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Two-laser multiphoton adiabatic passage in the frame of the Floquet theory. Applications to (1+1) and (2+1) STIRAP

S. Guérin[1,a] and H.R. Jauslin[2]

[1] Fachbereich Physik der Universität, 67653 Kaiserslautern, Germany
[2] Laboratoire de Physique, CNRS, Université de Bourgogne, BP 400, 21011 Dijon, France

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Abstract. We develop an adiabatic two-mode Floquet theory to analyse multiphoton coherent population transfer in N-level systems by two delayed laser pulses, which is a generalization of the three-state stimulated Raman adiabatic passage (STIRAP). The main point is that, under conditions of non-crossing and adiabaticity, the outcome and feasibility of a STIRAP process can be determined by the analysis of two features: (i) the lifting of degeneracy of dressed states at the beginning and at the end of the laser pulses, and (ii) the connectivity of these degeneracy-lifted branches in the quasienergy diagram. Both features can be determined by stationnary perturbation theory in the Floquet representation. As an illustration, we study the corrections to the RWA of the (1+1) STIRAP in strong fields and for large detunings. We analyse the possible breakdown of connectivity. In strong fields, the complete transfer is achieved, but the intermediate state, unpopulated within the RWA, can become populated during the process. In the (2+1) STIRAP, we show a residual degeneracy in a four-level system, that can be lifted by additional Stark shifts. The complete transfer is achieved under conditions of connectivity.

PACS. 42.50.Hz Strong-field excitation of optical transitions in quantum systems; multiphoton processes; dynamic Stark shift – 33.80.Be Level crossing and optical pumping – 42.65.Dr Stimulated Raman scattering; CARS

1 Introduction

It is well established that coherent population transfer is more efficient with adiabatic passage techniques than it is with single frequency π-pulse adjustment, since the former requires only adiabaticity as opposed to the strict adjustments of the length and the maximum amplitude of the laser pulse required for the latter process [1,2]. Adiabatic following can be produced by one laser pulse with swept frequency (chirping) [3,4], or by the application of two delayed laser pulses, the so-called STIRAP process (stimulated Raman adiabatic passage) [5–7]. The STIRAP process involves two delayed lasers (called usually the pump and Stokes lasers, which couple an intermediate state respectively to the initial and to the final state, see Fig. 1a). The two possible sequences of the laser pulses are referred to as "intuitive" (first pump and then Stokes laser) or as "counterintuitive" (reverse order). The complete population transfer in a three-level system is well understood in the frame of the rotating-wave approximation (RWA) [7,8]. The method consists in solving approximately the Schrödinger equation within the RWA, which allows one to

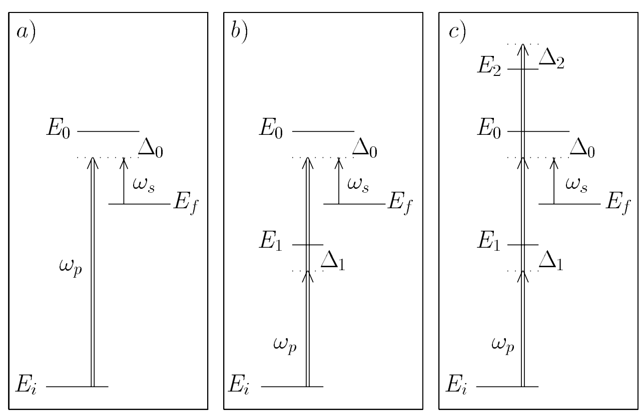

Fig. 1. The different schemes of the studied multiphoton STIRAP studied. a) The (1+1) STIRAP in a three-level system. For the one-photon detuning $\Delta_0 \neq 0$, we have a single-resonance process; for $\Delta_0 = 0$, we have a dual-resonance process. b) The (2+1) STIRAP in a four-level system. Δ_1 is the one-photon detuning to the intermediate level E_1 assisting the two-photon transition between E_i and E_0. c) The (2+1) STIRAP in a five-level system, introducing additional Stark shifts.

[a] e-mail: guerin@physik.uni-kl.de

calculate analytically the approximative dressed-states of the problem. An adiabatic following is then required for a complete population transfer. The calculations have been performed for a pure three-level system [5], a three-level system with other levels near the intermediate and final states [9–11], for a four-level system [12] with multi-photon STIRAP, and for multi-level systems with sequences of three-level STIRAP processes [13]. In the three-level systems, the analytic calculation of the RWA dressed states introduces a mixing angle, which expresses the connection of the dressed states to the initial and final bare states, and which gives a picture of the complete population transfer in the system. From this analysis one can deduce that, if the two laser fields are resonant, (i) the counterintuitive sequence for the pulses is required for the complete population transfer, (ii) the intermediate level is not populated with adiabatic evolution. In some experiments, where the lifetime of the intermediate level is comparable to the pulse length, this latter point is crucial to avoid incoherent losses. The essential point is that the population has to follow adiabatically one dressed state (the *transfer* state) which is connected to the initial state at the beginning of the process and to the final target state at the end. That is to say (i) the *connectivity* of the transfer state between the initial and final states is required, (ii) *the adiabatic passage* on this dressed state must be satisfied [5]. In the usual counterintuitive 3-level RWA, the transfer state is identify as a *trapped* state (it has no component in the intermediate level). It always connects the initial and final states. The connectivity problem has been investigated in absence of trapped state in a multi-level system [11] and for non-zero two-photon detuning in a three-level system [14–16]. Here we study the connectivity problem in absence of trapped state due to strong fields, large detunings (far from resonance) or Stark shifts induced by a multiphoton process for the pump or/and Stokes lasers. Non-adiabatic corrections have been considered in [17,18] and by considering superadiabatic basis in [19].

We present a more general analysis of the STIRAP problem, based on Floquet theory. This analysis goes beyond the RWA because (i) it allows to calculate simply with perturbative arguments the connectivity of the transfer state with the bare states without needing the complete solution of the problem, (ii) it gives exact (in a numerical sense) dressed eigenvalues during the whole process. We remark that item (i) holds also for the RWA (with small detunings) as long as the fields are weak. (In this case, Floquet states are well approximated by the RWA). The obtained corrections to the RWA exhibit cases for which there is not a unique one-to-one correspondence of laser frequencies and transitions, especially for high intensities and multilevel systems, where detunings can be large. The adiabatic Floquet theory for two modes is developed in two distinct steps: (i) We first apply degenerate stationary perturbation theory on Floquet states, which allows one to determine analytically the quasienergies for any multi-level system. The degeneracy breaking is analysed and related to the connectivity. (ii) The second step is the numerical calculation of the quasi-energies. This step produces "exact" dressed states in the sense that the approximations are only numerical and controlable [20]. This step can be used to study the validity of the RWA applied to the whole problem. This method allows one to explore the conditions (*e.g.* position and coupling of the main and auxiliary levels) that are favorable for complete population transfer. We remark that non-zero two-photon detuning can be studied with our approach by the usual extension of degenerate stationary perturbative theory to the quasi-degenerate case.

As an illustration of the approach, we study the STIRAP in a three-level system, and discuss the corrections to the RWA for some limiting cases (high intensities, large detunings). The main result for high intensities is that the transfer state can become non-constant (which was a main feature of the RWA), implying that the intermediate bare state becomes populated during the process, but not preventing the complete transfer (with a non-decaying intermediate level). We also show particular large detunings leading to the breakdown of connectivity.

We work with dimensionless variables (leading to $\hbar = 1$). They can be defined for example through a reference frequency (ω_0): $E \leftarrow E/\hbar\omega_0$, $t \leftarrow \omega_0 t$ and $\Omega_j \leftarrow \Omega_j/\omega_0$, where the Rabi frequency between two levels is defined as $\Omega_j = \mu\alpha_j/\hbar$ with μ the coupling between these two levels and α_j the amplitude of the j^{th} field. For example, if we assume a reference frequency $\omega_0 \sim 6\times10^{15}$ s^{-1} (U.V.), the pulse length of $T_p = 1000/\omega_0$ corresponds to a femtosecond regime ($T_p \sim 200$ fs) and the peak Rabi frequency $\Omega_p = 0.5\omega_0$ gives a peak intensity $I \sim 10^{15}$ W/cm^2. The approximate adiabatic criterion $\Omega_p T_p >> 1$ (deduced from the RWA analysis, if we assume the same Rabi frequencies and the same lengths for the Stokes and pump lasers [5]) can be satisfied for moderate intensities (Rabi frequencies are much smaller than the Bohr frequencies) and nanosecond pulses. In a femtosecond regime, the pulses being much shorter, higher intensities are required for adiabatic following. The predicted effects have been calculated for Rabi frequencies of the order of the Bohr frequencies of the system, implying quite large pulses area (several hundred). This kind of intensities can be experimentally achieved by femtosecond pulses. Of course, these large intensities can surely make other levels (and also continua) become relevant for the precise study of the dynamics. We are here interested by the qualitative deviations of the usual RWA in these extreme (but experimentally reachable) intensities. Short pulses imply that a further characteristic time (besides the pulse area) comes into play, namely the number of oscillations in a pulse: $n_p = T_P/(2\pi/\omega_p)$. The requirement of adiabaticity restricts the minimum length of the pulses in order to have n_p sufficiently large [21] and also restricts their strength so that the time during which the pulses envelope changes appreciably be long compared to $2\pi/\omega_p$. We have checked the adiabaticity with numerical studies involving $n_p \gtrsim 300$ and Rabi frequencies of the order of Bohr frequencies of the system.

We study the (2+1) STIRAP (two photons of the pump field are needed for the transfer) in a four- and five-level system. We show that the complete transfer is not possible in a four-level system except if the detuning involved in the two-photon process is large (far from resonance). Additional Stark shifts in a five-level system can make the complete transfer possible. The analysis of connectivity allows one to determine which types of level structures leads to this complete transfer. We first describe the STIRAP process in the frame of the Floquet theory and then expose the stationnary perturbation theory on Floquet states.

2 Two-mode Floquet formalism

We study a multi-level system with the Hamiltonian H_0 for which the initial population is in the state denoted φ_i (of energy E_i). The aim of the STIRAP process is to populate completely a final state φ_f (of energy E_f), which is not coupled directly with φ_i. This is achieved with the help of one or several intermediate states coupled in general with both φ_i and φ_f. The system is driven by two smooth pulse-shaped monochromatic laser fields, called pump and Stokes lasers. The respective variables will be denoted by the indices p and s. The envelopes, carrier frequencies and initial phases of the fields are respectively denoted: $\underline{\alpha} = (\alpha_p, \alpha_s)$, $\underline{\omega} = (\omega_p, \omega_s)$ and $\underline{\theta} = (\theta_p, \theta_s)$. The Hamiltonian reads

$$H^{\underline{\alpha}(t)}(\underline{\theta} + \underline{\omega}t) = H_0 + \mu\Big[\alpha_p(t)\sin(\theta_p + \omega_p t) + \alpha_s(t)\sin(\theta_s + \omega_s t)\Big], \qquad (1)$$

where H_0 describes the bare system and μ is the dipole moment operator. Denoting t_{0_p} and t_{0_s} the times for which the respective lasers are switched on, for the counterintuitive scheme (the Stokes laser before the pump laser), we have $t_{0_s} = 0$ and $t_{0_p} > 0$ represents the relative delay between the two lasers. The process occurs between the instants $t = 0$ and $t_f = t_{0_p} + T_p$, with T_p the duration of the pump pulse. For the intuitive scheme, we have $t_{0_p} = 0$ and $t_{0_s} > 0$. The process occurs between the instants $t = 0$ and $t_f = t_{0_s} + T_s$, denoting T_s the duration of the Stokes pulse.

To treat the two periodic time-dependences as supplementary degrees of freedom, we use the multi-mode (*i.e.* for two or more incommensurate frequencies) Floquet theory [22], generalizing the usual Floquet theory [23–25]. We construct the multi-mode Floquet theory working with the initial phases $\underline{\theta}$ of the fields [26–29] rather than with time. This conceptually clearer formulation avoids any confusion between the periodic (fast) time with the adiabatic (slow) time in formal developments. This representation is presented in detail for the periodic case in [26] and for the two-mode case in [27,28]. We remark that this representation appears naturally in the derivation of the Floquet Hamiltonian from the fully quantized Hamiltonian in a cavity. The phase $\underline{\theta}$ is closely connected to the phase of the photon field [20].

For a two-laser process, the quasienergy operator reads [26–29]

$$K^{\underline{\alpha}(t)}(\underline{\theta}) = H^{\underline{\alpha}(t)}(\underline{\theta}) - i\underline{\omega}\cdot\frac{\partial}{\partial\underline{\theta}}, \qquad (2)$$

where $\underline{\omega}\cdot\partial/\partial\underline{\theta} = \omega_p\partial/\partial\theta_p + \omega_s\partial/\partial\theta_s$. The quasienergy operator (2) acts on the enlarged space $\mathcal{K} = \mathcal{H}\otimes\mathcal{L}$, where $\mathcal{H}$ is the Hilbert space of the atom or molecule on which H acts and $\mathcal{L} = \mathcal{L}_2(d\theta_p/2\pi)\otimes\mathcal{L}_2(d\theta_s/2\pi)$ with $\mathcal{L}_2(S^1, d\theta_i/2\pi)$ the space of square integrable periodic functions with measure $d\theta_i/2\pi$. The Floquet theory allows one, for fixed values of α_p and α_s, to express the Schrödinger equation with time dependent Hamiltonian $i(\partial/\partial t)\phi = H^{\underline{\alpha}}(\underline{\theta}(t))\,\phi$ in terms of one with a time independent Hamiltonian $K^{\underline{\alpha}}$ (the quasienergy operator (2)): $i(\partial/\partial t)\psi = K^{\underline{\alpha}}(\underline{\theta})\,\psi$. The evolution operator of this latter equation, $\exp[-iK(t-t_0)]$, is connected to that of the original Schrödinger equation $U(t,t_0)$ by

$$\mathcal{T}_{-t}U(t,t_0)\mathcal{T}_{t_0} = e^{-iK(t-t_0)}, \qquad (3)$$

where the translation operator acts on functions $\xi(\underline{\theta})\in\mathcal{L}$ as

$$\mathcal{T}_t\xi(\underline{\theta}) = \xi(\underline{\theta} + \underline{\omega}t). \qquad (4)$$

The eigenvectors $\{\Psi_n(\underline{\theta})\}$ of $K^{\underline{\alpha}}$ (the Floquet states) form a complete orthonormal basis of $\mathcal{K}$ if the spectrum $\{\lambda_n\}$ (the quasienergies) is pure point. We emphasize that, even in the case where H_0 has a finite number of states, the spectrum of K is dense and can be continuous [28]. However, we will consider the spectrum as effectively pure point for the finite durations of the interaction provided by the pulses. This hypothesis is supported by the numerical simulations.

The quasienergies appear in families, which can be labeled by a positive integer m:

$$\lambda_n = \varepsilon_m + \underline{k}\cdot\underline{\omega}, \qquad n \equiv (m, \underline{k}), \qquad \underline{k} \equiv (k_p, k_s)$$

with k_p and k_s integers. Since the spectrum is in general dense, we can in principle reduce all the quasienergies to zone (composed of the set of one member per family for each family) as small as wanted. But numerically we limit the maximum numbers of photons for the two laser fields. This allows one to consider a *relative zone* compared to these maximum photon numbers, where all the dynamics can be described. In all the paper, we use the term "zone", meaning relative zone.

For each value of α_p and α_s, we can expand the solution of the time dependent Schrödinger equation in the basis of Floquet states and apply adiabatic principles for slow variations of the envelopes of the pulses [30]: If at time t_0 the system is an instantaneous Floquet state $\phi(t_0) = \Psi_n^{\underline{\alpha}(t_0)}(\theta(t_0))$, in the adiabatic limit ($T_p\to\infty$ and $T_s\to\infty$) the time evolution $\phi(t)$ stays for all t in an instantaneous Floquet eigenstate:

$$\phi(t) = e^{i\delta_n(t)}\Psi_n^{\underline{\alpha}(t)}(\underline{\theta}(t)) \quad (\delta_n\in\mathbb{R}). \qquad (5)$$

The phase $\delta_n(t)$ is the sum of the dynamical phase and Berry's geometric phase [31]. In our case where two parameters are varied adiabatically to form a closed loop between the instants $t = 0$ and t_f in the parameter space, the Berry phase can be non-zero at the end of the two pulses. But if we consider probabilities involving only one quasienergy branch, it is irrelevant. In the frame of Floquet theory, the quasienergies form surfaces as functions of the two amplitudes α_p and α_s. An optimized trajectory of the quasienergies on these surfaces can lead to complete population transfer. The goal of the following discussion is to find these trajectories to control complete transfer.

3 Degenerate stationary perturbation theory for two-mode Floquet states

In this section, we present the general analysis of the population transfer in a system H_0 driven by two pulse-shaped delayed lasers, resonant with the two unperturbed levels (E_i and E_f), in the sense that $\underline{N} \cdot \underline{\omega} \equiv N_p\omega_p + N_s\omega_s = E_f - E_i$, $\underline{N} = (N_p, N_s)$ with N_p and N_s integers (see Fig. 1 for examples: a) ($N_p = 1, N_s = 1$) STIRAP in a three-level system, ($N_p = 2, N_s = 1$) STIRAP in b) four-level and c) five-level systems). For each time t, the two resonant states give rise to two Floquet families $\left\{\lambda_{i,\underline{k}}^{\underline{\alpha}(t)}, \lambda_{f,\underline{k}}^{\underline{\alpha}(t)}\right\}$ ($\underline{k} = (k_p, k_s)$). In one zone, two Floquet states are degenerate for $\underline{\alpha} = (0,0)$. Other Floquet states can be degenerate, either accidentally during the process at some $\underline{\alpha} \neq (0,0)$, or if there are other resonances for $\underline{\alpha} = (0,0)$. We first consider the case, denoted as a single-resonance process for which only the above two quasienergies are degenerate (*i.e.* the couplings with the intermediate states are non-resonant). Later we will study the case of three degenerate Floquet eigenvalues at the beginning and at the end of the process, denoted as a dual-resonance process. The approach is based on the analysis, by stationary perturbation theory, of the degeneracy breaking at the beginning and at the the end of the pulses. Since the pulses are delayed, the degeneracy breaking at the beginning is determined by one of the lasers and by the other one at the end of the process. Since the frequencies are different, the degeneracies are lifted differently by the two lasers. From this perturbative analysis, we want to obtain conclusions on the feasibility of a complete population transfer. The perturbative analysis allows us to establish whether the following two conditions are satisfied for a given system and a given sequence of the delayed pulses:

(c1) The initial molecular state φ_i is associated to a single quasienergy branch emanating from the degenerate subspace.

(c2) This quasienergy branch, referred as the *transfer state* is connected to the final state φ_f at the end of the pulse.

In order for the conclusions to be valid, we have to assume the two following hypothesis about the non-perturbative regime:

(h1) There are no real crossings involving the transfer quasienergy level as a function of t (except at the initial and final times, when amplitudes are $\underline{\alpha} = 0$).

(h2) The dynamics on the transfer state is mainly adiabatic.

The two hypothesis cannot be checked by perturbative arguments, nor by the RWA in general. In the applications, we verify if they are satisfied by numerical computation of the full quasienergy diagram and by numerical solution of the time dependent Schrödinger equation. Assuming the hypothesis (h1), one can deduce from the perturbative analysis the connectivity properties between the levels at the beginning and at the end of the process. If there are some real crossings involving the transfer quasienergy, an independent analysis is necessary. Under the general assumptions (h1) and (h2), if the two conditions (c1) and (c2) are fulfilled, then the STIRAP process yields a complete population transfer. Beside the analysis of a given system, this approach gives a tool to explore different variations (*e.g.* of level structures, couplings, or laser frequencies and intensities) for which a STIRAP process can be feasible and efficient.

For the single-resonance process, we give a completely explicit formulation. The dual-resonance process is conceptually identical, but for clarity, we do not write down the complete formulas.

A characteristic assumption of the STIRAP process is that the initial and final states are not coupled directly, *i.e.* $\mu_{if} = 0$. We also assume $\mu_{ii} = \mu_{ff} = 0$.

3.1 Single-resonance process

We consider the case where two quasienergies are degenerate at the beginning and at the end of the process, which is produced by one multiphoton resonance in the system (*i.e.* N_p photons of the pump field plus N_s photons of the Stokes laser are resonant with $E_i - E_f$). For a single-resonance process, the relevant eigenvalues are, in one relative zone, $\left\{\lambda_{i,\underline{0}}^{\underline{\alpha}(t)}, \lambda_{f,-\underline{N}}^{\underline{\alpha}(t)}\right\}$, where $\lambda_{i,\underline{0}}^{(0)} = E_i$ and $\lambda_{f,-\underline{N}}^{(0)} = E_f - \underline{N} \cdot \underline{\omega}$ are degenerate at the beginning and at the end of the pulses, ($\alpha_p = \alpha_s = 0$). Because of the degeneracy, the corresponding Floquet states for $\underline{\alpha} = (0,0)$ can be any linear combination of φ_i and $\varphi_f e^{-i\underline{N}\cdot\underline{\theta}}$. The perturbation breaks up the degeneracy. Since the lasers are delayed, the degeneracy breakings are not equivalent at the end ($t = t_f$) and at the beginning ($t = 0$) of the process. We denote the zeroth order basis of linear combinations, adapted to this degeneracy breaking, as

$$\Psi_a^{begin}(\underline{\theta}) = a_i\varphi_i + a_f\varphi_f e^{-i\underline{N}\cdot\underline{\theta}}, \tag{6a}$$

$$\Psi_b^{begin}(\underline{\theta}) = -a_f^*\varphi_i + a_i^*\varphi_f e^{-i\underline{N}\cdot\underline{\theta}}, \tag{6b}$$

for $t = 0$, and as

$$\Psi_a^{end}(\underline{\theta}) = a'_i\varphi_i + a'_f\varphi_f e^{-i\underline{N}\cdot\underline{\theta}}, \tag{7a}$$

$$\Psi_b^{end}(\underline{\theta}) = -{a'_f}^*\varphi_i + {a'_i}^*\varphi_f e^{-i\underline{N}\cdot\underline{\theta}}, \tag{7b}$$

$$W^{(1)} = \begin{pmatrix} \mu_{ii}\int_0^{2\pi} d\theta_j \sin\theta_j \int_0^{2\pi} d\theta_k & \mu_{if}\int_0^{2\pi} d\theta_j\, e^{-iN_j\theta_j}\sin\theta_j \int_0^{2\pi} d\theta_k\, e^{-iN_k\theta_k} \\ \mu_{fi}\int_0^{2\pi} d\theta_j\, e^{iN_j\theta_j}\sin\theta_j \int_0^{2\pi} d\theta_k\, e^{iN_k\theta_k} & \mu_{ff}\int_0^{2\pi} d\theta_j \sin\theta_j \int_0^{2\pi} d\theta_k \end{pmatrix} = 0, \quad (19)$$

for $t = t_f$, with $|a_i|^2+|a_f|^2 = 1$ and $|a'_i|^2+|a'_f|^2 = 1$. When the initial degeneracy is lifted, the two states split into two branches corresponding to the Floquet states $\Psi_a^{\underline{\alpha}(t)}$ and $\Psi_b^{\underline{\alpha}(t)}$ associated to the quasi-energies that we denote $\lambda_a^{\underline{\alpha}(t)}$ and $\lambda_b^{\underline{\alpha}(t)}$. The adiabatic time evolution of the initial condition $\phi(0) = \varphi_i$ is thus

$$\phi(t) = a_i^* e^{i\delta_a(t)}\Psi_a^{\underline{\alpha}(t)}(\underline{\theta}+\underline{\omega}t) - a_f^* e^{i\delta_b(t)}\Psi_b^{\underline{\alpha}(t)}(\underline{\theta}+\underline{\omega}t). \quad (8)$$

At the end of the pulses $t = t_f$, the degeneracy of the two Floquet states appears again and we can develop the state at this time in terms of the two bare states:

$$\phi(t_f) = e^{i\delta_a(t_f)}\Big\{\Big[a_i^* a'_i + {a'_f}^* e^{i(\delta_b(t_f)-\delta_a(t_f))}\Big]\varphi_i + e^{-i\underline{N}\cdot(\underline{\theta}+\underline{\omega}t_f)}\Big[a_i^* a'_f - a_f {a'_i}^* e^{i(\delta_b(t_f)-\delta_a(t_f))}\Big]\varphi_f\Big\}, \quad (9)$$

and obtain the probabilities of inversion from φ_i to φ_f and of staying in φ_i:

$$P_{i\to f}(t_f) = \Big|a_i^* a'_f - a_f {a'_i}^* e^{i(\delta_b(t_f)-\delta_a(t_f))}\Big|^2, \quad (10a)$$

$$P_{i\to i}(t_f) = \Big|a_i^* a'_i + a_f {a'_f}^* e^{i(\delta_b(t_f)-\delta_a(t_f))}\Big|^2. \quad (10b)$$

If the two pulses are symmetric and not delayed, then $a_i = a'_i$ and $a_f = a'_f$, and the transition probability becomes

$$P_{i\to f}(t_f) = 4|a_i a_f|^2 \sin^2\left[\frac{1}{2}(\delta_a - \delta_b)\right]. \quad (11)$$

It is dependent of the path of the quasienergies and of the associated Berry phases. Otherwise, the complete transition is achieved, under the hypothesis of *adiabatic passage*, if one of the two following conditions is satisfied:

$$|a_i| \simeq 1 \text{ and } a_f \simeq 0,\ a'_i \simeq 0 \text{ and } |a'_f| \simeq 1, \quad (12a)$$

$$\text{or} \quad a_i \simeq 0 \text{ and } |a_f| \simeq 1,\ |a'_i| \simeq 1 \text{ and } a'_f \simeq 0, \quad (12b)$$

are fulfilled. We have here replaced the "equal" signs by "approximately equal" to keep in mind that we have non-adiabatic corrections (due to the finite length of pulses and to contibutions of other levels). The case (12a) means that, the zeroth order Floquet branch Ψ_a^{begin} being connected to the initial state: $\Psi_a^{begin} \equiv \varphi_i$, the complete population transfer from φ_i to φ_f by adiabatic passage requires that the zeroth order Floquet branch Ψ_a^{end} must be connected to φ_f: $\Psi_a^{end} \propto \varphi_f$. The case (12b) is similar, except that the population evolves on the branch $\Psi_b^{\underline{\alpha}(t)}$, its zeroth order beginning on φ_i: $\Psi_b^{begin} \equiv \varphi_i$ to end on φ_f: $\Psi_b^{end} \propto \varphi_f$. Each of these two cases corresponds to a different order of the sequence of the two delayed laser pulses, equations (12a) and (12b) are usually referred as the "intuitive" and "counterintuitive" laser sequences [5,32].

We apply the stationary perturbation method to calculate the degeneracy breaking of the Floquet states. The initial degenerate states φ_i and $\varphi_f e^{-i\underline{N}\cdot\underline{\theta}}$ (for $\underline{\alpha} = (0,0)$) generate a two-dimensional subspace $\mathcal{S}_0$ of the enlarged space $\mathcal{K}$. We denote K^{α_j} the α_j-dependent quasi-energy operator, corresponding to non-zero amplitude of the field α_j whith the other field kept at zero amplitude:

$$K^{\alpha_j} = K_0 + \alpha_j \hat{W}_j\,, \quad j = p, s, \quad (13)$$

with

$$K_0 := -i\underline{\omega}\cdot\frac{\partial}{\partial\underline{\theta}} + H_0 \quad (14)$$

and

$$\hat{W}_j := \mu\sin\theta_j \quad (15)$$

acting on $\mathcal{K}$ (we will omit the index j in $\hat{W}_j$, to simplify the notation). The eigenvalue problem $K^{\alpha_j}\Psi^{\alpha_j} = \lambda^{\alpha_j}\Psi^{\alpha_j}$ is solved by the perturbation method, *i.e.* in terms of powers of the small amplitude α_j:

$$\lambda^{\alpha_j} = \lambda^{(0)} + \alpha_j\lambda^{(1)} + \alpha_j^2\lambda^{(2)} + \cdots \quad (16)$$

$$\Psi^{\alpha_j} = |0\rangle + \alpha_j|1\rangle + \alpha_j^2|2\rangle + \cdots \quad (17)$$

where $|0\rangle \in \mathcal{S}_0$ represents the unknown linear combinations (6) and (7) in the zeroth order subspace generated by the two degenerate Floquet states

$$\big\{|\phi_i\rangle := |\varphi_i \otimes 1\rangle\,,\ |\phi_f\rangle := |\varphi_f \otimes e^{-i\underline{N}\cdot\underline{\theta}}\rangle\big\}$$

and $\lambda^{(0)} = E_i = E_f - \underline{N}\cdot\underline{\omega}$. The first order gives the eigenvalue problem restricted to the zeroth order subspace

$$\hat{W}^{(1)}|0\rangle = \lambda^{(1)}|0\rangle \quad (18)$$

with $\hat{W}^{(1)} = P_0\hat{W}P_0$, where P_0 is the projector on the zeroth order subspace. Written in the basis $\{|\phi_i\rangle\,,\ |\phi_f\rangle\}$, this operator is

See equation (19) *above*

and thus $\lambda^{(1)} = 0$. As a consequence, for any $N_p \neq 0$ and $N_s \neq 0$, the degeneracy is still present at the first order.

The second order eigenvalue problem restricted to the zeroth order subspace $\mathcal{S}_0$ is [33]

$$\hat{W}^{(2)}|0\rangle = \lambda^{(2)}|0\rangle, \quad (20)$$

with

$$\hat{W}^{(2)} = -P_0 \hat{W} Q_0 \left(K_0 - \lambda^{(0)}\right)^{-1} Q_0 \hat{W} P_0, \quad (21)$$

where $Q_0 = \mathbb{1} - P_0$. The eigenvalue problem (20) gives the second order correction of the eigenvalues and the associated zeroth order eigenvectors. The matrix elements of $W^{(2)}$ in the basis $\{|\phi_i\rangle, |\phi_f\rangle\}$ are given by $(\nu, \mu \in \{i, f\})$

$$W^{(2)}_{\nu\mu} = \sum_{(m,\underline{k})\neq\{(i,0,0),(f,-N_p,-N_s)\}} \frac{1}{E_i - \lambda^{(0)}_{m,\underline{k}}} \times \langle\phi_\nu|\hat{W}|\varphi_m e^{i\underline{k}\cdot\underline{\theta}}\rangle_{\mathcal{K}} \langle\varphi_m e^{i\underline{k}\cdot\underline{\theta}}|\hat{W}|\phi_\mu\rangle_{\mathcal{K}}, \quad (22)$$

which we can write more explicitly as

$$W^{(2)}_{\nu\nu} = \frac{1}{4}\sum_m |\mu_{\nu m}|^2 \times \left[\frac{1}{E_\nu - E_m + \omega_j} + \frac{1}{E_\nu - E_m - \omega_j}\right], \quad (23a)$$

$$W^{(2)}_{if} = 0, \quad (23b)$$

where $j \equiv p$ (resp. $j \equiv s$) for $\alpha_p \neq 0$ (resp. $\alpha_s \neq 0$) and $\alpha_s = 0$ (resp. $\alpha_p = 0$). The index m labels all the unperturbed levels of the system. The degeneracy is always lifted at this second order (barring exceptional cases). The eigenvalues λ_i and λ_f are thus

$$\lambda_i = E_i + W^{(2)}_{ii}\alpha_j^2 + \mathcal{O}\left(\alpha_j^3\right), \quad (24a)$$

$$\lambda_f = E_i + W^{(2)}_{ff}\alpha_j^2 + \mathcal{O}\left(\alpha_j^3\right). \quad (24b)$$

It is important to remark that the matrix $W^{(2)}$ is diagonal. Thus its eigenvectors are precisely $|\phi_i\rangle$ and $|\phi_f\rangle$, which are connected respectively to φ_i and to φ_f. The complete population transfer is in principle possible if the relevant Floquet state, connected to φ_i at the beginning of the process is connected to φ_f at the end to the process. But there can be deviation from adiabaticity (h2) if the perturbative regime does not lift the degeneracy fast enough compared to the speed of the process. This problem of matching between the perturbative and adiabatic regimes will be the subject of a forthcoming work. Here, we restrict to the situations in which the degeneracy is lifted fast enough by intermediate levels. This assumption will be checked by numerical simulations. Moreover, the presence of these intermediate levels are also required so that the initial and final states are coupled significantly, in order to satisfy the hypothesis of non-crossing (h1) during the whole process. Under the conditions of non-crossing (h1) and adiabaticity (h2), the complete population transfer from φ_i to φ_f, satisfying equations (12a) or equations (12b), requires only one of the following condition of connectivity:

$$\text{If} \quad \lambda_i^{begin} > \lambda_f^{begin} \quad \text{then} \quad \lambda_f^{end} > \lambda_i^{end}, \quad (25a)$$

$$\text{or if} \quad \lambda_i^{begin} < \lambda_f^{begin} \quad \text{then} \quad \lambda_f^{end} < \lambda_i^{end}. \quad (25b)$$

3.2 Dual-resonance process

We now consider the case where three quasienergies are degenerate at the beginning and at the end of the process, which is produced by two resonances in the system (*i.e.* the pump laser is resonant with $E_i - E_0$ and the Stokes laser with $E_f - E_0$) :

$$\underline{N}\cdot\underline{\omega} \equiv N_p\omega_p + N_s\omega_s = E_f - E_i, \quad (26a)$$

$$\underline{N}_i\cdot\underline{\omega} \equiv N_i\omega_p + N'_i\omega_s = E_0 - E_i, \quad (26b)$$

$$\underline{N}_f\cdot\underline{\omega} \equiv N_f\omega_p + N'_f\omega_s = E_f - E_0, \quad (26c)$$

$$N_i + N_f = N_p \quad N'_i + N'_f = N_s. \quad (26d)$$

We now consider the zone around E_0, *i.e.* $\lambda^{(0)}_{0,0,0} \equiv E_0$. The relevant eigenvalues are thus

$$\left\{\lambda^{\underline{\alpha}(t)}_{i,\underline{N}_i}, \lambda^{\underline{\alpha}(t)}_{f,-\underline{N}_f}, \lambda^{\underline{\alpha}(t)}_{0,0,0}\right\}.$$

The zeroth order subspace $\mathcal{S}_0$ is now generated by the three Floquet states

$$\left\{|\phi_i\rangle := |\varphi_i \otimes e^{i\underline{N}_i\cdot\underline{\theta}}\rangle, |\phi_0\rangle := |\varphi_0\rangle, |\phi_f\rangle := |\varphi_f \otimes e^{-i\underline{N}_f\cdot\underline{\theta}}\rangle\right\}.$$

In this zeroth order basis, the representation of the first order operator $\hat{W}^{(1)} = P_0\hat{W}P_0$ is:

$$W^{(1)} = \begin{pmatrix} 0 & W^{(1)}_{i0} & 0 \\ W^{(1)}_{0i} & 0 & W^{(1)}_{0f} \\ 0 & W^{(1)}_{f0} & 0 \end{pmatrix}, \quad (27)$$

with

$$W^{(1)}_{0i} = \left(W^{(1)}_{i0}\right)^* = \frac{\mu_{0i}}{2i}\Big[\delta_{j,p}\delta_{N'_i,0}\big(\delta_{N_i,-1} - \delta_{N_i,1}\big) + \delta_{j,s}\delta_{N_i,0}\big(\delta_{N'_i,-1} - \delta_{N'_i,1}\big)\Big], \quad (28a)$$

$$W^{(1)}_{f0} = \left(W^{(1)}_{0f}\right)^* = \frac{\mu_{f0}}{2i}\Big[\delta_{j,p}\delta_{N'_f,0}\big(\delta_{N_f,-1} - \delta_{N_f,1}\big) + \delta_{j,s}\delta_{N_f,0}\big(\delta_{N'_f,-1} - \delta_{N'_f,1}\big)\Big]. \quad (28b)$$

(We remind that $j = p, s$ depending on whether the analysed degeneracy lifting is produced by the pump or Stokes laser.) Thus, this matrix is non-zero only for the (1+1) STIRAP case, for which $N_i = 1$, $N'_i = 0$, $N_f = 0$ and $N'_f = -1$, leading to

$$W^{(1)}_{0i} = \left(W^{(1)}_{i0}\right)^* = -\frac{\mu_{0i}}{2i}\delta_{j,p}, \quad (29a)$$

$$W^{(1)}_{f0} = \left(W^{(1)}_{0f}\right)^* = \frac{\mu_{f0}}{2i}\delta_{j,s}. \quad (29b)$$

The degeneracy is then lifted linearly. Since this matrix is not diagonal, the condition (c1) is not always satisfied. It is satisfied for a certain order of the laser sequences (counterintuitive sequence [5]).

In the other $(N_p + N_s)$ STIRAP cases, we have $\hat{W}^{(1)} \equiv 0$ and the degeneracy is not lifted by the first order. The second order is needed (Eqs. (20) and (21)). The elements of the matrix $W^{(2)}$ are given by the formula (22) for $\nu, \mu \in \{i, 0, f\}$ and $(m, \underline{k}) \notin \{(i, \underline{N}_i), (0,0,0), (f, -\underline{N}_f)\}$ and where E_i is substituted by E_0. This gives for the diagonal elements

$$W^{(2)}_{\nu\nu} = \frac{1}{4}\sum_m |\mu_{\nu m}|^2 \left[\frac{1}{E_\nu - E_m + \omega_j} + \frac{1}{E_\nu - E_m - \omega_j}\right], \quad \nu \in \{i, 0, f\}, \tag{30}$$

where $j \equiv p$ (resp. $j \equiv s$) for $\alpha_p \neq 0$ (resp. $\alpha_s \neq 0$) and $\alpha_s = 0$ (resp. $\alpha_p = 0$). The off-diagonal elements, given by (22) are not zero in general. To study the lifting of the degeneracy, the matrix $W^{(2)}$ has to be analysed for each particular case.

The complete transition requires also in this case a condition of connectivity. For example, if the middle Floquet state, connected to the initial unperturbed state, is populated at the beginning of the process, then the Floquet state connected to the final unperturbed state, at the end of the process, has to be also the middle one.

4 The $(1+1)$ STIRAP. Corrections to RWA

In this section, we reinterpret the well-known (1+1) STIRAP process, yielding the complete population transfer in a three-level system, illustrating the above framework of adiabatic Floquet theory (see Fig. 1a). We compare in detail the single-resonance process [32] with the dual-resonance process [5]. The degeneracy is lifted linearly with the field amplitude in the latter case (first order) whereas it is quadratically lifted in the former case (second order). We study the effects of the corrections to the RWA to the connectivity problem and to the adiabaticity of the processes. We first remark that within the RWA analysis, the connectivity is always satisfied, for single- and dual-resonance processes. Here we obtain that for the dual-resonance process, the connectivity is also always satisfied, but that the adiabaticity can be affected for strong fields. We show a numerical example with complete population transfer in strong fields. Within the RWA analysis, the intermediate state is never populated during the process. The corrections to the RWA show that this intermediate state becomes populated during the process in strong fields. For the single-resonance process, the connectivity can be affected by the corrections, even for moderate fields, in the following particular cases: (i) The detuning to the intermediate level Δ_0 is comparable to the frequency difference $|\omega_p - \omega_s|$; (ii) The detuning Δ_0 is large in the sense that it is of the order of the frequency of one laser $-\omega_p$ or $-\omega_s$; (iii) The frequencies ω_p and ω_s are close. We examine some cases, showing the possible breakdown of connectivity and its consequences to the dynamics.

The intermediate state for the single-resonance process is denoted φ_0. For the dual-resonance process, φ_0 is resonant with the other states. Thus for the (1+1) STIRAP process, the single-resonance process can be referred to as a two-photon resonance, and the dual-resonance process to as two one-photon resonances. The intuitive process refers to an interaction in which the pump laser pulse arrives first, while in the counterintuitive process the Stokes pulse arrives first. We develop numerical simulations with energy levels $E_i = -2$ and $E_f = -0.39$, and the frequencies: $\omega_p = 2$ and $\omega_s = 0.39$. They are chosen so that for a rather small number of photons, the effects of the two frequencies are distinguishable. The coupling strengths are $\mu_{i0} = 0.25$ and $\mu_{f0} = 0.5$. We study interactions leading to the same peak Rabi frequencies for the pump and Stokes fields in order to have in all cases the optimum adiabatic passage for a given delay. We study short pulse lengths: $T_p = T_s = 1000$, with the counterintuitive or intuitive delays $t_{0_p} = 400$ or $t_{0_s} = 400$ (femtosecond regime). The pulses are taken of the form

$$\alpha_j(t) = \alpha_{m_j} \sin^2\left(\frac{\pi}{T_j}(t - t_{0_j})\right), \quad t_{0_j} \leq t \leq t_{0_j} + T_j, \quad j = p, s. \tag{31}$$

4.1 Dual-resonance process

We first consider the case where φ_0 is resonant with the initial and final states. This gives three degenerate Floquet states

$$\left\{|\phi_i\rangle := |\varphi_i \otimes e^{i\theta_p}\rangle, |\phi_0\rangle := |\varphi_0\rangle, |\phi_f\rangle := |\varphi_f \otimes e^{i\theta_s}\rangle\right\},$$

associated to the three identical quasienergies $\lambda^{(0)}_{i,1,0} = E_i + \omega_p$, $\lambda^{(0)}_{0,0,0} = E_0$, and $\lambda^{(0)}_{f,0,1} = E_f + \omega_s$. This generates a 3-dimensional zeroth order subspace $\mathcal{S}_0$. In this basis, the representation of the first order operator $\hat{W}^{(1)} = P_0 \hat{W} P_0$, for $\alpha_p \equiv 0$ and $\alpha_s \neq 0$, is given by equation (27) with $W^{(1)}_{0i} = 0$ (29a) and $W^{(1)}_{f0} = \mu_{f0}/2i$ (29b). It gives the three different first order contributions $\lambda^{(1)}$ to the eigenvalues (16) (denoted with the index s)

$$\lambda^{(s)}_a = -\frac{1}{2}|\mu_{0f}|, \quad \lambda^{(s)}_b = 0, \quad \lambda^{(s)}_c = \frac{1}{2}|\mu_{0f}|. \tag{32}$$

The degeneracy is thus linearly lifted with the field amplitude. This gives rise to three different branches. We determine the three zeroth order orthonormal eigenstates associated to these branches:

$$\begin{aligned} \Psi^{(s)}_a &= \frac{1}{\sqrt{2}}\left(\varphi_0 + i\varphi_f e^{i\theta_s}\right), \\ \Psi^{(s)}_b &= \varphi_i e^{i\theta_p}, \\ \Psi^{(s)}_c &= \frac{1}{\sqrt{2}}\left(i\varphi_0 + \varphi_f e^{i\theta_s}\right). \end{aligned} \tag{33}$$

For $\alpha_s \equiv 0$ and $\alpha_p \neq 0$, the first order matrix $W^{(1)}$ is given by equation (27) with $W^{(1)}_{0i} = -\mu_{0i}/2i$ (29a) and $W^{(1)}_{f0} = 0$ (29b), which gives the three different first order contributions to the eigenvalues (16)

$$\lambda^{(p)}_a = -\frac{1}{2}|\mu_{i0}|, \quad \lambda^{(p)}_b = 0, \quad \lambda^{(p)}_c = \frac{1}{2}|\mu_{i0}|. \tag{34}$$

The degeneracy is again lifted linearly. The three zeroth order orthonormal eigenstates are

$$\Psi_a^{(p)} = \frac{1}{\sqrt{2}}\left(i\varphi_i e^{i\theta_p} + \varphi_0\right),$$
$$\Psi_b^{(p)} = \varphi_f e^{i\theta_s},$$
$$\Psi_c^{(p)} = \frac{1}{\sqrt{2}}\left(\varphi_i e^{i\theta_p} + i\varphi_0\right). \qquad (35)$$

For $\alpha_p \equiv 0$ and $\alpha_s \to 0$, equations (33) imply that the instantaneous Floquet vector $\Psi_b^{\underline{\alpha}(t)}$, corresponding to the quasienergy λ_b in the middle, is connected to φ_i. For $\alpha_s \equiv 0$ and $\alpha_p \to 0$, equations (35) imply that this Floquet vector $\Psi_b^{\underline{\alpha}(t)}$ is connected to φ_f, if no real crossings involve it. This means that only the counterintuitive sequence for the laser fields leads to the complete population transfer for any shape of the lasers, provided that the passage is adiabatic and that no real crossings involve the followed Floquet state [5]. The obtained asymptotic first order eigenvalues are identical to the first order RWA eigenvalues. Thus the connectivity is well reproduced by the RWA, and the RWA quasienergies are correct at the first order of the field amplitude.

Numerical simulations. The system (with $E_0 = 0$) is perturbed by the fields of strong peak amplitudes: $\alpha_p = 2$ and $\alpha_s = 1$, giving the same peak Rabi frequencies $\Omega_p \equiv \mu_{i0}\alpha_p = 0.5$, $\Omega_s \equiv \mu_{f0}\alpha_s = 0.5$. We show a numerical calculation of the quasienergies (Fig. 2a). They are compared to the approximative RWA dressed energies. We have used 7-point discretizations for both fields, which are sufficient for convergence. On this diagram, only the three relevant quasienergies have been plotted. The RWA eigenvalues are qualitatively correct: the lifting of the degeneracy and thus the connectivity are well reproduced. We can observe only small corrections for high field amplitudes, these are due to the second and higher order effects which are not taken into account by RWA (They are due to the counter-rotating terms). These effects are analysed in detail in the following, for the single-resonance process, for which the degeneracy is lifted by the second order of the field amplitude. On this diagram, we compare numerical simulations with RWA and the full Hamiltonian for the counterintuitive process (Figs. 2b and c, with $P_{i\to n}(t) = |\langle\varphi_n|\phi(t)\rangle|^2$, $n = i, 0, f$): we observe that the corrections although changing the population during the process do not significantly affect the final populations: the population transfer is complete. One important difference is that the intermediate state during the process is populated. Within the RWA analysis, the transfer state is identical to the so-called *trapped state* [11], which is a linear combination of only the initial and final bare states. This implies that the intermediate state is never populated, except for adiabatic corrections [5]. The trapped quasienergy is a constant eigenvalue (thus independent of the field amplitudes). The deviations of the transfer quasienergy from the RWA trapped quasienergy is an indication that the transfer state gets a projection on the intermediate state during the process. This interprets the non negligible population on the intermediate state during the process, as shown in Figure 2c. Thus for the dual-resonance process, the criterion for adiabatic following can be affected by second and higher order corrections to RWA, but not the connectivity.

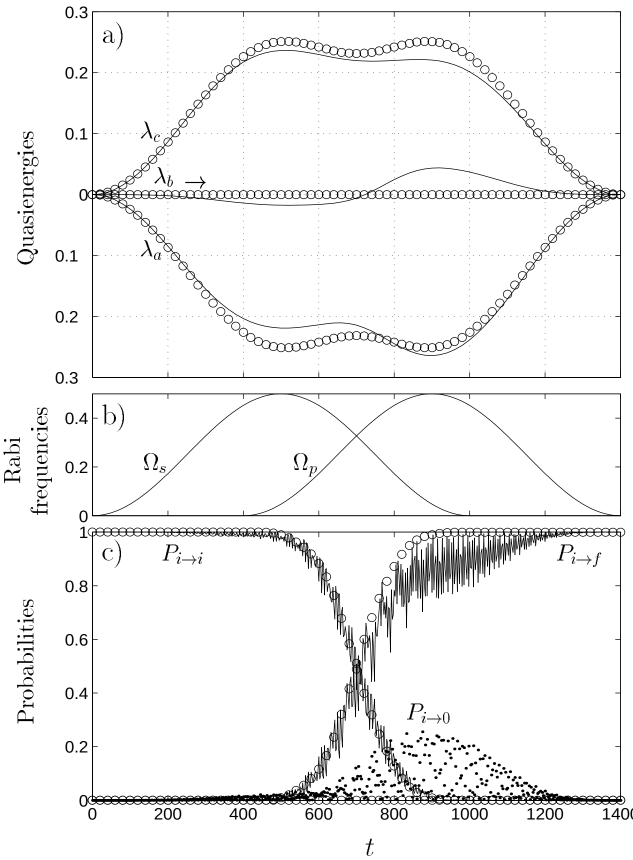

Fig. 2. The (1+1) STIRAP in strong field with a dual resonance ($\omega_p = 2$, $\omega_s = 0.39$, $\Delta_0 = 0$, $\Omega_{max} = 0.5$, $T_p = T_s = 1000$ and $t_{0_p} = 400$). a) The instantaneous exact (solid lines) and RWA (circle lines) quasienergies. The single arrow indicates the transfer state (λ_b). The deviation of the transfer state from the constant posisiton implies the non-zero population in the intermediate state during the process. b) The delayed instantaneous Rabi frequencies of the Stokes laser (Ω_s) and of the pump laser (Ω_p). c) Numerical simulation of the population transfer for the counterintuitive process: the exact solution (solid lines) and the RWA solution (circle lines). Population transfer is complete in both cases at the end of the process, although the intermediate solution is quite different.

We remark that, for the pulse lengths we used, the adiabatic error we observed is always very small (less than 1% of the total probability). It seems to be even smaller than the numerical error due to truncation of the basis to discretize the variables $\underline{\theta}$.

In Figure 3, we show the transfer Floquet state projected in the bare state basis (with the parameter $\underline{\theta} = 0$):

$$\mathcal{P}_n(t) = \left|\left\langle\varphi_n\middle|\Psi_T^{\underline{\alpha}(t)}(\underline{\theta}(t))\right\rangle\right|^2, \quad n = i, f, 0. \qquad (36)$$

We compare $\mathcal{P}_n(t)$ with the numerical solution projected in the bare state basis: $P_{i\to n}(t)$. The $\mathcal{P}_n$ fit the extrema

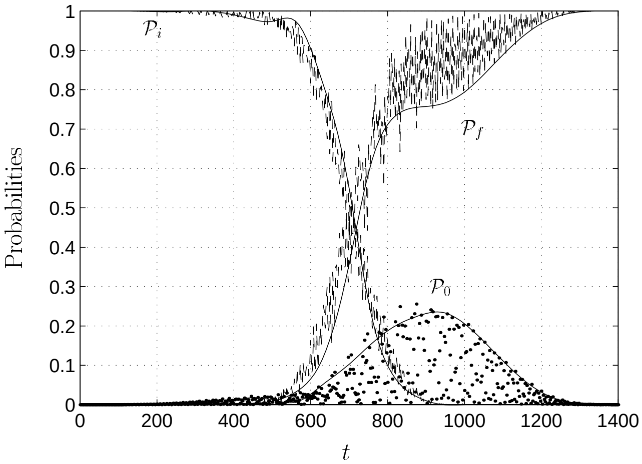

Fig. 3. Transfer Floquet state projected in the bare state basis $\mathcal{P}_n$ (solid line) and compared with $P_{i\to n}$, $n = i, 0, f$ (dashed line and dots, they come from the previous figure). The projections $\mathcal{P}_n$ approximately fit the extrema of $P_{i\to n}$.

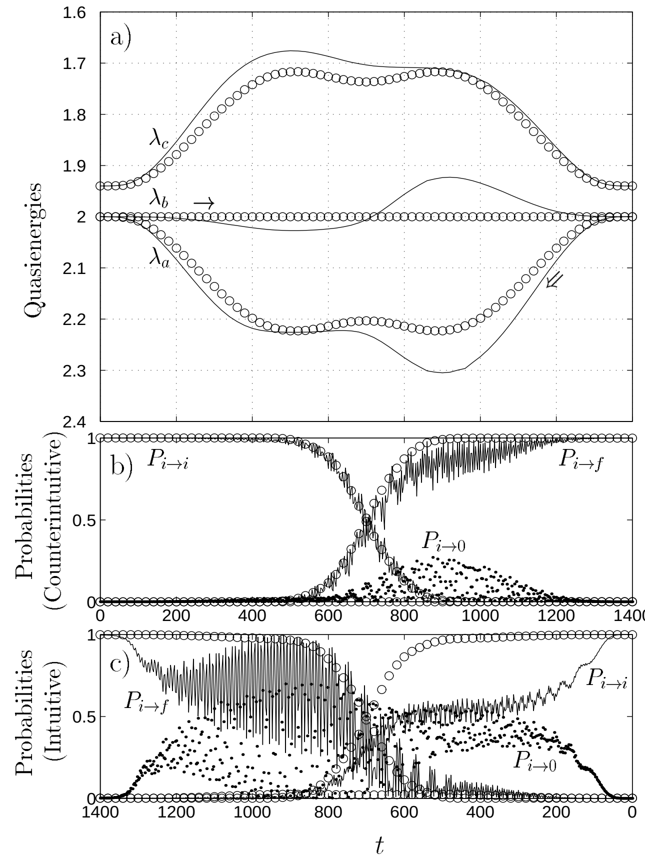

Fig. 4. The (1+1) STIRAP in strong field with one single resonance ($\omega_p = 2$, $\omega_s = 0.39$, $\Delta_0 = 0.06$, $\Omega_{max} = 0.5$ and $T_p = T_s = 1000$). a) The instantaneous exact quasienergies (solid lines) and RWA quasienergies (circle lines). The double arrow indicates the transfer state (λ_a) for the intuitive process ($t_{0_s} = 400$). The small deviations are essentially due to Stark shifts. b) Numerical simulation of the population transfer for the counterintuitive process ($t_{0_p} = 400$). Population transfer is complete. c) Numerical simulation of the population transfer for the intuitive process. Population transfer is complete, the intermediate state is highly populated during the process.

of $P_{i\to n}(t)$ approximately (the undesirable oscillations of the numerical evolution have disappeared). This shows the relevance of the Floquet basis to describe the evolution of the system. The transfer Floquet state determines to which extent the intermediate state is populated during the process.

4.2 Single-resonance process

We have seen that the first order of the perturbation theory does not lift the degeneracy, but that the second order can do it if $\hat{W}^{(2)}_{ii}$ is significantly different from $\hat{W}^{(2)}_{ff}$ (Eq. (23a)), that is if a level E_0 is such that $E_0 \approx E_i + \omega_p \approx E_f + \omega_s$. We denote Δ_0 the detuning between the intermediate level and the initial and the final levels:

$$\Delta_0 \equiv E_0 - (E_i + \omega_p) = E_0 - (E_f + \omega_s). \qquad (37)$$

We define Δ_p and Δ_s as the detunings respectively of the pump laser on the transition between E_f and E_0 and of the Stokes laser on the transition between E_i and E_0:

$$\Delta_p \equiv E_0 - (E_f + \omega_p) = \Delta_0 + \omega_s - \omega_p, \qquad (38a)$$

$$\Delta_s \equiv E_0 - (E_i + \omega_s) = \Delta_0 + \omega_p - \omega_s. \qquad (38b)$$

For any position of E_0 compared to $E_i + \omega_p$ and $E_f + \omega_s$, the connectivity condition (25a) is generally satisfied with the RWA analysis [5]. We remark that now the second order corrections to the RWA appear at the same order as the lifting of degeneracy. This means that connectivity can be affected by the corrections contrary to the dual-resonant case. In the general context (for which the connectivity is satisfied), the complete transfer from φ_i to φ_f requires only the condition of adiabatic following, for both counterintuitive and intuitive sequences of the laser pulses.

As an example, we analyse the system with the "small" detuned intermediate level $E_0 = 0.06$, in the sense that it preserves the connectivity as we will see. We show a numerical calculation of the quasienergies, which are compared to the RWA dressed states (Fig. 4a). This quasienergy diagram can interpret both the counterintuitive and the intuitive processes, depending on the direction it is read. The degeneracy breaking in this quasienergy diagram can be compared to the degeneracy breaking of the previous one (Fig. 2a). The lifting of degeneracy here is slower, since it appears in second order of amplitude, as opposed to second order in the single-resonance case. We denote the upper resonant quasienergy by λ_a and the lower one by λ_b. λ_a (resp. λ_b) corresponds to the Floquet branch $\Psi_a^{\alpha(t)}$ (resp. $\Psi_b^{\alpha(t)}$) connected to the state φ_i (resp. φ_f), for $\alpha_{m_s} \gtrsim 0$ and $\alpha_{m_p} = 0$ (the left part of the diagram 4a). We determine the second order quasienergies (24), for this part:

$$\lambda_a^{(s)} = E_i - \frac{1}{4}|\mu_{f0}|^2\left(\frac{1}{\Delta_0}+\frac{1}{\Delta_0+2\omega_s}\right)\alpha_s^2+\mathcal{O}(\alpha_s^3), \quad (39a)$$

$$\lambda_b^{(s)} = E_i - \frac{1}{4}|\mu_{i0}|^2\left(\frac{1}{\Delta_0+\omega_p-\omega_s}+\frac{1}{\Delta_0+\omega_p+\omega_s}\right)\alpha_s^2 +\mathcal{O}(\alpha_s^3), \quad (39b)$$

and, for $\alpha_{m_p} \gtrsim 0$ and $\alpha_{m_s} = 0$ (the right part of the diagram 4a),

$$\lambda_a^{(p)} = E_i - \frac{1}{4}|\mu_{i0}|^2\left(\frac{1}{\Delta_0}+\frac{1}{\Delta_0+2\omega_p}\right)\alpha_p^2+\mathcal{O}(\alpha_p^3), \quad (40a)$$

$$\lambda_b^{(p)} = E_i - \frac{1}{4}|\mu_{0f}|^2\left(\frac{1}{\Delta_0+\omega_s-\omega_p}+\frac{1}{\Delta_0+\omega_s+\omega_p}\right)\alpha_p^2 +\mathcal{O}(\alpha_p^3). \quad (40b)$$

For the right part, the Floquet branch $\Psi_a^{\underline{\alpha}(t)}$ (resp. $\Psi_b^{\underline{\alpha}(t)}$) is connected to the state φ_f (resp. φ_i). The condition of connectivity is

$$\text{If} \quad \lambda_a^{(s)} > \lambda_b^{(s)} \ (\text{resp. } \lambda_a^{(s)} < \lambda_b^{(s)})$$
$$\text{then} \quad \lambda_a^{(p)} > \lambda_b^{(p)} \ (\text{resp. } \lambda_a^{(p)} < \lambda_b^{(p)}). \quad (41)$$

These quasienergies have to be compared to the initially degenerate RWA quasienergies, which we write and develop into the second order (for the part where the Rabi frequencies are much smaller than the detuning Δ_0):

$$\lambda_a^{(p)} = E_i + \frac{\Delta_0}{2}\left[1-\sqrt{1+(\mu_{i0}\alpha_p/\Delta_0)^2}\right] = E_i - \frac{1}{4}\frac{|\mu_{i0}|^2}{\Delta_0}\alpha_p^2+\mathcal{O}(\alpha_p^4), \quad (42a)$$

$$\lambda_b^{(p)} = E_i. \quad (42b)$$

The corrections are of two types: The first one is due to the detuning Δ_0 which produces a *two-photon anti-resonant* term in addition to the resonant term. This correction holds only for a detuning non negligible compared to the Bohr frequencies. The second type of corrections is due to *Stark shifts* produced by the pump and Stokes laser fields on respectively the transitions φ_f/φ_0 and φ_i/φ_0 (with respect to the respective detunings Δ_p and Δ_s (38)). For a sufficiently small detuning Δ_0, we have, from equations (39) and (40), $|\lambda_b^s| >> |\lambda_a^s|$ and $|\lambda_b^p| >> |\lambda_a^p|$, with λ_b^s and λ_b^p of the same sign. This means that the connectivity (41) is thus satisfied in this case. For the counterintuitive process, the population follows adiabatically the quasienergy branch λ_a (Fig. 4b). For the intuitive process, it follows the quasienergy branch λ_b (Fig. 4b). Both schemes lead to a complete population transfer. The counterintuitive sequence is usually prefered in the nanosecond regime, because the intermediate state (φ_0, which can have a short lifetime) is less populated during this process.

The connectivity can be affected (independently of the amplitude of the fields) in one the following limit cases: (i) Δ_0 is comparable to $|\omega_p-\omega_s|$, (ii) Δ_0 is comparable to $-\omega_s$ or $-\omega_p$. For the dual-resonance process ($\Delta_0 = 0$) or for a small detuning Δ_0 compared to $|\omega_p-\omega_s|$, $-\omega_s$ and $-\omega_p$, these Stark shifts in general produce corrections on the quasienergies (see Figs. 2a, 4a) which do not affect the connectivity. They become more important if (i) the Rabi frequencies $\mu_{i0}\alpha_s$ or $\mu_{0f}\alpha_p$ become of the order of the detunings Δ_s or Δ_p, respectively, *i.e.* if the two frequencies ω_p and ω_s are close, or the Rabi frequencies are high (strong fields, see Fig. 2a.), or (ii) if ω_s or ω_p becomes small. In these cases where the corrections are not negligible, we cannot make a unique association of a specific pulse with a specific transition as it is approximated in the RWA.

Numerical simulations with breakdown of connectivity (Δ_0 of the order of $\omega_p-\omega_s$). We consider for simplicity $\omega_p > \omega_s$. The breakdown of connectivity occurs when (i) $\lambda_a^p < \lambda_b^p$, which is true for $\Delta_0 \gtrsim \omega_p-\omega_s$, and (ii) $\lambda_a^s > \lambda_b^s$. The last condition (ii) is always satisfied for $|\mu_{i0}|^2/|\mu_{f0}|^2 < 2$. We study the system with $E_0 = 1.7$ ($\Delta_0 = 1.7$), satisfying the previous conditions of breakdown of connectivity (Fig. 5). The peak field amplitudes are $\alpha_p = 2.2$, and $\alpha_s = 1.1$. We obtain no transfer as predicited (see Figs. 5b and c). For both the counterintuitive and intuitive processes, the population is back in the initial state at the end of the process.

We remark that for $\Delta_0 = \omega_p-\omega_s$, this process becomes dual-resonant, with the three degenerate eigenvalues for $\underline{\alpha} = 0$: $\lambda_{i,2,-1} = \lambda_{f,1,0} = \lambda_{0,0,0}$. Using the analysis of the Section 3.2, with in this case $N_f = -1$, $N_f' = 0$, $N_i = 2$ and $N_i' = -1$, we obtain a degeneracy lifting of first order between $\lambda_{f,1,0}$ and $\lambda_{0,0,0}$, and of second order between $\lambda_{i,2,-1}$ and $\lambda_{0,0,0}$. We can determine the conditions of connectivity (they depend on the size of $|\mu_{i0}|^2/|\mu_{f0}|^2$).

5 The $(2+1)$ STIRAP process in a four- and five-level system

We study the possibility of complete population transfer with a two-photon resonance for one of the lasers (we consider here a two-photon resonance for the pump laser, see Fig. 1b and c). We consider for simplicity that only one intermediate level (E_1) assists the two-photon transition. The study of this section interprets and completes the results of Oreg *et al.* [12] in a four-level system. They found with RWA analysis that adiabatic transfer is not possible when one of the intermediate levels is in resonance. We show that this transfer becomes possible if the intermediate state is far from resonance, provided that the connectivity is satisfied. We also show that a fifth level introducing Stark shifts in the process can make the transfer possible (still if the connectivity is satisfied).

The intermediate level E_1 defines the detuning

$$\Delta_1 \equiv E_1 - (E_i + \omega_p). \quad (43)$$

It can be close to the resonance (Δ_1 is small compared to the Bohr frequencies $E_1 - E_i$ and $E_0 - E_1$) or far from the resonance (Δ_1 comparable to or larger than the Bohr

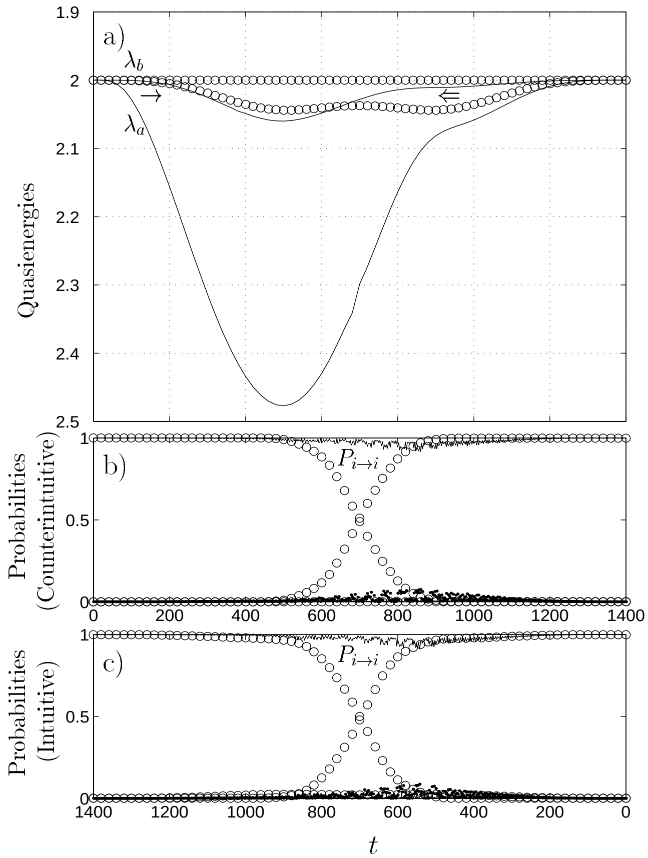

Fig. 5. The (1+1) STIRAP with one single resonance and with breakdown of connectivity ($\omega_p = 2$, $\omega_s = 0.39$, $\Delta_0 = 1.7$, $\Omega_{max} = 0.55$ and $T_p = T_s = 1000$). a) The instantaneous exact quasienergies (solid lines) and RWA quasienergies (circle lines). b) Numerical simulation of the population transfer for the counterintuitive process. c) Numerical simulation of the population transfer for the intuitive process. No transfer occurs in both processes because the connectivity is not satisfied, although complete transfer is predicted by the RWA.

frequencies). The resonant initial and final states of the system and the frequencies have the following numerical values for the simulations: $E_i = -2$, $E_f = -0.39$, $\omega_p = 1$, $\omega_s = 0.39$. All non-zero couplings are in this section are set to 1 for simplicity. For the (2+1) STIRAP, the two quasienergies for zero field $\lambda_{i,2,0} \equiv E_i + 2\omega_p$ and $\lambda_{f,0,1} \equiv E_f + \omega_s$ are degenerate. For the dual-resonance process, $\lambda_{0,0,0}$ is degenerate with the previous quasienergies.

5.1 Dual-resonance process

We consider that the energy $E_0 = 0$ is resonant with the initial level and the pump laser (two-photon resonance), and with the final level and the Stokes laser (one-photon resonance). For $\alpha_p \equiv 0$ and $\alpha_s \neq 0$, the first order matrix $W^{(1)}$ is the same one as in the (1+1) process, giving a linear breaking of the degeneracy. The first order contributions to the eigenvalues are given by (32) and the zeroth order eigenvector of interest, related to the transfer Floquet state, is the eigenvector connected to the initial state $\Psi_b^{(s)} = \varphi_i e^{i2\theta_p}$. Its associated eigenvalue (the zero eigenvalue) is always in-between the positive and negative eigenvalues. Thus, the complete population transfer can be obtained when the Stokes laser arrives first, *i.e.* in the counterintuitive order, since the other eigenvectors are linear combinations of φ_f and φ_0. The intuitive process fails in general as for the (1+1) STIRAP process.

For $\alpha_p \neq 0$ and $\alpha_s \equiv 0$, the degeneracy is lifted by the second order. We have to diagonalize $W^{(2)}$

$$W^{(2)} = \begin{pmatrix} W_{ii}^{(2)} & W_{i0}^{(2)} & 0 \\ W_{i0}^{(2)*} & W_{00}^{(2)} & 0 \\ 0 & 0 & W_{ff}^{(2)} \end{pmatrix}, \tag{44}$$

with

$$W_{ii}^{(2)} = -\frac{1}{4\Delta_1}\left|\mu_{i1}\right|^2\left[1+\frac{\Delta_1}{\Delta_1+2\omega_p}\right], \tag{45a}$$

$$W_{ff}^{(2)} = \frac{1}{4}|\mu_{f0}|^2\left[\frac{1}{E_f-E_0+\omega_p}+\frac{1}{E_f-E_0-\omega_p}\right], \tag{45b}$$

$$W_{00}^{(2)} = -\frac{1}{4\Delta_1}\left|\mu_{01}\right|^2\left[1+\frac{\Delta_1}{\Delta_1-2\omega_p}\right]-W_{ff}^{(2)}, \tag{45c}$$

$$W_{i0}^{(2)} = \frac{1}{4\Delta_1}\mu_{i1}\mu_{10}. \tag{45d}$$

The element $W_{ii}^{(2)}$ is the sum of resonant and anti-resonant terms between E_i and E_1. $W_{ff}^{(2)}$ contains Stark shifts of E_f due to the pump field and E_0. $W_{00}^{(2)}$ is the sum of resonant and anti-resonant terms between E_0 and E_1, and Stark shifts of E_0.

5.1.1 The four-level system with an intermediate level close to the resonance

In the case of an intermediate level close to resonance, the anti-resonant terms are negligible. We can also approximate $W_{ff}^{(2)} \approx 0$ if Stark shifts of E_f is negligible (*i.e.* the detuning between E_f and E_0 and the pump frequency is large compared to the pump Rabi frequency). This approximation is considered for the RWA, for which we exactly have $W_{ff}^{(2)} = 0$. The matrix (44) becomes approximately

$$W^{(2)} \approx -\frac{1}{4\Delta_1}\begin{pmatrix} \left|\mu_{i1}\right|^2 & -\mu_{i1}\mu_{10} & 0 \\ -\mu_{i1}^*\mu_{10}^* & \left|\mu_{01}\right|^2 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{46}$$

and gives the three approximate second order contributions to the eigenvalues (16)

$$\lambda_a^{(p)} \approx 0, \quad \lambda_b^{(p)} \approx 0, \quad \lambda_c^{(p)} \approx \frac{1}{4}\frac{\left|\mu_{i1}\right|^2+\left|\mu_{01}\right|^2}{E_0-E_1-\omega_p}\cdot \tag{47}$$

Thus two eigenvalues stay accidentally almost degenerate at this order. The degeneracy is only lifted when both

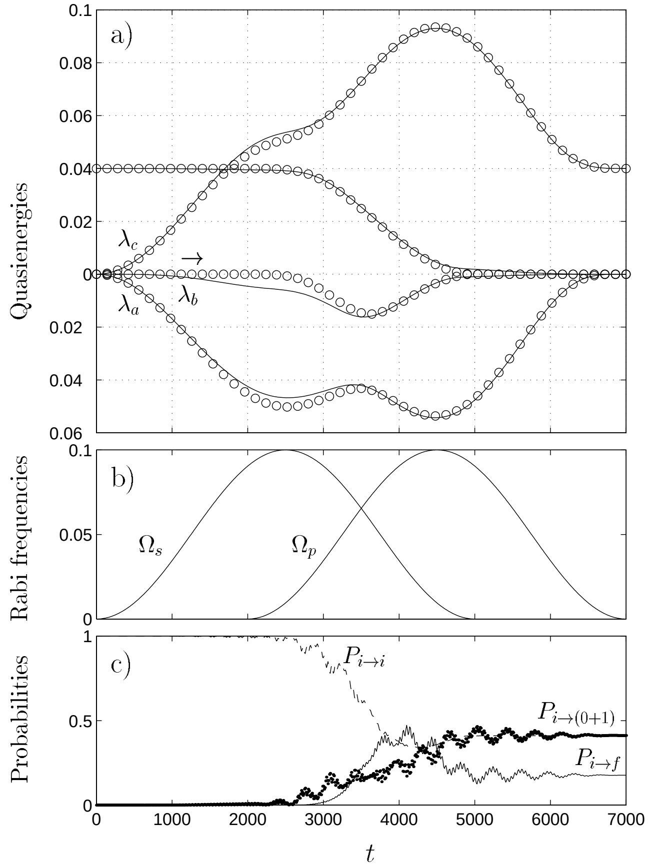

Fig. 6. The failed (2+1) STIRAP with a dual resonance for the counterintuitive process ($\omega_p = 1$, $\omega_s = 0.39$, $\Delta_1 = 0.04$, $\Delta_0 = 0$, $\Omega_{max} = 0.1$, $T_p = T_s = 5000$ and $t_{0_p} = 2000$). a) The instantaneous exact quasienergies (solid lines) and RWA quasienergies (circle lines). We remark the non-lifting of the degeneracy by the pump laser. b) The instantaneous Rabi frequencies. c) Numerical simulation of the population transfer. The transfer is not complete because of the residual degeneracy.

the laser fields are different from zero. The degeneracy is lifted, giving each branch connected to a superposition of bare levels and thus a complete transition cannot occur.

Numerical study. We study the counterintuitive process for a system containing the intermediate level $E_1 = -0.96$ ($\Delta_1 = 0.04$). The laser amplitudes are $\alpha_p = \alpha_s = 0.1$ (satisfying that the effective pump two-photon Rabi frequency is equal to the Stokes one-photon Rabi frequency [34]), with the pulse strengths $T_p = T_s = 5000$ and the delay of $t_{0_p} = 2000$. On the quasienergy diagram (Fig. 6a), we clearly see the near degeneracy of two quasienergies as long as α_s is zero (Fig. 6b). We observe the shared population on the bare states at the end of the process. We remark that the degeneracy remains strictly in the RWA analysis, but that the Floquet quasienergies have a small separation. This is due to small Stark shifts between E_f and E_0 produced by the pump laser. To achieve the complete transition, this degeneracy needs to be lifted for a small value of the pump laser amplitude. Several possibilities are successfully tested: (i) Additional Stark shifts are produced by the pump laser; (ii) The level E_0 is not resonant with the other ones at the beginning and the end of the process (single-resonance process).

5.1.2 The four-level system with a far from resonance intermediate level

In this case, we cannot neglect the Stark shift of E_f during the lifting of the degeneracy, since the detunings $E_f - (E_0 + \omega_p)$ and Δ_1 are both large. More generally, the Stark shift of E_f becomes relevant for a detuning Δ_1 of the order of or greater than $|\omega_p - \omega_s|$ (if the couplings $|\mu_{i1}|$ and $|\mu_{f0}|$ are of the same order). The connectivity requires that the quasienergy, associated to the Floquet state connected to the final state, is between the two other ones.

Numerical studies. We study this case with one far from resonance intermediate level E_1. For the dual-resonance process ($E_0 = 0.0$), we have obtained complete population transfer with the counterintuitive process for the following parameters (which satisfy the conditions of connectivity): $\Delta_1 = 1.6$, $\alpha_p = 0.4$, $\alpha_s = 0.05$, $T_p = T_s = 12000$ and $t_{0_p} = 4200$.

5.1.3 Additional Stark shifts

We consider again the four-level system with the intermediate level E_1 close to resonance. The residual degeneracy does not occur if another level E_2 (see Fig. 1c) contributes significantly to the lifting of the degeneracy by one of the following two possibilities: (i) E_2 contributes to the value of $W_{00}^{(2)}$:

$$E_2 \approx E_0 + \omega_p, \quad \Delta_2 = E_2 - (E_0 + \omega_p). \tag{48}$$

(ii) E_2 contributes to the value of $W_{ff}^{(2)}$:

$$E_2 \approx E_f + \omega_p, \quad \Delta_2 = E_2 - (E_f + \omega_p). \tag{49}$$

In the case (i), the condition of connectivity requiring that the Floquet state connecting to the final level φ_f has a quasienergy between the two others, gives

$$\frac{\Delta_2}{\Delta_1} > \frac{|\mu_{f1}|^2}{|\mu_{i1}|^2 + |\mu_{01}|^2}, \quad \Delta_1\Delta_2 > 0. \tag{50}$$

Numerical study. We show a numerical simulation for which the complete population is achieved (Fig. 7b), with the supplementary level satisfying the condition of connectivity (50): $E_2 = 0.92$ ($\Delta_2 = -0.08$), and the fields $\alpha_p = 0.1$ and $\alpha_s = 0.1$ satisfying the adiabatic passage. The pulse lengths are $T_p = T_s = 5000$ with a delay of $t_{0_p} = 2000$ (counterintuitive process). We clearly see the degeneracy breaking by the pump laser in the quasienergy diagram (Fig. 7a). Since the transfer Floquet state is different from the trapped state (constant eigenvalue), the

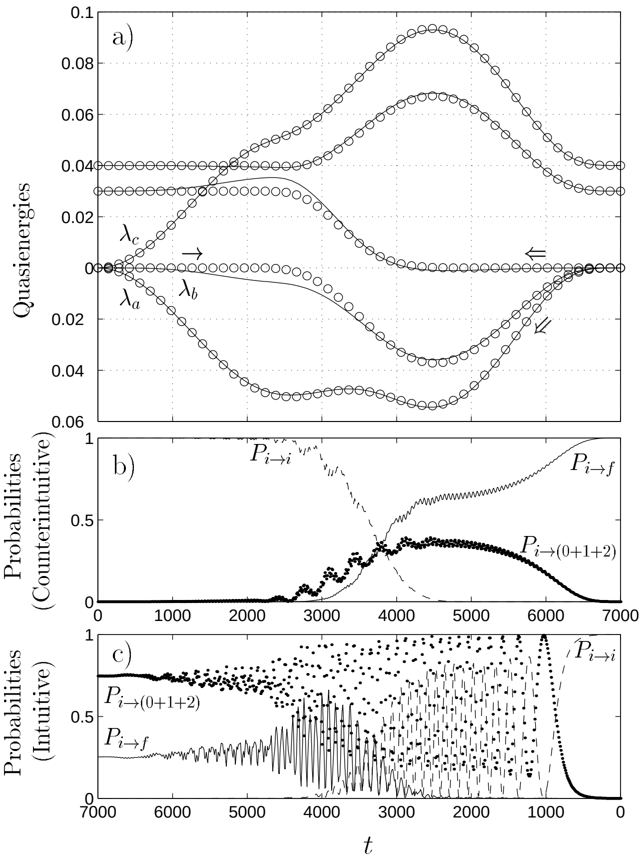

Fig. 7. The achieved (2+1) STIRAP with a dual resonance in a five-level system providing additional Stark shifts ($\omega_p = 1$, $\omega_s = 0.39$, $\Delta_1 = 0.04$, $\Delta_0 = 0$, $\Delta_2 = -0.08$, $\Omega_{max} = 0.1$ and $T_p = T_s = 5000$). a) The instantaneous exact quasienergies (solid lines) and RWA quasienergies (circle lines). The degeneracy is now lifted. b) Numerical simulation of the population transfer for the counterintuitive process ($t_{0_p} = 2000$). The complete population transfer is achieved. We observe a large population on the intermediate levels during the process because the quasienergy followed deviates from the zero eigenvalue. c) Numerical simulation of the population transfer for the intuitive process ($t_{0_s} = 2000$).

intermediate levels have a non-negligible population (maximum 40%) during the process. As predicted above, the intuitive process fails to produce complete transfer (Fig. 7c).

In the case (ii), the matrix becomes

$$W^{(2)} \approx -\frac{1}{4\Delta_1}\begin{pmatrix} |\mu_{i1}|^2 & -\mu_{i1}\mu_{10} & 0 \\ -\mu_{i1}^*\mu_{10}^* & |\mu_{01}|^2 + |\mu_{02}|^2\dfrac{\Delta_1}{\Delta_2} & 0 \\ 0 & 0 & 0 \end{pmatrix}. \tag{51}$$

The characteristic polynomial giving the eigenvalues of $W^{(2)}$ has always two different solutions (positive discriminant). The connectivity requires that the two non zero eigenvalues of $W^{(2)}$ (51) must have the opposite signs, *i.e.*

$$\Delta_1\Delta_2 < 0. \tag{52}$$

In this case, we have obtained numerically ($E_2 = 0.92$ has been used) a smaller population in the intermediate states during the process (maximum 25%), since the transfer Floquet state is now associated perturbatively to the constant zero eigenvalue, thus closer to the trapped state.

5.2 Single-resonance process

We consider the four-level system with the intermediate level E_1 close to the resonance. During the increasing (or decreasing) of the Stokes laser (with the amplitude of the pump laser at zero) we have from (23):

$$W_{ii}^{(2)} \approx 0\,, \quad W_{ff}^{(2)} \approx -\frac{|\mu_{f0}|^2}{4\Delta_0}, \tag{53}$$

and during the increasing (or decreasing) of the pump laser (with the amplitude of the Stokes laser at zero), we have

$$W_{ii}^{(2)} \approx -\frac{|\mu_{i0}|^2}{4\Delta_1}, \quad W_{ff}^{(2)} \approx 0. \tag{54}$$

The complete transfer requires the connectivity, *i.e.*

$$\Delta_0\Delta_1 > 0. \tag{55}$$

When the relation (55) is not satisfied, there is no population transfer. In fact the adiabatic passage implies that the population at the end of the process is still in the initial state φ_i.

Numerical studies. We show the complete population transfer with a numerical simulation (Fig. 8) for a system containing the levels E_0 and E_1 satisfying (55): $\Delta_1 = 0.04$ and $\Delta_0 = 0.04$, with the maximum fields $\alpha_p = \alpha_s = 0.05$ (This satisfies that the effective detuning, obtained by adiabatic elimination of the intermediate levels, is minimized [12]). We can switch first either the Stokes laser or the pump laser to achieve the transfer. The former case has the advantage to populate very little the intermediate levels during the process. We use for the pulse strengths $T_p = T_s = 5000$ with a delay of $t_{0_p} = 2000$ ($t_{0_p} = 0.4T_p$) for the counterintuitive process and $t_{0_s} = 2000$ for the intuitive one. We clearly see a residue of the previous accidental degeneracy (Fig. 6a) for the dual-resonance case: this gives now two close branches. We remark, in the quasienergy diagram (Fig. 8a), two close quasienergies which can cause deviation from adiabaticity if the passage is not slow enough.

We show the absence of complete population transfer (Fig. 9) when the relation (55) is not satisfied: $E_1 = -1.02$ ($E_1 \lesssim E_i + \omega_p$), the other quantities being unchanged. We remark that the two simulations (Fig. 9b and c) give roughly the same picture but reversed. This is because in this case, the counterintuitive and the intuitive processes follow exactly the same path in the quasienergy diagram, in opposite directions.

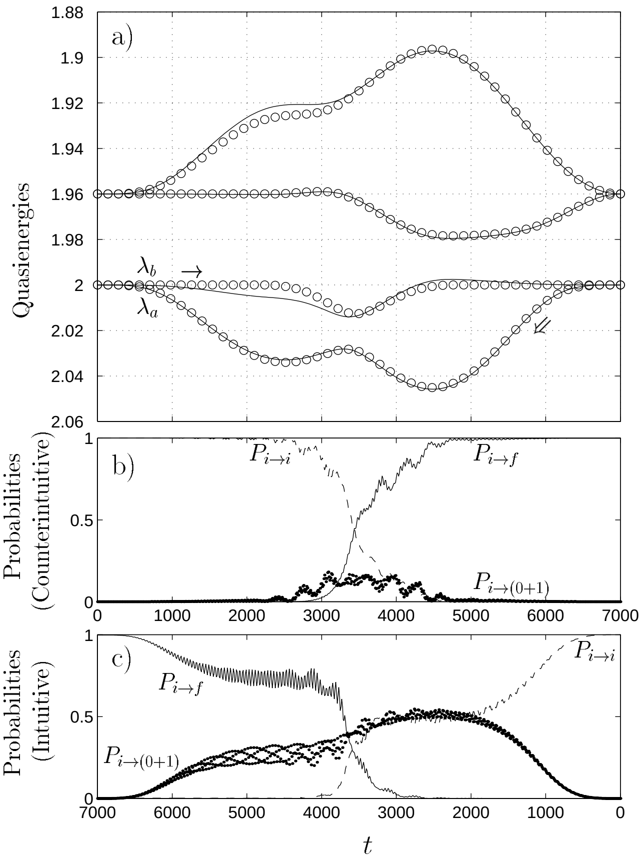

Fig. 8. The achieved (2+1) STIRAP with a single resonance. ($\omega_p = 1$, $\omega_s = 0.39$, $\Delta_1 = 0.04$, $\Delta_0 = 0.04$, $\Omega_{max} = 0.05$ and $T_p = T_s = 5000$). a) The instantaneous exact quasienergies (solid lines) and RWA quasienergies (circle lines). b) Numerical simulation of the population transfer for the counterintuitive process ($t_{0_p} = 2000$). c) Numerical simulation of the population transfer for the intuitive process ($t_{0_s} = 2000$). In both cases, the complete population transfer is achieved. We observe a larger population on the intermediate levels during the intuitive process.

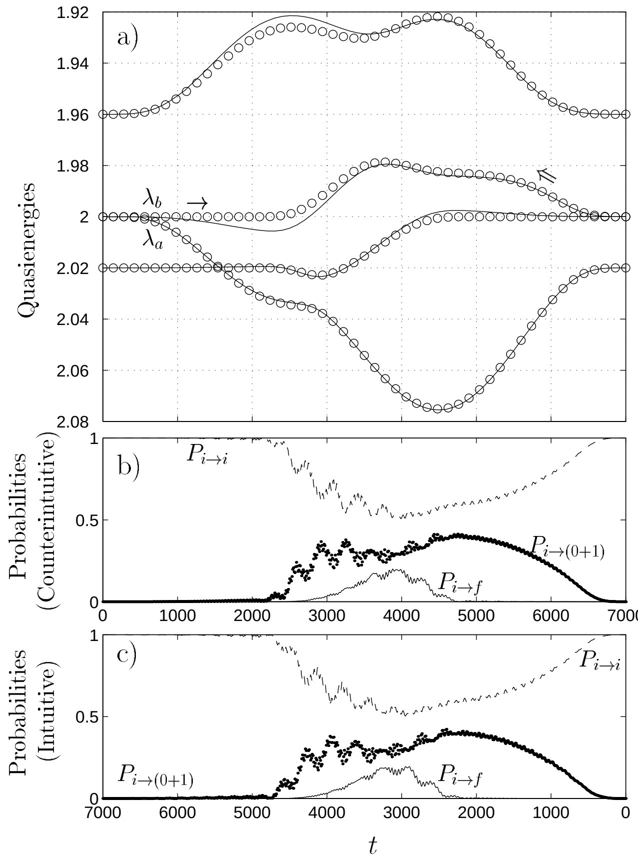

Fig. 9. The failed (2+1) STIRAP with a single resonance. ($\omega_p = 1$, $\omega_s = 0.39$, $\Delta_1 = -0.02$, $\Delta_0 = 0.04$, $\Omega_{max} = 0.05$ and $T_p = T_s = 5000$). a) The instantaneous exact quasienergies (solid lines) and RWA quasienergies (circle lines). b) Numerical simulation of the population transfer for the counterintuitive process ($t_{0_p} = 2000$). c) Numerical simulation of the population transfer for the intuitive process ($t_{0_s} = 2000$). In both cases, the population comes back to the initial state φ_i. We remark that the processes give the same picture because they follow the same path in reversed orders.

6 conclusion

In this article, we have discussed the STIRAP process in the frame of Floquet theory. We have shown corrections to the (1+1) STIRAP in strong field and with large detuning of the intermediate state. We have studied the feasibility of the (2+1) STIRAP in a four- and five-level system, determining the types of level structures which are well suited for complete population transfer. We remark that, essentially because of the two-photon process, (2+1) STIRAP in atoms can involve more than four levels and a continuum, which will produce Stark shifts and incoherent losses [35]. These Stark shifts should make the complete transfer feasible if the connectivity and the adiabatic following are satisfied. Incoherent losses will reduce the transfer.

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