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# The Different Designs of Molecule Logic Gates

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**Keywords:** quantum control, molecular logic, qubits, molecular circuits

**ABSTRACT:** From the bottom, it is demonstrated how all the known intramolecular single-molecule logic gate architectures – semi-classical circuits, quantum Hamiltonian circuits, and qubit circuits – are different versions of the quantum control of intramolecular processes. They only differ in the way the classical input data are encoded on the quantum molecular system and how the quantum-to-classical conversion proceeds to read the output.

## 1. Introduction: Molecule Logic Gates as Ultimate Advanced Materials

The miniaturization of functional materials undoubtedly creates many benefits in our lives, as demonstrated by cellular phone and portable computers. These functional miniatures would minimize energy consumption and waste, as well as incredibly speed up information conversion. Efforts to this end have been made continuously, with progress seen day-by-day. As illustrated by the amazing examples of atom-switches[1] and molecular machines,[2] one of the ultimate goals is ~~fabrication of devices of a~~ single molecule as information conversion materials. Molecular devices, including molecule logic gates, can be regarded as the most advanced future materials or systems.

A molecule logic gate is a single large molecule electronically interacting with $N$ atomic-scale metallic electrodes performing alone an "$M$ inputs – $P$ outputs" digital logic function.[3] This molecule is supposed to be adsorbed on the surface of a bulk material whose surface valence–conduction electronic band gap is large enough to avoid any leakage current between the surface atomic scale contacting electrodes, and to preserve the molecule electronic structure.[4] The bulk part of this supporting material is playing two important roles: the stabilization of the wiring of the molecule from the atomic scale to the micron scale[4] and the definition of packaging required to preserve the atomic scale precision of the molecule gate assemblage while exposed to ambient conditions.[5]

Carter[6] and, later, Aviram[7] were the first to propose how a single molecule can embed alone all the functionalities of a Boolean logic gate, followed in 2001 by Ellenbogen and Love's single full digital adder molecule.[8] In all these cases, the molecular chemical structure was intuitively designed using the classical Kirchhoff mesh and node electrical circuit laws. During the same period, it has been demonstrated that the classical circuit laws do not apply to intramolecular circuitry[9–11] due to the preservation of the electronic coherence along the circuit each time one electron is transferred through the molecule.[11] A well-known example of this violation of the Kirchoff laws is the absence of voltage drop across a given molecular chemical group that can be used along the molecule for different purposes, from wiring to amplifying.[3,11]

## 2. From Hybrid to Mono-Molecular Electronics

Many different architectures are possible for single-molecule logic devices. One molecule plus the number of required metallic nanopads, a "nanopads–molecule–nanopads" molecular device, can construct the simplest – and very classical molecular – device. **Figure 1** illustrates a molecular device based on the topological resemblance of a field-effect transistor (FET). In this $C_{60}$ electromechanical amplifier,[12] the gate effect is produced by intramolecular interference effects controlling the extension through the molecule of the penetration depth of the tunneling electron coming from the electrodes of the nanojunction. The transconductance cumulates the effect of reducing the tip-apex-to-surface distance and closing the $C_{60}$ ~~rapid~~ gap, due to molecular level repulsion effect, while pressing with the tip apex on the $C_{60}$ molecular cage. Calculations have indicated that a larger gain can be reached in an optimized junction configuration.[13] However, gains in such molecular devices are usually not high. The drawback of this classical design is that all the three terminal devices designed or experimented so far have a very small gain and a very large on-resistance. Therefore, the number of logic layers is very limited even in a crossbar-like architecture.

Another type of architecture is to integrate all the circuitry inside a single molecule, forcing the molecule to assume the shape of an electronic logic circuit. This semiclassical mapping leads to a simple molecule logic gate, like an OR, an AND, or

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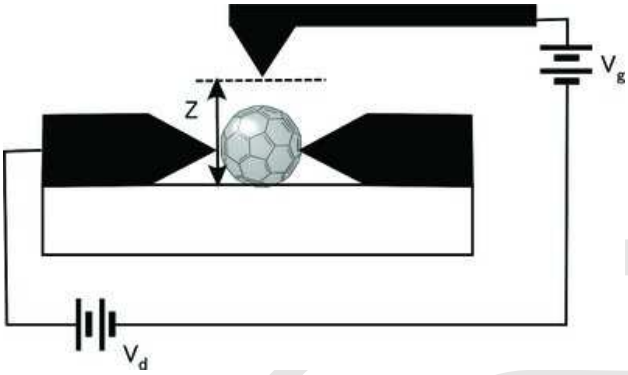


**Figure 1.** An example of a classically design molecular device. In this C60 single molecule amplifier, the $V_g$ gate voltage is controlling the $z$ distance of the tip apex to the surface. The variation of $I_d$ source–drain current intensity as a function of $z$ comes from the intramolecular electronic level repulsion triggered by the deformation of the C60 cage. A 0.1 nm deformation of the cage results in lifting of the C60 molecular level degeneracy, which cancels the destructive interference effect of the tunneling current through the C60 molecule.

an XOR gate, as presented here. The running current is very low, which forbids the extension of the lateral molecule size further than simple logic gates.

Instead of forcing a molecular structure to have the shape of an electrical circuit, we have proposed starting from the bottom and exploring how intramolecular quantum resources can be employed to fulfil a Boolean logic function[14,15] leading to the first experimental NOR single molecule logic gate operating with $M = 2$ atoms inputs.[16] Different approaches have been proposed in the same spirit, such as the qubits molecular implementation,[17] and have led to efficient arithmetic circuits in single molecules.

## 3. Advanced Quantum Logic Gates

In this short research news, we demonstrate that all the proposed designs – i.e., semi-classical intramolecular circuits,[18] intramolecular quantum Hamiltonian circuits,[19] and qubit molecular systems[17] logic gates – are all different versions of the quantum control of intramolecular processes. The semi-classical intramolecular circuit design is usually performed using a multi-channel multi-electrode scattering approach. This overshadows the elementary time-dependant electron transfer processes occurring through the intramolecular circuit. The molecular qubit approach generally describes the through bond interactions between the different intramolecular qubits by an effective coupling term without a full description of the electronic time-dependant response of the complete electronic system of the molecule. This may lead to a wrong evaluation of the intramolecular electronic decoherence effects so critical in a qubit approach. It is therefore important to bring back the quantum control concept as the centre of mono-molecular electronics, as described in the following.

For the demonstration, let us consider first an isolated quantum system (a large molecule) prepared in a non-stationary initial state $|\psi(0)>$. After this initial preparation, the intrinsic response of this quantum system is a spontaneous time evolution described by the well-known time-dependent state vector $|\psi(t)> = e^{-iHt/}|\psi(0)>$ where $H$ is the Hamiltonian of the molecule. This response can be represented by plotting the time dependant evolution of the $|\psi(t)><\psi(t)|$ density operator on a phase space called the quantum state space. $|\psi(t)><\psi(t)|$ is also known as the quantum trajectory of the quantum system. $|\psi(t)><\psi(t)|$ can be generally decomposed on a basis set leading to its real-valued coordinates. After this decomposition, a time parameterised curve is obtained which develops at the surface of a hypersphere named the Bloch sphere.[20] Any arithmetic or logic operations performed by the molecule can be interpreted as a consequence of a given $|\psi(t)><\psi(t)|$ trajectory control on its quantum state space. Because of the mathematical expression $|\psi(t)> = e^{-iHt/}|\psi(0)>$, the control parameters of the $|\psi(t)><\psi(t)|$ quantum trajectory can be located in the initial preparation or in the Hamiltonian $H$ of the molecule. These operations normally have to be performed before the decoherence of the initial $|\psi(0)>$ wave packet and before this initial preparation relaxes to the ground state of the molecule. But one solution to benefit from relaxation and decoherence is to perform a large number of $|\psi(0)>$ "preparation & decoherence (relaxation)" cycles that can be regularly or randomly spaced in time and to measure the average calculation result at the end of those cycles.

The electronic structure of the gate, i.e. the $H$ matrix elements, depends on the practical way chosen to encode the logical input and to measure the logical output of the gate. An $\{\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k\}$ digital input word can be encoded either on $|\psi(0)>$ or on some of the $H$ matrix elements. Of course if $|\psi(0)>$ encodes the logical inputs then $H$ is fixed and inversely.[20] A given $\{\beta_1,\ldots,\beta_i,\ldots,\beta_k'\}$ output word is associated with a given target state $|\phi>$. Specific characteristics of the $|<\phi|\psi(t)>|^2$ population, such as its maximum amplitude or its secular oscillation frequency, can be used to define an appropriate output measurement scheme as described below. In all the cases, the energy required to perform the logic operation is given by $|E_g - <\psi(0)|H|\psi(0)>|$ where $E_g$ is the ground state of the molecule. Depending on the selection of peculiar input and output encodings, all the known classes of single molecule logic gates can be constructed.

The first class of calculating molecules (**Figure 2**) is obtained by encoding the value of the logical input in different initial $|\psi(0)>$ states and by measuring the logical output on the population amplitude of well-selected target states. The truth table of those gates is defined by a linear and unitary transformation $B\,|\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k> = |\beta_1,\ldots,\beta_i,\ldots,\beta_k'>$. The $B$ operator is constructed for the result of the Boolean calculations to be measurable exactly at a series of time $t_n$ and the Hamiltonian reads: $H = (i/t_n)\log(B)$ because in a qubit design the $|\psi(t)><\psi(t)|$ quantum trajectory is fully periodic. Therefore, the distance between $|\psi(t)><\psi(t)|$ and the expected result $|\beta_1,\ldots,\beta_i,\ldots,\beta_k'> <\beta_k,\ldots,\beta_i,\ldots,\beta_1'|$ is exactly zero at each time $t_n$. This approach imposes that the input and output words have the same length with a one-to-one correspondence

  

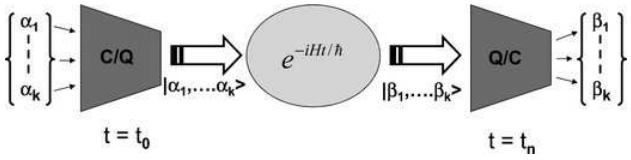


**Figure 2.** The formal set-up of a quantum computing logic gate where the central grey molecular system is essentially an organic molecule with qubits distributed along the molecular skeleton (not represented).[17] The $\{\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k\}$ classical digital input word is converted to quantum information (C/Q) encoded in the initial state vector $|\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k>$ at $t = t_0$. At each time $t = t_n$, a given $|\beta_1,\ldots,\beta_i,\ldots,\beta_{k'}>$ output state vector is reached. Its population measurement is obtained by a quantum-to-classical (Q/C) conversion which is generally a projective measurement.[17] More recently, the quantum vibration manifold of simple diatomic molecules was used to perform a fast Fourier transform demonstrating that our set-up is valid for intramolecular electronics and vibrationnal degrees of freedom.[24]

between the inputs and the outputs.[17] This first class of calculating molecules corresponds to the well-known qubit molecule approach where the qubits are positioned along the organic skeleton of the molecule and where the input and output accesses are performed by spectroscopy.[17] In this approach, a qubit can be, for example, the local magnetic moment of a transition metal in a metalo-organic transition metal complex.

The second class of calculating molecules is obtained by encoding the $\{\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k\}$ digital input words on the Hamiltonian, i.e. $H = H(\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k)$ while the $\{\beta_1,\ldots,\beta_i,\ldots,\beta_{k'}\}$ digital output words are probed on well-selected target states. Those logic gates are called Quantum Hamiltonian Computing (QHC) gates[20] and in principle only one initial driving state $|\psi(0)> = |\psi_d>$ is required (**Figure 3**a). Depending on the way the output status of the gate is measured, there are two types of QHC logic gate. The first type of QHC gate is inspired by the qubit approach: a logic output is set-up to "1" when the trajectory of the system reaches a given target state $|\phi_p>$ at $t = t_n$. A half-adder dinitro-anthracene molecule has been optimized following this approach, as presented in Figure 3b.[15] However, we have not found a systematic design rule for this design because of the extreme difficulty of getting an exact $|\psi(t)><\psi(t)|$ periodic trajectory with such quantum system.[20] In the second type of QHC gates, the $\omega_{dp}$ secular oscillation frequency between $|\psi_d>$ and a given $|\phi_p>$ target state is encoding for the output. A very fast oscillation encodes for a "1" and a very slow oscillation for a "0" logical outputs. Here, a symbolic analysis had been demonstrated which relates a $\omega_{dp}$ to a given $\{\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k\}$ digital input word.[21] The $|\psi(t)> = e^{-iHt/}|\psi(0)>$ time evolution is driven by oscillation frequencies whose values are simply the two by two differences of the $H(\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k)$ eigenvalues. Changing $H$ via the $\{\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k\}$ input words modifies the $H$ eigenvalues and therefore the $\omega_{dp}$ secular ~~frequencies~~. Notice that the $\{\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k\}$ input words are classical words that are converted into quantum information available on the system

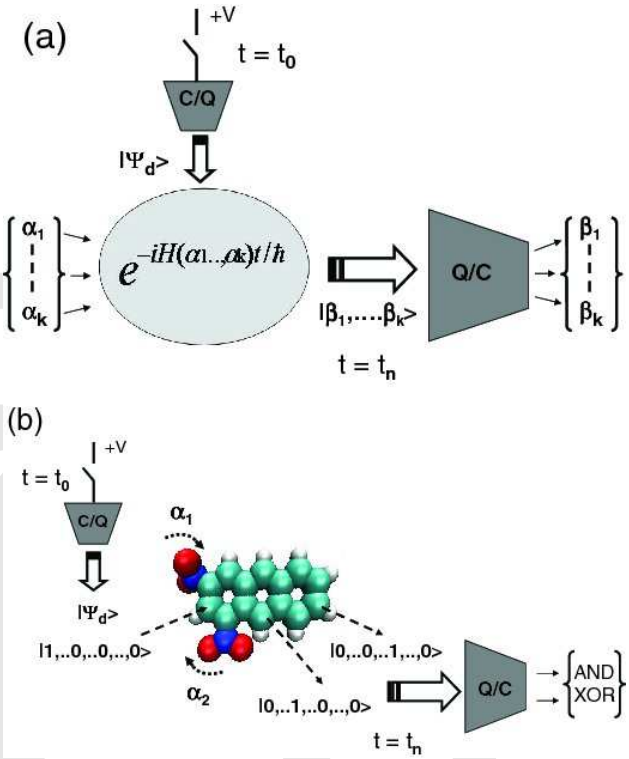


**Figure 3.** a) The formal set-up of a quantum Hamiltonian computing logic gate where the central grey molecular system is a specifically design molecule to fulfil a Boolean truth table when the $\{\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k\}$ classical digital input words are encoded on its Hamiltonian.[15] Here, the classical information is directly converted in quantum information via the change of the $H(\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k)$ eigenstates. The energy to run the logic gate is provided by the $|\psi_d>$ driving state independently of $(\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k)$. It is controlled classically to provide the $t_0$ (C/Q). At each time $t = t_n$, a given $|\beta_1,\ldots,\beta_i,\ldots,\beta_{k'}>$ output state vector is reached. Its population measurement is obtained by a quantum-to-classical (Q/C) conversion which is generally a projective measurement.[15] b) One example of a dinitro-anthracene molecule whose internal time dependent dynamic is fulfilling an half-adder digital operation. The $|\psi_d>$ initial state is supposed to be an electronic wave packet $p_z$ atomic orbital $|1,\ldots,0,\ldots,0,\ldots,0>$ localized on the indicated carbon atom. The two target states reached at the same time $t = t_n$ are also indicated on the figure. There are two well-chosen $p_z$ atomic orbitals of the indicated carbon atoms. The $\{\alpha_1,\alpha_2\}$ input digital word is the classical rotation of one of the two nitro groups. Each nitro rotation from a planar to a perpendicular conformation is changing the $\pi$ molecular orbitals system and is therefore controlling the population of the indicated carbon atom $p_z$ state.[15]

since the $H$ eigenvectors change each time a new input word is entered on $H$. All the two inputs–one output digital logic gate Hamiltonians were constructed this way.[21] From the eigenstates symmetry of those Hamiltonians, simple $\pi$ system topological Hückel matrices were established and enlarged respect-

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ing those symmetry and the position where to read the output on the quantum system. The ~~starphene NOR~~ gate molecule chemical structure (see **Figure 4**b) was determined following this procedure.[22]

One practical design of a molecule logic gate coming from this second type of QHC gates is obtained when $|\psi_d>$ and the $|\phi_p>$ are no more two non stationary quantum states of an isolated molecular system but the extended Bloch electronic eigenstates of metallic atomic scale wires interacting with the discrete electronic states of the molecule, as presented in Figure 4a. The quantum-to-classical output conversion occurs at the atomic scale electrode level and the elastic current intensity $I_{dp}$ passing from the $|\psi_d>$ ballistic channel to a given $|\phi_p>$ ballistic channel is simply proportional to the square of the corresponding $\omega_{dp}$.[23] The relation between $\omega_{dp}$ and $I_{dp}$ comes from the fact that the tunnel junction randomly prepares non-stationary $|\psi_d>$ states which are coupled electronically to $|\phi_p>$ through the molecule. The junction is acting as a band-pass filter for these Rabi-like oscillations and averages billions of such oscillation preparations per second, leading to the $I_{dp}$ measured tunnelling current.[23] Therefore, the $I_{dp}$ outputs are controlled by the effective electronic couplings between $|\psi_d>$ and the $|\phi_p>$ channels that is by the $H(\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k)$ matrix elements. The Boolean truth table results from a well-defined correspondence table between the classical $\{\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k\}$ input words and the $(I_{d1},\ldots,I_{dj},\ldots,I_{dk'})$ output current intensity words (see Figure 4b).

One inconvenience of this design is that the current flow has to pass through the complete molecular structure to capture the input configuration. This is due to the fact that there is only one $|\psi_d>$ driving channel for many $|\phi_p>$ output channels and in between a lot of canonical quantum states in charge of the gate truth table. Therefore, the current intensity is decaying exponentially with an increase of the spatial distance between $|\psi_d>$ and the $|\phi_p>$ leading to extremely small running current intensity for this type of gate.[24] This can be circumvented by increasing the number of $|\psi_d>$ driving channels as presented in **Figure 5**a, each $|\psi_d>$ being spatially localized as close as possible to a given $|\phi_p>$ to perform a very local quantum-to-classical conversion, as described above.[23] A given ($|\psi_p>$, $|\phi_p>$) couple (a very local tunnel junction) is prepared in a non-stationary state and oscillates (or not) through a little portion of molecular system at its $\omega_{pp}$ effective Rabi oscillation frequency before its decoherence. After connecting each driving $|\psi_p>$ ballistic channel electrode to the bias voltage, all the output junctions are prepared billion of time per second in a non-stationary state and the corresponding $I_p$ outputs are stabilised over time. One advantage of this approach is that when the $H(\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k)$ eigenvalues converts the $\{\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k\}$ classical words in quantum information dispatched over all the $H(\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k)$ eigenstates, this information is available all over the molecule and there is no need to pass a current through the full molecule to use this quantum information for performing a logic function as presented for the starphene NOR molecule gate Figure 5b.

Interestingly, this adaptation of our second type of QHC logic gates to improve its performances is related to the semi-classical intramolecular circuits design. By suppressing the

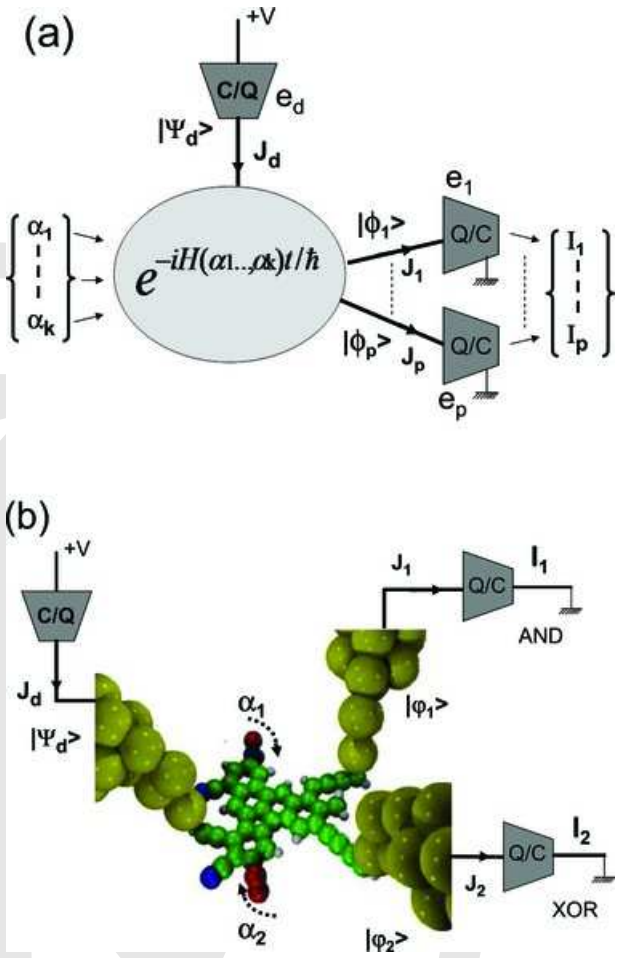


**Figure 4.** a) The formal set-up of a quantum Hamiltonian computing logic gate where the central grey molecular system is a specifically design molecule to fulfil a Boolean truth table when the $\{\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k\}$ classical digital input word is encoded on its Hamiltonian.[21] As in Figure 3, the classical information is directly converted into quantum information via the change of the $H(\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k)$ eigenstates. The energy to run the logic gate is provided by the $|\psi_d>$ driving state independently of $(\alpha_1,\ldots,\alpha_i,\ldots\alpha_k)$. Therefore, the $\omega_{dp}$ secular oscillation frequency between $|\psi_d>$ and $|\phi_p>$ is controlled by $(\alpha_1,\ldots,\alpha_i,\ldots\alpha_k)$.[20,21] The difference with Figure 3a is that $|\psi_d>$ is randomly prepared in time by the $(e_d,e_p)$ tunnelling junctions. As a consequence, the output is encoded in the tunnelling current word $\{I_1,\ldots,I_p\}$, each $I_p$ being proportional to the square of $\omega_{dp}$. In this set-up, $<r|\psi_d> = \exp(-ik\cdot r)$ and for every $p$, $<r|\phi_p> = t_p(E)\exp(-ik_p\cdot r)$ with for a low $V$ bias voltage $I_p = G_0\,|t_p(E_f)|^2$ V where $G_0$ is the quantum of conductance. The pseudo ballistic current density $J_d$ and $J_p$ can be calculated using $<r|\psi_d>$ and the $<r|\phi_p>$. b) An example of a conjugated dinitro molecule designed on purpose for the tunnelling current intensity through this molecule to be controlled by the classical $\{\alpha_1,\alpha_2\}$ input digital word. The $I_1$ output current is exactly proportional to $(\alpha_1\cdot\alpha_2)$, i.e., the AND Boolean function, and $I_2$ ia proportional to $(\alpha_1\oplus\alpha_2)$, i.e. the XOR Boolean function.[26]

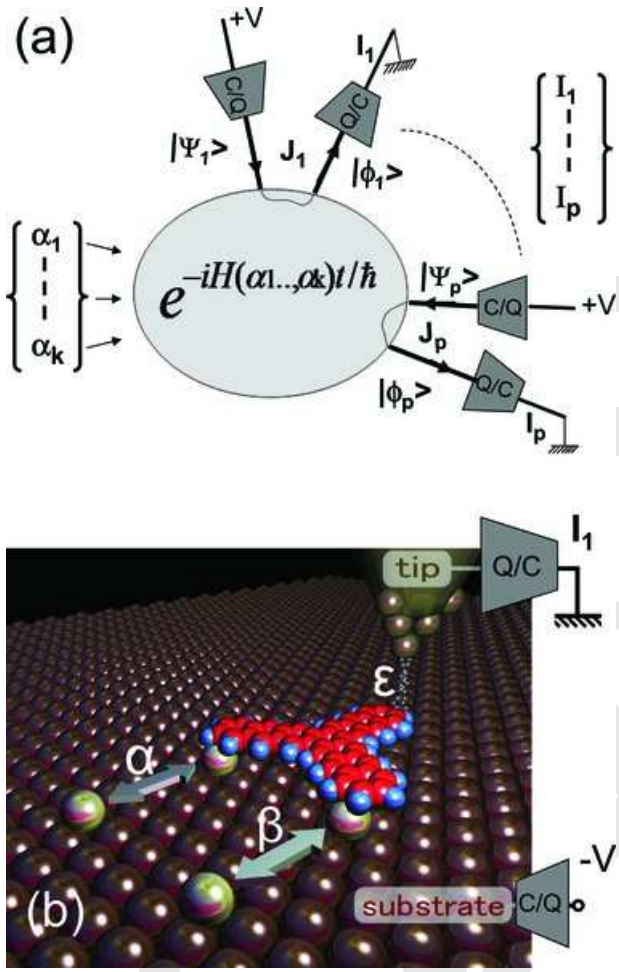


**Figure 5.** a) A more practical version of the Figure 4a formal set-up of a quantum Hamiltonian computing logic gate. To avoid Rabi oscillation frequency oscillation from $|\psi_d>$ to a $|\phi_p>$ through the full molecular system (see Figure 3b)), the tunnelling measuring junctions have been localised as close as possible to the output parts of the molecule. This essentially results from an increase of the number of $|\psi_d>$ states. The $\{\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k\}$ classical digital input word is still encoded in the Hamiltonian. b) The first experimental NOR gate molecule of this set-up where two atoms can be STM manipulated back and forth on the starphene Y-like molecule. Each time one atom is interacting with the π system of this molecule, its molecular states are modified. The consequence of this modification can be measured very locally, for example at the other end on the molecule.[16,22] Compared to the Figure 3 set-up, the tunnel current is only passing through a very small portion of the molecule. This opens the design of more complex logic gates where only quantum information is manipulated along the molecule.

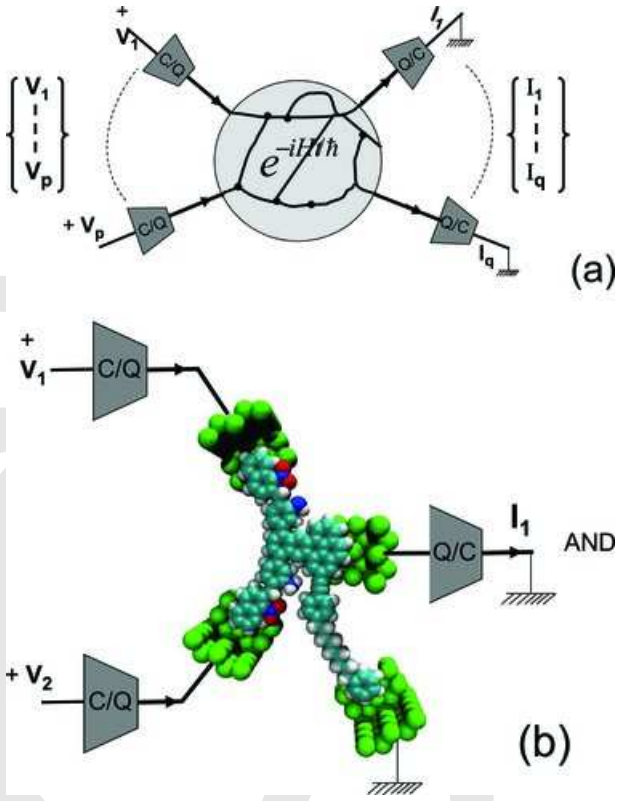


**Figure 6.** a) A semi-classical circuit version of the Figure 4a set-up where the classical input on the Hamiltonian have been suppressed and the circuit inside the molecule looks like a classical electrical circuit with nodes, meshes, and active intramolecular chemical groups (the black dots). This logic gate is still a single molecule interconnected to many atomic scale atomic wires. In the represented set-up, the information input is supposed to be encoded in the $\{V_1,\ldots,V_p\}$ classical word and the output on the $\{I_1,\ldots,I_p\}$ current intensity word. Well-adapted semi-classical circuits rules have to be applied to design such intramolecular circuits.[9–11] b) One simple example of an AND intramolecular circuits where two molecular rectifier chemical groups have been chemically bonded to long conjugated and saturated molecular wires for the $I_1$ tunnelling current intensity to be proportional to $V_1 \cdot V_2$.[18] Notice that much complex logic gates can certainly be designed this way. But the output tunnelling current intensity will not be practical, reaching intensities much lower that the aA.

$\{\alpha_1,\ldots,\alpha_i,\ldots,\alpha_k\}$ input words on the Hamiltonian, by simply encoding these words in the current intensity passing between certain ($|\psi_p>$, $|\phi_p>$) tunnel junctions and by considering also the cross-electronic coupling between a given $|\psi_p>$ and different $|\phi_m>$ channels lead to the construction of $H$ matrices whose structure can be mapped to a classical electronic circuit (see the formal set-up **Figure 6**a).[11] Therefore the semi-classical intramolecular circuits laws already demonstrated[9–11] can be easily deduced: i) by calculating the $\omega_{pm}$ Rabi oscillation frequencies per circuit elements, ii) by calculating the effective Rabi oscillation frequency of the full circuit from a $|\psi_p>$ and the different $|\phi_m>$ channels that is from branches to branches, and, iii) by using the quantum-to-classical conversion measurement per ($|\psi_p>$, $|\phi_m>$) tunnel junction to get the corresponding $I_{pm}$ per branch of the intramolecular circuit (See Figure 6b).

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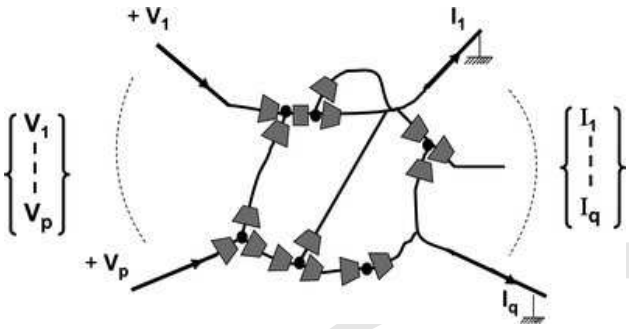


**Figure 7.** The well-known set-up of a hybrid molecular electronics circuit where all the active chemical groups of the Figure 6a design have been separated by classical non-ballistic wiring. On this figure, this is taken into account by inserting as many as required C/Q and Q/C convertors along the circuit. With this design, a lot of circuits have been proposed, like a full adder and an SRAM.[3]

Finally, when the functional entities of an intramolecular circuit are further separated by using long molecular wires, decoherence processes can be introduced along those molecular wires leading to a phase destruction of the electronic wave function in between those functional entities. In this case, nanoscale metallic wires can be used instead of molecular wires and a standard hybrid molecular electronic approach is recovered where one active device (a rectifier, a switch, or a transistor) is now a single molecule interconnected in the circuit by classical metallic nanoscale wires, as presented in **Figure 7**.

## 4. Conclusions

By addressing the rules of molecule logic gate design from the bottom, that is from a quantum control problem, we have demonstrated here that all the known molecule logic gate structures can be regrouped under the same formalism. The structure of a given class of gate is governed by the way the inputs are encoded on the molecule and by the way the output status is measured. For the simple two inputs–one output symmetric logic gates (NOR, AND, OR, NAND, XOR), only their practical implementation leads to the choice of one class of gate over the others. For more complex logic gates, like a full adder or more, the gate performance in term of current intensity, energy cost, and calculation time is a guide on how to design the logic gate molecular chemical structure. We anticipate that semi-classical circuits and qubit structured molecule logic gates may not be the best solution to achive high complexity with good logic gate performance. The seminal problem of the semi-classical intramolecular circuits is the exponential decay of their running tunnelling current intensity with their spatial lateral expansion. The qubit approach is limited from above by the necessary large increase of the number of qubits required to perform a complex logic function. This gives rise to the difficult decorence problem because their quantum spectrum is too regular. At the moment, our second QHC approach appears to be a good compromise between energy consumption, complexity, and the use of decoherence during the output measurements to run very complex single-molecule logic gates.

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