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Strong-field molecular alignment for quantum logic and quantum control

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We discuss an approach to quantum control based on initial adiabatic tuning of the field-free Hamiltonian by a strong laser field to optimize the system for the desired transitions induced by the control laser pulse. As an illustration, we describe single-qubit, two-qubit, and some qudit logical gates within rotational and vibrational states of a diatomic molecule. Gate operations use resonant Raman transitions, and the prior adjustment of the Hamiltonian is done by a strong nonresonant aligning field.

Strong-field molecular alignment [1,2] can be used as a tool to achieve specific goals of quantum control. Some examples include controlling the branching ratio between parallel and perpendicular electronic transitions [3] and controlled spinning of small molecules to rotational dissociation [4]. Alignment followed by coherent rotational revivals in a molecular gas can also be used to modify probe-laser pulses (upconversion, downconversion, and pulse compression [5]).

In this paper, we show how one can use strong-field alignment as a tool to adiabatically tune the molecular Hamiltonian, adjusting the rotational states to the specific task of quantum control one wants to perform. Conceptually, by modifying the Hamiltonian of the controlled system, one adds another knob to the coherent control scenarios which typically use the field-free states of the system [6]. When the field-free system is not ideally suited for a specific control task, this extra knob makes it possible to adiabatically adjust the Hamiltonian and facilitate the desired control operation in the dressed system. Note the two independent time scales present in the control scheme: adiabatic adjustment of the Hamiltonian acts on its own time scale that need not be the same as that of the desired control operation itself.

To illustrate this idea, we show how one can use an alignment to implement optical quantum logical gates within the space of rotational-vibrational states of a diatomic molecule. Here, rotations form one qubit and vibrations form the other. We consider single-qubit, two-qubit controlled-NOT (CNOT) operation and some qudit operations [7]. Note that when two degrees of freedom are associated with two different qubits (qudits), a single-qubit (qudit) operation implies transition between groups of levels. For example, switching the vibrational qubit (qudit) $v=0 \rightarrow v=1$ implies simultaneous transitions $|v=0,J\rangle \rightarrow |v=1,J\rangle$ for *all J*.

The ability to perform logical operations on coupled degrees of freedom as though they were completely independent, especially when the operating field explicitly couples them, has implications for both quantum information processing and quantum control. Indeed, if one wants to use several degrees of freedom in each node of a quantum computer, one has to learn to address each degree of freedom individually. The importance of logical operations for quantum control will be discussed in the conclusion of the paper.

Rather than relying on optimal control strategies [8], which use feedback and learning algorithms to find a pulse

that would make the desired transition [9], we apply the above-mentioned two-step strategy. In the first step, the adiabatically turned-on aligning field tunes the degree of angular localization of rotational states and thus controls matrix elements for rovibrational transitions. During the second step, another simple field induces transitions that correspond to a desired logical gate. We will focus on Raman transitions in a linearly polarized laser field, which are also the most convenient in this context due to available technology.

Consider a homonuclear diatomic molecule, such as ortho-N_2 or -D_2 in the ground electronic state. A linearly polarized field conserves the magnetic quantum number $M = J_z$, and for definiteness, we confine ourselves to the $M = 0$ subspace, with states characterized by $v = 0,1,2, \ldots$ and $J = 0,2,4, \ldots$ (odd J states are symmetry forbidden). If only the four lowest $M=0$ states, $v=0,1$ and $J=0,2$, could be initially populated (cold molecule), we can talk about single-bit and two-bit operations in this subspace (Fig. 1). Of course, our operations should cause no transitions between these four and any other state.
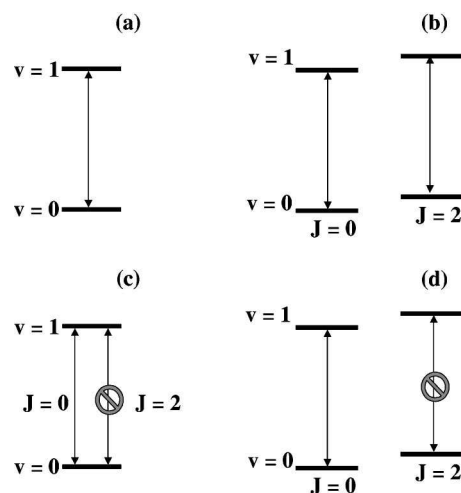

FIG. 1. Single-bit (a-b) and CNOT (c-d) operations in the 2×2 (v,J)-qubit system. Single-bit operation implies simultaneous identical $v=0 \rightarrow v=1$ transitions for both $J=0$ and $J=2$. The CNOT operation uses rotations as a control: the bit should be flipped for $J=0$ but not for $J=2$.

A single-bit operation would mean acting on one degree of freedom independent of the other. For example, we should be able to flip the vibrational qubit $v = 0,1$ not only without changing J, but also independent of J [Figs. 1(a) and 1(b)]. A CNOT operation would mean that we flip the vibrational qubit $v = 0,1$ depending on J, e.g., flip it for $J = 0$ but not for $J = 2$ [Figs. 1(c) and 1(d)]. If many v, J levels could be initially populated, we look at the blockwise (qudit) operations, which are discussed later in the paper.

At first glance, it might seem that there is no problem in manipulating the vibrational degree of freedom independent of the rotational one, since the rovibrational coupling in diatomic molecules is typically small. However, the exciting laser field simultaneously addresses both degrees of freedom since the interaction potential depends on the orientation of the molecule. For a Raman transition within the same electronic state, the laser-molecule interaction [2] is

$$V(R, \theta, t) = -\frac{1}{2} \langle \mathcal{E}^2(t) \rangle \Omega(\omega, R, \theta),$$

$$\Omega(\omega, R, \theta) = \alpha_\parallel(\omega, R)\cos^2\theta + \alpha_\perp(\omega, R)\sin^2\theta$$
$$= \alpha_\perp(\omega, R) + \Delta\alpha(\omega, R)\cos^2\theta, \quad (1)$$

where ω is the laser frequency, R is the internuclear distance, and $\langle \mathcal{E}^2(t) \rangle$ is the electric field, which is averaged over fast optical oscillations of the carrier but can include the beat at the Raman frequency. Finally, θ is the angle between the field polarization vector and the molecular axis, $\alpha_\parallel$ and $\alpha_\perp$ are parallel and perpendicular electronic polarizabilities, and $\Delta\alpha = \alpha_\parallel - \alpha_\perp$. Even if the angular momentum does not change during the $v = 0 \rightarrow 1$ transition, its matrix element includes $\langle J|\cos^2\theta|J \rangle$. Since the latter can differ for different J, the effective Rabi frequencies for flipping the vibrational bit will be different for different J, making a single-bit operation nontrivial.

This is where alignment is used. Adiabatic turn on of the aligning field transfers each free-rotor state J into a pendular state $\tilde{J}$. Logical operations are now performed on the states $v, \tilde{J}$. Changing the strength of the aligning field controls the matrix elements $\langle \tilde{J}|\cos^2\theta|\tilde{J} \rangle$. Once all logical operations are completed, adiabatic turn off puts the result back into the v, J basis.

To perform a single-bit operation between the states $v = 0$ and $v = 1$, we make the corresponding matrix elements equal for $\tilde{J} = 0$ and $\tilde{J} = 2$. The Rabi oscillations between $v = 0$ and $v = 1$ will then evolve independent of the particular $\tilde{J}$ state. For the CNOT operation with $\tilde{J}$ as a control, we adjust the aligning field so that the ratio of matrix elements for the correspondent transitions is k/n, e.g., 2/3 [10]:

$$\Omega(\tilde{J} = 2) \equiv \langle \tilde{J} = 2|\Omega(\omega, R, \theta)|\tilde{J} = 2 \rangle$$
$$= \frac{2}{3} \langle \tilde{J} = 0|\Omega(\omega, R, \theta)|\tilde{J} = 0 \rangle \equiv \Omega(\tilde{J} = 0). \quad (2)$$

In this case, the bit $v = 0,1$ for $\tilde{J} = 0$ will flip three times, while for $\tilde{J} = 2$ it will flip two times, returning to its initial state.

A single-bit operation on the rotational degree of freedom would mean flipping $\tilde{J} = 0$ and $\tilde{J} = 2$ independent of the vibrational quantum number. This operation is the easiest of the three: the matrix elements $\langle v, \tilde{J} = 0|\Omega(\omega, R, \theta)|v, \tilde{J} = 2 \rangle$ are virtually equal for $v = 0$ and $v = 1$ because the polarizability is almost the same for the two vibrational levels.

Let us illustrate how these ideas work. We assume that the laser pulse consists of a strong aligning field $\mathcal{E}_a\cos\omega t$ and a weaker field $\mathcal{E}_d$ that resonantly drives the transitions,

$$\mathcal{E}(t) = \mathcal{E}_a\cos\omega t + \mathcal{E}_d[\cos(\omega + \omega_R)t + \cos(\omega - \omega_R)t], \quad (3)$$

$$\langle \mathcal{E}^2(t) \rangle = \frac{1}{2}[\mathcal{E}_a^2 + 4\mathcal{E}_a\mathcal{E}_d\cos\omega_R t + 4\mathcal{E}_d^2\cos^2\omega_R t], \quad (4)$$

where ω_R is the frequency of the desired Raman excitation and ω is the carrier frequency. In the calculations, we assume an infrared ω, e.g., from a Ti:Sapphire laser, and use the static polarizability tensor $\alpha(R)$. In Eq. (4), $\mathcal{E}_a^2$ is responsible for alignment, $\mathcal{E}_a\mathcal{E}_d\cos\omega_R t$ drives the transitions, and $\mathcal{E}_d^2\cos^2\omega_R t$ is a potential source of errors.

Numerical calculations were done for the molecules N_2 and D_2. Given low vibrational excitation, the polarizability was approximated as $\alpha(R) = \alpha(R_{eq}) + (\partial\alpha/\partial R)(R - R_{eq})$, where R_{eq} is the equilibrium internuclear separation. The required data for N_2 and D_2 were taken from Refs. [11,12]. The linear term in this expansion is responsible not only for vibrational excitation, but also for laser-induced rovibrational coupling.

The calculation proceeded in two steps. First, the molecular Hamiltonian was diagonalized on the basis of uncoupled v, J states to find the eigenstates in the aligning field $\mathcal{E}_a$, including both field-free and field-induced rovibrational coupling. Off-diagonal matrix elements $\langle v|R - R_{eq}|v' \rangle$ were approximated by those for a harmonic oscillator; the required harmonic constant taken from Ref. [13]. Diagonal matrix elements are related to the rotational constant $B(v) = B_0 - \alpha_e(v + 1/2)$, where α_e is the rovibrational coupling constant. Namely,

$$B(v) = \left\langle v \left| \frac{1}{\mu R^2} \right| v \right\rangle = B_0 - \frac{2}{R_{eq}^3} \langle v|R - R_{eq}|v \rangle. \quad (5)$$

Second, transitions between the eigenstates of the aligned molecule induced by the field $4\mathcal{E}_a\mathcal{E}_d\cos\omega_R t + 4\mathcal{E}_d^2\cos^2\omega_R t$ were calculated, for a rectangular driving pulse. At this step, only the four main aligned states $v = 0,1$; $\tilde{J} = 0,2$ and their nearest neighbors with $v = 2$ and/or $\tilde{J} = 4$ were included. Populations of the latter states were monitored in order to control the escape from our two-bit system.

Relative behavior of the transition matrix elements between aligned states of N_2, $\langle \tilde{J}|\cos^2\theta|\tilde{J} \rangle$ and $\Omega(\tilde{J})\langle v = 0, \tilde{J}|\Omega(\omega, R, \theta)|\tilde{J}, v = 1 \rangle$ for $\tilde{J} = 0,2,4,6$ in N_2 is shown in Fig. 2. The state $\tilde{J} = 0$ is trapped first, and has the highest
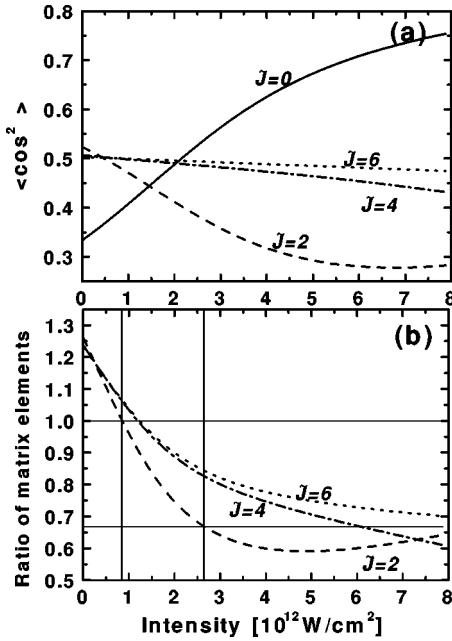
FIG. 2. The values of $\langle \tilde{J}|\cos^2\theta|\tilde{J}\rangle$ (a) and $\Omega(\tilde{J})/\Omega(\tilde{J}=0)$ (b) vs the intensity of the aligning field. Solid, dashed, dot-dashed, and dotted lines correspond to $\tilde{J}=0,2,4,6$, respectively. Two intensities marked with thin vertical lines correspond to $\Omega(2)=\Omega(0)$ and $\Omega(2)=2/3\Omega(0)$.

degree of angular localization in the strong field. For $\tilde{J}=2,4,\ldots$, the value of the matrix element $\langle \tilde{J}|\cos^2\theta|\tilde{J}\rangle$ first decreases and then starts to increase. Before the well is deep enough to trap the state, the wave function tends to localize near the maxima of the angular potential, where the corresponding classical motion is slower. The value of $\langle \tilde{J}|\cos^2\theta|\tilde{J}\rangle$ reaches its minimum when the well barely traps the state. In higher fields, the state is trapped and the alignment increases with the field.

Figure 3 shows single-bit and CNOT operations in N_2. For this molecule $R_{eq}=2.07$ a.u., and at this distance $\alpha_\perp=9.79$ a.u., $\Delta\alpha=4.88$ a.u., $\partial\alpha_\perp/\partial R=3.09$ a.u., $\partial\Delta\alpha/\partial R=7.74$ a.u. For the single-bit operation in v subspace, the
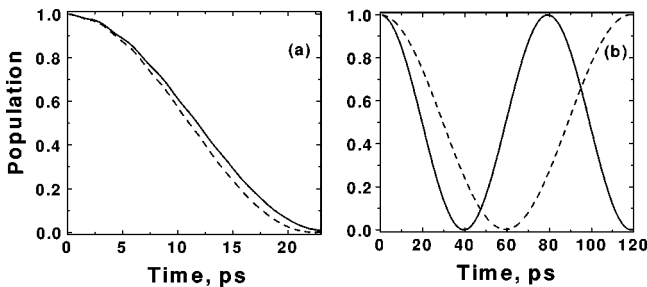

FIG. 3. (a) Single-bit operation in N_2. Solid line, population of the state $\tilde{J}=0$, $v=0$ in the case when this state was initially populated. Dashed line, same for the state $\tilde{J}=2$, $v=0$. (b) CNOT operation in N_2. Solid line, population of the state $\tilde{J}=0$, $v=0$ when the system is initially in this state; dashed line, population of the state $\tilde{J}=2$, $v=0$ when only this state is initially populated.

aligning field required to equalize the matrix elements is $\mathcal{E}_a=2.54\times10^7$ V/cm. Choosing the Raman beat frequency $\omega_R=4.39\times10^{14}$ s^{-1} and the driving field $\mathcal{E}_d=7.7\times10^6$ V/cm yields the operation with the fidelity of 99.3%. Changing the aligning field to $\mathcal{E}_a=4.48\times10^7$ V/cm (ratio of matrix elements for $\tilde{J}=2$ and $\tilde{J}=0$ equal to 3/2) yields the CNOT operation with the fidelity 99.8%. The driving field parameters are: $\mathcal{E}_d=2.32\times10^6$ V/cm, $\omega_R=4.39\times10^{14}$ s^{-1}. Note that since the operator inducing the transitions contains the product $\mathcal{E}_d\mathcal{E}_a$, for a stronger aligning field the driving field can be reduced. This reduces the errors due to population leakage to higher vibrational states. Furthermore, since we do not require *both* $\tilde{J}=0$ and $\tilde{J}=2$ to perform complete flips, we can further reduce the driving field and tune the frequency ω_R in resonance with the transition frequency for $\tilde{J}=0$.

High fidelity of the logical operations in N_2 is assisted by a low rovibrational coupling constant α_e in the field-free molecule. The larger value of α_e in D_2 sets tougher conditions for achieving high fidelity. Frequencies corresponding to the transition $v=0\leftrightarrow1$ differ for $J=0$ and $J=2$ by about $6\alpha_e$ [$B(v)=B_0-\alpha_e(v+1/2)$], and this detuning has to be compensated by the coupling strength. A stronger field causes stronger transitions to the states with $v=2$ and thus decreases fidelity of the logical operations.

For D_2 molecules, $R_{eq}=1.4$ a.u., and at this distance $\alpha_\perp=4.58$ a.u., $\Delta\alpha=1.8$ a.u., $\partial\alpha_\perp/\partial R=4.35$ a.u., and $\partial\Delta\alpha/\partial R=2.25$ a.u. To implement a CNOT gate in D_2, we chose the aligning field $\mathcal{E}_a=3.29\times10^8$ V/cm, which makes the ratio between the matrix elements for $\tilde{J}=0$ and $\tilde{J}=2$ equal to 5/6 (the ratios 3/2, 4/3, and 5/4 cannot be achieved in alignment of D_2). The driving field $\mathcal{E}_d=7.0\times10^6$ V/cm and $\omega_R=4.50\times10^{14}$ s^{-1} performs the gate in 5.6 ps with a fidelity of 98.5%. For the single-bit operation in the v domain, the aligning field $\mathcal{E}_a$ was set equal to 1.60×10^8 V/cm. Then the driving field with $\mathcal{E}_d=1.73\times10^7$ V/cm, $\omega_R=4.55\times10^{14}$ s^{-1} performs the operation with 96% fidelity in 8.9 ps.

As we already mentioned, a single-bit flip in the $\tilde{J}$ domain is a relatively simple operation. The matrix elements $\langle \tilde{J}=0|\cos^2\theta|\tilde{J}=2\rangle$ are virtually equal for $v=0$ and $v=1$, and their ratio varies in the range 0.986–0.998, depending on the field strength of the aligning field. Thus, any aligning field is satisfactory in terms of equalizing the Rabi frequencies for $v=0$ and $v=1$. Yet, there is an advantage in having an additional strong field in the system: in the presence of the aligning field, the transitions are driven by the perturbation $4\mathcal{E}_a\mathcal{E}_d\cos\omega_Rt$, and the noise is generated by a weaker term $4\mathcal{E}_d^2\cos^2\omega_Rt$. If the aligning field becomes weak then the relative contribution of the noise becomes larger. In the absence of the aligning field, one would have to rely on the resonance alone to suppress unwanted transitions.

In numerical calculations of the single-bit flip in the $\tilde{J}$ domain, we chose the aligning field to be the same as that used for a single-bit flip in the v domain. Then for the molecule N_2, the driving field of strength $\mathcal{E}_d=3.0\times10^6$ V/cm and frequency $\omega_R=2.12\times10^{12}$ s^{-1} performs the logical op-
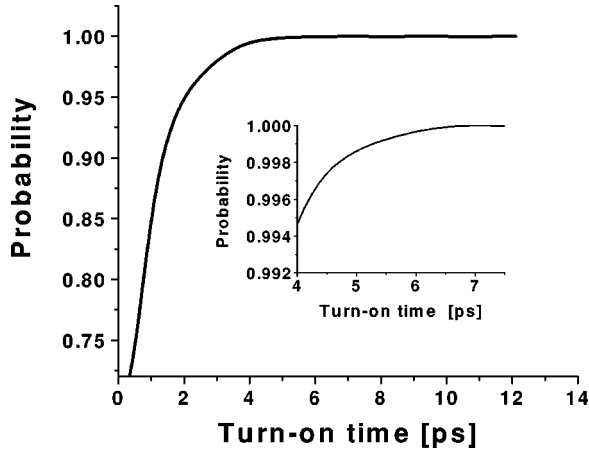
FIG. 4. Probability of populating the $|\tilde{J}=0\rangle$ state for different turn-on times of the aligning field. The initial state is $|J=0\rangle$ of the N_2 molecule. The peak field strength is $\mathcal{E}_a = 4.48 \times 10^7$ V/cm. The inset shows the expanded region between 4 and 8 ps.

eration with a fidelity of 99.7% in 17 ps. In D_2, a field with $\mathcal{E}_d = 2.30 \times 10^7$ V/cm, and $\omega_R = 3.10 \times 10^{13}$ s^{-1} implements the flip with 97% fidelity in 0.9 ps. Once again, the quality of the logical operation in D_2 is worse because of the larger difference between the frequencies of the corresponding transitions due to a larger value of the rovibrational coupling constant α_e.

The above considerations did not include fidelity loss due to transitions caused by the finite turn-on time of the aligning field. The smallest energy gap is between $J=0$ and $J=2$. Hence, most sensitive to the nonadiabaticity of the turn on are the transitions $|J=0\rangle \rightarrow |\tilde{J}=0\rangle$ and $|J=2\rangle \rightarrow |\tilde{J}=2\rangle$. Figure 4 shows the projection of the state created from $|J=0\rangle$ on the pendular state $|\tilde{J}=0\rangle$, for different turn-on times. Calculations were done for N_2, with a $\sin^2$-shaped turn on, and the same field strength as that required for the CNOT gate. One can see that already a 6 ps turn on transfers the molecule into $\tilde{J}$ state with the probability 0.9997.

Fidelity loss can also be caused by collisions with other molecules. The collisional time is of the order of 1 ns at room temperature and atmospheric pressure. Thus, keeping the density below 10^{-3} atm will ensure that collisions are completely negligible on a 100 ps time scale. Finally, radiative transitions in the ground electronic state for homonuclear diatomics are dipole forbidden and are hence negligible as a loss mechanism.

Now, let us discuss the possibility of blockwise logical gates. We shall consider two kinds of such operations: a single-qudit rotation in the v domain and a single-qudit rotation in the $\tilde{J}$ domain.

The first operation implies that we should choose two values of v, say, $v_1 = 0$ and $v_2 = 1$, and for all values of $\tilde{J}$ implement the same part of Rabi oscillation between the states $|v_1, \tilde{J}\rangle$ and $|v_2, \tilde{J}\rangle$. Similarly, a single-qudit flip in $\tilde{J}$

dimension implies implementing the same part of the Rabi oscillation between the states, say, $\tilde{J}=0$ and $\tilde{J}=2$ for all v.

The same physical procedure that makes the single-qubit rotation in the above 2×2 system can also produce the qudit rotations. Let us consider the v domain first. As seen in Fig. 2(b), the aligning field $\mathcal{E}_a^0$ that equalizes the matrix elements $\Omega(\tilde{J})$ for $\tilde{J}=0$ and $\tilde{J}=2$ also brings them quite close to those for $\tilde{J}=4,6$; for higher $\tilde{J}$ alignment is negligible and the matrix elements are very close to those for $\tilde{J}=6$. At $\mathcal{E}_a^0$, $\Omega(2)$ and $\Omega(4)$ differ only by 7%. By choosing the time of the operation to be $\tau = \pi/2\langle \Omega(\tilde{J}) \rangle$, where $\langle \Omega(\tilde{J}) \rangle$ is the average for $\tilde{J}=0,2$ and $\tilde{J}=4,6,\ldots$, we will choose the length of the Raman pulse with an accuracy of 3.5%. For the flip of populations, this will introduce an additional error of only 0.003. However, we stress that this technique will only work for a molecule with small rovibrational constant α_e, such as N_2, and not for D_2. A large value of α_e would lead to large detunings, making it impossible for the coupling strength to surpass all of them enough to ensure $\approx 100\%$ fidelity, without inducing unwanted transitions.

Similarly, the field that makes a single-qubit operation in the $\tilde{J}$ domain in the 2×2 system will produce the same operation between columns forming a qudit in the $\tilde{J}$ domain: the matrix elements for Raman transitions in the $\tilde{J}$ domain are virtually independent of v. High fidelity of this operation will also require a small rovibrational constant (ensuring small detunings between the frequencies of the $\tilde{J}=0$ to $\tilde{J}=2$ transitions for different values of v).

The specific examples considered here not only illustrate the two-knobs principle of quantum control, but also suggest a quantum computing perspective on quantum control. The goal of implementing logical gates may not necessarily be the achievement of quantum computation *per se*, but rather the construction of a general approach to controlling multidimensional wave packets. A complex problem of coherent control can be considered as a specific quantum computation, since both approaches design methods to obtain certain distributions of quantum amplitudes in dependence of the initial distribution. Hence, as in any quantum computation, a *logarithmically small* and universal set of logical gates might, in principle, be used in a quantum algorithm achieving the desired quantum state in a polynomial number of steps. This is why it may be useful to think of a general quantum control problem in terms of universal logical operations, such as CNOT and single-bit gates. Moreover, it is necessary to investigate logical gates which operate with large groups of levels rather than with two-level bitlike subsystems, since only such large "qudit" operations can provide the exponentially small basis for a multilevel quantum computation.

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