Eur. Phys. J. B **10**, 719–729 (1999)

THE EUROPEAN
PHYSICAL JOURNAL B

EDP Sciences

Dissipation, decoherence and preparation effects in the spin-boson system

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Received 23 October 1998 and Received in final form 26 February 1999

Abstract. The dynamics of the reduced density matrix of the driven dissipative two-state system is studied for a general diagonal/off-diagonal initial state. We derive exact formal series expressions for the populations and coherences and show that they can be cast into the form of coupled nonconvolutive exact master equations and integral relations. We show that neither the asymptotic distributions, nor the transition temperature between coherent and incoherent motion, nor the dephasing rate and relaxation rate towards the equilibrium state depend on the particular initial state chosen. However, in the underdamped regime, effects of the particular initial preparation, *e.g.* in an *off-diagonal* state of the density matrix, strongly affect the transient dynamics. We find that an appropriately tuned external ac-field can slow down decoherence and thus allow preparation effects to persist for longer times than in the absence of driving.

PACS. 05.30.-d Quantum statistical mechanics – 05.40.-a Fluctuation phenomena, random processes, noise and Brownian motion – 33.80.Be Level crossing and optical pumping

1 Introduction

The dissipative two-state system (TSS) can model a great variety of physical and chemical situations. The case in which the TSS is a spin 1/2 particle interacting with a heat bath is encountered in the context of spin magnetic resonance and relaxation [1]. A more general class concerns double-well systems where only the ground states of the two wells are occupied. The dissipative TSS can describe, *e.g.*, hydrogen tunneling in condensed media [2], tunneling of atoms between an atomic-force microscope tip and a surface [3], or of the magnetic flux in a rf-SQUID [4]. Recent experiments on submicrometer Bi wires have measured transition rates of two-level systems coupled to conduction electrons [5,6]. This model has been also applied to describe nonadiabatic chemical reactions in the condensed phase, such as electron transfer [7–11] or proton transfer reactions [12,13]. In these particular cases, the TSS describes the electronic or protonic motion between two diabatic potential energy surfaces. When the polarization of the fictitious spin is bilinearly coupled to a harmonic bath, one ends up with the so-termed spin-boson model [14–16]. For a symmetric bistable potential this problem is analogous to that of a spin 1/2 in a constant magnetic field in the $\mathbf{x}$ direction and with environmental fluctuating fields in the $\mathbf{z}$ direction, *cf. e.g.* [14]. The analogy holds also for an asymmetric double well potential under appropriate rotation of the "spin" axes.

In the spin-boson literature, the particle is usually prepared in a localized diagonal state of the reduced density matrix (RDM). In fact, if this initial condition is chosen, the dissipative TSS is a paradigm to investigate the interplay between quantum coherence phenomena and environmental influences. The coupling to the environment results in a reduction of the coherent tunneling motion by incoherent processes [14–16], and may lead to a transition to localization at zero temperature [17]. Finally, when the system is additionally subject to time-dependent external forces, diverse remarkable effects occur [18]. In the absence of coupling to a heat bath, complete destruction of tunneling can be induced by a coherent driving field with appropriately chosen frequency and amplitude [19,20]. In the presence of dissipation, this effect can still persist for many tunneling periods [21–24]. The transition temperature above which quantum coherence is destroyed by bath fluctuations is modified by a driving field [22–24]. Other aspects concern, *e.g.*, the possibility to control *a priori* the proton or electron transfer by an electric field [25,26], or the phenomenon of driving-induced large-amplitude coherent oscillations [27–31].

Here we study the reduced dynamics for *arbitrary* diagonal/off-diagonal initial preparation. Within the path-integral method, a set of exact equations for the elements of the RDM are obtained. In particular, the diagonal

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elements of the RDM satisfy a closed generalized (non-Markovian) master equation (GME) in which the particular initial preparation is in the inhomogeneity. Different approximations covering the interesting parameter regimes of temperature and bath coupling are discussed. We show that the asymptotic dynamics is independent of the particular initial state. However, preparation effects related to nonzero off-diagonal elements of the initial reduced density matrix are crucial at short times whenever the dynamics exhibits underdamped coherent oscillations. We address several features of the dissipative dynamics which are sensitive to this initial state. In particular, we show that a suitably tuned ac-field can slow down the decoherence induced by the environment, so that preparation effects can survive on a longer time-scale than in the absence of driving.

The "initial density matrix problem" is well known in the context of spin magnetic resonance and relaxation. Among the first microscopic theories are the pioneering works by Bloch [32], Redfield [33] and Fano [34] on the relaxation of dissipative spin systems subject to weak external magnetic fields. Common to these early works are the simplifying assumptions of a Markovian bath and of a weak interaction between the system and the bath. These assumptions lead to Markovian equations of motion for the RDM which can be solved in lowest order Born approximation. However, the Markov approximation for the case of open quantum systems weakly coupled to their environments may break the positivity of the reduced dynamics. It has been shown [35] that the weak-coupling Markovian equations of motion (Redfield equations) are a consistent approximation to the reduced dynamics only if supplemented by a slippage in the initial conditions. As already shown by Argyres and Kelley in 1964 [36], who started from the Liouville equation of motion for the global system, the description of the full time evolution of the reduced system generally involves the solution of non-Markovian equations of motion.

The problem of environment-induced decoherence has found attention in different contexts [16]. A theme of topical interest is the suppression of quantum coherence in magnetic grains or of a giant spin coupled to a phonon or spin bath environment [37]. Here, the case in which decoherence originates from the coupling to a harmonic thermal bath is considered.

In Section 2 we introduce the driven spin-boson model and the relevant dynamical quantities. In Section 3.1 we derive the exact formal solution for the RDM, and in Section 3.2 we obtain a set of exact integro-differential relations among the elements of the RDM. General features of the reduced dynamics, as well as of nonequilibrium correlation functions, are outlined in Section 3.3. In Section 4 we discuss several useful analytical approximations for the reduced dynamics, and in Section 5 we draw our conclusions.

2 The driven spin-boson system

To start with, we describe the externally driven spin-boson Hamiltonian $H(t) = H_{\rm TSS} + H_{\rm ext}(t) + H_{\rm B} + H_{\rm SB}$. The first term characterizes the isolated two-state system. It is conveniently written in the pseudospin form

$$H_{\rm TSS} = -\tfrac{1}{2}\hbar(\Delta_0\sigma_x + \epsilon_0\sigma_z). \qquad (2.1)$$

We choose basis states $|R\rangle$ (right) and $|L\rangle$ (left) as eigenstates of σ_z with eigenvalues $+1$ and -1, respectively. In the discrete representation, the position operator q is related to σ_z by $q = \frac{1}{2}\sigma_z d$, with d being the spatial distance between the localized states. The interaction energy $\hbar\Delta_0$ is the energy splitting of a symmetric ($\epsilon_0 = 0$) TSS due to quantum tunneling. The term $H_{\rm ext}(t)$ describes the interaction with external time-dependent fields. To be general, we introduce couplings to external fields which modulate the asymmetry energy between the two wells and the coupling energy between the localized states [18].

The general Hamiltonian for the driven TSS reads

$$H_{\rm TSS} + H_{\rm ext}(t) = -\tfrac{1}{2}\hbar[\Delta(t)\sigma_x + \varepsilon(t)\sigma_z]. \qquad (2.2)$$

Here, $\Delta(t)$ and $\varepsilon(t)$ may or may not be periodic, depending on the characteristics of the driving fields. Finally, the thermal bath is an ensemble of harmonic oscillators, $H_{\rm B} = \sum_i [p_i^2/2m_i + m_i\omega_i^2 x_i^2/2]$, and we consider bilinear couplings that are sensitive to the TSS position and to a collective bath coordinate X describing the bath polarization energy, $H_{\rm SB} := -\sigma_z X/2$, where $X = d\sum_i c_i x_i$. The time-dependent spin-boson Hamiltonian reads

$$H(t) = -\tfrac{1}{2}\hbar[\Delta(t)\sigma_x + \varepsilon(t)\sigma_z] - \tfrac{1}{2}\sigma_z X + H_{\rm B}. \qquad (2.3)$$

In this model all the bath influence is captured by the spectral density $J(\omega) = (\pi/2)\sum_i (c_i^2/m_i\omega_i)\delta(\omega - \omega_i)$. We assume a power-law form with an exponential cutoff,

$$J(\omega) = (2\pi\hbar/d^2)\alpha_s\tilde{\omega}^{1-s}\omega^s \exp(-\omega/\omega_c). \qquad (2.4)$$

Here, α_s is a dimensionless coupling constant, and $\tilde{\omega}$ a reference frequency. The case $s = 1$ describes an Ohmic bath coupling. We now study the RDM of the model (2.3).

2.1 The reduced density matrix of the driven spin-boson system

We wish to compute the RDM $\rho(t) = {\rm Tr_B}\{W(t)\}$ of the driven damped TSS, where $W(t)$ is the density matrix of the global system. We assume for the global system at time $t_0 = 0$ the product initial state $W(0) = \rho_0 W_{\rm B}$. Here $W_{\rm B}$ is the canonical density matrix of the bath, $W_{\rm B} = {\rm e}^{-\beta H_{\rm B}}/\,{\rm Tr}\,\{{\rm e}^{-\beta H_{\rm B}}\}$ ($\beta = 1/k_{\rm B}T$), while the TSS has been suddenly prepared in the general RDM state

$$\begin{aligned}\rho_0 &= \tfrac{1}{2}I + \tfrac{1}{2}P_0\sigma_z + a\sigma_x + b\sigma_y \\ &= \begin{pmatrix} p_{\rm R} & a - {\rm i}b \\ a + {\rm i}b & p_{\rm L}\end{pmatrix}.\end{aligned} \qquad (2.5)$$

Here, $P_0 := p_{\rm R} - p_{\rm L}$, where $p_{\rm R}$ and $p_{\rm L} = 1 - p_{\rm R}$ are the probabilities to find the particle in the right and left well, and a, b are the coherences. In the second form, the RDM $\rho_{\sigma,\sigma'}(t = 0)$ is written in the eigenstate basis of σ_z. With a, b real, the condition $\mathrm{Tr}\,\rho^2 \le 1$ leads to the constraint $a^2 + b^2 \le p_{\rm R} p_{\rm L}$. Knowledge of $\rho(t)$ at times $t > 0$ enables us to calculate the expectation value of *every* observable relevant for the TSS. The pseudospin form with matrices σ_j and I reads $\rho(t) = I/2 + \sum_{i=x,y,z} \langle\sigma_i\rangle_t \sigma_i/2$, where $\langle\sigma_i\rangle_t := \mathrm{Tr}\,\{\rho(t)\sigma_i\}$, yielding

$$\begin{aligned}\langle\sigma_z\rangle_t &= \rho_{1,1}(t) - \rho_{-1,-1}(t),\\ \langle\sigma_x\rangle_t &= \rho_{1,-1}(t) + \rho_{-1,1}(t),\\ \langle\sigma_y\rangle_t &= \mathrm{i}[\rho_{1,-1}(t) - \rho_{-1,1}(t)].\end{aligned} \tag{2.6}$$

The diagonal elements $\rho_{-1,-1}$ and $\rho_{1,1}$ are the populations, and the off-diagonal elements $\rho_{-1,1},\ \rho_{1,-1}$ are the coherences. The position expectation value

$$\langle\sigma_z\rangle_t := P(t) = \mathrm{Tr}\{\rho(t)\sigma_z\} = \mathrm{Tr}\{\sigma_z(t)\rho_0\} \tag{2.7}$$

provides central information on the dynamics of the TSS. For adiabatically varying driving fields, the difference $N(t)$ in the population of the energy levels is

$$N(t) := \cos[\theta(t)]\,\langle\sigma_z\rangle_t - \sin[\theta(t)]\,\langle\sigma_x\rangle_t, \tag{2.8}$$

where $\tan\theta = -\Delta/\varepsilon$. In particular, $N(t)$ coincides with $\langle\sigma_z\rangle_t$ for the case of a symmetric, undriven TSS.

We label the off-diagonal and diagonal states of the RDM by $\xi = \pm 1$ and $\eta = \pm 1$, respectively, and introduce the conditional RDM propagators $P(\mu, t; \mu_0, t_0)$ from the initial state μ_0 at time t_0 to the final state μ at time t, where $\mu, \mu_0 = \eta$ or ξ. Then, the expectation value of the position is expressed as

$$\begin{aligned}\langle\sigma_z\rangle_t = \sum_{\eta=\pm 1} \eta\, [&p_{\rm R} P(\eta, t; \eta_0 = 1, 0)\\ &+ p_{\rm L}\, P(\eta, t; \eta_0 = -1, 0)\\ &+ (a - \mathrm{i}b) P(\eta, t; \xi_0 = 1, 0)\\ &+ (a + \mathrm{i}b) P(\eta, t; \xi_0 = -1, 0)].\end{aligned} \tag{2.9}$$

Similarly, the coherence expectation value reads

$$\begin{aligned}\langle\sigma_x\rangle_t = \sum_{\xi=\pm 1} [&p_{\rm R} P(\xi, t; \eta_0 = 1, 0)\\ &+ p_{\rm L}\, P(\xi, t; \eta_0 = -1, 0)\\ &+ (a - \mathrm{i}b) P(\xi, t; \xi_0 = 1, 0)\\ &+ (a + \mathrm{i}b) P(\xi, t; \xi_0 = -1, 0)].\end{aligned} \tag{2.10}$$

In the absence of driving and dissipation, the RDM can easily be evaluated. One finds

$$\begin{aligned}\langle\sigma_z\rangle_t = (p_{\rm R} - p_{\rm L}) &\Big[\frac{\epsilon_0^2}{\nu_0^2} + \frac{\Delta_0^2}{\nu_0^2}\cos(\nu_0 t)\Big]\\ &+ 2a\frac{\Delta_0\epsilon_0}{\nu_0^2}[1 - \cos(\nu_0 t)] - 2b\frac{\Delta_0}{\nu_0}\sin(\nu_0 t),\end{aligned} \tag{2.11}$$

$$\begin{aligned}\langle\sigma_x\rangle_t = (p_{\rm R} - p_{\rm L}) &\frac{\epsilon_0\Delta_0}{\nu_0^2}\big[1 - \cos(\nu_0 t)\big]\\ &+ 2a\Big[\frac{\Delta_0^2}{\nu_0^2} + \frac{\epsilon_0^2}{\nu_0^2}\cos(\nu_0 t)\Big] + 2b\frac{\epsilon_0}{\nu_0}\sin(\nu_0 t),\end{aligned} \tag{2.12}$$

$$\langle\sigma_y\rangle_t = -\frac{1}{\Delta_0}\frac{\mathrm{d}}{\mathrm{d}t}\langle\sigma_z\rangle_t, \tag{2.13}$$

where $E_0 = \hbar\nu_0$ with $\nu_0 = (\Delta_0^2 + \epsilon_0^2)^{1/2}$ is the energy splitting. As expected, N is independent of t for any initial state, $N = (p_{\rm R} - p_{\rm L})\epsilon_0/\nu_0 + 2a\Delta_0/\nu_0$. By virtue of the expressions (2.11-2.13), the evolution for different initial preparations can be discussed. In the *standard* preparation, the TSS is set up at time $t_0 = 0$ in a (localized) eigenstate of σ_z, in particular when the tunneling dynamics is investigated. We then have $p_{\rm R} = 1$ (or $p_{\rm L} = 1$) and $a = b = 0$. After the system is released, $\langle\sigma_z\rangle_t$ and $\langle\sigma_x\rangle_t$ will undergo quantum coherent oscillations with frequency ν_0. In contrast, when the system is prepared in the ground state, we have $p_{\rm R} - p_{\rm L} = \epsilon_0/\nu_0$, $2a = \Delta_0/\nu_0$, and $b = 0$. In this case is $N(t) = 1$, $\langle\sigma_z\rangle_t = \epsilon_0/\nu_0$, and $\langle\sigma_x\rangle_t = \Delta_0/\nu_0$ for all t, and hence there is no dynamics. Beyond these limiting cases, one can think of other preparations. The maximum amplitude of the oscillations is reached when the system is prepared in an eigenstate of σ_z.

In the presence of driving and dissipation, the quantum coherent motion described by equations (2.11-2.13) is modified. Let us now investigate these modifications.

3 Path-integral solution for the RDM

3.1 Exact formal solution

For the model (2.3), the trace over the bath degrees of freedom can be performed exactly. The RDM is then written in terms of the spin path $\sigma(t) = 2q(t)/d$ as

$$\rho_{\sigma,\sigma'}(t) = \sum_{\sigma_0,\sigma_0'} \int \mathcal{D}\sigma \int \mathcal{D}\sigma'\, \mathcal{A}[\sigma]\mathcal{A}^*[\sigma']\mathcal{F}[\sigma,\sigma']\,\rho_{\sigma_0,\sigma_0'}(t_0).$$

The double spin path sum runs over all possible intermediate spin states ± 1, and the outer states are as indicated. The quantity $\mathcal{A}[\sigma]$ is the probability amplitude of the TSS to follow the path $\sigma(t')$ in the absence of fluctuating forces, and $\mathcal{F}[\sigma,\sigma']$ is the Feynman-Vernon influence functional describing the environmental influences [38]. It is convenient to introduce the linear combinations

$$\xi(t') = \tfrac{1}{2}[\sigma(t') - \sigma'(t')]\ , \qquad \eta(t') = \tfrac{1}{2}[\sigma(t') + \sigma'(t')].$$

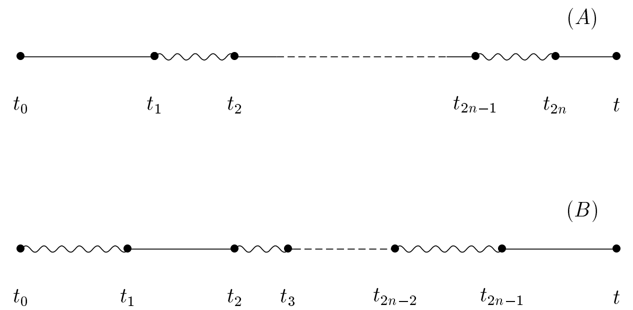

Fig. 1. Generic paths of class A and B contributing to the expectation value $\langle\sigma_z\rangle_t$. Paths of class A start and end in a sojourn state (straight line). Paths belonging to class B start in a blip state (curly line) and end in a sojourn state.

We then obtain $\mathcal{F}[\sigma,\sigma'] \to \mathcal{F}[\eta,\xi] = \exp \Phi[\eta,\xi]$ with

$$\begin{aligned}\Phi[\eta,\xi] = &\int_{t_0}^{t}\mathrm{d}t' \int_{t_0}^{t'}\mathrm{d}t'' \Big[\dot\xi(t')Q'(t'-t'')\dot\xi(t'') \\ &\qquad\qquad + \mathrm{i}\dot\xi(t')Q''(t'-t'')\dot\eta(t'')\Big] \\ &- \xi(t)\int_{t_0}^{t}\mathrm{d}t''\Big[Q'(t-t'')\dot\xi(t'') + \mathrm{i}Q''(t-t'')\dot\eta(t'')\Big] \\ &- \xi(t_0)\Big[\xi(t)Q'(t-t_0) - \int_{t_0}^{t}\mathrm{d}t'\dot\xi(t')Q'(t'-t_0)\Big] \\ &- \mathrm{i}\eta(t_0)\Big[\xi(t)Q''(t) - \int_{t_0}^{t}\mathrm{d}t'\dot\xi(t')Q''(t')\Big],\end{aligned}$$

where $Q(t) = Q^{'}(t) + \mathrm{i}Q^{''}(t)$ is given by

$$\begin{aligned}Q(t) = &\frac{d^2}{\hbar\pi}\int_0^\infty \mathrm{d}\omega \frac{J(\omega)}{\omega^2} \\ &\times\Big\{\coth\Big(\frac{\hbar\omega\beta}{2}\Big)\Big(1-\cos(\omega t)\Big) + \mathrm{i}\sin(\omega t)\Big\}.\end{aligned} \tag{3.1}$$

We have $\eta(t')\xi(t') = 0$. The periods the system spends in a diagonal state, $\eta(t') = \pm 1$, are referred to as *sojourns*. We have $\eta = +1$ for the state $\rho_{1,1}$, and $\eta = -1$ for $\rho_{-1,-1}$. The periods in which the system is off-diagonal , $\xi(t') = \pm 1$, have been dubbed *blips* [14,16]. We put $\xi = 1$ for the state $\rho_{1,-1}$, and $\xi = -1$ for the state $\rho_{-1,1}$.

We begin with studying $\langle\sigma_z\rangle_t$. Consider first the case where the system is initially and finally in a sojourn state (Fig. 1, top). We refer to the corresponding paths as class A. A general double path of class A with $2n$ transitions at flip times t_j, $j = 1, 2, .., 2n$ has $n+1$ sojourns and n blips. The influence function for this path reads

$$\begin{aligned}\Phi^{(2n)} &= H_n^A + \mathrm{i}\sum_{j=1}^{n}\sum_{k=0}^{j-1}\xi_j\eta_k\, X_{j,k}^A, \\ H_n^A &= -\sum_{j=1}^{n} Q'_{2j,2j-1} - \sum_{j=2}^{n}\sum_{k=1}^{j-1}\xi_j\xi_k\,\Lambda_{j,k}^A,\end{aligned} \tag{3.2}$$

where $Q_{j,k} := Q(t_j - t_k)$, $t_{2n+1} \equiv t$. The term H_n^A contains the intrablip and interblip correlations, and $\mathrm{Im}\,\Phi^{(2n)}$ all blip-sojourn correlations. We have for $k > 0$

$$\begin{aligned}\Lambda_{j,k}^A &= Q^{'}_{2j,2k-1} + Q^{'}_{2j-1,2k} - Q^{'}_{2j,2k} - Q^{'}_{2j-1,2k-1}, \\ X_{j,k}^A &= Q^{''}_{2j,2k+1} + Q^{''}_{2j-1,2k} - Q^{''}_{2j,2k} - Q^{''}_{2j-1,2k+1}.\end{aligned}$$

The correlation term involving the initial sojourn, $X_{j,0}^A$, keeps track of the particular initial preparation [16]. When the TSS is suddenly prepared in the state ρ_0, the bath is in the canonical state W_B, resulting in the correlations of the initial sojourn with the blip j

$$X_{j,0}^A = Q^{''}_{2j,1} - Q''_{2j-1,1} + Q^{''}_{2j-1,0} - Q^{''}_{2j,0}.$$

In contrast, when the TSS has been constrained in the state η_0 for a long period before it is released, the bath is in the shifted canonical state $\widetilde{W}_\mathrm{B} = W_\mathrm{B}\exp(\eta_0\beta X/2)$ at $t = 0$. Then we have $X_{j,0}^A \to \widetilde{X}_{j,0}^A = Q^{''}_{2j,1} - Q''_{2j-1,1}$.

All paths of class B start in a blip and end in a sojourn state after an odd number of steps (*cf.* Fig. 1, bottom). The corresponding influence function with n blips reads

$$\begin{aligned}\Phi^{(2n-1)} &= H_n^B + \mathrm{i}\sum_{j=2}^{n}\sum_{k=1}^{j-1}\xi_j\eta_k\, X_{jk}^B, \\ H_n^B &= -\sum_{j=0}^{n-1} Q'_{2j+1,2j} - \sum_{j=2}^{n}\sum_{k=1}^{j-1}\xi_j\xi_k\,\Lambda_{jk}^B.\end{aligned} \tag{3.3}$$

The division of the correlations is as for class A. We have

$$\begin{aligned}\Lambda_{j,k}^B &= Q^{'}_{2j-1,2k-2} + Q^{'}_{2j-2,2k-1} \\ &\quad - Q^{'}_{2j-1,2k-1} - Q^{'}_{2j-2,2k-2}, \\ X_{j,k}^B &= Q^{''}_{2j-1,2k} + Q^{''}_{2j-2,2k-1} - Q^{''}_{2j-1,2k-1} - Q^{''}_{2j-2,2k}.\end{aligned}$$

Finally, the sum over histories of paths is represented (i) by the sum over all possible numbers of steps, (ii) by the integrations over the corresponding flip times $\{t_j\}$, and (iii) by the sum over all possible arrangements of the blips $\{\xi_j = \pm 1\}$ and sojourns $\{\eta_j \pm 1\}$. For the time integrals and the associated transition amplitudes, we introduce the compact notation

$$\begin{aligned}\int_{t_0}^{t}\mathcal{D}_n\{t_j\}\cdots &= \int_{t_0}^{t}\mathrm{d}t_n\int_{t_0}^{t_n}\mathrm{d}t_{n-1}\ldots\int_{t_0}^{t_2}\mathrm{d}t_1\delta_n\{t_j\}\cdots, \\ \delta_n\{t_j\} &= \prod_{j=1}^{n}\Delta(t_j).\end{aligned}$$

The sum over the intermediate diagonal states can easily be performed. This leads to modified bias factors and influence functions. The influence of the time-dependent biasing forces is represented by the factors ($i = A, B$)

$$C_{n,i}^{(\mathrm{s})} = \cos\phi_n^i\,, \qquad C_{n,i}^{(\mathrm{a})} = \sin\phi_n^i, \tag{3.4}$$

$$\phi_n^A = \sum_{j=1}^{n}\xi_j\zeta(t_{2j},t_{2j-1}), \quad \phi_n^B = \sum_{j=1}^{n}\xi_j\zeta(t_{2j-1},t_{2j-2}),$$

where $\zeta(t,t') = \int_{t'}^{t} \mathrm{d}t''\varepsilon(t'')$. The superscript (s/a) labels terms symmetric/antisymmetric in the bias. The dissipative influences are described by the influence functions

$$F_{n,A}^{(+)} = \exp(H_n^A)\prod_{k=0}^{n-1}\cos\chi_{n,k}^A, \quad F_{n,A}^{(-)} = F_{n,A}^{(+)}\tan\chi_{n,0}^A,$$
$$F_{n,B}^{(+)} = \exp(H_n^B)\prod_{k=1}^{n-1}\cos\chi_{n,k}^B, \qquad (3.5)$$

in which the phases $\chi_{n,k}^i = \sum_{j=k+1}^{n}\xi_j X_{j,k}^i$ describe the sojourn-blip correlations.

Next, we introduce the partial expectation values

$$P_1^{(\mathrm{s})}(t) := \tfrac{1}{2}\mathrm{Tr}\,\{\sigma_z(t)\sigma_z\}, \quad P_1^{(\mathrm{a})}(t) := \tfrac{1}{2}\mathrm{Tr}\,\{\sigma_z(t)\},$$
$$P_2^{(\mathrm{s})}(t) := \tfrac{1}{2}\mathrm{Tr}\,\{\sigma_z(t)\sigma_y\}, \quad P_2^{(\mathrm{a})}(t) := \tfrac{1}{2}\mathrm{Tr}\,\{\sigma_z(t)\sigma_x\},$$

given in terms of the conditional propagators as

$$\sum_{\eta=\pm1}\eta\,P(\eta,t;\eta_0,0) = \eta_0\,P_1^{(\mathrm{s})}(t) + P_1^{(\mathrm{a})}(t),$$
$$\sum_{\eta=\pm1}\eta\,P(\eta,t;\xi_0,0) = +\mathrm{i}\xi_0 P_2^{(\mathrm{s})}(t) + P_2^{(\mathrm{a})}(t). \qquad (3.6)$$

Substituting the forms (3.6) into equation (2.9), we obtain for the general initial state (2.5) the position expectation as

$$\langle\sigma_z\rangle_t = (p_\mathrm{R} - p_\mathrm{L})\,P_1^{(\mathrm{s})}(t) \;+\; P_1^{(\mathrm{a})}(t)$$
$$+2aP_2^{(\mathrm{a})}(t) \;+\; 2bP_2^{(\mathrm{s})}(t). \qquad (3.7)$$

Collecting the various weight factors for the double spin path, we find the exact formal series

$$P_1^{(\mathrm{s})}(t) = 1 + \sum_{n=1}^{\infty}\Big(-\frac{1}{2}\Big)^n\int_0^t\mathcal{D}_{2n}\{t_j\}\sum_{\{\xi_j\}}F_{n,A}^{(+)}C_{n,A}^{(\mathrm{s})},$$
$$P_1^{(\mathrm{a})}(t) = -\sum_{n=1}^{\infty}\Big(-\frac{1}{2}\Big)^n\int_0^t\mathcal{D}_{2n}\{t_j\}\sum_{\{\xi_j\}}F_{n,A}^{(-)}C_{n,A}^{(\mathrm{a})},$$
$$P_2^{(\mathrm{a})}(t) = -\sum_{n=1}^{\infty}\Big(-\frac{1}{2}\Big)^n\int_0^t\mathcal{D}_{2n-1}\{t_j\}\sum_{\{\xi_j\}}\xi_1F_{n,B}^{(+)}C_{n,B}^{(\mathrm{a})},$$
$$P_2^{(\mathrm{s})}(t) = \sum_{n=1}^{\infty}\Big(-\frac{1}{2}\Big)^n\int_0^t\mathcal{D}_{2n-1}\{t_j\}\sum_{\{\xi_j\}}F_{n,B}^{(+)}C_{n,B}^{(\mathrm{s})}.$$

The sum $\sum_{\{\xi_j\}}$ is over the 2^n possible states for the n blips included in $F_{n,A}^{(\pm)}$ and $F_{n,B}^{(\pm)}$.

The initial condition $\langle\sigma_z\rangle_{t=0} = p_\mathrm{R} - p_\mathrm{L}$ is provided by the first term in equation (3.7). The residual terms are contributing for times $t > 0$ only. In the special case $a = b = P_\mathrm{L} = 0$, equation (3.7) reduces to the form $P(t) = P_1^{(\mathrm{s})}(t) + P_1^{(\mathrm{a})}(t)$, discussed in reference [14].

We refrain from writing down the corresponding series expressions for the coherences. We see from equation (2.10) that they involve the influence functional of those paths which start from a diagonal or off-diagonal state of the RDM and all end in an off-diagonal state. The explicit derivation for the standard initial preparation $p_\mathrm{L} = a = b = 0$ is given in reference [39].

Using $[\,H,\sigma_z]_- = \mathrm{i}\Delta\sigma_y$, we find for any initial state

$$\langle\sigma_y\rangle_t = -\frac{1}{\Delta(t)}\frac{\mathrm{d}}{\mathrm{d}t}\langle\sigma_z\rangle_t. \qquad (3.8)$$

Thus, $\langle\sigma_y\rangle_t$ can be interpreted as an average tunneling current. The exact formal solution for $\langle\sigma_x\rangle_t$ is more complicated and is discussed below.

The above exact expression for $\langle\sigma_z\rangle_t$, and the corresponding ones for $\langle\sigma_x\rangle_t$ and $\langle\sigma_y\rangle_t$ have a very complex form and cannot be evaluated analytically. Therefore one has to resort to suitable approximations and corresponding numerical computations. Before turning to approximations, we now show that the dynamics of $\langle\sigma_z\rangle_t$ can be expressed in terms of an exact master equation, and $\langle\sigma_x\rangle_t$ can be determined from an integral expression.

3.2 Exact master equations and integral expressions for the RDM

The kernels of master equations are the irreducible components. Irreducibility means that a kernel cannot be cut into two uncorrelated pieces at an intermediate sojourn without removing correlations across this sojourn. Following references [29,39], we define irreducible influence functions by subtracting all reducible components. We find for paths of type $i = A, B$ with n blips and time growing from right to left

$$\widetilde{F}_{n,i}^{(\pm)} = F_{n,i}^{(\pm)} - \sum_{j=2}^{n}(-1)^j$$
$$\times\sum_{m_1,\cdots,m_j}F_{m_1,i}^{(+)}F_{m_2,i}^{(+)}\cdots F_{m_j,i}^{(\pm)}\delta_{m_1+\cdots+m_j,n}.$$

The inner sum is over all positive integers $m_1,..,m_j$. By definition, each subtraction involves again time-ordered flips. In the subtracted terms, the bath correlations are only inside of the factors $F_{m_j,i}^{(\pm)}$, and there are no correlations between these factors. The $n = 2$ terms read

$$\tilde{F}_{2,A}^{(\pm)}(t_4,t_3,t_2,t_1) = F_{2,A}^{(\pm)}(t_4,t_3,t_2,t_1) \qquad (3.9)$$
$$-F_{1,A}^{(+)}(t_4,t_3)F_{1,A}^{(\pm)}(t_2,t_1)$$
$$\tilde{F}_{2,B}^{(\pm)}(t_3,t_2,t_1,t_0) = F_{2,B}^{(\pm)}(t_3,t_2,t_1,t_0) \qquad (3.10)$$
$$-F_{1,B}^{(+)}(t_3,t_2)F_{1,B}^{(\pm)}(t_1,t_0).$$

The flip times for the $n > 2$ terms can be taken from Figure 1. Removing from class A the outer sojourns, we find for the symmetric (s) and antisymmetric (a) kernels

$$K_A^{(\mathrm{s/a})}(t,t') = \tilde{K}_A^{(\mathrm{s/a})}(t,t') + \sum_{n=2}^{\infty}(-1)^{n-1}\int_{t'}^{t}\mathrm{d}t_{2n-1}\cdots\int_{t'}^{t_3}\mathrm{d}t_2 \times\delta_{2n}\{t_j\}\frac{1}{2^n}\sum_{\{\xi_j=\pm 1\}}\widetilde{F}_{n,A}^{(+/-)}C_{n,A}^{(\mathrm{s/a})}. \tag{3.11}$$

Here we have the identifications $t_1 = t'$ and $t_{2n} = t$. In lowest order in Δ, we obtain

$$\begin{aligned}\tilde{K}_A^{(\mathrm{s})}(t,t') &= \Delta(t)\Delta(t')h^{(+)}(t-t')\cos[\zeta(t,t')],\\ \tilde{K}_A^{(\mathrm{a})}(t,t') &= \Delta(t)\Delta(t')h^{(-)}(t-t')\sin[\zeta(t,t')],\end{aligned} \tag{3.12}$$

where

$$\begin{aligned}h^{(+)}(t) &= \mathrm{e}^{-Q'(t)}\cos\left[Q''(t)\right],\\ h^{(-)}(t) &= \mathrm{e}^{-Q'(t)}\sin\left[Q''(t)\right].\end{aligned} \tag{3.13}$$

The kernels connected with paths of class B are found by removing from these paths the final sojourn. The antisymmetric kernel is given by the series (we put $t_{2n-1} = t$)

$$K_B^{(\mathrm{a})}(t,t') = \tilde{K}_B^{(\mathrm{a})}(t,t') + \sum_{n=2}^{\infty}(-1)^{n-1}\int_{t'}^{t}\mathrm{d}t_{2n-2}\cdots\int_{t'}^{t_2}\mathrm{d}t_1 \times\delta_{2n-1}\{t_j\}\frac{1}{2^n}\sum_{\{\xi_j=\pm 1\}}\xi_1\widetilde{F}_{n,B}^{(+)}C_{n,B}^{(\mathrm{a})}. \tag{3.14}$$

The symmetric kernel $K_B^{(\mathrm{s})}(t,t')$ is found from equation (3.14) upon substituting

$$\xi_1\widetilde{F}_{n,B}^{(+)}C_{n,B}^{(\mathrm{a})} \to \widetilde{F}_{n,B}^{(+)}C_{n,B}^{(\mathrm{s})}. \tag{3.15}$$

The lowest order is again without internal steps,

$$\begin{aligned}\tilde{K}_B^{(\mathrm{s})}(t,t') &= \Delta(t)\mathrm{e}^{-Q'(t-t')}\cos[\zeta(t,t')],\\ \tilde{K}_B^{(\mathrm{a})}(t,t') &= \Delta(t)\mathrm{e}^{-Q'(t-t')}\sin[\zeta(t,t')].\end{aligned} \tag{3.16}$$

The full dynamics of $\langle\sigma_z\rangle_t$ is established by iterative succession of these kernels and an additional time integration. Next, we take the time derivative of equation (3.7) and rearrange the resulting series into sequences of irreducible clusters. The resulting series can be summed to the exact generalized master equation (GME),

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle\sigma_z\rangle_t = \int_0^t \mathrm{d}t'[K_A^{(\mathrm{a})}(t,t') - K_A^{(\mathrm{s})}(t,t')\langle\sigma_z\rangle_{t'}] + 2aK_B^{(\mathrm{a})}(t,0) - 2bK_B^{(\mathrm{s})}(t,0). \tag{3.17}$$

To calculate the dynamics of $\langle\sigma_x\rangle_t$, we introduce kernels $Y_{A/B}^{(\mathrm{s/a})}(t,t')$ which differ from $K_{A/B}^{(\mathrm{s/a})}(t,t')$ in the influence weights. For type A, we find (we put $t_1 = t'$, $t_{2n} = t$)

$$Y_A^{(\mathrm{s/a})}(t,t') = \widetilde{Y}_A^{(\mathrm{s/a})}(t,t') + \sum_{n=2}^{\infty}(-1)^{n-1}\int_{t'}^{t}\mathrm{d}t_{2n-1}\cdots\int_{t'}^{t_3}\mathrm{d}t_2 \times\delta_{2n-1}\{t_j\}\frac{1}{2^n}\sum_{\{\xi_j=\pm 1\}}\xi_n\widetilde{F}_{n,A}^{(-/+)}C_{n,A}^{(\mathrm{s/a})},$$

and the terms without internal steps read

$$\begin{aligned}\widetilde{Y}_A^{(\mathrm{s})}(t,t') &= \Delta(t')h^{(-)}(t-t')\cos[\zeta(t,t')],\\ \widetilde{Y}_A^{(\mathrm{a})}(t,t') &= \Delta(t')h^{(+)}(t-t')\sin[\zeta(t,t')].\end{aligned} \tag{3.18}$$

The symmetric kernel of type B is given by ($t_{2n-1} = t$)

$$Y_B^{(\mathrm{s})}(t,t') = \widetilde{Y}_B^{(\mathrm{s})}(t,t') + \sum_{n=2}^{\infty}(-1)^{n-1}\int_0^t\mathrm{d}t_{2n-2}\cdots\int_0^{t_2}\mathrm{d}t_1 \times\delta_{2n-2}\{t_j\}\frac{1}{2^n}\sum_{\{\xi_j=\pm 1\}}\xi_1\xi_n\widetilde{F}_{n,B}^{(+)}C_{n,B}^{(\mathrm{s})}. \tag{3.19}$$

The antisymmetric kernel of type B, $Y_B^{(\mathrm{a})}(t,t_0)$, is obtained from equation (3.19) upon substituting

$$\xi_1\xi_n\widetilde{F}_{n,B}^{(+)}C_{n,B}^{(\mathrm{s})} \to \xi_n\widetilde{F}_{n,B}^{(+)}C_{n,B}^{(\mathrm{a})}. \tag{3.20}$$

In lowest order, the system stays in the same blip state,

$$\begin{aligned}\widetilde{Y}_B^{(\mathrm{s})}(t,t') &= \mathrm{e}^{-Q'(t-t')}\cos[\zeta(t,t')],\\ \widetilde{Y}_B^{(\mathrm{a})}(t,t') &= \mathrm{e}^{-Q'(t-t')}\sin[\zeta(t,t')].\end{aligned} \tag{3.21}$$

The exact formal expression for $\langle\sigma_x\rangle_t$ is readily found as

$$\langle\sigma_x\rangle_t = \int_0^t \mathrm{d}t'[Y_A^{(\mathrm{s})}(t,t') + Y_A^{(\mathrm{a})}(t,t')\langle\sigma_z\rangle_{t'}] + 2aY_B^{(\mathrm{s})}(t,0) + 2bY_B^{(\mathrm{a})}(t,0). \tag{3.22}$$

Here, $\langle\sigma_z\rangle_{t'}$ may be calculated from equation (3.7) or equation (3.17). Thus, once we have calculated both $\langle\sigma_z\rangle_t$ and the kernels $Y_i^{(\mathrm{s/a})}(t,t')$, we obtain $\langle\sigma_x\rangle_t$ by quadrature.

3.3 General features of the reduced dynamics

In the absence of time-dependent driving, the expressions (3.17) and (3.22) are in convolutive form and can be studied by Laplace transformation. We now summarize several general features:

(i) The equilibrium state reached asymptotically is obtained from equations (3.17, 3.22) as

$$\begin{aligned}\langle\sigma_z\rangle_\infty &:= P_\infty = \frac{\Sigma^{(\mathrm{a})}(0)}{\Sigma^{(\mathrm{s})}(0)},\\ \langle\sigma_x\rangle_\infty &= \Sigma_x^{(\mathrm{s})}(0) + \Sigma_x^{(\mathrm{a})}(0)\,\langle\sigma_z\rangle_\infty,\end{aligned} \tag{3.23}$$

with the 'self-energies'

$$\Sigma^{(\mathrm{s/a})}(\lambda) = \int_0^\infty \mathrm{d}\tau \mathrm{e}^{-\lambda\tau} K_A^{(\mathrm{s/a})}(\tau),$$
$$\Sigma_x^{(\mathrm{s/a})}(\lambda) = \int_0^\infty \mathrm{d}\tau \mathrm{e}^{-\lambda\tau} Y_A^{(\mathrm{s/a})}(\tau). \qquad (3.24)$$

The states reached asymptotically are *independent* of the initial state, as expected. In the transient regime, the relevant dynamical quantities are the frequency and damping rate of the coherent oscillation, the coherent-incoherent transition temperature T^*, and the relaxation rate towards equilibrium. All these quantities are determined by the zeros of the equation

$$\lambda + \Sigma^{(\mathrm{s})}(\lambda) = 0. \qquad (3.25)$$

Since equation (3.25) is independent of the particular initial state, these quantities are *universal*.

(ii) Effects of the initial preparation on the dynamics drastically depend on the particular initial state chosen. We see from equations (3.17, 3.22) that off-diagonal preparation effects (a, $b \neq 0$) die out on the time scale τ_{K} on which the bath correlation function $Q'(t)$ decays. For strong enough damping, and/or high enough temperature, τ_{K} is very small compared to the time scale $1/\Sigma^{(\mathrm{s})}(0)$ for incoherent exponential relaxation into the equilibrium state. Thus, there is a huge time domain in which effects of the initial coherences have already died out, and the system relaxes with the rate $\Gamma_r := \Sigma^{(\mathrm{s})}(0)$ to the equilibrium state as

$$P(t) := \langle \sigma_z \rangle_t = [(p_{\mathrm{R}} - p_{\mathrm{L}}) - P_\infty]\, \mathrm{e}^{-\Gamma_r t} + P_\infty. \qquad (3.26)$$

Thus, only track of the diagonal states of the initial RDM is kept in the incoherent regime.

(iii) The particular initial preparation is crucial in the underdamped regime at short-to-intermediate times. We shall study this regime in the next section.

(iv) The above methodology for expectation values can be extended to the computation of correlation functions for a general *factorized* (*i.e.*, nonequilibrium) initial preparation of the global system. Here we study the family of correlation functions

$$C_{ij}^{+}(t) := \langle \sigma_i(t)\sigma_j(0)\rangle = \mathrm{Tr}\,\{\sigma_i(t)\sigma_j\rho_0\}$$
$$C_{ij}^{-}(t) := \langle \sigma_j(0)\sigma_i(t)\rangle = \mathrm{Tr}\,\{\sigma_i(t)\rho_0\sigma_j\}. \qquad (3.27)$$

Upon defining $\tilde{\rho}_j := \sigma_j\rho_0$, the correlation functions $C_{ij}^{\pm}(t)$ can be written in the form of expectation values,

$$C_{ij}^{+}(t) = \mathrm{Tr}\,\{\sigma_i(t)\tilde{\rho}_j\},$$
$$C_{ij}^{-}(t) = \mathrm{Tr}\,\{\sigma_i(t)\tilde{\rho}_j^{\dagger}\}. \qquad (3.28)$$

Using the form (2.5) for ρ_0, we obtain the expressions

$$\tilde{\rho}_z = \frac{1}{2}\sigma_z + \frac{P_0}{2} I + \mathrm{i}a\sigma_y - \mathrm{i}b\sigma_x = \begin{pmatrix} p_{\mathrm{R}} & a - \mathrm{i}b \\ -a - \mathrm{i}b & -p_{\mathrm{L}} \end{pmatrix},$$

$$\tilde{\rho}_x = \frac{1}{2}\sigma_x - \frac{\mathrm{i}P_0}{2}\sigma_y + aI + \mathrm{i}b\sigma_z = \begin{pmatrix} a + \mathrm{i}b & p_{\mathrm{L}} \\ p_{\mathrm{R}} & a - \mathrm{i}b \end{pmatrix},$$

$$\tilde{\rho}_y = \frac{1}{2}\sigma_y + \frac{\mathrm{i}P_0}{2}\sigma_x - \mathrm{i}a\sigma_z + bI = \begin{pmatrix} b - \mathrm{i}a & -\mathrm{i}p_{\mathrm{L}} \\ \mathrm{i}p_{\mathrm{R}} & b + \mathrm{i}a \end{pmatrix}.$$

We wish to emphasize that the $\{\tilde{\rho}_j\}$ are not proper density matrices since they are not positive definite. Therefore, the similarity of the correlation expression (3.28) with an expectation value is only formal.

Consider now explicitly the position autocorrelation function $C_{zz}^{\pm}(t)$. Substituting into equation (3.28) the partial probabilities $P_{1/2}^{(\mathrm{s/a})}(t)$, we obtain

$$C_{zz}^{\pm}(t) = P_1^{(\mathrm{s})}(t) + (p_{\mathrm{R}} - p_{\mathrm{L}})P_1^{(\mathrm{a})}(t)$$
$$\pm \mathrm{i}[\,2aP_2^{(\mathrm{s})}(t) - 2bP_2^{(\mathrm{a})}(t)\,]. \qquad (3.29)$$

In the special case $p_{\mathrm{R}} - p_{\mathrm{L}} = P_\infty$, we find

$$\mathrm{Re}\, C_{zz}^{\pm}(t) = P_1^{(\mathrm{s})}(t) + P_\infty P_1^{(\mathrm{a})}(t), \qquad (3.30)$$

originally discussed in reference [44].

Also the correlation functions $C_{zx}^{\pm}(t)$ and $C_{zy}^{\pm}(t)$ can be expressed in terms of the functions $P_1^{(\mathrm{s/a})}(t)$ and $P_2^{(\mathrm{s/a})}(t)$. Using the above forms for $\tilde{\rho}_j$, we find

$$C_{zx}^{\pm}(t) = P_2^{(\mathrm{a})}(t) + 2aP_1^{(\mathrm{a})}(t)$$
$$\pm \mathrm{i}[2bP_1^{(\mathrm{s})}(t) - (p_{\mathrm{R}} - p_{\mathrm{L}})P_2^{(\mathrm{s})}(t)], \qquad (3.31)$$

$$C_{zy}^{\pm}(t) = P_2^{(\mathrm{s})}(t) + 2bP_1^{(\mathrm{a})}(t)$$
$$\pm \mathrm{i}[(p_{\mathrm{R}} - p_{\mathrm{L}})P_2^{(\mathrm{a})}(t) - 2aP_1^{(\mathrm{s})}(t)]. \qquad (3.32)$$

We see that the correlation functions (3.29), (3.31), and (3.32) are complex in general and that the probabilities and coherences are mixing their roles. The symmetrized correlation functions are real, and the antisymmetrized parts $\chi_{ij}(t) = (\mathrm{i}/\hbar)\langle[\,\sigma_i(t), \sigma_j(0)\,]_-\rangle$ describe the linear response of $\langle\sigma_i\rangle_t$ to the δ-perturbation $\delta H = -\delta(t) f_j \sigma_j$,

$$\delta\langle\sigma_i\rangle_t = \chi_{ij}(t) f_j.$$

We now turn to useful approximations.

4 Approximate treatments

4.1 The noninteracting-blip approximation (NIBA)

The noninteracting-blip approximation (NIBA) [14] has found broad application in studies of the tunneling dynamics. The extension of the NIBA to the driven case is reviewed in reference [18]. In the NIBA, the blip-blip interactions $\Lambda_{j,k}$ are neglected, and also the sojourn-blip interactions $X_{j,k}$ are disregarded except those of neighbors, $k=j-1$, and they are approximated by $X_{j,j-1}=Q''(\tau_j)$, with blip length $\tau_j=t_{2j}-t_{2j-1}$ (class A) or $\tau_j=t_{2j-1}-t_{2j-2}$ (class B). Hence in the NIBA, the influence functions in equation (3.5) factorize into the individual blip influence factors. In the absence of interblip correlations, the irreducible influence functions $\widetilde{F}^{(\pm)}_{n,i}$ are zero for $n>1$. Then the series expressions (3.11-3.18) for the irreducible kernels reduce to the terms of lowest order in $\Delta(t)$, given by the kernels without internal flips $\tilde{K}^{(\mathrm{s/a})}_i$ in (3.12, 3.16), and $\tilde{Y}^{(\mathrm{s/a})}_i$ in (3.18, 3.21). Evaluation of equations (3.17, 3.22, 3.8) with these kernels gives the complete dynamics of the spin-boson model in the NIBA.

The NIBA is appropriate at sufficiently high temperature and/or large friction, and/or large bias. For zero bias and absence of driving, the interblip correlations in $\langle\sigma_z\rangle_t$ are of order damping strength squared. Then the NIBA for $\langle\sigma_z\rangle_t$ is also systematic for very weak damping down to $T=0$, whereas it is inconsistent for $\langle\sigma_x\rangle_t$ at low T (see below). This has been independently confirmed numerically by a direct comparison of NIBA results with those of the interacting-blip chain approximation (IBCA) [40], the real-time quantum Monte-Carlo simulation method [41], and the QUAPI method [42].

In the absence of driving, the NIBA predicts complete destruction of the coherent motion above the crossover temperature T^* [14,16,43]. Above T^*, the effects of off-diagonal preparation die out on a much shorter time scale than the characteristic time scale for incoherent decay, and therefore are irrelevant. In the NIBA, the TSS approaches the equilibrium state

$$\begin{aligned}\langle\sigma_z\rangle_\infty &= P_\infty = \tanh(\hbar\epsilon_0/2k_{\mathrm{B}}T),\\ \langle\sigma_x\rangle_\infty &= (\Delta_0/\epsilon_0)\,\tanh(\hbar\epsilon_0/2k_{\mathrm{B}}T),\end{aligned} \tag{4.1}$$

exponentially fast with the relaxation rate $\Sigma^{(\mathrm{s})}(0)=\Gamma_r$,

$$\Gamma_r = \Delta_0^2\int_0^\infty \mathrm{d}\tau h^{(+)}(\tau)\cos(\epsilon_0\tau). \tag{4.2}$$

For weak damping and low T, the NIBA breaks down for a biased system. We see from equation (4.1) that the NIBA predicts at $T=0$ incorrect *symmetry breaking* for arbitrarily small bias ϵ_0, $P_\infty=\mathrm{sgn}\,(\epsilon_0)$, and also a violation of the unitarity bound $\langle\sigma_x\rangle_\infty\le 1$.

Off-diagonal contributions in the initial state may influence the dynamics considerably in the coherent regime $T<T^*$. The related discussion is postponed after the discussion of the systematic weak-damping treatment which is free of the NIBA's flaws.

4.2 Systematic weak damping approximation

For weak damping, nonzero bias and low temperatures, the bath correlations $\Lambda_{j,k}$ and $X_{j,k\neq j-1}$ contribute already to terms which depend linearly on the spectral density $J(\omega)$. These correlations are dropped in the NIBA, and therefore the NIBA breaks down in this regime. The kernels $K^{(\mathrm{s/a})}_A(t,t')$ and $Y^{(\mathrm{s/a})}_A(t,t')$ in (3.11) and (3.18) have been studied for weak damping in references [29] and [39], respectively. The kernels $K^{(\mathrm{s/a})}_B(t,t')$ and $Y^{(\mathrm{s/a})}_B(t,t')$ are given by similar expressions. The weak-damping form of $\langle\sigma_z\rangle_t$ is obtained by solving the GME (3.17) with the related weak-damping kernels, $\langle\sigma_y\rangle_t$ is found from equation (3.8), and $\langle\sigma_x\rangle_t$ is obtained from the integral relation (3.22). Consider now first the undriven dynamics.

4.2.1 Expectation values

In the absence of driving the GME (3.17) is in convolutive form and is conveniently solved by Laplace transformation. Putting $t_0=0$, the population is found as

$$\begin{aligned}P(t) = {}&(N_1+n_1)\,\mathrm{e}^{-\Gamma_r t} + (N_2+n_2)\cos(\nu t)\,\mathrm{e}^{-\Gamma t}\\ &+(-2b\Delta/\nu+n_3)\sin(\nu t)\,\mathrm{e}^{-\Gamma t}+P_\infty,\end{aligned} \tag{4.3}$$

with tunneling frequency $\nu=(\Delta^2+\epsilon_0^2)^{1/2}$, and equilibrium value $P_\infty=(\epsilon_0/\nu)\tanh(\hbar\beta\nu/2)$. The adiabatically dressed tunneling coupling is in the Ohmic case, $\Delta=\Delta_0(\Delta_0/\omega_c)^{\alpha/(1-\alpha)}$, and in the super-Ohmic case

$$\Delta = \Delta_0\exp\left(-\frac{1}{\pi}\int_0^\infty \mathrm{d}\omega\,\frac{J(\omega)}{\omega^2}\right). \tag{4.4}$$

The relaxation rate Γ_r and dephasing rate Γ read

$$\begin{aligned}\Gamma_r &= (\Delta^2/2\nu^2)J(\nu)\coth(\hbar\beta\nu/2),\\ \Gamma &= \Gamma_r/2+2\pi\alpha\delta_{s,1}(\epsilon_0/\nu)^2k_{\mathrm{B}}T/\hbar.\end{aligned} \tag{4.5}$$

The second term contributes to Γ only in the Ohmic case. Finally, the amplitudes are

$$\begin{aligned}N_1 &= (\epsilon_0/\nu)^2P_0+2a\epsilon_0\Delta/\nu^2-P_\infty,\\ N_2 &= (\Delta/\nu)^2P_0-2a\epsilon_0\Delta/\nu^2,\\ n_1 &= -n_2=-4b\Gamma_r\Delta/\nu^2,\\ n_3 &= (\Gamma_rN_1+\Gamma N_2)/\nu,\end{aligned} \tag{4.6}$$

where the n_i are of linear order in the bath coupling.

The coherences are readily found upon substituting the solution (4.3) into equation (3.22) as

$$\begin{aligned}\langle\sigma_x\rangle_t = {}&(M_1+m_1)\,\mathrm{e}^{-\Gamma_r t}+(M_2+m_2)\cos(\nu t)\,\mathrm{e}^{-\Gamma t}\\ &+[2b\epsilon_0/\nu+m_3]\sin(\nu t)\,\mathrm{e}^{-\Gamma t}+\langle\sigma_x\rangle_\infty,\end{aligned} \tag{4.7}$$

with $\langle\sigma_x\rangle_\infty=(\Delta/\nu)\tanh(\hbar\beta\nu/2)$, and amplitudes

$$\begin{aligned}M_1 &= (\epsilon_0\Delta/\nu^2)P_0+2a(\Delta/\nu)^2-\langle\sigma_x\rangle_\infty,\\ M_2 &= -(\epsilon_0\Delta/\nu^2)P_0+2a\epsilon_0^2/\nu^2,\\ m_1 &= -m_2=4b\epsilon_0\Gamma/\nu^2\\ m_3 &= (\Gamma_rM_1+\Gamma M_2)/\nu.\end{aligned} \tag{4.8}$$

We gather from these results that the coherent dynamics of $P(t)=\langle\sigma_z\rangle_t$ and $\langle\sigma_x\rangle_t$ at short-to-intermediate times is strongly affected by the chosen initial state. The evolution of the population difference $P(t)$ is shown in Figure 2 for three different initial preparations. We consider (I) a "standard" initial state in which the system is released from the right well ($p_{\rm R} = 1$, $a = b = 0$), *i.e.*, an eigenstate of σ_z, (II) a preparation of the TSS in the ground state ($p_{\rm R} = 1/2 + \epsilon_0/2\nu$; $p_{\rm L} = 1/2 - \epsilon_0/2\nu$; $a = \Delta/2\nu$ and $b = 0$), and (III) a preparation in the excited state ($p_{\rm R} = 1/2 - \epsilon_0/2\nu$; $p_{\rm L} = 1/2 + \epsilon_0/2\nu$; $a = -\Delta/2\nu$ and $b = 0$). Figure 2 clearly shows that initial preparation in an off-diagonal state is distinguished in the dynamics only in the initial time domain. Effects related to an off-diagonal preparation show up only in the short-time underdamped dynamics. We have already noted in Section 3.3 that the equilibrium value and the damping rate are independent of the initial state chosen. Hence all curves approach the same equilibrium value on the same time scale. Preparation in an eigenstate of σ_z gives rise to large-amplitude damped oscillations (full curve). On the other side, when the TLS is prepared at low T in the ground state, the initial population P_0 is already close to the equilibrium population P_∞. In this case, $P(t)$ shows only small-amplitude damped oscillations around the asymptotic value $P_\infty \approx P_0$ (dashed-dotted curve). When the TSS is prepared in the excited state, we have $P_0 \approx -P_\infty$. Then the system performs small-amplitude damped oscillations superimposed to the slow exponential relaxation towards the equilibrium population (dashed curve).

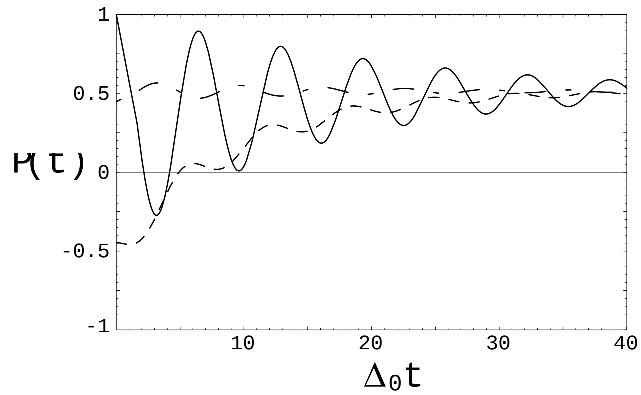

Fig. 2. The population $P(t) := \langle\sigma_z\rangle_t$ is sketched as a function of time for a standard preparation in the right state (full curve), preparation in the ground state (dashed curve), and preparation in the excited state (dashed-dotted curve). The parameters chosen are $\alpha = 0.05$, $\omega_c = 30\Delta_0$, $k_{\rm B}T = 0.05\hbar\Delta_0$, and $\epsilon_0 = 0.05\Delta_0$.

4.2.2 Nonequilibrium correlation functions

With the weak-damping expressions (4.3-4.6) for $P(t)$, it is straightforward to determine the partial expectation values. We find for $P_1^{\rm (s/a)}(t)$ the forms

$$P_1^{\rm (s)}(t) = (\epsilon_0^2/\nu^2)\,{\rm e}^{-\Gamma_r t} + (\Delta^2/\nu^2)\cos(\nu t)\,{\rm e}^{-\Gamma t} + [\,\epsilon_0^2\Gamma_r/\nu^3 + \Delta^2\Gamma/\nu^3\,]\sin(\nu t)\,{\rm e}^{-\Gamma t}, \quad (4.9)$$

$$P_1^{\rm (a)}(t) = P_\infty[\,1 - {\rm e}^{-\Gamma_r t} - (\Gamma_r/\nu)\sin(\nu t)\,{\rm e}^{-\Gamma t}\,], \quad (4.10)$$

and for the partial probabilities $P_2^{\rm (s/a)}(t)$

$$P_2^{\rm (s)}(t) = -2(\Gamma\Delta/\nu^2)\,{\rm e}^{-\Gamma_r t} + 2(\Gamma\Delta/\nu^2)\cos(\nu t)\,{\rm e}^{-\Gamma t} - (\Delta/\nu)\sin(\nu t)\,{\rm e}^{-\Gamma t}, \quad (4.11)$$

$$P_2^{\rm (a)}(t) = (\epsilon_0\Delta/\nu^2)\,{\rm e}^{-\Gamma_r t} - (\epsilon_0\Delta/\nu^2)\cos(\nu t)\,{\rm e}^{-\Gamma t} + [\,\epsilon_0\Delta(\Gamma_r - \Gamma)/\nu^3\,]\sin(\nu t)\,{\rm e}^{-\Gamma t}. \quad (4.12)$$

Substituting these forms into equations (3.29, 3.31), and into equation (3.32), we obtain the weak-damping expressions for the correlation functions $C_{zj}(t)$ ($j = x, y, z$).

4.2.3 Driven dynamics

Finally, let us study the effect of a monochromatic ac-field modulating the bias energy,

$$\varepsilon(t) = \epsilon_0 + \hat\epsilon\cos(\Omega t), \qquad \Delta(t) = \Delta_0. \quad (4.13)$$

The full dynamics can be worked out by generalizing the methods developed for the standard preparation [18] to the general initial state (2.5). Here we only wish to answer the intriguing question: Is it possible to slow down the bath-induced decoherence in such a way that preparation effects persist for longer times than in the absence of driving?

Consider high-frequency driving, $\Omega \gg \Delta_0$, ν, Γ, Γ_r, and the dynamics described by the field-averaged GME obtained from (3.17) upon substituting the field-averaged forms $\overline{K}_i^{\rm (s/a)}$ for the kernels $K_i^{\rm (s/a)}$. This is formally obtained upon performing in $K_i^{\rm (s/a)}$ the substitutions

$$\cos[\zeta(t,t')] \to J_0[f(t-t')]\cos[\epsilon_0(t-t')] \equiv \mathcal{C}(t-t'),$$
$$\sin[\zeta(t,t')] \to J_0[f(t-t')]\sin[\epsilon_0(t-t')] \equiv \mathcal{S}(t-t'),$$

where $f(t-t') = (2\hat\epsilon/\Omega)\sin[\Omega(t-t')/2]$. For example, the averaged weak-damping kernel $\overline{K}_A^{\rm (s)}$ takes the form

$$\overline{K}_A^{\rm (s)}(t-t') = \Delta_0^2\,\mathcal{C}(t-t')[1 - Q'(t-t')] + \Delta_0^4\int_{t'}^{t}{\rm d}t_2\int_{t'}^{t_2}{\rm d}t_1\,\mathcal{S}(t-t_2)\mathcal{P}(t_2-t_1)\mathcal{S}(t_1-t') \times[Q'(t-t') + Q'(t_2-t_1) - Q'(t_2-t') - Q'(t-t_1)]. \quad (4.14)$$

The first term represents the NIBA. In the residual contribution, the term $\mathcal{P}(t_2 - t_1)$ is sandwiched between two blips with intervals $t_1 - t'$ and $t - t_2$. This term describes all possible transitions of the *undamped* system between diagonal states during the period $t_2 - t_1$,

$$\mathcal{P}(t-t_0) = 1 + \sum_{n=1}^{\infty}(-1)^n\int_{t_0}^{t}\mathcal{D}_{2n}\{t_j\}\prod_{j=1}^{n}\mathcal{C}(t_{2j} - t_{2j-1}).$$

The resulting GME is convolutive. The respective pole condition is conveniently studied using the relation

$$J_0\left(\frac{2\hat{\epsilon}}{\Omega}\sin\frac{\Omega t}{2}\right) = \sum_{n=-\infty}^{\infty} J_n^2(\hat{\epsilon}/\Omega)\cos(n\Omega t), \qquad (4.15)$$

which suggests to introduce the channel tunneling frequencies $\Delta_{\mathrm{eff},n} = J_n(\hat{\epsilon}/\Omega)\Delta_0$, and side frequencies $\epsilon_n^\pm = \epsilon_0 \pm n\Omega$. The weak-damping dynamics can now be investigated by generalizing the line of reasoning proposed in reference [29], where to lowest order in $\hat{\epsilon}/\Omega$ only three effective tunneling frequencies and three side frequencies occurred, to an *infinite* set of effective tunneling and side frequencies. For small static bias $\epsilon_0 \ll \Omega$, and for the ratio $\hat{\epsilon}/\Omega$ chosen away from the zeros of $J_0(\hat{\epsilon}/\Omega)$, one finds that the dynamics behaves like for a static bias, but with effective tunneling matrix element $\Delta_{\mathrm{eff},0}$. Because $\Gamma_r, \Gamma \propto \Delta_{\mathrm{eff},0}^2 \le \Delta_0^2$, both the dephasing rate Γ and the relaxation rate Γ_r can be strongly reduced by a suitable choice of the parameters of the driving field. The same reasoning is applicable when an ac-field is applied which satisfies the resonance condition $\epsilon_n^- = 0$. In this case, when $\hat{\epsilon}/\Omega$ is away from the zeros of $J_n(\hat{\epsilon}/\Omega)$, the decay rates are proportional to $\Delta_{\mathrm{eff},n}^2 < \Delta_0^2$. This implies again that the *field-averaged* population difference $\overline{P}(t)$ and coherence $\overline{\langle\sigma_x\rangle}_t$ will be approximately given on a longer time scale by

$$\overline{P}(t) \approx p_{\mathrm{R}} - p_{\mathrm{L}}, \quad \overline{\langle\sigma_x\rangle}_t \approx 2a \qquad (4.16)$$

than in the absence of driving. Thus, the decay of the field-averaged RDM towards the average stationary state can be slower than in the absence of driving.

At the zeros of $J_n(\hat{\epsilon}/\Omega)$ (with n determined by the resonant or near resonance condition $|\epsilon_n^-| \ll \Omega$), a more careful analysis must be performed because several terms of the series expansion (4.15) may be relevant for the TSS dynamics. However, the important point now is that the decay towards equilibrium, as well as coherent tunneling, are strongly suppressed by the ac-field. The maximum suppression occurring at the zeros of $J_n(\hat{\epsilon}/\Omega)$. For a standard initial preparation ($p_{\mathrm{R}} = 1$), we recover from the first relation in (4.16) an effect which has been termed "coherent destruction of tunneling" [19]: a particle initially localized in one well will remain localized over several tunneling periods. The present results, however, are more general: in the absence of dissipation and for parameters at the zeros of $J_n(\hat{\epsilon}/\Omega)$, the ac-field acts in such a way that *any* particular initial preparation is maintained. This driving-induced effect also persists when the TSS is additionally coupled to a thermal bath. This effect can also be studied in the underdamped regime within the NIBA.

At this point, a more precise statement should me made. Upon averaging over the driving field, the field-induced oscillatory contributions about the field-averaged dynamics are disregarded. Hence, the field-averaged dynamics describes the actual dynamics only well if the amplitude of these oscillations is small. For particular parameter regimes, it turns out that this approximation may well describe the dynamics of $P(t)$ in the presence of a high frequency field, whereas the coherences may perform already large amplitude oscillations around their mean values [46]. In this case, time-dependent corrections to the average behavior of $\overline{\langle\sigma_x\rangle}_t$, equation (4.16), are relevant.

5 Conclusions

In conclusion, we have studied the dynamics of the driven dissipative two-state system for an arbitrary factorized initial state of the reduced density matrix. We have shown that complete information about the reduced dynamics is contained in the position expectation value $\langle\sigma_z\rangle_t$. Namely, $\langle\sigma_z\rangle_t$ obeys an exact generalized master equation, and $\langle\sigma_x\rangle_t$ is related to $\langle\sigma_z\rangle_t$ by an integral relation. The other coherence $\langle\sigma_y\rangle_t$ follows from $\langle\sigma_z\rangle_t$ by differentiation. We have also discussed the nonequilibrium correlation functions $C_{zj}^\pm(t) = \langle\sigma_z(t)\sigma_j(0)\rangle$. As a general feature, it turns out that characteristic quantities of the dynamics, such as the asymptotic expectation values, the dephasing and relaxation rates, and the transition temperature T^* above which quantum coherence is destroyed are "universal", *i.e.*, do not depend on the particular initial state chosen. Above T^*, only preparation effects which are related to a diagonal initial state of the reduced density matrix show up in the dynamics. In contrast, below T^* also the coherences of the initial RDM may have significant effects on the dynamics at short-to-intermediate times. To study these effects, we have presented different analytical approaches appropriate in the whole interesting temperature and damping regime. In particular, we have shown that a suitably tuned high-frequency ac-field may prolong *any* dynamical effect of the particular initial preparation. The phenomenon of driving-induced coherent suppression of tunneling, $P(t) = 1$ in reference [19], turns out to be a particular manifestation of the more general results given here.

The authors are grateful to G. Giaquinta, I. Goychuk, P. Hänggi, L. Hartmann, G. Lang and M. Thorwart for interesting discussions. E.P. acknowledges financial support by the INFM under the PRA-QTMD programme. M.G. is grateful to the Max-Planck-Institut für Festkörperforschung (Stuttgart) and to the INFM under the PRA-QTDM programme for financial support.

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