

Clocked Molecular Quantum-Dot Cellular Automata

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Abstract—Quantum-dot cellular automata (QCA) is an approach to computing that eliminates the need for current switches by representing binary information as the configuration of charge among quantum dots. For molecular QCA, redox sites of molecules serve as the quantum dots. The Coulomb interaction between neighboring molecules provides device–device coupling. By introducing clocked control of the QCA cell, power gain, reduced power dissipation, and computational pipelining can be achieved. We present an *ab initio* analysis of a simple molecular system, which acts as a clocked molecular QCA cell. The intrinsic bistability of the molecular charge configuration results in dipole or quadrupole fields that couple strongly to the state of neighboring molecules. We show how clocked control of the molecular QCA can be accomplished with a local electric field.

Index Terms—Molecular electronics, nanotechnology, quantum dots.

I. QUANTUM-DOT CELLULAR AUTOMATA

MODERN computing has achieved phenomenal success based on the use of binary numbers encoded as on or off states of current switches. However, scaling current switches down to the molecular level presents a host of problems, including complications arising from charge quantization, and high levels of power dissipation. An alternative approach, quantum-dot cellular automata (QCA) [1]–[4] retains the use of binary digits but represents the digits with the electronic charge configuration of a cell rather than states of current switches. This eliminates the need to use molecules as current switches and allows them to assume the more natural role of structured charge containers. For this scheme, charge quantization is an advantage because we are interested in the positions of individual electrons. It also has been shown that power dissipation in QCA can be reduced to a tolerable level, while true power gain is possible [5].

In the QCA approach, devices are composed of cells, each of which contains a small number of quantum dots. A dot, in this case, is simply a region in which charge is localized. Fig. 1(a) illustrates a schematic four-dot QCA cell. This cell has two extra mobile electrons (or holes), which tend to minimize their mutual Coulomb interaction by occupying opposite corners of the cell. The cell therefore has two degenerate ground states, which we associate with a binary “0” and “1.”

Computation requires interactions among bits or, in this case, among cells. If a second cell is placed near the first, the electrostatic interaction between the cells removes the degeneracy and

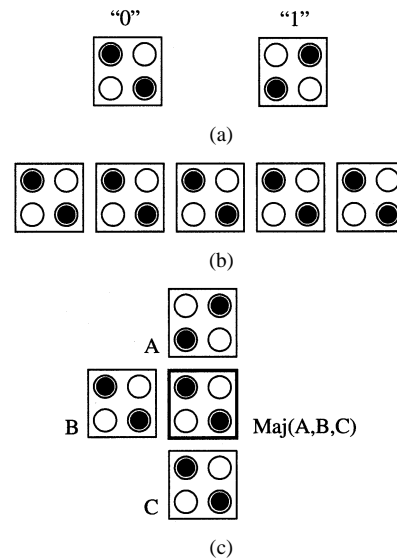


Fig. 1. Schematic of four-dot QCA cells and QCA devices. (a) Four-dot cell. Coulombic repulsion causes the two electrons (red circles) to occupy opposite corners of the cell. (b) QCA binary wire. Cells arranged in a line all take on the same value. (c) QCA majority gate. The three input cells A, B, and C vote on the state of the device cell (shown with a darker outline).

determines the ground state of the first cell. A key feature necessary for QCA operation is that the interaction between cells is nonlinear. The cells should “click” into essentially completely aligned configuration (“1” or “0” as appropriate) with even a small perturbation from a neighboring cell. It is also noteworthy that the interaction between cells involves no current flow from one cell to another.

QCA devices are constructed by arranging cells near each other an appropriate layout. Arranging cells in a line, as in Fig. 1(b), results in a QCA wire, which transmits binary information from one end to the other. The natural logic gate is the three-input majority gate shown in Fig. 1(c). Three input lines converge at a device cell, whose state is determined by the state of the majority of the inputs. Building from these, more complex circuits, such as adders [3] and even simple microprocessors [4] have been designed.

QCA devices using single-electron switching have been demonstrated using small metallic dots at low temperatures [7]–[14]. In these devices, the role of the dot is played by a small metal island connected to other islands by tunnel-junction barriers. The tunnel junctions act as capacitors that couple the potential on one dot to that of others and provide a path for quantum-mechanical tunneling. These experiments have shown that the rearrangement of single charges in one cell can indeed effectively change the state of neighboring cells and perform computational tasks. Majority logic gates, wires, memories, and shift-registers have all been demonstrated.

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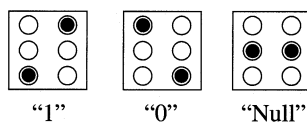


Fig. 2. Schematic of a clocked six-dot QCA cell. By varying the potential of the middle dots, we either pull the electrons onto the middle dots, resulting in a “null” state of the cell, or push the electrons off of the middle dots, resulting in a “1” or “0” state of the cell.

For large circuits it is desirable to add the ability to clock the movement of information from cell to cell. A clocked QCA cell is shown schematically in Fig. 2. A middle dot is added on each side of the cell and, with it, the ability to vary the potential on the middle dots. When the potential is large and attractive, the mobile charges are pulled onto the middle dots, as in Fig. 2(c). We refer to this as the “null” state of the cell; it contains no bit information. If the potential shifts to become large and repulsive, the charges move to the corners and the cell switches to a “0” or “1” state depending on the states of neighboring cells. A large repulsive potential acts as a barrier to charge tunneling, locking the cell in its state. Clocked QCA cells and more complex clocked circuits have been demonstrated in the metal-dot implementation [11], [12].

Clocking not only permits control of information flow around the circuit, it also enables true power gain in QCA devices. As information moves from stage to stage, signal energy is lost to the environment due to unavoidable dissipative processes (e.g. phonon emission). That energy must be restored at each stage or the information itself will be lost. Thus each stage must have actual power gain, augmenting a weak input signal to restore logic levels. In conventional transistor-based circuits, this restoration of energy in the signal path is accomplished by current from the power supply. In clocked QCA the energy comes from the clock without the flow of current. The clock does work on the cell to restore cells to the fully polarized “1” and “0” state. Power gain in QCA circuits has been analyzed theoretically for both the metal-dot [14] and molecular cases [5]. Power gain has been measured experimentally in clocked metal-dot QCA devices [14].

Important issues of reliability and defect tolerance in QCA systems have been studied [15]–[17], [37], though much more remains to be done. Another frontier involves rethinking computer architecture on the basis of QCA cells rather than transistors. Important steps have been taken in evolving both new strategies [18]–[21] and applications [22]–[24], but again, much work remains.

II. MOLECULAR QCA

The QCA paradigm is particularly well-matched to the challenge of molecular electronics [25]. Encoding binary information in the charge configuration of a molecule is a more natural match to molecular function than pressing molecules into service as current switches. Furthermore the most fundamental obstacle to realizing the full potential of molecular electronics is the need to dramatically lower the power dissipation per device. If we take the ultimate target of molecular electronics to be the development of single-molecule devices, then device densities could approach $10^{14}/\text{cm}^2$ (for 1 nm^2 devices). Any cur-

rent-switched device at reasonable speeds would melt the chip at those densities. The QCA approach has been shown to be able to reduce power dissipation by many orders of magnitude [5].

The role of dots in molecular QCA is played by redox centers within the molecule. A redox center can add an electron (be reduced) or lose an electron (be oxidized) without breaking chemical bonds. The redox center is characterized by low-lying nonbonding orbitals. We are particularly interested in two classes of redox centers, those whose nonbonding orbitals are comprised principally of s and p states, and those comprised principally of d states from transition metals. An example of the former is Si-phthalocyanine [26]; an example of the latter is the Ru-based Creutz–Taube ion [27]. Molecules with at least two redox centers, between which an electron can move, are called mixed-valence compounds, about which a considerable literature exists. It is from the class of mixed-valence compounds that we can seek QCA molecules. Synthetic approaches based both on transition metals and on π -bonded systems are currently under investigation.

In a pioneering contribution to molecular electronics, Aviram [28] examined a model mixed-valence compound. He considered a particularly simple model system composed of two allyl groups connected by an alkyl bridge. The allyl group is a 3-carbon complex which has a low-lying nonbonding π orbital and acts, in our terms, as a dot. This double-dot system was considered theoretically by Aviram and later by Hush *et al.* [29]. Aviram proposed a model system in which such a complex could act as the gate of a conducting molecular channel, resulting in a molecular field-effect transistor. Hush and coworkers used the two-allyl molecule to examine the interplay between field effects and the nuclear relaxation that accompanies the charge transfer.

Here, we use the same dot system as that first proposed by Aviram but consider a three-dot molecule that can act as a clocked molecular QCA cell. Molecule 1, which is shown in Fig. 3(a), is composed of three allyl groups connected by alkyl bridges in a “V” shape, shown schematically in Fig. 3(b). We consider the molecular cation, which has two allyl groups that are neutral and one allyl group that has a positive charge. The neutral allyl radical has a single unpaired electron in the nonbonding orbital so the transfer of an electron from one allyl group to another can be viewed as a hole moving. The three possible configurations of 1 are shown schematically in Fig. 4. The position of the green dot (representing a hole) shows which of the allyl groups is positively charged, with an empty nonbonding level. The other two allyl dots have an unpaired electron in the nonbonding level and are charge neutral.

Molecule 1 represents a clocked QCA half-cell. If the hole is in one of the upper dots, as shown in Fig. 4, it encodes a “1” or “0” bit. If the hole is in the lower dot, it represents a “null” state. For an isolated molecule the “1” and “0” configuration have the same energy by symmetry. The symmetry is broken by the presence of a similar nearby molecule or an input driver. The “null” state, with the hole in the central dot, may have a different energy. In this case, we find that its energy is actually lower than the two information-bearing states. The clocking field is a local electric field in the z -direction [see Fig. 5(a)], which can force the hole up into the active (“0” or “1”) state or back down into

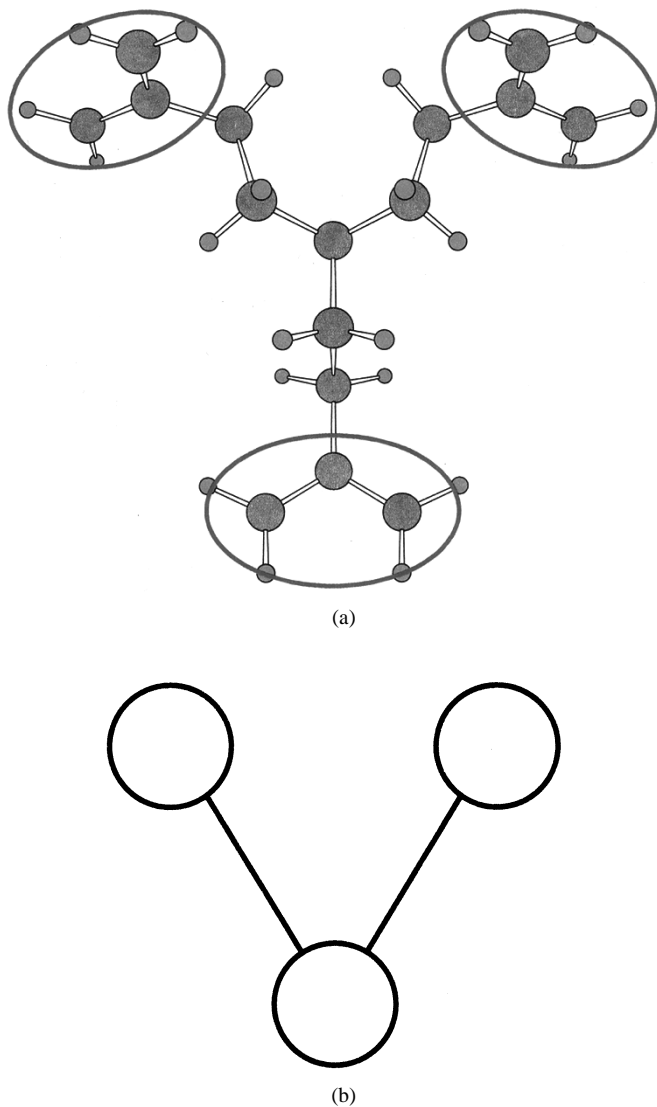


Fig. 3. Molecule 1. (a) Grey circles represent nuclei of carbon atoms; blue circles represent nuclei of hydrogen atoms. The molecule has three allyl groups (circled in red). These groups function as dots. (b) Schematic representation of molecule 1. Each circle represents a dot.

the “null” state. Because of the direction of the field, the clock is not biased toward the “0” or “1” state; that is determined by the interaction with the neighboring cells. We are not concerned here with the source of the clocking field. Reference [31] describes one approach to generating appropriate clocking fields using buried conductor wires. Importantly, the wires can be an order of magnitude larger than the molecules themselves and cause regions of the molecular circuit to be active at any one time.

Molecule 1 is a simplified system that captures some key QCA features; it is not a complete molecular QCA implementation. For a practical QCA device, we would need to add functional units for attachment of the molecule to a surface with the appropriate orientation. A neutral molecule, or an ion with a well-controlled counter-ion, would also be preferable to a charged species. Finally, one needs a means of inputting and reading data at the edges of the cell array. In the present work, we aim only to show that molecule 1 has the two most basic properties necessary for clocked QCA operation: It responds in

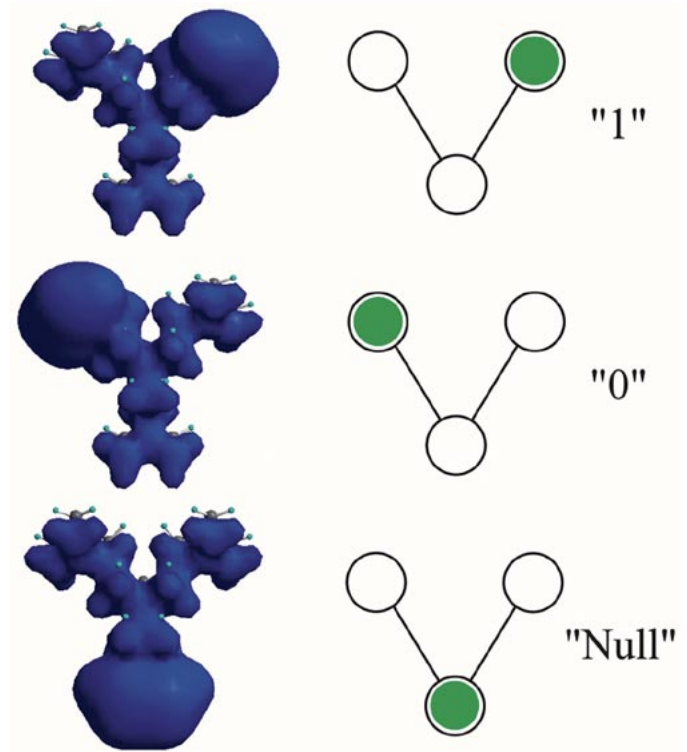


Fig. 4. Three charge configurations of molecule 1. The calculated isopotential surfaces of the three states are shown on the left, and schematic representations of the states are shown on the right. The three states correspond to occupation of each of the three allyl groups by the excess positive charge, or hole. In the schematic drawings, the hole is represented by a green circle. The presence of the hole on an allyl group causes a bulge in the iso-potential surface around that group.

a nonlinear fashion to the influence of a another molecule, and it can be clocked with a local electric field.

III. *AB INITIO* CALCULATIONS

All calculations reported here were performed with Gaussian 98 [32] using unrestricted Hartree–Fock theory and the STO-3G basis set. This theory level and basis set was used by Aviram and Hush and has the advantage of computational tractability. Improvements in computational power since those two studies allow us to solve for the molecule as a whole in a variety of conditions, rather than optimizing components separately. Higher levels of accuracy are not crucial to the present discussion because we are interested in the gross features of how charges respond to changes in the local field due to neighboring molecules in what is admittedly a simplified model system [33].

A. Response to Dipole Driver

We examine the response of 1 to the field produced by a single charge driver. The driver is meant to mimic the effect of another nearby molecule in a “1” or “0” state. We use a very simple approach, modeling the switching of the positive charge from one driver dot to another by moving a positive unit charge (hole) along a line segment. As shown in Fig. 5(a), the hole moves along a line parallel to the line connecting the charge centers on the upper two allyl groups (between the solid circle and dotted circle in the figure). The line along which the hole moves is

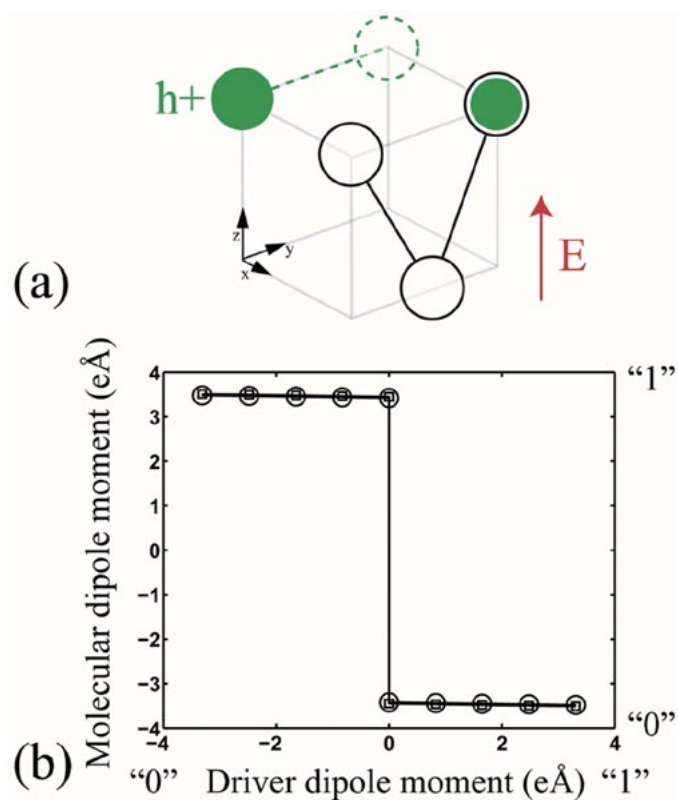


Fig. 5. Response of molecule 1 to a driver. (a) Arrangement of molecule, hole (driver), and electric field. The driver (labeled “h+”) moves along the dashed line. (b) Calculated response of molecule to driver. The induced dipole moment of the driver μ_{molecule} is plotted as a function of the driver dipole moment μ_{driver} . We associate a bit value of “1” with a positive dipole moment and a bit value of “0” with a negative dipole moment. Note that the response curve exhibits the nonlinear bistability required for QCA device operation. Data represented by circles correspond to the case when the nuclear positions are held fixed in a symmetric geometry, and the ground-state electronic configuration is calculated. Data represented by squares correspond to the case when the nuclear positions are allowed to relax self-consistently with the electronic energy minimization.

6.6 Å from the line connecting the charge centers on the allyl groups, the same as the distance between the charge centers on the allyl groups. The driver dots and test cell upper dots thus form a square. An applied clocking field in the z-direction is necessary to push the hole into the upper dots, putting the molecular cell in the active state where it can respond to the field of the neighbor.

Fig. 5(b) shows the calculated response of the molecule to the driver—the cell-cell response function. The state of the driver is characterized by the electric dipole moment μ_{driver} and the state of the molecule by μ_{molecule} [34]. For each position of the driver charge, we calculate the ground state configuration of the molecule and the corresponding induced dipole moment. For this calculation, a clocking field of 1.3 V/nm was applied. The nonlinearity evident in the response curve shown in Fig. 5(b) is ideal for QCA operation. Even a small driver dipole moment induces a large dipole moment in the molecule. Viewed as a device transfer function—output signal versus input signal—Fig. 5(b) corresponds to that of a high-gain inverter. A first approximation to the response of a chain of such molecular cells is obtained by iterating the transfer function, which here would result in a

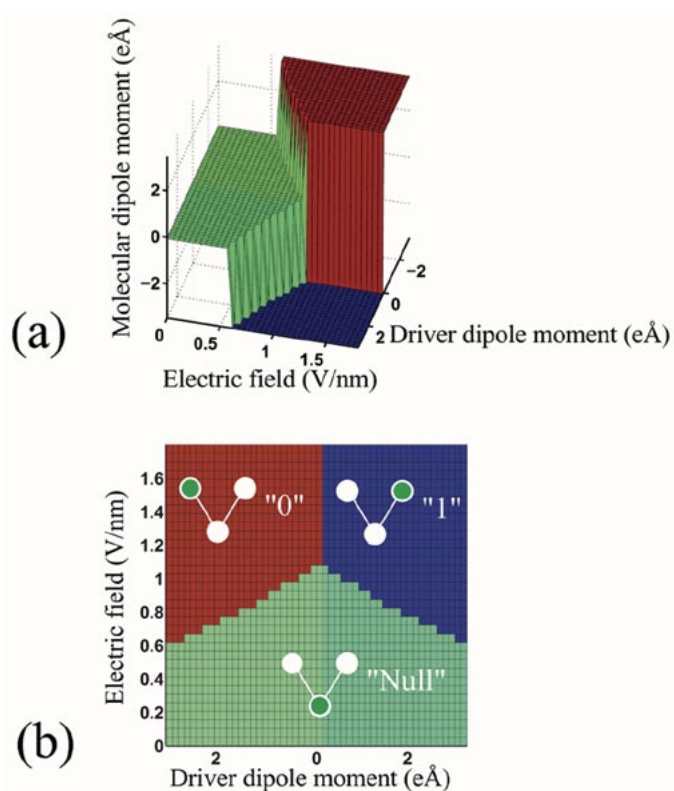


Fig. 6. Calculated response of molecule 1 to driver and varying electric field. (a) Molecular dipole moment μ_{molecule} as a function of the electric field E and the driver dipole moment μ_{driver} . (b) Phase diagram of the clocking response. The red area corresponds to the molecule in the “0” state; the blue area corresponds to the molecule in the “1” state; the green area corresponds to the molecule in the “null” state.

fully saturated inverter chain. Each molecular dipole would induce the opposite dipole in its neighbor.

When charge transfers between dots, there is a corresponding relaxation of the nuclear positions within the molecule. Data represented by circles correspond to the case when the nuclear positions are held fixed in a symmetric geometry, and the ground-state electronic configuration is calculated. Data represented by squares correspond to the case when the nuclear positions are allowed to relax self-consistently with the electronic energy minimization. It is clear that although nuclear relaxation may shift the absolute energy of the ground state, it has essentially no effect on the induced dipole moment.

The nonlinearity evident in the response curve shown in Fig. 5(b) is ideal for QCA operation. Even a small driver dipole moment induces a large dipole moment in the molecule. Viewed as a device transfer function—output signal versus input signal—Fig. 5(b) corresponds to that of a high-gain inverter. A first approximation to the response of a chain of such molecular cells is obtained by iterating the transfer function, which here would result in a fully saturated inverter chain. Each molecular dipole would induce the opposite dipole in its neighbor.

We consider the effect of varying the clocking field as well as the driver. Fig. 6 shows the induced molecular dipole moment as a function of both the electric field E and of the driver dipole moment μ_{driver} . For fields greater than about 1.1 V/nm, we obtain a response similar to the one shown in Fig. 5(b). However,

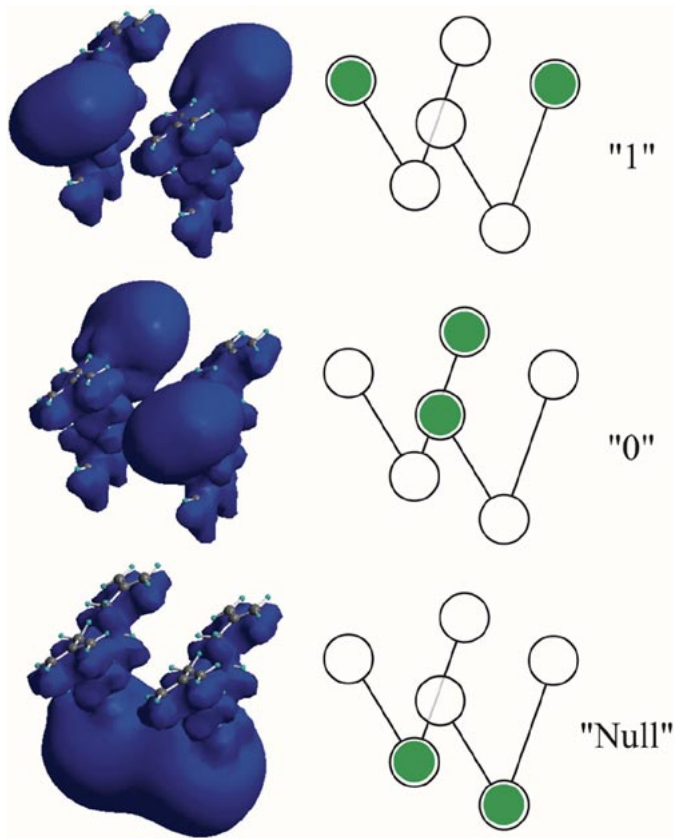


Fig. 7. Ground states of the double molecule. The calculated isopotential surfaces for the three states of the two-molecule cell are shown on the left, and schematic representations of the states are on the right. As for the single molecule, the holes (represented by green circles in the schematic drawings) give rise to bulges in the iso-potential surfaces.

for fields less than about 0.6 V/nm, the dipole moment of the molecule is essentially zero for all values of the driver dipole moment. The molecule is then in the null state, i.e. the positive charge is always on the lower allyl dot, and does not contribute to the dipole moment. This is exactly the situation required for clocking; the field determines whether the molecule is in the null state, unresponsive to inputs, or in the active state, when it responds in a nonlinear way to the input. By changing the nature of the lower “null” dot, molecules could be designed to be in a “normally-active” or “normally null” state. We note also that fields of this magnitude, though large, are quite attainable across single molecular layers [35].

B. Response to Molecule Driver

We now use two molecules of type 1 to construct a six-dot clocked cell like that shown schematically in Fig. 2. Our purpose is to verify that one molecule can switch another molecule. Fig. 7 shows the two molecules placed side-by-side, a distance 6.6 Å apart, again chosen so that the top dots form a square. Fig. 7 shows the calculated isopotential surfaces when the double-molecule is in the “null,” “1,” and “0” states. For the quantum chemistry calculations, the two-molecule cell is treated as a single molecule.

We consider the response of the cell to a driver. The state of the cell is now characterized by the electric quadrupole moment, evaluated in the plane of the four upper dots, with the coordi-

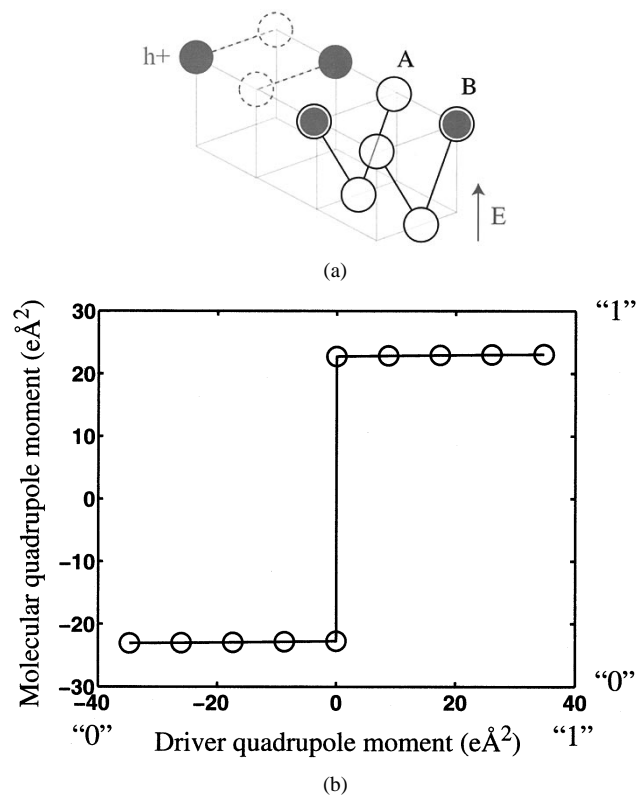


Fig. 8. Response of two-molecule cell to driver. (a) Geometry of two-molecule cell and driver. (b) Cell-cell response function. The induced quadrupole moment Q_{molecule} is shown as a function of the driver quadrupole moment Q_{driver} . A positive (negative) quadrupole moment is associated with a bit value of “1” (“0”). The nonlinear, bistable nature of this response function is ideal for QCA operation.

nate origin in the center of the square. We model a comparable molecular driver by two holes, as shown in Fig. 8(a): one on an axis 6.6 Å away from the nearest molecule (designated as molecule A in the figure) and the other on an axis 6.6 Å away from the first hole and 13.2 Å away from molecule A. This preserves the square aspect ratio of both the cell and the driver. As before, the driver charges move parallel to the line connecting the charge centers on the upper allyls of each molecule. As before, as well, we apply an electric field in the z-direction to model the clock.

Fig. 8(b) shows the calculated induced quadrupole moment of the double molecule Q_{molecule} as a function of the quadrupole moment of the driver Q_{driver} in the presence of an clocking field of 2.6 V/nm in the positive z-direction. Again, we see the strongly nonlinear bistable transfer characteristic that is so desirable for QCA operation. It is important here to note that the result shown in Fig. 8 is evidence that one molecule is switching another molecule. The driver’s direct effect on molecule A is to push it into the appropriate state. However, the direct effect of the driver would be to push molecule B into the “wrong” state. That B switches into the correct state, opposite that of molecule A, confirms that we are indeed seeing molecule-molecule coupling of precisely the sort required for QCA devices.

IV. DISCUSSION

The QCA paradigm presents a radical post-transistor approach to nanoscale computing, yet it retains the simplicity of

binary representation and the utility of general-purpose computation. Much work remains to be done in rethinking computing architectures appropriate for QCA-based computation. This work is integrally connected with the advancement of demonstrated functioning single-electron metal-dot QCA devices. These devices also act as prototypes of single-molecule devices that are currently being developed.

We have presented results on a simplified molecular system which does demonstrate that the key requirements for QCA operation can be met in mixed-valence molecules. QCA molecules can be designed with appropriate bistable switching behavior, and clocked control of molecules is possible using an applied electric field. While only an early step, these results encourage further theoretical and experimental work in this direction.

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