Monodromy matrix calculation by the precision finite difference method

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Introduction

The monodromy (or stability) matrix is defined as the derivative of a time-evolved coordinate in phase space (coordinate of either position or momentum) with respect an initial coordinate in phase space. That is:

$$\mathbf{M}_{ij} = \frac{\partial \xi_i(t)}{\partial \xi_j(0)}$$

Calculating the monodromy matrix is an essential part of the "stability analysis" required by many methods in semiclassical dynamics. The precision finite difference method is a new way to calculate the monodromy matrix which requires neither calculation of the Hessian nor propagation by matrix multiplication. Here, we introduce this method and compare it to other ways of calculating the monodromy matrix.

Pre-Existing Methods

Direct Propagation Method

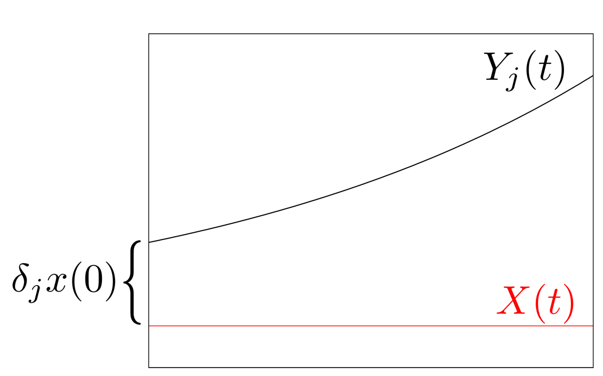
One of the most common ways to calculate the monodromy matrix is by calculating its time derivative and using standard molecular dynamics integrators to propagate through time. The time derivative is given by

$$\begin{pmatrix} \dot{M}_{qq} & \dot{M}_{qp} \\ \dot{M}_{pq} & \dot{M}_{pp} \end{pmatrix} = \begin{pmatrix} 0 & \mathbf{m}^{-1} \\ -\nabla\nabla V & 0 \end{pmatrix} \begin{pmatrix} M_{qq} & M_{qp} \\ M_{pq} & M_{pp} \end{pmatrix}$$

for a Hamiltonian of the form $p^2/2m + V(x)$. This method has the disadvantages of requiring the Hessian, and of requiring matrix multiplication to calculate the time derivative. However, it should be effectively exact, and we shall use is as the exact result for purposes of error comparisons.

Scaling: $O(F^3)$, plus Hessian

Naïve Finite Difference Method



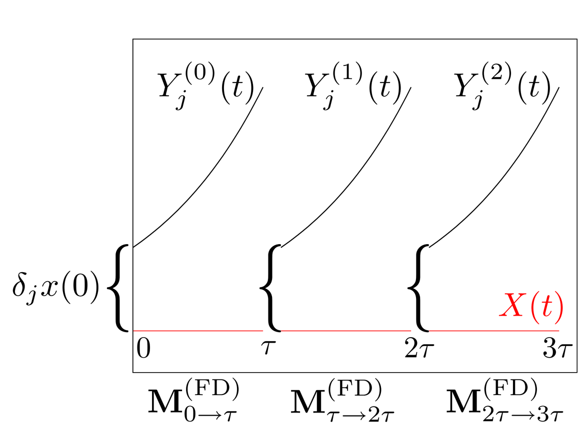
The definition of the monodromy matrix invites an attempt at a naïve finite difference approximation. Start with initial trajectory $X(t)$. For each dimension j in phase space, run an auxiliary trajectory $Y_j(t)$. Then the monodromy matrix can be calculated by finite difference.

$$\mathbf{M}^{(\mathrm{FD})}_{ij}(t) = \frac{(Y_j(t) - X(t))_i}{\delta_j x(0)}$$

However, this method is expected to fail for chaotic trajectories.

Scaling: $O(F^2)$, plus auxiliary trajectories

Garashchuk & Light's Method



Garashchuk and Light proposed a way of calculating the monodromy matrix without the Hessian.[1] They observed that the monodromy matrix is unitary; that is:

$$\mathbf{M}_{t_0\to t_2} = \mathbf{M}_{t_0\to t_1}\mathbf{M}_{t_1\to t_2}$$

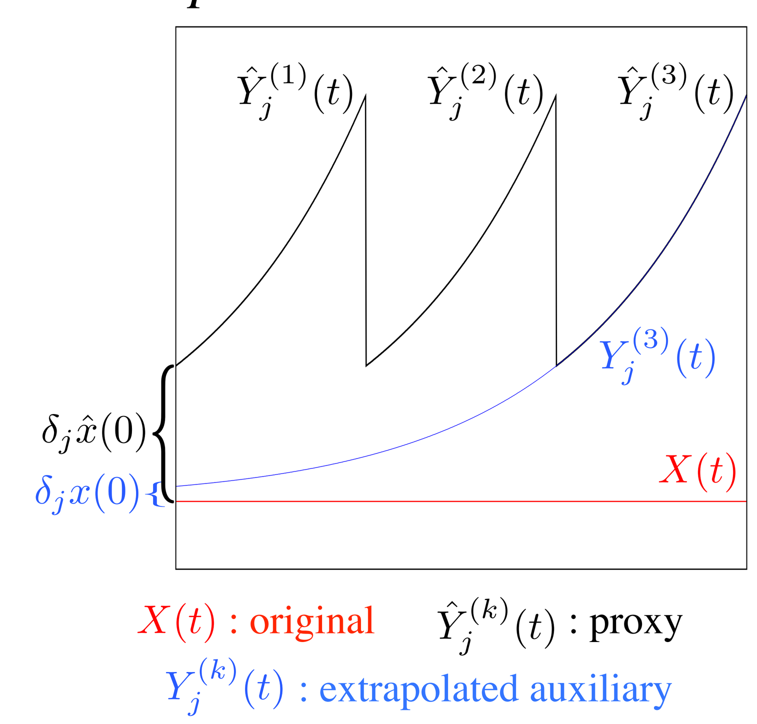
Their suggestion was to calculate short-time monodromy matrices by the naïve finite difference method, and then getting the long-time result by multiplying the short-time matrices together. However, this still requires many matrix multiplications.

$$\mathbf{M}(n\tau) = \prod_{i=0}^{n-1} \mathbf{M}^{(\mathrm{FD})}_{i\tau\to(i+1)\tau}$$

Scaling: $O(F^3)$, plus auxiliary trajectories

The Precision Finite Difference Method

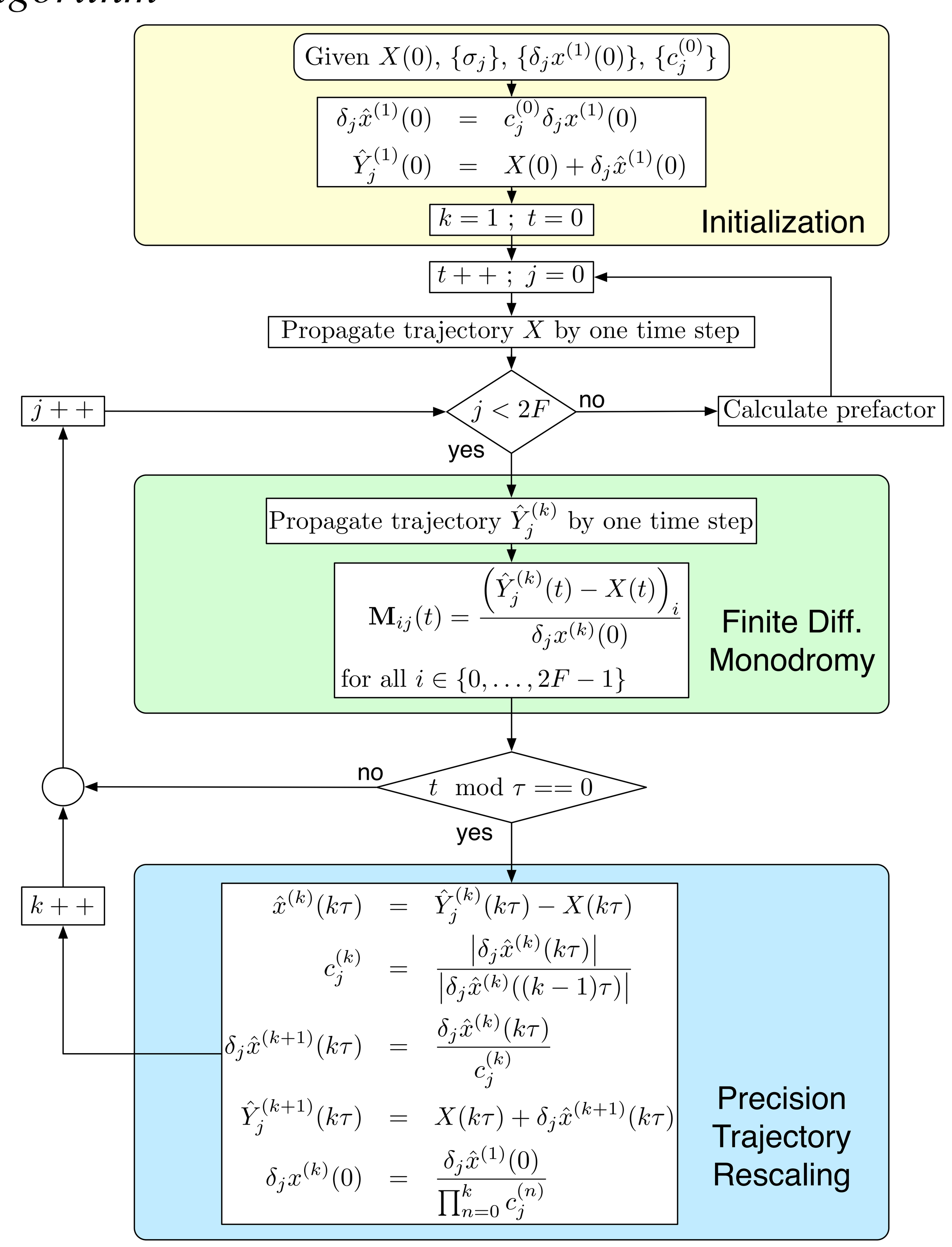
Concept



The precision finite difference method makes a finite difference approximation using a displacement smaller than would be possible for the naïve method. The trick is to use a trajectory with a larger displacement as a proxy for a trajectory with a smaller displacement (so long as both are within the linear regime.)

This idea was previously developed by Grünwald, Dellago, and Geissler for their "precision shooting" method in transition path sampling.[2]

Algorithm



Scaling: $O(F^2)$, plus proxy trajectories

Derivation of Precision Rescaling[2]

Define the solution operator $\Phi_t(X(0)) \equiv X(t)$ and let $\delta_j x(t) = Y(t) - X(t)$
Expanding $\delta_j x(t)$ to first order in $\delta_j x(0)$ we obtain:

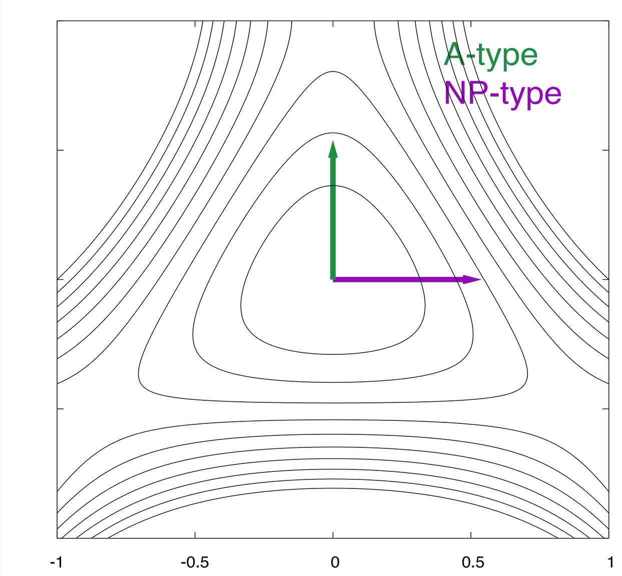
$$\delta_j x(t) \approx \Delta\delta_j x(0) \qquad \text{where } \Delta = \frac{\partial\Phi_t(X(0))}{\partial X(0)}$$

Take another trajectory in the linear regime, $\hat{Y}_j(t)$, where $\delta_j\hat{x}(0) = c_0\delta_j x(0)$
The same analysis applies, which means that:

$$\delta_j\hat{x}(t) \approx \Delta\delta_j\hat{x}(0) = \Delta c_0\delta_j x(0) \approx c_0\delta_j x(t)$$

This result means that the ratio of the displacement of two trajectories is constant in the linear regime. Therefore, the larger displacement can be used as a proxy for the smaller displacement. By repeatedly rescaling to follow proxy trajectories in the linear regime, we can extrapolate to an arbitrarily small initial displacement.

Application: The Hénon-Heiles Potential



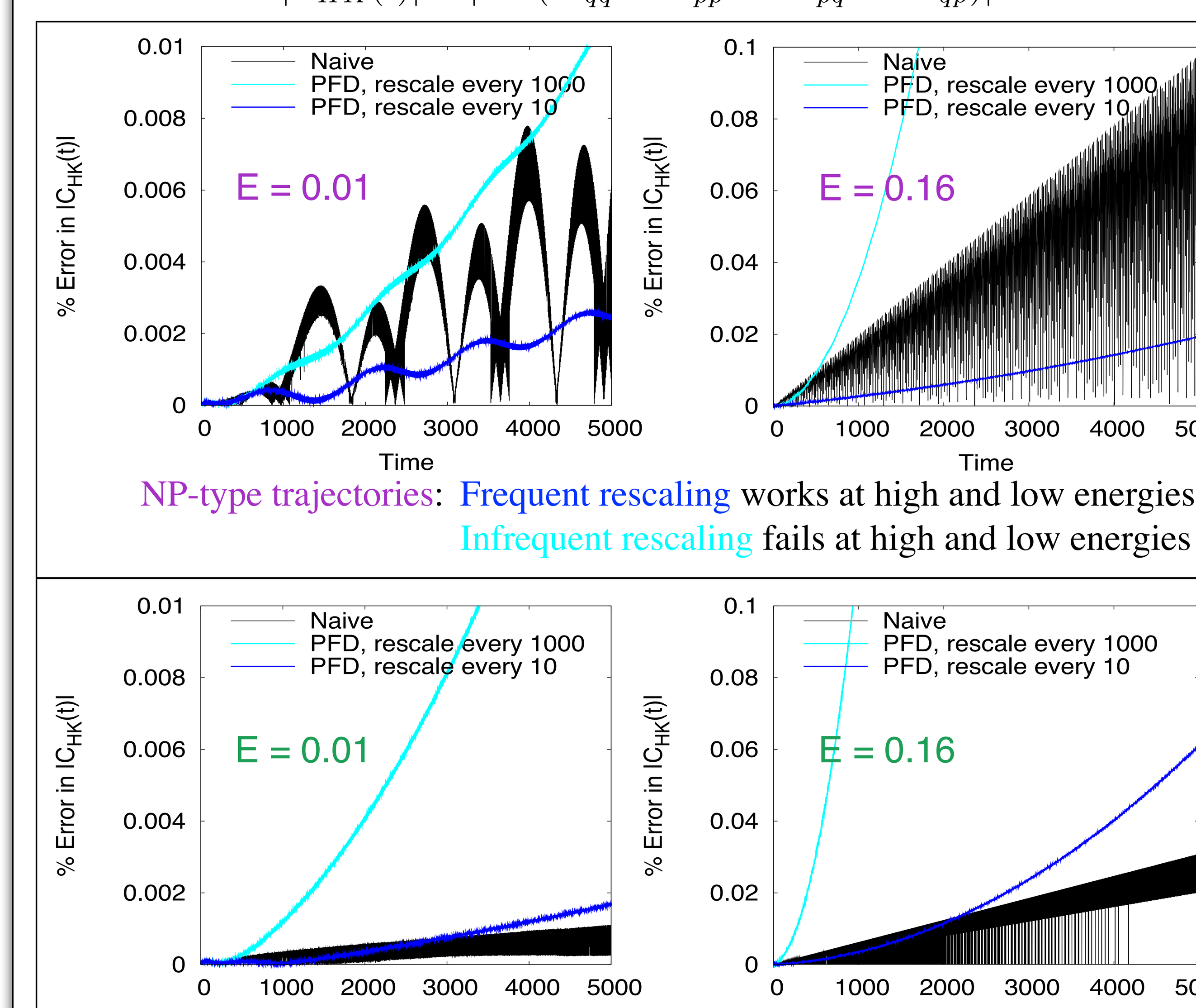
The Hénon-Heiles potential provides a challenging test system. It is a 2D potential with both stable and unstable trajectories. We used trajectories from two classes: non-periodic NP-type trajectories and periodic A-type trajectories (stable at low energy, unstable at high energy). Both begin at the origin with kinetic energy insufficient to cross the barrier.

$$V(x_1, x_2) = \frac{1}{2}\left(x_1^2 + x_2^2\right) + 2x_1^2 x_2 - \frac{1}{3}x_2^3$$

Results

Results shown are percent error in the magnitude of the Herman-Kluk prefactor:

$$|C_{HK}(t)| = |\det\left(M_{qq} + M_{pp} + iM_{pq} - iM_{qp}\right)|$$



NP-type trajectories: Frequent rescaling works at high and low energies
Infrequent rescaling fails at high and low energies

A-type trajectories: Even frequent rescaling fails at long time

Conclusions

1. The precision finite difference (PFD) method has better computational scaling than either Garashchuk & Light's method or direct propagation.
2. For non-periodic (typical) trajectories, PFD is more accurate than naïve finite difference when rescaling is frequent enough.
3. Element-by-element, the PFD monodromy matrix tends to be on the order of or better than the naïve method.
4. PFD is not as good for the A-type periodic trajectories (after several hundred periods). Other classes of periodic trajectories will be tested.
5. Naïve finite difference does quite well.
6. Unlike direct propagation or Garashchuk & Light's method, the PFD monodromy matrix can be calculated column by column.

References

1. Garashchuk and Light. *J. Chem. Phys.* **113**, 9390 (2000).
2. Grünwald, Dellago, and Geissler. *J. Chem. Phys.* **129**, 194101 (2008).

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