Real-Time Path Integral Approach to Nonequilibrium Many-Body Quantum Systems

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A real-time path-integral Monte Carlo approach is developed to study the dynamics in a many-body quantum system coupled to a phonon background until reaching a nonequilibrium stationary state. The approach is based on augmenting an exact reduced equation for the evolution of the system in the interaction picture which is amenable to an efficient path integral (worldline) Monte Carlo approach. Results obtained for a model of inelastic tunneling spectroscopy reveal the applicability of the approach to a wide range of physically important regimes, including high (classical) and low (quantum) temperatures, and weak (perturbative) and strong electron-phonon couplings.

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In recent years there has been considerable interest in the study of quantum mechanical systems at the nanometer scale that are driven out of equilibrium. Experimental breakthroughs on transport in molecular junctions have uncovered fascinating behavior in molecular systems far from equilibrium [1,2]. Much attention has been given to the study of the transport through strongly correlated systems in which electron correlations are dominant and lead to interesting physics such as the nonequilibrium Kondo effect [3–6] and Coulomb blockade [7] in quantum dots or molecules, tunneling in a Luttinger liquid [8,9], or inelastic effects induced by electron-phonon interactions [10] as probed by inelastic electron tunneling spectroscopy [11].

Exact theoretical treatment of such many-body systems in contact with a phonon background is rather sparse and includes only a small class of simplified model problems [12–14]. For a general solution mean field equations based on the many-body nonequilibrium Green's function (NEGF) approach can be formulated [15]. However, these approaches are quite limited since in many cases they are based on a perturbative scheme and the inclusion of higher order corrections is not always clear or systematic. Numerical approaches include auxiliary field Monte Carlo (MC) calculations [16,17] and a MC diagrammatic expansion of the partition function [18–20]. However, both approaches have only been applied to imaginary-time calculations. Thus, the development of a general approach suitable for the treatment of nonequilibrium many-body quantum systems with both bosonic as well as fermionic degrees of freedom (DOF) remains a grand challenge.

In this Letter we present a novel approach aimed at obtaining exact numerical results for various dynamical quantities such as the current, conductance, dot population, etc., in a many-body quantum system that is driven out of equilibrium. We focus on a well-studied model of inelastic tunneling spectroscopy [21–24], where a quantum dot is coupled to two fermionic reservoirs representing the left and right leads at chemical potentials of μ_L and μ_R, respectively, and to a bosonic bath representing the phonon environment. Motivated by the success of real-time path-integral Monte Carlo (PIMC) techniques developed for dissipative systems [25–28] and the diagrammatic MC approach for imaginary-time many-body systems [18–20], we combine both procedures and formulate an exact real-time path integral (PI) representation for the dynamical quantity of interest. To reduce the computational complexity we integrate out the fermionic leads and the bosonic environment and obtain expressions for their corresponding influence functionals [29,30]. We develop an adequate MC procedure where we propagate the density matrix from an initial factorized condition towards a steady-state.

A nonequilibrium noninteracting quantum dot coupled to a phonon environment can be described by the Hamiltonian

$$\begin{aligned} H &= H_{LR} + H^{(I)}_{D,LR} + H_D + H^{(I)}_{D,\mathrm{Ph}} + H_{\mathrm{Ph}} \\ &= \sum_{k\in L,R} \hbar\epsilon_k a_k^\dagger a_k + \sum_{k\in L,R} (t_k a_k^\dagger d + \mathrm{H.c.}) + \hbar\epsilon_D d^\dagger d \\ &\quad + d^\dagger d \sum_\alpha M_\alpha (b_\alpha^\dagger + b_\alpha) + \sum_\alpha \hbar\omega_\alpha (b_\alpha^\dagger b_\alpha + {}^1/_2). \end{aligned} \tag{1}$$

The time-dependent current from the left lead onto the dot can be written as

$$\begin{aligned} I_L(t) &= -e\frac{d}{dt}\langle N_L(t)\rangle = -2e\mathrm{Im}\sum_{k\in L}\frac{t_k}{\hbar}\langle a_k^\dagger(t) d(t)\rangle \\ &= -2e\mathrm{Im}\sum_{k\in L}\frac{t_k}{\hbar} e^{i\epsilon_k t}\mathrm{tr}\{W_0 U_I^\dagger(t) a_k^\dagger d_{H_0}(t) U_I(t)\}, \end{aligned} \tag{2}$$

where $W_0 = W^{(0)}_{LR} \times W^{(0)}_{D,\mathrm{Ph}}$ is the initial factorizing preparation, $U_I(t) = e^{iH_0 t/\hbar} e^{-iHt/\hbar}$ is the interaction picture time evolution operator with $H_0 = H_{LR} + H_{D,\mathrm{Ph}} = H_{LR} + H_D + H^{(I)}_{D,\mathrm{Ph}} + H_{\mathrm{Ph}}$, and $d_{H_0}(t) = e^{iH_0 t/\hbar} d e^{-iH_0 t/\hbar}$.

We present the approach for a quantum dot which is initially empty; the expressions for the case of an initially occupied dot (or a mixed preparation) can be obtained straightforwardly, as well as those for the right current

$I_R(t)$, the conductivity $g(t)$, the dot's population $P(t)$ etc. After expressing the time evolution operators in Eq. (2) by virtue of Dyson series, $I_L(t)$ can be written as an infinite sum over time-ordered integrals,

$$I_L(t) = 2e \sum_{n,n'=0}^{\infty} (-1)^{n+n'} \mathrm{Re} \int_0^t ds_{2n+1} \int_0^{s_{2n+1}} ds_{2n} \cdots \times \int_0^{s_2} ds_1 \int_0^t ds'_{2n'} \int_0^{s'_{2n'}} ds'_{2n'-1} \cdots \times \int_0^{s'_2} ds'_1 \mathcal{L}(\{\tilde{s}_j\}) \mathcal{G}(\{\tilde{s}_j\}),$$

$$\mathcal{L}(\{\tilde{s}_j\}) = \sum_{\{k_j\}} \exp\left\{ \sum_{j=1}^{2(n+n'+1)} (-1)^j \epsilon_{k_j} \tilde{s}_j \right\} \left[\prod_{j=1}^{n+n'+1} \frac{t_{k_{2j-1}}}{\hbar} \frac{t^*_{k_{2j}}}{\hbar} \right] \times \mathrm{tr}_{LR}\left\{ W^{(0)}_{LR} \left[\prod_{j=1}^{n+n'+1} a^\dagger_{k_{2j-1}} a_{k_{2j}} \right] \right\},$$

$$\mathcal{G}(\{\tilde{s}_j\}) = \mathrm{tr}_{D,\mathrm{Ph}}\left\{ W^{(0)}_{D,\mathrm{Ph}} \prod_{j=1}^{n+n'+1} d_{H_{D,\mathrm{Ph}}}(\tilde{s}_{2j-1}) d^\dagger_{H_{D,\mathrm{Ph}}}(\tilde{s}_{2j}) \right\}. \quad (3)$$

Here, $d_{H_{D,\mathrm{Ph}}}(t) = e^{iH_{D,\mathrm{Ph}}t/\hbar} d e^{-iH_{D,\mathrm{Ph}}t/\hbar} = d_{H_0}(t)$, and $\{\tilde{s}_j\} = \{s'_1, \ldots, s'_{2n'}, t, s_{2n+1}, \ldots, s_1\}$ is a contour-ordered sequence of time points residing on the forward (s_j) and backward ($s'_{j'}$) time axis. Without the presence of the phonons, Eq. (3) essentially resembles a continuation of the diagrammatic expansion of the partition function [18,19,31] from imaginary into real time. Analogously, the trace over the leads DOF can now be performed exactly, yielding

$$\mathcal{L}(t_1, \ldots, t_{2N}) = i^N \det(M), \quad (4)$$

where M is a matrix with elements $M_{ij} = [\Sigma^<_L(t_{2j-1} - t_{2i}) + \Sigma^<_R(t_{2j-1} - t_{2i})(1 - \delta_{t_{2j-1},t})(1 - \delta_{t_{2i},t})]$ for $j \le i \le N$ or $[\Sigma^>_L(t_{2j-1} - t_{2i}) + \Sigma^>_R(t_{2j-1} - t_{2i})(1 - \delta_{t_{2j-1},t}) \times (1 - \delta_{t_{2i},t})]$ for $j > i$. Here, $\Sigma^<_{L,R}(t)$ ($\Sigma^>_{L,R}(t)$) denotes the leads' lesser (greater) self energy in the time domain. $\mathcal{G}$ in Eq. (3) represents a $2(n + n' + 1)$-point correlation function of the dot-phonon subsystem along the Kadanoff-Baym contour $\mathcal{C}$:$s \in 0 \to t \to 0$. It is most conveniently evaluated in the framework of Feynman PIs, which allow to integrate out the phonon DOF exactly [29]. For this purpose, we first introduce the *dot path variables* $\sigma(s)$ and $\sigma'(s)$ denoting the state of the dot along the forward and backward branches of $\mathcal{C}$, respectively, with $\sigma^{(\prime)}(s) = 0$ (1) referring to an empty (occupied) dot. These dot forward and backward paths $\sigma(0 \to t)$ and $\sigma'(t \to 0)$, respectively, are uniquely defined by $\{\tilde{s}_j\}$, with

$$\sigma(s) = [1 - (-1)^{\sum_{j=1}^{2n+1} \theta(s - s_j)}]/2 \quad (5)$$

and $\sigma'(s)$ accordingly (cf. Fig. 1). Transitions between the two dot states can only be induced by the interaction part $H^{(I)}_{D,LR}$ acting at the times $\{\tilde{s}_j\}$. The latter thus represent the *kink times* of the dot path [32,33], between which $\sigma^{(\prime)}(s)$ remains constant; in this spirit, Eq. (3) closely resembles

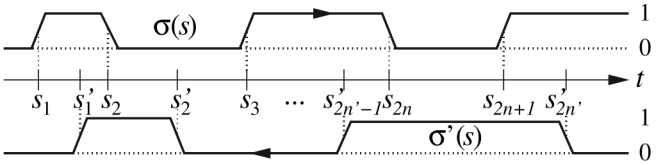

FIG. 1. Dot forward and backward paths $\sigma(0 \to t)$ and $\sigma'(t \to 0)$, respectively, and kink times $\{\tilde{s}_j\} = \{s_j, s'_j\}$.

the instanton expansion of the partition function in the spin-boson model [32]. In terms of σ and σ', $\mathcal{G}$ can now be expressed as

$$\mathcal{G} = \exp\left\{ i\epsilon_D \int_0^t ds[\sigma(s) - \sigma'(s)] \right\} \mathcal{F}[\sigma, \sigma'], \quad (6)$$

where $\mathcal{F}$ denotes the Feynman-Vernon influence functional [29] summarizing the influence of the phonons for an arbitrary distribution of the parameters M_α and ω_α.

While the expressions (4) and (6) for $\mathcal{L}$ and $\mathcal{G}$ allow for a rather compact expression of $I_L(t)$, they introduce retardation effects which are arbitrarily long ranged in time, making analytical progress rather cumbersome. To allow for a numerical evaluation of Eq. (3), the integrals therein are approximated according to

$$\int_0^t ds_N \int_0^{s_N} ds_{N-1} \cdots \int_0^{s_2} ds_1 f(s_1, \ldots, s_N) \simeq \tau^N \sum_{j_N=1}^{q} \sum_{j_{N-1}=1}^{j_N - 1} \cdots \sum_{j_1=1}^{j_2 - 1} f(j_1\tau, \ldots, j_N\tau), \quad (7)$$

where $\tau = t/q$. The systematic errors can be made arbitrarily small by increasing q accordingly. Performing the sums over all kink numbers and (discretized) kink times is now equivalent to summing over all possible (discretized) dot paths $\{\sigma_j = \sigma(j\tau), \sigma'_{j'} = \sigma'(j'\tau)\}$, such that Eq. (3) can be written as a discretized PI,

$$I_L(t) = 2e \sum_{\{\sigma_j, \sigma'_{j'}\}} (-\tau)^{\bar{n}-1} \mathcal{L}(\{\sigma_j, \sigma'_{j'}\}) \mathcal{G}(\{\sigma_j, \sigma'_{j'}\}), \quad (8)$$

where $\bar{n}$ denotes the number of kinks of the path $\{\sigma_j, \sigma'_{j'}\}$. Equation (8) can now readily be evaluated by means of PI (or worldline) MC calculations [28,33]. Note that while the systematic error can be avoided by employing continuous worldline MC calculations, the discretization scheme allows to compute and store all quantities needed to evaluate $\mathcal{L}$ and $\mathcal{G}$ prior to performing the MC moves, resulting in a very efficient sampling procedure.

We now turn to discuss the application of the proposed approach to the model system described by the Hamiltonian (1). In the present approach the effect of the leads is fully determined by the self energies $\Sigma^{</>}_{L/R}(t)$ [cf. Eq. (6)], which are defined in terms of $\Gamma(\epsilon) = \Gamma_L(\epsilon) + \Gamma_R(\epsilon)$ in Fourier space [15]. $\Gamma(\epsilon)$ is taken to be energy independent (wide band limit) with a soft cutoff at $\epsilon = \pm\epsilon_c$: $\Gamma_{L/R}(\epsilon) = \frac{\Gamma_{L/R}}{[1+e^{\nu(\epsilon-\epsilon_c)}][1+e^{-\nu(\epsilon+\epsilon_c)}]}$. In all results pre-

sented below, $\Gamma_L = \Gamma_R$, $\nu = 5\Gamma = 5(\Gamma_L + \Gamma_R)$, and $\epsilon_c = 10\Gamma$ or 20Γ to converge the results. Similarly, the phonon influence functional $\mathcal{F}$ is completely specified by the phonon spectral density [29], $J(\omega) = \frac{\pi}{\hbar^2}\sum_\alpha M_\alpha^2 \delta(\omega - \omega_\alpha)$. For a single phonon coupled to a secondary bath via a coupling constant γ, $J(\omega)$ becomes a Lorentzian: $J(\omega) = \frac{\gamma\omega}{[(\omega/\omega_0)^2-1]^2+[\hbar^2\gamma\omega_0\omega/(2M_0^2)]^2}$ [34]. We note in passing that the proposed simulation scheme does neither depend on the particular form of $\Sigma_{L/R}^{</>}(t)$ nor of $J(\omega)$.

Figure 2 shows the time-dependent current and dot population for different values of the model parameters for $T < \Gamma$ (quantum regime) and $M_0 > \Gamma$ (strong coupling). $I_{L/R}(t)$ (lower panels) are characterized by damped coherent oscillation with a long time exponential decay to a steady-state value. While the steady-state current can be extracted from an exponential fit to $I_{L/R}(t)$, the average current $I(t) = \frac{1}{2}[I_L(t) + I_R(t)]$ (upper left panel) typically decays much faster to steady state, such that in most cases it could be obtained as the corresponding plateau value. Our MC scheme provides converged results despite the notorious fermionic and dynamical sign problems. While the former has been reported to be practically absent in imaginary-time diagrammatic MC calculations [18,19], the latter is diminished by decoherence arising from the influence of the leads and the bosonic bath.

The fact that $I(t)$ approaches faster to steady state than $I_{L/R}(t)$ is consistent with other flux-based methods [35] and can be rationalized by looking at dynamical fluctuations under equilibrium conditions. This is depicted for the case where $\mu_L = \mu_R$ and the system decays to equilibrium. $I_{L/R}(t)$ exhibit pronounced coherent oscillations with finite values even at $t > 5\hbar/\Gamma$ while their average value vanishes for all times.

In Fig. 3 we plot the steady-state current I as a function of the bias $\mu_L - \mu_R = \mathrm{eV}$ for different electron-phonon couplings M_0 (lower panel), phonon frequencies ω_0 (middle panel), and couplings γ to a secondary bath (upper panel) for $k_BT < \Gamma$ (quantum regime), covering a wide parameter range from the Landauer inelastic case through the perturbative regime to the strong coupling limit. When the coupling γ between the primary oscillator and the secondary phonon bath is small we observe steps in $I(V)$ at integer values of $\mathrm{eV} = 2\hbar\omega_0$ [10]. As γ increases or for an Ohmic spectral density these steps diminish and eventually disappear, signifying the wide range of phonon frequencies contained in the spectral density. As M_0 increases the value of the current decreases from the Landauer inelastic single-channel value to lower values. Our approach clearly captures elastic effects at all values of $M_0 > 0$ as depicted by the lower values of the current and by the steps at twice the frequency of the primary phonon mode. Comparing the numerically exact PIMC results to an approximate method which is based on a generalization of the single particle approximation [36] to include the leading order term of the Fermi sea [21], we find quantitative agreement for small values of the voltage corresponding to the first step of $I(V)$. For larger values,

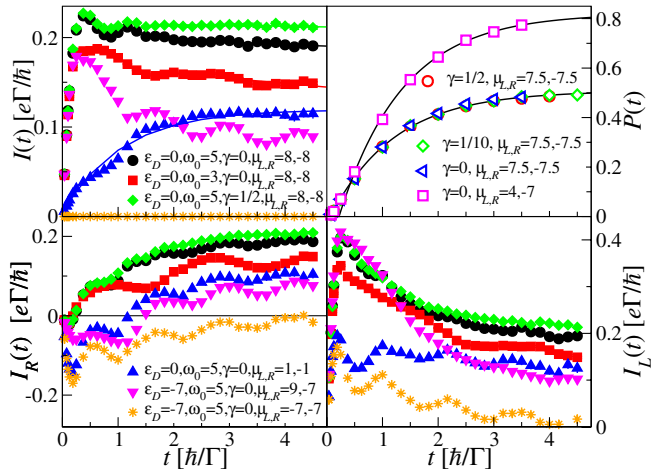

FIG. 2 (color online). Plots of $I_L(t)$ (lower left panel), $I_R(t)$ (lower right panel), average current $I(t)$ (upper left panel), and $P(t)$ (upper right panel) for $M_0 = 4$ and $k_BT = \frac{1}{5}$, in units of Γ.

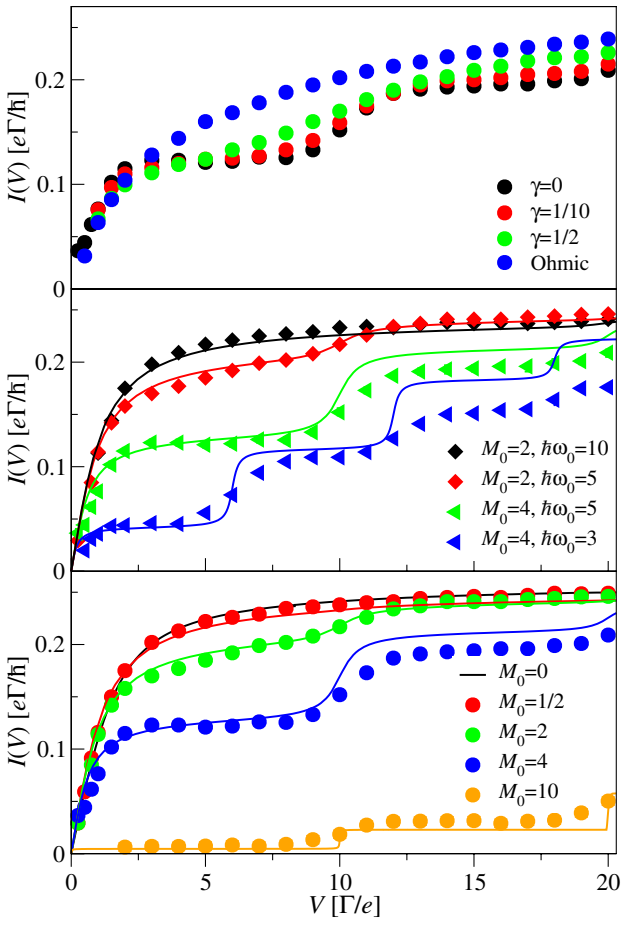

FIG. 3 (color online). Plots of the steady-state current I as a function of the bias voltage for $\hbar\epsilon_D = 0$, $\mu_{L,R} = \frac{\mathrm{eV}}{2}, -\frac{\mathrm{eV}}{2}$, and $k_BT = \frac{1}{5}$, in units of Γ. Circles, diamonds, and triangles denote QMC data while lines refer to the approximation of [21].

however, we observe significant deviations for $M_0/\Gamma \geq 4$. These deviations signify the importance of high order effects of the Fermi sea to the transport process, which are naturally included in the real-time PIMC approach. We note that other approximate methods, not discussed here, can also be applied to calculate for the steady-state current (see, e.g., Refs. [37,38]).

In conclusion, we have developed a novel real-time PIMC approach to study the dynamics in open quantum systems that are driven out of equilibrium. The approach is based on expressing the time evolution by virtue of Dyson series, before reducing the dynamics of the entire system by integrating out the fermionic and bosonic baths and introducing exact influence functionals. The remaining infinite sum over contour-ordered time integrals is then evaluated by PI (worldline) MC calculations. We believe that our approach is capable of resolving several shortcomings found in currently used approaches. First, it is not based on any perturbative treatment and can provide exact numerical results. Second, the real-time propagation scheme allows to study transient phenomena and time scales and also to include time-dependent fields. Finally, it can be applied to a general many-body problem, as long as an efficient sampling strategy can be found.

We have applied the approach to calculate the time-dependent current in a well-studied model of inelastic tunneling spectroscopy, where a quantum dot is coupled to two fermionic leads and to a bosonic phonon bath. Numerical results indicate that the approach is robust and can be used for a wide range of model parameters spanning the classical to quantum limits, a range of experimentally accessible chemical bias, different phonon frequencies, weak to strong electron-phonon couplings, a wide range of couplings between the primary phonon mode and a secondary phonon bath, and an arbitrary spectral density of the latter. The approach is still limited for regimes where the steady-state time scale and/or the decay time of coherent oscillations are similarly stretched. In addition, we expect an exponential scaling with number of electronic DOF, and correspondingly poor statistics. However, these limitations can be substantially attenuated by exploiting the large body of existing MC schemes for dissipative systems [25,26,28] combined with the advantages of diagrammatic MC approach for the fermionic sign problem [18,19].

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