PDEs in Mixed Quantum-Classical Dynamics and the *Koopmon* Method

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Motivation for mixed quantum-classical (MQC) models

Some applications

	Classical Subsystem	**Quantum** Subsystem
Foundations	measuring device	measured system
Cosmology	gravity	matter
Chemistry	nuclei	electrons

Overcome the **curse of dimensionality** (at least to some extent) in simulations of many-body quantum systems, e.g. in **molecular dynamics**.

Classical states – pure vs mixed

- **Classical pure states** are vectors in phase space (point particles)

  Hamilton's equations: $\boxed{\dot{q} = \partial_p H,\ \dot{p} = -\partial_q H}$

  $H\colon T^*Q \to \mathbb{R}$ is a phase-space function (Hamiltonian)

- **Classical mixed states** are positive densities $f > 0$ with $\int f(q,p)\,\mathrm{d}q\mathrm{d}p = 1$

  Liouville equation: $\boxed{\partial_t f = \{H, f\}}$

Note that $\{g,h\} = \partial_q g \partial_p h - \partial_p g \partial_q h$ denotes the canonical Poisson bracket.

Quantum states – pure vs mixed

- Quantum pure states are normalized wave functions in L^2

Schrödinger equation (TDSE): $\boxed{i\hbar\partial_t\psi = \hat{H}\psi}$

$\hat{H}: L^2 \to L^2$ is a self-adjoint operator, usually obtained by *quantization*, i.e.,

$$H(q,p) \to \hat{H}(\hat{q},\hat{p}), \quad \hat{q}\psi = q\psi,\, \hat{p}\psi = -i\hbar\nabla\psi, \quad [\hat{q},\hat{p}] = \hat{q}\hat{p} - \hat{p}\hat{q} = i\hbar$$

$\to$ **Unitary evolution**: $\psi(t) = U(t)\psi_0 = e^{-i\hat{H}t/\hbar}\psi_0$

- Quantum mixed states are positive-semidefinite operators $\hat{\rho}$ with $\mathrm{Tr}(\hat{\rho}) = 1$

von Neumann equation: $\boxed{i\hbar\partial_t\hat{\rho} = [\hat{H},\hat{\rho}]}$

Notational setup for the hybrid model

- Hamiltonian operator $\widehat{H} = \widehat{H}(z)$: an operator-valued function on phase space $\left(\text{e.g. } \hat{H}(z) = \hat{H}(q,p) = \frac{1}{2}\left(\frac{p^2}{m} + m\omega^2 q^2\right)\hat{\sigma}_0 + \gamma q \hat{\sigma}_z,\ m,\omega,\gamma > 0\right)$
- Hybrid density $\widehat{P} = \widehat{P}(z)$: (sufficiently smooth) distribution taking values in the space $\mathrm{Her}(\mathscr{H})$ of Hermitian operators on the quantum Hilbert space $\mathscr{H}$
- The **classical Liouville density** and **quantum density matrix** are recovered via

$$f = \operatorname{Tr}\widehat{P} \qquad \text{and} \qquad \hat{\rho} = \int_{T^*Q} \widehat{P}\,\mathrm{d}q\mathrm{d}p$$

Think of $\widehat{P}(z) = f(z)\hat{\rho}(z) = f(z)\psi(\bullet;z)\psi(\bullet;z)^\dagger$.

Aim: Propose MQC models that satisfy the following **consistency criteria**:

1) The classical subsystem is described by a **positive** probability density
2) The quantum subsystem is described by a **positive-semidefinite** density operator
3) In the absence of a coupling potential, the mixed dynamics reduces to **uncoupled quantum and classical flows**
4) The model **equations are covariant** under quantum unitary and classical canonical transformations
5) In the presence of an interaction potential, quantum purity $\mathrm{Tr}(\hat{\rho}^2)$ is not a constant of motion **(decoherence)**

→ **Challenging to satisfy all – currently, only one model on the market.**

Ehrenfest (PDE) model

$$i\hbar\frac{\partial \widehat{P}}{\partial t} + i\hbar\,\mathrm{div}\big(\widehat{P}\langle \mathsf{X}_{\widehat{H}}\rangle\big) = \big[\widehat{H}, \widehat{P}\big]$$

or equivalently,

$$\partial_t f + \mathrm{div}\left(f\langle \mathsf{X}_{\widehat{H}}\rangle\right) = 0, \qquad i\hbar\left(\partial_t + \langle \mathsf{X}_{\widehat{H}}\rangle\cdot\nabla\right)\widehat{\mathscr{P}} = [\widehat{H}, \widehat{\mathscr{P}}]$$

- **Satisfies all consistency requirements**; known to suffer from **overdecoherence**
- The quantum dynamics **fully decouples** (drawback of Ehrenfest dynamics)

$\mathsf{X}_{\widehat{H}} := (\partial_p\widehat{H}, -\partial_q\widehat{H})$, $\quad \widehat{\mathscr{P}} = \widehat{P}/f$, $\quad \langle \mathsf{X}_{\widehat{H}}\rangle = \mathrm{Tr}(\widehat{\mathscr{P}}\mathsf{X}_{\widehat{H}})$

Multi-trajectory Ehrenfest system

The equation for the classical density is solved by the **point-particle ansatz***

$$f(z,t) = \sum_{a=1}^{N} w_a \delta(z - \zeta_a(t)),$$

with $\zeta_a(t) = (q_a(t), p_a(t))$, $\dot{\zeta}_a = \langle X_{\widehat{H}} \rangle|_{z=\zeta_a}$, weights $w_a > 0$ and $\sum_a w_a = 1$.
$\langle X_{\widehat{H}} \rangle|_{z=\zeta_a}$ requires evaluating $\hat{\rho}_a(t) := \widehat{\mathscr{P}}(\zeta_a(t), t)$ at all times, so that $i\hbar\dot{\hat{\rho}}_a = [\widehat{H}_a, \hat{\rho}_a]$.

The resulting **multi-trajectory Ehrenfest system** reads

$$\dot{q}_a = \partial_{p_a} \langle \hat{\rho}_a | \widehat{H}_a \rangle, \qquad \dot{p}_a = -\partial_{q_a} \langle \hat{\rho}_a | \widehat{H}_a \rangle, \qquad i\hbar\dot{\hat{\rho}}_a = [\widehat{H}_a, \hat{\rho}_a],$$

with $\widehat{H}_a = \widehat{H}(\zeta_a)$ and $\langle \hat{\rho}_a | \widehat{H}_a \rangle = \mathrm{Tr}(\hat{\rho}_a \widehat{H}_a)$.

* **Computational particles** (not physical).

Construction of the new hybrid model

"Our new hybrid (PDE) model overcomes the **consistency issues***, enjoys both a variational and a Hamiltonian structure, and goes beyond Ehrenfest dynamics."*

Write classical mechanics in terms of Schrödinger-like wave functions!

→ In the following, we briefly outline the main steps of the construction. The detailed derivations are beyond the scope of this talk.

Step 1 (*Classical dynamics in a Hilbert space*)

The *Koopman–van Hove equation*

$$i\hbar\partial_t\Psi = \{i\hbar H, \Psi\} - (p\partial_p H - H)\Psi =: \widehat{\mathscr{L}_H}\Psi$$

yields classical Liouville dynamics $\partial_t D = \{H, D\}$ for the phase-space density

$$D = |\Psi|^2 + \hbar\,\mathrm{Im}\{\Psi^*, \Psi\} + \partial_p(p|\Psi|^2)$$

- Formally analogous to Schrödinger dynamics ($\widehat{\mathscr{L}_H}$ is self-adjoint)
- Note that in the **Koopman–von Neumann equation** the conserved energy $\int \Psi^*\{i\hbar H, \Psi\}\,\mathrm{d}q\mathrm{d}p = \int H\hbar\,\mathrm{Im}\{\Psi^*, \Psi\}\,\mathrm{d}q\mathrm{d}p \neq \int |\Psi|^2 H\,\mathrm{d}q\mathrm{d}p$ is **different from the physical energy** and **vanishes** if Ψ is real
- The sign of the classical density D is preserved in time

$\Psi = \Psi(z)$ is a square-integrable wave function on phase space.

Step 2 (Linear hybrid model)

Construction of **hybrid wave functions** and a linear hybrid PDE based on a classical two-particle (Koopman) wave function and **partial quantization**

$$\Psi(q,p,q',p') \to \Upsilon(z,x)$$

$$i\hbar\partial_t\Upsilon = \widehat{\mathscr{L}_{\widehat{H}}}\Upsilon := \{i\hbar\widehat{H},\Upsilon\} - \big(p\partial_p\widehat{H} - \widehat{H}\big)\Upsilon$$

$\to$ Quantum positivity is guaranteed; classical positivity is not guaranteed (no proof).

We need to work harder...

$\Upsilon(z,x)$ is a square-integrable function of both the classical and quantum coordinates z and x.
$\widehat{H} = \widehat{H}(z,\hat{x},\hat{p})$ is an operator-valued function on phase space.

Step 3 (Non-linear hybrid model)

The construction of the final non-linear PDE is based on the following steps:

- **Exact factorization**: $\Upsilon(z,x,t) = \chi(z,t)\psi(x,t;z)$
- **Madelung transform**: $\chi(z,t) = \sqrt{f(z,t)}e^{iS(z,t)/\hbar}$

  → The hydrodynamic formulation allows us to identify the **classical phase**
- Derive a **phase-invariant** variational principle

All these steps require a lot of work (Lagrangians, Euler–Poincaré reduction, group actions, gauge connections. . .)!

Equations of motion for the new MQC model

The **resulting equations** of the non-linear hybrid PDE read

$$\boxed{\partial_t f + \operatorname{div}\left(f\,\mathscr{X}\right) = 0, \qquad i\hbar\big(\partial_t + \mathscr{X}\cdot\nabla\big)\widehat{\mathscr{P}} = [\widehat{\mathscr{H}}, \widehat{\mathscr{P}}]}$$

where $\mathscr{X}$ and $\widehat{\mathscr{H}}$ include $\hbar$-order back-reaction corrections to the Ehrenfest quantities:

$$\mathscr{X} = \langle X_{\widehat{H}}\rangle + \frac{\hbar}{2f}\left(\left\langle X_{\widehat{H}}\cdot\nabla \,\middle|\, \widehat{\Gamma}\right\rangle - \left\langle \widehat{\Gamma}\cdot\nabla \,\middle|\, X_{\widehat{H}}\right\rangle\right),$$

$$\widehat{\mathscr{H}} = \widehat{H} + \frac{i\hbar}{2f}\left[2\nabla\widehat{\mathscr{P}} + \widehat{\mathscr{P}}\nabla f, X_{\widehat{H}}\right],$$

where $\widehat{\Gamma} = if[\widehat{\mathscr{P}}, \nabla\widehat{\mathscr{P}}] \to$ "$\hbar$-correction" to the **Ehrenfest model**

The new model overcomes consistency issues and goes beyond Ehrenfest dynamics

Nevertheless, the underlying PDE is **quite intimidating**.

How to solve it numerically?

$\rightarrow$ The presence of **inverses and gradients** in the equations does not allow for a direct trajectory-based closure (like for Ehrenfest) of the form

$$f = \sum_a w_a \delta(z - z_a), \quad \widehat{P} = \sum_a w_a \rho_a \delta(z - z_a).$$

Regularization and *Koopmons*

The *Koopmon method* uses a **variational regularization** to restore point-particle trajectories. The regularized Lagrangian reads

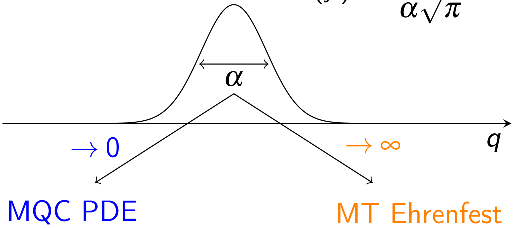
$$\bar{\ell} = \int_{T^*Q} \left(f \mathscr{A} \cdot \mathscr{X} + \langle \widehat{P}, i\hbar \hat{\xi} - \widehat{H} \rangle - a \underbrace{f^{-1}}_{\to \bar{f}^{-1}} \langle \bar{P}, i\hbar \{ \underbrace{P}_{\to \bar{P}}, \widehat{H} \} \rangle \right) \mathrm{d}z,$$

where $\bar{f} = K_\alpha * f$ and $\bar{P} = K_\alpha * \widehat{P}$ for some convolution kernel K_α in phase space.
The resulting regularized equations allow for delta-like expressions of f and $\widehat{P}$, returning trajectories called *Koopmons*.

The role of the regularization parameter α

The kernel function $K\colon T^*Q \to \mathbb{R}$ (in phase space) is chosen as a product of one-dimensional **normalized Gaussians**, *i.e.*, $K(q,p) = \tilde{K}(q)\tilde{K}(p)$ with

$$\tilde{K}(y) := \frac{1}{\alpha\sqrt{\pi}}\, e^{-y^2/\alpha^2}, \quad y \in \mathbb{R}.$$



MT = multi-trajectory. A **robust default** is $\alpha = 0.5$, $N = 500$.

Trajectory equations

$$\boxed{\dot{q}_a = w_a^{-1}\partial_{p_a} h, \quad \dot{p}_a = -w_a^{-1}\partial_{q_a} h, \quad i\hbar\dot{\rho}_a = w_a^{-1}[\partial_{\rho_a} h, \rho_a]}$$

where

$$h = \sum_a w_a \left\langle \rho_a, \widehat{H}_a + i\hbar \sum_b w_b [\rho_b, \mathscr{I}_{ab}] \right\rangle, \quad \partial_{\rho_a} h = \widehat{H}_a + i\hbar \sum_b w_b [\rho_b, \mathscr{I}_{ab} - \mathscr{I}_{ba}],$$

$$\mathscr{I}_{ab} = \frac{1}{2}\int_{T^*Q} \frac{K_a\{K_b, \widehat{H}\}}{\sum_c w_c K_c}\,\mathrm{d}z, \quad K_s(z,t) := K_\alpha(z - z_s(t))$$

Implementation

Spatial integration (phase-space integral $\mathscr{I}_{ab}$)

Implemented using the **composite trapezoidal rule** with a **time-dependent phase-space box** that adapts to the distribution at each time step.

Time integration

- Executed using the **fourth-order Runge–Kutta method (RK4)**
- A **time-adaptive variant** of RK4 is also available
- A **symplectic variant** is also available (fixed time steps)

Selected test cases studied

- *Tully 1-3* (high, intermediate, low momentum)
- *Double Arch* (high and low momentum)
- *Rabi* (ultrastrong, deep-strong, Jaynes–Cummings)

Quantities of interest

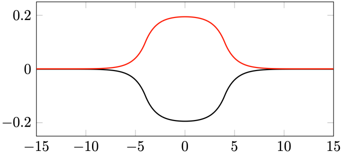
Liouville density (including projections), Bloch vector (incl. purity), populations

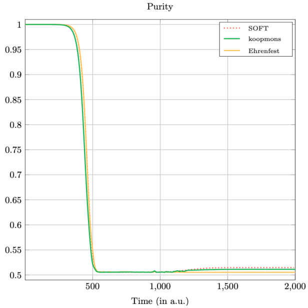
Quantum reference

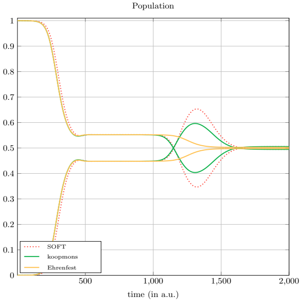
We benchmark against a **fully quantum TDSE solution** (*Split-Operator Fourier Transform*, SOFT) and visualize the corresponding **Wigner distributions**.

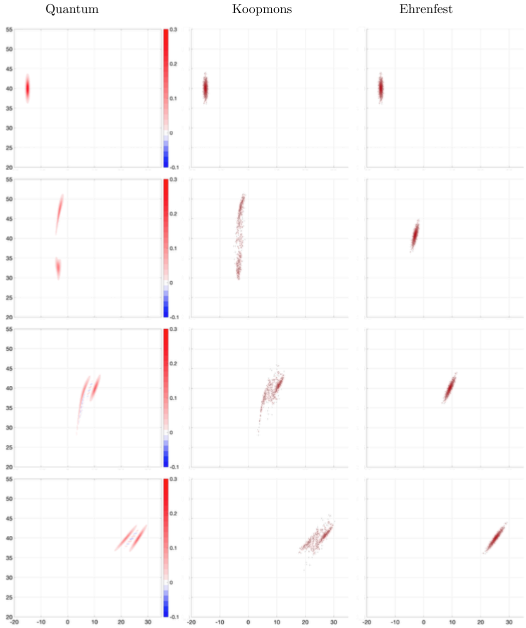
Double Arch (high momentum)

$N = 500$; $\alpha = 0.5$; $q_0 = -15$, $p_0 = 40$; $\rho_0 = [0, 0; 0, 1]$; $dt = 2$





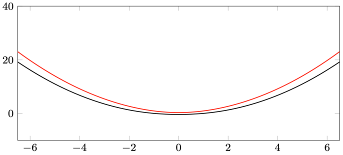


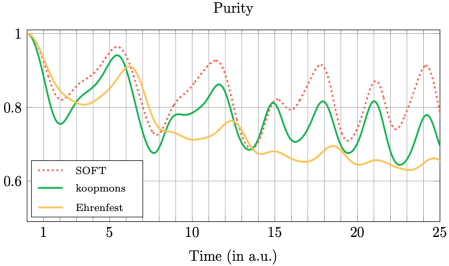


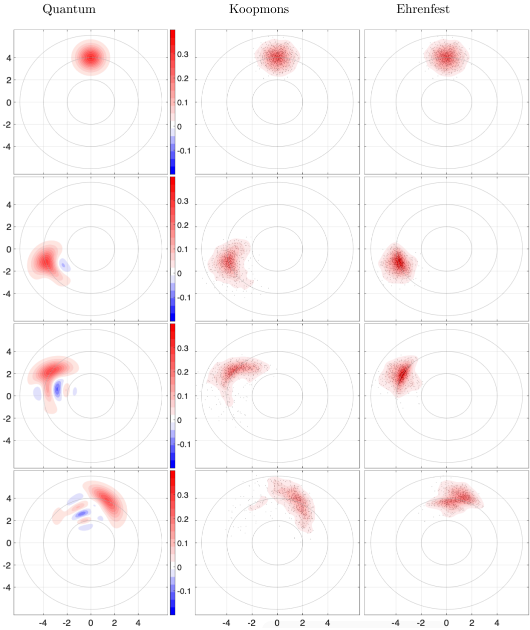
Snapshots at $t = 0$, 600, 1200, 2000

Rabi Hamiltonian (ultrastrong), $\hat{H}(q,p) = \frac{1}{2}\left(\frac{p^2}{m} + m\omega^2 q^2\right)\hat{\sigma}_0 + \gamma q \hat{\sigma}_z + B_0 \hat{\sigma}_x$

$N = 500$; $\alpha = 0.5$; $q_0 = 0$, $p_0 = 4$; $\rho_0 = [1,1;1,1]/2$; $dt = 0.05$







Snapshots at $t = 0,\ 10.5,\ 17.5,\ 25$

Work in progress (momentum coupling)

Rashba Hamiltonians (1D models for nanowires)

In the following, let $m, \omega > 0$, $\alpha_R > 0$ (Rashba coupling), $B_0 \in \mathbb{R}$ (magnetic field), and $g_e \in \mathbb{R}$ (Landé g-factor). We study Hamiltonians of the form

$$\hat{H}(q,p) = \frac{1}{2}\left(\frac{p^2}{m} + m\omega^2 q^2\right)\hat{\sigma}_0 + p\alpha_R\hat{\sigma}_y + \frac{1}{4}g_e B_0 \hat{\sigma}_x.$$

Classification

$$E_{SO} := \frac{m\alpha_R^2}{2}, \qquad E_Z := \frac{1}{4}g_e B_0.$$

Define $R := 2E_{SO}/|E_Z|$. The regime is

Zeeman dominated if $R < 1$, and **Rashba dominated** for $R > 1$.

We work in atomic units, i.e., $\hbar = 1$.

The following test cases have been studied

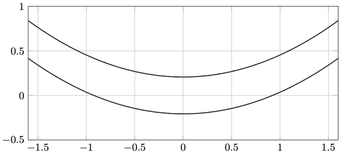
- Rashba dominated (ballistic and non-ballistic for *InAs*)
- Zeeman dominated (ballistic and non-ballistic for *InSb* & *GaAs*)

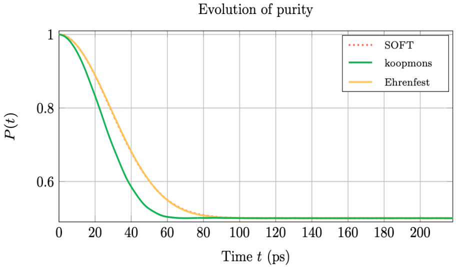
Quantities of interest

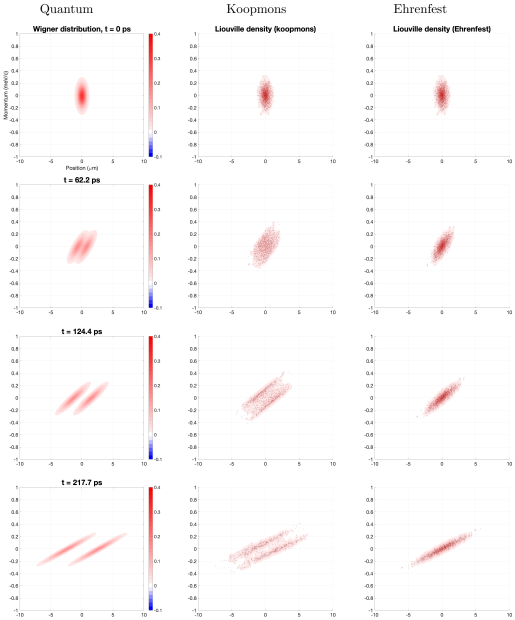
Liouville density (including projections), Bloch vector (incl. purity), uncertainty of the spin current

Zeeman dominated, ballistic (InSb)

$N = 500$; $\alpha = 0.5$; $q_0 = 0$, $p_0 = 0$; $\rho_0 = [1, 0; 0, 0]$; $dt = 217.7/400[ps]$



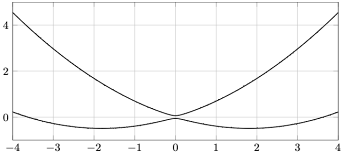


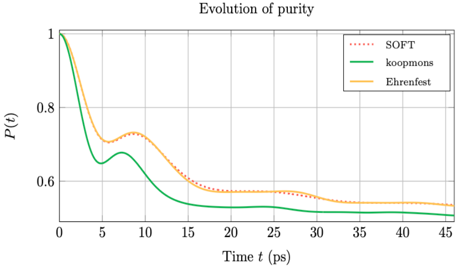


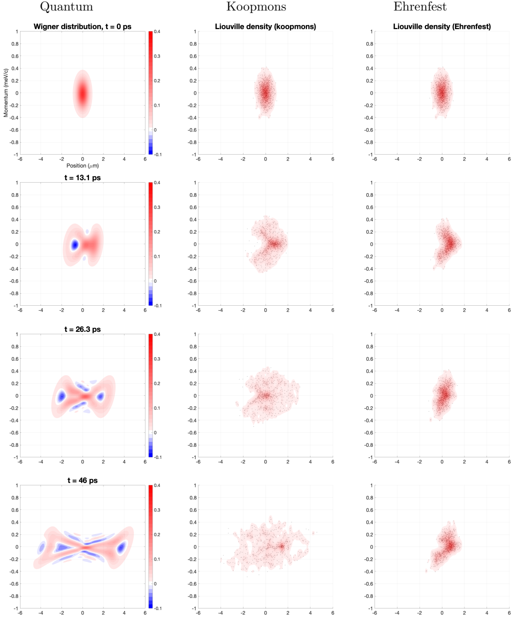
Snapshots at $t = 0$, 62.2, 124.4, 217.7 [ps]

Rashba dominated, ballistic (InAs)

$N = 500$; $\alpha = 0.5$; $q_0 = 0$, $p_0 = 0$; $\rho_0 = [1, 0; 0, 0]$; $dt = 46/400[ps]$







Snapshots at $t = 0$, 13.1, 26.3, 46 [ps]

Hybrid Ehrenfest–Koopman dynamics

Challenge

Koopmons simulations achieve accuracy levels (in both sectors) significantly beyond those of Ehrenfest dynamics. The price, however, is **higher computational cost**, which typically increases further as $\alpha \to 0$ (or in higher dimensions*).

Goal

Develop a numerical method that **switches between Ehrenfest and *Koopmons*** in order to significantly reduce computational cost.

*Note that for a broad class of Hamiltonians, the *Koopmons* scheme can be shown to admit an advantageous factorization property in dimensions $d \geq 2$ (linear combinations of integrals in phase space).

Problem

How can we identify regions where Ehrenfest dynamics is sufficiently accurate, and regions where the *Koopmons* **correction becomes essential**?

Idea

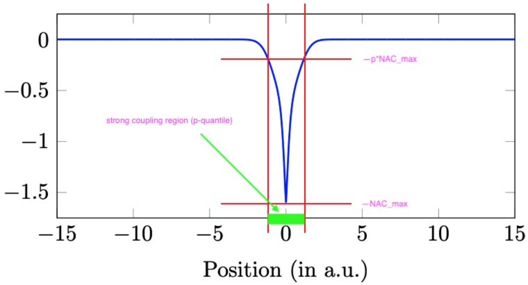
In nonadiabatic QC dynamics, the **nonadiabatic coupling (NAC) vectors** provide valuable information about regions of weak and strong coupling between the PESs.

Please note

The following strategy is our initial implementation for the *Tully models* and we are able to **significantly reduce computational cost**, while still obtaining reliable results in both the quantum and classical sectors. However, **feedback and suggestions for possible improvements are welcome** (e.g., approaches inspired by *surface hopping*).

Defining strong coupling via quantiles

- Define the strong-coupling region (SCR) via a quantile threshold $p \in [0,1]$
- We define: $\mathrm{SCR} := [p \cdot \mathrm{NAC}_{\max}, \mathrm{NAC}_{\max}]$



$\mathrm{NAC}_{\max} := \max_x |\mathrm{NAC}(x)|$

First Approach

- At each time step, compute the number of particles inside the SCR:

$$n_{\mathrm{SCR}}(t) = \#\Big\{ a \in \{1, \dots N\} \,:\, q_a(t) \in \mathrm{SCR} \Big\}$$

- If $n_{\mathrm{SCR}}(t) > N_{\mathrm{SCR}}$ for some given $N_{\mathrm{SCR}} > 0$, the simulation switches from the Ehrenfest to the *Koopmons* code

Implementation

- A trigger variable $\texttt{trigger} \in \{0, 1\}$ controls the switch
  ($\texttt{trigger} = 0$: Ehrenfest, $\texttt{trigger} = 1$: *Koopmons*)
  $\rightarrow$ All *Koopmons*-specific correction terms are skipped if $\texttt{trigger} = 0$

Experiment 1

Summary

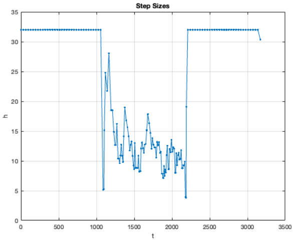
- Number of particles inside the SCR: $N_{\mathrm{SCR}} = 250$
- Quantile threshold: $p = 5\%$ $\quad\rightarrow\quad$ $\mathrm{SCR} = [-1.54, 1.54]$
- Switching times: $t_{\mathrm{on}} = 1,120$ a.u., $t_{\mathrm{off}} = 2,176$ a.u.
- Runtime of the hybrid code: 1,801 s

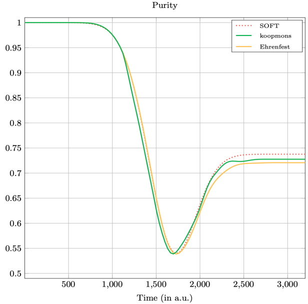
  $\approx 0.094\times$ ***Koopmons* reference** (19,228 s) [$0.42\times$ adaptive]

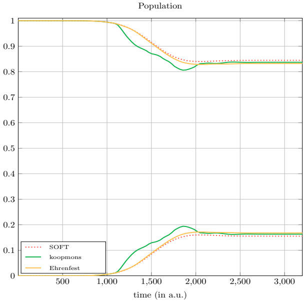
  $\approx 44\times$ Ehrenfest reference (40 s)

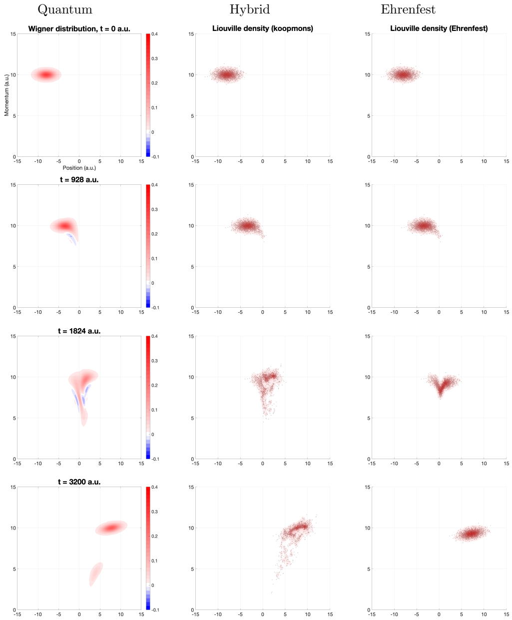
Tully 1 (low momentum)

$N = 1000$; $\alpha = 0.325$; $q_0 = -8$, $p_0 = 10$; $\rho_0 = [1, 0; 0, 0]$; $dt =$ "*adaptive*"









Snapshots at $t = 0$, 928, 1824, 3200 [au]

Experiment 2

Summary

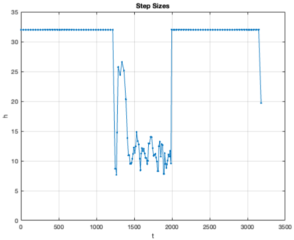
- Number of particles inside the SCR: $N_{\text{SCR}} = 450$
- Quantile threshold: $p = 5\%$ $\quad\rightarrow\quad$ $\text{SCR} = [-1.54, 1.54]$
- Switching times: $t_{\text{on}} = 1,280$ a.u., $t_{\text{off}} = 1,952$ a.u.
- Runtime of the hybrid code: 1,080 s

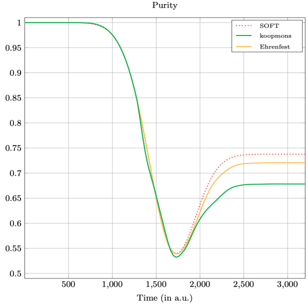
  $\approx 0.056\times$ ***Koopmons* reference** (19,228 s) [$0.25\times$ adaptive]

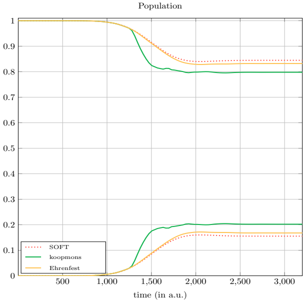
  $\approx 27\times$ Ehrenfest reference (40 s)

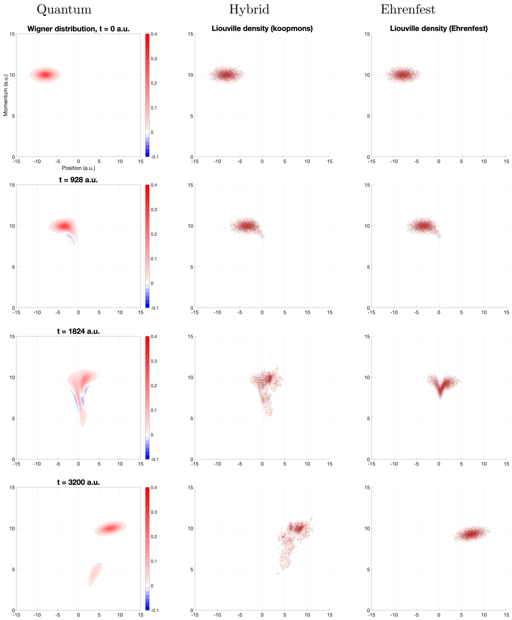
Tully 1 (low momentum)

$N = 1000$; $\alpha = 0.325$; $q_0 = -8$, $p_0 = 10$; $\rho_0 = [1, 0; 0, 0]$; $dt =$ "adaptive"









Snapshots at $t = 0$, 928, 1824, 3200 [au]

Final remarks

- New trajectory-based Hamiltonian approach for simulating MQC dynamics
- The *Koopmons* method **provides a solid foundation for developing closure methods** through the underlying variational structure
- Numerical tests for many different models (*Tully, Double Arch, Rabi, Rashba*,. . .)
- The hybrid Ehrenfest–Koopman dynamics show strong potential in first tests, but a rigorous switching strategy remains open

Thank you.