Non-Linear Correlation Functions and Zero-Point Energy Flow in Mixed Quantum-Classical Semiclassical Dynamics

Shreyas Malpathak and Nandini Ananth
Department of Chemistry and Chemical Biology, Baker Laboratory, Cornell University Ithaca, I4853 NY, USA

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Mixed Quantum Classical (MQC)-IVR is a recently introduced semiclassical framework that allows for selective quantization of the modes of a complex system. In the quantum limit, MQC reproduces the semiclassical Double Herman-Kluk IVR results, accurately capturing nuclear quantum coherences and conserving zero-point energy. However, in the classical limit, while MQC mimics the Husimi-IVR for real-time correlation functions with linear operators, it is significantly less accurate for non-linear correlation functions with errors even at time zero. Here, we identify the origin of this discrepancy in the MQC formulation and propose a modification. We analytically show that the modified MQC approach is exact for all correlation functions at time zero, and in a study of zero-point energy (ZPE) flow, we numerically demonstrate that it correctly obtains the quantum and classical limits as a function of time. Interestingly, while classical-limit MQC simulations show the expected, unphysical ZPE leakage, we find it is possible to predict and even modify the direction of ZPE flow through selective quantization of the system, with the quantum-limit modes accepting energy additions but preserving the minimum quantum mechanically required energy.

I. INTRODUCTION

Probing the effects of the quantum nature of nuclei and nuclear-electronic coupling in chemical and biological systems has been the focus of extensive research in recent years.\textsuperscript{[1]}\textsuperscript{[3]} Numerically exact methods to simulate quantum dynamics have found applications in chemical systems of modest sizes\textsuperscript{[24–26]} but their expensive scaling with system size remains a challenge. Path integral methods that rely on classical trajectories, such as Matsubara dynamics\textsuperscript{[6]} Ring Polymer Molecular Dynamics (RPMD)\textsuperscript{[8]} Centroid Molecular Dynamics (CMD)\textsuperscript{[10,11]} and Multistate Ring Polymer Molecular Dynamics\textsuperscript{[10,11]} have significantly more favorable scaling laws, but fail to capture nuclear quantum coherences.

Semiclassical (SC) Initial Value Representation (IVR) methods have emerged as a rigorous alternative for the simulation of quantum processes using near-classical trajectories. A hierarchy of SC approximations have been established with the most accurate, quantum-limit SC methods able to describe quantum effects such as zero point energy, tunneling, nonadiabatic and interference effects.\textsuperscript{[14,15]} Unfortunately, these quantum-limit methods require the evaluation of a complex oscillatory integrand and the resulting numerical sign problem limits applications to high-dimensional systems. Classical-limit SC methods, like Linearized SC-IVR\textsuperscript{[16,17]} do not capture interference effects but employ only classical trajectories making them suitable for the simulation of condensed phase processes where quantum coherences are short-lived.\textsuperscript{[18,20]}

The recently introduced mixed quantum-classical SC method, MQC-IVR\textsuperscript{[21,22]} has been shown to reduce the cost of quantum-limit SC correlation function calculations using a modified Filinov filtration technique\textsuperscript{[23,26]} to mitigate the effects of oscillatory phase. In the limit of small Filinov filter for all degrees of freedom (dofs) the MQC correlation function becomes identical to a quantum-limit SC-IVR correlation function, specifically the Double Herman-Kluk IVR (DHK-IVR).\textsuperscript{[27,29]} In the limit of large parameters, for linear operators, MQC corresponds to a classical-limit, linearized method, specifically the Husimi-IVR.\textsuperscript{[14,20,24]} As the name suggests, the MQC framework uniquely offers a path to selective quantization — by filtering the phase contribution from different dofs to different extents, it is possible to treat some modes in the quantum limit and others in the classical limit.

In previous work, for a range of 1D and 2D model systems, MQC has been numerically shown to gradually tune linear correlation functions from the quantum to the classical limit\textsuperscript{[21,22]} and to capture nuclear coherence effects in non-adiabatic scattering models.\textsuperscript{[15]} The analytic mixed quantum-classical IVR (AMQC-IVR) method was introduced more recently to treat a handful of system dofs in the quantum-limit while treating the rest in the classical-limit, enabling the calculation of thermal reaction rates in high-dimensional system-bath models.\textsuperscript{[30]} Despite these successes, a recent study showed that for correlation functions of operators that are not linear in position and momentum, MQC is less successful particularly in the classical limit where it yields inaccurate values even at time zero.\textsuperscript{[31]} Given that both DHK-IVR and Husimi-IVR are, in general, exact for correlation functions at time zero, this is a rather startling observation.

In this paper, we demonstrate the origin of the problem: for operators that are not linear in position and momentum, the classical-limit MQC correlation function does not coincide with the corresponding Husimi-IVR expression. We propose a general strategy to modify MQC such that its classical limit reproduces the Husimi-IVR expression for both linear and non-linear correlation functions. We then use this modified MQC expression to characterize zero-point energy (ZPE) flow in a series of model systems.

Quasi-classical and linearized SC methods typically exhibit ZPE ‘leakage’ in time, with high frequency modes losing zero-point energy to lower frequency ones.\textsuperscript{[31,32]} This non-
The function $f$ and the phase space function of Wigner density is expressed as,

\[ \rho_W(z) = \frac{1}{2\pi\hbar} \int d\Delta \left( \frac{\Delta}{2} \right) e^{-i p \Delta / \hbar}, \quad (5) \]

resulting in,

\[ B_W(z) = \int d\Delta \left( \frac{\Delta}{2} \right) e^{-i p \Delta / \hbar}. \quad (6) \]

Comparing Eq. [5] and Eq. [6] we see that the Wigner correspondence rules for the density operator and for general operators $\hat{B}$ differ only by a factor of $\left(2\pi\hbar\right)^{-1}$ that ensures the phase space density is normalized.

In the Husimi formulation, the correspondence rules are different with,

\[ f(\xi, \eta) = e^{-\frac{\xi^2}{\hbar^2} - \frac{\eta^2}{4\gamma^2}}, \quad (7) \]

resulting in,

\[ \rho_H(z) = \frac{1}{2\pi\hbar} (z|\hat{\rho}|z), \quad (8) \]

and,

\[ \hat{B}_H(z) = \frac{2\pi\hbar}{4\pi^2} \int_{-\infty}^{\infty} d\xi \int_{-\infty}^{\infty} d\eta \ e^{i\xi(\hat{x}-x)+i\eta(\hat{p}-p)} \hat{B} \]

\[ = \left(\frac{\gamma}{\pi}\right)^{1/4} e^{-\frac{(x-q)^2}{\gamma^2}+i p(x-q)/\hbar}, \quad (9) \]

referred to as the anti-Husimi transform of $\hat{B}$. Here, $|z\rangle$ is a coherent state with width $\gamma$ centered at $z$.

The Husimi density in Eq. [8] is proportional to the diagonal coherent state matrix element of the density operator, whereas this is not true for a general operator $\hat{B}$, except in special cases like $\hat{B} \equiv \hat{x}$ or $\hat{p}$. It is this difference that changes the accuracy of the the classical-limit of the MQC correlation function even at time zero, as shown in [11B].

To find the anti-Husimi transform of an operator, it is useful to establish a connection between the Wigner and Husimi functions:

\[ B_H(z) = \hat{G}(\gamma, q)|\hat{G}(\gamma^{-1} h^{-2}; p) B_W(z), \quad (11) \]

\[ \hat{B}_H(z) = \hat{G}^{-1}(\gamma, q)|\hat{G}^{-1}(\gamma^{-1} h^{-2}; p) B_W(z), \quad (12) \]

and,

\[ B_H(z) = \hat{G}\left(\gamma^{-1}, q\right) \hat{G}^{-1}\left(\gamma^{-1} h^{-2}; p\right) \hat{B}_H(z), \quad (13) \]

where $\hat{G}$ is a Gaussian convolution operator defined as,

\[ \hat{G}(\gamma, x) f(x) \equiv \left(\frac{\gamma}{\pi}\right)^{1/2} \int dx' e^{-\gamma(x-x')^2} f(x') \quad (14) \]

\[ = \frac{1}{\sqrt{2\pi}} e^{\frac{x^2}{\gamma^2}} f(x). \quad (15) \]
B. MQC Correlation Functions at Time Zero

A general, real-time quantum correlation function can be expressed as,

\[ C_{AB}(t) = \text{Tr} \left[ \hat{\rho} \hat{B}(t) \right] . \]

(16)

where \( \hat{H} \) is the system Hamiltonian, \( \hat{\rho} \) is the density, and \( \hat{A} \) and \( \hat{B} \) are the operators evaluated at time zero and time \( t \) respectively. Evaluated in a phase space formulation, this expression is,

\[ C_{AB}(t) = \int dz \left[ \hat{\rho} \right](z) \left[ \hat{B}(t) \right](z), \]

(17)

where \( [\cdot](z) \) is the phase space function of the operator. In the following sections, for simplicity, we discuss the time-dependence of expectation values (corresponding to \( \hat{A} = 1 \)) noting that all the expressions derived can be applied to correlation functions by replacing \( \hat{\rho} \) with \( \hat{\rho} \hat{A} \).

The MQC correlation function is obtained by Filinov filtering the phase of the quantum-limit DHK-IVR correlation function to obtain a phase space expression. Here we consider the MQC approximation to the time-dependent expectation value of a general operator \( \hat{B} \) for a 1D system

\[ \langle \hat{B}(t) \rangle_{\text{MQC}} = \frac{1}{(2\pi)^2} \int dz_0 \int dz_0' D(z_0, z_0'; c_p, c_q) \times \langle z_0 | \hat{B} | z_0' \rangle e^{i(S(z_0) - S(z_0'))} e^{-\frac{1}{2}c_p^2 \Delta z_0^2} e^{-\frac{1}{2}c_q^2 \Delta z_0^2}, \]

(18)

working in atomic units with \( h = 1 \) a.u. In Eq. (18) \( \hat{\rho} \) is the initial density operator for the system, \( D(z_0, z_0'; c_p, c_q) \) is the MQC prefactor, \( c_q \) and \( c_p \) are Filinov parameters, and \( \Delta z_0 = z_0 - z_0' \) for \( x \in \{ p, q \} \) are the difference variables. The phase space variables, \( z_0 \) and \( z_0' \), in Eq. (18) are obtained by propagating classical trajectories for time \( t \) under the classical Hamiltonian with initial conditions, \( z_0 \), and \( z_0' \), respectively, and \( S(z_0) \) and \( S(z_0') \) are the actions of forward and backward trajectories respectively.

At time zero, the MQC prefactor is simply

\[ D = \sqrt{(c_q + \gamma)(1 + c_p)}, \]

(19)

and the expectation value is,

\[ \langle \hat{B}(0) \rangle_{\text{MQC}} = \sqrt{(c_q + \gamma)(1 + c_p)} \int dz_0 \int dz_0' \langle z_0 | \hat{B} | z_0' \rangle e^{-\frac{1}{2}c_p^2 \Delta z_0^2} e^{-\frac{1}{2}c_q^2 \Delta z_0^2}. \]

(20)

For \( c = \{ c_p, c_q \} = 0 \), Eq. (20) reduces to the DHK expression,

\[ \langle \hat{B}(0) \rangle_{\text{DHK}} = \lim_{c \rightarrow 0} \langle \hat{B}(0) \rangle_{\text{MQC}} = \frac{1}{(2\pi)^2} \int dz_0 \int dz_0' \langle z_0 | \hat{B} | z_0' \rangle e^{-\frac{1}{2}c_p^2 \Delta z_0^2} e^{-\frac{1}{2}c_q^2 \Delta z_0^2}. \]

(21)

where we use the coherent state completeness relation,

\[ \hat{1} = \frac{1}{2\pi} \int dz_0 | z_0 \rangle \langle z_0 |. \]

(22)

In the classical limit (large Filinov parameters), the MQC expectation value is

\[ \lim_{c \rightarrow \infty} \langle \hat{B}(0) \rangle_{\text{MQC}} = \frac{1}{2\pi} \int dz_0 \langle z_0 | \hat{B} | z_0 \rangle = \langle \hat{B}(0) \rangle_{\text{exact}}, \]

(23)

where the inequality follows from the definition of \( \hat{B}_H \) in Eq. (9).

We have shown that while MQC is exact at time zero in the quantum limit (\( c \rightarrow 0 \)), it is not, in general, exact in the classical limit (\( c \rightarrow \infty \)). To understand the behaviour of MQC at time zero for finite non-zero values of the Filinov parameter, we insert the identity into Eq. (20) for both \( \hat{\rho} \) and \( \hat{B} \),

\[ \hat{B} = \frac{1}{2\pi} \int dz \hat{B}_H(z) | z \rangle \langle z | \]

(24)

Eq. (26) are functions of \( \gamma \) and the Filinov parameters, \( c_q \) and
c_p, respectively. Detailed definitions of $\hat{y}_q$ and $\hat{p}_p$ are provided in Appendix A along with details of the intermediate steps in the derivation.

The inequality in Eq. (26) indicates that for finite values of the Filinov parameter, MQC is not exact at zero time for a general operator $\hat{B}$. The exceptions are the position and momentum operators, $\hat{B} = \hat{x}$ or $\hat{p}$, where

$$\hat{G}(\hat{y}_q^1, q)\hat{G}(\hat{p}_p^{-1}, p)\hat{B}_H(z) = \hat{B}_H(z),$$

(28)

for all values of $c$, a finding that is consistent with previously published numerical results for correlation functions involving these operators.\cite{Filinov1979, Filinov1980}

We have shown that for a general operator, $\hat{B}$, the MQC correlation function is inaccurate at zero time for non-zero values of the Filinov parameter, and that its classical-limit does not reproduce the Husimi-IVR expression. One potential approach to this problem would be to reformulate MQC by modifying the choice of phase that is filtered. In the next section, we describe a second strategy that allows us to work within the present MQC formalism using a modified operator $\hat{B}$.

### C. Modifying the MQC Correlation Function

Comparing Eq. (26) and Eq. (27) suggests that we can correct the MQC correlation function by replacing operator $\hat{B}$ by a new operator $\hat{\mathcal{B}}$, such that

$$\hat{G}(\hat{y}_q^1, q)\hat{G}(\hat{p}_p^{-1}, p)\hat{B}_H(z) = \hat{B}_H(z).$$

(29)

or equivalently using the relationship between the Anti-Husimi transform and the Wigner transform in Eq. (12),

$$\hat{G}(\hat{y}_q^1, q)\hat{G}(\hat{p}_p^{-1}, p)\hat{B}_W(z) = \hat{B}_W(z).$$

(30)

The MQC expression with the modified operator is still exact in the quantum limit; when $c \to 0$, the Gaussian convolutions in Eq. (29) reduce to delta functions, such that

$$\hat{\mathcal{B}} = \hat{B}.$$  

(31)

In the classical limit, $c \to \infty$, the Filinov-dependent width parameters $\hat{y}_q \to \frac{2}{\gamma}$ and $\hat{p}_p \to 2\gamma$, and using Eq. (13) we obtain

$$\lim_{c \to \infty} \hat{B}_H(z) = \hat{B}_H(z).$$

(32)

The modified MQC expression thus achieves the correct form in both the quantum and classical limits, with

$$\lim_{c \to 0} \langle \hat{\mathcal{B}}(t) \rangle_{\text{MQC}} = \langle \hat{B}(t) \rangle_{\text{DHK}},$$

(33)

and

$$\lim_{c \to \infty} \langle \hat{\mathcal{B}}(t) \rangle_{\text{MQC}} = \langle \hat{B}(t) \rangle_{\text{Hus}}.$$  

(34)

Although our analysis and derivation of the modified operator is shown here for the expectation value, this idea can be simply extended to real-time correlation functions resulting in the modified MQC expression,

$$C_{AB}(t) = \frac{1}{(2\pi)^2} \int d\zeta_0 \int d\zeta'_0 D(\zeta_0, \zeta'_0, c_p, c_q)$$

$$\times \langle \zeta_0 | \hat{\mathcal{B}} | \zeta'_0 \rangle \langle \zeta'_0 | \hat{\mathcal{B}} | \zeta_0 \rangle e^{i[S(t_0) - S(t_0)]}$$

$$\times e^{-\frac{1}{2}c_p |\Delta_{t_0}|^2} e^{-\frac{1}{2}c_q |\Delta_{t_0}|^2}.$$  

(35)

Finding the modified operator $\hat{\mathcal{B}}$ can be non-trivial, but for simple operators it is possible to derive an analytic expression. For instance, the modified MQC correlation function when $\hat{B} = \hat{x}^2$ is obtained with the modified operator $\hat{\mathcal{B}}^2$,

$$\hat{x}^2 \to \hat{\mathcal{B}}^2 = \hat{x}^2 - \frac{c_p}{1 + c_q^2}.$$  

(36)

and similarly the modified MQC correlation function for $\hat{B} = \hat{p}^2$ is obtained with the modified operator $\hat{\mathcal{B}}^2$,

$$\hat{p}^2 \to \hat{\mathcal{B}}^2 = \hat{p}^2 - \frac{c_q^2}{\gamma + c_q}.$$  

(37)

### III. MODEL SYSTEM AND SIMULATION DETAILS

Inspired by a model previously used to study ZPE leakage in SC dynamics, we consider three different models of coupled harmonic oscillators with cubic couplings,

$$V(x_1, \ldots, x_F) = \sum_{i=1}^F \frac{1}{2} m \omega_i^2 x_i^2 + \frac{1}{2} \sum_{i<j} C_{ij} (x_i - x_j)^3,$$

(38)

where $F$ is number of degrees of freedom, all oscillators have mass $m = 1$ a.u., $\omega_i$ is the frequency of the $i^{th}$ oscillator, and $C_{ij}$ is the pair-wise coupling between oscillators. Model A is a two dimensional model — two harmonic oscillators with cubic coupling. Models B and C are three dimensional models where one oscillator is weakly coupled to a subsystem comprising two strongly coupled oscillators. In model C, two weakly coupled oscillators have the same frequency to explore the effect of having resonant modes treated with different levels of quantization. Frequencies of the oscillators and coupling constants for all models are listed in Table I.

TABLE I. Parameters for the three model systems (in atomic units).

| Model | F | $\omega_1$ | $\omega_2$ | $\omega_3$ | $C_{12}$ | $C_{23}$ | $C_{13}$ |
|-------|---|------------|------------|------------|-----------|-----------|----------|
| A     | 2 | 1.0        | 0.5        | 10^{-4}    | -         | -         | -        |
| B     | 3 | 1.0        | 0.5        | 0.25       | 10^{-4}   | 2 \times 10^{-4} | 0.0    |
| C     | 3 | 1.0        | 1.0        | 0.5        | 10^{-4}   | 2 \times 10^{-4} | 0.0    |

We chose the initial coherent state $|\psi_i\rangle = |z_i\rangle$ to correspond to the ground-state of the uncoupled harmonic oscillators, a product of coherent states with widths $\gamma = m \omega_i$, centered at $z_i = (0, 0)$. Since the magnitude of the coupling constants is very small, the overlap between the initial state and the ground state of the coupled system is nearly unity ensuring that only
the lowest vibrational state for each oscillator is populated. The total energy can then be written as,

\[ E = \sum_i E_i \]  

(39)

where the \( E_i \) is the ZPE of the \( i^{th} \) oscillator.

To track the flow of ZPE, we calculate the energy expectation value of each oscillator as a function of time,

\[ \langle \hat{E}_i(t) \rangle = \frac{1}{2m} \langle \hat{p}_i^2(t) \rangle + \frac{1}{2} m \omega_i^2 \langle \hat{x}_i^2(t) \rangle. \]

(40)

In our modified MQC implementation, the energy expectation value in Eq. (40) is calculated by evaluating two independent non-linear correlation functions using Eq. (35). Specifically, the \( \langle \hat{x}_i^2(t) \rangle \) term is obtained using modified operator \( \hat{B} = \hat{x}_i^2(t) \) defined in Eq. (36) and the \( \langle \hat{p}_i^2(t) \rangle \) term is obtained using modified operator \( \hat{B} = \hat{x}_i^2(t) \) defined in Eq. (37).

Numerical integrals are evaluated using standard Monte Carlo techniques with the sampling function,

\[ \rho \left( z_0, z_0' \right) = \mathcal{N} \left| \langle \psi_i | \psi_i' \rangle e^{-\frac{1}{2} \left( q_0 - q_0' \right)^T c_p \left( q_0 - q_0' \right) } \right| \times e^{-\frac{1}{2} \left( p_0 - p_0' \right)^T c_p \left( p_0 - p_0' \right) }, \]

(41)

where \( \mathcal{N} \) ensures proper normalization. Note that while the sampling is performed using sum and difference variables, we then transform back into the \( (z_0, z_0') \) for trajectory propagation and calculation of the estimator. Trajectories are propagated under the classical Hamiltonian using a fourth-order symplectic integrator with a timestep of 0.1 a.u. for Model A, and 0.175 a.u. for models B and C to ensure total energy conservation. All calculations presented here were converged to within the error bars shown using at most \( 10^5 \) trajectories.

Finally, we note that we renormalize the energy in each mode such that the total energy stays constant at all times,

\[ \sum_{i=1}^{Z} \langle \hat{E}_i(t) \rangle = \sum_{i=1}^{Z} \langle \hat{E}_i(0) \rangle. \]

This is required because SC methods used here are, at best, only approximately unitary.

IV. RESULTS AND DISCUSSION

Using similar model systems, previous work has established that DHK-IVR, a quantum-limit SC method, conserves ZPE whereas LSC-IVR, a classical-limit SC method does not. Here, we use the modified MQC method to establish the extent of ZPE leakage in dynamics that employ finite values of the Filinov parameter. Further, we investigate how selective quantization of modes influences ZPE flow, with the potential to limit the extent to which the unphysical flow of energy occurs within a subsystem.

In Fig. 1, we demonstrate that the modified MQC expression, unlike the original, reproduces the exact values for the energy expectation value of a single oscillator in 2D model A for all values of the Filinov parameter. We also show that the classical limit of the modified MQC also correctly reproduces the Husimi IVR result with significant ZPE leakage.

Having established the accuracy of the modified MQC expression in capturing both quantum-limit and classical-limit SC dynamics, we explore the extent to which this method conserves ZPE for different values of the Filinov parameter. In Fig. 2, we plot the average energies of the two oscillators in model A. As expected, DHK-IVR and quantum-limit MQC, \( c_1 = c_2 = c = 0.01 \), both show ZPE conservation for the length of the simulation. On the other hand, Husimi-IVR and classical-limit MQC, \( c = 100 \), both exhibit significant loss of ZPE with unphysical energy flow from the high frequency oscillator into the low frequency one. As the Filinov parameter is increased, we see a systematic onset of ZPE leakage from the high frequency mode, consistent with the idea that SC methods rely on interference to conserve ZPE. It is notable that despite increasing the Filinov parameter 10-fold, MQC with \( c = 0.1 \) still exhibits reasonable ZPE conservation and requires far few trajectories for convergence than the corresponding \( c = 0.01 \) MQC simulation.

Working with 2D model A, we also perform MQC simulations where one mode is treated in the quantum limit while the other is described in the classical limit. In Fig. 3, we demonstrate that the mode that is treated in the quantum limit does indeed conserve ZPE, whereas the classical-limit mode does not. Interestingly, when we quantize the low frequency oscillator, there is no observed change in ZPE flow with the classical-limit high-frequency mode continuing to lose energy. However, when we treat the high frequency mode in the quantum limit, we stem the loss of energy from this mode and instead see a reversal with energy flowing uphill from the classical-limit low-frequency mode.

Model B offers a more interesting test case for mixed quantization. For this model, in the quantum limit, all three oscillators conserve ZPE as shown in Fig. 4 and in the classical limit, we see rapid ZPE exchange between the two lower fre-
frequency oscillators that are strongly coupled, with the weakly coupled high frequency oscillator losing ZPE to the other two modes on a longer timescale. In the first study of mixed quantization, we treat the high frequency oscillator in the quantum limit while describing the rest in the classical limit. As shown in Fig. 4(a), we find that the classical ‘bath’ oscillators continue to exchange ZPE but the quantum-limit oscillator approximately conserves ZPE over a significantly longer timescale (by a factor of 2) with some increase in energy corresponding to leakage from the classical subsystem.
V. CONCLUSIONS

In this paper, we begin with a detailed study of the origin of the MQC SC method’s inaccuracy at time zero for non-linear correlation functions. We show that MQC with finite, non-zero values of the Filinov parameter is not exact at time zero for a general operator $\hat{B}$. Further, we show that the real-time MQC correlation functions in the classical limit does not to the Husimi IVR correlation function. We perform a detailed analysis of the Husimi and Wigner phase space formulation of quantum mechanics and identify a simple modification to correct for both these problems. Specifically, we propose that replacing operator $\hat{B}$ with a new operator that is obtained through inverse Gaussian convolutions of Wigner/Anti-Husimi functions of the original operator. We analytically derive the modified operator for $x^2$ and $\hat{p}^2$, and use the resulting expression to characterize ZPE flow in MQC simulations.

We construct a series of model systems comprising oscillators of different frequencies and with different coupling motifs chosen to mimic real system connectivities. We demonstrate that by selective quantization of weakly coupled modes, it is possible to improve the timescale on which ZPE is conserved for a subset of modes. This work establishes a promising path forward to real system simulations using the modified MQC approach proposed here. Next steps include establishing a path to deriving modified operators corresponding to other operators, including most notably the projection operator.

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Appendix A: Detailed derivation of the modified MQC method

Starting with Eq. (25), we note that the integrals over \( z_0 \) and \( z'_0 \) can be evaluated analytically to yield,
\[
\langle \hat{B}(0) \rangle_{MQC} = \sqrt{\frac{c_y + \gamma}{2\pi \gamma}} \int dz_0 \int dz'_0 \int dz'' \int dz''' \langle z_0, z' \rangle \rho_H(z') \langle z' | z'' \rangle \langle z'' | z''' \rangle \hat{B}_H(z''') \langle z''' | z_0 \rangle e^{-\frac{1}{2}c_y z_0^2} e^{-\frac{1}{2}c_p z_0^2}
\]
\[
= \frac{1}{2\pi} \sqrt{\frac{\gamma}{\pi}} \int dz' \int dz'' e^{-\gamma(x'-x'')^2} e^{-\gamma'(x'-p')^2} \rho_H(z') \hat{B}_H(z'')
\]
(A1)

where \( \gamma_p = \frac{\gamma y'}{2(2y' + \gamma')^2} \) and \( \gamma_p' = \frac{1}{2\gamma} \frac{\gamma}{\gamma' + \gamma} \). We then split the Gaussian convolution with width \( \gamma \) into two separate Gaussian convolutions each, \( \gamma_1^{-1} = \gamma_2^{-1} \). We now define \( \gamma_1 = \frac{\gamma y}{2(2y + \gamma)^2} \), and \( \gamma_1' = \frac{1}{2\gamma} \frac{\gamma}{\gamma' + \gamma} \), such that \( \gamma_1^{-1} = \gamma_1^{-1} + 2\gamma^{-1} \), (A3)

and

\[
\gamma_p = \gamma_p' + 2\gamma \quad \text{(A4)}
\]

to split the Gaussian convolutions in Eq. (A1) into two Gaussian convolutions each,
\[
\langle \hat{B}(0) \rangle_{MQC} = \frac{1}{2\pi} \int dz \left[ \frac{1}{2\pi} \int dz'' \int dz''' e^{-\frac{1}{2}(y-q)^2} e^{-\frac{1}{2}(p-p')^2} \rho_H(z') \right] \left[ \sqrt{\frac{\gamma}{\gamma'}} \int dz'' e^{-\gamma y(y-q)^2} e^{-\gamma'/(p-p')^2} \hat{B}_H(z'') \right]
\]
(A5)

To go from Eq. (A5) to Eq. (A6), we have used the definition of a Gaussian convolution from Eq. (14), and then used the relationship between the Husimi transform and the anti-Husimi transform of an operator, Eq. (17), to yield Eq. (A7), which is the same as Eq. (26).
