Time-optimal synthesis of unitary transformations in a coupled fast and slow qubit system

Robert Zeier,[1,*] Haidong Yuan,[2,†] and Navin Khaneja[1,‡]
[1]*Harvard School of Engineering and Applied Sciences, 33 Oxford Street, Cambridge, Massachusetts 02138, USA*
[2]*Department of Mechanical Engineering, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, Massachusetts 02139, USA*
(Received 3 October 2007; published 20 March 2008)

In this paper, we study time-optimal control problems related to a system of two coupled qubits where the time scales involved in performing unitary transformations on each qubit are significantly different. In particular, we address the case where unitary transformations produced by evolutions of the coupling take a much longer time as compared to the time required to produce unitary transformations on the first qubit, but a much shorter time as compared to the time to produce unitary transformations on the second qubit. We present a canonical decomposition of SU(4) in terms of the subgroup $SU(2)\times SU(2)\times U(1)$, which is natural in understanding the time-optimal control problem of such a coupled qubit system with significantly different time scales. A typical setting involves dynamics of a coupled electron-nuclear spin system in pulsed electron paramagnetic resonance experiments at high fields. Using the proposed canonical decomposition, we give time-optimal control algorithms to synthesize various unitary transformations of interest in coherent spectroscopy and quantum-information processing.

DOI: 10.1103/PhysRevA.77.032332 PACS number(s): 03.67.Lx

I. INTRODUCTION

The synthesis of unitary transformations using time-efficient control algorithms (e.g., pulse sequences) is a well studied problem in quantum-information processing and coherent spectroscopy. Design of time-efficient pulse sequences is of practical importance in experimental realizations of quantum computing, as they can reduce decoherence effects. The study of efficient control algorithms is related to the complexity of quantum circuits in an essential way (see, e.g., [1–3]).

Significant literature in this subject treat the case where unitary transformations on single qubits take negligible time compared to transformations involving interactions between different qubits. This particular assumption is very realistic for nuclear spins in nuclear magnetic resonance (NMR) spectroscopy. Under this assumption, Ref. [4] (see also [5–18]) presents time-optimal control algorithms to synthesize arbitrary unitary transformations on a system of two qubits. Further progress in the case of multiple qubits is reported in [5,10,16,18–25].

In this work, we study a coupled qubit system where local unitary transformations on the first qubit take significantly less time than local transformations on the second one. In addition, we assume that the coupling evolution is much slower than transformations on the first qubit but much faster than transformations on the second one. We denote this system as a coupled fast and slow qubit system. We present a canonical decomposition of SU(4) in terms of the subgroup $SU(2)\times SU(2)\times U(1)$ reflecting the significantly different time scales immanent in the system. Employing this canonical decomposition, we derive time-optimal control algorithms to synthesize various unitary transformations. These time-optimal control algorithms are qualitatively very different from the ones obtained for coupled qubits with fast local operations on both of the qubits (as in Ref. [4]). The latter system has been studied in depth in the context of coupled spin-1/2 systems.

Our methods are applicable to coupled electron-nuclear spin systems occurring in pulsed electron paramagnetic resonance (EPR) experiments at high fields, where the Rabi frequency of the electron at typical microwave power is much larger than the hyperfine coupling, which is further much larger than the Rabi frequency of the nucleus at typical rf power. In the context of quantum computing similar electron-nuclear spin systems appear in Refs. [26–42]. In the case of two qubits, we provide time-optimal control algorithms for coupled electron-nuclear spin systems at high fields.

The main results of this paper are as follows. Let S_μ and I_ν represent spin operators for the fast (electron spin) and slow (nuclear spin) qubit, respectively. Any unitary transformation $G\in SU(4)$ on the coupled spin system can be decomposed as

$$G = K_1 \exp(t_1 S^\beta I_x + t_2 S^\alpha I_x) K_2, \qquad (1)$$

where $S^\alpha I_x$ and $S^\beta I_x$ correspond to x rotations of the slow qubit, conditioned, respectively, on the up or down state of the fast qubit. The elements K_1 and K_2 are rotations synthesized by rapid manipulations of the fast qubit in conjunction with the evolution of the natural Hamiltonian. The elements K_1 and K_2 belong to the subgroup $SU(2)\times SU(2)\times U(1)$, and correspond in an appropriately chosen basis to block-diagonal special unitary matrices with 2×2-dimensional blocks of unitary matrices.

The minimum time to produce any unitary transformation G is the smallest value of $(|t_1|+|t_2|)/\Omega^I$, where Ω^I is the maximum achievable Rabi frequency of the nucleus and $(t_1,t_2)^T$ is a pair satisfying Eq. (1). Synthesizing K_1 and K_2

*zeier@eecs.harvard.edu
†haidong@mit.edu
‡navin@hrl.harvard.edu

takes negligible time on the time scale governed by Ω^I.

The paper is organized as follows. In Sec. II, we recall the physical details of our model system exemplified by a coupled electron-nuclear spin system. The Lie-algebraic structure of our model is described in Sec. III, which is used to derive control algorithms (e.g., pulse sequences) for synthesizing arbitrary unitary transformations in our coupled spin system. In Sec. IV, we present examples. We prove the time optimality of our control algorithms in Sec. V, and some details of the proof are given in the Appendix.

Our work draws some results from the theory of Lie groups, which are explained as needed. We refer to [43,44] for general reference. To make the paper broadly accessible, we work with explicit matrix representations of Lie groups and Lie algebras.

II. PHYSICAL MODEL

As our model system, we consider two coupled qubits. We introduce the operators S_μ and I_ν, which correspond to operators on the first and second qubit, respectively. In particular, these operators are defined by $S_\mu=(\sigma_\mu\otimes \mathrm{id}_2)/2$ and $I_\nu=(\mathrm{id}_2\otimes\sigma_\nu)/2$ (see [45]), where

$$\sigma_x:=\begin{pmatrix}0&1\\1&0\end{pmatrix},\ \sigma_y:=\begin{pmatrix}0&-i\\i&0\end{pmatrix},\ \text{and}\ \sigma_z:=\begin{pmatrix}1&0\\0&-1\end{pmatrix},$$

and are the Pauli matrices and

$$\mathrm{id}_2:=\begin{pmatrix}1&0\\0&1\end{pmatrix}$$

is the 2×2-dimensional identity matrix. In the remaining text, let $\mu,\nu\in\{x,y,z\}$ and $\gamma\in\{x,y\}$.

In an experimental setting using an electron-nuclear spin system, the first qubit is represented by the electron spin-1/2. Similarly, the second qubit is represented by the nuclear spin-1/2. We assume that in the presence of a static magnetic field pointing in the z direction, the free evolution is governed in the laboratory frame by a Hamiltonian of the form

$$H_0^{\mathrm{lab}}=\omega_S S_z+\omega_I I_z+J(2S_zI_z), \tag{2}$$

where ω_S and ω_I represent the natural precession frequency of, respectively, the first qubit and second qubit and J is the coupling strength. We assume that

$$\omega_S\gg\omega_I\gg J. \tag{3}$$

This assumption is motivated by coupled electron-nuclear spin systems occurring in EPR experiments at high fields (see, e.g., Sec. 3.5 of [46]). The time scales in Eq. (3) ensure that the hyperfine coupling Hamiltonian between the spins averages to the Ising Hamiltonian $2S_zI_z$, as in Eq. (2). This is the so-called high field limit.

The first and second qubit are controlled by transverse oscillating fields, which result in the corresponding control Hamiltonian given by $H_S^{\mathrm{lab}}+H_I^{\mathrm{lab}}$, where

$$H_S^{\mathrm{lab}}=2\Omega^S(t)\cos[\omega^S t+\phi_S(t)]S_x$$

is the control Hamiltonian of the first qubit and

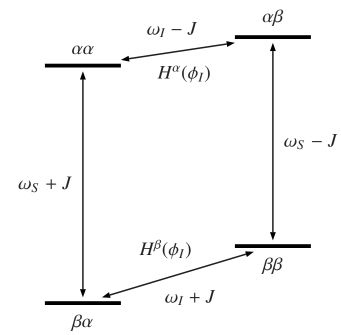

FIG. 1. The eigenstates of the Hamiltonian H_0^{lab} are shown. The α and β states of the spins denote their orientation along and opposite to the static magnetic field, respectively. The first and second index refer to the orientation of the electron and nuclear spin, respectively. The transitions $\alpha\alpha\leftrightarrow\alpha\beta$ and $\beta\alpha\leftrightarrow\beta\beta$ can be induced by $H^\alpha(\phi_I)$ and $H^\beta(\phi_I)$, respectively. Refer to the text for details.

$$H_I^{\mathrm{lab}}=2\Omega^I(t)\cos[\omega^I t+\phi_I(t)]I_x \tag{4}$$

is the control Hamiltonian of the second qubit. The amplitude, frequency, and phase of the control function with respect to the first qubit are represented by $\Omega^S(t)$, ω^S, and $\phi_S=\phi_S(t)$, respectively. Similarly, $\Omega^I(t)$, ω^I, and $\phi_I=\phi_I(t)$ represent the amplitude, frequency, and phase of the control function with respect to the second qubit. We use Ω^I and Ω^S to denote the maximal possible values of $\Omega^I(t)$ and $\Omega^S(t)$. In our model system, we assume that

$$\Omega^I\ll J\ll\Omega^S. \tag{5}$$

Therefore, we refer to the first qubit as the fast qubit and the second qubit as the slow qubit.

For our system there are two resonance frequencies for nuclear spin transitions. These are ω_I-J and ω_I+J, as shown in Fig. 1. We choose to irradiate on the nuclear spin with one of these frequencies. We subsequently show that this choice will lead to a time-optimal control algorithm. Thus, we choose $\omega^S=\omega_S$ and $\omega^I=\omega_I-J$. In a double rotating frame, rotating with the first and second qubit at frequency ω^S and ω^I, the transformations $U_{\mathrm{lab}}(t)$ and $U_{\mathrm{rot}}(t)$ describe, respectively, a unitary transformation in the laboratory frame and the double rotating frame related by

$$U_{\mathrm{lab}}(t)=\exp(-it\omega^S S_z)\exp(-it\omega^I I_z)U_{\mathrm{rot}}(t),$$

where $U_{\mathrm{lab}}(0)=U_{\mathrm{rot}}(0)$ is the identity transformation. Using the rotating wave approximation, the Hamiltonians H_0^{lab}, H_S^{lab}, and H_I^{lab} transform, respectively, to

$$H_0=JI_z+J(2S_zI_z), \tag{6}$$

$$H_S=\Omega^S(t)[S_x\cos\phi_S(t)+S_y\sin\phi_S(t)], \tag{7}$$

and

$$H_I=\Omega^I(t)[I_x\cos\phi_I(t)+I_y\sin\phi_I(t)].$$

In the absence of any irradiation on qubits, the system evolves under the free Hamiltonian $-iH_0$. From the time scales in Eq. (5), we can synthesize any unitary transformation of the form $\exp(-itS_\mu)$ in arbitrarily small time as compared to the evolution under H_0 or H_0+H_I. Using Eq. (7), we obtain the generators $-iS_x$ and $-iS_y$ by setting $\phi_S(t)=0$ and $\phi_S(t)=\pi/2$, respectively. Combining the evolution of these generators we can obtain the generator $-iS_z$.

Let us define the operators,

$$S^{\beta} = (\mathrm{id}_4/2 + S_z) = \begin{pmatrix} \mathrm{id}_2 & 0_2 \\ 0_2 & 0_2 \end{pmatrix}$$

and

$$S^{\alpha} = (\mathrm{id}_4/2 - S_z) = \begin{pmatrix} 0_2 & 0_2 \\ 0_2 & \mathrm{id}_2 \end{pmatrix},$$

where id_d is the $d \times d$-dimensional identity matrix and 0_2 is the 2×2-dimensional zero matrix. Note that $H_0 = 2JS^{\beta}I_z$, and we can rewrite H_0+H_I as

$$H_0 + H_I = 2JS^{\beta}I_z + \Omega^I(t)(S^{\alpha}+S^{\beta})(I_x \cos \phi_I + I_y \sin \phi_I).$$

Since $J \gg \Omega^I(t)$, and $S^{\beta}I_{\gamma}$ does not commute with $S^{\beta}I_z$, the above Hamiltonian gets in the first order approximation truncated to

$$H^{\alpha}(\phi_I) = 2JS^{\beta}I_z + \Omega^I(t)S^{\alpha}(I_x \cos \phi_I + I_y \sin \phi_I). \qquad (8)$$

Similarly, we can obtain the Hamiltonian

$$H^{\beta}(\phi_I) = 2JS^{\alpha}I_z + \Omega^I(t)S^{\beta}(I_x \cos \phi_I + I_y \sin \phi_I) \qquad (9)$$

by using $H^{\beta}(\phi_I)=\exp(i\pi S_x)H^{\alpha}(\phi_I)\exp(-i\pi S_x)$. In addition, it is possible to derive the Hamiltonian $H^{\beta}(\phi_I)$ on the same lines as we did for $H^{\alpha}(\phi_I)$ by choosing $\omega^S=\omega_S$ and $\omega^I=\omega_I+J$.

The Hamiltonians $H^{\alpha}(\phi_I)$ and $H^{\beta}(\phi_I)$, operate on the slow qubit and induce transitions $\alpha\alpha \leftrightarrow \alpha\beta$ and $\beta\alpha \leftrightarrow \beta\beta$ of the nuclear spin as shown in Fig. 1 (cf. Table 6.1.1 of [46]). The α and β states of the spins denote their orientation along and opposite to the static magnetic field, respectively. For the electron spin, the β state has lower energy than the α state as its gyromagnetic ratio is negative. Similarly, for the nuclear spin, the α state has lower energy than the β state as its gyromagnetic ratio is positive (as for a proton). We remark that the energy eigenstates $\beta\alpha$, $\beta\beta$, $\alpha\alpha$, and $\alpha\beta$ correspond, respectively, to the basis states 00, 01, 10, and 11. In Fig. 1, the first and second index in eigenstates refers to the orientation of the electron and nuclear spin, respectively. In this section, we have shown how to synthesize generators of the form $-iS_{\mu}$, $-iH^{\alpha}(\phi_I)$, and $-iH_0$.

For the main part of the paper we assume that we only irradiate on the transition $\alpha\alpha \leftrightarrow \alpha\beta$ [i.e., $-iH^{\alpha}(\phi_I)$]. In Sec. V B, we consider a more general model using irradiation on both transitions $\alpha\alpha \leftrightarrow \alpha\beta$ and $\beta\alpha \leftrightarrow \beta\beta$. In particular, we show (see Remark 4) that the minimum time to produce a unitary transformation cannot be reduced using the more general model. We defer the details to Sec. V B.

Remark 1. Motivated by the physical model [see, e.g., Eq. (5)], we neglect the time to produce operations on the fast qubit and the time of evolutions under the free Hamiltonian (see, e.g., Definition 5 of [4], for a formal definition of minimum time). Therefore, we define the time to produce a unitary transformation as the total time of evolution under the Hamiltonian $-iH^{\alpha}(\phi_I)$ [and $-iH^{\beta}(\phi_I)$].

III. LIE-ALGEBRAIC STRUCTURE OF THE MODEL SYSTEM

All transformations of our model system are contained in the Lie group $G=SU(4)$, which is the set of 4×4-dimensional unitary transformations of determinant one. The operators $-iI_{\mu}$, $-iS_{\nu}$, and $-i2I_{\mu}S_{\nu}$ are infinitesimal generators of the Lie group G, and they generate the 15-dimensional Lie algebra $\mathfrak{g}=\mathfrak{su}(4)$ given by the (real) vector space of 4×4-dimensional (traceless) skew Hermitian matrices. We have shown how to synthesize generators of the form $-iS_{\mu}$, $-iH^{\alpha}(\phi_I)$, and $-iH_0$. These generators are sufficient to produce any unitary transformation on the coupled qubit system, as described below.

Lemma 1. The Lie algebra generated by the elements $-iS_{\mu}$, $-iH^{\alpha}(\phi_I)$, and $-iH_0$, is equal to $\mathfrak{g}=\mathfrak{su}(4)$.

Therefore, a standard result (Theorem 7.1 of Ref. [47]) implies that the system is completely controllable, and any unitary transformation in $G=SU(4)$, can be synthesized by alternate evolution under the above Hamiltonians.

Lemma 2. The Lie algebra $\mathfrak{k}$, generated by the elements $-iS_{\mu}$ and $-iH_0$ consists of the elements $-iS_{\mu}$, $-i2S_{\nu}I_z$, and $-iI_z$.

The Lie algebra $\mathfrak{k}$ represents a class of generators that take significantly less time to be synthesized, as they only involve controlled rotations of the fast qubit and evolution of the free Hamiltonian $-iH_0$ (no controlled rotations of the slow qubit are involved). We can decompose

$$\mathfrak{g} = \mathfrak{k} \oplus \mathfrak{p}, \qquad (10)$$

where the subspace $\mathfrak{p}$ (of $\mathfrak{g}$) consists of the elements $-iI_{\gamma}$ and $-i2S_{\mu}I_{\gamma}$. The decomposition of Eq. (10) is a Cartan decomposition (see, e.g., [43], p. 213) as

$$[\mathfrak{k},\mathfrak{k}] \subset \mathfrak{k}, \quad [\mathfrak{k},\mathfrak{p}] \subset \mathfrak{p}, \quad \text{and} \quad [\mathfrak{p},\mathfrak{p}] \subset \mathfrak{k}, \qquad (11)$$

where $[g_1,g_2]=g_1g_2-g_2g_1$ is the commutator ($g_i \in \mathfrak{g}$).

Let $K=\exp(\mathfrak{k})$ denote the subgroup of $G=SU(4)$, which is infinitesimally generated by $\mathfrak{k}$. The elements of K can be synthesized only by the free evolution and employing controlled transformations on the fast qubit. Therefore, synthesizing transformations of K takes significantly less time as compared to general unitary transformations not contained in K. In particular, controlled transformations on the slow qubit are necessary to synthesize general unitary transformations. The Lie group $K=\exp(\mathfrak{k})$ is equal to $S[U(2)\times U(2)]$, which is sometimes referred to as $SU(2)\times SU(2)\times U(1)$.

Consider a maximal Abelian subalgebra $\mathfrak{a}$ contained in $\mathfrak{p}$. In our case, $\mathfrak{a}$ is spanned by the operators $-iS^{\beta}I_x$ and $-iS^{\alpha}I_x$. Any element $a\in\mathfrak{a}$ can be represented as $a_1(-iS^{\beta}I_x)+a_2(-iS^{\alpha}I_x)$, where $a_1,a_2\in\mathbb{R}$. As a matrix, a takes the form

$$-\frac{i}{2}\begin{pmatrix} 0 & a_1 & 0 & 0 \\ a_1 & 0 & 0 & 0 \\ 0 & 0 & 0 & a_2 \\ 0 & 0 & a_2 & 0 \end{pmatrix}.$$

We obtain the Lie group $A=\exp(\mathfrak{a})$ corresponding to the Abelian algebra $\mathfrak{a}$. From a Cartan decomposition of a real semisimple Lie algebra as satisfying Eqs. (10) and (11), we obtain a decomposition of the compact Lie group $\mathrm{G}=\mathrm{KAK}$ (see, e.g., [43], Chap. V, Theorem 6.7).

Lemma 3. Any element $G\in\mathrm{SU}(4)$ can be written as

$$G = K_1 \exp[t_1(-iS^{\beta}I_x) + t_2(-iS^{\alpha}I_x)]K_2, \tag{12}$$

where $t_1,t_2\in\mathbb{R}$ and $K_1,K_2\in\mathrm{K}$.

Remark 2. The computation of KAK decompositions was analyzed in Refs. [48–52]. In this work, we consider the Cartan decomposition, which corresponds to the type AIII in the classification of possible Cartan decompositions (see, e.g., pp. 451–452 of Ref. [43]).

Transforming all elements $G\in\mathrm{G}$ to $\mathrm{SWAP}\cdot G\cdot\mathrm{SWAP}$, where

$$\mathrm{SWAP} = \exp(-i\pi\mathbf{S}\cdot\mathbf{I}) = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix},$$

$\mathbf{S}=(S_x,S_y,S_z)^T$, and $\mathbf{I}=(I_x,I_y,I_z)^T$, the KAK decomposition is given in explicit matrices by

$$\begin{pmatrix} U_1 & 0_2 \\ 0_2 & U_2 \end{pmatrix} \exp\left[-\frac{i}{2} \begin{pmatrix} 0 & 0 & a_1 & 0 \\ 0 & 0 & 0 & a_2 \\ a_1 & 0 & 0 & 0 \\ 0 & a_2 & 0 & 0 \end{pmatrix} \right] \begin{pmatrix} U_3 & 0_2 \\ 0_2 & U_4 \end{pmatrix}$$
$$= \begin{pmatrix} U_1 & 0_2 \\ 0_2 & U_2 \end{pmatrix} \begin{pmatrix} c_1 & 0 & -is_1 & 0 \\ 0 & c_2 & 0 & -is_2 \\ -is_1 & 0 & c_1 & 0 \\ 0 & -is_2 & 0 & c_2 \end{pmatrix} \begin{pmatrix} U_3 & 0_2 \\ 0_2 & U_4 \end{pmatrix},$$

where $s_j=\sin(a_j/2)$ and $c_j=\cos(a_j/2)$. In particular, the Lie group K is given in this basis by block-diagonal unitary transformations, where 0_2 is the 2×2-dimensional zero matrix and U_1,U_2 (and U_3,U_4) are 2×2-dimensional unitary matrices such that the product of their determinants is one. The considered KAK decomposition is equivalent to the cosine-sine decomposition [53–55].

Remark 3. In Ref. [4], a different Cartan decomposition is considered. In that case, the subalgebra $\mathfrak{k}$ is given by the elements $-iS_\mu$ and $-iI_\nu$ and corresponds to unitary transformations on single qubits of a coupled two-qubit system. Synthesizing unitary transformations on single qubits is assumed in Ref. [4] to take significantly less time, as compared to unitary transformations which involve interactions between different qubits.

Since elements of K can be synthesized in negligible time, we obtain as the main result of this paper that the minimum time to synthesize any element $G\in\mathrm{SU}(4)$ is the minimum value of $(|t_1|+|t_2|)/\Omega^I$ such that $(t_1,t_2)^T$ is a pair satisfying Eq. (12). We defer the proof of this fact to Sec. V. Let us describe how to use the KAK decomposition of G, to synthesize an arbitrary transformation using only the generators $-iS_\mu$, $-iH^\alpha(\phi_I)$, and $-iH_0$.

The Lie algebra $\mathfrak{k}$ decomposes to $\mathfrak{k}_1\oplus\mathfrak{p}_1$, where $\mathfrak{k}_1$ is a subalgebra, composed of operators $-iS_\mu$ and $-i2S_\nu I_z$, and $\mathfrak{p}_1$ is generated by $-iI_z$, which commutes with all elements of $\mathfrak{k}_1$. The Lie algebra $\mathfrak{k}_1$ can be further subdivided by a Cartan decomposition $\mathfrak{k}_1=\mathfrak{k}_2\oplus\mathfrak{p}_2$. The subalgebra $\mathfrak{k}_2$ is generated by the operators $-iS_\mu$, and the subspace $\mathfrak{p}_2$ consists of the operators $-i2S_\mu I_z$. Therefore, similar as in Lemma 3, we obtain a decomposition of K.

Lemma 4. Each element $K_j\in\mathrm{K}$ can be decomposed as

$$\begin{aligned} K_j &= \exp(-i\tau_{2j-1}I_z)L_{2j-1}\exp(-i\tau_{2j}2S_zI_z)L_{2j} \\ &= \exp[-i(\tau_{2j-1}-\tau_{2j})I_z]L_{2j-1}\exp(-i\tau_{2j}H_0/J)L_{2j}, \end{aligned} \tag{13}$$

where $\tau_j\in\mathbb{R}$ and $L_j\in\mathrm{K}_2=\exp(\mathfrak{k}_2)$.

Using an Euler angle decomposition (see, e.g., pp. 454–455 of Ref. [56]), the elements $L_j\in\mathrm{K}_2$ are given as

$$\begin{aligned} L_j &= \exp(-i\theta_{j,1}S_z)\exp(-i\theta_{j,2}S_x)\exp(-i\theta_{j,3}S_z) \\ &= \exp[-i(\theta_{j,1}+\theta_{j,3})S_z]\exp[-i\theta_{j,2}R(\theta_{j,3})], \end{aligned} \tag{14}$$

where $R(\theta_{j,3})=S_x\cos\theta_{j,3}-S_y\sin\theta_{j,3}$.

Similarly, any element A of the subgroup A can be written as

$$\begin{aligned} A &= \exp[t_1(-iS^\beta I_x)+t_2(-iS^\alpha I_x)] \\ &= \exp\left[-i\frac{t_1}{\Omega^I}H^\beta(0)\right]e^{it_3I_z}e^{-it_4H_0/J} \\ &\quad\times\exp\left[-i\frac{t_2}{\Omega^I}H^\alpha(0)\right] \\ &= e^{it_3I_z}e^{-it_1H^\beta(t_3)/\Omega^I}e^{-it_4H_0/J}e^{-it_2H^\alpha(0)/\Omega^I}, \end{aligned} \tag{15}$$

for $t_3=2Jt_1/\Omega^I \bmod 4\pi$ and $t_4=J(t_1-t_2)/\Omega^I \bmod 2\pi\geq 0$. This follows by substituting for expressions of H_0, $H^\alpha(\phi_I)$, and $H^\beta(\phi_I)$ [see Eqs. (6)–(9)]. Combining Eqs. (13)–(15), a complete decomposition of an element $G\in\mathrm{SU}(4)$ can be written as

$$\begin{aligned} K_1AK_2 &= e^{-iv_0S_z}e^{-iwI_z}R_1e^{-i\tau_2H_0/J}R_2\exp\left[-i\frac{t_1}{\Omega^I}H^\beta(t_3+\tau)\right] \\ &\quad\times e^{-it_4H_0/J}\exp\left[-i\frac{t_2}{\Omega^I}H^\alpha(\tau)\right]R_3e^{-i\tau_4H_0/J}R_4, \end{aligned}$$

where all the transformations R_j operate on the fast qubit. In particular, we have $R_4=\exp[-i\theta_{4,2}R(\theta_{4,3})]$, $R_3=\exp[-i\theta_{3,2}R(v_3)]$, $R_2=\exp[-i\theta_{2,2}R(v_2)]$, $R_1=\exp[-i\theta_{1,2}R(v_1)]$, $v_3=\theta_{3,3}+\theta_{4,1}+\theta_{4,3}$, $v_2=\theta_{2,3}+\theta_{3,1}+v_3$, $v_1=\theta_{1,3}+\theta_{2,1}+v_2$, $v_0=\theta_{1,1}+v_1$, $\tau=\tau_4-\tau_3$, and $w=\tau_1-\tau_2+\tau_3-\tau_4-t_3$. The time to produce G is essentially $(t_1+t_2)/\Omega^I$. Note that

$$\exp(-iwI_z) = e^{-i\pi S_x}\exp[-iwH_0/(2J)]e^{i\pi S_x}\exp[-iwH_0/(2J)].$$

Transformations on the fast qubit such as $\exp(-iv_0S_z)$ are significantly faster. Figure 2 shows the canonical pulse sequence realizing any unitary transformation as a sequence of rotations under $-iH_0$, $-iH^\alpha(\phi_I)$, and $-iS_\mu$. The corresponding (minimum) time is $(t_1+t_2)/\Omega^I$.

IV. EXAMPLES

We introduce the unitary transformations CNOT[1,2], CNOT[2,1], and SWAP, which are given as follows (CNOT de-

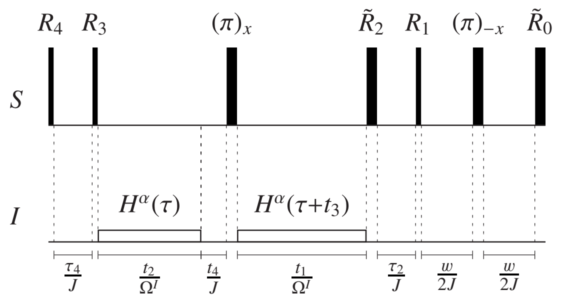

FIG. 2. The figure shows a canonical pulse sequence for synthesizing unitary transformations in the coupled qubit system. Let $\tilde{R}_2 = R_2 \exp(i\pi S_x)$ and $\tilde{R}_0 = \exp(-iv_0 S_z)\exp(-i\pi S_x)$. Since $1/J \ll 1/\Omega^I$, the length of the time intervals t_j/Ω^I is larger as depicted. Refer to the text for details.

notes "controlled-NOT"):

$$\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{pmatrix}, \quad \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \end{pmatrix}, \quad \text{and} \quad \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}.$$

Let $c \in \{1,3,-1,-3\}$. The elements of SU(4) corresponding to the transformation CNOT[2,1] are given by $\exp[c\pi \times (-i2S_xI_z + iS_x + iI_z)/2]$, which is equal to the transformation $\exp(ic\pi/4)\text{CNOT}[2,1]$. For CNOT[1,2] and SWAP we obtain the elements $\exp[c\pi(-i2S_zI_x + iS_z + iI_x)/2]$ and $\exp[c\pi(i2S_xI_x + i2S_yI_y + i2S_zI_z)/2]$, which are equal to $\exp(ic\pi/4)\text{CNOT}[1,2]$ and $\exp(ic\pi/4)\text{SWAP}$, respectively. These different instances of unitary transformations result from the irrelevance of the global phase in quantum mechanics and can be described mathematically by multiplying with elements of the (finite) center of G. The center consists of those elements which commute with all elements of G. To find the time-optimal control algorithm, we may have to consider multiplying with different elements of the center.

As $\exp(i\pi/4)\text{CNOT}[2,1]$ is an element of K, it takes negligible time to synthesize CNOT[2,1]. In strong contrast, $\exp(i\pi/4)\text{CNOT}[1,2]$ is not contained in K. Using the KAK decomposition, both elements $\exp(i\pi/4)\text{CNOT}[1,2]$ and $\exp(i\pi/4)\text{SWAP}$ correspond to the same generator of A, given by $\pi(-iS^\beta I_x) + 0(-iS^\alpha I_x)$, and the minimum time to synthesize each of them is equal to $t_{\min} = \pi$. This is still the optimal time if we consider multiplying with different elements of the center.

We explicitly state the control algorithms: The unitary transformation $\exp(i\pi/4)\text{CNOT}[1,2]$ is given by

$$\begin{aligned} &\exp(i\pi S_z/2)\exp(i\pi I_z)\exp(-i\pi S^\alpha I_x)\exp(-i\pi I_z) \\ &\quad = \exp(i\pi S_z/2)\exp(-it' H_0/J)\exp[-i\pi H^\alpha(\pi)/\Omega^I], \end{aligned}$$

where $t' = -\pi J/\Omega^I \bmod 2\pi \geq 0$. Similarly, the unitary transformation $\exp(i\pi/4)\text{SWAP}$ is given by

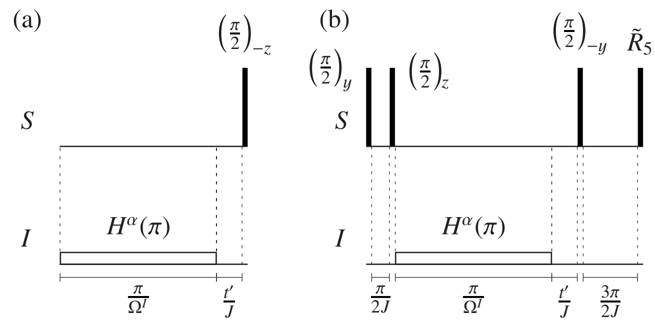

FIG. 3. The figure shows the pulse sequences for synthesizing the unitary transformations (a) $\exp(i\pi/4)\text{CNOT}[1,2]$ and (b) $\exp(i\pi/4)\text{SWAP}$, where $\tilde{R}_5 = \exp(i\pi S_z/2)\exp(-i\pi S_x/2)$. Since $1/J \ll 1/\Omega^I$, the length of the time intervals π/Ω^I is larger as depicted. Refer to the text for details.

$$\begin{aligned} &e^{i\pi/4}\text{CNOT}[2,1]e^{i\pi/4}\text{CNOT}[1,2]e^{-i\pi/4}\text{CNOT}[2,1] \\ &\quad = e^{i\pi S_z/2}e^{-i\pi S_x/2}e^{-i3\pi H_0/(2J)}e^{i\pi S_y/2}e^{-it' H_0/J} \\ &\qquad \times \exp[-i\pi H^\alpha(\pi)/\Omega^I]e^{-i\pi S_x/2}e^{-i\pi H_0/(2J)}e^{-i\pi S_y/2}. \end{aligned}$$

The corresponding pulse sequences are given in Fig. 3.

V. PROOF OF TIME OPTIMALITY

In this section, we prove the time optimality of the given control algorithms in order to synthesize unitary transformations in the coupled fast and slow qubit system. As expected, the maximal amplitude Ω^I [see Eq. (5)] determines the optimal time.

A. Simple case

All control algorithms, synthesizing a unitary transformation in time $t = \Sigma_j t_j$, can be written in the form

$$K'_{n+1} \exp[-it'_n H^\beta(\psi_n)]K'_n \cdots K'_2 \exp[-it'_1 H^\beta(\psi_1)]K'_1, \tag{16}$$

where $K'_j \in \mathrm{K}$ take negligible time to be synthesized as compared to the evolution under H^β, $t'_j = t_j/\Omega^I$, and $t_j, \psi_j \in \mathbb{R}$. We can rewrite Eq. (16) as

$$K_{n+1} \exp[-it_n S^\beta I_x]K_n \cdots K_2 \exp[-it_1 S^\beta I_x]K_1, \tag{17}$$

where $K_j \in \mathrm{K}$. Equation (17) can be rewritten as

$$\tilde{K}_{n+1} \exp(\tilde{p}_n) \cdots \exp(\tilde{p}_1), \tag{18}$$

where $\tilde{p}_j = \tilde{K}_j(-it_j S^\beta I_x)\tilde{K}_j^{-1}$ and $\tilde{K}_j$ are suitable elements of K. Observe that the elements $\tilde{p}_j$ are contained in $\mathfrak{p}$. This follows from the Campbell-Baker-Hausdorff formula (see, e.g., Appendix B.4 of Ref. [44]) and the fact that $[\mathfrak{k},\mathfrak{p}] \in \mathfrak{p}$ [see Eq. (11)]. It was shown in Ref. [4] that for all time-optimal control algorithms the elements $\tilde{K}_j$ can be chosen such that all $\tilde{p}_j$ commute. Therefore, all $\tilde{p}_j$ belong to a maximal Abelian subalgebra inside $\mathfrak{p}$, and we can find one $K_0 \in \mathrm{K}$ such that $K_0 \tilde{p}_j K_0^{-1} \in \mathfrak{a}$ for all j. Using this result and referring to Appendix A 1, we can rewrite Eq. (18) in the form

$$\bar{K}_2 \exp(t_n p_n) \cdots \exp(t_1 p_1) \bar{K}_1, \tag{19}$$

where $p_j = \beta_j(-iS^\beta I_x) + \alpha_j(-iS^\alpha I_x)$,

$$(\beta_j, \alpha_j)^T \in \{(-1,0)^T, (1,0)^T, (0,-1)^T, (0,1)^T\},$$

and $\bar{K}_1, \bar{K}_2 \in \mathrm{K}$. Equation (19) can be simplified to

$$\bar{K}_2 \exp[\bar{\beta}(-iS^\beta I_x) + \bar{\alpha}(-iS^\alpha I_x)]\bar{K}_1, \tag{20}$$

where $\bar{\alpha} = \Sigma_j \alpha_j t_j$ and $\bar{\beta} = \Sigma_j \beta_j t_j$. Assume that the unitary transformation to be synthesized is given by one of its KAK decompositions $\bar{K}_4 \exp[a_1(-iS^\beta I_x) + a_2(-iS^\alpha I_x)]\bar{K}_3$, where $a_j \in \mathbb{R}$ and $\bar{K}_3, \bar{K}_4 \in \mathrm{K}$. We remark that the KAK decomposition is not unique, and we prove in Appendix A 3 that all possible KAK decompositions $\bar{K}_6 \exp[a_1'(-iS^\beta I_x) + a_2'(-iS^\alpha I_x)]\bar{K}_5$ correspond to all values $a_j' = a_j + 2\pi z_j$, where $z_j \in \mathbb{Z}$ and $\bar{K}_5, \bar{K}_6 \in \mathrm{K}$. We can choose $\bar{a}_1$ and $\bar{a}_2$ as those values of a_1' and a_2' such that $|\bar{a}_1| + |\bar{a}_2|$ is minimum. If $|\bar{a}_1| + |\bar{a}_2| > t$, we cannot synthesize the unitary transformation in time t since all time-optimal control algorithms are equal to Eq. (20) and $|\bar{\alpha}| + |\bar{\beta}| = |\Sigma_j \alpha_j t_j| + |\Sigma_j \beta_j t_j| \le \Sigma_j(|\alpha_j| + |\beta_j|)t_j = \Sigma_j t_j = t$. For $|\bar{a}_1| + |\bar{a}_2| \le t$, we can use the control algorithm $\exp(-i\bar{a}_1 S^\beta I_x)\exp(-i\pi S_x)\exp(-i\bar{a}_2 S^\beta I_x)\exp(i\pi S_x)$ to synthesize the unitary transformation in time $|\bar{a}_1| + |\bar{a}_2|$.

B. General case

Until now, we have assumed that in Eq. (4), $\omega^I = \omega_I - J$, i.e., we irradiate on the transition $\alpha\alpha \leftrightarrow \alpha\beta$. More generally, under arbitrary irradiation on the nuclear spin, the resulting Hamiltonian in Eq. (4) can be written as

$$H_I^{\mathrm{lab}}(t') = 2\Omega^I(t')\{b_2 \cos[(\omega_I - J)t' + \phi_2(t')] + b_1 \cos[(\omega_I + J)t' + \phi_1(t')]\}I_x,$$

where $|b_1| + |b_2| \le 1$ (this ensures that the peak amplitude is $2\Omega^I$). The transition $\beta\alpha \leftrightarrow \beta\beta$ corresponds to the frequency $\omega_I + J$. We transform into a double rotating frame by

$$U_{\mathrm{lab}}(t') = \exp(-it'\omega_S S_z)\exp[-it'(\omega_I I_z + J2I_z S_z)]U_{\mathrm{rot}}(t'),$$

where $U_{\mathrm{lab}}(0) = U_{\mathrm{rot}}(0)$ is the identity transformation. Thus, the evolution under the control Hamiltonian for time t' (with constant $\Omega^I, \phi_1, \phi_2 \in \mathbb{R}$) generates a net rotation $K_1'' \exp[-it'\Omega^I(b_1 S^\beta I_p + b_2 S^\alpha I_q)]$, where $I_p = I_x \cos(\phi_1) + I_y \sin(\phi_1)$, $I_q = I_x \cos(\phi_2) + I_y \sin(\phi_2)$, and $K_1'' \in \mathrm{K}$. This can be rewritten as $K_1' \exp(-itb)K_2'$, where $b = b_1(-iS^\beta I_x) + b_2(-iS^\alpha I_x) \in \mathfrak{a}$, $t = t'\Omega^I$, and $K_1', K_2' \in \mathrm{K}$. Therefore, any control algorithm generates in time t, a transformation [written as in Eq. (17)]

$$K_{n+1} \exp[-it_n b]K_n \cdots K_2 \exp[-it_1 b]K_1,$$

where t_j is given in units of $1/\Omega^I$ and $\Sigma_j t_j = t$. This generalizes the case of $b_1 = 1$ and $b_2 = 0$, treated in Sec. V A.

Similarly as in Sec. V A, we obtain time-optimal control algorithms as in Eq. (18), where $\tilde{p}_j = \tilde{K}_j b \tilde{K}_j^{-1}$ and $\tilde{K}_j$ are suitable elements of K. Therefore, Eq. (18) can be transformed to Eq. (19), where the commuting elements $p_j = \beta_j(-iS^\beta I_x) + \alpha_j(-iS^\alpha I_x)$ are contained in the Weyl orbit $\mathcal{W}(b) = \{KbK^{-1} : K \in \mathrm{K}\} \cap \mathfrak{a}$, i.e., $(\beta_j, \alpha_j)^T$ is an element of the set (see Appendix A 1)

$$\{(b_1,b_2)^T, (b_1,-b_2)^T, (-b_1,b_2)^T, (-b_1,-b_2)^T, (b_2,b_1)^T, (-b_2,b_1)^T, (b_2,-b_1)^T, (-b_2,-b_1)^T\}. \tag{21}$$

As before, Eq. (19) can be simplified to Eq. (20), and we obtain $|\bar{\alpha}| + |\bar{\beta}| \le t(|b_1| + |b_2|)$. Furthermore, $\max\{|\bar{\alpha}|, |\bar{\beta}|\} \le t \max\{|b_1|, |b_2|\}$ holds. When the pairs $(a_1, a_2)^T$ and $(b_1, b_2)^T$ satisfy $\max\{|a_1|, |a_2|\} \le \max\{|b_1|, |b_2|\}$ and $|a_1| + |a_2| \le |b_1| + |b_2|$, then we say $(a_1, a_2)^T$ is r majorized by $(b_1, b_2)^T$, i.e., $(a_1, a_2)^T \prec_r (b_1, b_2)^T$. The notion of r majorization is equivalent to the condition that one element of $\mathfrak{a}$ is contained in the convex closure of the Weyl orbit of another one (for a proof see Appendix A 2).

Given any unitary transformation $G \in \mathrm{G}$, let t_{opt} be the smallest possible time such that

$$(a_1, a_2)^T \prec_r t_{\mathrm{opt}}(b_1, b_2)^T, \tag{22}$$

and $G = \bar{K}_2 \exp[a_1(-iS^\beta I_x) + a_2(-iS^\alpha I_x)]\bar{K}_1$ for some $\bar{K}_j \in \mathrm{K}$. Again, the KAK decomposition is not unique, and different KAK decompositions correspond to all values $a_j' = a_j + 2\pi z_j$, where $z_j \in \mathbb{Z}$ (see Appendix A 3). Let us choose a_j as an element of $[-\pi, \pi]$. We prove in Appendix A 4 that for such a choice of a_j, the equation $(a_1, a_2)^T \prec_r (a_1, a_2)^T + 2\pi(z_1, z_2)^T$ holds for all $z_1, z_2 \in \mathbb{Z}$. This implies that the smallest t_{opt} in Eq. (22) can be achieved for $a_1, a_2 \in [-\pi, \pi]$.

Then G cannot be synthesized in time t less than t_{opt}, as for such a control algorithm the equation $(\bar{\alpha}, \bar{\beta})^T \prec_r t(b_1, b_2)^T$ would hold, and this would contradict the minimality of t_{opt}. In addition, G can be synthesized in time t greater than or equal to t_{opt}: It follows from $(a_1, a_2)^T \prec_r t_{\mathrm{opt}}(b_1, b_2)^T$ that $(a_1, a_2)^T$ is contained in the convex closure of the Weyl orbit of $t_{\mathrm{opt}}(b_1, b_2)^T$ (see Appendix A 2) and we can synthesize G by convex combinations of elements of the Weyl orbit of $t_{\mathrm{opt}}(b_1, b_2)^T$.

Remark 4. Note, since $(b_1, b_2) \prec_r (1, 0)$, it follows that the minimum time to produce any unitary transformation can be obtained when all rf amplitude is used to irradiate only on one nuclear transition (say $\alpha\alpha \leftrightarrow \alpha\beta$) as described earlier (see Fig. 1), i.e., we do not use the second transition (say $\beta\alpha \leftrightarrow \beta\beta$). This justifies our initial choice of irradiating only on one nuclear transition.

VI. CONCLUSION

In this paper, we presented time-optimal control algorithms to synthesize arbitrary unitary transformations for the coupled fast and slow qubit system. These control algorithms are applicable to electron-nuclear spin systems in pulsed EPR experiments at high fields. Explicit examples were given for CNOT and SWAP operations. Our results can be considered as a first step to design time-optimal control algorithms for various systems in quantum-information processing which cannot be characterized by control systems with fast local controls. In doing so, we have to use Lie-group decompositions which reflect the inherent time scales in the given system.

Recently, controllability results have appeared for coupled electron-nuclear spin systems at low fields [57,58], where it is shown that it is possible to synthesize any unitary transformation on the electron-spin system by only manipulating the electron. New methods need to be developed to obtain time-optimal control algorithms in these settings.

ACKNOWLEDGMENTS

This work was supported by ONR Grant No. 38A-1077404, AFOSR Grant No. FA9550-05-1-0443, and NSF Grant No. 0133673.

APPENDIX A: PROOFS

1. Derivation of the Weyl orbit

Assume that $a=a_1(-iS^{\beta}I_x)+a_2(-iS^{\alpha}I_x)$ and $b=b_1(-iS^{\beta}I_x)+b_2(-iS^{\alpha}I_x)$ are elements of $\mathfrak{a}$. We compute the Weyl orbit $\mathcal{W}(a)=\{KaK^{-1}:K\in\mathrm{K}\}\cap\mathfrak{a}$ of a. We prove that b is contained in the Weyl orbit of a iff $(b_1,b_2)^T$ is an element of

$$\{(a_1,a_2)^T,(a_1,-a_2)^T,(-a_1,a_2)^T,(-a_1,-a_2)^T,(a_2,a_1)^T,\\ (-a_2,a_1)^T,(a_2,-a_1)^T,(-a_2,-a_1)^T\}. \tag{A1}$$

Let $K\in\mathrm{K}$ and consider a and b as matrices. It follows from $b=KaK^{-1}$ and $bb=KaK^{-1}KaK^{-1}$ that the equations $\|a\|_F=\|b\|_F$ and $\|a^2\|_F=\|b^2\|_F$ hold, where $\|q\|_F=\mathrm{Tr}(q^{\dagger}q)$. Thus, we get the equations $a_1^2+a_2^2=b_1^2+b_2^2$ and $a_1^4+a_2^4=b_1^4+b_2^4$. The corresponding solutions are given by all eight cases of Eq. (A1), and we obtain that $\mathcal{W}(a)$ is a subset of Eq. (A1). The proof follows since all these solutions can be derived by the map $(K,a)\mapsto KaK^{-1}$, where the elements $K\in\mathrm{K}$ are given by

$$\{\mathrm{id}_4,\exp(-i\pi S^{\alpha}I_z),\exp(-i\pi S^{\beta}I_z),\exp(-i\pi I_z),\\ \exp(-i\pi S_x),\exp(-i\pi S_x)\exp(-i\pi S^{\alpha}I_z),\\ \exp(-i\pi S_x)\exp(-i\pi S^{\beta}I_z),\exp(-i2\pi S_xI_z)\}. \tag{A2}$$

2. Convex closure of Weyl orbits

Assume that $a=a_1(-iS^{\beta}I_x)+a_2(-iS^{\alpha}I_x)$ and $b=b_1(-iS^{\beta}I_x)+b_2(-iS^{\alpha}I_x)$ are elements of $\mathfrak{a}$. We prove that $(a_1,a_2)^T$ is contained in the convex closure of the Weyl orbit of $(b_1,b_2)^T$ iff $(a_1,a_2)^T\prec_r(b_1,b_2)^T$.

Suppose $(a_1,a_2)^T$ is contained in the convex closure of the Weyl orbit of $(b_1,b_2)^T$. Assume that $|b_1|\geq|b_2|$. Then, $(a_1,a_2)^T=\Sigma_j w_j(b_{j,1},b_{j,2})^T$, where $(b_{j,1},b_{j,2})^T$ belongs to the set in Eq. (21) ($w_j\geq 0$ and $\Sigma_j w_j=1$). It follows that $|b_{j,1}|\leq|b_1|$ and $|b_{j,2}|\leq|b_1|$. Therefore, $|a_1|\leq|b_1|$ and $|a_2|\leq|b_1|$, implying $\max\{|a_1|,|a_2|\}\leq\max\{|b_1|,|b_2|\}$. Also note, $|a_1|+|a_2|\leq\Sigma_j w_j(|b_{j,1}|+|b_{j,2}|)=|b_1|+|b_2|$.

Suppose that $(a_1,a_2)^T\prec_r(b_1,b_2)^T$. The conditions $\max\{|a_1|,|a_2|\}\leq\max\{|b_1|,|b_2|\}$ and $|a_1|+|a_2|\leq|b_1|+|b_2|$ are equivalent to $(|a_1|,|a_2|)^T$ being weakly submajorized by $(|b_1|,|b_2|)^T$. Thus, we obtain from Proposition 4.C.2. of Ref. [59] that

$$\begin{aligned}(|a_1|,|a_2|)^T &= e_1(|b_1|,|b_2|)^T+e_2(|b_2|,|b_1|)^T+e_3(|b_1|,0)^T\\ &\quad+e_4(0,|b_1|)^T+e_5(|b_2|,0)^T+e_6(0,|b_2|)^T\\ &= f_1(|b_1|,|b_2|)^T+f_2(|b_2|,|b_1|)^T+f_3(|b_1|,-|b_2|)^T\\ &\quad+f_4(-|b_2|,|b_1|)^T+f_5(|b_2|,-|b_1|)^T\\ &\quad+f_6(-|b_1|,|b_2|)^T,\end{aligned}$$

where $e_j\geq 0$, $\Sigma_j e_j=1$, $f_1=e_1+(e_3+e_6)/2$, $f_2=e_2+(e_4+e_5)/2$, and $f_k=e_k/2$ for $k\in\{3,4,5,6\}$. In particular, we have that $f_j\geq 0$ (for all j) and $\Sigma_j f_j=1$. It follows that

$$\begin{aligned}(a_1,a_2)^T=(\epsilon_1|a_1|,\epsilon_2|a_2|)^T &= f_1(\epsilon_3 b_1,\epsilon_4 b_2)^T+f_2(\epsilon_5 b_2,\epsilon_6 b_1)^T\\ &\quad+f_3(\epsilon_7 b_1,\epsilon_8 b_2)^T+f_4(\epsilon_9 b_2,\epsilon_{10} b_1)^T\\ &\quad+f_5(\epsilon_{11} b_2,\epsilon_{12} b_1)^T+f_6(\epsilon_{13} b_1,\epsilon_{14} b_2)^T,\end{aligned}$$

for appropriate choices of $\epsilon_j\in\{1,-1\}$. We conclude the proof by consulting Eq. (21). A Lie-theoretic proof can be obtained by following Theorem 2 of Ref. [10].

3. KAK decomposition for elements of A

We prove that the elements $\exp(a')\in\mathrm{A}$ equal to $K_1\exp(a)K_2$ are given by the elements $(a_1',a_2')^T=(a_1,a_2)^T+2\pi(z_1,z_2)^T$, where $K_j\in\mathrm{K}$, $a'=a_1'(-iS^{\beta}I_x)+a_2'(-iS^{\alpha}I_x)$, $a=a_1(-iS^{\beta}I_x)+a_2(-iS^{\alpha}I_x)$, and $z_j\in\mathbb{Z}$.

We can choose a' as $a'=K(a+k)K^{-1}$, where K is an element of Eq. (A2) and $k\in\{q\in\mathfrak{a}|\exp(q)\in\mathrm{K}\}$ (cf. Ref. [10], Lemma 2, and Ref. [17], Proposition 4). Using the ansatz $\exp[a_1''(-iS^{\alpha}I_x)+a_2''(-iS^{\beta}I_x)]=\mathrm{id}_4$, where $a_1'',a_2''\in\mathbb{R}$, we obtain that $a_1'',a_2''\in\{4\pi z:z\in\mathbb{Z}\}$. It is a consequence of Theorem 8.5, Chap. VII, of Ref. [43] that $\{q\in\mathfrak{a}|\exp(q)\in\mathrm{K}\}$ is equal to the set $\{q_1(-iS^{\alpha}I_x)+q_2(-iS^{\alpha}I_x):q_1,q_2\in\{2\pi z:z\in\mathbb{Z}\}\}$. This completes the proof. We remark that $\exp[2\pi z_1(-iS^{\beta}I_x)+2\pi z_2(-iS^{\alpha}I_x)]=\exp[2\pi z_1(-iS^{\beta}I_z)+2\pi z_2(-iS^{\alpha}I_z)]$ for all $z_j\in\mathbb{Z}$, where $2\pi z_1(-iS^{\beta}I_z)+2\pi z_2(-iS^{\alpha}I_z)\in\mathfrak{k}$.

4. Proof of a majorization relation

We prove that $(a_1,a_2)^T\prec_r(a_1,a_2)^T+2\pi(z_1,z_2)^T$ holds for all $z_1,z_2\in\mathbb{Z}$, if we assume that $a_1,a_2\in[-\pi,\pi]$. As the case $z_1=z_2=0$ is trivial, we assume that $|z_1|>0$ or $|z_2|>0$. We get $\max\{|a_1+2\pi z_1|,|a_2+2\pi z_2|\}\geq 2\pi-\pi=\pi\geq\max\{|a_1|,|a_2|\}$, and the first condition in the definition of r majorization is satisfied. The second condition $|a_1+2\pi z_1|+|a_2+2\pi z_2|\geq|a_1|+|a_2|$ follows from the fact that $|a_j+2\pi z_j|\geq|a_j|$ is always true. In particular, this is trivial for $z_j=0$ and it is a consequence of $|a_j+2\pi z_j|\geq|(|2\pi z_j|-|a_j|)|\geq\pi\geq|a_j|$ in all other cases. The result follows.

[1] M. A. Nielsen, M. R. Dowling, M. Gu, and A. C. Doherty, Science **311**, 1133 (2006).
[2] M. A. Nielsen, Quantum Inf. Comput. **6**, 213 (2006).
[3] M. A. Nielsen, M. R. Dowling, M. Gu, and A. C. Doherty, Phys. Rev. A **73**, 062323 (2006).
[4] N. Khaneja, R. Brockett, and S. J. Glaser, Phys. Rev. A **63**, 032308 (2001).
[5] N. Khaneja and S. J. Glaser, Chem. Phys. **267**, 11 (2001).
[6] T. O. Reiss, N. Khaneja, and S. J. Glaser, J. Magn. Reson. **154**, 192 (2002).
[7] G. Vidal, K. Hammerer, and J. I. Cirac, Phys. Rev. Lett. **88**, 237902 (2002).
[8] K. Hammerer, G. Vidal, and J. I. Cirac, Phys. Rev. A **66**, 062321 (2002).
[9] A. M. Childs, H. L. Haselgrove, and M. A. Nielsen, Phys. Rev. A **68**, 052311 (2003).
[10] R. Zeier, M. Grassl, and T. Beth, Phys. Rev. A **70**, 032319 (2004).
[11] N. Khaneja, F. Kramer, and S. J. Glaser, J. Magn. Reson. **173**, 116 (2005).
[12] H. Yuan and N. Khaneja, Phys. Rev. A **72**, 040301(R) (2005).
[13] H. Yuan and N. Khaneja, in *Proceedings of the 44th IEEE Conference on Decision and Control, 2005 and 2005 European Control Conference, Seville, Spain* (IEEE, Piscataway, NJ, 2005), pp. 8006–8011.
[14] J. Swoboda, e-print arXiv:quant-ph/0601131v1.
[15] H. Yuan and N. Khaneja, Syst. Control Lett. **55**, 501 (2006).
[16] H. Yuan, Ph.D. thesis, Harvard University, 2006.
[17] G. Dirr, U. Helmke, K. Hüper, M. Kleinsteuber, and Y. Liu, J. Global Optim. **35**, 443 (2006).
[18] R. M. Zeier, Ph.D. thesis, Universität Karlsruhe, 2006 (unpublished).
[19] N. Khaneja, S. J. Glaser, and R. Brockett, Phys. Rev. A **65**, 032301 (2002).
[20] T. O. Reiss, N. Khaneja, and S. J. Glaser, J. Magn. Reson. **165**, 95 (2003).
[21] N. Khaneja, T. O. Reiss, C. Kehlet, T. Schulte-Herbrüggen, and S. J. Glaser, J. Magn. Reson. **172**, 296 (2005).
[22] T. Schulte-Herbrüggen, A. Spörl, N. Khaneja, and S. J. Glaser, Phys. Rev. A **72**, 042331 (2005).
[23] H. Yuan and N. Khaneja, *Proceedings of the 45th IEEE Conference on Decision and Control, San Diego, CA, 2006* (IEEE, Piscataway, NJ, 2006), pp. 3117–3120.
[24] N. Khaneja, B. Heitmann, A. Spörl, H. Yuan, T. Schulte-Herbrüggen, and S. J. Glaser, Phys. Rev. A **75**, 012322 (2007).
[25] H. Yuan, S. J. Glaser, and N. Khaneja, Phys. Rev. A **76**, 012316 (2007).
[26] M. Mehring, J. Mende, and W. Scherer, Phys. Rev. Lett. **90**, 153001 (2003).
[27] F. Jelezko, T. Gaebel, I. Popa, A. Gruber, and J. Wrachtrup, Phys. Rev. Lett. **92**, 076401 (2004).
[28] F. Jelezko, T. Gaebel, I. Popa, M. Domhan, A. Gruber, and J. Wrachtrup, Phys. Rev. Lett. **93**, 130501 (2004).
[29] M. Mehring, W. Scherer, and A. Weidinger, Phys. Rev. Lett. **93**, 206603 (2004).
[30] J. Mende, Ph.D. thesis, Universität Stuttgart, 2005.
[31] A. Heidebrecht, J. Mende, and M. Mehring, Solid State Nucl. Magn. Reson. **29**, 90 (2006).
[32] R. Rahimi Darabad, Ph.D. thesis, Osaka University, 2006.
[33] M. Mehring and J. Mende, Phys. Rev. A **73**, 052303 (2006).
[34] T. Gaebel *et al.*, Nat. Phys. **2**, 408 (2006).
[35] N. B. Manson, J. P. Harrison, and M. J. Sellars, Phys. Rev. B **74**, 104303 (2006).
[36] A. Heidebrecht, J. Mende, and M. Mehring, Fortschr. Phys. **54**, 788 (2006).
[37] F. Jelezko and J. Wrachtrup, Phys. Status Solidi A **203**, 3207 (2006).
[38] J. Wrachtrup and F. Jelezko, J. Phys.: Condens. Matter **18**, S807 (2006).
[39] L. Childress, M. V. Gurudev Dutt, J. M. Taylor, A. S. Zibrov, F. Jelezko, J. Wrachtrup, P. R. Hemmer, and M. D. Lukin, Science **314**, 281 (2006).
[40] A. Heidebrecht, Ph.D. thesis, Universität Stuttgart, 2006.
[41] D. D. Awschalom and M. E. Flatté, Nat. Phys. **3**, 153 (2007).
[42] M. V. Gurudev Dutt, L. Childress, L. Jiang, E. Togan, J. Maze, F. Jelezko, A. S. Zibrov, P. R. Hemmer, and M. D. Lukin, Science **316**, 1312 (2007).
[43] S. Helgason, *Differential Geometry, Lie Groups, and Symmetric Spaces* (American Mathematical Society, Providence, 2001), reprinted with corrections.
[44] A. W. Knapp, *Lie Groups Beyond an Introduction*, 2nd ed. (Birkhäuser, Boston, 2002).
[45] R. R. Ernst, G. Bodenhausen, and A. Wokaun, *Principles of Nuclear Magnetic Resonance in One and Two Dimensions* (Clarendon Press, Oxford, 1997), reprinted with corrections.
[46] A. Schweiger and G. Jeschke, *Principles of Pulse Electron Paramagnetic Resonance* (Oxford University Press, Oxford, 2001).
[47] V. Jurdjevic and H. J. Sussmann, J. Differ. Equations **12**, 313 (1972).
[48] S. S. Bullock, Quantum Inf. Comput. **4**, 396 (2004).
[49] S. S. Bullock and G. K. Brennen, J. Math. Phys. **45**, 2447 (2004).
[50] S. S. Bullock, G. K. Brennen, and D. P. O'Leary, J. Math. Phys. **46**, 062104 (2005).
[51] H. N. S. Earp and J. K. Pachos, J. Math. Phys. **46**, 082108 (2005).
[52] Y. Nakajima, Y. Kawano, and H. Sekigawa, Quantum Inf. Comput. **6**, 67 (2006).
[53] C. Van Loan, Numer. Math. **46**, 479 (1985).
[54] G. H. Golub and C. F. Van Loan, *Matrix Computations*, 2nd ed. (The Johns Hopkins University Press, Baltimore, 1989).
[55] C. C. Paige and M. Wei, Linear Algebr. Appl. **208-209**, 303 (1994).
[56] M. D. Shuster, J. Astronaut. Sci. **41**, 439 (1993).
[57] N. Khaneja, Phys. Rev. A **76**, 032326 (2007).
[58] J. S. Hodges, J. C. Yang, C. Ramanathan, and D. G. Cory, arXiv:0707.2956v1.
[59] A. W. Marshall and I. Olkin, *Inequalities: Theory of Majorization and Its Applications* (Academic Press, New York, 1979).