Laser-enhanced tunneling through resonant intermediate levels

Stéphane Guérin and Hans-Rudolf Jauslin
Laboratoire de Physique, Université de Bourgogne, CNRS, B.P. 400, F-21001 Dijon, France

(Received 11 July 1996)

We apply the tools of adiabatic Floquet theory to study the control of population transfer and tunneling processes by strong laser pulses. We show how tunneling can be enhanced by intermediate resonant levels by choosing appropriate pulse parameters. We obtain complete tunneling in times that are much shorter than the bare tunneling time or the times obtained without intermediate levels. [S1050-2947(97)02002-7]

PACS number(s): 42.50.Hz, 73.40.Gk, 03.65.−w

I. INTRODUCTION

The availability of intense pulsed laser sources has opened new possibilities in the control of molecular processes. This includes a variety of phenomena such as photodissociation and recombination of molecules, fast selective excitation of molecular vibrational states, and tunneling effects. All these tasks cannot be generally treated by perturbative methods because the high intensity of the field strongly modifies the level structure of the unperturbed system.

In the present paper, we use Floquet methods [1–5], which provide a generalization for periodic or quasiperiodic time-dependent systems of the notion of energy eigenvalues and eigenstates and leads to a generalization of the representation of the time evolution in an eigenfunction expansion. This formalism can also be extended to treat pulse-shaped laser fields by means of the addition of adiabatic principles [6,7]. One important role of these short laser pulses (in subpicosecond time domain) is to excite selectively one molecular vibrational state [8–14]. The main difficulty is the competition with the redistribution of energy over other degrees of freedom. More generally, short pulses avoid damage of the molecule caused by the high intensity. With the Floquet formalism, very interesting results of coherent effects have been obtained concerning (i) the prediction and control of population transfer in a Morse oscillator [12–14] and (ii) the prediction and control (enhancement or suppression) of the tunneling processes in a double-well potential [15–21].

In the present article we combine these two mechanisms, i.e., we enhance the tunneling using a controlled transition. The result is that the tunneling enhancement can be achieved with less intense laser pulses than those used in the previous mechanism. We point out that this lower intensity can be crucial to prevent damaging the molecule or activating processes not included in the model.

We treat the problem by solving numerically the time-dependent Schrödinger equation in a quartic double-well potential driven by a monochromatic laser pulse, using an accurate and fast pseudospectral numerical method [22–26] (with a Lanczos algorithm [27,28]). We choose the parameters to model the inversion of the NH_3 molecule [29].

In Sec. II we recall the method of control of the tunneling effect as proposed by Holthaus [18] and present the results corresponding to the NH_3 parameters. We describe the adiabatic Floquet formalism to interpret the results. We complete and extend several technical aspects of the adiabatic Floquet approach that had not been treated in the literature. In particular we clarify the mechanism of breaking of degeneracy in the case of N-photon resonances, in terms of degenerate perturbation theory applied to Floquet states.

In Sec. III, which contains the main results of this article, we use a pulse of the same length to achieve the same effect but using intermediate resonant or quasiresonant levels. We obtain the same final effect, but using a field intensity that is lower by a factor 40.

II. CONTROL OF TUNNELING DRIVEN BY A PULSE-SHAPED LASER FIELD

A. Tunneling model

The general system that we study is a particle in a quartic double-well potential driven by a smooth pulse-shaped monochromatic laser field. With the usual notations, in dimensionless units and denoting by θ the initial phase of the periodic force of frequency ω, the Hamiltonian is

$$H^{\alpha(t)}(x,p,\theta+\omega t)=H_0(x,p)+H_{\rm int}^{\alpha(t)}(x,\theta+\omega t), \quad (1)$$

where H_0 corresponds to the molecule

$$H_0=\frac{1}{2}p^2-\frac{1}{4}x^2+\frac{1}{64D}x^4 \quad (2)$$

and $H_{\rm int}$ describes the interaction, which we write in the dipole approximation as

$$H_{\rm int}=x\,\alpha(t)\sin(\theta+\omega t). \quad (3)$$

The pulse of duration T_p is taken to be of the form

$$\alpha(t)=\alpha_m\sin^2\left(\frac{\pi t}{T_p}\right), \quad 0\leq t\leq T_p. \quad (4)$$

The particular form of the pulse is not important for the phenomenon, the only relevant property is the slow variation. The particular analytic form (4) is chosen for the simplicity of its treatment. The constant D, which characterizes the height of the unperturbed barrier, is chosen to approximately correspond to the inversion of the NH_3 molecule [29] ($\hbar=1$) (Fig. 1). We denote the unperturbed eigenvalues and normalized eigenfunctions by $\{E_n,\varphi_n\}$ with $E_1<E_2<\cdots$.

1050-2947/97/55(2)/1262(14)/$10.00

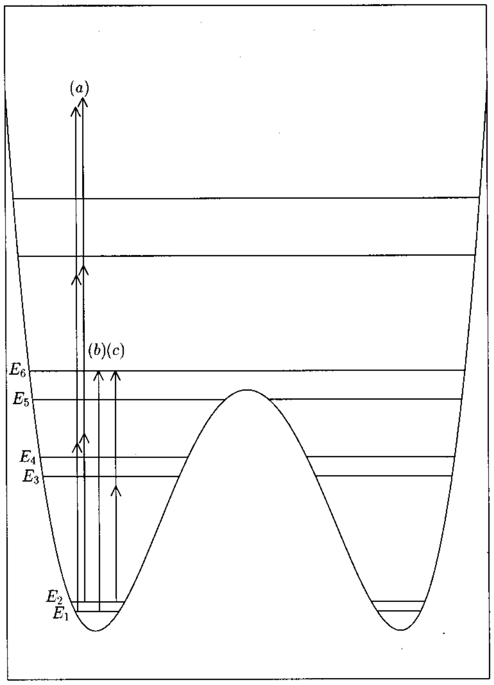

FIG. 1. Unperturbed double-well potential with a schematic indication of the eight lowest values of the unperturbed energies (not to scale, only the qualitative locations under or above the barrier are indicated). The values of the six lower energies are $E_1=-1.517\,04$, $E_2=-1.516\,85$, $E_3=-0.640\,819$, $E_4=-0.626\,364$, $E_5=-0.016\,503$, and $E_6=0.177\,458$. We consider (a) tunneling without resonance in Sec. II, (b) tunneling with a one-photon resonant transition between φ_1 and φ_6 in Sec. III B, and (c) a two-photon resonant transition between φ_2 and φ_6 in Sec. III C.

We characterize the tunneling effect by the crossing of the particle from one side to the other side of the well. In the unperturbed system, the tunneling can be described with the two linear combinations of the states φ_1 and φ_2 of the quasidegenerate lowest doublet (E_1,E_2)

$$\varphi_\pm=\frac{1}{\sqrt{2}}[\varphi_1(x)\pm\varphi_2(x)], \tag{5}$$

corresponding to the localization on the left- or the right-hand side, respectively. Choosing one of these states as the initial condition [e.g., $\phi(t=0)=\varphi_-$], the unperturbed system evolves naturally oscillating between φ_+ and φ_- with the bare frequency $\omega_b=(E_2-E_1)$. This defines the bare tunneling time τ_b

$$(E_2-E_1)\tau_b=(2n+1)\pi, \quad n\in\mathbb{Z}. \tag{6}$$

In the driven regime with an adiabatic pulse, Holthaus [18] established a similar formula, using an extension of the adiabatic theorem for the Floquet states.

B. Floquet formalism

In this section, we briefly summarize the main tools of Floquet theory for periodic Hamiltonians [30,31] of frequency ω and describe the extension needed for the addition of a slow time-dependent modulation $\alpha(t)$. We use units such that $\hbar=1$.

The problem with a time-dependent Hamiltonian can be reduced to an autonomous system by introducing a classical dynamical system $\theta(t)$: $H=H(x,\theta(t))$, where $\theta(t)$ represents the time evolution of the initial phase $\theta(t)=\theta+\omega t$ [32–35]. In the periodic case, $\theta(t)$ is thus a rotation on S^1, the circle of length 2π. We introduce the enlarged space $\mathcal{K}=\mathcal{H}\otimes L_2(S^1)$ where $L_2(S^1)$ is the space of square integrable periodic functions $\xi(\theta)$ and $\mathcal{H}$ the Hilbert space on which H acts. (This representation provides a simple generalization to the quasi-periodic case, i.e., the case with several incommensurable frequencies.)

The quasienergy operator K is defined as the infinitesimal generator of the unitary operator (acting on $\mathcal{K}$)

$$\mathcal{T}_{-t}U(t,t_0;\theta)\mathcal{T}_{t_0}:=e^{-iK(\theta)(t-t_0)}, \tag{7}$$

where the translation operator $\mathcal{T}_t$ acts on $L_2(S^1)$ as $\mathcal{T}_t\xi(\theta)=\xi(\theta(t))$ and $U(t,t_0;\theta)$ denotes the propagator of the Schrödinger equation. From this definition, the quasienergy operator K can be written as

$$K(\theta)=H(\theta)-i\omega\frac{\partial}{\partial\theta}. \tag{8}$$

This allows us to generalize the eigenfunction expansion of the time evolution for a periodic time-dependent system, considering the eigenvectors (*the Floquet states*) and the eigenvalues (*the quasienergies*) of K: if K has a pure point spectrum

$$K\Psi_n(\theta)=\lambda_n\Psi_n(\theta), \tag{9}$$

the eigenfunctions Ψ_n form an orthonormal basis of the enlarged space $\mathcal{K}$ and the time evolution of any initial condition $\phi(t=0)$ can be written as

$$\phi(t)=\mathcal{T}_t\sum_n c_n e^{-i\lambda_n t}\Psi_n(\theta)=\sum_n c_n e^{-i\lambda_n t}\Psi_n(\theta(t)), \tag{10}$$

where the coefficients c_n are determined from the initial condition by the scalar product in $\mathcal{K}$,

$$c_n=\langle\Psi_n|\phi(0)\otimes 1\rangle_{\mathcal{K}}=\int_0^{2\pi}\frac{d\theta}{2\pi}\langle\Psi_n|\phi(0)\rangle_{\mathcal{H}}. \tag{11}$$

[$\phi(0)\otimes 1$ is the initial condition (in $\mathcal{H}$) embedded into the enlarged space $\mathcal{K}$ by multiplying it with the function $f(\theta)=1\ \forall\theta$.] The index in the scalar product brackets indicates the Hilbert space to which it belongs.

For a given Floquet state Ψ_m associated with the quasienergy λ_m, it is easy to see that $\Psi_m e^{-ik\theta}$ is also an eigenfunction with eigenvalue $\lambda_m+k\omega$, for all positive or negative integers k. This implies that the quasienergies appear in families, labeled by the positive integer m, of the form

$$\lambda_n=\varepsilon_m+k\omega, \quad n\equiv(m,k) \quad \forall k\in\mathbb{Z}. \tag{12}$$

The quasienergy spectrum can thus be dense (everywhere or in some parts). The quasienergy families allow an equivalent simpler formulation of the generalized eigenfunction expansion (10), which we write using the labeling (12)

$$\phi(x,t)=\sum_m \tilde{c}_m(\theta)e^{-i\varepsilon_m t}\Psi_m(x,\theta(t)), \qquad (13)$$

with the coefficients

$$\tilde{c}_m(\theta)=\langle\Psi_m|\phi(x,0)\rangle_{\mathcal{H}} \qquad (14)$$

now determined by the scalar product in $\mathcal{H}$. Thus, by choosing for each m family an appropriate k, all the dynamics can be described in any single zone of quasienergy of size ω, e.g., the ''first Brillouin'' zone $-\omega/2\leq\lambda_{m,k}\leq\omega/2\ \forall m$. In this paper, we choose to represent the quasienergies in the zone around the energies E_1 and E_2 corresponding to the two initial unperturbed states φ_1 and φ_2.

One can treat the case of a periodic field modulated by a slowly varying envelope $\alpha(t)$ (e.g., a pulse) by combining the Floquet formalism with adiabatic techniques [7,36]. For the Hamiltonian $H^{\alpha(t)}(\theta(t))$ with a slowly varying parameter α, we define instantaneous quasienergy states at each time t calculated with an instantaneous quasienergy operator $K^{\alpha(t)}(\theta)$.

Extending the usual adiabatic theorem to these instantaneous Floquet states, one can formulate, under suitable conditions, the following adiabatic conjecture, in terms of the eigenfunctions [36]: If at time t_0 the system is an instantaneous Floquet state $\phi(t_0)=\Psi_n^{\alpha(t_0)}(\theta(t_0))$, in the adiabatic limit ($T_p\to\infty$) the time evolution determined by the Schrödinger equation $i(\partial/\partial t)\phi=H^{\alpha(t)}(\theta(t))\phi$ is such that $\phi(t)$ stays for all t in an instantaneous Floquet eigenstate:

$$\phi(t)=e^{i\delta_n(t)}\Psi_n^{\alpha(t)}(\theta(t)) \quad (\delta_n\in\mathbb{R}). \qquad (15)$$

The phase $\delta_n(t)$ is the superposition of the dynamical phase and Berry's geometric phase [37–39]. In our case, only one parameter [the amplitude $\alpha(t)$ of the field] is varied adiabatically to form a closed loop between the instants $t=0$ and $t=T_p$ in the parameter space. As a consequence, the geometric phase is zero at the end of the pulse and the phase $\delta_n(T_p)$ is then just the dynamical phase

$$\phi(T_p)=\exp\left(-i\int_0^{T_p}ds\lambda_n^{\alpha(s)}\right)\Psi_n^{\alpha(T_p)}(\theta(T_p)). \qquad (16)$$

The precise conditions for the validity of the adiabatic conjecture have not yet been proven in the general case, but it is supported by a fair amount of numerical studies. They mainly depend on the separation between the instantaneous quasienergy levels. The difficulty comes from the fact that the spectrum can be dense (a problem of small denominators). Without any relevant crossing or avoided crossing of quasienergy levels, this adiabatic conjecture has been proven for finite N-level models, where the spectrum of K is discrete with well-separated eigenvalues [40]. If there are crossings or avoided crossings, we have to add restrictions. We can apply a generalized version, considering no longer a single Floquet state but the Floquet subspace generated by the states involved in the crossing.

The quasienergies and Floquet states give a natural setting to study resonances and transitions. In the (ω,α) plane (denoting by α the amplitude of the ω-periodic force), a resonance corresponds to the crossing (degeneracy) of two (or more) Floquet eigenvalues at $\alpha=0$, giving rise to N-photon transitions in a perturbative regime. If the frequency is not resonant but close to the resonant one, we observe an avoided crossing with a minimal distance at some value $(\omega,\alpha_{\rm ac})$. In the adiabatic regime, the transitions between levels are essentially determined by the avoided crossings [6,43]. They can be treated by Landau-Zener analysis [7,41].

If there is some spatial symmetry in the problem (e.g., the quartic double well), the analysis of quasienergy crossings and Landau-Zener transitions has to be done for each parity class, since states of different symmetry are not coupled by the dipole interaction (3). Thus the quasienergy surfaces $\lambda(\omega,\alpha)$ of different parity class can cross without any consequences.

In this paper, we consider transitions induced by exact resonances or by near resonances. We give a precise theoretical analysis of the resonant transitions, applying stationary perturbation theory to the Floquet states. The quasiresonant case will be treated with the tools developed to interpret the transitions involving avoided crossings [6].

We remark that the preceding ideas on the combination of the Floquet picture and adiabatic techniques can be extended to more general time-dependent forces [42], in particular for quasiperiodic forces (i.e., two or more incommensurate frequencies): if we assume that the force contains N incommensurable frequencies, the set of the initial phases associated with the frequencies $\boldsymbol{\omega}=(\omega_1,\ldots,\omega_N)$ is represented by a vector $\boldsymbol{\theta}$. The time-evolved phase $\boldsymbol{\theta}(t)=(\theta_1+\omega_1 t,\ldots,\theta_N+\omega_N t)$ is a classical flow on a N-dimensional torus $\Omega=S^1\times\cdots\times S^1$. All the previous formulas are thus generalized by substituting ω and θ by $\boldsymbol{\omega}$ and $\boldsymbol{\theta}$, respectively, and $\boldsymbol{\omega}\cdot\partial/\partial\boldsymbol{\theta}=\omega_1\partial/\partial\theta_1+\cdots+\omega_N\partial/\partial\theta_N$.

C. Control of tunneling by laser without resonance

In this section we discuss the nonresonant mechanism of tunneling control treated in [18]. We set up the main formulas that we will use for the extension to the resonant mechanism. We also discuss the breakdown of the adiabatic regime due to the presence of avoided crossings. This effect, which in this case degrades the tunneling, is of the same nature as the mechanism of resonant population transfer that we will have to control in the resonant tunneling mechanism that we propose in Sec. III.

In the quartic double-well potential, the quasienergy operator is

$$K^{\alpha(t)}(\theta)=H_0+x\alpha(t)\sin\theta-i\omega\frac{\partial}{\partial\theta}. \qquad (17)$$

Applying the adiabatic conjecture to the initial condition $\phi(0)=\varphi_-$, we obtain the time evolution at the end of the pulse using Eq. (16),

$$\phi(T_p)=\frac{1}{\sqrt{2}}\left[\exp\left(-i\int_0^{T_p}\lambda_{1,0}^{\alpha(s)}ds\right)\Psi_1^{\alpha(T_p)}(\theta(T_p))\right.$$
$$\left.-\exp\left(-i\int_0^{T_p}\lambda_{2,0}^{\alpha(s)}ds\right)\Psi_2^{\alpha(T_p)}(\theta(T_p))\right]. \quad (18)$$

At the beginning and the end of the pulse, the Floquet states Ψ_1 and Ψ_2 are continuously connected $[\alpha(t)\to 0]$ with the unperturbed eigenfunctions φ_1 and φ_2. The tunneling probability is

$$|\langle\varphi_+|\phi(T_p)\rangle|^2=\sin^2\left[\frac{1}{2}\int_0^{T_p}ds(\lambda_{2,0}^{\alpha(s)}-\lambda_{1,0}^{\alpha(s)})\right] \quad (19)$$

which gives the complete tunneling condition

$$\int_0^{T_p}ds(\lambda_{2,0}^{\alpha(s)}-\lambda_{1,0}^{\alpha(s)})=(2n+1)\pi, \quad n\in\mathbb{Z}. \quad (20)$$

It is like the bare tunneling condition (6), but the unperturbed eigenvalues are replaced by the quasienergies with the appropriate integration over the pulse (note that $\lambda_{m,0}^{\alpha}\xrightarrow[\alpha\to 0]{}E_m$

To avoid any resonance at moderate field amplitudes, we choose the frequency $\omega=0.975$ (there are no states with energies close to $E_j+k\omega,\ j=1,2$, for moderate k). To verify the validity of the tunneling condition (20), we first solve the time-dependent Schrödinger equation numerically for a pulse of fixed length $T_p=741.8$ (this pulse contains about 115 oscillations). We then plot the probability for the system to be at the other side of the well [Fig. 3(b)].

In order to analyze the numerical result within the adiabatic Floquet framework, we solve the instantaneous eigenvalue problem (9) numerically, for α taken in the interval $0\leqslant\alpha\leqslant\alpha_m$. We use a basis of eigenfunctions of H_0 for the spatial part and Fourier series for the θ part [7] [see Fig. 2(b)]. We then calculate and plot the integral of Eq. (20) [Fig. 3(a)]. We find good agreement with the complete tunneling condition (20) for $n=0,1$ and for the complete destruction of tunneling [Eq. (20) for $n=1/2,3/2$]. We remark that complete destruction of tunneling refers only to the state at the end of the pulse; during the pulse evolution, the wave packet does indeed have a non-negligible projection on φ_+.

With the parameter $D=2.5$, this result has been obtained by Holthaus [18]. He pointed out that the tunneling time obtained in this way for $n=0$ can be shorter than the bare tunneling because the difference $\lambda_{2,0}^{\alpha}-\lambda_{1,0}^{\alpha}$ during the pulse becomes larger than the unperturbed difference E_2-E_1.

We obtain for the lowest field amplitude ($n=0$, i.e., $\alpha_m=0.332$) complete tunneling for a pulse length $T_p=741.8$, which is 20 times smaller than the bare tunneling time $\tau_b\approx 16574$. We remark that this ratio is not as good as the one in [18] because we use a smaller parameter $D=2$ (instead of $D=2.5$) that involves a larger separation of the doublet (E_1,E_2). We are moreover limited by a minimum pulse length in order to remain in adiabatic conditions.

The plot of Fig. 2(a) shows the difference $\lambda_{2,0}^{\alpha}-\lambda_{1,0}^{\alpha}$. We note the presence of two zeros at small values of α. Each of them corresponds to a very long tunneling time for the values of α in their neighborhood. This phenomenon was observed in numerical simulations by Grossmann *et al.*, who called it a *coherent destruction of tunneling* [19].

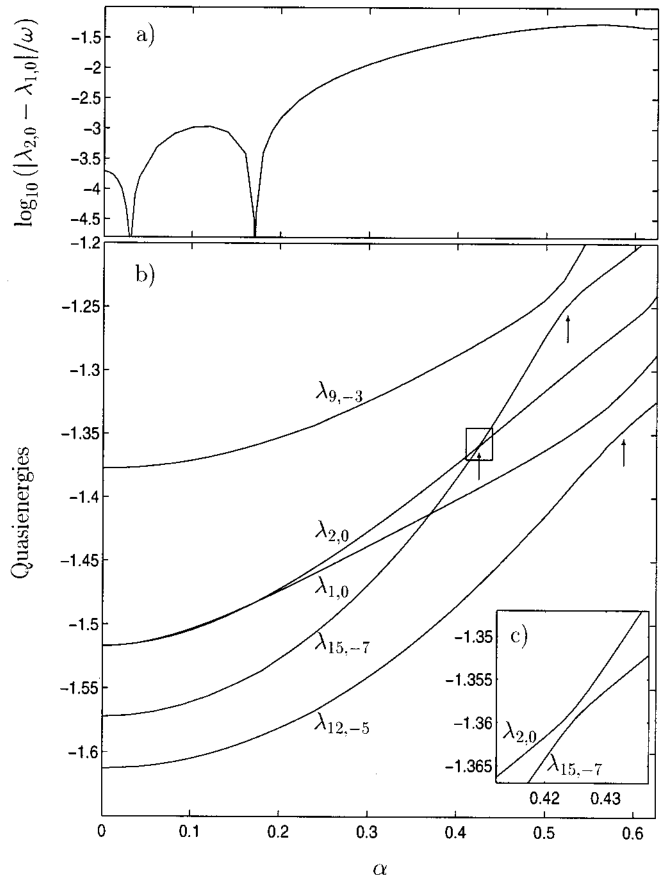

FIG. 2. (a) Logarithm of $|\lambda_{2,0}^{\alpha}-\lambda_{1,0}^{\alpha}|/\omega$ for the system involving no resonance ($\omega=0.975$). (b) Quasienergy diagram close to the unperturbed energies E_1 and E_2 (only the relevant quasienergies of our problem are plotted in this zone). We notice (i) two crossings between $\lambda_{1,0}^{\alpha}$ and $\lambda_{2,0}^{\alpha}$ involving the two singularities in (a), (ii) the three avoided crossings pointed by arrows: $\lambda_{2,0}^{\alpha}$ with $\lambda_{15,-7}^{\alpha}$ close to $\alpha=0.425$, indicating a seven-photon transition between the two unperturbed levels φ_2 and φ_{15}; $\lambda_{1,0}^{\alpha}$ with $\lambda_{12,-5}^{\alpha}$ close to $\alpha=0.58$, indicating a five-photon transition between the two unperturbed levels φ_1 and φ_{12}; and $\lambda_{2,0}^{\alpha}$ with $\lambda_{9,-3}^{\alpha}$. (c) The narrow avoided crossing of $\lambda_{2,0}^{\alpha}$ with $\lambda_{15,-7}^{\alpha}$ is magnified.

In Fig. 3(c) we have plotted the projection on the unperturbed states orthogonal to φ_1 and φ_2 after the end of the pulse, calculated from the numerical solution of the Schrödinger equation. This measures the deviation from adiabatic behavior. As expected, the validity gets worse for stronger fields.

We made the following somewhat unexpected observations. The deviation from adiabatic behavior seems to appear with two separated thresholds, but in between the system seems to return to a quasiperfect adiabaticity.

This can be qualitatively interpreted by the three avoided crossings appearing in the quasienergy plot [pointed by arrows in Fig. 2(b)]: the first one (for $\alpha\approx 0.425$) involves the quasienergies $\lambda_{2,0}^{\alpha}$ with $\lambda_{15,-7}^{\alpha}$ (seven-photon resonance) and the second and the third involve, respectively, $\lambda_{2,0}^{\alpha}$ with $\lambda_{9,-3}^{\alpha}$ and $\lambda_{1,0}^{\alpha}$ with $\lambda_{12,-5}^{\alpha}$ (five-photon resonance). This first avoided crossing induces transitions between the instanta-

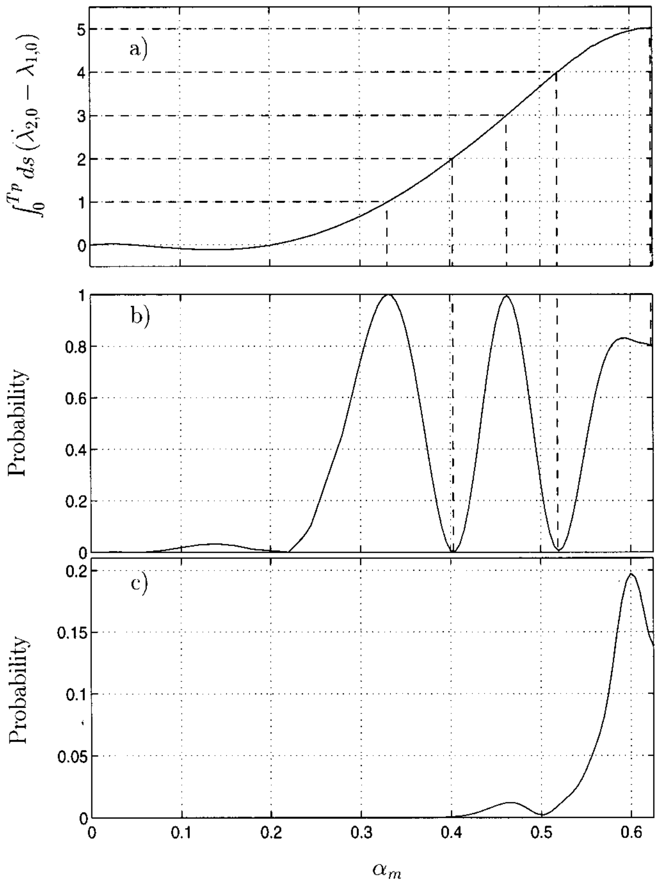

FIG. 3. (a) $\int_0^{Tp} ds(\lambda_{2,0}^{\alpha}-\lambda_{1,0}^{\alpha})$ calculated for each α_m with the values of the quasienergies of the diagram represented in Fig. 2(b) (plotted in units of π). (b) Tunneling probability $|\langle\varphi_+|\phi(T_p)\rangle|^2$ at the end of the pulse calculated with the numerical solution of the Schrödinger equation. (c) Projection of the numerical solution $\phi(T_p)$ for each α_m on the unperturbed states orthogonal to φ_1 and φ_2. We notice large values of the projection for $\alpha_m > 0.55$, which corresponds to the avoided crossings shown in Fig. 2(b) and implies large corrections to the adiabatic behavior of Eq. (18).

neous Floquet states $\Psi_{2,0}^{\alpha}$ and $\Psi_{15,-7}^{\alpha}$ For one passage through the avoided crossing, the transition probability depends on the speed of the evolution along the Floquet state compared to the minimal distance of the avoided crossing as in the usual Landau-Zener transitions [7]. Because of the shape of the pulse, the instantaneous states pass through the avoided crossing twice and the final transition probability depends also on the maximal amplitude that is reached beyond the avoided crossing. If we fix all the parameters except the maximal amplitude α_m and if we suppose that the pulse is sufficiently slow, the transition probability as a function of α_m can present oscillations between zero and a maximal value β (which depends on the other parameters). If we denote $P_{2\to 15}$ the transition probability from φ_2 to φ_{15} at the end of the pulse, in [6] it is shown that

$$P_{2\to 15}=\beta\sin^2\left[\int_{t_c}^{T_p-t_c} ds(\lambda_{15,-7}^{\alpha(s)}-\lambda_{2,0}^{\alpha(s)})\right], \qquad (21)$$

where t_c is the time of the avoided crossing. We can interpret the first small oscillation on Fig. 3(c) with the formula (21). The smallness of the maximal value β is due to the fact that the avoided crossing involving $\lambda_{2,0}$ with $\lambda_{15,-7}$ is very narrow compared to the speed of evolution along $\Psi_{2,0}^{\alpha}$: the evolution close to it is effectively highly nonadiabatic and most of the population jumps across (twice) [41]. This leads to a small component on φ_{15} at the end of the pulse. The return to adiabatic behavior (for $\alpha_m \approx 0.5$) is predicted by Eq. (21): we indeed calculate that the integral of this equation is equal to π for $\alpha_m \approx 0.51$, giving $P_{2\to 15}\approx 0$. After this value the two next avoided crossing appear and induce other transitions.

This mechanism will be used in a similar way to obtain tunneling with adiabatic conditions even in a resonance situation. In that case, we have an exact crossing near $\alpha_m=0$ (degeneracy of the Floquet states due to the exact resonance). The tunneling effect will be achieved if we can control the transition described above. We will then see that with this mechanism we can increase the difference $\lambda_{2,0}^{\alpha}-\lambda_{1,0}^{\alpha}$ even more using a resonance with intermediate levels.

III. ENHANCED TUNNELING BY A PULSE-SHAPED LASER FIELD WITH INTERMEDIATE RESONANT STATE

The mechanism discussed by Holthaus [18] involves only the two levels of the tunneling doublet. The frequency of the laser had indeed been chosen in order to avoid resonances with other unperturbed levels. Here we will study a different mechanism to control tunneling, which involves resonances with others levels. With the help of this effect, we wish to enhance tunneling, i.e., obtain controlled tunneling by a pulse-shaped laser field of length $T_p \ll \tau_b$ and with as weak an intensity as possible, to avoid damage of the system.

We consider two possible resonances: one-photon and two-photon population inversions. We first describe in Sec. III A how to control population inversion.

A. Control of the resonant population transfer in a nonperturbative regime

In this section we present the general results of the population transfer under a strong pulse-shaped laser that is resonant with two unperturbed levels of the system. We consider a general system with eigenvalues and normalized eigenfunctions denoted $\{E_1,\varphi_1\},\{E_2,\varphi_2\},\ldots$ and a N-photon resonance between two isolated levels $\{\varphi_i,E_i\}$ and $\{\varphi_f,E_f\}$.

We will calculate the time evolution of an initial condition $\phi(t=0)=\kappa\varphi_i$ with $|\kappa|\leq 1$ $(\kappa\in\mathbb{C})$ under a pulse-shaped field (4). (The reason to include the factor κ will become clear later on.) We will consider the general case of an N-photon resonance and then give more precise results for the cases of one-photon and two-photon resonances.

1. N-photon exact resonances

The frequency of the laser is chosen to produce a resonance between the two unperturbed levels $\{\varphi_i,E_i\}$ and $\{\varphi_f,E_f\}$:

$$\omega=\frac{E_f-E_i}{N}. \qquad (22)$$

For each time t, these two states give rise to two Floquet families $\{\lambda_{i,k}^{\alpha(t)},\lambda_{f,k}^{\alpha(t)}\}$ $(k\in\mathbb{Z})$. If the quasienergy zone $0\leq\lambda_{m,k}^{\alpha(t)}\leq\omega$ is considered and, without loss of generality, we suppose $0\leq E_i\leq\omega$, the relevant eigenvalues are then $\{\lambda_{i,0}^{\alpha(t)},\lambda_{f,-N}^{\alpha(t)}\}$, where $\lambda_{i,0}^{\alpha=0}=E_i$ and $\lambda_{f,-N}^{\alpha=0}=E_f-N\omega$. At the beginning and at the end of the pulse ($t=0$ and $t=T_p$), we have $\alpha=0$ and the two quasienergies $\lambda_{i,0}^{\alpha}$ and $\lambda_{f,-N}^{\alpha}$ are degenerate ($\lambda_{i,0}^{0}=\lambda_{f,-N}^{0}\equiv E_i$): the eigenfunctions are any linear combination of φ_i and $\varphi_f e^{-iN\theta}$.

The perturbation breaks up the degeneracy. We denote the zeroth-order basis of linear combinations adapted to the degeneracy breaking as

$$\Psi_a^{\alpha=0}(\theta)=a_i\varphi_i+a_f\varphi_f e^{-iN\theta}, \tag{23a}$$

$$\Psi_b^{\alpha=0}(\theta)=-a_f^*\varphi_i+a_i^*\varphi_f e^{-iN\theta}, \tag{23b}$$

with $|a_i|^2+|a_f|^2=1$. The initial degeneracy is lifted for $\alpha\neq0$: the two states split into two branches corresponding to the Floquet states $\Psi_a^{\alpha(t)}$ and $\Psi_b^{\alpha(t)}$ associated with the quasienergies that we denote $\lambda_a^{\alpha(t)}$ and $\lambda_b^{\alpha(t)}$.

We make the working hypothesis that the adiabatic evolution (15) connects these instantaneous Floquet states by continuity to the initial degenerate states $\Psi_a^{\alpha=0}$ and $\Psi_b^{\alpha=0}$ without transitions between the two Floquet branches $\Psi_a^{\alpha(t)}$ and $\Psi_b^{\alpha(t)}$, in spite of the fact that, at the beginning and at the end of the pulse, they are arbitrarily close to each other. This conjecture is strongly supported by the numerical evidence and can be expected to be justified by perturbative arguments, since the intensity is small when the two Floquet branches are close.

Inverting Eqs. (23), we obtain

$$\varphi_i=a_i^*\Psi_a^0-a_f\Psi_b^0, \tag{24a}$$

$$\varphi_f=e^{iN\theta}(a_f^*\Psi_a^0+a_i\Psi_b^0). \tag{24b}$$

The adiabatic time evolution of the initial condition $\phi(0)=\kappa\varphi_i$ is thus

$$\phi(t)=\kappa a_i^* e^{i\delta_a(t)}\Psi_a^{\alpha(t)}(\theta+\omega t)-\kappa a_f^* e^{i\delta_b(t)}\Psi_b^{\alpha(t)}(\theta+\omega t). \tag{25}$$

At the end of the pulse $t=T_p$, the degeneracy of the two Floquet states appears again and we can write using (23)

$$\begin{aligned}\phi(T_p)=&\kappa a_i^*\exp\left(-i\int_0^{T_p}ds\lambda_a\right)\Psi_a^0(\theta+\omega T_p)-\kappa a_f\\&\times\exp\left(-i\int_0^{T_p}ds\lambda_b\right)\Psi_b^0(\theta+\omega T_p)\\=&\exp\left(-i\int_0^{T_p}ds\lambda_a\right)\Bigg\{\Bigg[|a_i|^2+|a_f|^2\\&\times\exp\left(i\int_0^{T_p}ds(\lambda_a-\lambda_b)\right)\Bigg]\kappa\varphi_i+a_i^*a_f e^{-iN(\theta+\omega T_p)}\\&\times\left[1-\exp\left(i\int_0^{T_p}ds(\lambda_a-\lambda_b)\right)\right]\kappa\varphi_f\Bigg\},\end{aligned} \tag{26}$$

i.e., the time evolution can be again developed in the two functions $\{\varphi_i,\varphi_f\}$ at the end of the pulse:

$$\phi(T_p)=c_i(T_p)\varphi_i+c_f(T_p)\varphi_f. \tag{27}$$

The inversion from φ_i to φ_f at the end of the pulse is characterized by the probability $P_{i\to f}(T_p)\equiv|c_f(T_p)|^2$, which is obtained from Eq. (26):

$$P_{i\to f}(T_p)=4|\kappa a_i a_f|^2\sin^2\left[\frac{1}{2}\int_0^{T_p}ds(\lambda_a^{\alpha(s)}-\lambda_b^{\alpha(s)})\right]. \tag{28}$$

Note that this construction is not simply an approximation by a two-level model, since during the pulse evolution the wave packet (25) has components in other states beside $\{\varphi_i,\varphi_f\}$.

The expression (28) gives oscillations between the two unperturbed levels φ_i and φ_f (the maximal occupation probability on φ_f is not necessarily $|\kappa|^2$) for a fixed duration of the pulse $t=T_p$, as a function of the maximal amplitude α_m. This is due to the fact that the quasi-energy difference $\lambda_a^\alpha-\lambda_b^\alpha$ grows with α (there are no avoided crossings involving these quasienergies in the considered range of α). These oscillations will be characterized more precisely in the following sections for the cases of one- and two-photon resonances. They can be interpreted as follows. The initial resonant state φ_i splits into two orthonormal linear combinations $\Psi_a^{\alpha(0)=0}$ and $\Psi_b^{\alpha(0)=0}$ of the two Floquet states φ_i and $\varphi_f e^{-iN\theta}$. During the laser pulse, the components evolve adiabatically independently of each other, following instantaneous Floquet states and acquiring a phase. For $t=T_p$, the two resonant Floquet states interfere again and the difference of their two phases determines the occupation probability on the states φ_i and φ_f.

One can then conclude that (i) the inversion from φ_i to φ_f is complete if and only if the following two conditions are fulfilled:

$$|a_i|^2=|a_f|^2=\frac{1}{2}, \tag{29}$$

$$\int_0^{T_p}ds(\lambda_a^{\alpha(s)}-\lambda_b^{\alpha(s)})=(2n+1)\pi,\quad n\in\mathbb{Z}, \tag{30}$$

and (ii) the inversion from φ_i to φ_f is zero (complete Rabi cycle) if and only if the condition

$$\int_0^{T_p}ds(\lambda_a^{\alpha(s)}-\lambda_b^{\alpha(s)})=2n\pi,\quad n\in\mathbb{Z} \tag{31}$$

is fulfilled. In this latter case, inserting condition (31) into the time evolution (26), one obtains

$$\phi(T_p)=\exp\left(-i\int_0^{T_p}ds\lambda_a\right)\left[|a_i|^2+|a_f|^2\times\exp\left(-i\int_0^{T_p}ds(\lambda_b-\lambda_a)\right)\right]\kappa\varphi_i=\exp\left(-i\int_0^{T_p}ds\lambda_a\right)\kappa\varphi_i. \quad (32)$$

This last result has an important interpretation: in case of the complete Rabi cycle between two resonant states in adiabatic conditions (no avoided crossing involving these two states), the time evolution of the initial state leads back to this initial state with the phase given by the adiabatic conjecture, as if no resonance were involved.

Thus, in order to enhance the tunneling, we must necessarily fulfill the two conditions: the complete Rabi cycle from one of the two initial unperturbed states of the quartic double well through another resonant state and the tunneling condition. Another obvious condition is the hypothesis that all the other states are far enough and do not affect the adiabatic evolution of the two states involved in the resonance (nor the third state involved in the tunneling).

Breuer *et al.* [13,14] determined the coefficients $|a_i|=|a_f|=1/\sqrt{2}$ of the linear combinations (23) for a one-photon resonance within the rotating-wave approximation for a two-level system. In order to calculate analytically the coefficients a_i and a_f in a more general case, we apply the stationary perturbation method to the degenerate Floquet states, which gives the linear combinations adapted to the lifting of degeneracy for a small value of α. We will show the result for a two-photon resonance. We will expose in Sec. III B the result with enhancement of the tunneling for one-photon and two-photon resonances.

2. One-photon and two-photon exact resonance: Stationary perturbation theory for Floquet states

In this section we apply the stationary perturbation method to calculate the Floquet states in the enlarged space $\mathcal{K}$ from the initial degenerate states φ_i and $\varphi_f e^{-iN\theta}$ (for $\alpha=0$). We calculate up to first order for the one-photon resonance ($N=1$) and up to second order for the two-photon resonance ($N=2$), in order to obtain the coefficients a_i and a_f of the linear combinations (23) that lift the degeneracy for a small value of α.

We denote K^α the α-dependent quasienergy operator

$$K^\alpha=K_0+\alpha\hat{W}, \quad (33)$$

with $K_0:=-i\omega(\partial/\partial\theta)+H_0$ and $\hat{W}:=x\sin\theta$ acting on $\mathcal{K}$.

The eigenvalue problem $K^\alpha\Psi^\alpha=\lambda^\alpha\Psi^\alpha$ is solved by the perturbation method, i.e., in terms of powers of the small amplitude α:

$$\lambda^\alpha=\lambda^{(0)}+\alpha\lambda^{(1)}+\alpha^2\lambda^{(2)}+\cdots, \quad (34)$$

$$\Psi^\alpha=|0\rangle+\alpha|1\rangle+\alpha^2|2\rangle+\cdots, \quad (35)$$

where $|0\rangle\in\mathcal{S}_0$, $\mathcal{S}_0$ denoting the zeroth-order subspace generated by the two degenerate Floquet states $\{|\phi_i\rangle:=|\varphi_i\otimes\mathbb{1}\rangle,\ |\phi_f\rangle:=|\varphi_f\otimes e^{-iN\theta}\rangle\}$ and $\lambda^{(0)}=E_i=E_f-N\omega$. $|0\rangle$ represents the unknown linear combinations (23).

First, we use this method up to first order in the one-photon resonance case to determine the value of the coefficients a_i and a_f of the linear combinations involving the two degenerate states φ_i and $\varphi_f e^{-i\theta}$ [Eqs. (23) for $N=1$] with the eigenvalue $E_i=E_f-\omega$. The quasienergy operator K^α, projected on the degeneracy subspace and written in the zeroth-order basis $\{\varphi_i,\varphi_f e^{-i\theta}\}$, is

$$P_0K^\alpha P_0=\begin{pmatrix}E_i & 0\\ 0 & E_f-\omega\end{pmatrix}+\alpha\begin{pmatrix}\langle\varphi_i|x|\varphi_i\rangle\frac{1}{2\pi}\int_0^{2\pi}d\theta\sin\theta & \langle\varphi_i|x|\varphi_f\rangle\frac{1}{2\pi}\int_0^{2\pi}d\theta e^{-i\theta}\sin\theta\\ \langle\varphi_f|x|\varphi_i\rangle\frac{1}{2\pi}\int_0^{2\pi}d\theta e^{i\theta}\sin\theta & \langle\varphi_f|x|\varphi_f\rangle\frac{1}{2\pi}\int_0^{2\pi}d\theta\sin\theta\end{pmatrix}$$
$$=\begin{pmatrix}E_i & 0\\ 0 & E_f-\omega\end{pmatrix}+\frac{\alpha}{2i}\begin{pmatrix}0 & \langle\varphi_i|x|\varphi_f\rangle\\ -\langle\varphi_f|x|\varphi_i\rangle & 0\end{pmatrix}, \quad (36)$$

where P_0 is the projector on the zeroth-order subspace.

The first order gives the eigenvalue problem restricted to the zeroth-order subspace

$$\hat{W}^{(1)}|0\rangle=\lambda^{(1)}|0\rangle, \quad (37)$$

with $\hat{W}^{(1)}=P_0\hat{W}P_0$. Written in the zeroth-order basis, this operator gives the matrix

$$\hat{W}^{(1)}=\frac{1}{2}\begin{pmatrix}0 & -i\langle\varphi_i|x|\varphi_f\rangle\\ i\langle\varphi_f|x|\varphi_i\rangle & 0\end{pmatrix}, \quad (38)$$

which gives the two different first-order contributions to the eigenvalues

$$\lambda_a^{(1)}=\frac{1}{2}|\langle\varphi_i|x|\varphi_f\rangle|, \quad (39a)$$

$$\lambda_b^{(1)} = -\frac{1}{2}|\langle \varphi_i | x | \varphi_f \rangle|. \tag{39b}$$

The degeneracy is thus linearly lifted.

The calculation of the two zeroth-order orthonormal eigenstates gives the two linear combinations [Eqs. (23) for $N=1$] with the coefficients $a_i = -i a_f = 1/\sqrt{2}$:

$$\Psi_a = \frac{1}{\sqrt{2}}(\varphi_i + i\varphi_f e^{-i\theta}), \tag{40a}$$

$$\Psi_b = \frac{1}{\sqrt{2}}(i\varphi_i + \varphi_f e^{-i\theta}). \tag{40b}$$

These coefficients had been obtained in [12] for a two-level model using the rotating-wave approximation to calculate explicitly the Floquet states. The present approach is general since it applies to all systems with dipole coupling. These coefficients satisfy the necessary condition (29) (but not sufficient) for complete inversion from φ_i to φ_f. Thus, in the one-photon resonance case we can express the transfer probability as an integral of the instantaneous quasienergy difference

$$P_{i\to f}(T_p) = |\kappa|^2 \sin^2\left[\frac{1}{2}\int_0^{T_p} ds(\lambda_a^{\alpha(s)} - \lambda_b^{\alpha(s)})\right]. \tag{41}$$

In the two-photon resonance case, the two degenerate Floquet states, which generate the zeroth-order subspace denoted $\mathcal{S}_0$, are φ_i and $\varphi_f e^{-2i\theta}$ associated with the eigenvalue $E_i = E_f - 2\omega$. The degeneracy in this subspace [Eqs. (23) for $N=2$] is not lifted in first-order perturbation theory because projection of the quasienergy matrix in the zeroth-order basis is now

$$P_0 K^\alpha P_0 = \begin{pmatrix} E_i & 0 \\ 0 & E_f - 2\omega \end{pmatrix} \quad \forall \alpha, \tag{42}$$

i.e.

$$P_0 \hat{W} P_0 \equiv 0. \tag{43}$$

The second-order eigenvalue problem restricted to the zeroth-order subspace $\mathcal{S}_0$ is [44]

$$\hat{W}^{(2)}|0\rangle = \lambda^{(2)}|0\rangle, \tag{44}$$

with

$$\hat{W}^{(2)} = -P_0 \hat{W} Q_0 (K_0 - \lambda^{(0)})^{-1} Q_0 \hat{W} P_0, \tag{45}$$

where $Q_0 = \mathbb{1} - P_0$. This eigenvalue problem gives the second-order correction of the eigenvalues and the associated zeroth-order eigenvectors.

The elements of the matrix $\hat{W}^{(2)}$ are given by

$$\hat{W}^{(2)}_{\nu\mu} = \sum_{(m,k)\neq\{(i,0),(f,-2)\}} \frac{1}{E_i - \lambda^{(0)}_{m,k}} \langle \phi_\nu | \hat{W} | \varphi_m e^{ik\theta}\rangle_{\mathcal{K}} \langle \varphi_m e^{ik\theta} | \hat{W} | \phi_\mu \rangle_{\mathcal{K}}, \qquad \nu,\mu \in \{i,f\}, \tag{46}$$

which we can write more explicitly as

$$\hat{W}^{(2)}_{ii} = \frac{1}{4}\sum_{m\in\mathbb{Z}} |\langle \varphi_i | x | \varphi_m \rangle|^2 \left[\frac{1}{E_i - E_m + \omega} + \frac{1}{E_i - E_m - \omega}\right], \tag{47a}$$

$$\hat{W}^{(2)}_{if} = -\frac{1}{4}\sum_{m\in\mathbb{Z}} \frac{\langle \varphi_i | x | \varphi_m \rangle \langle \varphi_m | x | \varphi_f \rangle}{E_i - E_m + \omega} = (\hat{W}^{(2)}_{fi})^*, \tag{47b}$$

$$\hat{W}^{(2)}_{ff} = \frac{1}{4}\sum_{m\in\mathbb{Z}} |\langle \varphi_f | x | \varphi_m \rangle|^2 \left[\frac{1}{E_f - E_m + \omega} + \frac{1}{E_f - E_m - \omega}\right]. \tag{47c}$$

The degeneracy is lifted if the coupling between the two unperturbed states (φ_i and φ_f) and some other states is not negligible, giving in general two different eigenvalues for the matrix $\hat{W}^{(2)}$. The associated orthonormal eigenvectors give the coefficients a_i and a_f and the formula of the oscillations (28) can be completely determined. In the general case, the coefficients a_i and a_f do not satisfy the necessary condition (29) of the complete inversion from φ_i to φ_f. In Sec. III C, we will evaluate explicitly this lifting of the degeneracy for the two-photon resonance that we will apply to the enhancement of tunneling.

3. N-photon quasiresonance

If the frequency is quasiresonant between the two levels φ_i and φ_f, $\omega \approx (E_f - E_i)/N$, the quasienergy diagram λ as a function of α reveals an avoided crossing near $\alpha = 0$ induced by the crossing in the (ω,α) plane for $\omega=(E_f-E_i)/N$ and $\alpha=0$. The two Floquet states associated with these two states are not degenerate for $\alpha=0$. We can therefore denote without ambiguity the two Floquet states $\Psi_{i,0}^{\alpha(t)}$ and $\Psi_{f,-N}^{\alpha(t)}$ associated with the quasienergies $\lambda_{i,0}^{\alpha(t)}$ and $\lambda_{f,-N}^{\alpha(t)}$.

This avoided crossing suggests the use of the result giving the transition probability [6] that is calculated with a model of the two Floquet levels involved in the avoided crossing and with the hypothesis of rapid transitions near this avoided crossing. We conjecture that in this situation, Eq. (28) should be replaced by

$$P_{i\to f}(T_p)\sim\sin^2\left[\frac{1}{2}\int_{t_c}^{T_p-t_c}ds(\lambda_{i,0}^{\alpha(s)}-\lambda_{f,-N}^{\alpha(s)})\right], \qquad (48)$$

where t_c is the time corresponding to the avoided crossing denoted α_c in the quasienergy diagram: $\alpha_c=\alpha(t_c)$.

B. Enhanced tunneling with a one-photon resonant state

We consider the doublet (φ_1,φ_2) and the higher unperturbed level φ_6, which is above the unperturbed barrier. In this section, we apply a monochromatic pulsed laser field of frequency ω chosen very close to the difference E_6-E_1 that leads to resonance with the state φ_6.

We start with the same initial condition as in Sec. II: the wave function is localized on one side of the well $[\phi(0)=\varphi_-]$. Because the laser beam is pulse-shaped, the frequency ω has actually a certain width $\Delta\omega$ that gets thinner for larger pulse widths. The one-photon transition between the levels φ_2 and φ_6 is parity forbidden. Thus we expect transitions between φ_1 and φ_6, but φ_2 will stay undisturbed in the sense that, at the end of the pulse, the transitions between φ_2 and the other states will be negligible (although during the pulse φ_2 is actually perturbed).

We can combine the one-photon resonant considerations, described in Sec. III A 2 for the evolution of φ_1 and φ_6, with the evolution of the nonresonant state φ_2, which follows its instantaneous quasienergy eigenstate $\Psi_2^{\alpha(t)}(\theta(t))$ without interaction with the others. In this case, the initial degenerate quasienergy $\lambda_a^{\alpha=0}=\lambda_b^{\alpha=0}$ labels the degenerate energy $E_1=E_6-\omega$. The Floquet eigenstates can be written in this case

$$\Psi_a^{\alpha=0}(\theta)=a_1\varphi_1+a_6\varphi_6e^{-i\theta}, \qquad (49a)$$

$$\Psi_b^{\alpha=0}(\theta)=-a_6^*\varphi_1+a_1^*\varphi_6e^{-i\theta}, \qquad (49b)$$

$$\Psi_2^{\alpha=0}(\theta)=\varphi_2, \qquad (49c)$$

with the coefficients a_1 and a_6 given by Eq. (40): $a_1=-ia_6=1/\sqrt{2}$. Inverting these equations and inserting φ_1 and φ_6 in the adiabatic time evolution of the initial condition $\phi(0)=\varphi_-$, one obtains

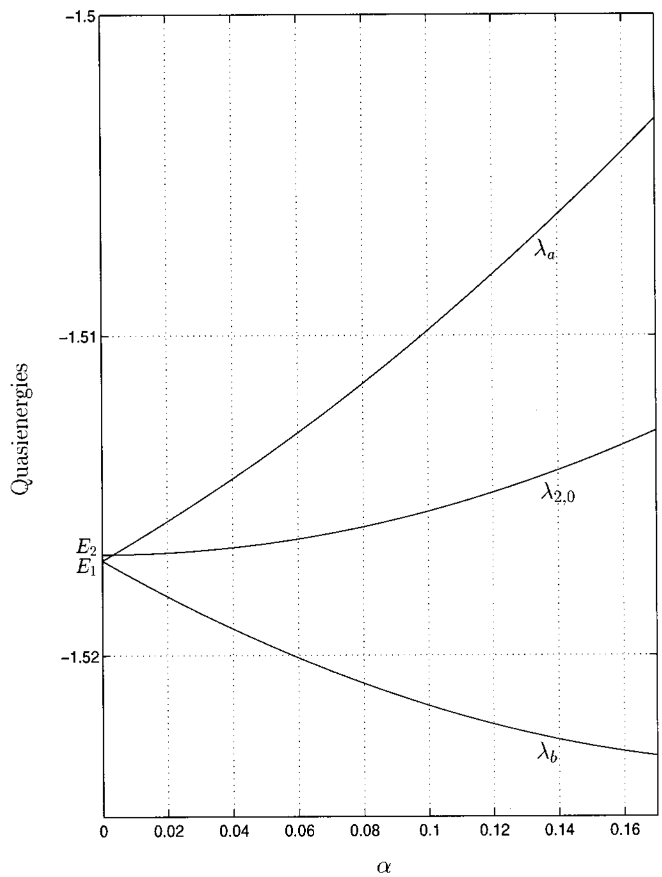

FIG. 4. For $\omega=1.6945\approx E_6-E_1$, quasienergy diagram close to the unperturbed energies E_1 and E_2. Notice the degeneracy of the Floquet eigenstates $\Psi_a^{\alpha=0}$ and $\Psi_b^{\alpha=0}$ with eigenvalue $E_1=E_6-\omega$, which is linearly lifted for $\alpha\neq0$.

$$\phi(T_p)=\frac{1}{\sqrt{2}}\exp\left(-i\int_0^{T_p}ds\lambda_a\right)\times\left\{\frac{1}{2}\left[1+\exp\left(i\int_0^{T_p}ds(\lambda_a-\lambda_b)\right)\right]\varphi_1+\frac{i}{2}e^{-iN(\theta+\omega T_p)}\left[1-\exp\left(i\int_0^{T_p}ds(\lambda_a-\lambda_b)\right)\right]\varphi_6-\exp\left(i\int_0^{T_p}ds(\lambda_a-\lambda_{2,0})\right)\varphi_2\right\}. \qquad (50)$$

The transition probability from φ_1 to φ_6 at the end of the pulse is then [formula (41) for $\kappa=1/\sqrt{2}$]

$$P_{1\to6}(T_p)=\frac{1}{2}\sin^2\left[\frac{1}{2}\int_0^{T_p}ds(\lambda_a^{\alpha(s)}-\lambda_b^{\alpha(s)})\right], \qquad (51)$$

which gives exactly the condition of complete Rabi cycle (31). As expected, the quasienergy diagram (Fig. 4) shows clearly the linear evolution of the quasienergies λ_a and λ_b for small values of α [Eq. (39)].

In Fig. 5(a), we have plotted the Rabi oscillations obtained by the numerical solution of the Schrödinger equation

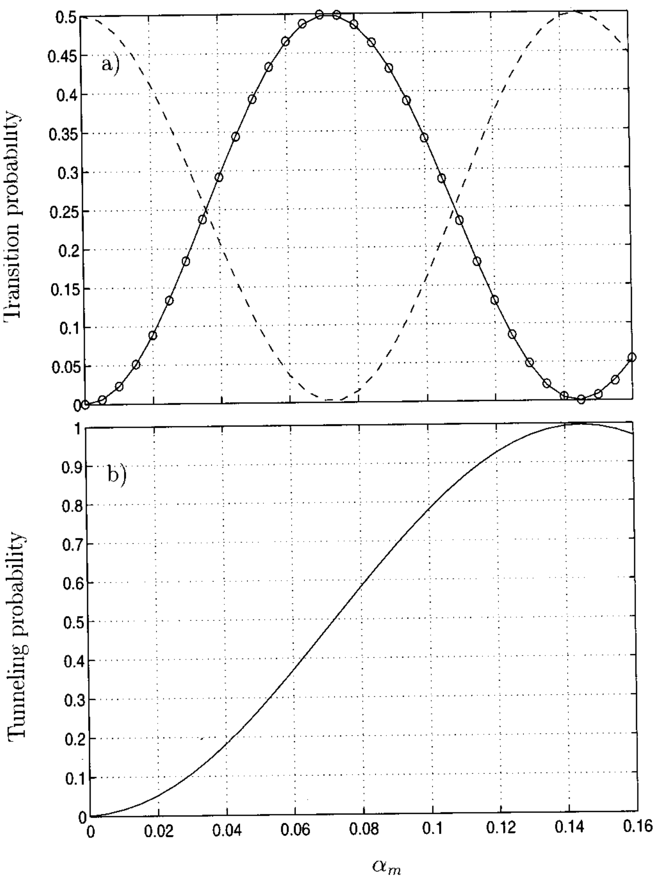

FIG. 5. (a) For $\omega=1.6945\approx E_6-E_1$, transition probabilities at the end of the pulse for $\varphi_1\rightarrow\varphi_6$ [i.e., $|\langle\varphi_6|\phi(T_p)\rangle|^2$], given by the numerical solution of the Schrödinger equation (solid line); $\varphi_1\rightarrow\varphi_6$, given by the predicting formula (41) with the quasienergies calculated numerically and shown in Fig. 4 (circles); and $\varphi_1\rightarrow\varphi_1$ [i.e., $|\langle\varphi_1|\phi(T_p)\rangle|^2$], given by the numerical solution of the Schrödinger equation (dashed line). (b) Tunneling probability at the end of the pulse. It is maximum (equal to 1) for the first complete Rabi cycle [i.e., $|\langle\varphi_6|\phi(T_p)\rangle|^2=0$].

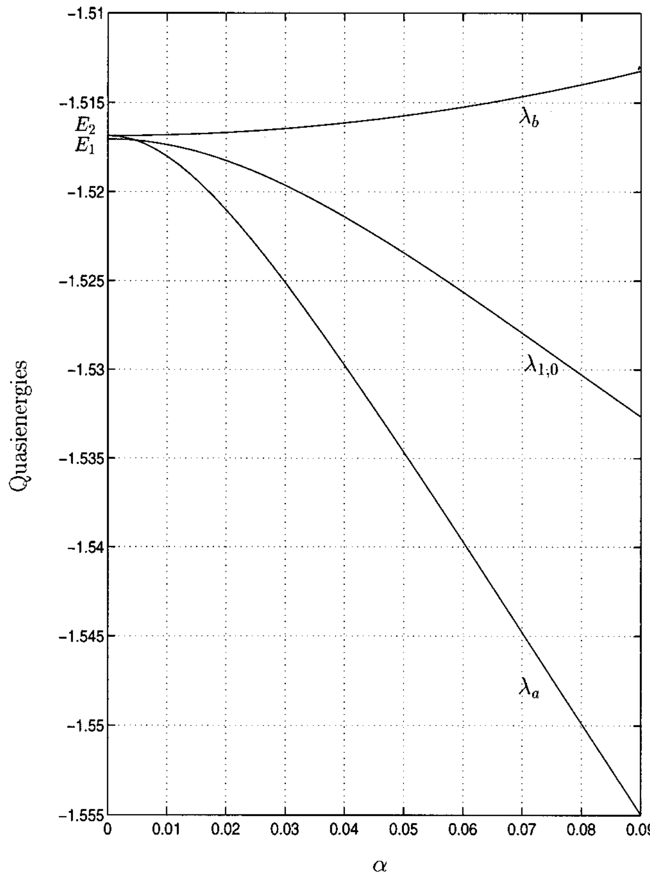

FIG. 6. For $\omega=0.847\,155\approx(E_6-E_2)/2$, quasienergy diagram close to the unperturbed energies E_1 and E_2. Notice the degeneracy of the Floquet eigenstates $\Psi_a^{\alpha=0}$ and $\Psi_b^{\alpha=0}$ with eigenvalue $E_2=E_6-2\omega$, which is quadratically lifted for $\alpha\neq 0$.

for our system and we compare it with the formula (51). The comparison shows very good agreement for the considered field amplitude range.

In order to obtain complete tunneling, we have necessarily to fulfill the condition of complete Rabi cycle (31) from φ_1 back to φ_1 at the end of the pulse. We now discuss the possibility to achieve tunneling with a one-photon resonance.

Inserting the condition (31) in Eq. (50) gives

$$\phi(T_p)=\frac{1}{\sqrt{2}}\exp\left(-i\int_0^{T_p}ds\lambda_a\right)\times\left[\varphi_1-\exp\left(i\int_0^{T_p}ds(\lambda_a-\lambda_{2,0})\right)\varphi_2\right], \quad (52)$$

which leads to a similar tunneling condition as in the nonresonant case (20)

$$\int_0^{T_p}ds(\lambda_a^{\alpha(s)}-\lambda_{2,0}^{\alpha(s)})=(2n+1)\pi, n\in\mathbb{Z}. \quad (53)$$

Thus the achievement of complete tunneling effect depends on the two conditions (31) and (53). In order to satisfy them, the two parameters (α_m, T_p) can be varied; we can also vary the frequency ω, keeping it, however, close to the value E_6-E_1 in order to stay close to resonance.

In order to find the parameters (α_m, T_p) for which these two conditions are satisfied, we perform a numerical calculation of the quasienergies for the fixed frequency $\omega=1.6945$, which is very close to the exact resonant value (quasienergy diagram in Fig. 4). For $T_p=741.8$ and $\alpha_m=0.145$, we predict complete tunneling with a good precision. We verify this prediction by the numerical simulation [see Fig. 5(b)]: the tunneling probability obtained from the quasienergy diagram (Fig. 4) is $\langle\varphi_+|\phi(T_p)\rangle=0.995$ and from the numerical simulation of the Schrödinger equation [Fig. 5(b)], we obtain $\langle\varphi_+|\phi(T_p)\rangle=0.996$. The maximum amplitude $(\alpha_{m(\text{one-photon-resonant})}=0.145)$ leading to the enhancement of tunneling gives the maximum intensity smaller by a factor 5 than the one obtained in the nonresonant case $(\alpha_{m(\text{nonresonant})}=0.332)$.

C. Enhanced tunneling with a two-photon resonant state

1. Two-photon exact resonance between the states φ_2 and φ_6

Now we consider the same laser pulse but of frequency ω very close to $(E_6-E_2)/2$. The selection rule makes now the $\varphi_1 \rightarrow \varphi_6$ transition parity forbidden. Thus we expect a two-photon transition between φ_2 and φ_6 that is favored by the presence of the unperturbed level φ_3 approximately equidistant between φ_2 and φ_6 (φ_4 is irrelevant because of the selection rules). The initial condition is again $\phi(0)=\varphi_-$. The initial degenerate quasienergy $\lambda_a^{\alpha=0}=\lambda_b^{\alpha=0}$ now labels the degenerate energy $E_2=E_6-2\omega$.

In this section we achieve tunneling with two-photon resonance from φ_2 back to itself, using exactly the same procedure exposed in Sec. III B. In this case, the initial Floquet states

$$\Psi_a^{\alpha=0}(\theta)=a_2\varphi_2+a_6\varphi_6 e^{-2i\theta}, \tag{54a}$$

$$\Psi_b^{\alpha=0}(\theta)=-a_6^*\varphi_2+a_2^*\varphi_6 e^{-2i\theta}, \tag{54b}$$

$$\Psi_1^{\alpha=0}(\theta)=\varphi_1 \tag{54c}$$

give the transition probability from φ_2 to φ_6 at the end of the pulse [formula (28) with $\kappa=1/\sqrt{2}$]

$$P_{2\rightarrow 6}(T_p)=2|a_2a_6|^2\sin^2\left[\frac{1}{2}\int_0^{T_p}ds(\lambda_a^{\alpha(s)}-\lambda_b^{\alpha(s)})\right]. \tag{55}$$

The values of the weights a_2 and a_6 are given by the eigenvectors of the second-order matrix $\hat{W}^{(2)}$ [matrix (46) including the coefficients (47) with the indices i and f replaced, respectively, by 2 and 6].

The dominant contribution to these matrix elements is approximately given by the unperturbed level φ_3:

$$\hat{W}^{(2)}\approx\frac{1}{4(E_2-E_3+\omega)}\begin{pmatrix}|\langle\varphi_2|x|\varphi_3\rangle|^2 & -\langle\varphi_2|x|\varphi_3\rangle\langle\varphi_3|x|\varphi_6\rangle\\ -\langle\varphi_6|x|\varphi_3\rangle\langle\varphi_3|x|\varphi_2\rangle & |\langle\varphi_6|x|\varphi_3\rangle|^2\end{pmatrix}. \tag{56}$$

The diagonalization gives the two different second-order contributions to the eigenvalues that lift the degeneracy

$$\lambda_a^{(2)}\approx\frac{|\langle\varphi_2|x|\varphi_3\rangle|^2}{4(E_2-E_3+\omega)}, \tag{57a}$$

$$\lambda_b^{(2)}\approx 0. \tag{57b}$$

The associated zeroth-order eigenvectors are

$$\Psi_a=\cos\frac{\delta}{2}\varphi_2+\sin\frac{\delta}{2}\varphi_6 e^{-2i\theta}, \tag{58a}$$

$$\Psi_b=-\sin\frac{\delta}{2}\varphi_2+\cos\frac{\delta}{2}\varphi_6 e^{-2i\theta}, \tag{58b}$$

with

$$\tan\delta\approx\frac{2|\langle\varphi_2|x|\varphi_3\rangle\langle\varphi_3|x|\varphi_6\rangle|}{|\langle\varphi_2|x|\varphi_3\rangle|^2-|\langle\varphi_6|x|\varphi_3\rangle|^2}. \tag{59}$$

The quasienergy diagram (see Fig. 6) confirms the quadratic dependence of λ_a and the absence of change of λ_b for small values of α. The numerical value of the maximum probability transition $P_{2\rightarrow 6}(T_p)$ [Eq. (55)] is: $2|a_2^*a_6|^2\approx 0.473$ [considering not only the dominant contributions but the exact elements (47) of the matrix $\hat{W}^{(2)}$].

To study the validity of the formula (55), leading to the condition of complete Rabi cycle (31), we compare it with the transition probability calculated explicitly by solving numerically the Schrödinger equation in Fig. 7(a). The agreement is quite good.

The condition of complete Rabi cycle gives the additional condition for complete tunneling

$$\int_0^{T_p}ds(\lambda_a^{\alpha(s)}-\lambda_{1,0}^{\alpha(s)})=(2n+1)\pi, \quad n\in\mathbb{Z}. \tag{60}$$

In order to determine if we can fulfill the two conditions (31) and (60), we calculate numerically the quasienergies for the fixed frequency $\omega=0.847\,155$, which is very close to the exact resonant value (quasienergy diagram in Fig. 6).

First, we fix the pulse length ($T_p=741.8$) and make the maximum amplitude α_m vary to achieve complete tunneling. Figure 7(b) shows that the first complete rabi cycle [$n=0$ of Eq. (31), i.e., $\alpha_m=0.052$] leads to incomplete tunneling (only about 90%). We do not find any value of α_m compatible with adiabatic behavior yielding complete tunneling.

This leads us to vary both parameters to look for complete tunneling. We find it for the maximum amplitude $\alpha_m=0.0424$ and a pulse length $T_p=9272.5$, which is only around 2 times smaller than the bare tunneling time. We conclude that in order to obtain a better result we have to optimize also the choice of the frequency. We obtain a better enhancement, i.e., tunneling for a shorter pulse length and a maximum amplitude of the same order of magnitude, varying the frequency around the resonant value. We present this final result in the next subsection.

We remark that in Fig. 10(a), we have plotted the projection of the time evolution on the three instantaneous Floquet states $\Psi_1^{\alpha(t)}(\theta(t))$, $\Psi_2^{\alpha(t)}(\theta(t))$, and $\Psi_6^{\alpha(t)}(\theta(t))$ for $\alpha_m=0.052$. We note that, numerically, we do not find the degeneracy of Ψ_2^0 and Ψ_6^0 for $\alpha=0$ because the frequency is not exactly resonant due to the finite numerical precision, but very close to the resonant value. This produces a rapid transition between $\Psi_2^{\alpha(t)}$ and $\Psi_6^{\alpha(t)}$ for $\alpha\approx 0$ instead of the degeneracy.

2. Two-photon quasiresonance between the states φ_2 and φ_6

In this section we choose a value of the frequency close to the previous value, which allows us to obtain tunneling for a value of α smaller than the one obtained in the one-photon resonance. φ_2 and φ_6 are now almost in resonance. In this case of two-photon quasiresonance between φ_2 and φ_6, the

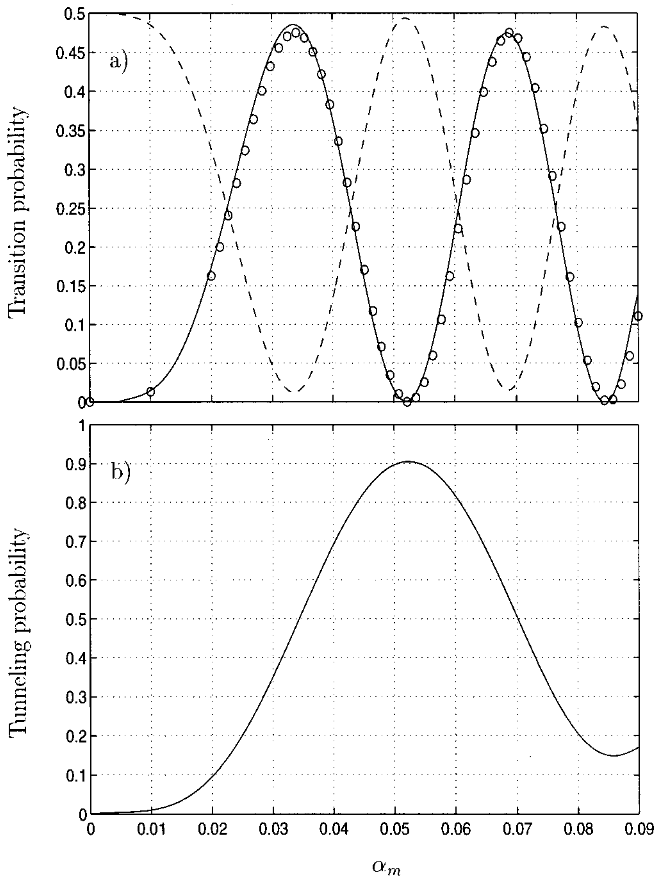

FIG. 7. (a) For $\omega = 0.847\ 155 \approx (E_6 - E_2)/2$, transition probabilities at the end of the pulse for $\varphi_2 \rightarrow \varphi_6$ [i.e., $|\langle \varphi_6 | \phi(T_p) \rangle|^2$], given by the numerical solution of the Schrödinger equation (solid line); $\varphi_2 \rightarrow \varphi_6$, as predicted by Eq. (55) with the quasienergies calculated numerically and shown in Fig. 6 (circles); and $\varphi_2 \rightarrow \varphi_2$ [i.e., $|\langle \varphi_2 | \phi(T_p) \rangle|^2$], given by the numerical solution of the Schrödinger equation (dashed line). (b) Tunneling probability at the end of the pulse. It is equal to 0.9 for the first complete Rabi cycle [i.e., $|\langle \varphi_6 | \phi(T_p) \rangle|^2 = 0$].

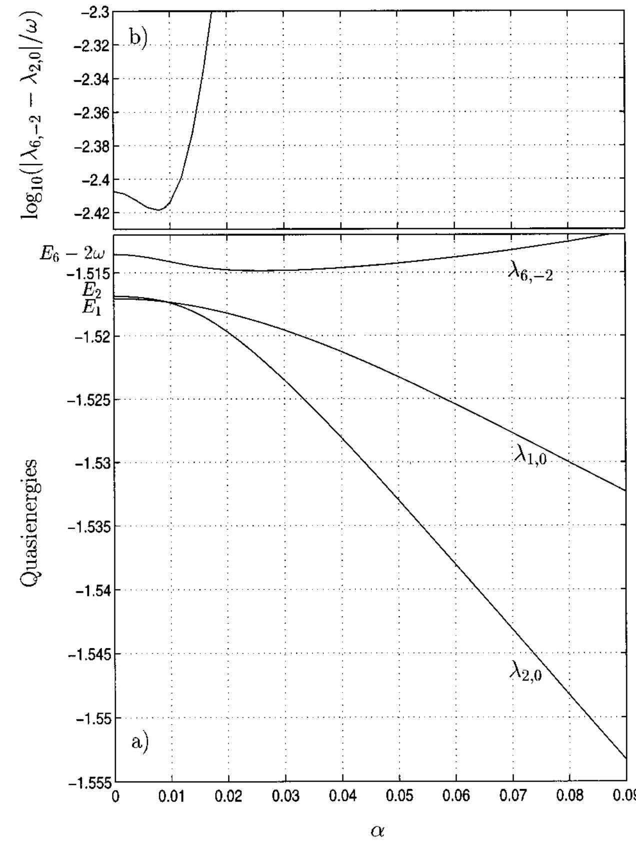

FIG. 8. (a) For $\omega = 0.8455$, quasienergy diagram close to the unperturbed energies E_1 and E_2. Notice the absence of degeneracy of the Floquet eigenstates $\Psi_2^{\alpha=0}$ and $\Psi_6^{\alpha=0}$. (b) Logarithm of $|\lambda_{6,-2}^{\alpha} - \lambda_{2,0}^{\alpha}|/\omega$, which shows the avoided crossing for $\alpha \approx 0.009$.

selection rules are unchanged from the case of exact resonance: the initial nonresonant state φ_1 evolves following its instantaneous Floquet state $\Psi_1^{\alpha(t)}(\theta(t))$ and we expect transitions between the states φ_2 and φ_6. However, the two Floquet states associated with these two states are not degenerate for $\alpha = 0$. The two branches on the quasienergy diagram [Fig. 8(a)] can be easily identified by continuity from the different energies E_2 and $E_6 - 2\omega$ for $\alpha = 0$ and labeled by $\Psi_2^{\alpha(t)}(\theta(t))$ and $\Psi_6^{\alpha(t)}(\theta(t))$. On this diagram [Fig. 8(a)] we observe the expected avoided crossing for $\alpha \approx 0.009$ [see the more detailed logarithmic plot in Fig. 8(b)]. We can interpret the transition between φ_2 and φ_6 with the formula (48), supposing that the transition is rapid enough close the avoided crossing:

$$P_{2\rightarrow 6}(T_p) \sim \sin^2\left[\frac{1}{2}\int_{t_c}^{T_p - t_c} ds(\lambda_2^{\alpha(s)} - \lambda_6^{\alpha(s)})\right], \qquad (61)$$

where t_c is the time corresponding to the avoided crossing denoted α_c in the quasienergy diagram: $\alpha_c = \alpha(t_c)$.

In Fig. 9(a) we have plotted the comparison between this formula (61) including the maximum calculated with the method for the exact resonance and the transition probability obtained from the numerical solution of the Schrödinger equation. We find that the formula (61) is a good approximation of this transfer of population, especially if the maximal amplitude is large compared to the point of the avoided crossing.

To achieve tunneling, we have moreover to suppose the validity of the property shown for the exact resonance (32) giving the phase of the time evolution of the initial state with the condition of complete Rabi cycle. In this case, the tunneling condition is exactly the same as the one obtained for the two-photon exact resonance (60).

In Fig. 10 we have plotted the projection of the time evolution on the three instantaneous Floquet states $\Psi_1^{\alpha(t)}(\theta(t))$, $\Psi_2^{\alpha(t)}(\theta(t))$, and $\Psi_6^{\alpha(t)}(\theta(t))$ for two-photon quasiresonant frequencies ($\alpha_m = 0.052$ is fixed). We note that the beginning of the $\Psi_2^{\alpha(t)} \rightarrow \Psi_6^{\alpha(t)}$ transition appears at a higher field intensity (close to the avoided crossing) for the less resonant frequency [Fig. 10(b)].

We predict and find enhancement of the tunneling (with the pulse length $T_p = 741.8$) for the maximum field amplitude $\alpha_{m(\text{two-photon-resonant})} = 0.052$ [see Fig. 9(b)], which gives the maximum intensity lower by a factor 40 than the maximum field intensity needed to enhance tunneling in the nonresonant case. The tunneling probability obtained from the

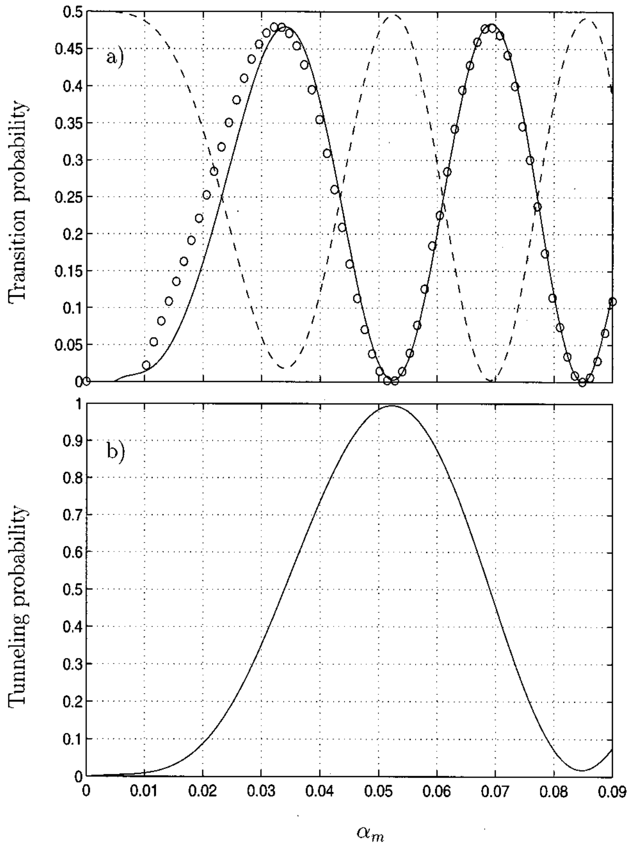

FIG. 9. (a) For $\omega = 0.8455 \approx (E_6 - E_2)/2$, but further away from resonance than the case in Fig. 7. Transition probabilities from the initial condition $\phi(0) = (\varphi_1 - \varphi_2)/\sqrt{2}$, at the end of the pulse for $\varphi_2 \to \varphi_6$ [i.e., $|\langle \varphi_6 | \phi(T_p) \rangle|^2$], given by the numerical solution of the Schrödinger equation (solid line); $\varphi_2 \to \varphi_6$, as predicted by Eq. (61) with the quasienergies calculated numerically as shown in Fig. 8 (circles); and $\varphi_2 \to \varphi_2$ [i.e., $|\langle \varphi_2 | \phi(T_p) \rangle|^2$], given by the numerical solution of the Schrödinger equation (dashed line). (b) Tunneling probability at the end of the pulse. It is equal to 0.99 for the first complete Rabi cycle [i.e., $|\langle \varphi_6 | \phi(T_p) \rangle|^2 = 0$].

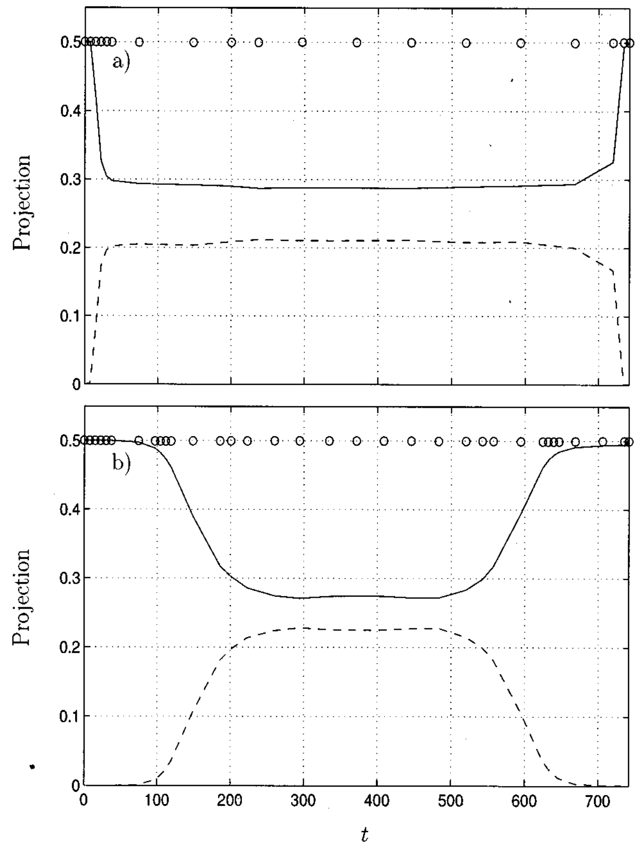

FIG. 10. Numerical solution of the Schrödinger equation projected at each time t on the instantaneous Floquet states for $\alpha_m = 0.052$: $|\langle \Psi_1^{\alpha(t)(\theta(t))} | \phi(t) \rangle|^2$, circles; $|\langle \Psi_2^{\alpha(t)}(\theta(t)) | \phi(t) \rangle|^2$, solid line; and $|\langle \Psi_6^{\alpha(t)}(\theta(t)) | \phi(t) \rangle|^2$, dashed line, for (a) the resonant frequency $\omega = 0.847\,155$ and (b) the quasiresonant frequency $\omega = 0.8455$.

quasienergy diagram (Fig. 8) is $\langle \varphi_+ | \phi(T_p) \rangle = 0.9942$, and from the numerical simulation of the Schrödinger equation [Fig. 9(b)], we obtain $\langle \varphi_+ | \phi(T_p) \rangle = 0.9941$.

IV. CONCLUSION

In this paper we obtain an enhancement of tunneling in a quartic double-well potential model of the NH_3 molecule by a pulse-shaped laser field, i.e. the tunneling is achieved in a time corresponding to the pulse length (T_p), which is much shorter than the bare tunneling time ($\tau_b \approx 16574$): $T_p = 741.8 \ll \tau_b$. The enhancement of tunneling is obtained for different ranges of maximum field intensity depending on the frequency of the laser field leading to qualitatively different processes: no resonance, one-photon resonance, and two-photon resonance. The interest of using the two-photon resonant process is that we can work with a lower intensity than for the nonresonant process. (The maximal amplitude of the pulse is lower by a factor 40.)

The tools we use to interpret the enhancement of tunneling are the adiabatic Floquet formalism for the theoretical prediction and the numerical solution of the time-dependent Schrödinger equation. The adiabatic Floquet theory has proven to be a very well adapted tool that allows us to develop a precise understanding of the mechanisms of N-photon resonant transitions.

ACKNOWLEDGMENTS

We would like to thank G. Jolicard for many helpful discussions. Support from the EC Contract No. ERBCHRXCT94-0460 for the project ‘‘stability and universality in classical mechanics’’ is acknowledged.

[1] J. H. Shirley, Phys. Rev. **138**, B979 (1965).
[2] H. Sambe, Phys. Rev. A **7**, 2203 (1973).
[3] S. Chu, Adv. Chem. Phys. **73**, 739 (1986).
[4] R. Blümel and U. Smilansky, Z. Phys. D **6**, 83 (1987).
[5] G. Casati and L. Molinari, Prog. Theor. Phys. (Suppl.) **98**, 286 (1989).
[6] H. P. Breuer, K. Dietz, and M. Holthaus, Z. Phys. D **8**, 349 (1988).

[7] H. P. Breuer and M. Holthaus, Z. Phys. D **11**, 1 (1989).
[8] W. Jakubetz, B. Just, J. Manz, and H. J. Schreier, J. Phys. Chem. **94**, 2294 (1990).
[9] J. Henkel and M. Holthaus, Phys. Rev. A **45**, 1978 (1992).
[10] K. Dietz, J. Henkel, and M. Holthaus, Phys. Rev. A **45**, 4960 (1992).
[11] J. P. Connerade, K. Conen, K. Dietz, and J. Henkel, J. Phys. B **25**, 3771 (1992).
[12] M. Holthaus and B. Just, Phys. Rev. A **49**, 1950 (1994).
[13] H. P. Breuer, K. Dietz, and M. Holthaus, J. Phys. B **24**, 1343 (1991).
[14] H. P. Breuer and M. Holthaus, J. Phys. Chem. **97**, 12 634 (1993).
[15] W. Lin and L. Balentine, Phys. Rev. Lett. **67**, 159 (1991).
[16] F. Grossmann, T. Dittrich, P. Jung, and P. Hänggi, Phys. Rev. Lett. **67**, 516 (1991).
[17] F. Grossmann, P. Jung, T. Dittrich, and P. Hänggi, Z. Phys. B **84**, 315 (1991).
[18] M. Holthaus, Phys. Rev. Lett. **69**, 1596 (1992).
[19] F. Grossmann, T. Dittrich, P. Jung, and P. Hänggi, J. Stat. Phys. **70**, 229 (1993).
[20] P. Hänggi, in *Quantum Dynamics of Submicron Structures*, edited by H. A. Cerdeira (Kluwer Academic, Dordrecht, 1995).
[21] T. H. Stoof and Y. V. Nazarov, Phys. Rev. B **53**, 1050 (1996).
[22] D. Gotlieb and S. Orszag, *Numerical Analysis of Spectral Methods: Theory and Applications* (SIAM, Philadelphia, 1977).
[23] D. Kosloff and R. Kosloff, J. Comput. Phys. **52**, 35 (1983).
[24] R. Kosloff, in *Numerical Grid Methods and Their Application to Schrödinger Equation*, Vol. 412 of *NATO Advanced Study Institute Series C*, edited by C. Cerjan (Kluwer Academic, Dordrecht, 1993).
[25] J. Boyd, *Chebyshev and Fourier Spectral Methods* (Springer, New York, 1989).
[26] C. Leforestier *et al.*, J. Comput. Phys. **94**, 59 (1991).
[27] C. Lanczos, J. Res. Natl. Bur. Stand. **45**, 255 (1950).
[28] T. Park and J. Light, J. Chem. Phys. **85**, 5870 (1986).
[29] A. Rauk, L. C. Allen, and E. Clementi, J. Chem. Phys. **52**, 4133 (1970).
[30] J. Bellissard, in *Trends in the Eighties*, edited by S. Albeverio and P. Blanchard (World Scientific, Singapore, 1985).
[31] J. Howland, Math. Ann. **207**, 315 (1974).
[32] T. S. Ho and S. I. Chu, J. Phys. B **17**, 2101 (1984).
[33] H. R. Jauslin and J. L. Lebowitz, Chaos **1**, 114 (1991).
[34] P. Blekher, H. R. Jauslin, and J. L. Lebowitz, J. Stat. Phys. **68**, 271 (1992).
[35] H. R. Jauslin, in *II Granada Lectures in Computational Physics*, edited by P. L. Garrido and J. Marro (World Scientific, Singapore, 1993).
[36] H. R. Jauslin, S. Guérin, and A. Déroussiaux, in *Chaos: The Interplay Between Stochastics and Deterministic Behaviour*, edited by P. Garbaczewski, A. Weron, and M. Wolf, Lecture Notes in Physics Vol. x (Springer, Berlin, 1995), pp. 161–168.
[37] M. V. Berry, Proc. R. Soc. London Ser. A **392**, 45 (1984).
[38] H. P. Breuer, K. Dietz, and M. Holthaus, Phys. Rev. A **47**, 725 (1993).
[39] D. J. Moore, Phys. Rep. **210**, 1 (1991).
[40] R. H. Young and W. J. Deal, Jr., J. Math. Phys. **11**, 3298 (1970).
[41] H. P. Breuer and M. Holthaus, Phys. Lett. A **140**, 507 (1989).
[42] S. Guérin and H. R. Jauslin (unpublished).
[43] A. Joye, Asymptotic Anal. **9**, 209 (1994).
[44] C. Piron, *Mécanique Quantique—Bases et Applications* (Presses Polytechniques et Universitaires Romandes, Lausanne, 1990).