Supporting Information: Ring-Polymer, Centroid and Mean-field Approximations to Multi-time Matsubara Dynamics

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CONTENTS

I. NON-CANONICAL CHANGE OF VARIABLES

By making the variable substitution

$$P_j \to \tilde{P}_j + im\omega_j Q_{-j} \tag{1}$$

and noting that

$$\omega_j = -\omega_{-j} \tag{2}$$

the Matsubara phase can be written as

$$\begin{aligned}
\theta_M(\boldsymbol{Q},\boldsymbol{P}) &= \sum_{j=-\bar{M}}^{\bar{M}} P_j\omega_j Q_{-j} \\
&= \sum_{j=-\bar{M}}^{\bar{M}} (\tilde{P}_j + im\omega_j Q_{-j})\omega_j Q_{-j} \\
&= \sum_{j=-\bar{M}}^{\bar{M}} (\tilde{P}_j\omega_j Q_{-j} + im\omega_j^2 Q_{-j}^2) \\
&= \sum_{j=-\bar{M}}^{\bar{M}} (\tilde{P}_j\omega_j Q_{-j} + im\omega_{-j}^2 Q_{-j}^2) \\
&= \theta_M(\boldsymbol{Q},\tilde{\boldsymbol{P}}) + 2iS_M(\boldsymbol{Q})
\end{aligned} \tag{3}$$

and the kinetic energy can be written as

$$\begin{aligned}
\frac{\boldsymbol{P}^2}{2m} &= \sum_{j=-\bar{M}}^{\bar{M}} \frac{P_j^2}{2m} \\
&= \sum_{j=-\bar{M}}^{\bar{M}} \frac{1}{2m}(\tilde{P}_j + im\omega_j Q_{-j})^2 \\
&= \sum_{j=-\bar{M}}^{\bar{M}} \left[\frac{\tilde{P}_j^2}{2m} + i\tilde{P}_j\omega_j Q_{-j} - \frac{m}{2}\omega_j^2 Q_{-j}^2\right] \\
&= \sum_{j=-\bar{M}}^{\bar{M}} \left[\frac{\tilde{P}_j^2}{2m} + i\tilde{P}_j\omega_j Q_{-j} - \frac{m}{2}\omega_{-j}^2 Q_{-j}^2\right] \\
&= \frac{\tilde{\boldsymbol{P}}^2}{2m} + i\theta_M(\boldsymbol{Q},\tilde{\boldsymbol{P}}) - S_M(\boldsymbol{Q})
\end{aligned} \tag{4}$$

Combining both results it follows that

$$\frac{\boldsymbol{P}^2}{2m} - i\theta_M(\boldsymbol{Q},\boldsymbol{P}) \to \frac{\tilde{\boldsymbol{P}}^2}{2m} + S_M(\boldsymbol{Q}) \tag{5}$$

and, therefore,

$$H_M(\boldsymbol{Q},\boldsymbol{P}) - i\theta_M(\boldsymbol{Q},\boldsymbol{P}) \to R_M(\boldsymbol{Q},\tilde{\boldsymbol{P}}) \tag{6}$$

Moreover, by noting that under the variable substitution

$$\begin{aligned}
\frac{\partial}{\partial Q_j} &\to \frac{\partial}{\partial Q_j} - im\omega_{-j}\frac{\partial}{\partial \tilde{P}_{-j}} \\
\frac{\partial}{\partial P_j} &\to \frac{\partial}{\partial \tilde{P}_j}
\end{aligned} \tag{7}$$

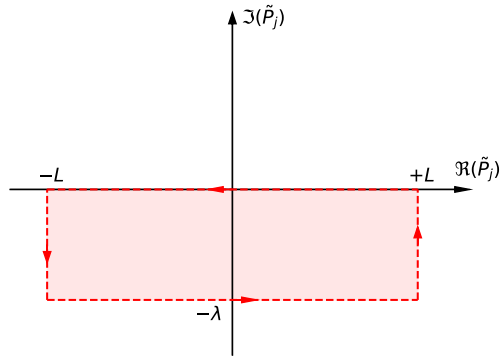

FIG. 1. Contour integration lines used for the analytic continuation in Eq. 10, with $L \to \infty$ and $\lambda_j = -im\omega_j Q_{-j}$.

the Matsubara Liovullian can be cast as

$$
\begin{aligned}
\mathcal{L}_M(\boldsymbol{Q},\boldsymbol{P}) &= \sum_{j=-\bar{M}}^{\bar{M}} \left[\frac{P_j}{m}\frac{\partial}{\partial Q_j} - \frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j}\frac{\partial}{\partial P_j} \right] \\
&= \sum_{j=-\bar{M}}^{\bar{M}} \left[\frac{1}{m}\left(\tilde{P}_j + im\omega_j Q_{-j}\right)\left(\frac{\partial}{\partial Q_j} - im\omega_{-j}\frac{\partial}{\partial \tilde{P}_{-j}}\right) - \frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j}\frac{\partial}{\partial \tilde{P}_j} \right] \\
&= \sum_{j=-\bar{M}}^{\bar{M}} \left[\frac{\tilde{P}_j}{m}\frac{\partial}{\partial Q_j} - i\omega_{-j}\tilde{P}_j\frac{\partial}{\partial \tilde{P}_{-j}} + i\omega_j Q_{-j}\frac{\partial}{\partial Q_j} + m\omega_j\omega_{-j}Q_{-j}\frac{\partial}{\partial \tilde{P}_{-j}} \right. \\
&\quad \left. - \frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j}\frac{\partial}{\partial \tilde{P}_j} \right] \\
&= \sum_{j=-\bar{M}}^{\bar{M}} \left[\frac{\tilde{P}_j}{m}\frac{\partial}{\partial Q_j} + i\omega_j\tilde{P}_j\frac{\partial}{\partial \tilde{P}_{-j}} - i\omega_j Q_j\frac{\partial}{\partial Q_{-j}} - m\omega_j^2 Q_j\frac{\partial}{\partial \tilde{P}_j} \right. \\
&\quad \left. - \frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j}\frac{\partial}{\partial \tilde{P}_j} \right] \\
&= \sum_{j=-\bar{M}}^{\bar{M}} \left[\frac{\tilde{P}_j}{m}\frac{\partial}{\partial Q_j} - \left(\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j} + m\omega_j^2 Q_j\right)\frac{\partial}{\partial \tilde{P}_j} \right] + \\
&\quad i\sum_{j=-\bar{M}}^{\bar{M}} \omega_j \left(\tilde{P}_j\frac{\partial}{\partial \tilde{P}_{-j}} - Q_j\frac{\partial}{\partial Q_{-j}} \right) \\
&\equiv \mathcal{L}_{RP}(\boldsymbol{Q},\tilde{\boldsymbol{P}}) + i\mathcal{L}_{\Im}(\boldsymbol{Q},\tilde{\boldsymbol{P}})
\end{aligned}
\tag{8}
$$

II. ANALYTIC CONTINUATION

After making the change of variables $P_j \to \tilde{P}_j + im\omega_j Q_{-j}$ for every Matsubara mode, the Matsubara approximation to the fully symmetrized (imaginary-time ordered) n-th order Kubo transformed multi-time correlation function can be exactly expressed as

$$K_M^{sym}(\boldsymbol{t}) = \frac{1}{(2\pi\hbar)^M Z_M} \int d\boldsymbol{Q} \prod_j \int_{-\infty - im\omega_j Q_{-j}}^{+\infty - im\omega_j Q_{-j}} d\tilde{P}_j \; e^{-\beta R_M(\boldsymbol{Q},\tilde{\boldsymbol{P}})} \times A_0(\boldsymbol{Q}) \prod_{k=1}^{n} e^{\mathcal{L}_M(\boldsymbol{Q},\tilde{\boldsymbol{P}})t_k} A_k(\boldsymbol{Q}). \tag{9}$$

Eq. 9 is an exact rewriting of Eq. 2 in the main text where the momenta $\tilde{P}_j$ are evaluated in the complex plane. However, since the integrals involved are of the form

$$\int_{-\infty-\lambda_j}^{+\infty-\lambda_j} d\tilde{P}_j f(\boldsymbol{Q},\tilde{\boldsymbol{P}},\boldsymbol{t}), \tag{10}$$

(with $\lambda_j = -im\omega_j Q_{-j}$) one can use the contours defined in Figure 1 to evaluate them. Provided that the function $f(\boldsymbol{Q},\tilde{\boldsymbol{P}},\boldsymbol{t})$ remains analytic (namely, free from singularities) inside the region (which is the case for any analytic Hamiltonian)[?] , by Cauchy's integral theorem the integral can be re-expressed as

$$\int_{-\infty-\lambda_j}^{+\infty-\lambda_j} d\tilde{P}_j f(\boldsymbol{Q},\tilde{\boldsymbol{P}},\boldsymbol{t}) = \int_{-\infty}^{+\infty} d\tilde{P}_j f(\boldsymbol{Q},\tilde{\boldsymbol{P}},\boldsymbol{t}) + \int_{+\infty}^{+\infty-\lambda_j} d\tilde{P}_j f(\boldsymbol{Q},\tilde{\boldsymbol{P}},\boldsymbol{t}) - \int_{-\infty}^{-\infty-\lambda_j} d\tilde{P}_j f(\boldsymbol{Q},\tilde{\boldsymbol{P}},\boldsymbol{t}) \tag{11}$$

The first term on the right hand side corresponds to the analytic continuation on the real axis, whereas the second and third represent the edges of the contour integration. These edge terms can be shown to be zero for a series of cases,[?] including the $t = 0$ limit, and therefore allow us to rewrite Eq. 9 as

$$\begin{aligned} K_M^{sym}(\boldsymbol{t}) &= \frac{1}{(2\pi\hbar)^M Z_M} \int d\boldsymbol{Q} \prod_j \int_{-\infty}^{+\infty} d\tilde{P}_j \; e^{-\beta R_M(\boldsymbol{Q},\tilde{\boldsymbol{P}})} A_0(\boldsymbol{Q}) \prod_{k=1}^{n} e^{\mathcal{L}_M(\boldsymbol{Q},\tilde{\boldsymbol{P}})t_k} A_k(\boldsymbol{Q}) \\ &= \frac{1}{(2\pi\hbar)^M Z_M} \int d\boldsymbol{Q} \int d\tilde{\boldsymbol{P}} \; e^{-\beta R_M(\boldsymbol{Q},\tilde{\boldsymbol{P}})} A_0(\boldsymbol{Q}) \prod_{k=1}^{n} e^{\mathcal{L}_M(\boldsymbol{Q},\tilde{\boldsymbol{P}})t_k} A_k(\boldsymbol{Q}). \end{aligned} \tag{12}$$

III. LIOUVILLIAN PROPERTIES

In this section we will summarize some of the properties of the Liovillian operators defined in the main text.

A. Ring-polymer Liouvillian

Action of the ring-polymer Liouvillian $\mathcal{L}_{RP}(\boldsymbol{Q},\boldsymbol{P})$

$$\mathcal{L}_{RP}(\boldsymbol{Q},\boldsymbol{P}) = \sum_{j=-\bar{M}}^{\bar{M}} \left[\frac{P_j}{m}\frac{\partial}{\partial Q_j} - \left(\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j} + m\omega_j^2 Q_j \right) \frac{\partial}{\partial P_j} \right], \tag{13}$$

on the centroid position Q_0 allows one to obtain the following relations

$$\mathcal{L}_{RP} Q_0 = \frac{P_0}{m}, \tag{14}$$

and

$$\begin{aligned} \mathcal{L}_{RP}^2 Q_0 &= \mathcal{L}_{RP}\left[\mathcal{L}_{RP} Q_0\right] \\ &= \mathcal{L}_{RP}\frac{P_0}{m} \\ &= -\frac{1}{m}\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_0}, \end{aligned} \tag{15}$$

and

$$\begin{aligned} \mathcal{L}_{RP}^3 Q_0 &= \mathcal{L}_{RP}\left[\mathcal{L}_{RP}^2 Q_0\right] \\ &= \mathcal{L}_{RP}\left(-\frac{1}{m}\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_0} \right) \\ &= -\frac{1}{m^2}\sum_{j=-\bar{M}}^{\bar{M}} P_j \frac{\partial^2 U_M(\boldsymbol{Q})}{\partial Q_j \partial Q_0}. \end{aligned} \tag{16}$$

Similarly the action of $\mathcal{L}_{RP}(\boldsymbol{Q},\boldsymbol{P})$ on the Matsubara potential gives

$$\mathcal{L}_{RP} U_M(\boldsymbol{Q}) = \sum_{j=-\bar{M}}^{\bar{M}} \frac{P_j}{m}\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j}, \tag{17}$$

and action on the ring-polymer spring potential gives

$$\begin{aligned} \mathcal{L}_{RP} S_M(\boldsymbol{Q}) &= \mathcal{L}_{RP}\left[\sum_{k=-\bar{M}}^{\bar{M}} \frac{1}{2} m\omega_k^2 Q_k^2 \right] \\ &= \sum_{j=-\bar{M}}^{\bar{M}} P_j \omega_j^2 Q_j. \end{aligned} \tag{18}$$

Finally, $\mathcal{L}_{RP}(\boldsymbol{Q},\boldsymbol{P})$ acting on the kinetic energy yields

$$\begin{aligned} \mathcal{L}_{RP}\frac{\boldsymbol{P}^2}{2m} &= \mathcal{L}_{RP}\left[\sum_{k=-\bar{M}}^{\bar{M}} \frac{{P_k}^2}{2m} \right] \\ &= \sum_{j=-\bar{M}}^{\bar{M}} \left[-\left(\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j} + m\omega_j^2 Q_j \right) \frac{P_j}{m} \right] \\ &= -\mathcal{L}_{RP}\left[U_M(\boldsymbol{Q}) + S_M(\boldsymbol{Q}) \right]. \end{aligned} \tag{19}$$

Combining the previous results it follows that

$$\mathcal{L}_{RP} R_M = \mathcal{L}_{RP} \left[\frac{\boldsymbol{P}^2}{2m} + S_M(\boldsymbol{Q}) + U_M(\boldsymbol{Q}) \right] = 0, \tag{20}$$

which implies that $\mathcal{L}_{RP}$ conserves the ring-polymer Boltzmann distribution $e^{-\beta R_M}$.

B. Imaginary Matsubara Liouvillian

Action of the imaginary Matsubara Liouvillian $\mathcal{L}_I(\boldsymbol{Q}, \boldsymbol{P})$

$$\mathcal{L}_I(\boldsymbol{Q}, \boldsymbol{P}) = \sum_{j=-\bar{M}}^{\bar{M}} \left[\omega_j \left(\tilde{P}_j \frac{\partial}{\partial \tilde{P}_{-j}} - Q_j \frac{\partial}{\partial Q_{-j}} \right) \right], \tag{21}$$

on the centroid position Q_0 gives

$$\mathcal{L}_I Q_0 = -\omega_0 Q_0 = 0, \tag{22}$$

and action on the centroid momenta P_0 gives

$$\mathcal{L}_I P_0 = \omega_0 P_0 = 0. \tag{23}$$

Additionally, it follows that

$$\begin{aligned}
\mathcal{L}_I \left[\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_0} \right] &= \sum_{j=-\bar{M}}^{\bar{M}} \left[\omega_j \left(P_j \frac{\partial}{\partial P_{-j}} - Q_j \frac{\partial}{\partial Q_{-j}} \right) \right] \left[\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_0} \right] \\
&= \sum_{j=-\bar{M}}^{\bar{M}} -\omega_j Q_j \frac{\partial^2 U_M(\boldsymbol{Q})}{\partial Q_{-j} \partial Q_0} \\
&= \frac{\partial}{\partial Q_0} \sum_{j=-\bar{M}}^{\bar{M}} -\omega_j Q_j \frac{\partial U_M(\boldsymbol{Q})}{\partial Q_{-j}} \\
&= \frac{\partial}{\partial Q_0} \sum_{j=-\bar{M}}^{\bar{M}} \omega_j Q_{-j} \frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j} \\
&= \frac{\partial}{\partial Q_0} \sum_{j=-\bar{M}}^{\bar{M}} -\frac{dQ_j}{d\tau} \frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j} \\
&= -\frac{\partial}{\partial Q_0} \frac{dU_M(\boldsymbol{Q})}{d\tau} = 0,
\end{aligned} \tag{24}$$

where we have used the fact that[?] $dQ_j/d\tau = -\omega_j Q_{-j}$ to obtain the next-to-last line and the last line follows from the imaginary time invariance (smooth nature) of the Matsubara potential.

$\mathcal{L}_I(\boldsymbol{Q},\boldsymbol{P})$ acting on the ring-polymer spring energy gives

$$\begin{aligned}
\mathcal{L}_I S_M(\boldsymbol{Q}) &= \mathcal{L}_I \left[\sum_{k=-\bar{M}}^{\bar{M}} \frac{1}{2} m\omega_k^2 Q_k^2\right] \\
&= \sum_{j=-\bar{M}}^{\bar{M}} \left[\omega_j \left(P_j \frac{\partial}{\partial P_{-j}} - Q_j \frac{\partial}{\partial Q_{-j}}\right)\right] \left[\sum_{k=-\bar{M}}^{\bar{M}} \frac{1}{2} m\omega_k^2 Q_k^2\right] \\
&= \sum_{j=-\bar{M}}^{\bar{M}} -m\omega_j \omega_{-j}^2 Q_j Q_{-j} \\
&= \sum_{j=-\bar{M}}^{\bar{M}} m\omega_{-j}^3 Q_j Q_{-j} = 0,
\end{aligned} \tag{25}$$

where the last line follows from symmetry about $j = 0$, i.e. the jth term will be canceled out by the $-j$th term (and $\omega_0 = 0$). Similarly, the imaginary Matsubara Liouvillian acting on the kinetic energy gives

$$\begin{aligned}
\mathcal{L}_I \frac{\boldsymbol{P}^2}{2m} &= \mathcal{L}_I \left[\sum_{k=-\bar{M}}^{\bar{M}} \frac{{P_k}^2}{2m}\right] \\
&= \sum_{j=-\bar{M}}^{\bar{M}} \left[\omega_j \left(P_j \frac{\partial}{\partial P_{-j}} - Q_j \frac{\partial}{\partial Q_{-j}}\right)\right] \left[\sum_{k=-\bar{M}}^{\bar{M}} \frac{{P_k}^2}{2m}\right] \\
&= \sum_{j=-\bar{M}}^{\bar{M}} \frac{\omega_j}{m} P_j P_{-j} = 0,
\end{aligned} \tag{26}$$

for the same reason as the previous case.

Additionally, $\mathcal{L}_I(\boldsymbol{Q},\boldsymbol{P})$ acting on the Matsubara potential gives

$$\begin{aligned}
\mathcal{L}_I U_M(\boldsymbol{Q}) &= \sum_{j=-\bar{M}}^{\bar{M}} \left[\omega_j \left(P_j \frac{\partial}{\partial P_{-j}} - Q_j \frac{\partial}{\partial Q_{-j}}\right)\right] U_M(\boldsymbol{Q}) \\
&= -\sum_{j=-\bar{M}}^{\bar{M}} \omega_j Q_j \frac{\partial U_M(\boldsymbol{Q})}{\partial Q_{-j}} \\
&= \sum_{j=-\bar{M}}^{\bar{M}} \omega_j Q_{-j} \frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j} \\
&= -\sum_{j=-\bar{M}}^{\bar{M}} \frac{dQ_j}{d\tau} \frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j} \\
&= -\frac{dU_M(\boldsymbol{Q})}{d\tau} = 0,
\end{aligned} \tag{27}$$

where we have used the fact that[?] $dQ_j/d\tau = -\omega_j Q_{-j}$ to obtain the next-to-last line and the last line follows from the imaginary time invariance (smooth nature) of the Matsubara potential. The steps used to obtain Eq. 27 can be generalized to show that the action of $\mathcal{L}_I$ on any permutationally invariant function of the form $f(\boldsymbol{Q})g(\boldsymbol{P})$ with $df/d\tau = dg/d\tau = 0$ will be zero.

Combining the previous results it follows that

$$\mathcal{L}_I R_M = \mathcal{L}_I \left[\frac{\boldsymbol{P}^2}{2m} + S_M(\boldsymbol{Q}) + U_M(\boldsymbol{Q}) \right] = 0, \tag{28}$$

which implies that $\mathcal{L}_I$ conserves the ring-polymer Boltzmann distribution $e^{-\beta R_M}$.

C. Matsubara Liouvillian

Action of the Matsubara Liouvillian $\mathcal{L}_M(\boldsymbol{Q},\boldsymbol{P}) = \mathcal{L}_{RP}(\boldsymbol{Q},\boldsymbol{P}) + i\mathcal{L}_I(\boldsymbol{Q},\boldsymbol{P})$ on the centroid position Q_0 gives

$$\begin{aligned} \mathcal{L}_M Q_0 &= \left[\mathcal{L}_{RP} + i\mathcal{L}_I\right] Q_0. \\ &= \mathcal{L}_{RP} Q_0 \end{aligned} \tag{29}$$

where we have used Eq. 22. Additionally,

$$\begin{aligned} \mathcal{L}_M^2 Q_0 &= \mathcal{L}_M \left(\mathcal{L}_M Q_0\right) \\ &= \mathcal{L}_M \left(\mathcal{L}_{RP} Q_0\right) \\ &= \left[\mathcal{L}_{RP} + i\mathcal{L}_I)\right] \left(\mathcal{L}_{RP} Q_0\right) \\ &= \mathcal{L}_{RP}^2 Q_0 + i\mathcal{L}_I \mathcal{L}_{RP} Q_0 \\ &= \mathcal{L}_{RP}^2 Q_0 + i\mathcal{L}_I \frac{P_0}{m} \\ &= \mathcal{L}_{RP}^2 Q_0, \end{aligned} \tag{30}$$

where we have used Eq. 14 and 23 to arrive at the final result. Also,

$$\begin{aligned} \mathcal{L}_M^3 Q_0 &= \mathcal{L}_M \left(\mathcal{L}_M^2 Q_0\right) \\ &= \mathcal{L}_M \left(\mathcal{L}_{RP}^2 Q_0\right) \\ &= \left[\mathcal{L}_{RP} + i\mathcal{L}_I\right] \left(\mathcal{L}_{RP}^2 Q_0\right) \\ &= \mathcal{L}_{RP}^3 Q_0 + i\mathcal{L}_I \mathcal{L}_{RP}^2 Q_0 \\ &= \mathcal{L}_{RP}^3 Q_0 + i\mathcal{L}_I \left[-\frac{1}{m} \frac{\partial U_M(\boldsymbol{Q})}{\partial Q_0} \right] \\ &= \mathcal{L}_{RP}^3 Q_0, \end{aligned} \tag{31}$$

where we have used Eqs. 15 and 24 to obtain the last result. Finally, it follows that

$$\begin{aligned}\mathcal{L}_M^4 Q_0 &= \mathcal{L}_M \left(\mathcal{L}_M^3 Q_0\right) \\ &= \mathcal{L}_M \left(\mathcal{L}_{RP}^3 Q_0\right) \\ &= \left[\mathcal{L}_{RP} + i\mathcal{L}_I\right] \left(\mathcal{L}_{RP}^3 Q_0\right) \\ &= \mathcal{L}_{RP}^4 Q_0 + i\mathcal{L}_I \mathcal{L}_{RP}^3 Q_0. \end{aligned} \tag{32}$$

D. Centroid Liouvillian

Action of the centroid Liouvillian $\mathcal{L}_C(Q_0, P_0)$

$$\mathcal{L}_C(Q_0, P_0) = \frac{P_0}{m}\frac{\partial}{\partial Q_0} - \frac{\partial W_0(Q_0)}{\partial Q_0}\frac{\partial}{\partial P_0}, \tag{33}$$

on the centroid position Q_0 gives

$$\mathcal{L}_C Q_0 = \frac{P_0}{m}, \tag{34}$$

and

$$\begin{aligned}\mathcal{L}_C^2 Q_0 &= \mathcal{L}_C \left(\mathcal{L}_C Q_0\right) \\ &= \mathcal{L}_C \frac{P_0}{m} \\ &= -\frac{1}{m}\frac{\partial W_0(Q_0)}{\partial Q_0}, \end{aligned} \tag{35}$$

and

$$\begin{aligned}\mathcal{L}_C^3 Q_0 &= \mathcal{L}_C \left(\mathcal{L}_C^2 Q_0\right) \\ &= \mathcal{L}_C \left(-\frac{1}{m}\frac{\partial W_0(Q_0)}{\partial Q_0}\right) \\ &= -\frac{P_0}{m^2}\frac{\partial^2 W_0(Q_0)}{\partial Q_0^2}. \end{aligned} \tag{36}$$

E. White-noise Fokker-Planck operator

Action of the white-noise Fokker-Planck operator

$$\mathcal{A}_{wn}(\boldsymbol{P}) = \nabla_{\boldsymbol{P}} \cdot \boldsymbol{\Gamma} \cdot \boldsymbol{P} + \frac{m}{\beta}\nabla_{\boldsymbol{P}} \cdot \boldsymbol{\Gamma} \cdot \nabla_{\boldsymbol{P}} \tag{37}$$

on the ring-polymer distribution $e^{-\beta R_M}$ gives

$$\begin{aligned}
\mathcal{A}_{wn} e^{-\beta R_M} &= \sum_{j,k=-\bar{M}}^{\bar{M}} \frac{\partial}{\partial P_j}\left[\Gamma_{jk} P_k e^{-\beta R_M}\right] + \frac{m}{\beta}\frac{\partial}{\partial P_j}\left[\Gamma_{ij}\frac{\partial}{\partial P_k}\left(e^{-\beta R_M}\right)\right] \\
&= \sum_{j,k=-\bar{M}}^{\bar{M}} \Gamma_{jk}\left[\delta_{jk} e^{-\beta R_M} + P_k e^{-\beta R_M}(-\beta\frac{P_j}{m})\right] + \frac{m}{\beta}\Gamma_{ij}\frac{\partial}{\partial P_j}\left[e^{-\beta R_M}(-\beta\frac{P_k}{m})\right] \\
&= \sum_{j,k=-\bar{M}}^{\bar{M}} \Gamma_{jk}\left[\delta_{jk} e^{-\beta R_M} + P_k e^{-\beta R_M}(-\beta\frac{P_j}{m})\right] - \Gamma_{ij}\left[\delta_{jk} e^{-\beta R_M} + P_k e^{-\beta R_M}(-\beta\frac{P_j}{m})\right] \\
&= 0
\end{aligned} \tag{38}$$

which proves that $\mathcal{A}_{wn}$ conserves the ring-polymer Boltzmann distribution.

IV. ERROR ANALYSIS FOR SINGLE-TIME CORRELATION FUNCTIONS

A. RPMD

For the case of single-time correlation functions, the general error term between RPMD and Matsubara dynamics become

$$f(l) = \left\langle A_0 A_1 A_2 \ldots \left(t_n^l \mathcal{L}^l A_n\right)\right\rangle_{RP} \tag{39}$$

for $l = 0, 1, 2, \ldots$ and $\mathcal{L} \equiv \mathcal{L}_{RP}$ or $\mathcal{L} \equiv \mathcal{L}_M$. Alternatively one can perform integration by parts along with making use of the fact that $\mathcal{L}_M e^{-\beta R_M} = \mathcal{L}_{RP} e^{-\beta R_M} = 0$ to rewrite Eq. 39 as

$$\begin{aligned}
f(l) = &-\left\langle \mathcal{L}^1 A_0 A_1 A_2 \ldots \left(t_n^l \mathcal{L}^{l-1} A_n\right)\right\rangle_{RP} \\
&-\left\langle A_0 \mathcal{L}^1 A_1 A_2 \ldots \left(t_n^l \mathcal{L}^{l-1} A_n\right)\right\rangle_{RP} \\
&-\left\langle A_0 A_1 \mathcal{L}^1 A_2 \ldots \left(t_n^l \mathcal{L}^{l-1} A_n\right)\right\rangle_{RP} \\
&- \ldots
\end{aligned} \tag{40}$$

This last expression shows that one can "redistribute" the Liouvillian associated with A_n (therefore reducing it order) to all other observables $A_{k\neq n}$. Recursive applications of integration by parts allows one to obtain similar expressions to Eq. 40 where additional reductions $l-2, l-3, \ldots$ can be performed albeit at an increase in the number of combinations of

terms. Armed with Eqs. 39 and 40, the short-time error of RPMD can be determined by looking for the first term in which $\mathcal{L}_M^l A_k \neq \mathcal{L}_{RP}^l A_k$.

For linear operators, since $\mathcal{L}_M^l Q_0 = \mathcal{L}_{RP}^l Q_0$ for $l \leq 3$, a straightforward analysis of Eq. 39 demonstrates that RPMD agrees with Matsubara up to order t^3. For $l \geq 4$, $\mathcal{L}_M^l Q_0 \neq \mathcal{L}_{RP}^l Q_0$. However, one can reduce the Liouvillian order in A_n by using Eq. 40 (up to three times), to show that for $l = 4, 5, 6$ RPMD and Matsubara agrees up to t^6.

For the case of nonlinear operators the disagreement between the RPMD and Matsubara Liouvillian occurs at $l = 2$. A similar analysis of Eqs. 39 and 40 reveals that RPMD TCF agrees up to order t^2 with Matsubara dynamics.

B. CMD

For the case of single-time correlation functions of centroid- dependent only operators (i.e. linear operators) the general error term between CMD and Matsubara dynamics becomes

$$f(l) = \left\langle A_0(Q_0) A_1(Q_0) A_2(Q_0) \ldots \left(t_n^l \mathcal{L}^l A_n(Q_0)\right)\right\rangle_{RP} \tag{41}$$

for $l = 0, 1, 2, \ldots$. In the previous equation, $A_k(Q_0) \equiv Q_0$, but we use the A_k notation to emphasize that a higher-order Kubo transform is being evaluated albeit all but one operator is time-independent.

Since $\mathcal{L}_M Q_0 = \mathcal{L}_c Q_0$, it is obvious to see that CMD agrees with Matsubara dynamics for $l = 1$.

For $l = 2$, $\mathcal{L}_M^2 Q_0 \neq \mathcal{L}_c^2 Q_0$. However, the non-centroid modes of the Matsubara Liouvillian

can be integrated out from Eq. 41 to give

$$
\begin{aligned}
f(2) &\sim \frac{1}{(2\pi\hbar)^M Z_M} \int d\boldsymbol{Q} \int d\tilde{\boldsymbol{P}}\; e^{-\beta R_M(\boldsymbol{Q},\tilde{\boldsymbol{P}})} \left[\prod_{k=0}^{n-1} A_k(Q_0)\right] \left(\mathcal{L}_M^2 A_n(Q_0)\right) \\
&\sim \frac{1}{(2\pi\hbar)^M Z_M} \int d\boldsymbol{Q} \int d\tilde{\boldsymbol{P}}\; e^{-\beta R_M(\boldsymbol{Q},\tilde{\boldsymbol{P}})} \left[\prod_{k=0}^{n-1} A_k(Q_0)\right] \left(-\frac{1}{m}\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_0}\right) \\
&\sim \frac{1}{(2\pi\hbar)^M Z_M} \int dQ_0 \int dP_0 \left[\prod_{k=0}^{n-1} A_k(Q_0)\right] \int d\boldsymbol{Q}' \int d\tilde{\boldsymbol{P}}'\; e^{-\beta R_M(\boldsymbol{Q},\tilde{\boldsymbol{P}})} \left(-\frac{1}{m}\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_0}\right) \\
&\sim \frac{1}{(2\pi\hbar)^M Z_M} \int dQ_0 \int dP_0 \left[\prod_{k=0}^{n-1} A_k(Q_0)\right] \int d\boldsymbol{Q}' \int d\boldsymbol{P}'\; \frac{1}{m\beta}\frac{\partial}{\partial Q_0} \left(e^{-\beta R_M(\boldsymbol{Q},\tilde{\boldsymbol{P}})}\right) \\
&\sim \frac{1}{(2\pi\hbar) Z_M} \int dQ_0 \int dP_0 \left[\prod_{k=0}^{n-1} A_k(Q_0)\right] \frac{1}{m\beta}\frac{\partial}{\partial Q_0} Z_0(Q_0, P_0) \\
&\sim \frac{1}{(2\pi\hbar) Z_M} \int dQ_0 \int dP_0 \left[\prod_{k=0}^{n-1} A_k(Q_0)\right] \frac{1}{m\beta}\frac{\partial}{\partial Q_0} e^{-\beta\left(\frac{P_0^2}{2m}+W_0(Q_0)\right)} \\
&\sim \frac{1}{(2\pi\hbar) Z_M} \int dQ_0 \int dP_0 \left[\prod_{k=0}^{n-1} A_k(Q_0)\right] e^{-\left(\frac{P_0^2}{2m}+W_0(Q_0)\right)} \left(-\frac{1}{m}\frac{\partial W_0(Q_0)}{\partial Q_0}\right) \\
&\sim \frac{1}{(2\pi\hbar)} \int dQ_0 \int dP_0\; \rho_c \left[\prod_{k=0}^{n-1} A_k(Q_0)\right] \left(\mathcal{L}_c^2 A_n(Q_0)\right)
\end{aligned}
\tag{42}
$$

where we have used Eqs. 30 and 15 to obtain the second line, the definition of Z_0 in terms of the potential of mean force W_0 in the sixth line and Eq. 35 to obtain the last line, demonstrating that CMD agrees with Matsubara dynamics for single-TCFs for $l = 2$. We remark that the key step in the derivation is the passage from the third to fourth line. Note that in the case of multi-TCF, the term $\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_0}$ will appear elevated to a power different than one which will invalidate the derivation, demonstrating the disagreement between CMD and Matsubara dynamics for $l = 2$.

Showing that CMD and Matsubara dynamics agrees for the case of $l = 3$ is trivial if one realizes that both $\mathcal{L}_M^3 Q_0$ (Eqs. 31 and 16) and $\mathcal{L}_c^3 Q_0$ (Eq. 36) give a function that is linear in P_j. Therefore, performing the Gaussian integrals over the momenta in Eq. 41 gives $f(3) = 0$ for both CMD and Matsubara dynamics.

V. MEAN-FIELD AVERAGE

A. Mean-Field Partition Function

The mean-field partition function is given by

$$
\begin{aligned}
Z_{MF}(\boldsymbol{Q}_N, \boldsymbol{P}_N) &= \frac{1}{(2\pi\hbar)^{M-N}} \int d\boldsymbol{Q}' \int d\boldsymbol{P}' \; e^{-\beta[H_M(\boldsymbol{Q},\boldsymbol{P}) - i\theta_M(\boldsymbol{Q},\boldsymbol{P})]} \\
&= \frac{1}{(2\pi\hbar)^{M-N}} \int d\boldsymbol{Q}' \int d\boldsymbol{P}' \; e^{-\beta\left[\frac{\boldsymbol{P}^2}{2m} + U_M(\boldsymbol{Q}) - i\theta_M(\boldsymbol{Q},\boldsymbol{P})\right]} \\
&= \frac{e^{-\beta\frac{\boldsymbol{P}_N^2}{2m}} e^{i\beta\theta_M(\boldsymbol{Q}_N,\boldsymbol{P}_N)}}{(2\pi\hbar)^{M-N}} \int d\boldsymbol{Q}' \int d\boldsymbol{P}' \; e^{-\beta\left[\frac{\boldsymbol{P}'^2}{2m} + U_M(\boldsymbol{Q}) - i\theta_M(\boldsymbol{Q}',\boldsymbol{P}')\right]} \\
&= \frac{e^{-\beta\frac{\boldsymbol{P}_N^2}{2m}} e^{i\beta\theta_M(\boldsymbol{Q}_N,\boldsymbol{P}_N)}}{(2\pi\hbar)^{M-N}} \int d\boldsymbol{Q}' \int d\tilde{\boldsymbol{P}}' \; e^{-\beta\left[\frac{\tilde{\boldsymbol{P}}'^2}{2m} + U_M(\boldsymbol{Q}) + S_M(\boldsymbol{Q}) - S_M(\boldsymbol{Q}_N)\right]} \\
&= e^{-\beta\frac{\boldsymbol{P}_N^2}{2m}} e^{i\beta\theta_M(\boldsymbol{Q}_N,\boldsymbol{P}_N)} \left(\frac{m}{2\pi\beta\hbar^2}\right)^{\frac{M-N}{2}} \int d\boldsymbol{Q}' \; e^{-\beta[U_M(\boldsymbol{Q}) + S_M(\boldsymbol{Q}) - S_M(\boldsymbol{Q}_N)]} \\
&= e^{-\beta\frac{\boldsymbol{P}_N^2}{2m}} e^{i\beta\theta_M(\boldsymbol{Q}_N,\boldsymbol{P}_N)} e^{-\beta W_{MF}(\boldsymbol{Q}_N)}
\end{aligned} \tag{43}
$$

where we have singled out the contribution of the N lowest Matsubara modes to the kinetic energy and phase factor in the third line, we have performed the change of variables $P_j \to \tilde{P}_j + im\omega_j Q_{-j}$ and used the contour-integration trick for the mean-field modes in the fourth line (recognizing that $\frac{\boldsymbol{P}'^2}{2m} - i\theta_M(\boldsymbol{Q}', \boldsymbol{P}') \to \frac{\tilde{\boldsymbol{P}}'^2}{2m} + S_M(\boldsymbol{Q}) - S_M(\boldsymbol{Q}_N)$), we have performed the Gaussian integrals over the $\tilde{\boldsymbol{P}}'$ modes in the fifth line, and we used the definition of potential of mean-force in the last line.

For the case of the centroid mean-field average, noting that $\omega_0 = 0$, Eq. 43 reduces to

$$
\begin{aligned}
Z_0(Q_0, P_0) &= \frac{e^{-\beta\frac{P_0^2}{2m}}}{(2\pi\hbar)^{M-1}} \int d\boldsymbol{Q}' \int d\tilde{\boldsymbol{P}}' \; e^{-\beta\left[\frac{\tilde{\boldsymbol{P}}'^2}{2m} + U_M(\boldsymbol{Q}) + S_M(\boldsymbol{Q})\right]} \\
&= e^{-\beta\frac{P_0^2}{2m}} \left(\frac{m}{2\pi\beta\hbar^2}\right)^{\frac{M-1}{2}} \int d\boldsymbol{Q}' \; e^{-\beta[U_M(\boldsymbol{Q}) + S_M(\boldsymbol{Q})]} \\
&= e^{-\beta\frac{P_0^2}{2m}} e^{-\beta W_0(Q_0)}.
\end{aligned} \tag{44}
$$

with W_0 the centroid potential of mean-force.

Note that the Matsubara partition function can be expressed in terms of mean-field

variables as

$$
\begin{aligned}
Z_M &= \frac{1}{(2\pi\hbar)^M}\int d\boldsymbol{Q}\int d\boldsymbol{P}\; e^{-\beta[H_M(\boldsymbol{Q},\boldsymbol{P})-i\theta_M(\boldsymbol{Q},\boldsymbol{P})]} \\
&= \frac{1}{(2\pi\hbar)^M}\int d\boldsymbol{Q}_N\int d\boldsymbol{P}_N\int d\boldsymbol{Q}'\int d\boldsymbol{P}'\; e^{-\beta[H_M(\boldsymbol{Q},\boldsymbol{P})-i\theta_M(\boldsymbol{Q},\boldsymbol{P})]} \\
&= \frac{1}{(2\pi\hbar)^N}\int d\boldsymbol{Q}_N\int d\boldsymbol{P}_N\; Z_{MF}(\boldsymbol{Q},\boldsymbol{P}) \\
&= \frac{1}{(2\pi\hbar)^N}\int d\boldsymbol{Q}_N\int d\boldsymbol{P}_N\; e^{-\beta\left[\frac{\boldsymbol{P}_N^2}{2m}-i\theta_M(\boldsymbol{Q}_N,\boldsymbol{P}_N)+W_{MF}(\boldsymbol{Q}_N)\right]} \\
&= \frac{1}{(2\pi\hbar)^N}\int d\boldsymbol{Q}_N\int d\tilde{\boldsymbol{P}}_N\; e^{-\beta\left[\frac{\tilde{\boldsymbol{P}}_N^2}{2m}+W_{MF}(\boldsymbol{Q}_N)\right]}e^{-\beta\sum_{j=-\bar{N}}^{\bar{N}}\frac{1}{2}m\omega_j^2Q_j^2}
\end{aligned}
\tag{45}
$$

where we have performed the change of variables $P_j \to \tilde{P}_j + im\omega_j Q_{-j}$ and used the contour-integration trick for the $\boldsymbol{P}_N$ modes in the last line, recognizing that $\frac{\boldsymbol{P}_N^2}{2m} - i\theta_M(\boldsymbol{Q}_N,\boldsymbol{P}_N) \to \frac{\tilde{\boldsymbol{P}}_N^2}{2m} + \sum_{j=-\bar{N}}^{\bar{N}}\frac{1}{2}m\omega_j^2Q_j^2$.

B. Mean-Field Liouvillian

The mean-field Liouvillian can be obtained by performing the mean-field average

$$
\mathcal{L}_{MF}(\boldsymbol{Q}_N,\boldsymbol{P}_N) = \frac{1}{(2\pi\hbar)^{M-N}Z_{MF}}\int d\boldsymbol{Q}'\int d\boldsymbol{P}'\; e^{-\beta[H_M(\boldsymbol{Q},\boldsymbol{P})-i\theta_M(\boldsymbol{Q},\boldsymbol{P})]}\mathcal{L}_M(\boldsymbol{Q},\boldsymbol{P}), \tag{46}
$$

on the Matsubara Liouvillian

$$
\mathcal{L}_M(\boldsymbol{Q},\boldsymbol{P}) = \sum_{j=-\bar{M}}^{\bar{M}}\mathcal{L}_j(\boldsymbol{Q},\boldsymbol{P}), \tag{47}
$$

where

$$
\mathcal{L}_j(\boldsymbol{Q},\boldsymbol{P}) = \frac{P_j}{m}\frac{\partial}{\partial Q_j} - \frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j}\frac{\partial}{\partial P_j}, \tag{48}
$$

represents the terms involving derivatives with respect to the k-th Matsubara normal mode. Therefore, the centroid mean-field average involves evaluating integrals of the form

$$
\langle P_j\rangle_{MF} \equiv \frac{1}{(2\pi\hbar)^{M-N}Z_{MF}}\int d\boldsymbol{Q}'\int d\boldsymbol{P}'\; e^{-\beta[H_M(\boldsymbol{Q},\boldsymbol{P})-i\theta_M(\boldsymbol{Q},\boldsymbol{P})]}P_j, \tag{49}
$$

and

$$
\left\langle\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j}\right\rangle_{MF} \equiv \frac{1}{(2\pi\hbar)^{M-N}Z_{MF}}\int d\boldsymbol{Q}'\int d\boldsymbol{P}'\; e^{-\beta[H_M(\boldsymbol{Q},\boldsymbol{P})-i\theta_M(\boldsymbol{Q},\boldsymbol{P})]}\left(\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j}\right), \tag{50}
$$

where we have introduced $\langle\cdot\rangle_{MF}$ as a shorthand notation for the mean-field average.

Averages of the form Eq. 49 can be evaluated directly. For a normal mode that has not been integrated out, namely $j \leq \bar{N}$, it is straightforward to show that

$$\begin{aligned}\langle P_j\rangle_{MF} &= \frac{1}{(2\pi\hbar)^{M-N}Z_{MF}}\int d\boldsymbol{Q}'\int d\boldsymbol{P}'\ e^{-\beta[H_M(\boldsymbol{Q},\boldsymbol{P})-i\theta_M(\boldsymbol{Q},\boldsymbol{P})]}P_j \\ &= P_j\frac{1}{(2\pi\hbar)^{M-N}Z_{MF}}\int d\boldsymbol{Q}'\int d\boldsymbol{P}'\ e^{-\beta[H_M(\boldsymbol{Q},\boldsymbol{P})-i\theta_M(\boldsymbol{Q},\boldsymbol{P})]} \\ &= P_j, \end{aligned} \tag{51}$$

where we have used the definition of Z_{MF} in the last step. For normal modes being integrated out, namely $j > \bar{N}$, it is straightforward to realize that $\langle P_j\rangle_{MF} = 0$ by noting that the integrals involve terms of the form

$$\begin{aligned}\langle P_j\rangle_{MF} &\sim \int dP_j\ e^{-\beta\left[\frac{P_j^2}{2m}-iP_j\omega_jQ_{-j}\right]}P_j \\ &= 0. \end{aligned} \tag{52}$$

Averages of the form Eq. 50 can be evaluate by noting that

$$\begin{aligned}\frac{\partial e^{-\beta[H_M-i\theta_M]}}{\partial Q_j} &= -\beta e^{-\beta[H_M-i\theta_M]}\left[\frac{\partial H_M}{\partial Q_j}-i\frac{\partial\theta_M}{\partial Q_j}\right] \\ &= -\beta e^{-\beta[H_M-i\theta_M]}\left[\frac{\partial U_M}{\partial Q_j}-i\omega_{-j}P_{-j}\right], \end{aligned} \tag{53}$$

For normal modes with $j < \bar{N}$, it is straightforward to show that

$$\begin{aligned}\left\langle -\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j}\right\rangle_{MF} &= \frac{1}{(2\pi\hbar)^{M-N}Z_{MF}}\int d\boldsymbol{Q}'\int d\boldsymbol{P}'\ e^{-\beta[H_M(\boldsymbol{Q},\boldsymbol{P})-i\theta_M(\boldsymbol{Q},\boldsymbol{P})]}\left(-\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j}\right) \\ &= \frac{1}{(2\pi\hbar)^{M-N}Z_{MF}}\int d\boldsymbol{Q}'\int d\boldsymbol{P}'\left[\frac{1}{\beta}\frac{\partial}{\partial Q_j}\left(e^{-\beta[H_M-i\theta_M]}\right)-i\omega_{-j}P_{-j}e^{-\beta[H_M-i\theta_M]}\right] \\ &= \frac{1}{\beta Z_{MF}}\frac{\partial Z_{MF}}{\partial Q_j}-i\omega_{-j}P_{-j} \\ &= \frac{1}{\beta}\frac{\partial\ \ln(Z_{MF})}{\partial Q_j}-i\omega_{-j}P_{-j} \\ &= -\frac{\partial W_{MF}(\boldsymbol{Q}_N)}{\partial Q_j}, \end{aligned} \tag{54}$$

where we have used the identity in Eq. 53 in the second line, and the definition of Z_{MF} in terms of the potential of mean force W_{MF} to obtain the last line. For the case of modes

with $j > \bar{N}$, a similar analysis gives

$$\begin{aligned}
\left\langle -\frac{\partial U_M(\boldsymbol{Q})}{\partial Q_j} \right\rangle_{MF} &= \frac{1}{(2\pi\hbar)^{M-N} Z_{MF}} \int d\boldsymbol{Q}' \int d\boldsymbol{P}' \; e^{-\beta[H_M(\boldsymbol{Q},\boldsymbol{P}) - i\theta_M(\boldsymbol{Q},\boldsymbol{P})]} \left(-\frac{\partial U_M}{\partial Q_j} \right) \\
&= \frac{1}{(2\pi\hbar)^{M-N} Z_{MF}} \int d\boldsymbol{Q}' \int d\boldsymbol{P}' \; e^{-\beta[H_M(\boldsymbol{Q},\boldsymbol{P}) - i\theta_M(\boldsymbol{Q},\boldsymbol{P})]} \left(-i\omega_{-j} P_{-j} \right) \\
&= -i\omega_{-j} \left\langle P_{-j} \right\rangle_{MF} \\
&= 0,
\end{aligned} \tag{55}$$

where we have again used the identity in Eq. 53 (noting that the partial derivative of $e^{-\beta(H_M - i\theta_M)}$ with respect to P_j can be integrated out) in the second line, and Eq. 52 to obtain the last line.

Combining the above results, the mean-field Liouvillian can therefore be written as

$$\begin{aligned}
\mathcal{L}_{MF}(\boldsymbol{Q}_N, \boldsymbol{P}_N) &= \sum_{j=-\bar{N}}^{\bar{N}} \left\langle \mathcal{L}_j \right\rangle_{MF} \\
&= \sum_{j=-\bar{N}}^{\bar{N}} \frac{P_j}{m} \frac{\partial}{\partial Q_j} - \frac{\partial W_{MF}(\boldsymbol{Q}_N)}{\partial Q_j} \frac{\partial}{\partial P_j}.
\end{aligned} \tag{56}$$

Note that for the case of centroid mean-field averages, the centroid Liouvillian is obtained

$$\mathcal{L}_c(Q_0, P_0) = \frac{P_0}{m} \frac{\partial}{\partial Q_0} - \frac{\partial W_0(Q_0)}{\partial Q_0} \frac{\partial}{\partial P_0}. \tag{57}$$

C. Mean-field Multi-Time Correlation Functions

For operators that only depends on the $\boldsymbol{Q}_N$ modes, by replacing the Matsubara Liouvillian with the mean-field Liouvillian (Eq. 56), the following approximation to the multi-time

correlation function can be obtained

$$
\begin{aligned}
K_{MF}^{sym}(\boldsymbol{t}) = & \frac{1}{(2\pi\hbar)^M Z_M} \int d\boldsymbol{Q} \int d\boldsymbol{P}\ e^{-\beta[H_M(\boldsymbol{Q},\boldsymbol{P})-i\theta_M(\boldsymbol{Q},\boldsymbol{P})]} A_0(\boldsymbol{Q}_N) \prod_{k=1}^{n} e^{\mathcal{L}_{MF}(\boldsymbol{Q}_N,\boldsymbol{P}_N)t_k} A_k(\boldsymbol{Q}_N) \\
= & \frac{1}{(2\pi\hbar)^M Z_M} \int d\boldsymbol{Q}_N \int d\boldsymbol{P}_N\ A_0(\boldsymbol{Q}_N) \prod_{k=1}^{n} e^{\mathcal{L}_{MF}(\boldsymbol{Q}_N,\boldsymbol{P}_N)t_k} A_k(\boldsymbol{Q}_N) \times \\
& \times \int d\boldsymbol{Q}' \int d\boldsymbol{P}'\ e^{-\beta[H_M(\boldsymbol{Q},\boldsymbol{P})-i\theta_M(\boldsymbol{Q},\boldsymbol{P})]} \\
= & \frac{1}{(2\pi\hbar)^N Z_M} \int d\boldsymbol{Q}_N \int d\boldsymbol{P}_N\ A_0(\boldsymbol{Q}_N) \prod_{k=1}^{n} e^{\mathcal{L}_{MF}(\boldsymbol{Q}_N,\boldsymbol{P}_N)t_k} A_k(\boldsymbol{Q}_N) Z_{MF}(\boldsymbol{Q}_N, \boldsymbol{P}_N) \\
= & \frac{1}{(2\pi\hbar)^N Z_M} \int d\boldsymbol{Q}_N \int d\boldsymbol{P}_N\ e^{-\beta\left[\frac{\boldsymbol{P}_N^2}{2m}+W_{MF}(\boldsymbol{Q}_N)-i\theta_M(\boldsymbol{Q}_N,\boldsymbol{P}_N)\right]} \\
& \times A_0(\boldsymbol{Q}_N) \prod_{k=1}^{n} e^{\mathcal{L}_{MF}(\boldsymbol{Q}_N,\boldsymbol{P}_N)t_k} A_k(\boldsymbol{Q}_N). \qquad (58)
\end{aligned}
$$

D. Centroid Molecular Dynamics

By replacing the Matsubara Liouvillian with the centroid Liouvillian, the CMD approximation is obtained

$$
\begin{aligned}
K_{CMD}^{sym}(\boldsymbol{t}) = & \frac{1}{(2\pi\hbar)^M Z_M} \int d\boldsymbol{Q} \int d\boldsymbol{P}\ e^{-\beta[H_M(\boldsymbol{Q},\boldsymbol{P})-i\theta_M(\boldsymbol{Q},\boldsymbol{P})]} A_0(Q_0) \prod_{k=1}^{n} e^{\mathcal{L}_c(Q_0,P_0)t_k} A_k(Q_0). \\
= & \frac{1}{(2\pi\hbar)^M Z_M} \int d\boldsymbol{Q} \int d\tilde{\boldsymbol{P}}\ e^{-\beta R_M(\boldsymbol{Q},\tilde{\boldsymbol{P}})} A_0(Q_0) \prod_{k=1}^{n} e^{\mathcal{L}_c(Q_0,P_0)t_k} A_k(Q_0). \\
= & \frac{1}{2\pi\hbar Z_M} \int dQ_0 \int dP_0\ Z_0(Q_0, P_0) A_0(Q_0) \prod_{k=1}^{n} e^{\mathcal{L}_c(Q_0,P_0)t_k} A_k(Q_0) \\
= & \frac{1}{2\pi\hbar Z_M} \int dQ_0 \int dP_0\ e^{-\beta\left[\frac{P_0^2}{2m}+W_0(Q_0)\right]} A_0(Q_0) \prod_{k=1}^{n} e^{\mathcal{L}_c(Q_0,P_0)t_k} A_k(Q_0) \qquad (59)
\end{aligned}
$$

where we have used the contour-integration trick to obtain the second line (note that Q_0 is not affected by the variable transformation), and the definition of Z_0 in terms of the potential of mean force W_0 to obtain the last lines.

VI. ADDITIONAL NUMERICAL RESULTS

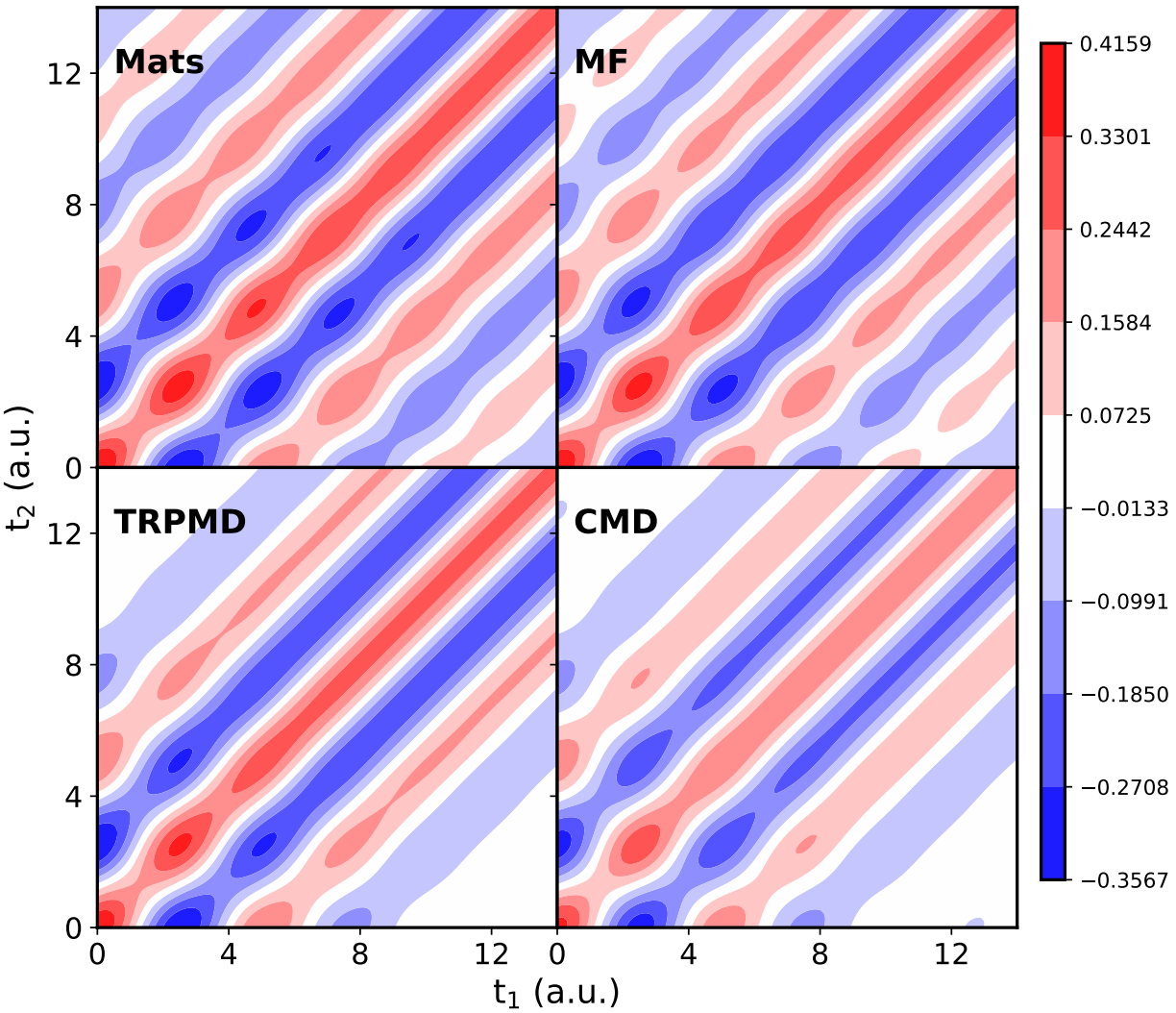

FIG. 2. Contour plots of the symmetrized double Kubo transformed $\langle \hat{q}^2 \hat{q}(t_1)\hat{q}(t_2)\rangle$ correlation function, for the quartic potential at $\beta = 2$, at different levels of theory.

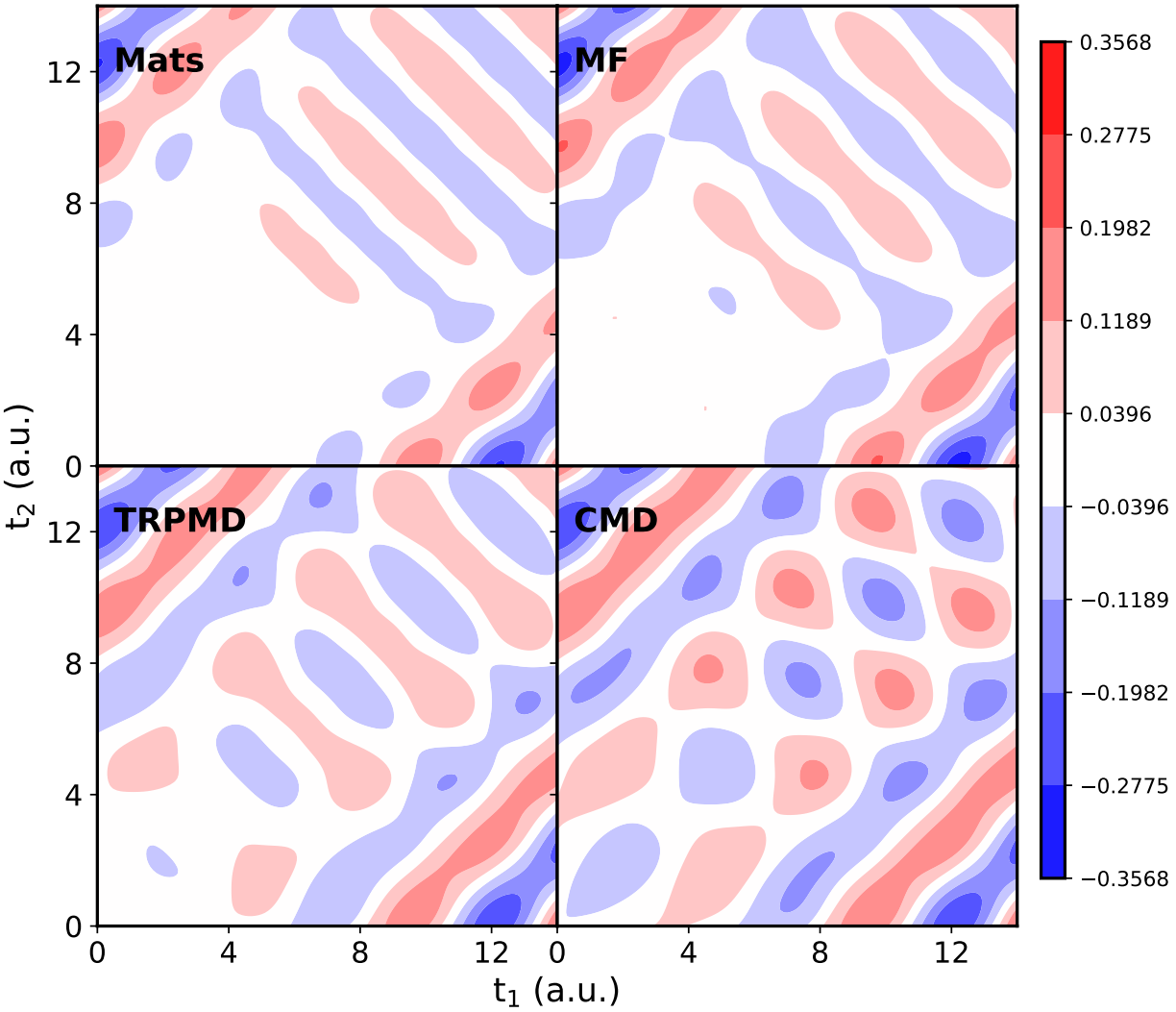

FIG. 3. Contour plots of the error between exact and approximated symmetrized double Kubo transformed $\left\langle \hat{q}^2 \hat{q}(t_1)\hat{q}(t_2) \right\rangle$ correlation function, for the quartic potential at $\beta = 2$, at different levels of theory.

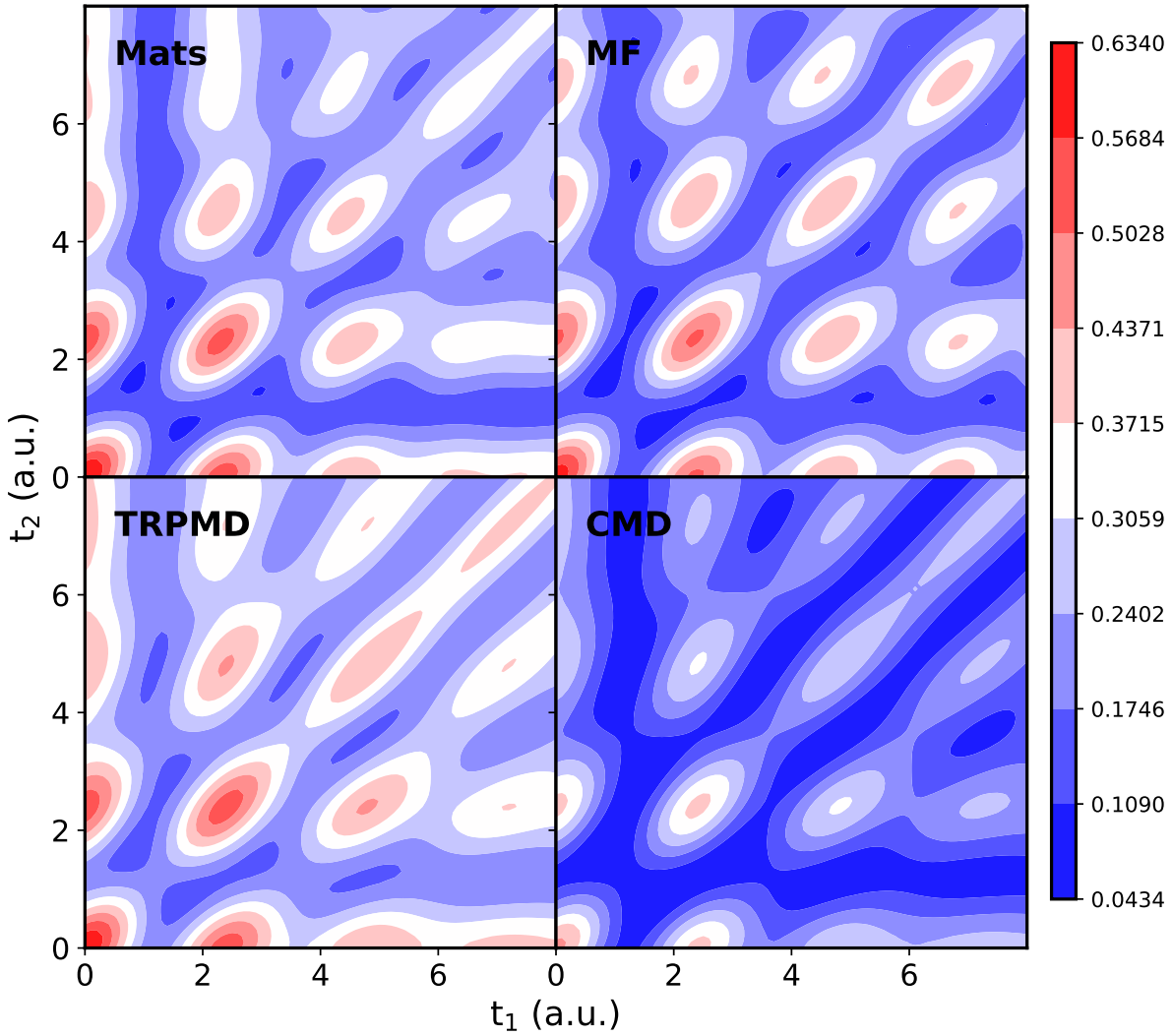

FIG. 4. Contour plots of symmetrized double Kubo transformed $\langle \hat{q}^2 \hat{q}^2(t_1) \hat{q}^2(t_2) \rangle$ correlation function, for the quartic potential at $\beta = 2$, at different levels of theory.

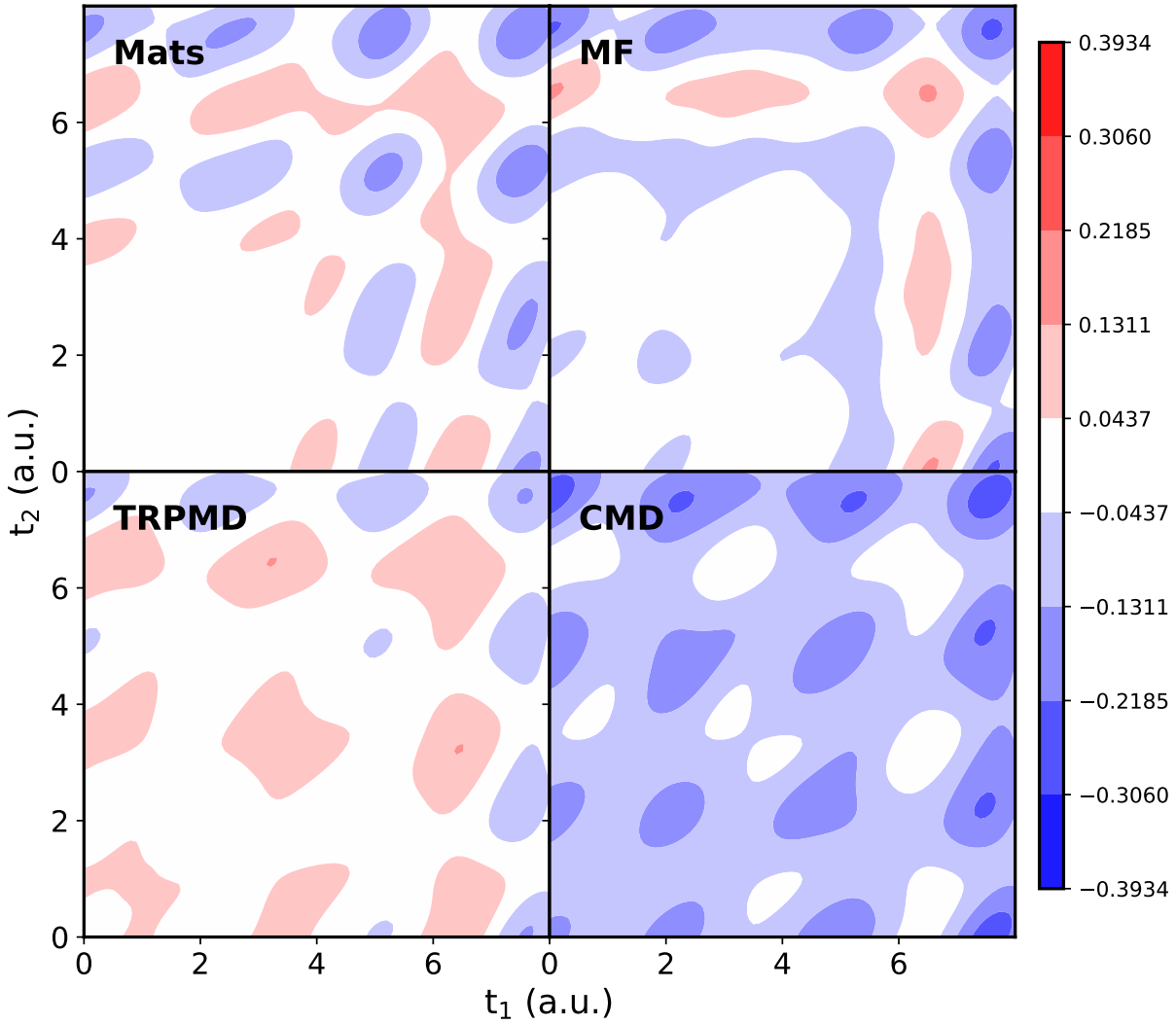

FIG. 5. Contour plots of the error between exact and approximated symmetrized double Kubo transformed $\langle \hat{q}^2 \hat{q}^2(t_1) \hat{q}^2(t_2) \rangle$ correlation function, for the quartic potential at $\beta = 2$, at different levels of theory.

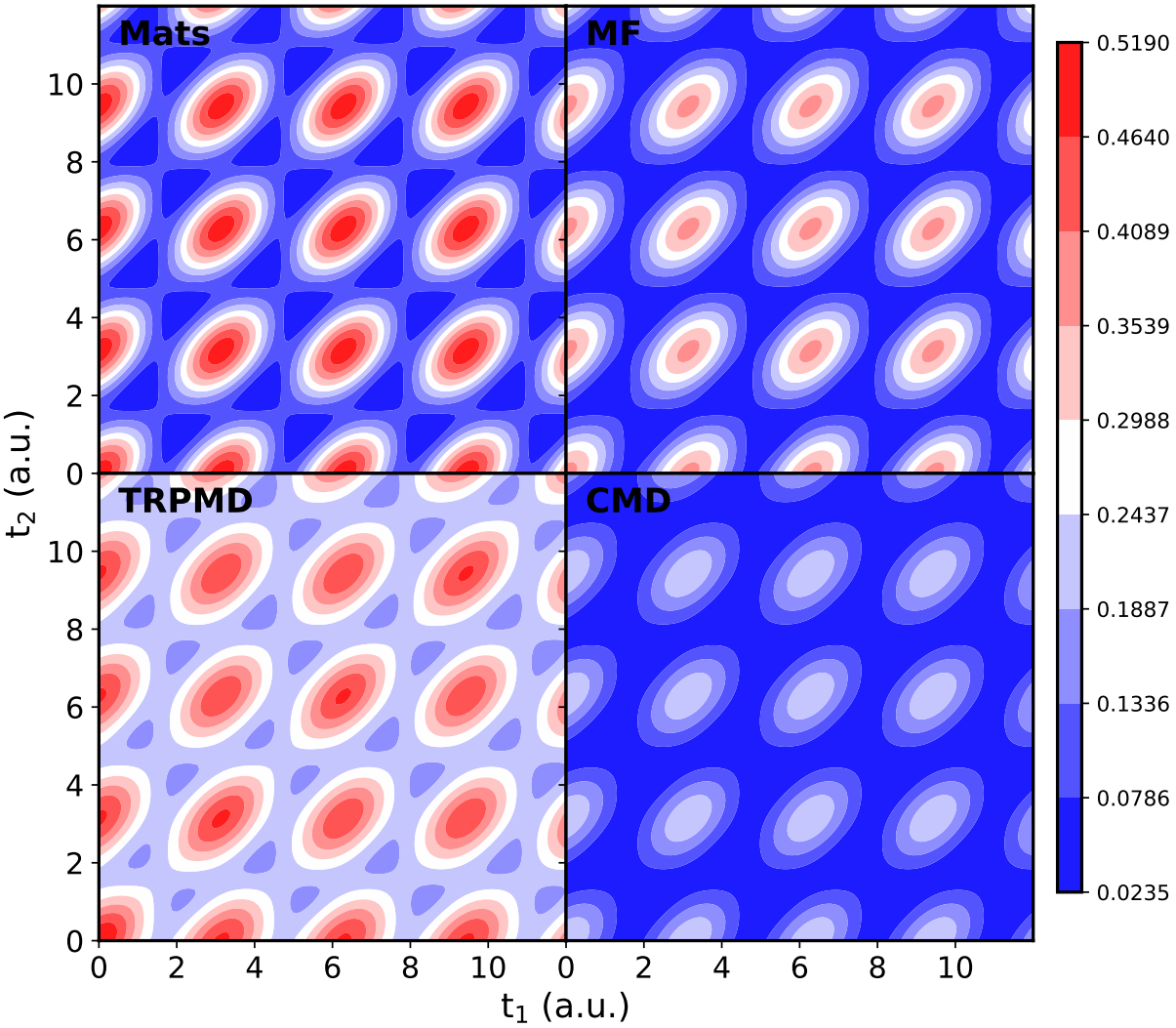

FIG. 6. Contour plots of symmetrized double Kubo transformed $\langle \hat{q}^2 \hat{q}^2(t_1) \hat{q}^2(t_2) \rangle$ correlation function, for the harmonic oscillator potential at $\beta = 4$, at different levels of theory.

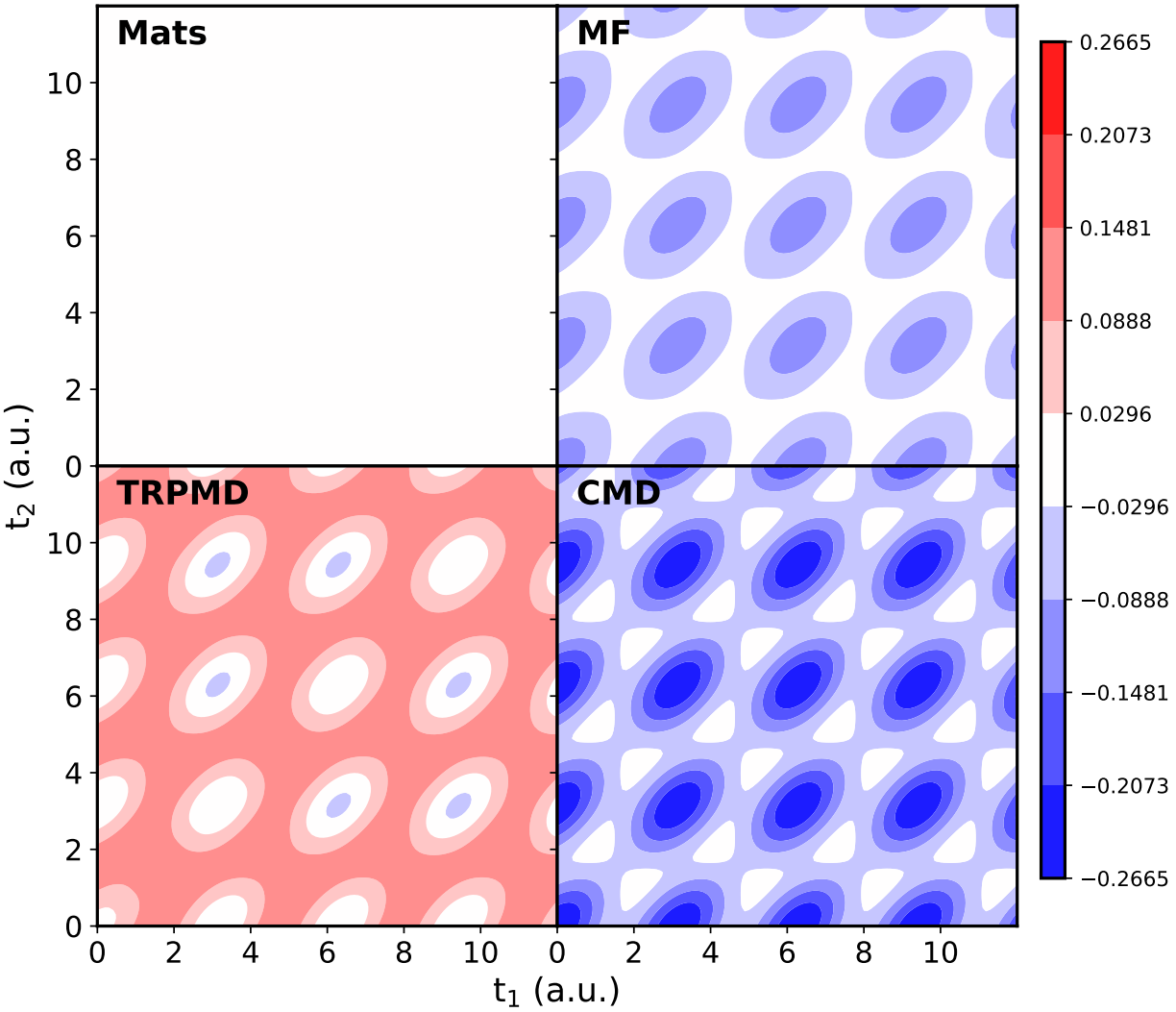

FIG. 7. Contour plots of the error between exact and approximated symmetrized double Kubo transformed $\langle \hat{q}^2 \hat{q}^2(t_1) \hat{q}^2(t_2) \rangle$ correlation function, for the harmonic oscillator potential at $\beta = 4$, at different levels of theory.