The generalized quantum master equation (GQME) formalism has recently been proven highly successful for the calculation of the reduced dynamics of complex many-body systems driven away from equilibrium. Two approaches exist: the Nakajima–Zwanzig–Mori time-convolution (TC) approach\(^{18–21}\) and the Tokuyama–Mori time-convolutionless (TCL) approach\(^{3–5}\). In both approaches, the complexity of solving the many-body quantum Liouville equation is replaced with the need to evaluate a time-dependent kernel/super-operator from which the dynamics of the reduced system can be inferred at all times. The formalism becomes advantageous when the characteristic decay time of the kernels is short compared to the approach to equilibrium or steady state, whereby brute-force numerically-converged solvers\(^{18–21}\) provide efficient schemes to obtain the kernels. Alternatively, the GQME is also an excellent starting point for approximate schemes based on semiclassical and mixed quantum-classical approaches\(^{15,22}\).

The TC memory kernel and the TCL kernel/generator are computed by means of auxiliary super-operators. For the TC memory kernel, this involves the solution of an integro-differential equation and requires the calculation of a super-operator whose form depends explicitly on the system-bath coupling\(^{18–21}\), which often becomes an intractable task. For the TCL kernel, the calculation is based on the so-called reduced system propagator super-operator which requires only generic system observables\(^{10,22}\) but requires an inversion of the reduced propagator which can lead to numerical instabilities.

Here, we outline a simple framework to obtain the TC memory kernel from the reduced system propagator alone, circumventing the need to obtain its inverse (TCL) or calculate higher order system-bath observables (TC). The form of the reduced system propagator is universal for any system-bath Hamiltonian, allowing for a reduction in the complexity of obtaining the TC memory kernel. We demonstrate this on the nonequilibrium generalized Anderson-Holstein impurity model. The framework also provides direct relations between the TC and TCL kernels in terms of the reduced system propagator.

Consider an open quantum system coupled to an environment (bath), described by \(\mathcal{H} = \mathcal{H}_S + \mathcal{H}_B + \mathcal{H}_{SB}\), where \(\mathcal{H}_S, \mathcal{H}_B\) are the system and bath Hamiltonians, and \(\mathcal{H}_{SB}\) the coupling between the two. The exact time evolution of the reduced density matrix (RDM), \(\sigma(t) = \text{Tr}_B \{ \rho(t) \} \) (\(\rho(t)\) is the full density matrix), within the TCL approach, is given in terms of a time-local kernel\(^{3,4}\) \(\frac{d}{dt} \sigma(t) = K(t) \sigma(t)\), where we assumed a non-correlated initial state, namely that \(\rho(0) = \sigma(0) \otimes \rho_B(0)\). A simple approach to obtain \(K(t)\), is based on the reduced system propagator, \(U_S(t) = \text{Tr}_B \{ e^{\mathcal{L}_t} \rho_B \} \neq e^{\mathcal{L}_t}\), where \(\mathcal{L} = -\frac{i}{\hbar} [\mathcal{H}, \cdots]\). Using \(U_S(t)\), the TCL generator is given by\(^{10,22}\)

\[
K(t) = \dot{U}_S (t) U_S^{-1} (t) \tag{1}
\]

The matrix elements of the super-operator \(U_S (t)\) can be obtained directly from the reduced density matrix elements, \(\sigma(t)\) evolved from different initial conditions of the system\(^{10}\). Since the time evolution of the reduced density operator in matrix form reads

\[
\sigma_{ij}(t) = \sum_{kl} U_{S,ij,kl} (t) \sigma_{kl}(0), \tag{2}
\]

it clearly follows that an initial state with \(\sigma_{nm}(0) = 1\) and the remaining values of \(\sigma_{kl}(0) = 0\), will give \(U_{S,ij,mm}(t) = \sigma_{ij}(t)\). For more details see Ref.\(^{10}\).

Importantly, the relation between \(U_S(t)\) and \(\sigma(t)\) holds for any system-bath Hamiltonian and thus, simplifies the calculation of \(K(t)\) for complex model systems. However, Eq. (1) is ill-defined when \(U_S(t)\) is singular (for example, when two system states are degenerate and couple to the bath in the same way), limiting its applicability.

An alternative to the TCL approach, describes the time evolution of the reduced density matrix using a non-local memory term\(^{19,20}\) \(\frac{d}{dt} \sigma(t) = \mathcal{L}_S \sigma(t) + \frac{1}{\hbar^2} \int_0^t dt' \kappa(t') \sigma(t - t')\). For this approach (TC), it is well known that one can rewrite the memory kernel \(\kappa(t)\) in terms of a Volterra equation of the second kind\(^{5,13}\)

\[
\kappa(t) = -\frac{\partial \Phi(t)}{\partial t} - \Phi(t) \mathcal{L}_S - \frac{1}{\hbar^2} \int_0^t dt' \Phi(t - t') \kappa(t'), \tag{3}
\]

where \(\Phi(t) = \hbar^2 \text{Tr}_B \{ \mathcal{L} e^{\mathcal{L} t} \rho_B \}\) is a super-operator that can be calculated by a variety of solvers\(^{11,17}\). However, unlike the reduced propagator, \(\Phi(t)\) depends explicitly on the form of the system-bath coupling via the full Liouvillian \(\mathcal{L} = \mathcal{L}_S + \mathcal{L}_B + \mathcal{L}_{SB}\), and requires the calculation of
The equation for the TC memory kernel may now be rewritten in terms of the reduced system propagator and the system Liouvillian alone:

$$\kappa (t) = \hbar^2 \dot{U}_S (t) - \hbar^2 \dot{U}_S (t) L_S - \int_0^t d\tau \dot{U}_S (t - \tau) \kappa (\tau).$$  \hfill (5)$$

Eq. (5) is the main result of this note. It provides a scheme to calculate the memory kernel for a general form of the coupling Hamiltonian without the need to calculate any terms involving the bath operators, as long as the time derivatives of $U_S (t)$ are obtained numerically. We demonstrate this for the generalized Anderson-Holstein model describing an impurity with on-site electron-electron (e-e) interactions, coupled to three baths: a phonon bath and two fermionic baths (leads) at different chemical potentials (full description given in Ref. 23). We deployed the multilayer multiconfiguration time-dependent Hartree (ML-MCTDH) method 24 to numerically compute the RDM at short times from independent initial system states. The reduced propagator was obtained from the ML-MCTDH results according to Eq. (2), and its time derivatives performed numerically (5 points finite difference). Finally, the memory kernel was computed according to Eq. (5).

Figure 1. The RDM and memory kernel for two values of the interaction energy $U$. Upper panels: The RDM elements propagated from an initially empty dot using the ML-MCTDH method (squares) and the TC GQME approach (solid lines). $\sigma_{nn}$ are the probabilities of the dot being empty (black), occupied by one electron (red), and occupied by two electrons (green). Lower panels: The seven distinct nonzero memory-kernel elements. All quantities are shown in units of $\hbar$. $U = 0 \Gamma$, $U = 2 \Gamma$.

In Fig. 1 we shown the results for two values of the on-site e-e repulsion, $U$. The elements of the memory kernel computed from the reduced system propagator are shown in the lower panels and the resulting elements of the reduced density are shown in the upper panels. The populations obtained by solving the TC GQME with the memory kernel given by Eq. (5) are in excellent agreement with the numerical results obtained directly from the ML-MCTDH method (upper panels), reassuring the numerical procedure to obtain $\kappa (t)$ from $U_S (t)$.

In summary, we have related the memory kernel $\kappa (t)$ in the Nakajima–Zwanzig–Mori TC formalism to the reduced system propagator $U_S (t)$, which can be obtained at short times from an impurity solver. Compared to previous formulations our approach provides a robust and simpler framework, circumventing the need to compute high-order system-bath observables. Moreover, unlike the Tokuyama–Mori TCL approach, the current formalism does not rely on the inversion of a super-operator which can be singular. We illustrated the correctness of the proposed approach for a model system describing both electron-electron and electron-phonon correlations and find excellent agreement between the accurate ML-MCTDH results and the generalized quantum master equation.

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