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Dissipative tunneling control by elliptically polarized fields

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The tunneling dynamics of a dissipative two-level system that is strongly driven by elliptically polarized electric fields is investigated. The dissipative dynamics is governed within the noninteracting-blip-approximation for the stochastic forces by a generalized master equation (GME). With the focus being on viscous friction, we compare exact numerical solutions of the GME with analytical approximations to both the transient and the asymptotic, long-time periodic dynamics. Novel phenomena are identified: These are a selective control on localization (or, as well, on delocalization) of the tunneling dynamics, or the *inversion* of an initially induced localization by a static bias via multiphoton-assisted tunneling. These effects can be selectively tuned as a function of the eccentricity parameter $p=E_y/E_x$ of corresponding field amplitudes. In particular, the case of a circularly polarized driving field with $p=\pm 1$ yields a dramatic enhancement of the relaxation rate at resonances, when an integer multiple of the angular driving frequency matches the asymmetry energy induced by a static bias. [S0021-9606(98)01931-X]

I. INTRODUCTION

The possibility to control the tunneling dynamics of dissipative multistable systems by time-dependent *linearly* polarized (LP) driving fields has been the objective of many analytical and numerical investigations during the last years.[1–16] Interest in this topical research area is apparent via the ample variety of physical and chemical situations which can be described with a dissipative, driven two-level system (TLS). A TLS can model intrinsic two-level systems, such as spin 1/2 particles or two-level atoms. More generally, it describes the motion of a quantum particle at low temperatures in an effective double well potential in the case that only the lowest energy doublet is occupied. In this latter case, the dissipative TLS can describe, for example, hydrogen tunneling in condensed media,[17] tunneling of atoms between an atomic-force microscope tip and a surface,[18] or tunneling of the magnetic flux in a superconducting quantum interference device (SQUID).[19] Moreover, since the seminal works by Marcus[20] and Levich,[21] this very same model has been applied to describe nonadiabatic chemical reactions in condensed phases, such as electron-transfer[22–25] or proton-transfer reactions.[26] Finally, the change of molecular chirality of an optically active molecule may be regarded as a tunneling process of a charged particle in a double well potential.[27,28]

In this work we investigate the dissipative as well as the nondissipative TLS dynamics in the presence of *elliptically* polarized (EP) driving fields. Moreover, we study the influence of an additionally present dc-bias. With EP fields characterized by two amplitude strengths E_x and E_y, the degree of eccentricity $p=E_y/E_x$ provides an additional parameter to selectively control *a priori* the tunneling dynamics. The nondissipative dynamics of an initially localized TLS driven by circularly polarized (CP) fields has been recently investigated by Shao and Hänggi.[28] They have demonstrated that, as in the case of LP fields, an appropriately chosen CP driving can induce coherent suppression of tunneling. A necessary condition for this to occur is that the driving frequency coincides with the Rabi frequency. We find that in the presence of an additional dc-bias this localization effect is preserved at small-to-moderate dc-bias strengths. However, as the dc-bias strength is increased even further complete delocalization may occur. Moreover, the resulting dynamics can be periodic or almost-periodic.

The effect of decoherence on a CP-driven TLS has been investigated in Ref. 29 within a phenomenological approach based on the stochastic Schrödinger equation. To gain more insight on the role of decoherence versus dissipation effects, we investigate the dissipative tunneling dynamics of an EP-field driven TLS within a microscopic, systematic approach to dissipation. In doing so, we generalize the familiar spin-boson model[30,31] to the case of external EP radiation fields. In this model, dissipation emerges via the contact of the two-state particle to a heat bath described by an ensemble of harmonic oscillators with a continuum of normal mode frequencies. The predictions of this model in the presence of LP-fields have been discussed previously by many authors.[2–16]

Here we generalize the real-time path-integral approach for a LP-driven dissipative TLS[3,5,14] to the case of EP-driving. The environmental influence is investigated within the noninteracting-blip-approximation (NIBA) for the stochastic forces, leading to a generalized master equation (GME) for the tunneling dynamics. For the specific case of viscous friction (i.e., Ohmic dissipation), we compare exact numerical solutions of the GME with analytical approximations to the transient as well as to the asymptotic, periodic long-time dynamics. Several effects, such as the possibility to localize or strongly delocalize the tunneling motion, or the inversion of localization by appropriately tuning of the driving field parameters are discussed. We also contrast our findings for the TLS dynamics in the presence of EP-fields to the commonly studied case of LP-driving.

The paper is organized as follows. In Sec. II we introduce the model and the relevant dynamical quantities. In Sec. III A we obtain a formally exact solution for the driven and dissipative dynamics in the form of a series expression in the number of tunneling transitions. These results are simplified in Sec. III B where we discuss the noninteracting-blip approximation (NIBA) together with the corresponding generalized master equation (GME). Numerical and analytical results for the driven dynamics in the presence and in the absence of dissipation are presented in Sec. IV. Finally, in Sec. V we summarize our findings and present some conclusions.

II. THE DISSIPATIVE TLS DRIVEN BY ELLIPTICALLY POLARIZED FIELDS

To start with, we consider a driven, symmetric[32] nondissipative two-level-system described by the Hamiltonian $H(t)=H_0-\boldsymbol{\mu}\cdot\boldsymbol{\mathcal{E}}(t)$, where H_0 is the unperturbed TLS Hamiltonian, $\boldsymbol{\mu}$ is the TLS dipole moment, and $\boldsymbol{\mathcal{E}}(t)$ is the external elliptically polarized radiation field whose direction rotates in the plane perpendicular to the z axis. In the basis of the energy eigenstates $|1\rangle$, $|2\rangle$ of H_0 this driven TLS Hamiltonian reads

$$H(t)=-\frac{\hbar\Delta}{2}\sigma_z-\mu[\mathcal{E}_x(t)\sigma_x-\mathcal{E}_y(t)\sigma_y],\tag{1}$$

where the σ_i denote the Pauli spin matrices, $\hbar\Delta$ is the bare energy splitting and $\mu=\langle 1|\mu_x|2\rangle=i\langle 1|\mu_y|2\rangle$ is the transition dipole moment. In particular, EP fields are described by $\mathcal{E}_x(t)=E_x\cos(\Omega t)$, $\mathcal{E}_y(t)=E_y\sin(\Omega t)$. Circularly polarized (CP) radiation is obtained when $|E_x|=|E_y|$. Finally, LP fields are described by $\mathcal{E}_x=E_x\cos(\Omega t)$, $\mathcal{E}_y=0$. Hence by introducing the eccentricity parameter

$$p=E_y/E_x,\tag{2}$$

CP fields and LP fields are characterized by $p=\pm 1$ and $p=0$, respectively. In particular, left-circularly polarized light corresponds to $p=1$, and right-circularly polarized light to $p=-1$.

To investigate tunneling problems it is more convenient to express the driven TLS Hamiltonian in the basis of the vectors $|R\rangle=(|1\rangle+|2\rangle)/\sqrt{2}$ and $|L\rangle=(|1\rangle-|2\rangle)/\sqrt{2}$, with $|R\rangle$ and $|L\rangle$ localized states in the right and left well, respectively, of the underlying double well potential. This can be obtained by performing the unitary transformation $R=\exp(-i\pi\sigma_y/4)$ on the Hamiltonian (1). Setting $\hbar\nu_1(t)=-2\mu\mathcal{E}_x(t)$ and $\hbar\nu_2(t)=-2\mu\mathcal{E}_y(t)$, the transformed Hamiltonian $H_{\rm loc}(t)=RH(t)R^{-1}$ reads

$$H_{\rm loc}(t)=-\frac{\hbar}{2}[\Delta\sigma_x+\nu_1(t)\sigma_z+\nu_2(t)\sigma_y].\tag{3}$$

In this representation we find that $\sigma_z|R\rangle=|R\rangle$, and $\sigma_z|L\rangle=-|L\rangle$, so that σ_z plays the role of the position operator of the two-state system. Correspondingly, the effect of EP driving results in a time-dependent modulation $\nu_1(t)$ of the TLS asymmetry energy, as in the familiar LP case, and in a *complex-valued, time-dependent* tunneling matrix element

$$V(t)=\Delta-i\nu_2(t)\tag{4}$$

between the localized states. We observe that the effect of a static dc-field $\mathcal{E}_{\rm dc}$ which breaks the inversion symmetry of the unperturbed Hamiltonian H_0 is described in the driven Hamiltonian (3) upon substituting $\nu_1(t)=-2\mu\mathcal{E}_x(t)$ with $\nu_1(t)=-2\mu[\mathcal{E}_x(t)+\mathcal{E}_{\rm dc}]$.

Finally, to investigate the effects of dissipation on the tunneling dynamics we consider a bath described by an ensemble of harmonic oscillators with a bilinear coupling in the TLS-bath coordinates.[31] This yields the EP-driven spin-boson Hamiltonian, i.e.,

$$H_{\rm SB}(t)=H_{\rm loc}(t)+\frac{1}{2}\sum_i\left[\frac{p_i^2}{m_i}+m_i\omega_i^2x_i^2-c_ix_id\sigma_z\right],\tag{5}$$

where d denotes the tunneling distance between the localized states. As long as we are interested in the reduced dynamics of the TLS alone, the influence of the heat bath is completely characterized by the environmental spectral density, i.e.,

$$J(\omega)=\frac{\pi}{2}\sum_i\frac{c_i^2}{m_i\omega_i}\delta(\omega-\omega_i).\tag{6}$$

The dynamical quantity of interest is the expectation value

$$P(t):=\mathrm{Tr_B}\{W(t)\sigma_z\}\tag{7}$$

of the discretized position operator σ_z. It describes the time evolution of the population difference between the localized states. Here $\mathrm{Tr_B}$ denotes the trace over the bath degrees of freedom with $W(t)$ being the full density matrix of the system-plus-reservoir. We assume that at time $t=0$ the particle is held at the site $|R\rangle$, with the bath being in thermal equilibrium at temperature T. We next consider the system evolution for this factorized initial condition.

III. DYNAMICAL EQUATIONS FOR THE EP-DRIVEN DYNAMICS

A. Exact formal solution

To investigate the TLS dynamics we generalize the real-time path-integral approach for the LP-driven dissipative TLS[3,5,9] to include also the effects of a time-dependent, *complex-valued* matrix element. Summing over the history of the system's visits of the four states of the reduced density matrix, the exact formal solution for the dynamical quantity $P(t)$ can be found in the form of a series in the number of time-ordered tunneling transitions. By introducing

$$\delta_n=\prod_{j=1}^{n}V(t_{2j})V^*(t_{2j-1}),\quad \Phi_n=\sum_{j=1}^{n}\xi_j\int_{t_{2j-1}}^{t_{2j}}dt'\,\varepsilon_1(t'),\tag{8}$$

the solution for the population difference $P(t)$ reads

$$P(t)=1+\sum_{n=1}^{\infty}(-1)^n\int_0^t dt_{2n}\int_0^{t_{2n}}dt_{2n-1}\cdots\int_0^{t_2}dt_1\times 2^{-n}\sum_{\{\xi_j=\pm 1\}}\delta_n e^{i\Phi_n}(F_n^{(+)}+iF_n^{(-)}),\tag{9}$$

with the ξ-charges labeling the two off-diagonal states of the reduced density matrix. The quantities δ_n and Φ_n in (9) derscribe the influence of the time-dependent biasing forces, while the dissipative influences are captured by the functions $F_n^{(\pm)}$. To express the latter 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, respectively, of the bath correlation function ($\beta\equiv 1/k_bT$), i.e.,

$$Q(t)=\frac{d^2}{\hbar\pi}\int_0^\infty d\omega\,\frac{J(\omega)}{\omega^2}\,\frac{\cosh(\hbar\omega\beta/2)-\cosh[\omega(\hbar\beta/2-it)]}{\sinh(\hbar\omega\beta/2)}. \tag{10}$$

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. 15, 30, and 31), the functions $\Lambda_{j,k}$ describe the inter-blip 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 intra-blip and inter-blip 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

$$F_n^{(+)}=G_n\prod_{k=0}^{n-1}\cos(\eta_{n,k}),\quad F_n^{(-)}=F_n^{(+)}\tan(\eta_{n,0}). \tag{11}$$

Thus far our results are (formally) exact. Unfortunately, the exact series expression is still too complicated to use for quantitative calculations. Thus approximation schemes must be invoked.

B. Dynamics within the NIBA for elliptically polarized driving fields

In the following we shall evaluate the driven dynamics within the so termed noninteracting-blip-approximation (NIBA) for the stochastic forces.[30] The NIBA is based on the assumption that the average time spent by the system in a diagonal state of the reduced density matrix (a sojourn) is very long compared to the average time spent in an off-diagonal state (a blip). This assumption leads to the possibility of neglecting bath-induced correlations between different blip time intervals, as well as designated blip-sojourns correlations in the exact series expression. The NIBA is then formally obtained by neglecting the interblip correlations ($\Lambda_{j,k}=0$), and all blip-sojourn correlations ($X_{j,k}=0$ for $j\neq k+1$). Note, however, that in the NIBA the history of the deterministic driving force is completely accounted for. As demonstrated in Ref. 9 for LP driving, the exact formal series expression for the TLS dynamics described by $P(t)$ [i.e., the equivalent of relation (9) for LP driving] can be recast into the form of a generalized master equation (GME). Within the NIBA, the GME for EP driving reads

$$\dot{P}(t)=\int_0^t dt'[K^{(-)}(t,t')-K^{(+)}(t,t')P(t')], \tag{12}$$

with integral kernels $K^{(\pm)}(t,t')=h^{(\pm)}(t-t')\mathcal{F}^{(\pm)}(t,t')$, where

$$\mathcal{F}^{(+)}(t,t')=\mathrm{Re}\{\delta(t,t')\exp[i\eta(t,t')]\}, \tag{13}$$

$$\mathcal{F}^{(-)}(t,t')=\mathrm{Im}\{\delta(t,t')\exp[i\eta(t,t')]\}, \tag{14}$$

and with $\eta(t,t')=\int_{t'}^{t}dt''\nu_1(t'')$, and $\delta(t,t'):=\delta_1(t,t')=V(t)V^*(t')$. Moreover, we have introduced the functions

$$h^{(+)}(t)=\exp[-Q'(t)]\cos[Q''(t)],$$

$$h^{(-)}(t)=\exp[-Q'(t)]\sin[Q''(t)]. \tag{15}$$

The validity of the NIBA for the static case has been discussed in Refs. 30 and 31, and for the LP-driven case in Refs. 3 and 5. Generally, the NIBA assumptions are fulfilled for large enough friction and/or high temperatures, although the range of validity depends on the specific form chosen for the spectral density $J(\omega)$ of the medium. Moreover, the NIBA can be justified for any kind of spectral density as long as the system exhibits overdamped exponential relaxation. In the limit $\nu_2\to 0$ the NIBA kernels for a dissipative TLS with LP driving are recovered.[4,9,10] Finally, in the absence of dissipation we find $h^{(+)}(t)=1$, $h^{(-)}(t)=0$, and the GME (12) with the corresponding kernels describes the nondissipative dynamics at zero temperature *exactly*.

Up to here our results have been valid for *arbitrary* forms of time-dependent fields modulating the asymmetry energy as well as the tunneling matrix element of the TLS. In the following we shall consider the case of a TLS driven both by a dc-field, causing an asymmetry energy, and by a *monochromatic* EP field, yielding

$$\nu_1(t)=\epsilon_0+\epsilon_1\cos(\Omega t),\quad \nu_2(t)=\epsilon_2\sin(\Omega t), \tag{16}$$

where $\hbar\epsilon_0=-2\mu\mathcal{E}_{\mathrm{dc}}$, and $\hbar\epsilon_1=-2\mu E_x$, $\hbar\epsilon_2=-2\mu E_y$. This driving form implies that

$$\mathcal{F}^{(+)}(t,t')=[\Delta^2+\epsilon_2^2\sin(\Omega t)\sin(\Omega t')]\cos[\eta(t,t')]-\Delta\epsilon_2[\sin(\Omega t)-\sin(\Omega t')]\sin[\eta(t,t')],$$

$$\mathcal{F}^{(-)}(t,t')=[\Delta^2+\epsilon_2^2\sin(\Omega t)\sin(\Omega t')]\sin[\eta(t,t')]+\Delta\epsilon_2[\sin(\Omega t)-\sin(\Omega t')]\cos[\eta(t,t')]. \tag{17}$$

Note that the driven TLS is sensible to the sign of ϵ_2, or equivalently, the dynamics is different depending on whether the EP-field is left-polarized or right-polarized. Moreover, we observe that the dc-field plus CP-field driving case is described by $\epsilon_0\neq 0$ and $p=\pm 1$, i.e., $\epsilon_1=\pm\epsilon_2$ in (16); a dc-field plus LP-driving is obtained with $\epsilon_0\neq 0$ and $p=0$, i.e., $\epsilon_2=0$. We now investigate the driven dynamics in different parameter regimes.

IV. CONTROL OF TUNNELING

A. Coherent dynamics

To understand the effects introduced by dissipation, we first discuss the coherent, nondissipative dynamics. For the special case of a zero dc-field and CP driving with $p=1$, i.e., $\epsilon_0=0$ and $\epsilon_1=\epsilon_2=\epsilon$ in (16), an exact analytic solution has recently been discussed in Ref. 28. There, it has been predicted that a CP field can indeed induce coherent destruction of tunneling (CDT). A necessary condition for this to occur is that the driving frequency Ω equals the Rabi frequency $\Omega_R=\sqrt{(\Delta-\Omega)^2+\epsilon^2}$. This condition implies that

$$\Omega_R=\Omega=\Delta(1+\nu^2)/2, \quad \nu=\epsilon/\Delta. \tag{18}$$

In particular, complete localization requires that $\nu\rightarrow\infty$, i.e., an infinitely strong CP field at an infinite high frequency. To reveal the localization mechanism it is convenient to use the Floquet theory. The quasienergies for the nondissipative system with zero dc-field are given by $\tilde{\epsilon}_{\pm,n}=[\pm\Omega_R/2+(n+1/2)\Omega]\mathrm{mod}\Omega$, with n an integer number. The condition for localization $\Omega_R=\Omega$ coincides with the exact level crossing: $\tilde{\epsilon}_{+,0}=\tilde{\epsilon}_{-,1}$. This may be understood as follows. When driven by the periodic CP field, the two eigenstates are surrounded by the field to become the dressed states with the quasienergies as their energies. As in the undriven case, when the level crossing happens, coherence is suppressed. It should be stressed again that the level crossing of the quasienergies yields a necessary (but not sufficient) criterion for suppression of coherence. Therefore just as for the linearly polarized case,[15,33] a similar interference mechanism inducing coherent suppression of tunneling applies to the circularly polarized light.

Here, we investigate the effect of a finite dc-bias $\epsilon_0\neq 0$ on the CDT in the presence of CP-driving, cf. Fig. 1. As shown in Fig. 1(a), a dc-field of low intensity preserves CDT, as it occurs in the zero dc-field limit, cf. inset in Fig. 1(a). The resulting dynamics, although still localized, exhibits a different periodicity as compared to the zero dc-field case. For comparison, the predictions for the dc-bias case ($\epsilon_0\neq 0$ and $\epsilon_1=\epsilon_2=0$), and for the LP case in the presence of the dc-field ($\epsilon_0\neq 0,\epsilon_1\neq 0$, and $\epsilon_2=0$) are also shown. It is seen that the sole dc-driven dynamics gets modified only slightly if an additional LP field is added (whose frequency Ω and strength ϵ match the localization condition (18) for CP fields). As shown in Fig. 1(b), a dramatic change occurs when a stronger dc-bias is assumed. The dc-bias is chosen to be large enough so that in the absence of additional fields the TLS dynamics is *fully localized* (see dashed curve). When an additional LP field is applied the resulting dynamics becomes less localized. In contrast, an additional CP field satisfying the localization condition (18) is now sufficient to *fully delocalize* the dynamics.

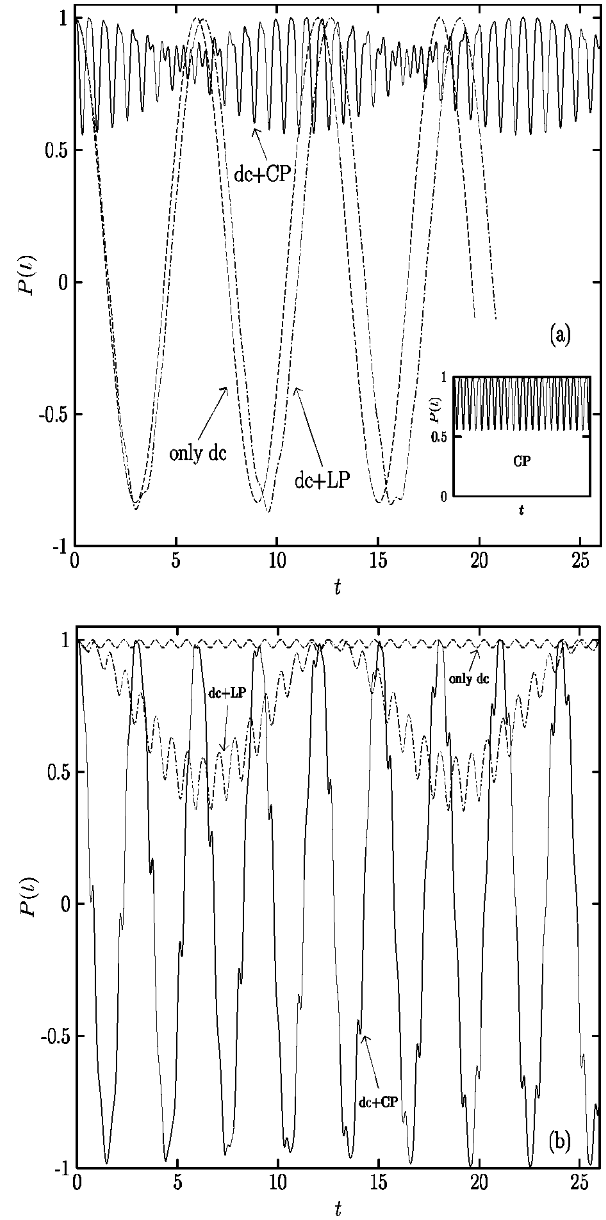

FIG. 1. Coherent, nondissipative time evolution of the population difference $P(t)$ between the localized states of a TLS driven both by a dc-field and by time-dependent CP or LP fields. The driving frequency is $\Omega=8.5\Delta$ and the CP driving strength is $\epsilon_1=\epsilon_2=4.0\Delta$, whereas in the LP case $\epsilon_1=4.0\Delta$ and $\epsilon_2=0$. In panel (a) a small dc-bias $\epsilon_0=0.3\Delta$ is considered, while in panel (b) the dc-bias strength is raised to $\epsilon_0=8.0\Delta$. (a) For small dc-bias strength ϵ_0 a CP-field induced coherent destruction of tunneling (CDT) does occur. A necessary condition for CDT is that the driving frequency Ω equals the Rabi frequency. The inset shows the occurrence of CP-driving induced CDT in the *absence* of the dc-bias. (b) CDT is destroyed for larger dc-fields strengths. In contrast, the dynamics becomes *fully* delocalized. Note, however, that in the sole presence of the dc-field, or of the dc-field plus LP field, the TLS dynamics is completely localized.

B. Dissipative driven dynamics

Thus far our results are valid for arbitrary dissipation. In order to make quantitative predictions, we specify a form of the spectral density $J(\omega)$ of the bath that is suitable to describe realistic physical or chemical systems. In the following the bath is assumed to have a viscous Navier-Stokes form described by an ''Ohmic'' spectrum, i.e.,

$$J(\omega)=(2\pi\hbar/d^2)\alpha\omega e^{-\omega/\omega_c}. \tag{19}$$

Here ω_c is an exponential cutoff frequency corresponding to the smallest relaxation time scale $\tau_c=1/\omega_c$ of the medium. The friction strength $\alpha=E_r/2\hbar\omega_c$ is a dimensionless cou-

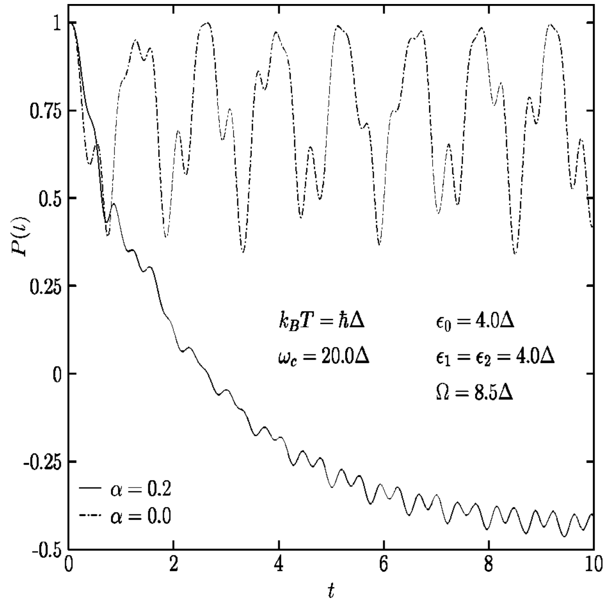

FIG. 2. Dissipative TLS dynamics in the presence of a generic CP-field plus dc-field driving within the NIBA. Viscous friction (i.e., ''Ohmic'' dissipation) is considered. For comparison, the nondissipative, coherent dynamics (dot-dashed curve) is also depicted. The dissipative population difference $P(t)$ exponentially decays towards an asymptotic nonstationary state for characteristic parameters of the CP driving. In contrast to the nondissipative case where the driven dynamics can be periodic or almost periodic, the asymptotic dynamics always becomes periodic assuming the periodicity of the external field. Here and in the following figures α denotes the dimensionless viscous friction coefficient, and ω_c the cutoff frequency that characterize the Ohmic spectral density in Eq. (19).

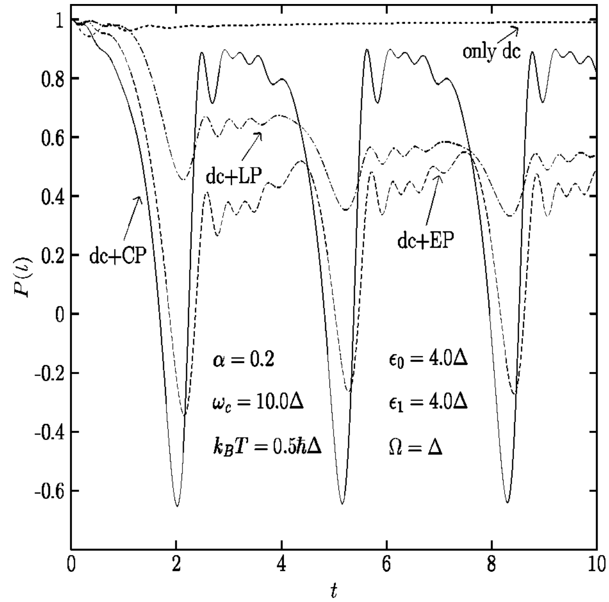

FIG. 3. Large amplitude oscillations characterize the asymptotic dynamics of a dissipative TLS that is driven simultaneously by a strong dc-field and by a CP, LP, or EP field. The dc-bias strength ϵ_0 is chosen sufficiently large so that the system is completely localized in the sole presence of the dc-field. Additional ac-fields cause a periodic population transfer between the two states (i.e., delocalization). The time-dependent field strengths are chosen to be $\epsilon_1 = \epsilon_2$ for CP driving, $\epsilon_1 = 2\epsilon_2$ for EP driving, and $\epsilon_2 = 0$ for LP driving, respectively.

pling constant formed by the ratio of the medium reorganization energy $E_r = (d^2/\pi)\int_0^\infty d\omega J(\omega)/\omega$ to twice the cutoff frequency.

The spectral form (19) as been used, e.g., to describe long-range electron transfer reactions as well as proton transfer reactions in molecular solids. For typical electron or proton transfer reactions τ_c is in the range of 1 ps, while the medium reorganization energy typically exceeds 10 cm^{-1} $\simeq 1.25\times 10^{-3}$ eV.[2,34] In the following, we choose moderate friction $\alpha=0.2$ and a cutoff frequency $\omega_c = 20.0\Delta$, leading to a reorganization energy $E_r = 80$ cm^{-1} for an unperturbed tunneling splitting energy $\hbar\Delta$ of 10 $\mathrm{cm}^{-1} \simeq 1.25\times$meV. Finally, addressing the temperature regime $k_B T \approx \hbar\Delta$, this corresponds to the temperature regime around $T\approx 15$ K.

From (19) the bath correlation functions $Q(t)$ in (10) can be evaluated exactly, yielding

$$Q''(t) = 2\alpha \arctan(\omega_c t),$$

$$Q'(t) = 2\alpha \ln\left\{ \sqrt{1+\omega_c^2 t^2} \frac{\Gamma^2(1+1/\hbar\beta\omega_c)}{|\Gamma(1+1/\hbar\beta\omega_c + it/\hbar\beta)|^2} \right\}, \tag{20}$$

where $\Gamma(x)$ denotes the Gamma function. Some characteristics of the driven dynamics obtained by a numerical integration of the NIBA master equation (12) are depicted in Figs. 2 and 3. As shown with Fig. 2, the interaction with the heat bath qualitatively changes the driven TLS dynamics. In the absence of dissipation (dot-dashed curve) the population dynamics in the presence of a *generic* CP field can be periodic or almost-periodic (i.e., the temporal dynamics is governed by two incommensurate quasienergies). The environmental influence is twofold (cf. full-line): (i) Dissipation destroys quantum coherence leading to an exponential decay towards an asymptotic, nonstationary state. (ii) The asymptotic dynamics reached at long times is *periodic* with the periodicity of the external CP-field. Figure 3 depicts an example of control of tunneling in the presence of different kinds of driving fields: The dynamics of the population difference which can be fully *localized* in the presence of a dc-field, can be strongly *delocalized* when additional ac-fields of intermediate angular frequency Ω are acting. As shown in Fig. 3, large amplitude oscillations in the asymptotic dynamics of $P(t)$ can be observed for LP fields as well as for EP and CP fields. The resulting effect of these driving-induced large amplitude oscillations is a periodic population transfer between the two localized states of the system, being most pronounced in the case of CP-driving. The periodicity is in all of the three cases given by the driving frequency Ω of the ac-field. Moreover, a fine resonance structure is superimposed on top of the oscillatory behavior. To gain better insight to the results of Fig. 2 and Fig. 3 we apply suitable approximations to the GME in (12).

1. Markovian regime

In the following we investigate the Markovian limit $\tau(\Omega,\epsilon_1,\epsilon_2) \ll \{2\pi/\Omega, \gamma_{\mathrm{H}}^{-1}, \dots\}$, where τ denotes the characteristic memory time of the kernels in Eq. (14). Generally, it depends on the bath parameters as well as on the driving field parameters. That is, in the Markovian regime τ is assumed to be the smallest time scale of the system dynamics.

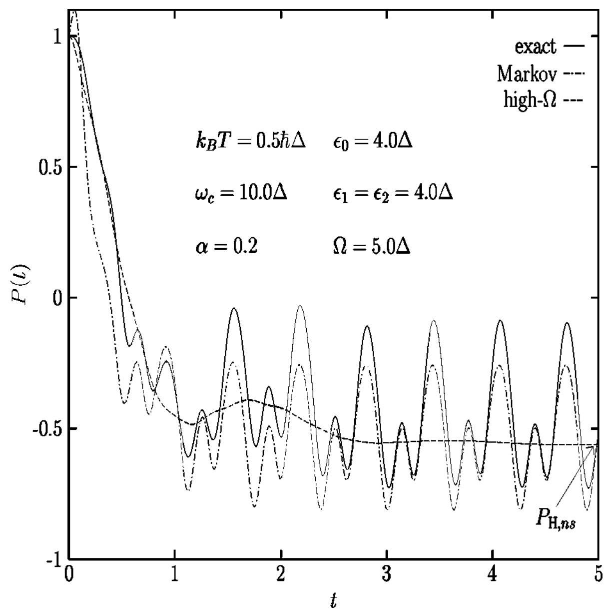

FIG. 4. Comparison between the predictions of the NIBA generalized master equation for the population difference $P(t)$, cf. (12), and the Markov and high-frequency approximations, Eqs. (21) and (24), respectively. A CP driving field of intermediate frequency Ω is used.

We have denoted by $\gamma_{\rm H}$ the time-average of the time-dependent relaxation rate $\gamma_{\rm M}(t)$, cf. (28) below, governing the transient long-time dynamics, cf. (22) below. Then, the long-time behavior, within the NIBA, is intrinsically incoherent. The Markovian approximation $P_{\rm M}(t)$ to $P(t)$ obeys the rate equation

$$\dot{P}_{\rm M}(t) = -\gamma_{\rm M}(t)[P_{\rm M}(t) - P_{{\rm M},ns}(t)], \tag{21}$$

with the time-dependent rate given by

$$\gamma_{\rm M}(t) = \int_0^\infty d\tau\, h^{(+)}(\tau)\mathcal{F}^{(+)}(t,t-\tau), \tag{22}$$

and time-dependent, nonstationary equilibrium value $P_{{\rm M},ns}(t) = \rho_{\rm M}(t)/\gamma_{\rm M}(t)$, with

$$\rho_{\rm M}(t) = \int_0^\infty d\tau\, h^{(-)}(\tau)\mathcal{F}^{(-)}(t,t-\tau). \tag{23}$$

In Fig. 4 the predictions of the Markovian approximation given by Eq. (21) are compared with those of the exact NIBA equation (12) for the case of moderate friction α, and for an intermediate driving frequency Ω of the CP field. The predictions of the high-frequency approximation discussed in the next section are also reported. One clearly observes that in this parameter regime the Markovian approximation provides good results for the long-time dynamics. However, it may predict an incorrect behavior at short propagation times. For example, for the parameters chosen in Fig. 4, unphysical values of $P(t)$ exceeding one are assumed at very short propagation times.

We conclude with some remarks on the validity of the Markovian approximation. In the absence of time-dependent fields it is well established[30,31] that the TLS exhibits an incoherent Markovian dynamics—and correspondingly the NIBA is justified—for strong enough damping $\alpha > 1$, for high enough temperatures $k_BT \geqslant \hbar\Delta/\alpha$ $(\alpha < 1)$, or in the presence of a strong dc-field $\epsilon_0 \gg \Delta$. These conditions become modified in the presence of an additional ac-field. For example, for a very slowly varying EP-field the overall effect is an adiabatic modulation of the asymmetry energy of the TLS, as well as of the tunneling matrix element. Correspondingly, the TLS will exhibit an overdamped dynamics when $|\nu_1(t)| > |V(t)|$, and/or when $\alpha > 1$, and/or when $k_BT \geqslant \hbar |V(t)|/\alpha$ $(\alpha < 1)$. When the driving frequency is increased, driving-induced correlations invalidate these simple quasi-static prescriptions for the validity of the Markovian approximation. We discuss this moderate-to-high-frequency regime in the next section.

2. High frequency regime

In the high-frequency regime $\Omega \gg \{\gamma_{\rm H}, \Delta, \epsilon_2\}$, the driving field oscillates too fast to account for the details of the dynamics within a single period. A valid approximation to the dynamics described by Eq. (12) amounts to approximate the kernels $K^{\pm}(t,t')$ in Eq. (12) with their average $\langle K^{(\pm)}(t,t')\rangle_\Omega := K_{\rm H}^{(\pm)}(t-t')$ over a driving period. Hence time translation invariance is recovered by the averaging procedure, and the essential dynamics of $P(t) \to P_{\rm H}(t)$ is governed by the convolutive equation

$$\dot{P}_{\rm H}(t) = \int_0^t dt' [K_{\rm H}^{(-)}(t-t') - K_{\rm H}^{(+)}(t-t')P_{\rm H}(t')]. \tag{24}$$

The evaluation of the time-averaged kernels is accomplished readily by observing that this time average has only to be carried out on the field-dependent contributions. Therefore we obtain

$$K_{\rm H}^{(+)}(t) = \Delta^2 h^{(+)}(t)\cos(\epsilon_0 t)\mathcal{L}(t),$$
$$K_{\rm H}^{(-)}(t) = \Delta^2 h^{(-)}(t)\sin(\epsilon_0 t)\mathcal{L}(t), \tag{25}$$

where, with the form (17) of the driving functions $\mathcal{F}^{(\pm)}$, the field-dependent contribution $\mathcal{L}(t)$ reads

$$\mathcal{L}(t) = \left[J_0\left(\frac{2\epsilon_1}{\Omega}\sin\frac{\Omega t}{2}\right) - 2\frac{\epsilon_2}{\Delta}\sin(\Omega t/2) J_1\left(\frac{2\epsilon_1}{\Omega}\sin\frac{\Omega t}{2}\right) + \frac{\epsilon_2^2}{2\Delta^2}\cos(\Omega t) J_0\left(\frac{2\epsilon_1}{\Omega}\sin\frac{\Omega t}{2}\right) + \frac{\epsilon_2^2}{2\Delta^2} J_2\left(\frac{2\epsilon_1}{\Omega}\sin\frac{\Omega t}{2}\right)\right], \tag{26}$$

with $J_\nu(z)$ denoting the Bessel function of first kind of order ν. Because (24) is of convolutive type, a solution is conveniently obtained by Laplace transformation techniques. In particular, the resulting long-time dynamics of the time-averaged population difference $P_{\rm H}(t)$ is Markovian, and governed by the equation

$$P_{\rm H}(t) = (1 - P_{{\rm H},ns})\exp(-\gamma_{\rm H} t) + P_{{\rm H},ns}, \tag{27}$$

where $P_{{\rm H},ns} = \rho_{\rm H}/\gamma_{\rm H}$, and

$$\gamma_{\rm H} = \int_0^\infty d\tau\, K_{\rm H}^{(+)}(\tau), \quad \rho_{\rm H} = \int_0^\infty d\tau\, K_{\rm H}^{(-)}(\tau). \tag{28}$$

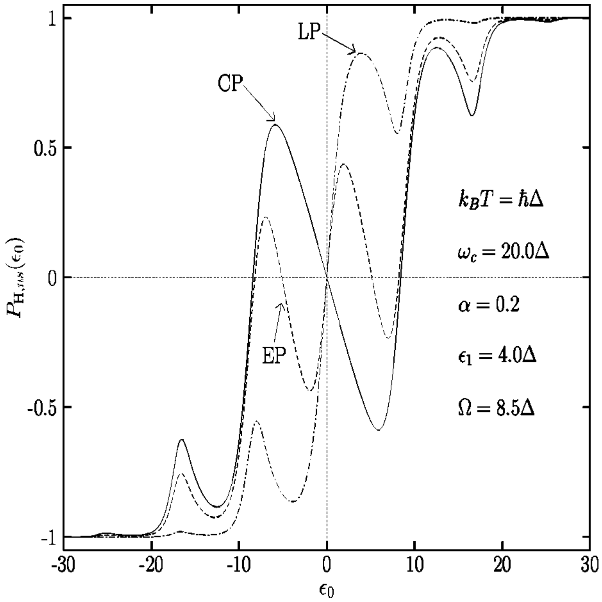

FIG. 5. Inversion of localization: For CP and EP driving the time-averaged nonstationary equilibrium value $P_{\mathrm{H},ns}$ becomes negative even though a positive dc-bias is applied. For CP driving $\epsilon_2=\epsilon_1$, while $\epsilon_2=0$ for LP driving, and $\epsilon_2=0.25\epsilon_1$ for EP driving. This inversion originates from multiphoton absorption and emission processes as described by (30).

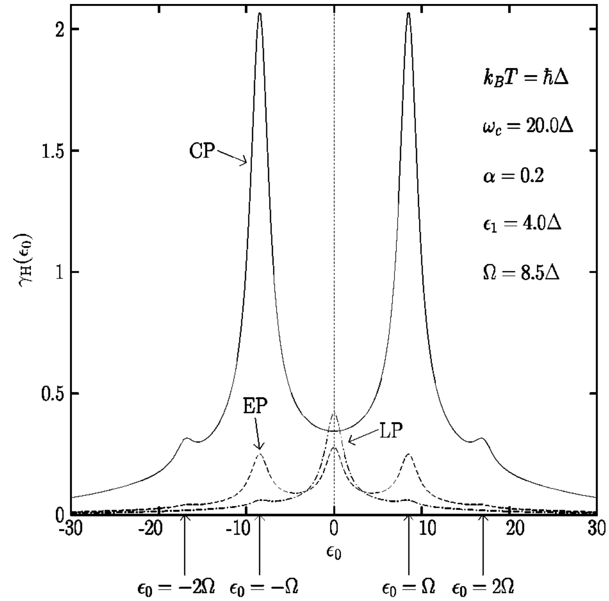

FIG. 6. Photon assisted tunneling. The high-frequency rate γ_{H} plotted versus the dc-bias ϵ_0 exhibits characteristic resonance peaks at multiple integers of the driving angular frequency Ω. The driving-induced new channels occur in the presence of CP, EP as well as LP driving fields. The weights $\mathcal{C}_n$ of these channels are *different* for the three types of ac-driving. For the chosen parameters, the CP-driven high-frequency dynamics is dominated by the first CP-emission channel $\mathcal{C}_1$, while $\mathcal{C}_0\simeq 0$. Correspondingly, the decay rate at $\epsilon_0=\Omega$ is strongly enhanced. The parameters are chosen as in Fig. 5.

Thus a fast field suppresses the periodic long-time oscillations, yielding an exponential decay with high-frequency relaxation rate γ_{H} towards the time-averaged, nonstationary equilibrium value $P_{\mathrm{H},ns}$. We observe that [as follows from (27)], the time-averaged forward rate γ_f and backward rate γ_b between the localized states can be conveniently expressed as $2\gamma_f=\gamma_{\mathrm{H}}+\rho_{\mathrm{H}}$, and $2\gamma_b=\gamma_{\mathrm{H}}-\rho_{\mathrm{H}}$, respectively. Hence the quantity $\rho_{\mathrm{H}}=\gamma_f-\gamma_b$ can be interpreted as the averaged tunneling current from the right to the left localized state.

The predictions of (24) are shown in Fig. 4 and compared with the numerical solution of the GME (12). A CP field of intermediate frequency Ω is assumed. Even though the frequency is not very high, the high-frequency approximation (24) correctly reproduces the field-averaged dynamics of $P(t)$. The system exponentially reaches at long times the asymptotic averaged equilibrium value $P_{\mathrm{H},ns}$, around which the exact NIBA solution oscillates.

The time-averaged, nonstationary equilibrium value $P_{\mathrm{H},ns}$, and the tunneling rate γ_{H}, are depicted versus the applied static bias ϵ_0 in Figs. 5 and 6, respectively. Some interesting features relating to the selective control of tunneling are revealed: As depicted in Fig. 5, suitably chosen CP-fields can induce an *inversion of localization*, i.e., $P_{\mathrm{H},ns}$ can become *negative* even though a *positive* dc-bias is applied. In the investigated parameter regime, the region where this inversion of localization occurs becomes smaller for EP-driving, and in the LP case (eccentricity $p=0$) no inversion is revealed. However, for other parameter regimes, an inversion of localization can also be induced by LP driving fields. For large asymmetries ϵ_0, the three driving types induce a similar behavior, and for extremely large asymmetries ϵ_0 the expected limiting values ± 1 are reached. In Fig. 6 the high-frequency rate γ_{H} in (28) is depicted versus dc-bias ϵ_0 for the parameters chosen in Fig. 5. Characteristic maxima, due to multiphoton absorption or emission processes, occur at multiple integers of the driving frequency for CP as well as for EP and LP driving. For the chosen parameters, the CP-field rate is strongly enhanced as compared to the EP-field, or LP-field rates. The maximal enhancement occurs in correspondence to the first resonance peak. Because the dc-bias dependence of the rate in the absence of ac-fields closely resembles that of the LP-field rate (not shown), the CP-field rate is also larger than the corresponding rate in the presence of a sole dc-bias. Finally, we observe that the LP-field rate becomes larger than the CP-field rate only around zero bias $\epsilon_0=0$, where the CP-field rate exhibits a minimum, rather than a maximum.

In the following we analyze the reason for this interesting behavior of γ_{H} and $P_{\mathrm{H},ns}$. To do so, we look for an appropriate series representation for the high-frequency rate γ_{H} as well as for the tunneling current ρ_{H}. By making use of the formula[35]

$$J_\nu\left(2z\,\sin\frac{a}{2}\right)=\sum_{n=-\infty}^{\infty} J_n(z)J_{n+\nu}(z)e^{i[na+\nu(a-\pi)/2]}, \quad (29)$$

we obtain

$$\mathcal{R}(\epsilon_0,\epsilon_1,\epsilon_2;\alpha)=\sum_{n=-\infty}^{\infty}\mathcal{R}_0(\epsilon_0+n\Omega;\alpha)\times\left[J_n\left(\frac{\epsilon_1}{\Omega}\right)+\frac{\epsilon_2}{2\Delta}J_{n-1}\left(\frac{\epsilon_1}{\Omega}\right)-\frac{\epsilon_2}{2\Delta}J_{n+1}\left(\frac{\epsilon_1}{\Omega}\right)\right]^2, \quad (30)$$

where $\mathcal{R}=\gamma_{\rm H}$ or $\rho_{\rm H}$, and $\mathcal{R}_0(\epsilon_0;\alpha):=\mathcal{R}(\epsilon_0,\epsilon_1=0,\ \epsilon_2=0;\alpha)$ denotes the corresponding quantity in the absence of EP-fields. Hence, the EP-field produces new channels for dc-tunneling due to multiphoton emission $(n<0)$ and multiphoton absorption $(n>0)$, each weighted by the factor $\mathcal{C}_n:=[J_n(\epsilon_1/\Omega)+(\epsilon_2/2\Delta)J_{n-1}(\epsilon_1/\Omega)-(\epsilon_2/2\Delta)J_{n+1}(\epsilon_1/\Omega)]^2$. The structure of $P_{{\rm H},ns}$, or of the high-frequency rate $\gamma_{\rm H}$, can be explained by analyzing the complex interference pattern between the shifted dc-replicas $\mathcal{R}_0(\epsilon_0+n\Omega;\alpha)$, each weighted by the factor $\mathcal{C}_n$. Let us focus on the inversion of localization observed in Fig. 5 for CP-driving. Because $P_{{\rm H},ns}$ is given by the ratio between $\rho_{\rm H}$ and $\gamma_{\rm H}$, the inversion of localization occurs, with $\gamma_{\rm H}>0$, when the averaged tunneling current $\rho_{\rm H}$ becomes negative. The sign of $\rho_{\rm H}$ is determined by the competition between the channels with $n=0$, $n>0$, each of which contributing a positive weight to the total current, and higher order channels with $n<0$ which may yield a negative contribution. With the help of the blocking effect of the $n=0$ channel (i.e., $\mathcal{C}_0\simeq 0$), it can thus be more favorable to emit n photons rather than to absorb n of them, even if the weight of the sidebands is the *same*, and given by $\mathcal{C}_n$. For example, for the CP-field case in Fig. 5 the main contribution to the average value $P_{{\rm H},ns}$ is found to come from the first negative channel $(n=-1)$. The structure of the high frequency rate $\gamma_{\rm H}$ can be explained along the same line of reasoning. For example, the maxima at $\epsilon_0=n\Omega$ can be understood by noting that the static rate $\gamma_0(\epsilon_0;\alpha)$ exhibits a maximum at $\epsilon_0=0$. Finally we observe [as clearly seen from (30)] that this photon-sideband structure arises for EP as well as for LP fields. With only LP fields acting, the corresponding weight $\mathcal{C}_n$ of the sidebands equals $J_n^2(\epsilon_1/\Omega)$.[6]

V. CONCLUSION

In this work we investigated the role of elliptically polarized driving on the tunneling dynamics of a two-level system that generally is subject to quantum dissipation. The eccentricity parameter p characterizing elliptically polarized field driving, and a static bias, provide a tunable mechanism for controlling driven, dissipative tunneling. In the absence of dissipation, coherent destruction of tunneling as well as bias-induced delocalization can occur with circularly polarized fields, when the monochromatic angular driving frequency matches the Rabi frequency. Dissipation alters this coherent tunneling dynamics, leading to an oscillatory decay of coherence towards an asymptotic long-time dynamics that is periodic with the periodicity of the external driving field. Most striking features of CP-driving controlled dissipative tunneling are the possibility of inversion of bias-induced localization, or the strong delocalization phenomenon via selective EP-driving of an initial, bias-induced localization. The high-frequency, long-time regime is characterized by an average Markovian relaxation rate which exhibits resonances when an integer multiple of the EP-driving angular frequency matches the static bias. Both the inversion of localization and the resonancelike features of the decay rate of coherence are governed by multiphoton tunneling events that can be tuned *a priori* by an appropriate choice of the driving parameters. One should point out that all of these features can qualitatively also be realized, in a different parameter regime, by a suitably chosen linearly polarized driving field. However, both the inversion of localization and the resonant structure of the decay rate are more pronounced effects for elliptically polarized than for linearly polarized driving. Therewith, there is a wide parameter regime, as well as different forms of driving fields, placed at the disposal to observe the novel features. These latter are expected to become observable for proton and electron transfer reactions in condensed phases at sufficiently low temperatures.

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