Universal delocalization rate in driven dissipative two-level systems at high temperature

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We study the lifetime of localized states in a two-level system coupled to a dissipative bath and driven by strong time-periodic monochromatic fields. At high temperature, moderate friction and high frequency driving the dynamics is practically exponential characterized by a rate constant. By mapping the driven dissipative two-level system onto a dissipative multilevel curve-crossing problem and applying semiclassical nonadiabatic rate theory we show that strong fields can stabilize localized states over long time intervals and that the delocalization rate exhibits a ''universal'' plateau whose value depends only on the intensity of the driving field. Numerical path integral results confirm our theoretical predictions. [S1063-651X(97)06103-5]

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The competition among tunneling, time-dependent driving and dissipation can lead to very rich dynamical behavior. A central issue concerns the possibility of preserving localization in driven symmetric two-level systems (TLS). In the absence of dissipative mechanisms earlier work has shown [1] that tunneling can be suppressed entirely under certain ''optimal localization'' conditions which amount to degeneracies in the driven TLS quasienergy spectrum. In addition, appropriate laser pulses can lead to localization of an initially delocalized TLS state [2]. Dissipation generally opposes perfect localization via destruction of phase coherence [3,4]. It is, therefore, of interest to examine whether any amount of control can be achieved in the common experimental situation of moderate dissipation strength and temperature, where phase relations no longer prevail.

In this paper we study the evolution of initially (left- or right-) localized states in a symmetric two-level system coupled to a harmonic dissipative bath and driven by a monochromatic field according to the Hamiltonian

$$H(t) = -\hbar\Delta\sigma_x + \sum_j \frac{p_j^2}{2m_j} + \frac{1}{2}m_j\omega_j^2\left(x_j + \frac{c_j\sigma_z}{m_j\omega_j^2}\right)^2 + \sigma_z V_0 \cos \omega_0 t \qquad (1)$$

Here σ_x and σ_z are the 2×2 Pauli spin matrices, $2\hbar\Delta$ is the splitting of the tunneling doublet in the absence of driving and of dissipation, and V_0 is the driving amplitude. The coupling to a large number of harmonic bath degrees of freedom produces a dissipative environment whose influence on the TLS dynamics is encompassed in the spectral density [5]

$$J(\omega) = \frac{\pi}{2}\sum_j \frac{c_j^2}{m_j\omega_j}\delta(\omega-\omega_j). \qquad (2)$$

We refer to the overall strength of the TLS bath coupling as the ''friction'' or ''dissipation'' parameter.

At temperatures that are high with respect to the TLS tunneling splitting and with high driving frequency the evolution of localized states is essentially exponential characterized by a decay constant k. To calculate the decay constant we resort to the quantized representation of the radiation field in which the driven dissipative TLS becomes equivalent to the following time-independent Hamiltonian

$$H_{\mathrm{QM}} = -\hbar\Delta\sigma_x + \sum_j \frac{p_j^2}{2m_j} + \frac{1}{2}m_j\omega_j^2\left(x_j + \frac{c_j\sigma_z}{m_j\omega_j^2}\right)^2 + (a^\dagger a + 1/2)\hbar\omega_0 + C\sigma_z(a+a^\dagger). \qquad (3)$$

Here a and $a^\dagger$ represent field annihilation and creation operators, respectively, and C is a measure of the field-TLS interaction strength. In order for Eq. (3) to be equivalent to Eq. (1) in the semiclassical limit, the coupling constant C must satisfy

$$C = \frac{V_0}{2\sqrt{n+1/2}}, \qquad (4)$$

where n is the quantum number that specifies the photon state. In the semiclassical limit where the effects of the radiation field are equivalent to those of a time-dependent driving term, $n \gg 1$. In the canonical quantization of the radiation field (see, for example, [6]) the ''position'' variable $q = \sqrt{\hbar/2\omega_0}(a+a^\dagger)$ is proportional to the electric field component, while the conjugate ''momentum'' describes the magnetic field.

In the absence of a bath, the electric field energy in Eq. (3) represents two parabolic diabatic curves which are coupled via the constant term $\hbar\Delta$ and which intersect at the origin (see Fig. 1). The remaining terms in Eq. (3) couple this two-curve system to a dissipative harmonic bath. This way the driven dissipative TLS is mapped on a time-independent dissipative multilevel curve-crossing problem and transitions between left- and right-localized TLS states correspond to nonadiabatic surface hopping events. Note that dissipative terms in Eq. (3) involve coupling of the bath to the discrete TLS operator and the field coordinate experiences friction only indirectly.

Within the semiclassical approximation, the nature of the dynamics depends largely on the value of the Landau-Zener adiabaticity parameter [7–9] which for a symmetric system is equal to

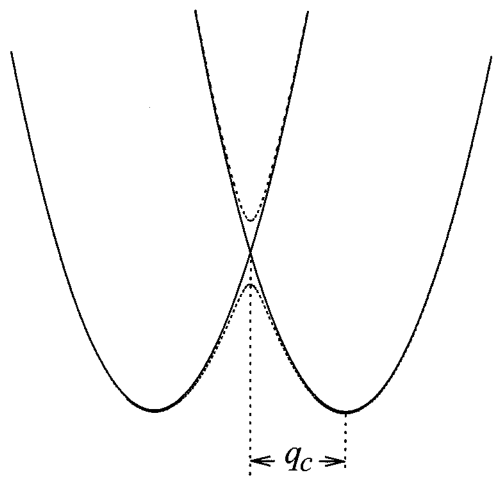

FIG. 1. Electric field energy curves for a TLS coupled to a quantized radiation field. The adiabatic curves are also shown as dotted lines.

$$\delta = \frac{\pi(\hbar\Delta)^2}{\hbar \nu \lambda}. \tag{5}$$

Here $\pm\lambda$ are the slopes of the two electric field curves at the crossing point and ν is the corresponding classical ''velocity,'' which corresponds here to the magnitude of the magnetic field component. If $\delta > 1$ the motion is essentially confined to one of the ''adiabatic'' curves (see Fig. 1). In the opposite limit where $\delta \ll 1$ frequent transitions between curves occur and the dynamics is nonadiabatic.

To evaluate the adiabaticity parameter in the case of the driven TLS characterized by the Hamiltonian of Eq. (3), note that the slopes of the electric field curves at the crossing point are

$$\lambda_{\pm} = \pm\left(\frac{\omega_0}{(2n+1)\hbar} V_0\right)^{1/2}, \tag{6}$$

while the ''velocity'' can be obtained from the energy conservation condition

$$\tfrac{1}{2}\nu^2 + \tfrac{1}{2}\omega_0^2 q_c^2 = (n+1/2)\hbar\omega_0, \tag{7a}$$

where

$$q_c = \frac{\sqrt{2}}{\sqrt{\hbar\omega_0^3}} C \tag{7b}$$

is the distance of the crossing point from the minimum of one of the harmonic curves in Eq. (3) (cf. Fig. 1). Since $n \gg 1$ in the semiclassical limit, the electric field component at the crossing point is small compared to the total field energy and can be dropped, leading to the result

$$\nu = [(2n+1)\hbar\omega_0]^{1/2}. \tag{8}$$

Substitution of Eqs. (6) and (8) into Eq. (5) gives the adiabaticity parameter for a TLS driven by a monochromatic field:

$$\delta = \frac{\pi\hbar\Delta^2}{\omega_0 V_0}. \tag{9}$$

The TLS coupling mixes the diabatic electric field curves over a length determined by the Landau-Zener adiabaticity parameter. Because of this mixing, the motion of the field coordinate is dissipative in the Landau-Zener region where hopping events are most likely to occur. In the limit of large quantum number, the fractional energy loss of the field to the bath is expected to be very small on the time scale of TLS delocalization, such that the field coordinate reaches a time-dependent steady state where the site populations are equal on average. At high temperature and sufficiently strong dissipation phase coherence is destroyed and the crossing events follow Poisson statistics. In that case the dynamics should be exponential characterized by a rate constant. Since $n \gg 1$ a semiclassical surface hopping treatment is justified for the crossing rate.

We therefore employ qualitative semiclassical ideas developed in the context of nonadiabatic rate theory [10] to treat the TLS dynamics in the quantized field representation for $\omega_0 V_0 \gg \hbar\Delta^2$. Earlier work has shown [10,11] that the curve crossing rate exhibits at high temperatures a plateau spanning a wide range of friction. This behavior is the result of cancellation [10] between the Kramers adiabatic factor [12] that tends to diminish the rate at sufficiently strong dissipation and the Landau-Zener probability that enhances transitions between diabatic potential curves. Since the field energy is in the classical limit much larger than the energy at the curve crossing point, the activation factor is unity. Within the quasiclassical surface hopping model, the forward curve-crossing rate is then given by the frequency of passing through the curve crossing region times the Landau-Zener hopping probability

$$k_{\mathrm{SC}}^{+} = 2\left(\frac{\omega_0}{2\pi}\right) P_{\mathrm{LZ}}, \tag{10a}$$

where

$$P_{\mathrm{LZ}} = 1 - e^{-\delta} \tag{10b}$$

is the Landau-Zener factor. Substitution of the adiabaticity parameter in Eq. (10) leads to the following result for the forward curve-crossing rate:

$$k_{\mathrm{SC}}^{+} = \frac{\omega_0}{\pi}\left[1 - \exp\left(-\frac{\pi(\hbar\Delta)^2}{\hbar\omega_0 V_0}\right)\right]. \tag{11}$$

Finally, the overall decay constant that characterizes TLS delocalization is

$$k_{\mathrm{SC}} = 2k_{\mathrm{SC}}^{+} = 2\frac{\omega_0}{\pi}\left[1 - \exp\left(-\frac{\pi(\hbar\Delta)^2}{\hbar\omega_0 V_0}\right)\right] \approx \frac{2\hbar\Delta^2}{V_0}. \tag{12}$$

This result is independent of the parameters of the environment and of the frequency of the driving field.

Therefore, by recognizing the analogy between delocalization in a driven dissipative TLS and nonadiabatic rate theory we have established the existence of a plateau in the decay constant of a localized state at high temperature. Comparison of Eq. (12) with the result of the noninteracting blip approximation [5] for the force-free TLS coupled to an ohmic bath shows that the decay of localized states can be *delayed* substantially via proper driving at moderate values of the dissipation parameter. The decay constant in this regime is *universal*, as it depends only on the intensity of the driving field. On the other hand, the portion of parameter space exhibiting a rate plateau depends on the value of the adiabaticity parameter, increasing with the intensity of the field. We note that increase of the field frequency does not necessarily favor the rate plateau, as the effects of driving average out to zero when the field and tunneling frequencies are adiabatically separated.

Dakhnovskii [13], as well as Grifoni *et al.* [14], have extended the noninteracting blip approximation (NIBA) [5] to treat the TLS dynamics in the presence of time-dependent driving. These works resulted in kinetic equations for the decay of localized TLS states subject to high-frequency driving. Although the full NIBA result obtained in Ref. [13] generally predicts qualitatively correct behaviors with regard to the driven TLS dynamics, Dakhnovskii's prediction of field-induced increase in the TLS decay rate is in disagreement with the present findings at weak or moderate friction. This discrepancy arises primarily from breakdown of the approximations employed in Refs. [13] and [16] to simplify the NIBA result. Other recent analysis of the NIBA result [17] predicted a decrease of the TLS delocalization rate due to driving, in harmony with our results at weak or moderate dissipation. None of these earlier works revealed the existence of a medium- and frequency-independent rate regime.

At very weak or very strong dissipation the driven TLS decay constant is expected to deviate from the result of Eq. (12). At small values of the friction phase coherence between resonant energy levels on the diabatic potentials should generally lead to increase of the decay rate above its plateau value [18]. Deviations from this trend may occur if an additional symmetry exists that prevents the accumulation of phase which leads to rate enhancement. For example, if the field parameters correspond to a degeneracy of the left- and right-displaced photon states [19,20], transfer between diabatic potentials is possible only via coupling to the bath and the decay constant should decrease as the friction parameter approaches zero. Such effects are intimately connected to degeneracies in the driven TLS quasienergy spectrum that are known to cause in the absence of dissipation exact localization at times which are multiples of the Floquet period [1]. Such effects will be examined in more detail in another publication [21].

To confirm the above theoretical analysis we have performed accurate numerical calculations on the decay of initially localized states in a symmetric TLS coupled to generic dissipative environments. The calculations employ an iterative path integral scheme developed earlier in our group [22,23] (for a review see [24]) generalized to time-dependent potentials [4]. We choose a driving field of amplitude $V_0=30\hbar\Delta$ and present results for the generic driving frequency $\omega_0=15\Delta$ and for the optimal localization condition $\omega_0=10.87\Delta$. The second set of field parameters leads to suppression of tunneling for the dissipationless TLS. All calculations presented below are carried out at the temperature $\beta^{-1}=10\hbar\Delta$.

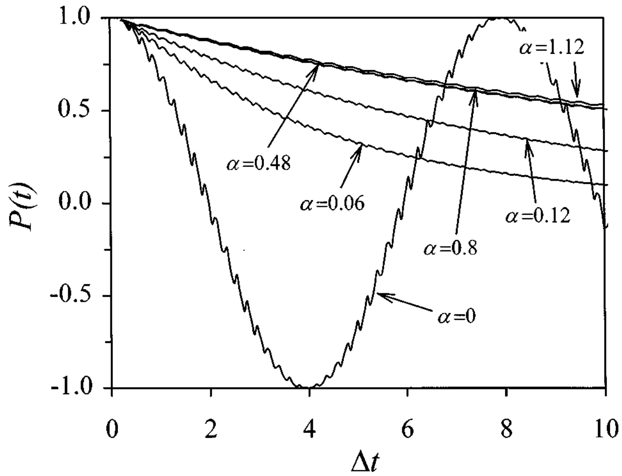

FIG. 2. The expectation value $\langle\sigma_z(t)\rangle$ as a function of time with the generic driving field $V_0=30\hbar\Delta$, $\omega_0=15\Delta$ at $\hbar\beta\Delta=0.1$ and for various values of the Kondo parameter.

First we present path integral results for the delocalization rate in the case of an ohmic bath with exponential cutoff [5]. The spectral density has the form

$$J(\omega)=\frac{\pi}{2}\hbar\alpha\ \omega\ e^{-\omega/\omega_c}, \tag{13}$$

where α is the dimensionless Kondo parameter that characterizes the friction strength. The TLS is initially in the $+1$ (right-localized) state. We choose $\omega_c=20\Delta$.

Figure 2 shows the evolution of the average TLS position $P(t)\equiv\langle\sigma_z(t)\rangle=\mathrm{Tr}[\tilde{\rho}(t)\sigma_z]$ (where $\tilde{\rho}(t)$ represents the TLS reduced density matrix) for generic field driving at several values of the Kondo parameter. While the dissipationless driven TLS exhibits coherent oscillations, the localized state decays practically exponentially even at fairly small values of the Kondo parameter. It is thus clear that the concept of a rate is appropriate in this regime.

The dissipative TLS decay constant at the same temperature is plotted in Fig. 3 as a function of the Kondo parameter in the cases of a generic driving, under optimal localization conditions as well as in the absence of driving. It is seen that the lifetime of a localized state is significantly extended in both driven systems. With both driving fields the TLS decay constant displays a broad plateau where the computed decay rate is in excellent agreement with the prediction of Eq. (12). The qualitatively distinct effects of the above fields on the tunneling dynamics of the bare TLS are responsible for the different trends observed in Fig. 3 at weak friction. These trends are in qualitative agreement with the arguments presented above.

Similar behavior is observed in Fig. 4, which shows the TLS decay rate for a superohmic bath characterized by the spectral density

$$J(\omega)=\frac{\pi}{2}\hbar\alpha\ \omega\left(\frac{\omega}{\Delta}\right)e^{-\omega/\omega_c}. \tag{14}$$

We choose $\omega_c=3\Delta$. The maximum of this spectral density occurs at $\omega=2\omega_c$ and therefore lies well above the bare TLS tunneling frequency. Results are shown for the generic driv-

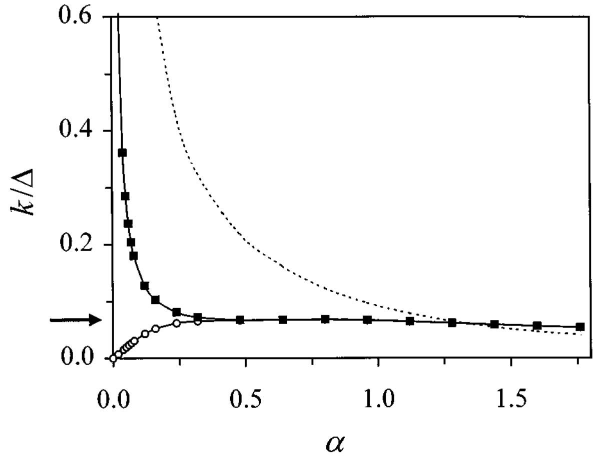

FIG. 3. The decay rate of an initially localized state as a function of the Kondo parameter α for an ohmic bath [cf. Eq. (13)] at $\hbar\beta\Delta = 0.1$. Dotted line: force-free TLS. Solid squares: TLS driven by a generic driving field with $V_0 = 30\hbar\Delta$, $\omega_0 = 15\Delta$. Hollow circles: TLS driven by a localizing field with $V_0 = 30\hbar\Delta$, $\omega_0 = 10.87\Delta$. The arrow indicates the theoretical result of Eq. (12).

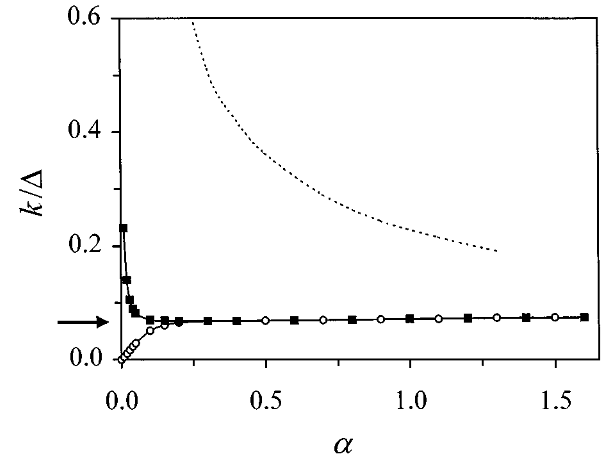

FIG. 4. The decay rate of an initially localized state as a function of the Kondo parameter α for a superohmic bath [cf. Eq. (14)] at $\hbar\Delta\beta = 0.1$. Dotted line: force-free TLS. Solid squares: TLS driven by a generic driving field with $V_0 = 30\hbar\Delta$, $\omega_0 = 55\Delta$. Hollow circles: TLS driven by a localizing field with $V_0 = 30\hbar\Delta$, $\omega_0 = 6.933\Delta$. The arrow indicates the theoretical result of Eq. (12).

ing frequency $\omega_0 = 5.5\Delta$ and for the value $\omega_0 = 6.933\Delta$ which leads to localization of the dissipationless system. It is seen that the driven TLS exhibits incoherent relaxation characterized by a rate constant which again depends only on the overall field amplitude over a wide range of dissipation strength, in line with the theoretical analysis presented above.

From a practical point of view, the existence of a ‘‘universal’’ small delocalization rate at high temperature implies that significant control of tunneling dynamics can be achieved via irradiation with strong monochromatic fields under diverse conditions. Although simple arguments about phase coherence may lead to the conclusion that localization should be destroyed at high temperature and/or large friction, the theoretical analysis presented in this paper shows a different trend: while weak friction opposes perfect localization, further increase of the dissipation parameter leads to a plateau where the decay rate can be small, yet independent of most parameters and thus achievable without fine tuning of experimental conditions. This robust phenomenon may find a novel application to the control of charge oscillations in semiconductor double quantum well nanostructures.

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