Koopmon Trajectories in Nonadiabatic Quantum-Classical Dynamics

Paul Bergold*

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Joint work with *W. Bauer* (Surrey), *F. Gay-Balmaz*, (NTU Singapore),
G. Manfredi (CNRS Strasbourg), and *C. Tronci* (Surrey)

*Institute of Mathematics, University of Gdańsk, Poland

Motivation of 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 extend) in simulations of many-body quantum systems, e.g. in **molecular dynamics**.

Setting the stage

Quantum & Classical (mixed states)

- x denotes quantum coordinates
- Quantum dynamics: **von Neumann equation**
- $i\hbar\partial_t\hat{\rho} = [\widehat{H},\hat{\rho}]$, where $\hat{\rho}$ is a *density matrix*, $\hat{\rho} \in \mathrm{Her}(\mathscr{H})$, $\mathrm{Tr}(\hat{\rho}) = 1$

- $z = (q,p) \in T^*Q$ (phase space) denotes classical coordinates
- Classical dynamics: **Liouville equation**
- $\partial_t f = \{H,f\}$, where $f > 0$ and $\int_{T^*Q} f(q,p)\,\mathrm{d}q\mathrm{d}p = 1$

$\{\cdot,\cdot\}$ is the canonical Poisson bracket, $[\cdot,\cdot]$ the commutator.

Setup for the hybrid model

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

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

The hybrid density operator can be thought of as $\widehat{P}(z) = f(z)\hat{\rho}(z) = f(z)\psi(\bullet;z)\psi(\bullet;z)^\dagger$.

Aim: Propose MQC models satisfying all 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 both quantum unitary transformations 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)**

$\rightarrow$ **Difficult to fulfil them all at the same time! Only <u>one</u> model on the market.**

Ehrenfest 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,

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

- **Fulfils all consistency requirements**, known to suffer from overdecoherence
- The quantum dynamics **decouples entirely** (drawback of Ehrenfest dynamics)

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

Multi-trajectory Ehrenfest system

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

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

with $\zeta_a(t) = (q_a(t), p_a(t))$, $\dot{\zeta}_a = \langle X_{\widehat{H}} \rangle|_{z=\zeta_a}$, and weights $w_a > 0$ such that $\sum_a w_a = 1$.
$\langle X_{\widehat{H}} \rangle|_{z=\zeta_a}$ 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 $\langle \hat{\rho}_a | \widehat{H}_a \rangle = \mathrm{Tr}(\hat{\rho}_a \widehat{H}_a)$.

These are **computational particles**, not physical particles!

Strategy for a MQC method

Blend **Koopman's classical mechanics** with methods in **symplectic geometry**!

- Based on **hybrid density operators**
- **Basis-independent** formulation
- Enjoys **variational** and **Hamiltonian structure** (conservation laws, closure)

All these steps require a lot of work! (as shown by C. Tronci)

(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

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

where $\mathscr{X}$ and $\widehat{\mathscr{H}}$ include $\hbar$-corrections (backreaction terms) to the Ehrenfest quantities, that is:

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

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

where $\widehat{\Gamma} = if[\widehat{\mathscr{P}}, \nabla\widehat{\mathscr{P}}]$

$\rightarrow$ "$\hbar$-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?

→ 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* exploits a **variational regularization** in order 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*.

In the limit $\alpha \to 0$, we ask for K_α to tend to the delta distribution (Dirac sequence), thereby recovering the **original Lagrangian**. In the limit $\alpha \to \infty$, we obtain **Ehrenfest**!

K_α smooth kernel function, e.g. normalized Gaussian with variance $\alpha > 0$

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

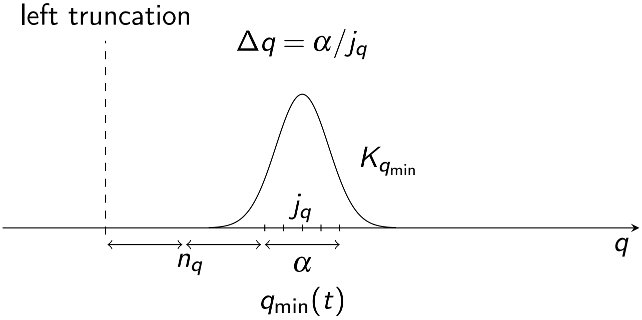
$$h = \sum_a w_a \Big\langle \rho_a, \widehat{H}_a + i\hbar \sum_b w_b [\rho_b, \mathscr{I}_{ab}] \Big\rangle, \quad \partial_{\rho_a} h = \widehat{H}_a + i\hbar \sum_b w_b [\rho_b, \mathscr{I}_{ab} - \mathscr{I}_{ba}],$$

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

Implementation

Spatial integration

Executed using the **composite trapezoidal rule**, employing a **time-dependent phase-space box** that dynamically adjusts with the distribution in each time step.



Analogously for right truncation and corresponding truncation in momentum space

Time integration

- Executed using the **fourth-order Runge–Kutta method (RK4)**.
- **Time adaptive** embedded RK methods (with *Delyan Zhelyazov*).
- **Symplectic variant** also available.

Code Structure (Pseudocode)

Input: Hamiltonian; number of particles; factorized kernel functions (α); initial data

for $k = 1, 2, \ldots$

Compute the time evolved parameters $q^{(k)}, p^{(k)}, \rho^{(k)}$

Cost $\rightarrow$ time integrator (RK4): $20N$ phase-space integrals

Check if total energy is conserved

Cost $\rightarrow$ N phase-space integrals

Implemented in MATLAB on a laptop machine.

Numerical experiments

Classical sector

Visualization of the time-evolved (Liouville) phase-space density.

Quantum sector

Visualization of decoherence levels, expressed in terms of **quantum purity** $\mathrm{Tr}(\hat{\rho}^2)$.

"Reference" solver (there is no true reference)

We compare our results with a **fully quantum solution** by solving the TDSE using a *split operator Fourier transform* (SOFT) method. We then plot the Wigner distribution.

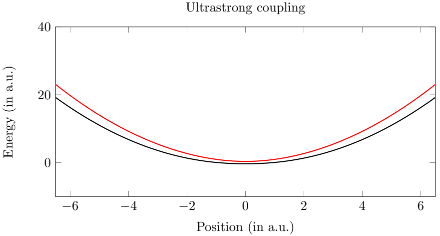
Rabi Hamiltonian in the **ultrastrong** coupling regime

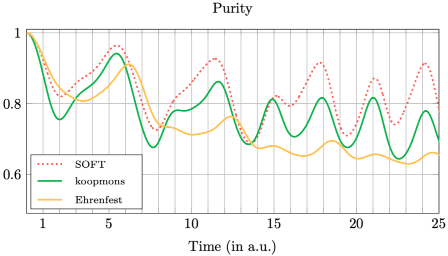
$$\hat{H}(q,p) = \frac{1}{2}\left(\frac{p^2}{m} + m\omega^2 q^2\right)\hat{\sigma}_0 + \gamma q \hat{\sigma}_z + B_0 \hat{\sigma}_x$$

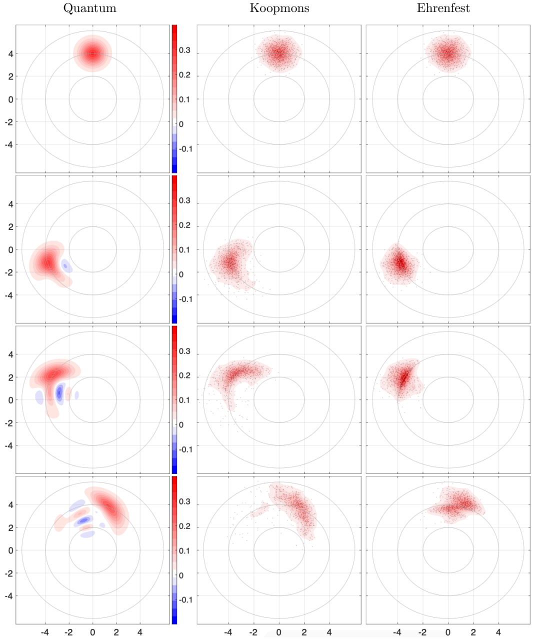
for $m = \omega = 1$, $\gamma = 0.29$, $B_0 = 0.35$.

$\hat{\sigma}_0 = [1, 0; 0, 1]$, $\hat{\sigma}_x = [0, 1; 1, 0]$, $\hat{\sigma}_z = [1, 0; -1, 0]$

Input: $N = 500$; $\alpha = 0.5$; $q_0 = 0$, $p_0 = 4$; $\rho_0 = [1, 1; 1, 1]/2$; $t_{\text{fin}} = 25$; $dt = 0.05$

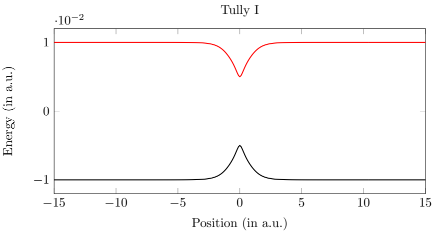


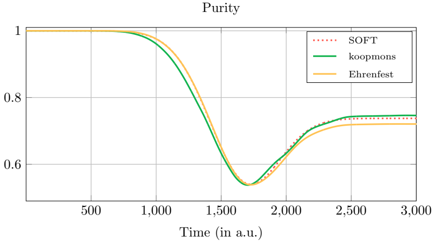


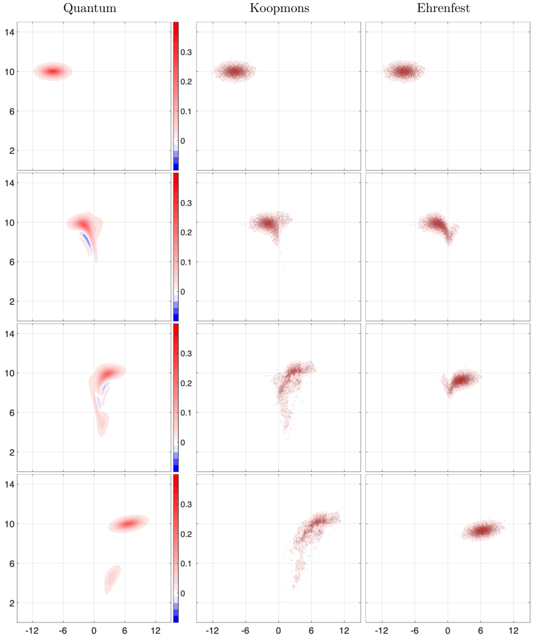


Standard values for N and α; snapshots at $t = 0,\ 10.5,\ 17.5,\ 25$

Input: $N = 1000$; $\alpha = 0.325$; $q_0 = -8$, $p_0 = 10$; $\rho_0 = [1, 0; 0, 0]$; $t_{\text{fin}} = 3000$; $dt = 2$

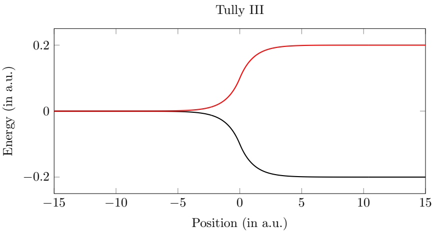


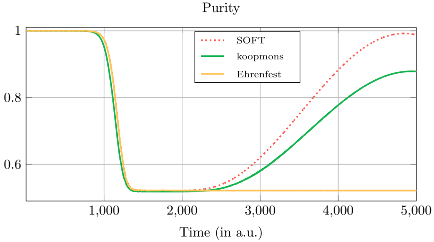


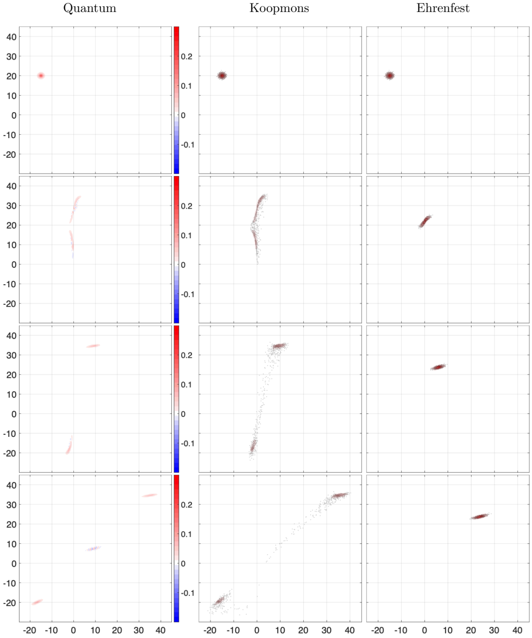


Tuned values for N and α; snapshots at $t = 0$, 1280, 2130, 3000

Input: $N = 1000$; $\alpha = 0.325$; $q_0 = -15$, $p_0 = 20$; $\rho_0 = [0, 0; 0, 1]$; $t_{\text{fin}} = 3500$; $dt = 2$







Tuned values for N and α; snapshots at $t = 0$, 1500, 2000, 3500

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)

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

The physical parameters m and α_R are material dependent.

$\rightarrow$ Good test cases available **without HO potential**

Classification

In the following, we work with $\hbar = 1$. The spin-orbit energy is defined as

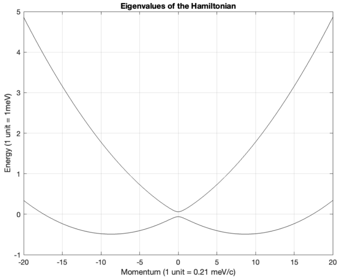
$$E_{SOC} := \frac{m\alpha_R^2}{2}.$$

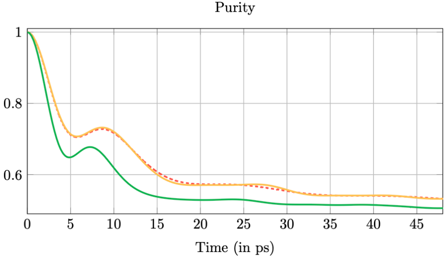
Let $R := 2E_{SOC}/|B|$. The coupling regime is called

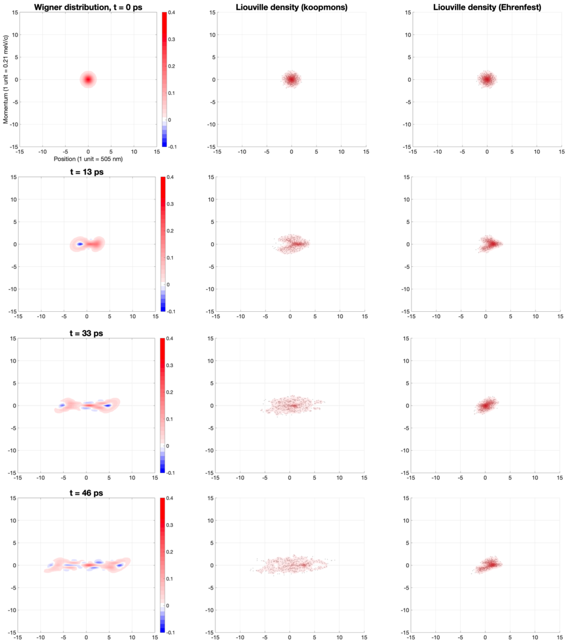
Zeeman dominated if $R < 1$, and **Rashba dominated** for $R > 1$.

Input: $N = 500$; $\alpha = 0.5$; $q_0 = 0$, $p_0 = 0$; $\rho_0 = [0, 0; 0, 1]$; $t_{\text{fin}} = 46\,\text{ps}$; $dt = t_{\text{fin}}/100$

Model: No HO; InAs; $m = 0.023$; $\alpha_R = 0.0396$; $B_0 = 140\,\text{mT}$; Rashba dominated ($R \approx 16$)



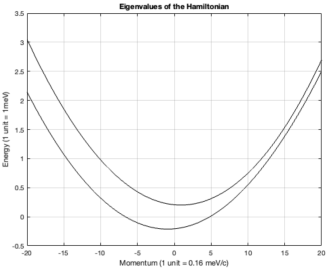


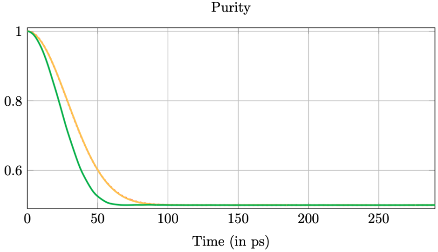


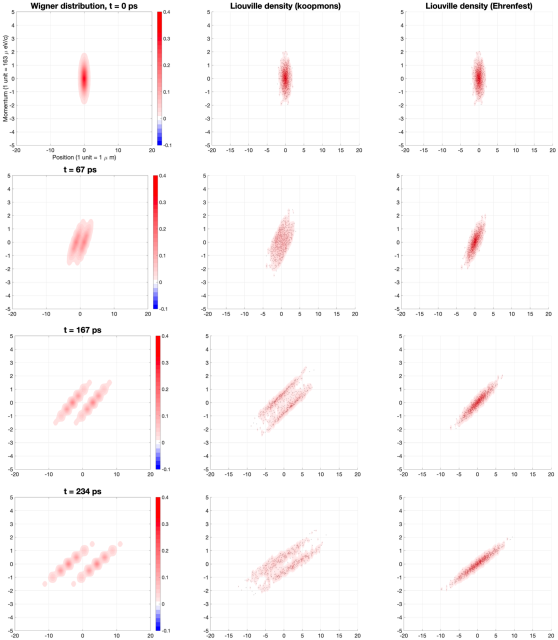
Standard values for N and α; InAs accounts for large coupling

Input: $N = 500$; $\alpha = 0.5$; $q_0 = 0$, $p_0 = 0$; $\rho_0 = [0,0;0,1]$; $t_{\text{fin}} = 234\,\text{ps}$; $dt = t_{\text{fin}}/100$

Model: No HO; InSb; $m = 0.014$; $\alpha_R = 0.0021$; $B_0 = 140\,\text{mT}$; Zeeman dominated ($R \approx 0.008$)



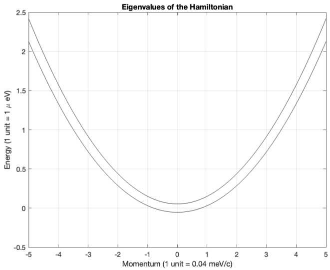


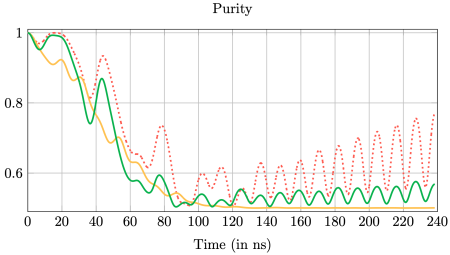


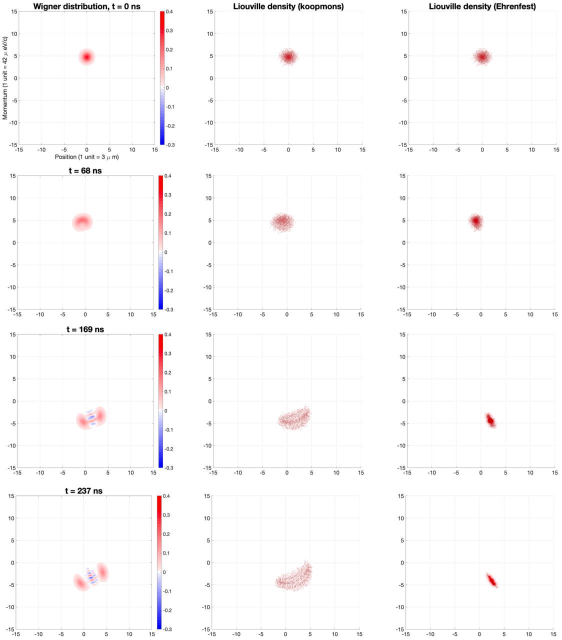
Standard values for N and α; InSb accounts for small coupling

Input: $N = 500$; $\alpha = 0.5$; $q_0 = 0$, $p_0 = 1e-4$; $\rho_0 = [0,0;0,1]$; $t_{\text{fin}} = 237\,\text{ns}$; $dt = t_{\text{fin}}/100$

Model: HO; $\omega = 44\,\text{MHz}$; GaAs; $m = 0.067$; $\alpha_R = 4.7159e-05$; $B_0 = 4.2\,\text{mT}$; Zeeman dominated ($R \approx 0.08$)







Standard values for N and α; GaAs accounts for small coupling, frequency yet to small

Final remarks

- New trajectory-based Hamiltonian approach for simulating MQC dynamics
- The *koopmon* method provides a **ground for developing closure methods** through the underlying variational structure
- Numerical tests for many different models (currently: Rashba Hamiltonians)
- The *koopmon* method can be **extended to higher dimensions**
  $\rightarrow$ Please do ask me about this!

Thank you.

Backup Slides

Koopmons in higher dimensions

The *koopmon* method can be extended to higher dimensions by using only sums of products of integrals in $2d$.

Consider two degrees of freedom $z_1 = (q_1, p_1)$ and $z_2 = (q_2, p_2)$. For a given multi-index $\kappa \in \{(a,b) : a,b = 1,\dots,N\}$ we assume the singular solution ansatz in two dimensions

$$\widehat{P}(z_1, z_2, t) = \sum_\kappa w_\kappa \hat{\rho}_\kappa(t) \delta(z_1 - \zeta_1^{(\kappa)}(t)) \delta(z_2 - \zeta_2^{(\kappa)}(t)).$$

In addition, we restrict this ansatz to consider the case $w_\kappa = w_{(a,b)} = w_a w_b$, along with

$$\zeta_1^{(a,b)} = \zeta_1^{(a,b')} \qquad \forall a, b, b' \qquad \text{and} \qquad \zeta_2^{(a,b)} = \zeta_2^{(a',b)} \qquad \forall b, a, a'.$$

Furthermore, we pick a kernel such that $K(z-z') = K_1(z_1-z_1')K_2(z_2-z_2')$, so that

$$\begin{aligned}\bar{P}(z_1,z_2,t) &= \sum_{a,b} w_a w_b \hat{\rho}_{ab}(t) K_1(z_1-\zeta_1^{(a)}(t))K_2(z_2-\zeta_2^{b}(t)) \\ &=: \sum_{a,b} w_a w_b \hat{\rho}_{ab}(t) K_1^{(a)}(z_1,t) K_2^{(b)}(z_2,t),\end{aligned}$$

as well as $\bar{f} = \sum_{a,b} w_a w_b K_1^{(a)} K_2^{(b)} = \left(\sum_a w_a K_1^{(a)}\right)\left(\sum_b w_b K_2^{(b)}\right)$.

Based on the following class of Hamiltonians,

$$\widehat{H}(z_1, z_2) = H_c(z_1, z_2)1 + \widehat{H}_Q + h_2(z_2)\widehat{H}_1(z_1) + h_1(z_1)\widehat{H}_2(z_2),$$

the backreaction integrals

$$\widehat{I}_{aba'b'} := \iint \left(\frac{K_1^{(a)} K_2^{(b)} K_2^{(b')} \{K_1^{(a')}, \widehat{H}\}_1}{\sum_{c,d} w_c w_d K_1^{(c)} K_2^{(d)}} + \frac{K_1^{(a)} K_2^{(b)} K_1^{(a')} \{K_2^{(b')}, \widehat{H}\}_2}{\sum_{c,d} w_c w_d K_1^{(c)} K_2^{(d)}} \right) \mathrm{d}^2 z_1 \mathrm{d}^2 z_2$$

can be decomposed into a sum of products of integrals, each of dimension 2.

$$\widehat{I}_{aba'b'} = \widehat{I}_1^{(aa')} J_2^{(bb')} + I_1^{(aa')} \widehat{J}_2^{(bb')} + I_2^{(bb')} \widehat{J}_1^{(aa')} + \widehat{I}_2^{(bb')} J_1^{(aa')},$$

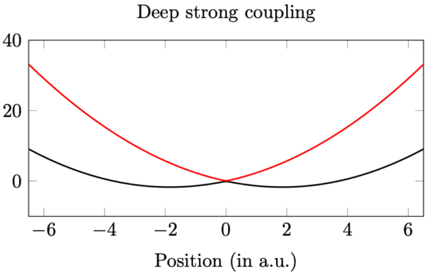
along with the definitions

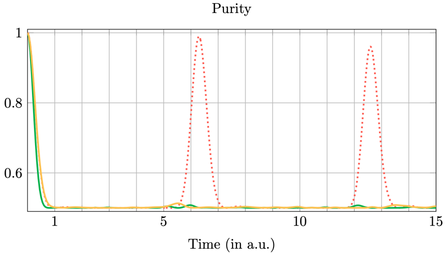
$$I_\ell^{(ss')} := \int \frac{K_\ell^{(s)} \{K_\ell^{(s')}, h_\ell\}_\ell}{\sum_c w_c K_\ell^{(c)}} \, \mathrm{d}^2 z_\ell, \qquad J_\ell^{(ss')} := \int \frac{K_\ell^{(s)} K_\ell^{(s')} h_\ell}{\sum_d w_d K_\ell^{(d)}} \, \mathrm{d}^2 z_\ell,$$

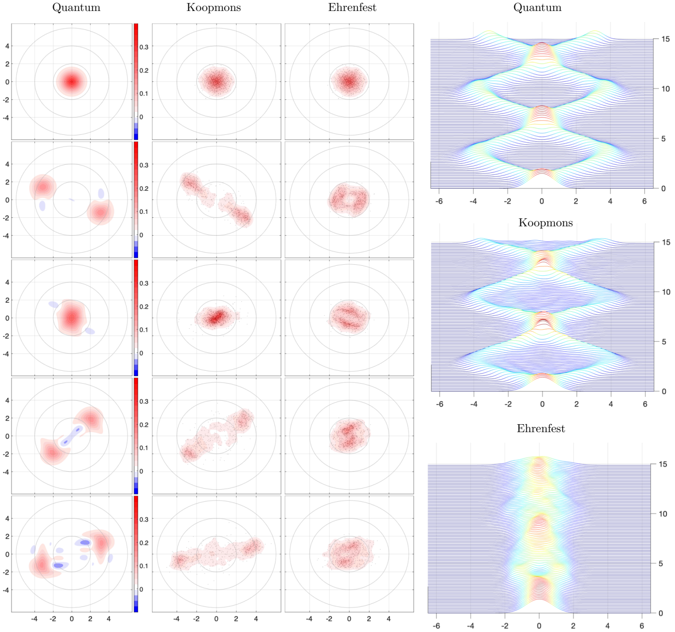
$$\widehat{I}_\ell^{(ss')} := \int \frac{K_\ell^{(s)} \{K_\ell^{(s')}, \widehat{H}_\ell\}_\ell}{\sum_c w_c K_\ell^{(c)}} \, \mathrm{d}^2 z_\ell, \qquad \widehat{J}_\ell^{(ss')} := \int \frac{K_\ell^{(s)} K_\ell^{(s')} \widehat{H}_\ell}{\sum_d w_d K_\ell^{(d)}} \, \mathrm{d}^2 z_\ell,$$

for $\ell = 1, 2$ and $s = a, b$.

Input: $N = 500$; $\alpha = 0.5$; $q_0 = 0$, $p_0 = 0$; $\rho_0 = [1, 1; 1, 1]/2$; $t_{\text{fin}} = 15$; $dt = 0.05$







Standard values for N and α; snapshots at $t = 0$, 4, 6, 8, 15