Stabilization of localized states in dissipative tunneling systems interacting with monochromatic fields

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We study the dynamics of an initially localized symmetric two-level system coupled to high-temperature dissipative environments and driven by a strong time-periodic force which corresponds to high-frequency monochromatic light. Qualitative arguments based on the quantized representation of the radiation field predict a wealth of intriguing behaviors which are confirmed and quantified via accurate numerical path integral calculations. With the exception of very strong friction we find that high-frequency driving always helps stabilize localized states. At intermediate friction the delocalization rate approaches a "universal" limiting value which is largely independent of the parameters of the environment and of the specifics of the driving force, depending only on its overall strength. This robust behavior implies that localized states can be stabilized in these systems without much finetuning of external conditions. In the weak friction regime the interplay between phase interference and dissipation results in nonmonotonic variation of the decay rate with friction and driving frequency. The path integral results are compared to those obtained earlier via analytical treatments. © 1997 American Institute of Physics.

I. INTRODUCTION

The interaction of electromagnetic radiation with tunneling systems can lead to a variety of intriguing patterns and has been a subject of numerous studies. For simplicity, often only the lowest tunneling doublet is considered explicitly, giving rise to a two-level system (TLS) which (within the classical treatment of the radiation field) is driven by a time-dependent force. Coupling of the driven TLS to a dissipative environment results in even more complex dynamical effects whose exploration has begun only recently. Because the dissipative TLS provides an adequate model of such diverse phenomena as electron transfer reactions, impurity tunneling in crystals, and charge oscillations in semiconductor double quantum well structures, its understanding is vital in several areas of chemistry and physics; the addition of coherent radiation introduces new parameters, offering the intriguing possibility of manipulating such processes in a desirable manner. Aside from theoretical and perhaps technological interest in exploring the different behaviors that can arise from the interplay among tunneling, coherent driving and dissipation, the driven two-level system in contact with a heat bath constitutes the simplest "control" problem whose study may shed light onto the possibility of sustaining large-amplitude coherent oscillations in polyatomic systems.

Most previous work on driven two-level systems has been concerned with the issue of localization. In the absence of dissipation Grossman et al. have demonstrated that addition of a simple oscillatory driving term can under certain conditions quench tunneling of a symmetric TLS, maintaining indefinitely spatial localization of a left- (or right-) localized TLS state. Bavli and Metiu have shown that appropriate laser pulses can be used to localize a TLS which has been prepared in a delocalized state. In addition, several previous studies have found that coupling to a heat bath results in destruction of localization at a rate that can be small compared to that for thermalization of the free dissipative TLS; in general terms, survival of a localized state is aided by strong fields, low temperature and weak dissipation. Other recent work has suggested that external driving can alter dramatically the localized state lifetime and even increase the decay rate.

In the limit where the field frequency exceeds considerably the tunneling splitting of the bare TLS and at high temperature, localized states decay essentially exponentially even with fairly weak friction. Recent work by Makri and Wei has shown that the decay rate of an initially localized state is largely insensitive to the characteristics of the environment over a wide range of friction. In this regime, the lifetime of localized states can be extended significantly with the use of strong monochromatic fields. This fact implies that the prospect of decreasing the tunneling rate in such systems is not as unfavorable as it may appear from extrapolation of the low-temperature, weak friction data.

Another interesting question that has been addressed is related to the possibility of sustaining large-amplitude coherent tunneling oscillations in driven two-level systems, thus preventing approach of equilibrium. Makarov and Makri have demonstrated that such effects are indeed achievable with weak continuous-wave radiation nearly resonant with the tunneling doublet by exploiting the phenomenon of quantum stochastic resonance and should be readily observable in double-quantum-well structures. Evans et al. have also reported a similar effect in models of electron transfer induced by strong alternating fields.

In the present paper we explore further the dynamics of localized states in a symmetric two-level system coupled to a high-temperature harmonic dissipative bath and driven by a
Here, \(\sigma_s\) and \(\sigma_z\) are the 2×2 Pauli spin matrices, \(2\hbar\Omega\) is the splitting of the bare TLS tunneling doublet, \(\{x_j\}\) are the coordinates of the harmonic bath degrees of freedom responsible for dissipation, and \(V(t)\) is the driving field applied to the TLS which represents in the semiclassical limit its interaction with radiation. The high temperature regime is most relevant to practical applications: since the energy scale is set primarily by the splitting of the tunneling doublet, the TLS may be in the “high temperature” regime even though the absolute temperature is actually rather low.

Our approach combines simple theoretical arguments based on a time-independent model that employs the quantized electromagnetic field, as well as accurate numerical path integral calculations. Convergence of the path integral for the long times required by the nature of the problem at hand are possible with an iterative procedure developed earlier in our group. This scheme is based on Feynman’s path integral representation of quantum mechanics and exploits the finite length of memory interactions that characterize macroscopic environments. By ignoring negligible long-range interactions in the dissipative influence functional, the multidimensional discretized path integral is converted into a series of lower-dimensional operations which employ a propagator tensor. The tensor multiplication scheme, which constitutes the generalization of conventional matrix multiplication algorithms to dissipative systems, is stable and accurate over long propagation periods and thus ideally suited to the study of laser-induced dynamics of crystalline solids.

Section II summarizes the iterative tensor multiplication scheme. Section III discusses the dynamics of localized TLS states at high temperatures in terms of theoretical arguments as well as accurate numerical calculations. The starting point of our analysis is the quantized electromagnetic field. This procedure maps the driven TLS onto a time-independent curve crossing problem whose behavior can be understood by a combination of semiclassical surface hopping ideas and phase interference considerations. Numerical results illustrate these ideas and provide a quantitative picture. The predictions of the quantized radiation model are found to be in excellent agreement with the numerical path integral results. Section IV compares our findings to those obtained via earlier analytical treatments. Finally, Sec. V summarizes the present results and presents concluding remarks.

II. ITERATIVE EVALUATION OF THE DISSIPATIVE PATH INTEGRAL

Coupling of the driven TLS to a large number of harmonic bath degrees of freedom in Eq. (1.1) produces a dissipative environment whose influence on the TLS dynamics is specified by the spectral density function:

\[
J(\omega) = \frac{\pi}{2} \sum_j \frac{c_j^2}{m_j\omega_j} \delta(\omega - \omega_j). \tag{2.1}
\]

Time evolution can be expressed in terms of the propagator \(U(t',t)\) for the time-dependent Hamiltonian of Eq. (2.1). Since it is not interested in recording the detailed dynamics of each bath degree of freedom but only the overall influence of the environment on the TLS state, we integrate out the bath and focus on the evolution of the reduced density matrix:

\[
\tilde{\rho}(t) = \text{Tr}_b[U(t,0)\rho(0)U^{-1}(t,0)], \tag{2.2}
\]

where \(\text{Tr}_b\) denotes the trace with respect to the harmonic bath.

In order to obtain propagators accurate over fairly large time increments it is useful to identify a meaningful low-dimensional reference system. In the present case we choose a time dependent reference that includes the TLS Hamiltonian along with the driving term:

\[
H_0(t) = -\hbar\Omega\sigma_s + V(t)\sigma_z. \tag{2.3}
\]

The remaining terms in the Hamiltonian of Eq. (1.1) correspond to harmonic oscillators that are displaced by an amount which depends on the TLS site. Using a symmetric splitting of the time evolution operator and assuming that the system and bath are uncoupled at \(t=0\) such that the initial density matrix can be written as a product of system and bath density matrices,

\[
\rho(0) = \tilde{\rho}(0)\rho_b(0) \tag{2.4}
\]

the reduced density matrix can be expressed in the following discretized path integral form:

\[
\langle \sigma^\dagger \tilde{\rho}(t) \sigma' \rangle = \sum_{\sigma_0^0=\pm 1} \sum_{\sigma^1_1=\pm 1} \cdots \sum_{\sigma^{N-1}_N=\pm 1} \sum_{\sigma^0_N=\pm 1} \sum_{\sigma^1_N=\pm 1} \sum_{\sigma^N_N=\pm 1} \langle \sigma^\dagger U_0(t,t-\Delta t) \sigma^+ \rangle \cdots \times \langle \sigma^\dagger U_0(\Delta t,0) \sigma^0 \rangle \langle \sigma^\dagger \rho_b(0) \sigma^0 \rangle \langle \sigma^\dagger U_0^{-1}(\Delta t,0) \sigma^+ \rangle \cdots \times \langle \sigma^-_{N-1} U_0^{-1}(t,t-\Delta t) \sigma' \rangle F(\sigma^0, \sigma^1, ..., \sigma^N, \sigma^-_{N-1}, \sigma^+_{N-1}, \sigma^+_{N-1}, \sigma^-_{N-1}, \sigma^+, \sigma^-; \Delta t). \tag{2.5}
\]
The possibility of iterative evaluation of the path integral expression, Eq. (2.5), arises from the fact that the bath response function characterizing dissipative environments drops off rapidly with time. This feature is a consequence of dephasing in media with broad spectra and implies that “long memory” terms make negligible contribution to the dynamics. Retaining terms with \(|k-k'| \leq \Delta k_{\text{max}}\), where \(\Delta k_{\text{max}}\Delta t\) is the effective memory length, leads to quasi-Markovian dynamics for a higher dimensional quantity and an iterative scheme. Specifically, an augmented reduced density tensor \(\mathbf{R}\) of rank \(\Delta k_{\text{max}}\) can be constructed whose evolution is given by the iterative procedure \(^{22}\)

\[
R(\sigma_{k+1}, \ldots, \sigma_{k+\Delta k_{\text{max}}}; (k+1)\Delta t) = \sum_{\sigma_k^* = \pm 1} \Lambda(\sigma_k^*, \ldots, \sigma_{k+\Delta k_{\text{max}}}; k\Delta t) \times R(\sigma_k^*, \ldots, \sigma_{k+\Delta k_{\text{max}}}; k\Delta t),
\]

where \(\Lambda\) is an appropriate time-dependent propagator tensor. The path integral interactions included in a single iteration of this scheme are indicated in the diagram of Fig. 1. Projection of \(\mathbf{R}\) yields the reduced density matrix \(\hat{\rho}\) of the TLS.

\[
\hat{\rho}(\sigma_k^*; k\Delta t) = \langle \sigma_k^* | \hat{\rho}(k\Delta t) | \sigma_k^- \rangle = R(\sigma_k^*, \sigma_{k+1}^*; \ldots; 0; k\Delta t).
\]

The above procedure can be iterated to yield the reduced density matrix over long propagation intervals. Stability is guaranteed, as the tensor multiplication scheme preserves the trace of the reduced density matrix.\(^{21}\)

The iterative procedure outlined above avoids global evaluation of the multidimensional discretized path integral, which is responsible for the failure of conventional Monte Carlo path integral approaches to yield converged results at times longer than two or three periods of motion.\(^{28,29}\) It is easily generalizable to systems described by continuous potentials via the use of discrete variable representations.\(^{30}\)

For a TLS, propagation of the augmented reduced density tensor requires storage of \(2^{2(\Delta k_{\text{max}}+1)}\) complex numbers. This is an easy task for \(\Delta k_{\text{max}}\) up to about 10. The above version of the tensor multiplication scheme is not practical if the medium-induced memory spans many more time steps. In addition, storage considerations may become critical even in short memory processes if the system of interest has more than two sites. To address such situations, Sim and Makri have recently proposed use of a Monte Carlo procedure that filters out those spin sequences whose weight in the path integral is lower than a desired threshold.\(^{31,32}\) This procedure usually results in dramatic reduction of the required storage and makes calculations feasible in cases of long-time nonlocality. In the present work, though, we restrict consideration to a model spectral density with relatively short memory and do not need to resort to filtering techniques.

### III. DRIVEN TLS DYNAMICS IN HIGH-TEMPERATURE ENVIRONMENTS

In this section we discuss the dynamics of a driven symmetric TLS coupled to harmonic dissipative baths. In the absence of coupling to a bath, the most pronounced effect of driving is the suppression of tunneling along a one-dimensional manifold in the \(V_0, \omega_0\) parameter space\(^6\) to which we refer as “optimal localization condition.” This phenomenon is related to a degeneracy in the driven TLS quasienergy spectrum generally when \(2V^2/\hbar\omega_0 = z_j\), where \(z_j\) are nodes of the zero-order Bessel function and leads to rigorous survival of localized states at times which are multiples of the field period. Interaction with a dissipative bath offers the possibility of delocalization via destruction of phase coherence which generally leads to a time-dependent steady state whose time-average corresponds (for a symmetric TLS) to Boltzmann equilibrium.

The effects of weak dissipation on the driven TLS dynamics have been explored earlier by means of master equation approaches\(^7,8\) and also via path integral calculations in our group.\(^9\) These studies concluded that dissipation eventually destroys localization. However, when the parameters of the driving term do not satisfy precisely the above condition for suppression of tunneling, the lifetime of an initially localized state can be increased by weakly coupling the TLS to a dissipative environment.

In a recent article\(^{15}\) Makri and Wei argued that the lifetime of localized states can be extended very significantly even at high temperature and moderate friction. When the
dissipation parameter is sufficiently large to suppress phase interference the rate of delocalization was found to depend only on the overall field strength. Here, we present a more detailed account of the theory along with additional numerical calculations that illustrate this phenomenon and its deviations from the "universal rate" regime. In the next section we compare our results to those of earlier analytical work.

Below we explore the evolution of initially localized states toward thermal equilibrium under conditions that lead to incoherent dynamics. In agreement with earlier work, we find that the decay of a localized state in a driven dissipative TLS is practically exponential (with superposed small amplitude oscillations) at intermediate to high temperature and/or with sufficiently strong friction, at least as long as the driving frequency is sufficiently high with respect to the tunneling splitting. Under such conditions we calculate the rate of delocalization and its inverse, the mean lifetime of a localized state, over a wide range of temperature and system–bath coupling strength.

The results that we present employ harmonic dissipative environments characterized by three different spectral densities. The first is an Ohmic spectral density with exponential cutoff

\[ J_1(\omega) = m_0 \gamma \omega e^{-\omega/\omega_c}. \]  

(3.1a)

Here, \( m_0 \) is the effective mass of the tunneling system, \( \gamma \) is the friction coefficient, and \( \omega_c \) is a cutoff which ensures that the phonon spectrum of the environment decays to zero at high frequencies. We choose \( \omega_c = 20 \Omega \); this magnitude of the cutoff frequency is typical of semiconductor double quantum well structures where \( \Omega \sim 10–50 \text{ cm}^{-1} \) while the crystal Debye frequency is a few hundred \text{ cm}^{-1}. The second bath model involves a superohmic spectral density of the form

\[ J_2(\omega) = m_0 \gamma \omega \frac{\omega}{\Omega} e^{-\omega/\omega_c}. \]  

(3.1b)

The quadratic variation of this function at small frequencies is characteristic of acoustic phonons in two-dimensional Debye solids. We choose the cutoff frequency such that the maximum of this spectral density occurs at \( 2\omega_c = 6 \Omega \) and therefore lies well above the bare TLS tunneling frequency. Finally, we choose as a third model a bath characterized by Drude friction, for which the spectral density takes the form

\[ J_3(\omega) = \frac{m_0 \gamma \omega}{1 + \lambda^2 \gamma^2 \omega^2}. \]  

(3.1c)

with \( \lambda = 1.3 m_0 \gamma / \Omega \). In all cases, the strength of the dissipation is commonly characterized by the dimensionless Kondo parameter \( \alpha = 2 m_0 \gamma / \pi \hbar \).

The TLS is initially in the \( +1 \) (right-localized) state. The driving term

\[ V(t) = V_0 \cos \omega_0 t \]  

(3.2)

represents the classical limit of a monochromatic continuous-wave electromagnetic field. The amplitude of the driving term is chosen in the range \( V_0 \approx 30 \hbar \Omega \) which (for typical values of the system dipole moment) corresponds to laser fields of intermediate to large strength.

A. Rate plateau at intermediate friction

In this subsection we focus on the intermediate friction regime. We begin with the choice \( V_0 = 20 \hbar \Omega \) and present results at two driving frequencies, \( \omega_0 = 12 \hbar \Omega \) and \( \omega_0 = 16.714 \hbar \Omega \). At the first of the above driving frequencies the bare (\( \alpha = 0 \)) TLS exhibits coherent tunneling oscillations, while the choice \( \omega_0 = 16.714 \hbar \Omega \) constitutes an optimal localization condition.

First we present numerical path integral results at temperature such that \( k_B T = \beta^{-1} = 10 \hbar \Omega \). This temperature is high with respect to the TLS tunneling splitting but satisfies \( \hbar \omega_0 \beta^{-1} \). In this regime (and for \( \alpha < 1 \) but not too close to zero) the force-free TLS in contact with an ohmic bath undergoes incoherent relaxation with a rate which within the non-interacting blip approximation is given by the expression

\[ k_{\text{NIBA}} = 2 \sqrt{\frac{\pi}{\Gamma(\alpha + 1/2)}} \frac{\Omega^2}{\hbar \beta \omega_c} \left( \frac{\pi}{\hbar \beta \omega_c} \right)^{2\alpha - 1}. \]  

(3.3)

Figure 2 displays the evolution of the average TLS position \( \langle \sigma(t) \rangle = \text{Tr} [ \hat{\rho}(t) \sigma_z ] \) for the Ohmic TLS at \( \hbar \Omega \beta^{-1} = 0.1 \) in the absence of driving and with the two fields specified above at friction values characterized by \( \alpha = 0 \) (no dissipation), \( \alpha = 0.16 \) and \( \alpha = 0.64 \). With finite friction it is seen that the localized state in the periodically driven TLS displays essentially incoherent dynamics with a small-amplitude oscillatory component superposed on the exponential decay. In addition, the lifetime of a localized state is extended when the TLS is driven by a sinusoidal field. However, although the decay rates differ significantly when different driving frequencies are used in the case of \( \alpha = 0.16 \), indistinguishable dynamics result at a higher value of the dissipation parameter. At first glance this observation is surprising.

Below we present a qualitative account of the behaviors observed in driven dissipative tunneling systems. To this end we resort to the quantized representation of the radiation field in which the Hamiltonian of Eq. (1.1) is time-independent

\[ H = -\hbar \omega_0 \sigma_z + \sum_j \left( \frac{p_j^2}{2 m_j} + \frac{1}{2} m_j \omega_j^2 \right) \left( x_j + \frac{c_j \sigma_z}{m_j \omega_j} \right)^2 + (a^\dagger a + \frac{1}{2}) \hbar \omega_0 + C \sigma_z (a + a^\dagger). \]  

(3.4)

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. (3.4) to be equivalent to Eq. (1.1) in the semiclassical limit, the coupling constant \( C \) must satisfy

\[ C = \frac{V_0}{2 \sqrt{n + \frac{1}{2}}}. \]  

(3.5)

where \( n \) is a quantum number that specifies the number of photons. 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 absence of a bath, Eq. (3.4) represents two parabolic "adiabatic" potential surfaces which are coupled via the
constant term $\hbar \Omega$ and which intersect at the origin (see Fig. 3). The remaining terms in Eq. (3.4) couple this two-surface system to a dissipative harmonic environment. This way the driven dissipative TLS is mapped on a time-independent dissipative multilevel curve-crossing problem, although the system–bath coupling differs from that employed in conventional system–bath models. As will become clear below, the details of the system–bath interaction are generally unimportant except at weak friction where dissipative processes are slow and mode specificity prevails.

By virtue of the quantized field analog, transitions between left and right states of the driven TLS correspond to hopping between diabatic curves in the time-independent quantized field Hamiltonian. Under conditions that lead to exponential decay, the situation is similar to that in electron transfer reactions with the field position $q$ playing the role of a ‘‘reaction coordinate.’’ However, there is an important difference between canonical reactions and the problem at hand: In order to give rise to constant amplitude driving, the field oscillator should maintain essentially constant energy. This is indeed the case if the field oscillator is in a state of very large quantum number, a necessary condition for validity of the quantum-classical correspondence. Thus the ‘‘reaction coordinate’’ does not reach thermal equilibrium in this case. Instead, a steady state is reached at long time, in which the populations of the two diabatic curves become equal on average and the average TLS position is zero.

With the above peculiarities of the present problem in mind, we proceed with a reasoning similar to that of Frauenfelder and Wolynes in their qualitative semiclassical discussion of nonadiabatic rate theory. The effects of friction on the classical reaction rate are largely determined by the average number of trajectory recrossings of the transition state region. Although the field coordinate is not directly coupled to the bath in the present case, the two-curve system as a whole experiences friction during curve crossing. Mixing of the two potential surfaces dilutes this friction, spreading it over the entire ‘‘Landau–Zener’’ region around the crossing point. We conclude that the reaction coordinate experiences friction throughout the range over which curve-crossing events are likely.

The Landau–Zener region in which the diabatic curves interact strongly extends roughly over the coordinate range $|q|<q_{LZ}$ in which the energy difference between diabatic potentials does not exceed twice the coupling strength $\hbar \Omega$. Using the coupling relation between TLS and field, a straightforward calculation gives

$$q_{LZ} = \sqrt{\frac{2 \hbar \Omega}{\omega_0 V_0}} \sqrt{2n+1},$$

where the mass of the field oscillator has been set equal to unity. Division of the Landau–Zener length $2q_{LZ}$ by the average classical velocity gives the average time $\tau_{LZ}$ spent in the vicinity of the crossing point during each half cycle of
vibration. One can estimate the average velocity from its value \( \nu_c \) at the crossing point from the energy conservation condition

\[
\frac{1}{2} \dot{\nu}_c^2 + \frac{1}{2} \dot{\omega}_0^2 q_{\text{min}}^2 = (n + \frac{1}{2}) \hbar \omega_0
\]

(3.7a)

where

\[
q_{\text{min}} = \frac{\hbar V_0}{\sqrt{2} \sqrt{2n + 1} \hbar \omega_0}
\]

(3.7b)

is the distance of the crossing point from a potential minimum. Since \( n \gg 1 \) in the semiclassical limit, the potential energy term is small at the crossing point and can be dropped, leading to the following result for the classical velocity:

\[
\nu_c = [(2n + 1) \hbar \omega_0]^{1/2}
\]

(3.8)

Combining the last equation with Eq. (3.6) leads to the following result for the average time spent in the Landau–Zener region

\[
\tau_{\text{LZ}} = \frac{4 \Omega}{V_0 \omega_0}
\]

(3.9)

Within the semiclassical approximation and in the absence of coupling to a heat bath, the nature of the dynamics depends on the value of the Landau–Zener adiabaticity parameter,

\[
\delta = \frac{2 \pi (\hbar \Omega)^2}{\hbar \nu_c |\Delta \lambda|}
\]

(3.10)

where \( \Delta \lambda \) is the difference in the slopes of the two diabatic curves at the crossing point. Use of Eq. (3.8) gives the adiabaticity parameter for a TLS driven by a monochromatic field

\[
\delta = \frac{\pi \hbar \Omega^2}{\omega_0 V_0}
\]

(3.11)

From this, \( \delta \ll 1 \) if the time spent in the Landau–Zener region, Eq. (3.9), is short compared to the tunneling period. In that case the dynamics is nonadiabatic. In the opposite limit where \( \delta \gg 1 \) the motion is confined to the adiabatic potentials (see Fig. 3).

Since the motion is dissipative over the entire Landau–Zener region where population transfer takes place, the results of reaction rate theory are applicable. When the adiabaticity parameter is small, earlier theoretical work has predicted the existence at high temperature of a “golden rule plateau” where the rate is independent of dissipation over a wide range of friction strength. The rate plateau appears when the friction is sufficiently large to destroy phase memory. Eventually, when the mean collision-free time becomes smaller than the time spent in the Landau–Zener region, \( \nu_c \ll \tau_{\text{LZ}} \), the rate decreases monotonically with further increase of the dissipation parameter. The nonadiabatic rate plateau has been confirmed by stochastic surface hopping and path integral calculations.

In the present case where \( n \gg 1 \) availability of the “activation energy”

\[
V_c = \frac{V_0^2}{4(n + \frac{1}{2}) \hbar \omega_0}
\]

is guaranteed and the forward curve-crossing rate is given semiclassically by the frequency of passing through the curve crossing point times the Landau–Zener hopping probability \( P_{\text{LZ}} \)

\[
k_{\text{SC}} = 2 \left( \frac{\omega_0}{\pi} \right) P_{\text{LZ}}
\]

(3.12)

where

\[
P_{\text{LZ}} = 1 - e^{-\delta}
\]

(3.13)

is the Landau–Zener hopping probability. Substitution of Eq. (3.11) in the Landau–Zener probability leads to the following result for the forward crossing rate:

\[
k_{\text{SC}} = \frac{\omega_0}{\pi} \left[ 1 - \exp \left( - \frac{\pi (\hbar \Omega)^2}{\hbar \omega_0 V_0} \right) \right]
\]

(3.14)

Finally, the overall decay rate of a localized state in the plateau regime is

\[
k_{\text{SC}} = 2k_{\text{SC}} = 2 \left( \frac{\omega_0}{\pi} \right) \left[ 1 - \exp \left( - \frac{\pi (\hbar \Omega)^2}{\hbar \omega_0 V_0} \right) \right] = \frac{2 \hbar \Omega^2}{V_0}
\]

(3.15)

This result is valid if the adiabaticity parameter, Eq. (3.11), is sufficiently small for the Landau–Zener factor to be accurate. In addition, validity of the arguments presented above rests on the assumption that the TLS-field coupling should be sufficiently strong. For small field amplitudes \( V_0 \), the effect of driving should disappear; similarly, if \( \omega_0 \gg \Omega \) the driving force averages out to zero and is not felt by the TLS. In both cases the driven dissipative TLS reverts to the standard spin-boson problem. According to Eq. (3.7b), the weak field and high driving frequency limits are associated within the quantized field picture with small separation of the diabatic curves, therefore weak interaction between TLS and field. Finally, at constant field amplitude we expect the rate plateau to shrink if the bath spectral density is extended to very high frequencies and/or the temperature is lowered, as the Kramers turnover rate curve of the corresponding adiabatic problem will then shift toward smaller friction values.

Therefore, use of the quantized field representation predicts the existence of a “universal” delocalization rate for the driven dissipative TLS which spans a wide range of friction. With large field amplitudes this limiting rate is generally small compared to its value in the absence of driving. This fact is easily seen in the case of an Ohmic bath by comparing Eq. (3.15) to the result of the noninteracting blip approximation. For example, for \( \alpha = 0.5 \) (a typical friction strength in the plateau regime) Eq. (3.3) simplifies to \( k_{\text{NIBA}} = 2 \pi \Omega^2 / \omega_c \) from which it follows that \( k_{\text{SC}} < k_{\text{NIBA}} \) if \( V_0 \gg \hbar \omega_c / \pi \).

The above predictions of the quantized field model are confirmed by accurate path integral calculations displayed below. Figure 4(a) shows the driven Ohmic TLS decay constant \( k \) as a function of the Kondo parameter in the absence

\[
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\]
of driving for the generic strong field characterized by $V_0=20\hbar\Omega$, $\omega_0=12\Omega$ and under the “optimal localization” condition $V_0=20\hbar\Omega$, $\omega_0=16.714\Omega$ at $\hbar\Omega\beta=0.1$. It is seen that both driving conditions enhance significantly the lifetime of a localized state. Most importantly, the delocalization rate displays the same plateau that spans the friction range $0.3<\alpha<0.9$ in both systems driven by the same intensity field, irrespective of the precise values of the driving frequencies and in spite of the fact that the dissipationless dynamics is vastly different in these two cases. The friction-independent value of the rate in the plateau regime is in excellent agreement with the prediction of Eq. (3.15) which was obtained from a semiclassical treatment of the quantized field model. At large values of the dissipation parameter the delocalization rate begins to drop off, in accord with the theoretical arguments presented above.

Figure 4(b) displays the TLS delocalization rate at a lower temperature, $\hbar\Omega\beta=2$, as a function of the Kondo parameter for the two driving fields specified earlier as well as in the absence of driving. Here, the onset of the constant rate regime occurs at stronger friction and the rate plateau is overall less pronounced. This is a consequence of more persisting phase relations at lower temperature which favor state-specific behaviors (see the next subsection) and invalidate the classical surface hopping treatment up to larger dissipation values. In addition, it is seen that driving can increase the decay rate at strong friction. This phenomenon can occur when the strength of the driving field is adequate to support a rate plateau that is sufficiently wide to intersect the monotonically decreasing force-free TLS decay rate.

Further confirmation of the analysis presented above is offered by Fig. 5 which shows the dependence of the TLS delocalization rate on temperature for the “generic” driving field specified by $V_0=20\hbar\Omega$, $\omega_0=12\Omega$ at $\alpha=0.64$ with Ohmic spectral density. As seen from Fig. 4(a), this value of the Kondo parameter lies well within the rate plateau regime. The flatness of the driven TLS rate in Fig. 5 presents a striking contrast to the results for the force-free dissipative TLS, showing that the driven TLS delocalization rate is also independent of temperature in this regime.

Finally, we discuss the driven TLS decay constant in the intermediate-to-strong friction regime under driving by different intensity fields for the case of Ohmic dissipation. Figure 6(a) shows the TLS delocalization rate for a driving field of amplitude $V_0=30\hbar\Omega$ at a high temperature $k_B T=10\hbar\Omega$. Results are shown for a generic driving frequency $\omega_0=15\Omega$ and for the value $\omega_0=10.87\Omega$ which corresponds to an optimal localization condition. It is seen that the rate plateau spans an even larger friction range at this high field intensity, extending up to $\alpha=1.5$. Again, the plateau value of the rate is in excellent agreement with the analytical quantized field result.

Figure 6(b) presents the path integral results for the delocalization rate of an Ohmic TLS driven by a weak field of amplitude $V_0=5\hbar\Omega$ at a high temperature $\hbar\Omega\beta=0.1$. We choose $\omega_0=6\Omega$ as a generic field frequency. At this low field intensity optimal localization occurs with $\omega_0=4.28\Omega$; in agreement with earlier numerical findings the optimal localization frequency deviates slightly in this case from the analytical prediction $2V_0/\hbar\omega_0=2.4048$. Here, the rate exhibits a pronounced maximum as a function of friction when driven by the optimal field. The maximum rate occurs at a smaller value of the dissipation parameter compared to the case of a high intensity field and is again in excellent agreement with Eq. (3.15). With the generic driving field corre-
The variation of the TLS decay rate with field amplitude at constant driving frequency and intermediate friction strength is examined in Fig. 7 which shows path integral results for Ohmic friction at various combinations of $a$, $v_0$, and $b$. It is seen that the delocalization rate decreases monotonically with field strength as predicted by the quantized field treatment, irrespective of the specific value of the driving frequency. At sufficiently large values of the field intensity the numerical results are in excellent agreement with the prediction of Eq. (3.15). With weaker fields, when the plateau is expected to disappear, the quantized field result overestimates the decay rate. At very strong friction (data not shown in Fig. 7) the decay rate grows very slowly with $V_0$.

The analysis leading to the existence of a rate plateau implies that the TLS delocalization rate is also independent of the mechanism of dissipation in this regime. To confirm this prediction, we show in Fig. 8 path integral results for the TLS decay rate in the cases of superohmic and Drude dissipation characterized by the spectral densities of Eqs. (3.1b) and (3.1c), respectively. In both cases, the driving amplitude is $V_0=30\hbar\Omega$, while the generic and optimal driving frequencies are chosen as $\omega_0=5.5\Omega$ and $\omega_0=6.933\Omega$. It is seen that the driven TLS exhibits at moderate friction the same universal rate plateau in all cases, whose value is in excellent agreement with the prediction of Eq. (3.15).
agreement with the semiclassical result of Eq. (3.15), even though the field-free dynamics exhibit vastly different behaviors.

B. Weak friction

In the intermediate friction plateau region and at high temperature the TLS delocalization rate depends on the field intensity alone. As shown in Figs. 4, 6, and 7 the decay constant deviates from that pattern at small values of the dissipation parameter, exhibiting behavior sensitive to the parameters of the field as well as those of the environment.

At weak dissipation phase relations become important. Within the quantized field picture constructive phase interference between resonant eigenstates of the diabatic potentials is expected to increase the rate above its plateau value in the case of a symmetric TLS. This expectation is in agreement with the numerical results of Figs. 4, 6, and 7 in the case of “generic” driving conditions.

In the absence of dissipation and for \( \omega_0 \gg \Omega \), the tunneling matrix element is renormalized by the Franck–Condon overlap between resonant states of the diabatic curves. As the separation \( 2q_{\text{min}} \) between these curves becomes nonzero when a field is applied, the Franck–Condon overlap drops below unity and the renormalized tunneling splitting is decreased. This effect results in slower delocalization compared to the field-free result in the weak friction regime. However, the Franck–Condon overlap depends nonmonotonically on the diabatic potential separation and therefore on the field amplitude such that it goes through zero when \( 2V_0/\hbar \omega_0 \) coincides with a zero of the zeroth order Bessel function \( J_0 \); the resulting degeneracy is responsible for exact localization in the absence of dissipation. Weak interaction with a dissipative bath results in slow delocalization via broadening of the degenerate levels.

Due to the nonmonotonic variation of Franck–Condon factors and the uneven spacing of the nodes of \( J_0 \) the lifetime of a TLS localized state exhibits an interesting “antiresonance” pattern at small friction values. Figure 9 shows the lifetime \( \tau=1/k \) of a localized state as a function of the driving frequency with a constant field amplitude \( V_0=20\hbar\Omega \) at temperature characterized by \( \hbar\Omega\beta=0.1 \) for an Ohmic spectrum at several values of the Kondo parameter. The lifetime displays several maxima which become more pronounced as the field frequency is increased up to about \( V_0/\hbar \); i.e., the longest lifetime corresponds to the first node of the Bessel function, \( 2V_0/\hbar \alpha_0=\pi_1 \), where \( \pi_1=2.4048 \), while other local maxima occur at \( 2V_0/\hbar \alpha_0=\pi_i \), \( i=2,3,... \). At zero dissipation even small departure from the rigorous localization condition results eventually in destruction of localization and the lifetime peaks become delta functions. With finite friction these peaks acquire considerable width. Thus the lifetime of localized states can be extended even with significant deviation from optimal conditions. At small driving frequencies neighboring peaks overlap, resulting in small fluctuations of the TLS delocalization rate about the plateau value. At high driving frequency the lifetime decreases monotonically to the driving-free spin-boson value. Finally, when the dissipation becomes sufficiently strong, state-specific effects induced by phase interference are wiped out and the lifetime becomes only very weakly dependent on driving frequency.

Figure 10 displays the dependence of the decay rate on field intensity for the driving frequency \( \omega_0=5\Omega \) at \( \alpha=0.05 \). The delocalization rate exhibits an oscillatory pattern (corresponding to the antiresonance structure discussed in the previous paragraph) superposed on an overall decaying curve.

The increasing width of these antiresonance structures as dissipation gets stronger gives rise to a nonmonotonic varia-

FIG. 9. The lifetime of a localized state as a function of driving frequency for the field amplitude \( V_0=20\hbar\Omega \) obtained by iterative evaluation of the path integral. The TLS is coupled to an Ohmic bath and the temperature is \( \hbar\Omega\beta=0.1 \). (a) \( \alpha=0.025 \). (b) \( \alpha=0.05 \). (c) \( \alpha=0.10 \). (d) \( \alpha=0.20 \).
tation of the delocalization rate with friction as the field parameters approach an optimal localization condition. This effect is seen in Fig. 11, which shows the TLS decay rate constant as a function of the Kondo parameter for $V_0=20\hbar\Omega$ at a few driving frequencies near $\omega_0=16.714\Omega$. At these frequencies, which for $\alpha<0.1$ lie near the center of the lifetime curves of Fig. 9, the decay rate first decreases as the dissipation becomes weaker but grows again as the Kondo parameter is lowered even further, causing narrowing of the antiresonance lines which (at fixed frequency) implies relatively large deviation from optimal localization.

IV. COMPARISON WITH EARLIER WORK

A number of recent articles have analyzed the dynamics of dissipative two-level systems driven by oscillatory fields by extending the noninteracting blip approximation (NIBA) to include time-dependent driving. Use of this approximation led to master equations that describe the effects of the time-dependent field on the decay characteristics of an initially localized state. In this section we compare the NIBA results for the delocalization rate of a periodically driven symmetric TLS to those obtained in Sec. III via accurate path integral calculations.

The driven TLS decay rate expression derived for $\omega_0\gg\Omega$ within the NIBA by Dakhnovskii\textsuperscript{10} as well as by Grifoni \textit{et al.}\textsuperscript{14} reads (after correction of arithmetic errors in Ref. 10) as follows:

$$k_{\text{NIBA}} = 4\Omega^2 \int_0^\infty \frac{J_0}{\omega_0^2} \sin(\omega_0 t/2) \cos(4Q(t)/\pi\hbar)$$

$$\times \exp[-4Q_2(t)/\pi\hbar] dt,$$

where

$$Q_1(t) = \int_0^\infty \frac{J(\omega)}{\omega^2} \sin(\omega t) d\omega,$$

$$Q_2(t) = \int_0^\infty \frac{J(\omega)}{\omega^2} (1 - \cos(\omega t)) \coth(\hbar\omega\beta/2) d\omega.$$

For an Ohmic spectral density the first of these integrals can be evaluated exactly, while the second can be approximated at low temperature to give\textsuperscript{1}

$$Q_1(t) = m_0 \gamma \tan^{-1}(\omega_0 t),$$

$$Q_2(t) \approx \frac{1}{\pi} m_0 \gamma \ln(1 + \omega_0^2 t^2) + m_0 \gamma \ln\left[\frac{\hbar\beta}{\pi t}\right].$$

By employing further approximations to simplify Eq. (4.1) in the Ohmic case, Dakhnovskii concluded\textsuperscript{10–12} that the TLS decay rate increases in general with the ratio $V_0/\omega_0$. That result, which is in direct contradiction to the results presented in the previous section for weak or moderate values of the dissipation parameter, is mainly due to breakdown of the approximations introduced by Dakhnovskii to Eq. (4.1).\textsuperscript{32} On the other hand, using the fact that $|J_0(x)| \ll 1$, Grifoni and Hänggi concluded\textsuperscript{48} that for Ohmic friction and with large $\omega_0$ driving always results in reduction of the rate for a symmetric TLS. However, this statement is correct only if $\omega_0$ is strictly infinite. With finite cutoff frequency Eq. (4.1) predicts a definite decrease of the driven TLS decay rate only in the case of an Ohmic spectral density and for $\alpha<\frac{1}{2}$, in which case the product $\cos[4Q_1(t)/\pi\hbar] \exp[-4Q_2(t)/\pi\hbar]$ is non-negative\textsuperscript{1} for all values of $t$. At larger values of the Kondo parameter, and for any value of friction with other spectral densities, the oscillatory nature of the Bessel function in the NIBA combined with the remaining of the integrand which may also alternate in sign may lead to enhancement of the rate with respect to that of the bare dissipative TLS, in qualitative agreement with the numerical path integral results presented in the previous section.

Even though the presence of the Bessel function in Eq. (4.1) may appear to imply a strong sensitivity of the NIBA decay rate to the ratio $V_0/\omega_0$, the frequency dependence is strongly suppressed at moderate dissipation strengths. We have evaluated Eq. (4.1) via numerical integration and found that the full NIBA rate is only weakly dependent on the driving frequency and the parameters of the environment at

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**Figure 10.** Path integral results for the TLS delocalization rate as a function of field amplitude for the driving frequency $\omega_0=5\Omega$. The TLS is coupled to an Ohmic bath of Kondo parameter $\alpha=0.05$ and temperature $k_B T=10\hbar\Omega$.

**Figure 11.** Path integral results for the decay rate of a localized state in an Ohmic TLS as a function of the Kondo parameter for $V_0=20\hbar\Omega$ and several driving frequencies. Solid squares: $\omega_0=16.714\Omega$ (optimal localization condition); hollow squares: $\omega_0=17.21\Omega$; solid circles: $\omega_0=17.5\Omega$; hollow circles: $\omega_0=18\Omega$; solid triangles: $\omega_0=19\Omega$; hollow triangles: $\omega_0=20\Omega$. The temperature is $k_B T=10\hbar\Omega$. J. Chem. Phys., Vol. 106, No. 6, 8 February 1997
moderate dissipation strengths for the Ohmic and superohmic spectral densities employed in our path integral calculations. However, the existence of a rate plateau in the driven TLS, hidden in the rather intricate structure of the integrand of Eq. (4.1) within the range of validity of the NIBA, has not been noted previously.

To illustrate the above remarks we present in Fig. 12 the ratio of the driven and force-free TLS decay rates as given by the NIBA result obtained via numerical evaluation of Eq. (4.1) for finite and zero fields, respectively, in the case of an Ohmic spectrum with \( \omega_0 = 20 \Omega \) at a high and an intermediate temperature. Unlike approximate evaluations of Eq. (4.1) valid only in limiting regimes, the full NIBA expression reproduces semiquantitatively the results obtained via path integral calculations.

Before concluding this section, we point out an apparent similarity (within a numerical factor of the derived rate expression, Eq. (3.15), to a result obtained by Morillo and Cukier \(^{49} \) for the average TLS decay rate in a disordered classical medium in the limit of strong, nearly static fields (\( V_0 \gg \pi \alpha \hbar c \), \( \omega_0 \ll \omega_e \)). In that work, averaging over the TLS dipole orientation with respect to the external field cancels the friction dependence in the weak dissipation limit. Before such averaging is performed and/or at significant friction strength the delocalization rate obtained by these authors exhibits pronounced dependence on dissipation. By contrast, the rate plateau identified in the present paper exists at constant amplitude driving and thus this behavior is relevant to crystalline materials, exciton transport in semiconductors or single-molecule tunneling in anisotropic media.

**V. CONCLUDING REMARKS**

We have presented a variety of behaviors that arise when a dissipative TLS is driven by a periodic force which corresponds in the semiclassical limit to a continuous-wave monochromatic radiation field. In the parameter regimes where localized states exhibit incoherent relaxation the delocalization rate displays a variety of patterns which can be understood by mapping the periodically driven TLS onto a time-independent curve crossing problem. While this mapping involves the quantized radiation field, the intermediate friction behavior can be analyzed in terms of simple semiclassical ideas, although quantum mechanical effects dominate the dynamics at weak dissipation.

For strong fields and with driving frequencies that are larger than the tunneling splitting the driven TLS is well within the nonadiabatic regime. In that case the high-temperature delocalization rate takes on a “universal” value which depends only on the overall field intensity over a wide range of friction. The “plateau” rate is usually much smaller than that realized under driving-free conditions. At moderate friction it decreases as the inverse of the field amplitude, with an additional oscillatory component at weak dissipation. With strong fields the rate plateau can persist over a large range of friction, such that the driven TLS rate can exceed that in the absence of driving at large values of the dissipation parameter. As the field amplitude decreases, the “adiabaticity” parameter in the curve crossing analogy increases, leading to larger delocalization rate and gradual vanishing of the rate plateau. At very small field intensities and/or very high driving frequencies the system reverts to the well-understood force-free spin-boson Hamiltonian. Finally, at lower temperature and/or very high bath cutoff frequencies the rate plateau shrinks and eventually vanishes if the field amplitude is kept fixed.

At low temperatures and weak dissipation, control of the tunneling dynamics is intimately connected with delicate phase relations which are generally disrupted as the coupling to a macroscopic environment is increased and one might expect the localizing effects of oscillatory fields to disappear when \( \hbar \Omega \beta \ll 1 \) or if the Kondo parameter exceeds a certain small value. As shown in this paper, though, not only do localization effects persist at high temperature even under strong damping conditions; such effects become more robust there, leading to a lifetime of the localized state that is essentially independent of the parameters of the environment and of the driving frequency. Stabilization of localized states is not opposed by dissipative processes.

The behaviors established via accurate path integral calculations are generally in harmony with those predicted by the full NIBA result, Eq. (4.1). Previous qualitatively incorrect conclusions regarding the variation of the TLS decay rate with various parameters are mainly due to incorrect approximations of this expression. In this sense, the NIBA provides (within the range of its validity) a successful description of the symmetric TLS decay rate in the incoherent regime realized at high driving frequencies. On the other hand, the simple semiclassical analysis presented in this paper provides a physically appealing picture that predicts the existence at moderate friction of a universal rate regime where the TLS delocalization rate drops off monotonically with increasing driving amplitude.

The slow relaxation of the driven TLS under diverse conditions in the high-temperature, moderate friction regime is very encouraging from the point of view of controlling the
tunneling dynamics. By applying a simple monochromatic field of appropriate strength one can generally decelerate substantially the delocalization of a symmetric TLS.

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47. This explanation of the discrepancy between Dakhnovskii’s the numerical results was originally communicated by the referee.