Supporting Information: An exact imaginary-time path-integral phase-space formulation of multi-time correlation functions

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(Dated: February 4, 2023)

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CONTENTS

I. GENERALIZED WIGNER-WEYL TRANSFORMS IDENTITIES

We begin by proving some identities for the ring-polymer generalized Wigner-Weyl transforms. The ring-polymer phase-space function $[\hat{O}]_N(\mathbf{q},\mathbf{p})$ is the generalized Wigner-Weyl transform defined, as follows:

$$\left[\hat{O}\right]_N(\mathbf{q},\mathbf{p}) \equiv \frac{1}{N}\sum_{j=1}^{N}\left[\hat{O}\right]_W(q_j,p_j), \tag{1}$$

where

$$\left[\hat{O}\right]_W(q,p) \equiv \int d\Delta\; e^{\frac{i}{\hbar}p\Delta}\,\langle q-\frac{\Delta}{2}|\,\hat{O}\,|q+\frac{\Delta}{2}\rangle\,, \tag{2}$$

is the one-dimension Wigner-Weyl transform.[1–4] On the other hand, the generalized Wigner Boltzmann transform $[e^{-\beta\hat{H}}]_{\overline{N}}(\mathbf{q},\mathbf{p})$ is the Wigner-Weyl transform defined as

$$\left[e^{-\beta\hat{H}}\right]_{\overline{N}}(\mathbf{q},\mathbf{p}) \;\equiv\; \int d\boldsymbol{\Delta}\;\prod_{l=1}^{N}e^{\frac{i}{\hbar}p_l\Delta_l}\,\langle q_{l-1}-\frac{\Delta_{l-1}}{2}|\,e^{-\beta_N\hat{H}}\,|q_l+\frac{\Delta_l}{2}\rangle\,, \tag{3}$$

with $\beta_N=\beta/N$.

To keep the presentation clear, we will use the shorthand notation

$$\left[\hat{O}\right]_{W,j} \;\equiv\; \left[\hat{O}\right]_W(q_j,p_j), \tag{4}$$

to denote the dependence on the j-th coordinates of the (one-dimension) Wigner-Weyl transform. Additionally, we will use the following well-known properties of the Wigner transforms: [1–5]

- Moyal or Sine bracket

$$\left(\frac{i}{h}\right)\left[[\hat{O}_1,\hat{O}_2]\right]_W \;=\; \left(\frac{2}{h}\right)\left[\hat{O}_1\right]_W\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}\right)\left[\hat{O}_2\right]_W. \tag{5}$$

- Baker or Cosine bracket

$$\left(\frac{1}{2}\right)\left[[\hat{O}_1,\hat{O}_2]_+\right]_W \;=\; \left[\hat{O}_1\right]_W\cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}\right)\left[\hat{O}_2\right]_W. \tag{6}$$

- Wigner-Moyal series

$$\left[\hat{O}(t)\right]_W = e^{\mathcal{L}t}\left[\hat{O}\right]_W . \tag{7}$$

In the previous equations,

$$\overleftrightarrow{\Lambda} = \frac{\overleftarrow{\partial}}{\partial p}\frac{\overrightarrow{\partial}}{\partial q} - \frac{\overleftarrow{\partial}}{\partial q}\frac{\overrightarrow{\partial}}{\partial p}, \tag{8}$$

represents the negative Poisson bracket and $\mathcal{L}$ is the quantum Liouvillian operator defined as

$$\begin{aligned}\mathcal{L} &\equiv \frac{2}{\hbar}\left[\hat{H}\right]_W \sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}\right)\\ &= \frac{p}{m}\frac{\overrightarrow{\partial}}{\partial q} - \frac{2}{\hbar}V(q)\sin\left(\frac{\hbar}{2}\frac{\overleftarrow{\partial}}{\partial q}\frac{\overrightarrow{\partial}}{\partial p}\right).\end{aligned} \tag{9}$$

Finally, we will make extensive use of the integral representation of the delta function,

$$\delta(\Delta) = \frac{1}{2\pi\hbar}\int dp\; e^{\frac{i}{\hbar}p\Delta}, \tag{10}$$

and the operator identity

$$\begin{aligned}\hat{1} &= \int dq\;\; |q\rangle\langle q|\\ &= \int dq\int d\Delta\;\delta(\Delta)\,|q+\frac{\Delta}{2}\rangle\langle q-\frac{\Delta}{2}|\\ &= \frac{1}{2\pi\hbar}\int dq\int d\Delta\int dp\; e^{\frac{i}{\hbar}p\Delta}\;\; |q+\frac{\Delta}{2}\rangle\langle q-\frac{\Delta}{2}|\,.\end{aligned} \tag{11}$$

A. Ring-Polymer Partition Function

Here we show that the ring-polymer partition function, defined as

$$Z_N \equiv \frac{1}{(2\pi\hbar)^N}\int d\mathbf{q}\int d\mathbf{p}\;\left[e^{-\beta\hat{H}}\right]_{\overline{N}}(\mathbf{q},\mathbf{p}), \tag{12}$$

is equivalent to the partition function in Hilbert space, namely

$$Z_N = Z, \tag{13}$$

where

$$Z \equiv Tr\left[e^{-\beta\hat{H}}\right]. \tag{14}$$

The proof follows from

$$
\begin{aligned}
Z_N &= \frac{1}{(2\pi\hbar)^N}\int d\mathbf{q}\int d\mathbf{p}\ \left[e^{-\beta\hat{H}}\right]_{\overline{N}}(\mathbf{q},\mathbf{p}) \\
&= \frac{1}{(2\pi\hbar)^N}\int d\mathbf{q}\int d\mathbf{p}\int d\boldsymbol{\Delta}\ \prod_{l=1}^{N} e^{\frac{i}{\hbar}p_l\Delta_l}\,\langle q_{l-1}-\frac{\Delta_{l-1}}{2}|\,e^{-\beta_N\hat{H}}\,|q_l+\frac{\Delta_l}{2}\rangle \\
&= \frac{1}{(2\pi\hbar)^N}\int d\mathbf{q}\int d\mathbf{p}\int d\boldsymbol{\Delta}\ e^{\frac{i}{\hbar}p_1\Delta_1}e^{\frac{i}{\hbar}p_2\Delta_2}\cdots e^{\frac{i}{\hbar}p_N\Delta_N}\,\langle q_N-\frac{\Delta_N}{2}|\,e^{-\beta_N\hat{H}}\,|q_1+\frac{\Delta_1}{2}\rangle \\
&\quad\times\langle q_1-\frac{\Delta_1}{2}|\,e^{-\beta_N\hat{H}}\,|q_2+\frac{\Delta_2}{2}\rangle\cdots\langle q_{N-1}-\frac{\Delta_{N-1}}{2}|\,e^{-\beta_N\hat{H}}\,|q_N+\frac{\Delta_N}{2}\rangle \\
&= \frac{1}{2\pi\hbar}\int dq_N\int dp_N\int d\Delta_N\ e^{\frac{i}{\hbar}p_N\Delta_N}\,\langle q_N-\frac{\Delta_N}{2}|\,e^{-\beta\hat{H}}\,|q_N+\frac{\Delta_N}{2}\rangle \\
&= \int dq_N\int d\Delta_N\ \delta(\Delta_N)\,\langle q_N-\frac{\Delta_N}{2}|\,e^{-\beta\hat{H}}\,|q_N+\frac{\Delta_N}{2}\rangle \\
&= \int dq_N\ \langle q_N|\,e^{-\beta\hat{H}}\,|q_N\rangle \\
&= Tr\left[e^{-\beta\hat{H}}\right] \\
&= Z.
\end{aligned}
\tag{15}
$$

We used the definition of Z_N [Eq. (12)] at the first equality and the the definition of $[e^{-\beta\hat{H}}]_{\overline{N}}$ [Eq. (3)] at the second equality. At the third equality, we expand the productorial, noting that $q_0 = q_N$ and $\Delta_0 = \Delta_N$. To obtain the fourth equality, we used the identity Eq. (11) to integrate out the variables p_k, q_k and Δ_k for $k \neq N$, and recognized that $\left(e^{-\beta_N\hat{H}}\right)^N = e^{-\beta\hat{H}}$. At the fifth equality, we performed the integral over p_N to obtain the delta function $\delta(\Delta_N)$ [Eq. (10)]. The sixth equality follows from the integration over Δ_N. At the seventh equality, we recognized a trace in position basis. The final equality follows from the definition of the partition function in Hilbert space. This completes the proof of Eq. (13).

B. Ring-Polymer Phase-Space Averages

Here we prove that the ring-polymer average, defined as

$$
\left\langle \hat{O}(t)\right\rangle_N \equiv \frac{Z_N^{-1}}{(2\pi\hbar)^N}\int d\mathbf{q}\int d\mathbf{p}\ \left[e^{-\beta\hat{H}}\right]_{\overline{N}}(\mathbf{q},\mathbf{p})\left[\hat{O}(t)\right]_N(\mathbf{q},\mathbf{p}), \tag{16}
$$

is equivalent to an average in Hilbert space, namely

$$
\left\langle \hat{O}(t)\right\rangle_N = \left\langle \hat{O}(t)\right\rangle, \tag{17}
$$

where

$$\left\langle \hat{O}(t) \right\rangle \equiv Z^{-1} Tr \left[e^{-\beta \hat{H}} \hat{O}(t) \right]. \tag{18}$$

The proof follows from

$$\begin{aligned}
\left\langle \hat{O}(t) \right\rangle_N &= \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \ \left[e^{-\beta \hat{H}} \right]_{\overline{N}} (\mathbf{q},\mathbf{p}) \left[\hat{O}(t) \right]_N (\mathbf{q},\mathbf{p}) \\
&= \frac{1}{N} \sum_{j=1}^{N} \frac{Z^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \int d\mathbf{\Delta} \ \left[e^{-\beta \hat{H}} \right]_{\overline{N}} (\mathbf{q},\mathbf{p}) \left[\hat{O}(t) \right]_{W,j} \\
&= \frac{1}{N} \sum_{j=1}^{N} \frac{Z^{-1}}{(2\pi\hbar)^2} \int dq_N \int dq_j \int dp_N \int dp_j \int d\Delta_N \int d\Delta_j \ e^{\frac{i}{\hbar} p_N \Delta_N} e^{\frac{i}{\hbar} p_j \Delta_j} \\
&\quad \times \langle q_N - \frac{\Delta_N}{2} | \left(e^{-\beta_N \hat{H}} \right)^j | q_j + \frac{\Delta_j}{2} \rangle \langle q_j - \frac{\Delta_j}{2} | \left(e^{-\beta_N \hat{H}} \right)^{N-j} | q_N + \frac{\Delta_N}{2} \rangle \left[\hat{O}(t) \right]_{W,j} \\
&= \frac{1}{N} \sum_{j=1}^{N} \frac{Z^{-1}}{(2\pi\hbar)^2} \int dq_N \int dq_j \int dp_N \int dp_j \int d\Delta_N \int d\Delta_j \int d\Delta_j' \\
&\quad \times e^{\frac{i}{\hbar} p_N \Delta_N} e^{\frac{i}{\hbar} p_j \Delta_j} e^{\frac{i}{\hbar} p_j \Delta_j'} \\
&\quad \times \langle q_N - \frac{\Delta_N}{2} | \left(e^{-\beta_N \hat{H}} \right)^j | q_j + \frac{\Delta_j}{2} \rangle \langle q_j - \frac{\Delta_j}{2} | \left(e^{-\beta_N \hat{H}} \right)^{N-j} | q_N + \frac{\Delta_N}{2} \rangle \\
&\quad \times \langle q_j - \frac{\Delta_j'}{2} | \hat{O}(t) | q_j + \frac{\Delta_j'}{2} \rangle \\
&= \frac{1}{N} \sum_{j=1}^{N} \frac{Z^{-1}}{2\pi\hbar} \int dq_N \int dq_j \int dp_N \int d\Delta_N \int d\Delta_j \int d\Delta_j' \ e^{\frac{i}{\hbar} p_N \Delta_N} \delta(\Delta_j + \Delta_j') \\
&\quad \times \langle q_N - \frac{\Delta_N}{2} | \left(e^{-\beta_N \hat{H}} \right)^j | q_j + \frac{\Delta_j}{2} \rangle \langle q_j - \frac{\Delta_j}{2} | \left(e^{-\beta_N \hat{H}} \right)^{N-j} | q_N + \frac{\Delta_N}{2} \rangle \\
&\quad \times \langle q_j - \frac{\Delta_j'}{2} | \hat{O}(t) | q_j + \frac{\Delta_j'}{2} \rangle \\
&= \frac{1}{N} \sum_{j=1}^{N} \frac{Z^{-1}}{2\pi\hbar} \int dq_N \int dq_j \int dp_N \int d\Delta_N \int d\Delta_j \ e^{\frac{i}{\hbar} p_N \Delta_N} \\
&\quad \times \langle q_N - \frac{\Delta_N}{2} | \left(e^{-\beta_N \hat{H}} \right)^j | q_j + \frac{\Delta_j}{2} \rangle \langle q_j - \frac{\Delta_j}{2} | \left(e^{-\beta_N \hat{H}} \right)^{N-j} | q_N + \frac{\Delta_N}{2} \rangle \\
&\quad \times \langle q_j + \frac{\Delta_j}{2} | \hat{O}(t) | q_j - \frac{\Delta_j}{2} \rangle \\
&= \frac{1}{N} \sum_{j=1}^{N} \frac{Z^{-1}}{2\pi\hbar} \int dq_N \int dp_N \int d\Delta_N \int dx_+ \int dx_- \ e^{\frac{i}{\hbar} p_N \Delta_N} \\
&\quad \times \langle q_N - \frac{\Delta_N}{2} | \left(e^{-\beta_N \hat{H}} \right)^j | x_+ \rangle \langle x_+ | \hat{O}(t) | x_- \rangle \langle x_- | \left(e^{-\beta_N \hat{H}} \right)^{N-j} | q_N + \frac{\Delta_N}{2} \rangle \\
&= \frac{1}{N} \sum_{j=1}^{N} \frac{Z^{-1}}{2\pi\hbar} \int dq_N \int dp_N \int d\Delta_N \ e^{\frac{i}{\hbar} p_N \Delta_N}
\end{aligned}$$

$$
\begin{aligned}
&\times \langle q_N - \frac{\Delta_N}{2}| (e^{-\beta_N \hat{H}})^j \hat{O}(t) (e^{-\beta_N \hat{H}})^{N-j} |q_N + \frac{\Delta_N}{2}\rangle \\
&= \frac{Z^{-1}}{N} \sum_{j=1}^{N} \int dq_N \ \langle q_N| (e^{-\beta_N \hat{H}})^j \hat{O}(t) (e^{-\beta_N \hat{H}})^{N-j} |q_N\rangle \\
&= \frac{Z^{-1}}{N} \sum_{j=1}^{N} Tr \left[(e^{-\beta_N \hat{H}})^j \hat{O}(t) (e^{-\beta_N \hat{H}})^{N-j} \right] \\
&= \frac{Z^{-1}}{N} \sum_{j=1}^{N} Tr \left[e^{-\beta \hat{H}} \hat{O}(t) \right] \\
&= \left\langle \hat{O}(t) \right\rangle .
\end{aligned} \tag{19}
$$

We used the definition of $\left\langle \hat{O}(t) \right\rangle_N$ [Eq. (16)] at the first equality. At the second equality, we used the definition of $[\hat{O}(t)]_N(\mathbf{q}, \mathbf{p})$ [Eq. (1)] and the fact that $Z_N = Z$ [Eq. (13)]. At the third equality, we use the definition of $[e^{-\beta\hat{H}}]_{\overline{N}}$ [Eq. (3)], we expand the productorial (noting that $q_0 = q_N$ and $\Delta_0 = \Delta_N$) and we used the identity Eq. (11) to integrated out the variables p_k, q_k and Δ_k for $k \neq j$, $k \neq N$ [see derivation of Eq. (15)].[1] We used the definition of the one-dimensional Weyl-Wigner transform [Eq. (2)] to obtain the fourth equality. At the fifth equality, we performed the integral over p_j to obtain the delta function $\delta(\Delta_j + \Delta'_j)$ [Eq. (10)]. We performed the integral over Δ'_j at the sixth equality. At the seventh equality, we performed the change of variables $q_j \pm \frac{\Delta_j}{2} \to x_\pm$. We performed the integral over $x_\pm$ at the eighth equality. At the ninth equality, we performed the integral over p_N [giving a delta function $\delta(\Delta_N)$] followed by the integral over Δ_N. We recognized a trace at the tenth line. To obtain the eleventh equality we used the invariance of the trace to cyclic permutations to reorder the operators inside the trace. The final equality follows from the definition of an average in Hilbert space. This completes the proof of Eq. (17).

C. Explicit Expression for Generalized Boltzmann Transform

In the $N \to \infty$ limit, an explicit expression for the generalized Wigner Boltzmann transform can be obtained by noting that

$$
\begin{aligned}
\langle y| e^{-\beta_N \hat{H}} |x\rangle &= \langle y| e^{-\frac{\beta_N}{2}\hat{V}} e^{-\beta_N \hat{T}} e^{-\frac{\beta_N}{2}\hat{V}} |x\rangle \\
&= e^{-\frac{\beta_N}{2}(V(y)+V(x))} \langle y| e^{-\beta_N \hat{T}} |x\rangle
\end{aligned}
$$

[1] Note that for the case $j = N$, we are abusing the notation.

$$
\begin{aligned}
&= e^{-\frac{\beta_N}{2}(V(y)+V(x))} \int dp \, \langle y|p\rangle \, e^{-\beta_N \frac{p^2}{2m}} \, \langle p|x\rangle \\
&= e^{-\frac{\beta_N}{2}(V(y)+V(x))} \frac{1}{2\pi\hbar} \int dp \; e^{\frac{i}{\hbar}p(y-x)} e^{-\beta_N \frac{p^2}{2m}} \\
&= \left(\frac{m}{2\pi\beta_N\hbar^2}\right)^{1/2} e^{-\frac{\beta_N}{2}(V(y)+V(x))} e^{-\frac{m}{2\beta_N\hbar^2}(y-x)^2},
\end{aligned} \tag{20}
$$

where we used the Trotter splitting to symmetrically split the Boltzmann operator at the first equality and evaluated the corresponding matrix elements of the potential and kinetic operator on the position and momentum basis, respectively. Employing Eq. (20) to evaluate the $\langle q_{l-1} - \frac{\Delta_{l-1}}{2}| \, e^{-\beta_N \hat{H}} \, |q_l + \frac{\Delta_l}{2}\rangle$ matrix elements in the definition of the generalized Wigner Boltzmann transform [Eq. (3)], it follows that

$$
\begin{aligned}
\left[e^{-\beta\hat{H}}\right]_{\overline{N}} = & \left(\frac{m}{2\pi\beta_N\hbar^2}\right)^{N/2} \int d\boldsymbol{\Delta} \, \exp\left(\frac{i}{\hbar}\sum_{l=1}^{N} p_l\Delta_l\right) \\
& \times \exp\left(-\frac{\beta_N}{2}\sum_{l=1}^{N}\left[V(q_l - \frac{\Delta_l}{2}) + V(q_l + \frac{\Delta_l}{2})\right]\right) \\
& \times \exp\left(-\frac{m}{2\beta_N\hbar^2}\sum_{l=1}^{N}\left[q_l - q_{l-1} + \frac{\Delta_l + \Delta_{l-1}}{2}\right]^2\right).
\end{aligned} \tag{21}
$$

Note that upon integration over momenta $\mathbf{p}$, the generalized Boltzmann-Wigner transform reduces to

$$
\begin{aligned}
\int d\mathbf{p} \; \left[e^{-\beta\hat{H}}\right]_{\overline{N}} = & \left(\frac{m}{2\pi\beta_N\hbar^2}\right)^{N/2} \int d\mathbf{p} \int d\boldsymbol{\Delta} \, \exp\left(\frac{i}{\hbar}\sum_{l=1}^{N} p_l\Delta_l\right) \\
& \times \exp\left(-\frac{\beta_N}{2}\sum_{l=1}^{N}\left[V(q_l - \frac{\Delta_l}{2}) + V(q_l + \frac{\Delta_l}{2})\right]\right) \\
& \times \exp\left(-\frac{m}{2\beta_N\hbar^2}\sum_{l=1}^{N}\left[q_l - q_{l-1} + \frac{\Delta_l + \Delta_{l-1}}{2}\right]^2\right) \\
= & \left(\frac{2\pi m}{\beta_N}\right)^{N/2} \int d\boldsymbol{\Delta} \, \left(\prod_{l=1}^{N}\delta(\Delta_l)\right) \\
& \times \exp\left(-\frac{\beta_N}{2}\sum_{l=1}^{N}\left[V(q_l - \frac{\Delta_l}{2}) + V(q_l + \frac{\Delta_l}{2})\right]\right) \\
& \times \exp\left(-\frac{m}{2\beta_N\hbar^2}\sum_{l=1}^{N}\left[q_l - q_{l-1} + \frac{\Delta_l + \Delta_{l-1}}{2}\right]^2\right) \\
= & \left(\frac{2\pi m}{\beta_N}\right)^{N/2} \exp\left(-\beta_N\left[\sum_{l=1}^{N} V(q_l) + \frac{m}{2\beta_N^2\hbar^2}\sum_{l=1}^{N}(q_l - q_{l-1})^2\right]\right),
\end{aligned} \tag{22}
$$

where we performed the integration over momenta $\mathbf{p}$ to obtain the delta functions $\delta(\Delta_j)$ [Eq. (10)] at the second equality, and we performed the integrals over $\boldsymbol{\Delta}$ at the final equality.

D. Alternative Definition of the Generalized Wigner-Weyl Transforms

The generalized Wigner-Weyl transformed introduced by Eq. (1) can be written in different but equivalent notations. For example, in Ref. [6] (see also Refs. [7, 8]) we defined it as:

$$\left[\hat{O}(t)\right]_N(\mathbf{q},\mathbf{p}) = \int d\boldsymbol{\Delta}\int d\mathbf{z}\; O(\mathbf{z})\prod_{l=1}^{N} e^{\frac{i}{\hbar}p_l\Delta_l}\left\langle q_l-\frac{\Delta_l}{2}\right| e^{\frac{i}{\hbar}\hat{H}t}\left|z_l\right\rangle\left\langle z_l\right| e^{-\frac{i}{\hbar}\hat{H}t}\left|q_l+\frac{\Delta_l}{2}\right\rangle, \tag{23}$$

with

$$O(\mathbf{z}) = \frac{1}{N}\sum_{j=1}^{N} O(z_j). \tag{24}$$

Here we prove that Eq. (1) and (23) are equivalent for observables that are only functions of the position (namely, $\hat{O}=O(\hat{q})$).

The proof follows from the fact that $N-1$ of the forward-backward propagators in Eq. (23) are identities, as:

$$\begin{aligned}
\left[\hat{O}(t)\right]_N(\mathbf{q},\mathbf{p}) &= \int d\boldsymbol{\Delta}\int d\mathbf{z}\; O(\mathbf{z})\prod_{l=1}^{N} e^{\frac{i}{\hbar}p_l\Delta_l}\left\langle q_l-\frac{\Delta_l}{2}\right| e^{\frac{i}{\hbar}\hat{H}t}\left|z_l\right\rangle\left\langle z_l\right| e^{-\frac{i}{\hbar}\hat{H}t}\left|q_l+\frac{\Delta_l}{2}\right\rangle \\
&= \frac{1}{N}\sum_{j=1}^{N}\int d\boldsymbol{\Delta}\int d\mathbf{z}\; O(z_j)\prod_{l=1}^{N} e^{\frac{i}{\hbar}p_l\Delta_l}\left\langle q_l-\frac{\Delta_l}{2}\right| e^{\frac{i}{\hbar}\hat{H}t}\left|z_l\right\rangle\left\langle z_l\right| e^{-\frac{i}{\hbar}\hat{H}t}\left|q_l+\frac{\Delta_l}{2}\right\rangle \\
&= \frac{1}{N}\sum_{j=1}^{N}\int d\boldsymbol{\Delta}\int d\mathbf{z}\; e^{\frac{i}{\hbar}p_j\Delta_j}\left\langle q_j-\frac{\Delta_j}{2}\right| e^{\frac{i}{\hbar}\hat{H}t}\hat{O}\left|z_j\right\rangle\left\langle z_j\right| e^{-\frac{i}{\hbar}\hat{H}t}\left|q_j+\frac{\Delta_j}{2}\right\rangle \\
&\quad\times\prod_{\substack{l=1\\ l\neq j}}^{N} e^{\frac{i}{\hbar}p_l\Delta_l}\left\langle q_l-\frac{\Delta_l}{2}\right| e^{\frac{i}{\hbar}\hat{H}t}\left|z_l\right\rangle\left\langle z_l\right| e^{-\frac{i}{\hbar}\hat{H}t}\left|q_l+\frac{\Delta_l}{2}\right\rangle \\
&= \frac{1}{N}\sum_{j=1}^{N}\int d\boldsymbol{\Delta}\; e^{\frac{i}{\hbar}p_j\Delta_j}\left\langle q_j-\frac{\Delta_j}{2}\right| e^{\frac{i}{\hbar}\hat{H}t}\hat{O}e^{-\frac{i}{\hbar}\hat{H}t}\left|q_j+\frac{\Delta_j}{2}\right\rangle \\
&\quad\times\prod_{\substack{l=1\\ l\neq j}}^{N} e^{\frac{i}{\hbar}p_l\Delta_l}\left\langle q_l-\frac{\Delta_l}{2}\middle|q_l+\frac{\Delta_l}{2}\right\rangle \\
&= \frac{1}{N}\sum_{j=1}^{N}\int d\boldsymbol{\Delta}\; e^{\frac{i}{\hbar}p_j\Delta_j}\left\langle q_j-\frac{\Delta_j}{2}\right|\hat{O}(t)\left|q_j+\frac{\Delta_j}{2}\right\rangle\prod_{\substack{l=1\\ l\neq j}}^{N} e^{\frac{i}{\hbar}p_l\Delta_l}\delta(\Delta_l)
\end{aligned}$$

$$= \frac{1}{N}\sum_{j=1}^{N}\int d\Delta_j \; e^{\frac{i}{\hbar}p_j\Delta_j}\,\langle q_j - \frac{\Delta_j}{2}|\,\hat{O}(t)\,|q_j + \frac{\Delta_j}{2}\rangle$$

$$= \frac{1}{N}\sum_{j=1}^{N}\left[\hat{O}(t)\right]_{W,j}. \tag{25}$$

We used the alternative definition of the generalized Wigner transform [Eq. (23)] at the first equality and we used the definition of $O(\mathbf{z})$ [Eq. (24)] at the second equality. At the third equality, we used the fact that for position-dependent operators, $\hat{O}\,|z\rangle = O(z)\,|z\rangle$. At the fourth equality, we integrated out the $\mathbf{z}$ variables. We recognized the time-evolved operator $\hat{O}(t) = e^{\frac{i}{\hbar}\hat{H}t}\hat{O}e^{-\frac{i}{\hbar}\hat{H}t}$ at the fifth equality. We performed the integrals over Δ_k for $k \neq j$ to arrive at the sixth equality. The final equality follows from the definition of the Wigner-Weyl transform [Eq. (2)]. This completes the proof that the definitions of the generalized Wigner-Weyl transform provided by Eqs. (1) and (23) are equivalent (for position-dependent operators). We remark that the generalized Wigner transform definition adopted in this paper provides a more general definition that can be applied to arbitrary operators of position and momenta $\hat{O} = O(\hat{q}, \hat{p})$.

E. Ring-Polymer Quantum Liouvillian

Here we prove that the time evolution of the generalized Wigner transform is given by

$$e^{\mathcal{L}_N t}\left[\hat{O}\right]_N(\mathbf{q},\mathbf{p}) \;=\; \left[\hat{O}(t)\right]_N(\mathbf{q},\mathbf{p}), \tag{26}$$

where the ring-polymer quantum Liouvillian $\mathcal{L}_N$ is defined as

$$\mathcal{L}_N \;\equiv\; \frac{2N}{\hbar}\left[\hat{H}\right]_N \sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right), \tag{27}$$

and the ring-polymer Janus operator as

$$\overleftrightarrow{\Lambda}_N \equiv \sum_{j=1}^{N}\frac{\overleftarrow{\partial}}{\partial p_j}\frac{\overrightarrow{\partial}}{\partial q_j} - \frac{\overleftarrow{\partial}}{\partial q_j}\frac{\overrightarrow{\partial}}{\partial p_j} = \sum_{j=1}^{N}\overleftrightarrow{\Lambda}_j. \tag{28}$$

We begin by proving that the ring-polymer quantum Liouvillian $\mathcal{L}_N$ can be written in terms of single-bead terms as

$$\mathcal{L}_N = \sum_{j=1}^{N}\left[\frac{p_j}{m}\frac{\overrightarrow{\partial}}{\partial q_j} - \frac{2}{\hbar}V(q_j)\sin\left(\frac{\hbar}{2}\frac{\overleftarrow{\partial}}{\partial q_j}\frac{\overrightarrow{\partial}}{\partial p_j}\right)\right] = \sum_{j=1}^{N}\mathcal{L}_j. \tag{29}$$

To prove Eq. (29), we notice that since the ring-polymer Janus operator is a sum of one-dimensional operators involving derivatives [Eq. (28)], the action of $\overleftrightarrow{\Lambda}_N$ on a function that only depends on one coordinate reduces to the action of the one-dimensional Janus operator, namely

$$\begin{aligned}
\overleftrightarrow{\Lambda}_N f(q_k,p_k) &= \overleftrightarrow{\Lambda}_k f(q_k,p_k),\\
f(q_k,p_k)\overleftrightarrow{\Lambda}_N &= f(q_k,p_k)\overleftrightarrow{\Lambda}_k,\\
f(q_k,p_k)\overleftrightarrow{\Lambda}_N g(q_l,p_l) &= f(q_k,p_k)\overleftrightarrow{\Lambda}_k g(q_l,p_l)\delta_{kl}.
\end{aligned} \tag{30}$$

From these properties of the Janus operator, and the Sine Taylor expansion

$$\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) = \frac{\hbar}{2}\overleftrightarrow{\Lambda}_N + \cdots\,, \tag{31}$$

it follows that

$$\begin{aligned}
\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) f(q_k,p_k) &= \sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_k\right) f(q_k,p_k),\\
f(q_k,p_k)\,\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) &= f(q_k,p_k)\,\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_k\right),\\
f(q_k,p_k)\,\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) g(q_l,p_l) &= f(q_k,p_k)\,\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_k\right) g(q_l,p_l)\delta_{kl}.
\end{aligned} \tag{32}$$

Similarly, for the Cosine function

$$\cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) = 1 + \cdots\,, \tag{33}$$

the following relations hold:

$$\begin{aligned}
\cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) f(q_k,p_k) &= \cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_k\right) f(q_k,p_k),\\
f(q_k,p_k)\,\cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) &= f(q_k,p_k)\,\cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_k\right),\\
f(q_k,p_k)\,\cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) g(q_l,p_l) &= f(q_k,p_k)\,\cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_k\right) g(q_l,p_l)\delta_{kl}\\
&\quad + f(q_k,p_k)g(q_l,p_l)(1-\delta_{kl}).
\end{aligned} \tag{34}$$

The proof of Eq. (29) follows as

$$\begin{aligned}
\mathcal{L}_N &= \frac{2N}{\hbar}\left[\hat{H}\right]_N \sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right)\\
&= \frac{2N}{\hbar}\left[\frac{\hat{p}^2}{2m} + V(\hat{q})\right]_N \sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right)
\end{aligned}$$

$$
\begin{aligned}
&= \frac{2}{\hbar}\sum_{j=1}^{N}\left[\frac{p_j^2}{2m}+V(q_j)\right]\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right)\\
&= \frac{2}{\hbar}\sum_{j=1}^{N}\left[\frac{p_j^2}{2m}+V(q_j)\right]\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_j\right)\\
&= \sum_{j=1}^{N}\left[\frac{p_j}{m}\overrightarrow{\frac{\partial}{\partial q_j}}-\frac{2}{\hbar}V(q_j)\sin\left(\frac{\hbar}{2}\overleftarrow{\frac{\partial}{\partial q_j}}\overrightarrow{\frac{\partial}{\partial p_j}}\right)\right]\\
&= \sum_{j=1}^{N}\mathcal{L}_j. \qquad (35)
\end{aligned}
$$

We used the definition of the generalized quantum Liouvillian [Eq. (27)] at the first equality, and the definition of the Hamiltonian $\hat{H}=\hat{p}^2/2m+V(\hat{q})$ at the second equality. At the third equality, we used the definition of the generalized Wigner transform [Eq. (1)] and evaluated the one-dimensional Wigner transforms of the Hamiltonian. We used the property Eq. (32) to obtain the fourth equality. At the final equality, we used the Taylor expansion of the Sine function. This completes the proof of Eq. [29].

Using Eq. (35), it is straightforward to show that

$$
\begin{aligned}
e^{\mathcal{L}_N t}\left[\hat{O}\right]_N(\mathbf{q},\mathbf{p}) &= \frac{1}{N}\sum_{j=1}^{N}e^{\mathcal{L}_N t}\left[\hat{O}\right]_{W,j}\\
&= \frac{1}{N}\sum_{j=1}^{N}e^{\left(\sum_{k=1}^{N}\mathcal{L}_k\right)t}\left[\hat{O}\right]_{W,j}\\
&= \frac{1}{N}\sum_{j=1}^{N}e^{\mathcal{L}_j t}\left[\hat{O}\right]_{W,j}\\
&= \frac{1}{N}\sum_{j=1}^{N}\left[\hat{O}(t)\right]_{W,j}\\
&= [\hat{O}(t)]_N(\mathbf{q},\mathbf{p}). \qquad (36)
\end{aligned}
$$

We used the definition of the generalized Wigner transform [Eq. (1)] at the first equality. At the second equality, we used Eq. (35). We used the property Eq. (32) to obtain the third equality. The fourth equality follows from the (one-dimensional) Moyal series expansion of the quantum Liouvillian [Eq. (7)]. The final equality follows from the definition of the generalized Wigner transform [Eq. (1)]. This completes the proof of Eq. [26].

F. Ring-Polymer Sine and Cosine Couplings

1. Sine Coupling

Defining

$$\overleftrightarrow{s} \equiv \left(\frac{2N}{\hbar}\right) \sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) \tag{37}$$

as the Sine coupling (note the inclusion of the $2N/\hbar$ factor in the definition), it follows that

$$\begin{aligned} \left[\hat{O}_1\right]_N \overleftrightarrow{s} \left[\hat{O}_2\right]_N &= \left(\frac{2}{\hbar}\right) \frac{1}{N} \sum_{j,k=1}^{N} \left[\hat{O}_1\right]_{W,j} \sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) \left[\hat{O}_2\right]_{W,k} \\ &= \left(\frac{2}{\hbar}\right) \frac{1}{N} \sum_{j=1}^{N} \left[\hat{O}_1\right]_{W,j} \sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_j\right) \left[\hat{O}_2\right]_{W,j} \\ &= \left(\frac{i}{\hbar}\right) \frac{1}{N} \sum_{j=1}^{N} \left[[\hat{O}_1, \hat{O}_2]_-\right]_{W,j}. \end{aligned} \tag{38}$$

At the first equality, we used the definition of the generalized Wigner transform [Eq. (1)]. We used the property Eq. (32) to obtain the second equality. The last equality follows from the definition of the one-dimensional Moyal bracket [Eq. (5)]. Note that the Sine coupling between two generalized Wigner transforms is just the sum of one-dimensional Wigner-Weyl transform of the commutator $[\hat{O}_1, \hat{O}_2]_-$ evaluated at a particular bead coordinate.

2. Cosine Coupling

Defining

$$\overleftrightarrow{c} \equiv \cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) \tag{39}$$

as the Cosine coupling, it follows that

$$\begin{aligned} \left[\hat{O}_1\right]_N \overleftrightarrow{c} \left[\hat{O}_2\right]_N &= \frac{1}{N^2} \sum_{j,k=1}^{N} \left[\hat{O}_1\right]_{W,j} \cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) \left[\hat{O}_2\right]_{W,k} \\ &= \frac{1}{N^2} \left\{ \sum_{j=1}^{N} \left[\hat{O}_1\right]_{W,j} \cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_j\right) \left[\hat{O}_2\right]_{W,j} + \sum_{\substack{j,k=1 \\ j\neq k}}^{N} \left[\hat{O}_1\right]_{W,j} \left[\hat{O}_2\right]_{W,k} \right\} \end{aligned}$$

$$= \frac{1}{N^2}\left\{\sum_{j=1}^{N}\frac{1}{2}\left[[\hat{O}_1,\hat{O}_2]_+\right]_{W,j} + \sum_{\substack{j,k=1\\ j\neq k}}^{N}\left[\hat{O}_1\right]_{W,j}\left[\hat{O}_2\right]_{W,k}\right\}. \tag{40}$$

At the first equality, we used the definition of the generalized Wigner transform [Eq. (1)]. We used the property Eq. (34) to obtain the second equality. The last equality follows from the definition of the one-dimensional Baker bracket [Eq. (6)]. Note that the Cosine coupling between two generalized Wigner transforms, is composed of two terms: the first one is a sum of one-dimensional Wigner-Weyl transforms of the anti-commutator $[\hat{O}_1, \hat{O}_2]_+$ evaluated at a particular bead coordinate; the second term, on the other hand, consists of sums of products between $[\hat{O}_1]_W$ and $[\hat{O}_2]_W$ evaluated at *different* bead coordinates. We remark that this "bead decoupling" is reminiscent of a (discrete) Kubo integral.

3. Multiple Sine and Cosine Couplings

It is straightforward to generalize the formulas for the Sine and Cosine coupling to the case of multiples generalized Wigner transforms.

Specifically, the Sine-Sine coupling between generalized Wigner functions is given by

$$\begin{aligned}
\left\{\left[\hat{O}_1\right]_N \overleftrightarrow{s} \left[\hat{O}_2\right]_N\right\} \overleftrightarrow{s} \left[\hat{O}_3\right]_N &= \left(\frac{2i}{\hbar^2}\right)\frac{1}{N}\left\{\sum_{j,k=1}^{N}\left[[\hat{O}_1,\hat{O}_2]_-\right]_{W,j}\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right)\left[\hat{O}_3\right]_{W,k}\right\} \\
&= \left(\frac{2i}{\hbar^2}\right)\frac{1}{N}\left\{\sum_{j=1}^{N}\left[[\hat{O}_1,\hat{O}_2]_-\right]_{W,j}\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_j\right)\left[\hat{O}_3\right]_{W,k}\right\} \\
&= \left(\frac{i}{\hbar}\right)^2\frac{1}{N}\left\{\sum_{j=1}^{N}\left[[[\hat{O}_1,\hat{O}_2]_-,\hat{O}_3]_-\right]_{W,j}\right\}.
\end{aligned} \tag{41}$$

We used Eq. (38) and the definition of the generalized Wigner transform [Eq. (1)] at the first equality. We used the property Eq. (32) to obtain the second equality. The last equality follows from the definition of the one-dimensional Moyal bracket [Eq. (5)]. Note that the Sine-Sine coupling involves a sum of one-dimensional Wigner-Weyl transforms of the double commutator operator $[[\hat{O}_1, \hat{O}_2]_-, \hat{O}_3]_-$ evaluated at a particular bead coordinate.

Similarly, the Sine-Cosine coupling between generalized Wigner functions is given by

$$\left\{\left[\hat{O}_1\right]_N \overleftrightarrow{s} \left[\hat{O}_2\right]_N\right\} \overleftrightarrow{c} \left[\hat{O}_3\right]_N = \left(\frac{i}{\hbar}\right)\frac{1}{N^2}\left\{\sum_{j,k=1}^{N}\left[[\hat{O}_1,\hat{O}_2]_-\right]_{W,j}\cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right)\left[\hat{O}_3\right]_{W,k}\right\}$$

$$
\begin{aligned}
&= \left(\frac{i}{\hbar}\right)\frac{1}{N^2}\left\{\sum_{j=1}^{N}\left[[\hat{O}_1,\hat{O}_2]_-\right]_{W,j}\cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_j\right)\left[\hat{O}_3\right]_{W,j}\right.\\
&\qquad\left.+\sum_{\substack{j,k=1\\ j\neq k}}^{N}\left[[\hat{O}_1,\hat{O}_2]_-\right]_{W,j}\left[\hat{O}_3\right]_{W,k}\right\}\\
&= \left(\frac{i}{\hbar}\right)\frac{1}{N^2}\left\{\sum_{j=1}^{N}\frac{1}{2}\left[[[\hat{O}_1,\hat{O}_2]_-,\hat{O}_3]_+\right]_{W,j}\right.\\
&\qquad\left.+\sum_{\substack{j,k=1\\ j\neq k}}^{N}\left[[\hat{O}_1,\hat{O}_2]_-\right]_{W,j}\left[\hat{O}_3\right]_{W,k}\right\}.
\end{aligned}\tag{42}
$$

We used Eq. (38) and the definition of the generalized Wigner transform [Eq. (1)] at the first equality. We used the property Eq. (34) to obtain the second equality. At the last equality, we used the definition of the Baker bracket [Eq. (6)] for the first term. Note that the Sine-Cosine coupling is composed of two terms: the first one is a sum of one-dimensional Wigner-Weyl transforms evaluated at a particular bead coordinate of the (symmetrized) operator $[\hat{O}_1,\hat{O}_2]_-\hat{O}_3$; the second term, on the other hand, consists of sums of products between the commutator $[[\hat{O}_1,\hat{O}_2]_-]_W$ and $[\hat{O}_3]_W$ evaluated at different bead coordinates. We remark that the "bead decoupling" and structure of the Sine-Cosine coupling is reminiscent of a (discrete) Kubo integral between $[\hat{O}_1,\hat{O}_2]_-$ and $[\hat{O}_3]$.

We remark that one could define an alternative ordering of couplings between generalized Wigner functions giving rise to the Cosine-Sine expression (compare to Eq. (42))

$$
\begin{aligned}
\left\{\left[\hat{O}_1\right]_N\overleftrightarrow{c}\left[\hat{O}_2\right]_N\right\}\overleftrightarrow{s}\left[\hat{O}_3\right]_N &= \left(\frac{2}{\hbar}\right)\frac{1}{N^2}\left\{\sum_{j,l=1}^{N}\frac{1}{2}\left[[\hat{O}_1,\hat{O}_2]_+\right]_{W,j}\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right)\left[\hat{O}_3\right]_{W,l}\right.\\
&\qquad\left.+\sum_{\substack{j,k,l=1\\ j\neq k}}^{N}\left[\hat{O}_1\right]_{W,j}\left[\hat{O}_2\right]_{W,k}\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right)\left[\hat{O}_3\right]_{W,l}\right\}\\
&= \left(\frac{2}{\hbar}\right)\frac{1}{N^2}\left\{\sum_{j=1}^{N}\frac{1}{2}\left[[\hat{O}_1,\hat{O}_2]_+\right]_{W,j}\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_j\right)\left[\hat{O}_3\right]_{W,j}\right.\\
&\qquad+\sum_{\substack{j,k=1\\ j\neq k}}^{N}\left[\hat{O}_1\right]_{W,j}\left[\hat{O}_2\right]_{W,k}\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_k\right)\left[\hat{O}_3\right]_{W,k}
\end{aligned}
$$

$$
+ \sum_{\substack{j,k=1 \\ j \neq k}}^{N} \left[\hat{O}_1\right]_{W,j} \left[\hat{O}_2\right]_{W,k} \sin\left(\frac{\hbar}{2} \overleftrightarrow{\Lambda}_j\right) \left[\hat{O}_3\right]_{W,j} \Bigg\}
$$

$$
= \left(\frac{i}{\hbar}\right) \frac{1}{N^2} \Bigg\{ \sum_{j=1}^{N} \frac{1}{2} \left[[[\hat{O}_1, \hat{O}_2]_+, \hat{O}_3]_-\right]_{W,j}
+ \sum_{\substack{j,k=1 \\ j \neq k}}^{N} \left[\hat{O}_1\right]_{W,j} \left[[\hat{O}_2, \hat{O}_3]_-\right]_{W,k}
+ \sum_{\substack{j,k=1 \\ j \neq k}}^{N} \left[\hat{O}_2\right]_{W,k} \left[[\hat{O}_1, \hat{O}_3]_-\right]_{W,j} \Bigg\}. \tag{43}
$$

We used Eq. (40) and the definition of the generalized Wigner transform [Eq. (1)] at the first equality. We used the property Eq. (32) to obtain the second equality. At the final equality, we used the definition of the Moyal bracket [Eq. (5)] to evaluate the sine couplings. Note, however, that since $[[\hat{O}_1, \hat{O}_2]_+, \hat{O}_3]_- = [[\hat{O}_1, \hat{O}_3]_-, \hat{O}_2]_+ + [[\hat{O}_2, \hat{O}_3]_-, \hat{O}_1]_+$ the Cosine-Sine coupling can be expressed in terms of Sine-Cosine couplings instead (see Eq. (55))

Finally, the Cosine-Cosine coupling between generalized Wigner functions, is given by

$$
\left\{ \left[\hat{O}_1\right]_N \overleftrightarrow{c} \left[\hat{O}_2\right]_N \right\} \overleftrightarrow{c} \left[\hat{O}_3\right]_N = \frac{1}{N^3} \Bigg\{ \sum_{j,l=1}^{N} \frac{1}{2} \left[[\hat{O}_1, \hat{O}_2]_+\right]_{W,j} \cos\left(\frac{\hbar}{2} \overleftrightarrow{\Lambda}_N\right) \left[\hat{O}_3\right]_{W,l}
+ \sum_{\substack{j,k,l=1 \\ j \neq k}}^{N} \left[\hat{O}_1\right]_{W,j} \left[\hat{O}_2\right]_{W,k} \cos\left(\frac{\hbar}{2} \overleftrightarrow{\Lambda}_N\right) \left[\hat{O}_3\right]_{W,l} \Bigg\}
$$

$$
= \frac{1}{N^3} \Bigg\{ \sum_{j=1}^{N} \frac{1}{2} \left[[\hat{O}_1, \hat{O}_2]_+\right]_{W,j} \cos\left(\frac{\hbar}{2} \overleftrightarrow{\Lambda}_j\right) \left[\hat{O}_3\right]_{W,j}
+ \sum_{\substack{j,l=1 \\ j \neq l}}^{N} \frac{1}{2} \left[[\hat{O}_1, \hat{O}_2]_+\right]_{W,j} \left[\hat{O}_3\right]_{W,l}
+ \sum_{\substack{j,k=1 \\ j \neq k}}^{N} \left[\hat{O}_1\right]_{W,j} \left[\hat{O}_2\right]_{W,k} \cos\left(\frac{\hbar}{2} \overleftrightarrow{\Lambda}_k\right) \left[\hat{O}_3\right]_{W,k}
+ \sum_{\substack{j,k=1 \\ j \neq k}}^{N} \left[\hat{O}_1\right]_{W,j} \left[\hat{O}_2\right]_{W,k} \cos\left(\frac{\hbar}{2} \overleftrightarrow{\Lambda}_j\right) \left[\hat{O}_3\right]_{W,j}
$$

$$
+ \sum_{\substack{j,k,l=1 \\ j\neq k, j\neq l, k\neq l}}^{N} \left[\hat{O}_1\right]_{W,j} \left[\hat{O}_2\right]_{W,k} \left[\hat{O}_3\right]_{W,l} \Bigg\}
$$

$$
= \frac{1}{N^3} \Bigg\{ \sum_{j=1}^{N} \frac{1}{4} \left[[[\hat{O}_1, \hat{O}_2]_+, \hat{O}_3]_+\right]_{W,j}
+ \sum_{\substack{j,l=1 \\ j\neq l}}^{N} \frac{1}{2} \left[[\hat{O}_1, \hat{O}_2]_+\right]_{W,j} \left[\hat{O}_3\right]_{W,l}
+ \sum_{\substack{j,k=1 \\ j\neq k}}^{N} \frac{1}{2} \left[\hat{O}_1\right]_{W,j} \left[[\hat{O}_2, \hat{O}_3]_+\right]_{W,k}
+ \sum_{\substack{j,k=1 \\ j\neq k}}^{N} \frac{1}{2} \left[\hat{O}_2\right]_{W,k} \left[[\hat{O}_1, \hat{O}_3]_+\right]_{W,j}
+ \sum_{\substack{j,k,l=1 \\ j\neq k, j\neq l, k\neq l}}^{N} \left[\hat{O}_1\right]_{W,j} \left[\hat{O}_2\right]_{W,k} \left[\hat{O}_3\right]_{W,l} \Bigg\}. \tag{44}
$$

We used Eq. (40) and the definition of the generalized Wigner transform [Eq. (1)] at the first equality. We used the property Eq. (34) to obtain the second equality. At the final equality, we used the definition of the Baker bracket [Eq. (6)] to evaluate the cosine couplings. Note that the Cosine-Cosine coupling presents a more complex structure in which, depending on the sum term, different operators are evaluated at particular (equal or different) bead coordinates. We remark that the "bead decoupling" and structure of the Cosine-Cosine coupling is reminiscent of a symmetrized (discrete) Double Kubo integral [6, 9, 10] between $\hat{O}_1$, $\hat{O}_2$ and $\hat{O}_3$.

G. Properties of Sine/Cosine Couplings

1. Adjoint property of Janus operator

Noting that the Janus operator involves partial derivatives with respect to $\mathbf{q}$ and $\mathbf{p}$, it follows that for arbitrary functions $a(\mathbf{q}, \mathbf{p})$ and $b(\mathbf{q}, \mathbf{p})$ of ring-polymer variables the following equality holds for arbitrary integer n:

$$
a(\mathbf{q}, \mathbf{p}) \overleftrightarrow{\Lambda}_N^n b(\mathbf{q}, \mathbf{p}) = (-1)^n \; b(\mathbf{q}, \mathbf{p}) \overleftrightarrow{\Lambda}_N^n a(\mathbf{q}, \mathbf{p}) \tag{45}
$$

2. *Anti-self-adjoint property of Sine*

The anti-self-adjoint property of the Sine coupling

$$a(\mathbf{q},\mathbf{p})\overleftrightarrow{s}b(\mathbf{q},\mathbf{p}) \;=\; -b(\mathbf{q},\mathbf{p})\overleftrightarrow{s}a(\mathbf{q},\mathbf{p}) \tag{46}$$

with $a(\mathbf{q},\mathbf{p})$ and $b(\mathbf{q},\mathbf{p})$ arbitrary functions of ring-polymer variables, follows from

$$\begin{aligned} a\overleftrightarrow{s}b \;&=\; \left(\frac{2N}{\hbar}\right) a\,\sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) b \\ &=\; \left(\frac{2N}{\hbar}\right)\sum_{n=0}^{\infty}\frac{(-1)^n}{(2n+1)!}\left(\frac{\hbar}{2}\right)^{2n+1}\left[a\;\overleftrightarrow{\Lambda}_N^{2n+1}b\right] \\ &=\; \left(\frac{2N}{\hbar}\right)\sum_{n=0}^{\infty}\frac{(-1)^n}{(2n+1)!}\left(\frac{\hbar}{2}\right)^{2n+1}\left[(-1)b\;\overleftrightarrow{\Lambda}_N^{2n+1}a\right] \\ &=\; (-1)b\overleftrightarrow{s}a \end{aligned} \tag{47}$$

where we have used Eq. (45) for even powers at the third equality.

3. *Self-adjoint property of Cosine*

The self-adjoint property of the Cosine coupling

$$a(\mathbf{q},\mathbf{p})\overleftrightarrow{c}b(\mathbf{q},\mathbf{p}) \;=\; b(\mathbf{q},\mathbf{p})\overleftrightarrow{c}a(\mathbf{q},\mathbf{p}) \tag{48}$$

with $a(\mathbf{q},\mathbf{p})$ and $b(\mathbf{q},\mathbf{p})$ arbitrary functions of ring-polymer variables, follows from

$$\begin{aligned} a\overleftrightarrow{c}b \;&=\; a\,\cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) b \\ &=\; \sum_{n=0}^{\infty}\frac{(-1)^n}{(2n)!}\left(\frac{\hbar}{2}\right)^{2n}\left[a\;\overleftrightarrow{\Lambda}_N^{2n}b\right] \\ &=\; \left(\frac{2N}{\hbar}\right)\sum_{n=0}^{\infty}\frac{(-1)^n}{(2n)!}\left(\frac{\hbar}{2}\right)^{2n}\left[b\;\overleftrightarrow{\Lambda}_N^{2n}a\right] \\ &=\; b\overleftrightarrow{c}a \end{aligned} \tag{49}$$

where we have used Eq. (45) for odd powers at the third equality.

4. *Properties under integral sign*

The properties of Sine and Cosine coupling under the integral sign

$$\int d\mathbf{q}\int d\mathbf{p}\; a(\mathbf{q},\mathbf{p})\left\{b(\mathbf{q},\mathbf{p})\overleftrightarrow{s}c(\mathbf{q},\mathbf{p})\right\} \;=\; \int d\mathbf{q}\int d\mathbf{p}\;\left\{a(\mathbf{q},\mathbf{p})\overleftrightarrow{s}b(\mathbf{q},\mathbf{p})\right\}c(\mathbf{q},\mathbf{p}) \tag{50a}$$

$$\int d\mathbf{q} \int d\mathbf{p}\ a(\mathbf{q},\mathbf{p}) \left\{ b(\mathbf{q},\mathbf{p}) \overleftrightarrow{c}\, c(\mathbf{q},\mathbf{p}) \right\} = \int d\mathbf{q} \int d\mathbf{p}\ \left\{ a(\mathbf{q},\mathbf{p}) \overleftrightarrow{c}\, b(\mathbf{q},\mathbf{p}) \right\} c(\mathbf{q},\mathbf{p}) \quad (50b)$$

with $a(\mathbf{q},\mathbf{p})$, $b(\mathbf{q},\mathbf{p})$ and $c(\mathbf{q},\mathbf{p})$ arbitrary functions of ring-polymer variables, follows from the fact that for each term in the Taylor expansion of the Sine and Cosine the following equality holds

$$\int d\mathbf{q} \int d\mathbf{p}\ a(\mathbf{q},\mathbf{p}) \left\{ b(\mathbf{q},\mathbf{p}) \overleftrightarrow{\Lambda}^{n}_{N} c(\mathbf{q},\mathbf{p}) \right\} = \int d\mathbf{q} \int d\mathbf{p}\ \left\{ a(\mathbf{q},\mathbf{p}) \overleftrightarrow{\Lambda}^{n}_{N} b(\mathbf{q},\mathbf{p}) \right\} c(\mathbf{q},\mathbf{p}) \tag{51}$$

for any integer n.

We will prove Eq. (51) by induction. For $n = 1$ (base case), we have that

$$\begin{aligned}
\int d\mathbf{q} \int d\mathbf{p}\ a \left\{ b \overleftrightarrow{\Lambda}_{N} c \right\} &= \int d\mathbf{q} \int d\mathbf{p}\ a \left\{ \sum_k \frac{\partial b}{\partial p_k}\frac{\partial c}{\partial q_k} - \frac{\partial b}{\partial q_k}\frac{\partial c}{\partial p_k} \right\} \\
&= -\int d\mathbf{q} \int d\mathbf{p}\ \left\{ \sum_k \frac{\partial b}{\partial p_k}\frac{\partial a}{\partial q_k} - \frac{\partial b}{\partial q_k}\frac{\partial a}{\partial p_k} \right\} c \\
&= -\int d\mathbf{q} \int d\mathbf{p}\ \left\{ b \overleftrightarrow{\Lambda}_{N} a \right\} c \\
&= \int d\mathbf{q} \int d\mathbf{p}\ \left\{ a \overleftrightarrow{\Lambda}_{N} b \right\} c
\end{aligned} \tag{52}$$

where in the second line we have used integration by parts, assumed that the surface term vanishes and recognized that terms containing mixed derivatives $\frac{\partial^2 b}{\partial p_k \partial q_k}$ cancel out, and in the last line, we used Eq. (45).

Now we take the inductive step and prove that if Eq. (51) holds for n, then it also holds for $n + 1$:

$$\begin{aligned}
\int d\mathbf{q} \int d\mathbf{p}\ a \left\{ b \overleftrightarrow{\Lambda}^{n+1}_{N} c \right\} &= \int d\mathbf{q} \int d\mathbf{p}\ a \left\{ b \overleftrightarrow{\Lambda}^{n}_{N} \overleftrightarrow{\Lambda}_{N} c \right\} \\
&= \sum_k \int d\mathbf{q} \int d\mathbf{p}\ a \left\{ \frac{\partial b}{\partial p_k} \overleftrightarrow{\Lambda}^{n}_{N} \frac{\partial c}{\partial q_k} - \frac{\partial b}{\partial q_k} \overleftrightarrow{\Lambda}^{n}_{N} \frac{\partial c}{\partial p_k} \right\} \\
&= -\sum_k \int d\mathbf{q} \int d\mathbf{p}\ \frac{\partial a}{\partial q_k} \left\{ \frac{\partial b}{\partial p_k} \overleftrightarrow{\Lambda}^{n}_{N} c \right\} - \frac{\partial a}{\partial p_k} \left\{ \frac{\partial b}{\partial q_k} \overleftrightarrow{\Lambda}^{n}_{N} c \right\} \\
&= -\sum_k \int d\mathbf{q} \int d\mathbf{p}\ \left\{ \frac{\partial a}{\partial q_k} \overleftrightarrow{\Lambda}^{n}_{N} \frac{\partial b}{\partial p_k} \right\} c - \left\{ \frac{\partial a}{\partial p_k} \overleftrightarrow{\Lambda}^{n}_{N} \frac{\partial b}{\partial q_k} \right\} c \\
&= \int d\mathbf{q} \int d\mathbf{p}\ \left\{ a \overleftrightarrow{\Lambda}^{n}_{N} \overleftrightarrow{\Lambda}_{N} b \right\} c \\
&= \int d\mathbf{q} \int d\mathbf{p}\ \left\{ a \overleftrightarrow{\Lambda}^{n+1}_{N} b \right\} c
\end{aligned} \tag{53}$$

where in the third line we have used integration by parts, assumed that the surface term vanishes and recognized that terms containing mixed derivatives $[\frac{\partial^2 b}{\partial p_k \partial q_k} \overleftrightarrow{\Lambda}_N^{(n)} c]$ cancel out, and in the fourth line used Eq. (51) that we assume holds for n and arbitrary functions of ring-polymer variables (such as $\frac{\partial a}{\partial p_k}$ or $\frac{\partial b}{\partial q_k}$).

5. *Association rules*

The Sine and Cosine coupling operators satisfy the association rules

$$\left([\hat{O}_1]_N \overleftrightarrow{s} [\hat{O}_2]_N\right) \overleftrightarrow{s} [\hat{O}_3]_N = [\hat{O}_1]_N \overleftrightarrow{s} \left([\hat{O}_2]_N \overleftrightarrow{s} [\hat{O}_3]_N\right) + \left([\hat{O}_1]_N \overleftrightarrow{s} [\hat{O}_3]_N\right) \overleftrightarrow{s} [\hat{O}_2]_N \tag{54}$$

$$\left([\hat{O}_1]_N \overleftrightarrow{c} [\hat{O}_2]_N\right) \overleftrightarrow{s} [\hat{O}_3]_N = [\hat{O}_1]_N \overleftrightarrow{c} \left([\hat{O}_2]_N \overleftrightarrow{s} [\hat{O}_3]_N\right) + \left([\hat{O}_1]_N \overleftrightarrow{s} [\hat{O}_3]_N\right) \overleftrightarrow{c} [\hat{O}_2]_N \tag{55}$$

$$\left([\hat{O}_1]_N \overleftrightarrow{c} [\hat{O}_2]_N\right) \overleftrightarrow{c} [\hat{O}_3]_N = [\hat{O}_1]_N \overleftrightarrow{c} \left([\hat{O}_2]_N \overleftrightarrow{c} [\hat{O}_3]_N\right) \tag{56}$$

with follows from Eqs. (41)-(44). Note that the first relation resembles the Jacobi identity

$$[[\hat{O}_1, \hat{O}_2]_-, \hat{O}_3]_- = [\hat{O}_1, [\hat{O}_2, \hat{O}_3]_-]_- + [[\hat{O}_1, \hat{O}_3]_-, \hat{O}_2]_- \tag{57}$$

in ring-polymer phase-space.

II. KUBO IDENTITY IN RING-POLYMER PHASE-SPACE

In this section, we will prove that in the $N \to \infty$ limit the following identity holds between Sine and Cosine coupling operators

$$\left[e^{-\beta \hat{H}}\right]_{\overline{N}} \overleftrightarrow{s} \left[\hat{O}(t)\right]_N = \beta \left[e^{-\beta \hat{H}}\right]_{\overline{N}} \overleftrightarrow{c} \frac{d}{dt} \left[\hat{O}(t)\right]_N . \tag{58}$$

We remark that Eq. (58) resembles the Kubo identity in ring-polymer phase space.

We start by recognizing that

$$\begin{aligned}
\left[e^{-\beta \hat{H}}\right]_{\overline{N}} \overleftrightarrow{s} \left[\hat{O}(t)\right]_N &= \left(\frac{2N}{\hbar}\right) \left[e^{-\beta \hat{H}}\right]_{\overline{N}} \sin\left(\frac{\hbar}{2} \overleftrightarrow{\Lambda}_N\right) \left[\hat{O}(t)\right]_N \\
&= \frac{2}{\hbar} \sum_{j=1}^{N} \left[e^{-\beta \hat{H}}\right]_{\overline{N}} \sin\left(\frac{\hbar}{2} \overleftrightarrow{\Lambda}_N\right) \left[\hat{O}(t)\right]_{W,j} \\
&= \frac{2}{\hbar} \sum_{j=1}^{N} \left[e^{-\beta \hat{H}}\right]_{\overline{N}} \sin\left(\frac{\hbar}{2} \overleftrightarrow{\Lambda}_j\right) \left[\hat{O}(t)\right]_{W,j}
\end{aligned}$$

$$= \frac{1}{i\hbar}\sum_{j=1}^{N}\left[e^{-\beta\hat{H}}\right]_{\overline{N}}\left(e^{\frac{i\hbar}{2}\overleftrightarrow{\Lambda}_j} - e^{-\frac{i\hbar}{2}\overleftrightarrow{\Lambda}_j}\right)\left[\hat{O}(t)\right]_{W,j}, \tag{59}$$

and that

$$\begin{aligned}
\left[e^{-\beta\hat{H}}\right]_{\overline{N}}\overleftrightarrow{c}\frac{d}{dt}\left[\hat{O}(t)\right]_N &= \left[e^{-\beta\hat{H}}\right]_{\overline{N}}\cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right)\left[\dot{\hat{O}}(t)\right]_N \\
&= \frac{1}{N}\sum_{j=1}^{N}\left[e^{-\beta\hat{H}}\right]_{\overline{N}}\cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right)\left[\dot{\hat{O}}(t)\right]_{W,j} \\
&= \frac{1}{N}\sum_{j=1}^{N}\left[e^{-\beta\hat{H}}\right]_{\overline{N}}\cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_j\right)\left[\dot{\hat{O}}(t)\right]_{W,j} \\
&= \frac{1}{2N}\sum_{j=1}^{N}\left[e^{-\beta\hat{H}}\right]_{\overline{N}}\left(e^{\frac{i\hbar}{2}\overleftrightarrow{\Lambda}_j} + e^{-\frac{i\hbar}{2}\overleftrightarrow{\Lambda}_j}\right)\left[\dot{\hat{O}}(t)\right]_{W,j}.
\end{aligned} \tag{60}$$

We used the definition of the Sine and Cosine coupling operators at the first equality. At the second equality, we used the definition of the generalized Wigner transform [Eq. (1)]. To obtain the third equality, we used the properties Eqs. (32) and (34) to express the Sine/Cosine in terms of the one-dimension Janus operator. Note that in Eq. (60) we used the shorthand notation $\dot{\hat{O}}(t) = \frac{d}{dt}\hat{O}(t)$.

The evaluation of the terms in Eqs. (59) and (60) can be performed by realizing that the generalized Boltzmann transform can be expressed in terms of one-dimensional Wigner transforms as

$$\begin{aligned}
&\left[e^{-\beta\hat{H}}\right]_{\overline{N}} \\
&= \int d\boldsymbol{\Delta}\prod_{l=1}^{N} e^{\frac{i}{\hbar}p_l\Delta_l}\langle q_{l-1} - \frac{\Delta_{l-1}}{2}|\, e^{-\beta_N\hat{H}}\,|q_l + \frac{\Delta_l}{2}\rangle \\
&= \int d\Delta_j\; e^{\frac{i}{\hbar}p_j\Delta_j}\langle q_j - \frac{\Delta_j}{2}|\left\{\int d\boldsymbol{\Delta}'\prod_{l=j+1}^{j+N-1} e^{\frac{i}{\hbar}p_l\Delta_l}\; e^{-\beta_N\hat{H}}\,|q_l + \frac{\Delta_l}{2}\rangle\langle q_l - \frac{\Delta_l}{2}|\right\} e^{-\beta_N\hat{H}}\,|q_j + \frac{\Delta_j}{2}\rangle \\
&= \left[\left\{\int d\boldsymbol{\Delta}'\prod_{l=j+1}^{j+N-1} e^{\frac{i}{\hbar}p_l\Delta_l}\; e^{-\beta_N\hat{H}}\,|q_l + \frac{\Delta_l}{2}\rangle\langle q_l - \frac{\Delta_l}{2}|\right\} e^{-\beta_N\hat{H}}\right]_{W,j},
\end{aligned} \tag{61}$$

where we have used the definition of the generalized Boltzmann transform in the first line, reorder and regrouped terms in the second line (by singling out the j-th contribution to the integral), and used the definition of the one-dimensional Wigner function in the third line. Therefore, it follows that

$$\left[e^{-\beta\hat{H}}\right]_{\overline{N}} e^{\frac{i\hbar}{2}\overleftrightarrow{\Lambda}_j}\left[\hat{O}\right]_{W,j} = \left[\left\{\int d\boldsymbol{\Delta}'\prod_{l=j+1}^{j+N-1} e^{\frac{i}{\hbar}p_l\Delta_l}\; e^{-\beta_N\hat{H}}\,|q_l + \frac{\Delta_l}{2}\rangle\langle q_l - \frac{\Delta_l}{2}|\right\} e^{-\beta_N\hat{H}}\hat{O}\right]_{W,j}$$

$$
\begin{aligned}
&= \int d\boldsymbol{\Delta} e^{\frac{i}{\hbar}p_j\Delta_j} \;\; \langle q_{j-1} - \frac{\Delta_{j-1}}{2}|\, e^{-\beta_N\hat{H}}\hat{O}\,|q_j + \frac{\Delta_j}{2}\rangle \\
&\quad \times \prod_{\substack{l=1\\ l\neq j}}^{N} e^{\frac{i}{\hbar}p_l\Delta_l} \;\; \langle q_{l-1} - \frac{\Delta_{l-1}}{2}|\, e^{-\beta_N\hat{H}}\,|q_l + \frac{\Delta_l}{2}\rangle \\
&= \int d\boldsymbol{\Delta} \; \prod_{l=1}^{N} e^{\frac{i}{\hbar}p_l\Delta_l}\, \langle q_{l-1} - \frac{\Delta_{l-1}}{2}|\, e^{-\beta_N\hat{H}}\hat{O}_j\,|q_l + \frac{\Delta_l}{2}\rangle \,, \qquad (62)
\end{aligned}
$$

and

$$
\begin{aligned}
\left[e^{-\beta\hat{H}}\right]_{\overline{N}} e^{-\frac{i\hbar}{2}\overleftrightarrow{\Lambda}_j} \left[\hat{O}\right]_{W,j} &= \left[\hat{O}\left\{\int d\boldsymbol{\Delta}' \prod_{l=j+1}^{j+N-1} e^{\frac{i}{\hbar}p_l\Delta_l}\; e^{-\beta_N\hat{H}}\,|q_l + \frac{\Delta_l}{2}\rangle\,\langle q_l - \frac{\Delta_l}{2}|\right\} e^{-\beta_N\hat{H}}\right]_{W,j} \\
&= \int d\boldsymbol{\Delta} e^{\frac{i}{\hbar}p_j\Delta_j} \;\; \langle q_j - \frac{\Delta_j}{2}|\, \hat{O}e^{-\beta_N\hat{H}}\,|q_{j+1} + \frac{\Delta_{j+1}}{2}\rangle \\
&\quad \times \prod_{\substack{l=1\\ l\neq j}}^{N} e^{\frac{i}{\hbar}p_l\Delta_l} \;\; \langle q_l - \frac{\Delta_l}{2}|\, e^{-\beta_N\hat{H}}\,|q_{l+1} + \frac{\Delta_{l+1}}{2}\rangle \\
&= \int d\boldsymbol{\Delta} \; \prod_{l=1}^{N} e^{\frac{i}{\hbar}p_l\Delta_l}\, \langle q_l - \frac{\Delta_l}{2}|\, \hat{O}_j e^{-\beta_N\hat{H}}\,|q_{l+1} + \frac{\Delta_{l+1}}{2}\rangle \\
&= \int d\boldsymbol{\Delta} \; \prod_{l=1}^{N} e^{\frac{i}{\hbar}p_l\Delta_l}\, \langle q_{l-1} - \frac{\Delta_{l-1}}{2}|\, \hat{O}_{j-1} e^{-\beta_N\hat{H}}\,|q_l + \frac{\Delta_l}{2}\rangle \,, \qquad (63)
\end{aligned}
$$

where we have used the one-dimensional Moyal product in the first lines, and used the definition of the one-dimensional Wigner transform to reorder and regrouped terms to obtain the final expressions. To simplify notation, in the last lines of Eqs. (62) and (63) we have introduced the operator $\hat{O}_j$ that only acts on the j-th path integral bead.

From Eqs. (62) and (63) it, therefore, follows that

$$
\begin{aligned}
&\sum_{j=1}^{N} \left[e^{-\beta\hat{H}}\right]_{\overline{N}} \left(e^{\frac{i\hbar}{2}\overleftrightarrow{\Lambda}_j} \pm e^{-\frac{i\hbar}{2}\overleftrightarrow{\Lambda}_j}\right) \left[\hat{O}\right]_{W,j} \\
&= \sum_{j=1}^{N} \int d\boldsymbol{\Delta} \; \prod_{l=1}^{N} e^{\frac{i}{\hbar}p_l\Delta_l}\, \langle q_{l-1} - \frac{\Delta_{l-1}}{2}|\, [e^{-\beta_N\hat{H}}, \hat{O}_j]_{\pm}\,|q_l + \frac{\Delta_l}{2}\rangle \,. \qquad (64)
\end{aligned}
$$

Using the Kubo identity[11]

$$
\begin{aligned}
[e^{-\lambda\hat{H}}, \hat{O}] &= i\hbar\; e^{-\lambda\hat{H}} \int_0^{\lambda} d\lambda' \dot{\hat{O}}(-i\hbar\lambda') \\
&= i\hbar \int_0^{\lambda} d\lambda' \dot{\hat{O}}(+i\hbar\lambda')\; e^{-\lambda\hat{H}}, \qquad (65)
\end{aligned}
$$

it is possible to relate the commutators and anti-commutators $[e^{-\beta_N \hat{H}}, \hat{O}_j]_\pm$ in the $N \to \infty$ limit as

$$[e^{-\beta_N \hat{H}}, \hat{O}_j]_- = i\hbar \frac{\beta_N}{2} \left[\frac{1}{\beta_N} \int_0^{\beta_N} d\lambda \; e^{-\beta_N \hat{H}} \dot{\hat{O}}_j(-i\hbar\lambda) + \dot{\hat{O}}_j(+i\hbar\lambda) e^{-\beta_N \hat{H}} \right]$$
$$\overset{N\to\infty}{=} i\hbar \frac{\beta_N}{2} \left[e^{-\beta_N \hat{H}} \dot{\hat{O}}_j + \dot{\hat{O}}_j e^{-\beta_N \hat{H}} \right]$$
$$\overset{N\to\infty}{=} i\hbar \frac{\beta_N}{2} \left[e^{-\beta_N \hat{H}}, \dot{\hat{O}}_j \right]_+ , \tag{66}$$

where we have used the fact that

$$\lim_{x\to 0} \frac{1}{x} \int_0^x dx' \; f(x') = f(0) \tag{67}$$

to linearize the Kubo integral in the $N \to \infty$ limit.

Combining relation Eq. (66) with Eq. (64), it follows that in the $N \to \infty$ limit

$$\sum_{j=1}^{N} \left[e^{-\beta \hat{H}} \right]_{\overline{N}} \left(e^{\frac{i\hbar}{2} \overleftrightarrow{\Lambda}_j} - e^{-\frac{i\hbar}{2} \overleftrightarrow{\Lambda}_j} \right) \left[\hat{O} \right]_{W,j} = i\hbar \frac{\beta_N}{2} \sum_{j=1}^{N} \left[e^{-\beta \hat{H}} \right]_{\overline{N}} \left(e^{\frac{i\hbar}{2} \overleftrightarrow{\Lambda}_j} + e^{-\frac{i\hbar}{2} \overleftrightarrow{\Lambda}_j} \right) \left[\dot{\hat{O}}(t) \right]_{W,j} \tag{68}$$

and, therefore, from Eqs. (59) and (60),

$$\left[e^{-\beta \hat{H}} \right]_{\overline{N}} \overleftrightarrow{s} \left[\hat{O} \right]_N = \beta \left[e^{-\beta \hat{H}} \right]_{\overline{N}} \overleftrightarrow{c} \left[\dot{\hat{O}}(t) \right]_N . \tag{69}$$

III. ALTERNATIVE EXPRESSION FOR CORRELATION FUNCTIONS

Due to the properties of the Sine and Cosine operators, there are different but equivalent forms of expressing time correlation functions in ring-polymer phase-space. We remark that previously derived path-integral expressions for the Kubo transformed and Double Kubo transformed correlation functions by others[7, 8] and us[6] are expressed in these alternative forms. The notation used in this paper, however, provides the most general form of correlation functions, allowing to apply the theory to general non-linear operators and non-equilibrium systems, permitting the natural extension of the theory to multi-point correlation functions, and highlighting the similarities and symmetries of the different correlation functions.

To demonstrate the equivalence of notation, consider the ring-polymer phase-space representation of the Kubo transform correlation function given by the Cosine two-point time correlation function

$$\left\langle \hat{B}(t) \overleftrightarrow{c} \hat{A} \right\rangle_N = \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \; \left[e^{-\beta \hat{H}} \right]_{\overline{N}} \left\{ \left[\hat{B}(t) \right]_N \overleftrightarrow{c} \left[\hat{A} \right]_N \right\} . \tag{70}$$

Using the Cosine operator self-adjoint property [Eq. (48)] and property under the integral sign [Eq. (50)], we can rewrite the Cosine correlation as

$$\left\langle \hat{B}(t)\overleftrightarrow{c}\hat{A}\right\rangle_N = \frac{Z_N^{-1}}{(2\pi\hbar)^N}\int d\mathbf{q}\int d\mathbf{p}\ \left\{\left[e^{-\beta\hat{H}}\right]_{\overline{N}}\overleftrightarrow{c}\left[\hat{A}\right]_N\right\}\left[\hat{B}(t)\right]_N. \quad (71)$$

But, since [see Section II, Eqs. (60), (64)]

$$\begin{aligned}\left[e^{-\beta\hat{H}}\right]_{\overline{N}}\overleftrightarrow{c}\left[\hat{A}\right]_N &= \frac{1}{2N}\sum_{j=1}^{N}\int d\boldsymbol{\Delta}\ \prod_{l=1}^{N}e^{\frac{i}{\hbar}p_l\Delta_l}\,\langle q_{l-1}-\frac{\Delta_{l-1}}{2}|\,[e^{-\beta_N\hat{H}},\hat{A}_j]_+\,|q_l+\frac{\Delta_l}{2}\rangle \\ &= \int d\boldsymbol{\Delta}\ \prod_{l=1}^{N}e^{\frac{i}{\hbar}p_l\Delta_l}\,\langle q_{l-1}-\frac{\Delta_{l-1}}{2}|\,\frac{1}{2}\left(\hat{\overline{A}}e^{-\beta_N\hat{H}}+e^{-\beta_N\hat{H}}\hat{\overline{A}}\right)|q_l+\frac{\Delta_l}{2}\rangle \\ &\equiv \left[e^{-\beta\hat{H}}\hat{\overline{A}}\right]_{\overline{N}},\end{aligned} \quad (72)$$

where, to simplify the notation, we have defined the operator

$$\hat{\overline{A}} = \frac{1}{N}\sum_{j=1}^{N}\hat{A}_j, \quad (73)$$

we can rewrite the Kubo correlation as

$$\left\langle \hat{B}(t)\overleftrightarrow{c}\hat{A}\right\rangle_N = \frac{Z_N^{-1}}{(2\pi\hbar)^N}\int d\mathbf{q}\int d\mathbf{p}\ \left[e^{-\beta\hat{H}}\hat{\overline{A}}\right]_{\overline{N}}\left[\hat{B}(t)\right]_N. \quad (74)$$

Note that for operators $\hat{A}=A(\hat{q})$ that only depend on position

$$\left[e^{-\beta\hat{H}}\hat{\overline{A}}\right]_{\overline{N}} = \int d\boldsymbol{\Delta}\ \frac{1}{2}\left(A(\mathbf{q}-\frac{\boldsymbol{\Delta}}{2})+A(\mathbf{q}+\frac{\boldsymbol{\Delta}}{2})\right)\prod_{l=1}^{N}e^{\frac{i}{\hbar}p_l\Delta_l}\,\langle q_{l-1}-\frac{\Delta_{l-1}}{2}|\,\frac{1}{2}e^{-\beta_N\hat{H}}\,|q_l+\frac{\Delta_l}{2}\rangle. \quad (75)$$

Moreover, for linear operators

$$\left[e^{-\beta\hat{H}}\hat{\overline{A}}\right]_{\overline{N}} = \int d\boldsymbol{\Delta}\ A(\mathbf{q})\prod_{l=1}^{N}e^{\frac{i}{\hbar}p_l\Delta_l}\,\langle q_{l-1}-\frac{\Delta_{l-1}}{2}|\,\frac{1}{2}e^{-\beta_N\hat{H}}\,|q_l+\frac{\Delta_l}{2}\rangle. \quad (76)$$

We remark that the path-integral expression for the Kubo transform correlation function given by Eqs. (74) and (75) [or (76)], - equivalent to the Cosine correlation function [Eq. (70)] - has previously appeared in the literature[6–8, 12, 13].

IV. EXACT RING-POLYMER PHASE-SPACE REPRESENTATION OF MULTI-POINT CORRELATION FUNCTIONS

Here, we present a step-by-step proof of the connection and equivalence between the ring-polymer phase-space and Hilbert space representation of time-correlation functions.

A. Auxiliary integrals

For future convenience, we introduce some general integrals in ring-polymer phase space and their connection to Hilbert space.

Consider the "type 1" integral defined as

$$I_1 = \frac{Z_N^{-1}}{(2\pi\hbar)^N}\int d\mathbf{q}\int d\mathbf{p}\ \left[e^{-\beta\hat{H}}\right]_{\overline{N}}\left(\frac{1}{N}\sum_{j=1}^{N}\left[\hat{O}\right]_{W,j}\right), \tag{77}$$

where $\hat{O}$ represents an *arbitrary* operator. Note that the integral defined above involves a ring-polymer phase-space average of a sum of one-dimensional Wigner transforms evaluated at particular phase-space points (q_j, p_j). However, recognizing the invariance of the Boltzmann operator $[e^{-\beta\hat{H}}]_{\overline{N}}$ to cyclic permutations of the variables (i.e. invariance to the change $q_l \to q_{l+1}$), the sum can be simplified to obtain an equivalent expression for I_1, namely

$$I_1 = \frac{Z_N^{-1}}{(2\pi\hbar)^N}\int d\mathbf{q}\int d\mathbf{p}\ \left[e^{-\beta\hat{H}}\right]_{\overline{N}}\left[\hat{O}\right]_{W,N}. \tag{78}$$

It is straightforward to perform the integration over the ring-polymer phase-space with the help of identity Eq. (11) to obtain

$$\begin{aligned}
I_1 &= \frac{Z_N^{-1}}{(2\pi\hbar)^N}\int d\mathbf{q}\int d\mathbf{p}\ \left[e^{-\beta\hat{H}}\right]_{\overline{N}}(\mathbf{q},\mathbf{p})\left[\hat{O}\right]_{W,N} \\
&= \frac{Z^{-1}}{2\pi\hbar}\int dq_N\int dp_N\int d\Delta_N\ e^{\frac{i}{\hbar}p_N\Delta_N}\langle q_N-\frac{\Delta_N}{2}|\, e^{-\beta\hat{H}}\,|q_N+\frac{\Delta_N}{2}\rangle \\
&\quad\times\left[\hat{O}\right]_{W,N} \\
&= \frac{Z^{-1}}{2\pi\hbar}\int dq_N\int dp_N\int d\Delta_N\ e^{\frac{i}{\hbar}p_N\Delta_N}\langle q_N-\frac{\Delta_N}{2}|\, e^{-\beta\hat{H}}\,|q_N+\frac{\Delta_N}{2}\rangle \\
&\quad\times\left\{\int d\Delta'_N e^{\frac{i}{\hbar}p_N\Delta'_N}\langle q_N-\frac{\Delta'_N}{2}|\,\hat{O}\,|q_N+\frac{\Delta'_N}{2}\rangle\right\} \\
&= Z^{-1}\int dq_N\int d\Delta_N\int d\Delta'_N\ \delta(\Delta_N+\Delta'_N)\,\langle q_N-\frac{\Delta_N}{2}|\, e^{-\beta\hat{H}}\,|q_N+\frac{\Delta_N}{2}\rangle \\
&\quad\times\langle q_N-\frac{\Delta'_N}{2}|\,\hat{O}\,|q_N+\frac{\Delta'_N}{2}\rangle \\
&= Z^{-1}\int dq_N\int d\Delta_N\ \langle q_N-\frac{\Delta_N}{2}|\, e^{-\beta\hat{H}}\,|q_N+\frac{\Delta_N}{2}\rangle \\
&\quad\times\langle q_N+\frac{\Delta_N}{2}|\,\hat{O}\,|q_N-\frac{\Delta_N}{2}\rangle \\
&= Z^{-1}\int dx_+\int dx_-\ \langle x_-|\, e^{-\beta\hat{H}}\,|x_+\rangle\langle x_+|\,\hat{O}\,|x_-\rangle \\
&= Z^{-1}Tr\left[e^{-\beta\hat{H}}\hat{O}\right].
\end{aligned} \tag{79}$$

We used the identity Eq. (11) to integrate out the variables p_k, q_k and Δ_k for $k \neq N$ (and recognized that $Z_N = Z$) in the second line [see the derivation of Eq. (15)], used the definition of Wigner-Weyl transform in the third line, performed the integral over p_N (giving a delta function) in the fourth line, performed the integral over Δ'_N in the fifth line, performed the change of variables $q \pm \frac{\Delta}{2} = x_\pm$ in the sixth line, and recognized a trace in the final line. We remark that type-1 integrals correspond to averages in Hilbert space.

Similarly, consider the "type-2" integral defined as

$$I_2 = \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \ \left[e^{-\beta\hat{H}}\right]_{\overline{N}} \left(\frac{1}{N} \sum_{\substack{j,k=1 \\ j\neq k}}^{N} \left[\hat{O}\right]_{W,j} \left[\hat{P}\right]_{W,k} \right), \tag{80}$$

where $\hat{O}$ and $\hat{P}$ represent *arbitrary* operators. Note that the integral defined above involves a ring-polymer phase-space average of a sum of products of one-dimensional Wigner transforms evaluated at different phase-space points (q_j, p_j) and (q_k, p_k), with $j \neq k$. However, recognizing the invariance of the Boltzmann operator $[e^{-\beta\hat{H}}]_{\overline{N}}$ to cyclic permutations of the variables, the sum can be simplified to obtain an equivalent expression for I_2, namely

$$\begin{aligned} I_2 &= \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \ \left[e^{-\beta\hat{H}}\right]_{\overline{N}} \left(\sum_{j=1}^{N-1} \left[\hat{O}\right]_{W,j} \right) \left[\hat{P}\right]_{W,N} \\ &= \sum_{j=1}^{N-1} I_2(j). \end{aligned} \tag{81}$$

Using the identity Eq. (11), the integration over ring-polymer phase-space can be performed, giving

$$\begin{aligned} I_2(j) &= \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \ \left[e^{-\beta\hat{H}}\right]_{\overline{N}} (\mathbf{q}, \mathbf{p}) \left[\hat{O}\right]_{W,j} \left[\hat{P}\right]_{W,N} \\ &= \frac{Z^{-1}}{(2\pi\hbar)^2} \int dq_j \int dq_N \int dp_j \int dp_N \int d\Delta_j \int d\Delta_N \ e^{\frac{i}{\hbar}p_j\Delta_j} e^{\frac{i}{\hbar}p_N\Delta_N} \\ &\quad \times \langle q_N - \frac{\Delta_N}{2}| \, (e^{-\beta_N\hat{H}})^j \, |q_j + \frac{\Delta_j}{2}\rangle \, \langle q_j - \frac{\Delta_j}{2}| \, (e^{-\beta_N\hat{H}})^{N-j} \, |q_N + \frac{\Delta_N}{2}\rangle \\ &\quad \times \left[\hat{O}\right]_{W,j} \left[\hat{P}\right]_{W,N} \\ &= \frac{Z^{-1}}{(2\pi\hbar)^2} \int dq_j \int dq_N \int dp_j \int dp_N \int d\Delta_j \int d\Delta_N \ e^{\frac{i}{\hbar}p_j\Delta_j} e^{\frac{i}{\hbar}p_N\Delta_N} \\ &\quad \times \langle q_N - \frac{\Delta_N}{2}| \, (e^{-\beta_N\hat{H}})^j \, |q_j + \frac{\Delta_j}{2}\rangle \, \langle q_j - \frac{\Delta_j}{2}| \, (e^{-\beta_N\hat{H}})^{N-j} \, |q_N + \frac{\Delta_N}{2}\rangle \\ &\quad \times \left[\hat{O}\right]_{W,j} \int d\Delta'_N e^{\frac{i}{\hbar}p_N\Delta'_N} \, \langle q_N - \frac{\Delta'_N}{2}| \, \hat{P} \, |q_N + \frac{\Delta'_N}{2}\rangle \end{aligned}$$

$$
\begin{aligned}
&= \frac{Z^{-1}}{2\pi\hbar}\int dq_j\int dq_N\int dp_j\int d\Delta_j\int d\Delta_N\int d\Delta'_N\; e^{\frac{i}{\hbar}p_j\Delta_j}\delta(\Delta_N+\Delta'_N)\\
&\quad\times\langle q_N-\frac{\Delta_N}{2}|\,(e^{-\beta_N\hat{H}})^j\,|q_j+\frac{\Delta_j}{2}\rangle\,\langle q_j-\frac{\Delta_j}{2}|\,(e^{-\beta_N\hat{H}})^{N-j}\,|q_N+\frac{\Delta_N}{2}\rangle\\
&\quad\times\left[\hat{O}\right]_{W,j}\langle q_N-\frac{\Delta'_N}{2}|\,\hat{P}\,|q_N+\frac{\Delta'_N}{2}\rangle\\
&= \frac{Z^{-1}}{2\pi\hbar}\int dq_j\int dq_N\int dp_j\int d\Delta_j\int d\Delta_N\; e^{\frac{i}{\hbar}p_j\Delta_j}\\
&\quad\times\langle q_N-\frac{\Delta_N}{2}|\,(e^{-\beta_N\hat{H}})^j\,|q_j+\frac{\Delta_j}{2}\rangle\,\langle q_j-\frac{\Delta_j}{2}|\,(e^{-\beta_N\hat{H}})^{N-j}\,|q_N+\frac{\Delta_N}{2}\rangle\\
&\quad\times\left[\hat{O}\right]_{W,j}\langle q_N+\frac{\Delta_N}{2}|\,\hat{P}\,|q_N-\frac{\Delta_N}{2}\rangle\\
&= Z^{-1}\int dq_j\int dq_N\int d\Delta_j\int d\Delta_N\\
&\quad\times\langle q_N-\frac{\Delta_N}{2}|\,(e^{-\beta_N\hat{H}})^j\,|q_j+\frac{\Delta_j}{2}\rangle\,\langle q_j-\frac{\Delta_j}{2}|\,(e^{-\beta_N\hat{H}})^{N-j}\,|q_N+\frac{\Delta_N}{2}\rangle\\
&\quad\times\langle q_j+\frac{\Delta_j}{2}|\,\hat{O}\,|q_j-\frac{\Delta_j}{2}\rangle\,\langle q_N+\frac{\Delta_N}{2}|\,\hat{P}\,|q_N-\frac{\Delta_N}{2}\rangle\\
&= Z^{-1}\int dx_+\int dx_-\int dy_+\int dy_-\\
&\quad\times\langle x_-|\,(e^{-\beta_N\hat{H}})^j\,|y_+\rangle\,\langle y_+|\,\hat{O}\,|y_-\rangle\,\langle y_-|\,(e^{-\beta_N\hat{H}})^{N-j}\,|x_+\rangle\,\langle x_+|\,\hat{P}\,|x_-\rangle\\
&= Z^{-1}Tr\left[(e^{-\beta_N\hat{H}})^j\hat{O}(e^{-\beta_N\hat{H}})^{N-j}\hat{P}\right].
\end{aligned}
\tag{82}
$$

We have used the identity Eq. (11) to integrate out the variables p_k, q_k and Δ_k for $k\neq j$, $k\neq N$ (and recognized that $Z_N = Z$) in the second line [see the derivation of Eq. (15)], used the definition of the Wigner-Weyl transform of $\hat{O}_2$ in the third line, performed the integral over p_N (giving a delta function) in the fourth line, performed the integral over Δ'_N in the fifth line, performed a similar integration over p_j and Δ'_j (arising from the definition of the Wigner-Weyl transform of $\hat{O}_1$) in the sixth line, performed the change of variables $q_N \pm \frac{\Delta_N}{2} = x_\pm$ and $q_j \pm \frac{\Delta_j}{2} = y_\pm$ in the seventh line, and recognized a trace in the final line. Therefore, it follows that

$$
\begin{aligned}
I_2 &= \sum_{j=1}^{N-1} Z^{-1}Tr\left[(e^{-\beta_N\hat{H}})^j\hat{O}(e^{-\beta_N\hat{H}})^{N-j}\hat{P}\right]\\
&= \sum_{j=1}^{N-1} Z^{-1}Tr\left[(e^{-\beta_N\hat{H}})^{N-j}\hat{O}(e^{-\beta_N\hat{H}})^{j}\hat{P}\right].
\end{aligned}
\tag{83}
$$

We remark that type-2 integrals correspond to discrete Kubo transform correlations between arbitrary observables $\hat{O}$ and $\hat{P}$. [2]

[2] Technically speaking, the end points of the Kubo integral discretization are not accounted for.

Similarly, consider the "type-3" integral defined as

$$I_3 = \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta \hat{H}}\right]_{\overline{N}} \left(\frac{1}{N} \sum_{\substack{j,k,l=1 \\ j\neq k\neq l}}^{N} \left[\hat{O}\right]_{W,j} \left[\hat{P}\right]_{W,k} \left[\hat{Q}\right]_{W,l} \right), \tag{84}$$

where $\hat{O}$, $\hat{P}$ and $\hat{Q}$ represent *arbitrary* operators. Note that the integral defined above involves a ring-polymer phase-space average of a sum of products of one-dimensional Wigner transforms evaluated at different phase-space points (q_j, p_j), (q_k, p_k) and (q_l, p_l) with $j \neq k$, $j \neq l$, $k \neq l$. However, recognizing the invariance of the Boltzmann operator $[e^{-\beta \hat{H}}]_{\overline{N}}$ to cyclic permutations of the variables, the sum can be simplified to obtain an equivalent expression for I_3, namely

$$\begin{aligned} I_3 &= \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta \hat{H}}\right]_{\overline{N}} \left(\sum_{\substack{j,k=1 \\ j\neq k}}^{N-1} \left[\hat{O}\right]_{W,j} \left[\hat{P}\right]_{W,k} \right) \left[\hat{Q}\right]_{W,N} \\ &= \sum_{\substack{j,k=1 \\ j\neq k}}^{N-1} I_3(j,k) \\ &= \sum_{k=1}^{N-1} \sum_{j=1}^{k-1} I_3(j,k) + \sum_{k=1}^{N-1} \sum_{j=k+1}^{N-1} I_3(j,k). \end{aligned} \tag{85}$$

Employing the identity Eq. (11), integration over the ring-polymer phase-space coordinates can be performed. For the case $j < k$, one obtains

$$\begin{aligned} I_3(j,k;j<k) &= \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta \hat{H}}\right]_{\overline{N}} (\mathbf{q},\mathbf{p}) \left[\hat{O}\right]_{W,j} \left[\hat{P}\right]_{W,k} \left[\hat{Q}\right]_{W,N} \\ &= \frac{Z^{-1}}{(2\pi\hbar)^3} \int dq_j \int dq_k \int dq_N \int dp_j \int dp_k \int dp_N \int d\Delta_j \int d\Delta_k \int d\Delta_N \\ &\quad \times e^{\frac{i}{\hbar} p_j \Delta_j} e^{\frac{i}{\hbar} p_k \Delta_k} e^{\frac{i}{\hbar} p_N \Delta_N} \langle q_N - \frac{\Delta_N}{2} | \, (e^{-\beta_N \hat{H}})^j \, | q_j + \frac{\Delta_j}{2} \rangle \\ &\quad \times \langle q_j - \frac{\Delta_j}{2} | \, (e^{-\beta_N \hat{H}})^{k-j} \, | q_k + \frac{\Delta_k}{2} \rangle \langle q_k - \frac{\Delta_k}{2} | \, (e^{-\beta_N \hat{H}})^{N-k} \, | q_N + \frac{\Delta_N}{2} \rangle \\ &\quad \times \left[\hat{O}\right]_{W,j} \left[\hat{P}\right]_{W,k} \left[\hat{Q}\right]_{W,N} \\ &= \frac{Z^{-1}}{(2\pi\hbar)^3} \int dq_j \int dq_k \int dq_N \int dp_j \int dp_k \int dp_N \int d\Delta_j \int d\Delta_k \int d\Delta_N \\ &\quad \times e^{\frac{i}{\hbar} p_j \Delta_j} e^{\frac{i}{\hbar} p_k \Delta_k} e^{\frac{i}{\hbar} p_N \Delta_N} \langle q_N - \frac{\Delta_N}{2} | \, (e^{-\beta_N \hat{H}})^j \, | q_j + \frac{\Delta_j}{2} \rangle \\ &\quad \times \langle q_j - \frac{\Delta_j}{2} | \, (e^{-\beta_N \hat{H}})^{k-j} \, | q_k + \frac{\Delta_k}{2} \rangle \langle q_k - \frac{\Delta_k}{2} | \, (e^{-\beta_N \hat{H}})^{N-k} \, | q_N + \frac{\Delta_N}{2} \rangle \end{aligned}$$

$$
\begin{aligned}
&\times \left[\hat{O}\right]_{W,j} \left[\hat{P}\right]_{W,k} \left\{ \int d\Delta'_N e^{\frac{i}{\hbar} p_N \Delta'_N} \langle q_N - \frac{\Delta'_N}{2} | \, \hat{Q} \, | q_N + \frac{\Delta'_N}{2} \rangle \right\} \\
= \;& \frac{Z^{-1}}{(2\pi\hbar)^2} \int dq_j \int dq_k \int dq_N \int dp_j \int dp_k \int d\Delta_j \int d\Delta_k \int d\Delta_N \int d\Delta'_N \\
&\times e^{\frac{i}{\hbar} p_j \Delta_j} e^{\frac{i}{\hbar} p_k \Delta_k} \delta(\Delta_N + \Delta'_N) \langle q_N - \frac{\Delta_N}{2} | \, (e^{-\beta_N \hat{H}})^j \, | q_j + \frac{\Delta_j}{2} \rangle \\
&\times \langle q_j - \frac{\Delta_j}{2} | \, (e^{-\beta_N \hat{H}})^{k-j} \, | q_k + \frac{\Delta_k}{2} \rangle \langle q_k - \frac{\Delta_k}{2} | \, (e^{-\beta_N \hat{H}})^{N-k} \, | q_N + \frac{\Delta_N}{2} \rangle \\
&\times \left[\hat{O}\right]_{W,j} \left[\hat{P}\right]_{W,k} \langle q_N - \frac{\Delta'_N}{2} | \, \hat{Q} \, | q_N + \frac{\Delta'_N}{2} \rangle \\
= \;& \frac{Z^{-1}}{(2\pi\hbar)^2} \int dq_j \int dq_k \int dq_N \int dp_j \int dp_k \int d\Delta_j \int d\Delta_k \int d\Delta_N \\
&\times e^{\frac{i}{\hbar} p_j \Delta_j} e^{\frac{i}{\hbar} p_k \Delta_k} \langle q_N - \frac{\Delta_N}{2} | \, (e^{-\beta_N \hat{H}})^j \, | q_j + \frac{\Delta_j}{2} \rangle \\
&\times \langle q_j - \frac{\Delta_j}{2} | \, (e^{-\beta_N \hat{H}})^{k-j} \, | q_k + \frac{\Delta_k}{2} \rangle \langle q_k - \frac{\Delta_k}{2} | \, (e^{-\beta_N \hat{H}})^{N-k} \, | q_N + \frac{\Delta_N}{2} \rangle \\
&\times \left[\hat{O}\right]_{W,j} \left[\hat{P}\right]_{W,k} \langle q_N + \frac{\Delta_N}{2} | \, \hat{Q} \, | q_N - \frac{\Delta_N}{2} \rangle \\
= \;& \frac{Z^{-1}}{2\pi\hbar} \int dq_j \int dq_k \int dq_N \int dp_j \int d\Delta_j \int d\Delta_k \int d\Delta_N \\
&\times e^{\frac{i}{\hbar} p_j \Delta_j} \langle q_N - \frac{\Delta_N}{2} | \, (e^{-\beta_N \hat{H}})^j \, | q_j + \frac{\Delta_j}{2} \rangle \\
&\times \langle q_j - \frac{\Delta_j}{2} | \, (e^{-\beta_N \hat{H}})^{k-j} \, | q_k + \frac{\Delta_k}{2} \rangle \langle q_k - \frac{\Delta_k}{2} | \, (e^{-\beta_N \hat{H}})^{N-k} \, | q_N + \frac{\Delta_N}{2} \rangle \\
&\times \left[\hat{O}\right]_{W,j} \langle q_k + \frac{\Delta_k}{2} | \, \hat{P} \, | q_k - \frac{\Delta_k}{2} \rangle \langle q_N + \frac{\Delta_N}{2} | \, \hat{Q} \, | q_N - \frac{\Delta_N}{2} \rangle \\
= \;& Z^{-1} \int dq_j \int dq_k \int dq_N \int d\Delta_j \int d\Delta_k \int d\Delta_N \\
&\times \langle q_N - \frac{\Delta_N}{2} | \, (e^{-\beta_N \hat{H}})^j \, | q_j + \frac{\Delta_j}{2} \rangle \\
&\times \langle q_j - \frac{\Delta_j}{2} | \, (e^{-\beta_N \hat{H}})^{k-j} \, | q_k + \frac{\Delta_k}{2} \rangle \langle q_k - \frac{\Delta_k}{2} | \, (e^{-\beta_N \hat{H}})^{N-k} \, | q_N + \frac{\Delta_N}{2} \rangle \\
&\times \langle q_j + \frac{\Delta_j}{2} | \, \hat{O} \, | q_j - \frac{\Delta_j}{2} \rangle \langle q_k + \frac{\Delta_k}{2} | \, \hat{P} \, | q_k - \frac{\Delta_k}{2} \rangle \langle q_N + \frac{\Delta_N}{2} | \, \hat{Q} \, | q_N - \frac{\Delta_N}{2} \rangle \\
= \;& Z^{-1} \int dx_+ \int dx_- \int dy_+ \int dy_- \int dz_+ \int dz_- \, \langle x_- | \, (e^{-\beta_N \hat{H}})^j \, | y_+ \rangle \langle y_+ | \, \hat{O}_1 \, | y_- \rangle \\
&\times \langle y_- | \, (e^{-\beta_N \hat{H}})^{k-j} \, | z_+ \rangle \langle z_+ | \, \hat{O}_2 \, | z_- \rangle \langle z_- | \, (e^{-\beta_N \hat{H}})^{N-k} \, | x_+ \rangle \langle x_+ | \, \hat{O}_3 \, | x_- \rangle \\
= \;& Z^{-1} Tr \left[(e^{-\beta_N \hat{H}})^j \hat{O} (e^{-\beta_N \hat{H}})^{k-j} \hat{P} (e^{-\beta_N \hat{H}})^{N-k} \hat{Q} \right]. \qquad (86)
\end{aligned}
$$

We have used the identity Eq. (11) to integrated out the variables p_l, q_l and Δ_l for $l \neq j, k, N$ in the second line [see derivation of Eq. (15)], used the definition of the Wigner-Weyl transform of $\hat{O}_3$ in the third line, performed the integral over p_N (giving a delta function) in the fourth line, performed the integral over Δ'_N in the fifth line, performed the integral over p_k and Δ'_k (arising from the definition of the Wigner-Weyl transform of $\hat{O}_2$) in the sixth line, performed the integral over p_j and Δ'_j (arising from the definition of the Wigner-Weyl transform of $\hat{O}_1$) in the seventh line, performed the change of variables $q_N \pm \frac{\Delta_N}{2} = x_\pm$, $q_j \pm \frac{\Delta_j}{2} = y_\pm$ and $q_k \pm \frac{\Delta_k}{2} = z_\pm$ in the eighth line, and recognized a trace in the last line.

Following a similar analysis, for $j > k$ it follows that

$$I_3(j,k;j>k) \;=\; Z^{-1}Tr\left[(e^{-\beta_N\hat{H}})^k\hat{P}(e^{-\beta_N\hat{H}})^{j-k}\hat{O}(e^{-\beta_N\hat{H}})^{N-j}\hat{Q}\right]. \tag{87}$$

Notice that

$$\begin{aligned}\sum_{k=1}^{N-1}\sum_{j=1}^{k-1} I_3(j,k) \;&=\; \sum_{k=1}^{N-1}\sum_{j=1}^{k-1} Z^{-1}Tr\left[(e^{-\beta_N\hat{H}})^j\hat{O}(e^{-\beta_N\hat{H}})^{k-j}\hat{P}(e^{-\beta_N\hat{H}})^{N-k}\hat{Q}\right]\\ &=\; \sum_{j=1}^{N-1}\sum_{k=1}^{j-1} Z^{-1}Tr\left[(e^{-\beta_N\hat{H}})^{N-j}\hat{O}(e^{-\beta_N\hat{H}})^{j-k}\hat{P}(e^{-\beta_N\hat{H}})^{k}\hat{Q}\right], \end{aligned} \tag{88}$$

and

$$\begin{aligned}\sum_{k=1}^{N-1}\sum_{j=k+1}^{N-1} I_3(j,k) \;&=\; \sum_{k=1}^{N-1}\sum_{j=k+1}^{N-1} Z^{-1}Tr\left[(e^{-\beta_N\hat{H}})^k\hat{P}(e^{-\beta_N\hat{H}})^{j-k}\hat{O}(e^{-\beta_N\hat{H}})^{N-j}\hat{Q}\right]\\ &=\; \sum_{j=1}^{N-1}\sum_{k=j+1}^{N-1} Z^{-1}Tr\left[(e^{-\beta_N\hat{H}})^{N-k}\hat{P}(e^{-\beta_N\hat{H}})^{k-j}\hat{O}(e^{-\beta_N\hat{H}})^{j}\hat{Q}\right], \end{aligned} \tag{89}$$

where we have reordered the sum and relabeled the dummy indexes to obtain the last equalities in the previous equations.

Combining the terms, it follows that

$$\begin{aligned} I_3 \;=\; &\sum_{j=1}^{N-1}\sum_{k=1}^{j-1} Z^{-1}Tr\left[(e^{-\beta_N\hat{H}})^{N-j}\hat{O}(e^{-\beta_N\hat{H}})^{j-k}\hat{P}(e^{-\beta_N\hat{H}})^{k}\hat{Q}\right]\\ &+\sum_{j=1}^{N-1}\sum_{k=j+1}^{N-1} Z^{-1}Tr\left[(e^{-\beta_N\hat{H}})^{N-k}\hat{P}(e^{-\beta_N\hat{H}})^{k-j}\hat{O}(e^{-\beta_N\hat{H}})^{j}\hat{Q}\right]. \end{aligned} \tag{90}$$

We remark that type-3 integrals are related to (discrete) symmetrized Double-Kubo transform correlations involving arbitrary operators $\hat{O}$, $\hat{P}$ and $\hat{Q}$.

We remark that the procedure to define and evaluate "type-n" integrals I_n can be generalized to any order. However, as the order of the integral increase, the number of terms and possible permutations to be considered in the sums quickly grow, and the derivation of the expressions become tedious. Nevertheless, clear trends can be identified: a type-n integral consist of *ordered* sums involving different imaginary-time steps $e^{\beta_N\hat{H}}$ between all the possible permutations of operators [e.g., Eqs. (83) and (90)]. As such, type-n integrals are discrete versions of symmetrized n-order Kubo transforms.

B. Two-point correlation functions

1. *"Sine" correlation functions*

Consider the "Sine" ring-polymer correlation defined as

$$\begin{aligned}\left\langle \hat{O}_1 \overset{\leftrightarrow}{s} \hat{O}_2 \right\rangle_N &\equiv \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta \hat{H}} \right]_{\overline{N}} \left\{ \left[\hat{O}_1 \right]_N \overset{\leftrightarrow}{s} \left[\hat{O}_2 \right]_N \right\} \\ &= \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta \hat{H}} \right]_{\overline{N}} (\mathbf{q}, \mathbf{p}) \\ &\quad \times \left\{ \left[\hat{O}_1 \right]_N (\mathbf{q}, \mathbf{p}) \left(\frac{2N}{\hbar} \right) \sin \left(\frac{\hbar}{2} \overset{\leftrightarrow}{\Lambda}_N \right) \left[\hat{O}_2 \right]_N (\mathbf{q}, \mathbf{p}) \right\} . \end{aligned} \tag{91}$$

Introducing the expressions for the Sine coupling [Eq. (38)] into Eq. (91), the Sine correlation can be expressed as

$$\begin{aligned}\left\langle \hat{O}_1 \overset{\leftrightarrow}{s} \hat{O}_2 \right\rangle_N &= \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta \hat{H}} \right]_{\overline{N}} (\mathbf{q}, \mathbf{p}) \\ &\quad \times \left\{ \left(\frac{i}{\hbar} \right) \frac{1}{N} \sum_{j=1}^{N} \left[[\hat{O}_1, \hat{O}_2] \right]_{W,j} \right\} . \end{aligned} \tag{92}$$

Besides an additional $i/\hbar$ factor, the Sine correlation in Eq. (92) represents a type-1 integral [Eq. (77)] with $\hat{O} = [\hat{O}_1, \hat{O}_2]$. Therefore, it follows from Eq. (79) that

$$\left\langle \hat{O}_1 \overset{\leftrightarrow}{s} \hat{O}_2 \right\rangle_N = \left(\frac{i}{\hbar} \right) \left\langle [\hat{O}_1, \hat{O}_2] \right\rangle , \tag{93}$$

namely, Sine correlation functions are an exact ring-polymer phase-space representation of a correlation involving a commutator between two operators. We remark that this equality holds for any finite N. Note that for $\hat{O}_1 = \hat{B}(t_1)$ and $\hat{O}_2 = \hat{A}(t_0)$, one obtains the correlation functions appearing in linear response theory.

2. *"Cosine" correlation functions*

Consider the "Cosine" ring-polymer correlation defined as

$$\begin{aligned}\left\langle \hat{O}_1 \overset{\leftrightarrow}{c} \hat{O}_2 \right\rangle_N &\equiv \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta \hat{H}} \right]_{\overline{N}} \left\{ \left[\hat{O}_1 \right]_N \overset{\leftrightarrow}{c} \left[\hat{O}_2 \right]_N \right\} \\ &= \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta \hat{H}} \right]_{\overline{N}} (\mathbf{q}, \mathbf{p}) \\ &\quad \times \left\{ \left[\hat{O}_1 \right]_N (\mathbf{q}, \mathbf{p}) \cos \left(\frac{\hbar}{2} \overset{\leftrightarrow}{\Lambda}_N \right) \left[\hat{O}_2 \right]_N (\mathbf{q}, \mathbf{p}) \right\} . \end{aligned} \tag{94}$$

Introducing the expressions for the Cosine coupling [Eq. (40)] into Eq. (94), the Cosine correlation can be decomposed into two terms, namely

$$\left\langle \hat{O}_1 \overleftrightarrow{c} \hat{O}_2 \right\rangle_N = \frac{1}{2} T_1 + T_2, \tag{95}$$

with

$$T_1 = \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta\hat{H}} \right]_{\overline{N}} \left\{ \frac{1}{N^2} \sum_{j=1}^{N} \left[[\hat{O}_1, \hat{O}_2]_+ \right]_{W,j} \right\}, \tag{96a}$$

and

$$T_2 = \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta\hat{H}} \right]_{\overline{N}} \left\{ \frac{1}{N^2} \sum_{\substack{j,k=1 \\ j\neq k}}^{N} [\hat{O}_1]_{W,j} [\hat{O}_2]_{W,k} \right\}. \tag{96b}$$

Besides an additional $1/N$ factor, the T_1 term in Eqs. (96) represents a type-1 integral [Eq. (77)] with $\hat{O} = [\hat{O}_1, \hat{O}_2]_+$, whereas the T_2 term represents a type-2 integral [Eq. (80)] with $\hat{O} = \hat{O}_1$ and $\hat{P} = \hat{O}_2$. Therefore, it follows from Eqs. (79) and (83) that

$$\left\langle \hat{O}_1 \overleftrightarrow{c} \hat{O}_2 \right\rangle_N = \frac{Z^{-1}}{N} {\sum_{j=0}^{N}}' Tr \left[(e^{-\beta_N \hat{H}})^{N-j} \hat{O}_1 (e^{-\beta_N \hat{H}})^j \hat{O}_2 \right], \tag{97}$$

where the prime in $\sum'$ indicates that the first and last indexes are weighted by one-half. Recognizing that the sum over j is just the trapezoidal rule for the discretization of a Riemann integral, it follows that in the $N \to \infty$ limit

$$\begin{aligned} \lim_{N\to\infty} \left\langle \hat{O}_1 \overleftrightarrow{c} \hat{O}_2 \right\rangle_N &= \frac{Z^{-1}}{\beta} \int_0^\beta d\lambda Tr \left[e^{-(\beta-\lambda)\hat{H}} \hat{O}_1 e^{-\lambda\hat{H}} \hat{O}_2 \right] \\ &= \frac{1}{\beta} \int_0^\beta d\lambda \left\langle \hat{O}_1(-i\hbar\lambda) \hat{O}_2 \right\rangle \\ &= \left\langle \hat{O}_1 \bullet \hat{O}_2 \right\rangle, \end{aligned} \tag{98}$$

namely, Cosine correlation functions are an exact ring-polymer phase-space representation of correlations involving a Kubo integral between two operators. Note that for $\hat{O}_1 = \hat{B}(t_1)$ and $\hat{O}_2 = \hat{A}(t_0)$, one obtains the correlation functions appearing in linear response theory.

C. Three-point correlation functions

1. "Sine-Sine" correlation functions

Consider the "Sine-Sine" ring-polymer defined as

$$\begin{aligned}
\left\langle \left\{ \hat{O}_1 \overleftrightarrow{s} \hat{O}_2 \right\} \overleftrightarrow{s} \hat{O}_3 \right\rangle_N &\equiv \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \left[e^{-\beta \hat{H}} \right]_{\overline{N}} \left\{ \left\{ \left[\hat{O}_1 \right]_N \overleftrightarrow{s} \left[\hat{O}_2 \right]_N \right\} \overleftrightarrow{s} \left[\hat{O}_3 \right]_N \right\} \\
&= \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \left[e^{-\beta \hat{H}} \right]_{\overline{N}} (\mathbf{q}, \mathbf{p}) \\
&\times \left\{ \left\{ \left[\hat{O}_1 \right]_N (\mathbf{q}, \mathbf{p}) \left(\frac{2N}{\hbar} \right) \sin \left(\frac{\hbar}{2} \overleftrightarrow{\Lambda}_N \right) \left[\hat{O}_2 \right]_N (\mathbf{q}, \mathbf{p}) \right\} \right. \\
&\times \left. \left(\frac{2N}{\hbar} \right) \sin \left(\frac{\hbar}{2} \overleftrightarrow{\Lambda}_N \right) \left[\hat{O}_3 \right]_N (\mathbf{q}, \mathbf{p}) \right\}. \qquad (99)
\end{aligned}$$

Introducing the expressions for the Sine-Sine coupling [Eq. (41)] into Eq. (99), the Sine-Sine correlation can be expressed as

$$\begin{aligned}
\left\langle \left\{ \hat{O}_1 \overleftrightarrow{s} \hat{O}_2 \right\} \overleftrightarrow{s} \hat{O}_3 \right\rangle_N &= \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \left[e^{-\beta \hat{H}} \right]_{\overline{N}} (\mathbf{q}, \mathbf{p}) \\
&\times \left\{ \left(\frac{i}{\hbar} \right)^2 \frac{1}{N} \sum_{j=1}^{N} \left[[[\hat{O}_1, \hat{O}_2], \hat{O}_3] \right]_{W,j} \right\}. \qquad (100)
\end{aligned}$$

Besides an additional $(i/\hbar)^2$ factor, the T_1 term in Eq. (100) represents a type-1 integral [Eq. (77)] with $\hat{O} = [[\hat{O}_1, \hat{O}_2], \hat{O}_3]$. Therefore, it follows from Eq. (79) that

$$\left\langle \left\{ \hat{O}_1 \overleftrightarrow{s} \hat{O}_2 \right\} \overleftrightarrow{s} \hat{O}_3 \right\rangle_N = \left(\frac{i}{\hbar} \right)^2 \left\langle \left[\left[\hat{O}_1, \hat{O}_2 \right], \hat{O}_3 \right] \right\rangle, \qquad (101)$$

namely, Sine-Sine correlation functions are an exact ring-polymer phase-space representation of correlation functions involving a double-commutation relation between operators. We remark that this equality holds for any finite N. Note that for $\hat{O}_1 = \hat{C}(t_2)$, $\hat{O}_2 = \hat{B}(t_1)$, and $\hat{O}_3 = \hat{A}(t_0)$ one obtains the correlation function appearing in second-order response theory.

2. "Sine-Cosine" correlation functions

Consider the "Sine-Cosine" ring-polymer correlation defined as

$$\begin{aligned}
\left\langle \left\{ \hat{O}_1 \overleftrightarrow{s} \hat{O}_2 \right\} \overleftrightarrow{c} \hat{O}_3 \right\rangle_N &\equiv \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \left[e^{-\beta \hat{H}} \right]_{\overline{N}} \left\{ \left\{ \left[\hat{O}_1 \right]_N \overleftrightarrow{s} \left[\hat{O}_2 \right]_N \overleftrightarrow{c} \right\} \left[\hat{O}_3 \right]_N \right\} \\
&= \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \left[e^{-\beta \hat{H}} \right]_{\overline{N}} (\mathbf{q}, \mathbf{p})
\end{aligned}$$

$$\times \left\{ \left\{ \left[\hat{O}_1\right]_N (\mathbf{q},\mathbf{p}) \left(\frac{2N}{\hbar}\right) \sin\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) \left[\hat{O}_2\right]_N (\mathbf{q},\mathbf{p}) \right\} \right.$$
$$\left. \times \cos\left(\frac{\hbar}{2}\overleftrightarrow{\Lambda}_N\right) \left[\hat{O}_3\right]_N (\mathbf{q},\mathbf{p}) \right\}. \tag{102}$$

Introducing the expressions for the Sine-Cosine coupling [Eq. (42)] into Eq. (102), the Sine-Cosine correlation can be decomposed into two terms, namely

$$\left\langle \left\{ \hat{O}_1 \overleftrightarrow{s} \hat{O}_2 \right\} \overleftrightarrow{c} \hat{O}_3 \right\rangle_N = \frac{1}{2} T_1 + T_2, \tag{103}$$

with

$$T_1 = \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta\hat{H}}\right]_{\overline{N}} \left\{ \left(\frac{i}{\hbar}\right) \frac{1}{N^2} \sum_{j=1}^{N} \left[[[\hat{O}_1,\hat{O}_2]_-,\hat{O}_3]_+\right]_{W,j} \right\} \tag{104a}$$

$$T_2 = \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta\hat{H}}\right]_{\overline{N}} \left\{ \left(\frac{i}{\hbar}\right) \frac{1}{N^2} \sum_{\substack{j,k=1\\ j\neq k}}^{N} \left[[\hat{O}_1,\hat{O}_2]\right]_{W,j} [\hat{O}_3]_{W,k} \right\} \tag{104b}$$

Besides an additional $i/(\hbar N)$ factor, the T_1 term in Eqs. (104) represents a type-1 integral [Eq. (77)] with $\hat{O} = [[\hat{O}_1,\hat{O}_2]_-,\hat{O}_3]_+$, whereas the T_2 term represents a type-2 integral [Eq. (80)] with $\hat{O} = \hat{O}_2$ and $\hat{P} = \hat{O}_3$. Therefore, it follows from Eqs. (79) and (83) that

$$\left\langle \left\{ \hat{O}_1 \overleftrightarrow{s} \hat{O}_2 \right\} \overleftrightarrow{c} \hat{O}_3 \right\rangle_N = \left(\frac{i}{\hbar}\right) \frac{Z^{-1}}{N} {\sum_{j=0}^{N}}' Tr\left[(e^{-\beta_N \hat{H}})^{N-j} [\hat{O}_1,\hat{O}_2] (e^{-\beta_N \hat{H}})^j \hat{O}_3 \right], \tag{105}$$

where the prime in $\sum'$ indicates that the first and last indexes are weighted by one-half. Recognizing that the sum over j is just the trapezoidal rule for the discretization of a Riemann integral, it follows that in the $N \to \infty$ limit

$$\begin{aligned} \left\langle \left\{ \hat{O}_1 \overleftrightarrow{s} \hat{O}_2 \right\} \overleftrightarrow{c} \hat{O}_3 \right\rangle_N &= \left(\frac{i}{\hbar}\right) \frac{Z^{-1}}{\beta} \int_0^\beta d\lambda Tr\left[e^{-(\beta-\lambda)\hat{H}} [\hat{O}_1,\hat{O}_2] e^{-\lambda\hat{H}} \hat{O}_3 \right] \\ &= \left(\frac{i}{\hbar}\right) \left\langle [\hat{O}_1,\hat{O}_2] \bullet \hat{O}_3 \right\rangle, \end{aligned} \tag{106}$$

namely, Sine-Cosine correlation functions are exact ring-polymer phase space representation of correlation functions involving a commutator relation and Kubo integral between three operators. Note that for $\hat{O}_1 = \hat{A}(t_0)$, $\hat{O}_2 = \hat{B}(t_1)$, and $\hat{O}_3 = \hat{C}(t_2)$, one obtains the correlation function appearing in second-order response theory.

3. "Cosine-Cosine" correlation functions

Consider the "Cosine-Cosine" ring-polymer defined as

$$\begin{aligned}\left\langle \hat{O}_1 \overleftrightarrow{c} \hat{O}_2 \overleftrightarrow{c} \hat{O}_3 \right\rangle_N &\equiv \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta\hat{H}}\right]_{\overline{N}} \left\{ \left[\hat{O}_1\right]_N \overleftrightarrow{c} \left[\hat{O}_2\right]_N \overleftrightarrow{c} \left[\hat{O}_3\right]_N \right\} \\ &= \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta\hat{H}}\right]_{\overline{N}} (\mathbf{q},\mathbf{p}) \\ &\quad \times \left\{ \left[\hat{O}_1\right]_N (\mathbf{q},\mathbf{p}) \cos\left(\frac{\hbar}{2} \overleftrightarrow{\Lambda}_N\right) \left[\hat{O}_2\right]_N (\mathbf{q},\mathbf{p}) \right. \\ &\quad \left. \times \cos\left(\frac{\hbar}{2} \overleftrightarrow{\Lambda}_N\right) \left[\hat{O}_3\right]_N (\mathbf{q},\mathbf{p}) \right\}. \end{aligned} \tag{107}$$

Introducing the expressions for the Cosine-Cosine coupling [Eq. (44)] into Eq. (107), the Cosine-Cosine correlation can be expressed as

$$\left\langle \hat{O}_1 \overleftrightarrow{c} \hat{O}_2 \overleftrightarrow{c} \hat{O}_3 \right\rangle_N = \frac{1}{4} T_1 + \frac{1}{2}(T_2 + T_2' + T_2'') + T_3, \tag{108}$$

with

$$T_1 = \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta\hat{H}}\right]_{\overline{N}} (\mathbf{q},\mathbf{p}) \left\{ \frac{1}{N^3} \sum_{j=1}^{N} \left[[[\hat{O}_1, \hat{O}_2]_+, \hat{O}_3]_+ \right]_{W,j} \right\}, \tag{109a}$$

$$T_2 = \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta\hat{H}}\right]_{\overline{N}} (\mathbf{q},\mathbf{p}) \left\{ \frac{1}{N^3} \sum_{\substack{j,l=1 \\ j\neq l}}^{N} \left[[\hat{O}_1, \hat{O}_2]_+ \right]_{W,j} [\hat{O}_3]_{W,l} \right\}, \tag{109b}$$

$$T_2' = \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta\hat{H}}\right]_{\overline{N}} (\mathbf{q},\mathbf{p}) \left\{ \frac{1}{N^3} \sum_{\substack{j,k=1 \\ j\neq k}}^{N} [\hat{O}_1]_{W,j} \left[[\hat{O}_2, \hat{O}_3]_+ \right]_{W,k} \right\}, \tag{109c}$$

$$T_2'' = \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta\hat{H}}\right]_{\overline{N}} (\mathbf{q},\mathbf{p}) \left\{ \frac{1}{N^3} \sum_{\substack{j,k=1 \\ j\neq k}}^{N} [\hat{O}_2]_{W,k} \left[[\hat{O}_1, \hat{O}_3]_+ \right]_{W,j} \right\}, \tag{109d}$$

and

$$T_3 = \frac{Z_N^{-1}}{(2\pi\hbar)^N} \int d\mathbf{q} \int d\mathbf{p} \, \left[e^{-\beta\hat{H}}\right]_{\overline{N}} (\mathbf{q},\mathbf{p}) \left\{ \frac{1}{N^3} \sum_{\substack{j,k,l=1 \\ j\neq k, j\neq l, k\neq l}}^{N} [\hat{O}_1]_{W,j} [\hat{O}_2]_{W,k} [\hat{O}_3]_{W,l} \right\} \tag{109e}$$

Besides an additional $1/N^2$ factor, the T_j terms in Eqs. (109) represent different type of integrals introduced in Sec. IV A. For instances, T_1 is a type-1 integral [Eq. (77)] with $\hat{O} = [[\hat{O}_1, \hat{O}_2]_+, \hat{O}_3]_+$. The T_2 terms represent type-2 integrals [Eq. (80)] with $\hat{O} = [\hat{O}_1, \hat{O}_2]_+$ and $\hat{P} = \hat{O}_3$ for T_2, $\hat{O} = \hat{O}_1$ and $\hat{P} = [\hat{O}_2, \hat{O}_3]_+$ for T_2', and $\hat{O} = \hat{O}_2$ and $\hat{P} = [\hat{O}_1, \hat{O}_3]_+$ for T_2''. The term T_3 represent a type-3 integral [Eq. (84)] with $\hat{O} = \hat{O}_1$, $\hat{P} = \hat{O}_2$ and $\hat{Q} = \hat{O}_3$.

Combining all the T_j terms together, it follows from Eqs. (79), (83) and (90) that

$$
\begin{aligned}
\left\langle \hat{O}_1 \overset{\leftrightarrow}{c} \hat{O}_2 \overset{\leftrightarrow}{c} \hat{O}_3 \right\rangle_N &= \frac{Z^{-1}}{N^2} \left\{ \sum_{j=0}^{N}{}' \sum_{k=0}^{j}{}' Tr\left[(e^{-\beta_N \hat{H}})^{N-j} \hat{O}_1 (e^{-\beta_N \hat{H}})^{j-k} \hat{O}_2 (e^{-\beta_N \hat{H}})^{k} \hat{O}_3 \right] \right. \\
&\quad \left. + \sum_{j=0}^{N}{}' \sum_{k=j}^{N}{}' Tr\left[(e^{-\beta_N \hat{H}})^{N-k} \hat{O}_2 (e^{-\beta_N \hat{H}})^{k-j} \hat{O}_1 (e^{-\beta_N \hat{H}})^{j} \hat{O}_3 \right] \right\}, \qquad (110)
\end{aligned}
$$

where the prime in $\sum'$ indicates that the first and last indexes are weighted by one-half. Recognizing that the sum over j, k is just the trapezoidal rule for the discretization of an iterative double Riemann integral, in the $N \to \infty$ limit it follows that

$$
\begin{aligned}
\left\langle \hat{O}_1 \overset{\leftrightarrow}{c} \hat{O}_2 \overset{\leftrightarrow}{c} \hat{O}_3 \right\rangle_N &= \frac{Z^{-1}}{\beta^2} \int_0^{\beta} d\lambda_0 \int_0^{\lambda_0} d\lambda_1 Tr\left[e^{-(\beta-\lambda_0)\hat{H}} \hat{O}_1 e^{-(\lambda_0-\lambda_1)\hat{H}} \hat{O}_2 e^{-\lambda_1 \hat{H}} \hat{O}_3 \right] \\
&\quad + \frac{Z^{-1}}{\beta^2} \int_0^{\beta} d\lambda_0 \int_{\lambda_0}^{\beta} d\lambda_1 Tr\left[e^{-(\beta-\lambda_1)\hat{H}} \hat{O}_2 e^{-(\lambda_1-\lambda_0)\hat{H}} \hat{O}_1 e^{-\lambda_0 \hat{H}} \hat{O}_3 \right] \\
&= \frac{1}{\beta^2} \int_0^{\beta} d\lambda_0 \int_0^{\lambda_0} d\lambda_1 \left\langle \hat{O}_1(-i\hbar\lambda_0) \hat{O}_2(-i\hbar\lambda_1) \hat{O}_3 \right\rangle \\
&\quad + \frac{1}{\beta^2} \int_0^{\beta} d\lambda_0 \int_{\lambda_0}^{\beta} d\lambda_1 \left\langle \hat{O}_2(-i\hbar\lambda_1) \hat{O}_1(-i\hbar\lambda_0) \hat{O}_3 \right\rangle \\
&= \frac{1}{\beta^2} \int_0^{\beta} d\lambda_0 \int_0^{\beta} d\lambda_1 \left\langle \hat{T}_\beta \hat{O}_1(-i\hbar\lambda_0) \hat{O}_2(-i\hbar\lambda_1) \hat{O}_3 \right\rangle \\
&= \left\langle \hat{O}_1 \bullet \hat{O}_2 \bullet \hat{O}_3 \right\rangle, \qquad (111)
\end{aligned}
$$

where we have introduced the imaginary time-ordering operator $\hat{T}_\beta$ to freely commute operators inside the integral sign. [6, 9] In other words, Cosine-Cosine correlation functions are exact ring-polymer phase space representations of (symmetrized) Double-Kubo transformed correlation functions[6, 9]. Note that for $\hat{O}_1 = \hat{A}(t_0)$, $\hat{O}_2 = \hat{B}(t_1)$, and $\hat{O}_3 = \hat{C}(t_2)$, one obtains the correlation function appearing in second-order response theory.

V. ADDITIONAL REFERENCES

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