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Molecular Diodes and Applications

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Abstract: Due to the huge power consumption and expensive fabrication methods required, down scaling silicon devices to sub-100 nm dimensions is becoming very unattractive. On the other hand, it is easier to build electronic circuits using molecules since they are small and their properties can be tuned. In this review, we first discuss the building blocks of molecular electronics. We then describe how these building blocks can be used to build single molecule based digital logic such as AND, OR and XOR gates. The distinction of these molecular electronic building blocks is that for first time, (i) the Tour wires are used as the conductive backbone for the rectifying junctions, (ii) donor/acceptor principles are implemented in the molecular wire itself and (iii) the logic gates are realized using molecular rectifying diodes embedded in the molecular conducting wire itself.

Keywords: Molecular electronics, molecular diode, molecular RTD, digital logic, AND, OR, XOR.

INTRODUCTION

The fast paced developments in the field of semiconductor technology in the recent past have proved without doubt that the efforts to miniaturize the transistors will reach the physical limits earlier than anticipated due to the laws of quantum mechanics and limitations of fabrication methods [1]. Scaling down silicon devices to sub 100 nm and packing millions of them in a chip will not be an attractive idea as these systems consume huge power and will become very expensive to fabricate. Therefore, we need to look beyond silicon to make ultra-high density electronic systems and it is important that alternate methods such as using molecules to make devices at the nanoscale are explored and exploited [2]. Silicon devices operate based on the movement of a large number of electrons n bulk matter while molecular devices take advantage of the quantum mechanical effects taking place at the nanometer scale. The building blocks of molecular electronics are single or small packets of molecules. While efforts are on to replace the conventional wires and semiconductor devices with molecules, what would be desirable is to build molecular architectures with addressable electronic properties [3-5]. The main advantage of molecular electronics is the lower cost, compatibility with flexible substrates and simpler packaging when compared to the conventional inorganic electronics. A number of molecular electronics based digital circuits have already been reported [6,7] and larger scales of integration (up to 864 switches per circuit) have been realized in clocked sequential complementary circuits [8].

Since molecules are small, their functionality can be tuned because of their special properties. Either synthesis or self- assembly processes can be used to manipulate the molecules. The most important molecular backbones are: 1) polyphenylene-based chains and 2) carbon nanotubes. Unlike in the case of silicon technology, the bottom-up-approach of manipulating molecules is expected to be not only cheap but also will result in higher speeds of information processing with less power. Making electronic rectifiers using molecules was first proposed by Aviram and Ratner [9]. There are two types of molecular diodes: rectifying diodes and resonant tunneling diodes which can both be used in realizing monomolecular digital logic circuits [10]. In addition, we also need molecular wires [11-15] to connect the molecular devices into a complex circuit with specific applications [2,7,16]. In this paper, we will first discuss the above building blocks of molecular electronics and based on two recent inventions [17,18] explain their application in realizing important molecular digital electronic gates such as AND, OR and XOR gates. The distinction of the above two inventions lies in the fact that for the first time, the Tour wires are used as the conductive backbone for the rectifying junctions and the donor/acceptor principles are implemented in the molecular wire itself. In addition, this invention also reports for the first time the realization of simple logic gates using molecular rectifying diodes embedded in the molecular conducting wire itself.

MOLECULAR CONDUCTORS AND INSULATORS

There are two types of challenges in molecular electronics: (i) Building molecular structures which act as switches and (ii) combining these molecules into a complex circuit to perform a given function. Both the above tasks require reproducible organic molecular conductors and insulators [19]. The two types of molecular scale conductors that have often been suggested are polyphenylene based conductors [20] and carbon nanotubes [21].

Polyphenylene based molecular conductors are made up of organic aromatic benzene rings with bonds (Fig. **1**) or tied up by acetylene spacers (Fig. **2**). Both these conductors have been shown to conduct small electrical currents.

Polyphenylene conductors are made from two or more phenyl groups. When one hydrogen atom is removed from benzene (C6H6) (Fig. **3**), the phenyl group is formed (Fig. **4**). However, the removal of two hydrogen atoms from benzene results in a phenylene ring (Fig. **4**). A polypheny-

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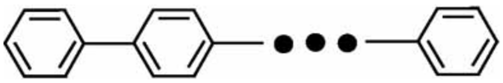

Fig. (1). Schematic representation of a polyphenylene chain [17].

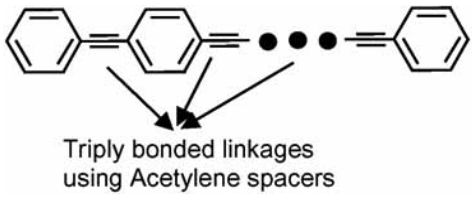

Fig. (2). Schematic representation of the polyphenylene chain in which phenyl and phenylene groups are seperated [17].

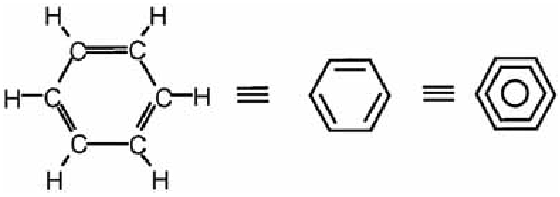

Fig. (3). Single benzene ring and its equivalent symbols [17].

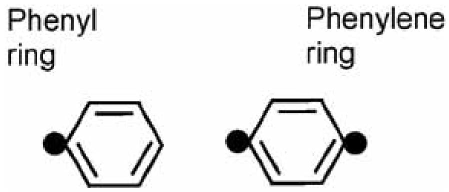

Fig. (4). Phenyl and phenylene rings [17].

lene molecule can be formed by bonding these phenylene groups resulting in a chain like molecular structure and the manipulation of these molecules has been well understood by the organic chemists. Methods are now available for producing reproducible conductive polyphenylene molecular wires, known as Tour wires, which are compatible to the molecular electronic devices so that appropriate interconnections can be established at the molecular level [22]. Polyphenylene wires conduct because of the conjugated pi-orbital lying above and below the plane of the molecule (Fig. **5**) forming a channel for electron transport when the wire is biased by an external voltage.

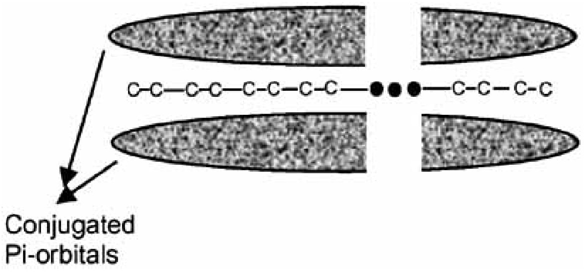

Fig. (5). Side view of the benzenoid structure showing the conjugated Pi-orbitals.

Another potential candidate for gigascale interconnection in molecular electronics is the metallic single wall carbon nanotube (SWCNT) which acts as an ideal quantum wire with current conduction taking place only in one dimension [23,24] (Fig. **6**). CNTs are also known as buckminsterfullerene tube or buckytube. Unlike in the case of conventional metallic conductors such as copper, in CNT carriers can only be backscattered giving rise long range mean free path length [25,26] Therefore, in a CNT conduction is possible only in forward or backward direction since there is significantly reduced phase space for small angle scattering events in a 1-D system. Monolayer carbon nanotubes have been shown to outperform copper interconnects [27]. Nanotubes can be connected in parallel to reduce resistance and inductance.

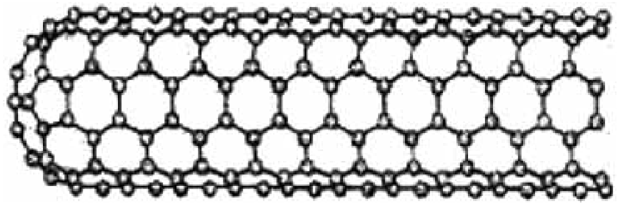

Fig. (6). Single wall carbon nanotube.

A typical organic insulator which contains singly bonded molecules (not benzene rings) are known as the aliphatic group (Fig. **7**). In an aliphatic organic molecule with only sigma bonds, the conducting channel outside the plane of the nuclei has interruptions resulting in discontinuities in the electron density at the positions of the nuclei. As a result, in the presence of an applied voltage, these molecules cannot conduct current and act as insulators. Therefore, in the polyphenylene chain, the conductive channel can be broken up by inserting an aliphatic molecule leading to the formation of insulators or barriers to the electron transport.

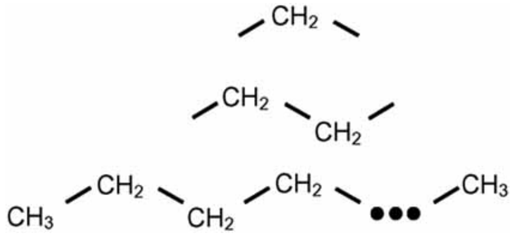

Fig. (7). Example of aliphatic molecular structures [17].

MOLECULAR DIODES

There are two types of molecular diodes - rectifying diodes and resonant tunneling diodes. In both these devices, when the applied voltage exceeds a critical value, electrons are driven through one or more potential barriers.

A. Molecular Rectifying Diode

A diode or a rectifier, which conducts only in one direction, is the building block of any three terminal semiconductor electronic device such as a bipolar transistor or a field effect transistor. Diode based logic circuits using AND/OR gates are well known for building logic families by using the rectifying diodes at the input and connecting a resistor between the supply or the ground. A molecular diode too contains two terminals and functions like a semiconductor pn junction and has electronic states which can be clearly distinguished between highly conductive state (ON) and less conductive state (OFF).

The seminal work of Aviram and Ratner in 1974 led to several experimental attempts to build molecular diodes. Aviram and Ratner have suggested that electron donating constituents make conjugated molecular groups having a large electron density (N-type) and electron withdrawing constituents make conjugated molecular groups poor in electron density (P-type). According to them, a non-centrosymmetric molecule having appropriate donor and acceptor moieties linked with a σ-bridge and connected with suitable electrodes will conduct current only in one direction - acting as a rectifier. They showed that in this *D*-σ-*A* molecule, the lowest unoccupied molecular orbital (LUMO) and highest occupied molecular orbital (HOMO) can be aligned in such a way that electronic conduction is possible only in one direction making it function like a molecular diode. Asymmetric current-voltage characteristics for a σ-bridged system were first reported in 1990 [28,29].

The structure of the mono-molecular diode proposed in [17] is shown in Fig. **8** and its schematic representation is shown in Fig. **9**. This diode is based on a molecular conducting wire consisting of two identical sections (S1, S2) separated by an insulating group R. Section S1 is doped by at least one electron donating group (X e.g. ¾ NH_2, ¾ OH, ¾ CH_3, ¾ CH_2CH_3) and section S2 is doped by at least one electron withdrawing group (Y e.g. ¾ NO_2, ¾ CN, ¾ CHO). The insulating group R (such as ¾ CH_2 ¾, ¾ CH_2CH_2 ¾) can be incorporated into the molecular wire by bonding a saturated aliphatic group (no pi-orbitals). To adjust the voltage drop across R, multiple donor/acceptor sites can be incorporated. The single molecule ends are connected to the contact electrodes e.g. gold. The band diagram of the mono-molecular diode under zero-bias conditions is shown in Fig. **10**. We notice that there are three potential barriers - one corresponding to the insulating group (middle barrier) and two corresponding to the contact between the molecule and the electrode (left and right barriers). These potential barriers provide the required isolation between various parts of the structure. The occupied energy levels in the metal contacts and the Fermi energy level E_F are also shown. On the left of the central barrier all the pi-type energy levels (HOMO as well as LUMO) are elevated due to the presence of the electron donating group X and similarly on the right of the central barrier the energy levels are lowered due to the presence of the electron withdrawing group Y. This causes a built-in potential to develop across the barrier represented by the energy difference ΔE_{LUMO}. For current to flow electrons must overcome the potential barrier from electron acceptor doped section (S2) to electron donor doped section (S1) and this forms the basis for the formation of the mono-molecular rectifying diode. The energy band diagram under forward bias conditions (left hand contact at higher potential than the right hand contact) is shown in Fig. **11**. Here, electrons are induced to flow by tunneling through the three potential barriers from right to left causing a forward current flow from left to right.

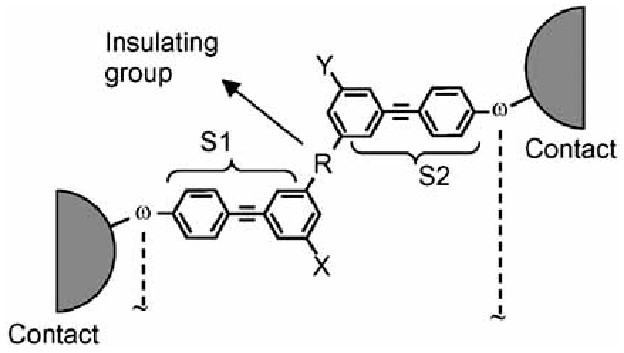

Fig. (8). Structure of molecular diode integrally embedded in a poly-phenylene based molecular conducting wire [17].

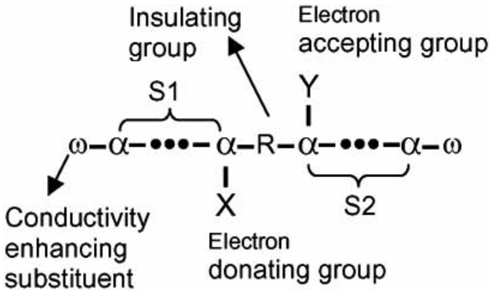

Fig. (9). Schematic representation of the molecular diode [17].

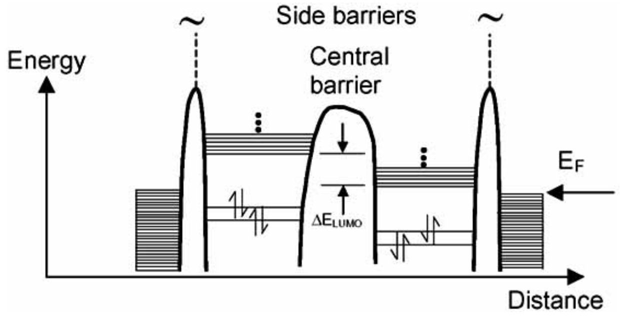

Fig. (10). Orbital energy diagram of the polyphenylene monomolecular rectifying diode under zero bias conditions [17].

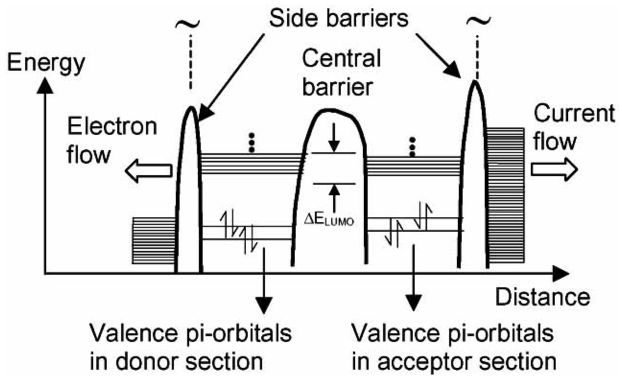

Fig. (11). Orbital energy diagram of the polyphenylene monomolecular rectifying diode under forward bias conditions [17].

The band diagram under reverse bias conditions (left hand contact at lower potential than the right hand contact) is shown in Fig. **12**. As a result, electrons from the left contact would try to flow towards the right contact which is at a higher potential. However, conduction is not possible because the there is still an energy difference between the Fermi energy E_F of the left contact and the LUMO energy of the electron donor doped section. It is assumed that both the applied forward and reverse bias potentials are identical. For a higher reverse bias, however, it is possible for the Fermi energy E_F of the left contact to come in resonance with the LUMO energy of the electron donor doped section causing a large current to flow in reverse direction and this is akin to the breakdown condition in a diode.

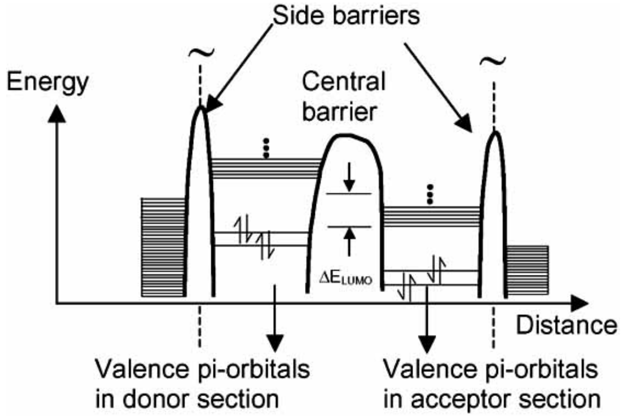

Fig. (12). Orbital energy diagram of the polyphenylene monomolecular rectifying diode under reverse bias conditions [17].

The above mono-molecular diode can be used as a building block for realizing molecular digital logic circuits.

B. Molecular Resonant Tunneling Diode

Resonant tunneling diodes or RTDs are well known [30] and have been studied widely because of their potential in very high speed/functionality circuits [31]. The RTD has been intensely researched in last three decades as a promising nanoelectronic device for both analog and digital applications [32-35]. RTDs have been fabricated using the semiconductor heterostructure epitaxial techniques to realize the peak current-voltage characteristics that are typical of tunnel diodes but without the associated problems of large junction capacitance.

The structure of a molecular resonance tunneling diode based on a molecular conducting wire backbone is shown in Fig. **13** [36,37]. In this structure, two aliphatic methylene groups (CH_2) are inserted on both sides of the benezene ring. Since aliphatic groups act as insulators, they create potential barriers to the flow of electrons in the molecular conducting wire. The only way for the current to flow in the presence of an applied voltage is when the electrons are forced to pass through the benezene whose width is only about 0.5 nm. The operation of the molecular RTD can be understood using the band diagram shown in Fig. **14** which shows (i) potential barriers due to the two aliphatic methylene groups, (ii) energy levels in the benzene ring through electron must traverse and (iii) the orbital energy levels in the molecular conducting wire.

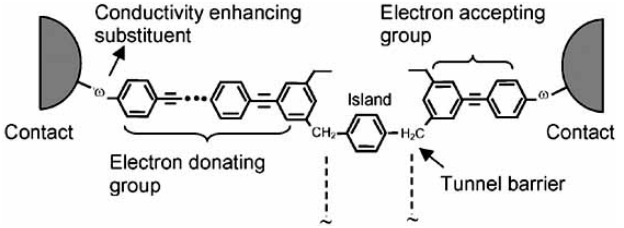

Fig. (13). Structure of molecular resonant tunneling diode [18].

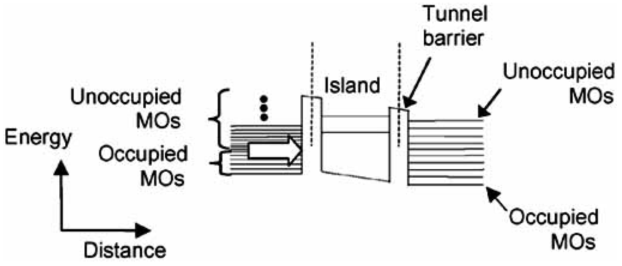

Fig. (14). Band diagram of the molecular RTD showing the 'OFF' state [18].

In the presence of an applied voltage across the molecule, the molecular RTD will not conduct any current or will remain in OFF state since, as shown in Fig. **14**, the kinetic energy of the incoming electrons is different from that of the empty energy levels in side the benzene ring sandwiched between the two thin potential barriers. However, by changing the applied voltage a resonance situation can be created i.e. the kinetic energy of the incoming electrons can be made to match with one of the unoccupied energy levels inside the benzene ring. The device will now turn ON since electrons can traverse through the wire Fig. **15**. The molecular RTD shows a negative differential resistance with two operating points following the standard mechanism of a solid state resonant tunneling diode.

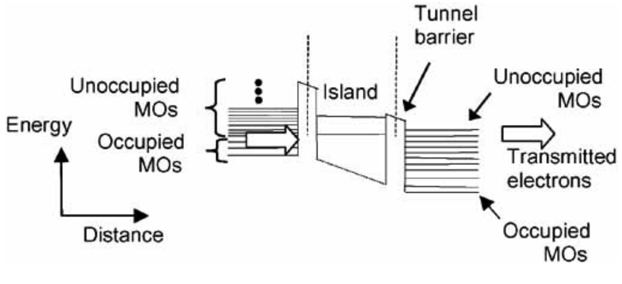

Fig. (15). Band diagram of the molecular RTD showing the 'ON' state [18].

The above discussed molecular diode and molecular RTD can be used as the basic building blocks for synthesizing a single molecule exhibiting simple Boolean logic such as OR, AND and XOR gates. By building larger single molecule structures using the above building blocks, we can even realize more complex digital circuits such as HALF and FULL adders and their derivatives [17,18].

MONOMOLECULAR DIGITAL LOGIC STRUCTURES

As an alternative to silicon device miniaturization, in the recent past, significant research efforts are going on in using molecular devices such as molecular diodes, molecular wires and molecular transistors to build basic digital building blocks such as the logic gates (AND, NOT, OR, XOR, etc.) [38]. Building complex integrated circuits at the molecular level still remains a technological challenge because of the inherent presence of defective components in any chemically assembled molecular circuit due to the imperfect yield of the chemical reactions [39] Improving the yield is important for realizing future nanoprocessors and molecular computers. Unlike in the case of conventional electronics, the molecular electronics devices and logic circuits operate based on different mechanisms such as acid/base reactions, conformation changes, photoinduced electron and energy transfer mechanisms, photoinduced isomerizations, redox processes (based on chemical species whose oxidation number, and hence electronic structure, can be changed reversibly) [40] and supramolecular chemistry. Any of these mechanisms can be used to efficiently establish the signal communication between molecular switches and logic gates. Based on the inventions made in [17,18], we describe below examples of monomolecular logic gates (AND, OR and XOR) formed for the first time using monomolecular rectifying structures realized using selective doping of the molecular conducting wire.

A. Realization of Diode Logic Molecular AND Gate [17]

The circuit representation of a diode logic AND gate is shown in (Fig. **16**) and the schematic representation of the diode logic molecular AND gate is shown in (Fig. **17**). The schematic of the exemplary poly-phenylene diode logic molecular AND gate is shown in (Fig. **18**) and uses the previously discussed rectifying wires as the building blocks. The molecular AND gate consists of two inputs A and B and one output C connected to the respective contacts. This structure exhibits the classical semiconductor AND gate behaviour. As shown in (Fig. **17**), the single molecule AND gate consists of (i) two conducting wires each having a donating section (with at least one electron donating group) as well an accepting section (with at least one electron accepting group), and (ii) a respective insulating group R inserted between the accepting and donating sections. The central barrier height can be increased by using more number of donor and acceptor sites. A conductivity enhancing substituent ω can be utilized to improve the conductivity between the molecule and the contact pad. Both the conducting wires of the molecular diodes are joined together forming a common node Q to which an aliphatic chain can be attached. Another conducting molecular wire is connected to this common node so that it can be attached to a contact pad forming the output C. This completes the formation of the larger single molecule. This entire construction of the molecular diode AND logic (or other complex logic gates) in a single molecule can be accomplished using the standard insertion and substitution techniques or other techniques such as scanning electron microscope or nano-probes.

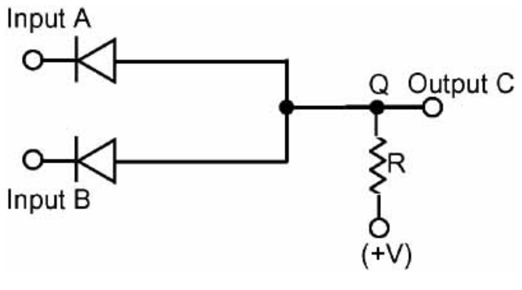

Fig. (16). Circuit diagram of the diode logic AND gate [18].

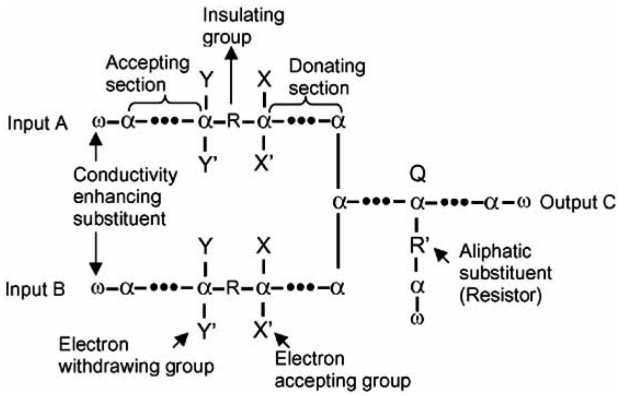

Fig. (17). Schematic representation of the diode logic molecular AND gate [18].

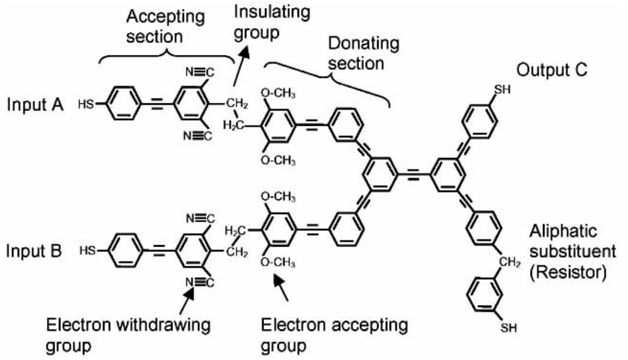

Fig. (18). Schematic of the exemplary poly-phenylene diode logic molecular AND gate [18].

What is important about the above molecular AND gate is that it is extremely small measuring only 3 nm × 4 nm in area which is at least a million times smaller than the gates realized using conventional semiconductor technology [17].

B. Realization of Diode Logic Molecular OR Gate [17]

The circuit representation of a diode logic OR gate is shown in (Fig. **19**). The schematic of the exemplary poly-phenylene diode logic molecular OR gate is shown in (Fig. **20**) and uses the previously discussed rectifying wires as the building blocks. The molecular OR gate consists of two

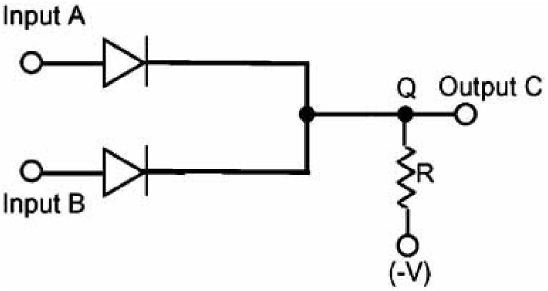

Fig. (19). Circuit diagram of the diode logic OR gate [18].

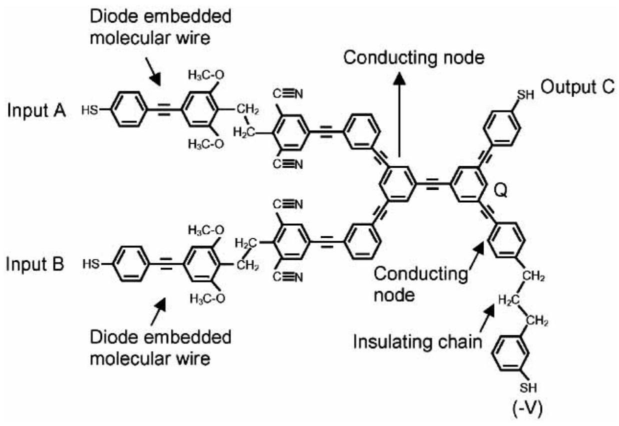

Fig. (20). Schematic representation of the diode logic molecular OR gate [18].

inputs A and B and one output C connected to the respective contacts. This structure exhibits the classical semiconductor OR gate behaviour. As shown in (Fig. **20**), the single molecule OR gate consists of (i) two conducting wires each having a donating section (with at least one electron donating group) as well an accepting section (with at least one electron accepting group), and (ii) a respective insulating group R inserted between the accepting and donating sections. The acceptor sections of the conducting wires are bonded together forming a conducting node to which an insulating aliphatic group is connected and extended by a conducting wire such as the polyphenylene based wire to act as the output C. A conductivity enhancing substituent ω can be utilized to improve the conductivity between the molecule and the contact pad. This completes the formation of the larger single molecule functioning as the classical OR gate. Just as in the case of the monomolecular AND gate, the above single molecule OR gate size is only 3 nm × 4 nm [17].

C. Realization of Molecular XOR Gate Using a Molecular RTD [17]

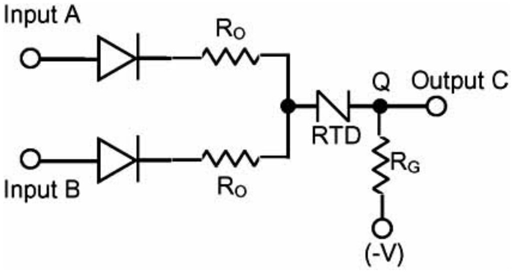

Fig. (21). Circuit diagram of the diode logic XOR gate using the molecular RTD [18].

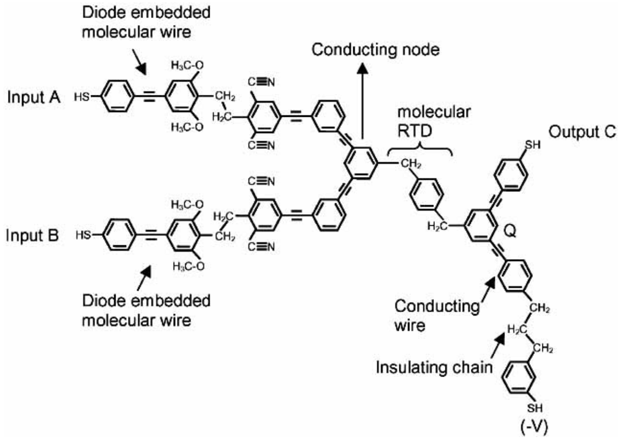

Fig. (22). Schematic representation of the diode logic molecular XOR gate using the molecular RTD [18].

The schematic gate of the XOR gate is shown in Fig. **21** and the exemplary molecular implementation of the XOR gate is shown in Fig. **22** which is similar to that of the molecular OR gate discussed above with the exception of the presence of the resonant tunneling diode. The molecular XOR gate consists of two diode-embedded molecular wires with an effective resistance R_0 and their acceptor sections are chemically bonded to a common connecting node. The resistance R_G is made of an insulating chain. One end of R_G is connected to the negative potential and the other end is connected to point Q through a conducting wire. The molecular RTD is connected between the connecting node and point Q. The molecular diode has the current-voltage behaviour as shown in Fig. **23**. Another polyphenylene conducting wire is connected between point Q and the output node C of the XOR gate of which the input nodes are A and B.

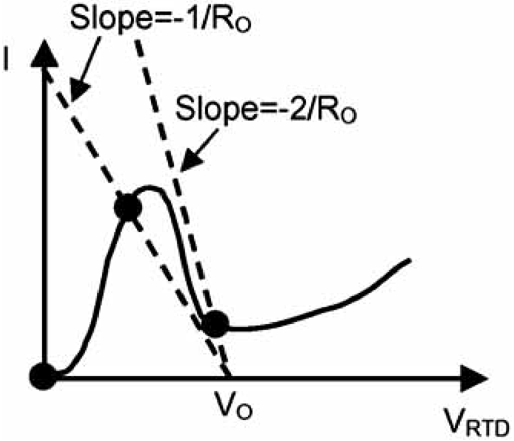

Fig. (23). Current-voltage characteristics of an RTD incorporated into the molecular XOR gate [17].

By combining the above described XOR gate with molecular AND and OR gates, it is possible to realize more complex logic circuits [17,18]. It may be noted that the molecular RTD has two operating points: one on the peak of the RTD operating curve and the second in the valley of the RTD operating curve. The input to the gates will decide in which of these two operating points the RTD works. If only one of the inputs of the XOR gate is logic 1, the RTD will turn ON forcing the output of the gate to be logic 1. If both inputs to the XOR gate are logic 1, the RTD is OFF forcing the output to be logic 0. The presence of RTD and its two operating points in the I-V curve make the molecular XOR gate to behave like the conventional semiconductor XOR gate.

CURRENT & FUTURE DEVELOPMENTS

The complete set of single molecule structures (diodes, RTDs, conductors and insulators and the logic gates based on these) can in fact be used to build more complex Boolean functions. For example, by combining the molecular AND gate and molecular XOR gate, a single molecule HALF ADDER can be built. By combining two molecular HALF ADDERS, a single molecule FULL ADDER can be built. Using the above approach, it is also possible to build an electronic switching device with power gain by adding a gate structure to a molecular diode forming a single molecule which functions as a three terminal transistor similar to the a conventional silicon field effect transistor [18]. As a result, it is now possible to realize in a large single molecule to provide molecular inverters with power gain, a possibility with immense applications in digital circuit design.

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