Quantum computing based on vibrational eigenstates: Pulse area theorem analysis

Taiwang Cheng and Alex Brown[a)]
Department of Chemistry, University of Alberta, Edmonton, Alberta T6G 2G2, Canada

(Received 21 July 2005; accepted 13 December 2005; published online 19 January 2006)

In a recent paper [D. Babikov, J. Chem. Phys. **121**, 7577 (2004)], quantum optimal control theory was applied to analyze the accuracy of quantum gates in a quantum computer based on molecular vibrational eigenstates. The effects of the anharmonicity parameter of the molecule, the target time of the pulse, and the penalty function on the accuracy of the qubit transformations were investigated. We demonstrate that the effects of all the molecular and laser-pulse parameters can be explained utilizing the analytical pulse area theorem, which originates from the standard two-level model. Moreover, by analyzing the difference between the optimal control theory results and those obtained using the pulse area theorem, it is shown that extremely high quantum gate fidelity can be achieved for a qubit system based on vibrational eigenstates. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2164457]

I. INTRODUCTION

The field of quantum computing[1–3] has emerged as an intriguing and exciting new research area that physicists, mathematicians, engineers, and chemists have started to explore. In order to realize a quantum computer several problems need to be addressed: the identification of a physical system to represent quantum bits (qubits), the maintenance of coherence, and the implementation of mechanisms for performing quantum logic gate operations on the qubits. While several different physical systems have been proposed or utilized to realize quantum computing algorithms, e.g., NMR,[4–6] recent interest has turned to using the vibrational states of molecules to represent the qubits and tailored femtosecond laser pulses to implement quantum gate operations.[7–18] It has been shown that the vibrational, or rovibrational, states of molecules are reasonable candidates for encoding quantum information and applying quantum information processing. When using the vibrational states of a molecule as the qubit basis, the number of available qubits is proportional to the number of vibrational degrees of freedom: $3N-6$ for a nonlinear molecule containing N atoms. Current physical systems studied for molecular quantum computing have been small gas-phase molecules for one-, two-, and three-qubit problems. For example, there have been theoretical explorations in model diatomics,[7] I_2,[13,14,18] C_2H_2,[8–11] NH_3,[12] and an experimental study for Li_2.[15] Decoherence for molecules in the gas phase can be due to collisions, coupling between vibrational and other degrees of freedom (i.e., electronic or rotational), and, for polyatomic molecules, resonances between vibrational modes. In general, the number of collisions can be kept very low and the typical lifetimes of vibrations are orders of magnitude greater than the durations of the femtosecond pulses used to perform the gate operations. The quantum gate operations on the vibrational qubits are performed using shaped ultrashort laser pulses, which are usually determined theoretically by applying the optimal control theory[8,16,17,19] (OCT) although other proposals have been made, e.g., time-frequency resolved coherent anti-Stokes Raman scattering.[13,14]

One of the most used algorithms within the optimal control theory as applied to molecular processes is the one developed by Zhu *et al.*[19] Their algorithm is an extension of the one due to Krotov.[20] The method is based on a forward-backward iterative scheme to improve the control field, and it has been shown[19] that this method exhibits quadratic and monotonic convergences. This method has proven to be very efficient and thus is widely used when treating the problem of steering a quantum system from an initial state to final states,[21–23] i.e., state-to-state control. Tesch and de Vivie-Riedle[8] have generalized the original algorithm[19] into a multitarget formulation, which can find the optimized field for steering simultaneously a set of initial pure states to a set of final states. Such a generalization has direct applicability in quantum computing, where an algorithm implemented as a unitary transformation operating on a set of states has to be carried out irrespective of the input. In this application, both input and output are encoded as a superposition of these states.[16]

Using the multitarget formulation of the optimal control theory, de Vivie-Riedle and co-workers[8–11] have examined extensively quantum logic gate operations implemented for a two-qubit model system based on vibrational normal modes in C_2H_2. Their two-qubit model is based on the asymmetric CH-stretching mode and the *cis*-bending mode. They have shown that pulses can be designed to implement one- and two-qubit operations, i.e., local and global NOT gates, Hadamard gate, and CNOT gate, with reasonable high gate fidelities. Of course, in a polyatomic molecule such as C_2H_2, decoherence due to coupling with other vibrational states outside those composing the qubits is of critical importance and it has been demonstrated that these processes can be

[a)]Electronic mail: alex.brown@ualberta.ca

suppressed by optimized laser pulses.[10] A similar theoretical calculation of tailored pulses for realizing Hadamard and CNOT gate operation in a two-dimensional model of NH_3 has recently been published.[12] The studies involving C_2H_2 and NH_3 have cast the problem in terms of wave functions. Recently, Ohtsuki[18] has formulated the design of gate pulses using the optimal control theory within the density-matrix formalism. The formulation allows the inclusions of relaxation (decoherence effects), although relaxation was neglected in the initial study of I_2.[18] These theoretical studies have highlighted the feasibility of using molecular vibrational states for quantum computing and have addressed, or begun to address, some of the major concerns, e.g., decoherence. While many of the studies of molecular computing have been theoretical, the experimental demonstration in Li_2 of the Deutsch-Jozsa algortihm[24] by Vala *et al.*[15] provides impetus for pursuing molecular quantum computing.

The current studies of molecular quantum computing are promising, but one primary question to ask is what would be the best possible choice of a molecule for realizing practical quantum computation. In a recent paper, Babikov[7] has begun to seek an answer to this question by addressing the problem of gate fidelity (accuracy) using a diatomic molecule as a model for performing quantum gate operations with tailored infrared femtosecond pulses. Gate fidelity is of critical importance if practical quantum computing is to be performed since tens (or more) of sequential gate operations may be required to implement a particular algorithm. Babikov[7] showed via numerical exploration that general trends exist in the dependence of gate fidelity on the anharmonicity of the system and the pulse length. These dependencies can be used as guidelines for choosing an appropriate molecular system for implementing quantum information processing. It was demonstrated that when all parameters are carefully chosen, the accuracy of quantum gates based on the vibrational states of a diatomic model could reach as high as 99.9%.

In this paper, we readdress the problem of designing laser pulses for performing quantum logic gate operations. We formulate the design of the laser pulse in terms of the *analytic* pulse area theorem rather than in the context of the *numerical* optimal control theory. By formulating the problem with the pulse area theorem, the importance of the various molecular and laser parameters can be predicted analytically and the choice of suitable systems for molecular quantum computing can be readily evaluated. In Sec. II A, the optimal control theory and the pulse area theorem are reviewed briefly in the context of creating a NOT gate. The simple one-dimensional Morse oscillator used as a model system is presented in Sec. II B. In Sec. III, analytical π pulse results are used to interpret different aspects of the implementation of quantum gate operations in vibrational systems. The power of the π pulse in predicting the influence of both molecular and laser parameters is emphasized. Some brief conclusions and discusssion are given in Sec. IV.

II. THEORY

The two lowest-energy bound vibrational eigenstates of the OH molecule are considered as a practical realization of a qubit, and a shaped external laser field is used to apply the quantum logic gates. First, the methods for determining the laser parameters required to perform the quantum gate operation are outlined. Second, the physical system used for the qubit is briefly introduced.

A. The NOT gate: Optimal control theory and the π pulse

In this paper, only the NOT gate is considered explicitly. For the NOT gate, the laser pulse must perform the following logical operations:

$$\begin{aligned} &\mathrm{NOT}|0\rangle \rightarrow |1\rangle, \\ &\mathrm{NOT}|1\rangle \rightarrow |0\rangle. \end{aligned} \tag{1}$$

To set up the quantum NOT gate for the simple one-dimensional Morse oscillator system considered here, see Sec. II B, two choices for determining the laser field are examined: (i) a laser pulse constructed *numerically* via the optimal control theory and (ii) a laser pulse determined *analytically* using the pulse area theorem. These two methodologies are reviewed briefly.

The goal of the optimal control theory for logical gate operations is to find the laser pulse $\varepsilon(t)$ that maximizes population transfer for the two transitions of Eq. (1). The interaction of the laser field with the dipole moment of the molecule (dipole approximation) is considered in this paper. The optimization of the population transfer should occur after the time interval T, which is commonly referred to as the target time. Also, the optimization of the population transfer must be achieved while constraining the total pulse energy, $\int_0^T |\varepsilon(t)|^2 dt$. One common way to accomplish these two goals is to maximize the following objective functional:[8]

$$\begin{aligned} K_{fi} = &\sum_{k=1,2} (|\langle \psi_i^k(T)|\phi_f^k\rangle|^2) - \int_0^T \alpha(t)|\epsilon(t)|^2 dt \\ &- \sum_{k=1,2} 2\,\mathrm{Re}\left\{ \langle \psi_i^k(T)|\phi_f^k\rangle \int_0^T \langle \psi_f^k(t)|i[H_0 - \mu\varepsilon(t)] \right. \\ &\left. + (\partial/\partial t)|\psi_i^k(t)\rangle dt \right\}, \end{aligned} \tag{2}$$

where the sum is over the two transitions of interest. In Eq. (2), $\psi_i^k(t)$ is the time-dependent wave function for the system driven by the laser pulse $\varepsilon(t)$. ϕ_i^k and ϕ_f^k are the initial state and the desired final (target) state, respectively, for the kth transition of interest. The first term in the functional serves to maximize the gate fidelity, which is related to the overlap of the initial state after interaction with the field of duration T. The second term is a penalty term utilized to minimize the energy of the laser field in performing gate operation. The last term is used to satisfy the time-dependent Schrödinger equation for molecule-field interaction. Here the penalty term in Eq. (2) is chosen to impose restrictions on the total pulse energy only, but other restrictions could be enforced on the optimized field, such as frequency biasing to ensure spectral simplicity. There have been various alternate choices of the objective functional in the literature,[22,25,26] and we

choose the current one for its simplicity as well as its wide applicability.

In Eq. (2), $\alpha(t)$ is a penalty function

$$\alpha(t) = \frac{\alpha_0}{s(t)}, \tag{3}$$

where α_0 is the constant penalty factor and $s(t)$ plays the role of pulse envelope. The constant penalty factor α_0, which is set at the beginning of the algorithm, plays a critical role in determining the overall yield and, also, the complexity of the pulse.[27] Low values of $\alpha_0(<0.1)$, which impose only a weak restriction on the total pulse energy, allow high yields to be obtained but lead to extremely complex pulses. de Vivie-Riedle and co-workers have demonstrated[27] how the algorithm can be modified to use large values of $\alpha_0(\geqslant 1000)$, while still achieving high yields but with much simplified pulses. In the examples considered in this paper, the constant penalty factor $\alpha_0=1$ unless stated otherwise. When chosen appropriately, the function $s(t)$ forces the optimized field to switch on and off smoothly.[21] Here, as in Ref. 7, we use the form $s(t)=\sin^2(\pi t/T)$.

The goal of determining the optimal pulse $\varepsilon(t)$ is accomplished by maximizing Eq. (2) with respect to ψ_f^k, ψ_i^k, and $\varepsilon(t)$, which results in the following system of coupled equations:

$$i\hbar\frac{\partial}{\partial t}\psi_i^k(t) = [H_0 - \mu\varepsilon(t)]\psi_i^k(t), \quad \psi_i^k(0) = \phi_i^k, k = 1,2, \tag{4}$$

$$i\hbar\frac{\partial}{\partial t}\psi_f^k(t) = [H_0 - \mu\varepsilon(t)]\psi_f^k(t), \quad \psi_f^k(T) = \phi_f^k, k = 1,2, \tag{5}$$

and

$$\varepsilon(t) = -\frac{s(t)}{\hbar\alpha_0}\mathrm{Im}\left[\sum_{k=1,2}\langle\psi_i^k(t)|\psi_f^k(t)\rangle\langle\psi_f^k(t)|\mu|\psi_i^k(t)\rangle\right]. \tag{6}$$

The system of coupled equations, Eqs. (4)–(6), is solved numerically using the iterative method proposed by Zhu *et al.*[19]

Optimal control theory has proven very successful in designing pulses for maximizing population transfer in a variety of molecular systems,[8,19,21] and the construction of experimentally feasible pulses has also been highlighted.[21,22] However, difficulties arise in the interpretation of the physical mechanism for controlled population transfer as the interference of a large number of optical pathways can contribute to the control. Therefore, one cannot readily determine how laser parameters, e.g., pulse length, intensity, frequency, and/or molecular properties (e.g., transition dipole moments and energy level spacing), will affect the excitation process. To better understand these effects, we constructed the logic gate using an analytical π pulse[28–31] based on the pulse area theorem rather than a numerical pulse determined via the optimal control theory. We will focus on the original π pulse results from a two-level model, although more general π pulses beyond the pulse area theorem have been developed for multilevel systems.[30,31] It is demonstrated that this simple π pulse, along with the pulse area theorem, is able to clarify most of the underlying mechanism in the quantum NOT gate operation.

Here we consider a simple laser field of the form

$$\varepsilon(t) = \varepsilon_0 s(t)\sin(\omega_0 t), \tag{7}$$

where there is a single frequency component ω_0 equal to the energy difference between the two qubit states, E_1-E_0; ε_0 is the maximum laser field strength; and the pulse envelope $s(t)$ has the same form as for the optimal control theory field.

The pulse area theorem states that within the rotating wave approximation (RWA) and for an on-resonance excitation frequency, the final transition probability for a two-level system initially in one state will only be determined by the area of the pulse, A. From this theorem, the final population transfer P is

$$P = |\langle\psi_i(T)|\phi_f\rangle|^2 = \sin^2(A/2), \tag{8}$$

where

$$A = \int_0^T \mu_{01}\varepsilon_0 s(t)dt, \tag{9}$$

with μ_{01} being the transition dipole moment connecting the two states of interest. If we assume that the optimized pulse is of the form given in Eq. (7), then the pulse area $A=\varepsilon_0\mu_{01}T/2$. For a pulse with an area π, 100% of the population will be transferred from the initial state to the final state. Therefore, the field strength ε_0 for the NOT gate is determined analytically from the condition that the pulse area should be π, i.e., $\varepsilon_0=2\pi/(T\mu_{12})$. It has been proven[32] that within the RWA, the π pulse is the *globally* optimized field for a two-level system. Note that the π pulse is also the initial trial field for the optimal control algorithm.

B. The molecular model

The ground-state potential for the OH diatomic is approximated by an analytic Morse oscillator of the following form:

$$V(r) = -D + D[e^{-a(r-r_e)} - 1]^2. \tag{10}$$

The eigenvalues of the Morse oscillator are given by

$$E_v = -D + \omega\left(v + \frac{1}{2}\right) - \Delta\left(v + \frac{1}{2}\right)^2, \tag{11}$$

where ω is harmonic frequency,

$$\omega = a\sqrt{\frac{2D}{m}}, \tag{12}$$

and Δ is the anharmonicity parameter,

$$\Delta = \frac{a^2}{2m}. \tag{13}$$

The standard set of Morse parameters for the OH diatomic are $D=0.1994$ hartree, $r_e=1.821$ bohr, and $a=1.189$ bohr^{-1}, and thus the harmonic frequency ω is 3964 cm^{-1} and the anharmonicity Δ is 89.76 cm^{-1}. In addition to this standard OH model, the effects of both increased and decreased anharmonicity, i.e., larger and smaller Δ, are considered in Sec. III.

The dipole moment function has the analytic form

$$\mu(r) = \mu_0 r e^{-r/r_0}, \tag{14}$$

with μ_0=3.088 a.u. (7.848 D) and r_0=0.6 bohr.

The above potential is represented by a one-dimensional grid and the eigenvalues and eigenvectors are calculated with the window operator method[33] and a recursive scheme,[34] respectively. The eigenenergies obtained from the grid agree with the analytical results in Eq. (11). The transition dipole moments, $\mu_{ij}=\langle\psi_i|\mu(r)|\psi_j\rangle$, are evaluated numerically from the eigenvectors and the dipole moment, Eq. (14). The permanent dipole moments are neglected in all calculations.

Unless otherwise stated, only the first five vibrational states ($0\leqslant v\leqslant 4$) are included in the calculations and this five-level model has been used to approximate the full vibrational manifold for OH, as well as for the other model systems with different anharmonicities. The eigenstates will be termed as $|i\rangle$ for the ith eigenstate. The qubit is chosen to be formed by the ground state $|0\rangle$ and first excited vibrational state $|1\rangle$.

We note here that the second-order procedure in Ref. 19 for the optimal control algorithm has been utilized, i.e., the term related with $[\mu(r),H_0]=\mu(r)H_0-H_0\mu(r)$ has been included in our calculations, see Eqs. (48)–(51) of Ref. 19. Although this term has been introduced in Ref. 19, its exact form was not given. For the OH model, defined by Eqs. (10) and (14), it takes the following form:

$$[\mu(r),H_0] = \frac{\mu_0}{m} e^{-r/r_0}\left\{\left[\frac{r_0-r}{r}\right]\frac{\partial}{\partial r} - \frac{1}{r_0} + \frac{r}{2r_0^2}\right\}. \tag{15}$$

In our discrete level representation, the effect of this term is taken into account by introducing an effective dipole coupling term $d\mu_{ij}=(i\Delta t/2)\langle\psi_i|[\mu(r),H_0]|\psi_j\rangle$.

To explain the decrease of gate fidelity with the decrease in anharmonicity, see Sec. III, it is necessary to go beyond the two-level system. If the anharmonicity is large, the system is effectively a two-level one as all transitions, except that between the qubit states, are far off resonance. When the anharmonicity decreases, the effects of other states have to be considered and here a three-level system is used to approximate the effect of anharmonicity. As long as the pulse length satisfies $T\gg(2\pi/\omega_0)$, i.e., the adiabatic condition is justified, one can use the RWA. For typical vibrational energy spacings (100–5000 cm^{-1}), the adiabatic condition will be fulfilled unless one wishes to use ultrafast pulses for excitation, i.e., pulse durations of less than 100 fs. The effective Hamiltonian for a three-level system using the RWA is

$$H = \frac{1}{2}\begin{pmatrix} 0 & \mu_{01}\varepsilon_0 s(t) & 0 \\ \mu_{01}\varepsilon_0 s(t) & 0 & \mu_{12}\varepsilon_0 s(t) \\ 0 & \mu_{12}\varepsilon_0 s(t) & -4\Delta \end{pmatrix}. \tag{16}$$

In order to simplify the problem, the model is only extended to a three-level system since from the numerical results shown in Ref. 7 and theoretical considerations, the next vibrational state $|2\rangle$ will have the most significant (detrimental) effect on the $|0\rangle\rightarrow|1\rangle$ and $|1\rangle\rightarrow|0\rangle$ excitation processes. According to Eq. (11), under the RWA, the detuning of the vth state scales as $2(v-1)\Delta$, so $v=2$ will have the smallest detuning. The detuning of other states will increase with an increase of the state number, and thus their roles will be much less significant.

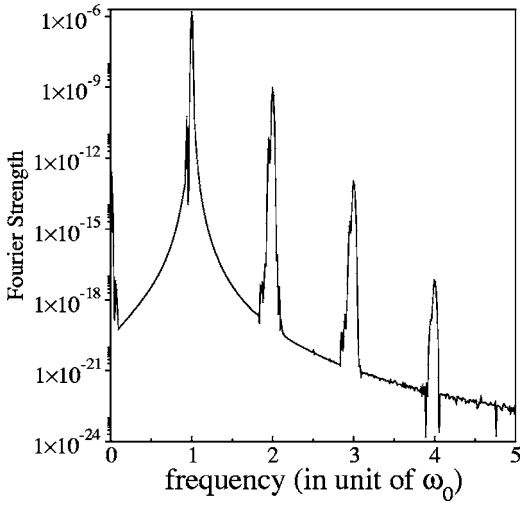

FIG. 1. The Fourier components of the pulse obtained via the optimal control theory algorithm for the OH molecule.

III. RESULTS AND DISCUSSION

We first compare the optimal control theory results and the π pulse predictions for the NOT gate as implemented for the OH molecule. The target time T is set to 750 fs, as was done in Ref. 7. Our numerical results for OH, as obtained using the optimal control theory algorithm,[7] are in quantitative agreement with the original work. For the NOT gate shown in Fig. 2 of Ref. 7, the fidelity is given as 0.9948, while our result is 0.9947. The small difference could be attributed to minor differences between the numerical parameters and propagation methods used. The results obtained using the five-level model of OH have been tested by comparing them with those obtained using direct wave-packet propagation and/or a full 22-level model and good agreement between the five-level model and these more sophisticated models is found.

The field strength defined from Eq. (9) for the π pulse is 0.005 461 a.u., which is roughly in accord with the maximum field strength shown in Fig. 2 of Ref. 7. The gate fidelity using the π pulse prediction for the field in an exact calculation is determined to be 0.989 64, and thus the optimal control theory algorithm only improves the fidelity marginally compared with the π pulse. Also the maximum frequency components of the optimized field are nearly on resonance with ω_0; see Fig. 1, which shows the power spectrum of the field resulting from the OCT calculation. From this example, it is clear that the π pulse captures most of the physical signatures of the optimized pulse, at least for the model OH system. Although the resemblance between the optimal control theory and π pulse results is to be expected for an anharmonic system such as OH, the analytic nature of the π pulse, or more generally, the pulse area theorem, provides direct physical insight into the roles of different molecular and laser parameters in quantum gate operations.

The dependence of the gate fidelity on the anharmonicity parameter is one of the principal results in Ref. 7. Thus, the

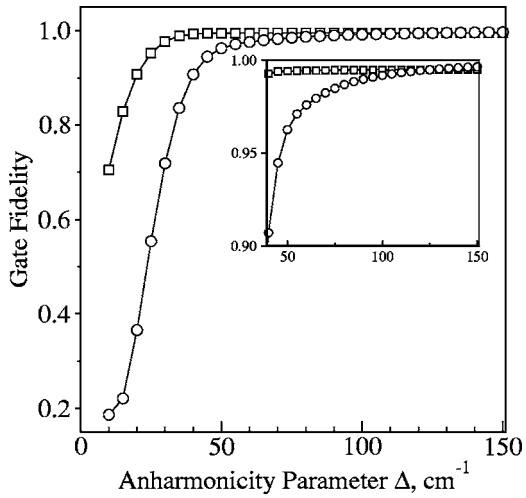

FIG. 2. Fidelity of the NOT gate as a function of the anharmonicity parameter of the model system. Results are illustrated for the π pulse (circles) and optimal control theory (squares). The inset shows an expanded view of the high-anharmonicity regime.

ability of the π pulse to predict this dependence is considered. The target time is again set to be 750 fs. The gate fidelities obtained as a function of the anharmonicity parameter using the analytic π pulse predictions for the field are shown in Fig. 2, along with the numerical optimal control theory pulses for comparison. For the smallest anharmonicities of $\Delta=10$, 15, and 20 cm^{-1}, a seven-level model was utilized to ensure that the maximum population of the highest state was below 0.001. From Fig. 2, the dependence of the gate fidelity on anharmonicity, i.e., decrease of fidelity with decrease of anharmonicity parameter, can be reproduced qualitatively by the π pulse. However, the gate fidelities obtained via a single frequency π pulse are not as high, i.e., less than 90%, in the low-anharmonicity regime below 40 cm^{-1}, where using OCT significantly improves the gate fidelity. In the range of $50\ \mathrm{cm}^{-1} \leqslant \Delta \leqslant 110\ \mathrm{cm}^{-1}$ suggested by Babikov[7] as appropriate for achieving the high fidelities necessary for quantum computation, the fidelities from the π pulse prediction are almost as good as the results obtained using the optimal control theory algorithm.

The use of the analytic theory allows a direct qualitative estimate of the anharmonicity regime where the optimal control theory will significantly improve the π pulse prediction, which in turn means the anharmonicity regime where quantum computation is difficult, see Fig. 2. For any given pulse duration T, the field strength required to produce a π pulse can be determined via $\varepsilon_0=2\pi/(T\mu_{01})$, and thus the spectral full width at half maximum of the most detrimental $|1\rangle$ to $|2\rangle$ resonance can be approximated by $\mu_{12}\varepsilon_0$. The $|1\rangle$ to $|2\rangle$ transition is detuned from the single laser frequency of the π pulse by an amount 2Δ. Therefore, when the spectral width of the π pulse is comparable or larger than 2Δ, we would expect the two-level approximation to fail and the quantum gate fidelity to be much lower than 90%.

While the above expression for spectral width is the continuous-wave laser field estimate, it does provide a reasonable qualitative estimate of the pulsed laser field one. For the laser and field parameters used to generate Fig. 2, the half width at half maximum of the $|1\rangle$ to $|2\rangle$ resonance is approximately 30 cm^{-1}. For anharmonicities of this magnitude or less, the effect of state $|2\rangle$ in the generalized three-level system will begin to be significant and it is clear that the π pulse predictions become quite poor in this regime.

Another important observation from Fig. 2 is that the gate fidelity determined from the optimal control theory shows a plateau for high anharmonicities. On the other hand, the π pulse results increase slowly but definitely in the high-anharmonicity regime. Therefore, the fidelity from the π pulse prediction will eventually exceed that from the optimal control field, see inset of Fig. 2. Figure 2 clearly shows that the optimal control theory results are not the global maximum in the sense of the final yield (gate fidelity), and thus the significance of the "accuracy" of quantum gates obtained via the optimal control theory must be examined carefully. For example, for the OH parameters, by decreasing the penalty parameter α_0 from 1 to 0.5, we can obtain a gate fidelity of 0.9986, which is considerably higher than that of the $\alpha_0=1$ result of 0.9947. The maximum field amplitude for the $\alpha_0=0.5$ case increases by 2.3% compared with the $\alpha_0=1$ case, while the total pulse energy increases by only 4.8%. This amount of energy increase should be readily achieved experimentally, and thus higher fidelity can be achieved.

The reason for the increase in gate fidelity with the decrease in α_0 can be well understood using the pulse area theorem analysis. For optimization using the objective function of Eq. (2), we assume that the optimized pulse has a pulse area of $A=\pi-\delta$, where δ is a small increment. Using this pulse area, the maximum field strength is $\varepsilon_0=2(\pi-\delta)/(T\mu_{12})$. If δ is small, then this optimized pulse will generate a population transfer P that scales as $P\approx 1-(\delta/2)^2$. Since the time-dependent wave function satisfies the Schrödinger equation, the last term in the objective function of Eq. (2) will be identically zero. The sum of the first two terms can then be determined using a small δ approximation, i.e., only the δ and δ^2 terms are retained. After a relatively straightforward derivation, the objective function K_{fi} (within the RWA and for a small value of δ) is given by

$$K_{fi}=\left(2-\frac{\alpha_0\pi^2}{\mu_{01}^2T}\right)-\frac{1}{\mu_{01}^2T}\left(\left[\frac{\mu_{01}^2}{2}T-\alpha_0\right]\delta^2-2\alpha_0\pi\delta\right). \tag{17}$$

The increment $\delta_{\max}$ that maximizes the objective function can be determined to be

$$\delta_{\max}=\frac{2\alpha_0\pi}{\mu_{01}^2T+2\alpha_0}. \tag{18}$$

It is related to the transition moment between the qubit states μ_{01}, the target time T, and, most importantly, the (time-independent) penalty function parameter α_0. Within these approximations, the maximal gate fidelity that can be obtained using the optimal control theory will be $1-(\delta_{\max}/2)^2$. In the high-anharmonicity regime of Fig. 2, where the two-level model is most valid, i.e., $\Delta \geqslant 50\ \mathrm{cm}^{-1}$, Eq. (18) provides near quantitative predictions for the gate fidelity. For example, for the OH parameters where $\Delta=89.76\ \mathrm{cm}^{-1}$, Eq. (18) predicts a gate fidelity of 0.9951 for $\alpha_0=1$ and 0.9987 for $\alpha_0=0.5$.

These can be compared with the gate fidelities determined numerically using the optimal control theory algorithm of 0.9947 and 0.9986, respectively. In fact, by decreasing α_0 further to 0.1, the gate fidelity can be determined to be higher than 99.99% using the optimal control theory algorithm. Clearly, using the objective function given by Eq. (2), one must be careful in the choice of the penalty function parameter α_0, as it can critically affect the predicted gate fidelities.

Equation (18) also makes it clear that while the π pulse captures most of the physics in the high-anharmonicity regime, the optimized pulse will not be the π pulse unless $\alpha_0=0$. Since the optimal control algorithm penalizes the pulse energy, see the second term of Eq. (2), it would eventually deem an increase in pulse amplitude as unfavorable. Thus, while the pulse determined via the optimal control theory would approach the π pulse limit, it would never reach this limit as the increase in pulse amplitude required would not be numerically warranted due to the penalty function. Clearly, how close one comes to this limit is a function of the choice of penalty parameter α_0 and any nonzero penalty on the field, with fixed target time, will prevent the fidelity from reaching the "true" maximum. We expect the α_0 effect shown above from pulse area analysis to be general. In Ref. 35, it is shown that the unrestricted optimal control will always lead to perfect control or no control. It is also shown that the maxima in the control plane that might be accessible via the optimal control algorithm have "flat tops" (see Fig. 1 of Ref. 35) in parameter spaces. This is similar to the flat tops in Eq. (8) near $A=\pi$. So, in general, when the optimal control algorithm explicitly penalizes the pulse energy, as in Eq. (2), it would eventually deem an increase in pulse amplitude as unfavorable due to the decrease in the objective function from the pulse energy term.

While the focus has been on the dependence of quantum gate fidelity on the anharmonicity of the molecular system, other effects such as the target time can be understood using the pulse area analysis. For all the anharmonicities considered and for a target time of $T=750$ fs, we have $\mu_{01}^2 T \geqslant 2\alpha_0(\alpha_0=1)$; the transition dipole moment has a very small dependence on the anharmonicity of the system. Therefore, in Eq. (18), the α_0 term in the denominator can be omitted and it can be seen that increasing the target time T has the same effect as decreasing α_0. For example, using the OH parameters and $\alpha_0=1$, Eq. (18) predicts that an increase in the target time from 750 to 1000 fs will result in an increase in gate fidelity from 0.9951 to 0.9972; the corresponding numerical results determined using the optimal control theory algorithm are 0.9947 and 0.9971, respectively. Clearly, the simple pulse area analysis provides a physical insight into the effects of the target time (pulse length) on the gate fidelity. Physically, increasing the target time results in a decrease in the maximum field strength, as discussed above. Hence, the effective spectral bandwidth for the $|1\rangle$ to $|2\rangle$ transition is reduced relative to that for the shorter pulse, and the molecular system can be better approximated as a two-level system even for smaller anharmonicities.

The difference between Hadamard gate and NOT gate can also be well explained from Eq. (18). The Hadamard gate corresponds to the following operation

$$\begin{aligned} \mathrm{HAD}|0\rangle &\rightarrow \frac{1}{\sqrt{2}}[|0\rangle + |1\rangle, \\ \mathrm{HAD}|1\rangle &\rightarrow \frac{1}{\sqrt{2}}[|0\rangle - |1\rangle]. \end{aligned} \tag{19}$$

Within the pulse area theorem treatment of the problem, the change from a NOT gate to a Hadamard gate corresponds to a change in pulse area from π to $\pi/2$. Therefore, the maximal gate fidelity that can be obtained using the optimal control theory will be $1-(\delta_{\max}/2)^2$, where $\delta_{\max}$ is given by Eq. (18) with π replaced by $\pi/2$. For the OH parameters, the fidelity of the Hadamard gate[7] (0.9988) can be accurately reproduced using this approximation (0.9988).

The above discussions provide analytical explanations for the results in the high-anharmonicity regime, $\Delta \geqslant 50$ cm^{-1} for $T=750$ fs. For the purposes of quantum logic gate operations, where extremely high gate fidelities are required, this is the only regime of interest. However, for the low-anharmonicity regime, the improvement of the optimal control result over the simple π pulse prediction is quite significant. For very harmonic systems, i.e., low anharmonicities, transitions to states outside the two-level system can be significant. In order to compensate for these detrimental transitions, the optimized pulse must have more than a single (time-independent) frequency component, i.e., it cannot be the single frequency π pulse. Interference between different transitions can be utilized to maximize the final yield of a desired transition. The understanding of these interferences in the low-anharmonicity regime is outside the scope of this paper, but here we demonstrate that an analytical investigation of these mechanisms is possible.

The Fourier analysis of the optimized pulse for OH, see Fig. 1, shows that the spectrum contains a series of frequency components located near the one-photon and overtone frequencies. The magnitudes of the peaks decrease quickly (note the logarithmic scale) and thus only the one-photon peak is significant. A Fourier analysis of the optimized pulse for the lowest anharmonicity considered in this paper, $\Delta=10$ cm^{-1}, shows a spectrum (not shown here) that is similar to that presented in Fig. 1, but the overtone transitions have increased in magnitude and become numerically significant. In order to investigate the role of the overtone transitions in the control process, a new pulse is constructed numerically using only the frequency components around the first peak (in the range of $[0.8\omega, 1.1\omega]$). We find that by only using these frequency components, good agreement with the complete optimal control field is obtained, see Fig. 3, even for the most harmonic case. Therefore, most of the control can be obtained by using a pulse with frequencies around the one-photon frequency, rather than one with many high-frequency components. This fact enables the utilization of the RWA when examining optimal control and the use of these simplified pulses for optimal control problems is the subject of on-going investigations.

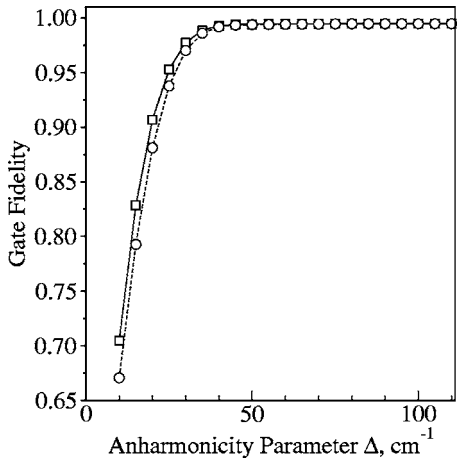

FIG. 3. Fidelities of the NOT gate as a function of the anharmonicty parameter of the model system. Results are illustrated for the reconstructed pulse (circles) and for the laser pulse with the full spectrum taken into account (squares). See text for more details.

IV. CONCLUSION AND DISCUSSION

The effects of different laser and molecular parameters on the implementation of qubit gate operations based on molecular vibrational states have been investigated. By constructing the *analytically* optimized pulse based on the pulse area theorem, we have demonstrated that the simple π pulse captures the physics of single-qubit gate operations for large anharmonicities, $\Delta \geqslant 50\ \mathrm{cm}^{-1}$ for a target time of 750 fs. A simple analytical expression, Eq. (18), has been derived that allows the dependence of the gate fidelity on the target time and penalty function parameter α_0 to be determined. It has been shown that α_0, which can be chosen arbitrarily, plays a critical role in the determination of the final gate fidelity. Although the importance of the penalty parameter α_0 on the optimal control theory results has been qualitatively discussed before[27] based on numerical investigations, to the best of our knowledge, Eqs. (17) and (18) represent the first analytical expressions derived to show this importance.

The discussion in this paper shows that one is generally prevented from attaining 100% gate fidelity using an objective function of the form given by Eq. (2) due to the penalty on the total pulse energy. Theoretically, this difficulty can be overcome by introducing the field in the previous iteration as a reference field[17] and then penalizing the difference between the new field and the reference field in the objective function. However, this choice of reference field may be problematic as the field strength is not restricted. More elaborate choices of the reference field are being developed to address the problem of incorporating various constraints on the optimized field in a uniform way, and it will be discussed separately.[36]

Overall, the utility of simple analytical expressions for interpreting the roles each of the laser and molecular parameters plays in the use of vibrational eigenstates for molecular quantum computing has been emphasized. These analytical expressions complement the numerical calculations based on quantum optimal control algorithms and are very useful for predicting and explaining quantum gate operation in this one-dimensional diatomic model. We expect that the use of sophisticated quantum optimal control algorithms will be crucial for obtaining optimized fields to control dynamics in polyatomic molecules and/or to implement more complex quantum computing algorithms. For example, the tailored pulses may have to account for decoherence effects.[10] Decoherence has been neglected in the present one-dimensional model, since the pulse duration for the gate operations is much shorter than the lifetimes of the vibrational states defining the qubit. If the model considered in the present work was chosen to represent a single vibrational mode in a polyatomic molecule, decoherence effects could be introduced with a phenomenological state lifetime. One could utilize a more sophisticated density-matrix approach, as considered by Ohtsuki,[18] but the addition of a phenomenological lifetime into the current analytical model would provide a qualitative understanding of the underlying physics including decoherence. While the model presented is basic, analogous ideas, e.g., two two-level systems with interactions between them, should help in the understanding of the implementation of multiqubit operations, e.g., the CNOT gate, in polyatomic molecules. Work in this area is in progress.

ACKNOWLEDGMENTS

The authors thank the Natural Sciences and Engineering Research Council of Canada and the University of Alberta for financial support.

[1] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, New York, 2000).
[2] G. Benenti, G. Casati, and G. Strini, *Basic Concepts*, Principles of Quantum Computation and Information Vol. I (World Scientific, Singapore, 2004).
[3] R. Feynman, Int. J. Theor. Phys. **21**, 467 (1982).
[4] J. A. Jones and M. Mosca, J. Chem. Phys. **109**, 1648 (1998).
[5] D. G. Cory, A. F. Fahmy, and T. F. Havel, Proc. Natl. Acad. Sci. U.S.A. **94**, 1634 (1997).
[6] N. A. Gershenfeld, Science **275**, 350 (1997).
[7] D. Babikov, J. Chem. Phys. **121**, 7577 (2004).
[8] C. Tesch and R. de Vivie-Riedle, Phys. Rev. Lett. **89**, 157901 (2002).
[9] C. Tesch, L. Kurtz, and R. de Vivie-Riedle, Chem. Phys. Lett. **343**, 633 (2001).
[10] U. Troppmann, C. Tesch, and R. de Vivie-Riedle, Chem. Phys. Lett. **378**, 273 (2003).
[11] U. Troppmann and R. de Vivie-Riedle, J. Chem. Phys. **122**, 154105 (2005).
[12] S. Suzuki, K. Mishima, and K. Yamashita, Chem. Phys. Lett. **410**, 358 (2005).
[13] R. Zadoyan, D. Kohen, D. A. Lidar, and V. A. Apkarian, Chem. Phys. **266**, 323 (2001).
[14] Z. Bihary, D. R. Glenn, D. A. Lidar, and V. A. Apkarian, Chem. Phys. Lett. **360**, 459 (2002).
[15] J. Vala, Z. Amitay, B. Zhang, S. R. Leone, and R. Kosloff, Phys. Rev. A **66**, 062316 (2002).
[16] J. P. Palao and R. Kosloff, Phys. Rev. Lett. **89**, 188301 (2002).
[17] J. P. Palao and R. Kosloff, Phys. Rev. A **68**, 062308 (2003).
[18] Y. Ohtsuki, Chem. Phys. Lett. **404**, 126 (2005).
[19] W. Zhu, J. Botina, and H. Rabitz, J. Chem. Phys. **108**, 1953 (1998).
[20] D. Tannor, V. Kazakov, and V. Orlov, in *Time Dependent Quantum Molecular Dynamics*, edited by J. Broeckhove and L. Lathouwers (Plenum, New York, 1992), p. 403.
[21] K. Sundermann, M. Motzkus, and R. de Vivie-Riedle, J. Chem. Phys. **110**, 1896 (1999).
[22] T. Hornung, M. Motzkus, and R. de Vivie-Riedle, J. Chem. Phys. **115**, 3105 (2001).
[23] Y. Ohtsuki, G. Turnici, and H. Rabitz, J. Chem. Phys. **120**, 5509 (2004).
[24] D. Deutsch and R. Jozsa, Proc. R. Soc. London, Ser. A **439**, 553 (1992).

[25] J. M. Geremia, W. Zhu, and H. Rabitz, J. Chem. Phys. **113**, 10841 (2000).
[26] G. G. Balint-Kurti, F. R. Manby, Q. Ren, M. Artamonov, T.-S. Ho, and H. Rabitz, J. Chem. Phys. **122**, 084110 (2005).
[27] T. Hornung, M. Motzkus, and R. de Vivie-Riedle, Phys. Rev. A **65**, 021403 (2002).
[28] N. Rosen and C. Zener, Phys. Rev. **40**, 502 (1932).
[29] G. F. Thomas, Phys. Rev. A **27**, 2744 (1983).
[30] M. Holthaus and B. Just, Phys. Rev. A **49**, 1950 (1994).
[31] J. Cao, C. J. Bardeen, and K. R. Wilson, Phys. Rev. Lett. **80**, 1406 (1998).
[32] M. Garcia and I. Grigorenko, J. Phys. B **37**, 2569 (2004).
[33] K. J. Schafer and K. C. Kulander, Phys. Rev. A **42**, 5794 (1990).
[34] H. Kobeissi, J. Comput. Phys. **61**, 351 (1985).
[35] H. Rabitz, M. Hsieh, and C. Rosenthal, Science **303**, 1998 (2004).
[36] T. Cheng and A. Brown (unpublished).