Frequency-modulated pulses for quantum bits coupled to time-dependent baths

Benedikt Fauseweh,[1,*] Stefano Pasini,[2,†] and Götz S. Uhrig[1,‡]
[1] *Lehrstuhl für Theoretische Physik I, TU Dortmund, Otto-Hahn Straße 4, DE-44221 Dortmund, Germany*
[2] *Forschungszentrum Jülich, DE-52425 Jülich, Germany*

(Received 2 December 2011; published 7 February 2012)

We consider the coherent control of a quantum bit by the use of short pulses with finite duration τ_p. By shaping the pulse, we perturbatively decouple the dynamics of the bath from the dynamics of the quantum bit during the pulse. Such shaped pulses provide single quantum bit gates robust against decoherence which are useful for quantum-information processing. We extend previous results in two ways: (i) we treat frequency-modulated pulses and (ii) we pass from time-independent baths to analytically time-dependent baths. First- and second-order solutions for π and $\pi/2$ pulses are presented. They are useful in experiments where amplitude modulation is difficult to realize.

DOI: 10.1103/PhysRevA.85.022310   PACS number(s): 03.67.Pp, 82.56.Jn, 76.60.Lz, 03.65.Yz

I. INTRODUCTION

The occurrence of decoherence in quantum systems is one of the main difficulties to be overcome in modern experiments. Especially quantum-information processing (QIP) requires the quantum system to remain in fixed phase relations between the application of quantum gates. Otherwise one cannot benefit from the quantum parallelism making QIP such a powerful technique.

A generic example for a quantum bit (qubit) is a spin $S = 1/2$. One of the first implementations of a quantum algorithm was realized with nuclear magnetic resonance (NMR) [1]. The qubits are encoded in the nuclear spin degrees of freedom. We also use the spin language to describe the states and dynamics of the qubit. The state $\uparrow$ is identified with the logical $|1\rangle$ and the state $\downarrow$ with the logical $|0\rangle$.

The loss of coherence is induced by the coupling of the spin to its environment, the so-called bath. One way to suppress this coupling is the application of suitable control pulses first introduced by Hahn in 1950 [2] for NMR experiments. This idea led to the development of sequences for control pulses ranging from the Carr-Purcell-Meiboom-Gill (CPMG) cycle [3,4] to more and more complex control schemes [5]. In QIP, this approach is known under the name of dynamic decoupling (DD) [6–8]. Theoretically, dynamic decoupling can be achieved to infinite order in the duration T of the total pulse sequence [9,10]. One particularly efficient way to deal with pure dephasing decoherence is the use of theoretically optimized DD (Uhrig DD) [11–13]. It has been successfully implemented experimentally [14,15].

The pulses considered in theoretical studies of DD schemes are mostly ideal in the sense that they have an infinite amplitude and that they act instantaneously in time corresponding to Dirac δ pulses. Of course, this property cannot be realized in experiments. If the finite pulse duration is taken into account in calculations, it turns out to be a nuisance in most cases (see, for instance, Refs. [10,16]). Hence, there is an abundant literature on pulse shaping and optimization which we can mention only partly [16–31] (for a book see Ref. [32]). We emphasize, however, that a suitably shaped pulse can be integrated into a DD sequence such that the high-order suppression of decoherence is hardly hampered [33,34].

The majority of the existing theoretical studies of pulse shaping consider pulses acting on the two-dimensional Hilbert space. The goal is to design robust pulses which tolerate a maximum of frequency offset or other inaccuracies of the pulse [17–23,26]. The next stage of complexity includes random time-dependent classical noise, which is still described by classical fields coupled to the spin [25]. The maximum stage of complexity considers a fully quantum mechanical bath which means that the qubit is coupled to a macroscopic quantum mechanical system by noncommuting operators [16,24,27–31]. It is on this level that our present study is situated. We stress that a quantum mechanical pulse, which is robust against a coupling to its environment, constitutes an appropriate single-qubit gate.

In particular, we extend previous work [30] in two ways: (i) We allow for analytically time-dependent bath operators, both in the spin-bath coupling and in the Hamiltonian of the bath. Such time dependence may, for instance, arise from a time-dependent reference frame [35]. (ii) We propose frequency-modulated pulses while before only amplitude-modulated pulses were studied [16,24,27,28,30,31] except in the general no-go theorem in Ref. [29]. We stress that in the NMR context amplitude and phase-modulated pulses have been discussed intensively [21–23]. But to our knowledge these investigations do not comprise quantum mechanical baths nor dynamic classical noise.

Explicitly, we compute continuous solutions for π and $\pi/2$ pulses realized by frequency modulation (see also Ref. [23]). The consideration of frequency modulation is motivated from experimental situations where the frequency of a pulse can be controlled more accurately or more easily than its amplitude. Thus the present study is complementary to preceding ones.

The paper is organized as follows: In Sec. II, we give an overview of the model under study and motivate our ansatz for the time evolution of the whole system. In Sec. III we derive the perturbative expansion for a generic time-dependent bath. We require that the time dependence is analytical in order to be able to apply a perturbative approach. Then we introduce the frequency-modulated ansatz in Sec. IV and specialize the

*benedikt.fauseweh@tu-dortmund.de
†s.pasini@fz-juelich.de
‡goetz.uhrig@tu-dortmund.de

general equations for this specific case. The solutions found for first- and second-order pulses are discussed in Sec. V and we finally conclude in Sec. VI.

II. MODEL AND ANSATZ

We consider the general case of a spin coupled to a time-dependent bath

$$H(t) = H_{\mathrm{b}}(t) + \vec{\sigma} \cdot \vec{A}(t), \tag{1}$$

where $H_{\mathrm{b}}(t)$ denotes the part of the Hamiltonian that acts only on the bath. We refer to it as the bath Hamiltonian. The vector of Pauli matrices $\vec{\sigma}$ acts on the Hilbert space of the spin $S = 1/2$ while $\vec{A}(t)$ is a vector of bath operators to which the spin is coupled. No special operator structure is assumed for the bath operators; i.e., the commutators $[A_i(t'), A_i(t)]$, $[A_i(t'), A_j(t)]$, and $[A_i(t'), H_{\mathrm{b}}(t)]$ do not need to vanish.

The model (1) comprises typical cases such as a bosonic bath or a spin network. Relevant experimental systems comprise the electronic spin in a quantum dot coupled to the bath of nuclear isotope spins [36] or the spin of a nitrogen vacancy center in diamond interacting again with a bath of nuclear isotope spins [37]. For our purposes, we require that the time dependence of the operators $H_{\mathrm{b}}(t)$ and $\vec{A}(t)$ is analytical so that they can be expanded in time:

$$\vec{A}(t) = \vec{A}_0 + \vec{A}_1 t + \vec{A}_2 t^2 + \cdots , \tag{2a}$$

$$H_{\mathrm{b}}(t) = H_{\mathrm{b},0} + H_{\mathrm{b},1} t + H_{\mathrm{b},2} t^2 + \cdots . \tag{2b}$$

This analyticity is often fulfilled, e.g., in rotating reference frames or in the operator interaction picture. For fast time-dependencies, however, the above expansion is not useful because the derivatives are large. Very fast oscillatory time-dependencies are better treated by average Hamiltonian theory.

Our model includes the common case of a purely dephasing bath, i.e., a spin coupled only along the σ_z direction to the bath. This model is justified in experiments where the dephasing time T_2 is significantly lower than the longitudinal relaxation time T_1. This is the case if the energetic splitting between the states with $\sigma_z = 1$ and $\sigma_z = -1$ is large.

The coupling strength between the spin and the bath is given by $\lambda := ||\vec{A}(t)||$ while the energy of the bath is defined to be $\omega_{\mathrm{b}} := ||H_{\mathrm{b}}(t)||$. If these operators are not bounded, that means if λ and ω_{b} cannot be defined by the operator norms, we refer by λ and ω_{b} to the generic energy scales of the corresponding operators. For instance, in a bosonic bath ω_{b} is the upper cutoff of the bosonic energy spectrum. The energy scales serve as reference values for τ_{p}. That means that we aim at an expansion in the dimensionless ratios $\omega_{\mathrm{b}}\tau_{\mathrm{p}}$ and $\lambda\tau_{\mathrm{p}}$.

Applying the control pulse to the system, the term

$$H_v(t) = \vec{\sigma} \cdot \vec{v}(t) \tag{3}$$

is added to the Hamiltonian $H(t)$. Here $\vec{v}(t)$ is a vector of amplitudes describing the controllable time-dependent shape of the pulse. The normalized vector $\vec{v}(t)/|\vec{v}(t)|$ is the current axis of rotation at time t while the norm $|\vec{v}(t)|$ describes the magnitude of the control term which determines the velocity of rotation. Without loss of generality, we assume that the pulse starts at $t = 0$ and ends at $t = \tau_{\mathrm{p}}$. The time evolution between 0 and τ_{p} of the combined system reads

$$U(\tau) = T\left[\exp\left(-i\int_0^{\tau} H(t)dt - i\int_0^{\tau} H_v(t)dt\right)\right], \tag{4}$$

where T stands for the standard time ordering.

Our aim is to perturbatively decouple the time evolution of the spin from the time evolution of the bath during the pulse. This motivates the following ansatz for the time evolution of the whole system

$$U(\tau_{\mathrm{p}}) = U_{\mathrm{b}}(\tau_{\mathrm{p}})P(\tau_{\mathrm{p}})U_{\mathrm{c}}(\tau_{\mathrm{p}}), \tag{5}$$

where

$$U_{\mathrm{b}}(t) = T\exp\left(-i\int_0^{t} H_{\mathrm{b}}(t')dt'\right), \tag{6a}$$

$$P(t) := T\exp\left(-i\vec{\sigma}\cdot\int_0^{t} \vec{v}(t')dt'\right). \tag{6b}$$

The unitary operator $U_{\mathrm{b}}(\tau_{\mathrm{p}})$ describes the time evolution of the bath and $P(\tau_{\mathrm{p}})$ the rotation of the spin due to the pulse. Note that the ansatz $U_{\mathrm{b}}(\tau_{\mathrm{p}})P(\tau_{\mathrm{p}})$ does not comprise any coupling between spin and bath. It is close to the goals of many previous studies aiming at robust pulses [17–23,25,26] and it corresponds to the ansatz used in previous studies separating the pulse from a classcial [25] or a quantum mechanical dynamics of the bath [16,24,27,30,31]. We emphasize that an ansatz which separates the pulse from the dynamics of the spin plus bath system can be shown not to succeed beyond leading order [28,29].

Since the spin-bath coupling is not included in $U_{\mathrm{b}}(\tau_{\mathrm{p}})P(\tau_{\mathrm{p}})$ we introduced the correction unitary operator $U_{\mathrm{c}}(\tau_{\mathrm{p}})$ in Eq. (5). We want to shape the pulse so that the correction term is as close to the identity as possible. A perfect decoupling would imply $U_{\mathrm{c}}(\tau_{\mathrm{p}}) = \mathbb{1}$. But this is unrealistic to achieve. Hence we pursue the perturbative approach to make as many terms of an expansion in τ_{p} as possible vanish. Then $U(\tau_{\mathrm{p}}) \approx U_{\mathrm{b}}(\tau_{\mathrm{p}})P(\tau_{\mathrm{p}})$ represents a valid approximation and one can neglect the spin-bath coupling during the pulse. We remark that pulses shaped in this way constitute robust single-qubit gates.

III. DERIVATION

The derivation of the perturbative conditions for the shaped pulses is very similar to the derivation given in Ref. [30]. Yet we present a brief outline here in order to keep the present article self-contained and because we extend the previous derivation to analytically time-dependent baths. We start from the pulse Hamiltonian in Eq. (3). We describe the time-dependent pulse operator as a global rotation about the axis $\hat{a}(t)$:

$$P(t) = \exp\left(-i\vec{\sigma}\cdot\hat{a}(t)\frac{\psi(t)}{2}\right), \tag{7}$$

where $|\hat{a}(t)| = 1$. The spin is turned by the angle $\psi(t)$ at the time t. Every unitary operator acting only on the Hilbert space of the spin can be written in the form of Eq. (7). In particular, a pulse that turns the spin by an angle χ satisfies

$$\psi(\tau_{\mathrm{p}}) = \chi. \tag{8}$$

We stress the difference between the current axis of rotation $\vec{v}(t)/|\vec{v}(t)|$ and the effective axis $\hat{a}(t)$ describing the total rotation of the spin from its position at time 0 to its current position at time t.

By definition the pulse operator fulfills the Schrödinger equation

$$i\partial_t P(t) = H_v(t)P(t), \tag{9}$$

which implies [29]

$$\begin{aligned} 2\vec{v}(t) = \psi'(t)\hat{a}(t) + \hat{a}'(t)\sin\psi(t) \\ -[1-\cos\psi(t)][\hat{a}'(t)\times\hat{a}(t)]. \end{aligned} \tag{10}$$

This differential equation is solved numerically for the frequency-modulated ansatz below. The time evolution of the whole system is given by

$$i\partial_t U(t) = [H_b(t) + \vec{\sigma}\cdot\vec{A}(t) + H_v(t)]U(t). \tag{11}$$

Inserting the ansatz (5) and solving for $\partial_t U_c(t)$ yields

$$i\partial_t U_c(t) = G(t)U_c(t) \tag{12}$$

$$G(t) = P^{-1}(t)U_b^{-1}(t)\,\vec{\sigma}\cdot\vec{A}(t)\,U_b(t)P(t). \tag{13}$$

Thus the unitary correction is determined by a Schrödinger equation with $G(t)$ as its time-dependent Hamiltonian. The formal solution of Eq. (12) in terms of the standard time ordering operator is

$$U_c(t) = T\left[\exp\left(-i\int_0^t G(\tau)d\tau\right)\right]. \tag{14}$$

Aiming at an expansion of $U_c(t)$ in powers of τ_p it is convenient to use the Magnus expansion [38] to express the time-ordered exponential

$$U_c(\tau_p) = \exp[-i\tau_p(G^{(1)} + G^{(2)} + \cdots)], \tag{15}$$

where $G^{(i)} = O(\tau_p^{i-1})$. The first two terms read

$$\tau_p G^{(1)} = \int_0^{\tau_p} dt\, G(t), \tag{16a}$$

$$\tau_p G^{(2)} = -\frac{i}{2}\int_0^{\tau_p} dt_1 \int_0^{t_1} dt_2 [G(t_1),G(t_2)]. \tag{16b}$$

Next, we need an expansion of $G(t)$ in powers of time. To this end, we consider the representation (7) which implies

$$\begin{aligned} P^{-1}(t)\vec{\sigma}\cdot\vec{A}(t)P(t) &= [\cos\psi\vec{A} - \sin\psi(\hat{a}\times\vec{A}) \\ &\quad + (1-\cos\psi)(\hat{a}\cdot\vec{A})\hat{a}]\cdot\vec{\sigma} && (17a) \\ &= \vec{n}_{A(t)}(t)\cdot\vec{\sigma} && (17b) \\ &= \sum_{i,j} n_{i,j}(t)A_j(t)\sigma_i, && (17c) \end{aligned}$$

where the time dependencies on the right-hand side of Eq. (17a) are omitted to lighten the notation. The vector operator $\vec{n}_{A(t)}(t)$ is the vector $\vec{A}(t)$ after a rotation about the axis $\hat{a}(t)$ by the angle $-\psi(t)$. The corresponding rotation matrix $D_{\hat{a}}(-\psi)$ is given by its matrix elements $n_{i,j}(t)$; for their explicit form see Appendix C. Due to the orthogonality of $D_{\hat{a}}(-\psi)$ the moduli of all its matrix elements are bounded by unity.

Note that there are two different kinds of time dependence in Eq. (17b). On the one hand, the time dependence of $\vec{A}(t)$ becomes weaker and weaker as the pulse duration τ_p is taken to zero because we assume that $\vec{A}(t)$ is analytical. This is exploited below. On the other hand, the time dependence of the $n_{i,j}(t)$ scales with τ_p, which means that $\tilde{n}_{i,j}(s) := n_{i,j}(s\tau_p)$ is completely independent of τ_p because the pulse has to be completed at $t = \tau_p$ whatever the pulse duration is.

We proceed by introducing the vector operator $\tilde{A}(t)$ and expanding it in powers of t:

$$\begin{aligned} \tilde{A}(t) &:= U_b^{-1}(t)A(t)U_b(t) && (18a) \\ &= \vec{A}_0 + it[H_{b,0},\vec{A}_0] + t\vec{A}_1 + O(t^2). && (18b) \end{aligned}$$

Here the main differences to the derivation in Ref. [30] arises. In Ref. [30], the term proportional to $\vec{A}_1$ did not appear because the bath was considered to be time-independent. Using the vector operator $\vec{n}_{A(t)}(t)$ from Eq. (17b) we rewrite $G(t)$ concisely as

$$\begin{aligned} G(t) &= P^{-1}(t)\vec{\sigma}\cdot\tilde{A}(t)P(t) && (19a) \\ &= \vec{n}_{\tilde{A}(t)}(t)\cdot\vec{\sigma}. && (19b) \end{aligned}$$

This form of $G(t)$ can be expanded in powers of t such that the neglected terms are of second order in τ_p for $t\in[0,\tau_p]$:

$$G(t) = \vec{n}_{A_0}\cdot\vec{\sigma} + t\left(i\left[H_{b,0}, i\vec{\sigma}\cdot\vec{n}_{A_0}\right] + \vec{n}_{A_1}\cdot\vec{\sigma}\right) + O\left(\tau_p^2\right). \tag{20}$$

Note that the time dependence stemming from the pulse rotation is not expanded because it does not change on $\tau_p \to 0$. Physically this means that one can expand in $\tau_p H(t)$, i.e., in $\lambda\tau_p$ and in $\omega_b\tau_p$, but *not* in $\tau_p H_v(t)$ because the magnitude of $H_v(t)$ is increased on $\tau_p \to 0$ to realize the desired pulse.

Inserting Eq. (20) in the terms of the Magnus expansion (16) eventually yields

$$U_c(\tau_p) = \exp[-i(\eta^{(1)} + \eta^{(2)} + \cdots)], \tag{21a}$$

$$\eta^{(1)} = \sum_{i,j}\sigma_i A_{j,0}\int_0^{\tau_p} n_{i,j}(t)dt, \tag{21b}$$

$$\eta^{(2)} = \sum_i \sigma_i\left(\eta_i^{(2a)} + \eta_i^{(2b)}\right) + \eta^{(2c)}, \tag{21c}$$

where $\eta^{(i)} \propto \tau_p^i$. Explicitly, one has

$$\eta_i^{(2a)} = \sum_j([H_{b,0},A_{j,0}] - iA_{j,1})\int_0^{\tau_p} t\, n_{i,j}(t)dt, \tag{22a}$$

$$\begin{aligned} \eta_i^{(2b)} = \sum_{l,m}[A_{l,0},A_{m,0}]_+ \int_0^{\tau_p} dt_1 \int_0^{t_1} dt_2 \\ \times \sum_{j,k}\epsilon_{ijk}n_{j,l}(t_1)n_{k,m}(t_2), \end{aligned} \tag{22b}$$

$$\begin{aligned} \eta^{(2c)} = \sum_{i,j<k}[A_{j,0},A_{k,0}]\int_0^{\tau_p} dt_1 \int_0^{t_1} dt_2 \\ \times[n_{i,j}(t_1)n_{i,k}(t_2) - n_{i,j}(t_2)n_{i,k}(t_1)]. \end{aligned} \tag{22c}$$

In these equations the anticommutator $[\cdot,\cdot]_+$ and the completely antisymmetric Levi-Civita tensor ϵ_{ijk} appear. The indices i, j, k, l, and m take one of the values x, y, or z. Most of the Eqs. (21b) and (22) are identical to those obtained in Ref. [30]. Only in $\eta_i^{(2a)}$ does the time dependence of the bath appear additionally. It is encoded in the operators $A_{j,1}$, which are zero for a time-independent bath.

The second-order equations (22) do not show a dependence on pure bath terms of an order higher than $H_{\mathrm{b},0}$. This means that the time dependence of the pure bath Hamiltonian is irrelevant up to second order. This is reasonable because even for a time-independent bath the actual bath dynamics induced from H_b appears only in second-order conditions. This does not apply to the vector operator $\vec{A}(t)$ which is already relevant in first-order pulses.

In the general case of a completely generic bath, all the expressions $\eta^{(\alpha)}$, $\alpha \in \{1,2a,2b,2c\}$, have to vanish in order to fulfill $U_\mathrm{c} = \mathbb{1} + O(\tau_p^3)$. The pulse shape determines the time evolution of the matrix elements $n_{i,j}$. Hence, in order to fulfill all the conditions the operator-independent integrals in Eqs. (21b) and (22) must disappear. The resulting 39 scalar equations are identical to those obtained in Ref. [30].

This is our first key result. It proves the applicability of the previously obtained pulses even in the presence of a nontrivial time dependence of the bath which may stem from special reference frames or from the interaction picture of fast modes. For specific cases, such as the pure dephasing model or if $[H_\mathrm{b}(t),\vec{A}(t)] = 0$ the number of scalar equations to be fulfilled for $U_\mathrm{c} = \mathbb{1} + O(\tau_\mathrm{p}^3)$ is reduced significantly. Pulses with less complexity can be used. This is studied in the sequel.

IV. FREQUENCY MODULATED ANSATZ

To solve Eq. (10) we choose an ansatz for $\vec{v}(t)$. In this paper we focus on a frequency-modulated pulse acting only in the $\sigma_x\sigma_y$ plane with a fixed amplitude $V_0 := |\vec{v}|$ and the current axis of rotation

$$\vec{v}(t) = \begin{pmatrix} V_0 \cos[\Phi(t)] \\ V_0 \sin[\Phi(t)] \\ 0 \end{pmatrix}, \tag{23}$$

where $\Phi(t)$ is a time-dependent phase. Note the difference to pulses with a time-dependent amplitude and a fixed axis discussed, for instance, in Refs. [16,27,30]. We focus here on frequency modulation in complement to previous work because there may be experimental setups where frequency modulation is much easier (or more accurately) implemented than amplitude modulation. Note that the ansatz (23) assumes that the control pulse can be switched on instantaneously. Transients are assumed to be sufficiently steep to be taken as jumps. The consideration of continuous amplitudes and frequency modulation is left to future research.

To point out the relation of the ansatz (23) to the experimental realization in the laboratory framework we consider a spin with a Larmor frequency ω_L in the NMR language [39]:

$$H_z = -\frac{\omega_\mathrm{L}}{2}\sigma_z. \tag{24}$$

Of course, this description is not restricted to nuclear spins. Any two-level system with an energy splitting can be considered. The control field is realized by applying a field perpendicular to the σ_z axis rotating with the Larmor frequency:

$$H_\mathrm{rf} = V_0\{\sigma_x \cos[\omega_\mathrm{L} t - \Phi(t)] - \sigma_y \sin[\omega_\mathrm{L} t - \Phi(t)]\}. \tag{25}$$

We include a time-dependent phase $\Phi(t)$ to shape the pulse. Its derivative $\partial_t\Phi(t)$ is the deviation of the frequency from the Larmor frequency. In this sense Eq. (25) describes a frequency-modulated pulse. Next, H_rf is transformed into the rotating framework in which H_z vanishes. Using the unitary time evolution induced by H_z

$$U_\mathrm{rot}(t) = \exp\left(i\frac{\omega_\mathrm{L}}{2}t\sigma_z\right), \tag{26}$$

we obtain $H_\mathrm{rot}(t) = U_\mathrm{rot}^\dagger H_\mathrm{rf} U_\mathrm{rot}$, which reads

$$H_\mathrm{rot}(t) = \begin{pmatrix} V_0\cos[\Phi(t)] \\ V_0\sin[\Phi(t)] \\ 0 \end{pmatrix} \cdot \begin{pmatrix} \sigma_x \\ \sigma_y \\ \sigma_z \end{pmatrix} = \vec{v}(t)\cdot\vec{\sigma}. \tag{27}$$

In order to find $\hat{a}(t)$ and $\psi(t)$ appearing in the parametrization in Eq. (7) of the pulse one has to solve the differential equation (10). Because $\hat{a}(t)$ is a unit vector, it is convenient to describe it by two angles $\varphi(t)$ and $\theta(t)$:

$$\hat{a}(t) = \begin{pmatrix} \sin[\theta(t)]\cos[\varphi(t)] \\ \sin[\theta(t)]\sin[\varphi(t)] \\ \cos[\theta(t)] \end{pmatrix}. \tag{28}$$

Solving Eq. (10) for the time derivatives of $\psi(t)$, $\varphi(t)$, and $\theta(t)$, we find

$$\partial_t\psi = 2V_0\sin\theta\,[\sin\Phi\sin\varphi + \cos\Phi\cos\varphi], \tag{29a}$$

$$\partial_t\varphi = V_0\frac{[\cos\frac{\psi}{2}\sin(\Phi-\varphi) - \sin\frac{\psi}{2}\cos\theta\cos(\Phi-\varphi)]}{\sin\frac{\psi}{2}\sin\theta}, \tag{29b}$$

$$\partial_t\theta = V_0\frac{[\cos\frac{\psi}{2}\cos\theta\cos(\Phi-\varphi) + \sin\frac{\psi}{2}\sin(\Phi-\varphi)]}{\sin\frac{\psi}{2}}. \tag{29c}$$

The seeming singularities for vanishing angles on the right-hand sides of Eqs. (29b) and (29c) have no *physical* reason, but they only result from the choice of spherical coordinates and from the chosen parametrization in Eq. (7). Note that the global axis of rotation $\hat{a}$ is not uniquely defined if ψ is a multiple of 2π.

At the very beginning at $t = 0$ the current axis of rotation $\vec{v}$ and the global one $\vec{a}$ coincide. The former lies by construction in the $\sigma_x\sigma_y$ plane. Hence we have the initial conditions

$$\lim_{t\to 0}\theta(t) = \frac{\pi}{2}, \tag{30a}$$

$$\lim_{t\to 0}\psi(t) = 0, \tag{30b}$$

$$\lim_{t\to 0}\varphi(t) = \Phi(0), \tag{30c}$$

where the latter two equations represent our deliberate choice. Inspecting the limit $t \to 0$ one additionally finds

$$2\partial_t\varphi|_{t=0} = \partial_t\Phi(t)|_{t=0}, \tag{31a}$$

$$\partial_t\theta\,|_{t=0} = 0. \tag{31b}$$

The derivative $\partial_t \psi$ follows trivially from Eq. (29a). In the next section, we provide solutions for this ansatz and a specific case of spin-bath coupling.

V. RESULTS

We are interested in the experimentally important case of a purely dephasing model, i.e., a bath coupled only via σ_z to the spin

$$\vec{A}(t) = A(t)\vec{e}_z. \tag{32}$$

Hence the coupling becomes simpler, but the bath dynamics itself is still kept in full generality. Spin flips do not occur in this model so that T_1 is infinite. But decoherence of the T_2 type is entirely kept. This assumption is justified in many experimental realizations. Moreover, the simplification of the coupling is advantageous for pulse shaping because it reduces the number of integral conditions derived from Eqs. (21b) and (22) in second order to be fulfilled from 39 to 3 first-order conditions and 6 second-order conditions which are given explicitly in Appendices A and B.

In the following, we present continuous pulses which fulfill the first-order conditions (first-order pulses) and pulses which fulfill all first- and second-order conditions (second-order pulses) for pure dephasing as in Eq. (32). Thereby, we provide optimized pulses that decouple the spin from the bath during the duration of the pulse up to $O(\tau_\mathrm{p}^3)$.

In order to consider a continuous frequency modulation we use the Fourier series ansatz

$$\Phi(t)=\sum_n b_{2n-1}\sin(2\pi nt/\tau_\mathrm{p})+b_{2n}[\cos(2\pi nt/\tau_\mathrm{p})-1] \tag{33}$$

for $\Phi(t)$. We consider π and $\pi/2$ pulses because of their frequent use in QIP and NMR. Therefore, the pulse has to fulfill

$$\psi(\tau_\mathrm{p}) = \pi \quad \text{or} \quad \pi/2 \tag{34}$$

for π pulses and $\pi/2$ pulses, respectively, according to Eq. (8). The value $\theta(\tau_\mathrm{p})$ is fixed by the fact that the final axis of rotation has to be perpendicular to σ_z to rotate the spin by the full angle $\psi(\tau_\mathrm{p})$. Thus we require

$$\theta(\tau_\mathrm{p}) = \frac{\pi}{2}. \tag{35}$$

For a given ansatz, Eq. (33), the numerical procedure to find solutions is straightforward. We solve the differential equations (29) using a fourth-order Runge-Kutta algorithm. For this solution the conditions (21b), (22), (8), and (35) are evaluated. We search for roots using the Powell hybrid method in the GNU scientific library [40].

Of two different pulses the one with the lower amplitude V_0 is preferable in experiment because less power is needed to realize it. For an experimentally realizable maximum amplitude this implies that the theoretical pulse with lower amplitude can be made shorter, which is definitely advantageous.

TABLE I. Overview of the pulses satisfying all first-order equations (21b). FM-1-PI denotes the frequency-modulated π pulse. FM-1-PI2 denotes the frequency-modulated $\pi/2$ pulse. The dimensionless coefficients b_n belong to the ansatz in Eq. (33). The amplitudes V_0 are given in units of $1/\tau_\mathrm{p}$. With all eight digits given the conditions are fulfilled for the π pulse within 10^{-10} and for the $\pi/2$ pulse within 10^{-9}. With only two digits they are fulfilled within 10^{-3} and 10^{-2}, respectively.

First-order pulses			
FM-1-PI		FM-1-PI2	
V_0	3.751 466 09	V_0	4.928 924 84
b_1	0.000 114 42	b_1	0.000 098 74
b_2	−1.093 471 12	b_2	−0.943 316 59
b_3	0.000 124 43	b_3	0.000 025 30
b_4	−0.594 525 72	b_4	−0.120 876 63

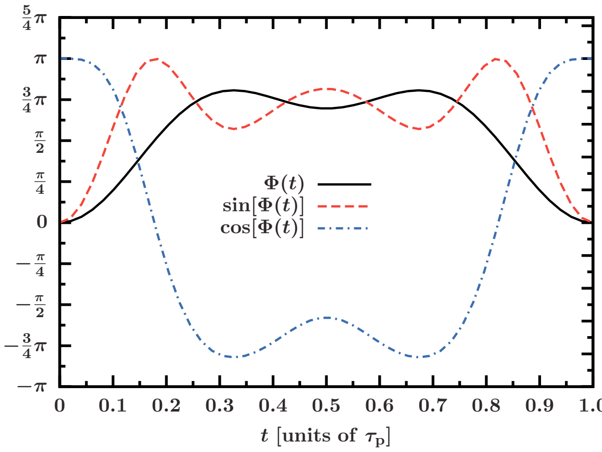

FIG. 1. (Color online) First-order π pulse FM-1-PI. We also plot $\sin\Phi(t) \propto v_y(t)$ and $\cos\Phi(t) \propto v_x(t)$ to illustrate the pulse shape in spin space. The left scale refers to $\Phi(t)$ and the right scale to $\sin\Phi(t)$ and $\cos\Phi(t)$, respectively. The Fourier coefficients for this pulse are given in Table I.

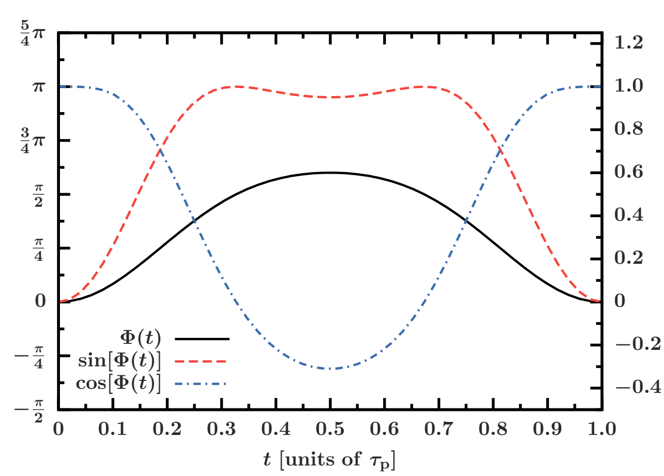

FIG. 2. (Color online) First-order $\pi/2$ pulse FM-1-PI2. We also plot $\sin\Phi(t) \propto v_y(t)$ and $\cos\Phi(t) \propto v_x(t)$ to illustrate the pulse shape in spin space. The left scale refers to $\Phi(t)$ and the right scale to $\sin\Phi(t)$ and $\cos\Phi(t)$, respectively. The Fourier coefficients for this pulse are given in Table I.

TABLE II. Overview of the pulses satisfying all first- and second-order equations, Eqs. (21b) and (III). FM-2-PI and FM-2-PI2 denote the frequency-modulated π pulse and $\pi/2$ pulse, respectively. The dimensionless coefficients b_n belong to the ansatz in Eq. (33). The amplitudes V_0 are given in units of $1/\tau_p$. With all eight digits given the conditions are fulfilled for the π pulse within 10^{-10} and for the $\pi/2$ pulse within 10^{-11}. With only two digits they are fulfilled within 10^{-2} and 10^{-2}, respectively.

Second-*f* order pulses			
FM-2-PI		FM-2-PI2	
V_0	12.834 329 79	V_0	12.256 193 90
b_1	0.114 751 39	b_1	1.730 718 40
b_2	0.172 485 87	b_2	0.735 299 59
b_3	0.482 625 21	b_3	0.232 425 23
b_4	−1.144 948 51	b_4	−0.248 293 10
b_5	−0.208 790 91	b_5	−0.071 022 04
b_6	0.253 780 13	b_6	−0.131 923 80
b_7	0.203 068 35	b_7	1.079 482 26
b_8	−0.167 480 22	b_8	0.122 200 06
b_9	−0.320 522 54	b_9	0.046 089 86
b_{10}	0.325 862 03	b_{10}	−0.153 656 17

Hence we search for pulses with lower amplitude among the second-order pulses. This is done by using an additional coefficient b_m in the ansatz (33) and minimizing the amplitude V_0 of the resulting solutions by varying this additional coefficient.

A. First-order pulses

For first-order pulses and the pure dephasing model, the set of conditions (21b) comprises only three equations given in Appendix A. Adding conditions (34) and (35) five parameters are necessary to construct first-order pulses. One parameter is the amplitude V_0 and the others are the coefficients b_n in ansatz (33). The characteristics of the pulses are reported in Table I. The pulses are plotted in Figs. 1 and 2. Note that the composite and continuous amplitude-modulated pulses found in Ref. [30] have comparable amplitudes for first-order pulses.

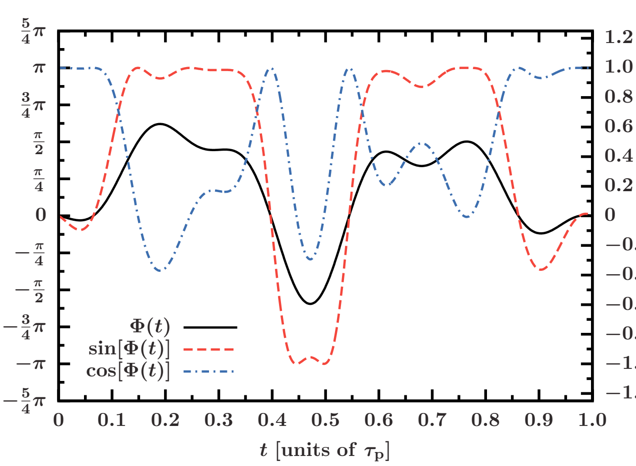

FIG. 3. (Color online) Second-order π pulse FM-2-PI. We also plot $\sin\Phi(t) \propto v_y(t)$ and $\cos\Phi(t) \propto v_x(t)$ to illustrate the pulse shape in spin space. The left scale refers to $\Phi(t)$ and the right scale to $\sin\Phi(t)$ and $\cos\Phi(t)$, respectively. The Fourier coefficients for this pulse are given in Table II.

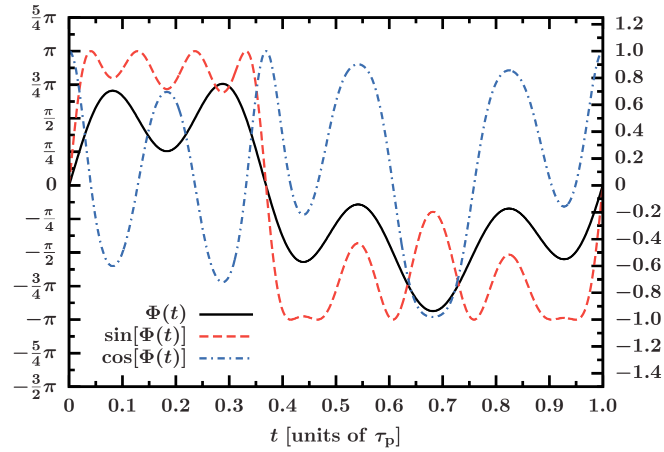

FIG. 4. (Color online) Second-order $\pi/2$-pulse FM-2-PI2. We also plot $\sin\Phi(t) \propto v_y(t)$ and $\cos\Phi(t) \propto v_x(t)$ to illustrate the pulse shape in spin space. The left scale refers to $\Phi(t)$ and the right scale to $\sin\Phi(t)$ and $\cos\Phi(t)$, respectively. The Fourier coefficients for this pulse are given in Table II.

B. Second-order pulses

Second-order pulses additionally have to fulfill conditions (22). These equations again simplify for a purely dephasing bath leading to six additional integral conditions besides the first-order terms see Appendix B. Note that more equations are to be fulfilled than for amplitude modulation [30] because the frequency-modulated pulses involve all three spin directions. The solutions for π and $\pi/2$ pulses are given in Table II and they are displayed in Figs. 3 and 4. Numerically, the double integrals in Eqs. B2 are particularly demanding. Full quantum mechanical studies of higher-order pulses will be hampered by even higher dimensional integrals occurring in the Magnus expansion [38]. An alternative route, which may be numerically more efficient, consists of the direct solution of the Schrödinger equation [24]. The mathematical existence of higher-order pulses is known [31].

Since we are interested in pulses with low amplitudes, we aim at minimizing the amplitude. To this end, we add another Fourier coefficient to the ansatz (33) and vary this additional parameter. In this way, we obtained the pulses FM-2-MIN-PI and FM-2-MIN-PI2 given in Table III and plotted in Figs. 5 and 6. Empirically it turned out to be more efficient to consider b_{14} instead of b_{11} as an additional coefficient. It is expected that even lower amplitudes can be achieved by using further coefficients. But our calculations with different coefficients, not shown here, indicate that this route would improve the amplitude only by 1%–2% at the expense of a more complex pulse shape.

By using only one free coefficient (b_{14}) we found $\pi/2$ pulses with amplitudes lower than $9.0/\tau_p$ to be compared with the amplitude-modulated pulses [30] with amplitude $11.5/\tau_p$. For π pulses we need $V_0 = 10.7/\tau_p$ in comparison

TABLE III. Overview of the pulses satisfying all first- and second-order equations, Eqs. (21b) and (22), with minimized amplitude. FM-2-PI and FM-2-PI2 denote the frequency-modulated π pulse and $\pi/2$ pulse with minimized amplitude, respectively. The dimensionless coefficients b_n belong to the ansatz in Eq. (33). The amplitudes V_0 are given in units of $1/\tau_p$. With all eight digits given the conditions are fulfilled for the π pulse within 10^{-10} and for the $\pi/2$ pulse within 10^{-9}. With only two digits they are fulfilled within 10^{-3} and 10^{-1}, respectively.

Minimized second-order pulses			
FM-2-MIN-PI		FM-2-MIN-PI2	
V_0	10.707 114 54	V_0	8.435 414 12
b_1	0.000 020 87	b_1	−1.820 415 07
b_2	1.387 689 38	b_2	−0.352 491 97
b_3	−0.000 199 22	b_3	0.030 548 74
b_4	−0.706 689 98	b_4	0.520 935 76
b_5	−0.000 015 88	b_5	−0.555 044 40
b_6	0.137 730 85	b_6	−0.388 155 68
b_7	0.000 087 70	b_7	0.451 673 61
b_8	0.688 943 31	b_8	−0.194 450 80
b_9	−0.000 114 08	b_9	−0.161 948 06
b_{10}	−0.697 440 86	b_{10}	−0.282 233 30
b_{14}	0.465 019 91	b_{14}	0.045 858 97

to $11.0/\tau_p$ for amplitude modulation. The amplitudes of the amplitude-modulated pulses refer to piecewise constant pulses; for continuous pulses they are even higher. Of course, the reduction of the amplitudes for frequency-modulated pulses is not spectacular. But it is remarkable that pulses with relatively low amplitudes can be found despite the larger number of conditions to be fulfilled: the frequency-modulated pulse fulfills $9 + 2 = 11$ equations including the conditions for the angles, and the amplitude-modulated pulse fulfills $5 + 1 = 6$ conditions including the condition for the rotation angle.

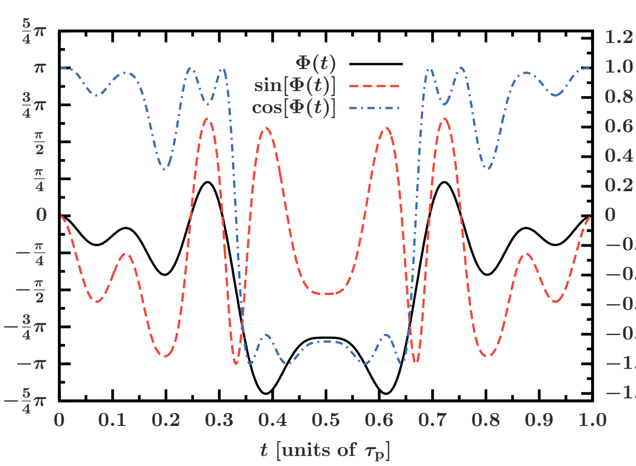

FIG. 5. (Color online) Minimized second-order π pulse FM-2-MIN-PI; $\sin\Phi(t) \propto v_y(t)$ and $\cos\Phi(t) \propto v_x(t)$ show the pulse shape in spin space. The left scale refers to $\Phi(t)$ and the right scale to $\sin\Phi(t)$ and $\cos\Phi(t)$, respectively. The Fourier coefficients for this pulse are given in Table III.

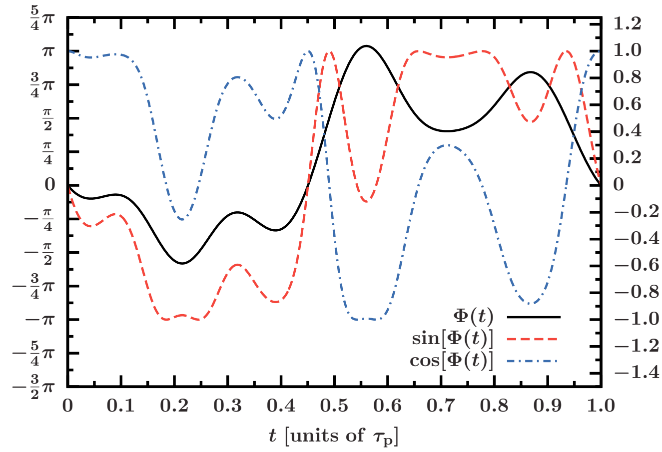

FIG. 6. (Color online) Minimized second-order $\pi/2$ pulse FM-2-MIN-PI2. We also plot $\sin\Phi(t) \propto v_y(t)$ and $\cos\Phi(t) \propto v_x(t)$ to illustrate the pulse shape in spin space. The left scale refers to $\Phi(t)$ and the right scale to $\sin\Phi(t)$ and $\cos\Phi(t)$, respectively. The Fourier coefficients for this pulse are given in Table III.

VI. CONCLUSIONS

In this paper we extended the existing perturbative approach to decouple a spin from a quantum mechanical bath by means of short control pulses in two ways.

First, we allowed for a time-dependent bath, which means both the bath Hamilton operator and the coupling operators may have an explicit, analytical time dependence. Yet, we found that this time dependence does not alter the requirements for the pulse shape which were derived previously for time-independent baths [30]. Hence, the pulses found previously are also applicable for time-dependent environments as they arise, for instance, in time-dependent reference frames or in the interaction picture of otherwise time-independent Hamiltonians. This is our first key result.

Second, we studied frequency-modulated pulses in first order and in second order in the pulse duration τ_p for quantum mechanical baths. Previously, only amplitude modulation was considered explicitly for quantum mechanical baths [16,27,30]. Frequency modulation was so far studied for static baths only [23]. We provide explicit solutions for continuous frequency-modulated pulses with amplitudes which have been minimized empirically. Such pulses are expected to be useful in experiments where no amplitude modulation can be realized or where the achievable accuracy for frequency modulation is superior to the accuracy of amplitude modulation. For instance, they can be used to implement realistic optimized dynamic decoupling [33,34] where the dynamic decoupling sequence is adapted to pulses of finite length. The frequency-modulated pulses constitute our second key result.

We emphasize that modulated pulses correspond to quantum gates which are robust against decoherence in the framework of quantum-information processing, e.g., the $\pi/2$ pulse about σ_y preceded by a π pulse about σ_z realizes the important Hadamard gate up to a global factor i.[1]

[1]This fact was stated in Refs. [28] and [29] in a too shortened way, leaving out the π pulse.

Further work should concentrate on higher-order terms not studied here. Such terms comprise higher-dimensional integrals so that the numerical effort increases considerably. Another promising route is to extend the model from pure dephasing to general decoherence. This would allow for systems with finite T_1 as well, at the expense of more complex pulses.

But at the present stage, it is also called for to verify the performance of the proposed pulses experimentally in order to assess how promising further extensions would be.

ACKNOWLEDGMENTS

We thank Christopher Stihl, Nils Drescher, Frederik Keim, and Leonid Pryadko for useful discussions and comments. The study of frequency modulation was triggered by a discussion with Michael Biercuk and Hermann Uys. We acknowledge financial support of the DFG under Project UH 90/5-1.

APPENDIX A: FIRST-ORDER CONDITIONS

For the first-order conditions we inspect Eq. (21b) to find the corresponding scalar equations. In a purely dephasing bath the sum over j collapses to $j = z$, resulting in the scalar equations

$$\eta_{11}= \int_0^{\tau_p} a_y(t)\sin[\psi(t)]-[1-\cos(\psi)]a_x(t)a_z(t)dt, \quad \text{(A1a)}$$

$$\eta_{12}= \int_0^{\tau_p} a_x(t)\sin[\psi(t)]+[1-\cos(\psi)]a_y(t)a_z(t)dt, \quad \text{(A1b)}$$

$$\eta_{13} = \int_0^{\tau_p} \cos[\psi(t)] + [1-\cos(\psi)]a_z(t)^2 dt. \quad \text{(A1c)}$$

The a_i are the components of the global axis of rotation parametrized in Eq. (28).

APPENDIX B: SECOND-ORDER CONDITIONS

For the second-order conditions, we additionally have to consider Eqs. (22) to find the corresponding scalar equations. Again certain sums collapse due to the purely dephasing bath model and we eventually obtain

$$\eta_{21} =\int_0^{\tau_p} t\{a_y(t)\sin[\psi(t)]-[1-\cos(\psi)]a_x(t)a_z(t)\}dt, \quad \text{(B1a)}$$

$$\eta_{22} =\int_0^{\tau_p} t\{a_x(t)\sin[\psi(t)]+[1-\cos(\psi)]a_y(t)a_z(t)\}dt, \quad \text{(B1b)}$$

$$\eta_{23} = \int_0^{\tau_p} t\{\cos[\psi(t)] + [1-\cos(\psi)]a_z(t)^2\}dt, \quad \text{(B1c)}$$

and

$$\eta_{24} = \int_0^{\tau_p} dt_1 \int_0^{t_1} dt_2[n_{yz}(t_1)n_{zz}(t_2)-n_{zz}(t_1)n_{yz}(t_2)], \quad \text{(B2a)}$$

$$\eta_{25} = \int_0^{\tau_p} dt_1 \int_0^{t_1} dt_2[n_{zz}(t_1)n_{xz}(t_2)-n_{xz}(t_1)n_{zz}(t_2)], \quad \text{(B2b)}$$

$$\eta_{26} = \int_0^{\tau_p} dt_1 \int_0^{t_1} dt_2[n_{xz}(t_1)n_{yz}(t_2)-n_{yz}(t_1)n_{xz}(t_2)]. \quad \text{(B2c)}$$

The matrix elements $n_{ij}(t)$ occurring here are those of the rotation matrix $D_{\hat{a}}(-\psi)$ given explicitly in Eq. (C1). The components a_i are parametrized in Eq. (28).

APPENDIX C: ROTATION MATRIX

To derive the matrix $D_{\hat{a}}(-\psi)$ we refer the reader to Ref. [30]. It is calculated by comparison of the coefficients in Eq. (17a). We obtain the matrix (C1) below, where the time dependencies of $\psi(t)$ and $\hat{a}(t)$ are omitted for clarity:

$$D_{\hat{a}}(-\psi) = \begin{pmatrix} \cos\psi + (1-\cos\psi)a_x^2 & a_z\sin\psi + (1-\cos\psi)a_xa_y & -a_y\sin\psi + (1-\cos\psi)a_xa_z \\ a_z\sin\psi + (1-\cos\psi)a_xa_y & \cos\psi + (1-\cos\psi)a_y^2 & a_x\sin\psi + (1-\cos\psi)a_ya_z \\ a_y\sin\psi + (1-\cos\psi)a_xa_z & -a_x\sin\psi + (1-\cos\psi)a_ya_z & \cos\psi + (1-\cos\psi)a_z^2 \end{pmatrix}. \quad \text{(C1)}$$

[1] L. M. K. Vandersypen, M. Steffen, G. Breyta, C. S. Yannoni, M. H. Sherwood, and I. L. Chuang, Nature (London) **414**, 883 (2001).
[2] E. L. Hahn, Phys. Rev. **80**, 580 (1950).
[3] H. Y. Carr and E. M. Purcell, Phys. Rev. **94**, 630 (1954).
[4] S. Meiboom and D. Gill, Rev. Sci. Instrum. **29**, 688 (1958).
[5] U. Haeberlen, *High Resolution NMR in Solids: Selective Averaging* (Academic Press, New York, 1976).
[6] L. Viola and S. Lloyd, Phys. Rev. A **58**, 2733 (1998).
[7] M. Ban, J. Mod. Opt. **45**, 2315 (1998).
[8] L. Viola, E. Knill, and S. Lloyd, Phys. Rev. Lett. **82**, 2417 (1999).
[9] K. Khodjasteh and D. A. Lidar, Phys. Rev. Lett. **95**, 180501 (2005).
[10] K. Khodjasteh and D. A. Lidar, Phys. Rev. A **75**, 062310 (2007).
[11] G. S. Uhrig, Phys. Rev. Lett. **98**, 100504 (2007); **106**, 129901(E) (2011).
[12] G. S. Uhrig, New J. Phys. **10**, 083024 (2008); **13**, 059504(E) (2011).
[13] W. Yang and R.-B. Liu, Phys. Rev. Lett. **101**, 180403 (2008)
[14] M. J. Biercuk, H. Uys, A. P. Van Devender, N. Shiga, W. M. Itano, and J. J. Bollinger, Nature (London) **458**, 996 (2009).
[15] J. Du, X. Rong, N. Zhao, Y. Wang, J. Yang, and R. B. Liu, Nature (London) **461**, 1265 (2009).
[16] L. P. Pryadko and P. Sengupta, Phys. Rev. A **78**, 032336 (2008).
[17] R. Tycko, Phys. Rev. Lett. **51**, 775 (1983).
[18] M. H. Levitt, Prog. Nucl. Magn. Reson. Spectrosc. **18**, 61 (1986).

[19] H. K. Cummins and J. A. Jones, New J. Phys. **2**, 6 (2000).
[20] H. K. Cummins, G. Llewellyn, and J. A. Jones, Phys. Rev. A **67**, 042308 (2003).
[21] T. E. Skinner, T. O. Reiss, B. Luy, N. Khaneja, and S. J. Glaser, J. Magn. Reson. **163**, 8 (2003).
[22] K. Kobzar, T. E. Skinner, N. Khanejac, S. J. Glaser, and B. Luy, J. Magn. Reson. **170**, 236 (2004).
[23] T. E. Skinner, K. Kobzar, B. Luy, M. R. Bendall, W. Bermel, N. Khaneja, and S. J. Glaser, J. Magn. Reson. **179**, 241 (2006).
[24] P. Sengupta and L. P. Pryadko, Phys. Rev. Lett. **95**, 037202 (2005).
[25] M. Möttönen, R. de Sousa, J. Zhang, and K. B. Whaley, Phys. Rev. A **73**, 022332 (2006).
[26] W. G. Alway and J. A. Jones, J. Magn. Reson. **189**, 114 (2007).
[27] L. P. Pryadko and G. Quiroz, Phys. Rev. A **77**, 012330 (2008).
[28] S. Pasini, T. Fischer, P. Karbach, and G. S. Uhrig, Phys. Rev. A **77**, 032315 (2008).
[29] S. Pasini and G. S. Uhrig, J. Phys. A: Math. Theor. **41**, 312005 (2008).
[30] S. Pasini, P. Karbach, C. Raas, and G. S. Uhrig, Phys. Rev. A **80**, 022328 (2009).
[31] K. Khodjasteh, D. A. Lidar, and L. Viola, Phys. Rev. Lett. **104**, 090501 (2010).
[32] M. H. Levitt, *Spin Dynamics: Basics of Nuclear Magnetic Resonance* (Wiley & Sons, Chichester, 2005).
[33] G. S. Uhrig and S. Pasini, New J. Phys. **12**, 045001 (2010).
[34] S. Pasini, P. Karbach, and G. S. Uhrig, Europhys. Lett. **96**, 10003 (2011).
[35] S. Pasini and G. S. Uhrig, J. Phys. A: Math. Theor. **43**, 132001 (2010).
[36] J. Schliemann, A. Khaetskii, and D. Loss, J. Phys.: Condens. Matter **15**, R1809 (2003).
[37] F. Jelezko and J. Wrachtrup, Phys. Status Solidi A **203**, 3207 (2006).
[38] S. Blanes, F. Casas, J. Oteo, and J. Ros, Phys. Rep. **470**, 151 (2009).
[39] L. M. K. Vandersypen and I. L. Chuang, Rev. Mod. Phys. **76**, 1037 (2005).
[40] M. Galassi, *GNU Scientific Library Reference Manual*, 3rd ed. (Network Theory, Bristol, UK, 2009).