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TUNNELING CONTROL OF A DISSIPATIVE DOUBLE-DOUBLET-SYSTEM BY EXTERNAL FIELDS

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The possibility to control the quantum dynamics of a dissipative double-doublet-system by external time-dependent driving fields is investigated. The system consists of two tunneling doublets separated by an energy gap, as it is for example the case of a double-well potential in which the tunneling dynamics can no longer be restricted to the lowest tunneling doublet. A real-time path-integral approach is used to investigate, both analytically and numerically, the vibrational as well as the tunneling dynamics in an *unified* way. It is shown that the process of thermalization is selectively controlled by a time-dependant field.

1 Introduction

Dissipative tunneling in effective bistable systems finds widespread applications in many physical and chemical situations (for a review see e.g. Ref. [1]). Moreover, the development of strong laser and maser sources has opened a doorway to control the time evolution of nonlinear quantum systems. With the exception of only a few works[2,3,4], all previous treatments of driven bistable tunneling systems have restricted the dynamics to the lowest tunneling-doublet[5]. This so-termed two-level system (TLS) approximation describes well the dynamics at very low temperatures; but it becomes increasingly invalid at higher temperatures and for resonant driving fields that couple different, well separated tunneling doublets.
With the two-level approximation abandoned, the dynamics involves both tunneling between the metastable quantum wells, as well as vibrational intrawell transitions among different tunneling doublets. The prime focus of this work is the development of a consistent treatment wherein tunneling (T), driving and vibrational relaxation (VR) are treated on a common footing. This becomes possible if one uses the representation in which the system-bath coupling operator is diagonal, i.e. the so termed *discrete variable representation* (DVR). A real-time path integral approach is used to derive a set of coupled generalized master equations (GME) within a noninteracting cluster approximation for the combined VR and T dynamics (VRT-NICA).

2 The model

For the sake of clarity only, but without loss of generality, we shall restrict the following discussion to the case that only two tunneling doublets contribute significantly to the driven dynamics. We consider the Hamiltonian $H(t) = H_{\rm DDS} + H_{\rm ext}(t) + H_{\rm B}$. The first term $H_{\rm DDS}$ denotes the Hamiltonian of the isolated double-doublet system. This can be derived from the energy spectrum E_n (energy eigenstates $|n\rangle$) of a double-well potential which is restricted to the two lowest lying doublets $\hbar\Delta_1 = E_2 - E_1$ and $\hbar\Delta_2 = E_4 - E_3$. In analogy with the TLS case, we define a new basis $\{|R_1\rangle, |L_1\rangle, |R_2\rangle, |L_2\rangle\}$ of appropriate linear superpositions of the energy eigenstates, with the new states being localized in the left or right well, respectively. Then, the isolated Hamiltonian takes the form

$$H^{loc}_{\rm DDS} = -\sum_{i=1,2} \frac{\hbar\Delta_i}{2}\Big(|L_i\rangle\langle R_i| + |R_i\rangle\langle L_i|\Big) + \hbar\overline{\omega}_0 I_2 \,, \tag{1}$$

where $I_2 = |L_2\rangle\langle L_2| + |R_2\rangle\langle R_2|$ and $\overline{\omega}_0$ is the energy gap separating the two doublets. The external field-control is characterized by $H_{\rm ext}(t) = -(\mathcal{E}_0 + s\sin\Omega t)\, q$. In this localized representation the discrete position operator of the system reads $q = \sum_{i,j=1,2} a_{ij}(|R_i\rangle\langle R_j| - |L_i\rangle\langle L_j|)$. With $a_{11} = \langle 1|q|2\rangle$, $a_{22} = \langle 3|q|4\rangle$ and $a_{12} = a_{21} = (\langle 1|q|4\rangle + \langle 2|q|3\rangle)/2$ it is – in clear contrast to the TLS - case – *nondiagonal*. Finally, we model quantum dissipation by an ensemble of harmonic oscillators that are bilinearly coupled to the system [1], i.e., $H_{\rm B} = \frac{1}{2}\sum_{i=1}^{\infty}\left[\frac{p_i^2}{m_i} + m_i\omega_i^2\left(x_i - \frac{c_i}{m_i\omega_i^2}q\right)^2\right]$. Then, the environmental influence on the system is fully characterized by the bath spectral density $J(\omega)$. In this work we consider the explicit Ohmic form with exponential cut-off ω_c, namely $J(\omega) = \gamma\,\omega\, e^{-\omega/\omega_c}$, with γ being the friction coefficient. We wish to evaluate the probability $P_{\rm L}(t) := \sum_i \langle L_i|\rho(t)|L_i\rangle$ to find at time $t > t_0$ the system in the left well, for a factorized initial state $W(t_0) = \rho(t_0)\, W_{\rm B}$ of the global density matrix. Here $\rho(t) = {\rm Tr}_{Bath} W(t)$ denotes the reduced density matrix (RDM) of the system, while $W(t)$ is the density matrix of the system-plus-reservoir. At time t_0, we assume the bath to be in thermal equilibrium at temperature T. For an evaluation of the double path integral representation of the RDM, we need to express the isolated Hamiltonian in the eigenbasis of that operator which couples the system to the bath, i.e. the position operator q. With $q|u_k\rangle = u_k|u_k\rangle$ (k=1,..,4) the DDS Hamiltonian in this so called *discrete variable representation* reads

$$H^{DVR}_{\rm DDS} = \sum_{k,l=1,..,4} |u_k\rangle\langle u_k|H^{loc}_{\rm DDS}|u_l\rangle\langle u_l| \,. \tag{2}$$

Within this new representation the double path integral expression for the RDM can be evaluated analytically and numerically.

3 The method: Real-time path-integral evaluation of the RDM

By introducing the notation $\rho_{ii'} := \langle u_i|\rho(t)|u_{i'}\rangle$ the formally exact path integral expression for the RDM in the DVR basis reads

$$\rho_{ii'}(t) = \sum_{j,j'=1}^{4} \int \mathcal{D}q \int \mathcal{D}q' \mathcal{A}[q]\mathcal{A}^*[q']\mathcal{F}[q,q']\rho_{jj'}(t_0) \ , \qquad (3)$$

with the paths subject to the constraint $q(t) = u_i$, $q'(t) = u_{i'}$, and $q(t_0) = u_j$, $q'(t_0) = u_{j'}$. The quantity $\mathcal{A}[q]$ is the probability amplitude to follow the path $q(s)$ for zero bath coupling. The bath influence is captured by the Feynman-Vernon influence functional $\mathcal{F}[q,q']$ [1] which couples each path to every other one.

By introducing the symmetric and antisymmetric paths $\xi_\pm(s) = q(s) \pm q'(s)$ the double path-integral is expressed as a single path-integral over the sixteen states of the RDM in the (q,q')-plane. Any path can then be viewed a sequence of 'sojourns' (S) and 'clusters' (C), with S/C being the time-intervals spent in a diagonal/off-diagonal state of the RDM. In general, the influence functional induces interactions among different C, among C and S, as well as inside a same C. Neglecting the intercluster interactions (for details see Ref. [6]), a generalized master equation (GME) can be derived. It reads

$$\dot{\rho}_{ii}(t) = I_i(t,t_0) + \sum_j \int_{t_0}^{t} dt' H_{ij}(t,t')\rho_{jj}(t') \ . \qquad (4)$$

4 Results

The predictions of the GME (4) for sequential VR-T dynamics are shown in in Fig. 1a where the population $P_L(t)$ of the left well is depicted. $P_L(t)$ is compared against precise numerical results obtained from an iterative evaluation of the real-time path integral (3) within the quasi-adiabatic propagator path integral (QUAPI) method[4,7]. In the chosen parameter regime, the agreement is excellent. At lower temperatures deviations occur when higher order coherent paths contribute to the VR-T dynamics. An example of *control* of tunneling is shown in Fig. 1b. The system is prepared at initial time $t_0 = 0$ in the left-down state $|L_1\rangle$. In presence of a heat bath, the undriven DDS thermalizes to equilibrium (lower curve). However, the process of thermalization can be considerably slowed down in the presence of a suitably tuned external ac-field (middle curve). The field parameters are chosen such that the two quasienergy levels belonging to the upper doublet Δ_2 of the isolated driven DDS do cross (upper curve). In this latter case tunneling is suppressed, i.e.,

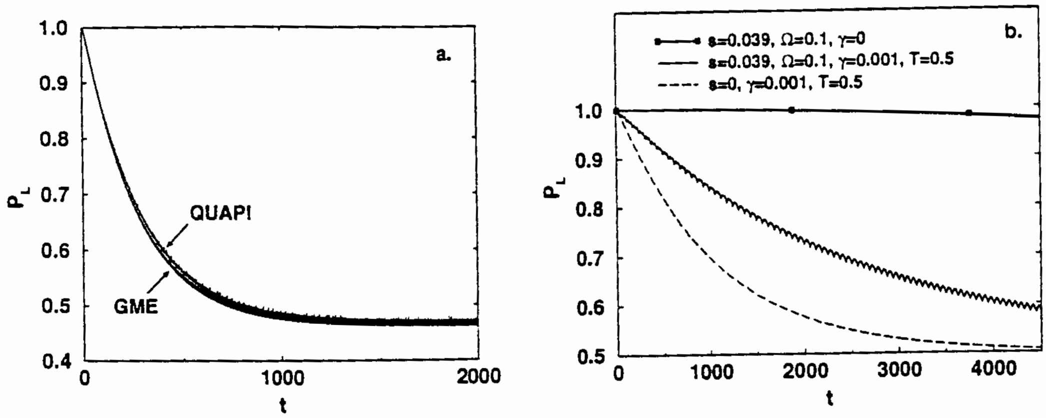

Figure 1: Fig. 1a.: The population $P_L(t)$ of the left well is shown vs. time t. The agreement between analytical (GME) and numerical (QUAPI) results is striking. The parameters are $\Delta_1 = 0.0037, \Delta_2 = 0.12, \overline{\omega}_0 = 0.81, \mathcal{E}_0 = 0.2, s = 0.5, \Omega = 1.0, T = 5.0, \gamma = 0.1, \omega_c = 10.0$. The parameters are measured with respect to the classical oscillation frequency ω_0 in the full DW-potential. Fig. 1b.: Same as Fig. 1a. (only QUAPI results) for the symmetric system $\mathcal{E}_0 = 0$.

a coherent destruction of tunneling (CDT) occurs (see the review in Ref. [5]). Thus, the CDT phenomenon is suppressed by dissipation. The thermalization process, however, is considerably slower than in the undriven case.

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