Phonon dynamics in correlated quantum systems driven away from equilibrium

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A general form of a many-body Hamiltonian is considered, which includes an interacting fermionic subsystem coupled to noninteracting extended fermionic and bosonic systems. We show that the exact dynamics of the extended bosonic system can be derived from the reduced density matrix of the subsystem alone, despite the fact that the latter contains information about the subsystem only. The advantage of the formalism is immediately clear: While the reduced density matrix of the subsystem is readily available, the formalism offers access to observables contained in the full density matrix, which is often difficult to obtain. As an example, we consider an extended Holstein model and study the nonequilibrium dynamics of the so-called “reaction mode” for different model parameters. The effects of the phonon frequency, the strength of the electron-phonon couplings, and the source-drain bias voltage on the phonon dynamics across the bistability are discussed.

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Strongly correlated systems show a remarkable range of interesting phenomena, some of which can be explained with the current arsenal of theoretical tools [1,2]. Their behavior when driven away from equilibrium (e.g., by a finite bias voltage) is less well understood [3]. This is because theoretical tools that provide a reliable and accurate description under equilibrium conditions are difficult to converge for open quantum systems driven away from equilibrium [4,5]. For example, Kondo physics in equilibrium has been understood within renormalization group theory for several decades [6], while the spectral properties under bias [7] were recently solved exactly by a numerical real-time quantum Monte Carlo formalism [8,9], confirming the voltage splitting of the Kondo peak [10]. The lack of a robust theoretical framework to nonequilibrium many-body physics has been the driving force for developing theoretical tools to understand both the dynamics and the approach to a steady state when strong correlations are dominant.

One such powerful tool is based on the Nakjima-Zwanzig reduced density matrix (RDM) formalism [11–13] combined with a proper impurity solver to obtain the memory kernel [14]. This approach has been applied recently to study charge and spin relaxation near the Kondo crossover temperature of the Anderson impurity model [15] and to study localization and bistability in the nonequilibrium extended Holstein model [16,17]. The Nakjima-Zwanzig formalism, by construction, provides access to the dynamics of observables within the reduced space only. Here, we expand the methodology and show how to extract the dynamics of a certain class of observables that were traced out. Our approach is particularly suitable for systems with strong electron-phonon couplings, which give rise to highly interesting phenomena [18–22]. In light of this, we apply the formalism to analyze nonequilibrium phonon dynamics in the extended Holstein model. While the nonequilibrium phonon distribution in the steady state of this model has been analyzed in great detail (see, e.g., [23–27] and references therein), so far there are only very few theoretical studies of time-dependent phonon dynamics, which all involve significant approximations [28–31]. The methodology presented in this paper facilitates a numerically converged treatment of this nonequilibrium many-body problem.

To outline the reduced density matrix formalism, consider a general Hamiltonian for a many-body quantum system comprising bosons and fermions:

\[
H = H_f + H_b + V_{sf} + V_{sb},
\]

where \(H_f = \sum_{ij} \epsilon_{ij} d_i^\dagger d_j + \sum_{ijmn} V_{ijmn} d_i^\dagger d_j^\dagger d_m d_n\) and \(H_b = \sum_{\alpha q} \epsilon_{\alpha q} c_\alpha^\dagger c_\alpha + \sum_{\alpha} \hbar \omega_{\alpha} (b_\alpha^\dagger b_\alpha + \frac{1}{2})\) are the interacting and noninteracting parts of the Hamiltonian for the fermionic degrees of freedom and the bosonic degrees of freedom. The coupling between the subspace “s” and “f” is described by \(V_{sf}\) and often is chosen in the form of a hopping between sites, \(\sum_{\alpha q} (i_k d_i^\dagger c_\alpha + \text{H.c.})\), but the formalism developed below is not limited to this choice. The coupling between the interacting fermions and bosons is taken to the lowest order in dimensionless boson coordinates, \(x_\alpha = \frac{1}{\sqrt{2}} (b_\alpha^\dagger + b_\alpha)\):

\[
V_{sb} = \sum_{ij,\alpha} M_{ij,\alpha}^\alpha d_i^\dagger d_j x_\alpha.
\]

Here, \(d_i^\dagger / d_i\) and \(c_\alpha^\dagger / c_\alpha\) are fermionic creation/annihilation operators at sites \(i\) and \(k\), respectively, and \(b_\alpha^\dagger b_\alpha\) are bosonic creation/annihilation operators for mode \(\alpha\). The above many-body Hamiltonian is a general form covering different generic models, e.g., the Fermi-Bose-Hubbard model [32], the spin-boson model [33], and the Anderson-Holstein quantum impurity model [34,35]. Thus, developing an approach to extract the time-dependent solution of this generic model is of great importance. Using the projection operator \(P = \rho_f(0) \otimes \rho_b(0) \text{Tr}_{f,b}\), where \(\rho_f(0) \otimes \rho_b(0)\) is the initial density matrix in the “s” and “f” subspaces and the trace \(\text{Tr}_{f,b}\) is performed only for these degrees of freedom, one can derive an exact...
function, obtained from the RDM [or from the lesser two-time Green
the positions and momenta of the bosonic variables can be
degrees of freedom in the system part. More specifically, we
is often difficult (or perhaps impossible) to calculate.
formalism for nonsystem operators is that each observable
introduction of an additional memory term with a longer decay
which depends both on $s$ and $f$ and
operators requires the
|\rangle = \sum_{n_{1},n_{2},...,n_{N}} \delta_{n_{1},0} \delta_{n_{2},0} \delta_{n_{j},1} \cdot (8)
Equations (7) and (8) imply that if the RDM of the system is
known the average positions and momenta of the boson modes
can be obtained by solving for Eq. (7) with the RDM given as
an input. This is the main result of this paper. We now illustrate
this for the extended Holstein model.
In this model, $H_s = \varepsilon d^\dagger d$ includes a single level, $H_f = \sum_{k \in L,R} \varepsilon_k \psi^\dagger_k \psi_k$, and $H_b = \sum_{n} \hbar \omega_a (b_n^\dagger + b_n) + \frac{\lambda}{2}$. The
coupling between the system and the fermionic and bosonic
baths is given by $V_{sf} = \sum \tau d^\dagger \psi_k^\dagger c_{k} + \text{H.c.}$ and $V_{fb} = d^\dagger \sum M_{a} (b_{n}^\dagger + b_{n})$, respectively. $t_f$ and $M_a$
determine the strength of the hybridization and electron-phonon
couplings, respectively. The former is modeled by a tight-binding spectral
terms with a longer decay
time scale is typically governed by a large energy scale), i.e., the system
“forgets” its history rapidly [36]. Therefore, one can develop
suitable quantum impurity solvers to calculate the memory
until it decays and obtain the dynamics of the RDM at all
times using Eq. (3).
The Nakajima-Zwanzig formalism, by construction, provides
access to the dynamics of observables within the reduced
space only. Observables that depend also on nonsystem
degrees of freedom (e $f,h$) can, in principle, be calculated
by introducing additional sets of Nakajima-Zwanzig-like
equations. For example, for open quantum systems, the current
which depends both on $s$ and $f$ operators requires the
introduction of an additional memory term with a longer decay
time [36]. The drawback of this extended Nakajima-Zwanzig
formalism for nonsystem operators is that each observable
requires the introduction of an additional memory term, which
is often difficult (or perhaps impossible) to calculate.
We propose an alternative formalism suitable for a certain
class of observables that does not require any additional
calculation of memory terms or the inclusion of the boson
degrees of freedom in the system part. More specifically, we
show that the time evolution of the expectation values of
the positions and momenta of the bosonic variables can be
obtained from the RDM [or from the lesser two-time Green
function, $G^\tau (t,t')$] of the system alone, despite the fact that
$\sigma (t)$ [or $G^\tau (t,t')$] does not contain any information about the
bosonic bath that was traced out. The derivation given below is
rather simple but the result is powerful. It offers a way to extract
information which is not directly accessible. We illustrate the
approach for the extended nonequilibrium Holstein model
and discuss the correlations between the phonon and electron
dynamics.

Consider the Heisenberg equation of motion for $b_a^\dagger (t)$ and
$b_a (t)$ generated by the general Hamiltonian of Eq. (1):

$$\dot{b}_a (t) = -i \omega_a b_a (t) - \frac{i}{\sqrt{2}} \sum_{ij} M_{ij} b_i^\dagger (t) d_j (t),$$

$$\dot{b}_a^\dagger (t) = i \omega_a b_a^\dagger (t) + \frac{i}{\sqrt{2}} \sum_{ij} M_{ij}^* d_i^\dagger (t) d_j (t),$$

where the dimensionless position and momentum of each bo-
son mode is given by $x_a (t) = \frac{i}{\sqrt{2}} [b_a^\dagger (t) + b_a (t)]$ and $p_a (t) = \frac{i}{\sqrt{2}} [b_a^\dagger (t) - b_a (t)]$, respectively. Taking the expectation value
over the initial density matrix $\rho (0)$, we find that

$$\langle \dot{x}_a (t) \rangle = \omega_a \langle p_a (t) \rangle,$$

$$\langle \dot{p}_a (t) \rangle = -\omega_a \langle x_a (t) \rangle - \frac{1}{\hbar} \sum_{ij} M_{ij} \langle d_i^\dagger (t) d_j (t) \rangle,$$

where $\langle \cdots \rangle \equiv \text{Tr} [\rho (0) \cdots ]$. The expectation value of the site
populations and coherences can be expressed in terms of the
RDM (for the same sake by the elements of the Green function
of the system) by

$$\langle d_i^\dagger d_i \rangle = \sum_{n_1,n_2,...,n_N} \delta_{n_1,0} \delta_{n_2,0} \delta_{n_i,1} \cdot (7)$$

$$\langle d_i^\dagger d_j \rangle = \sum_{n_1,n_2,...,n_N} \delta_{n_i,0} \delta_{n_j,1} \delta_{n_1,0} \delta_{n_2,0} \delta_{n_i,1} \cdot (8)$$

Equations (7) and (8) imply that if the RDM of the system is
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baths is given by $V_{sf} = \sum \tau d^\dagger \psi_k^\dagger c_{k} + \text{H.c.}$ and $V_{fb} = d^\dagger \sum M_{a} (b_{n}^\dagger + b_{n})$, respectively. $t_f$ and $M_a$
determine the strength of the hybridization and electron-phonon
couplings, respectively. The former is modeled by a tight-binding spectral
density with an overall coupling determined by $\Gamma$ while
the latter is modeled by an Ohmic spectral density $J(\omega) = \frac{\eta}{\hbar} \omega \omega \exp(-\frac{\omega}{\hbar \omega})$, where the dimensionless Kondo parameter, $\eta = \frac{\Delta}{\hbar \omega}$, determines the overall electron-phonon couplings, $\omega_\text{c}$ is
the characteristic phonon bath frequency, and $\lambda = \sum_{n} \frac{M_{a}^2}{\hbar \omega_n}$ is
$\frac{1}{T} \int d\omega J(\omega)$ is the reorganization energy (or polaron shift),
which also determines the shifting of the dot energy upon
charging.

The reduced density matrix was recently solved for this
model [16,17] by employing two different approaches to
calculate the memory kernel and solving Eq. (3) for $\sigma (t)$: (i) a
two-time nonequilibrium Green function (NEGF) [37] method
within the self-consistent Born approximation (SCBA) [17]
suitable for weak electron-phonon couplings and (ii) the
multilayer multiconfiguration time-dependent Hartree (ML-
MCTDH) [38,39], which is numerically exact but more demanding.
The results obtained in a wide range of parameters
revealed dynamics on multiple timescales. In addition to the short and intermediate timescales associated with the separate electronic and phononic degrees of freedom, the electron-phonon coupling introduces longer timescales related to the adiabatic or nonadiabatic tunneling between the two charge states \((d^1d) \equiv 1\) and \((d^1\bar{d}) \equiv 0\). The analysis shows, furthermore, that the value of the dot occupation may depend on the initial preparation of the phonon degrees of freedom, suggesting the existence of bistability [16,17,23,40,41]. Intriguingly, the phenomenon of bistability persists even on timescales longer than the adiabatic/nonadiabatic tunneling time.

In Fig. 1 we show the correlation between the dynamics of the average dot occupation, the reaction mode, \(\langle Q(t) \rangle = \sum_\alpha M_\alpha \langle x_\alpha(t) \rangle / \sqrt{\sum_\alpha M_\alpha^2}\), and its corresponding momentum, \(\langle P(t) \rangle = \langle Q(t) \rangle / \Omega\), where \(\Omega = \int \frac{d\omega}{2\pi} J(\omega) = \omega_c\) is the reaction mode frequency. These results were obtained for weak electron-phonon couplings by solving the memory kernel required to obtain the RDM using the two-time NEGF within the SCBA. Within this limit, the NEGF-SCBA provides an accurate description of the RDM in comparison to the numerically exact ML-MCTDH approach [38,39]. We consider four different initial conditions for the system and boson bath, namely, all combinations of initial occupied/unoccupied \([n_d(0) = 0/1]\) dot and shifted \([\langle x_\alpha(0) \rangle = -\frac{2\omega_c}{\Gamma_1}\] unshifted \([\langle x_\alpha(0) \rangle = 0]\) phonon modes. These shifted/unshifted values of \(\langle x_\alpha(0) \rangle\) correspond to the location of the minimum of the diabatic potential energy of the occupied/unoccupied dot.

At long times, the dot population (lower panels of Fig. 1) decays to a unique value (closer to the empty state) regardless of the initial preparation of the system or phonon bath, with a typical decay time inversely proportional to \(\Omega\) for the shifted bath and to \(\Gamma\) for the unshifted bath. The average position \(\langle Q(t) \rangle\) and its corresponding momentum \(\langle P(t) \rangle\) follow the population dynamics. At \(t = 0\), \(\langle Q(0) \rangle\) assumes two different values corresponding to the left/right potential minimum. Regardless of the initial conditions, the motion of the reaction mode is overdamped (i.e., no oscillations are observed). This is known to occur for the reaction mode of a bosonic bath with Ohmic spectral density. At long times, the average position decays to values corresponding to the more stable well, consistent with the behavior of the dot populations. The typical time scale for approaching the steady-state value is given by \(\Omega\) (and not by \(\Gamma\)) regardless of the initial conditions and varies only slightly for an unoccupied initial dot.

The qualitative behavior of the dot population changes when the coupling to the boson bath increases. In Fig. 2 we show the results for a larger value of \(\lambda = \epsilon_d\) and different bias voltages \(\Delta\mu\), still within the validity of the NEGF-SCBA. In this case, both potential minima are degenerate and the related spin-boson model (at equilibrium) shows a localization transition at temperature \(T = 0\), which is broadened and finally disappears as \(T\) is increased. The appearance of two distinct values of the dot population at long times at small bias voltages \(\Delta\mu\) is consistent with the equilibrium results for the spin-boson model. The bias voltage plays a similar role of temperature, and as its value increases the bistability disappears.

Turning to discuss the transient behavior of \(\langle Q(t) \rangle\) and \(\langle P(t) \rangle\), we find that similar to the previous case of weaker electron-phonon couplings, the average position of the reaction
coordinate follows closely the transient behavior of the dot population. At long times, \( \langle Q(t) \rangle \) assumes two values roughly corresponding to the two minima of the potential energy along the reaction mode, with vanishing differences as \( \Delta \mu \) increases. The corresponding average momenta always decay to zero at long times, regardless of the initial conditions of the dot and boson bath, suggesting that on the time scale observed \( \langle P(t) \rangle \) decays to its vanishing steady-state value.

The relation between the behavior of \( n_d(t) \) and \( \langle Q(t) \rangle \) at steady state can be derived analytically. Since at steady-state \( \langle \rho_d(t) \rangle = 0 \), one finds from Eq. (7) that each boson mode must satisfy the relation \( x_{\alpha} = -\frac{\lambda_{\alpha}}{M_{\alpha}^2} n_d \), and thus, the difference in \( x_{\alpha} \) between the two different initial distributions of the phonon modes is given by \( M_{\alpha} \Delta x_{\alpha} = -\frac{M_{\alpha}^2}{\lambda_{\alpha}} \Delta n_d \), where \( \Delta n_d \) is the corresponding difference between the two dot populations at steady state. Summing both sides over \( \alpha \), the reaction mode difference, \( \Delta Q = \sum_{\alpha} M_{\alpha} \Delta x_{\alpha}/\sqrt{\sum_{\alpha} M_{\alpha}^2} \), must satisfy the relation

\[
\Delta Q = -\frac{\lambda}{\sqrt{\sum_{\alpha} M_{\alpha}^2}} n_d,
\]

where as before \( \lambda = \sum_{\alpha} \frac{M_{\alpha}^2}{\lambda_{\alpha}} \). This is in agreement with the result obtained in Fig. 2.

So far, we have discussed the appearance of two bistable solutions in the so-called adiabatic limit, where \( \omega_c \ll \Gamma \). In Fig. 3 we show results for larger values of \( \omega_c \) in the regime where \( \omega_c \approx \Gamma \). The value of the electron-phonon coupling (reorganization energy) is somewhat larger than the perturbation regime for which the NEGF-SCBA is accurate. Therefore, we obtain the input required to generate the memory kernel and the RDM from the numerically exact ML-MCTDH approach [38,39]. Similar to the adiabatic limit with weaker electron-phonon couplings (shown in Fig. 2), the long time behavior of \( n_d(t) \) depends on the initial conditions of the phonon bath. However, unlike the adiabatic limit, here we find an additional time scale at long times which is associated with the transition from one diabatic potential surface to the other. Intriguingly, however, the bistability prevails at times longer than the tunneling between the two diabatic surfaces. As the phonon frequency increases, the value of \( \Delta n_d \) decreases and eventually disappears when \( \omega_c \gg \Gamma \).

Similarly to the adiabatic limit, \( Q(t) \) shows the same behavior as \( n_d(t) \), including the long time decay associated with the aforementioned tunneling between the diabatic surfaces, and the long time value of \( \Delta Q \) is correlated with that of \( \Delta n_d \), in agreement with Eq. (9). Unlike the transient behavior of the reaction coordinate, its corresponding momentum decays to its steady-state value on a much faster time scale (typically on a time scale given by \( \Sigma^{-1} \)) and does not show the long time relaxation behavior associated with the phonon tunneling. This implies that the tunneling process is not driven by inertia, but is rather in the overdamped limit.

In summary, we have expanded our recently developed nonequilibrium quantum dynamics methodology, which combines reduced density matrix theory with an impurity solver to obtain the memory kernel, to describe phonon dynamics in correlated open quantum systems. Although the phonon degrees of freedom are formally not part of the reduced system, the structure of the equations of motion allows the calculation of phonon observables based solely on the density matrix and memory kernel of the reduced system. The application to a Holstein-type model for phonon-coupled electron transport in nanosystems reveals the intricate interplay between electron and phonon dynamics in these systems, including the phenomenon of bistability.

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