Efficient synthesis of quantum gates on indirectly coupled spins

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Experiments in coherent nuclear and electron magnetic resonance, and quantum computing in general correspond to control of quantum mechanical systems, guiding them from initial to final target states by unitary transformations. The control inputs (pulse sequences) that accomplish these unitary transformations should take as little time as possible so as to minimize the effects of relaxation and decoherence and to optimize the sensitivity of the experiments. Here, we derive a time-optimal sequences as fundamental building blocks for synthesize unitary transformations. Such sequences can be widely implemented on various physical systems, including the simulation of effective Hamiltonians for topological quantum computing on spin lattices. Experimental demonstrations are provided for a system consisting of three nuclear spins.

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I. INTRODUCTION

The control of quantum systems has important applications in physics and chemistry. In particular, the ability to steer the state of a quantum system (or an ensemble of quantum systems) from a given initial state to a desired target state forms the basis of spectroscopic techniques such as nuclear magnetic resonance (NMR) and electron spin resonance (ESR) spectroscopy [1, 2] and laser coherent control [3] and quantum computing [4, 5]. Experiments in coherent nuclear and electron magnetic resonance, and optical spectroscopy correspond to control of quantum mechanical ensembles, guiding them from initial to final target states by unitary transformations. The control inputs (pulse sequences) that accomplish these unitary transformations should take as little time as possible so as to minimize the effects of relaxation and decoherence and to optimize the sensitivity of the experiments. The time-optimal synthesis of unitary operators is now well understood for coupled two-spin systems [10–15]. This problem has also been recently studied in the context of linear three-spin topologies [16, 18–24]. In this article, we use optimal control technique to design pulse sequences to efficiently generate a class of quantum gates on three-spin systems, and show that such pulse sequences have significant savings in the implementation time of trilinear Hamiltonians and synthesis of couplings between indirectly coupled qubits over conventional methods. Such pulse sequences also have applications on various other systems. The article is organized as following; in section [II] we first review the previous results on the linearly coupled three-spin system; in section [III] we use optimal control techniques to design the new pulse sequences and show significant savings in the implementation time and the wide applications of such pulse sequences; in section [IV] we show the experiment implementation of these pulse sequences on NMR; section [V] concludes.

II. TIME OPTIMAL CONTROL FOR THREE LINEARLY COUPLED SPINS

In this section, we give a brief introduction of previous results on three linearly coupled spins [16], to which our new results is be compared with.

Consider a chain of three spins coupled by scalar couplings ($J_{13} = 0$). Furthermore assume that it is possible to selectively excite each spin (perform one qubit operations in context of quantum computing). The goal is to produce a desired unitary transformation $U \in SU(8)$, from the specified couplings and single spin operations in shortest possible time. The unitary propagator $U$, describing the evolution of the system in a suitable rotating frame is well approximated by

![FIG. 1: Three linearly coupled spins](image-url)
where

\[ H_d = 2\pi J_{12}I_{1z}I_{2z} + 2\pi J_{23}I_{2z}I_{3z}, \]
\[ H_1 = 2\pi I_{1x}, \]
\[ H_2 = 2\pi I_{1y}, \]
\[ H_3 = 2\pi I_{2x}, \]
\[ H_4 = 2\pi I_{2y}, \]
\[ H_5 = 2\pi I_{3x}, \]
\[ H_6 = 2\pi I_{3y}. \]

We use the notation \( I_{\ell\nu} = \bigotimes_j I_{\ell j} \), where \( a_j = \nu \) for \( j = \ell \) and \( a_j = 0 \) otherwise (see [11]). The matrices \( I_2 := \frac{1}{2} \left[ \begin{array}{cc} 1 & 0 \\ 0 & 1 \end{array} \right] \), \( I_3 := \frac{1}{2} \left[ \begin{array}{cc} 0 & -1 \\ 1 & 0 \end{array} \right] \), and \( I_0 := \left[ \begin{array}{cc} 1 & 0 \\ 0 & 1 \end{array} \right] \) are the Pauli spin matrices and \( I_0 \) is the 2 \times 2-dimensional identity matrix. The symbol \( J_{12} \) and \( J_{23} \) represents the strength of scalar couplings between spins (1, 2) and (2, 3) respectively, here we will treat the important case of this problem when the couplings are both equal \((J_{12} = J_{23} = J)\). We will be most interested in a unitary propagator of the form

\[ U = \exp(-i\theta I_{1z}I_{2z}I_{3z}). \]

These propagators are hard to produce as they involve trilinear terms in the effective Hamiltonian. We will refer to such propagators as trilinear propagators.

We assume that we can selectively rotate each spin at a rate much faster than the evolution of the couplings, i.e., the single spin operations can be done in negligible time.

**Theorem 1** [11] Given the spin system in (1), with \( J_{12} = J_{23} = J \) and \( J_{13} = 0 \), the minimum time \( t^*(U_F) \) required to produce a propagator of the form \( U_F = \exp(-i\theta I_{1z}I_{2z}I_{3z}) \), \( \theta \in [0, 4\pi] \) is given by

\[ t^*(U_F) = \frac{\sqrt{2\pi\theta - (\theta/2)^2}}{2\pi J} = \frac{\sqrt{\kappa(4 - \kappa)}}{2J}, \]

where \( \kappa = \frac{\theta}{2\pi} \). The pulse sequence that produces the propagator \( U_F \) is as follows.

\[ U_F = \exp(-i\pi J_{1z}I_{2y}) \exp(-i\pi J_{2x}) \exp(T[2\pi J(I_{1z}I_{2z} + I_{2z}I_{3z}) + i\beta I_{2x}]) \exp(i\pi J_{2y}) \]

\[ \exp(T[2\pi J(I_{1z}I_{2z} + I_{2z}I_{3z}) + i\beta I_{2x}]) \exp(i\pi J_{2y}) \]

where \( \beta = 2\pi - \theta/2 \) and \( T = \frac{\sqrt{\kappa(4 - \kappa)}}{2J} \).

### III. Quantum Gates Between Indirectly Coupled Spins

#### A. Time optimal sequences

In this section we derive a time optimal sequence for a control problem arises from simulation of trilinear couplings of indirectly coupled spins. First we consider to generate the trilinear term \( e^{i\theta S_i} \), where \( S_i = -4i[I_{1x}I_{2z}I_{3y} + I_{1y}I_{2z}I_{3x}] \).

Let

\[ S_2 = -2i[I_{1x}I_{2x} + I_{2x}I_{3z}], \]
\[ S_3 = -2i[I_{1y}I_{2y} + I_{2y}I_{3y}], \]

here \( S_2, S_3 \) are locally equivalent to the coupling Hamiltonian \( H_d \) (up to a re-scaling of time, from now on we assume the time unit is \( \frac{1}{\sqrt{J}} \)).

\[ S_2 = e^{-\frac{\pi}{2}(I_{1y}I_{2x} + I_{2x}I_{3z})}(-iH_d)e^{\frac{\pi}{2}(I_{1y}I_{2x} + I_{2x}I_{3z})} \]
\[ S_3 = e^{\frac{\pi}{2}(I_{1y}I_{2x} + I_{2x}I_{3z})}(-iH_d)e^{-\frac{\pi}{2}(I_{1y}I_{2x} + I_{2x}I_{3z})} \]

Notice that \( S_1, S_2, S_3 \) form an so(3) algebra, i.e.

\[ [S_1, S_2] = S_3, \]
\[ [S_2, S_3] = S_1, \]
\[ [S_3, S_1] = S_2, \]

thus we can map \( S_1, S_2, S_3 \) to \( \Omega_x, \Omega_y, \Omega_z \), where

\[ \Omega_x = \left( \begin{array}{ccc} 0 & 0 & 0 \\ 0 & 0 & -1 \\ 0 & 1 & 0 \end{array} \right), \]
\[ \Omega_y = \left( \begin{array}{ccc} 0 & 0 & 1 \\ 0 & 0 & 0 \\ -1 & 0 & 0 \end{array} \right), \]
\[ \Omega_z = \left( \begin{array}{ccc} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{array} \right). \]

Then we reformulate the problem as following:

\[ \frac{d}{dt} \Omega = A\Omega, \]

where \( u(t), v(t) \) can be \( \{\pm 1, 0\} \) as we can change the sign of the Hamiltonian by local controls, and it is 0 when it is not switched on. Also at each instant of time, only one Hamiltonian can be switched on, so \( \forall t, |u(t)| + |v(t)| = 1 \).
Now the question becomes how to generate $\Omega(T) = e^{i\Omega_T}$ in minimum time under the dynamics governed by Eq.\[3\] with the initial condition $\Omega(0) = I$. This is equivalent to find the optimal sequences

$$\exp(\pm\Omega_y t_1) \exp(\pm\Omega_z t_2) \exp(\pm\Omega_y t_3) \exp(\pm\Omega_z t_4) \cdots$$

to generate $\exp(\alpha\Omega_x)$, such that $T = \sum_i t_i$ is minimized. The conventional method is to use the Baker-Campbell-Hausdorff (BCH) formula

$$e^{\alpha\Omega_x} = e^{\frac{\alpha}{2}\Omega_x} e^{\alpha\Omega_x} e^{-\frac{\alpha}{2}\Omega_x},$$

which corresponds to set $u(t) = 0, v(t) = 1$ when $t \in [0, \frac{\pi}{2}], u(t) = 1, v(t) = 0$ when $t \in [\frac{\pi}{2}, \frac{3\pi}{2}]$ and $u(t) = 0, v(t) = -1$ when $t \in [\frac{3\pi}{2} + \alpha, \pi + \alpha]$, the total duration is $\pi + \alpha$ units of time. While this is optimal when $\alpha$ is larger than $\frac{\pi}{2}$, for small $\alpha$, it is far from optimal since there is always an offset $\pi$ for the total time.

The minimum time can actually be achieved by adding one switch. For $\alpha \in [0, \frac{\pi}{2}]$, the time optimal sequence takes the form

$$e^{\alpha\Omega_x} = e^{\frac{\alpha}{2}\Omega_x} e^{-\delta t\Omega_y} e^{-\delta t\Omega_z} e^{\frac{\alpha}{2}\Omega_x}, \quad \text{(6)}$$

while

$$t_2 = t_1 = \arccos \frac{1}{\sin \frac{\alpha}{2} + \cos \frac{\alpha}{2}}, \quad \delta t = \arccos (\cos \frac{\alpha}{2} - \sin \frac{\alpha}{2}). \quad \text{(7)}$$

The total time to generate $e^{\alpha\Omega_x}$ is

$$f(\alpha) = 2[\arccos \frac{1}{\sin \frac{\alpha}{2} + \cos \frac{\alpha}{2}} + \arccos (\cos \frac{\alpha}{2} - \sin \frac{\alpha}{2})].$$

Symmetrically, when $\alpha \in [-\frac{\pi}{2}, 0]$,

$$f(\alpha) = f(-\alpha).$$

So for $\alpha \in [-\frac{\pi}{2}, \frac{\pi}{2}]$,

$$f(\alpha) = 2[\arccos \frac{1}{\sin \frac{\alpha}{2} + \cos \frac{\alpha}{2}} + \arccos (\cos \frac{\alpha}{2} - \sin \frac{\alpha}{2})]. \quad \text{(8)}$$

The detailed derivation of this sequence can be found in the appendix, here we just give an intuitive picture of how the sequence works. Geometrically $\{e^{i\Omega_x}\}$ corresponds to the a rotation on a sphere which moves the point $(0, \sin \alpha, \cos \alpha)$ to $(0, 0, 1)$ while keeps the $x$-axis fixed. The sequence in Eq.\[6\] corresponds to four steps that achieve such a rotation. As shown in Fig.\[2\] it is first rotated around the $y$-axis for $t_1$ time, which rotates the point $a = (0, \sin \alpha, \cos \alpha)$ to $b = (\cos \alpha \sin t_1, \sin \alpha, \cos \alpha \cos t_1)$; then it’s rotated around $(-z)$-axis for $\delta t$ time, which rotates point $b$ to the point $c = (\sqrt{\cos^2 \alpha \sin^2 t_1 + \sin^2 \alpha}, 0, \cos \alpha \cos t_1)$ which is on the XZ-plane; after that it is rotated around $(-y)$-axis for $\delta t$ time to the point $d = (0, 0, 1)$; since $x$-axis is orthogonal to $\vec{a}$ and rotations does not change the angles, so after these rotations, the $x$-axis has moved to somewhere which is orthogonal to $\vec{d}$, i.e., somewhere on the XY-plane, so one needs to make another rotation around the $z$-axis for $t_2$ time, that moves the $x$-axis back to the original position.

It needs to be noted that when writing the manuscript, it was brought to the authors attention that Dirk Mettlenhuber had a similar construction of the time optimal sequences\[3\].

B. Quantum gates on indirectly coupled spins

The sequence can be directly used to simulate the trilinear terms $e^{i\theta S_1}$ on the linearly coupled three-spin system, which is

$$e^{\alpha S_1} = e^{\frac{\alpha}{2}S_3} e^{-\delta t S_2} e^{-\delta t S_3} e^{\frac{\alpha}{2}S_1}, \quad \text{(9)}$$

where $S_1, S_2, S_3$ are as defined in last section. The minimum time needed to generate $e^{i\theta S_1}, \theta \in [-\frac{\pi}{2}, \frac{\pi}{2}]$, is

$$f(\theta) = 2[\arccos \frac{1}{\sin \frac{\theta}{2} + \cos \frac{\theta}{2}} + \arccos (\cos \frac{\theta}{2} - \sin \frac{\theta}{2})].$$

(10)

It is not only shorter than the conventional method using BCH formula, but is also shorter than the strategy described in section\[1\]. In Fig.\[3\] we plotted the total time needed for different strategies.

We can also construct various other gates between indirectly coupled spin 1 and spin 3 in the linearly coupled three-spin system by concatenating the optimal sequence. For example, we can build gates $e^{-2i\theta [I_1 I_3 + I_3 I_1]}, \theta \in$
Combining them, we can efficiently simulate the Heisenberg coupling $e^{-i\delta \theta [I_1 I_2 + I_3 I_4]}$ between indirectly coupled spins.
Let’s first index the four spins $B_p$ act on with number 1, 2, 3, 4, and assume they have couplings to nearest neighbor: $\sigma_1, \sigma_2, \sigma_3, \sigma_4$, where $\sigma_1, \sigma_2$ denote an operator acting $\sigma_z$ on spin 1 and 2 and identity on other spins. To simulate $\exp(-i\theta B_p)$, we can implement the time optimal sequence as following:

$$\exp(-i\theta B_p) = \exp(-it_1\sigma_1\sigma_2\sigma_3) \exp(it_2\sigma_3\sigma_4) \exp(it_2\sigma_1\sigma_2\sigma_3) \exp(-it_1\sigma_3\sigma_4),$$

where

$$t_1 = \arccos \frac{1}{\sin \frac{\theta}{2} \cos \frac{\theta}{2}},$$

$$t_2 = \arccos (\cos \frac{\theta}{2} - \sin \frac{\theta}{2}).$$

Here $\exp(-it_1\sigma_1\sigma_2\sigma_3)$ can be generated by the sequence

$$\exp(-it_1\sigma_1\sigma_2\sigma_3) = \exp(-it'_1\sigma_1\sigma_2) \exp(it'_2\sigma_2\sigma_3) \exp(it'_2\sigma_1\sigma_3) \exp(-it'_1\sigma_2\sigma_3),$$

where terms containing $\sigma_x$ and $\sigma_y$ are obtained from single spin operations on Ising coupling and

$$t'_1 = \arccos \frac{1}{\sin \frac{t_1}{2} \cos \frac{t_1}{2}},$$

$$t'_2 = \arccos (\cos \frac{t_1}{2} - \sin \frac{t_1}{2}).$$

And the term $\exp(it_2\sigma_1\sigma_2\sigma_3y$) can be similarly generated. Thus by repeatedly using the four-pulses sequence, one can efficiently simulate $B_p$. Please note that the concatenation of the optimal sequence may not keep its optimality, nevertheless it is much more efficient than conventional method using BCH formula.

**IV. EXPERIMENT**

We experimentally demonstrated the implementation of the unitary transformation $e^{2\pi i(l_{1x}l_{2x}+l_{1y}l_{2y}+l_{1z}l_{2z})}$ on a BRUKER AVANCE 500M NMR spectrometer. The sample is the amino moiety of 15N acetamide ($NH_2COCH_3$). Two protons in the spin system $-NH_2$ present the spins 1 and 3. The chemical shift difference between the two protons is 306 Hz. Nuclear 15N denotes spin 2. The J-coupling constants among the three spins are $J_{12} = J_{23} = 88Hz$, $J_{13} = 2.6Hz$. The pulse sequence used in the experiment is shown in Fig.6. In this experiment we take $\theta = 2\pi$, $t_1 = t_2 = 2.84ms$, $\delta t = 5.68ms$. The whole duration of this pulse is 17ms. We choose six different initial states to observe the final states after the pulses are applied. For the six different initial states

$I_{1x}, I_{1y}, I_{3x}, I_{3y}, I_{1z}, I_{3z}$, the corresponding final states should be

$I_{1x}I_{2x}I_{3x}, -I_{1x}I_{2x}I_{3y}, I_{1x}I_{2z}I_{3z}, -I_{1y}I_{2x}I_{3z}$,
FIG. 6: The pulse sequence diagram for the unitary transformation $e^{2\pi i(I_1 I_2 I_3 + I_1 I_2 I_3)}$, where thin vertical lines denote 90° pulses and 180° pulses (the wide vertical lines) are inserted for refocusing of frequency offset effects. The durations $t_1, t_2$ and $\delta t$ can be calculated according to the above theory. That is $t_1 = t_2 = \arccos \frac{1}{\sqrt{1 + \cos \theta}}$, $\delta t = \arccos (\cos \frac{\theta}{2} - \sin \frac{\theta}{2})$.

$I_3$ and $-I_1$, respectively. Fig. IV shows the experimental spectra (for the state $I_iz_i$ ($i=1, 3$), a 90 degree reading pulse along y axis has been applied before acquisition) and Fig. A in the appendix shows the simulations using the theory. The consistency between theory and experimental spectra indicates that the pulse sequence works accurately and the unitary transformation has been experimentally implemented.

V. CONCLUSION

We derived a time optimal sequences to generate quantum gates, which can be widely used on various physical systems, for example, it can be used to generate various quantum gates on indirectly coupled spins and can also be used to simulate topological quantum computing on spin lattice.

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Appendix A: Time optimal pulse sequences

In this Appendix, we derive the solution of the optimal control problem for

$$\frac{d}{dt} \Omega = A \Omega$$

(A1)

$$A = u(t)\Omega_z + v(t)\Omega_y$$

$u(t), v(t) \in \{1, -1, 0\}$ and $\forall t, |u(t)| + |v(t)| = 1$. $\Omega(0) = I$, we want to generate $\Omega(T) = e^{i\Omega_T}$ in minimum time. For the rest of the derivation, we will omit $t$ for ease of notation.

We first apply the maximum principle[41]. Since the system has a fat distribution, there is no singular

extremal[42]. The control hamiltonian then can be written as

$$H = -1 + tr(\lambda^T A \Omega),$$

where $\lambda$ is an auxiliary 3-dim vector variable and $\dot{\lambda} = A\lambda$, the optimal control $u, v$ should maximize the control Hamiltonian, i.e., $u, v = \arg\max\{H\}$. 

FIG. 7: NMR spectra of different initial and final states, where left column are spectra of different initial states and right column are spectra of corresponding output states.
\[ H = -1 + tr(\lambda^T A\Omega) \]
\[ = -1 + tr(A\Omega\lambda^T) \]
\[ = -1 + tr(A\frac{\Omega\lambda^T - \lambda\Omega^T}{2} + A\frac{\Omega\lambda^T + \lambda\Omega^T}{2}) \]
\[ = -1 + tr(A\frac{\Omega\lambda^T - \lambda\Omega^T}{2}) \],

where the last step holds because \( A \) is skew symmetric and \( S = \frac{\Omega\lambda^T + \lambda\Omega^T}{2} \) is symmetric, as

\[ tr(AS) = tr[(AS)^T] = tr(-SA) = -tr(AS), \]

thus \( tr(AS) = 0 \). Now let

\[ M = \frac{\Omega\lambda^T - \lambda\Omega^T}{2} = m_x\Omega_x + m_y\Omega_y + m_z\Omega_z, \]

then

\[ H = -1 + tr(um_z\Omega_x^2) + tr(vm_y\Omega_y^2) \]
\[ = -1 - 2um_z - 2vm_y. \] (A3)

From the definition of \( M \), we get

\[ \hat{M} = [A, M], \]

expanding \( M \) and \( A \), it gives

\[ \hat{m}_x\Omega_x + \hat{m}_y\Omega_y + \hat{m}_z\Omega_z = [u\Omega_x + v\Omega_y, m_x\Omega_x + m_y\Omega_y + m_z\Omega_z], \]

we thus get

\[ \hat{m}_x = -um_y + vm_z, \]
\[ \hat{m}_y = um_x, \]
\[ \hat{m}_z = -vm_x, \] (A4)

as \( u, v = \text{argmax}\{H\} \), we get

\[ u = -sgn(m_z), v = 0 \text{ if } |m_z| > |m_y|, \]
\[ u = 0, v = -sgn(m_y) \text{ if } |m_z| < |m_y|, \]

if \( |m_z| = |m_y| \), then \( u, v \) can be either.

Assume that initially \( m_z(0) > m_y(0) > 0, m_x(0) > 0 \) (solutions under other initial conditions can be similarly worked out), then initially

\[ u = -sgn(m_z(0)) = -1, v = 0 \]

From Eq. A4 we get

\[ \hat{m}_x = m_y, \]
\[ \hat{m}_y = -m_x, \]
\[ \hat{m}_z = 0. \] (A5)

It evolves as in Fig. 8(a).

Case I: If \( m_z^2(0) + m_y^2(0) < m_z^2(0) \), then the controls are constant, \( u = -sgn(m_z(0)) = -1, v = 0 \) through out.

Case II: If \( m_z^2(0) + m_y^2(0) > m_z^2(0) \), then after evolving for some time, \( |m_y| \) will exceed \( |m_z| \), so we need to switch the controls at time point \( t_1 \), where \( m_y(t_1) = -m_z(0) \), to \( v = -sgn(m_y) = 1, u = 0 \), then from Eq. A4 we get

\[ \hat{m}_x = m_z, \]
\[ \hat{m}_y = 0, \]
\[ \hat{m}_z = -m_x. \] (A6)

FIG. 8: The evolution trajectory of \( m_z, m_y, m_z \).

which evolves as in fig. 8(b) until \( m_z(t_2) = -m_z(0) \) at some point \( t_2 \), where we switch the controls to \( u = -sgn(m_z) = 1, v = 0 \) and the dynamics changes to

\[ \hat{m}_x = -m_y, \]
\[ \hat{m}_y = m_x, \]
\[ \hat{m}_z = 0. \] (A7)

which evolves as in fig. 8(c) until \( m_y(t_3) = m_z(0) \) where we switch the controls to \( v = -sgn(m_y) = -1, u = 0 \) and the dynamics becomes

\[ \hat{m}_x = -m_z, \]
\[ \hat{m}_y = 0, \]
\[ \hat{m}_z = m_x. \] (A8)

which will evolves till \( m_z(t_4) = m_z(0) \) at some time point \( t_4 \), where we switch back to Eq. A5 and the process repeats. Other initial conditions of \( m_x, m_y, m_z \) give similar
periodical controls. So the optimal sequences in this case have the following pattern:

\[
\begin{array}{cccccc}
1 & 2 & 3 & 4 & 5 & \ldots & n \\
\delta t & \delta t & \delta t & \delta t & \delta t & \ldots & \delta t \\
\end{array}
\]

\[
\begin{array}{cccccc}
t_1 & t_2 & t_3 & t_4 & t_5 & \ldots & t_n \\
u,v & u,v & u,v & u,v & u,v & \ldots & u,v \\
1,0 & 1,0 & 0,1 & 1,0 & 0,1 & \ldots & 0,1 \\
0,-1 & 0,-1 & 0,1 & 0,-1 & 0,1 & \ldots & 0,1 \\
-1,0 & -1,0 & 1,0 & -1,0 & 1,0 & \ldots & 1,0 \\
1,0 & 1,0 & 0,1 & 1,0 & 0,1 & \ldots & 0,1 \\
0,-1 & 0,-1 & 1,0 & 0,-1 & 1,0 & \ldots & 1,0 \\
-1,0 & -1,0 & 0,1 & -1,0 & 0,1 & \ldots & 1,0 \\
\end{array}
\]

and also the sequences with \( u,v \) switched. Here \( t_1, t_2 \leq \delta t < \pi \), are the corresponding evolution periods of each step, the evolution periods for the intermediate steps are equal.

Case III: If \( m_x^2(0) + m_y^2(0) = m_z^2(0) \), the process starts similarly, first \( u = -\text{sgn}(m_z(0)) = -1, v = 0 \),

\[
\begin{align*}
m_x &= m_y, \\
m_y &= -m_x, \\
m_z &= 0.
\end{align*}
\]  

It evolves till \( m_y(t_1) = -m_z(0) \) at some time point \( t_1 \). But since \( m_x^2(0) + m_z^2(0) = m_y^2(0) \), so at time \( t_1, m_x(t_1) = 0 \). From Eq. \((A4)\), at time \( t_1 \)

\[
\begin{align*}
m_x &= -um_y + vm_z, \\
m_y &= um_x, \\
m_z &= -vm_x = 0.
\end{align*}
\]  

This is a singular point and we can choose \( u,v \) such that \( m_x = -um_y + vm_z = 0 \), which can be achieved by rapidly changing between \( u = -1, v = 0 \) and \( u = 0, v = 1 \). If it continues with \( u = -1, v = 0 \), then it will evolve till \( t_2 \) such that \( m_y(t_2) = m_z(0) \) and reach another singular point, where we can switching rapidly between \( u = -1, v = 0 \) and \( u = 0, v = -1 \), which is equivalent to evolve along \( (\Omega_y + \Omega_z) \). If it continues with \( u = 0, v = 1 \), then it follows the dynamics

\[
\begin{align*}
m_x &= m_z, \\
m_y &= 0, \\
m_z &= -m_x.
\end{align*}
\]  

This sequence is switching between regular points(\( \Omega_z \pm \Omega_y \)) and the regular points in the middle of sequence have to evolve for \( \pi \) units of time each. For the optimal sequence, we can just consider the sequences with at most one singular point, as if it appears twice, for example if the optimal sequence contains \( e^{\delta t_2(\Omega_y - \Omega_z)} e^{\Omega_z t} e^{\delta t_3(\Omega_y + \Omega_z)} \), then we can replace it by

\[
e^{\delta t_2(\Omega_y - \Omega_z)} e^{\Omega_y t} e^{\delta t_3(\Omega_y + \Omega_z)} e^{\Omega_z t}
\]

which has only one singular point with same time cost.

With these information, we can determine the optimal sequence to generate \( e^{\alpha \Omega} \), \( \alpha \in \mathbb{R} \). The element \( \{e^{\alpha \Omega}\} \) has a one to one correspondence to the action on sphere of rotating the point \( (0, \sin \alpha, \cos \alpha) \) to \( (0,0,1) \) while keeping the \( x \)-axis fixed. We now study the time optimal way to generate such actions.

Clearly the constant controls can’t rotate the point \( (0, \sin \alpha, \cos \alpha) \) to \( (0,0,1) \) and using two rotations we can’t rotate the point \( (0, \sin \alpha, \cos \alpha) \) to \( (0,0,1) \) while keeping the \( x \)-axis fixed. There are a few ways to generate \( e^{\Omega t} \) using three rotations, they are all essentially equivalent to

\[
e^{\alpha \Omega_x} e^{\beta (\Omega_z + \Omega_y)} e^{\gamma \Omega_z},
\]

or

\[
e^{\alpha \Omega_z} e^{\beta (\Omega_x + \Omega_y)} e^{\gamma \Omega_x}.
\]

All these pulses turn out taking longer time than the four rotations presented below, so we will not present the detail of the calculation on these pulses.

![FIG. 9: Rotations that moves \( a = (0, \sin \alpha, \cos \alpha) \) to \( d = (0,0,1) \) via the point \( b = (\cos \alpha \sin t_1, \sin \alpha, \cos \alpha \cos t_1) \) and \( c = (\sqrt{\cos^2 \alpha \sin^2 t_1 + \sin^2 \alpha}, 0, \cos \alpha \cos t_1) \).](attachment:image.png)

We now give the strategy with four rotations that moves \( (0, \sin \alpha, \cos \alpha) \) to \( (0,0,1) \) and keeps the \( x \)-axis fixed. As shown in Fig.9 it is first rotated around the \( y \)-axis for \( t_1 \) time, which rotates the point \( a = (0, \sin \alpha, \cos \alpha) \) to \( b =
(cos α sin t₁, sin α, cos α cos t₁); then it is rotated around 
(-z)-axis for δt time, which rotates point b to point
c = (\sqrt{cos^2 α sin^2 t₁ + sin^2 α, 0, cos α cos t₁}) which is on
the XZ-plane; after that it is rotated around the (-y)-axis
for δt time to the point d = (0, 0, 1); since the x-axis is or-
thogonal to \(\mathbf{a} \mathbf{b} \mathbf{d}\) and rotations does not change the angles,
so after these rotations, the x-axis has moved to some-
where which is orthogonal to \(\mathbf{a} \mathbf{d}\), i.e., somewhere on the
XY-plane, so we need to make another rotation around
the z-axis for \(t₂\) time, which moves the x-axis back to
the original position. After combining these rotations, we get
\[ e^{αΩz} = e^{δtΩz} e^{-δtΩs} e^{-δtΩy} e^{δtΩy} e^{δtΩs} e^{δtΩz}. \] (A14)

From the second and third rotations, we get two equa-
tions on δt and \(t₁\)
\[ \tan(δt) = \frac{\sin α}{\cos α \sin t₁}, \] (A15)
\[ \tan(δt) = \frac{\sqrt{\cos^2 α \sin^2 t₁ + \sin^2 α}}{\cos α \cos t₁}, \]
which we can solve to get the value of \(t₁\) and δt:
\[ t₁ = \arccos \left( \frac{1}{\sin^2 t₁ + \cos^2 \frac{α}{2}} \right), \]
\[ δt = \arccos(\cos \frac{α}{2} - \sin \frac{α}{2}). \] (A16)

To calculate the value of \(t₂\), we need to figure
out the trajectory of the x-axis: it is first ro-
tated from (1, 0, 0) to (cos \(t₁\), 0, -sin \(t₁\)), then it is
rotated to (cos \(t₁\) cos δt, -cos \(t₁\) sin δt, -sin \(t₁\)),
then to (\(\sqrt{\cos^2 t₁ \cos^2 δt + \sin^2 t₁, -cos t₁ \sin δt, 0}\))
back to (1,0,0), i.e.,
\[ \cos t₂ = \sqrt{\cos^2 t₁ \cos^2 δt + \sin^2 t₁} = \frac{1}{\sin^2 t₁ + \cos^2 \frac{α}{2}}, \]
so
\[ t₂ = t₁ = \arccos \left( \frac{1}{\sin^2 t₁ + \cos^2 \frac{α}{2}} \right). \]

So the total time to generate \(e^{αΩz}\) is
\[ f(α) = 2[arccos \left( \frac{1}{\sin^2 \frac{α}{2} + \cos^2 \frac{α}{2}} \right) + arccos(\cos \frac{α}{2} - \sin \frac{α}{2})]. \] (A17)
Symmetrically when \(α \in [-\frac{π}{2}, 0]\),
\[ f(α) = f(-α). \]
So for \(α \in [-\frac{π}{2}, \frac{π}{2}]\),
\[ f(α) = 2[arccos \left( \frac{1}{\sin^2 \frac{α}{2} + \cos^2 \frac{α}{2}} \right) + arccos(\frac{|α|}{2} - \sin \frac{|α|}{2})]. \] (A18)

This is actually the optimal sequences, adding more
switches would not help [2]. For completeness, we
sketch the proof in [3] showing a representative sequence
\(e^{Ωs} e^{Ωy} e^{Ωy} e^{-Ωs} e^{-Ωy} e^{-Ωy}\) can not be part of optimal sequence,
thus optimal sequences can not have more than
four rotations. First note that
\[ e^{Ωy} e^{Ωy} e^{-Ωs} e^{-Ωy} e^{-Ωs} e^{-Ωy}\] can be time optimal.
As \(3s > 2π - s\) when \(s > \frac{π}{4}\), thus only for \(s \leq \frac{π}{4}\),
e\(e^{Ωs} e^{Ωy} e^{-Ωs} e^{-Ωy} e^{-Ωy}\) can be time optimal.
For \(s = \frac{π}{4}\),
\[ e^{Ωs} e^{Ωy} e^{-Ωs} e^{-Ωy} e^{-Ωy} = e^{Ωs} e^{-Ωy} = e^{Ωs} e^{-Ωy} = e^{Ωs} e^{-Ωy} = e^{Ωs} e^{-Ωy}, \] (A19)
\[ e = e^{t(\frac{3π}{4})} e^{Ωs} e^{Ωy} e^{Ωy}, \]
as \(\frac{π}{2} - t + \frac{π}{4} + \frac{π}{4} < t + \frac{3π}{4}\) thus the sequences with \(s = \frac{π}{4}\)
can not be optimal.
For \(s \in (0, \frac{π}{4})\), it can be shown that for \(t\) sufficiently
small, there exists \(r₁(t), r₂(t), r₃(t)\) such that
\[ e^{Ωs} e^{-Ωs} e^{Ωy} e^{-Ωy} e^{Ωy} e^{-Ωy} e^{-Ωy} \]
and \(r₁ + 2r₂ + r₃ < 2t + 3s\). Thus \(e^{Ωs} e^{Ωy} e^{-Ωs} e^{-Ωy} e^{Ωy}\) can not be optimal or part of optimal sequences.

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FIG. 10: Simulation of NMR spectra of different initial and final states, where left column are spectra of different initial states and right column are spectra of corresponding output states.