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Driven tunneling dynamics: Bloch-Redfield theory versus path-integral approach

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In the regime of weak bath coupling and low temperature we demonstrate numerically for the spin-boson dynamics the equivalence between two widely used but seemingly different roads of approximation, namely, the path-integral approach and the Bloch-Redfield theory. The excellent agreement between these two methods is corroborated by an efficient *analytical* high-frequency approach: it well approximates the decay of quantum coherence via a series of damped coherent oscillations. Moreover, a suitably tuned control field can selectively enhance or suppress quantum coherence.

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The dynamics of driven quantum systems which interact with a large number of environmental degrees of freedom [1–3] plays an increasingly prominent role: its vast applicability ranges from tunneling phenomena in solid state physics, the study of electron and proton transfer in condensed phases, to the gate operation in quantum computing devices [4], to name but a few. In particular, the use of properly tailored external driving forces enables one to selectively manipulate a quantum transport process. The various communities typically rely on different methods of description. The two most popular approaches for a portrayal of the time evolution of the corresponding reduced density matrix (RDM) are either based on the system-bath coupling expansion obtained by use of a projector operator method (commonly known as the *Bloch-Redfield formalism*), or on the expansion in the coupling matrix element Δ (such as a tunnel splitting) by use of (real-time) *path-integral methods*. Nevertheless, there exists practically little crosstalk between the practitioners of the two approaches, and even more, not much of detailed comparison between the two seemingly different roads of approximation needed for practical calculations.

For the archetype quantum system of a driven spin-boson dynamics, namely, the driven dissipative two-state system (TSS) dynamics (TSS) [3], the application of the so termed noninteracting blip approximation (NIBA), i.e., the leading order result in the tunnel coupling Δ^2, produced many impressive successes in entangling the complexity of driven open quantum systems. This scheme works best in the regime of strong friction and/or high thermal temperatures. Much less is presently known, however, about the corresponding complexity of the driven dynamics in the deep quantum regime at low temperatures and weak system-bath coupling, where the NIBA is failing and higher order terms in the series in Δ must be accounted for [5,6]. In practice, this latter regime is of relevance for many situations such as, e.g., for the challenge of ''battling decoherence'' in quantum computing schemes [4].

Our main objective with this work is to enlight the advantages and disadvantages of the two approaches. In doing so we present three major findings: (i) We numerically demonstrate the equivalence for the driven tunneling dynamics between the path-integral method beyond NIBA and the coupled set of nonstationary, Markovian Bloch-Redfield equations. (ii) Starting from the generalized master equation (GME) for the RDM, obtained within the path-integral approach, we arrive at an *analytic* high driving frequency approximation that compares well with comprehensive numerical findings. (iii) With this analytical result one can efficiently determine the optimal control of quantum coherence.

Our starting point is the *driven* spin-boson Hamiltonian [3] where the TSS is bilinearly coupled to an ensemble of harmonic oscillators, i.e.,

$$\hat{H}(t) = -\hbar[\Delta\hat{\sigma}_x + \epsilon(t)\hat{\sigma}_z]/2 + \sum_i \hbar\omega_i(\hat{b}_i^\dagger\hat{b}_i + 1/2) + \hat{\sigma}_z \sum_i c_i(\hat{b}_i + \hat{b}_i^\dagger)/2, \qquad (1)$$

with $\hat{\sigma}_i$ being Pauli spin matrices. Here Δ describes the coupling between the two states, and $\epsilon(t)$ is the external, time-dependent control field. The basis states are chosen such that $|R\rangle$ (right) and $|L\rangle$ (left) are the localized eigenstates of the ''position'' operator $\hat{\sigma}_z$. All effects of the Gaussian bath on the TSS are captured by the force autocorrelation function [1–3] $\mathcal{M}(t) = (1/\pi)\int_0^\infty d\omega J(\omega)[\cosh(\hbar\omega/2k_BT - i\omega t)/\sinh(\hbar\omega/2k_BT)]$, where the spectral density of the environment, $J(\omega) = \pi\hbar^{-2}\Sigma_i c_i^2 \delta(\omega-\omega_i) = 2\pi\alpha\omega e^{-\omega/\omega_c}$, is assumed to be of Ohmic form with exponential cutoff and dimensionless coupling strength α. The dynamical quantities of interest are the expectation values $\sigma_i(t) := \mathrm{Tr}\{\hat{\rho}(t)\hat{\sigma}_i\}$ which, together with the unit matrix $\hat{I}$, comprise the complete reduced density matrix $\hat{\rho}(t) = \hat{I}/2 + \Sigma_{i=x,y,z}\sigma_i(t)\hat{\sigma}_i/2$. In the following we assume that at time $t=0$ the particle is held at the right site $\sigma_z = +1$, with the bath being in thermal equilibrium.

Path-integral approach. For a harmonic bath the exact formal solution for the evolution of the $\sigma_i(t)$ can be expressed in terms of real-time double path integrals [1–3]. This procedure yields the formally exact set of equations [3,5,6]

$$\dot{\sigma}_z(t)=\int_0^t dt'[K_z^{(-)}(t,t')-K_z^{(+)}(t,t')\sigma_z(t')],$$
$$\sigma_x(t)=\int_0^t dt'[K_x^{(+)}(t,t')+K_x^{(-)}(t,t')\sigma_z(t')], \tag{2}$$

and $\sigma_y(t)=-\dot{\sigma}_z(t)/\Delta$. Here, the kernels $K_i^{(\pm)}$, $i=x,z$ are found in the form of a series expansion in Δ. Because the exact series expression cannot be evaluated to all orders, approximation schemes necessarily must be invoked. A familiar scheme is the noninteracting-blip approximation (NIBA) [1–3], which corresponds to a truncation of the series expansion to lowest order in Δ. The NIBA is approximatively valid only for the dynamics of $\sigma_z(t)$ if on average $\langle\epsilon(t)\rangle=0$. However, in the presence of a static asymmetry component, it breaks down for weak damping *and* low temperatures [2,3]. A systematic weak damping approximation for the kernels $K_i^{(\pm)}$ in Eq. (2), which circumvents the weaknesses of the NIBA has been discussed in [5,6]. By keeping track of the bath-induced correlations to *linear* order in α, the whole series expansion in Δ can be summed analytically. The kernels $K_z^{(\pm)}$ read

$$\begin{aligned}K_z^{(+)}(t,t')=&\Delta^2\cos[\zeta(t,t')][1-Q'(t-t')]\\&+\int_{t'}^t dt_2\int_{t'}^{t_2}dt_1\Delta^4\sin[\zeta(t,t_2)]\\&\times P_0(t_2,t_1)\sin[\zeta(t_1,t')][Q'(t-t')\\&+Q'(t_2-t_1)-Q'(t_2-t')-Q'(t-t_1)],\\K_z^{(-)}(t,t')=&\Delta^2\sin[\zeta(t,t')]Q''(t-t')\\&-\int_{t'}^t dt_2\int_{t'}^{t_2}dt_1\Delta^4\sin[\zeta(t,t_2)]P_0(t_2,t_1)\\&\times\cos[\zeta(t_1,t')][Q''(t-t')-Q''(t_2-t')].\end{aligned} \tag{3}$$

Here, the first term in $K_z^{(\pm)}$ represents the weak-coupling form of NIBA. In the remaining contribution the term $P_0(t_2,t_1)$ accounts for all tunneling events during the time interval $[t_1,t_2]$ that are not influenced by damping. Hence, $P_0(t_2,t_1)$ solves the generalized master equation (GME) for $\sigma_z(t)$ (2) with the zero-damping kernels $K^{(+)}(t,t')=\Delta^2\cos[\zeta(t,t')]$ and $K^{(-)}(t,t')=0$, where $\zeta(t,t')=\int_{t'}^t dt''\epsilon(t'')$ captures the effects of the external force. The bath-influence is encapsulated in the functions $Q'(t)$ and $Q''(t)$ being the real and imaginary part, respectively, of the twice integrated bath correlation function $\mathcal{M}(t)$.

Bloch-Redfield formalism. In the Nakajima-Zwanzig theory [7] it is well known how to construct an exact generalized master equation for the reduced density matrix with the help of projection operators. For intermediate to high temperatures and/or strong damping, but for arbitrary driving, a master equation for $\hat{\rho}(t)$ can be obtained within the small polaron theory, yielding equations that are equivalent to the NIBA [8,9]. For weak coupling to the bath the projection operator technique yields the GME in Born approximation that can be further simplified to the Markovian kinetic equations without loss of accuracy to the leading order in dissipative coupling. For strong harmonic driving this objective was first achieved in 1964 by Argyres and Kelley [10]. Following the reasoning in [10] the kinetic equations for the RDM of a stochastically driven TSS were found in [11(a)] and in a different way in [11(b)]. Generalizing [10,11] to the case of a spin-boson problem with an *arbitrary* control field we find the coupled equations

$$\begin{aligned}\dot{\sigma}_x(t)&=\epsilon(t)\sigma_y-\Gamma_{xx}(t)\sigma_x-\Gamma_{xz}(t)\sigma_z-A_x(t),\\\dot{\sigma}_y(t)&=-\epsilon(t)\sigma_x+\Delta\sigma_z-\Gamma_{yy}(t)\sigma_y-\Gamma_{yz}(t)\sigma_z-A_y(t),\end{aligned} \tag{4}$$

with $\Gamma_{yy}(t)=\Gamma_{xx}(t)$ and $\dot{\sigma}_z=-\Delta\sigma_y$. Here the time-dependent rates $\Gamma_{ij}(t)=\int_0^t dt'\mathcal{M}'(t-t')b_{ij}(t,t')$, together with the inhomogeneities $A_x(t)=\mathrm{Im}\,F(t)$, $A_y(t)=\mathrm{Re}\,F(t)$, with $F(t)=2\int_0^t dt'\mathcal{M}''(t-t')U_{RR}(t,t')U_{RL}(t,t')$ determine the dissipative action of the thermal bath on the TSS. The functions $\mathcal{M}'$ and $\mathcal{M}''$ are the real part and imaginary part, respectively, of the correlation function $\mathcal{M}$. The quantities $U_{RR}(t,t')=\langle R|U(t,t')|R\rangle$ and $U_{RL}(t,t')=\langle R|U(t,t')|L\rangle$ are matrix elements of the time evolution operator $U(t,t')$ of the *nondissipative* driven TSS. The functions b_{ij} read $b_{xx}=|U_{RR}|^2-|U_{RL}|^2$, $b_{xz}=2\,\mathrm{Re}\,U_{RR}U_{RL}$, and $b_{yz}=-2\,\mathrm{Im}\,U_{RR}U_{RL}$. This main result in Eq. (4) yields a consistent Bloch-Redfield-type description of the externally driven spin-boson dynamics. Equations of the form (4) were derived by Bloch and Redfield in 1957 [12] to describe spin relaxation in nuclear magnetic resonance, and in [13] for the dynamics of the undriven spin-boson problem. Our set of Eqs. (4) generalizes [13] to general driving forces. Note that these derived equations are valid in the parameter region $\alpha\ln(\omega_c/\Delta)\ll1$, where the frequency corrections to the dynamics incurred due to the dissipation are small, and the perturbative treatment is fair. One can show that for the undriven case, $\epsilon(t)=\epsilon_0$, the analytic solution of Eq. (4) in first order in α reproduces the analytical path-integral weak-damping results, cf. [2,6] and Eq. (5) below with zero ac-field.

Analytic high-frequency solution. Up to now no assumptions on the deterministic control field have been made. Next, we focus our attention on a monochromatic field of the form $\epsilon(t)=\epsilon_0+s\cos\Omega t$. Moreover, we restrict our investigations on the $\sigma_z(t)$-dynamics, as this quantity is of prime interest for describing tunneling properties. Because the path-integral approach yields a closed integro-differential equation for $\sigma_z(t)$, we start from the generalized master equation (2). In the high-frequency regime $[\Omega\gg\{\Delta,\epsilon_0,\Gamma_R\}$, with Γ_R defined in Eq. (6) below] a good approximation to the dynamics of $\sigma_z(t)$ amounts to perform the substitution $\mathcal{R}[\zeta(t,t')]\to\langle\mathcal{R}[\zeta(t,t')]\rangle=J_0(\chi)\mathcal{R}[\epsilon_0(t-t')]$ into the kernels $K_z^{(\pm)}(t,t')$, where $\mathcal{R}=\cos$ or $\sin$ and $\chi=(2s/\Omega)\sin[\Omega(t-t')/2]$. Here $\langle\,\rangle$ denotes time-averaging, and J_0 is the zero order Bessel function. The resulting generalized master equation is in the form of a time-convolution. A solution is conveniently obtained by use of Laplace transformation techniques, upon generalizing a line of reasoning proposed in Ref. [5]. By expanding the Bessel function $J_0(\chi)$ in Fourier series, and upon introducing the field-dressed tunneling splittings $\Delta_n=|J_n(s/\Omega)|\Delta$ and the photon-induced asymmetries $\epsilon_n=\epsilon_0-n\Omega$, we end up with

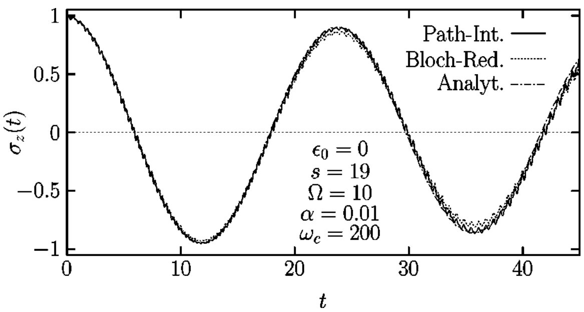

FIG. 1. Matching between path integral and Bloch-Redfield. The comparison of the dynamical Eqs. (2), (4), and (5) for unbiased TSS-dynamics depicts excellent agreement. For this resonant condition ($\epsilon_0 = n^*\Omega$, $n^* = 0$) the dynamics is well described by Eq. (5) with the single-mode frequency $\tilde{\theta}_0 = \tilde{\Delta}_0$. Here and in the following figures frequencies are expressed in units of Δ, times in units of Δ^{-1}. The temperature is zero throughout.

$$\sigma_z(t) = P_\infty + (\mathcal{P}_0 - P_\infty) e^{-\Gamma_R t} + \sum_{n=-\infty}^{\infty} \mathcal{C}_n \cos(\tilde{\theta}_n t) e^{-\Gamma_n t}. \tag{5}$$

Conservation of probability yields $\mathcal{P}_0 + \Sigma_n \mathcal{C}_n = 1$, with $\mathcal{P}_0 = \Pi_n(\epsilon_n/\theta_n)^2$, and $\mathcal{C}_n = \Pi_m(\theta_n^2 - \epsilon_m^2)/[\theta_n^2 \Pi_{m\neq n}(\theta_n^2 - \theta_m^2)]$. The damping rates and the averaged nonequilibrium value P_∞ read [14]

$$\Gamma_R = 2\sum_n \Gamma_n, \quad \Gamma_n = \frac{1}{4}\mathcal{C}_n f_n^2 S(\theta_n), \tag{6}$$

$$P_\infty = \frac{1}{2\Gamma_R} \sum_n \sqrt{\mathcal{P}_0} f_n \mathcal{C}_n J(\theta_n) \sum_m \frac{\Delta_m^2}{\theta_n^2 - \epsilon_m^2}. \tag{7}$$

Here $f_n = \sqrt{\mathcal{P}_0}\,\theta_n \Sigma_m \Delta_m^2/[\epsilon_m(\theta_n^2 - \epsilon_m^2)]$, and $S(\theta) = J(\theta)\coth(\hbar\theta/2k_BT)$. The infinite set of frequencies θ_n is determined by the pole equation for the undamped TSS

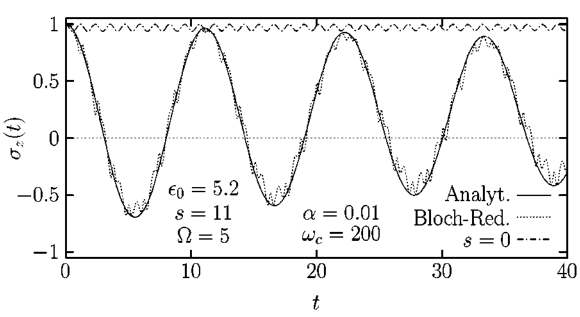

FIG. 2. Driving induced quantum coherence phenomena. In the presence of a quasiresonant high-frequency field away from the zeros of $J_{n^*}(s/\Omega)$, the population difference $\sigma_z(t)$ exhibits a coherent oscillatory decay which is dominated by a *single* mode oscillation frequency $\tilde{\theta}_{n^*}$. A comparison between the predictions of the analytical solution (5) with just the single-mode frequency $\tilde{\theta}_1$, for a near-resonant field (i.e., $n^* = 1$ with $\epsilon_1 = |\epsilon_0 - \Omega| = 0.2\Delta$) with the Bloch-Redfield result in Eq. (4) is depicted. Note that in the undriven situation ($s = 0$) the TSS dynamics is almost completely *localized.*

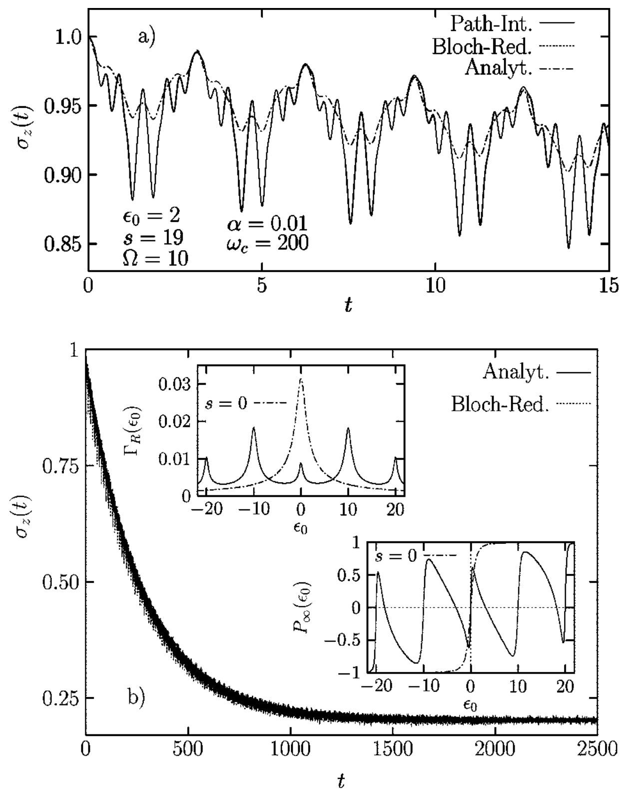

FIG. 3. Controlling tunneling. In the presence of an *off-resonance* no net separation of time scales occurs and the population $\sigma_z(t)$ shows a complex interference pattern (a). Note that the numerical solutions of Bloch-Redfield and path-integral equations coincide within linewidth. The TSS dynamics is dominated by an *incoherent* decay towards its asymptotic limit (b), so that quantum coherence is lost. The incoherent decay rate Γ_R, however, can be strongly diminished. This is demonstrated in the upper left inset where the *photon assisted decay rate* Γ_R is plotted vs the dc-bias ϵ_0. It exhibits characteristic resonance peaks at multiple integers of the driving frequency Ω. These peaks are shifted replicas of the dc-driven ($s = 0$) rate with different weights. Thus, a suitable chosen bias can enhance or suppress the decay of populations. Finally, the lower right inset shows the *averaged nonequilibrium* population difference P_∞. It exhibits a nonmonotonic dependence on the dc-bias when combined with a high-frequency field. For appropriate values of the dc-field a population inversion ($P_\infty < 0$ when $\epsilon_0 > 0$, and vice versa) can occur.

$$\prod_n (\epsilon_n^2 - \theta^2) + \sum_n \Delta_n^2 \prod_{m;m\neq n} (\epsilon_m^2 - \theta^2) = 0. \tag{8}$$

Finally, to approximately take into account bath-induced frequency-shifts the tunneling frequencies $\tilde{\theta}_n$ are evaluated from Eq. (8) upon substituting $\Delta_n \to \Delta_n[1 - \alpha\ln(\omega_c/\Delta)] := \tilde{\Delta}_n$. Thus, in this high-frequency regime the system generally still exhibits damped coherent oscillations, as in the undriven case, although, an *infinite* set of oscillation frequencies $\tilde{\theta}_n$ with corresponding damping rates Γ_n enters this driven dynamics. Superimposed to these coherent oscillations there occurs an incoherent decay with rate Γ_R towards P_∞.

In Figs. 1–3 we depict comparisons amongst the numerical predictions of the Born-Markov equations (4), the path-integral GME (2), and the analytical solution (5) for small Ohmic friction and zero temperature. For the driven dynamics the agreement is remarkable. It increases further with increasing temperature (not shown). To achieve a convergence of Eq. (5) a truncation of the pole equation (8) to five (or less, cf. Figs. 1 and 2) modes, characterized by (Δ_n, ϵ_n), turned out to be sufficient. In Fig. 1 the influence of an *unbiased* $(\epsilon_0=0)$ control field is investigated. This corresponds to a resonant $(\epsilon_0=n^*\Omega)$ field with $n^*=0$. In Figs. 2 and 3 the case of a finite bias $\epsilon_0\neq 0$ is depicted. Figure 2 depicts the near-resonant situation $|\epsilon_0-n^*\Omega|=|\epsilon_{n^*}| \ll\{\tilde{\Delta}_{n^*},|\epsilon_0|\}$ away from the zeros of $J_{n^*}(s/\Omega)$: the coherent dynamics is now already well captured by the *single resonant mode* frequency $\tilde{\theta}_{n^*}=\sqrt{\tilde{\Delta}^2_{n^*}+\epsilon^2_{n^*}}$. This finding generalizes the small-dc-bias analysis in [15]. In addition, we deduce from the parameters chosen in Fig. 2 that our approach can even work for intermediate driving frequencies $(\Omega \approx\epsilon_0)$. Due to the fact that $\tilde{\Delta}_{n^*}\leqslant\Delta$, and $\epsilon_{n^*}<\epsilon_0$, the field-induced oscillation frequency $\tilde{\theta}_{n^*}$ can be much smaller than in the undriven case. In the *off-resonance* situation $(|\epsilon_n|>\Delta_n$ for all $n)$ of Figs. 3(a) and 3(b), $\sigma_z(t)$ exhibits a complex interference pattern with quantum coherence suppressed. Moreover, the decay towards the nonstationary equilibrium value occurs on a much longer time scale as compared to the case with $s=0$. This result, observed recently in [16], can be understood via close inspection of the upper left inset in Fig. 3(b), where the averaged decay rate Γ_R is plotted versus ϵ_0. For the chosen parameters the decay rate is strongly diminished. Moreover, the lower right inset depicts the averaged nonequilibrium value P_∞ versus the dc-bias ϵ_0. Here, photon assisted tunneling rules the possible *inversion* of asymptotic population (i.e., $P_\infty<0$, for $\epsilon_0>0$, and vice versa).

In conclusion we maintain that in the perturbative regime $[\alpha \ln(\omega_c/\Delta)\ll 1]$ it is numerically advantageous to evaluate the weak coupling tunneling dynamics by using the nonstationary Markovian Bloch-Redfield equations, as compared to the non-Markovian path-integral GME. We find numerically perfect agreement as depicted with Figs. 1–3. Within the time scale of tunneling we find *no* observable non-Markovian effects. On physical grounds, the same remarks apply to the time evolution of the full density matrix. Note, however, that for the scaling regime (i.e., cutoff $\omega_c\to\infty$) Bloch-Redfield theory increasingly fails; it then is necessary to correct even the path-integral GME with an additional, concocted renormalization scheme [6]. Finally, our analytical scheme may prove prominent in order to optimize quantum coherence.

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