Optimal adiabatic passage by shaped pulses: Efficiency and robustness

S. Guérin,* V. Hakobyan, and H. R Jauslin

Laboratoire Interdisciplinaire Carnot de Bourgogne UMR 5209 CNRS, Université de Bourgogne, BP 47870, F-21078 Dijon, France

(Received 1 May 2011; published 27 July 2011)

We explore the efficiency and robustness of population transfer in two-state systems by adiabatic passage (i) when the driving pulse is optimally designed in order to lead to parallel adiabatic passage or (ii) with a linear chirping. We show how one could practically implement the corresponding designs of the pulses in the spectral domain. We analyze the robustness of the two shapings taking into account fluctuations of the phase, amplitude, and the area of the pulse. We show the overall superiority of the parallel adiabatic passage especially when one faces the issue of a pulse area that is not well known. We show that the robustness of parallel adiabatic passage is not improved when it is complemented by a correcting field that cancels out the nonadiabatic losses.

DOI: 10.1103/PhysRevA.84.013423 PACS number(s): 32.80.Qk, 42.50.Hz, 42.50.Ex

I. INTRODUCTION

Manipulating the state of a quantum system by external fields is an important issue in a wide variety of problems [1–5]. Modern applications, such as quantum computations processing, necessitate a fine control corresponding typically to an admissible error of at most 10^{-4} [5]. Such control should also feature robustness with respect to variations or an imperfect knowledge of the experimental parameters. Finally, fast processes that are not subject to dissipation, nor to decoherence, are desirable.

In a two-state system, the fastest process to achieve a complete population transfer corresponds to a resonant pulse with area π for the Rabi frequency, and any time dependence of the (diagonal) detuning cannot accelerate it [6]. This process is, however, nonrobust with respect to variations of the area that are often difficult to avoid in practice. In nuclear magnetic resonance (NMR), a series a π pulses with well-defined static phases, known as composites pulses, have been proposed to compensate unknown errors in the parameters [7]. This technique is being investigated in quantum optics [8].

On the other hand, adiabatic passage and its variations [9,10] allow robustness of the transfer as one increases the pulse area. But it leads in principle to an incomplete transfer reaching 1 only asymptotically in the adiabatic limit (i.e., $T \to \infty$, where T is a measure of the duration of the process). One can estimate the efficiency of the transfer for a concrete model using a complex time method leading to the Davis-Dykhne-Pechukas (DDP) formula [11–14].

Modern technologies allow the shaping at will of the field amplitude and phase even in the ultrafast femtosecond regime. In this case, the field is shaped in the frequency domain through the spatial separation and manipulation of the spectral components [15,16]. Finding an optimal shape that offers the best compromise between process speed, (i.e., featuring an area as close as possible to π) and robustness is thus important for applications.

Such an optimization has been proposed on the basis of the DDP formula resulting in the parallel adiabatic passage (PLAP) technique in which the fields produce eigenenergies which are parallel to each other [17–19]. The use of an additional field that cancels the nonadiabatic coupling has also been proposed [20,21]. We remark, however, that this technique is expected to have, in practice, a limited advantage regarding robustness since it requires having explicit knowledge of the (small) nonadiabatic coupling. In particular, we show in this paper that the use of this technique does not give better results than PLAP with respect to an imperfect knowledge of the field area (Sec. VI).

In this paper, we investigate the technique of optimization which is based on the DDP formula. The population transfer by adiabatic passage is here defined as optimal for the parameters with the smallest coupling area for which the DDP formula gives a complete population transfer. Despite the mathematical need of an adiabatic limit $T \to \infty$, where T corresponds to the duration of the interaction with the field, which corresponds in the context of pulsed coupling to an infinite pulse area, the DDP formula is known to be already very accurate for a finite and relatively small area (see, for instance, [22]). In practice, it is an important issue to determine the needed value of this area to get an efficient population transfer (that has to be quantitatively defined depending on the problem that is studied) while preserving the robustness of the process. We remark that the DDP formula does not give any direct information about robustness of the process, which is expected to be better for a more adiabatic process, and this has to be analyzed through numerical simulation for a concrete model.

The DDP formula shows that the population transfer is complete when (i) the eigenvalues are dynamically parallel (PLAP) [17], or (ii) when the nonadiabatic components cancel by interference. The latter situation technically requires two transition points in the complex-time plane (see below for a more precise statement), and is referred to as adiabatic passage complemented by destructive interference (DIAP). Since it is based on specific conditions of interference, it is expected to have a limited robustness. In this paper, we compare the robustness of these two techniques and we show that the robustness is superior for PLAP in a concrete model of interest.

The paper is organized as follows: We first recall the DDP formula. Next we define the PLAP (Sec. III) and DIAP (Sec. IV) techniques on a concrete example with a Gaussian pulse, which allows one to deduce two types of optimal shaping. Their implementation in the frequency domain is shown in Sec. V. Their respective robustness is analyzed in Sec. VI. We conclude in Sec. VII.

*sguerin@u-bourgogne.fr

II. THE MODEL AND THE DDP FORMULA

We study the population transfer between an initially populated ground state $|g\rangle$ and an excited state $|e\rangle$ (of respective energies $\hbar\omega_g$ and $\hbar\omega_e$) that can be modeled in the resonant approximation (and up to terms proportional to the identity) by a two-state effective dressed Hamiltonian of the type [1] (in the dressed-state basis $\{|g;0\rangle,|e;1\rangle\}$, where the second label stands for a relative number of photons)

$$\mathsf{H}^{[\Omega,\Delta]}=\frac{\hbar}{2}\begin{bmatrix}0 & \Omega\\ \Omega & 2\Delta\end{bmatrix}, \tag{1}$$

with the two time-dependent parameters $\Omega\equiv\Omega(t)$ (the effective Rabi frequency) and $\Delta\equiv\Delta(t)$ (the detuning), that can be *a priori* varied as wished. Here the effective Rabi frequency (assumed real and positive for simplicity) reads $\Omega=\mu\mathcal{E}/\hbar$ with μ being the dipole coupling and $\mathcal{E}(t)$ being the amplitude of the field of instantaneous frequency $\omega(t)=\omega_0+\dot{\phi}(t)$ (with $\omega_0 t+\phi(t)$ being the phase of the field) such that $\Delta(t)=\omega_e-\omega_g-\omega(t)$.

Adiabatic passage means that, in the adiabatic limit, the dynamics projects at all times, up to a phase, on the instantaneous eigenvector of $H(t)$ that is continuously connected to the initial state. It leads to a population transfer when this eigenvector finally connects to the target excited state. This typically occurs in the so-called crossing models, corresponding to pulsed interactions whose instantaneous frequency crosses the resonance; that is, for a detuning that changes sign during the interaction. For a finite time of interaction (characterized by T), the preceding statement becomes only approximate and deviations from it are generally referred to as nonadiabatic losses that lead to some population being brought back to the initial state at the end of the interaction. The DDP formalism allows one to determine the efficiency of the population transfer at the end of the interaction. For a crossing model, the DDP formula gives more precisely the probability P_g of return to the initial ground state. The population transfer to the excited state is thus $P_e=1-P_g$. In the adiabatic limit, the probability of return is given by a coherent sum:

$$P_g=\left|\sum_{k=1}^{N}\Gamma_k e^{i\mathcal{D}(t_k)}\right|^2, \tag{2}$$

with the phase factors $\Gamma_k=\pm1$ for a real Ω, and

$$\mathcal{D}(t)=\int_0^t\lambda(z)dz,\quad \lambda(t)=\sqrt{\Omega^2(t)+\Delta^2(t)}. \tag{3}$$

This takes into account all the (complex) N transition points t_k, $k=0,\dots,N-1$, defined as the complex zeros of the eigenenergy splitting:

$$\lambda(t_k)=0, \tag{4}$$

lying on the Stokes line γ in the upper complex plane, defined as

$$\begin{aligned}&\mathrm{Im}[\mathcal{D}(\gamma)]\\ &\quad=\text{const.}=\mathrm{Im}[\mathcal{D}(t_0)]=\cdots=\mathrm{Im}[\mathcal{D}(t_{N-1})]\end{aligned} \tag{5}$$

and closest to the real axis.

The conditions of validity of the DDP formula are (i) $\lambda(t)\neq0$ for all real t (non-degeneracy condition) and (ii) $\lambda(z)$ is analytic and single valued in a complex domain that includes the Stokes line closest to the real axis and the real axis.

The DDP formula had been initially established for the generic case with a single transition point [11–13]. It was then formulated [12] and proven [14] for multiple transition points, which is the situation often encountered in practice, in particular when one considers symmetric (i.e., odd or even) pulses and detunings.

The complete population transfer (in the adiabatic limit) occurs when $P_g=0$, which is in principle exactly satisfied from Eq. (2) either when (i) the transition points go to infinity [i.e., when the eigenvalues are parallel at all times [17] (PLAP)] or when (ii) the coherent sum destructively interferes (DIAP).

We remark that the popular Allen-Eberly model [23] with $\Omega=\Omega_0\,\mathrm{sech}(t/T)$ and $\Delta=\Delta_0\tanh(t/T)$ possesses singularities in the complex plane which prevent the transition points to go to infinity when we force the eigenvalues to be parallel: the first transition points merge instead to the first singularity in this case [17]. Thus, this model is not expected to show a better efficiency for the situation of parallel eigenvalues.

Below we briefly recall the technique of parallel adiabatic passage. We next show that single-parameter linear chirp allows complete population transfer by DIAP. The robustness of the two techniques are compared in the next section.

III. PARALLEL ADIABATIC PASSAGE

A. Definition

PLAP is satisfied when the dynamics follows a trajectory in the parameter space (Ω,Δ) given by

$$\Omega^2+\Delta^2=\Omega_0^2. \tag{6}$$

Assuming a given pulse shape $0\leqslant\Lambda_T(t)\leqslant1$ for the coupling

$$\Omega(t)=\Omega_0\Lambda_T(t), \tag{7}$$

one can easily extract Δ for PLAP as a function of this shape:

$$\Delta_\pm(t)=\pm\Omega_0\sqrt{1-\Lambda_T^2(t)}. \tag{8}$$

Assuming that the field is maximum at $t=0$ [i.e., $\Lambda_T(0)=1$], we can choose for convenience $\Delta(t)=\Delta_+(t)$ for $t>0$ and $\Delta(t)=\Delta_-(t)$ for $t<0$, which leads to

$$\Delta(t)=\Omega_0 g(t),\quad g(t)=\mathrm{sgn}(t)\sqrt{1-\Lambda_T^2(t)}. \tag{9}$$

We consider here a Gaussian shape $\Lambda_T(t)=e^{-(t/T)^2}$. The left frame of Fig. 1 shows the transfer efficiency after the interaction, by numerical solution of the Schrödinger equation, as functions of Ω_0T and Δ_0T with such a Gaussian shape and a detuning of the form $\Delta(t)=\Delta_0 g(t)$. The transfer efficiency is better for a darker zone.

The left frame of Fig. 1 shows, for $\Delta_0=0$, the Rabi oscillations where the transfer is complete when the Rabi frequency has an area of an odd multiple of π. They extend as roughly vertical lines of highly efficient transfer which merge approximately (and better for larger area) to a zone surrounding the PLAP line ($\Delta_0=\Omega_0$). An important feature is that the width of the region of efficient population around the PLAP line becomes larger for larger pulse areas. On the other hand, the widths of the vertical lines located below the

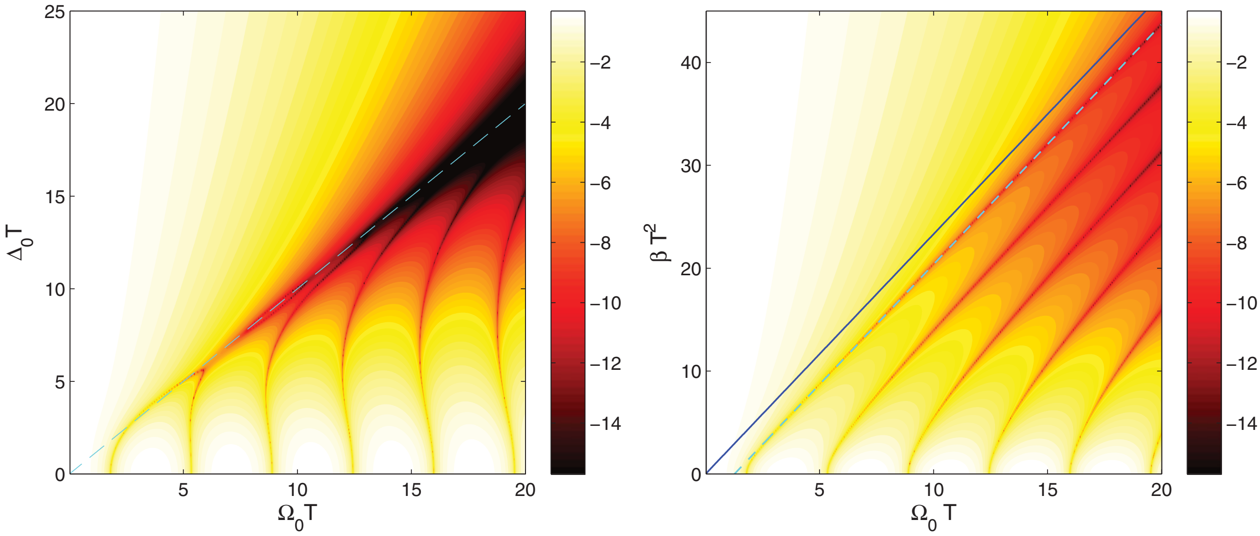

FIG. 1. (Color online) Contour plot (decimal logarithmic scale) of the probability of return to the ground state at the end of the interaction for a coupling of Gaussian shape $\Omega(t) = \Omega_0\Lambda_T(t)$, $\Lambda_T(t) = e^{-(t/T)^2}$, as a function of $\Omega_0 T$ and (i) $\Delta_0 T$ with $\Delta(t) = \Delta_0 g(t)$ (left frame) and (ii) βT^2 with a linear chirp $\Delta(t) = \beta t$ (right frame). The dashed line $\Delta_0 = \Omega_0$ [Eq. (22)] of the left (right) frame corresponds to PLAP (DIAP with the minimum Rabi frequency area). The full line (right frame) is the transition line (15) between the zones of single and double transition points (see text).

PLAP line are much smaller. From these observations, one can anticipate the high robustness with respect to the pulse area of the PLAP technique. This is analyzed in Sec. VI.

The efficiency of the PLAP technique is already very good from $\Omega_0 T = 2.15$, which corresponds to an area of $3.8 \approx 1.2\pi$ (to be compared to the area π that is the minimal area that leads to a complete population transfer [6]). For this value, the error is less than 1%. One can get an ultrahigh efficiency with an error less than 10^{-4} from $\Omega_0 T = 2.53$, which corresponds to an area of $4.5 \approx 1.45\pi$.

B. Transitionless parallel adiabatic passage

As confirmed in the left frame of Fig. 1, despite the remarkably large region of efficient transfer surrounding the PLAP line, the transfer is, in general, not strictly complete on the PLAP line (but is close to it). One can improve it by suppressing the nonadiabatic losses as originally suggested in [20] for three-state systems and reformulated in [21] as a transitionless quantum driving technique. We thus construct the transitionless parallel adiabatic passage (T-PLAP) which transforms the PLAP line to a line of strictly complete population transfer.

One proposed version of the technique is based on adding a corrector-driving Hamiltonian $H_c(t)$ to the original Hamiltonian in order to compensate each time the nonadiabatic coupling inducing unwanted transitions. This leads to the new Hamiltonian

$$\hat{H}(t) = \mathsf{H}^{[\Omega,\Delta]} + H_c(t) \tag{10}$$

with the corrector Hamiltonian

$$H_c(t) = \frac{\hbar}{2}\begin{pmatrix} 0 & -i\Omega_c(t) \\ i\Omega_c(t) & 0 \end{pmatrix}, \quad \Omega_c(t) = \frac{\Omega\dot{\Delta} - \dot{\Omega}\Delta}{\Omega^2 + \Delta^2}. \tag{11}$$

The initially real Rabi frequency becomes complex: $\Omega(t) \to \Omega(t) - i\Omega_c(t)$. This corresponds to a field decomposed into two parts of equal polarization but with one part in quadrature phase with respect to the other one. We can apply the technique on the PLAP in order to force it to lead to an exact population transfer for any pulse area of the original Rabi frequency. This leads to a correcting Rabi frequency independent of Ω_0:

$$\Omega_c(t) = -\mathrm{sgn}(t)\frac{\dot{\Lambda}_T(t)}{\sqrt{1 - \Lambda_T^2(t)}}. \tag{12}$$

One remarks that the area of this correcting Rabi frequency is π, as expected, such that when $\Omega_0 = \Delta_0 = 0$, it ensures the complete transfer.

We will show more precisely in Sec. VI on a concrete example with an average over various pulse areas that the transitionless parallel adiabatic passage does not improve PLAP.

IV. ADIABATIC PASSAGE COMPLEMENTED BY DESTRUCTIVE INTERFERENCE: THE CASE OF GAUSSIAN PULSE WITH LINEAR CHIRPING

The simplest field shaping that features DIAP is the one that leads to a linearly time-dependent detuning; a so-called linear chirping:

$$\Delta(t) = \beta t. \tag{13}$$

This model and its DDP analysis have been studied in detail in Ref. [24]. Following this reference, we determine below the value of β that leads to the DIAP as a function of the peak Rabi frequency Ω_0 considering its smallest value (since, for a given β, there are several values of Ω_0 producing the DIAP; see Fig. 1, right frame).

We introduce the variable $s = t/T$. The transition points s_k, solutions of Eq. (4), read

$$s_k = \sqrt{\frac{1}{2}W(-\alpha)}, \quad \alpha = \frac{2\Omega_0^2}{(\beta T)^2}, \tag{14}$$

where $W(x)$ is known as the Lambert W function, defined as the inverse function of $f(W) = We^W$. $W(x)$ is real for $x \geqslant -1/e$, and $W(x) \leqslant 0$ for $-1/e < x \leqslant 0$. We have $W(-1/e) = -1$. One can identify a single transition point s_0 lying on the Stokes line closest to the real axis, and of smallest imaginary part, when its real part is zero, which arises when $W(\cdot)$ in Eq. (14) is real and negative (i.e., for $\alpha < 1/e$). There are two transition points (on the Stokes line closest to the real axis), denoted $s_\pm$, when $W(\cdot)$ in Eq. (14) is not real (i.e., for $\alpha > 1/e$). Thus, the branch

$$\beta T = \sqrt{2e}\Omega_0 \tag{15}$$

corresponding to $\alpha = 1/e$, for which we denote the transition point as $s_{0,0} = i/\sqrt{2}$, separates in the plane $(\Omega_0,\beta T)$ the zones of single and double transition points. The two transition points have opposite real parts: $\mathrm{Re}(s_-) = -\mathrm{Re}(s_+)$ and identical imaginary parts: $\mathrm{Im}(s_-) = \mathrm{Im}(s_+)$. This implies $\mathrm{Im}[\widetilde{\mathcal{D}}(s_-)] = \mathrm{Im}[\widetilde{\mathcal{D}}(s_+)]$ and $\mathrm{Re}[\widetilde{\mathcal{D}}(s_-)] = -\mathrm{Re}[\widetilde{\mathcal{D}}(s_+)]$, where we have denoted $\widetilde{\mathcal{D}}(s) = \mathcal{D}(t)$. The probability of population return reads in the case of two transition points (in the adiabatic limit):

$$P_g = 4e^{-2\mathrm{Im}[\mathcal{D}(t_+)]}\cos^2 \mathrm{Re}[\mathcal{D}(t_+)], \tag{16}$$

where we have also used $\Gamma_- = \Gamma_+ = 1$ (since we consider an even coupling and an odd detuning).

The smallest peak coupling Ω_0, for a given β, that leads to DIAP is thus a solution of

$$\mathrm{Re}[\widetilde{\mathcal{D}}(s_+)] = \frac{\pi}{2}. \tag{17}$$

One cannot solve this equation exactly but only approximately. We achieve this by remarking that the corresponding transition point $s_+ = t_+/T$ is located close to $s_{0,0} = i/\sqrt{2}$ corresponding to the branch (15). Using a series expansion of $W(x)$, for $x := -\alpha \lesssim -1/e$, denoting $\epsilon = -1/e - x > 0$, we obtain to a very good accuracy (with an error less than 2% as checked numerically):

$$s_+ \simeq s_{0,0} + \frac{1}{2}\sqrt{e\epsilon}\left(1 - \frac{5}{36}e\epsilon\right) - i\frac{1}{\sqrt{2}}\frac{e}{12}\epsilon. \tag{18}$$

Next we decompose the integral (3) as follows:

$$\widetilde{\mathcal{D}}(s_+) = \beta T^2\left(\int_0^{s_{0,0}} + \int_{s_{0,0}}^{s_+}\right)\sqrt{z^2 + \frac{\alpha}{2}e^{-2z^2}}dz. \tag{19}$$

The first integral leads to an imaginary value, hence

$$\mathrm{Re}[\widetilde{\mathcal{D}}(s_+)] = \beta T^2\int_{s_{0,0}}^{s_+}\sqrt{z^2 + \frac{\alpha}{2}e^{-2z^2}}dz. \tag{20}$$

At the lowest order of ϵ, for large β and Ω_0, we find for the solution of Eq. (17):

$$\beta T \sim \sqrt{2e}\Omega_0, \quad \beta \to \infty, \tag{21}$$

which gives a line parallel to (and below) the branch (15) separating the zones of single and double transition points.

A numerical analysis of the integral (20) allows one to determine an approximate equation of this line for finite values of β:

$$\beta T \approx \sqrt{2e}(\Omega_0 - 1.25/T). \tag{22}$$

This line is shown in the right frame of Fig. 1, where the error of the transfer probability to the excited state is numerically determined as a function of $\Omega_0 T$ and βT^2. It fits very well the zone of efficient population transfer of smallest area (for a given β), and is parallel to the transition line (15) between the zones of single and double transition points.

For respectively $\Delta_0 = 0$ and $\beta = 0$, the two frames of Fig. 1 show the same Rabi oscillations. A salient feature is that the zones of complete transfer can be extended for both frames: They all merge to the PLAP line in the left frame, while only the one of smallest Ω_0 (i.e., of smallest pulse area) approximately coincides (and better for larger $\Omega_0 T$) to the DIAP line. The extensions of the other zones of complete transfer for larger pulse areas are approximately parallel to the DIAP line. One notices that, in both cases, these extensions are surrounded by larger zones of efficient transfer for larger $\Omega_0 T$, which clearly indicates an expected better robustness for larger $\Omega_0 T$. However, the size of this zone of efficient transfer is shown to be much larger around the PLAP line. We thus anticipate a better robustness of the PLAP technique with respect to the DIAP technique. This is analyzed in detail in Sec. VI.

V. IMPLEMENTATIONS BY SPECTRAL SHAPING

The implementation of adiabatic techniques with chirped fields can be achieved in the femtosecond regime by a spectral shaping [15,16].

The DIAP with a Gaussian pulse and a linear chirping (13) analyzed in the preceding section can be simply implemented in practice since, when the mean frequency of the initial field matches the transition frequency, it requires a device which shapes only the spectral phase, which can be achieved using a grating [25] or a single spatial light modulator [26]. On the other hand, the PLAP requires a shaping of both the spectral phase and amplitude as shown below. This can be produced, for instance, with a double-layer liquid-crystal spatial light modulator such as the one used in [27].

We assume an input field of Gaussian shape with the mean frequency ω_0 and the full width at half maximum (FWHM; for the corresponding intensity) $T_{\mathrm{in,FWHM}} = T_{\mathrm{in}}\sqrt{2\ln 2}$:

$$\mathcal{E}_{\mathrm{in}}(t) = \mathcal{E}_{0\mathrm{in}}\Lambda_{T_{\mathrm{in}}}(t)e^{i\omega_0 t}, \quad \Lambda_{T_{\mathrm{in}}}(t) = e^{-(t/T_{\mathrm{in}})^2}. \tag{23}$$

The output pulse $\mathcal{E}(t)$ that subsequently interacts with the system is chosen to be also of Gaussian shape:

$$\mathcal{E}(t) = \mathcal{E}_0\Lambda_T(t)e^{i[\omega_0 t + \phi(t) - \theta]}, \tag{24}$$

with a phase θ to be defined and the instantaneous frequency $\omega(t) = \omega_0 + \dot{\phi}(t)$ giving the relation between the phase $\phi(t)$ and the one-photon detuning of our initial problem:

$$\phi(t) = (\omega_e - \omega_g - \omega_0)t - \int^t \Delta(s)ds. \tag{25}$$

This choice for the output pulse to be Gaussian is arbitrary; it is here chosen for its simplicity. The spectral shaping allows the transformation in the frequency domain of the input field into the output field through a transparency coefficient $0 \leqslant \mathcal{T}(\omega) \leqslant 1$ and a phase $\varphi(\omega)$ as follows:

$$\widetilde{\mathcal{E}}(\omega) = \mathcal{T}(\omega) e^{i\varphi(\omega)} \widetilde{\mathcal{E}}_{\text{in}}(\omega), \tag{26}$$

with $\widetilde{\mathcal{E}}(\omega)$ denoting the Fourier transform of $\mathcal{E}(t)$. Two masks are generally used: one operates on the transparency while the other one operates on the phase. The duration T of the output Gaussian pulse has to be carefully chosen such that there exists a solution for the transparency $\mathcal{T}(\omega)$ and the phase $\varphi(\omega)$ of (26) that leads to the desired output field (24) and, more precisely, that this solution works well within the input Gaussian spectrum. A smooth solution that is easily implementable is also desirable.

A. Gaussian pulse and linear chirping

When the frequency of the input field matches the transition frequency (i.e., $\omega_0 = \omega_e - \omega_g$), it is well known that a linear chirping results from a single modulator with a quadratic spectral phase:

$$\mathcal{T}(\omega) = 1, \quad \varphi(\omega) = \gamma(\omega - \omega_0)^2. \tag{27}$$

This leads to the output field which is of maximum amplitude when it is exactly resonant:

$$\mathcal{E}(t) = \mathcal{E}_{0\text{in}} \sqrt{\frac{T_{\text{in}}}{T}} e^{-(t/T)^2} e^{i[\omega_0 t + \phi(t) - \theta]}, \tag{28}$$

with the instantaneous frequency

$$\omega(t) \equiv \omega_0 + \dot{\phi}(t) = \omega_0 - \frac{8\gamma}{T_{\text{in}}^4 + 16\gamma^2} t \tag{29a}$$

$$\simeq \omega_0 - \frac{1}{2\gamma} t \text{ for } \gamma \gtrsim T_{\text{in}}^2, \tag{29b}$$

the phase

$$\theta = \arg \sqrt{T_{\text{in}}^2 - 4i\gamma}, \tag{30}$$

and the duration

$$T = \frac{4\gamma}{T_{\text{in}}} \sqrt{1 + \left(\frac{T_{\text{in}}^2}{4\gamma} \right)^2} \tag{31a}$$

$$\simeq \frac{4\gamma}{T_{\text{in}}^2} T_{\text{in}} \text{ for } \gamma \gtrsim T_{\text{in}}^2. \tag{31b}$$

The width of the chirp that can be characterized by $|\dot{\phi}(T/2) - \dot{\phi}(-T/2)| = |\dot{\phi}(T)|$ is thus, in practice, limited by the spectrum of the laser:

$$\dot{\phi}(T) \lesssim \frac{2}{T_{\text{in}}}, \tag{32}$$

reaching its asymptotic value $4/T_{\text{in}}$ for $\gamma \gtrsim T_{\text{in}}^2$, corresponding to the duration $T \gtrsim 4T_{\text{in}}$.

We can determine a relation between the slope β of the chirp (13) and the coefficient γ of the quadratic phase (27) as

$$\beta T^2 = 8 \frac{\gamma}{T_{\text{in}}^2} \tag{33a}$$

$$\simeq 2 \frac{T}{T_{\text{in}}} \text{ for } \gamma \gtrsim T_{\text{in}}^2. \tag{33b}$$

This means that any value βT^2 can be obtained by appropriately adjusting γ, which amounts to choosing T [from Eq. (31)]. Since one can also produce any value $\Omega_0 T$ by choosing the peak intensity of the field, any region of the right part of Fig. 1 can be, in principle, obtained from a concrete implementation with a pulse of limited bandwidth. A practical limitation will be a limited field intensity to avoid unwanted destructive effects such a ionization.

If there is a mismatch between the mean laser frequency ω_0 and the transition frequency $\omega_1 := \omega_e - \omega_g$, and if one wants an effective frequency that is resonant (i.e., $\Delta = 0$) when the output field is maximum in the time domain, we have to shape the spectral amplitude as

$$\begin{aligned} \mathcal{T}(\omega) &= e^{\frac{1}{4}[(\omega-\omega_0)^2 T_{\text{in}}^2 - (\omega-\omega_1)^2 T_a^2]}, \\ \varphi(\omega) &= \gamma(\omega - \omega_1)^2, \end{aligned} \tag{34}$$

with the requirement that the shaping operates well within the bandwidth:

$$|\omega_0 - \omega_1| \lesssim \left(\frac{1}{T_{\text{in}}} - \frac{1}{T_a} \right) \sqrt{2 \ln 2}, \quad T_a > T_{\text{in}}. \tag{35}$$

This leads to the output field which is of maximum amplitude when it is exactly resonant:

$$\mathcal{E}(t) = \mathcal{E}_{0\text{in}} \sqrt{\frac{T_a}{T}} e^{-(t/T)^2} e^{i[\omega_1 t + \phi(t) - \theta]}, \tag{36}$$

with the instantaneous frequency

$$\omega(t) \equiv \omega_1 + \dot{\phi}(t) = \omega_1 - \frac{8\gamma}{T_a^4 + 16\gamma^2} t \tag{37a}$$

$$\simeq \omega_0 - \frac{1}{2\gamma} t \text{ for } \gamma \gtrsim T_a^2, \tag{37b}$$

the phase

$$\theta = \arg \sqrt{T_a^2 - 4i\gamma}, \tag{38}$$

and the duration

$$T = \frac{4\gamma}{T_a} \sqrt{1 + \left(\frac{T_a^2}{4\gamma} \right)^2} \tag{39a}$$

$$\simeq \frac{4\gamma}{T_a} \text{ for } \gamma \gtrsim T_a^2. \tag{39b}$$

B. PLAP

The achievement of PLAP by a spectral shaping necessitates a shaping both in phase and amplitude even when the mean frequency of the input field matches with the transition frequency, $\omega_0 = \omega_e - \omega_g$ (that is the situation we consider here for simplicity). One cannot determine in closed form the transparency and phase shaping of (26), but they can be obtained numerically by applying the Fourier transform of a given output pulse:

$$\begin{aligned} \mathcal{T}(\omega) &= |\widetilde{\mathcal{E}}(\omega)/\widetilde{\mathcal{E}}_{\text{in}}(\omega)|, \\ \varphi(\omega) &= \arg[\widetilde{\mathcal{E}}(\omega)/\widetilde{\mathcal{E}}_{\text{in}}(\omega)]. \end{aligned} \tag{40}$$

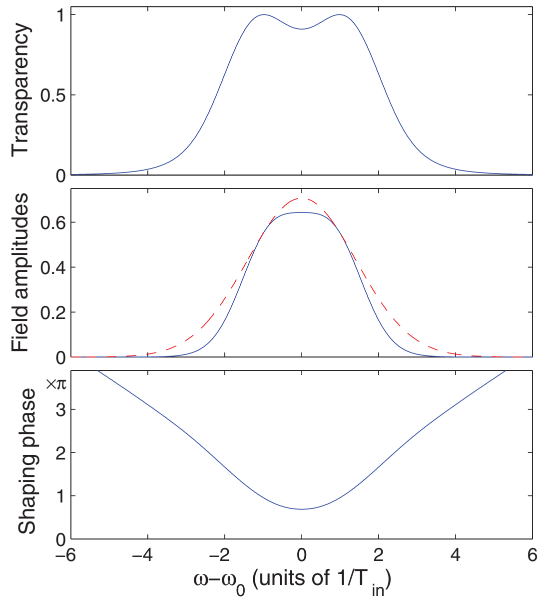

FIG. 2. (Color online) Spectral shaping corresponding to a parallel adiabatic passage as a function of the angular frequency for Gaussian input (23) and output (42) fields with $\Delta_0 = \Omega_0 = 1.5/T_{\rm in}$, and $T = 3T_{\rm in}$. Upper frame shows the transparency $\mathcal{T}(\omega)$, middle frame shows the Fourier transform of the input and output field shapes (units of $T_{\rm in}$); $\widetilde{\Lambda}_{T_{\rm in}}(\omega)$ (dashed line) and $\mathcal{T}(\omega)\widetilde{\Lambda}_{T_{\rm in}}(\omega)$ (full line). Lower frame shows the phase $\varphi(\omega)$. Here the coefficient κ of (42) is found to be $\kappa \approx 0.53$.

The finite spectrum of the input field imposes for the amplitude of the chirp $\Delta_0 = \Omega_0$:

$$\Omega_0 \lesssim 2/T_{\rm in}, \tag{41}$$

such that the maximum of the transparency is well located within the spectrum.

In practice, we choose the amplitude of the chirp $\Delta_0 = \Omega_0$ and the duration $T > T_{\rm in}$ of the output field of the form

$$\mathcal{E}(t) = \mathcal{E}_{0\rm in}\,\kappa e^{-(t/T)^2} e^{i[\omega_0 t + \phi(t)]}, \tag{42}$$

with the phase

$$\phi(t) = -\Omega_0 \int^t \mathrm{sgn}(s)\sqrt{1-\Lambda_T^2(s)}ds. \tag{43}$$

The additional factor κ in the amplitude of the field (42) has to be fixed such that $\mathcal{T}(\omega) \leqslant 1$.

Figure 2 shows an example of the resulting shaping. The transparency and the phase of the shaping are shown to be smooth functions that are expected to be easily implemented from a practical point of view.

VI. COMPARATIVE STUDY OF ROBUSTNESS

In this section we analyze and compare the robustness of DIAP and PLAP with respect to fluctuations of the instantaneous amplitude and detuning and also with respect to fluctuations of the pulse area.

The robustness with respect to instantaneous fluctuations of PLAP and DIAP is *a priori* questionable since these techniques are based on the use of the Davis-Dykhne-Pechukas formula in the complex-time plane requiring analytic functions as the pulse parameters.

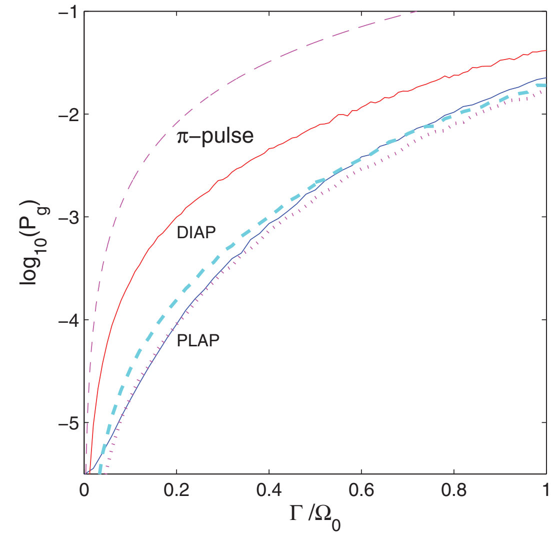

FIG. 3. (Color online) Infidelity (decimal logarithmic scale) for the PLAP (lower full line) and DIAP (upper full line) techniques for $\Omega_0 T = 5$ (corresponding to a pulse area $\int \Omega(t)dt = 5\sqrt{\pi}$), and for the π pulse (upper dashed line) with respect to an imperfect knowledge of the pulse area. The transitionless PLAP (T-PLAP) technique is shown as dotted and lower dashed lines for $\int \Omega(t)dt = 5\sqrt{\pi}$ (larger area for T-PLAP) and $\int |\Omega(t) - i\Omega_c(t)|dt = 5\sqrt{\pi}$ (same area for PLAP and T-PLAP), respectively.

A. Pulse area fluctuation

The robustness of the process with respect to an imperfect knowledge of the pulse areas is shown in Fig. 3 through ensemble averaging. We have determined the final populations by averaging over many realizations of an ensemble of systems with different peak Rabi frequencies uniformly distributed over the range $\Omega_0 \pm \Gamma/2$. Their peak Rabi frequency $\Omega_{0,j}$ is chosen as

$$\Omega_{0,j} = \Omega_0(1 + r_j\Gamma/\Omega_0), \tag{44}$$

where $-0.5 \leqslant r_j < 0.5$ is a uniformly distributed random number, Ω_0 is the average Rabi frequency, and Γ is the width of the probability distribution.

Such an averaging gives the following for the π-pulse population return:

$$\bar{P}_g^{(\pi)} = \frac{1}{2}\left(1 - \frac{2}{\Gamma\sqrt{\pi}}\sin\frac{\Gamma\sqrt{\pi}}{2}\right). \tag{45}$$

Figure 3 shows that the robustness of DIAP and PLAP is much improved with respect to the π-pulse technique. It also proves that PLAP is, in general, much superior (except for very small area fluctuations), despite the fact that DIAP leads, for the considered situation, to a better population transfer in the absence of fluctuation. In particular, we can see the remarkable result that the infidelity is smaller or equal to the benchmark of 10^{-4} for Γ/Ω_0 as large as 0.2 for PLAP. For this rate, the fidelity of PLAP is better by more than one order of magnitude than that of DIAP.

We have also tested the transitionless PLAP (T-PLAP); namely, with the use of an additional field that cancels the nonadiabatic coupling. The robustness with respect to an imperfect knowledge of the pulse area is displayed in Fig. 3 as dotted and lower dashed lines corresponding to a correcting Rabi frequency, giving an additional area with respect to the simple PLAP, and to the same total area in absolute value for PLAP and T-PLAP, respectively. This shows that T-PLAP does not improve PLAP overall (even when T-PLAP uses an additional area), but is on the contrary deteriorated over a large range of width (Γ) when the same area is taken.

B. Amplitude fluctuation

We model the instantaneous fluctuations of the field envelope with Gaussian white noise considering a relative deviation $\xi(t)$ as a stochastic variable of average and correlation:

$$\langle\xi(t)\rangle = 0, \quad \langle\xi(t)\xi(t')\rangle = 2\Gamma\delta(t-t'), \tag{46}$$

where the brackets $\langle\cdot\rangle$ denote an ensemble average and $\delta(t)$ is the Dirac δ function. Each sequence $\{\xi(t_i)\}$ of the ensemble is generated at discrete times t_i separated by the step Δt according to

$$\xi(t_i) = \sqrt{\frac{2\Gamma}{\Delta t}}\, r_n(t_i), \tag{47}$$

where $r_n(t_i)$ is a random number generated from a normal distribution with mean 0 and standard deviation 1. We have determined the instantaneous populations by averaging over many realizations of time histories.

For a sequence j of the ensemble, the Rabi frequency is more precisely defined at a discrete time t_i as

$$\Omega_j(t_i) = \Omega_0\Lambda(t_i)[1+\xi(t_i)], \tag{48}$$

with $\Omega_0\Lambda(t_i)$ being the ensemble average of the Rabi frequency at time t_i (i.e., without fluctuations).

Figure 4 shows the infidelity of the transfer as a function of Γ normalized by Ω_0. The PLAP technique is slightly better. The infidelity is smaller than 10^{-4} for $\Gamma \lesssim 10^{-4}\Omega_0$. We make the remarkable observation that the infidelity is nearly the same for PLAP and the π-pulse technique (except for very low noise rate).

Figure 5 displays a dynamics of PLAP for a single realization of the fluctuating Rabi frequency (48). Despite the relative smallness of Γ/Ω_0, one can notice the relatively large fluctuations of the Rabi frequency due to the small time step.

C. Phase fluctuation

1. Gaussian white noise

We can model the instantaneous fluctuations of the detunings as above with Gaussian white noise but considering now an instantaneous frequency offset for the stochastic variable $\xi(t)$ (46). This corresponds to a Wiener-Levy process for the corresponding phase [28]. This procedure is known to be equivalent to the use of the density matrix equation for the time evolution with the stochastic variables replaced by the constant dephasing rate Γ [29].

Figure 6 displays the infidelity for such a case. One can make the following observations: The three techniques rapidly

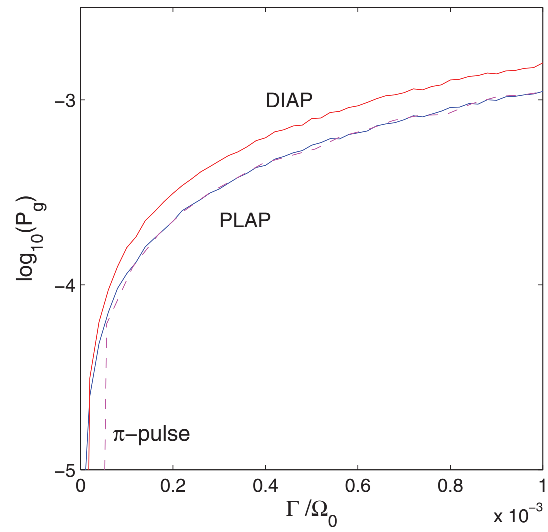

FIG. 4. (Color online) Infidelity (in decimal logarithmic scale) for the PLAP (lower full line) and DIAP (upper full line) techniques for $\Omega_0 T = 5$ and for the π-pulse technique (corresponding to $\Omega_0 T = \sqrt{\pi}$ and $\Delta = 0$, dashed line) for an ensemble average over a white-noise fluctuating field amplitude of rate Γ versus Γ/Ω_0 (with Ω_0 taken as the respective one).

fail even for relatively small dephasing rate (with respect to Ω_0) and the π-pulse technique is better than the DIAP and PLAP techniques. This latter result can be easily interpreted in terms of the dephasing rate: The dephasing corresponds to a destruction of the coherence of the superposition of state necessarily occurring during the dynamics leading to the population inversion. Its effect is thus reduced for a shorter duration of interaction which is the case for the π pulse (for the same given Ω_0). We can conclude that a dephasing process is more detrimental for adiabatic passage of longer duration.

If the dephasing rate is known, one can improve the efficiency of PLAP by modifying the dynamics such that it alternatively follows an ellipse in the parameter space (Ω,Δ) to accelerate it when the superposition is created, as shown in [30]. However, (i) this improvement is significant only for relatively large dephasing rates and (ii) it does not allow the benchmark 10^{-4} for the infidelity to be reached. For small dephasing rates such as the one considered in Fig. 6, the improvement is not significant.

A transfer of high efficiency even for appreciable dephasing rates can be recovered with the use of optimal control theory, as analyzed in Ref. [31]. Its adiabatic counterpart is an open question.

2. Gaussian exponentially correlated noise

The preceding Gaussian white noise can be made less destructive for adiabatic passage if one considers correlations. A typical model is the Ornstein-Uhlenbeck process with zero mean and exponentially correlated noise for the stochastic variable (see, for instance, [32]):

$$\langle\xi(t)\rangle = 0, \quad \langle\xi(t)\xi(t')\rangle = D\Gamma\exp(-\Gamma|t-t'|). \tag{49}$$

For large values of $\Gamma \gg D$, one recovers the Gaussian white noise (46). The opposite extreme case $\Gamma \ll D$ corresponds

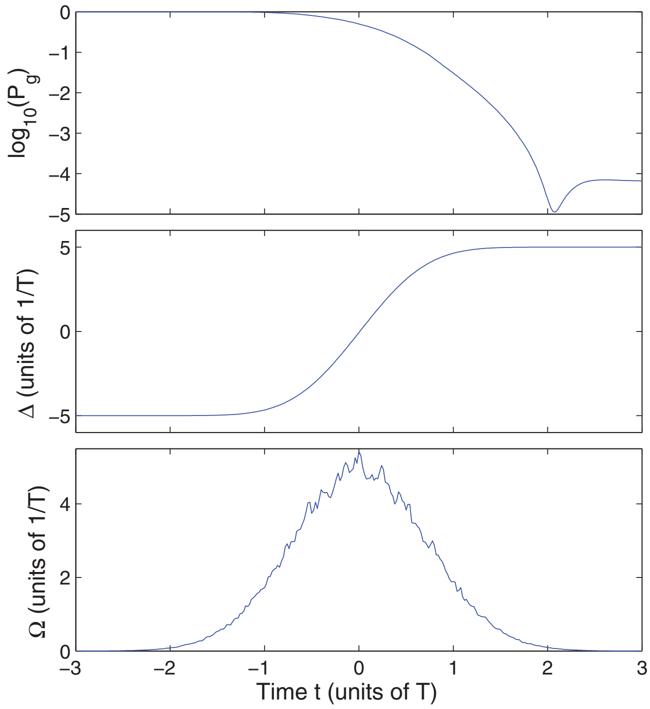

FIG. 5. (Color online) Dynamical infidelity (upper frame) for a realization of PLAP dynamics for $\Omega_0 T = 5$ and a white-noise fluctuating field amplitude (corresponding to the instantaneous Rabi frequency shown in the lower frame) of rate $\Gamma = 10^{-4}\Omega_0$. The detuning (shown in the middle frame) is assumed to be without fluctuation.

to an ensemble of fields with constant frequencies that obey Gaussian statistics with variance $D\Gamma$.

Figure 7 shows that, when the correlation is of larger width (i.e., for a smaller ΓT and a given product $D\Gamma T^2$), the DIAP technique is more efficient than the π-pulse technique, and the PLAP technique becomes well superior to the two other techniques.

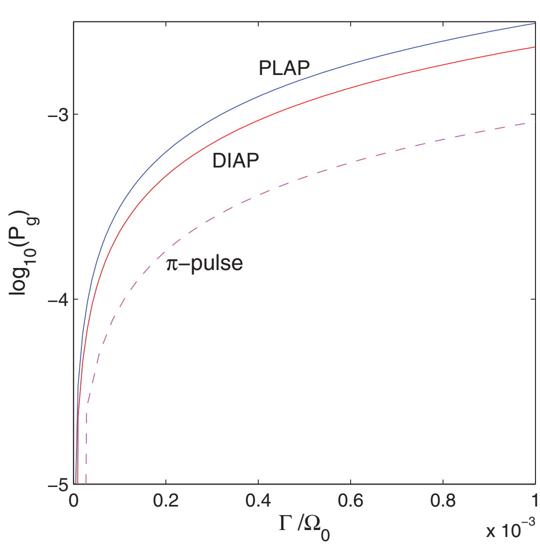

FIG. 6. (Color online) Same as Fig. 4 but for an ensemble average over a white-noise fluctuating detuning of rate Γ.

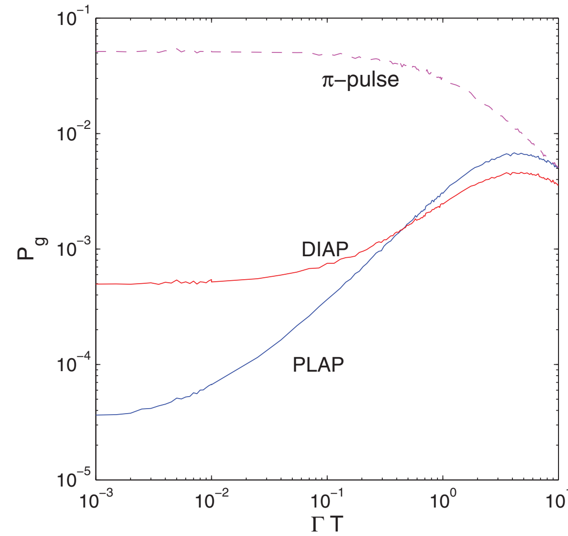

FIG. 7. (Color online) Infidelity (decimal log-log scale) for the PLAP and DIAP techniques for $\Omega_0 T = 5$ (full lines) and for the π-pulse technique (dashed line), for an ensemble average over an Ornstein-Uhlenbeck fluctuating field detuning as a function of ΓT for $D\Gamma T^2 = 0.1$.

VII. CONCLUSION AND DISCUSSION

In conclusion, we have investigated and compared two ways to reach population transfer of high fidelity by adiabatic passage; namely, the PLAP and DIAP techniques. Both techniques are based on a DDP analysis. PLAP is such that the eigenvalues are dynamically parallel, while DIAP corresponds to an adiabatic passage which is complemented by a destructive interference of the nonadiabatic transitions. One can remark that DIAP can be seen as an extension to a chirped-frequency interaction of the Rabi π-pulse transition since the latter can be interpreted as an interference (which is destructive for the probability of population return) of the two components, from the initial state split onto the two eigenstates, having acquired a dynamical phase [33,34].

In femtosecond regimes, these techniques could be implemented by a spectral shaping. DIAP with a Gaussian pulse and a linear chirping, when the mean frequency of the initial field matches the transition frequency, requires a device which shapes only the spectral phase. On the other hand, PLAP is more complicated to produce since it requires a shaping of both the spectral phase and amplitude in general.

We have analyzed the sensitivity of the techniques for various types of fluctuations. We have considered instantaneous fluctuations of the amplitudes and phases, and also an averaging upon randomly distributed Rabi-frequency areas. In practical implementation, we expect the latter to be the most critical issue due to an imperfect knowledge of the interaction details (through the area of the pulse itself, the position or the volume of the considered quantum system, ...). We have shown that PLAP is much more robust than DIAP (with a fidelity of more than one order of magnitude for rates of fluctuation that lead to an infidelity of order 10^{-4}, see Fig. 3) with respect to the lack of knowledge of the Rabi-frequency area. We have shown that one cannot improve the robustness of PLAP with respect to pulse areas if one complements it with a field that cancels the nonadiabatic coupling.

On the other hand, uncorrelated instantaneous fluctuations, even if they are expected to be relatively well controlled in practice, have been shown to be, in general, very detrimental for any coherent techniques. We have shown this for amplitude (Fig. 4) and phase (Fig. 6) white-noise fluctuations. The phase white-noise fluctuations have been shown to be more detrimental for adiabatic processes that always need more time to operate than the π-pulse technique (for a given field amplitude peak), since this noise corresponds to a dephasing decoherence that destroys the transient superpositions. If we introduce sufficient correlations in the noise, we have shown that we recover the superiority of the PLAP technique (Fig. 7).

Extending the results of this paper to systems with more than two levels necessitates, in principle, the derivation of a DDP formula for multilevel systems. Even for three-state systems, this has been shown to be complicated and not generically solvable due to numerous crossings in the complex plane [12]. Only specific symmetries in the Hamiltonian allow this extension. In the simplest case, the result can be interpreted as a local Landau-Zener analysis of the consecutive avoided crossings between pairs of levels assumed separated and that do not involve interfering paths, such that the final probability is the product of the probabilities corresponding to the consecutive avoided crossings [35,36]. Extending DIAP would thus require a model beyond this simple result. The PLAP technique has already been considered in a three-state system in a Raman configuration: The dynamics is designed such that the three corresponding eigenstates are parallel at all times [19]. Numerics has shown a robust transfer of very high accuracy.

For an N state with $N > 3$, finding parameters that would allow N parallel eigenstates is expected to be a difficult problem involving the design of many parameters. This could be solvable numerically for specific cases. A much weaker constraint, supported by a local Landau-Zener analysis, could be to drive the dynamics such that the eigenstate adiabatically transporting the population would correspond to an eigenvalue parallel to the closest one. This idea will be investigated; in particular, to guide the adiabatic path in a two-parameter space [37] in the context of state selectivity [38].

With the advances in producing ultrashort pulse of uv–xuv frequency, one could also consider the extension of such techniques with interacting pulses of a few cycles. In such an interaction, the rotating-wave approximation is itself questionable and PLAP should be investigated within the more general adiabatic Floquet theory [10].

ACKNOWLEDGMENTS

We acknowledge support from the French Agence Nationale de la Recherche (Project CoMoC), the European Marie Curie Initial Training Network Grant No. CA-ITN-214962-FASTQUAST, and from the Conseil Régional de Bourgogne.

[1] B. W. Shore, *The Theory of Coherent Atomic Excitation* (Wiley, New York, 1990); B. W. Shore, Acta Physica Slovaca **58**, 243 (2008).
[2] M. Shapiro and P. Brumer, *Principles of the Quantum Control of Molecular Processes* (Wiley, New York, 2003).
[3] D. Tannor, *Introduction to Quantum Mechanics: A Time-Dependent Perspective* (University Science Books, Sausalito, 2007).
[4] P. Král, I. Thanopulos, and M. Shapiro, Rev. Mod. Phys. **79**, 53 (2007).
[5] M. A. Nielson and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, Cambridge, 2000).
[6] U. Boscain, G. Charlot, J.-P. Gauthier, S. Guérin, and H. R. Jauslin, J. Math. Phys. **43**, 2107 (2002).
[7] M. H. Levitt, Prog. Nucl. Magn. Reson. Spectrosc. **18**, 61 (1986); R. Freeman, *Spin Choreography* (Spektrum, Oxford, 1997).
[8] S. S. Ivanov and N. V. Vitanov, Opt. Lett. **36**, 1275 (2011).
[9] N. V. Vitanov, T. Halfmann, B. W. Shore, and K. Bergmann, Annu. Rev. Phys. Chem. **52**, 763 (2001).
[10] S. Guérin and H. R. Jauslin, Adv. Chem. Phys. **125**, 147 (2003).
[11] A. M. Dykhne, Zh. Eksp. Teor. Fiz. **41**, 1324 (1962) [Sov. Phys. JETP **14**, 941 (1962)].
[12] J. P. Davis and P. Pechukas, J. Chem. Phys. **64**, 3129 (1976); J.-T. Hwang and P. Pechukas, *ibid.* **67**, 4640 (1977).
[13] A. Joye, H. Kuntz, and Ch-Ed. Pfister, Ann. Phys. **208**, 299 (1991).
[14] A. Joye, G. Mileti, and Ch-Ed. Pfister, Phys. Rev. A **44**, 4280 (1991).
[15] A. M. Weiner, Rev. Sci. Instrum. **71**, 1929 (2000).
[16] A. Monmayrant and B. Chatel, Rev. Sci. Instrum. **75**, 2668 (2004).
[17] S. Guérin, S. Thomas, and H. R. Jauslin, Phys. Rev. A **65**, 023409 (2002).
[18] G. S. Vasilev, A. Kuhn, and N. V. Vitanov, Phys. Rev. A **80**, 013417 (2009).
[19] G. Dridi, S. Guérin, V. Hakobyan, H. R. Jauslin, and H. Eleuch, Phys. Rev. A **80**, 043408 (2009).
[20] R. G. Unanyan, L. P. Yatsenko, K. Bergmann, and B. W. Shore, Opt. Commun. **139**, 48 (1997).
[21] M. V. Berry, J. Phys. A **42**, 365303 (2009); Xi Chen, I. Lizuain, A. Ruschhaupt, D. Guéry-Odelin, and J. G. Muga, Phys. Rev. Lett. **105**, 123003 (2010).
[22] G. S. Vasilev and N. V. Vitanov, Phys. Rev. A **70**, 053407 (2004).
[23] L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Dover, New York, 1987).
[24] G. S. Vasilev and N. V. Vitanov, J. Chem. Phys. **123**, 174106 (2005).
[25] B. Broers, H. B. van Linden van den Heuvell, and L. D. Noordam, Phys. Rev. Lett. **69**, 2062 (1992); D. J. Maas, C. W. Rella, P. Antoine, E. S. Toma, and L. D. Noordam, Phys. Rev. A **59**, 1374 (1999).
[26] M. Krug, T. Bayer, M. Wollenhaupt, C. Sarpe-Tudoran, T. Baumert, S. S. Ivanov, and N. V. Vitanov, New J. Phys. **11**, 105051 (2009).

[27] S. Zhdanovich, E. A. Shapiro, J. W. Hepburn, M. Shapiro, and V. Milner, Phys. Rev. A **80**, 063405 (2009).

[28] N. G. van Kampen, *Stochastic Processes in Physics and Chemistry*, 3rd ed. (Elsevier, Amsterdam, The Netherlands, North-Holland, 2007).

[29] G. S. Agarwal, Phys. Rev. A **18**, 1490 (1978).

[30] X. Lacour, S. Guérin, and H. R. Jauslin, Phys. Rev. A **78**, 033417 (2008).

[31] D. Sugny, C. Kontz, and H. R. Jauslin, Phys. Rev. A **76**, 023419 (2007).

[32] L. P. Yatsenko, V. I. Romanenko, B. W. Shore, and K. Bergmann, Phys. Rev. A **65**, 043409 (2002).

[33] M. Holthaus and B. Just, Phys. Rev. A **49**, 1950 (1994).

[34] L. P. Yatsenko, S. Guérin, and H. R. Jauslin, Phys. Rev. A **70**, 043402 (2004).

[35] C. E. Caroll and F. T. Hioe, J. Phys. A **19**, 1151 (1986).

[36] S. S. Ivanov and N. V. Vitanov, Phys. Rev. A **77**, 023406 (2008).

[37] S. Guérin, L. P. Yatsenko, and H. R. Jauslin, Phys. Rev. A **63**, 031403 (2001); L. P. Yatsenko, S. Guérin, and H. R. Jauslin, *ibid.* **65**, 043407 (2002).

[38] S. Thomas, S. Guérin, and H. R. Jauslin, Phys. Rev. A **71**, 013402 (2005).