RAPID COMMUNICATIONS

PHYSICAL REVIEW E VOLUME 48, NUMBER 6 DECEMBER 1993

Quantum versus classical control of tunneling

Dmitrii E. Makarov

School of Chemical Sciences, University of Illinois, 505 South Mathews Avenue, Urbana, Illinois 61801

(Received 9 August 1993)

Coherent tunneling can be suppressed or enhanced by exciting a vibrational mode coupled to the tunneling coordinate. The localization in a two-level system (TLS) driven by a periodic force is obtained as a semiclassical limit for such a mode. We derive and test a semiclassical quantization rule that relates the quasienergies of a classically driven TLS with the energy splittings in the model of a TLS coupled to a quantum oscillator. The dissipative stabilization of a quasilocalized state and the possibility of localization by a quasiperiodic force are analyzed from this perspective.

PACS number(s): 05.45.+b, 03.65.−w, 74.50.+r, 73.40.Gk

The effect of an external driving force on tunneling in a symmetric double well has received a great deal of attention in the last few years after Grossmann *et al.* [1] discovered that tunneling can be completely suppressed by a periodic force. This effect is opposite the enhancement of tunneling by a periodic force observed earlier by Lin and Ballentine [2] and later investigated in the framework of the Floquet formalism by Holthaus [3]. Tunneling can be suppressed when the frequency of the driving force is small enough that the force does not induce transitions to higher energy levels of the double well. Under these circumstances the relevant time scale of the problem is much longer than the time scale of the corresponding classical problem, and the coherent suppression of tunneling should be thought of as a purely quantum-mechanical effect which cannot be ascribed to the changes in the classical phase space of the system. Moreover, as was shown in the subsequent studies [4,5] most features of the coherent suppression of tunneling are preserved when the energy spectrum of the double well is truncated to the lowest doublet so that the Hamiltonian reads

$$H=\Delta\sigma_x+V(t)\sigma_z\ , \tag{1}$$

where $V(t)$ is a periodic function; e.g., $V(t)=V_0\cos(\omega t+\phi)$. The states where the eigenvalues of σ_z are $+1$ and -1 correspond to populating the right and left wells, respectively. Based on this two-level system (TLS) approximation, simple analytical theories were applied to show that localization occurs when the ratio of the amplitude of the driving force to its frequency is approximately constant.

Since a real experimental situation is always plagued by noise, in order to realize this tunneling control scheme in practice one needs to know the answer to the question: Is the localization phenomenon robust enough to be observed when the driving force is not exactly periodic and/or there is energy dissipation? Bavli and Metiu [6] studied the possibility of creating a localized state in a double well by gradually turning on a laser pulse. Farelly and Milligan [7] showed that more efficient localization can be achieved by driving the system with two fields with commensurate frequencies. Morillo and Cukier [8] discussed the possibility of controlling a tunneling chemical reaction by an external periodic field. Grossmann *et al.* [9] and Dittrich, Oelschlagel, and Hanggi [10] investigated, numerically, the effect of contaminating the periodic force by noise, and also studied the effect of dissipation using a master-equation approach. In the both cases they found an intriguing phenomenon: localization was stabilized by sufficiently strong dissipation or noise instead of the naturally expected destruction of the localized state. A qualitative explanation of this phenomenon is still lacking. Gomez Llorente and Plata [5], based on perturbative treatment of the driving force, predicted the possibility of localization by a quasiperiodic force and indicated a criterion for such localization.

In this paper we wish to point out that the same localization effect may be observed when the classical periodic external force is replaced by the effect of a *quantum* oscillator. Based on this observation, we study the quantum-mechanical model and show, in particular, that all the results known for the classically driven two-level system may be reproduced as the limit of large quantum numbers for the quantum oscillator. Using a semiclassical quantization scheme, we establish the correspondence between the quasienergies in the classically driven TLS and the energy splittings in the quantum model. We also show the possibility of localization by a quasiperiodic force, which corresponds to a multidimensional oscillator in our quantum-mechanical model, and give a simple explanation of the stabilization effect due to dissipation.

To describe a tunneling system interacting with a vibration, we consider the Hamiltonian

$$H=\Delta\sigma_x+Cx(t)\sigma_z+\frac{p^2}{2m}+V(x)\ . \tag{2}$$

Though the results obtained below apply to a rather general form of potential $V(x)$, our particular example will be the harmonic oscillator, $V(x)=m\omega^2x^2/2$, which corresponds to the sinusoidal driving force in the model (1). For this case, the Hamiltonian (2) represents two coupled parabolic surfaces, $V_{1,2}=m\omega^2(x\pm C/m\omega^2)^2/2-C^2/2m\omega^2$, which cross each other at $x=0$. In the absence of tunneling ($\Delta=0$) each energy level of the harmonic oscillator, $E_n=\hbar\omega(n+\frac{1}{2})$, would be doubly degenerate. Tunneling lifts this degeneracy, splitting each energy level into a doublet. However, at some n the energy level may turn out to be degenerate again or to have a very small splitting. This, in particular, follows from scattering theory for the curve-crossing problem, which shows that the transition probability between the two surfaces is a

 ©1993 The American Physical Society

nonmonotonic function of energy that vanishes at certain energies [11]. The degeneracy means that the system prepared on one of the surfaces never arrives at the other. In other words, tunneling in the two-level system is suppressed completely.

Using nearly degenerate perturbation theory, one obtains the splitting of the nth energy level to be proportional to the overlap of the harmonic-oscillator wave functions for each surface:

$$2\Delta_n = 2\Delta \int dx\, \psi_n(x + C/m\omega^2)\psi_n(x - C/m\omega^2)$$
$$= 2\Delta \exp(-a/2) L_n(a) , \quad (3)$$

where $a = 2C^2/\hbar m\omega^3$ and L_n is the Laguerre polynomial. The localized states correspond to zeros of L_n. We note first that L_0 has no zeros, so that the ground state is always split and the tunneling is never suppressed by an oscillator in the ground state. This remains true for an anharmonic oscillator with an arbitrary potential $V(x)$ because its ground-state wave function has no zeros and the overlap integral never vanishes. However, for any excited state the tunneling may be completely suppressed by an appropriate choice of C. To establish the correspondence between the models (1) and (2), we consider the limit of large quantum numbers, $n \gg 1$. In this case the zeros of $L_n(a)$ are given by $a_j = z_j^2/(4n+2)$, where z_j are the zeros of the Bessel function $J_0(z)$. Assuming in this limit the amplitude of the oscillator x_0 to be semiclassically given by

$$m\omega^2 x_0^2/2 = \hbar\omega(n + \tfrac{1}{2}) , \quad (4)$$

so that the amplitude of the perturbation exerted by this oscillator on the two-level system equals

$$V_0 = Cx_0 , \quad (5)$$

one obtains the localization criterion in the form

$$2V_0/\hbar\omega = z_j , \quad (6)$$

which is identical to the result for a classically driven TLS obtained in [4,5].

In order for the perturbation theory exploited above to be valid, the doublets with different n should not overlap with each other, that is, Δ_n should be smaller than the energy-level spacing of the unperturbed oscillator. Since the overlap integral of the two wave functions in (3) is always less than unity, for the harmonic oscillator the validity of our description is provided by the condition $\Delta \ll \hbar\omega$, which does not depend on the quantum number, and which is also the condition for (nearly) complete localization in the model (1) [5]. For a dissociative potential $V(x)$, though, the energy-level spacing decreases with increasing n, which may invalidate the independent doublet picture at large n.

It is noted that no degeneracy is expected when the coupling between the two-level system is of the form $Cx\sigma_x$, or when there is an asymmetry (a term $\delta\sigma_z$ in the Hamiltonian). This result is in line with the symmetry observations made in [9].

To gain better insight into the connection between the models (1) and (2), we next quantize the system (2) using the semiclassical approximation for the x coordinate. The energy levels can be determined in a standard way as the poles of the spectral function $g(E) = \mathrm{Tr}(H-E)^{-1}$ (see, e.g., [12]), which can be represented as a path integral [13,14]

$$g(E) = i\hbar^{-1} \int_0^\infty d\tau\, e^{iE\tau/\hbar} \int dx(0) \int_{x(0)=x(\tau)} D[x(t)] e^{iS_0[x(t)]/\hbar} \mathrm{Tr}_\sigma \hat{T} \exp\left[-i\int_0^\tau dt\, H_\sigma(x(t))/\hbar\right] , \quad (7)$$

where $\hat{T}$ is the time ordering operator, $S_0[x(t)] = \int_0^\tau dt[m\dot{x}^2/2 - V(x)]$ is the classical action of the x subsystem, and the time-dependent Hamiltonian of the forced two-level system, $H_\sigma(x(t))$, is given by (1) with $V(t) = Cx(t)$. The closed paths $x(t)$ can be periodically continued beyond the segment $0 \le t \le \tau$. The time evolution operator for the system (1) becomes diagonal in the Floquet basis (see, e.g., [15]), which gives

$$\mathrm{Tr}_\sigma \hat{T} \exp\left[-i\int_0^\tau dt\, H_\sigma(x(t))/\hbar\right] = \sum_{n=0,1} e^{-i\varepsilon_n \tau/\hbar} = 2\cos(\varepsilon\tau/\hbar) , \quad (8)$$

where $\varepsilon_{1,2} = \pm\varepsilon$ are the quasienergies of the two-level system. Substituting Eq. (8) into Eq. (7) and then following the standard route of the periodic orbit theory for semiclassical quantization [12], one finds that coupling to a TLS shifts the poles of the semiclassical spectral function for the free oscillator, $g_{\mathrm{osc}}(E)$,

$$g(E) = g_{\mathrm{osc}}(E - \tilde{\varepsilon}(E)) + g_{\mathrm{osc}}(E + \tilde{\varepsilon}(E)) . \quad (9)$$

Here $\pm\tilde{\varepsilon}(E)$ are quasienergies of the driven TLS calculated for the single periodic orbit $x(t)$ of the oscillator with energy E. For small enough Δ_n, we then obtain iteratively that each energy level of the oscillator, E_n, given by the standard Bohr-Sommerfled quantization condition, is split into a doublet,

$$E = E_n \pm \Delta_n, \quad \Delta_n = \tilde{\varepsilon}(E_n) . \quad (10)$$

It should be noted that this quantization rule holds for any form of the coupling between the oscillator and TLS as well as for arbitrary $V(x)$.

The quantization rule (10) answers the question about the relationship between the model (1) and our model (2), demonstrating that interaction between the oscillator and TLS replaces energies of the latter by the corresponding quasienergies. It is well known [4,5] that the localization in the system (1) takes place when the solution to the Schrödinger equation becomes periodic so that the quasienergy doublet is degenerate, this condition being identical to the localization criterion in the semiclassically quantized model (2).

It is interesting to test the formula (10) numerically. In Fig. 1 the numerically exact values of the splittings of energy levels ($n = 0$–16) obtained by direct numerical diag-

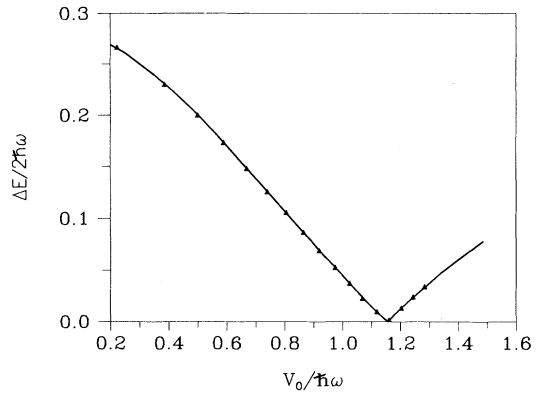

FIG. 1. Half-splitting of energy levels $n=0-16$ for the model (2) (separate points) and quasienergy for the model (1) (solid line) plotted against the parameter $V_0/\hbar\omega$ for $C^2/\hbar m\omega^3=0.05$ and $\omega=3.5\Delta$. The correspondence between the two models is given by Eqs. (4) and (5), or equivalently by the quantization rule (10).

onalization of the Hamiltonian (2) (separate points) are compared with those calculated using Eq. (10), i.e., with the quasienergy splitting of the corresponding classically driven TLS. For the parameters chosen, Eq. (10) turns out to be practically exact even at $n=0$. Localization is achieved at $n=13$, which corresponds to $V_0/\hbar\omega\approx 1.14$, in accord with Eq. (6).

In Fig. 2 the dependence of the numerically found splitting of the vibrational levels $n=0,1,2$ on the parameter $V_0/\hbar\omega$ is presented and compared with the semiclassical result, Eq. (10) (solid line). For $n=0$ (dashed line), Eq. (10) only works at sufficiently small coupling C (i.e., small $V_0/\hbar\omega$); at higher couplings the classically driven system exhibits localization, while the model (2) does not for the reasons explained above. For $n=1,2$ the tunneling splitting vanishes at n values of $V_0/\hbar\omega$, which are to a good approximation described by our quantization formula, Eq. (10). Therefore, the region of the coupling parameter C in which suppression of tunneling is observed is well described by our semiclassical quantization formu-

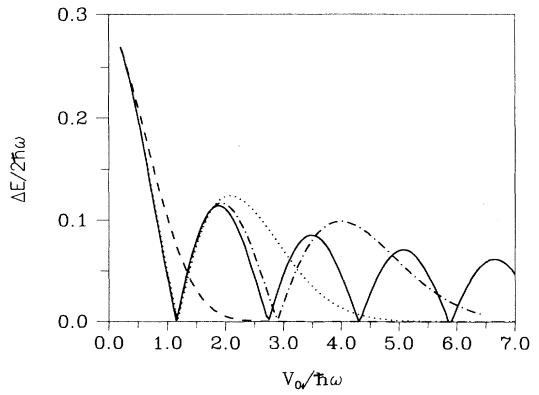

FIG. 2. Half-splitting of energy levels calculated using Eq. (10) (solid line) and by exact numerical diagonalization of the Hamiltonian (2) for $n=0$ (dashed line), $n=1$ (dotted line), and $n=2$ (dashed-dotted line), plotted against the parameter $V_0/\hbar\omega$ for $\omega=3.5\Delta$.

la with the accuracy improving as n increases.

At stronger couplings the splitting decreases monotonically, in contrast to Eq. (10). In this region, increasing the quantum number always increases the splitting. This region is characterized by the overlapping of nonoscillatory tails of the oscillator wave functions in Eq. (3), i.e., by tunneling along the x coordinate through the reorganization energy barrier, V_r ($V_r=C^2/2m\omega^2$ for the harmonic-oscillator model), which obviously has no analogue in the model (1). Therefore, the validity of Eq. (10) necessitates that there be no tunneling along x, which leads to the condition $V_r<\hbar\omega(n+\frac{1}{2})$. For the parameters adopted in Fig. 1 this condition is fulfilled even for $n=0$, which explains why Eq. (10) is successful for small quantum numbers. The analysis of Fig. 2 shows that, indeed, this condition well describes the application scope of Eq. (10).

So far in this paper we have been pursuing the analogy between the two systems, showing that, indeed, there is a full quantum-classical correspondence. In the remaining discussion we will exploit this correspondence to show how the questions about dissipative driven tunneling and about tunneling driven by several periodic fields can be addressed from the point of view of the model (2).

To take dissipation into account, the Hamiltonian (2) is supplemented with a set of harmonic oscillators coupled linearly to the two-level system,

$$H'=H+\sum_j p_j^2/2m_j+m_j\omega_j^2q_j^2/2+C_jq_j\sigma_z\ . \tag{11}$$

We assume that there is an upper cutoff ν_c for the bath spectral density, $J(\nu)=\pi\sum_j C_j^2\delta(\nu-\omega_j)/2m_j\omega_j$, such that $J(\nu)$ vanishes for $\nu\geq\nu_c<\omega$. This condition prohibits the phonon-induced transitions between the doublets of the Hamiltonian H in low orders of perturbation theory. For this reason, one can consider the dynamics within a single doublet independently of all other doublets. In other words, if the system is initially prepared in the nth doublet, we can replace the Hamiltonian H in (11) by an effective TLS, $H=\Delta_n\sigma_x$. The solution to the resulting spin-boson problem has been the subject of a good deal of literature (see, e.g., the review [16] and references therein). Just with an illustrative purpose, we use below the Redfield theory (Bloch equations) approximation, where the expectation $\langle\sigma_z\rangle$ satisfies a Langevin-type equation

$$\langle\ddot{\sigma}_z\rangle+\eta\langle\dot{\sigma}_z\rangle+\Delta_n^2\langle\sigma_z\rangle=0\ . \tag{12}$$

The friction coefficient here is the sum of the rate constants for intradoublet transitions and is equal to $\eta=2\hbar^{-1}J(\Delta_n)\coth(\hbar\Delta_n/2k_BT)$. If $\eta/2<\Delta_n$, $\langle\sigma_z\rangle$ exhibits damped oscillations with the damping coefficient equal to $\eta/2$ and therefore increasing with increasing friction. However, if $\eta/2>\Delta_n$, $\langle\sigma_z\rangle$ decays exponentially with the relaxation rate decreasing as η increases (in the limit $\eta\gg\Delta_n$ this rate is Δ_n^2/η). Formulated in terms of our original problem, this is nothing but the effect of stabilization of the localized state by dissipation, observed in [9,10]. A more careful nonperturbative treatment [16] of the spin-boson Hamiltonian confirms the

qualitative conclusion that dissipation slows down the delocalization. For example, in the Ohmic case, $J(\nu)=\gamma\nu\exp(-\nu/\nu_c)$, the relaxation rate is proportional to $T^{2\alpha-1}$, where $\alpha=2\gamma/\pi\hbar$ is the Kondo parameter. If $\alpha<\frac{1}{2}$, the relaxation rate decreases with increasing temperature, in accord with the result obtained in [10].

The two-state model studied here does not explain the *increase* in the relaxation rate at temperatures above a certain critical temperature, observed in [10]. We believe this increase can only be accounted for if one goes beyond the TLS model for tunneling. In an actual double well, increasing T populates higher energy levels, which enhances the transitions between the wells, thereby more effectively destroying the localized state.

We proceed next to the possibility of localization by a quasiperiodic force. For the model (1) we set $V(t)=\sum_{i=1}^{N}V_i\cos(\omega_i t+\phi_i)$ with incommensurate frequencies ω_i. The Hamiltonian is no longer periodic and the standard periodicity arguments [4] cannot be invoked to explain localization.

For the model (2) one replaces the one-dimensional harmonic oscillator by an N-dimensional oscillator with eigenfrequencies ω_i, so that Eqs. (4) and (5) are satisfied separately for each mode. If we adopt the two-state treatment for each doublet, the formula (3) can be used, save that a single overlap integral will be replaced by the product of overlap integrals for each oscillator. This leads us to a somewhat unexpected conclusion: as long as the doublets are considered well separated, localization is observed whenever the localization condition [Eq. (6) in the case of large quantum numbers] is fulfilled for one of the oscillators, no matter what the parameters of the remaining oscillators are. This conclusion is, however, in agreement with the considerations presented in [5] for the classically driven system. To demonstrate this, we use the perturbative formula derived in that paper. If at $t=0$ the system is entirely in the left state, then the probability of finding it in the right state after time t is

$$p(t)=\Delta^2\left|\int_0^t dt'\exp\left[-2i\int_0^{t'}dt''V(t'')/\hbar\right]\right|^2$$
$$=\Delta^2\left|\int_0^t dt'\exp\left[2i\sum_{i=1}^{N}\frac{V_i}{\hbar\omega_i}\sin(\omega_i t'+\phi_i)\right]\right|^2. \quad (13)$$

According to [5], the two-level system can be localized if the zero Fourier component of the integrand in Eq. (13) vanishes, that is, $\int_{-\infty}^{\infty}dt'\exp[2i\sum_{i=1}^{N}(V_i/\hbar\omega_i)\sin(\omega_i t'+\phi_i)]=0$. Using the Fourier series,

$$e^{i\alpha\sin\theta}=\sum_{k=-\infty}^{\infty}J_k(\alpha)e^{ik\theta}, \quad (14)$$

this integral can be transformed to

$$\int_{-\infty}^{\infty}dt'\exp\left[2i\sum_{i=1}^{N}(V_i/\hbar\omega_i)\sin(\omega_i t'+\phi_i)\right]$$
$$=\sum_{k_1,\ldots,k_N}J_{k_1}\left[2\frac{V_1}{\hbar\omega_1}\right]\cdots J_{k_N}\left[2\frac{V_N}{\hbar\omega_N}\right]$$
$$\times\exp\left[i\sum_{i=1}^{N}k_i\phi_i\right]\delta\left[\sum_{i=1}^{N}k_i\omega_i\right]. \quad (15)$$

Because all frequencies are incommensurate, the only term that survives in this sum is proportional to the product, $\prod_{i=1}^{N}J_0[2(V_i/\hbar\omega_i)]$, which vanishes whenever any pair, V_i and ω_i, satisfies the localization condition (6), in accord with our prediction for the model (2).

As a final remark, we mention that while the possibility of controlling tunneling experimentally by applying a classical driving field is presently being speculated in the literature, the quantum control of tunneling has virtually been realized in the experiments on mode specificity of tunneling [17,18] (see the recent review [14] for a more detailed reference list). By analyzing the laser fluorescence excitation spectra for jet-cooled tropolone [17,18], it was concluded that the excitation of some nonreactive vibrational modes partially or even completely suppresses proton tunneling, while other vibrations promote it. Whether the observed tunneling splitting increases or decreases in a progression of vibrational levels depends on the symmetry of the excited mode. All these experimental findings are clearly in harmony with the results for the model (2) described here.

I am grateful to Professor Victor A. Benderskii for drawing my attention to the mode specificity of tunneling and to Professor Nancy Makri for many insightful discussions.

[1] F. Grossmann, T. Dittrich, P. Jung, and P. Hanggi, Phys. Rev. Lett. **67**, 516 (1991).
[2] W. A. Lin and L. E. Ballentine, Phys. Rev. Lett. **65**, 2927 (1990).
[3] M. Holthaus, Phys. Rev. Lett. **69**, 1596 (1992).
[4] F. Grossmann and P. Hanggi, Europhys. Lett. **18**, 571 (1992).
[5] J. M. Gomez Llorente and J. Plata, Phys. Rev. A **45**, R6958 (1992).
[6] R. Bavli and H. Metiu, Phys. Rev. Lett. **69**, 1986 (1992).
[7] D. Farelly and J. A. Milligan, Phys. Rev. E **47**, R2225 (1993).
[8] M. Morillo and R. I. Cukier, J. Chem. Phys. **98**, 4548 (1993).
[9] F. Grossmann, T. Dittrich, P. Jung, and P. Hanggi, J. Stat. Phys. **70**, 229 (1993).
[10] T. Dittrich, B. Oelschlagel, and P. Hanggi, Europhys. Lett. **22**, 5 (1993).
[11] H. Nakamura, J. Chem. Phys. **87**, 4031 (1987).
[12] R. Rajaraman, Phys. Rep. **21**, 227 (1975).
[13] V. A. Benderskii and D. E. Makarov, Phys. Lett. A **161**, 535 (1992).
[14] V. A. Benderskii, V. I. Goldanskii, and D. E. Makarov, Phys. Rep. **233**, 195 (1993).
[15] J. H. Shirley, Phys. Rev. **138**, B979 (1965).
[16] A. J. Leggett, S. Chakravarty, A. T. Dorsey, M. P. A. Fisher, A. Garg, and M. Zwerger, Rev. Mod. Phys. **59**, 1 (1987).
[17] R. L. Redington, Y. Chen, G. J. Scherer, and R. W. Field, J. Chem. Phys. **88**, 627 (1988).
[18] H. Sekiya, Y. Nagashima, and Y. Nishima, J. Chem. Phys. **92**, 5761 (1990); H. Sekiya, Y. Nagashima, T. Tsuji, Y. Nishimura, A. Mori, and H. Takeshita, J. Phys. Chem. **95**, 10 311 (1991).