Molecular Quantum Computing by an Optimal Control Algorithm for Unitary Transformations

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Abstract

Quantum computation is based on implementing selected unitary transformations which represent algorithms. A generalized optimal control theory is used to find the driving field that generates a prespecified unitary transformation. The approach is illustrated in the implementation of one and two qubits gates in model molecular systems.

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A universal model of a quantum computer can be constructed from an array of two level systems used as registers (qubits). Any general quantum gate can be decomposed to a one and two qubit unitary transformation \cite{1}. Thus a physical realization of a quantum computer should be able control such unitary transformations by use of external driving fields. Realizations based on nuclear magnetic resonance techniques already have been demonstrated for non-trivial quantum gates. For example, a three qubit quantum Fourier transform has been performed by a sequence of pulses corresponding to one and two qubit transformations \cite{2}. This implementation was based on addressing each qubit independently by their spectral separation. However, when many qubits become involved in a computation, the spectrum becomes congested so that it becomes more difficult to address each qubit individually. The total fidelity of the algorithm will depend on the accumulation of errors at each step. Another difficulty results from decoherence processes which are unavoidable due to coupling to the external environment. These processes will degrade the performance in proportion to the time required to carry out the computation task. Thus it is desirable to minimize the number of computation steps and the total computation time.

An alternative possibility for implementing quantum computing is the use of molecules driven by shaped light pulses. A hypothetical model can be based on using the vibrational and rotational states as registers. A shaped light pulse can write data as amplitudes on these states. The computing algorithm is then implemented by performing a unitary transformation employing a second shaped pulse. In the end the output can be read by a probe pulse. This approach implies changing the computation model by tying together many single and two qubit operations into one resulting in a combined unitary transformation.

Such an approach has been used for the experimental implementation of elements of coherent computation in Li$_2$ \cite{3,4}. For example, two states of the ro-vibrational manifold of an electronic state can form a single qubit. By taking advantage of multiple electronic states, the evolution of the molecular system can be controlled using electromagnetic fields in the optical region realized by shaped laser pulses. The fast development of pulse shaping technology can make possible the implementation of simple quantum gates in molecules. This program has to overcome interference from the large number of other molecular levels coupled to the field but not assigned to the qubits. The task becomes therefore to implement the quantum algorithm on the molecular levels used as registers while avoiding the intervention of other states from the same system.
Tesh et. al. [5], proposed the use of optimal control theory (OCT) to calculate the field which can induce a specific transformation used for quantum computation. Originally, OCT was designed as a method to obtain a light field which could induce a specific state to state transformation [6, 7]. In the quantum computing context, the goal is to obtain the optimal pulse that induces a given unitary transformation, irrespective of the initial state of the system. The present approach generalized OCT to obtain directly the driving field that induces a target unitary transformation in the system. The approach is based on the equation of motion of the unitary transformation by making use of the main ingredients of OCT. This allows the utilization of the large number of tools that have been developed in this field.

The model system consists of a free Hamiltonian $\hat{\mathbf{H}}_0$ controlled by an external field $\epsilon(t)$,

$$
\hat{\mathbf{H}}(t) = \hat{\mathbf{H}}_0 - \hat{\mathbf{\mu}} \epsilon(t), \tag{1}
$$

where $\hat{\mathbf{\mu}}$ is a system operator. In the molecular system, $\hat{\mathbf{\mu}}$ is the transition dipole operator and $\epsilon(t)$ describes a shaped short light pulse. For simplicity, the field $\epsilon(t)$ is assumed real but the generalization to an electromagnetic field with two independently controlled polarizations [8] is straightforward.

The algorithm is represented in the target time $T$ by a unitary transformation $\hat{\mathbf{U}}(T)$ generated from the Hamiltonian Eq. (1);

$$
\frac{\partial\hat{\mathbf{U}}(t)}{\partial t} = -\frac{i}{\hbar} \hat{\mathbf{H}}(t) \hat{\mathbf{U}}(t), \tag{2}
$$

with the initial condition $\hat{\mathbf{U}}(t = 0) = 1$, where $1$ denotes the identity operator. The objective is to obtain the optimal driving field $\epsilon(t)$ that induces a given unitary transformation $\hat{\mathbf{O}}$ at $t = T$, i.e., $\hat{\mathbf{U}}(T) = e^{i\phi} \hat{\mathbf{O}}$. $\phi$ denotes a physically irrelevant global phase that is related to the energy origin and is therefore uncontrollable by the field.

The task is a typical inversion problem which can be solved by employing a variational procedure maximizing the projection of the generated operator on the target operator,

$$
|\tau| = |\text{Tr}\{\hat{\mathbf{O}}^\dagger \hat{\mathbf{U}}(T)\}|, \tag{3}
$$

where the projection $(\hat{\mathbf{A}} \cdot \hat{\mathbf{B}})$ is defined by $\text{Tr}\{\hat{\mathbf{A}}^\dagger \hat{\mathbf{B}}\}$. The functional $\tau$ is a complex number inside the circle $|\tau| \leq N_H$, where $N_H$ is the dimension of the Hilbert space of the system. Equality is reached only when the argument of the trace is $e^{i\phi} 1$ and then a maximum
of $|\tau|$ is equivalent to $\hat{U}(T) = e^{i\phi}\hat{O}$. The optimal solution is then found by maximizing the functional Eq. (3), with respect to the control field $\epsilon(t)$. Since a direct algorithm to maximize $|\tau|$ was not found, a working alternative is used based on formulating the problem as the optimization of $\text{Re}[\tau]$, or of $\text{Im}[\tau]$, or of a linear combination of both. For simplicity, the optimization of the real part represented by the functional $J = \text{Re}[\text{Tr}\{\hat{O}^\dagger\hat{U}(T)\}]$ is considered. Two constraints are introduced [6, 7], the first restricts the dynamics to obey the Schrödinger equation Eq. (2), and the second restricts the total field energy. Using Lagrange multipliers a modified functional is obtained,

$$\bar{J} = \text{Re} \left[ \tau - \int_0^T \text{Tr} \left\{ \left( \frac{\partial \hat{U}(t)}{\partial t} + \frac{i}{\hbar} \hat{H}(t) \hat{U}(t) \right) \hat{B}(t) \right\} dt \right] - \lambda \int_0^T \frac{1}{s(t)}|\epsilon(t)|^2 dt, \quad (4)$$

where $\hat{B}(t)$ is an operator Lagrange multiplier, $\lambda$ is a scalar Lagrange multiplier and $s(t)$ is a shape function which turns the pulse on and off [8]. The use of more elaborate constraints and choices of $\lambda$, allows a higher degree of control on the shape of the optimal pulse [10, 11].

Applying the calculus of variations, $\delta \bar{J} = 0$ with respect to $\hat{B}$, $\hat{U}$, and $\epsilon$, a set of equations is obtained: a) The Schrödinger equation Eq. (2) with the initial condition $\hat{U}(t = 0) = \mathbb{1}$ for $\hat{U}$; b) The inverse Schrödinger equation

$$\frac{\partial \hat{B}(t)}{\partial t} = \frac{i}{\hbar} \hat{B}(t) \hat{H}(t) \quad (5)$$

with the condition $\hat{B}(t = T) = \hat{O}^\dagger$ for $\hat{B}$; c) The field equation:

$$\epsilon(t) = -\frac{s(t)}{2\lambda \hbar} \text{Im}[\text{Tr}\{\hat{B}(t) \hat{\mu} \hat{U}(t)\}], \quad (6)$$

Eq. (2) and (3) represent two counter currents with information from the initial condition and the target unitary transformation respectively. The equations are solved iteratively, the Krotov method [12], similar to the methods described in Ref. [13], was found to be the most efficient. The input is a “guess” field, $\epsilon^{(0)}(t)$, so that in the $k$ iteration ($k = 1, 2,...$): (i) $\hat{B}^{(k-1)}(t)$ is propagated backwards from $t = T$ to $t = 0$ using Eq. (5) and $\epsilon^{(k-1)}$; and (ii) $\hat{U}^{(k)}(t)$ is propagated forward using Eq. (2) and $\epsilon^{(k)}$ is evaluated using

$$\epsilon^{(k)}(t) = -\frac{s(t)}{2\lambda \hbar} \text{Im}[\text{Tr}\{\hat{B}^{(k-1)}(t) \hat{\mu} \hat{U}^{(k)}(t)\}]. \quad (7)$$

The procedure is repeated until the desired convergence has been reached. The hard numerical task is the propagation of the operators $\hat{U}$ and $\hat{B}$ with the time dependent Hamiltonian for which a second order Newton polynomial integrator [14] was used.
A direct use of Eq. (7) in the algorithm leads to saturation. This is because the constraint related with the field energy becomes more important than the original objective. A remedy is to interpret the right hand side of Eq. (7), denoted by $\Delta \epsilon^{(k)}$, as a correction of the field in the previous interaction [15]. The field after the $k$ iteration is then given by $\epsilon^{(k)}(t) = \epsilon^{(k-1)}(t) + \Delta \epsilon^{(k)}(t)$. The OCT procedure was first applied to a 1 qubit operation the Hadamard rotation (Eq. [10]). In a similar fashion, 2-qubit operation such as the “controlled not” were obtained. The optimal fields generating unitary operations with eight (3-qubits) were also attempted.

It was found that the number of iterations required to converge the results increased approximately exponentially (or factorially) with the number of states in the problem. A possible reason for this scaling is that not only a specific state to state transition has to be forced but this has to be carried out without disturbing the other state to state transitions in $\hat{U}$. As a result the scaling becomes $O(n!)$ which is consistent with numerical experience.

In a molecular environment, obtaining the optimal field to carry out an algorithm is more involved. The register levels which are used to write the input and output are only part of a much larger manifold of molecular energy levels. Considering the advances in pulse shaping techniques in the visible region of the spectrum the transitions of choice are electronic. For such a molecular construction, imposing a unitary transformation on the total set of levels which are addressed by the field is too restrictive. Relying on the experience that the convergence is close to factorial, an extremely large number of iterations would be required to converge.

The strategy is therefore to restrict the target objective to only the states used directly as registers keeping the other states in the system as passive observers. The reduced objective is obtained by changing the previous expressions $\text{Tr}\{\hat{A}\hat{B}\}$ to $\sum_i \langle i|R\hat{A}\hat{B}|i\rangle_R$, where $\{|i\rangle_R\}$ is a basis of the subspace of registers. The subindex $R$ is used to denote the operators in that subspace, for example $\hat{O}_R$ denotes the target unitary transformation. The substitution of the restricted condition instead of the trace in Eq. (3) keeps the maximum condition when the unitary transformation $\hat{U}_R(T)$ is equal to $\hat{O}_R$. The maximum value becomes equal to the dimension of the subspace $N_R$. In the condition for $\hat{B}$ at time $T$, a particular dependence must be specified for all the levels. For simplicity the identity in the passive subspace has been chosen.

The approach is illustrated using two models. For the first model, the implementation of a unitary transformation in one qubit while minimizing the population transfer to other
levels in the molecule is studied. The model consists of a molecule with two electronic surfaces described by the Hamiltonian,

$$\hat{H} = \begin{pmatrix} \hat{H}_g & -\hat{\mu}(t) \\ -\hat{\mu}(t) & \hat{H}_e \end{pmatrix}, \quad (8)$$

where $\hat{H}_g$ and $\hat{H}_e$ are the ground and excited surface Hamiltonians. The electronic surfaces are coupled by the transition dipole operator $\hat{\mu}$, controlled by the shaped field $\epsilon(t)$. The two first levels of the ground electronic surface are chosen as the registers representing the qubit. The model includes 15 rovibronic levels in the ground electronic state and 5 in the excited state described by the Hamiltonians,

$$\hat{H}_g = \sum_{i=1}^{15} E_{gi} |g_i\rangle\langle g_i|; \quad \hat{H}_e = \sum_{j=1}^{5} E_{ej} |e_j\rangle\langle e_j|.$$  \quad (9)

shown in Fig. 1. Next, a transition dipole operator with equal coupling strength between levels was chosen, $\hat{\mu} = \mu_0 \sum_{i=1}^{15} \sum_{j=1}^{5} |g_i\rangle\langle e_j| + \text{H.c.}$, where H.c. denotes the Hermitian conjugate.

In Fig. 1 the optimized field is shown when the target unitary transformation is a Hadamard rotation given by,

$$\hat{O}_H^R = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix}, \quad (10)$$

restricted to the $2 \times 2$ qubit subspace. The shape function was chosen as $s(t) = \sin^2(2\pi t/T)$ and the initial guess was $\epsilon(0)(t) = s(t) \cos(\Omega t)$, where $\Omega$ is the frequency of the 00-line of the electronic transition. Fig. 1 shows that the dominant frequencies are the ones related to the known vibronic molecular transitions. In the time domain the field is split into a symmetric sequence of sub pulses. A phase relation correlating the dominant frequencies is observed in a Wigner plot (not shown). These phase relations guarantee that no population is lost to the excited levels.

In the second model the two electronic surfaces, Eq. (8), have two orthogonal vibrational modes, denoted as $\alpha$ and $\beta$. The first two levels of each vibrational mode in the ground electronic surface are chosen as the the physical implementation of the two qubits. The goal is to induce an operation that involves entanglement between them. In this model each vibrational mode of the ground surface is coupled by the field to the corresponding
FIG. 1: (a) Schematic representation for executing the Hadamard rotation. Units are chosen such \( \hbar = 1 \). The solid lines represent the two levels associated with the qubit. The levels in the ground surface are equally spaced with \( \omega_g = 1 \) (for Li\(_2\) in the A state \( \omega_g = 255 \text{ cm}^{-1} \)) and in the excited surface with \( \omega_e = 0.9 \) (for Li\(_2\) in the E state \( \omega_e = 241 \text{ cm}^{-1} \)). The other model parameters are \( \Omega = 15 \) (for Li\(_2\) the A to E transitions \( \Omega = 12400 \text{ cm}^{-1} \)) and \( \mu_0 = 0.1 \). The arrows are two of the possible transitions induced by the driving field. (b) Optimized field that induces a Hadamard rotation in the qubit at \( T = 70 \) (for Li\(_2\) \( T \sim 1.5 \text{ psec} \)). (c) Fourier transform of the field.

mode in the excited electronic surface. The modes in the excited surface are coupled by a static term modeling Duschinsky rotation. This last term which is not controlled by the field can generate entanglement between the qubits. The model consists of two levels for each vibrational normal mode denoted by \( \alpha \) and \( \beta \). The electronic surface Hamiltonians are

\[
\hat{H}_g = \hat{H}_{g\alpha} \otimes \mathbb{1}_\beta + \mathbb{1}_\alpha \otimes \hat{H}_{g\beta} \quad \text{and} \quad \hat{H}_e = \hat{H}_{e\alpha} \otimes \mathbb{1}_\beta + \mathbb{1}_\alpha \otimes \hat{H}_{e\beta} + \hat{V}_{\alpha\beta}
\]

with,

\[
\hat{H}_{g\nu} = (E_{g\nu}|g_0\rangle_\nu \langle g_0|_\nu + E_{g\nu}|g_1\rangle_\nu \langle g_1|_\nu),
\]

\[
\hat{H}_{e\nu} = (E_{e\nu}|e_0\rangle_\nu \langle e_0|_\nu + E_{e\nu}|e_1\rangle_\nu \langle e_1|_\nu),
\]

\[
\hat{V}_{\alpha\beta} = \delta_{\alpha\beta} (|e_0\rangle_\alpha \otimes |e_1\rangle_\beta \langle e_1|_\beta \otimes |e_0\rangle_\alpha + |e_1\rangle_\alpha \otimes |e_0\rangle_\beta \langle e_0|_\beta \otimes |e_1|_\alpha),
\]

(11)
with $\hat{V}_{\alpha\beta}$ the Duschinsky term that couples the vibrational modes in the excited surface. A schematic representation of the levels is given in Fig. 2. The transition dipole operator is chosen as $\hat{\mu} = \hat{\mu}_\alpha \otimes 1_\beta + 1_\alpha \otimes \hat{\mu}_\beta$, with $\hat{\mu}_\nu = \mu_\nu (|e_0\rangle_\nu + |e_1\rangle_\nu)(\langle g_0|_\nu + \langle g_1|_\nu) + \text{H.c.}$ Operators in the combined Hilbert space of this system are represented by $16 \times 16$ matrices.

The target unitary transformation is a two qubits quantum Fourier transform $\hat{O}^{FT}_R$, at $T = 320$.

The target unitary transformation is a two qubits quantum Fourier transform $\hat{O}^{FT}_R$, at $T = 320$.

\[
\hat{O}^{FT}_R = \frac{1}{2} \begin{pmatrix} 1 & 1 & 1 & 1 \\ 1 & i & -1 & -i \\ 1 & -1 & 1 & -1 \\ 1 & -i & -1 & i \end{pmatrix}
\]

\__(12)\_
where the operator is represented in the basis $|g_i\rangle_\alpha \otimes |g_j\rangle_\beta$. Notice that operators in the subspace of interest are represented by $4 \times 4$ matrices. The spectra of the field resulting from the optimization is shown in Fig. 2. The frequencies are confined in the region of the vibronic transitions. The structure of the spectra is much more complex than in the previous model. An obvious reason is the larger dimension of the subspace of interest. In addition the complicated mechanism required to create entanglement is indirect. During the pulse significant population is transferred to the excited electronic states. The resulting more complex field is reflected by also in the Wigner distribution, where the field shows a very high degree of correlation between time and frequency.

In conclusion, Optimal Control Theory has been generalized to obtain the driving field that generates any target unitary transformation. In principle the scheme allows the implementation of any quantum gate using a single step. Compared to the implementation of the gate using a sequence of known simple pulses, the loss by the possible complexity of the shaped pulse can be more than compensated by the faster implementation. Convergence is efficient for a small number of levels. Besides, with the right choice of the guess field, the method can obtain the driving field for more complex systems where a large number of levels are involved and the pulses for simple operations are unknown, as in molecular systems. The advantage of such systems is the short time in which these algorithms can be executed. Using Li$_2$ as an example, the Hadamard rotation can be executed in $\sim 1.5$ psec. The 2-qubit Fourier transform could be executed in a molecule for example OCS in approximately $\sim 6$ psec. These fast timescales give hope that the quantum computation can be carried out before decoherence processes take place.

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