Explicit generation of unitary transformations in a single atom or molecule

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A constructive procedure for generating a prescribed unitary transform via the optically driven evolution of a multilevel atom is described. Assuming a clean separation of the coupled levels, the procedure employs the rotating wave approximation together with a decomposition of a unitary matrix into simpler matrix factors with specified structure. Applications to state preparation and observation are also provided.

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I. INTRODUCTION

The preparation of an *a priori* prescribed finite-dimensional unitary transformation via the evolution of an externally driven quantum system is a theoretical and technological challenge that plays an important role in several fields including atomic and molecular manipulation [1–4], quantum computation [5,6], and quantum cryptography [7]. This paper will describe a systematic technique to generate arbitrary special unitary transformations in a quantum system that draws on concepts from control theory. The results described here are applicable to the broader domain of quantum system manipulation.

The principal concept exploited here is that any special unitary matrix can be decomposed into products of transformations with a particular structure. Similar decompositions have recently been suggested as well. Reck *et al.* [8] use the decomposition of any $m\times m$ unitary matrix into a product of simpler matrices [i.e., tensor products of an arbitrary 2×2 block and a complementary block consisting of the identity matrix of dimension $(m-2)$ obtained in [9]] to produce an optical implementation of quantum cryptography schemes. They assume no structure within the factors and then show that any special unitary matrix can be written as a product of such factors. In this regard, Law and Eberly [10] considered a decomposition for controlling the quantum state of a cavity field. There is a relationship between their factors and those in the present paper, but the number of factors needed are different. Finally, DiVincenzo has demonstrated that any unitary matrix can be written as a product of unitary matrices that are either a tensor product of an arbitrary 2×2 block and I_{M-2} or a special 4×4 block and I_{M-4} [13]. This latter decomposition was motivated by a multiparticle implementation of quantum logic gates. Here we will consider the fundamental case that the nontrivial sub-block is of size 2×2. Unlike the conventional decomposition where the 2×2 blocks are in U(2), the approach presented in this paper respects the structure. Moreover, it can be tailored to take into account practical laboratory constraints.

The specific ingredients of our approach to creating the unitary matrices has the following steps.

(1) Employ an atom or other simple quantum system whose energy levels are well separated, such that there are no interfering resonances with any pair of optically coupled levels. Assurance of this condition may call for the introduction of suitable static external fields. Spectral separation permits the system to be controlled by sequentially addressing pairs of levels. Furthermore, this circumstance allows one to employ the rotating wave approximation when considering control field designs. Controllability is also possible under conditions more relaxed than this clean spectral separation, but each such case must be individually analyzed for its applicability.

(2) Decompose the prescribed $N\times N$ unitary matrix into a product of $N\times N$ unitary matrices that are nontrivial only in a sub-block. Here we will consider the simplest case where the sub-block is of size 2×2. This decomposition will respect the selection rules appropriate to the system, and will also be tailored to take into account the rotating wave approximation and any limitations on the matrix elements of the electric dipole operator, which is assumed to be the coupling operator. The decomposition used in this paper is different from the standard one where the 2×2 block can be any element of U(2) (see [9]).

(3) An explicit characterization of the exponential of an arbitrary real linear combination of the Pauli matrices will be utilized. This formulation will be useful in parametrizing the 2×2 blocks of the previous steps to provide an explicit design for laboratory control.

The balance of this paper is organized as follows. Section

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II presents a derivation of the decomposition needed for the physical system. Next, a constructive factorization of any SU(2) matrix into the factors prescribed by this decomposition is derived. In addition, it is shown how selection rules can be respected during the decomposition process. Remark 2.1 explains how the number of factors may be minimized and also discusses an alternative derivation of the desired SU(2) decomposition suggested to us by Harter [12]. Section III discusses how the concepts in the paper may be applied to state preparation and observation. Finally Sec. IV addresses the extent to which the ideas in this paper may be generalized to other applications.

II. GENERATING ATOMIC SCALE UNITARY TRANSFORMATIONS

Consider an atom irradiated by a sequence of electromagnetic pulses with the aim of guiding its evolution initially from the identity matrix to a desired final unitary matrix. The atom-pulse interaction is taken to be semiclassical. The atom is assumed to have the following properties: (1) The atom has M accessible levels that are well separated for controllability based on frequency discrimination. (2) The matrix elements of the dipole operator (with respect to the basis consisting of the eigenfunctions of the free Hamiltonian) are all real and the diagonal elements are identically zero. This latter assumption can be relaxed in certain cases. (3) The atom-pulse pair is ''controllable'' in the sense that there is always a connection between any two levels, i.e., if selection rules preclude accessing level i directly from j, then there is a set of levels (possibly more than one) forming a ladder, such that state i can be accessed at least indirectly from state j.

The multilevel atom will be irradiated by a sequence of tailored pulses. Each of the pulses above will address only a prescribed pair of levels. There is flexibility in the pulse structure and each should be chosen to be as simple as possible. The action of the entire sequence of pulses is described below as

$$i\dot{U}(t,0)=(H_0-\mu\epsilon)U(t,0), \tag{2.1}$$

where μ is the induced dipole operator and $\epsilon(t)$ is the control field. Defining $\Omega(t,0)$ by

$$U(t,0)=\exp(-iH_0t)\Omega(t,0)$$

leads to the interaction representation

$$i\dot{\Omega}(t,0)=-\epsilon(t)e^{iH_0t}\mu e^{-iH_0t}\Omega(t,0).$$

The control pulse is taken to be $\epsilon(t)=A(t)\cos(\omega t+\phi)$, where ϕ is a phase to be chosen and $A(t)$ is assumed to be slowly time varying compared to ω^{-1}, and the frequency ω is resonant with levels i and j to be coupled.

The $M\times M$ matrix $\epsilon(t)e^{iH_0t}\mu e^{-iH_0t}$ will have all allowed couplings given by μ, but, given the conditions stated earlier, we may neglect all terms except the (i,j)th and (j,i)th entries, which, respectively, are $\mu_{ij}e^{i\phi}A(t)$ and $\mu_{ij}e^{-i\phi}A(t)$ (recall that all entries of the matrix representation of μ are assumed to be real and the diagonal entries are all zero).

The pulse is applied for a time t_1 with the only restriction that the integral $\int_0^{t_1}A(t)dt$ be insensitive to a slight change in t_1. Applying the rotating wave approximation yields

$$\Omega(t_1,0)=V_1,\Omega(0,0)=Id_M,$$

where the matrix V_1 is an $M\times M$ matrix that is the identity except for a 2×2 block, which is the exponential of a 2×2 matrix of the form

$$\begin{pmatrix} 0 & i\gamma_1 \\ i\overline{\gamma_1} & 0 \end{pmatrix}. \tag{2.2}$$

$\overline{\gamma_1}$ is the complex conjugate of the complex number γ_1, which has the following polar representation:

$$\gamma_1=\mu_{ij}e^{i\phi}\int_0^{t_1}A(t)dt. \tag{2.3}$$

It will be seen later in Eq. (2.6) that the magnitude of the complex number γ_1 enters into our considerations only as the argument of trigonometric functions. Hence $A(t)$ and ϕ are chosen so that γ_1's magnitude can take any desired value within $[0,2\pi]$ to achieve the necessary action to generate V_1. This point is useful for practical laboratory considerations.

After the application of this first pulse the unitary generator is given by

$$U(t_1,0)=e^{-iH_0t_1}V_1.$$

Now another pulse of length t_2 is applied that is resonant with only some other allowed pair of levels. We then have

$$U(t,0)=U(t,t_1)U(t_1,0).$$

Once again setting $U(t,t_1)=e^{-iH_0(t-t_1)}\Omega(t,t_1)$ gives

$$i\dot{\Omega}(t,t_1)=-\epsilon(t)e^{iH_0(t-t_1)}\mu e^{-iH_0(t-t_1)}\Omega(t,t_1).$$

Premultiplying both sides of the last equation by $e^{iH_0t_1}$, using the rotating wave approximation, and then integrating up to time $t=t_2$ yields

$$e^{iH_0t_1}\Omega(t_2,t_1)=V_2e^{iH_0t_1}\Omega(t_1,t_1)$$

for a 2×2 block which is the exponential of a 2×2 matrix similar in structure to that in Eq. (2.2). As $\Omega(t_1,t_1)$ is the identity matrix, the last equation becomes

$$\Omega(t_2,t_1)=e^{-iH_0t_1}V_2e^{iH_0t_1}.$$

Hence,

$$U(t_2,t_1)=e^{-iH_0(t_2-t_1)}\Omega(t_2,t_1)=e^{-iH_0t_2}V_2e^{iH_0t_1},$$

and

$$U(t_2,0)=U(t_2,t_1)U(t_1,0)=e^{-iH_0t_2}V_2V_1.$$

If a total of k such coherently locked pulses are applied (starting at $t=0$ and lasting for a total period of time $T=t_1+t_2+\cdots+t_k$) we get

$$U(T,0)=e^{-iH_0T}V_kV_{k-1}\cdots V_2V_1, \tag{2.4}$$

where the V_i, $i=1,\ldots,k$ are $M\times M$ matrices with each being the identity except for a 2×2 block, which is the exponential of a 2×2 matrix similar in structure to that in Eq. (2.2), with γ_1 replaced by suitable complex numbers γ_i. If it is desired that $U(T,0)$ be a prescribed matrix V, then the question that needs to be addressed is whether V can be decomposed as in the last equation by a suitable choice of pulses. Equivalently, since e^{-iH_0T} is already known, the question is whether any $M\times M$ unitary matrix can be decomposed as the product $\Pi_{i=1}^{k}V_i$ for some choice of k. If the answer is affirmative, then one can consider optimizing the sequence. This optimization would seek to minimize k, while simultaneously considering the pulse-shaping capabilities of the apparatus.

It is necessary to show that V can be written as $e^{-iH_0T}\Pi_{k=1}^{L}V(\gamma_k)$. Equivalently we have to show that every unitary matrix $\tilde{V}=e^{iH_0T}V$ can be written as $\Pi_{k=1}^{L}V(\gamma_k)$. It will be shown below that this can always be achieved if $\tilde{V}$ is in SU(M). As any $\tilde{W}\in \mathrm{U}(M)$ can be written as $e^{i\Gamma}\tilde{V}$ with Γ a real scalar and $\tilde{V}\in \mathrm{SU}(M)$, this shows that the goal of creating V, up to a scalar phase factor that is irrelevant, has been attained.

In the remainder of the paper we will assume that $V\in \mathrm{SU}(M)$ and denote by V^H its Hermitian conjugate. Towards the end of demonstrating that V has the desired factorization, we premultiply V^H by a product of matrices with each factor in the product being a matrix that is the identity except for a 2×2 block of the form $V(\gamma_k)$, so that the result is the identity $M\times M$ matrix.

Before proceeding to the general situation we first illustrate, via examples, how selection rules are respected in the process of reducing V^H to the identity by left multiplication. Assume that the atom has $M=4$ levels and that the only allowed transitions are $1\rightarrow 2$ and $1\rightarrow 3$ and $1\rightarrow 4$. Any two levels are still accessible from one another, albeit via level 1.

Denoting $V^H=(a_{ij})$, $i,j=1,\ldots,4$, we will first reduce column 4 to $(0,0,0,1)^T$, then column 3 to $(0,0,1,0)^T$, then column 2 to $(0,1,0,0)^T$ and then finally column 1 to $(1,0,0,0)^T$. The reduction will be achieved by premultiplying V^H by suitable matrices that are tensor products of I_2 with $V(\gamma_k)$. Notice that this order is not the only possible one. All that is important is that column 1 be reduced last. In order to appreciate this point, suppose on the contrary that column 1 has been transformed first to $(1,0,0,0)^T$, thereby leaving a matrix of the form

$$\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & a_1 & b_1 & c_1 \\ 0 & a_2 & b_2 & c_2 \\ 0 & a_3 & b_3 & c_3 \end{pmatrix}$$

with the 3×3 matrix on the bottom right corner an element of SU(3). Once this is achieved we will not be able to reduce the remaining columns (if any) to unit columns because level 1 will no longer have any population in it to effect any further intermediate transitions required between the remaining levels.

Returning now to the procedure suggested above. Consider the reduction of column 4 to the corresponding unit column. We may view the fourth column as a particular state of a four-level system. First transfer all of the amplitude from the a_{24} element to the a_{14} element by using the $1\rightarrow 2$ transition. This can be effected by premutiplying V^H by a matrix in SU(4) of the form

$$\begin{pmatrix} a_1 & b_1 & 0 & 0 \\ c_1 & d_1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}.$$

The submatrix

$$\begin{pmatrix} a_1 & b_1 \\ c_1 & d_1 \end{pmatrix}$$

is in SU(2) whose elements encode information about a_{24} and a_{14} (see remark 2.1 below). In the process of this premultiplication, V^H will be transformed into a special unitary matrix with its (2,4) entry equal to 0. Next use the $1\rightarrow 3$ transition to transfer all population from the (3,4) entry of the new matrix to its (1,4) entry. This may be effected by premultiplying the new matrix by a matrix in SU(4) of the form

$$\begin{pmatrix} a_2 & 0 & b_2 & 0 \\ 0 & 1 & 0 & 0 \\ c_2 & 0 & d_2 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}.$$

The submatrix

$$\begin{pmatrix} a_2 & b_2 \\ c_2 & d_2 \end{pmatrix}$$

in SU(2) encodes information about the (3,4) and (1,4) entries of the new matrix, as before. This premultiplication will result in a SU(4) matrix whose (2,4) and (3,4) entries are both zero. Finally the $1\rightarrow 4$ transition is used to transform the the fourth column of the last matrix to the column $(0,0,0,1)^T$. Once again this may be achieved by premutiplying the last matrix by a matrix in SU(4) of the form

$$\begin{pmatrix} a_3 & 0 & 0 & b_3 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ c_3 & 0 & 0 & d_3 \end{pmatrix},$$

where the matrix

$$\begin{pmatrix} a_3 & b_3 \\ c_3 & d_3 \end{pmatrix}$$

in SU(2) is specifically tailored to achieve the desired effect. After this sequence of premultiplications the net effect is a matrix in SU(4) whose last column is $(0,0,0,1)^T$. Unitarity requires that the fourth row be (0,0,0,1). Special unitarity now forces the remaining 3×3 block $\tilde{V}$ to be in SU(3). We now proceed to reduce the third column of $\tilde{V}$ to $(0,0,1)^T$ by making use of transition $1\rightarrow 2$ to transform all the amplitude to level 1, and then using the transition $1\rightarrow 3$ to reduce the third column of the resultant matrix to $(0,0,1)^T$. As before both of these transitions are achieved through premultiplication by matrices in SU(4) whose last column and last row are $(0,0,0,1)^T$ and (0,0,0,1). The balance of these matrices are tensor products of I_1 and the matrix in SU(2) specifically tailored to achieve the desired effects.

Remark 2.1. Let us consider the structure of the matrices in SU(2) that achieves the desired effects above. For example, in nulling out the a_{24} element of V^H, we have several choices for the matrix in SU(2). Let r be the norm of the vector

$$\begin{pmatrix} a_{14} \\ a_{24} \end{pmatrix},$$

and then seek a matrix $V_{12}\in \mathrm{SU}(2)$ such that

$$V_{12}\begin{pmatrix} a_{14} \\ a_{24} \end{pmatrix} = \begin{pmatrix} re^{i\theta} \\ 0 \end{pmatrix}.$$

Usually θ can be taken as arbitrary. Only in the last step in converting any column to the corresponding unit vector does θ have to be zero. Fixing θ leads to a unique V_{12} that will achieve the above result. However, the freedom in the choice of θ (except in the last step) is useful in minimizing the number of $V(\gamma_k)$'s needed. Indeed it will be shown below that any matrix in SU(2) is a product of at most three matrices of the form $V(\gamma_k)$. So the freedom in choosing θ is useful in minimizing the number of $V(\gamma_k)$'s. In fact, there is a matrix of the form $V(\gamma)$ such that

$$V(\gamma)\begin{pmatrix} p \\ q \end{pmatrix} = \begin{pmatrix} \sqrt{|p|^2+|q|^2}e^{i\mu} \\ 0 \end{pmatrix},$$

where the complex numbers p and q have polar representations $p=|p|e^{i\mu}$ and $q=|q|e^{i\zeta}$. Notice that the phase of p is the same as that of

$$V(\gamma)\begin{pmatrix} p \\ q \end{pmatrix}.$$

This will be demonstrated later in remark (2.2). *Thus not insisting that θ be zero (except in one step) reduces the number of $V(\gamma_k)$'s to be used and thus the number of pulses to be applied.*

The general case of an M-level atom can be handled by directly extending the above procedure to express any matrix in SU(M) as a product of matrices that are tensor products of I_{M-2} and an element of SU(2). Thus, it will be shown below that every element of SU(2) can be written as a product of $V(\gamma_k)$'s.

Considering $\gamma=re^{i\phi}$, then the typical $V(\gamma)$ may be expressed as

$$\begin{pmatrix} \cos r & 0 \\ 0 & \cos r \end{pmatrix} + \frac{i\sin r}{r}\begin{pmatrix} 0 & re^{i\phi} \\ re^{-i\phi} & 0 \end{pmatrix}. \tag{2.5}$$

Equation (2.5) is a straightforward consequence of the formula for the exponential of an arbitrary linear combination of the Pauli matrices [equivalently the Darboux coordinates of an SU(2) matrix, [11]]. To be successful in factoring any SU(2) matrix in the desired manner, a first candidate to try would be $e^{i\lambda\sigma_z}$ (i.e., the exponential of the remaining Pauli matrix). Using Eq. (2.5) a simple calculation shows that $e^{i\lambda\sigma_z}=V(\gamma_2)V(\gamma_3)$, with $\gamma_2=(\pi/2)e^{i\sigma_1}$ and $\gamma_3=(\pi/2)e^{i\sigma_2}$, where σ_1 and σ_2 are any real numbers such that $\lambda=\sigma_1-\sigma_2+\pi$. Finally, any $S\in \mathrm{SU}(2)$ admits the following representation:

$$S=\begin{pmatrix} e^{i\lambda}\cos\alpha & e^{i\mu}\sin\alpha \\ e^{i(\pi-\mu)}\sin\alpha & e^{-i\lambda}\cos\alpha \end{pmatrix} \tag{2.6}$$

with λ, $\mu\in[0,2\pi)$ and $\alpha\in[0,\pi/2]$. Equation (2.6) can be obtained by writing the entries of S in polar coordinates and using the definition of SU(2). From Eqs. (2.5) and (2.6) it is easily deduced that $S=V(\gamma_1)e^{i\lambda\sigma_z}$, where $\gamma_1=\alpha e^{i\theta_1}$, $\theta_1=\lambda+\mu-\pi/2$. This constructively yields the desired factorization of SU(2). Note that the pulse area of each of the three pulses is bounded by $\pi/2$ in absolute value.

Remark 2.2. The factorization of SU(2) matrices into factors involving only x and y rotations may also be derived by other techniques [12]. Regardless of the approach it should be noted that S is the unique SU(2) matrix that conveys the vector (1 0) on the sphere (equivalently the spin-up z state) to the vector that is represented by the first column of S. Thus, the factorization of S discussed above may also be viewed as generating this unique transformation using only x and y rotations. Using this perspective it is possible to geometrically derive [12] a sequence of x and y rotations that amounts to a factorization of S. This viewpoint also enables a constructive verification of the claim in remark (2.1). Indeed, S^H is the unique matrix that takes the first column of S to (1 0). It is possible to go from $(e^{ia}\ 0)$ to (1 0) via the matrix $e^{-ia\sigma_z}$ for any real a. The relation shown above $S=V(\gamma_1)e^{i\lambda\sigma_z}$ implies that there is a matrix of the form $V(\gamma)$ that conveys the first column of S be the first column of some SU(2) matrix; thus the claim of remark (2.1) is verified. Since $V(\gamma)$ is $V(\gamma_1)^H$, there is an explicit formula for γ in relation to the entries $(p\ q)$ of the given vector on the sphere, viz., $\gamma=\alpha e^{i\theta}$, where $\cos\alpha=|p|/(|p|^2+|q|^2)$ and $\theta=\mu-\lambda-\pi/2$.

III. APPLICATIONS TO STATE PREPARATION AND OBSERVATION

This section will consider applications of the generation of special unitary matrices to (a) state preparation and (b)

state observation. Case (a) will be discussed from the perspective of controllability [14] and case (b) aims to show that the preparation of certain sequences of special unitary matrices leads to the determination of the state of the system.

A. State preparation

The case of state preparation is implicitly present in a previous paper [14], where the problem of controllability of molecular systems was analyzed by considering the system defined by the corresponding unitary generator. However, a slight refinement is needed to utilize that analysis, since in this paper we have only shown how special unitary matrices may be prepared. Given two vectors u and v on the unit sphere of C^M there is an entire family of unitary matrices U such that $Uu=v$. To construct them we first consider the unitary matrices U_u and U_v, where U_u and U_v have as their first columns the vectors u and v. The remaining columns can be any vectors that render U_u and U_v unitary (e.g., those obtained by applying the Gram-Schmidt process). Clearly $U_u p=u$ and $U_v p=v$, where $p=(1,0,\ldots,0)^T$. Then the matrix $U=U_v U_u^H$ satisfies $U_u=v$.

Now consider the case where the matrix acting on u to give v is desired to be special unitary. Construct $S_u=AU_u^H$ and $S_v=U_vB$ according to $A=\mathrm{diag}(1,e^{i\delta_u},1,\ldots,1)$, where $e^{i\delta_u}=\det U_u$, and $B=\mathrm{diag}(1,e^{-i\delta_v},1,\ldots,1)$ with $e^{i\delta_v}=\det U_v$. Then both S_u and S_v are special unitary and thus the product $S=S_vS_u$ is also special unitary and satisfies $Su=v$, since $Uu=v$. Hence by preparing the special unitary matrix S and acting on the initial state u it is possible to create the desired state v. In most situations $u=(1,0,\ldots,0)^T$.

It would be interesting to examine this approach for initial state creation for low-dimensional systems. The ability to create initial states for low-dimensional systems can be combined with other approaches such as optimal control [2,15,3,4] to create desired states for higher-dimensional systems.

B. State observation

State observation is fundamental to control engineering [16], and it is equally significant for quantum systems [17]. The ability to determine that a desired state has been actually obtained is needed to assess the success of the control process and its use in a variety of applications. We assume that the measurement process can determine the population of some particular level N. Thus, if $\psi_{\mathrm{final}}=\sum_{i=1}^{M}c_i e^{i\phi_i}\psi_i$, where ψ_i, $i=1,\ldots,M$ is some basis (e.g., eigenfunctions of the free Hamiltonian), then we are able to measure $|c_N|^2$. By preparing the special unitary matrix that results in the exchange of populations between levels N and n, it is possible to measure $|c_n|^2$, for all $n=1,\ldots,M$. The only remaining quantities to be determined are the phases ϕ_i, $i=1,\ldots,M$. Since only relative phases are relevant, we assume that the reference phase ϕ_N is 0. Next the special unitary matrix that is the identity except for the following 2×2 block in the (N,n) position is prepared:

$$\frac{1}{\sqrt{2}}\begin{pmatrix}1 & 1\\ -1 & 1\end{pmatrix}.$$

When applied to ψ_{final} it results in a vector that is identical to ψ_{final} except that c_N and c_n are replaced by $\zeta_N=(c_N+c_n)/\sqrt{2}$ and $\zeta_n=(c_N-c_n)/\sqrt{2}$. By assumption, we can read out $|\zeta_N|^2$. Since,

$$|\zeta_N|^2=\tfrac{1}{2}\{|c_N|^2+|c_n|^2+c_Nc_n^*+c_N^*c_n\}$$

it is possible to determine

$$M_n=\frac{|\zeta_N|^2-\frac{1}{2}(|c_N|^2+|c_n|^2)}{2|c_N||c_n|}=\frac{e^{i(\phi_N-\phi_n)}+e^{-i(\phi_N-\phi_n)}}{2}=\cos(\phi_N-\phi_n).$$

However, this does not determine ϕ_n since the cosine function is double valued on $(0,2\pi)$.

To remedy this latter problem we prepare the special unitary matrix that is the tensor product of I_{M-2} and the following 2×2 block in the (N,n) position:

$$\frac{1}{\sqrt{2}}\begin{pmatrix}1 & i\\ i & 1\end{pmatrix}.$$

When applied to ψ_{final}, the transformation results in a vector that is identical to ψ_{final} except that c_N and c_n are replaced by $\zeta_N'=(c_N+ic_n)/\sqrt{2}$ and $\zeta_n'=(ic_N+c_n)/\sqrt{2}$. Once again, by assumption we are able to measure $|\zeta_N'|^2$. Since the quantity

$$M_n'=\frac{-ie^{i(\phi_N-\phi_n)}+ie^{-i(\phi_N-\phi_n)}}{2}=\sin(\phi_N-\phi_n)$$

can be measured, it is clear that $(\phi_N-\phi_n)$ can be determined for each n, and hence so can ϕ_n, $n=1,\ldots,M$.

A total of three $(M-1)$ measurements are required on a sequence of controlled systems for the two $(M-1)$ unknowns $|c_i|^2$, ϕ_i. The above scheme is a special case of determining the elements of a density matrix via projection measurements developed in [17].

IV. CONCLUSIONS

In this paper a constructive scheme for the creation of any special unitary matrix has been analyzed. Although the physical system used here is a single particle interacting with an electromagnetic field, there are some general features to the scheme that are relevant for other quantum systems [18]. These features are as follows: The quantum system in question can be controlled by addressing subsystems of dimension two sequentially; the logarithm of the unitary generator, obtained after the controlling mechanism is applied to any of these two-dimensional subsystems, lies in the span of any two of the Pauli matrices.

The first point above suggests that we represent our desired special unitary matrix as the products of factors that are nontrivial only in a 2×2 block. If no special structure on this

2×2 block is desired, then this can always be achieved [9]. If a special structure is desired, then a preliminary question is what special structures are feasible. The second point yields the answer. The reason is that the Lie algebra generated by any two of the Pauli matrices equals the entire Lie algebra of SU(2). Thus, if the second point is valid for the quantum system being studied, then any matrix in SU(2) can be decomposed, in principle at least, into a product of matrices with the prescribed special structure. The very important practical questions of obtaining this decomposition in an algorithmic fashion and also minimizing the number of factors in the decomposition depend on the particular system. However, if the quantum system is such that in the second point above the logarithm belongs to the span of $i\sigma_x$ and $i\sigma_y$, then the constructive algorithm introduced here is applicable to it as well.

Although quantum computation [5,6] was not the primary focus of this work, its provides useful motivation. Any proposal to fabricate a quantum computer encompasses three steps: (i) preparation of the initial state that will serve as the input; (ii) preparation of the unitary transformation that will function as the logic gate; and (iii) reading the final output. With every concrete protocol proposed for a quantum computer the physical means for achieving these three steps must be specified. In addition the nature of each step depends on the particular problem or quantum algorithm. This paper considered a single atom (i.e., a particle in the limit of one electron) as a realization. There are many practical limitations associated with this choice (as is the case with any other choice thus far presented), but there are also strong arguments to suggest that an atom would be an ideal system for practical laboratory study. Single particles are attractive from the point of view of isolating the system from external influences, and thereby enhancing the lifetime (provided states of sufficient radiative lifetime are involved). Furthermore, both the theoretical and experimental aspects of controlling molecular and atomic dynamics are advancing very rapidly. All three steps above can be subsumed into the problem of creating a prescribed unitary matrix from the evolution of a controlled atom. It is possible to achieve entanglement within a single molecule by simultaneously manipulating two degrees of freedom, such as rotation and vibration, or radial and angular degrees of freedom in a single atom, etc. A further critical matter concerns the structure of the algorithms used in quantum computation. The current algorithms are designed to work with multiple particles. However, this circumstance does not preclude working with multilevel systems. The nature of the state observation is also very important. Section III considered full state observation, but other less demanding observations are desirable in keeping with the quantum computation algorithms proposed so far. These topics go beyond the scope of this paper, but the work here should provide motivation for exploring these questions further.

Finally, it would be interesting to examine the field of molecular control [2,15] from the perspective of this paper. Most problems in molecular control can be cast as state preparation for multilevel systems. Section III showed that this goal can be subsumed into the problem of creating one out of a family of special unitary matrices. The main feature of our algorithm is that it does not require any costly computations, although the pulse sequence still needs to be determined.

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[1] R. Gordon and S. Rice, Annu. Rev. Phys. Chem. **48**, 601 (1997).
[2] W. Warren, H. Rabitz, and M. Dahleh, Science **259**, 1581 (1993).
[3] K. Wilson *et al.*, J. Phys. Chem. **97**, 2320 (1995).
[4] A. Butkovsky and Y. Samoilenko, Control of Quantum Mechanical Processes and Systems (Kluwer Academic, Dordrecht, 1990).
[5] R. Feynman, Optics News **11**, 11 (1985).
[6] P. Shor, in *Proceedings of the 25th Annual ACM Symposium on Foundations of Computer Science*, edited by S. Goldwasser (IEEE Computer Society, Los Alamitos, CA, 1994), p. 124.
[7] A. Sudberry, Nature (London) **362**, 586 (1993).
[8] M. Reck, A. Zeilinger, H. Bernstein, and P. Bertani, Phys. Rev. Lett. **73**, 58 (1994).
[9] F. Murnaghan, The Unitary and Rotation Groups (Spartan, New York, 1960).
[10] C. Law and J. Eberly, Phys. Rev. Lett. **76**, 1055 (1996).
[11] W. G. Harter, *Principles of Symmetry, Dynamics and Spectroscopy* (Wiley-Interscience, New York, 1993).
[12] W. Harter (private communication).
[13] P. DiVincenzo, Phys. Rev. A **51**, 1015 (1995).
[14] V. Ramakrishna, M. Salapaka, M. Dahleh, H. Rabitz, and A. Peirce, Phys. Rev. A **51**, 960 (1995).
[15] S. Shi and H. Rabitz, J. Chem. Phys. **92**, 364 (1990).
[16] J. Zabcyk, Mathematical Control Theory (Birkhauser, Basel, Switzerland, 1990).
[17] O. Steuernagel and J. Vaccaro, Phys. Rev. Lett. **75**, 3201 (1995).
[18] V. Ramakrishna, R. Ober, and H. Rabitz, in *Proceedings of the Mathematical Theory of Networks and Systems Conference*, Padova, Italy, 1998, edited by A. Beghi, L. Finnesso, and G. Picci (Il Poligrafo, Padova, Italy, 1998).