Constructive control of a spin system via periodic control

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Abstract

Constructive procedures for the control of a class of coupled spin systems with bounded amplitude sinusoidal pulses are described. Full details are first illustrated for a single spin $\frac{1}{2}$ particle. The key steps for a pair of such particles are then sketched. The fact that the amplitudes can be arbitrarily bounded implies that the separation between the Larmor frequencies of the individual spin $\frac{1}{2}$ particles can be quite small and yet the system can be controlled by addressing each spin individually.

I Introduction

Spin systems arise in a variety of applications, [4, 2]. Their manipulation, therefore, is a problem which arouses considerable interest. In this paper we will consider first a single spin system and show that one can prepare any desired unitary generator from the evolution of such a system when it is probed by a piecewise sinusoidal magnetic field. The frequency of this field is tuned at the Larmor frequency of the spin. It remains, therefore, to specify the duration, the amplitude and the phase of each piece of this piecewise sinusoidal field. This is achieved in two steps. First, by a proper choice of phase, it is shown that the system is transformed into a system controlled by constant inputs in a suitable rotating frame. Next, the amplitude and duration of each piece is determined by finding that piecewise constant control which will drive the system in the rotating frame to a desired state. For the second step a certain Euler angle decomposition of SU(2) is used. The entire procedure is fully constructive. Extension to the case of two spins is also provided. Some of the details are sketched at the insistence of a churlish (or perhaps web shy) referee even though every detail can be found in the public domain, [8]. In principle, there is no obstruction for three or more spins, but the requisite calculations of exponentials of matrices, render the method less constructive than the case of one or two spins. It is interesting that even though the direct problem of solving the differential equations for a two-level system driven by a known sinusoidal field in closed form does not seem to have a solution, the inverse problem of finding a sinusoidal field which will drive the state of such a system to another desired state is completely solvable in closed form. The calculations for the single spin systems extend easily to similar classical systems evolving on SO(3).

The main steps involved in the constructive methodology for the single particle case are as follows: i) The system is transformed into a suitable rotating frame, so that in this rotating frame the system, when probed by a periodic field, becomes one of a family of singleinput systems without drift, controlled by constant inputs. These constant inputs are the amplitudes of the original sinusoidal control field. The frequency of each piece of this field is the Larmor frequency of the spin. We remark that merely passing to a rotating frame and choosing the frequency to be the Larmor frequency is not enough. The phases of each piece of the sinusoidal field has to be chosen appropriately to achieve the final target. and ii) A suitable Euler decomposition is used to determine the amplitudes and duration of each piece of the control field.

For the case of two spin systems, the overall strategy is similar. However, the rotating frame does not kil-I the drift, so that the system in the rotating frame is one of a family of single input systems with drfit. Further, while the analogue of Euler angle decompositions for SU(4) is reasonably straightforward to establish, an explicit calculation of the angles themselves is non-trivial problem. Secondly, the determination of amplitudes and durations of the pulses is now a harder problem, since the system in the rotating frame is no longer driftless. In this paper, we will indicate briefly how both problems are resolved. For reasons of length, full details for the two spin system cannot be provided here. Details are available on the Los Alamos archive (quant-ph/0012019). In this introduction, we will only comment on the latter of these two problems. One method of avoiding the problems posed by drift is to use hard pulses. These are extremely high amplitude pulses applied for very short durations. It turns out that the corresponding Euler decomposition of SU(4) factors every target as a product of exponentials of the drift and exponentials of the control matrix. The former can clearly be obtained by free evolution. It is for the latter that hard pulses have been suggested. The rationale is as follows. Suppose it is desired to prepare e^{bB} , where B is the control matrix and $b \in R$. For this apply a control,

$$u(t) = \frac{b}{\delta}$$

for δ units of time, where δ is a small number. Then the system's evolution is $\exp(\delta A + bB)$, where A is the drift. In the limit $\delta \to 0$, this tends to e^{bB} . Clearly this is not desirable on at least two counts. First, it yields e^{bB} only approximately. Secondly, to improve the accuracy δ has to be made ever smaller. This is both impractical and even for any reasonable tolerance requires high amplitudes. Such amplitudes have the effect of coupling other spins, neglected in the assumed model, to the system. In our approach, we do not view B as the control matrix, but rather as a certain iterated commutator of the drift and the control matrix (which it happens to be). This enables (after some guiles) to produce fields which not only prepare e^{bB} exactly but also can be arbitrarily bounded in amplitude.

The idea of using decompositions of unitary matrices to control quantum systems has suddenly become popular. The papers, [7, 9, 10], describe the utility of such decompositions for molecular systems. The papers, [11, 12], address quantum optics and cavity QED respectively. Neither paper, however, provides formulae for the parameters entering each factor of the factorizations used. The papers, [16, 17, 20, 8] address spin systems. The first studies spin systems in the weak coupling limit - which is also the limit this paper will use. The second studies a pair of spins in the strong coupling limit. However, the z component of the magnetic field is taken to be available to manipulate. In particular, it is supposed that this component can be taken to be zero. However, this is unrealistic since the z component is the static strong magnetic field. Without this static field the Larmor frequencies become null and with it the entire spin dynamics essentially vanishes. Both papers, [16, 17] use hard pulses and further do not supply formulae for the factors entering the factorization. In this paper (see [8] for full details) these problems are overcome. In [20] the author extends the constructive procedure described in [8] for two spin to the case of two coupled strongly homonuclear particles. The techniques mimick those in [8] (such as passage to a rotating frame and using sinusoidal fields to switch between different systems). However, the method for producing bounded amplitude controls is different from that in [8].

The balance of this paper is organized as follows. In the next section some notation is reviewed. The next section presents the results for a single spin system. Extensions to the case of two spins is presented in the fourth section. The final section offers some conclusion-

2 Some Terminology and Notation

Throughout this paper the following notations will be used. The Pauli matrices will be denoted as:

$$\sigma_x = \left(egin{array}{cc} 0 & 1 \ 1 & 0 \end{array}
ight)$$

$$\sigma_y = \left(\begin{array}{cc} 0 & -i \\ i & 0 \end{array}\right)$$

and

$$\sigma_z = \left(\begin{array}{cc} 1 & 0 \\ 0 & -1 \end{array}\right)$$

In terms of this definition of the Pauli matrices, a few important 4×4 Hermitian matrices can also be defined:

$$I_{1k} = \sigma_k \otimes I_2; I_{2k} = I_2 \otimes \sigma_k, \ k = x, y, z; \tag{2.1}$$

3 Single Spin System

The dynamics of a single spin in atomic units with Larmor frequency, ω , under an external radio-frequency field, may be described via, [4]

$$\dot{V} = -\frac{i}{2}(\omega\sigma_z V + b\sigma_x u_1(t)V + b\sigma_y u_2(t)V), \quad (3.2)$$

with $V \in SU(2)$, $V(0) = I_2$. Here b is a constant called the gyromagnetic ratio and the $u_i(t)$, i = 1, 2 are piecewise sinusoidal fields to be designed. We will choose each piece of these fields to take the form:

$$u_1(t) = c\cos(\omega t + \phi); u_2(t) = c\sin(\omega t + \phi) \qquad (3.3)$$

Thus, the frequency of each piece of both controls is the Larmor frequency, ω , of the spin. The amplitude and phase of each piece has to be chosen. The Larmor frequency is typically large. This is because it is directly proportional to the strength of a static magnetic field which is typically large and is applied parallel to the z- axis. The proportionality constants are related to the spin, while the strength is, of course, apparatus dependent.

From One Input to Two Inputs: In the spectroscopy literature, one does not think of the $u_i(t)$ as two independent controls. Rather the system is probed by a radiofrequency field with some frequency and some

phase, which is normally polarized along the x-axis. This linearly oscillating field can be decomposed into two counter-rotating terms, one of which can be neglected when the static magnetic field is high (which it usually is). The net effect of this that the dynamics behaves as if there were two periodic controls, $u_i(t)$ with the parametric form described above. Insofar as we are aware, the $u_i(t)$ cannot be taken to be any more independent, i.e., the functional form of the two inputs has to be given by Equation (3.3).

Let us first address the determination of the phase of each piece. For this we will pass to a rotating frame. The purpose of this rotating frame is to to render the dynamics in the rotating frame both autonomous and related to the Euler decomposition chosen. Specifically let

$$U(t) = e^{tF}V(t)$$

The rotating frame, e^{tF} , is the exponential of

$$F = \frac{i}{2}\omega\sigma_z$$

A direct calculation now yields

$$\dot{U} = -rac{i}{2}cb_1\Delta$$

where the matrix, Δ is parametrized in the following fashion by the phase, ϕ :

$$\Delta = \left(\begin{array}{cc} 0 & e^{-i\phi} \\ e^{i\phi} & 0 \end{array} \right)$$

Thus, by choosing $\phi=0$ one can ensure that $\Delta=\sigma_x$, whereas choosing $\phi=\frac{3\pi}{2}$ gives $\Delta=\sigma_y$. This then yields the following single-input driftless system(s) in the rotating frame:

$$\dot{U} = -\frac{i}{2}cb_1\sigma_x, \text{ if } \phi = 0$$
 (3.4)

or

$$\dot{U} = -\frac{i}{2}cb_1\sigma_y$$
, if $\phi = \frac{3\pi}{2}$ (3.5)

This makes the preparation of any $S \in SU(2)$ clear. Simply factorize S in its σ_x, σ_y Euler angles:

$$S = e^{iD\sigma_x}e^{iE\sigma_y}e^{iF\sigma_x}$$

To generate the factor $e^{iD\sigma_x}$ (resp. $e^{iF\sigma_x}$) choose the phase of the field to be 0 and the power of the field (i.e., amplitude times the duration) to be $-2\frac{D}{b_1}$ (resp. $-2\frac{D}{b_1}$). This gives one considerable liberty in choosing the duration and the amplitude. In particular, the duration can always be chosen to be positive. Likewise, to generate $e^{iE\sigma_y}$ choose the phase of the field to be $\frac{3\pi}{2}$ and the power of the field to be $-2\frac{E}{b_1}$. This generates S in the rotating frame in some time T_S . Thus, $U(T_S) = S$.

To prepare S, in the original frame the sequence of sinusoidal fields has to be followed by free evolution for the requisite time, T_S , Once we have made a choice of the duration of each of the pulses, this requisite time can also be clearly determined.

The Euler angles D, E and F can be explicitly determined from the Cayley-Klein coordinates - which are nothing but the entries of S written in polar coordinates. It is easier to work directly with $i\sigma_x$ and $i\sigma_y$, than by starting with more familiar Euler angles and using them to obtain D, E and F, [15].

Arbitrary Bounds on the Power and Amplitude:

The structure of the foregoing procedure makes it clear that any bound on the amplitude or the power of <u>each</u> piece of the field can be accommodated. For this all that has to be done is to factorize each factor of the Euler factorization further into exponentials of $i\sigma_x$ or $i\sigma_x$.

Different Choices for Phase: Choosing ϕ , different from 0 and $\frac{\pi}{2}$ leads to arbitary linear combinations the matrices, $i\sigma_x$ and $i\sigma_y$, for the matrix of the driftless system in the rotating frame. This is useful because it is possible to explicitly factorize any $\in SU(2)$ as a product of exponentials of linear combinations $i\sigma_x$ and $i\sigma_y$, [7, 9]. One advantage of using both these matrices is that the resulting amplitude is typically lower than that obtained from using the Euler decompositions.

Postmultiplication of the Rotating Frame: It may seem profitable to define $U(t) = V(t)e^{tF}$ for certain objectives. One such objective is obtaining a desired value of the expectation of an observable. It is not difficult to see that if the initial density matrix is diagonal, i.e., has no coherences, then e^{tF} has no effect on the expectation value. This seems attractive, since we do not have to use the free evolution that was needed when e^{tF} premultiplied V(t). However, now it is not possible to obtain a time-independent system in the rotating frame.

Extension to SO(3): The above procedure can be mimicked verbatim in the following cases: i) Systems on SU(2) with two inputs, where the drift and the control matrices are orthogonal elements of su(2) (under the standard inner product); ii) Systems on SO(3) with two inputs, where the drift and control matrices are images of the corresponding system on SU(2) as in i), under the standard Lie algebra isomorphism $\psi: su(2) \to so(3)$. The basic idea is illustrated for i). Using the orthogonality of the drifts and control matrices, we obtain a Lie algebra isomorphism $\psi: su(2) \to su(2)$, where ψ maps the drift and control of i) to the drift and control matrices of the system (3.2). This exponentiates to a Lie group homomorphism $\phi: SU(2) \to SU(2)$. Given a desired target T for i), we choose a target S for (3.2) such

that $\phi(S)=T$. Now, we find a sinusoidal field which prepares S for (3.2). Then the same field prepares T for i). This last argument requires some care. If we were preparing S by piecewise constant fields, then the fact that ϕ exponentiates ψ would yield the conclusion. For piecwise sinusoidal fields, we approximate the field that prepares S by piecewise constant fields and then follow the previous line to obtain the conclusion.

4 The Two Spin Case

Next, the extension of the foregoing section to the case of a pair of coupled spin $\frac{1}{2}$ particles will be described. Technical details can be found in [8]. Let the Larmor frequencies of the two particles be ω_1 and ω_2 . It is assumed that i) There are no resonances between ω_1 and ω_2 ; and ii) that the difference between them, $|\omega_1 - \omega_2| \ge 2\pi J$, where J is the coupling constant between the two spins. The coupling constant, J and the Larmor frequencies depend both on the molecular species and the experimental apparatus. Typically, J is quite small. Thus, the second of the two assumptions above does not require a wide separation between the two Larmor frequencies.

Homonuclearity Incorporated: It is important to notice that is not being assumed a priori that the difference between the two Larmor frequencies is much more than the amplitude of the pulse, as is frequently assumed to render the argument, that each spin can be addressed individually, valid [4]. To the contrary, we will see that regardless of the difference between the two frequencies, we can obtain any desired bound on the amplitude of the pulse. This means that even though the system will be controlled by individually addressing each spin, the difference between the Larmor frequencies can be rather small. Thus, the methodology of this manuscript is applicable to homonuclear molecules, while the hard pulse techniques necessarily require the molecule to be heteronuclear. Thus, for instance our methods are capable of handling a system of two protons whose Larmor frequencies are slightly different because of different "shielding effects", [4]. In other words, molecules whose gyromagnetic ratios are the same (i.e., homonulcear molecules) are susceptible to the methods in [8]. For the case of "strongly homonuclear" molecules, i.e., the case when the Larmor frequencies are equal, see [20]. The constructive algorithms in [20] are similar in many respects to those in [8]. However, the technique in [20] to incorporate bounds on amplitude is different from that in [8].

The assumption that $\omega 1$ and ω_2 are not resonant means that the system can be controlled by addressing each spin individually, as long as the amplitude of the field, c satsifies $|\omega_1 - \omega_2| \ge c$, [4]. Let us first assume that we are addressing the first spin. In other words,

we choose our fields, $u_1(t)$ and $u_2(t)$ to be sinusoidal with frequency equal to ω_1 . Then, the assumption that $|\omega_1 - \omega_2| \ge 2\pi J$ means that the evolution of the two spins in atomic units can be modelled according to:

$$\dot{V} = -\frac{i}{2} (2\pi J I_{1z} I_{2z} + \omega_1 I_{1z} + \omega_2 I_{2z} + b_1 I_{1x} u_1(t) + b_1 I_{1y} u_2(t))V$$
(4.6)

with $V \in SU(4)$, $V(0) = I_4$. The constant b_1 is the gryomagnetic ratio of the first spin.

If the second spin was being addressed (which means that the frequency of the $u_i(t)$ is taken to be ω_2), then the resulting model would be similar to the last equation except that $I_{1k}, k = x, y$ would be replaced by $I_{2k}, k = x, y$ and b_2 , the gyromagnetic ratio of the second particle, would be used instead of b_1 .

Choice of the Rotating Frame: As in the previous section the system will be transformed to a rotating frame. It might seem tempting to use e^{tA} , where A is the drift of the system as the rotating frame. However, this is not useful because the matrices $-iI_{ij}$, i=1,2,j=x,y generate only $su(2)\otimes su(2)$ and not all of su(4). Thus, unless the target is in $SU(2)\otimes SU(2)$ killing the drift is injurious to our goal. Further, using e^{tA} will lead to a time-dependent system in the rotating frame. The choice of the rotating frame, which will be made, is dictated by the following decomposition of any $S \in SU(4)$:

$$S = \prod_{k=1}^{Q} e^{it_k M_k} \tag{4.7}$$

Here each M_k is any one of the matrices $I_{1z}I_{2z}$ and $I_{ij}i=1,2,j=x,y$. Further details regarding the t_k,M_k and Q appears later. One of the important contributions of the authors' Los Alamos archive paper, [8], is indeed to determine the t_k algorithmically from the entries of S. Without such a procedure any "algorithm" based on (4.7) is not even remotely algorithmic. For the moment however, Equation (4.7) suggests that e^{tF} , with

$$F = \frac{i}{2}(\omega_1 I_{1z} + \omega_2 I_{2z})$$

is a useful rotating frame. Choosing the fields to be:

$$u_1(t) = c\cos(\omega t + \phi); u_2(t) = c\sin(\omega t + \phi)$$

with $\omega=\omega_1$ and $\phi=0$ (resp. $\phi=\frac{3\pi}{2}$) gives the following evolution for $U\left(U(t)=e^{tF}V(t)\right)$

$$\dot{U} = -\frac{i}{2}(2\pi J I_{1z} I_{2z} + c b_1 \Delta) U \tag{4.8}$$

where $\Delta = I_{1x}(\text{resp.}I_{1y})$. Likewise, choosing $\omega = \omega_2$ and $\phi = 0(\text{resp.}\frac{3\pi}{2})$ gives:

$$\dot{U} = -\frac{i}{2}(2\pi J I_{1z}I_{2z} + cb_1\Delta)U$$

with $\Delta = I_{2x}(\text{resp.}I_{2y})$. Thus, by choosing the frequency and phase as per the recipe above yields one of

four single-input systems with drift, controlled by constant inputs, in the rotating frame. The drift of each of the four systems is $-\frac{i}{2}(2\pi J I_{1z}I_{2z})$, while the control matrix is upto a constant one of the four matrices, $-iI_{ij}$, i=1,2,j=x,y.

Now equation (4.7) immediately suggests that if for a certain k, M_k is $I_{1z}I_{2z}$ then that factor can be produced by free evolution - this clear if $t_k > 0$. For negative, t_k it just needs to be noted that the matrix $-iI_{1z}I_{2z}$ is periodic (only for free evolution terms do we use periodicity). Thus, the constant t_k in $e^{-it_kI_{1z}I_{2z}}$ may always be presumed to be positive. So it remains only to address the factors where the M_k is one of I_{ij} , i = 1, 2, j = x, y.

The following result, therefore, finishes the constructive control, via bounded amplitude controls, for this coupled two spin system provided the parameters, t_k can be calculated explicitly:

Proposition 4.1 Let $L \in R$, the matrix $e^{-iLI_{ij}}$, i = 1, 2, j = x, y can be explicitly factorized as $\prod_{k=1}^3 exp(-ia_k J I_{1z} I_{2z} - ib_k I_{ij})$, with $a_k > 0$. The constants, a_k and b_k can be written down in terms of the t_k in Equation (4.7). Further, there is an explicit refinement of of the factorization to meet any ampltude bound $|\frac{b_k}{a_k}| C$.

The proof of this proposition follows some inspired matrix manipulations. The matrix manipulations, referred to, are facilitated by the following crucial observation:

$$(i\alpha JI_{1z}I_{2z}+i\beta I_{ij})^2=-k^2I_4, \alpha, \beta \in R, k \text{ some constant}$$

Similarly, the square of the commutator of the drift and the control matrix is also, upto a constant, equal to $-I_4$. These observations enable us to factor explicitly the exponentials, $e^{-iLI_{ij}}, L \in R$, as products of free evolution terms and a single control pulse term. The key control theoretic insight, motivating this factorization, is that $-iI_{ij}$, i = 1, 2, j = x, y is, upto a constant, an iterated commutator of $-iI_{1z}I_{2z}$ with $-I_{ij}$ itself. In this commutator $-iI_{1z}I_{2z}$, the free Hamiltonian, appears twice, while $-iI_{ij}$ appears once. Thus, it seems plausible, on basis of our prior work [9], that the target $e^{-iLI_{ij}}$ could be prepared by three pulses, two of which are free evolution terms and one is a constant input, applied to the system: $\dot{U} = -iII_{1z}I_{2z}U - iI_{ij}Uu(t)$ This is indeed true. While, this phenomenon is probably false in general it seems to be holding true for many other classes of spin systems. For instance, some numerical experiments that we performed seem to suggest that this true if the system is treated in the strong coupling limit also. In this limit, the drift and control matrices have more complicated structure, which (pending further research) preclude explicit calculations of their exponentials in closed form.

Remark 4.1 Undoing the Rotating Frame: If $U(T_S) =$ S, then in the original frame, the target $e^{-T_S F}S$ has been prepared. Even if one uses hard pulses, T_S will typically be bounded from below by a positive time. So this error must be rectified. In [17] a rotating frame is not used - perhaps because the design is piecewise constant and further the z component of the field is taken to be zero. Similarly, [16], does not mention this issue even though a passage to a rotating frame is used. The solution to this problem is to prepare the target $e^{T_0F}S$ in the rotating frame for a parameter, T_0 , which is chosen to satisfy $T_1 + T_S = T_0$, where T_1 is the time it takes to prepare e^{T_0F} in the rotating frame. For this T_0 has to satisfy a certain transcendental equation. If J >> 2 in atomic units (this is typically the case), then a good approximation to T_0 is $T_S + \frac{21+\sqrt{2}}{T}$.

Determining the Analogues of the Euler Angles: To make the entire procedure genuinely constructive, the "Euler" angles, t_k in (4.7) have to be related to the entries of S. To gain some perspective on this question, let us see how Equation (4.7) arises. It is well known that the Lie algebra su(4) admits a Cartan decomposition in terms of $su(2) \otimes su(2)$ and its complement in su(4), [19]. In [16] this fact and some calculations are used to arrive at Equation (4.7). However, they do not give any method to find the t_k . Using some Kronceker calculus, it can be shown, specifically that:

$$\begin{split} S &= e^{iD_1I_{1z}}e^{iE_1I_{1y}}e^{iF_1I_{1z}}e^{iD_2I_{2z}}e^{iE_2I_{2y}}\\ &e^{iF_2I_{2z}}e^{-i\frac{\pi}{4}I_{1y}}e^{-i\frac{\pi}{4}I_{2y}}e^{-i\theta_1I_{1z}I_{2z}}e^{-i\frac{7\pi}{4}I_{1y}}\\ &e^{-i\frac{7\pi}{4}I_{2y}}e^{-i\frac{7\pi}{4}I_{1z}}e^{-i\frac{7\pi}{4}I_{2z}}e^{-i\theta_2I_{1z}I_{2z}}e^{-i\frac{\pi}{4}I_{1z}}\\ &e^{-i\frac{\pi}{4}I_{2z}}e^{-i\theta_3I_{1z}I_{2z}}e^{iD_3I_{1z}}e^{iE_3I_{1y}}e^{iF_3I_{1z}}\\ &e^{iD_2I_{2z}}e^{iE_2I_{2y}}e^{iF_2I_{2z}} \end{split}$$

In [8] the problem of finding the Euler angle analogues is successfully tackled. The main idea is to use a modification of the Givens decomposition to constructively factor S as a product

$$S = \prod_{k=1}^6 S_k$$

Here $S_k, k=1,\ldots,5$ is a matrix which is, upto permutation, a block matrix whose blocks are I_2 and an explicitly determined SU(2) matrix. S_6 is the Kronecker product $e^{i\frac{\pi}{2}\sigma_y}\otimes S(\alpha,\zeta,\mu)$, where $S(\alpha,\zeta,\mu)$ is the unique SU(2) matrix which conveys the vector (d_1,d_2) to $(||(d_1,d_2)||,0)$. Here (d_1,\ldots,d_4) is the fourth column of S^{\dagger} . In [8] the fifteen real parameters, $(D_i,E_i,F_i), i=1,\ldots,4,\theta_k, k=1,\ldots,3$ were determined explicitly for each of the $S_k, k=1,\ldots,6$. This then provides a factorization of S of the form (4.7) with all the parameters, t_k fully determined from the entries S. The advantage over working directly with S is twofold. First, each of the Givens factors is easy to compute from the entries of S. Second, the only non-trivial part of a Givens matrix is a 2×2 , SU(2) submatrix. SU(2) matrices admit

several easily computed parametrizations in sharp contrast to SU(4) matrices and thus, the determination of the (D_i, E_i, F_i) , $i = 1, \ldots, 4$ and $\theta_k, k = 1, \ldots, 3$ for a Givens matrix can be carried out in closed form.

Further Extensions: In principle, there is no obstacle to extending these results to both the case of additional spins and stroung coupling limits. The main difficulty for the case of strong coupling limit is to choose a suitable rotating frame in which the system is autonomous. In the weak coupling limit, when the number of spins is three or more, analogous Euler decompositions exist. However, some of the terms cannot be expressed as the exponential of the free Hamiltonian. Even if these problems were ignored, the goal of constructive state generation is still elusive. This is mainly because the matrix exponential calculations are substantially more complicated. While it is possible to write down an explicit formula for the exponential of an su(4) matrix, this formula is so longwinded that extracting information out of it is a daunting task. Thus, even for two particles unless special assumptions are imposed on the Hamiltonian, a fully constructive technique, along the lines in this paper, will require more investigation.

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