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Quantum Quenches and Driven Dynamics in Interacting Nanodevices

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Abstract

The remarkable progress of recent years in the fabrication and design of nanodevices has revolutionized the study of strong electronic correlations in such systems. While earlier focus has centered mainly on linear response, current interest has largely shifted to true nonequilibrium dynamics, whether under steady-state conditions or in response to quantum quenches and driven dynamics. The purpose of the proposed research is to theoretically investigate the nonequilibrium dynamics of interacting nanodevices with a special emphasize on single-molecule transistors (SMTs). Such devices have gained enormous interest in recent years both as the basic building blocks of molecular electronics and as a platform for studying the electron-phonon coupling at the nanoscale. Using the generic model for SMTs consisting of a single resonant level coupled by displacement to a single vibrational mode we shall (1) examine the response of the device to a broad class of quantum quenches and ac drives, (2) compute the zero-temperature conductance of an Aharonov-Bohm interferometer with a single-molecule transistor embedded in one of its arms, and (3) compute the nonequilibrium heat current between an electronic and a bosonic reservoir in a suitable variant of the Aharonov-Bohm interferometer. The analytical insights gained will later be used as guidance for analyziug and interpreting numerical computations covering arbitrary coupling strengths.

1 Introduction

The description of strong electronic correlations far from thermal equilibrium constitutes one of the major open questions of modern condensed matter physics. Even under the most favorable conditions of nonequilibrium steady state, many of the concepts and techniques that have proven so successful in equilibrium are simply inadequate. Recent advancements in a broad range of systems, from time-resolved spectroscopies [1, 2] to cold atoms [3, 4] and driven nanostructures [5, 6], have opened new and exciting possibilities for studying the nonequilibrium dynamics in response to quantum quenches and forcing fields. Depending on the physical context one is interested in questions of both basic and practical nature, such as what are the underlying time scales governing the dynamics, how long is coherence maintained, and whether and how does the system equilibrate at

long times. Some questions, e.g., the issue of equilibration, often require nonperturbative treatments even if the system is tuned to weak coupling.

Recent years have witnessed the development of an array of powerful numerical techniques aimed at tracking the real-time dynamics of interacting low-dimensional systems. In the more specific context of quantum impurity systems these methodologies include time-dependent variants of the density-matrix renormalization group [7, 8], the time-dependent numerical renormalization group [9, 10], different continuous-time Monte Carlo approaches [11, 13, 12, 14], and sparse polynomial space representations [15]. Despite notable successes, part of these methods are subject to finite-size effects and discretization errors, while others are confined to rather short time scales. Analytical efforts in this realm have focused mainly on suitable adaptations of perturbative renormalization-group [16, 17] and flow-equation [18] ideas, which in turn neglect higher order terms. Exact analytical solutions, when available, are thus invaluable both for setting a benchmark and for gaining unbiased understanding of the underlying physics. Unfortunately such exact solutions are restricted at present to very special models whose coupling constants must be carefully tuned [19, 20].

The aim of the proposed research is to investigate the nonequilibrium dynamics of interacting nanodevices, with a special emphasize on molecular devices. Single-molecule devices have attracted considerable interest lately due to the technological promise of molecular electronics [21]. From a basic-science perspective they offer an outstanding platform to study the electron-phonon coupling at the nano-scale. In a typical molecular bridge, molecular orbitals are coupled simultaneously to the lead electrons and to the vibrational modes of the molecule, with the former degrees of freedom reduced to a single effective band in the absence of a bias voltage [22]. A minimal model for an unbiased single-molecule device therefore consists of a single resonant level coupled by displacement to a single vibrational mode, as described by the Hamiltonian of Eqs. (1)–(3) below. This Hamiltonian has been extensively used in recent years to model single-molecule transistors and will occupy a significant part of our proposed research. In particular, focusing on the limit where the level is broad as compared to the electron-phonon coupling, we shall

1. present an asymptotically exact solution for the nonequilibrium dynamics of the model for a broad class of quantum quenches and ac drives,

2. compute the zero-temperature conductance of an Aharonov-Bohm interferometer with a single-molecule transistor embedded in one of its arms, and

3. compute the asymptotically exact nonequilibrium heat current between an electronic and a bosonic reservoir in a suitable variant of the Aharonov-Bohm interferometer.

The physical understanding gained from these analytic solutions will later be used as guidance for analyzing and interpreting numerical calculations that apply to much broader parameter regimes. Below we present in detail our proposed research plan.

2 Nonequilibrium Dynamics of Single-Molecule Transistors

One of the common models used to describe single-molecule transistors is that of a resonant level coupled to a single vibrational mode. The corresponding Hamiltonian, expected to describe the system away from Coulomb-blockade valleys where a single unpaired spin resides on the molecule, consists of a single spinless electronic level $d^\dagger$ with energy ϵ_d, which is coupled by displacement to a local vibrational mode $b^\dagger$ with frequency ω_0. The level is further coupled to a band of spinless electrons via the hopping matrix t, as described by the Hamiltonian

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\rm dot}, \tag{1}$$

where

$$\mathcal{H}_0 = \sum_k \epsilon_k c_k^\dagger c_k + t\sum_k (d^\dagger c_k + c_k^\dagger d), \tag{2}$$

$$\mathcal{H}_{\rm dot} = \epsilon_d \hat{n}_d + \omega_0 b^\dagger b + g(b^\dagger + b)\left(\hat{n}_d - \frac{1}{2}\right), \tag{3}$$

with $\hat{n}_d = d^\dagger d$.

The traditional treatment of this Hamiltonian is based either on perturbation theory in g in the weak-coupling regime, or on the Lang-Frisov transformation [23] and the polaronic approximation when t is small. Recently, Dóra and Halbritter [24] have derived a particularly elegant nonperturbative approach to the model, building on the observation that the original Hamiltonian of Eqs. (1)-(3) can be mapped onto an exactly solvable bosonic form in the limit where the electronic level is broad. Using this mapping, these authors proceeded to compute the temperature-dependent conductance of a molecular bridge under resonance conditions. Our first goal is to extend this approach to compute the nonequilibrium dynamics in response to a broad class of quantum quenches and drives.

2.1 Mapping onto an Exactly Solvable Form

The mapping onto an exactly solvable Hamiltonian is done in two steps: (i) Conversion of the Hamiltonian to a continuum-limit form and (ii) Application of Abelian bosonization. The mapping onto a continuum-limit form gives rise to a new ultraviolet cutoff — the hybridization width $\Gamma = \pi\rho_0 t^2$ — and an associated short-distance cutoff $a = 2v_F/\Gamma$. The coupling constants in the contiuuum-limit Hamiltonian are renormalized according to the new cutoff:

$$\mathcal{H} = -iv_F \int \psi^\dagger(x)\partial_x\psi(x) + \omega_0 b^\dagger b + \left[\lambda(b^\dagger + b) + \tilde{\epsilon}_d\right] :\psi^\dagger(0)\psi(0): \tag{4}$$

with

$$\lambda = ga = 2\frac{v_F}{\Gamma}g,$$
$$\tilde{\epsilon}_d = \epsilon_d a = 2\frac{v_F}{\Gamma}\epsilon_d. \tag{5}$$

Here $:\cdots:$ stands for normal-ordering with respect to the filled Fermi sea. Having a continuum-limit Hamiltonian at hand one can apply Abelian bosonization [25] to convert the Hamiltonian to a form quadratic in bosons:

$$\mathcal{H} = \sum_k \epsilon_k a_k^\dagger a_k + \omega_0 b^\dagger b + \left[\lambda(b^\dagger + b) + \tilde{\epsilon}_d\right] \sum_k \xi_k \left(a_k^\dagger + a_k\right), \tag{6}$$

where $a_k^\dagger$ are bosonic modes, and the coefficients ξ_k are given by

$$\xi_k = \sqrt{\frac{k}{2\pi L}} e^{-ak/2\pi} \tag{7}$$

(L being the size of the system). This Hamiltonian, whose construction is controlled by the smallness of the electron-phonon coupling g, the polaronic shift g^2/ω_0 and the level energy $|\epsilon_d|$ as compared to the level width Γ, is quadratic in bosonic operators and can therefore be diagonalized exactly. We use scattering theory to compute the eigenmodes $\alpha_k^\dagger$ of the bosonic Hamiltonian, which are given by

$$\begin{aligned} \alpha_k^\dagger &= a_k^\dagger + \lambda g(\epsilon_k + i\eta)\xi_k \Big[(\epsilon_k - \omega_0)b + (\epsilon_k + \omega_0)b^\dagger \\ &\quad + 2\omega_0\lambda \sum_{q>0} \xi_q \left(\frac{a_q^\dagger}{\epsilon_k + i\eta - \epsilon_q} + \frac{a_q}{\epsilon_k + i\eta + \epsilon_q}\right)\Big]. \end{aligned} \tag{8}$$

Here

$$g(z) = \left[z^2 - \omega_0^2 - 2\omega_0\Sigma(z)\right]^{-1} \tag{9}$$

is related to the phononic Green function and

$$\Sigma(z) = \lambda^2 \sum_{k>0} \xi_k^2 \left(\frac{1}{z - \epsilon_k} - \frac{1}{z + \epsilon_k}\right) \tag{10}$$

is the corresponding self-energy. One can expand operators representing physical observables in terms of these eigenmodes, and since the time evolution of the eigenmodes is known, this expansion allows the calculation of the real-time dynamics of these observables. The specific calculation depends on the initial state in which the system was prepared. The most relevant observables are the phononic occupancy and displacement

$$\begin{aligned} \hat{n}_b &= b^\dagger b, \\ \hat{Q} &= \frac{b^\dagger + b}{\sqrt{2}}, \end{aligned} \tag{11}$$

and the occupancy of the electronic level

$$\hat{n}_d = d^\dagger d = \frac{1}{2} + a \sum_{q>0} \xi_q \left(a_q^\dagger + a_q\right). \tag{12}$$

2.2 Quench Scenarios

Having these expansions at hand, several physical scenarios can be considered in which the system is prepared in some initial state when a quantum quench is applied. The time-dependent response of the system to the sudden quench can then be calculated.

We consider several such scenarios. In the first scenario, the electron-phonon interaction g is abruptly switched on. Namely, the system is prepared at time $t = 0$ in a disentangled state that consists of the filled Fermi sea of the conduction electrons and some arbitrary phononic state. The electron-phonon interaction is then abruptly switched on and the system evolves according to the full Hamiltonian. Our interest is to follow the response of the system to that quench and examine the relaxation back to equilibrium. Of special interest are the time scales and frequencies that characterize the response of different observables to the quench, and their dependence on parameters such as the coupling constant g. In addition, we can investigate the role of the initial state in which the system is prepared prior to the quench. The ability to explore the effect of different initial states on the behavior of the observables in question is an advantage of the solution based on scattering states, whereas the Keldysh approach to nonequilibrium restricts the initial state of the system to be a thermal one.

The response of the system is found to be characterized by a softened frequency ω and a decay time τ which can be computed from the Green function $g(z)$. The expectation value of the phononic occupancy $\langle \hat{n}_b \rangle$, plotted in Fig. 1, oscillates with frequency 2ω and decays with the relaxation time $\tau/2$ whereas the displacement of the phonon oscillates with frequency ω and decays with time τ. This distinction, which is common to all scenarios considered, stems from the nature of the eigenmode expansion: quantities that are linear in the eigenmodes will oscillate and decay with frequency ω and time τ while quantities that are quadratic in the eigenmodes will oscillate twice as fast and decay twice as rapidly.

As a second scenario we consider an abrupt change of the energy level ϵ_d: At $t = 0$ the system occnpies the ground state of the Hamiltonian with $\epsilon_d = 0$ when the value of ϵ_d is abruptly changed to some nonzero value. This scenario, which breaks particle-hole symmetry, can be realized experimentally as the energy level ϵ_d can be controlled quite efficiently using suitable gate voltages. The breaking of particle-hole symmetry unpins the level occupancy $n_d = \langle \hat{n}_d \rangle$ from one-half and the phononic displacement from zero. The response of n_d to the quench, plotted in Fig. 2, consists of short-time dynamics, mediated by the conduction electrons occupying the levels near the cutoff Γ, followed at longer times by damped oscillations with frequency ω and the decay time τ.

The last quench scenario we consider is an abrupt change of the phonon frequency ω_0. In this scenario the system is initially held at the ground state of the full Hamiltonian with some ω_0 when the phonon frequency is abruptly changed to $\omega_1 = \omega_0 + \delta\omega$. The system then relaxes back to equilibrium, at which point physical quantities obtain the same values they would have had had the system been originally prepared with the phonon frequency ω_1. This serves again as a demonstration of the thermalization property of the system, as the final state is independent of the preparation process.

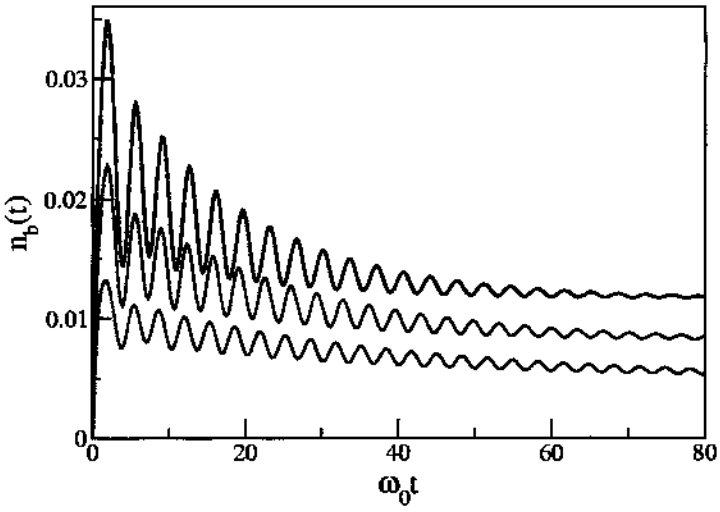

Figure 1: Time evolution of the phonon occupancy $n_b(t)$ following an abrupt switching on of the electron-phonon coupling g at time $t = 0$, starting from an empty phonon state. Here g/Γ equals 0.229 (green), 0.28 (red) and 0.324 (black).

2.3 Driven Dynamics

Quantum control of nanodevices often involves driven dynamics, where a periodic drive is applied to the system. Such cases are usually a challenge to describe since the system not only remains permanently far from equilibrium but it never even reaches steady state. Remarkably, our approach allows us to describe a rather broad class of driven dynamics where the drive couples linearly to the bosonic degrees of freedom. This class includes at least two physically relevant scenarios where periodic forcing with frequency Ω is applied either to the localized phonon or the electronic level.

The response of the system to such drives is characterized by a transition between a combination of several components oscillating with different frequencies at short times ($t \ll \tau$) and an oscillation with a single frequency at long times ($t \gg \tau$). Quantities that are linear in the bosonic eigenmodes, such as the phonon displacement and the level occupancy, have a short-time behavior that is composed of two components oscillating with frequencies ω and Ω while at long times only the latter component survives. Quantities that are quadratic in the bosonic eigenmodes, e.g., the phononic occupancy, have four oscillating components at short times, oscillating with frequencies 2ω, $\Omega \pm \omega$ and 2Ω, where once again only the latter component survives at long times. The fact that the system oscillates with a single frequency at long times, instead of developing all harmonics of the driving frequency Ω, is due to the restriction to the weak-coupling regime. We expect all harmonics to appear as the electron-phonon interaction is increased beyond the applicability of our approach.

The amplitude of the oscillations at long time depends not only on the strength of the driving force but also on the driving frequency Ω, with a sharp resonance at $\Omega = \omega$ and a vanishing amplitude at $\Omega = \omega_0$. In Fig. 3 we have plotted the amplitude of the long-time oscillations of the level occupancy as a function of the driving frequency, when the drive

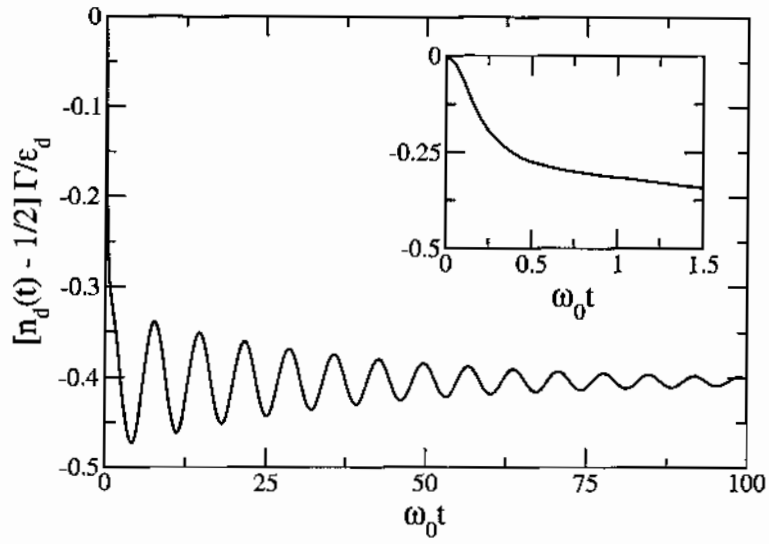

Figure 2: Time evolution of the occupancy $n_d(t)$ of the localized electronic level, following an abrupt change in its energy from $\epsilon_d = 0$ to $\epsilon_d \neq 0$. Here $g/\Gamma = 0.324$. Note that $n_d(t) - 1/2$ depends linearly on ϵ_d. Inset: A zoom on the short-time behavior which is well described by a Lorentzian form (not shown).

is applied to the level itself.

3 Aharonov-Bohm interferometer with a molecular bridge placed on one of its arms

Following the study of quench and driven dynamics in a single-molecule transistor our next goal is to apply the same methodology to another setting of current interest — an Aharonov-Bohm interferometer with a single-electron transistor embedded in one of its arms (see Fig. 4 for a schematic sketch of the device). The study of Aharonov-Bohm interferometers with a quantum dot in one of its arms has been a thriving field of research over the past fifteen years [26], driven by the measurements of phase rigidity [27] and phase lapses [28] in the transmitted current. By replacing the quantum dot with single-molecule transistor one can extend these studies to the effect of the electron-phonon coupling. The corresponding Hamiltonian reads

$$\begin{aligned}\mathcal{H} &= -iv_F \sum_{\alpha=L,R} \int_{-\infty}^{\infty} \psi_\alpha^\dagger(x)\partial_x\psi_\alpha(x)dx + \omega_0 b^\dagger b + \epsilon_d d^\dagger d + g(b^\dagger + b)(d^\dagger d - 1/2) \\ &+ \sum_\alpha t_\alpha\{\psi_\alpha^\dagger(0)d + d^\dagger\psi_\alpha(0)\} + t_{LR}\left\{e^{i\varphi}\psi_L^\dagger(0)\psi_R(0) + e^{-i\varphi}\psi_R^\dagger(0)\psi_L(0)\right\}, \qquad (13)\end{aligned}$$

where $\psi_\alpha^\dagger(x)$ is the field operator for lead α, t_{LR} is the direct tunneling amplitude between the leads representing the lower (free) arm of the interferometer, and $\varphi = 2\pi\Phi/\Phi_0$ is the flux Φ threading the ring measured in units of the flux quantum $\Phi_0 = hc/e$.

We plan to consider two variants of this setting, distinguished by how efficiently is the vibrational mode coupled to additional environment degrees of freedom other than the

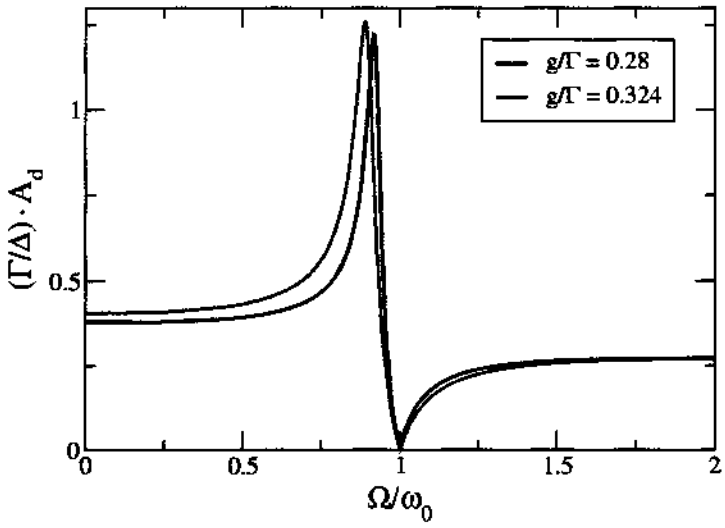

Figure 3: The asymptotic long-time amplitude of oscillations A_d of the level occupancy vs. the driving frequency Ω for two different strengths of the electron-phonon coupling g.

electronic ones specified in Eq. (13). In the first case the vibrational mode on the molecule will be taken to be isolated from the environment, while in the second scenario it will be assumed to be coupled to its own bosonic bath, which may represent, e.g., the substrate phonons. In the latter case the Hamiltonian of Eq. (13) should be supplemented with the Hamiltonian of the bosonic bath and its coupling to the molecular vibrational mode, for which we shall use the generic Hamiltonian

$$\mathcal{H}_{\rm bath} = \sum_n \omega_n \beta_n^\dagger \beta_n + (b^\dagger + b) \sum_n \lambda_n \left(\beta_n^\dagger + \beta_n\right). \tag{14}$$

Here $\beta_n^\dagger$ represent the bosonic modes of the bath, coupled by displacement to the phonon $b^\dagger$. All information of the coupling to the bath is contained in the coupling function

$$J(\omega) = \sum_n \lambda_n^2 \delta(\omega - \omega_n), \tag{15}$$

for which we shall take the standard power-law form

$$J(\omega) = 2\pi\alpha\omega_c \left(\frac{\omega}{\omega_c}\right)^s \theta(\omega_c - \omega) \tag{16}$$

with the high-energy cutoff ω_c. The power $s = 1$ corresponds to an ohmic bath.

A separate question pertains to each variant of the device. In the isolated case we are interested in the conductance in response to a chemical potential difference between the leads, and in particular in the effect of the electron-phonon coupling on it. In the case of a bosonic bath we seek to compute the heat current that is exchanged between the bosonic and fermionic baths when a temperature gradient is applied between the two. Both questions have attracted considerable interest lately [29, 30, 31].

Focusing again on the limit of a broad level we succeeded, as a first step, to map each of the two variants of the Aharonov-Bohm interferometer onto a continuum-limit Hamiltonian similar to that of Eq. (1), which in turn can be mapped using bosonization onto a

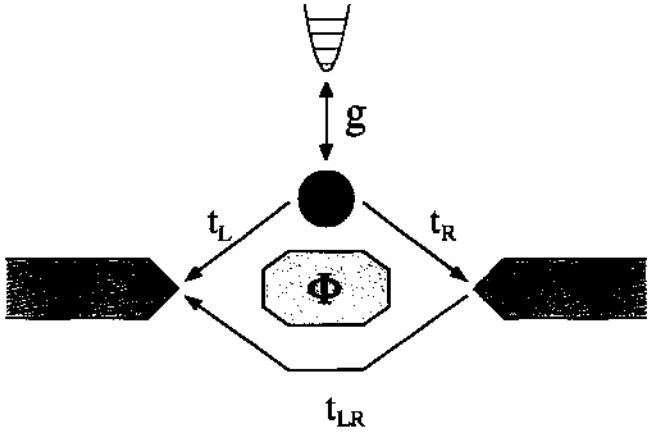

Figure 4: Schematic sketch of an Aharonov-Bohm Interferometer with a Single-Molecule Transistor Embedded in One of its Arms.

form quadratic in bosons. In case of coupling to a bosonic bath our mapping supports a finite temperature gradient between the electronic and bosonic baths, providing thereby a rare example where asymptotically exact results can be obtained for the nonlinear heat transport in an interacting nanodevice. Our current goal is to complete the calculation of the quantities of interest and their analysis along the lines described below.

In the isolated case, the zero-temperature conductance can be related to the retarded dot Green function, evaluated at zero energy [32]. The latter can be related, through the Friedel-Langreth sum rule [33], to the occupancy of the dot, which can be computed from our solution. The case involving the bosonic bath requires application of the Keldysh technique in order to calculate the relevant Green functions pertaining to the heat flow. Taking advantage of the quadratic nature of the bosonized Hamiltonian these functions can be computed exactly.

4 Numerical Evaluation of Quench Dynamics

In section 2 we described our asymptotically exact approach for computing the nonequilibrium dynamics of a single-molecule transistor. This solution provides both analytical predictions and an analytical framework for analyzing and interpreting such dynamics. The solution is confined, however, to the limit of a broad level and near-resonance conditions. In a subsequent study we plan to extend our investigation to arbitrary couplings. To this end, we plan to employ the time-dependent numerical renormalization group [9, 10], which is a powerful tool for tracking the quench dynamics of quantum impurity systems. Our main goal is to examine which aspects of our solution persist away from weak coupling, and what are the new qualitative features that are introduced as the electron-phonon coupling is increased. Particularly interesting is the regime where the phonon is strongly damped, where we expect a significant change in the damped oscillations depicted in Fig. 1.

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