

Schemes for robust quantum computation with polar molecules

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We show how ultracold polar molecules, suggested as a new platform for quantum computation, can be manipulated to switch “on” and “off” their strong dipole-dipole interactions. This can be accomplished through selective excitation of states with considerably different dipole moments. We discuss different schemes for quantum gates using real molecules: CO, LiH, and CaF, as examples of polar molecules which are being experimentally studied at ultracold temperatures. These schemes can be realized in several recently proposed architectures.

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Quantum computing is one of the most rapidly developing areas in physics today. For certain tasks, quantum computers have significant potential to outperform classical computers [1]. Quantum-information processing makes use of quantum superposition in which the fundamental piece of information, called a *qubit*, consists of a superposition of quantum states, denoted $|0\rangle$ and $|1\rangle$. One important building block of a quantum computer is a 2-qubit “gate” operation, in which a coherent change in the state of one qubit can be brought about through a carefully controlled interaction with another qubit, and the result is dependent on the state of the second qubit. In order to implement reversible quantum logic operations it is essential to address these quantum states coherently. A variety of physical systems have been proposed as possible platforms for quantum computing. A key challenge in all of these approaches is to identify *strong* and *controllable* interactions that would allow for the creation of fast quantum operations with minimal decoherence.

Polar molecules present a promising new platform for quantum computation [2,3], because they incorporate the prime advantage of both neutral atoms [4] and trapped ions [5], i.e., long coherence times and strong interactions, respectively [6]. Here we explore the possibility of switching “on” and “off” the strong interactions between polar molecules by taking advantage of the fact that the magnitude (and sign) of a molecular dipole moment can change depending on the state of the molecule. By exciting the molecule between a state that exhibits a strong dipole moment and a state with a dipole moment of zero, the interactions can be effectively turned on and off, thus helping to simplify phase gates and minimize decoherence. This approach is very different from others using molecules, e.g., via vibrational eigenstates [7] and optimal control [8].

It should be noted that our scheme could be integrated into condensed matter physics architectures, using, for example, molecule chips [9] or microtraps connected to superconducting wires [6,10]. In a recent article [6], the experimental implementation of quantum information processing using superconducting stripline resonators has been studied in detail; our approach based on dipolar switching can be readily adapted to this architecture. Advances in cooling [11] and storing [12] techniques are beginning to make the precise manipulation of single molecules possible.

In this paper, we investigate the implementation of universal two-qubit logic gates in realistic ultracold polar molecules. In this “dipolar switching” scheme, we use the fact that all heteronuclear molecules have (a) different dipole moments depending on their electronic, vibrational, or rotational states, and (b) zero expectation value for the dipole moment in the $N=0$ rotational state.

We first describe the generic setup to obtain a phase gate, or universal two-qubit operation, in Fig. 1. We assume that the molecules are individually addressable by optical or microwave fields, and choose $|0\rangle$ and $|1\rangle$ as, for example, hyperfine states, within a zero-dipole-moment manifold in a level with a long coherence time and $|e\rangle$ is a metastable state in a large-dipole-moment manifold. Single-qubit rotations can be accomplished with optical or microwave fields. The initial states of two individual sites A and B can be prepared in a superposition state, e.g. using $\pi/2$ Raman pulses. A one- or two-photon transition couples $|1\rangle$ and $|e\rangle$ coherently, but not $|0\rangle$ and $|e\rangle$. This can always be accomplished by either polarization or frequency selection. The molecules interact via a dipole-dipole interaction only if both are in the $|e\rangle$ state, and acquire a phase $\phi(t)$. After a time $t=\tau$ such that $\phi=\pi$, we coherently stimulate the states $|e\rangle$ back to $|1\rangle$. This can be summarized by

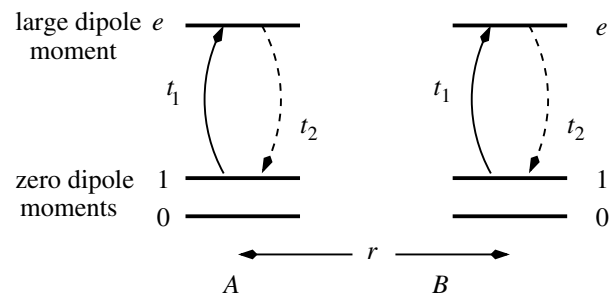


FIG. 1. Phase gate: Two molecules A and B separated by r are prepared in a superposition of states $|0\rangle$ and $|1\rangle$. At $t_1=0$, we excite $|1\rangle$ of both into $|e\rangle$: Both interact via dipole-dipole interactions and acquire a phase ϕ . At time $t_2=\tau$ such that $\phi=\pi$, we stimulate coherently both $|e\rangle$ back to $|1\rangle$.

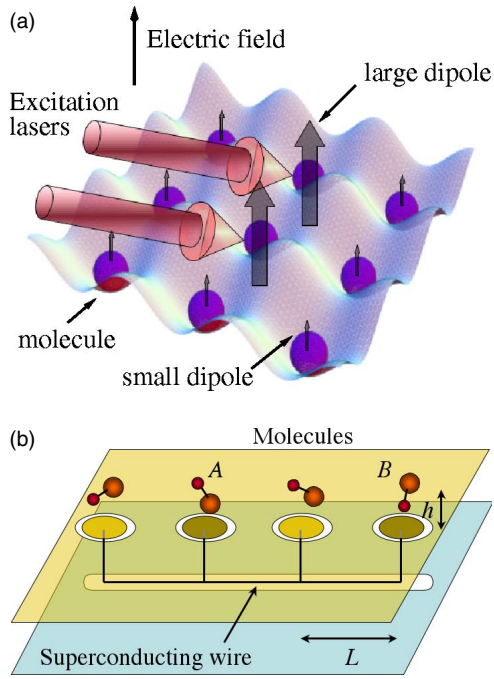


FIG. 2. (Color online) Setups: (Top) Molecules individually addressable by lasers are stored in an optical lattice; (bottom) superconducting wires are used to “deliver” the interaction. In both, molecules are selectively excited, and interact only if both are in $|e\rangle$.

	π -pulse		dip-dip		π -pulse	
$ 00\rangle$	\rightarrow	$ 00\rangle$	\rightarrow	$ 00\rangle$	\rightarrow	$ 00\rangle$
$ 01\rangle$	\rightarrow	$ 0e\rangle$	\rightarrow	$ 0e\rangle$	\rightarrow	$ 01\rangle$
$ 10\rangle$	\rightarrow	$ e0\rangle$	\rightarrow	$ e0\rangle$	\rightarrow	$ 10\rangle$
$ 11\rangle$	\rightarrow	$ ee\rangle$	\rightarrow	$- ee\rangle$	\rightarrow	$- 11\rangle$

The resulting transformation corresponds to a phase gate. We desire the wave function, expressed as $e^{-iE\Delta t/\hbar}$, to acquire a phase shift of π , thus becoming $e^{-i\pi}$, as A and B experience dipole-dipole interaction in state $|e\rangle$. The π -phase shift produced in the time τ between the exciting and deexciting π -pulses is given by

$$\phi = \pi = \frac{1}{\hbar} \int_0^\tau d\tau' \frac{d^2}{r^3} (3 \cos^2 \theta - 1) \rho_e^2(\tau'), \quad (1)$$

where d and ρ_e are the dipole moment and fractional population in the excited state, r the distance between molecules A and B , and θ the angle between the dipole moments. This formulation allows for finite excitation and deexcitation times and imperfect π pulses.

To implement the scheme, molecules are stored in a 1D or 2D array so that their dipole moments could be aligned by an electric field perpendicular to the array. We assume the full development of the storage and addressing capabilities of two recently proposed architectures (see Fig. 2). The first is an optical lattice with a lattice spacing of about $1 \mu\text{m}$, as suggested by DeMille [2]. Using a dc field for dipole alignment during trapping naturally allows the repulsive dipole-dipole interaction to aid with homogeneous distribution in

the lattice. In this case, addressing single qubits can be accomplished by either using the inhomogeneous dc electric fields proposed by DeMille to create individualized transition frequencies or by individual addressing with light in the visible part of the frequency spectrum. The second architecture is based on a “stripline wire” architecture, as suggested at Yale and Harvard [6,10]; molecules sit on their own small microwave traps which also serve for addressing, and are connected via a superconducting wire that allows for long-range dipole-dipole interaction, effectively replacing the $1/r^3$ term in Eq. (1) with $1/h^2r$, where h is the distance of the molecules from the wire. Here, all the fields need to be in the microwave range.

We now describe three possible setups utilizing variations of our switchable phase-gate scheme. The first system is based on carbon monoxide (CO). As far as dipolar molecules are concerned, CO is an anomaly; while its electronic ground state $X^1\Sigma^+$ has a very small dipole moment ($\mu \approx 0.1$ D in the vibrational ground state which is expected to be the easiest to trap), there exists a very long-lived ($\tau_{\text{life}} \approx 10\text{--}1000$ ms) excited electronic state $a^3\Pi$ with a large dipole moment, $\mu \approx 1.5$ D. As “0” and “1,” we choose, for example, two hyperfine states of $X^1\Sigma^+$, $v=0$, $N=0$ of ^{13}CO [13]. With a hyperfine splitting of about 1 MHz, selective excitation from $|1\rangle$ to $|e\rangle$ is possible. The transition frequency between $X^1\Sigma^+$ and $a^3\Pi$ is in the UV (about $48\,000 \text{ cm}^{-1}$), and the optical lattice architecture would be the ideal choice. With a coherence time in an optical lattice of a few seconds [2] and possibly much less [14] and a necessary dipole-dipole interaction time of several milliseconds, there can be about 10^3 operations. The scheme, however, is very straightforward, the techniques are in place or nearly so, and CO is a very well studied molecule [15].

A more common situation can be found in molecules such as alkali hydrides or mixed alkali dimers, e.g., LiH or LiCs. These molecules have large permanent dipole moments μ (as large as 7 D) in their ground electronic state $X^1\Sigma^+$ (for $|0\rangle$ and $|1\rangle$), and a metastable electronic state $a^3\Sigma^+$ (for $|e\rangle$) for which the potential well is located at large nuclear separation and supports at least one bound state; in most cases, these triplet states have permanent dipole moments close to zero. These properties can be used to implement a scheme in all important points similar to the CO scheme, except for three details. First, the phase gate would be “inverted,” i.e., $|00\rangle \rightarrow -|00\rangle$, $|01\rangle \rightarrow |01\rangle$, $|10\rangle \rightarrow |10\rangle$, and $|11\rangle \rightarrow |11\rangle$. Second, it requires the molecules to be stored, with the help of an aligning dc electric field, in the large-dipole state which would most likely lead to seriously shortened coherence times. In addition, the interaction would happen for *all* molecules, not just the two we wish to be coupled by a phase gate. However, this can be mitigated by switching on an aligning dc field only during interaction times, and for exactly a 2π phase shift.

For any molecule in a pure $N=0$ rotational state, the expectation value of its dipole moment is zero. Such states can acquire a dipole moment by the application of an electric field that mixes $N=0$ and $N=1$ states. So, by adding together the 2π phase shift using a dc field and the “negative” π phase shift for the molecules in the $|e\rangle$ state, the phase gate is given by

	exc+dc	π	deexc	dc
$ 00\rangle$	\rightarrow	$ 00\rangle$	\rightarrow	$- 00\rangle$
$ 01\rangle$	\rightarrow	$ 0e\rangle$	\rightarrow	$ 0e\rangle$
$ 10\rangle$	\rightarrow	$ 0e\rangle$	\rightarrow	$ e0\rangle$
$ 11\rangle$	\rightarrow	$ ee\rangle$	\rightarrow	$ ee\rangle$

Note that the scheme described for CO could be adapted for these molecules by using two different vibrational states of $a^3\Sigma^+$ as $|0\rangle$ and $|1\rangle$, and a low-level vibrational state of $X^1\Sigma^+$ as $|e\rangle$.

The last setup we propose here is the “rotational scheme.” It utilizes the fact that in the rotational ground state $N=0$ the dipole moment μ is, in fact, zero. We choose for all states the electronic and vibrational ground state. While $|0\rangle, |1\rangle$ are also in the rotational ground state $N=0$, $|e\rangle$ is the *superposition* of neighboring rotational states $|e\rangle = |e_1\rangle + |e_2\rangle$, as shown in Fig. 3 [16]. Because both $|0\rangle$ and $|1\rangle$ are in the absolute ground state with exactly zero dipole moment, this system has several advantages: Maximum coherence time, ease of storage, and no residual dipole-dipole interaction. Moreover, any polar molecule can be used with this scheme, as long as it has at least two hyperfine states. Interesting choices would be NaCl or CaF with a dipole moment of up to 10 D.

Given the fact that rotational levels are spaced in the GHz range and thus only low-frequency photons are required, this scheme is suitable for both the optical lattice and the superconducting wire architectures; for a dipole moment $\mu = 10$ D, $r = 10 \mu\text{m}$, and $h = 0.1 \mu\text{m}$, the necessary interaction time is of the order of $3 \mu\text{s}$. With a coherence time of the order of 100 ms–1 s, this setup would thus allow for 10^5 – 10^6 operations.

If the sites can be addressed individually and the dipole-dipole interactions are very strong, the previous schemes could take advantage of the so-called dipole *blockade* mechanism. This mechanism has been introduced for quantum-information processing with Rydberg atoms [17], and generalized to mesoscopic ensembles [18] as well as van der Waals interactions [19]. The original dipole blockade proposal [17,18] relies on a rapid “hopping” of the excitation between the energy levels of two Rydberg atoms, leading to an effective splitting of the doubly excited state. When this

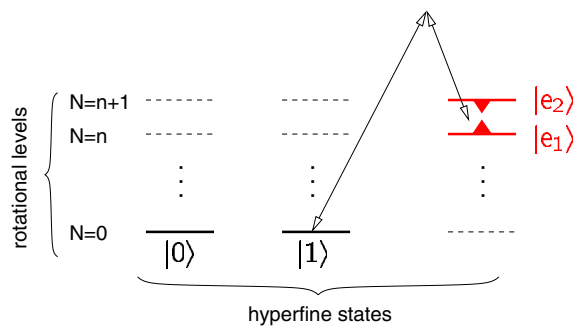


FIG. 3. (Color online) Example of level system for “rotational scheme”: All states are part of the electronic and vibrational ground state. $|0\rangle$ and $|1\rangle$ are in different hyperfine states. $|0\rangle, |1\rangle$ are $|N=0\rangle$ states; $|e\rangle \propto |e_1\rangle + |e_2\rangle$ is a superposition between two adjacent rotational states $|N=n\rangle$ and $|N=n+1\rangle$.

splitting is sufficiently large, the energy levels are shifted far away from the unperturbed atomic resonance, effectively eliminating the transition to this doubly excited state; one atom can be excited into a Rydberg state, but additional Rydberg excitations are prevented by the large energy shifts.

In a similar fashion, the blockade mechanism can be generalized to polar molecules. If the dipole-dipole interaction is strong enough, i.e., larger than the bandwidth of the excitation laser, the doubly excited state corresponding to $|ee\rangle$ will be shifted out of resonance and never excited. If both sites A and B are addressable individually, the ability to drive a 2π transition in site B depends on whether site A is excited (see Fig. 4). At t_1 , we apply a π pulse to molecule A and populate the state $|e\rangle$. At t_2 we apply a second pulse (2π) to molecule B . If A is already in $|e\rangle_A$, the dipole-dipole interaction shifts the state $|e\rangle_B$, the photon is off-resonance; hence no transition. If A is not in $|e\rangle_A$, B acquires a phase of π after the process. At t_3 , we deexcite A with another π pulse; in summary,

	t_1	t_2	t_3	
$ 00\rangle$	$ 00\rangle$	$ 00\rangle$	$ 00\rangle$	
$ 01\rangle$	$ 01\rangle$	\rightarrow	$- 01\rangle$	
$ 10\rangle$	\rightarrow	$i e0\rangle$	\rightarrow	$- 10\rangle$
$ 11\rangle$	\rightarrow	$i e1\rangle$	\rightarrow	$- 11\rangle$

This scheme is robust with respect to the separation between the molecules; as long as the excitation is blockaded, the exact separation is not important.

A key operation at the end of several qubit operations is the readout of the quantum registers. Several approaches could be employed with polar molecules. For example, selective ionization of one of the states (0 or 1) and the detection of molecular ions can be readily accomplished. However, this is a destructive method, since the molecule is lost after the readout, and the site would need to be refilled. A different method uses a “cycling” fluorescent transition in which the molecules decay after irradiation directly back into the state from which they came. Although this might be more difficult for molecules than for atoms because of the large number of molecular levels, it offers the advantage of being “nondestructive.” Another approach based on recent work on evanescent-wave mirrors for polar molecules might yield promising results [20]; while “0” would stick to the wall, “1” could be reflected. Because reflection takes place far away from the surface of the mirror, it might help to minimize decoherence due to shorter range interactions with the surface. The schemes described above may suffer from various sources of error. As our schemes rely only on internal mo-

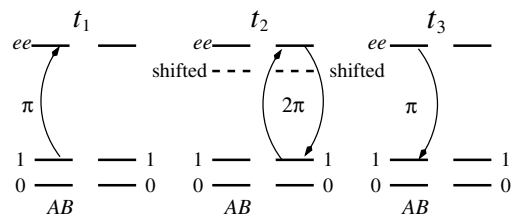


FIG. 4. Principle of the dipole blockade (see text).

molecular states, there is a possibility that some of the molecules may be translationally “hot.” If molecules are not in the motional ground state of the trap, there can be considerable uncertainty and variation in the separation between molecules, which can affect the exact phase. For example, during $a \sim 1 \mu\text{s}$ gate time, the motion of RbCs molecules at $10 \mu\text{K}$ can lead to $\sim 3\%$ variation in the phase. We can control and reduce such error, e.g., using molecules with larger dipole moments, larger separations, shorter gate times, or lower temperatures. Note that decoherence and uncertainty due to molecular motion can be completely eliminated using *dipole blockade*, leading to higher fidelity [18].

Finally, several technical issues may affect the implementation of our schemes, such as turning on and off electric fields, misalignments of dipoles, decoherence in an optical lattice (e.g., incoherent photon scattering or ionization), or imperfect excitation pulses; these can be overcome, e.g., by trapping molecules with evanescent-wave mirrors, using stimulated Raman adiabatic passage or chirped pulses. Other physical effects such as dc and ac Stark mixing, spontaneous

decay of metastable molecular states, or rovibrational quenching if sites contain more than one molecule, can be avoided by a judicious choice of molecules and states, and careful loading of sites.

In summary, we propose a different platform that combines the advantages of both neutral atoms, such as long coherence times, and trapped ions, such as strong interactions. Contrary to ions, the interactions can be made “switchable,” a feature helping to simplify phase gates considerably and thus to minimize decoherence. Using these techniques, up to 10^6 operations should be obtainable in the available coherence time. Finally, the possibility exists to bridge the gap with condensed matter devices, using polar molecules instead of quantum dots in circuits with superconducting wires that convey the interaction.

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