CORA Cork Open Research Archive
Cartlann Taighde Oscailte Chorcaí

Title	Robust quantum control by a single-shot shaped pulse
Authors	Daems, D.;Ruschhaupt, Andreas;Sugny, Dominique;Guerin, Stéphane
Publication date	2013
Original Citation	Daems, D., Ruschhaupt, A., Sugny, D. and Guérin, S. (2013) 'Robust quantum control by a single-shot shaped pulse', Physical Review Letters, 111(5), 050404 (5pp). doi: 10.1103/PhysRevLett.111.050404
Type of publication	Article (peer-reviewed)
Link to publisher's version	https://journals.aps.org/prl/abstract/10.1103/PhysRevLett.111.050404 - 10.1103/PhysRevLett.111.050404
Rights	© 2013, American Physical Society
Download date	2026-11-16 08:03:10
Item downloaded from	https://hdl.handle.net/10468/4616



UCC
University College Cork, Ireland
Coláiste na hOllscoile Corcaigh

Robust Quantum Control by a Single-Shot Shaped Pulse

D. Daems,[1] A. Ruschhaupt,[2] D. Sugny,[1] and S. Guérin[1,*]
[1]Laboratoire Interdisciplinaire Carnot de Bourgogne, CNRS UMR 6303, Université de Bourgogne, BP 47870, 21078 Dijon, France
[2]Department of Physics, University College Cork, Cork, Ireland
(Received 17 April 2013; published 31 July 2013)

Considering the problem of the control of a two-state quantum system by an external field, we establish a general and versatile method allowing the derivation of smooth pulses which feature the properties of high fidelity, robustness, and low area. Such shaped pulses can be interpreted as a single-shot generalization of the composite pulse-sequence technique with a time-dependent phase.

DOI: 10.1103/PhysRevLett.111.050404 PACS numbers: 03.65.Aa, 32.80.Qk, 42.50.Dv, 42.50.Ex

Modern applications of quantum control, such as quantum information processing [1], require time-dependent schemes featuring three important issues: The transfer to the target state should be achieved (i) with a high fidelity, typically with an admissible error lower than 10^{-4} [1], (ii) in a robust way with respect to the imperfect knowledge of the system or to variations in experimental parameters, and (iii) with a minimum time of interaction and low field energy in order to prevent unwanted destructive intensity effects.

The Rabi method (see, e.g., [2]), corresponding to an exact resonant coupling between two quantum states, leads to an inversion $P = 1$ by a Rabi frequency of area $A = \pi$ (π-pulse technique). This defines the transfer quantum speed limit in the sense that the π area of the Rabi frequency is the smallest area that gives the inversion [3]. An extra energy can be used to satisfy additional constraints such as robustness with respect to the variations of parameters. In this framework, adiabatic techniques are famous examples [4]; they, however, require, in principle, a large pulse area and do not lead to an exact transfer. Improvements to optimize its efficiency by parallel adiabatic passage [5] or by shortcuts to adiabaticity [6] have been proposed.

A practical measure of the robustness can be defined through the deviation of the excitation profile as a function of the considered parameters. The use of composite pulses [7], replacing the single resonant pulse by a sequence of pulses with well-defined static phases, is a popular method for self-compensation of errors. These phases and the number of pulses can be determined so that the derivatives of the excitation profile are nullified order by order. Recent extensions with smooth pulses, such as an algebraic design of the composite sequence [8] and the use of an adiabatic phase [9], have been proposed. Typically, the profile P of the inversion deviates around $A = \pi$ as $P \sim 1 - (A - \pi)^{2n}$ with n the (odd) number of composite resonant π pulses. Techniques of optimal control are also actively developed for this purpose of robustness [10], but they only lead to numerical solutions.

In this Letter, we establish a strategy that allows a robust and precise transfer to a given target state by a pulse specifically shaped in phase and amplitude. This can be viewed as a generalization of the strategy relying on composite pulses to a single-shot pulse of a time-dependent phase. We first show that the issue of robustness can be reduced to nullifying integrals that cancel out the derivatives of the excitation profile order by order. The central result of this work is that this can be, in general, achieved by an oscillatory parametrization of the phase of the wave function. This continuous trigonometric basis is the key difference with respect to known methods which make use of stepwise functions (e.g., for the phase in the case of composite pulses). This allows an explicit derivation of the components of the shaped pulse. Our technique is explicitly shown for the robust inversion with respect to the pulse area, to the detuning, or to both parameters. This approach is, however, versatile and can be applied to other types of robustness and to the transfer to more complicated targets, such as fully robust quantum gates in view of applications in quantum information processing.

The resulting smooth shaped pulses feature the required properties of high fidelity, robustness, and low area. Furthermore, they have an explicit analytic form, much simpler than the ones usually obtained [11,12], with only a few parameters to adjust, contrary to numerical optimal control procedures (see, for instance, [13]).

Our discussion is based on a resonant system (rotating wave approximation) between two states $|1\rangle$ and $|2\rangle$ (of respective energies $\hbar\omega_1$ and $\hbar\omega_2$), for which the most general Hamiltonian governing the dynamics is of the following form (apart from a term proportional to the identity) [14]:

$$\hat{H}(\Delta, \Omega, \eta) = \frac{\hbar}{2}\begin{bmatrix} -\Delta(t) & \Omega(t)e^{-i\eta(t)} \\ \Omega(t)e^{i\eta(t)} & \Delta(t) \end{bmatrix}. \quad (1)$$

In the context of atoms interacting with a laser field [of phase $\omega_0 t - \eta(t)$ with ω_0 the mean frequency], the Rabi frequency is decomposed into an absolute value $\Omega(t) > 0$ (proportional to the field amplitude for a one-photon

 © 2013 American Physical Society

transition), of area $A = \int \Omega(t)dt$ and a phase $\eta(t)$. The detuning between the mean frequency of the field and the transition is then static: $\Delta(t) \equiv \delta = \omega_2 - \omega_1 - \omega_0$.

The errors in reaching the target state arise from the imperfect knowledge of the area A (for instance through an imperfect knowledge of the coupling constant), of the static detuning δ (corresponding to an inhomogeneous broadening of an ensemble), or to dynamical fluctuations of the pulse shape or of its instantaneous phase (corresponding to fluctuations of the time-dependent part of the detuning) [15]. For simplicity, we focus our discussion on the deviation with respect to the area A of the Rabi frequency and the static detuning δ. Robust methods are designed to improve the quadratic deviation benchmark of the Rabi method.

The solution of the time-dependent Schrödinger equation (TDSE) $i\hbar(\partial/\partial t)\phi = \hat{H}\phi$ can be parametrized with two angles $\theta \equiv \theta(t) \in [0, \pi]$, $\varphi - \eta \equiv \varphi(t) - \eta(t) \in]-\pi, \pi]$ on the Bloch sphere of Cartesian coordinates $\rho_x = \rho_{21} + \rho_{12} = \sin\theta\cos\varphi$, $\rho_y = i(\rho_{21} - \rho_{12}) = \sin\theta\sin\varphi$, $\rho_z = \rho_{11} - \rho_{22} = \cos\theta$ (with $\rho_{mn} \equiv \langle m|\phi\rangle \times \langle\phi|n\rangle$), and with a global phase $\gamma \equiv \gamma(t)$ as

$$\phi = \begin{bmatrix} e^{i\varphi/2}\cos(\theta/2) \\ e^{-i\varphi/2}\sin(\theta/2) \end{bmatrix} e^{-i\gamma/2}. \tag{2}$$

The phase transformation $T = \mathrm{diag}[e^{-i\eta/2}, e^{i\eta/2}]$ allows one to deal with a real symmetric Hamiltonian $T^\dagger \hat{H}(\delta, \Omega, \eta)T - i\hbar T^\dagger(dT/dt) = \hat{H}(\delta + \dot{\eta}, \Omega, 0)$ of solution $\psi = T^\dagger\phi$. This shows that the phase η can be incorporated in the detuning and interpreted as the rotation of the axes x and y about the z axis of the Bloch sphere. One can thus consider without loss of generality, the Hamiltonian equation (1) of the form $\hat{H}[\Delta(t), \Omega(t), 0]$. Inserting Eq. (2) in the TDSE, we get

$$\dot{\theta} = \Omega\sin\varphi, \tag{3a}$$

$$\dot{\varphi} = \Delta + \Omega\cos\varphi\,\mathrm{cotan}\theta, \tag{3b}$$

$$\dot{\gamma} = \Omega\frac{\cos\varphi}{\sin\theta} = \dot{\theta}\frac{\mathrm{cotan}\varphi}{\sin\theta}. \tag{3c}$$

We assume $\phi(t_i) = |1\rangle$ as an initial condition, corresponding to the initial conditions $\theta(t_i) \equiv \theta_i = 0$ (north pole), $\varphi(t_i) \equiv \varphi_i = \gamma(t_i) \equiv \gamma_i$ (not specified by the initial state). We consider the inversion, which is achieved for the final condition $\theta(t_f) \equiv \theta_f = \pi$ (south pole). For any trajectory featured by $\varphi(t)$, $0 \leftarrow \theta(t) \rightarrow \pi$ on the Bloch sphere, one can integrate Eq. (3a): $\theta_f - \theta_i = \pi = \int_{t_i}^{t_f} ds\Omega(s)\sin\varphi(s)$. Since $\int_{t_i}^{t_f} ds\Omega(s)\sin\varphi(s) \leq \int_{t_i}^{t_f} ds\Omega(s)$, one concludes that the inversion is achieved when $\int_{t_i}^{t_f} ds\Omega(s) \geq \pi$, which is consistent with Ref. [3]. The minimum area π is obtained for the meridian $\varphi = \pi/2$, implying $\gamma_i = \pi/2$, $\dot{\gamma} = 0$ from Eq. (3c) and $\Delta = 0$ from Eq. (3b), which corresponds to the Rabi transfer. On the other hand, the ideal adiabatic solution is derived when $A \gg 1$ giving $\varphi \rightarrow 0, \pi$ [in order to have a bounded $\dot{\theta}$ from Eq. (3a)] and the well-known dynamical phase $\gamma(t)/2 \rightarrow \pm\int_{t_i}^{t}\sqrt{\Omega(s)^2 + \Delta(s)^2}ds/2$ from Eq. (3c).

More generally, the transfer to a given target state up to a (global) phase corresponds to the desired final conditions θ_f and φ_f. The control of the phase of the target state would require the additional condition $\gamma(t_f) \equiv \gamma_f$, which will not be considered here. We will additionally assume $\Omega(t_i) = \Omega(t_f) = 0$ for practical implementation, and a strictly monotonic increasing of θ, i.e., $\dot{\theta} > 0$, with $\Omega \geq 0$, which leads to $0 \leq \varphi \leq \pi$ from Eq. (3a).

The goal of the control consists in finding a particular solution of Eq. (3) reaching the target state, which is robust and of lowest Ω area. The strategy is similar to the one used for generating composite pulses: One nullifies the derivatives of the transfer profile with respect to the considered parameters. This is here achieved with the Hamiltonian of the form

$$H_{\alpha,\delta}(t) = \frac{\hbar}{2}\begin{bmatrix} -\Delta(t) & \Omega(t) \\ \Omega(t) & \Delta(t) \end{bmatrix} + \frac{\hbar}{2}\begin{bmatrix} -\delta & \alpha\Omega(t) \\ \alpha\Omega(t) & \delta \end{bmatrix}. \tag{4}$$

The corresponding solution $\phi_{\alpha,\delta}(t)$, parametrized by α and δ, depends on the functions $\Omega(t)$ and $\Delta(t)$, which are chosen such that the complete transfer to the target state ϕ_T is achieved at $t = t_f$ for $\delta = 0$ and $\alpha = 0$: $\phi_{0,0}(t_f) \equiv \phi_0(t_f) = \phi_T$. The program detailed below can be summarized as follows: We first determine at the end of the process $\phi_{\alpha,\delta}(t_f)$ perturbatively, from the exact transfer ϕ_T as the zeroth order, which gives an expansion of the profile $|\langle\phi_T|\phi_{\alpha,\delta}(t_f)\rangle|^2$ [see Eq. (5)]. We calculate the general expression of the terms of the perturbation at any order [see (8) for the second order, and the symbolic path diagram in Fig. 1 for higher orders]. The functions $\Omega(t)$ and $\Delta(t)$, among all of those that satisfy the transfer for $\alpha = 0$ and $\delta = 0$, are next chosen to nullify, at a given order, the derivatives of the profile with respect to α and δ. This task is analytically solved by an inverse engineering of a family of solution $\phi_0(t)$ which we choose by an appropriate parametrization of the global phase γ [see (10)]. This parametrization features free parameters that have to be adjusted to nullify the derivatives.

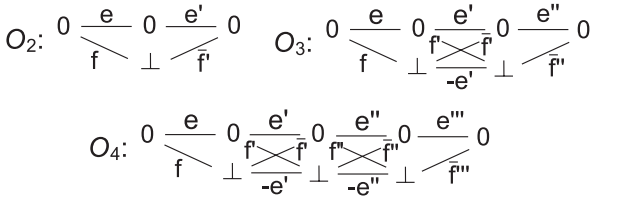


FIG. 1. Symbolic path diagrams giving the construction of the O_n integrals. The symbol e stands for $e(t)$, e' for $e(t')$, and so on. For instance, for $n = 3$ ($n = 4$), the diagram features four (eight) paths. Its extension for larger n is direct.

The perturbative expansion of $\phi_{\alpha,\delta}(t_f)$ with respect to α and δ taking the second matrix term of Eq. (4) as a perturbation denoted V reads $\langle\phi_T|\phi_{\alpha,\delta}(t_f)\rangle = 1 + O_1 + O_2 + O_3 + \cdots$, where O_n denotes the term of total order n. It gives for the excitation profile

$$|\langle\phi_T|\phi_{\alpha,\delta}(t_f)\rangle|^2 = 1 + \tilde{O}_1 + \tilde{O}_2 + \tilde{O}_3 + \cdots, \quad (5)$$

with $\tilde{O}_n$ the term of order n. The first two terms read

$$O_1 = -i\int_{t_i}^{t_f}\langle\phi_0(t)|V(t)|\phi_0(t)\rangle dt \equiv -i\int_{t_i}^{t_f} e(t)dt, \quad (6a)$$

$$O_2 = (-i)^2\int_{t_i}^{t_f} dt\int_{t_i}^{t} dt'[e(t)e(t') + f(t)\bar{f}(t')], \quad (6b)$$

with $\tilde{O}_1 = O_1 + \bar{O}_1$, $e = -(1/2)(\delta\cos\theta - \alpha\dot{\gamma}\sin^2\theta)$,

$$f = \langle\phi_0|V|\phi_\perp\rangle = \frac{1}{2}\left[\delta\sin\theta + \alpha\left(\frac{1}{2}\dot{\gamma}\sin 2\theta - i\dot{\theta}\right)\right]e^{i\gamma}, \quad (7)$$

and the orthogonal solution of the TDSE $\phi_\perp(t) = [e^{i\varphi/2}\sin(\theta/2), -e^{-i\varphi/2}\cos(\theta/2)]^T e^{i\gamma/2}$ such that $\langle\phi_\perp(t)|\phi_0(t)\rangle = 0$. The other terms can be determined from the symbolic diagrams depicted in Fig. 1. Since $e(t)$ is real, there is no first-order deviation for the excitation profile. One can simplify the second order as

$$\tilde{O}_2 \equiv O_2 + \bar{O}_2 + \bar{O}_1 O_1 = -\left|\int_{t_i}^{t_f} f(t)dt\right|^2 \quad (8)$$

using the property $\int_\tau^T dt\int_\tau^t dt'[a(t)b(t') + a(t')b(t)] = \int_\tau^T a(t)dt\int_\tau^T b(t)dt$. This property also implies $\int_\tau^T dta(t)\times\int_\tau^T dtb(t)\int_\tau^t dt'c(t') = \int_\tau^T dt\int_\tau^t dt'\int_\tau^{t'} dt''[a(t)b(t')c(t'') + b(t)a(t')c(t'') + b(t)c(t')a(t'')]$ and extends as $\int_\tau^T dt\times\int_\tau^t dt'\int_\tau^{t'} dt''\sum_\sigma a(t^{[\sigma(0)]})b(t^{[\sigma(1)]})c(t^{[\sigma(2)]}) = \int_\tau^T a(t)dt\times\int_\tau^T b(t)dt\int_\tau^T c(t)dt$, where $\sum_\sigma$ means the summation over the six permutations of the number of primes (between 0 and 2). This is used to determine relatively simple integrals in a similar way for higher orders. The third order reads $\tilde{O}_3 = -4\int_{t_i}^{t_f} dt\int_{t_i}^{t} dt'\int_{t_i}^{t'} dt''\mathrm{Im}[\bar{f}(t)e(t')f(t'')]$. Robustness at a given order n is obtained when the parameters of the field are chosen such that they allow nullifying the integrals $\tilde{O}_m$ for $m \le n$. The second-order robustness issue corresponds to the two equations:

$$\int_{t_i}^{t_f} e^{i\gamma}\sin\theta dt = 0, \qquad \int_{\theta_i}^{\theta_f} e^{i\tilde{\gamma}}\sin^2\theta d\theta = \frac{1}{4}[e^{i\tilde{\gamma}}\sin 2\theta]_{\theta_f}^{\theta_i}, \quad (9)$$

for the robustness with respect to the detuning δ and to the pulse area, respectively. If one considers the robustness only with respect to the pulse area, the corresponding equation involves an integral which is independent of the particular temporal parametrization of θ. For some integrals, such as, for instance, the second order robustness with respect to the detuning [left equation of Eq. (9)], we need additionally an explicit time parametrization of θ. For the Rabi method ($\Delta = 0$), the equations in (9) cannot be satisfied for any pulse shape: It is robust up to the first order for the excitation profile. Below, we explicitly derive solutions for the issue of inversion (with θ varying from 0 to π). The same technique applies for other target states.

These equations (9) (and the ones corresponding to higher orders) can be solved by an oscillatory parametrization (Fourier series) of the global phase as a function of θ, $\gamma(t) \equiv \tilde{\gamma}(\theta)$ (with the label a or b to identify it):

$$\tilde{\gamma}_a(\theta) = \varphi_i + 2\theta + C_1\sin(2\theta) + \cdots + C_n\sin(2n\theta) + \cdots, \quad (10a)$$

$$\tilde{\gamma}_b(\theta) = \varphi_i + \theta + C_1\sin(2\theta) + \cdots + C_n\sin(2n\theta) + \cdots, \quad (10b)$$

with the choice $\theta(t) = \pi[\mathrm{erf}(t/T) + 1]/2$ (giving a smooth pulse), and T featuring the duration of interaction. The choice of these parametrizations has been guided by the simple following result: For Eq. (10a) [(10b)], the imaginary [real] parts of Eq. (9) are nullified for all C_n's. They also allow a simple expression in view of controlling the phases, since, from Eq. (3c), we have $\mathrm{cotan}\varphi = \sin(\theta)d\tilde{\gamma}/d\theta$. They give $\gamma_f = \varphi_i + 2\pi[\varphi_i + \pi]$ for parametrization, Eq. (10a) [(10b)]. In both cases, due to the oscillatory nature of the parametrization, one has to adjust only a few coefficients C_n in order to nullify all the required integrals at a given order.

TABLE I. Robustness of order n (i.e., $\tilde{O}_{m\le n} = 0$) with respect to the area (referred to as type A), to the detuning (type δ), or both (type $A\delta$), with the parametrizations, Eq. (10a) (a) or Eq. (10b) (b), and the coefficients C_j, $j = 1, 2, 3$, ($C_{j>3} = 0$). We have considered parametrizations leading to low pulse areas.

Type	Parametrization	Order	C_1	C_2	C_3	Pulse area ($\times\pi$)
A	a	3	-1	0	0	2.16
A	b	3	-1.6788	0	0	2.09
δ	a	3	-0.2305	0	0	1.78
$A\delta$	b	2	-1.189	0.7285	0	2.23
A	a	5	-2.4864	-0.74	0	3.14
A	a	7	-3.46	-1.365	-0.5	3.86

Some obtained coefficients are summarized in Table I. The resulting pulse area $\int_{t_i}^{t_f}\Omega(t)dt = \int_0^{\pi}\sqrt{1+(\dot{\tilde{\gamma}})^2\sin^2\theta}d\theta$ is mentioned. One can notice increasing areas when more coefficients different from zero are considered. Table I shows, for instance, that one obtains a pulse area of only 1.78π for the *third order robustness* solely with respect to the detuning taking the parametrization, Eq. (10a), with $C_1 = -0.2305$. Alternatively, we obtain at best a pulse area of 2.09π for the third order robustness solely with respect to the pulse area taking the parametrization, Eq. (10b). We emphasize that this latter choice allows a smooth pulse of a slightly smaller area than the nonsmooth pulse proposed in [15] (with $C_1 = -1$, see first line of Table I). In both cases, the third order terms $\tilde{O}_3$ are systematically nullified when the coefficients are adjusted to nullify the second order, Eq. (8), due to the symmetry of the parametrization. One can alternatively force robustness with respect to both the detuning δ and the pulse area, nullifying both terms of Eq. (9) (see the fourth line of Table I). This shows the remarkable result that the obtained pulse area is only slightly larger than the one obtained above for robustness solely with respect to the pulse area (but for a robustness of order 2).

If one considers robustness at high order, one has to nullify the higher order terms. The results for the robustness with respect to the pulse area at orders 5 and 7 are shown in Table I.

The robustness with respect to the pulse area at different orders is demonstrated in Fig. 2: It shows that the transfer profile as a function of α [as defined in Eq. (4) with $\delta = 0$] becomes flatter and flatter when one nullifies higher order terms in the perturbative expansion, Eq. (5). We can notice that the profile is not symmetric with respect to $\alpha = 0$:

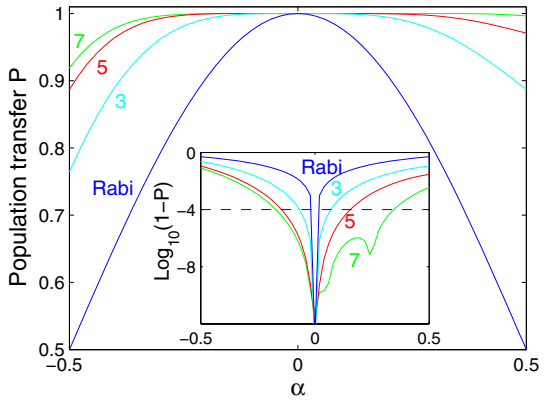


FIG. 2 (color online). Robustness with respect to the pulse area: Population transfer P at the end of the pulse as a function of the relative deviation α (dimensionless) of the area. Different orders of robustness (from order 3 to 7, as indicated next to the curves) with the coefficients indicated in Table I (type A, parametrization a) are compared to the standard Rabi method. Inset: Logarithm to the basis 10 of the deviation.

The transfer is less robust for a smaller area (corresponding to negative values of α). The inset of Fig. 2 depicts the deviation of the excitation profile at a logarithmic scale. It shows the remarkable result that the inversion is accomplished with the 10^{-4} high-fidelity accuracy benchmark (indicated by the horizontal dashed line) even with an error in area up to 17% for the highest order solution (limited by negative deviations).

Figure 3 displays the derived temporal shapes of the Rabi frequencies: They feature a more and more broadened profile which oscillates more and more for higher order robustness, being reminiscent to the sequence of composite pulses which would not be completely off between the peaks.

In conclusion, we have derived a robust transfer technique with a single-shot shaped pulse which appears as a fast alternative technique to composite pulses. We have reduced the robustness issue to a problem of nullifying integrals, achieved for an oscillatory parametrization of the global phase with only a few parameters to be adjusted. The resulting smooth shaped pulse features properties of high fidelity, high-order robustness, and low area. For robustness with respect to the pulse area, to the detuning, or to both parameters, we have derived pulses with an explicit and relatively simple form, easily implementable experimentally. We emphasize that our analysis allows the treatment of already complex systems, beyond isolated two-level systems. For instance it allows the study of an ensemble (possibly infinite) of uncoupled $1/2$ spins driven by rf fields (nuclear magnetic resonance), each interacting with different amplitude and detuning due to the field inhomogeneity. Such a system is usually treated fully numerically (see [11,13]). The derived solution could find important applications in this context, such as magnetic resonance imaging [16]. The presence of other nonresonant states can be treated in an effective way by adiabatic elimination, inducing Stark shifts [14]. This corresponds to another type of perturbation with respect to which robustness can be analyzed using the present technique. For the case of a resonant multilevel system, it has very recently been shown that one can eliminate unwanted transitions while controlling the transfer between desired

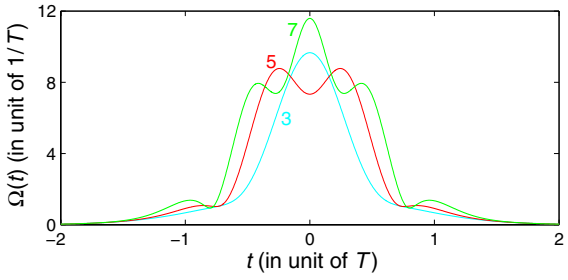


FIG. 3 (color online). Derived Rabi frequency pulse shapes for robustness of orders 3, 5, and 7 with respect to the pulse area (with the parameters of Table I).

transitions by a sequence of composite pulses [17]. This aim could be alternatively reached by a single-shot pulse with our method. The techniques which we present are thus versatile and can be applied to other types of robustness and targets. For instance, reaching a robust coherent superposition of state [with θ varying from 0 to the desired value, and φ_f chosen from Eq. (3c)] would also need a few coefficients to be adjusted for the pulse. Ultimately, adapting the parametrization, one could also produce robust propagators (gates), suitable for quantum information processing. In this case, more integrals to be nullified will be involved, which will need more coefficients C_n to be adjusted: Algorithms of optimal control could then be used to optimize these coefficients [18]. This task will, however, be relatively easy in comparison with traditional optimal control techniques which need, in general, more than a hundred coefficients to be adjusted.

The derived robust pulses could also find applications in ultrafast (femtosecond) processes, requiring their production in the spectral domain [19].

We acknowledge support from the European Marie Curie Initial Training Network GA-ITN-214962-FASTQUAST and from the Conseil Régional de Bourgogne.

*sguerin@u-bourgogne.fr

[1] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, Cambridge, 2000); D. Bouwmeester, A. Ekert, and A. Zeilinger, *The Physics of Quantum Information: Quantum Cryptography, Quantum Teleportation, Quantum Computation* (Springer-Verlag, Berlin, 2000).

[2] L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Dover, New York, 1987).

[3] U. Boscain, G. Charlot, J.-P. Gauthier, S. Guérin, and H. R. Jauslin, J. Math. Phys. (N.Y.) **43**, 2107 (2002).

[4] N. V. Vitanov, M. Fleischhauer, B. W. Shore, and K. Bergmann, Adv. At. Mol. Opt. Phys. **46**, 55 (2001); N. V. Vitanov, T. Halfmann, B. W. Shore, and K. Bergmann, Annu. Rev. Phys. Chem. **52**, 763 (2001); S. Guérin and H. R. Jauslin, Adv. Chem. Phys. **125**, 147 (2003).

[5] S. Guérin, S. Thomas, and H. R. Jauslin, Phys. Rev. A **65**, 023409 (2002); G. Dridi, S. Guérin, V. Hakobyan, H. R. Jauslin, and H. Eleuch, *ibid.* **80**, 043408 (2009); S. Guérin, V. Hakobyan, and H.-R. Jauslin, *ibid.* **84**, 013423 (2011).

[6] R. G. Unanyan, L. P. Yatsenko, K. Bergmann, and B. W. Shore, Opt. Commun. **139**, 48 (1997); M. V. Berry, J. Phys. A **42**, 365303 (2009); X. Chen, I. Lizuain, A. Ruschhaupt, D. Guéry-Odelin, and J. G. Muga, Phys. Rev. Lett. **105**, 123003 (2010).S. Ibanez, X. Chen, E. Torrontegui, J. G. Muga, and A. Ruschhaupt, Phys. Rev. Lett. **109**, 100403 (2012).X. Chen, A. Ruschhaupt, S. Schmidt, A. del Campo, D. Guéry-Odelin, and J. G. Muga, Phys. Rev. Lett. **104**, 063002 (2010).

[7] M. H. Levitt, Prog. Nucl. Magn. Reson. Spectrosc. **18**, 61 (1986); R. Tycko, Phys. Rev. Lett. **51**, 775 (1983); S. Wimperis, J. Magn. Reson. **109**, 221 (1994); R. Freeman, *Spin Choreography* (Spektrum, Oxford, 1997).

[8] B. T. Torosov and N. V. Vitanov, Phys. Rev. A **83**, 053420 (2011).

[9] B. T. Torosov, S. Guérin, and N. V. Vitanov, Phys. Rev. Lett. **106**, 233001 (2011).

[10] X. Wang, L. S. Bishop, J. P. Kestner, E. Barnes, K. Sun, and S. D. Sarma, Nat. Commun. **3**, 997 (2012); P. Owrutsky and N. Khaneja, Phys. Rev. A **86**, 022315 (2012).

[11] T. E. Skinner, T. O. Reiss, B. Luy, N. Khaneja, and S. J. Glaser, J. Magn. Reson. **163**, 8 (2003).

[12] G. Turinici and H. Rabitz, Phys. Rev. A **70**, 063412 (2004).

[13] K. Kobzar, T. E. Skinner, N. Khaneja, S. J. Glaser, and B. Luy, J. Magn. Reson. **170**, 236 (2004).

[14] B. W. Shore, *The Theory of Coherent Atomic Excitation* (Wiley, New York, 1990); *Manipulating Quantum Structure Using Laser Pulses* (Cambridge University Press, New York, 2011).

[15] A Ruschhaupt, X. Chen, D. Alonso, and J. G Muga, New J. Phys. **14**, 093040 (2012).

[16] M. Lapert, Y. Zhang, M. A. Janich, S. J. Glaser, and D. Sugny, Sci. Rep. **2**, 589 (2012).

[17] G. T. Genov and N. V. Vitanov, Phys. Rev. Lett. **110**, 133002 (2013).

[18] T. E. Skinner and N. I. Gershenzon, J. Magn. Reson. **204**, 248 (2010).

[19] M. Wollenhaupt, V. Engel, and T. Baumert, Annu. Rev. Phys. Chem. **56**, 25 (2005); M. Wollenhaupt, A. Assion, and T. Baumert, in *Springer Handbook of Lasers and Optics*, edited by F. Träger (Springer, New York, 2007), Chap. 12.