Pulse-Shape-Controlled Tunneling in a Laser Field

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It is shown that appropriately shaped smooth pulses of an external oscillating force lead to population transfer in a bistable quantum system on a time scale that can be orders of magnitude shorter than the bare tunneling time. Those pulse shapes for which this phenomenon is achieved can accurately be determined from a tunneling functional Δ_T which is constructed explicitly.

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The problem of tunneling through the barrier of a double well in the presence of an external ac field has received a considerable amount of attention recently [1-4]. It is of great practical importance in several areas of physics, e.g., in solid-state physics in the study of tunneling phenomena in periodically driven semiconductor heterostructures, or in the attempt to control a chemical reaction by laser pulses in chemical physics. However, the majority of previous investigations remained restricted to a constant amplitude of the driving force. In a number of important cases, the physical situation is different: For instance, the study of nonperturbative phenomena in the dynamics of molecules interacting with laser fields may require field strengths that cannot be obtained with a cw laser, and it is indispensable to work with very short laser pulses if it is necessary to beat fast molecular processes such as the redistribution of vibrational energy [5].

This Letter addresses the theoretical description of the tunneling process for a particle in a bistable potential which is subjected to an oscillating force with an amplitude that changes smoothly in time. It will be shown that carefully shaping the pulses opens a new possibility of controlling the tunneling process: Because the quantum dynamics remains very regular even for high driving amplitude, there is a systematic way of designing pulses such that practically complete population transfer in a bistable potential is achieved in times which can be orders of magnitude shorter than the tunneling time of the undriven system, and there is a functional of the pulse shape which accurately describes this phenomenon. This functional, in turn, allows the theoretical prediction of the parameters of optimal pulses. Thus, it can serve as a further tool in the field of laser-assisted molecular control [6-8].

A paradigmatic model for the situation under discussion is provided by a particle in a quartic double-well potential which is driven by a classical oscillating force. Adopting the notation of Grossmann *et al.* [3,4], the Hamiltonian of this system may be expressed in dimensionless units to read

$$H(p,x,t)=\frac{p^2}{2}-\frac{1}{4}x^2+\frac{1}{64D}x^4+\lambda(t)x\sin\omega t\,. \qquad (1)$$

In the absence of the driving force, D determines the number of double-well eigenstates with negative energy. For the present study, the value $D=2.5$ has been employed which means that three pairs of states fall "below" the barrier.

If E_1 and E_2 denote the energies of the two lowest double-well eigenstates $\varphi_1(x)$ and $\varphi_2(x)$, the quantum mechanics of the tunneling process in the undriven double well can be formulated in well-known, simple terms: The linear combinations

$$\varphi_\pm(x):=\frac{1}{\sqrt{2}}[\varphi_1(x)\pm\varphi_2(x)] \qquad (2)$$

are functions "localized" in the left or right well, respectively. If one chooses the initial condition $\psi(x,t=0)=\varphi_+(x)$, the wave function evolves in time as

$$\psi(x,t)=\frac{1}{\sqrt{2}}e^{-iE_1t}[\varphi_1(x)+\varphi_2(x)e^{-i(E_2-E_1)t}]\,, \qquad (3)$$

and after the "tunneling time"

$$\tau_b:=\frac{\pi}{E_2-E_1}\,, \qquad (4)$$

both components of the wave function have acquired a relative factor of −1: A particle initially localized in one of the wells has tunneled into the other.

The task now is to transfer this simple picture to the case where there is a driving force $\lambda(t)x\sin\omega t$. For very strong driving fields, a theoretical analysis in terms of unperturbed double-well eigenstates and their energies obviously is no longer sensible. But for arbitrarily high *constant* driving amplitudes λ, there are Floquet states $u_\alpha^\lambda(x,t)$ and quasienergies $\varepsilon_\alpha^\lambda$ defined by the eigenvalue equation

$$\left[-\frac{1}{2}\frac{\partial^2}{\partial x^2}-\frac{1}{4}x^2+\frac{1}{64D}x^4+\lambda x\sin\omega t-i\partial_t\right]u_\alpha^\lambda(x,t)=\varepsilon_\alpha^\lambda u_\alpha^\lambda(x,t)\,, \qquad (5)$$

with periodic boundary conditions in time: $u_\alpha^\lambda(x,t)=u_\alpha^\lambda(x,t+2\pi/\omega)$. These Floquet states are true stationary states of periodically time-dependent quantum systems [9]. They can be determined by generalized Bohr- Sommerfeld quantization rules [10] if the classical phase space is regular, and they obey an adiabatic principle [11–13]: If the system is initially prepared in a Floquet state and if the parameters of the Hamiltonian are then varied sufficiently slowly, the system remains in the Floquet state connected to the initial one. In other words, if the quasienergies are considered as functions of the parameters, they form quasienergy surfaces on which the Floquet states evolve in a Born-Oppenheimer like fashion.

This statement is by no means trivial: If ε_n^λ is an eigenvalue of Eq. (5), so is $\varepsilon_n^\lambda + m\omega$ for every (positive or negative) integer m. Hence, each of the (infinitely many) eigenstates of the unperturbed double well leads to a quasienergy eigenvalue in the "first Brillouin zone" $-\omega/2 \le \varepsilon < +\omega/2$: The quasienergy spectrum is dense. This fact, in turn, implies that the usual gap condition of the standard adiabatic theorems has to be replaced by a condition on the ineffectiveness of resonances [14].

Such a technically difficult modification of the adiabatic theorem guarantees the adiabatic response to a change of the amplitude λ which takes place on a time scale that is long compared to the period $T = 2\pi/\omega$ of the external oscillating force. Hence, when the field is turned on, the initial double-well eigenstates φ_i evolve into the "connected" Floquet states u_i^λ with phase factors which are determined by the quasienergies, and the initial wave function $\psi(x,t=0) = \varphi_+(x)$ is shifted into

$$\psi(x,t) = \frac{1}{\sqrt{2}}\left[u_1^{\lambda(t)} \exp\left(-i\int_0^t \varepsilon_1^{\lambda(t')}dt'\right) + u_2^{\lambda(t)} \exp\left(-i\int_0^t \varepsilon_2^{\lambda(t')}dt'\right)\right]. \quad (6)$$

Accordingly, under the influence of a smooth pulse $\lambda(t)$ which starts from $\lambda = 0$ at $t = 0$, reaches a maximal strength $\lambda_{\max}$, and decreases back to $\lambda = 0$ after a total pulse time t_p, both parts of the wave function acquire a relative phase

$$\Delta_T(t_p) = \int_0^{t_p} dt\,(\varepsilon_2^{\lambda(t)} - \varepsilon_1^{\lambda(t)}) \quad (7)$$

during the course of the pulse, and if the conditions are such that

$$\Delta_T(t_p) = \pi(2n+1), \quad n = 0, \pm 1, \pm 2, \pm 3, \ldots, \quad (8)$$

the total relative phase factor is -1: The initial wave function $\varphi_+(x)$ has (up to an irrelevant overall phase factor) evolved into $\varphi_-(x)$, and the pulse has forced the particle to tunnel through the barrier. Such a mechanism has a further, possibly attractive feature: Provided the quasienergy difference $\varepsilon_2^\lambda - \varepsilon_1^\lambda$ becomes larger than the original tunnel splitting $E_2 - E_1$, the actual tunneling time τ, which now has to be defined by

$$\Delta_T(\tau) = \pi, \quad (9)$$

can be much shorter than the "bare" tunneling time (4) of the undriven system.

In the following, the viability of this mechanism will be demonstrated for a frequency $\omega = 1.5$. This frequency is of the order of the spacing between the lowest two pairs of double-well eigenstates; the spacing between the lowest two states is much smaller: $D = 2.5$ amounts to $E_2 - E_1 = 1.507 \times 10^{-5}$; the bare tunneling time (4) is $2.085 \times 10^5 = 49766T$ with $T = 2\pi/\omega$. Thus, in the absence of the driving field, the tunneling time is equivalent to roughly 5×10^4 cycles.

Figure 1 shows a plot of the logarithm of the absolute value of the numerically determined difference between the instantaneous quasienergies ε_2^λ and ε_1^λ. In the perturbative regime, the quasienergies approach each other and actually cross, but for high driving strength their difference increases strongly such that for $\lambda = 1$ it is more than 3 orders of magnitude larger than for $\lambda = 0$. Whereas the quasienergy crossing is associated with the "coherent destruction of tunneling" [3,4], the large quasienergy difference in the strong field regime leads to comparatively short tunneling times. Intuitively speaking, such a large quasienergy difference indicates that the Floquet states originating from the lowest two double-well eigenstates acquire admixtures from excited states which have energies closer to the top of the barrier and, therefore, tunnel faster.

The crucial point now is the direct verification of Eq. (8). Two things are needed: On the one hand, the time-dependent Schrödinger equation has to be solved with initial condition $\psi(x,t=0) = \varphi_+(x)$ for given pulses $\lambda(t)$; on the other hand, the corresponding phase integrals (7)

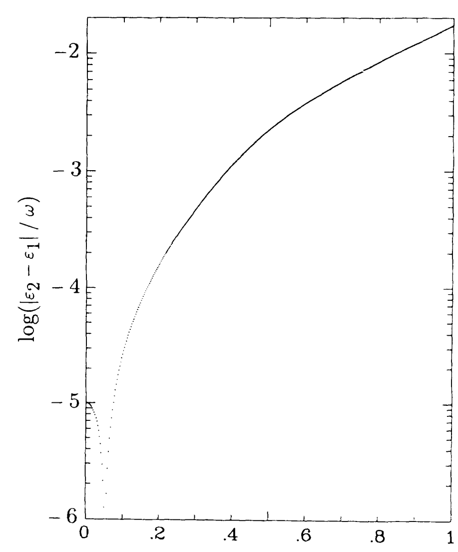

FIG. 1. Logarithm of the absolute value of the quasienergy difference $|\varepsilon_2^\lambda - \varepsilon_1^\lambda|/\omega$ for the system (5) with $D = 2.5$ and $\omega = 1.5$. Note that for $\lambda = 1$ this difference is more than 3 orders of magnitude larger than for $\lambda = 0$; the singularity near $\lambda = 0.05$ indicates a crossing.

have to be evaluated. A convenient choice is

$$\lambda(t)=\lambda_{\max}\sin^2\left(\frac{\pi t}{t_p}\right),\quad 0\le t\le t_p\,, \tag{10}$$

but it is clear that any other smooth pulse would do equally well.

The lower half of Fig. 2 shows the tunneling probability $|\langle\varphi_-|\psi(t_p)\rangle|^2$ for such pulses (10) with a fixed length of $t_p=500T$ as a function of $\lambda_{\max}$, whereas the upper half shows the tunneling phase Δ_T. In view of the simple line of reasoning, the agreement of the numerical result with the adiabatic theory could hardly be any better. Precisely as predicted by Eq. (8), a population transfer $\varphi_+\rightarrow\varphi_-$ occurs when Δ_T becomes equal to an odd multiple of π, although in the present example the pulse length t_p is 2 orders of magnitude shorter than the bare tunneling time τ_b.

But the most important fact is that Eq. (8) can be employed to "design" optimal pulses for laser-assisted tunneling. From the definition (7) it is obvious that Δ_T actually is a functional of the pulse shape $\lambda(t)$, and once the instantaneous quasienergies $\varepsilon_1^\lambda,\varepsilon_2^\lambda$ have been calculated (or determined experimentally) in the range $0\le\lambda\le\lambda_{\max}$, this functional can easily be evaluated for arbitrary smooth pulse shapes. The requirement

$$\Delta_T[\lambda(t)]=\pi \tag{11}$$

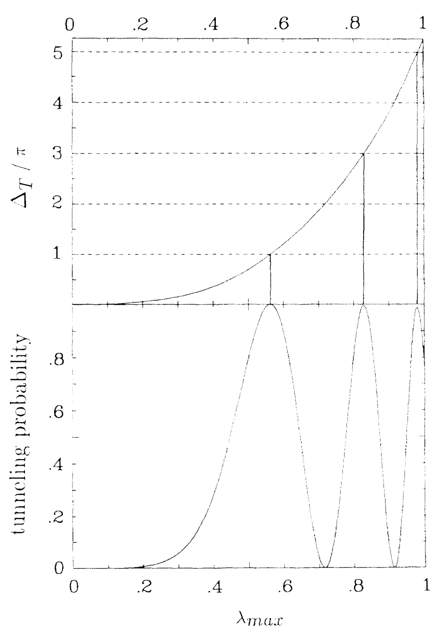

FIG. 2. Upper half: Tunneling functional Δ_T (absolute value, in units of π) evaluated for pulses (10) with $\omega=1.5$ and $t_p=500\times2\pi/\omega$. Predicted maximal amplitudes $\lambda_{\max}$ for complete population transfer are indicated. Lower half: Tunneling probability $|\langle\varphi_-|\psi(t_p)\rangle|^2$ plotted as function of $\lambda_{\max}$, as calculated from numerical solutions of the Schrödinger equation.

then determines those pulses which lead to complete population transfer with a minimal driving strength [i.e., $n=0$ in Eq. (8)]. In fact, pulses characterized by (11) can be interpreted as generalizations of the familiar "π pulses" for two-level systems [15]; the condition (11) replaces the usual "area theorem." From a practical point of view, it is important that this condition is not very restrictive: One can specify a suitable pulse length t_p in the adiabatic regime and work with smooth (e.g., Gaussian) envelopes that can be realized without much effort in an actual experiment.

It is instructive to compare the tunneling functional Δ_T with the functional Δ_{if} which has recently been developed to describe the selective excitation of molecular vibrational states [16,17]. Although both functionals appear very similar, there is a significant difference: In the case of selective excitation, an initially resonant state is split into a superposition of two Floquet states which then evolve separately on their own quasienergy surfaces until they interfere at the end of the laser pulse. In the present case of laser-assisted tunneling in a symmetric double well, there is no "splitting and interference," but both components of the initial wave function $\varphi_+(x)$ evolve independently (to the extent that the adiabatic approximation is valid) and never interact during the whole pulse. (In fact, in the present example both components even evolve in sectors of different symmetry: For fixed λ, the quasienergy operator is invariant under the combined operation $x\rightarrow -x$ *and* $t\rightarrow t+T/2$; and for $E_2-E_1<\omega$ the instantaneous Floquet states u_1^λ and u_2^λ have different parity under this operation. This fact also allows the quasienergies ε_1^λ and ε_2^λ to cross.)

It should be emphasized that the mechanism for pulse-shape-controlled population transfer in a bistable quantum system is quite general. As long as the relevant quasienergies do not show "reactive" avoided crossings where Landau-Zener transitions occur with notable probability [13], the dynamics is simply that of a two-level system in the slowly moving frame of reference which is provided by the instantaneous Floquet states $u_1^{\lambda(t)}$ and $u_2^{\lambda(t)}$.

In conclusion, it has been shown that the tunneling process of a particle in a symmetric bistable potential can efficiently be controlled by the shape of a pulse of an external periodic force. Theoretically speaking, the key point is the manipulation of the relative phase between two adiabatically moving components of the wave function; from a practical point of view it is important that this can be done by pulses with smooth envelopes. Practically complete population transfer is possible on time scales which are orders of magnitude shorter than the bare tunneling time, and the shape of those pulses that achieve this phenomenon can be theoretically predicted by the tunneling functional Δ_T. It would be of interest to investigate the effect of strong laser pulses on the tunneling effect in "engineered" semiconductor double-quantum-well structures [18]. However, a more important application of the mechanism outlined in this Letter may be found in the laser-assisted study of molecular dynamics [5-8,19]: The present results indicate that specifically designed smooth laser pulses can overcome potential barriers in a systematic and controllable manner.

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