Bipartite entanglement and control in multiqubit systems

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Abstract

In this paper, the effect of the control on bipartite entanglement is discussed from a geometric viewpoint for a nuclear magnetic resonance (NMR) system as a model of the \( n \)-qubit control system. The Hamiltonian of the model is the sum of the drift and control Hamiltonians, each of which describes the interaction between pairs of qubits (or \( \frac{1}{2} \)-spins) and between one of qubits and an external magnetic field, respectively. According to the bipartite partition \((\mathbb{C}^2)^{\otimes m} = (\mathbb{C}^2)^{\otimes \ell} \otimes (\mathbb{C}^2)^{\otimes m}\) with \( \ell + m = n \), the Schrödinger equation for the NMR system is put in the matrix form. This paper gives a solution to the Schrödinger equation with the assumptions of small coupling among qubits and of constant controls. The solution is put in the form of power series in small parameters. In particular, in the case of the two-qubit NMR system, the drift and control Hamiltonians are shown to be coupled to work for entanglement promotion, by examining solutions to the Schrödinger equation in detail. The concurrence, a measure of entanglement, is evaluated along the solution for a small time interval in order to observe that the control effect appears, not at the first-order terms in \( t \), but at the higher-order terms in \( t \). The evaluated concurrence also suggests which control makes the two-qubit more entangled or less.

*Key words: bipartite entanglement, control, local transformation group, NMR system.

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1 Introduction

Entanglement plays many roles in quantum computation and quantum information theory, and is a resource for quantum communication. Recently, the geometry of entanglement has been developed [9,11–13,18–22]. The algebraic study of entanglement also has been made to seek for invariants that quantify multiqubit entanglement [4,10,26]. The geometry of entanglement in few-qubit systems was studied in [18,22] and in [20,21] in terms of the Hopf fibration and the complex projective spaces, respectively. The first author [11] showed that the concurrence for the two-qubit system can be characterized as the coordinate of the factor space of the state space by the action of the local transformation group. He continued to study bipartite entanglement in multiqubit systems [12]. In particular, the sets of separable and maximally entangled states in bipartite entanglement are identified, and the concurrence is extended to a measure of bipartite entanglement in multiqubit systems. Further, the distance between the set of states of prescribed entanglement and the set of separable states is measured. The bipartite entanglement is measured also for the process of Grover’s search algorithm [13]. For the bipartite entanglement, see also [9,19].

On the other hand, from a practical standpoint, it is of much importance to control quantum states in order to create entangled states. Many of papers on control problems for multiqubit systems deal with control systems on unitary groups [1–3,5,6,15–17,23]. The theory of control on compact Lie groups has been established [14]. A sufficient condition for controllability of the control system on \( SU(N) \) was given in [3]. The Cartan decomposition of the Lie algebra and the symmetric space are techniques frequently used for studying control systems [15–17].

Two notions as to controllability of multiqubit systems were defined and studied by Albertini and D’Alessandro [1,2], which are “pure state controllability” and “equivalent state controllability”. In [2], equivalent state controllability was shown to be equivalent to pure state controllability. In the case of multiqubit systems lying in electro-magnetic fields, they [1] gave a necessary and sufficient condition for the system to be (pure state) controllable. The condition is described in terms of the graph associated with the interactions of qubits, which is related with the Lie algebraic structure of the system. Their method is based on the control theory on compact Lie groups [6,14]. As will be pointed out in Appendix B, their model Hamiltonian is a bit different from that adopted in this article.
The aim of this article is to study entanglement and control of nuclear magnetic resonance (NMR) multiqubit systems from a geometric viewpoint. Since the notion of entanglement cannot be defined in terms of the whole transformation group, the control theory on compact Lie groups will not fit in with the study of entanglement and control. The state space of a finite-dimensional quantum system is a finite complex vector space with a constraint required for the probability density. In this article, the state space is a linear space of complex matrices with the constraint from the probability density. The reason for the choice of the present state space is that the entanglement measure like the concurrence is easy to treat on this state space. The state space admits the two-sided action of the local transformation group $U(2^\ell) \otimes U(2^m)$, where $m$ and $\ell$ are determined by the bipartite partition of the multiqubit $(\mathbb{C}^2)^\otimes n = (\mathbb{C}^2)^\otimes \ell \otimes (\mathbb{C}^2)^\otimes m$. As will be stated later, the entanglement of the multiqubit system is invariant by the action of the local transformation group. Hence, in order to make the multiqubit more entangled, the control is required to move the state in the direction transverse to the orbit of the local transformation group. However, for NMR systems to be treated in this article, the control vectors generated by the control Hamiltonian are tangent to the orbit mentioned above, so that they would not make the system more entangled, if no coupling were made with other vector fields. Since the total Hamiltonian of the NMR system is the sum of the control and drift Hamiltonians, one may expect the coupling between them to occur. The objectives in this article are to set up the control problem on the above-mentioned state space and to show, by solving the Shrödinger equation with constant controls, that the control and drift Hamiltonian are coupled together to give rise to vector fields transverse to the group orbit for the entanglement promotion of the multiqubit system.

The organization of this paper is as follows: Section 2 contains a geometric setting up of $n$-qubit systems. According to the bipartite partition $(\mathbb{C}^2)^\otimes n = (\mathbb{C}^2)^\otimes \ell \otimes (\mathbb{C}^2)^\otimes m$ with $\ell + m = n$, the state space $M$ is viewed as the space of $2^\ell \times 2^m$ normalized matrices $C$, on which the local transformation group $G = U(2^\ell) \otimes U(2^m)$ acts. The action of $G$ leaves invariant the bipartite entanglement of the $n$-qubit. Linear operators on the $n$-qubit system are mapped to vector fields on the state space $M$. Lie brackets among those vector fields are given explicitly. In Section 3, the Hamiltonian of an NMR model is given, which is split into two operators, the drift Hamiltonian $\hat{H}_d$ and control Hamiltonian $\hat{H}_c$. The $\hat{H}_d$ and $\hat{H}_c$ describe the interactions between pairs of $n$ qubits and between one of qubits and an external magnetic field, respectively. In addition,
a solution to $n$-qubit NMR systems with a certain type of drift Hamiltonians is given with the assumption that the coupling constants between qubits are small enough. The solution is put in the form of power series in small parameters related to the coupling constants. It will be shown that the vector fields associated with the drift and control Hamiltonians are coupled actually. In Section 4, the coupling between those vector fields is extensively studied on the two-qubit system to observe that entanglement is actually promoted. Discussion runs as follows: A solution $C(t)$ of the Schrödinger equation in the matrix form is obtained as a power series in the small coupling constant. The solution is examined in detail to show that the vector fields associated with the control and drift Hamiltonians are coupled indeed to induce vector fields transverse to the $G$-orbit. The effect of the control on entanglement is verified by estimating the difference between the concurrences of the solutions for the controlled and uncontrolled systems for a small time interval in the case that the initial state is a diagonal matrix $\Lambda = \text{diag} (\lambda_1, \lambda_2)$. The result is that the effect of the control is so slow that it emerges at the third-order term in $t$. Concluding remarks are made in Section 5, in which the Cartan decomposition method is related to the present method. Appendix A gives a list of Lie brackets among vector fields related with the drift and control Hamiltonians for the two-qubit NMR system. Appendix B contains the proof of the controllability of the $n$-qubit NMR system. The method for proof is different from that in [1].

2 Geometric setting for bipartite entanglement

2.1 The state space

We start with geometric setting for the $n$-qubit system after [12]. The Hilbert space for the $n$-qubit system is $(\mathbb{C}^2)^\otimes n = \mathbb{C}^2 \otimes \cdots \otimes \mathbb{C}^2$, whose state vectors $|\Psi\rangle$ are put in the form 

$$|\Psi\rangle = \sum_{J=(j_1, \ldots, j_n)\in \{0,1\}^n} c_J |j_1\rangle \otimes \cdots \otimes |j_n\rangle \quad \text{with} \quad \sum_J |c_J|^2 = 1,$$

where $\{|0\rangle, |1\rangle\}$ denotes the computational basis of a single qubit system $\mathbb{C}^2$. We denote by $\{|J\rangle\}_{J\in \{0,1\}^n}$ the standard orthonormal basis $|j_1\rangle \otimes \cdots \otimes |j_n\rangle$ of $(\mathbb{C}^2)^\otimes n$. The Hermitian inner product of $|\Psi\rangle = \sum_J c_J |J\rangle$ and $|\Phi\rangle = \sum_J d_J |J\rangle$ is then given by $\langle \Psi | \Phi \rangle = \sum_J \overline{c_J} d_J$.

According to a bipartite partition of the $n$ qubits, the Hilbert space $(\mathbb{C}^2)^\otimes n$ is decomposed into the tensor product $(\mathbb{C}^2)^\otimes \ell \otimes (\mathbb{C}^2)^\otimes m$ with $\ell + m = n$. We here assume...
that \(0 < \ell \leq m\) without loss of generality. A state vector \(|\Psi\rangle\) of the \(n\)-qubit system is separable with respect to the bipartite decomposition \((\mathbb{C}^2)^\otimes n = (\mathbb{C}^2)^\otimes \ell \otimes (\mathbb{C}^2)^\otimes m\), if there are two unit vectors \(|\Psi_1\rangle \in (\mathbb{C}^2)^\otimes \ell\) and \(|\Psi_2\rangle \in (\mathbb{C}^2)^\otimes m\) such that \(|\Psi\rangle = |\Psi_1\rangle \otimes |\Psi_2\rangle\).

A vector \(|\Psi\rangle\) is said to be entangled if it is not separable.

According to the bipartite decomposition \((\mathbb{C}^2)^\otimes n = (\mathbb{C}^2)^\otimes \ell \otimes (\mathbb{C}^2)^\otimes m\), a state vector \(|\Psi\rangle\) can be rewritten in the form

\[
|\Psi\rangle = \sum_{A \in \{0,1\}^\ell} \sum_{B \in \{0,1\}^m} c_{AB} |A\rangle \otimes |B\rangle,
\]

where \(A = (j_1, \cdots, j_\ell)\) and \(B = (j_{\ell+1}, \cdots, j_n)\). This expression gives rise to the isomorphism of \((\mathbb{C}^2)^\otimes n\) to the vector space \(\mathbb{C}^{2^\ell \times 2^m}\) of \(2^\ell \times 2^m\) complex matrices,

\[
\iota: (\mathbb{C}^2)^\otimes n \rightarrow \mathbb{C}^{2^\ell \times 2^m}; \quad |\Psi\rangle = \sum_{A,B} c_{AB} |A\rangle \otimes |B\rangle \mapsto C = (c_{AB}). \tag{2.1}
\]

The \(\mathbb{C}^{2^\ell \times 2^m}\) is endowed with the inner product given by \(\text{tr}(C_1^* C_2)\) for \(C_1, C_2 \in \mathbb{C}^{2^\ell \times 2^m}\).

Since \(|\Psi\rangle\) is normalized, the corresponding matrix \(C = \iota(|\Psi\rangle)\) is subject to the condition \(\text{tr}(C^* C) = 1\). Thus, the state space for the \(n\)-qubit system is defined to be

\[
M := \{C \in \mathbb{C}^{2^\ell \times 2^m} \mid \text{tr}(C^* C) = 1\};
\]

which is diffeomorphic with the \((2^n+1-1)\)-dimensional sphere. The state space \(M\) becomes a Riemannian manifold equipped with the standard metric

\[
\langle X, Y \rangle_C := \frac{1}{2} \text{tr}(X^* Y + Y^* X), \quad X, Y \in T_CM, \tag{2.2}
\]

where the tangent space to \(M\) at \(C\) is identified with

\[
T_CM = \{X \in \mathbb{C}^{2^\ell \times 2^m} \mid \text{tr}(X^* C + C^* X) = 0\}.
\]

### 2.2 Bipartite entanglement

In this subsection, we make a review of a measure of bipartite entanglement introduced by the first author [11, 12].

The separability condition of a state vector \(|\Psi\rangle\) can be expressed as rank \(CC^* = 1\), where \(C\) is the matrix corresponding to \(|\Psi\rangle\). Since the \(2^\ell \times 2^\ell\) matrix \(CC^*\) is positive semi-definite together with \(\text{tr}(CC^*) = 1\) and \(\ell \leq m\), the condition of rank \(CC^* = 1\) is equivalent to \(\det(I_{2^\ell} - CC^*) = 0\), where \(I_{2^\ell}\) denotes the \(2^\ell \times 2^\ell\) identity matrix. The first author [11, 12] showed that the quantity

\[
F(C) := \det(I_{2^\ell} - CC^*) \tag{2.3}
\]
serves as a measure of bipartite entanglement, and as an extension of the concurrence which is well-known as a measure of entanglement in two-qubit system. In fact, one has $F(C) = \det(CC^*)$, the square of the concurrence, for the two-qubit with $\ell = m = 1$. The quantity $F(C)$ is non-negative, and vanishes if and only if $C$ is separable. Further, the $F$ takes the maximal value $(1 - 1/2^\ell)^2$ if and only if all eigenvalues of $CC^*$ coincide. If $F$ attains the maximal value at $C$, the matrix $C \in M$ or the corresponding state vector $|\Psi\rangle$ is said to be maximally entangled with respect to the bipartite decomposition $(C^2)^\otimes_m = (C^2)^\otimes_\ell \otimes (C^2)^\otimes_m$.

2.3 $U(2^\ell) \otimes U(2^m)$-action

The action of the group $G = U(2^\ell) \otimes U(2^m)$ on $M \subset \mathbb{C}^{2^\ell \times 2^m}$ is defined by

$$C \mapsto gCh^T,$$

where $g \in U(2^\ell)$ and $h \in U(2^m)$. This action is isometric with respect to (2.2). We remark here that the map $\iota$ is $G$-equivariant, namely, $\iota^{-1}(gCh^T) = (g \otimes h)\iota^{-1}(C)$. As is easily verified, the function $F$ is invariant under the $G$-action, so that the $G$-action (2.4) does not change bipartite entanglement.

We now consider the quotient space $M/G$. An arbitrary matrix $C \in M$ is decomposed into the product

$$C = g(\Lambda, 0)h^T, \quad g \in U(2^\ell), h \in U(2^m), \Lambda = \text{diag}(\lambda_1, \ldots, \lambda_{2^\ell}),$$

where $0$ is the $2^\ell \times (2^m - 2^\ell)$ zero matrix, and $\lambda_1, \lambda_2, \ldots, \lambda_{2^\ell}$ are the eigenvalues of $CC^*$. In the following, we assume that $\lambda_1 \geq \cdots \geq \lambda_{2^\ell} \geq 0$. The quantities $\lambda_1, \cdots, \lambda_{2^\ell}$ are called the singular values of $C$. The expression (2.5) is equivalent to the Schmidt decomposition of a state vector $|\Psi\rangle$. From this decomposition, the quotient space $M/G$ can be identified with

$$\left\{(\lambda_1, \cdots, \lambda_{2^\ell}) \in \mathbb{R}^{2^\ell} \mid \lambda_1 \geq \cdots \geq \lambda_{2^\ell} \geq 0, \lambda_1^2 + \cdots + \lambda_{2^\ell}^2 = 1\right\}.$$ 

The sets of separable and maximally entangled states project through the natural projection $M \to M/G; C \mapsto (\lambda_1, \cdots, \lambda_{2^\ell})$ to the points $(1, 0, \cdots, 0)$ and $(1/\sqrt{2^\ell}, \cdots, 1/\sqrt{2^\ell}) \in \mathbb{R}^{2^\ell}$, respectively, both of which lie on the boundary of the quotient $M/G$.

The state space $M$ is stratified into strata, according to $G$-orbit types [12], which are determined by the isotropy subgroups $G_C := \{g \otimes h \in G \mid gCh^T = C\}$. The isotropy subgroup and the orbits are already studied for four-qubit systems in detail, and for multiqubit systems in brief [12]. We now summarize the results in a refined form.
Proposition 2.1. Let $N$ be the number of distinct singular values of $C$, and $m_j$ the multiplicity of its $j$-th largest singular value.

(1) If $CC^*$ is non-singular, the isotropy subgroup $G_C$ is isomorphic to

$$\left(U(m_1) \times \cdots \times U(m_N)\right) \otimes U(2^m - 2^\ell),$$

where the product $U(m_1) \times \cdots \times U(m_N)$ is viewed as a subgroup of $U(2^\ell)$, and the $G$-orbit $O_C := \{(g \otimes h)C | g \otimes h \in G\}$ is diffeomorphic to

$$G/G_C \approx U(2^\ell) \times \left(U(m_1) \times \cdots \times U(m_N)\right) V_{2^\ell}(\mathbb{C}^{2m}).$$

(1-1) In the generic case that singular values of $C$ are all distinct and non-zero, $G_C$ and $O_C$ take, respectively, the form

$$G_C \cong T^{2^\ell} \otimes U(2^m - 2^\ell) \quad \text{and} \quad O_C \approx U(2^\ell) \times_{TV^{2^\ell}} V_{2^\ell}(\mathbb{C}^{2m}).$$

(1-2) For a maximally entangled state $C$, $G_C$ and $O_C$ become, respectively,

$$G_C \cong U(2^\ell) \otimes U(2^m - 2^\ell) \quad \text{and} \quad O_C \approx V_{2^\ell}(\mathbb{C}^{2m}).$$

(2) If $CC^*$ is singular, the group $G_C$ is isomorphic to

$$\left(U(m_1) \times \cdots \times U(m_{N-1}) \times U(m_N)\right) \otimes U(2^m - 2^\ell + m_N),$$

and the orbit $O_C$ is diffeomorphic to

$$G/G_C \approx V_{2^\ell-m_N}(\mathbb{C}^{2^\ell}) \times \left(U(m_1) \times \cdots \times U(m_{N-1})\right) V_{2^\ell-m_N}(\mathbb{C}^{2m}).$$

(2-1) In particular, if $C$ is separable, $G_C$ and $O_C$ are, respectively, of the form

$$G_C \cong U(2^\ell - 1) \times U(2^m - 1), \quad \text{and} \quad O_C \approx S^{2^\ell+1-1} \times_{U(1)} S^{2m+1-1}.$$

2.4 Vertical and horizontal subspaces

This subsection deals with the infinitesimal action of $G = U(2^\ell) \otimes U(2^m)$. For $\xi \in u(2^\ell)$ and $\eta \in u(2^m)$, the fundamental vector field associated with $\xi \otimes I_{2^m} + I_{2^\ell} \otimes \eta \in \mathfrak{g} = \text{Lie}(G)$ is determined through

$$X_{\xi \otimes I_{2^m} + I_{2^\ell} \otimes \eta}(C) := \frac{d}{dt} \bigg|_{t=0} e^{i\xi C} e^{i\eta^T} = \xi C + C \eta^T.$$
The vertical subspace $V_C$ is defined to be the tangent space $T_C\mathcal{O}_C$ at $C$ to the orbit $\mathcal{O}_C$, and formed by the fundamental vector fields evaluated at $C$,

$$V_C = \{X_\xi \otimes I_2m + I_2m \otimes \eta(C) = \xi C + C\eta^T \mid \xi \in \mathfrak{u}(2^\ell), \eta \in \mathfrak{u}(2^m)\}.$$  

The dimension of $V_C$ is given by $\dim V_C = 2^{\ell+1} + 2^{m+1} - 3$, if $C$ is separable,

$$\dim V_C = 2^{\ell+1} + 2^{m+1} - 2^{2\ell}, \quad \text{if } C \text{ is maximally entangled.} \quad (2.11)$$

The horizontal subspace $H_C$ of $T_C M$ is defined to be the orthogonal complement of $V_C$ with respect to the metric (2.2). A vector $X \in T_C M$ is horizontal, if and only if $\langle X, \xi C + C\eta^T \rangle = 0$ for any $\xi, \eta \in \mathfrak{u}(2^\ell)$, so that the horizontal subspace at $C \in M$ is expressed as

$$H_C = \{X \in T_C M \mid XC^* - CX^* = 0, C^*X - X^*C = 0\}.$$  

From (2.11) together with $\dim H_C = 2^{\ell+m+1} - 1 - \dim V_C$, we have

$$\dim H_C = \begin{cases} 
2(2^\ell - 1)(2^m - 1), & \text{if } C \text{ is separable,} \\
2^{\ell+1} - 1, & \text{if } C \text{ is maximally entangled.}
\end{cases}$$

### 2.5 The Lie algebra of linear vector fields on $M$

In discussing control problems, we need to calculate Lie brackets of vector fields on $\mathbb{C}^{2^\ell \times 2^m}$. In particular, we have to work with the Lie brackets of vector fields of the form

$$X_{A \otimes B}(C) := ABC^T, \quad C \in \mathbb{C}^{2^\ell \times 2^m},$$

where $A \in \mathbb{C}^{2^\ell \times 2^\ell}$ and $B \in \mathbb{C}^{2^m \times 2^m}$ do not need to be Hermitian or skew Hermitian at present. The Lie bracket of $A_1 \otimes B_1, A_2 \otimes B_2 \in \mathbb{C}^{2^\ell \times 2^\ell} \otimes \mathbb{C}^{2^m \times 2^m}$ is defined to be

$$[A_1 \otimes B_1, A_2 \otimes B_2] := [A_1, A_2] \otimes B_1B_2 + A_2A_1 \otimes [B_1, B_2]. \quad (2.12)$$

Since this bracket is verified to satisfy the Jacobi identity, the linear space $\mathbb{C}^{2^\ell \times 2^\ell} \otimes \mathbb{C}^{2^m \times 2^m}$ is endowed with a Lie algebraic structure. This Lie algebra is isomorphic to $\mathfrak{gl}(2^n, \mathbb{C})$ through

$$f : \mathbb{C}^{2^\ell \times 2^\ell} \otimes \mathbb{C}^{2^m \times 2^m} \longrightarrow \mathbb{C}^{2^n \times 2^n} : A \otimes B \longrightarrow \begin{pmatrix} a_{11}B & \cdots & a_{12^\ell}B \\
\vdots & & \vdots \\
a_{2^\ell 1}B & \cdots & a_{2^\ell 2^\ell}B \end{pmatrix}.$$
In fact, $f$ is shown to be bijective and homomorphic, $f([A_1 \otimes B_1, A_2 \otimes B_2]) = [f(A_1 \otimes B_1), f(A_2 \otimes B_2)]$.

Now we are in a position to state the following proposition:

**Proposition 2.2.** For $A_1 \otimes B_1, A_2 \otimes B_2 \in \mathbb{C}^{2^\ell \times 2^\ell} \otimes \mathbb{C}^{2^m \times 2^m}$, the Lie bracket of the associated vector fields $X_{A_1 \otimes B_1}$ and $X_{A_2 \otimes B_2}$ on $\mathbb{C}^{2^\ell \times 2^\ell}$ is given by

$$[X_{A_1 \otimes B_1}, X_{A_2 \otimes B_2}] = -X_{[A_1 \otimes B_1, A_2 \otimes B_2]}.$$  

This means that the correspondence $A \otimes B \mapsto X_{A \otimes B}$ gives rise to an anti-isomorphism of $(\mathbb{C}^{2^\ell} \otimes \mathbb{C}^{2^m}, [\cdot, \cdot])$ to the Lie algebra $\{X_{A \otimes B} \mid A \in \mathbb{C}^{2^\ell \times 2^\ell}, B \in \mathbb{C}^{2^m \times 2^m}\}$ of linear vector fields on $\mathbb{C}^{2^\ell \times 2^\ell}$.

**Corollary 2.3.** In the two-qubit system with $\ell = m = 1$, one has

$$[X_{i\sigma_j \otimes \sigma_k}, X_{i\sigma_{j'} \otimes \sigma_{k'}}] = -X_{[i\sigma_j \otimes \sigma_k, i\sigma_{j'} \otimes \sigma_{k'}]} = X_{[\sigma_j, \sigma_j'] \otimes \sigma_k \otimes \sigma_{k'}} + X_{\sigma_{j'} \sigma_j \otimes [\sigma_k, \sigma_{k'}]} \quad (2.13)$$

for $j, j', k, k' = 0, 1, 2, 3$. Here the $\sigma_j, j = 1, 2, 3$, denote the Pauli matrices defined as

$$\sigma_1 := \frac{1}{2} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma_2 := \frac{1}{2} \begin{pmatrix} 0 & i \\ -i & 0 \end{pmatrix}, \quad \sigma_3 := \frac{1}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.$$  

**Proof of Proposition 2.2.** Let $\varphi_{A \otimes B}^t$ denote the flow generated by the vector field $X_{A \otimes B}$ on $\mathbb{C}^{2^\ell \times 2^m}$. We identify $(\mathbb{C}^2)^{\otimes n}$ with the space $\mathbb{C}^{2^n}$ of $2^n$-dimensional column vectors, and the isomorphism $\iota : (\mathbb{C}^2)^{\otimes n} \to \mathbb{C}^{2^\ell \times 2^m}$ with an isomorphism of $\mathbb{C}^{2^n}$ to $\mathbb{C}^{2^\ell \times 2^m}$. We note here that the following diagram commutes,

$$\begin{array}{ccc}
\mathbb{C}^{2^\ell \times 2^m} & \xrightarrow{\text{action of } A \otimes B} & \mathbb{C}^{2^\ell \times 2^m} \\
\downarrow \iota^{-1} & & \downarrow \iota^{-1} \\
\mathbb{C}^{2^n} & \xrightarrow{\text{action of } f(A \otimes B)} & \mathbb{C}^{2^n}.
\end{array}$$

This means that

$$(\iota^{-1})^* X_{A \otimes B}(C) = Y_{f(A \otimes B)}(\iota^{-1}(C)) \quad \text{and} \quad \iota^{-1} \circ \varphi_{A \otimes B}^t = e^{t f(A \otimes B)} \circ \iota^{-1}, \quad (2.14)$$

where $Y_{f(A \otimes B)}(c) = f(A \otimes B)c$ with $c \in \mathbb{C}^{2^n}$. 


By the definition of the Lie bracket together with (2.1), we obtain

\[
[X_{A_1 \otimes B_1}, X_{A_2 \otimes B_2}](C)
\]

\[
= \frac{\partial^2}{\partial t \partial s} \bigg|_{t=s=0} \left( \varphi^{-s}_{A_2 \otimes B_2} \circ \varphi^{-t}_{A_1 \otimes B_1} \circ \varphi^{s}_{A_2 \otimes B_2} \circ \varphi^{t}_{A_1 \otimes B_1} \right)(C)
\]

\[
= \frac{\partial^2}{\partial t \partial s} \bigg|_{t=s=0} \left( \iota \circ (e^{-sf(A_2 \otimes B_2)} \circ e^{-tf(A_1 \otimes B_1)} \circ e^{sf(A_2 \otimes B_2)} \circ e^{tf(A_1 \otimes B_1)}) \circ \iota^{-1}(C) \right)
\]

\[
= \iota_\ast [Y_{f(A_1 \otimes B_1)}, Y_{f(A_2 \otimes B_2)}](\iota^{-1}(C)).
\]

Further, by the formula \([Y_{\Xi_1}, Y_{\Xi_2}] = -Y_{[\Xi_1, \Xi_2]}\) for any \(\Xi_1, \Xi_2 \in \mathbb{C}^{2^n \times 2^n}\), one has

\[
[X_{A_1 \otimes B_1}, X_{A_2 \otimes B_2}](C)
\]

\[
= \iota_\ast [Y_{f(A_1 \otimes B_1)}, Y_{f(A_2 \otimes B_2)}](\iota^{-1}(C)) = \iota_\ast (-Y_{[f(A_1 \otimes B_1), f(A_2 \otimes B_2)]})(\iota^{-1}(C))
\]

\[
= -\iota_\ast Y_{f([A_1 \otimes B_1, A_2 \otimes B_2])}(\iota^{-1}(C)) = -X_{[A_1 \otimes B_1, A_2 \otimes B_2]}(C).
\]

Since the map \(A \otimes B \mapsto Y_{f(A \otimes B)}\) is bijective to the space of linear vector fields on \(\mathbb{C}^{2^n}\), so is the map \(A \otimes B \mapsto X_{A \otimes B}\) to the space of linear vector fields on \(\mathbb{C}^{2^n \times 2^m}\). This ends the proof. \(\square\)

We now apply (2.13) for the two-qubit system. We take, for instance, \(A_1 \otimes B_1 = i\sigma_3 \otimes \sigma_3\) and \(A_2 \otimes B_2 = i\sigma_1 \otimes I\). By Corollary 2.3, the Lie bracket of the associated linear vector fields is given by \([X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_1 \otimes I}] = X_{[\sigma_3, \sigma_1] \otimes \sigma_3} + X_{\sigma_1 \sigma_3 \otimes [\sigma_3, I]} = -X_{i\sigma_2 \otimes \sigma_3}\). Other Lie brackets will be found in Appendix A.

### 3 Controls in NMR systems

Sections 3 and 4 deal with control problems in \(n\)- and 2-qubit systems, respectively. In this paper, we adopt a nuclear magnetic resonance (NMR) system as a quantum computation model from \(n\)-qubit systems [16].

The total Hamiltonian operator \(\hat{H}\) of the NMR system is the sum of two operators,

\[
\hat{H} := \hat{H}_d + \hat{H}_c,
\]

where \(\hat{H}_d\) and \(\hat{H}_c\) are called the drift and control Hamiltonians, and expressed as

\[
\hat{H}_d := \sum_{1 \leq \alpha < \beta \leq n} J_{\alpha \beta} \sigma_{3}^{(\alpha, n)} \sigma_{3}^{(\beta, n)}, \quad \text{(3.1a)}
\]

\[
\hat{H}_c := \sum_{\alpha=1}^{n} \left( v_1^{(\alpha)} \sigma_{1}^{(\alpha, n)} + v_2^{(\alpha)} \sigma_{2}^{(\alpha, n)} \right), \quad \text{(3.1b)}
\]
respectively. Here, $J_{\alpha\beta} \in \mathbb{R}$ are coupling constants determining the strength of the interactions between the $\alpha$-th and $\beta$-th qubits. The $v_1^{(\alpha)}$, $v_2^{(\alpha)} : \mathbb{R} \to \mathbb{R}$ are (time-dependent) controls acting on the $\alpha$-th qubit. The $\sigma_j^{(\alpha,n)}$ are defined as

$$\sigma_j^{(\alpha,n)} := I \otimes \cdots \otimes I \otimes \sigma_j \otimes I \otimes \cdots \otimes I, \quad (3.2)$$

for $j = 1, 2, 3$, and $\alpha = 1, \cdots, n$, with $I$ the $2 \times 2$ identity matrix. The controls $v_1^{(\alpha)}$ and $v_2^{(\alpha)}$ are assumed to be piecewise constant functions in $t$.

The Schrödinger equation for the NMR system is expressed as

$$\frac{d}{dt} |\Psi\rangle = -i\hat{H}|\Psi\rangle, \quad (3.3)$$

where the natural unit system has been adopted, so that the Plank constant $\hbar$ is equal to one. Since we are interested in bipartite entanglement, we have to put the Schrödinger equation in the form of matrix equation. According to the isomorphism $(\mathbb{C}^2)^\otimes n \cong \mathbb{C}^{2\ell \times 2m}$ with $n = \ell + m$, the vector fields associated with the drift and control Hamiltonians are expressed, respectively, as

$$-iH_d(C) = -i \sum_{1 \leq \alpha < \beta \leq \ell} J_{\alpha\beta} [\sigma_3^{(\alpha,\ell)} \sigma_3^{(\beta,\ell)}] C - i \sum_{\ell+1 \leq \alpha < \beta \leq \ell+m} J_{\alpha\beta} C [\sigma_3^{(\alpha,m)} \sigma_3^{(\beta,m)}]^T$$

$$-i \sum_{\ell+1 \leq \alpha < \beta \leq \ell+m} J_{\alpha\beta} [\sigma_3^{(\alpha,\ell)}] C [\sigma_3^{(\beta-m,\ell)}]^T, \quad (3.4a)$$

$$-iH_c(C) = \sum_{1 \leq \alpha \leq \ell} [I \otimes (\alpha-1) \otimes \xi_{\alpha} \otimes I \otimes (\ell-\alpha)] C + \sum_{\ell+1 \leq \beta \leq \ell+m} C [I \otimes (\ell-1) \otimes \xi_{\beta} \otimes I \otimes (m-\beta+\ell)]^T, \quad (3.4b)$$

where

$$\xi_{\alpha} = -i(v_1^{(\alpha)} \sigma_1 + v_2^{(\alpha)} \sigma_2),$$

and where the symbols with square brackets like $[\sigma_3^{(\alpha,\ell)}]$ denote the Kronecker products of matrices corresponding to the tensor products concerned. The Schrödinger equation on $M$ is then put in the form

$$\frac{dC}{dt} = -iH_d(C) - iH_c(C). \quad (3.5)$$

As is easily verified, $-iH_d$ and $-iH_c$ are vector fields on $M$. In what follows, we will refer to $-iH_d(C)$ and $-iH_c(C)$ as the drift and control vector fields, respectively.
3.1 A solution to the NMR system

We show that Eq. (3.5) can be solved with the assumption that the coupling constants are small enough and that the controls are constant. In view of (3.4), we may put the drift and control vector fields in the form,

\[-iH_d(C) = -i \sum_{k=1}^{N} \varepsilon_k \Sigma_1^{(k)} C (\Sigma_2^{(k)})^T,\]
\[-iH_c(C) = \xi_1 C + C \xi_2^T,\]

respectively, where \(\varepsilon_k, k = 1, \cdots, N\), are small parameters, where \(\Sigma_1^{(k)} \in \mathbb{C}^{2^t \times 2^m}, \Sigma_2^{(k)} \in \mathbb{C}^{2^t \times 2^m}, k = 1, \cdots, N\), are constant Hermitian matrices, and where \(\xi_1 \in \mathfrak{su}(2^t), \xi_2 \in \mathfrak{su}(2^m)\) are constant controls. The Schrödinger equation we are to solve is

\[
\dot{C} = -i \sum_{k=1}^{N} \varepsilon_k \Sigma_1^{(k)} C (\Sigma_2^{(k)})^T + \xi_1 C + C \xi_2^T
\]
\[
= - \sum_{k=1}^{N} \varepsilon_k X_{\Sigma_1^{(k)} \otimes \Sigma_2^{(k)}}(C) + X_{\Sigma_1 \otimes I_{2^m}}(C) + X_{I_{2^t} \otimes \xi_2}(C). \tag{3.7}
\]

Since the coupling constants \(\varepsilon_k\) are sufficiently small, we may suppose that the solution \(C(t)\) to (3.7) can be expanded into a power series in \(\varepsilon_k, k = 1, \cdots, N\),

\[
C(t) = \sum_{n=(n_1, \cdots, n_N) \in (\mathbb{Z}_{\geq 0})^N} \varepsilon^n C_n(t) \quad \text{with} \quad \varepsilon^n := \varepsilon_1^{n_1} \cdots \varepsilon_N^{n_N}.
\]

By substituting this expansion into (3.7) and comparing the left and right hand-sides, we obtain the series of differential equations

\[
\dot{C}_0 = \xi_1 C_0 + C_0 \xi_2^T,
\]
\[
\dot{C}_n = -i \sum_{k=1, \cdots, N, n_k \neq 0} \Sigma_1^{(k)} C_{n-e_k} (\Sigma_2^{(k)})^T + \xi_1 C_n + C_n \xi_2^T \quad \text{for} \quad n \neq 0, \tag{3.8}
\]

where \(e_1 = (1, 0, \cdots, 0), \cdots, e_N = (0, \cdots, 0, 1) \in (\mathbb{Z}_{\geq 0})^N\).

To solve the above equations, we introduce new unknown functions by \(Q_n(t) = (e^{-t\xi_1} \otimes e^{-t\xi_2}) \cdot C_n(t)\). Then \(Q_n(t)\) with \(n \in (\mathbb{Z}_{\geq 0})^N\) prove to satisfy the equations

\[
\dot{Q}_0 = 0,
\]
\[
\dot{Q}_n = -i \sum_{k=1, \cdots, N, n_k \neq 0} \left( e^{-t\xi_1} \Sigma_1^{(k)} e^{t\xi_1} \right) Q_{n-e_k} \left( e^{-t\xi_2} \Sigma_2^{(k)} e^{t\xi_2} \right)^T
\]
\[
= -i \sum_{k=1, \cdots, N, n_k \neq 0} \text{Ad}_{e^{-t\xi_1} \otimes e^{-t\xi_2}} (\Sigma_1^{(k)} \otimes \Sigma_2^{(k)}) Q_{n-e_k} \quad \text{for} \quad n \neq 0.
\]
These equations for \( Q_n \) are inductively integrable. The first \((N+1)\) of solutions are given by \( Q_0(t) = Q_0(0) \) and

\[
Q_{e_k}(t) = Q_{e_k}(0) - i \int_0^t \Ad_{e^{-s \xi_1} e^{-s \xi_2}} (\Sigma_1^{(k)} \otimes \Sigma_2^{(k)}) Q_0(0) \, ds, \quad k = 1, \ldots, N.
\]

In order to write down solutions in a compact form, we introduce \( N \) linear operators

\[
T_k : C^\infty(\mathbb{R}; \mathbb{C}^{2^m}) \to C^\infty(\mathbb{R}; \mathbb{C}^{2^m}), \quad k = 1, \ldots, N,
\]

defined by

\[
(T_k F)(t) := -i \int_0^t \Ad_{e^{-s \xi_1} e^{-s \xi_2}} (\Sigma_1^{(k)} \otimes \Sigma_2^{(k)}) F(s) \, ds, \quad F \in C^\infty(\mathbb{R}; \mathbb{C}^{2^m}).
\]

Then, in terms of these operators, \( Q_{e_k}(t), k = 1, \ldots, N \), take the form

\[
Q_{e_k}(t) = Q_{e_k}(0) + (T_k Q_0(0))(t), \quad k = 1, \ldots, N,
\]

where \( Q_0(0) \) in the right-hand side is viewed as a constant function.

We proceed to, say, \( Q_{e_1+e_2}(t) \). The equation for \( Q_{e_1+e_2}(t) \) is expressed as

\[
\dot{Q}_{e_1+e_2} = -i \Ad_{e^{-t \xi_1} e^{-t \xi_2}} (\Sigma_1^{(2)} \otimes \Sigma_2^{(2)}) Q_{e_1} - i \Ad_{e^{-t \xi_1} e^{-t \xi_2}} (\Sigma_1^{(1)} \otimes \Sigma_2^{(1)}) Q_{e_2},
\]

and integrated to yield

\[
Q_{e_1+e_2}(t) = Q_{e_1+e_2}(0) + (T_2 Q_{e_1})(t) + (T_1 Q_{e_2})(t)
\]

\[
= Q_{e_1+e_2}(0) + (T_2 Q_{e_1}(0))(t) + (T_1 Q_{e_2}(0))(t) + (T_2 \circ T_1 Q_0(0))(t) + (T_1 \circ T_2 Q_0(0))(t).
\]

In a similar manner, the \( Q_n(t) \) is inductively integrated and expressed as

\[
Q_n(t) = \sum_{m=(m_1, \ldots, m_N) \in (\mathbb{Z}_{\geq 0})^N} \sum_{m_k \leq n_k, k=1, \ldots, N} \left( T_K Q_{n-m}(0) \right)(t),
\]

where \( |m| := m_1 + \cdots + m_N \), and where \( T_K : C^\infty(\mathbb{R}; \mathbb{C}^{2^m}) \to C^\infty(\mathbb{R}; \mathbb{C}^{2^m}) \) are defined to be

\[
T_K := T_{k_1} \circ \cdots \circ T_{k_{|m|}}, \quad K = (k_1, \ldots, k_{|m|}) \in \{1, \ldots, N\}^{|m|}.
\]

Hence, we have obtained the following.

**Proposition 3.1.** The solution \( C(t) \) to (3.7) is put in the form of power series in \( \varepsilon_k \), \( k = 1, \ldots, N \),

\[
C(t) = (e^{t \xi_1} \otimes e^{t \xi_2}) \sum_n \sum_m \sum_K \varepsilon^n (T_K Q_{n-m}(0))(t)
\]

\[
= (e^{t \xi_1} \otimes e^{t \xi_2}) \sum_{m \in (\mathbb{Z}_{\geq 0})^N} \sum_{K \in \{1, \ldots, N\}^{|m|}, \# \{ \nu | k_{\nu} = k \} = m_k, k=1, \ldots, N} \varepsilon^m (T_K C(0))(t),
\]

where we have used the fact that \( C(0) = \sum_n \varepsilon^n C_n(0) = \sum_n \varepsilon^n Q_n(0) \).

If the control is piecewise constant in \( t \), the continuation along time of solutions of the form (3.10) with respective constant controls will yield a solution to (3.7).
3.2 Controllability of the NMR systems

From Eq. (3.6), we observe that the control vector field are vertical, so that the measure defined in (2.3) does not change along this vector field. Hence, the control would not make the NMR system entangled without the coupling with the drift vector field. With this observation in mind, we gain insight into (3.10) to understand how the coupling occurs. We first look into the operators $T_k$ defined in (3.9). Writing out the integrand of (3.9) in the form of a power series in $s$, we obtain

\[
(T_k F)(t) = -i \int_0^t \left\{ \sum_{i=1}^k F(s)(\Sigma_2^{(k)})^T - s\left( [\xi_1, \Sigma_1^{(k)}] F(s) (\Sigma_2^{(k)})^T + \Sigma_1^{(k)} F(s) [\xi_2, \Sigma_2^{(k)}]^T \right) \\
+ \frac{s^2}{2!} \left( [\xi_1, [\xi_1, \Sigma_1^{(k)}]] F(s) (\Sigma_2^{(k)})^T + 2[\xi_1, \Sigma_1^{(k)}] F(s) [\xi_2, \Sigma_2^{(k)}]^T \right) \\
+ \Sigma_1^{(k)} F(s) [\xi_2, [\xi_2, \Sigma_2^{(k)}]^T] + \cdots \right\} ds.
\]

By applying Proposition 2.2, we can put the above equation in the form

\[
(T_k F)(t) = \int_0^t \left\{ -X_{i \Sigma_1^{(k)} \otimes \Sigma_2^{(k)}} (F(s)) + s \left( [X_{\xi_1 \otimes I}, -X_{i \Sigma_1^{(k)} \otimes \Sigma_2^{(k)}}] (F(s)) + [X_{I \otimes \xi_2}, -X_{i \Sigma_1^{(k)} \otimes \Sigma_2^{(k)}}] (F(s)) \right) \\
+ \frac{s^2}{2!} \left( [X_{\xi_1 \otimes I}, [X_{\xi_1 \otimes I}, -X_{i \Sigma_1^{(k)} \otimes \Sigma_2^{(k)}}]] (F(s)) + 2[X_{\xi_1 \otimes I}, [X_{I \otimes \xi_2}, -X_{i \Sigma_1^{(k)} \otimes \Sigma_2^{(k)}}]] (F(s)) \\
+ [X_{I \otimes \xi_2}, [X_{I \otimes \xi_2}, -X_{i \Sigma_1^{(k)} \otimes \Sigma_2^{(k)}}]] (F(s)) \right) + \cdots \right\} ds. \tag{3.11}
\]

Since the expansion (3.11) means that the drift and control vector fields are coupled indeed, and since the operator $T_k$ is the composition of these $T_k$, Eq. (3.10) shows that the drift and control vector fields work together to give rise to the solution. If the coupling between the drift and control vector fields generates vector fields with non-vanishing horizontal components, the NMR system will get more entangled by controls.

Incidentally, if the NMR system is controllable, then any initial state can be transferred into, say, a maximally entangled state by some controls. This means that the control vector field should be coupled with the drift vector field in the case of controllable NMR systems.

According to [2], the system (3.3) is said to be pure state controllable if, for any initial and final states, there are controls $\xi_\alpha, \alpha = 1, \ldots, n$, and a finite time $T > 0$ such that the initial state is transferred into the final one on the finite time $T$. Further, the system (3.3) is pure state controllable if and only if the Lie algebra $L_H$ generated
by \( \{-i\hat{H}_d\} \cup \{i\sigma_j^{(\alpha,n)}\}_{j=1}^{\alpha=1,\ldots,n} \) is isomorphic to \( \text{su}(2^n) \). For the time being, we are interested in the coupling between the drift and control vector fields, and postpone the controllability problem to Appendix B, in which we will give a necessary and sufficient condition for controllability.

However, it is of great help to apply the controllability theorem to the NMR system. To state the theorem, we need to introduce the notion of spin graph. Here, the spin graph associated with a given drift Hamiltonian \( \hat{H}_d \) is defined to be the un-oriented graph that has \( n \) nodes and edges joining the nodes labeled as \( \alpha \) and \( \beta \) such that \( J_{\alpha\beta} \neq 0 \), where nodes and edges represent spin-\( \frac{1}{2} \) particles and interactions among spin-\( \frac{1}{2} \) particles such that \( J_{\alpha\beta} \neq 0 \), respectively. In terms of the spin graph, the controllability theorem is stated as follows (see Theorem B.8): The NMR system is controllable, if and only if the associated spin graph is connected. In view of this, we may set some of \( J_{\alpha\beta} \) in (3.4) to be zero. We here take

\[
\begin{align*}
J_{1\beta} &\neq 0, & \ell + 1 \leq \beta \leq \ell + m, \\
J_{\alpha,\ell+1} &\neq 0, & 1 \leq \alpha \leq \ell, \\
J_{\alpha\beta} &= 0, & \text{otherwise}
\end{align*}
\]

Then, the associated spin graph is connected, so that the NMR system with these coupling constants is controllable. The drift vector field in (3.4) is now expressed as

\[
-i\hat{H}_d(C) = -i[\sigma_3^{(1,\ell)}]C\left( \sum_{\ell+1 \leq \beta \leq \ell + m} J_{1\beta}[\sigma_3^{(\beta - \ell, m)}]^T \right) - i\left( \sum_{2 \leq \alpha \leq \ell} J_{\alpha,\ell+1}[\sigma_3^{(\alpha, \ell)}]\right)C[\sigma_3^{(1,m)}]^T.
\]

In this case, one has \( N = 2 \), and

\[
\Sigma_1^{(1)} = [\sigma_3^{(1,\ell)}], \quad \Sigma_2^{(1)} = \sum_{\ell+1 \leq \beta \leq \ell + m} J_{1\beta}[\sigma_3^{(\beta - \ell, m)}]^T, \quad \varepsilon_1 J_{1\beta}^{(1)} = J_{1\beta},
\]

\[
\Sigma_1^{(2)} = \sum_{2 \leq \alpha \leq \ell} J_{\alpha}^{(2)}[\sigma_3^{(\alpha, \ell)}], \quad \Sigma_2^{(2)} = [\sigma_3^{(1,m)}]^T, \quad \varepsilon_2 J_{\alpha}^{(2)} = J_{\alpha,\ell+1},
\]

\[
\xi_1 = \sum_{1 \leq \alpha \leq \ell} [I^{\otimes (\alpha-1)} \otimes \xi_\alpha \otimes I^{\otimes (\ell-\alpha)}], \quad \xi_2 = \sum_{\ell+1 \leq \beta \leq \ell + m} [I^{\otimes (\beta-\ell-1)} \otimes \xi_\beta \otimes I^{\otimes (m-\beta+\ell)}]^T,
\]

where \( \xi_\alpha, \xi_\beta \in \text{span}_R\{i\sigma_1, i\sigma_2\} \).

### 4 Controls in two-qubit NMR systems

In this section, we deal with the two-qubit case. Our system is expressed as

\[
\frac{dC}{dt} = -JX_{i\sigma_3 \otimes \sigma_3} + X_{\xi_1 \otimes I} + X_{I \otimes \xi_2} = -iJ\sigma_3 C\sigma_3^T + \xi_1 C + C\xi_2^T,
\]

(4.1)
where \( J > 0 \) is the coupling constant between the two qubits, and where \( \xi_1, \xi_2 \) are constant controls taking values in \( \text{span}_R \{ i\sigma_1, i\sigma_2 \} \). In comparison to (3.7), this equation is easier to solve, and we can investigate solutions in detail to understand how controls make the NMR system more entangled.

4.1 Two-qubit NMR systems without control

To understand why one needs controls to make the NMR systems entangled, we look into the uncontrolled system with \( \xi_1 = \xi_2 = 0 \) in (4.1). If \( \xi_1 = \xi_2 = 0 \), Eq. (4.1) is easily integrated to give the solution

\[ C(t) = \varphi_{-i\sigma_3}^J (C(0)) = \begin{pmatrix} e^{-iJt/4} c_{00}(0) & e^{iJt/4} c_{01}(0) \\ e^{iJt/4} c_{10}(0) & e^{-iJt/4} c_{11}(0) \end{pmatrix}. \] (4.2)

Then the measure \( F(C(t)) = \det(C(t)^* C(t)) \) is evaluated as

\[ F(C(t)) = |c_{00}(0)c_{11}(0)|^2 + |c_{01}(0)c_{10}(0)|^2 - \left( e^{-iJt} c_{00}(0)c_{01}(0)c_{10}(0)c_{11}(0) + e^{iJt} c_{00}(0)c_{01}(0)c_{10}(0)c_{11}(0) \right), \]

which gives rise to the inequality

\[ F(C(t)) \leq (|c_{00}(0)c_{11}(0)| + |c_{01}(0)c_{01}(0)|)^2 =: D(C(0)). \] (4.3)

Eq. (4.3) implies that the quantity \( D(C(0)) \) determines whether the state driven by (4.1) with the initial state \( C(0) \) reaches a maximally entangled state without any controls or not.

Let us be reminded of the fact that the measure \( F \) ranges over \( 0 \leq F(C) \leq 1/4 \) for the two-qubit, where the minimum or the maximum occurs, according to whether \( C \) is separable or maximally entangled. If \( D(C(0)) = 1/4 \), the state \( C(t) \) evolves to reach a maximally entangled state without control even if \( C(0) \) is separable. For instance, starting with the separable state

\[ C(0) = \frac{1}{2} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix}, \]

the solution curve \( C(t) \) passes through the maximally entangled state at \( t = \pi/J \),

\[ C \left( \frac{\pi}{J} \right) = \frac{1}{2} \begin{pmatrix} e^{-i\pi/4} & e^{i\pi/4} \\ e^{i\pi/4} & e^{-i\pi/4} \end{pmatrix}. \]
We note in addition that \( F(C(t)) = \frac{1 - \cos Jt}{8} \) in this case. In contrast with this, if \( D(C(0)) = 0 \), then \( C(0) \) is separable, and the state \( C(t) \) is always separable because of \( 0 \leq F(C(t)) \leq D(C(0)) = 0 \). An example of such an initial state is

\[
C(0) = \text{diag}(e^{i\theta}, 0). \tag{4.5}
\]

This initial state never gets entangled without controls. From these facts, we observe that the separable states \( \frac{1}{2}(|0\rangle + |1\rangle) \otimes (|0\rangle + |1\rangle) \) and \( e^{i\theta}|0\rangle \otimes |0\rangle \) are of different nature with respect to the drift Hamiltonian.

### 4.2 Behaviours of solutions in the two-qubit NMR system

We turn to the controlled system (4.1) with \( \xi_1 \) and \( \xi_2 \) non-vanishing constants. Since the associated spin graph is connected, the two-qubit NMR system is controllable, so that any separable state can get entangled. This implies that the coupling between the drift and control vector fields must occur.

We solve the Schrödinger equation (4.1) as a power series of the coupling constant \( J > 0 \). As in the previous section, we assume that the \( J \) is small enough, and that the solution \( C(t) \) of (4.1) can be expanded into a power series in \( J \) as

\[
C(t) = \sum_{n=0}^{\infty} J^n C_n(t). \tag{4.6}
\]

These equations can be integrated inductively, like (3.8). In fact, from the above equations, differential equations for \( e^{-t\xi_1} C_n(t)e^{-t\xi_2} \) are easily obtained and integrated to give

\[
e^{-t\xi_1} C(t)e^{-t\xi_2} = \sum_{n=0}^{\infty} J^n e^{-t\xi_1} C_n(t) e^{-t\xi_2} = \sum_{n=0}^{\infty} (-iJ)^n \int_0^t ds_n \text{Ad}_{e^{-s_n \xi_1}}(\sigma_3) \int_0^{s_n} ds_{n-1} \text{Ad}_{e^{-s_{n-1} \xi_1}}(\sigma_3) \cdots \int_0^{s_2} ds_1 \text{Ad}_{e^{-s_2 \xi_1}}(\sigma_3) \times C(0) \text{Ad}_{e^{s_2 \xi_2}}(\sigma_3^T) \cdots \text{Ad}_{e^{s_{n-1} \xi_2}}(\sigma_3^T) \text{Ad}_{e^{s_n \xi_2}}(\sigma_3^T), \tag{4.7}
\]

where we have used the assumption that \( C(0) = \sum_{n=0}^{\infty} J^n C_n(0) \). The following proposition is easy to prove.

**Proposition 4.1.** The power series (4.7) uniformly converges to a function \( P(t) \), with which the solution of the NMR system (4.1) is given by \( C(t) = e^{t\xi_1} P(t)e^{t\xi_2} \).
So far we have found the solution as the power series \((4.7)\) in the coupling constant \(J\). We now analyze the solution in order to see in detail that the coupling between the drift and control vectors actually occurs to make the NMR system more entangled. We denote the \(n\)-th order term in \(J\) of \((4.7)\) by \(P_n(t)\). Then, the solution is expressed as
\[
C(t) = \sum_{n=0}^{\infty} J^n e^{i\xi_1 \phi} P_n(t) e^{it^2 T}.
\]
This explains how the controls \(\xi_1\) and \(\xi_2\) contribute to entanglement promotion of the system. The zeroth-order term does not make the system more entangled because of \(F(e^{it\xi_1 \phi} P_0(0) e^{it^2 T}) = F(C(0))\). Contrarily, the higher-order terms in \(J\) are expected to make the NMR system entangled in general. We look into \((4.7)\) in detail. The first-order term \(P_1(t)\) of \((4.7)\) is put in the form \((T_1 C(0))(t)\).

Then, as is seen from \((3.11)\), \(P_1(t)\) is expressed as
\[
P_1(t) = - t X_{\sigma_3 \otimes \sigma_3} (C(0)) + \frac{t^2}{2!} \left( [X_{\xi_1 \otimes I}, -X_{\sigma_3 \otimes \sigma_3}] + [X_{I \otimes \xi_2}, -X_{\sigma_3 \otimes \sigma_3}] \right) \big|_{C(0)}
+ \frac{t^3}{3!} \left( [X_{\xi_1 \otimes I}, [X_{\xi_1 \otimes I}, -X_{\sigma_3 \otimes \sigma_3}]] + [X_{\xi_1 \otimes I}, [X_{I \otimes \xi_2}, -X_{\sigma_3 \otimes \sigma_3}]]
+ [X_{I \otimes \xi_2}, [X_{\xi_1 \otimes I}, -X_{\sigma_3 \otimes \sigma_3}]] + [X_{I \otimes \xi_2}, [X_{I \otimes \xi_2}, -X_{\sigma_3 \otimes \sigma_3}]] \right) \big|_{C(0)} + \cdots.
\]

This shows that the drift and control vector fields are coupled indeed. We note that \([X_{I \otimes \xi_2}, [X_{\xi_1 \otimes I}, -X_{\sigma_3 \otimes \sigma_3}] = [X_{\xi_1 \otimes I}, [X_{I \otimes \xi_2}, -X_{\sigma_3 \otimes \sigma_3}]\] by the Jacobi identity and that \([X_{\xi_1 \otimes I}, X_{I \otimes \xi_2}] = 0\).

By a similar computation, the second-order term \(P_2(t)\) of \((4.7)\) is written out as
\[
P_2(t) = (T_1 \circ T_1 C(0))(t)
= \frac{t^2}{2!} (-i\sigma_3 (-i\sigma_3 C(0)\sigma_3^T)\sigma_3^T) - \frac{t^3}{3!} \sigma_3 ([\xi_1, \sigma_3] C(0)\sigma_3^T + \sigma_3 C(0)[\xi_2, \sigma_3] \sigma_3^T) \sigma_3^T + \cdots.
\]
We formally denote the first and second terms of the right-hand side of the above equation by \(\frac{t^2}{2!} (-X_{\sigma_3 \otimes \sigma_3})^2 \big|_{C(0)}\) and \(\frac{t^3}{3!} (-X_{\sigma_3 \otimes \sigma_3}) \big([X_{\xi_1 \otimes I}, -X_{\sigma_3 \otimes \sigma_3}] + [X_{I \otimes \xi_2}, -X_{\sigma_3 \otimes \sigma_3}]\big) \big|_{C(0)}\), respectively. The first term \(\frac{t^2}{2!} (-X_{\sigma_3 \otimes \sigma_3})^2\) comes from the Taylor expansion of the flow \(\phi_{-\sigma_3 \otimes \sigma_3}\) with respect to \(t\).

It is inductively shown that the \(n\)-th order term \(P_n(t)\) is expanded into a power series of \(t\) with the lowest-order term \(\frac{t^n}{n!} (-X_{\sigma_3 \otimes \sigma_3})^n\), which comes from the Taylor expansion \(\phi_{-\sigma_3 \otimes \sigma_3}(C(0)) = \sum_{n=0}^{\infty} \frac{t^n}{n!} (-X_{\sigma_3 \otimes \sigma_3})^n \big|_{C(0)}\). Hence, we have

**Proposition 4.2.** If \(t\) is sufficiently small, the solution \(C(t)\) of \((4.1)\) allows of the
approximation of the form

\[ e^{-i\xi_1 C(t)} e^{-i\xi_2 T} - \varphi_{-i\sigma_3 \otimes \sigma_3}^J (C(0)) \]

\[ \sim \frac{J t^2}{2!} \left( [X_{\xi_1 \otimes I}, -X_{i\sigma_3 \otimes \sigma_3}] + [I \otimes X_{\xi_2}, -X_{i\sigma_3 \otimes \sigma_3}] \right) \bigg|_{C(0)} \\
+ \frac{J^2 t^3}{3!} (-X_{i\sigma_3 \otimes \sigma_3}) \left( [X_{\xi_1 \otimes I}, -X_{i\sigma_3 \otimes \sigma_3}] + [I \otimes X_{\xi_2}, -X_{i\sigma_3 \otimes \sigma_3}] \right) \bigg|_{C(0)} \\
+ \frac{J^3 t^3}{3!} \left( [X_{\xi_1 \otimes I}, [X_{\xi_1 \otimes I}, -X_{i\sigma_3 \otimes \sigma_3}]] + [X_{\xi_1 \otimes I}, [I \otimes X_{\xi_2}, -X_{i\sigma_3 \otimes \sigma_3}]] + [X_{I \otimes \xi_2}, [X_{\xi_1 \otimes I}, -X_{i\sigma_3 \otimes \sigma_3}]] + [X_{I \otimes \xi_2}, [I \otimes X_{\xi_2}, -X_{i\sigma_3 \otimes \sigma_3}]] \right) \bigg|_{C(0)} + \cdots , \quad (4.9) \]

where \( \varphi_{-i\sigma_3 \otimes \sigma_3}^J (C(0)) \) is the solution of the uncontrolled system \( dC/dt = -J X_{i\sigma_3 \otimes \sigma_3} \) with the initial state \( C(0) \).

### 4.3 Bases of vertical and horizontal subspaces

So far we have shown that the drift and control vector fields are coupled. We now wish to show that the coupling generates vector fields with non-vanishing horizontal components. To this end, we have to look into vertical and horizontal vector fields in detail.

Since the subspaces \( V_C \) and \( H_C \) depend on the stratum to which \( C \) belongs, we make a brief review of a stratification of the state space \( M \) for the two-qubit system after [12]. According to singular values \( \lambda_1(C) \geq \lambda_2(C) \) of \( C \in M \), the state space \( M \) of the two-qubit system is stratified into three \( G \)-invariant strata as

\[
M = M_0 \sqcup M_1 \sqcup M_2,
\]

where

\[
M_0 = \{ C \in M \mid \lambda_1(C) = 1, \lambda_2(C) = 0 \},
\]

\[
M_1 = \{ C \in M \mid \lambda_1(C) > \lambda_2(C) > 0 \}, \quad \text{(4.10)}
\]

\[
M_2 = \{ C \in M \mid \lambda_1(C) = \lambda_2(C) = 1/\sqrt{2} \}.
\]

The strata \( M_0 \) and \( M_2 \) are the sets of separable and maximally entangled states, respectively.

We now deals with bases of the subspaces \( V_C \) and \( H_C \). We take up a typical matrix \( \Lambda = \text{diag} (\lambda_1, \lambda_2) \) with \( \lambda_1 \geq \lambda_2 \), to which all the matrices with the same singular values are translated by the \( U(2) \otimes U(2) \) action. Let us start with the vertical subspace \( V_\Lambda \).

Proposition 2.1 with (4.10) implies that

\[
\dim V_\Lambda = \begin{cases} 
6, & \text{if } \Lambda \in M_1, \\
5, & \text{if } \Lambda \in M_0, \\
4, & \text{if } \Lambda \in M_2. 
\end{cases} \quad \text{(4.11)}
\]
Tangent vectors $X_{iI} (\Lambda), X_{i\sigma_j I} (\Lambda)$ and $X_{iI} (\Lambda)$ for $j = 1, 2, 3$, defined in (2.10), span the tangent space $V_\Lambda$. However, some of these vectors coincide with one another,

1. $X_{i\sigma_3 I} (\Lambda) = X_{iI} (\Lambda)$ at any diagonal matrix $\Lambda$,
2. $X_{i\sigma_3 I} (\Lambda) = X_{iI} (\Lambda) = X_{iI} (\Lambda)$ at $\Lambda = \text{diag} (1, 0) \in M_0$,
3. $X_{i\sigma_1 I} (\Lambda) = X_{iI} (\Lambda)$ and $X_{i\sigma_2 I} (\Lambda) = -X_{iI} (\Lambda)$, at $\Lambda = I / \sqrt{2} \in M_2$.

Thus, respective bases of $V_\Lambda$ are obtained as follows:

**Proposition 4.3.** Let $\Lambda = \text{diag} (\lambda_1, \lambda_2)$.

(i) If $\Lambda$ lies in the principal stratum $M_1$, then $V_\Lambda$ has the basis $X_{iI} (\Lambda), X_{i\sigma_3 I} (\Lambda), X_{i\sigma_1 I} (\Lambda), X_{i\sigma_2 I} (\Lambda), X_{iI} (\Lambda), X_{iI} (\Lambda)$.

(ii) If $\Lambda$ is a separable state, i.e., $\Lambda = \text{diag} (1, 0) \in M_0$, then $V_\Lambda$ has the basis $X_{iI} (\Lambda), X_{i\sigma_3 I} (\Lambda), X_{i\sigma_1 I} (\Lambda), X_{i\sigma_2 I} (\Lambda), X_{iI} (\Lambda)$.

(iii) If $\Lambda$ is maximally entangled, i.e., $\Lambda = I / \sqrt{2} \in M_2$, then $V_\Lambda$ has the basis $X_{iI} (\Lambda), X_{i\sigma_3 I} (\Lambda), X_{i\sigma_1 I} (\Lambda), X_{i\sigma_2 I} (\Lambda), X_{iI} (\Lambda)$.

Since the $G$-action (2.4) is isometric, the singular value decomposition (2.5), viewed as a map, $\Lambda \mapsto C = (g \otimes h) \cdot \Lambda$, gives rise to a basis of $V_C$ from that of $V_\Lambda$ by the differential map $(g \otimes h)_*$.

The next task is to find a basis of the horizontal subspace $H_C$. From (4.11), the dimension of $H_C$ proves to be

$$\dim H_C = \begin{cases} 
1, & \text{if } C \in M_1, \\
2, & \text{if } C \in M_0, \\
3, & \text{if } C \in M_2.
\end{cases} \quad (4.12)$$

The following proposition is easily verified by a straightforward computation.

**Proposition 4.4.** Let $\Lambda = \text{diag} (\lambda_1, \lambda_2)$.

(i) If $\Lambda$ lies in the principal stratum $M_1$, then the horizontal subspace $H_\Lambda$ is a one-dimensional vector space spanned by $X_{i\sigma_1 \sigma_2} (\Lambda) = X_{i\sigma_2 \sigma_1} (\Lambda)$.

(ii) If $\Lambda$ is separable, i.e., $\Lambda \in M_0$, or $\Lambda = \text{diag} (1, 0)$, then $H_\Lambda$ has the basis $X_{i\sigma_1 \sigma_2} (\Lambda), X_{i\sigma_1 \sigma_2} (\Lambda) = X_{i\sigma_2 \sigma_1} (\Lambda)$.
(iii) If $\Lambda$ is maximally entangled, i.e., $\Lambda \in M_2$, or $\Lambda = I/\sqrt{2}$, then $H_\Lambda$ has the basis

$$iX_{i\sigma_1 \otimes I}(\Lambda), iX_{i\sigma_2 \otimes I}(\Lambda), iX_{i\sigma_3 \otimes I}(\Lambda).$$

A basis of $V_C$ are formed from that of $V_\Lambda$ by the differential map $(g \otimes h)_*$. 

From these propositions, we observe that when $\Lambda = I/\sqrt{2} \in M_2 \cong U(2)$, the vertical subspace $V_\Lambda$ is identified with the space $u(2)$ of $2 \times 2$ skew Hermitian matrices, and that the horizontal subspace $H_\Lambda$ with $\Lambda = I/\sqrt{2}$ is identified with the space $i\mathfrak{su}(2)$ of traceless Hermitian matrices. Further, the basis vectors of $H_\Lambda$ given in Proposition 4.3 (iii) are alternatively expressed as

$$iX_{i\sigma_1 \otimes I}(\frac{1}{\sqrt{2}} I) = iX_{iI \otimes \sigma_1}(\frac{1}{\sqrt{2}} I) = 2X_{i\sigma_3 \otimes \sigma_2}(\frac{1}{\sqrt{2}} I) = 2X_{i\sigma_2 \otimes \sigma_3}(\frac{1}{\sqrt{2}} I),$$

$$iX_{i\sigma_2 \otimes I}(\frac{1}{\sqrt{2}} I) = -iX_{iI \otimes \sigma_2}(\frac{1}{\sqrt{2}} I) = 2X_{i\sigma_1 \otimes \sigma_3}(\frac{1}{\sqrt{2}} I) = -2X_{i\sigma_3 \otimes \sigma_1}(\frac{1}{\sqrt{2}} I),$$

$$iX_{i\sigma_3 \otimes I}(\frac{1}{\sqrt{2}} I) = iX_{iI \otimes \sigma_3}(\frac{1}{\sqrt{2}} I) = 2X_{i\sigma_1 \otimes \sigma_2}(\frac{1}{\sqrt{2}} I) = 2X_{i\sigma_2 \otimes \sigma_1}(\frac{1}{\sqrt{2}} I).$$

Remark. The vector fields $X_{i\sigma_2 \otimes \sigma_1}$ and $X_{i\sigma_1 \otimes \sigma_1}$ are generated, by taking the Lie brackets among $X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_1 \otimes I}$ and $X_{iI \otimes \sigma_2}$,

$$X_{i\sigma_2 \otimes \sigma_1} = [X_{iI \otimes \sigma_2}, [X_{i\sigma_1 \otimes I}, X_{i\sigma_3 \otimes \sigma_3}]], \quad X_{i\sigma_1 \otimes \sigma_1} = -[X_{iI \otimes \sigma_2}, [X_{i\sigma_2 \otimes \sigma_2}, X_{i\sigma_3 \otimes \sigma_3}]],$$

respectively. The first and second equations of the above imply that, if doubly coupled, the drift and control vector fields generates a horizontal vector at $C = \Lambda \in M_1$ and another horizontal vector at $C = \Lambda \in M_0$, respectively.

### 4.4 The concurrence and control

We now investigate the right-hand side of (4.9) to examine the effect of the control on entanglement promotion. We suppose that the initial state $C(0)$ is a diagonal matrix $\Lambda = \text{diag}(\lambda_1, \lambda_2)$.

The terms in the $O(Jt^2)$-term of the right-hand side of (4.9) are expanded as

$$[X_\xi_{I \otimes I}, -X_{i\sigma_3 \otimes \sigma_3}] = -x_1 X_{i\sigma_2 \otimes \sigma_3} + y_1 X_{i\sigma_1 \otimes \sigma_3},$$

$$[X_{iI \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}] = -x_2 X_{i\sigma_3 \otimes \sigma_2} + y_2 X_{i\sigma_3 \otimes \sigma_1},$$

(4.15)

where $\xi_\alpha = ix_\alpha \sigma_1 + iy_\alpha \sigma_2, \alpha = 1, 2$, with $x_\alpha, y_\alpha \in \mathbb{R}$. These vector fields are all vertical at $\Lambda$ if $\Lambda \neq I/\sqrt{2}$. Indeed, the tangent vectors $X_{i\sigma_2 \otimes \sigma_3}(\Lambda), X_{i\sigma_1 \otimes \sigma_3}(\Lambda), X_{i\sigma_3 \otimes \sigma_2}(\Lambda),$
and $X_{i\sigma_3 \otimes \sigma_1}(\Lambda)$ are written as linear combinations of the vertical vectors $X_{i\sigma_1 \otimes I}(\Lambda)$, $X_{i\sigma_2 \otimes I}(\Lambda)$, $X_{iI \otimes \sigma_1}(\Lambda)$ and $X_{iI \otimes \sigma_2}(\Lambda)$. For instance, one has

$$X_{i\sigma_2 \otimes \sigma_3}(\Lambda) = \frac{\lambda_1^2 + \lambda_2^2}{2(\lambda_1^2 - \lambda_2^2)}X_{i\sigma_2 \otimes I}(\Lambda) + \frac{\lambda_1 \lambda_2}{\lambda_1^2 - \lambda_2^2}X_{iI \otimes \sigma_2}(\Lambda),$$  \hspace{1cm} (4.16a)

$$X_{i\sigma_1 \otimes \sigma_3}(\Lambda) = \frac{\lambda_1^2 + \lambda_2^2}{2(\lambda_1^2 - \lambda_2^2)}X_{i\sigma_1 \otimes I}(\Lambda) - \frac{\lambda_1 \lambda_2}{\lambda_1^2 - \lambda_2^2}X_{iI \otimes \sigma_1}(\Lambda),$$  \hspace{1cm} (4.16b)

if $\Lambda \neq I/\sqrt{2}$. If $\Lambda = I/\sqrt{2}$, they are found to be horizontal on account of (4.13).

The $O(J^2t^3)$-term is also vertical at $\Lambda$, as is shown by

$$-X_{i\sigma_3 \otimes \sigma_3}[X_{\xi_1 \otimes I}, -X_{i\sigma_3 \otimes \sigma_1}] = -\frac{1}{8}X_{\xi_1 \otimes I}, \quad -X_{i\sigma_3 \otimes \sigma_3}[X_{I \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}] = -\frac{1}{8}X_{I \otimes \xi_2}.$$

The $O(J^3t)$-term in (4.14) is significant for entanglement promotion. The vector fields $[X_{\xi_1 \otimes I}, [X_{\xi_1 \otimes I}, -X_{i\sigma_3 \otimes \sigma_1}]]$, $[X_{I \otimes \xi_2}, [X_{I \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}]]$ and $[X_{\xi_1 \otimes I}, [X_{I \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}]]$ are expressed as

$$[X_{\xi_1 \otimes I}, [X_{\xi_1 \otimes I}, -X_{i\sigma_3 \otimes \sigma_1}]] = (x_1^2 + y_1^2)X_{i\sigma_3 \otimes \sigma_3},$$

$$[X_{I \otimes \xi_2}, [X_{I \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}]] = (x_2^2 + y_2^2)X_{i\sigma_3 \otimes \sigma_3},$$

$$[X_{\xi_1 \otimes I}, [X_{I \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}]] = -x_1 x_2 X_{i\sigma_2 \otimes \sigma_2} + x_1 y_2 X_{i\sigma_2 \otimes \sigma_1} + y_1 x_2 X_{i\sigma_1 \otimes \sigma_2} - y_1 y_2 X_{i\sigma_1 \otimes \sigma_1},$$

respectively. Since $C(0) = \Lambda$ is diagonal, one has $X_{i\sigma_3 \otimes \sigma_3}(\Lambda) = X_{I \otimes I}(\Lambda) \in V_\Lambda$, so that the tangent vectors $[X_{\xi_1 \otimes I}, [X_{\xi_1 \otimes I}, -X_{i\sigma_3 \otimes \sigma_3}]]_\Lambda$ and $[X_{I \otimes \xi_2}, [X_{I \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}]]_\Lambda$ are vertical. In contrast with this, from Proposition 4.4 and Remark after it, it turns out that the tangent vector $[X_{\xi_1 \otimes I}, [X_{I \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}]]_\Lambda$ contains the horizontal components $X_{i\sigma_2 \otimes \sigma_1}(\Lambda), X_{i\sigma_1 \otimes \sigma_2}(\Lambda)$. In particular, if $\xi_1$ and $\xi_2$ are taken as $\xi_1 = i\sigma_1$ and $\xi_2 = i\sigma_2$, respectively, the tangent vector in question is horizontal,

$$[X_{i\sigma_1 \otimes I}, [X_{I \otimes \sigma_2}, -X_{i\sigma_3 \otimes \sigma_3}]]_\Lambda = X_{i\sigma_2 \otimes \sigma_1}(\Lambda) \in H_\Lambda.$$

This shows that the drift vector field $X_{i\sigma_3 \otimes \sigma_3}$ and the control vector fields $X_{\xi_1 \otimes I}, X_{I \otimes \xi_2}$ are coupled to generate a horizontal vector at $\Lambda$. We note further that the horizontal vector emerges at the third-order term in $t$, if $\Lambda \neq I/\sqrt{2}$.

We wish to evaluate the entanglement to confirm that the two-qubit system gets more entangled. As our entanglement measure and the concurrence are equivalent for the two-qubit system on account of $\sqrt{F(C(t))} = |\det C(t)|$, we here use the concurrence as a measure. From Eq. (4.13), the concurrence is approximately evaluated as

$$\sqrt{F(C(t))} \sim \sqrt{F(\varphi_{-i\sigma_3 \otimes \sigma_3}(\Lambda))} + \frac{Jt^3}{3!\sqrt{F(\Lambda)}}(dF)_\Lambda\left([X_{\xi_1 \otimes I}, [X_{I \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}]]\right),$$  \hspace{1cm} (4.17)
where we have used the following facts; (i) $F(e^{-t\xi_1}C(t)e^{-t\xi_2}) = F(C(t))$, (ii) $[X_{\xi_1 \otimes I}, -X_{i\sigma_3 \otimes \sigma_3}]$ and $[X_{I \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}]$ are vertical at $\Lambda$ if $\Lambda$ is not the maximally entangled state $I/\sqrt{2}$, (iii) $-X_{i\sigma_3 \otimes \sigma_3}[X_{\xi_1 \otimes I}, -X_{i\sigma_3 \otimes \sigma_3}]$ and $-X_{i\sigma_3 \otimes \sigma_3}[X_{I \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}]$ are vertical at $\Lambda$, (iv) $[X_{\xi_1 \otimes I}, [X_{\xi_1 \otimes I}, -X_{i\sigma_3 \otimes \sigma_3}]]$ and $[X_{I \otimes \xi_2}, [X_{I \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}]]$ are vertical at $\Lambda$, and (v) $[X_{\xi_1 \otimes I}, [X_{\xi_1 \otimes I}, -X_{i\sigma_3 \otimes \sigma_3}]] = [X_{\xi_1 \otimes I}, [X_{I \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}]]$. Since the derivative of the measure $F$ is given by

$$dF = 8\text{Re} \left( \det C^* \text{ tr} \left( \sigma_2 C \sigma_2 dC^T \right) \right),$$

one has

$$(dF)_\Lambda \left( [X_{\xi_1 \otimes I}, [X_{I \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}]] \right) = \frac{-x_1 y_2 + y_1 x_2}{2} \lambda_1 \lambda_2 (\lambda_1^2 - \lambda_2^2).$$

Hence, substituting this into (4.17), we have the following estimate of the concurrence,

$$\sqrt{F(C(t))} \sim \sqrt{F(\varphi_{-i\sigma_3 \otimes \sigma_3}(\Lambda))} + \frac{Jt^3}{2 \cdot 3!} (-x_1 y_2 + y_1 x_2)(\lambda_1^2 - \lambda_2^2)$$

for a sufficiently small $t > 0$. It is to be noted that the effect of controls on the concurrence emerges actually at the third-order term in $t$. We have to note that for $\Lambda = \text{diag (1, 0)}$, one has $F(\varphi_{-i\sigma_3 \otimes \sigma_3}(\Lambda)) = 0$, so that $\sqrt{F(C(t))} \sim \frac{Jt^3}{2 \cdot 3!} (-x_1 y_2 + y_2 x_1)$. This means that the state $\Lambda = \text{diag (1, 0)}$ gets entangled slowly by the controls.

The growth rate of the concurrence at the third order in $t$ depends on the quantity $-x_1 y_2 + y_1 x_2$, where $x_\alpha, y_\beta$ are control parameters given by $\xi_\alpha = ix_\alpha \sigma_1 + iy_\alpha \sigma_2$. It would be reasonable to restrict the magnitude of controls to $\sum_{\alpha=1}^2 (x_\alpha^2 + y_\alpha^2) = 1$. If this is the case, the maximal growth ratio is realized when $x_1 = -y_2, x_2 = y_1$. This means that the most efficient control for entanglement is given by $\xi_1 = ix_1 \sigma_1 + ix_2 \sigma_2, \xi_2 = ix_2 \sigma_1 - ix_1 \sigma_2$ with $x_1^2 + x_2^2 = 1/2$. However, if we choose the control given by $\xi_1 = ix_1 \sigma_1 - ix_2 \sigma_2, \xi_2 = ix_2 \sigma_1 + ix_1 \sigma_2$ with $x_1^2 + x_2^2 = 1/2$, the two-qubit gets into a less entangled state. If $\xi_1 = ix_1 \sigma_1 + ix_2 \sigma_2, \xi_2 = ix_2 \sigma_1 + ix_1 \sigma_2$ with $x_1^2 + x_2^2 = 1/2$, no change will occur in the entanglement of the two-qubit.

We turn to the case that the initial state $C(0)$ is not diagonal. Let us be reminded of the fact that when the $C(0)$ is diagonal, the $O(Jt^2)$-term in (4.19) is vertical. However, if $C(0)$ is not diagonal, the $O(Jt^2)$-term is not vertical, so that Eqs. (4.15) and (4.18) are put together to yield the contribution to entanglement promotion by

$$-\frac{Jt^2}{2 \sqrt{F(C(0))}} (dF)_{C(0)} \left( [X_{\xi_1 \otimes I}, -X_{i\sigma_3 \otimes \sigma_3}] + [X_{I \otimes \xi_2}, -X_{i\sigma_3 \otimes \sigma_3}] \right)$$

$$= -\frac{Jt^2}{4} \text{Re} \left( e^{-i\theta}(c_{01} + c_{10})(z_1 c_{00} + z_2 c_{11}) \right),$$

(4.20)
where $z_\alpha = x_\alpha + iy_\alpha$ for $\alpha = 1, 2$, and where $e^{i\theta}$ is defined by $\det C = e^{i\theta} |\det C| = e^{i\theta} \sqrt{F(C)}$. From (4.20), the $O(J t^2)$-term identically vanishes for arbitrary $\xi_1$ and $\xi_2$ if $c_{00} = c_{11} = 0$ or $c_{01} + c_{10} = 0$, but it takes a non-zero value in general, if $c_{01} + c_{10} \neq 0$ and $(c_{00}, c_{11}) \neq (0, 0)$.

5 Concluding remarks

We have discussed how the control and drift vector fields are coupled to make the NMR system more entangled on the two-qubit model, by examining the solution in detail in terms of horizontal and vertical vector fields and by evaluating the concurrence. As was pointed out in Sec. 4.1, the initial state $C(0) = \text{diag} \left(e^{i\theta}, 0\right)$ or $e^{i\theta} |0\rangle \oplus |0\rangle$ never gets entangled without controls, but we have shown in Sec. 4.4 that it can be entangled if controls are taken. However, the effect of the control on entanglement is so slow that it emerges at the third-order term in $t$. In fact, as is shown in Sec. 4.4, only vertical vector fields are generated in the second-order term in $t$ by the coupling between the drift and control vector fields, and further vector fields with non-vanishing horizontal components emerge at the third-order term in $t$.

It would be of help to state the relation of our theory to the Cartan decomposition of the Lie algebra. As for the two-qubit systems, the associated Lie algebra is $\mathfrak{su}(4)$, of which the Cartan decomposition is given by

$$\mathfrak{su}(4) = \mathfrak{k} \oplus \mathfrak{p},$$

where

$$\mathfrak{k} = \text{span}\{iI \otimes \sigma_j, i\sigma_k \otimes I\}_{j,k=1,2,3}, \quad \mathfrak{p} = \text{span}\{2i\sigma_j \otimes \sigma_k\}_{j,k=1,2,3}.$$  

The subalgebra $\mathfrak{k}$ generates vertical tangent vectors, and some of elements in $\mathfrak{p}$ span the horizontal subspace at $\Lambda = \text{diag} \left(\lambda_1, \lambda_2\right)$. For example, if $\lambda_1 \neq \lambda_2$, one has a horizontal vector $X_{i\sigma_1 \otimes \sigma_2}(\Lambda)$ (see Props. 4.3 and 4.4). However, the vectors associated with $\mathfrak{p}$ are not always horizontal. As is seen from (4.16), $X_{i\sigma_2 \otimes \sigma_3}(\Lambda)$ and $X_{i\sigma_1 \otimes \sigma_3}(\Lambda)$ are vertical if $\Lambda \neq I/\sqrt{2}$. In [27], as to non-local operation, they state that $\mathfrak{k}$ can be viewed as the local part in $\mathfrak{su}(4)$ and $\mathfrak{p}$ as the non-local part. In this sense, the horizontal vectors are of non-local nature, and the horizontal subspace makes the non-locality quite sharp. This is because the entanglement gets promoted most efficiently in the direction of horizontal vectors.
We comment on the motivation behind this study, which is related to the decomposition method for the analysis of quantum control problems on compact Lie groups. The decomposition method is of much use to design a control in order to construct a desired unitary operator [15–17], and then has been developed [7, 8]. In these situations, the control (or local operation) works instantaneously, and the drift (or non-local) Hamiltonian is assumed to be negligible while the control works. Under this assumption, the time evolution operator $U_t = e^{-i\hat{H}t}$ can be decomposed into the product of the local and non-local operators,

$$U_t = L_0 e^{-i\tau_1 \hat{H}_d} L_1 \cdots e^{-i\tau_N \hat{H}_d} L_N$$  \hspace{1cm} (5.1)

where $L_0, \cdots, L_N \in SU(2^\ell) \otimes SU(2^m)$ are generated by the control Hamiltonian, and where $\tau_1, \cdots, \tau_N > 0$ are time intervals for which the control Hamiltonian vanishes. In this view, the bipartite entanglement is not concerned by the coupling between the drift and control Hamiltonians in the time evolution. Our motivation is to consider the coupling in the evolution without the above assumption, and to investigate the relation of the coupling to the bipartite entanglement. The work [24] by Romano shares the motivation with us.

In [24], the decomposition of the unitary group SU(4) is used to study entanglement magnification. The time evolution $U_t$ is decomposed into $U_t = L_t A_t K_t$, where $L_t, K_t \in SU(2) \otimes SU(2)$ and $A_t \in H$ with $H$ the Cartan subgroup of SU(4). The author says that $L_t$ is irrelevant, and claims that the non-local operator $A_t$ makes a contribution to entanglement magnification together with a help of $K_t$. The decomposition method is sharp to study the entanglement in the evolution, because the evolution operator is completely separated into the “entangling part” $A_t$ and the two local operators $L_t$ and $K_t$. But, the process of entanglement promotion does not explicitly appear in the decomposition, since not only $A_t$ but also $L_t, K_t$ take in the coupling between the drift and control Hamiltonians. In fact, the $A_t, L_t$ and $K_t$ are all unknown until the time evolution operator $U_t$ is integrated in an explicit form.

On the other hand, the method of this article is represented in a familiar form with bipartite entanglement, and explains how the coupling between the drift and control Hamiltonians occurs in the time evolution. See (3.10), (3.11), (4.7) and (4.9). In particular, the right hand-side of (4.9) is not discussed in the context of (5.1). In a comparison to [24], our method allows one not to solve the time evolution of the system, although it is not sharp in the sense that the terms in (4.9) contain vertical (or local) components.
The formulas, (3.10) together with (3.11), and (4.9) also show how the effect of the controls spreads among qubits in the dynamical system. In contrast with this, the proof of the controllability of the NMR system would show the spread of the controls among qubits in a “static” situation. For the sake of self-containedness, we give the proof of the controllability of our NMR system (see Theorem B.8) in a method different from that in [1]. As is pointed out in App. B, the NMR system is controllable if and only if the associated spin graph is connected.

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A straightforward calculation along with Corollary 2.3 provides

\[
\begin{align*}
[X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_1 \otimes I}] &= -X_{i\sigma_2 \otimes \sigma_3}, & [X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_2 \otimes I}] &= X_{i\sigma_1 \otimes \sigma_3}, \\
[X_{i\sigma_3 \otimes \sigma_3}, X_{iI \otimes \sigma_1}] &= -X_{i\sigma_3 \otimes \sigma_2}, & [X_{i\sigma_3 \otimes \sigma_3}, X_{iI \otimes \sigma_2}] &= X_{i\sigma_3 \otimes \sigma_1}, \\
[X_{i\sigma_1 \otimes I}, X_{i\sigma_2 \otimes I}] &= -X_{i\sigma_3 \otimes I}, & [X_{iI \otimes \sigma_1}, X_{iI \otimes \sigma_2}] &= -X_{iI \otimes \sigma_3},
\end{align*}
\]
and the other Lie brackets prove to vanish. Further, double Lie brackets are given as follows:

\[
[X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_1 \otimes I}] = -[X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_2 \otimes \sigma_3}] = -X_{i\sigma_1 \otimes I}, \\
[X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_1 \otimes I}] = -[X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_1 \otimes \sigma_3}] = -X_{i\sigma_2 \otimes I}, \\
[X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_3 \otimes \sigma_1}, X_{i\sigma_2 \otimes \sigma_3}] = -[X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_3 \otimes \sigma_2}] = -X_{i\sigma_1 \otimes \sigma_1}, \\
[X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_3 \otimes \sigma_1}, X_{i\sigma_1 \otimes \sigma_2}] = -[X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_3 \otimes \sigma_1}] = -X_{i\sigma_1 \otimes \sigma_2}, \\
[X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_3 \otimes \sigma_1}, X_{i\sigma_1 \otimes \sigma_2}] = -[X_{i\sigma_3 \otimes \sigma_3}, X_{i\sigma_3 \otimes \sigma_1}] = -X_{i\sigma_3 \otimes \sigma_3},
\]

## B Controllability of the NMR system

As stated in Sec.3, we study the controllability of our NMR system. The drift Hamiltonian we treat is given by

\[
\hat{H}_d = \hat{H}_0 = \sum_{1 \leq \alpha < \beta \leq n} J_{\alpha\beta} \sigma_3^{(\alpha)} \sigma_3^{(\beta)},
\quad (B.1)
\]

where, for simplicity, we have denoted the drift Hamiltonian by \(\hat{H}_0\) and abbreviated \(\sigma_j^{(\alpha, n)}\) to \(\sigma_j^{(\alpha)}\), where \(\sigma_j^{(\alpha, n)}\) were defined in (3.2). For our convenience, we express the
control Hamiltonian as follows:
\[ \hat{H}_c = \sum_{\mu=1}^{n} v_{1}^{(\mu)} \hat{H}_\mu + \sum_{\mu=n+1}^{2n} v_{2}^{(\mu-n)} \hat{H}_\mu, \]
where
\[ \hat{H}_\mu := \begin{cases} \sigma_1^{(\mu)}, & \mu = 1, \ldots, n, \\ \sigma_2^{(\mu-n)}, & \mu = n+1, \ldots, 2n, \end{cases} \quad (B.2) \]

Our control system with the Hamiltonian \( \hat{H} = \hat{H}_d + \hat{H}_c \) is (pure state) controllable if and only if the Lie algebra generated by \( i\hat{H}_0, i\hat{H}_\mu, \mu = 1, \ldots, 2n \), is equal to \( \mathfrak{su}(2^n) \).

For comparison’s sake, we here quote the drift and control Hamiltonians studied in [1], which are given by
\[ \hat{H}'_0 = \sum_{\alpha<\beta} (M_{\alpha\beta} \sigma_1^{(\alpha)} \sigma_1^{(\beta)} + N_{\alpha\beta} \sigma_2^{(\alpha)} \sigma_2^{(\beta)} + P_{\alpha\beta} \sigma_3^{(\alpha)} \sigma_3^{(\beta)}), \quad (B.3) \]
and
\[ \hat{H}'_j = \sum_{\alpha=1}^{n} r_{\alpha} \sigma_j^{(\alpha)}, \quad j = 1, 2, 3, \quad (B.4) \]
respectively, where \( M_{\alpha\beta}, N_{\alpha\beta}, P_{\alpha\beta} \in \mathbb{R} \) are coupling constants and \( r_{\alpha} \in \mathbb{R} \) are the gyromagnetic ratio of the \( \alpha \)-th spin-\( \frac{1}{2} \) particle (or qubit). In our case of (B.2), it is assumed that each of spin-\( \frac{1}{2} \) particles can be stimulated by the only two components of the magnetic field. In contrast with this, in the case of (B.4), all the spin-\( \frac{1}{2} \) particles are assumed to be stimulated simultaneously by all the three components of the magnetic field with possibly different gyromagnetic ratios. While our drift Hamiltonian (B.1) contains only \( \sigma_3 \) factors, but theirs (B.3) has all the \( \sigma_j \) factors. In [1], they prove that the NMR system with the Hamiltonian \( \hat{H}'_0 + \sum_{j=1}^{3} u_j \hat{H}'_j \) is controllable if and only if the associated spin graph is connected.

Our objective in what follows is to show that our NMR system is controllable if and only if the associated spin graph is connected. The main point is to show that the Lie algebra generated by the operators \( -i\hat{H}_\mu, \mu = 0, 1, \ldots, 2n \), is equal to \( \mathfrak{su}(2^n) \). In this respect, what we have to do for proof is the same as that in [1]. However, as our Hamiltonian is different from that in [1], the method for proof should be different from that in [1].

### B.1 A decomposition of \( \mathfrak{su}(2^n) \)

Before calculating commutators among \( \{ -i\hat{H}_\mu \}_{\mu=0,1,\ldots,2n} \), we decompose \( \mathfrak{su}(2^n) \) into the sum of subspaces. We denote by \( \mathcal{B} \) the Lie algebra \( \mathfrak{su}(2^n) \),
\[ \mathcal{B} = \text{span}_\mathbb{R} \{ i\sigma_{j_1} \otimes \sigma_{j_2} \otimes \cdots \otimes \sigma_{j_n} | j_1, j_2, \ldots, j_n \in \{ 0, 1, 2, 3 \} \} \setminus \{ iI^{\otimes n} \}, \quad (B.5) \]
and break it up into subspaces $\mathcal{B}^{(k)}$, $k = 0, 1, \cdots, n - 1$, all the basis elements of which have as many as $k$ $I$-factors. Since $\dim \mathcal{B}^{(k)} = \binom{n}{k} 3^{n-k}$, and since $\dim \mathfrak{su}(4) = 4^n - 1 = \sum_{k=0}^{n-1} \binom{n}{k} 3^{n-k}$, the vector space $\mathcal{B}$ is broken up into

$$\mathcal{B} = \bigoplus_{k=0}^{n-1} \mathcal{B}^{(k)}. \quad (B.6)$$

For $k = 0$, $\mathcal{B}^{(0)}$ is expressed as

$$\mathcal{B}^{(0)} = \text{span}_\mathbb{R} \{i\sigma_{j_1} \otimes \sigma_{j_2} \otimes \cdots \otimes \sigma_{j_n} \mid j_1, j_2, \cdots, j_n \in \{1, 2, 3\}\}.$$  

Among $\mathcal{B}^{(k)}$ with $k = 1, \cdots, n$, we write down $\mathcal{B}^{(k)}$ only for $k = n - 1$;

$$\mathcal{B}^{(n-1)} = \text{span}_\mathbb{R} \{i\sigma_j^{(1)}, i\sigma_j^{(2)}, \cdots, i\sigma_j^{(n)} \mid j = 1, 2, 3\}.$$  

Further, we set

$$\mathcal{B}_3 := \text{span}_\mathbb{R} \{i\sigma_{j_1} \otimes \sigma_{j_2} \otimes \cdots \otimes \sigma_{j_n} \mid j_1, j_2, \cdots, j_n \in \{0, 3\}\} \setminus \{iI^{(n)}\}. \quad (B.7)$$

The following lemma shows that $\mathcal{B}_3$ holds a key position in calculating commutators.

**Lemma B.1.** Commutators among $\{-i\hat{H}_\mu\}_{\mu=1,\cdots,2n} \cup \mathcal{B}_3$ generate all the basis operators of $\mathcal{B}$.

**Proof.** Note that we have $\mathcal{B}_3 = \bigcup_{k=0}^{n-1} (\mathcal{B}_3 \cap \mathcal{B}^{(k)})$ from (B.6) and (B.7). For $k = 0$, we take an operator $i\sigma_3^{(n)} \in \mathcal{B}_3 \cap \mathcal{B}^{(0)}$. The commutators between $i\sigma_3^{(n)}$ and $-i\hat{H}_\mu$ are given by

$$[i\sigma_3^{(n)}, -i\hat{H}_\mu] = \begin{cases} 
  i\sigma_3^{(\mu-1)} \otimes \sigma_2 \otimes \sigma_3^{(n-\mu)}, & \mu = 1, \cdots, n, \\
  -i\sigma_3^{(\mu-n-1)} \otimes \sigma_1 \otimes \sigma_3^{(2n-\mu)}, & \mu = n + 1, \cdots, 2n.
\end{cases}$$

Taking successive commutators $[[i\sigma_3^{(n)}, -i\hat{H}_\mu], -i\hat{H}_\nu], \mu, \nu = 1, \cdots, 2n$, and so on, we can obtain the operators of the form, up to $\pm$ sign,

$$i\sigma_{j_1} \otimes \sigma_{j_2} \otimes \cdots \otimes \sigma_{j_n}, \; j_1, j_2, \cdots, j_n \in \{1, 2, 3\}.$$  

If we start with $iI \otimes \sigma_3^{(n-1)} \in \mathcal{B}_3 \cap \mathcal{B}^{(1)}$ and follow the same procedure as above, we can obtain

$$iI \otimes \sigma_{j_2} \otimes \cdots \otimes \sigma_{j_n}, \; j_2, \cdots, j_n \in \{1, 2, 3\}.$$  

Through the process of taking the commutator of an operator in $\mathcal{B}_3 \cap \mathcal{B}^{(k)}$ with $-i\hat{H}_\mu$, the resultant tensor product operator may have the $I$-factors fixed and $\sigma_3$-factors in the site $\mu$ or $\mu - n$ changed to $\sigma_1$ or $\sigma_2$, according to whether $\mu \in \{1, \cdots, n\}$, or $\mu \in \{n+1, \cdots, 2n\}$. Taking successive commutators, we can obtain the tensor product operators with the $(n-k)$ $\sigma_3$-factors replaced by $\sigma_1$ or $\sigma_2$ and the $k$ $I$-factors fixed, so that we have all the basis elements of $\mathcal{B}^{(k)}$ in the form of commutators among $\{-i\hat{H}_\mu\}_{\mu=1,\cdots,2n} \cup (\mathcal{B}_3 \cap \mathcal{B}^{(k)})$, $k = 0, 1, \cdots, n - 1$. This ends the proof.  

\[\square\]
B.2 Controllability with the complete spin graph

We now assume that $J_{\alpha \beta} \neq 0$ for all $1 \leq \alpha < \beta \leq n$, or that the associated spin graph is complete, to show that the commutators among $\{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n}$ span $\mathfrak{su}(2^n)$. On account of Lemma [B.1], we have only to show that commutators among $\{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n}$ can span $\mathcal{B}_3$. We first show that by taking commutators among $\{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n}$, the term $-i\sigma_3^{(\alpha)} \sigma_3^{(\beta)}$ of the drift Hamiltonian $-i\hat{H}_0$ can be singled out, if $J_{\alpha \beta} \neq 0$. For $-i\hat{H}_0 = -i \sum_{\alpha < \beta} J_{\alpha \beta} \sigma_3^{(\alpha)} \sigma_3^{(\beta)}$ and $-i\hat{H}_\mu = -i\sigma_1^{(\mu)}$, $\mu = 1, \ldots, n - 1$, the commutators between them are given by

$$[-i\hat{H}_0, -i\hat{H}_\mu] = -i \sum_{\alpha=1}^{\mu-1} J_{\alpha \mu} \sigma_3^{(\alpha)} \sigma_3^{(\mu)} - i \sum_{\beta=\mu+1}^n J_{\mu \beta} \sigma_3^{(\mu)} \sigma_3^{(\beta)}.$$  

We calculate further commutators to obtain, for $1 \leq \mu < \nu \leq n$,

$$[-i\hat{H}_0, -i\hat{H}_\mu, -i\hat{H}_\nu] = -i J_{\mu \nu} \sigma_3^{(\mu)} \sigma_3^{(\nu)}.$$  

Moreover, a calculation with the right-hand side of the above provides, for $1 \leq \mu < \nu \leq n$,

$$[-i\hat{H}_\mu, -i\hat{H}_\nu, -iJ_{\mu \nu} \sigma_2^{(\mu)} \sigma_3^{(\nu)}] = -i\sigma_1^{(\mu)}, -iJ_{\mu \nu} \sigma_2^{(\mu)} \sigma_3^{(\nu)} = -i J_{\mu \nu} \sigma_3^{(\mu)} \sigma_3^{(\nu)}.$$  

If $J_{\mu \nu} \neq 0$, we obtain $-i\sigma_3^{(\mu)} \sigma_3^{(\nu)}$ as a commutator among $\{i\hat{H}_0, \ldots, i\hat{H}_n\}$. Thus, we have proved the following

**Lemma B.2.** If $J_{\alpha \beta} \neq 0$ for $\alpha$ and $\beta$ with $1 \leq \alpha < \beta \leq n$, the operator $-i\sigma_3^{(\alpha)} \sigma_3^{(\beta)}$ can be realized as a commutator among $\{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n}$.

We now show that the $\mathcal{B}_3$ can be generated from $\{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n}$, by using Lemma B.2 under the assumption that $J_{\alpha \beta} \neq 0$ for all $1 \leq \alpha < \beta \leq n$. Let $\alpha_i$, $i = 1, \ldots, m$, be positive integers such that

$$1 \leq \alpha_1 < \alpha_2 < \cdots < \alpha_{m-1} < \alpha_m \leq n \quad \text{with } m \geq 2. \quad (B.8)$$

For a given sequence of $\alpha_i$, Lemma B.2 provides us with the operators

$$-i\sigma_3^{(\alpha_1)} \sigma_3^{(\alpha_2)}, -i\sigma_3^{(\alpha_2)} \sigma_3^{(\alpha_3)}, \ldots, -i\sigma_3^{(\alpha_{m-1})} \sigma_3^{(\alpha_m)}$$

in the form of commutators among $\{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n}$. We can form commutators among these operators and $\{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n}$ to obtain

$$F_1 := [-i\sigma_3^{(\alpha_1)} \sigma_3^{(\alpha_2)}, -i\hat{H}_{n+\alpha_2}] = -i\sigma_1^{(\alpha_1)} \sigma_3^{(\alpha_2)};$$

$$F_k := [-i\sigma_3^{(\alpha_k)} \sigma_3^{(\alpha_{k+1})}, -i\hat{H}_{\alpha_k}, -i\hat{H}_{n+\alpha_{k+1}}] = -i\sigma_2^{(\alpha_k)} \sigma_1^{(\alpha_{k+1})},$$

$$F_{m-1} := [-i\sigma_3^{(\alpha_{m-1})} \sigma_3^{(\alpha_m)}, -i\hat{H}_{\alpha_{m-1}}] = -i\sigma_2^{(\alpha_{m-1})} \sigma_3^{(\alpha_m)};$$
where \( k = 2, \cdots, m - 2 \), if \( m \geq 3 \). From \( F_1, F_2, \cdots, F_{m-1} \), we can form commutators, for \( m = 3, \cdots, n \),

\[
C_m := [ \cdots [[F_1, F_2], \cdots, F_{m-2}], F_{m-1}] = -i\sigma_3^{(a_1)} \sigma_3^{(a_2)} \cdots \sigma_3^{(a_{m-1})} \sigma_3^{(a_m)},
\]

which is in \( \mathcal{B}_3 \cap \mathcal{B}^{(n-m)} \). For \( m = 1, 2 \), the operators \( C_1, C_2 \) can be expressed, respectively, as

\[
C_1 := -i\sigma_3^{(a)} = [-i\hat{H}_1, -i\hat{H}_{n+1}], \quad 1 \leq a \leq n, \tag{B.9}
\]

\[
C_2 := -i\sigma_3^{(a_1)} \sigma_3^{(a_2)}, \tag{B.10}
\]

where \( C_1 \in \mathcal{B}_3 \cap \mathcal{B}^{(n-1)} \) and \( C_2 \in \mathcal{B}_3 \cap \mathcal{B}^{(n-2)} \), and these can be expressed as commutators among \( \{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n} \) as well.

Taking all possible sequences of \( \alpha_i \) subject to \( (B.8) \), we can construct all possible tensor product operators with \( m \) \( \sigma_3 \)-factors and \( (n - m) \) \( I \)-factors as commutators among \( \{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n} \). In other words, we can form all the basis elements of \( \mathcal{B}_3 \cap \mathcal{B}^{(n-m)} \) in the form of commutators among \( \{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n} \). Since \( \mathcal{B}_3 = \sum_{m=1}^n (\mathcal{B}_3 \cap \mathcal{B}^{(n-m)}) \), we obtain the following

**Lemma B.3.** If \( J_{a\beta} \neq 0 \) for all \( \alpha \) and \( \beta \) with \( 1 \leq \alpha < \beta \leq n \), the \( \mathcal{B}_3 \) are generated by commutators among \( \{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n} \).

From Lemmas [B.1] and [B.3] it turns out that if \( J_{a\beta} \neq 0 \) for all \( \alpha \) and \( \beta \) with \( 1 \leq \alpha < \beta \leq n \), \( \mathcal{B} \) is generated by taking commutators among \( \{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n} \). Hence, we obtain

**Proposition B.4.** If \( J_{a\beta} \neq 0 \) for all \( \alpha \) and \( \beta \) with \( 1 \leq \alpha < \beta \leq n \), or if the spin graph associated with the drift Hamiltonian is complete, the Lie algebra \( \mathfrak{su}(2^n) \) is generated from \( \{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n} \).

### B.3 Controllability with a connected spin graph

In the following, we consider the case where some of \( J_{a\beta} \)'s may vanish, but the spin graph is connected. Suppose that \( J_{a\beta}, J_{\beta\gamma} \neq 0 \), and \( J_{a\gamma} = 0 \). We may assume that \( \alpha < \beta < \gamma \). By Lemma [B.2] we can put the operators \( -i\sigma_3^{(a)} \sigma_3^{(\beta)}, -i\sigma_3^{(\beta)} \sigma_3^{(\gamma)} \) in the form of commutators among \( \{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n} \). We then take commutators to obtain

\[
D := [\{-i\sigma_3^{(a)} \sigma_3^{(\beta)}, -i\hat{H}_\beta\}, -i\sigma_3^{(\beta)} \sigma_3^{(\gamma)}] = -i\sigma_3^{(a)} \sigma_1^{(\beta)} \sigma_3^{(\gamma)}.
\tag{B.11}
\]
From $D, -i\sigma_3^{(\beta)}\sigma_3^{(\gamma)}, -i\hat{H}_{n+\beta}$, and $-i\hat{H}_n$, we obtain
\[
D' := 4[[D, -i\hat{H}_\gamma], [-i\hat{H}_{n+\beta}, -i\sigma_3^{(\beta)}\sigma_3^{(\gamma)}]] = -i\sigma_3^{(\alpha)}\sigma_3^{(\gamma)}.
\] (B.12)

Further calculation provides
\[
D'' := [D', -i\hat{H}_{n+\gamma}] = -i\sigma_3^{(\alpha)}\sigma_3^{(\gamma)}.
\]

Thus we have found that the operators $-i\sigma_3^{(\alpha)}\sigma_3^{(\gamma)}$ can be described as commutators taken among $-i\sigma_3^{(\alpha)}\sigma_3^{(\beta)}, -i\sigma_3^{(\beta)}\sigma_3^{(\gamma)}$, and $-i\hat{H}_\mu, \mu = 1, \cdots, 2n$. This implies that if there are interactions between particles $\alpha$ and $\beta$ and between particles $\beta$ and $\gamma$, an interaction between particles $\alpha$ and $\gamma$ is induced, though $J_{\alpha\gamma} = 0$ at the beginning, by taking commutators among $\{-i\hat{H}_\mu\}_{\mu=0,1,\cdots,2n}$. We may interpret this fact as follows: In the process of making commutators among $\{-i\hat{H}_\mu\}_{\mu=0,1,\cdots,2n}$, two edges $(\alpha, \beta), (\beta, \gamma)$ of the spin graph give rise to a new edge $(\alpha, \gamma)$ which represents an interaction between two particles $\alpha$ and $\gamma$.

So far we have studied the case where there are two edges between the nodes $\alpha$ and $\gamma$. We may apply the same procedure to the case where there are $r$ edges between the nodes $\alpha$ and $\gamma$, where $r = 2, \cdots, n - 1$. Suppose we have, say, three edges $(\alpha, \beta_1), (\beta_1, \beta_2), (\beta_2, \gamma)$ with $J_{\alpha\beta_1}, J_{\beta_1\beta_2}, J_{\beta_2\gamma} \neq 0$. The same procedure as above yields the edge $(\alpha, \beta_2)$ from $(\alpha, \beta_1)$ and $(\beta_1, \beta_2)$, and consequently $(\alpha, \gamma)$ from $(\alpha, \beta_2)$ and $(\beta_2, \gamma)$. Then, it turns out that if two particles $\alpha$ and $\gamma$ are linked with a sequence of two-particle interactions, the operator $-i\sigma_3^{(\alpha)}\sigma_3^{(\gamma)}$ describing an interaction between the particles $\alpha$ and $\gamma$ is induced by taking commutators among $\{-i\hat{H}_\mu\}_{\mu=0,1,\cdots,2n}$. Since the spin graph is connected, there is a sequence of edges between any pair of nodes $\alpha$ and $\gamma$, so that one can obtain the operator $-i\sigma_3^{(\alpha)}\sigma_3^{(\gamma)}$ expressed as a commutator among $\{-i\hat{H}_\mu\}_{\mu=0,1,\cdots,2n}$. Thus we have reached the same conclusion as in Lemma B.2 with an assumption weaker than that in Lemma B.2.

**Lemma B.5.** If the associated spin graph is connected, all the interaction operators $-i\sigma_3^{(\alpha)}\sigma_3^{(\beta)}, 1 \leq \alpha < \beta \leq n$, are generated from the drift and control Hamiltonians.

Thus Prop. B.4 is refined as follows:

**Proposition B.6.** If the spin graph associated with the drift Hamiltonian is connected, the Lie algebra $\mathfrak{su}(2^n)$ is generated from $\{-i\hat{H}_\mu\}_{\mu=0,1,\cdots,2n}$. Thus, the NMR system is controllable if the associated spin graph is connected.
B.4 A necessary and sufficient condition for controllability

We now consider the case where the spin graph $S$ is disconnected. We assume that the graph $S$ is broken up into two disjoint subgraphs, $S_1$ and $S_2$, each of which is connected. Suppose that $S_1$ and $S_2$ have nodes 1 to $r$, and $r+1$ to $n$, respectively. According to the decomposition, $S = S_1 \cup S_2$, of the spin graph, the drift Hamiltonian is also decomposed into the sum of two terms,

$$
\hat{H}_0 = \hat{H}'_0 + \hat{H}''_0, \quad \hat{H}'_0 = \sum_{1 \leq \alpha < \beta \leq r} J_{\alpha \beta} \sigma_3^{(\alpha)} \sigma_3^{(\beta)}, \quad \hat{H}''_0 = \sum_{r+1 \leq \alpha < \beta \leq n} J_{\alpha \beta} \sigma_3^{(\alpha)} \sigma_3^{(\beta)}.
$$

Since $S_1$ is connected, in the same procedure as taken in proving Prop. B.6, the Lie algebra $L_1$ generated from $-i\hat{H}'_0$ and $\{-i\hat{H}_\alpha, -i\hat{H}_{n+\alpha}\}_{\alpha=1,\ldots,r}$ proves to be $L_1 = su(2^r) \otimes I \otimes (n-r)$. In the same manner, the operators $-i\hat{H}''_0$ and $\{-i\hat{H}_{r+\alpha}, -i\hat{H}_{n+r+\alpha}\}_{\alpha=1,\ldots,n-r}$ generate the Lie algebra $L_2 = I \otimes su(2^{n-r})$. Since two operators each of which is in the respective Lie algebras $L_1$ and $L_2$ commute, the Lie algebra generated by the whole operators $\{-i\hat{H}_\mu\}_{\mu=0,1,\ldots,2n}$ splits into the direct sum $L_1 \oplus L_2$, which is a subalgebra of $su(2^n)$ but not equal to the whole $su(2^n)$. We may generalize this fact to the case where the spin graph is broken up into more than two disjoint subgraphs, but the generalization is easy to perform. So far we have shown the following

**Proposition B.7.** If the spin graph associated with the drift Hamiltonian is disconnected, the NMR system (3.3) is not controllable.

From Props B.6 and B.7 we obtain the following theorem;

**Theorem B.8.** The NMR system (3.3) with the drift and control Hamiltonians (3.1) is controllable, if and only if the spin graph associated with the drift Hamiltonian is connected.