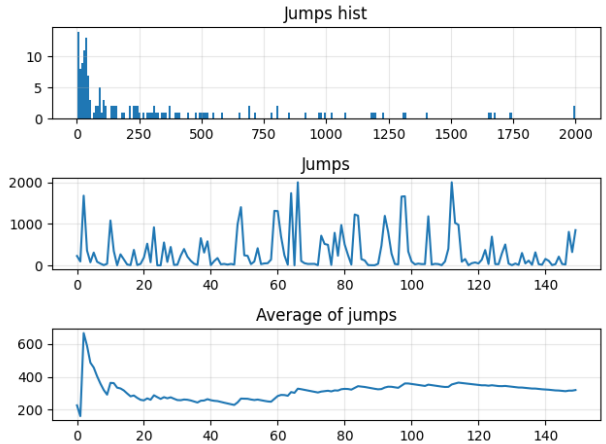
Coarse trajectory is wrong: $T_{\text{mean}} = 91 \times \delta t$, confid. interval $[81; 100] \times \delta t$

Parareal results, statistical accuracy ($\delta_{\text{conv}} = 10^{-3}$)

Reference results: $T_{\text{mean}} = 373 \times \delta t$, confidence interval $[278; 469] \times \delta t$



Parareal results: $T_{\text{mean}} = 320 \times \delta t$, confidence interval $[246; 394] \times \delta t$



Very good statistical accuracy (overlapping confidence intervals), while no pathwise accuracy for this value of δ_{conv}. Gain ≈ 5

Conclusions

- Non-intrusive implementation within LAMMPS, which allows to consider realistic systems
- We found a regime of intermediate δ_{conv} where
  - significant computational gains
  - no pathwise accuracy but excellent statistical accuracy

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Available postdoc position: please contact me for details!