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Dynamical Hysteresis in Bistable Quantum Systems

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We are studying bistable quantum systems coupled to a thermal environment in the presence of an external controlling force. The impact of thermal and quantum effects on the switching hysteresis is studied by evaluating a time-dependent real-time double path integral with the recently developed iterative tensor multiplication scheme for the *quasiadiabatic path integral propagator method* for temperatures ranging from well above to well below quantum-classical crossover temperature. The switching dynamics and its intimate connection to quantum stochastic resonance is studied here for the first time *without* limitation to a two-state description and/or small external forces. [S0031-9007(97)02709-9]

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Ideal macroscopic switches are bistable devices which are driven from one state (*on*) to another state (*off*) by an external force. Once the switch is in one of the states and the force is released, the switch should stay practically forever in this position. This requirement of stability demands a dissipative (i.e., hysteretic) switching mechanism. In this paper, we are studying the dynamical hysteresis of small switches, operating under conditions where thermal and quantum effects are not negligible or even dominate the switching dynamics.

For the following discussion, we consider an overdamped *classical* bistable system (coordinate x) with an external control force $F(t)$ with $-A < F(t) < A$. The potential minima correspond to the positions *on*/*off*, respectively. Sweeping the potential in the absence of thermal fluctuations adiabatically slowly (sweeping rate $\Omega = 2\pi/\tau_0$) back and forth, the system switches from *on* to *off* when the potential minimum corresponding to *on* disappears, and vice versa. The time interval during which the potential is bistable is denoted by $\tau_{bi} < \tau_0$. In a parametric $x(t)$-$F(t)$ diagram, one observes a hysteresis loop because the system stays in the metastable minimum. At increasing temperatures, the noise-free area of the hysteresis loop is reduced due to thermal hopping out of the metastable minimum, preferably shortly before the deterministic switching point is reached. For a given strength of the fluctuations, the area of the hysteresis loop vanishes for adiabatically slow sweeping, since the system relaxes fast into local equilibria [1,2]. At increasing sweeping rates, the area of the hysteresis loop is increasing until it reaches a maximum, when half the time interval τ_{bi} during which the system is bistable (within one sweeping cycle $\tau_0 = 2\pi/\Omega$) balances the mean dwell-time τ_d in one state. At very large frequencies, the area vanishes due to dynamical cutoff.

Making the switch small so that quantum effects become relevant, a third time scale, namely tunneling time, enters the game. To our knowledge, this case has not been studied before. The questions we are addressing in this paper are: How does the area of the hysteresis loop depend on the frequency of the control force, on the temperature, and the dissipative coupling to the heat bath? How do quantum effects influence the dynamics?

Model.—For a quantitative study, we use as a working model a quantum particle with mass M in a quartic bistable potential

$$V(q) = \frac{M^2\omega_0^4}{64\Delta U}\, q^4 - \frac{M\omega_0^2}{4}\, q^2, \tag{1}$$

with the barrier height ΔU and the curvature at the minima $M\omega_0^2$. The system is coupled with coupling constants c_i to an ensemble of harmonic oscillators with masses m_i and frequencies ω_i (bath). The control force is modeled by a sinusoidal external forcing with variable

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amplitude A and frequency Ω. The Hamiltonian of the compound system is given by

$$\mathbf{H} = \frac{\mathbf{p}^2}{2M} + V(\mathbf{q}) + \mathbf{q}A\sin(\Omega t) + \sum_i \left[\frac{\mathbf{p}_i^2}{2m_i} + \frac{1}{2} m_i \omega_i^2 \left(\mathbf{x_i} - \frac{c_i \mathbf{q}}{m_i \omega_i^2} \right)^2 \right], \quad (2)$$

where $\mathbf{q}, \mathbf{p}$ are the coordinate and momentum operators of the subsystem of interest and $\mathbf{x_i}, \mathbf{p_i}$ those of the bath oscillators (see, e.g., [3]). Eliminating the bath oscillators, the Heisenberg equation of motion for the position operator $\mathbf{q}$ is given by

$$\ddot{\mathbf{q}} + \int_0^t \gamma(t - t')\dot{\mathbf{q}}(t')\, dt' + \frac{1}{M} V'(\mathbf{q}) = \mathbf{\Gamma}(t), \quad (3)$$

where $\mathbf{\Gamma}(t)$ is a time-dependent fluctuational operator containing all the initial values of the bath oscillator position and momentum operators, and where $\gamma(s)$ is the damping kernel

$$\gamma(s) = \frac{1}{M} \sum_j \frac{c_j^2}{m_j \omega_j^2} \cos(\omega_j s). \quad (4)$$

Throughout this paper, we assume an Ohmic bath with a continuous spectral density $J(\omega)$ and an exponential cutoff at ω_c

$$J(\omega) = M\omega \ \mathrm{Re}[\tilde{\gamma}(\omega)] = M\gamma\omega \exp\left(- \frac{\omega}{\omega_c} \right), \quad (5)$$

where $\tilde{\gamma}(\omega)$ is the one-sided Fourier transform of the damping kernel $\gamma(s)$. Analytical approximate solutions in the presence of external periodic driving have been obtained recently by Grifoni *et al.* [4] within linear response theory (for small control forces) and semiclassical approximations around the crossover temperature. Within a two-state approach (spin-Boson-system), which is a good approximation for a slowly operating control force ($\Omega \ll \omega_0$) with a small amplitude A ($Ax_0 \ll \hbar\omega_0$, where x_0 is the characteristic length scale of the system) and for low temperatures ($k_B T \ll \hbar\omega_0$), the periodically driven damped quantum system has been studied in [5–8]. For a strong external driving and for a nonadiabatic sweeping frequency Ω, the two-state description fails, and we are forced to employ numerical methods.

Solution with real-time path integrals.—The reduced density matrix (traced over the bath variables) can be written according to Feynman and Vernon [9] as

$$\rho(q_f, q_f', t) = \int dq_i dq_i' J(q_f, q_f'; q_i, q_i'; t) \rho(q_i, q_i'; 0), \quad (6)$$

with

$$J(q_f, q_f'; q_i, q_i'; t) = \int \mathcal{D}_{\boldsymbol{q}} \mathcal{D}_{\boldsymbol{q}'} \exp\left\{ \frac{i}{\hbar} (S_S[q] - S_S[q']) \right\} \times \mathcal{F}_{\mathbf{FV}}[q, q']. \quad (7)$$

Here, $\mathcal{F}_{FV}[q, q']$ denotes the influence functional for the Ohmic heat bath (see, e.g., [10]), given by

$$\mathcal{F}_{FV}[q, q'] = \exp\left(- \frac{1}{\hbar} \Phi_{FV}[q, q'] \right),$$

$$\Phi_{FV}[q, q'] = \int_0^t dt' \int_0^{t'} dt'' [q(t') - q'(t')] \times [L(t' - t'')q(t'') - L^*(t' - t'')q'(t'')] - \frac{i}{\pi} M\gamma\omega_c \int_0^t dt' [q^2(t') - q'^2(t')], \quad (8)$$

with the autocorrelation function of the heat bath

$$L(t) = \frac{1}{\pi} \int_0^\infty d\omega \times J(\omega) \left[\coth\left(\frac{\hbar\omega}{2k_B T} \right) \cos(\omega t) - i \sin(\omega t) \right], \quad (9)$$

and the classical action functional $S_S[q]$ of the system-variable q along a path $q(t)$. For a numerical evaluation of the double path integral, we have to introduce a discretization in time and space. Conventional discretization in time, i.e., trapezoidal discrete path integrals, requires a fine time slicing to obtain accurate long-time results. Together with a fine enough grid in space, this yields high-dimensional integrals which in general cannot be carried through, even on large scale computers. Monte Carlo sampling, on the other hand, is known to be plagued by phase cancellation problems due to fast oscillating terms. In this paper, we apply an iterative tensor multiplication scheme for the *quasiadiabatic propagator path integral*. This scheme was recently developed by Makri and Makarov [11]. We split the time evolution operator of the compound system symmetrically into a product of an environmental propagator (sum of the Hamiltonians of the bath and the coupling), the system propagator and again an environmental propagator. We truncate the bath memory (this requires careful testing for each set of parameters) and choose a basis given by the eigenfunctions of the position operator on the subspace spanned up by the M lowest energy eigenfunctions of the system Hamiltonian. In our calculations, we have used the five lowest energy eigenfunctions ($M = 5$, yielding five grid points in space), a time slicing $\delta t \approx 10^{-2} t_0$ (see below), and three grid points for the memory kernel of the bath. For the details of this technique, the reader is referred to the original literature [11].

For the numerical calculations, we have introduced dimensionless variables and parameters. The characteristic time scale is $t_0 \equiv 1/\omega_0$, where ω_0 is the angular frequency at the potential minimum. The dimensionless parameters are given by $D = \Delta U/(\hbar\omega_0)$, $\bar{T} = k_B T/(\hbar\omega_0)$, $\bar{\gamma} = \gamma/\omega_0$, $\bar{\Omega} = \Omega/\omega_0$, $\bar{A} = Ax_0/(\hbar\omega_0)$ where $x_0 = \sqrt{\hbar/(M\omega_0)}$. The bars are dropped from

now on for convenience. An important temperature is the crossover temperature T_{co}. In the semiclassical limit ($D \gg \hbar\omega_0$), this temperature is given (in dimensionless units) by $T_{co} = (1/2\pi)(\sqrt{\gamma^2/4 + 1/2} - \gamma/2)$ (see, e.g., [12]). This temperature separates the regime $T > T_{co}$, where thermal hopping dominates over quantum coherence, from the regime $T < T_{co}$, where quantum tunneling dominates over thermal hopping. This is in our case only a rough estimate because we are in the deep quantum regime ($D \approx \hbar\omega_0$) with only one doublet of energy eigenvalues lying below the barrier. The initial density matrix is constructed from the symmetric ground state of the system Hamiltonian. After a transient relaxation time, the reduced density matrix and the mean value $\langle q \rangle(t)$ approach their steady states (independent of initial conditions) being periodic with the period of the external control force. The average area of the hysteresis loop is given by

$$\langle H \rangle = -A\Omega \int_0^{\tau_0} \langle q \rangle(t) \cos(\Omega t)\, dt\,. \qquad (10)$$

Results.—Throughout this paper, we have used a dimensionless barrier height $D = 1.0$ and a cutoff for the bath modes $\omega_c = 7.5$. For studying hysteresis, we choose the amplitude of the control force $A = 0.8$ being large enough to actually *switch* the system between the positions *on*/*off*. The area of the hysteresis loop is shown as a function of the sweeping rate $\Omega = 2\pi/\tau_0$ in Fig. 1. In contrast to the classical switch, the curves (and especially the optimal frequency $\omega_{\max}$) depend only very weakly on the temperature for low temperatures. Similar than in the classical case, we argue that the maximum is determined by the time-scale matching condition $\tau_T = 1/2\tau_{bi} \propto 1/\Omega$, with the decay time τ_T to cross the barrier due to incoherent tunneling [13], which is at low temperatures according to semiclassical results (see, e.g., [12]) indeed only weakly depending on the temperature. Given $\omega_{\max} \approx 0.04$, the time scale matching argument yields for the inverse decay time $1/\tau_T \approx 0.0267$ being larger than the bare tunnel splitting $\Delta \approx 0.0239$. For higher temperatures, we find that $\omega_{\max}$ is shifted to *lower* values, which is surprising since it implies a decrease of the decay rate for increasing temperatures. We do not have a satisfactory explanation for this, but mention that semiclassics is not applicable to our system with a shallow barrier with only the doublet below the barrier.

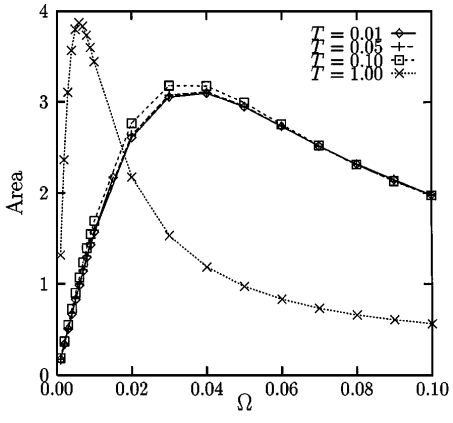

FIG. 1. The area of the hysteresis loop is shown as a function of the sweeping rate Ω at $\gamma = 1.0$, $A = 0.8$, and temperatures $T = 0.01$, 0.05, 0.1, and 1.0.

Quantum stochastic resonance for large amplitudes.—In Fig. 2(a), the area of the hysteresis loop is shown as a function of the temperature. We find a maximum at a finite temperature indicating quantum stochastic resonance (see also [5,7,8]), although the amplitude of the control force is *larger* than the switching threshold. An effect like this has been observed in crystals of high-spin molecules [14], where the magnetization goes through a maximum when the temperature is varied. In classical systems, the area of the hysteresis loop at such large amplitudes A decreases monotonically for increasing temperatures [2]. Furthermore, we observe that for decreasing damping, the maximum of the area of the hysteresis loop disappears in favor of a slow monotonous decrease of the area for increasing temperatures. In the following, we explain the observed behavior.

As described above, in classical systems, the area of the hysteresis loop is increasing for decreasing temperature to reach at zero noise the largest value. In quantum systems, however, as the temperature decreases, tunneling operating on a finite time scale τ_T facilitates escape out of the potential well before the switching point is reached if $\tau_T < \tau_{bi}/2$. The area of the hysteresis loop can therefore—as observed—decrease as the temperature

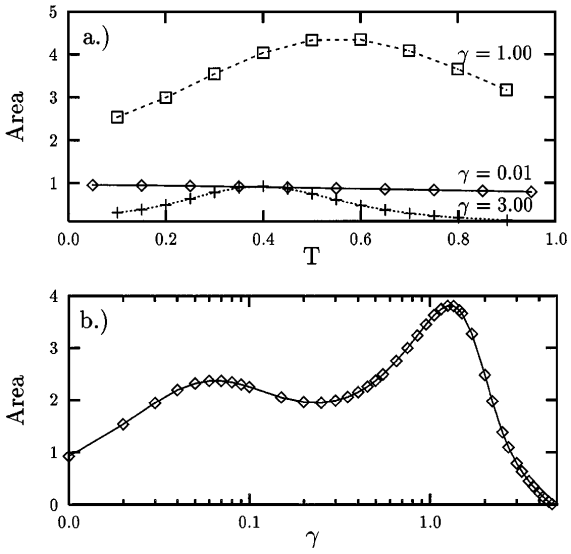

FIG. 2. The area of the hysteresis loop is shown (a) as a function of the temperature at $\Omega = 0.015$ and $A = 0.8$ for various damping constants γ and (b) as a function of the damping $T = 0.3$, $A = 0.8$, and $\Omega = 0.015$.

is decreasing, yielding a maximum of the area, although the amplitude is large enough for deterministic switching.

Why does this effect disappear for weak damping? At weak damping, the crossover temperature T_{co} becomes large and quantum tunneling determines the area of the hysteresis loop up to large temperatures. The curve for $\gamma = 0.01$ in Fig. 2(a) is therefore almost temperature independent without a peak. A similar effect is also observed for small amplitudes where the potential remains bistable throughout the sweeping cycle. In Fig. 3, we show the area of the hysteresis loop in such a case as a function of the temperature (see also [7] for a related effect in a two-state approximation).

Synchronization of tunneling by tuning the damping.— Although the area of the hysteresis loop does not have a maximum as a function of temperature in the tunneling dominated regime, one observes a strong nonmonotonic behavior as a function of the damping. In Fig. 2(b), we show the area of the hysteresis loop at $T = 0.3$ as a function of the damping γ in a tunneling-dominated regime. The values of the damping at which one finds maxima depend on the switching frequency and amplitude. At a given frequency and amplitude, the tunneling process is best synchronized with the control force at those values of the damping where the area shows maxima. The reason for the two maxima is that as a function of damping, the quantum (and classical) transition rate first increases linearly from $\gamma = 0$ for weak damping and decreases with $1/\gamma$ for large damping [12]. Between these two limits, there is a turnover at some intermediate damping value. For a given driving frequency, this yields therefore two optimal values for the damping γ, at which the matching condition for tunneling and driving is fulfilled.

In conclusion, the area of the hysteresis loop of a sweeped bistable quantum switch shows a maximum as a function of the sweeping frequency. For low temperatures, the sweeping frequency at which the maximum occurs is determined by the tunneling time scale. For high temperatures, we find that this synchronization frequency is shifted to lower values. Stochastic resonance is shown to be effective for not too small damping, but also—in contrast to classical systems—for *large* driving amplitudes. The results presented here are of relevance for a number of fields such as chemical physics, quantum wells, and especially for the recent observations of quantum steps and their temperature dependence in the magnetization of high-spin molecules [14].

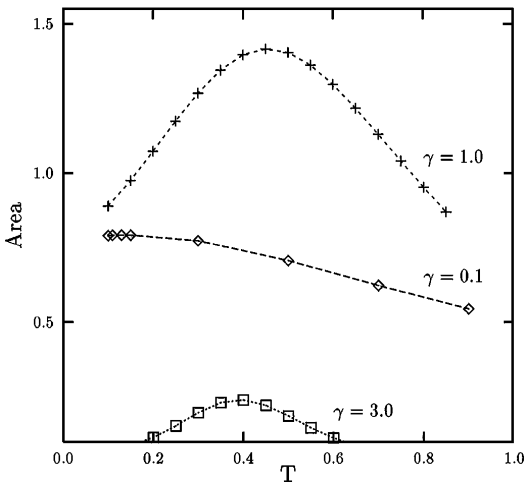

FIG. 3. The area of the hysteresis loop is shown as a function of the temperature for $A = 0.4$ for different damping constants between $\gamma = 0.1$ and $\gamma = 3.0$ ($\Omega = 0.015$).

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