Exact Calculation of the Time Convolutionless Master Equation Generator: Application to the Nonequilibrium Resonant Level Model

Lyran Kidon,1,2 Eli Y. Wilner,3 and Eran Rabani4,2
1) School of Chemistry, The Sackler Faculty of Exact Sciences, Tel Aviv University, Tel Aviv 69978, Israel
2) The Sackler Center for Computational Molecular and Materials Science, Tel Aviv University, Tel Aviv, Israel 69978
3) School of Physics and Astronomy, The Sackler Faculty of Exact Sciences, Tel Aviv University, Tel Aviv 69978, Israel
4) Department of Chemistry, University of California and Materials Science Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA

The generalized quantum master equation provides a powerful tool to describe the dynamics in quantum impurity models driven away from equilibrium. Two complementary approaches, one based on Nakajima–Zwanzig–Mori time-convolution (TC) and the other on the Tokuyama–Mori time-convolutionless (TCL) formulations provide a starting point to describe the time-evolution of the reduced density matrix. A key in both approaches is to obtain the so called “memory kernel” or “generator”, going beyond second or fourth order perturbation techniques. While numerically converged techniques are available for the TC memory kernel, the canonical approach to obtain the TCL generator is based on inverting a super-operator in the full Hilbert space, which is difficult to perform and thus, all applications of the TCL approach rely on a perturbative scheme of some sort. Here, the TCL generator is expressed using a reduced system propagator which can be obtained from system observables alone and requires the calculation of super-operators and their inverse in the reduced Hilbert space rather than the full one. This makes the formulation amenable to quantum impurity solvers or to diagrammatic techniques, such as the nonequilibrium Green’s function. We implement the TCL approach for the resonant level model driven away from equilibrium and compare the time scales for the decay of the generator with that of the memory kernel in the TC approach. Furthermore, the effects of temperature, source-drain bias, and gate potential on the TCL/TC generators are discussed.

I. INTRODUCTION

The development of accurate and efficient schemes to calculate the dynamic response in quantum impurity models remains one of the greatest challenge in chemical and condensed matter physics. Significant advances have been made in the context of the spin-boson model[1] in which a single impurity spin is coupled to a bosonic bath at equilibrium.[2] Addressing the dynamics in quantum impurity models driven away from equilibrium by the application of a bias voltage, is much more challenging, and most brute-force real-time techniques are limited to relatively short times.[3,4]

This short-coming associated with the well-known dynamical sign problem can be lifted by combining real-time impurity solvers with a reduced-density matrix formalism.[5,6] The basic idea is that the total system can be decomposed into two parts - an interesting part we call “the system” and an environment collectively called “the bath” to which the quantum system is coupled. The exact calculation of the dynamics within this formulation is often given in terms of the Nakajima–Zwanzig–Mori time-convolution (TC) approach[7,8,9,10] derived from the unitary dynamics of the full Hilbert space using projection operator techniques. In this formalism, the complexity of solving the many-body quantum Liouville–von-Neumann equation is reduced to the evaluation of a super-operator so-called the “memory kernel” which fully determines the non-Markovian dynamics of the system.[11] Since the decay of the system typically exceeds the characteristic decay time of the memory kernel, brute-force numerical impurity solvers limited to short times can be used to extract the kernel, from which the long-time dynamics can be obtained.[12,13] This approach has proven fruitful for spin magnetization dynamics,[14] in the nonequilibrium Anderson impurity model[15] driven by electron–electron correlations and for nonadiabatic dynamics and bistability.[16,17] in the nonequilibrium extended Holstein model,[18] driven by electron-phonon correlations. This formalism is also widely used as a starting point to derive perturbative schemes for fermionic systems[19–22] and has gained attention in other fields as well.[23–25]

An alternative to the TC generalized quantum master equation is based on Tokuyama–Mori time-convolutionless (TCL) quantum master equation.[26] The technique eliminates the dependence of the time evolution on the history of the system and replaces the integro-differential TC equation with a first order differential equation, which is exact and local in time.[27–29] In the TCL quantum master equation, the TC memory kernel is replaced by a time-local super-operator referred to as the TCL generator or kernel. A direct calculation of the TCL generator is extremely difficult since it relies on finding

---

[1] Current address: Department of Physics, Columbia University, New York, NY 10027, USA.
an inverse to a super-operator in the full Hilbert space.\textsuperscript{23} Thus, in nearly all applications of the TCL approach, a perturbative scheme in the system-bath coupling is used to obtain the TCL generator.\textsuperscript{20,23} An attempt to formulate a non-perturbative path-integral approach to calculate the kernel has been proposed recently.\textsuperscript{24}

In this work we adopt a different approach to calculate the TCL kernel based on the reduced density propagator\textsuperscript{11} and show that the matrix elements of this 4-ranked tensor can be obtained directly from the population and coherences of the reduced system alone. While this connection may seem redundant, in fact, it offers two important advantages: First, since the TCL generator decays on a faster time-scale compared to the decay of the populations and coherences themselves, one can use numerically converged impurity solvers to obtain the TCL generator at short-times and infer about the converged system dynamics on any time scale, similar to the cutoff method used in the TC approach.\textsuperscript{13,24} Second, an alternative diagrammatic expansion based on the nonequilibrium Green’s function (NEGF) approach offers means to systematically expand the kernel beyond second or fourth order in the system-bath coupling.

We implement the TCL approach to study the population dynamics in the resonant level model\textsuperscript{13} and analyze the time scales for the decay of the TCL kernel as a function of temperature ($T$), source-drain bias ($V_{\text{SD}}$) and gate voltage ($\varepsilon$), and compare it to the decay of the TC memory kernel. We find that both kernels decay on an identical time scale and thus, in this respect, neither formulations is advantageous. However, the TCL formulation requires only system dependent quantities as input, quantities that are often easier to generate using a numerically converged impurity solvers.

\section{II. REDUCED DENSITY MATRIX
FORMALISM: TC AND TCL APPROACHES}

\subsection{A. The reduced density matrix formalism}

Consider an open quantum system coupled to an environment representing the “bath” degrees of freedom, described by the full Hamiltonian:

$$H = H_0 + V = H_S + H_B + V.$$  \textsuperscript{(1)}

where $H_0 = H_S + H_B$ describes the system and bath Hamiltonians, and $V$ the coupling between the two. The time evolution of the full density matrix is given by the Liouville–von-Neumann equation, $\dot{\rho}(t) = -\frac{i}{\hbar} [H, \rho(t)] = \mathcal{L}_0(t)$, for which a numerical converged solution is impossible to obtain due to the exponential scaling complexity with the size of the system and bath. However, in most situations the dynamics of the bath is not interesting and thus, one is concerned with the time evolution of the system alone, described by the reduced density matrix, $\sigma(t)$:

$$\rho_B \otimes \sigma(t) = P \rho(t) \equiv \rho_B \otimes \text{Tr}_B \rho(t),$$ \textsuperscript{(2)}

where $\rho_B$ is the density matrix of the initial state of the bath and $P = \rho_B \otimes \text{Tr}_B \cdots$ is a projection operator onto the system subspace. There are two different approaches to describe the time evolution of $\sigma(t)$: (a) the Nakajima–Zwanzig–Mori time-convolution approach\textsuperscript{35,22} and (b) the Tokuyama–Mori time-convolutionless approach.\textsuperscript{23}

For the former (TC), the equation of motion for $\sigma(t)$ is given by

$$\frac{\partial}{\partial t} \sigma(t) = \mathcal{L}_S \sigma(t) + \int_0^t d\tau \kappa(\tau) \sigma(t - \tau)$$ \textsuperscript{(3)}

where $\mathcal{L}_S = -\frac{i}{\hbar} [H_S, \cdots]$ and the memory kernel (a super-operator in the system subspace) is given by

$$\kappa(t) = \text{Tr}_B \left\{ \mathcal{L} e^{\mathcal{Q} \mathcal{L} t} \mathcal{L} \rho_B \right\}$$ \textsuperscript{(4)}

with $P + \mathcal{Q} = \mathcal{I}$ ($\mathcal{I}$ is the unit operator). For the latter (TCL), the equation of motion for the reduced density operator is given in terms of a time-local kernel\textsuperscript{23}

$$\frac{\partial}{\partial t} \sigma(t) = \mathcal{K}(t) \sigma(t).$$ \textsuperscript{(5)}

In the above equation, the TCL kernel (a super-operator in the system subspace) is defined by

$$\mathcal{K}(t) = \text{Tr}_B \left\{ \mathcal{L} \left( 1 - \Sigma(t) \right)^{-1} \rho_B \right\}$$ \textsuperscript{(6)}

where

$$\Sigma(t) = \int_0^t d\tau e^{\mathcal{Q} \mathcal{L} \tau} \mathcal{L} \mathcal{P} e^{-\mathcal{L} \tau}$$ \textsuperscript{(7)}

is a super-operator in the full Hilbert space of the system and bath. In the above equations, we have assumed a factorized initial condition, $\rho(0) = \rho_B(0) \otimes \sigma(0)$. For a correlated initial state, an additional term should be included in both the TC and TCL equations for $\sigma(t)$.\textsuperscript{23}

The complication of solving for the reduced density operator is manifested through the TC or TCL kernels. In fact, it seems more complicated to solve for the kernels than obtaining the full density operator $\rho(t)$, since both $\kappa(t)$ and $\mathcal{K}(t)$ involve projected propagation $e^{\mathcal{Q} \mathcal{L} t}$ and furthermore, the TCL kernel requires an inversion of a super-operator in the full Hilbert space. However, the reduced density operator formalism offers two main advantages. First, when formulated in the interaction picture, the TC and TCL approaches are useful as a starting point for approximate methods, such as the second order perturbation in the system-bath coupling leading to the Redfield equations. Second, if the dynamics governing $\kappa(t)$ or $\mathcal{K}(t)$ are short lived, then obtaining the dynamics of $\sigma(t)$ on all time scales can be achieved by calculating the kernels at short time only. In fact, this has been a useful approach to obtain the memory kernel in the Nakajima-Zwanzig-Mori TC approach, as recently illustrated for nonequilibrium impurity models\textsuperscript{25} and for other condensed phase systems.\textsuperscript{26}
B. The TCL Generator in terms of the reduced system propagator

In the Nakajima-Zwanzig-Mori TC approach, one can rewrite the memory kernel in terms of a Volterra equation of the second kind, removing the complexity of the projected dynamics of Eq. (4),

\[ \kappa (t) = \frac{\partial \Phi (t)}{\partial t} - \Phi (t) L_S - \int_0^t d\tau \Phi (t - \tau) \kappa (\tau) \tag{8} \]

where \( \Phi (t) = Tr_B \{ L_V e^{L t} \rho_B \} \) and \( L_V = -i \hbar [V, \cdots] \). The super-operator \( \Phi (t) \) can now be calculated by a variety of numerically exact techniques, since it does not involve projected dynamics. A similar approach for the TCL generator seems difficult to derive, since one has to invert the super-operator \( 1 - \Sigma (t) \) which spans the entire Hilbert space. Indeed, most applications of the TCL approach were based on expanding \( (1 - \Sigma (t))^{-1} \) in a Taylor series with the condition that \( \| \Sigma (t) \| \leq 1 \), such that \( \kappa (t) \) is obtained perturbatively,

\[ \kappa (t) = \frac{\partial \Phi (t)}{\partial t} - \Phi (t) L_S - \int_0^t d\tau \Phi (t - \tau) \kappa (\tau) \tag{8} \]

Here, we adopt a simple formalism to obtain \( \kappa (t) \) circumventing the need to invert a super-operator in the full Hilbert space by rewriting the TCL generator in terms of system observables only. The approach is thus, amenable to impurity solvers of the kind used in the TC approach. We begin by redefining the expression for \( \sigma (t) \) as,

\[ \sigma (t) = Tr_B \{ \rho (t) \} = Tr_B \{ U (t) \rho (0) \} \tag{9} \]

where \( U (t) = e^{L t} \) is the full propagator. For factorized initial conditions,

\[ \sigma (t) = Tr_B \{ U (t) \sigma (0) \otimes \rho_B (0) \} = Tr_B \{ e^{iHt} \sigma (0) \otimes \rho_B (0) e^{-iHt} \} = U_S (t) \sigma (0) \tag{10} \]

where \( U_S (t) \) is the propagator of the system (dot) only \( (U_S (t) \neq e^{L_S t}) \). By reversing the equation and performing a time derivative on it, we obtain,

\[ \dot{\sigma} (t) = U_S (t) \sigma (0) = \dot{U}_S (t) U_S^{-1} (t) \sigma (t) \tag{11} \]

This equation has the form of the TCL quantum master equation for the reduced density matrix (Eq. (5)) where by analogy the TCL generator \( \kappa (t) \) is given by,

\[ \kappa (t) = \dot{U}_S (t) U_S^{-1} (t) \tag{12} \]

Thus, in order to obtain the TCL generator one has to compute \( U_S (t) \) as defined in Eq. (10) and also invert \( U_S (t) \). As will become clear below, obtaining the matrix elements of \( U_S (t) \) is straightforward and so is the inversion. To see this, we rewrite the above equations in a basis set and provide explicit expression for the elements of the reduced propagator in the following subsection.

C. Matrix representation

Choosing a basis for the system subspace as the eigenstates of the system decoupled from the bath, \( |i \rangle \), an operator \( O \) will be represented by its matrix elements \( O_{ij} = \langle i | O | j \rangle \) and a super-operators \( \mathcal{O} \) will be represented by the elements of a “tetradic” (specified by four subscripts) \( O_{ijkl} = Tr_S \{ \langle i | \langle j | O | k \rangle | l \rangle \} \). A super-operator is an operator that works on other operators rather than on quantum states and thus, it turns a matrix into a new matrix in the following way:

\[ (\mathcal{O} \mathcal{O})_{ij} = \sum_{kl} O_{ij,kl} O_{kl} \tag{13} \]

Following this definition, it can be shown that the product of two super-operators is given by:

\[ (\mathcal{O} \mathcal{R})_{ij,kl} = \sum_{mn} O_{ij,mn} \mathcal{R}_{mn,kl} \tag{14} \]

Therefore, the time evolution of the reduced density matrix elements is given by:

\[ \dot{\sigma}_{ij} (t) = \sum_{kl} U_S^{*} (t) \sigma_{kl} (0) , \tag{15} \]

the time-local master equation for the reduced density matrix in matrix form reads:

\[ \frac{\partial}{\partial t} \sigma_{ij} = \sum_{kl} K_{ijkl} (t) \sigma_{kl} (t) , \tag{16} \]

and the TCL generator elements are given by:

\[ K_{ijkl} (t) = \sum_{mn} \left( \dot{U}_S (t) \right)_{ij,mn} \left( U_S^{-1} (t) \right)_{mn,kl} \tag{17} \]

For simplicity, instead of writing operators in the system sub-space as \( N \times N \) matrices, where \( N \) is the dimension of the sub-space, and super-operators as tensors of dimensions \( N^4 \), we may represent the first by vectors of size \( N^2 \) and the latter by \( N^2 \times N^2 \) matrices. This representation preserves the definitions in Eqs. (13) and (14) and simplifies certain operations, such as finding the inverse of \( U_S \).

D. System propagator matrix elements

The exact form of the system propagator matrix elements can be written explicitly in terms of the matrix elements of the full propagator, as derived in Appendix A. The derivation is completely general and therefore, also constitutes a proof that such a tetradic representation (Eq. (15)) in fact exists. However, this tetradic form given in the Appendix may be difficult to treat. Here, instead we represent the matrix elements of \( U_S (t) \) in terms of the reduced density matrix elements
directly. This is done by propagating the system from different initial conditions of $\sigma(0)$. For example, if we take $\sigma_{nm}(0) = 1$, where the subscript $m$ denotes one of the many-body states of the system, and the remaining values of $\sigma_{ij}(0) = 0$, then from Eq. 15 it follows that

$$\mathcal{U}_{S,ij,mm}(t) = \sigma_{ij}(t).$$  \hspace{1cm} (18)

Hence, the elements $\mathcal{U}_{S,ij,mm}(t)$ can be obtained directly from the matrix elements of the reduced density operator with the above specified initial condition. Since $\sigma_{ij}(t)$ can be generated using a proper impurity solver at short times, the TCL kernel can be generated without the need to invert a super-operator in the full Hilbert space or use a perturbation expansion to obtain $\mathcal{K}(t)$.

Rather, the TCL kernel is generated by inverting $\mathcal{U}_S(t)$ in the system subspace only. This amounts to a computational complexity that is similar to that used to generate the memory kernel in the TC approach.

Similarly, for the elements $\mathcal{U}_{S,ij,mm}(t)$ with $m \neq n$, one has to take two different initial conditions for the system. The first is $\sigma_{nm}(0) = 1, \sigma_{mn}(0) = \sigma_{nm}(0) = z$ and the remaining values of $\sigma_{ij}(0) = 0$ and the second only differs in $\sigma_{mn}(0) = \sigma_{nm}(0) = z'$, where $z$ and $z'$ are complex numbers ($z \neq z'$) and at least one is not purely real or imaginary. With the above initial conditions, we find that:

$$\sigma_{ij}^z(t) = \mathcal{U}_{S,ij,mm}(t) + z\mathcal{U}_{S,ij,mm}(t) + z^*\mathcal{U}_{S,ij,mm}(t)$$

$$\sigma_{ij}^{z'}(t) = \mathcal{U}_{S,ij,mm}(t) + z'\mathcal{U}_{S,ij,mm}(t) + z'^*\mathcal{U}_{S,ij,mm}(t).$$  \hspace{1cm} (19)

where the superscript $z$ or $z'$ indicates which initial condition is used to generate $\sigma(t)$. Extracting $\mathcal{U}_{S,ij,mm}(t)$ and $\mathcal{U}_{S,ij,mm}(t)$ can now be achieved by solving the above linear equations. As before, $\sigma_{ij}^z(t)$ and $\sigma_{ij}^{z'}(t)$ can be generated at short times from an impurity solver.

III. EXPLICIT FORMULATION OF THE TCL APPROACH FOR THE RESONANT LEVEL MODEL

A. Model and factorized initial conditions

We now turn to demonstrate the approach outlined above to calculate the TCL kernel and compare the time scales governing it with the TC memory formalism for a transport model system. We focus on the noninteracting resonant level model since the TCL and TC kernels can be obtained in terms of an exact nonequilibrium Green’s function approach. However, more complicated models can be treated in the same manner with the approach presented above. The Hamiltonian describing this open quantum system is already given in the format of Eq. 1, where

$$H_S = \varepsilon d^d d$$  \hspace{1cm} (20)

is the system Hamiltonian with fermionic creation/annihilation operators $d^d/d$, respectively, and energy $\varepsilon$. The bath Hamiltonian represents the noninteracting leads:

$$H_B = H_L + H_R = \sum_{k \in L} \varepsilon_k c_k^\dagger c_k + \sum_{k \in R} \varepsilon_k c_k^\dagger c_k,$$  \hspace{1cm} (21)

with fermionic creation/annihilation operators $c_k^\dagger/c_k$, respectively for the left ($L$) or right ($R$) leads. The hybridization between the system and the leads is given by

$$V = \sum_{k \in L,R} t_k (c_k^\dagger d + h.c.)$$  \hspace{1cm} (22)

with coupling strength $t_k = \sqrt{\delta \varepsilon J(\varepsilon_k)/2\pi}$ and $\delta \varepsilon$ is the band discretization width. For all applications reported below, we assume a wide band spectral function of the form:

$$J(\varepsilon) = \frac{\Gamma/2}{(1 + e^{\gamma(\varepsilon - \varepsilon_C)}) (1 + e^{-\gamma(\varepsilon + \varepsilon_C)})},$$  \hspace{1cm} (23)

where $\gamma = 4\Gamma$ is the cutoff length and $\varepsilon_C = 40\Gamma$ is the band cutoff energy.

We take a factorized form for the initial condition described by

$$\rho(0) = \sigma(0) \otimes \rho_L(0) \otimes \rho_R(0)$$

where $\sigma(0)$ will be specified below and

$$\rho_{L/R}(0) = \frac{1}{Z_{L/R}} \exp \left(-\beta \left(H_{L/R} - \mu_{L/R} N_{L/R} \right) \right) \hspace{1cm}$$

with $Z_{L/R}$ the corresponding normalization, $\beta = 1/k_B T$ is the inverse temperature, $\mu_{L/R}$ are the left lead and the right lead chemical potentials, respectively, such that the source-drain bias is given by $V_{SD} = \mu_L - \mu_R$. In the above, $N_{L/R} = \sum_{k \in L/R} c_k^\dagger c_k$.

B. Calculation of the TCL kernel

For the above model, the system is spanned by two levels, an empty ($\langle 0 \rangle$) and occupied ($\langle 1 \rangle = d^d |0\rangle$) dot. Thus, the reduced density matrix of the system, $\sigma(t) = \text{Tr}_B \{\rho(t)\}$, is a $2 \times 2$ matrix and the system propagator $\mathcal{U}_S(t)$ is a super-matrix with $2^d$ elements. For simplicity we will represent $\sigma(t)$ with a 4-dimensional vector and $\mathcal{U}_S(t)$ as a $4 \times 4$ matrix, as explained in Sec. II C.

The diagonal matrix elements and coherences dynamics are independent in the resonant level model, since coherences do not couple different Fock spaces. Therefore, the system propagator has a block structure with vanishing off-diagonal block-elements. Since we are interested in
the populations only, we limit the discussion to the
diagonal elements of the reduced density matrix, given by
the relevant part of Eq. (10):
\[
\sigma_{00}(t) = U_{S,00,00}(t) \sigma_{00}(0) + U_{S,00,11}(t) \sigma_{11}(0),
\]
\[
\sigma_{11}(t) = U_{S,11,00}(t) \sigma_{00}(0) + U_{S,11,11}(t) \sigma_{11}(0).
\]
(24)
Since \(\sigma_{11}(t) = 1 - \sigma_{00}(t)\) and thus \(U_{S,00,00}(t) + U_{S,11,00}(t) = 1\) and \(U_{S,00,11}(t) + U_{S,11,11}(t) = 1\), we can
express the relevant part of the reduced propagator in
terms of two matrix elements alone:
\[
U_S(t) = \begin{pmatrix} U_{S,00,00}(t) & U_{S,00,11}(t) \\ 1 - U_{S,00,00}(t) & 1 - U_{S,00,11}(t) \end{pmatrix}.
\]
(25)
From Eq. (24), it is clear that \(U_{S,00,00}(t) = \sigma_{00}(t)\) for
an initial empty dot and \(U_{S,00,11}(t) = \sigma_{00}(t)\) for
an initial occupied dot. Thus, to obtain the two independent
elements of \(U_S(t)\), we carry out two separate calculations
corresponding to these two initial dot preparations,
where \(\sigma_{00}(t) = \langle d(t) d(t) \rangle\) can be calculated in a
myriad of ways (exactly, for this model). Here, we use the
NEGF approach to obtain \(\langle d(t) d(t) \rangle\).

The TCL kernel matrix elements, \(K_{00,00}(t)\) and
\(K_{00,11}(t)\), can be obtained from Eq. (12), and are given by:
\[
K_{00,00}(t) = \frac{\dot{U}_{S,00,00}(1 - U_{S,00,11}) - \dot{U}_{S,00,11}(1 - U_{S,00,00})}{U_{S,00,00} - U_{S,00,11}},
\]
\[
K_{00,11}(t) = \frac{\dot{U}_{S,00,11}U_{S,00,00} - \dot{U}_{S,00,00}U_{S,00,11}}{U_{S,00,00} - U_{S,00,11}}.
\]
(26)
The above sum rules for \(\sigma(t)\) and \(U_S(t)\) translate to the
following sum rules for \(K(t)\):
\[
K_{00,00}(t) = -K_{11,00}(t),
\]
\[
K_{00,11}(t) = -K_{11,11}(t).
\]
(27)

C. Temperature and Bias Voltage Dependence
of \(K(t)\)

The temperature and bias voltage dependence of \(K(t)\) is
complicated, however
\[
K_{00,00}(t) - K_{00,11}(t) = \frac{\dot{U}_{S,00,00}(t) - \dot{U}_{S,00,11}(t)}{U_{S,00,00}(t) - U_{S,00,11}(t)}
\]
is independent of \(T\) or \(V_{SD}\). This can be shown by using
the approach outlined in Ref. [50] to express \(U_{S,00,00}(t)\)
and \(U_{S,00,11}(t)\). We begin by defining a correlation ma-
trix with elements \(C_{ij}(t) = \langle c_i^\dagger(t) c_j(t) \rangle\), where \(i, j = 0\)
refers to the dot \((c_0 \equiv d)\), \(i, j = 1, \ldots, n\) refers to the
left lead and \(i, j = n + 1, \ldots, 2n\) refers to the right lead. For
a non-correlated initial state, \(C(0)\) is a diagonal matrix
with elements \(\langle c_i^\dagger(0) c_0(0) \rangle = n_d \equiv f_0\), where \(n_d\) is the
initial population of the dot, \(\langle c_i^\dagger(0) c_i(0) \rangle = f_L(\varepsilon_i) \equiv f_i\)
for \(1 \leq i \leq n\), and \(\langle c_i^\dagger(0) c_i(0) \rangle = f_R(\varepsilon_i) \equiv f_i\)
for \(n + 1 \leq i \leq 2n\). Here, \(f_{L/R}(\varepsilon) = (1 + e^{\beta(\varepsilon - \mu_{L/R})})^{-1}\) is
the Fermi Dirac distribution of the left/right lead, respec-
tively. Next, we define a new set of fermionic operators
\(a_\alpha = \sum_{j=0}^{2n} M_{\alpha j} c_j\) where \(M\) is the matrix that
diagonalizes the one-body Hamiltonian
\[
H_1 = \begin{pmatrix}
\varepsilon & t_1^L & t_2^L & \cdots & t_n^R \\
t_1^L & \varepsilon & & & \\
1 & t_2^L & \varepsilon & & \\
\vdots & \ddots & \ddots & \ddots & \\
t_n^L & & & \varepsilon & \\
\end{pmatrix}
\]
(29)
with eigenvalues \(\varepsilon_\alpha = (M^\dagger \cdot H_1 \cdot M)_{\alpha \alpha}\) and \(a_\alpha(t) = e^{-\frac{\varepsilon_\alpha}{2} t} a_\alpha(0)\).
Using these relations, \(\sigma_{11}(t) = 1 - \sigma_{00}(t)\) is
given by:
\[
\sigma_{11}(t) = \langle c_0^\dagger(t) c_0(t) \rangle = \sum_{\alpha \beta} M_{0\alpha} M_{\beta 0}^* \langle a_\alpha^\dagger(t) a_\beta(t) \rangle
\]
\[
= \sum_{\alpha} M_{0\alpha} M_{\beta 0}^* e^{\frac{1}{2}(\varepsilon_\alpha - \varepsilon_\beta)t} \langle a_\alpha^\dagger(0) a_\beta(0) \rangle.
\]
(30)
Rewriting the above in terms of the original set of fermionic
operators, we find:
\[
\sigma_{11}(t) = \sum_{\alpha \beta} M_{0\alpha} M_{\beta 0}^* e^{\frac{1}{2}(\varepsilon_\alpha - \varepsilon_\beta)t} \cdot \sum_{ij} M_{i\alpha}^* M_{j\beta} \langle c_i^\dagger(0) c_j(0) \rangle
\]
\[
= \sum_{i=0}^{2n} f_i \left| \sum_{\alpha} M_{0\alpha}^* e^{-\frac{\varepsilon_\alpha}{2} t} M_{\alpha i} \right|^2.
\]
(31)
Using Eq. (18) we may now recall that for a choice of
an empty initial state, that is for \(f_0 = n_d = 0\),
\(U_{S,00,00}(t) = \sigma_{00}(t)\) and for an initial occupied dot
\(U_{S,00,11}(t) = \sigma_{00}(t)\). Thus, for \(n_d = 0\):
\[
U_{S,00,00}(t) = 1 - \sum_{i=1}^{2n} f_i \left| \sum_{\alpha} M_{0\alpha}^* e^{-\frac{\varepsilon_\alpha}{2} t} M_{\alpha i} \right|^2
\]
(32)
and for \(n_d = 1\):
\[
U_{S,00,11}(t) = U_{S,00,00}(t) - \sum_{\alpha} |M_{0\alpha}|^2 e^{-\frac{\varepsilon_\alpha}{2} t}.
\]
(33)
From the above it is clear that \(U_{S,00,00}(t) - U_{S,00,11}(t) =
\left| \sum_{\alpha} |M_{0\alpha}|^2 e^{-\frac{\varepsilon_\alpha}{2} t} \right|^2\), which is \(T\) and \(V_{SD}\) independent.
IV. RESULTS FOR THE RESONANT LEVEL MODEL

![Graphs showing results for different ε values](image)

Figure 1. Plots of the steady-state dot population as a function of the inverse cutoff time (lower panels), the time dependence of the dot population (middle panels), and \(\kappa_{00,00}(t)\) and \(\partial\kappa_{00,00}/\partial t\) (upper panels). Left, middle and right panels are for different values of the dot energy, as indicated. The source-drain bias voltage is \(V_{SD} = \Gamma\). The green, red and black curves are for \(T = \Gamma/10\), \(T = \Gamma\), and \(T = 10\Gamma\), respectively.

To compare the TC and TCL approaches, we have generated the corresponding kernels for the resonant level model using a two-time NEGF approach, which is exact for non-interacting electrons. For the resonant level model, the coherences are decoupled from the populations\(^{[18]}\) and here we focus on the latter only. Hence, both the TC and TCL kernels have only two independent elements, \(\kappa_{00,00}(t)\) and \(\kappa_{01,11}(t)\) were obtained by solving Eq. \((38)\), where the input \(\Phi(t)\) was generated from the two-time lesser Green’s function of the lead-dot \((G_k^<(t,t') = \frac{1}{\hbar} \langle d^\dagger(t')c_k(t) \rangle\). \(\kappa_{00,00}(t)\) and \(\kappa_{01,11}(t)\) were obtained from Eqs. \((18)\) and \((20)\), where the dot population was generated from dot lesser Green's function \((G^<(t,t') = \frac{1}{\hbar} \langle d^\dagger(t')d(t) \rangle)\).

In the upper panels of Figs. 1, 2 and 3 we compare the ‘00,00’ element of the TC memory kernel to the time-derivative of the corresponding TCL element for several values of the dot energy \(\varepsilon\), for three temperatures, and for different values of the bias-voltage \((V_{SD})\). Since the TCL generator decays to a constant, we plot its time-derivative, providing a more direct comparison to the TC kernel. \(\kappa_{00,00}(t)\) and \(\partial\kappa_{00,00}/\partial t\) have the same sign, and thus for clarity we plot \(-\kappa_{00,00}(t)\), however, even when we compare the two quantities directly, they show a slightly different transient behavior. Interestingly, both \(\kappa(t)\) and \(\dot{\kappa}(t)\) decay on an identical time scale, regardless of the dot energy, temperature or source-drain bias voltage.

In the middle panels of Figs. 1, 2 and 3 we show the transient dot population \((\sigma_{11}(t))\) using the TC and TCL approaches. In addition, we also plot the exact dot population obtained directly from the NEGF calculation. Thus, each panel includes 9 curves, but only 3 are clearly distinguishable, corresponding to different parameters. The other curves overlap signifying the excellent agreement between the TC, TCL and the exact NEGF results for the cutoff time \((t_c)\) used. Furthermore, it is clearly evident that the decay of the dot population occurs on longer time scales compared to the decay of \(\kappa_{00,00}(t)\) or \(\partial\kappa_{00,00}/\partial t\). This is true as long as the source-drain bias voltage is not too large and the temperature not too small. This separation of time scales is central to the use of the TC and TCL approaches.

In the lower panels of Figs. 1, 2 and 3 we plot the steady state value of \(\sigma_{11}(t \to \infty)\) as obtained directly from the kernel elements:

\[
\sigma_{11}(\infty) = \frac{\kappa_{00}}{\kappa_{00} - \kappa_{01}},
\]

where for the TC \(K_{ij} = \int_0^{t_c} dt \kappa_{ij,jj}(t)\) and for the TCL \(K_{ij} = K_{ii,jj}(t_c)\). For all cases studied, the TC and TCL show exactly the same behavior with the cutoff time, \(t_c\). This implies that neither formulation has an advantage over the other with respect to the time scales needed to generate the kernel using a proper impurity solver. Whether this is a general result or specific to the resonant level model is still an open question, which will be addressed in future studies.

V. CONCLUDING REMARKS

We have adopted the reduced density propagator formalism to obtain the TCL generator, circumventing the need to invert a super-operator in the full Hilbert space and thereby use a perturbative scheme in the system-
bath coupling. The elements of the reduced density propagator can be deduced from the time-dependent reduced density matrix alone, which can be generated using a proper impurity solver or a diagrammatic technique. The formalism provides a clear advantage for situations where the TCL generator decays much faster than the reduced density matrix itself.

We have implemented the approach for an open non-interacting quantum system driven away from equilibrium by a source-drain bias potential. Comparison of the present TCL approach to our previously developed TC approach reveals that the relaxation time for both kernels is identical, suggesting that neither one is superior in this respect. However, the current formalism requires only system observables to generate the kernel. This may become an advantage, depending on the specific choice of the impurity solver used to calculate the kernel.

ACKNOWLEDGMENTS

We would like to thank David Reichman and Michael Thoss for insightful discussions and suggestions. E.Y.W. is grateful to The Center for Nanoscience and Nanotechnology at Tel Aviv University for a doctoral fellowship.

Appendix A: Tetradic representation of the system propagator

To find the tetradic representation for the reduced system propagator, let us work in the uncoupled base, with Latin letters for the dot many-body states $i, j, k, l, \ldots$ and Greek for the leads many-body states $\alpha, \beta, \gamma, \delta \ldots$. Then, we may write:

$$\rho(t) = U(t) \rho(0)$$

$$\rho_{\alpha \beta \gamma \delta}(t) = \sum_{k, \gamma, l, \delta} U_{\alpha \beta \gamma \delta; k, \gamma, l, \delta}(t) \rho_{k \gamma, l \delta}(0) \quad \ldots \quad (A1)$$

where we used the explicit tetradic form of the Liouville propagator, as shown in reference [51] (C.6 P.105):

$$(e^{iHt})_{ij,kl} = (e^{iHt})_{ik} (e^{-iHt})_{lj} \quad (A2)$$

and the factorized initial conditions:

$$\rho_{k \gamma, l \delta}(0) = \langle k | \gamma \rho(0) | l, \delta \rangle$$

$$= \langle k | \otimes \langle \gamma | (\rho_B(0) \otimes \sigma(0)) | l \otimes \delta \rangle$$

$$= \langle \gamma | \rho_B(0) \delta \rangle \langle k | \sigma(0) | l \rangle = \rho_{B, \gamma \delta}(0) \sigma_{kl}(0). \quad (A3)$$

Now performing the trace over the bath is simply a matter of summing the expression in the last line of Eq. (A1) over $\alpha$ with $\delta_{\alpha \beta}$, and we have:

$$\sigma_{ij}(t) = (\text{Tr}_B \{\rho(t)\})_{ij}$$

$$\quad = \sum_{\alpha} \sum_{k, \gamma, l, \delta} (e^{iHt})_{\alpha, k \gamma} (e^{-iHt})_{l \delta, j \alpha} \rho_{B, \gamma \delta}(0) \sigma_{kl}(0)$$

$$\quad = \sum_{k l} \left[ \sum_{\alpha, \gamma, \delta} (e^{iHt})_{\alpha, k \gamma} (e^{-iHt})_{l \delta, j \alpha} \rho_{B, \gamma \delta}(0) \right] \sigma_{kl}(0)$$

$$\quad = \sum_{k l} U_{\alpha, \beta \gamma \delta}(t) \sigma_{kl}(0) \quad (A4)$$

where the tetradic form of the system propagator is given by:

$$U_{\alpha, \beta \gamma \delta}(t) = \sum_{\alpha, \beta, \gamma} (e^{iHt})_{\alpha, \beta \gamma} \rho_{B, \beta \gamma}(0) (e^{-iHt})_{\gamma, j \alpha} \quad (A5)$$

REFERENCES

1. A. J. Leggett, S. Chakravarty, A. Dorsey, M. P. Fisher, A. Garg, and W. Zwerger, Rev. Mod. Phys. 59, 1 (1987).
2. C. H. Mak, Phys. Rev. Lett. 68, 899 (1992).
3. R. Egger and C. Mak, Phys. Rev B 50, 15210 (1994).
4. N. Makri and D. E. Makarov, J. Chem. Phys. 102, 4600 (1995).
