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Dynamical resonances and the topology of the multiphoton adiabatic passage

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We analyze adiabatic transfer processes in two-level systems driven by bichromatic delayed laser pulses. We show that the robust transfer of atomic population and the exchange of momentum with the laser fields can be understood on the basis of topological properties of dressed energy surfaces that are determined by dynamical resonances. The analysis shows a topological quantization of the multiple absorption and emission of photons.

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I. INTRODUCTION

Adiabatic passage techniques provide a robust method to control atomic and molecular processes by interaction with laser pulses. Different versions have been proposed and realized experimentally [1–7]. The main effect that can be achieved is the complete transfer of atomic population to a selected excited state. Another effect is, e.g., the control of momentum transfer from the laser fields to an atomic beam, leading to a deflection of the beam [8–12]. The phenomenology of single- and two-photon adiabatic passage is a fairly well understood field. However, this is not so for multiphoton processes. In this paper we consider these processes with pulse-shaped bichromatic fields in two-level systems. The models are analyzed in terms of dressed states, which are the eigenstates of the coupled atom-plus-field system. The theoretical analysis of these processes indicates that the main conceptual ingredients are the adiabatic following of dressed states and connectivity properties of the paths linking initial and target states. These processes are robust because they do not depend on the precise shapes of the laser pulses or on precise tuning of laser frequencies.

The goal of the present paper is twofold. First we show in the context of a concrete example that under adiabatic conditions the dynamics of the process can be described completely by the topology of dressed-state energy surfaces. This topological aspect is the key to the robustness of the process. The second goal is to show that the topology of these surfaces is completely determined by the resonances of the system. These resonances can be of two kinds: (i) zeroth-order resonances, which arise when a laser frequency (or an integer combination of frequencies) is equal to a Bohr frequency of the atom. This type of resonance has an effect for arbitrarily weak-field amplitudes, provided that the time of interaction is long enough; (ii) *dynamical or nonlinear resonances* that are *induced by the field*, and appear only when the fields becomes stronger than some well-defined *thresholds* (see, e.g., [13]). The effect of nonlinear resonances, which cannot be treated by simple perturbation expansions, is analyzed with the general method developed in [14]. It consists of a combination of perturbative techniques formulated in terms of an iteration of contact transformations (which are the quantum equivalent of Kolmogorov-Arnold-Moser transformations [15]) and specific unitary transformations that are adapted to the dynamics of the resonances. One of the main results is that, for processes based on adiabatic following on the energy surfaces, the perturbative corrections provided by the contact transformations are irrelevant, in the sense that the end effect of the transfer does not depend on them. Our approach can be used to select and optimize the transfer of population and of momentum by the suitable choice of the delay between the pulses and the peak amplitudes of the two lasers.

II. EFFECTIVE DRESSED HAMILTONIAN

We consider the Hamiltonian

$$\mathsf{H}(\underline{\omega}t+\underline{\theta})=\mathsf{H}_0-\mathbf{d}\cdot\left[\sum_{i=1}^{2}\mathbf{e}_i\alpha_i(t)\cos(\omega_i t+\theta_i)\right], \qquad (1)$$

where H_0 is the Hamiltonian of the free two-level system of energies $E_1<E_2$. The states $\{|1\rangle,|2\rangle\}$ span the Hilbert space $\mathcal{H}=\mathbb{C}^2$ on which H_0 and the dipole moment operator $\mathbf{d}$ act. The total electric field, containing two carrier frequencies $\underline{\omega}=(\omega_1,\omega_2)$, is characterized by unit vectors $\mathbf{e}_j$, smooth pulsed-shaped envelope functions of time $\underline{\alpha}(t)=[\alpha_1(t),\alpha_2(t)]$, and the initial phases $\underline{\theta}=(\theta_1,\theta_2)$. The interaction is thus characterized by the time-dependent Rabi frequencies $\Omega_j(t)=-\langle 1|\mathbf{d}\cdot e_j|2\rangle\alpha_j(t)/\hbar$, $j=1,2$. We consider solutions $|\phi(t)\rangle$ of the time-dependent Schrödinger equation with the initial condition $|\phi(-\infty)\rangle=|1\rangle$. Both frequencies are different and off-resonant, as depicted in Fig. 1: $\Delta_i\equiv(E_2-E_1)/\hbar-\omega_i$, $i=1,2$. We introduce the relevant beat frequency $\delta\equiv\omega_1-\omega_2=\Delta_2-\Delta_1$. In the following, we will consider for simplicity the case $\Delta_1=-\Delta_2$ so that $\delta=-2\Delta_1$. We study a strong-field regime in the sense that the nonperturbative regime $|\delta|\lesssim\max_t\{|\Omega_j(t)|\}\ll(E_2-E_1)/\hbar$, $j=1,2$ is considered. Since for frozen field amplitudes the Hamiltonian (1) is periodic with two frequencies, it is natural to introduce the strong-field dressed Hamiltonian (the so-called Floquet or quasienergy Hamiltonian) [16–18], which we formulate in a way that derives naturally from the theory of quantized dressed states in a cavity [19–21]:

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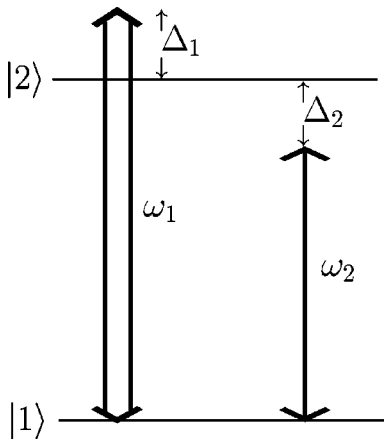

FIG. 1. Diagram of linkage patterns between two atomic states.

$$\mathsf{K} = -i\hbar\omega_1 \frac{\partial}{\partial\theta_1} - i\hbar\omega_2 \frac{\partial}{\partial\theta_2} + \mathsf{H}(\underline{\theta}). \tag{2}$$

The Floquet Hamiltonian allows us to take into account the photon exchanges between the atom and the laser fields. It is formally constructed on the initial phases $\underline{\theta}$ of the fields that are treated as dynamical variables acting on $\mathcal{L} = \mathcal{L}_2(d\theta_1/2\pi) \otimes \mathcal{L}_2(d\theta_2/2\pi)$ where each $\mathcal{L}_2(d\theta_i/2\pi)$ is the Hilbert space of 2π-periodic functions of the angle θ_i. The quasienergy operator (2) acts thus on the enlarged space $\mathcal{K} = \mathcal{H} \otimes \mathcal{L}$. The eigenelements of K can be calculated numerically in a truncated Fourier decomposition for each frequency. In the regime considered here the Floquet Hamiltonian (2) can be approximated, after a rotating wave transformation $\left[\begin{smallmatrix} 1 & 0 \\ 0 & e^{i\theta_1} \end{smallmatrix}\right] K \left[\begin{smallmatrix} 1 & 0 \\ 0 & e^{-i\theta_1} \end{smallmatrix}\right]$, by an effective dressed Hamiltonian given by [11]

$$\mathsf{K}_{\text{eff}} = -i\hbar\delta \frac{\partial}{\partial\theta} + \frac{\hbar}{2}\begin{bmatrix} 0 & \Omega_1 \\ \Omega_1 & 2\Delta_1 \end{bmatrix} + \frac{\hbar\Omega_2}{2}\begin{bmatrix} 0 & e^{-i\theta} \\ e^{i\theta} & 0 \end{bmatrix}, \tag{3}$$

with $\theta \equiv \theta_1 - \theta_2$. This effective model is valid only if the two frequencies ω_1 and ω_2 are not equal. The derivative term represents the relative number of photon pairs, one ω_1 photon minus one ω_2 photon. Thus the absorption of one ''effective photon'' of frequency δ in the effective model (3) corresponds in the complete model (2) to the absorption of one photon of frequency ω_1 and the emission of one photon of frequency ω_2. If the two laser fields are counterpropagating perpendicularly to the atomic beam, this double photon exchange results in a net transfer of momentum to the atom of $\hbar(\omega_1+\omega_2)/c$, which manifests as a deflection of the beam. The second term of the effective Hamiltonian (3) is the usual rotating wave approximation (RWA) Hamiltonian (associated with the ω_1 field), with eigenvalues $2\lambda^0_\pm = \hbar\Delta_1 \pm \hbar\sqrt{(\Delta_1)^2 + (\Omega_1)^2}$. The third term can be viewed as a perturbation of this usual RWA Hamiltonian.

Our method to analyze the dynamics globally consists of (i) the calculation of the dressed eigenenergy surfaces of the effective quasienergy operator as a function of the two Rabi frequencies Ω_1 and Ω_2, (ii) the analysis of their topology, and (iii) the application of adiabatic principles to determine the dynamics of processes in view of the topology of the surfaces.

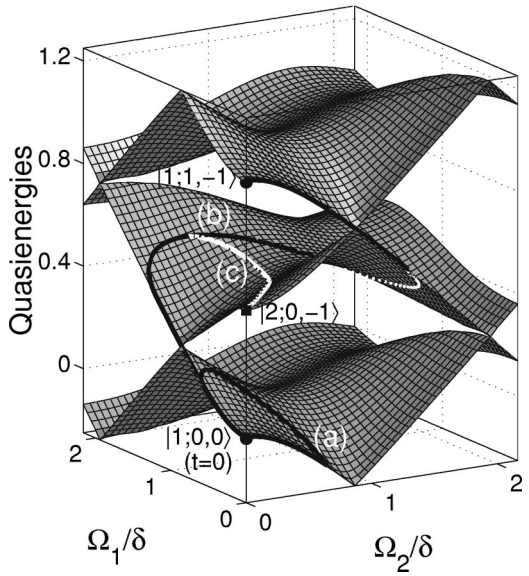

FIG. 2. Quasienergy surfaces (in units of δ) as functions of Ω_1 and Ω_2 for $\delta = -2\Delta_1 = 2\Delta_2$. Three different paths [denoted (a), (b), and (c)] depending on the temporal evolution of the pulses are depicted.

III. QUASIENERGY SURFACE TOPOLOGY

For frozen values of the two fields Ω_1 and Ω_2, we calculate dressed states and dressed energies by diagonalizing K_{eff}. The eigenelements can be labeled with two indices: one, denoted n, refers to the levels of the atom, and another one denoted k, to the relative photon numbers. The index k stands for the number of ω_1 photons absorbed and the number of ω_2 photons emitted. The eigenvalues and eigenvectors have the following property of periodicity: $\lambda_{n;k,-k} = \lambda_{n;0,0} + k\hbar\delta$ and thus $|n;k,-k\rangle_{\text{eff}} = |n;0,0\rangle_{\text{eff}} \exp(ik\theta)$. The eigenelements appear as two families, each of which consists of an infinite set of eigenvalues with equal spacing $\hbar\delta$. The eigenstates of K_{eff} $|1;k,-k\rangle_{\text{eff}}$ and $|2;k,-k\rangle_{\text{eff}}$ can thus be labeled by $|1;k,-k\rangle$ and $|2;-1+k,-k\rangle$, $k \in \mathbb{Z}$ in the original basis of Eq. (2).

In Fig. 2, we display quasienergy surfaces, calculated numerically, as functions of the scaled Rabi frequencies Ω_1/δ and Ω_2/δ. Together with the adiabatic analysis, the topology of these surfaces gives insight into the various atomic population and photon transfers that can be produced by choosing appropriately the temporal evolution of the pulses. We start with the dressed state $|1;0,0\rangle$, i.e., the lowest atomic state with zero ω_1 and ω_2 photons. Its energy is shown as the starting point of various paths. Since the envelopes of the pulses vary slowly, the adiabatic theorem implies that the solution of the time-dependent *dressed* Schrödinger equation follows the instantaneous dressed eigenvectors continuously connected to the initial state. This describes accurately the dynamics if the time dependence of the envelopes is slow

enough according to the Landau-Zener analysis. If two (uncoupled) eigenvalues cross, the adiabatic analysis shows that the dynamics follows the crossing. We define the *transfer state* as the dressed eigenvector that is adiabatically followed, i.e., on which the dressed population resides during the dynamics. *The knowledge of the topology of the quasienergy surfaces gives the adiabatic connectivity between the initial and target dressed states*. There are two infinite families of quasienergy surfaces that are constructed by the translations by $\hbar\,\delta k$, $k\in\mathbb{Z}$ of two surfaces. Any two neighboring surfaces have points of contact that are conical intersections. In the present model all the points of intersection are located either at the line $\Omega_1=0$ or at the line $\Omega_2=0$, corresponding to the situations where only one of the laser fields is interacting with the atom. This has the crucial consequence that the adiabatic following across these intersections can be done in a robust way, by making sure that the second field is not active when the amplitude of the first one goes across the intersection. This is the mechanism that allows a robust controlled adiabatic passage from one dressed state surface to the next. Besides these true crossings, the quasienergy surfaces display avoided crossings. These true crossing and avoided crossings are associated with *dynamical resonances*, as we will show below.

We consider the action of two smooth pulses, associated with Rabi frequencies $\Omega_1(t)$ and $\Omega_2(t)$, which act on the two-level atom with a time delay τ. A sequence of such pulses corresponds to a closed loop in the parameter plane Ω_1 and Ω_2. Figure 2 shows three examples of adiabatic paths depending on the peak amplitudes of the Rabi frequencies and leading to three different final atomic population and photon transfers. Each of the two black curves (labeled a and b) correspond to a sequence of two smooth pulses of equal length T and equal peak Rabi frequencies $\Omega_{\max}$, separated by a delay such that the pulse 1 is switched on before the pulse 2. For curve (a), the shifts of the eigenvalues are smaller than the energy of the first intersections. As a consequence, the path stays on a single surface, and at the end the system returns to the initial state, without any final transfer of photons or of the atomic population.

Curve (b) corresponds to shifts that are larger than the first intersections. The crossing of the first intersection as Ω_1 increases with $\Omega_2=0$ brings the dressed system into the first upper quasienergy surface. Turning on and increasing the amplitude Ω_2 (while Ω_1 decreases) moves the path across this surface. When the second field Ω_2 decreases, the curve crosses an intersection (with $\Omega_1=0$) that brings the system to the third level surface, on which the curve stays until the end of the pulse Ω_2. The transfer state is finally connected to state $|1;1,-1\rangle$: there is no transfer of atomic population, but one ω_2 photon has been absorbed and one ω_1 photon has been emitted at the end of the process. This path has been redrawn as a function of time in Fig. 3(b) [using truncated $\sin^2$ envelopes of length $T=100/\delta$ and delayed by $\tau=T/3$, shown on Fig. 3(a)]. Each of the two dynamical resonances is crossed twice; each appears as one true crossing and one avoided crossing. They can be described as follows: the field 1 dynamically shifts the eigenvalues that become resonant with field 2. This resonance is mute when field 2 is off (left true crossing) and becomes effective when field 2 is on (left avoided crossing). The second dynamical resonance occurs symmetrically from a dynamical Stark shift due to field 2 that makes the eigenvalues resonant with field 1.

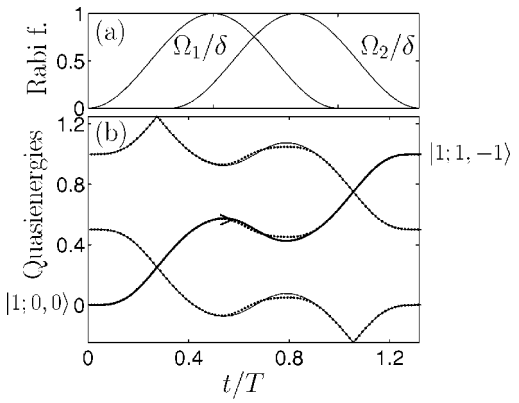

FIG. 3. (a) Rabi frequencies (in units of δ) from squared trig function envelopes. (b) Quasienergy curves (in units of δ), corresponding to the path b of Fig. 2 ($\Omega_{\max}=1.5\delta$) from formula (4) (dotted lines) and the exact numerical result (full line). The arrow indicates the adiabatic path (big line).

If the peak amplitudes are taken even larger, such that two dynamical resonances are crossed (corresponding to the true crossings when Ω_1 rises with $\Omega_2=0$ and when Ω_2 decreases with $\Omega_1=0$), the final state is $|1;2,-2\rangle$, i.e., there is no atomic population transfer but an absorption of two ω_2 photons and an emission of two ω_1 photons. This kind of process can be generalized to paths yielding the connectivity of the transfer state to $|1;k,-k\rangle$, i.e., the emission of k ω_1 photons and the absorption of k ω_2 photons (k positive for pulse 1 before pulse 2 and negative for pulse 2 before pulse 1), with no atomic population transfer. This result is shown in Fig. 4 where we have diagrammed the final average effective number of exchanged photons k as a function of the peak Rabi frequencies, calculated numerically by solving the dressed time-dependent Schrödinger equation. The dips are due to nonadiabatic Landau-Zener transitions when the pulse overlapping is in the neighborhood of the intersections. With a configuration of counterpropagating laser fields, perpendicular to the atomic beam, this translates into the possibility of deflection of the beam by the transfer of a momentum $k\hbar(\omega_1+\omega_2)/c$.

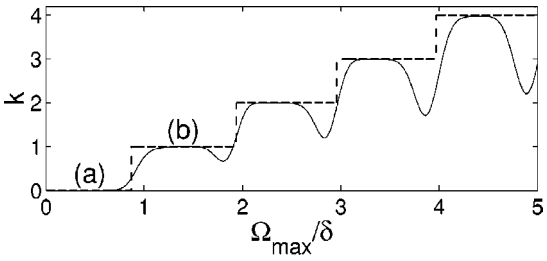

FIG. 4. Comparison of the number k of effective photons emitted at the end of the process [Eq. (5)] (dashed line) with the average number of effective photons from the exact numerical result (full line). The plateaus labeled (a) and (b) refer to the two paths of Fig. 2 for pulse length $T=100/\delta$ and delay $\tau=T/3$.

Inspection of the surface topology shows the possibility of a pulse sequence leading to atomic population transfer in this system. As illustrated by the path (c) in Fig. 2, this process requires two pulses of different peak amplitude. The first resonance is crossed by the raising pulse 1 (with $\Omega_2=0$). The second pulse is chosen with a smaller peak amplitude in order to avoid the passage through the resonance that would lead the system to the third level surface. In this way we obtain a path that ends at $|2;0,-1\rangle$: the atomic population is completely transferred to the excited level, accompanied with absorption of one ω_2 photon. This can also be generalized for upper and lower paths: the connectivity leads to $|2;-1+k,-k\rangle$, with k positive (pulse 1 before smaller pulse-2 amplitude) or negative or zero integer (pulse 2 before smaller pulse-1 amplitude).

The topology of the quasienergy surfaces thus shows which appropriate delays and peak amplitudes induce desired atomic population and photon transfers. In the adiabatic regime, these loops can be classified into topologically inequivalent classes. If the evolution is adiabatic, all paths of a given class lead to the same end effect. This property underlies the robustness of the process.

The explicit consideration of the small perturbative corrections from the full model (2) does not change the topology of the surfaces in the sense that the conical intersections are not removed but only slightly shifted.

IV. ANALYTIC RESULTS

With the technique combining the *rotating wave transformations* (RWT) and *contact transformations* developed in [14], one can treat accurately the dynamical resonances. If we take into account the first two dynamical resonances, that are associated with the path (b), one obtains the following explicit expression for the dressed energy surfaces:

$$\frac{\lambda_{\pm,k}}{\hbar}=\frac{\Delta_1}{2}+k\delta\mp\left[\frac{1}{4}(\sqrt{A}-\delta)^2+\frac{(\varepsilon^2\Omega_1\lambda_-^0)^2}{\hbar^2 A}\right]^{1/2} \tag{4}$$

with $A=\{[(\Delta_1)^2+(\Omega_1)^2]^{1/2}-\delta\}^2+4(\varepsilon\lambda_-^0/\hbar)^2$ and $2\varepsilon=-\Omega_2/\sqrt{(\Delta_1)^2+(\Omega_1)^2}$. Figure 3(b) displays these eigenvalues which are in close agreement with the exact eigenvalues calculated numerically. Figure 3 shows that the formula (4) determines the qualitative feature of the spectrum and especially the connectivity. This systematic method can also be applied to treat the next dynamical resonances occurring for higher field amplitudes.

Since the connectivity of the transfer state to $|1;k,-k\rangle$ is based on the crossings, we can determine analytically the final number of effective photons k as a function of the peak Rabi frequencies $\Omega_{\max}/\delta$ (taken equal) in the purely adiabatic regime:

$$k=[\text{integer part of } \sqrt{(\Omega_{\max}/\delta)^2+(\Delta_1/\delta)^2}]. \tag{5}$$

It predicts the adiabatic plateaux of Fig. 4. They can be interpreted as a topological quantization of the number of exchanged photons.

In conclusion, we remark that the tools presented in this paper can be applied to a large variety of systems and control processes.

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