Comment on "Transient dynamics of Ohmic dissipative two-level systems driven by dc-ac fields" by Wang and Zhao

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Abstract

Recent results on the dynamics of a two-state system driven by dc-ac fields are discussed. We show that the approximation put forward in the work by Wang and Zhao [Phys. Lett. A 217 (1996) 232] gives *qualitative* incorrect results for the dynamics when effects of the dc field in the presence or absence of the ac field are considered.

The dynamics of a dissipative two-level system (TLS) driven by strong laser fields has been the object of intense investigations in past years [1–8]. Here we report on the recent advances on the dynamics of the dissipative two-level system as developed in Refs. [2–8]. By use of a direct comparison, we demonstrate that the approximative results put forward by Wang and Zhao [9] are in disagreement with nonapproximated results of the noninteracting-blib approximation (NIBA) in Refs. [2,5–7].

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

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

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 $\hbar\Delta$, while the asymmetry energy is $\hbar\varepsilon(t) = \hbar(\epsilon_0 + \hat{\epsilon}\cos\Omega t)$ and describes the coupling with external dc-ac fields. As far as regards the influence of the bath on the TLS dynamics, all information is captured in the twice-integrated bath correlation function [10,11] ($\beta = 1/k_{\rm B}T$), i.e.,

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

where $J(\omega) = \frac{1}{2}\pi\sum_\alpha(c_\alpha^2/m_\alpha\omega_\alpha)\delta(\omega-\omega_\alpha)$ is the bath spectral density. For Ohmic dissipation it has the form $J(\omega) = (2\pi\hbar^2/d^2)\alpha\omega{\rm e}^{-\omega/\omega_c}$, with α denoting the dimensionless coupling strength and ω_c a cut-off frequency.

PII S0375-9601(96)00902-4

Finally, the quantity of interest to investigate the TLS dynamics is the expectation value $\langle\sigma_z(t)\rangle \equiv P(t,t_0)$ where we suppose that the particle was held at the site $\sigma_z = 1$ at times $t < t_0$ with the bath having a thermal distribution. An exact path-integral solution for the expectation value $\langle\sigma_z(t)\rangle$ has first been obtained in Ref. [2], together with closed form analytical solutions valid at low-frequency driving fields, within the NIBA. On the other hand, a master equation for the case of a symmetric TLS that covers only the transient dynamics was obtained within the NIBA first in Ref. [3], by addressing the high-frequency regime. The generalization of the NIBA master equation for high frequencies and a biased TLS was discussed afterwards by Dakhnovskii in Ref. [4,5]. The *exact* (i.e. valid beyond the NIBA) non-Markovian master equation that governs the TLS dynamics has recently been obtained in Ref. [7]. The NIBA master equation for the asymmetric TLS reads [4–8]

$$\dot{P}(t,t_0) = \int_{t_0}^{t} dt' [K^{(-)}(t,t') - K^{(+)}(t,t')P(t',t_0)], \tag{2}$$

with the kernels $K^{(\pm)}(t,t')$ given by

$$K^{(+)}(t,t') = \Delta^2 e^{-Q'(t-t')} \cos[Q''(t-t')] \cos[\eta(t,t')],$$
$$K^{(-)}(t,t') = \Delta^2 e^{-Q'(t-t')} \sin[Q''(t-t')] \sin[\eta(t,t')], \tag{3}$$

where $\eta(t,t') = \epsilon_0(t-t') + \hat{\epsilon}/\Omega(\sin\Omega t - \sin\Omega t')$. Here $Q'(t)$ and $Q''(t)$ are the real and imaginary parts of the bath correlation function $Q(t)$, respectively. It corresponds to Eq. (6) [1] of Ref. [9]. The authors of Ref. [9] then attempt to obtain, following Refs. [3,4], an approximation to (2) of convolutive type, being valid for the case of an high-frequency driving field. In doing so, one intrinsically neglects the oscillatory longtime dynamics. In this way the authors of Ref. [9] arrive at their Eq. (13) in Ref. [9]. This Eq. (13), which constitutes the central starting point of the further analysis in Ref. [9], however, is incorrect. Likewise, the same mistake appears in Eq. (37) by Dakhnovskii in Ref. [4]. In fact, a static bias $\hbar\epsilon_0$ does break the spatial inversion symmetry of the dynamics. This leads, for example, to different backward and forward relaxation rates and to an equilibrium (or quasi-equilibrium for the case of fast ac fields) value, being different from zero. In particular, within the NIBA and in the absence of the ac field, the forward (γ^+) and backward (γ^-) relaxation rates obey the detail balance relation $\gamma^+ = e^{\hbar\beta\epsilon_0}\gamma^-$, thus $P(t,t_0)$ will reach at long times the thermal equilibrium value $P_{eq} = \tanh(\hbar\beta\epsilon_0/2)$, cf. Refs. [10,11]. This relation no longer holds in the presence of asymmetry ($\epsilon_0 \neq 0$) and nonzero driving ($\hat{\epsilon}\cos\Omega t$). A discussion of how the detailed balance symmetry is broken in the presence of dc-ac fields is also given in Ref. [8]. This error has been noted by Dakhnovskii and Coalson, which they consequently corrected in later work [5]. Analogous results have been obtained by use of a path integral formulation by Grifoni et al. [6,7], and recently by Goychuck et al. [8] within a polaron approach.

The correct high-frequency master equation is readily obtained from (2) if we observe that the essential dynamics of $P(t,t_0)$ is described by its average value $p_0(t-t_0)$ over a period, of the ac field. It reads [5–8]

$$\dot{p}_0(t-t_0) = \int_{t_0}^{t} dt' [k_0^{(-)}(t-t') - k_0^{(+)}(t-t')p_0(t'-t_0)], \tag{4}$$

with the kernels $k_0^{(\pm)}(t-t')$ representing the average of $K^{(\pm)}(t,t')$ over a period

$$k_0^{(+)}(\tau) = h(\tau) e^{-Q'(\tau)} \cos[Q''(\tau)] \cos(\epsilon_0\tau),$$
$$k_0^{(-)}(\tau) = h(\tau) e^{-Q'(\tau)} \sin[Q''(\tau)] \sin(\epsilon_0\tau), \tag{5}$$

where $h(\tau) = \Delta^2 J_0[(2\hat{\epsilon}/\Omega)\sin(\Omega\tau/2)]$ and $J_0(z)$ is the zero order Bessel function.

[1] Note that Eq. (6) in Ref. [9] contains a mistake: The minus sign in the inhomogeneous term (first line) should read plus.

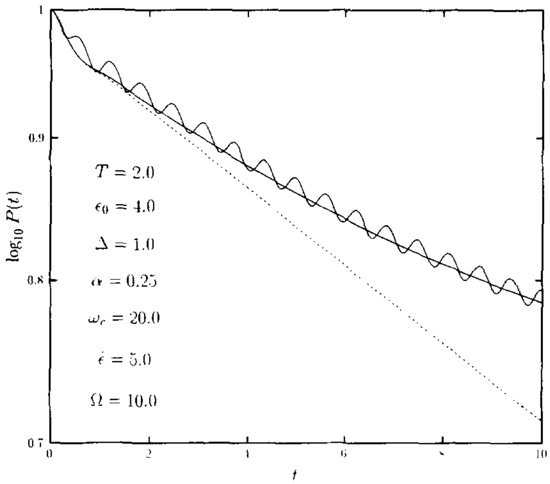

Fig. 1. The logarithm (base 10) of expectation value $P(t) = P(t, t_0 = 0)$ of the exact NIBA equation in (2) (solid oscillatory line) is compared with its high-frequency approximation in (4) (solid line). The incorrect approximation, Eq. (13) of Ref. [9], is also depicted by the dashed line. The parameters are given in the figure.

The numerical solution of the full NIBA equation in (2) is compared with the correct high-frequency approximation in (4) in Fig. 1. At asymptotic long times the oscillatory parts in Fig. 1 survive; they describe the periodic longtime dynamics [6]. We find good agreement between the exact NIBA in (2) and its high-frequency approximation in (4). In contrast, we also compare our results with the high frequency result in (13) of Ref. [9]. It can be obtained from (4) setting the inhomogeneous contribution $k_0^{(-)}(\tau) \equiv 0$ and $k_0^{(+)}(\tau) = h(\tau) \exp[-Q'(\tau)] \cos[Q''(\tau) + \epsilon_0 \tau]$. Apart from a limiting regime at very short initial times, we find distinct differences over the whole regime of small-to-moderate-to-long times. This comparison explicitly demonstrates the shortcomings inherent in Eq. (13) of Ref. [9] where the incorrect result (dashed line) decays exponentially towards the incorrect longtime value $P(t \to \infty) = 0$. The failings of this high-frequency approximation in turn impact also their analysis regarding the behavior of the rate coefficient τ^{-1}, see Eq. (23) in Ref. [9], or the behavior of the transition temperature T^*, see Eq. (34) in Ref. [9].

Moreover, we observe from Fig. 1 that the exact NIBA in (2) and its high-frequency approximation in (4) do not exhibit a decaying behavior that is single exponential like.

In summary, the relaxation of the transient dynamics at high-frequency driving is governed by Eq. (4), which differs from Eq. (13) in Ref. [9] by the nonzero inhomogeneous contribution $k_0^{(-)}(t - t')$ and by a different transition kernel $k_0^{(+)}(t - t')$.

M.G. and P.H. gratefully acknowledge financial support by the Deutsche Forschungsgemeinschaft under contract (Ha1517/14-1).

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