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CONTROL OF SOME QUANTUM SYSTEMS - ROLE OF PHASES

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Abstract: Constructive procedures for the exact control of a class of molecular and spin systems with sinusoidal pulses are described. It is shown that the phases of these fields plays a crucial role. It is also shown that the fields can be bounded in amplitude and that the number of phases required to be at the disposal of the experimenter is quite small. The fact that the amplitudes can be bounded in advance means, amongst other things, that the conditions needed for the underlying model to be valid, can be guaranteed to hold. *Copyright © 2001 IFAC*

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1. INTRODUCTION

One often hears the adage that, while an overall phase is irrelevant in quantum mechanics, relative phases are important for a wide swathe of quantum phenomena. Therefore, it is reasonable to expect that by a suitable choice of phase in an external, sinusoidal field, a quantum system may be controlled to great finesse. In this paper two examples which confirm this expectation are provided. The first example comes from molecular control and the second from NMR spectroscopy. These systems play an important role in many technologies. See Butkovsky and Samoilenko (1980); Rabitz et al., (2000) and Silverman (2000).

Though the two examples address different physical phenomena, they share some similar features. First, both examples may be thought of as systems of interconnected oscillators. The proposed control methodology in both cases consists of addressing each oscillator in the system individually, via a sinusoidal field tuned to the frequency of the oscillator in question. For this to be successful, two ingredients are required. First, there should be no resonances amongst the various oscillators. Secondly, the pulse area (which is the integral of the field) should be much smaller than the frequency of the oscillator in question. It is known experimentally for both systems that, under these assumptions, each individual oscillator may indeed be addressed separately with negligible interference from the others. In this paper external sinusoidal fields whose pulse area can be bounded arbitrarily will be devised. The next point of similarity is that the *phase of the sinusoidal field* plays a crucial role in the efficacy of the control. The final point of similarity is that the key mathematical constructs are structured decompositions of the unitary matrices. In this paper most proofs are omitted. They may be found in Ramakrishna et al., (1999) and Ramakrishna et al., (2000b). Before finishing this introductory section, it is worth stressing an important point about the second

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example. Mathematically, the problem reduces to the control of a single input system with drift on $SU(4)$. One technique to to control such systems consists of extremely high amplitudes applied for very short times. Such pulses are called hard pulses. From a controls point of view, a hard pulse argument is an argument which neglects the drift at the cost of approximate state steering. Not only is this arduous to implement in the laboratory, but it is leads to violation of the basic model as the amplitude is increased in a desire to improve accuracy. In contrast, in this paper we will achieve bounded pulse area via fields whose amplitudes are not large and the state is nevertheless exactly prepared. *We are able to do this because the structure of the drift is utilized in the field design.*

Notation: It is assumed that the reader is familiar with the Pauli matrices, $\sigma_i, i = x, y, z$. Related to these are the following tensor products: $I_{ik} = \sigma_k \otimes I_2; I_{2i} = I_2 \otimes \sigma_k,\ i = x, y, z$

2. THE MOLECULAR EXAMPLE

Consider the generation of any desired special unitary transformation within a single N-level atom/molecule, or more generally a single particle, interacting with an electromagnetic field which is treated semi-classically. The evolution of the system is described by $\dot{U} = AU + BU\epsilon(t); U \in U(N)$ Here A is a diagonal matrix $diag(i\lambda_1, \ldots, i\lambda_N)$. The frequencies of the system are given by the differences, $\lambda_i - \lambda_j$. B is the matrix representation of the dipole operator and is a skew-Hermitian. Further assumptions on the molecule are as follows. i) There are no resonances amongst the frequencies of the system. ii) The entries of B are purely imaginary and all its diagonal entries are zero. iii) If selection rules (see Silverman, 2000) preclude a certain level being accessed directly from another, then there must be a ladder of levels linking the two, so that the two states can be accessed from each other indirectly. This can be restated as follows. Consider the undirected graph, $G = (V, E)$, whose vertex set, V, consists of N indices $\{1, \ldots, N\}$ and two vertices i and j are connected by an edge iff the (i, j)th entry of B is non-zero. The assumption, then, is the same as requiring that the undirected graph, G, is connected. Under these assumptions, it was shown in Ramakrishna et al., (1999) that if such an N-level atom was subjected to a series of Q coherently locked sinusoidal fields, $\epsilon_k(t) = A_k(t)\cos(\omega_k t + \phi_k)$ with (slowly varying) amplitude $A_k(t)$, frequency, ω_k, resonant with a pair of levels and phase, ϕ_k, applied for $T = t_1 + t_2 + \ldots t_Q$ units of time, then under the rotating wave approximation (see Silverman, 2000) its unitary generator after T units of time is given by

$$U(T) = e^{-iAT} V_Q V_{Q-1} \ldots V_2 V_1 \qquad (1)$$

The $N \times N$ matrices V_k are, after a suitable permutation, block matrices whose top $(N-2) \times (N-2)$ block is the $(N-2) \times (N-2)$ identity matrix and the remaining 2×2 block is an $SU(2)$ matrix of the form: $V(\gamma_k) = \begin{pmatrix} 0 & i\gamma_k \\ i\bar{\gamma}_k & 0 \end{pmatrix}$ The complex number, γ_k, is related to the kth pulse via, $\gamma_k = -ib_{nm}e^{i\phi_k}a_k$ where ϕ_k and a_k are the phase and the pulse area of the kth pulse, while b_{nm} is the matrix element of B corresponding to the pair of levels addressed by the kth pulse. Since the times t_i can be chosen subject only to the restriction that the pulse area, $\int_0 t_k A_k(t)dt$ is not too sensitive to them, it is clear that the matrix e^{-iAT} is known in advance. Thus, the preparation of a desired unitary generator reduces to the question of whether every unitary matrix can be decomposed into a product of matrices of the form $V(\gamma_i)$. Since the matrices $V(\gamma_i)$ are special unitary, it would seem that only those $U \in U(N)$ such that $e^{iAT}U$ is in $SU(N)$ can be so prepared. However, every unitary matrix is an overall phase factor times a special unitary matrix, and such phase factors are experimentally irrelevant, unless detecting such a phase, when the N level particle is viewed as part of a larger system, was the purpose of the experiment, (cf., Silverman, 2000). Thus, the question of preparing a desired unitary generator reduces to being able to factor every matrix in $SU(N)$ as a product of $V(\gamma_k)$'s. This question was answered affirmatively and constructively in two steps in Ramakrishna et al., (1999): i) **S1:** First it was shown that every $SU(N)$ matrix can be decomposed into a product of block matrices which are the identity matrix, except in a 2×2 block, where they are some $SU(2)$ matrix. In the absence of selection rules that this is a standard fact. The novelty in Ramakrishna et al., (1999) was to show how to obtain such a decomposition while simultaneously respecting forbidden transitions imposed by the selection rules. ii) **S2:** The main step was to show that every $SU(2)$ matrix could be factored as a product of 3 matrices of the form $V(\gamma_k)$. Note that, as the complex numbers, γ_k need not be either purely real or purely imaginary, this decomposition is *not* the usual Euler decomposition. *Further, explicit formulae were given for the complex numbers, γ_k, in terms of the entries of $S \in SU(2)$ represented in polar coordinates.* Since, the absolute value and phase of the numbers, γ_k, are themselves the pulse area and the phase of the incident fields, a completely constructive procedure for preparing any desired unitary generator was thereby obtained. Thus, in summary any unitary generator could be prepared by sinusoidal pulses, resonant with a pair of levels, each with pulse area no more than $\frac{\pi}{2}$ as long as the means to prepare *any* phase is at

the experimenter's disposal. The bound, $\frac{\pi}{2}$, on the pulse area of a single pulse, comes from the details in Ramakrishna et al., (1999). The requirement that any phase be at the experimenter's disposal arises from the fact that the 2×2 block in **S1**, could in principle, be any $SU(2)$ matrix. Thus, for a wide class of atoms/molecules - those which meet the conditions imposed in Ramakrishna et al., (1999) - there is a simple, but explicit, procedure to generate any unitary transformation via interaction with sinusoidal pulses. The second step, **S2**, can be viewed as constructive path planning for a driftless two input system on $SU(2)$, viz., $\dot{V} = i\sigma_x V u_1(t) + i\sigma_y V u_2(t), V \in SU(2)$. Indeed, the matrix $V(\gamma)$ is the exponential of a suitable (real) combination of $i\sigma_x$ and $i\sigma_y$. However, it is important to realize that u_1 and u_2 have nothing to do with the actual controls in the physical problem. Similarly, the coefficients of $i\sigma_x$ and $i\sigma_y$ in this exponential have no relation to the time and magnitude of the control applied to the actual system.

Bounding the Pulse Area and Minimizing the Number of Phases: Since there is a bound of $\frac{\pi}{2}$ on the pulse area of an individual pulse in Ramakrishna et al., (1999). methodology could not be directly applied to atoms whose energy levels were not widely separated. This is because, the rotating wave approximation which was invoked in Ramakrishna et al., (1999) requires, in addition to the no resonance condition, that the pulse area be much lower than the frequency of the oscillator addressed,(see Silverman, 2000). However, the first author showed, in Ramakrishna, (2000a), how not only the pulse area could be bounded arbitrarily but also the number of phases required could be chosen to be atmost four. This significantly increases the class of molecules to which the control strategy proposed in Ramakrishna et al., (1999). can be applied, while at the same time minimizes the resources (for phase creation) required. The proof of this was achieved by proving constructively a bang-bang result for the following system : $\dot{V} = i\sigma_y V + i\sigma_x V u(t), V \in SU(2)$ This is a system with drift (if one preferred to deal with the driftless system in the previous remark, then the number of phases needed by this strategy would be six). Once again, $u(t)$, is not the control actually applied. The same construction can be modified to achieve arbitrary pulse area bounds. This is done by applying the same value of the bang-bang control to this system on $SU(2)$ for short periods of time, instead of applying this value for a longer time at one shot (e.g., apply the control, $u = 1$, for ten times for one minute instead of one time for ten minutes!). This may seem counterintuitive. However, it is worth emphasizing that the duration of the control, $u(t)$, for this last $SU(2)$ system is not really the duration of the actual control. Rather, it is related to the pulse area of the actual control. Thus, minimizing pulse area of a control pulse for the actual system turns out to be equivalent to minimizing the duration an individual piece of the fictitious bang-bang control, $u(t)$, for the above $SU(2)$ system.

3. THE SPIN EXAMPLE

Technical details which are omitted here for brevity may be found in Ramakrishna et al., (2000b). It is assumed that there are no resonances amongst the two Larmor frequencies of the two spins. Under this assumption it is possible to address each spin indvidually with negligible contribution from the other spin, as long as the amplitude of the field can be bounded (see Evans, 2000). A common model for the evolution of such a system is $\dot{V} = -\frac{i}{2}(\tilde{A}V + u_1(t)B_1V + u_2(t)B_2V), V \in SU(4)$ Here, $\tilde{A}$ is $\omega_1\sigma_z \otimes I_2 + \omega_2 I_2 \otimes \sigma_z + J\sigma_z \otimes \sigma_z$, and the interaction Hamiltonians are $B_1 = b_1 I_{1x}$ and $B_2 = b_1 I_{1y}$. The $b_i, i = 1, 2$ (b_2 appears below) are constants related to the gyromagnetic ratios, and the $\omega_i, i = 1, 2$ are the Larmor frequencies. J is also a constant. We take $u_1(t) = c\cos(\omega t + \phi)$ and $u_2(t) = c\sin(\omega t + \phi)$. The amplitude, c, frequency, ω and phase, ϕ are each to be designed in a piecewise constant manner. The frequency will always be taken to be ω_1 (or ω_2, if the other spin is being addressed). The above equation was derived assuming that the first spin was being addressed. If the second spin is being addressed, then B_1, B_2 would be replaced by $b_2 I_{2x}$ and $b_2 I_{2y}$ respectively, while the frequency, ω, of the field would be replaced by ω_2.

Now passing to a rotating frame, $U(t) = e^{tF}V$ with $F = \frac{i}{2}(\omega_2 I_2 \otimes \sigma_z + \omega_1 \sigma_z \otimes I_2)$ and choosing the phase, ϕ, to be 0 yields $\dot{U} = -\frac{i}{2}(JI_{1z}I_{2z})U - \frac{i}{2}(cb_1\sigma_x \otimes I_2)U$ If, on the other hand, the phase of the incident fields, ϕ, was equal to $\frac{\pi}{2}$, one would have had $\dot{U} = -\frac{i}{2}(JI_{1z}I_{2z})U - \frac{i}{2}(cb_1\sigma_y \otimes I_2)U$. Similarly, the *same* rotating frame can be used to address the other spin (with the frequency of the field, $\omega = \gamma_2$) to obtain the following equation $\dot{U} = -\frac{i}{2}(JI_{1z}I_{2z})U - \frac{i}{2}(cb_2 I_2 \otimes \sigma_x)U$ if the phase, ϕ equals 0. Choosing $\phi = \frac{\pi}{2}$ similarly yields $\dot{U} = -\frac{i}{2}(JI_{1z}I_{2z})U - \frac{i}{2}(cb_2 I_2 \otimes \sigma_y)U$. *Thus, by choosing the frequency of the field to be one of the Larmor frequencies and by choosing the phase in an appropriate manner to equal one of a few valuesi* and then passing to a unique rotating frame leads to the following system, which is controlled by constant inputs: $\dot{U} = -\frac{i}{2}AU - \frac{i}{2}dBU, U \in SU(4)$ with $A = J\sigma_z \otimes \sigma_z$ and B one of the matrices $I_{1x}, I_{2x}, I_{1y}, I_{2y}$. The constant, d, is related to the amplitude of the field, c, and other constants of the system. So, to complete the constructive control methodology all that remains

is to specify the duration and the amplitude, c, of the control fields $u_1(t)$ and $u_2(t)$. For this, it suffices to provide the duration and the amplitude d. Indeed, using the Cartan decomposition of $SU(4)$ in terms of $SU(2) \otimes SU(2)$, it can be shown that every matrix in $SU(4)$ can be written in the following form:

$$S = \Pi_{k=1}^{Q} exp(-it_k A_k) \qquad (2)$$

The A_k are one of $I_{1z}I_{2z}, I_{1z}, I_{2z}, I_{1y}, I_{2y}$. One specific form of Equation (2) was given in Khaneja and Glaser (2000), whereby $S \in SU(4)$ was represented as a product of 21 such factors. Of these 15 were parametrized by 15 parameters, (D_i, E_i, F_i) and $\theta_k, i = 1, \ldots, 4; k = 1, \ldots, 3$. *Without providing any technique for finding these 15 parameters*, it is suggested in Khaneja and Glaser (2000) that, this decomposition can be used to control the system with piecewise constant hard pulses to account for the exponential of the control coupling and by free evolution for exponential of the drift.

In Ramakrishna et al., (2000b) there are three significant innovations. First, phases are used to design piecewise sinusoidal fields whose amplitudes will be the piecewise constant controls for the system in the rotating frame. This is in contrast to assuming that the system is already in a rotating frame. Mere passage to a rotating frame will not suffice - the correct choice of phase is absolutely *essential*. Secondly, instead of using high ampliutde pulses to generate the exponentials of the $I_{ij}, i = 1, 2, j = x, y$ controls whose amplitude can be bounded arbitrarily were provided for such factors. These amplitudes were provided explicitly, assuming that the real parameters, $(D_i, E_i, F_i), i = 1, \ldots, 4$ and $\theta_k, k = 1, \ldots, 3$ are already available. Third, these parameters were *in fact constructively determined in terms of* S. This third step, which is needed if any control technology based on the Cartan decomposition is to be completely constructive, makes an unexpected contact with the mathematics of the molecular example. Indeed, the target matrix S was first factored into a Givens decomposition of the type in **S1** described in the previous section (with some appropriate modifications). Then the parameters $(D_i, E_i, F_i), i = 1, \ldots, 4$ and $\theta_k, k = 1, \ldots, 3$ were determined explicitly for the factors in the Givens decomposition. 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. The first of these three innovations was described above in detail. The remaining may be found in Ramakrishna et al., (2000b).

4. CONCLUSIONS

Relative phases are all important in quantum mechanics. In this paper two physically important situations, wherein one can completely and constructively control quantum systems, via sinusoidal external fields whose phases played an important role, were given. While the ability to create these phases exists in laboratories, it was also shown that the number of phases needed constitute a very small list. Equally importantly it was shown that the amplitude and pulse areas of the fields can be bounded *a priori*. The signifcance of this goes beyond minimizing expense. It means, crucially, that the basic assumptions which go into the model can be met. In other words, there is no analogue of the spillover effect in contrast to hard pulse techniques. Furthermore, the techniques of these paper are an improvement over these latter techniques because the former provide exact path planning.

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