Dynamics of the dissipative two-state system under ac modulation of bias and coupling energy

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We consider a driven dissipative two-state system subject to periodic modulation of *both* of its two parameters. The exact master equation that governs the dynamics is derived. Closed form analytical solutions are presented within the noninteracting-blip approximation for the stochastic forces, and for the *exactly* solvable case $\alpha=1/2$ of the Ohmic viscosity. We apply these results to a tight-binding particle with zero intrinsic bias. Selection rules are obeyed for arbitrary dissipation. For Ohmic dissipation, the effects of fast asymmetry modulation result in an overall *reduction* of quantum coherence and in a possible localization. On the contrary, a multiplicative modulation of the tunneling coupling leads to an exponential *enhancement* of tunneling. [S1063-651X(96)50910-4]

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The problem of a quantum particle coupled to a thermal bath and tunneling through the barrier of a slightly asymmetric double-well potential is ubiquitous in many physical and chemical systems. It can model for example long range electron-transfer reactions [1], the tunneling of atoms between an atomic-force microscope tip and a surface [2], or the magnetic flux in a superconducting quantum interference device (SQUID) [3]. At sufficiently low temperatures the dynamics only involves the ground states of the potential minima, and the system can be effectively restricted to the two-dimensional Hilbert space spanned by the two ground states. This two-level system (TLS), when isolated from the thermal bath, is the simplest system exhibiting quantum interference effects, as it can be prepared to oscillate clockwise between the eigenstates in the left and right well. Quite generally, the stochastic influence results in a reduction of the coherent tunneling motion by incoherent processess [4,5], and may even lead to a transition to self-trapping at zero temperature [6]. An important question is to which degree the tunneling dynamics is influenced by externally applied time-dependent fields. Up to now, only the effects of an external ac field modulating the asymmetry energy between the localized states were considered [7–15]. In particular, a complete destruction of tunneling can be induced by a coherent driving field of appropriate frequency and strength [7]. This effect can be stabilized in the presence of dissipation [8,10]. The transition temperature, above which quantum coherence is destroyed by a stochastic environment, is modified by a driving field [10]. A novel non-Markovian dynamics may arise due to driving induced correlations between tunneling transitions [9,11,12]. The response to the weak coherent signal [13,14] or the asymptotic tunneling amplitude [15] may be enhanced for optimal values of the stochastic forces.

In this work we generalize the model to include the possibility of external (*multiplicative* or *additive*) modulation of the coupling energy between the localized states. As a working model we consider the time-dependent spin-boson Hamiltonian where the bath is described as an ensemble of harmonic oscillators with a bilinear coupling in the TLS-bath coordinates

$$H(t)=-\frac{\hbar}{2}[\Delta(t)\sigma_x+\varepsilon(t)\sigma_z] +\frac{1}{2}\sum_\alpha\left(\frac{p_\alpha^2}{m_\alpha}+m_\alpha\omega_\alpha^2x_\alpha^2-c_\alpha x_\alpha d\sigma_z\right). \qquad (1)$$

Here the σ's are Pauli matrices, and the eigenstates of σ_z are the basis states in a localized representation where d is the tunneling distance. The tunneling splitting energy is given by $\hbar\Delta(t)$ while the asymmetry energy is $\hbar\varepsilon(t)$. This time dependence of the coupling parameter could arise, for example, from an ac modulation of the barrier height or width of the underlying double-well potential, and could be realized in a superconducting loop with two Josephson junctions [3]. The case of a dichotomically fluctuating tunneling coupling in bridge-assisted electron-transfer has recently been investigated in [16]. We observe that, as expressed by Eq. (8) below, while the effect of asymmetry modulation is additive, a barrier modulation results in a *multiplicative* (exponential) modification of the coupling parameter [17].

In the following we derive exact formal solutions for the dissipative dynamics and discuss closed form solutions within the noninteracting-blip approximation (NIBA) for the stochastic forces. Selection rules for the asymptotic dynamics are found. Further, for the special case $\alpha=1/2$ of the Ohmic viscosity, we obtain *exact* results. We then apply our findings to study the influence of a periodic modulation of the tunneling splitting of a *symmetric* TLS in an Ohmic environment. In contrast to the case of asymmetry driving, which implies an overall *reduction* of quantum coherence by fast ac fields and a possible localization in one of the tight-binding states, we find a possible *enhancement* of quantum coherence, and always an increase in the relaxation rate.

Suppose now that at times $t<0$ the particle is held at the site $\sigma_z=1$ with the bath having a thermal distribution. We then compute the probability $\langle\sigma_z(t)\rangle\equiv P(t)$ at times $t\geqslant 0$ for this factorizing initial state. After tracing out the thermal bath, all environmental effects are captured by the twice-integrated bath correlation function [4,5] ($\beta=1/k_BT$)

$$Q(t)=\frac{d^2}{\pi}\int_0^\infty d\omega\,\frac{J(\omega)}{\omega^2}\,\frac{\cosh[\omega\beta/2]-\cosh[\omega(\beta/2-it)]}{\sinh[\omega\beta/2]},$$

where $J(\omega)=(\pi/2)\Sigma_\alpha(c_\alpha^2/m_\alpha\omega_\alpha)\delta(\omega-\omega_\alpha)$ is the spectral density of the heat bath. Upon summing over the history of the system's visits of the four states of the reduced density matrix, we can generalize previous findings for the evolution of an asymmetry driven damped system [9,11,12] to find the formal solution in the form of a series in the number of time-ordered tunneling transitions. Introducing the notation $\delta_n=\Pi_{j=1}^{2n}\Delta(t_j)$, it reads

$$P(t)=1+\sum_{n=1}^{\infty}(-1)^n\int_0^t dt_{2n}\int_0^{t_{2n}}dt_{2n-1}\cdots\int_0^{t_2}dt_1$$

$$\times 2^{-n}\sum_{\{\xi_j=\pm1\}}(F_n^{(+)}C_n^{(+)}-F_n^{(-)}C_n^{(-)}),\tag{2}$$

$$C_n^{(+)}=\delta_n\cos\Phi_n\,,C_n^{(-)}=\delta_n\sin\Phi_n\,.\tag{3}$$

Here the ξ charges label the two off-diagonal states of the reduced density matrix. The phase Φ_n describes the influence of the time-dependent biasing forces,

$$\Phi_n=\sum_{j=1}^{n}\xi_j[g(t_{2j})-g(t_{2j-1})],\tag{4}$$

where $g(t)=\int^t dt'\varepsilon(t')$. All the dissipative influences are in the functions $F_n^{(\pm)}$ To express them in compact form, we introduce the functions $Q_{j,k}=Q(t_j-t_k)$ and

$$\Lambda_{j,k}=Q'_{2j,2k-1}+Q'_{2j-1,2k}-Q'_{2j,2k}-Q'_{2j-1,2k-1}\,,$$

$$X_{j,k}=Q''_{2j,2k+1}+Q''_{2j-1,2k}-Q''_{2j,2k}-Q''_{2j-1,2k+1}\,,$$

where $Q'(t)$ and $Q''(t)$ are the real and imaginary part of the bath correlation function $Q(t)$, respectively. Denoting as *sojourns* the periods $t_{2j}<t'<t_{2j+1}$ in which the system is in a diagonal state, and as *blips* the periods $t_{2j-1}<t'<t_{2j}$ in which the system stays in one of the two off-diagonal states (cf. Refs. [4,5]), the function $\Lambda_{j,k}$ describes the interblip correlations of the blip pair $\{j,k\}$, while the function $X_{j,k}$ describes the correlations of the blip j with a preceding sojourn k. Then, all intrablip and interblip correlations of n blips are combined in the expression

$$G_n=\exp\left(-\sum_{j=1}^{n}Q'_{2j,2j-1}\sum_{j=2}^{n}\sum_{k=1}^{j-1}\xi_j\xi_k\Lambda_{j,k}\right).$$

Upon introducing the influence phases describing the correlations between the kth sojourn and the $n-k$ succeeding blips, $\eta_{n,k}=\Sigma_{j=k+1}{}^{n}\xi_jX_{j,k}$, the full influence functions take the form

$$F_n^{(+)}=G_n\prod_{k=0}^{n-1}\cos\eta_{n,k}\,,\quad F_n^{(-)}=F_n^{(+)}\tan\eta_{n,0}\,.\tag{5}$$

Up to now our results are exact. Further, having captured the bath and driving correlations in the influence functions $F_n^{(\pm)}$ and in the coefficients $C_n^{(\pm)}$ respectively, the exact master equation for the probability $P(t)$ can be derived from Eq. (2) as prescribed in Ref. [12]. It reads

$$\dot{P}(t)=\int_0^t dt'[K^{(-)}(t,t')-K^{(+)}(t,t')P(t')],\tag{6}$$

where the kernels $K^{(\pm)}(t,t')$ are defined by a series expression in δ_n. In particular, within the NIBA [4], which is formally obtained neglecting the interblip correlations ($\Lambda_{j,k}=0$) and all blip-sojourn correlations ($X_{j,k}=0$ for $j\neq k+1$), the kernels in Eq. (6) reduce to the expressions

$$K^{(+)}(t,t')=e^{-Q'(t-t')}\cos[Q''(t-t')]C_1^{(+)}(t,t'),$$

$$K^{(-)}(t,t')=e^{-Q'(t-t')}\sin[Q''(t-t')]C_1^{(-)}(t,t').\tag{7}$$

It is interesting to observe that the polaron transformation approach discussed in [10,16] leads [18], if applied to the Hamiltonian (1), to a master equation analogous to Eq. (6), and with kernels (7).

Let us now focus on the effect of ac modulation of the TLS parameters in the driven Hamiltonian (1). We shall assume the analytic forms

$$\varepsilon(t)=\epsilon_0+\epsilon\cos\Omega_\epsilon t,\quad \Delta(t)=\Delta_0\exp(\delta\cos\Omega_\delta t),\tag{8}$$

where ϵ_0 and Δ_0 represent the asymmetry energy and the tunneling coupling in the absence of driving fields, respectively. The effects of asymmetry modulation [i.e., $\delta=0$ in Eq. (8)] have been the object of intense research in the past years [8–12]. This we shall denote as situation I. For comparison, we performed the analysis of the dissipative TLS dynamics under (on physical grounds small) modulation of the tunneling splitting [$\epsilon=0$, $\delta<1$ in Eq. (8)]. This we shall indicate as situation II. Further, we introduce a subscript $\zeta=\epsilon$ or δ which, refers to quantities in case I or II, respectively. Equation (6) is conveniently solved by Laplace transformation. Introducing the Laplace transform $\hat{P}(\lambda)=\int_0^\infty dte^{-\lambda t}P(t)$ of $P(t)$, one finds

$$\lambda\hat{P}(\lambda)=1+\int_0^\infty dte^{-\lambda t}[\hat{K}_\lambda^{(-)}(t)-\hat{K}_\lambda^{(+)}(t)P(t)],\tag{9}$$

where $\hat{K}_\lambda^{(\pm)}(t)=\int_0^\infty dt'e^{-\lambda t'}K^{(\pm)}(t+t',t)$. For case I or II the kernels $\hat{K}_\lambda^{(\pm)}(t)$ have the periodicity of the external field and can be expanded in Fourier series,

$$\hat{K}_\lambda^{(\pm)}(t)=\sum_{m=-\infty}^{\infty}k_m^{\pm}(\lambda)e^{-im\Omega_\zeta t},\tag{10}$$

hence allowing a recursive solution [11]. Some features of the two different realizations can now be discussed

(i) For a TLS with intrinsic bias $\epsilon_0\neq0$, the asymptotic dynamics is in both cases periodic in time with the periodicity $2\pi/\Omega_\zeta$ of the driving force, i.e., $\lim_{t\to\infty}P(t)=P^{(\mathrm{as})}(t)=\Sigma_m p_m e^{-im\Omega_j t}$ where

$$p_0=\frac{k_0^-(0)}{k_0^+(0)}-\sum_{m\neq0}\frac{k_m^+(0)}{k_0^+(0)}p_m\,,$$

and for $m\neq0$

$$p_m=\frac{i}{m\Omega_\zeta}\left(k_m^-(-im\Omega_\zeta)-\sum_n k_{m-n}^+(-im\Omega_\zeta)p_n\right).$$

For case I, within the NIBA, the functions $k_m^\pm$ are explicitly given in [11]. For case II we obtain

$$k_m^-(\lambda)=(-1)^m\Delta_0^2\int_0^\infty d\tau e^{-\lambda\tau-Q'(\tau)}$$
$$\times e^{-im\Omega_\delta(\tau/2)}\sin[Q''(\tau)]\sin(\epsilon_0\tau)I_m\left(2\delta\cos\frac{\Omega_\delta\tau}{2}\right),\tag{11}$$
$$k_m^+(\lambda)=(-1)^m\Delta_0^2\int_0^\infty d\tau e^{-\lambda\tau-Q'(\tau)}$$
$$\times e^{-im\Omega_\delta(\tau/2)}\cos[Q''(\tau)]\cos(\epsilon_0\tau)I_m\left(2\delta\cos\frac{\Omega_\delta\tau}{2}\right),$$

where $I_m(z)$ denotes the modified Bessel function.

(ii) For a *symmetric* TLS, spatial symmetry properties of the kernels $k_m^\pm(\lambda)$ imply that all the Fourier components of $P^{(as)}(t)$ with *even* index vanish in case I [11]. We find here that *all* the harmonics vanish in case II. We note that the same selection rules are known to hold in driven classical symmetric systems [19].

(iii) As shown by Eqs. (6) or (9), the transient as well as the long-time dynamics depends on a complicated interplay between the stochastic and driving forces. Some simplifications are allowed when a separation of time scales is possible. Here we shall focus on the interesting high frequency regime $\Omega_\zeta\gg\tau_K^{-1}$ where τ_K is the characteristic time of the transient dynamics. In this approximation, the kernels $\hat{K}_\lambda^{(\pm)}\times(t)$ in Eq. (9) can be substituted with their average $k_0^\pm(\lambda)$ over a period. One obtains (for convenience we explicitly indicate the field dependence),

$$\hat{P}(\lambda)=\frac{1+k_0^-(\lambda;\zeta)/\lambda}{\lambda+k_0^+(\lambda;\zeta)},\tag{12}$$

where the condition $\Omega_\zeta\gg\tau_K^{-1}\simeq|\lambda|$ has to be proofed self-consistently. Thus, a fast field suppresses the periodic long-time oscillations, and the TLS approaches incoherently the steady value $p_0=k_0^-(0;\zeta)/k_0^+(0;\zeta)$ with relaxation rate $k_0^+(0;\zeta)\equiv\Gamma_\zeta$. For case II the relaxation rate Γ_δ is immediately evaluated from Eq. (11). For case I, the corresponding rate Γ_ϵ is obtained again from Eq. (11) by substituting $I_0(2\delta\cos(\Omega_\delta\tau/2))$ with $J_0((2\epsilon/\Omega_\epsilon)\sin(\Omega_\epsilon\tau/2))$, where $J_0(z)$ is the zero order Bessel function of first kind. Finally, in the limit $\zeta\to0$ the modified rates Γ_ζ reduce to the static one Γ_0 [4,5].

To make quantitative predictions, we consider the case of Ohmic dissipation where the spectral density takes the form $J(\omega)=(2\pi\hbar^2/d^2)\alpha\omega e^{-\omega/\omega_c}$, with α the dimensionless coupling strength and ω_c a cut-off frequency. Then the bath correlation functions assume the form $Q'(\tau)=\alpha\ln[1+\omega_c^2\tau^2]+2\alpha\ln[(\hbar\beta/\pi\tau)\sinh(\pi\tau/\hbar\beta)]$, $Q=2\alpha\tan^{-1}(\omega_c\tau)$ [4,5]. Further, we restrict ourselves to the case of a TLS with zero intrinsic bias ($\epsilon_0=0$).

As a first feature, because $|J_0(z)|\leqslant1$, it is apparent that for a symmetric TLS the effect of a fast asymmetry modulation is an overall reduction of the incoherent tunneling rate Γ_ϵ as compared to the static one Γ_0 whenever $\alpha<1/2$. On the contrary, because $I_0(z)\geqslant1$, the tunneling rate Γ_δ is always *increased* in case II.

Second, we study the modification of the quantum coherent motion by stochastic and driving forces, i.e., we explicitly determine the poles of Eq. (12) resulting from the equation $\lambda+k_0^+(\lambda;\zeta)=0$. For our purposes it is convenient to express the kernels $k_0^+(\lambda;\zeta)$ in terms of the static one $\mathcal{K}(\lambda)\equiv\lim_{\zeta\to0}k_0^+(\lambda;\zeta)$ as

$$k_0^+(\lambda;\zeta)=\sum_{n=-\infty}^{\infty}H_n(\zeta)\mathcal{K}(\lambda+in\Omega_\zeta),\tag{13}$$

where $H_n(\epsilon)=J_n^2(\epsilon/\Omega_\epsilon)$, $H_n(\delta)=I_n^2(\delta)$, and

$$\mathcal{K}(\lambda)=\frac{\Delta_e}{\pi}\left(\frac{\hbar\beta\Delta_e}{2\pi}\right)^{1-2\alpha}\frac{h(\lambda)}{\alpha+\hbar\beta\lambda/2\pi},\tag{14}$$

$$h(\lambda)=\Gamma(1+\alpha+\hbar\beta\lambda/2\pi)/\Gamma(1-\alpha+\hbar\beta\lambda/2\pi).\tag{15}$$

Here we made the high frequency approximation $\omega_c\tau\gg1$ in the correlation functions $Q'(\tau)$ and $Q''(\tau)$, and a necessary condition for Eq. (12) becomes $\hbar\beta\Omega_\zeta\gg2\pi\alpha$. Here $\Gamma(z)$ denotes the gamma function and $\Delta_e=\Delta_0(\Delta_0/\omega_c)^{\alpha/(1-\alpha)}\times[\cos(\pi\alpha)\Gamma(1-2\alpha)]^{1/(2-2\alpha)}$ is the bath-renormalized tunneling splitting when $\alpha<1$. Using Eq. (14), the pole equation predicts for the static case with $\alpha\leqslant1/2$ a destruction of quantum coherence by bath-induced incoherent transitions above a transition temperature $T_0(\alpha)$ [4,5]. For $\alpha\geqslant1/2$ the dynamics is incoherent down to $T=0$. For weak Ohmic damping $\alpha\ll1$ one has from Eq. (15) that $h(\lambda)\simeq1$. Hence, the pole equation becomes just a quadratic equation in λ and the transition temperature is determined by the condition of the solutions being real and degenerate [4,5]. Such a situation is expected to be strongly modified if the additional influence of ac field is considered. Taking into account the high-frequency condition $\Omega_\zeta\gg|\lambda|$, up to the order $O(|\lambda|/\Omega_\zeta)^4$ and for $\alpha\ll1$, we find

$$k_0^+(\lambda;\zeta)=\mathcal{K}(\lambda)\left[H_0(\zeta)+\left(\frac{\alpha+\hbar\beta\lambda/2\pi}{\hbar\beta\Omega_\zeta/2\pi}\right)^2\sum_{n\neq0}\frac{H_n(\zeta)}{n^2}\right].$$

This equation is of fundamental importance in understanding the role of driving fields on the tunneling dynamics. Its two addenda act, in fact, in preserving or suppressing quantum coherence, respectively. When the first contribution dominates, the effect of a fast field is roughly to renormalize the effective tunneling matrix element Δ_e as $\Delta_e\to\Delta_\zeta$, where $\Delta_\epsilon=\Delta_e|J_0(\epsilon/\Omega_\epsilon)|^{1/1-\alpha}$ and $\Delta_\delta=\Delta_eI_0(\delta)^{1/1-\alpha}$ for case I or II, respectively. Hence, from static considerations, we find the transition temperature $T_\zeta(\alpha)\simeq\hbar\Delta_\zeta/\pi\alpha k_B$ when $\alpha\ll1$. Because $T_\epsilon(\alpha)\leqslant T_0(\alpha)$, the effect of asymmetry driving is an overall reduction of quantum coherence [10]. Near the zeroes of $J_0(\epsilon/\Omega_\epsilon)$ quantum coherence is strongly suppressed and the particle tunnels incoherently with rate $\Gamma_\epsilon=\Gamma_0(2\pi\alpha/\hbar\beta\Omega_\epsilon)^2\Sigma_{n\neq0}J_n^2(\epsilon/\Omega_\epsilon)/n^2$ down to $T=0$. Because $\Gamma_\epsilon<\Gamma_0\ll\Omega_\epsilon$, suppression of tunneling may be stabilized for weak dissipation over several periods of the driving force in accordance with previous findings [8]. On the other hand, because $I_0(\delta)\geqslant1$, this simple effect of renormalization of tunneling always holds true in case II. Being $T_\delta(\alpha)\geqslant T_0(\alpha)$, quantum coherence is always enhanced in case II.

Finally, we conclude by discussing the *exactly* solvable case of strong friction $\alpha=1/2$, which is imperative to understanding the role of driving-induced correlations on the TLS dynamics. Generalizing the technique discussed in Ref. [9], the series (3) can indeed be summed up exactly to give for $P(t)=P^{(a)}(t)+P^{(s)}(t)$ the result

$$P^{(a)}(t)=\int_0^t dt_2 \exp\left(-\int_{t_2}^t ds\,\gamma(s)\right)\int_0^{t_2} dt_1 \Delta(t_2)\Delta(t_1) \times e^{-Q'(t_2-t_1)}\exp\left(-\int_{t_1}^{t_2} ds\,\gamma(s)/2\right)\sin[g(t_2)-g(t_1)],$$

$$P^{(s)}(t)=\exp\left(-\int_0^t ds\,\gamma(s)\right), \tag{16}$$

where we introduced the relaxation rate $\gamma(t)=\pi\Delta^2(t)/2\omega_c$. Here, $P^{(a)}(t)$ represents the contribution to $P(t)$ antisymmetric with respect to the bias inversion $\varepsilon\to-\varepsilon$, and determines the long-time dynamics (cf. [9] for case I). The investigation of the symmetric contribution $P^{(s)}(t)$ is, on the other hand, useful to get information on the transient dynamics. As expected, Eq. (16) predicts that in the absence of driving a symmetric TLS undergoes, for *any* temperature, incoherent relaxation (straight line of Fig. 1) with rate $\Delta_0^2/2\omega_c=\Delta_e$. Further, $P^{(s)}(t)$ is not sensible to asymmetry modulation. The effect of modulation of coupling is shown in Fig. 1, where $P^{(s)}(t)$ is depicted for different driving frequencies. When $\Omega_\delta>\Delta_e$ driving-induced coherent oscillations are superimposed onto the incoherent motion (dashed and dotted lines). These oscillations successively vanish as the driving frequency is decreased (dash-dotted line). This results in an incoherent relaxation towards equilibrium, which is faster with respect to the static case.

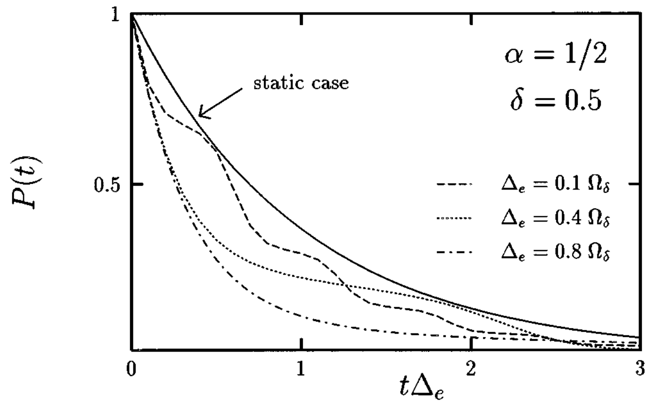

FIG. 1. Time-dependent population of a symmetric TLS under modulation of tunneling coupling for different driving frequencies and for Ohmic strength $\alpha=1/2$.

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