Quantum Computing by an Optimal Control Algorithm for Unitary Transformations

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Quantum computation is based on implementing selected unitary transformations representing algorithms. A generalized optimal control theory is used to find the driving field that generates a prespecified unitary transformation. The approach is independent of the physical implementation of the quantum computer and it is illustrated for one and two qubit gates in model molecular systems, where only part of the Hilbert space is used for computation.

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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 into a one and two qubit unitary transformation [1]. Thus a physical realization of a quantum computer should be able to control such unitary transformations by the use of external driving fields. 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 over 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. To determine the field that induces a given unitary transformation turns to be a crucial task when the number of qubits and higher spurious levels increases.

Tian and Lloyd [2] propose a method to avoid the undesired transition to the higher levels when the external driving field that induces the unitary transformation in the qubit subspace is known. Tesch \textit{et al.} [3] proposed the use of optimal control theory (OCT) to calculate the field which can induce a specific transformation in the context of molecular quantum computation. Originally, OCT was designed as a method to obtain a light field which could induce a specific state to state transformation [4,5]. 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, taking in account all the qubits and higher spurious levels coupled to it. 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 $\hat{H}_0$ controlled by an external field $\epsilon(t)$,

\begin{equation}
\hat{H}(t) = \hat{H}_0 - \hat{\mu} \cdot \epsilon(t),
\end{equation}

where $\hat{\mu}$ is a system operator. In a molecular system, $\hat{\mu}$ 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 [6] is straightforward.

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

\begin{equation}
\frac{\partial \hat{U}(t)}{\partial t} = -\frac{i}{\hbar} \hat{H}(t)\hat{U}(t),
\end{equation}

with the initial condition $\hat{U}(t = 0) = \mathbb{1}$, where $\mathbb{1}$ denotes the identity operator. The objective is to obtain the optimal driving field $\epsilon(t)$ that induces a given unitary transformation $\hat{O}$ at $t = T$, i.e., $\hat{U}(T) = e^{i\phi} \hat{O}$. $\phi$ 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,

\begin{equation}
|\tau| = |\text{Tr}[\hat{O}^\dagger \hat{U}(T)]|,
\end{equation}

where the projection $(\hat{A} \cdot \hat{B})$ is defined by $\text{Tr}[\hat{A}^\dagger \hat{B}]$. 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 $e^{i\phi} \mathbb{1}$ and then a maximum of $|\tau|$ is equivalent to $\hat{U}(T) = e^{i\phi} \hat{O}$. Then $|\tau|/N_H$ gives the fidelity of the implementation of the unitary transformation.

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: optimization of $\text{Re}[\tau]$, or of
Im[\tau], or both. For simplicity, the optimization of the real part represented by the functional \( J = \text{Re}[\text{Tr}[\hat{\mathbf{O}}^\dagger \hat{\mathbf{U}}(T)]] \) is considered. Two constraints are introduced [4,5], the first restricts the dynamics to obey the Schrödinger equation (2), and the second restricts the total field energy. Using Lagrange multipliers a modified functional is obtained,

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

(4)

where \( \hat{\mathbf{B}}(t) \) 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 [7]. The use of more elaborate constraints and choices of \( \lambda \) allows a higher degree of control on the shape of the optimal pulse [8].

Applying the calculations of variations, \( \delta J = 0 \) with respect to \( \hat{\mathbf{B}}, \hat{\mathbf{U}}, \) and \( \epsilon \), a set of equations is obtained: (a) the Schrödinger equation (2) with the initial condition \( \hat{\mathbf{U}}(T) = 1 \) for \( \hat{\mathbf{U}} \); (b) the inverse Schrödinger equation

\[
\frac{\partial \hat{\mathbf{B}}(t)}{\partial t} = \frac{i}{\hbar} \hat{\mathbf{B}}(t) \hat{\mathbf{H}}(t)
\]

(5)

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

\[
\epsilon(t) = -\frac{s(t)}{2\lambda} \text{Im}[\text{Tr}[\hat{\mathbf{B}}(t) \hat{\mu} \hat{\mathbf{U}}(t)]].
\]

(6)

Equations (2) and (5) represent two counter currents with information from the initial condition and the target unitary transformation, respectively. The equations are solved iteratively. The Krotov method [9], similar to the methods described in Ref. [10], was found to be the most efficient. The input is a "guess" field, \( \epsilon^{(0)}(t) \), so that in the \( k \) iteration (\( k = 1, 2, \ldots \) ): (i) \( \hat{\mathbf{B}}^{(k-1)}(t) \) is propagated backwards from \( t = T \) to \( t = 0 \) using Eq. (5) and \( \epsilon^{(k-1)} \); and (ii) \( \hat{\mathbf{U}}^{(k)}(t) \) is propagated forward using Eq. (2) and \( \epsilon^{(k)} \) is evaluated using

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

(7)

The procedure is repeated until the desired convergence has been reached. The hard numerical task is the propagation of the operators \( \hat{\mathbf{U}} \) and \( \hat{\mathbf{B}} \) with the time dependent Hamiltonian for which a second order Newton polynomial integrator [11] was used.

A direct use of Eq. (7) in the algorithm leads to saturation. This is because the constraint related to 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 [12]. 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 one qubit operation, the Hadamard rotation [Eq. (10)]. In a similar fashion, a two qubit operation such as the “controlled NOT” was obtained. The optimal fields generating unitary operations with eight states (three qubits) were also attempted.

It was found that the number of iterations required to converge the results increased at least exponentially (or factorially) with the dimension of the Hilbert space 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 \( \hat{\mathbf{U}} \). 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 \( \text{Tr}[\hat{\mathbf{B}}] \) to \( \sum_{R} \langle \hat{\mathbf{B}}_{R} \rangle_{R} \), where \( \langle \hat{\mathbf{B}}_{R} \rangle_{R} \) is a basis of the subspace of registers. The subindex \( R \) is used to denote the operators in that subspace, for example, \( \hat{\mathbf{O}}_{R} \) denotes the target unitary transformation. The substitution of the restricted condition instead of the trace in Eq. (3), denoted as \( \tau_{R} \), keeps the maximum condition when the unitary transformation \( \hat{\mathbf{U}}_{R}(T) \) is equal to \( \hat{\mathbf{O}}_{R} \). The maximum value becomes equal to the dimension of the subspace \( N_{R} \), and the fidelity of the restricted problem is given by \( \tau_{R}/N_{R} \). In the condition for \( \hat{\mathbf{B}} \) 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 molecular models. Molecules driven by shaped light pulses are an alternative possibility for implementing quantum computing [13,14]. Such an approach has been used for the experimental implementation of elements of coherent computation in \( \text{Li}_2 \) [15].

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,

\[
\hat{\mathbf{H}} = \begin{pmatrix}
\hat{\mathbf{H}}_{g} & -\hat{\mu} \epsilon(t) \\
-\hat{\mu} \epsilon(t) & \hat{\mathbf{H}}_{e}
\end{pmatrix},
\]

(8)

where \( \hat{\mathbf{H}}_{g} \) and \( \hat{\mathbf{H}}_{e} \) are the ground and excited surface
Hamiltonians. The electronic surfaces are coupled by the transition dipole operator \( \hat{\mu} \), controlled by the shaped field \( e(t) \). The first two 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 five in the excited state described by the Hamiltonians,

\[
\hat{H}_g = \sum_{i=1}^{15} E_{gi} |g_i\rangle \langle g_i|, \quad \hat{H}_e = \sum_{j=1}^{5} E_{ej} |e_j\rangle \langle e_j|, \tag{9}
\]

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 \( \text{H.c.} \) denotes the Hermitian conjugate.

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

\[
\hat{\mathcal{O}}^H_R = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix}, \tag{10}
\]

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 \( e^{(0)}(t) = s(t) \cos(\Omega t) \), where \( \Omega \) is the frequency of the 00 line of the electronic transition. The target fidelity obtained \( 1 - |\tau_R|^2/N_R < 10^{-6} \). Figure 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 subpulses. 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 [12].

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 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 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 \( \hat{H}_g = \hat{H}_{g\alpha} \otimes \mathbb{1}_\beta + \mathbb{1}_\alpha \otimes \hat{H}_{g\beta} \) and \( \hat{H}_e = \hat{H}_{e\alpha} \otimes \mathbb{1}_\beta + \mathbb{1}_\alpha \otimes \hat{H}_{e\beta} + \hat{V}_{\alpha\beta} \)

with \( \hat{V}_{\alpha\beta} \) 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 \( \hat{\mu} = \hat{\mu}_\alpha \otimes \mathbb{1}_\beta + \mathbb{1}_\alpha \otimes \hat{\mu}_\beta \), with \( \hat{\mu}_\alpha = \mu_0 \langle \langle e_0| \rangle \langle e_0| \rangle + \langle e_1| \rangle \langle e_1| \rangle + \text{H.c.} \). 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 [16],

\[
\hat{\mathcal{O}}^{\text{FT}}_R = \frac{1}{2} \begin{pmatrix} 1 & 1 & 1 & 1 \\ 1 & i & -1 & -i \\ 1 & -i & 1 & -1 \\ 1 & -i & -1 & i \end{pmatrix}, \tag{12}
\]

where the operator is represented in the basis \( |g_i\rangle \otimes |e_j\rangle \). Notice that operators in the subspace of interest are represented by \( 4 \times 4 \) matrices. The spectrum of the field resulting from the optimization is shown in Fig. 2. The target fidelity obtained \( 1 - |\tau_R|^2/N_R < 10^{-6} \). The frequencies are confined in the region of the vibronic transitions. The structure of the spectrum 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 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. The known sequence can always be used as a guess field and OCT will always improve the result. The advantage of molecular 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 two qubit Fourier transform could be executed in a molecule, for example, OCS in approximately $\sim 6$ psec. As the decoherence time scale is on the order of nsec, these fast time scales give hope that the quantum computation can be carried out before decoherence processes take place.

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