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A View of the Future of Molecular Electronics

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A VIEW OF THE FUTURE OF MOLECULAR ELECTRONICS

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Abstract

The article describes some prevailing thoughts and an approach to the development of molecular circuit elements for molecular electronics. Three basic principles are proposed and exemplified by a particular example of a suitable molecule. The model relies upon the current state of the art in science and technology.

Introduction

The term molecular electronics (ME) as used in this article refers to the possible future use of molecules as electronic circuit elements in signal-processing circuits, and the science and technology associated with this goal. For years, the notion of utilizing molecules to do the chores of transistors and electronic switches has been a tantalizing idea, and has recently become the target of many research programs around the world. The activity in the area is gauged by the ever increasing number of researchers who associate themselves with the field, the number of conferences and workshops dedicated to it and the outlay of capital earmarked for it around the world.

It has been shown over a period of many years that advances in miniaturization of electronic components lead to improved electronic products and services. Current commercial silicon technology is based on individual transistors and capacitors that have line widths of 0.8 micrometer. On the other hand, experimental silicon field-effect devices have been reported to have reached 1000 Å size (1), with raw inverter

delay of 13 psec. Table 1 lists one commercial company's published plans for future product announcements (2). From this table it is evident that even by year 2005 the limits of miniaturization will not be reached. Therefore, there seems to be plenty of room for further miniaturization, beyond the current technology. Numerous studies that have explored the theoretical limits of miniaturization in conventional Si devices, have come to the conclusion that the actual limit of device size in an integrated circuit (IC), could be as low as a few hundred Å. Below these dimensions semiconductor devices may not work properly because there won't be enough dopant atoms in depleted layers to maintain a potential barrier between the different regions of a device (3). Smaller dimensions could lead also to cross-talk between the components. Meanwhile other concepts for small devices have been proposed, which are different in principle from the current experimental Si devices. These include quantum interference in disordered metals (4) and resonant tunneling through successive barriers (4). These proposed devices are referred to as mesoscopic devices, and are envisioned to be of submicron dimensions. The quest for molecular electronics can be understood in the context of the trend for miniaturization. Molecular electronics is perceived to approach the ultimate limits of possible miniaturization.

Technology (in microns)	Products		Development start	Introduction date	Peak output
	SRAM	DRAM			
1.8	64 Kbit	256 Kbit	1977	1982-83	1988
1.2	256 Kbit	1 Mbit	1980	1985-86	1991
0.8	1 Mbit	4 Mbit	1983	1988-89	1994
0.5	4 Mbit	16 Mbit	1986	1991-92	1997
0.35	16 Mbit	64 Mbit	1989	1994-95	2000
0.25	64 Mbit	256 Mbit	1992	1997-98	2003
0.15	256 Mbit	1 Gbit	1995	2000-01	2006

Table 1 The projected DRAM product plan of the Motorola company

The physics and technology of conventional electronic devices and of the mesoscopic devices are completely different, yet they share in common an important feature that raises a question about the economic benefits that might be derived by switching from today's conventional technology to a mesoscopic technology. This commonality is the method of fabrication. The fabrication method of every current IC or mesoscopic experimental IC is based on the stepwise assembly of the individual electronic components that make up the circuit directly on the chip. It was found empirically that the number of fabrication steps is increasing in proportion to the density. Table 2 illustrates this point. The table lists the number of lithographic steps that have been used in the production of commercial ICs up to the present 4 Mbit dynamic random access memories (DRAMs). Bearing in mind that each lithographic step comprises of five (or more) substeps, one realizes that production of large scale integration (LSI) circuits is an involved process. In the likely event that the trend continues, it can be expected that further advances in compaction will require a yet a higher number of lithographic steps. Associated with each lithographic step must be some possibility of error which leads to a low yield of acceptable devices. As dimensions decrease, the need for fine tolerances increases.

DEVICE	REPORTED # OF MASKS	TECHNOLOGY
256 KB	6 - 8	MMOS - 1 LEVEL METALLURGY
1 MB	8 - 10	CMOS - MMOS 1 LEVEL METALLURGY
4 MB	11 - 13	CMOS - 2 LEVEL METALLURGY
16 MB	12 - 14	CMOS - 2 LEVEL METALLURGY

Table 2 The number of fabrication steps used to produce IC products.

Therefore, it would be advantageous in the future to decrease the number of production steps. Such a decrease is hard to expect from the current approach of incremental advances in the production of LSI microchips, if silicon or mesoscopic technologies would be employed.

In contrast, molecular electronics differs completely in the basic concept and in the envisioned fabrication methods from all other technologies. In principle, the components are preformed and embodied in the structure and the electronic configuration of the molecules used. Consequently, in assembling the molecular IC one does not have to build the individual components on the chip, but rather one synthesizes components (the molecules) as bulk chemicals and then, by some appropriate surface attachment techniques, interconnects them into an electronic circuit. The result of this short-cut is fewer fabrication steps, a decrease in inspection steps, for a cost effective performance. Other advantages that are perceived to be offered by ME are the highest possible density and a promise of exceptionally high speed of operation, that is associated with electron transfer events in molecules.

Mapping a Strategy for Molecular Electronics.

It is obvious that ME presents a challenge to scientists and engineers because it is based on principles and techniques that lie at the frontiers of science and technology. By coincidence, several developments have occurred in the last decade that provide an access to the molecular world. These are the invention of the scanning tunneling microscope (STM), new surface-science analytical methods, the development of surface binding techniques, such as self assembly (6), advances in synthetic metals (7), the advent of new synthetic methods for the preparation of materials that can be categorized as molecular electronic switches (8), and spectacular advances in nanolithographic techniques (9).

In addition to having the proper tools, it is also necessary to have a proper strategy that would lead to integrated molecular circuitry. The proposed strategy brought forward in this article is based on three principles that are envisioned to provide a bridge between the current state of the art and the molecular IC.

1 Planarity. Despite the fact that nature utilizes molecules in three-dimensional interconnecting networks, the first principle is that we develop a planar approach to ME. In doing so we would take advantage of the knowledge that has been gathered in constructing planar ICs by employing lithographic methods for constructing circuit connections. We would also be able to test better the ME circuit, when it is stretched in two dimensions

2 A Prepared Surface. The idea here is that all the metallurgy necessary to interconnect the molecules for the integrated ME circuit should be deposited on the surface in advance. The deposition of the " wiring " should be made with provisions of gaps that would be filled by molecules at a later stage. These spaces should be tailored to fit the dimensions of the molecules. Then, in the next step, the molecules would be added to complete the circuit. This last stage could be accomplished by utilizing the self-assembly techniques mentioned above (6).

3 One Kind of Molecule. The last principle is to provide a single kind of molecule that would perform all the needed electronic functions. This means that the electronic function would be determined by the mode of interconnections rather than the specific structure of the molecule. Thus, in one kind of interconnection two identical molecules could serve as "AND" logic gates and in another mode the same two molecules could serve as "OR" logic gates. The need for this principle is clear when considering the alternative prospect of building an IC based on only the first two principles and the requirement to bind specifically different kinds of molecules in various points of the plane, on a molecular scale, a process that would probably be very complicated.

Implementation of the Strategy

Implementing the first two principles looks rather simple in concept. The contention is that the embodiment of these principles can be realizable with today's knowledge and experience of science and technology. The news from the field of nanolithography continues to astound. It has been demonstrated in many laboratories that it is possible to fabricate patterns with line dimensions of 150 angstroms

(10). These patterns are best prepared with electron-beam lithographic techniques. Other methods of pattern formation, such as utilization of an STM are not yet ready for the task, primarily because the scanning is very slow. However, as a research tool, the STM is very valuable, since impressive demonstrations of nano-lithography using the STM, have been reported (11).

The metallurgy should be chosen in a way that would make possible the future binding of the molecular switches. The choice of the particular metal to be employed is closely related to the particular self-assembly method that one wishes to employ. For instance, from the chemistry point of view, it is possible to synthesize molecules of specific length such as, for example, 250 Å that would fit precisely into the prepared gaps that the circuit designer and the chip architect can provide. Furthermore, if the method of choice for surface attachment of the molecules is the thiol/gold coupling, then the designer would opt to provide molecular switches with some thiol end groups and a circuit metallurgy that would consist of appropriate gold metal patterns (6). Other couples of specific interaction have been developed (12) and are available for this application.

Implementation of the third principle is also within reach. Theoretical groundwork has been done that will permit the implementation of this principle (13). In addition, the chemicals that were proposed in the literature cited, have in part been synthesized in a number of independent laboratories. The total synthesis of the required compounds will probably be completed in a number of laboratories soon (14). This will open the door to the experimental physics effort that will undoubtedly follow.

A Molecular Electronic Switch based on an Insulator to Conductor Transition

A class of molecules that could be utilized for the implementation of the third principle are the spiro switches (8,13). Figure 1 shows the structure of one possible embodiment of the switch. Close examination of the molecule reveals that there are several structural principles that have been incorporated into the design. It should be noticed that we are dealing with a $\pi - \sigma - \pi$ type of molecule. This kind of structure has been shown in many studies, both theoretical (15) and experimental

(16), to retain the electronic configuration of the individual π moieties because of the so called $\sigma - \pi$ separation. The σ bridge acts as an insulator (15), because of the difference between the orbital energies for the two type of bonds, yet, at the same time, the σ bridge has the remarkable property of participating in the transfer of electrons from side to side by a mechanism that came to be known as through-bond conductivity (17).

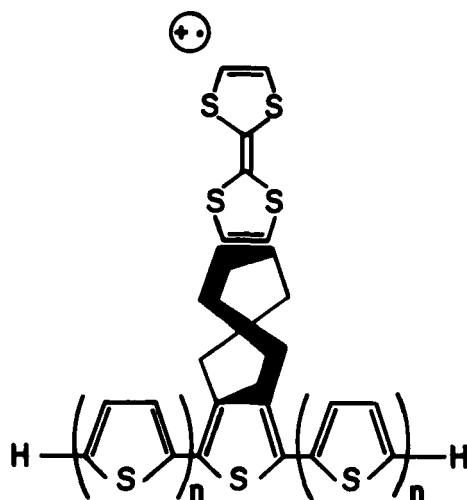


Figure 1

The π moieties in the molecule belong to a particular class of organic molecules that has been shown to include many electrically conductive polymers or oligomers. These "Synthetic Metals" have structural and electronic stability and the remarkable feature of very high electrical conductivity, comparable to metals (18). In some cases, room-temperature conductivities of these materials were registered to be as high as $10^3 (\text{ohm} - \text{cm})^{-1}$, which are almost as high as the conductivity of bismuth. Another observable feature is the unfilled orbital in one of the π groups. This feature is responsible for creating two simultaneous effects. First, it creates a situation whereby the molecule becomes a double-well-potential molecule. This comes about because one of the two π moieties has one electron more than its counterpart. This electron finds itself in a box that has the shape of a double-well potential. The physics of a particle in such a box is well understood, and as far as the molecular switches are concerned, the quantum mechanical aspects of these molecules are

rather well developed (13, 19). Second, it has the potential of rendering the π chain conductive, much in the same way that doping of polythiophene or polyacetylene with dopants that remove electrons from the polymeric chains produces conductivity in those polymers (insulator to conductor transition (8)). This will occur when an electron from the π group will migrate to the group.

The purpose of the specific stereochemistry that was incorporated into the design of the molecular switch is to localize the electron in one of the wells. Since an electron is a light particle, it tends to oscillate in double wells, due to tunneling. This oscillation can be at frequencies as high as one oscillation per picosecond. However, in the spiro switch, the spatial orthogonality between the initial and final states (the two flexomers) causes the oscillation to slow down considerably (20). This crucial effect gives meaning to the notion of an electronic switch, because of the longevity of the initial and final states. When the two π groups are identical the double well potential is symmetric and the switch can be used for memory purposes. If π and π are as shown in figure 1, the switch has the property of a transistor.

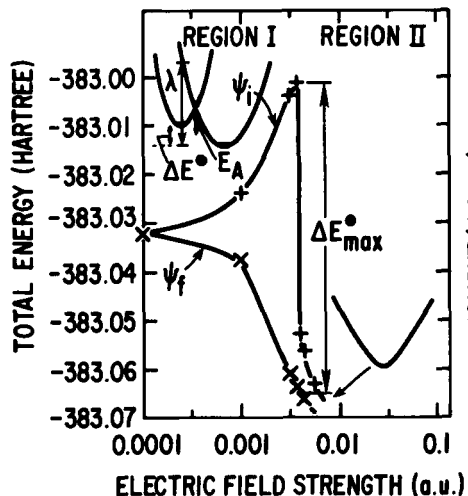


Figure 2

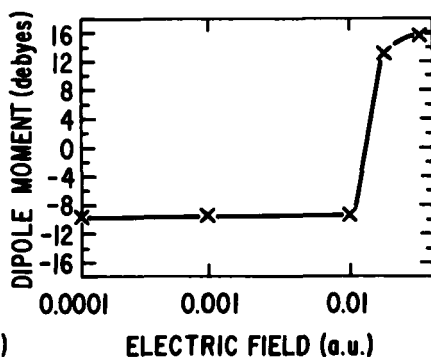


Figure 3

The Switching

Quantum mechanical MO calculations of a spiro molecule similar to the one shown in figure 1 revealed an interesting behavior in electric fields. Application of a field

along the axis that crosses the spiro carbon in the bridge and the two π rings adjacent to the bridge, causes the energy of the two states (initial and final) to shift energetically from the field free situation (8, 13); One state goes up in energy while the other goes down by the same amount. As the field is increased, this trend persists until a threshold is reached. Figure 2 illustrates this point for the molecule presented in reference 8. It is interesting to note that the dipole moment of the molecule flips by 180° when the threshold field is exceeded, indicating that an electron has transferred. The dependence of the dipole moment on the field is plotted in figure 3. When an electron transfers, the electrically insulating chain becomes conductive (8). At this stage the insulator to conductor transition occurs.

The tunneling phenomenon in a double-well potential emerged as the dominant feature in the design of molecular-electronic switches. This led to considerable activity aimed at increasing our understanding of the processes involved, particularly, the study of double-well-potential molecules in electric fields (8,13). It was found that the "inverted" effect in electron transfer reactions, plays an important role in electric field switching, since it is expected that there would be an applied electric field (F_{crit}), at which the electron transfer rate is at a maximum. This is apparent from the Marcus expression for electron transfer rates (21), for the condition where the exponent is equal to zero.

$$k = A \exp \left[\frac{-(\Delta G + \lambda)^2}{4\lambda k_B T} \right] \quad A = \frac{2\pi}{\hbar} |T_{ab}|^2 \left(\frac{1}{\sqrt{4\pi\lambda k_B T}} \right)$$

Here λ is the reorganization energy, T_{ab} , the tunneling matrix element, and ΔG , the standard free energy of the reaction. The free energy is directly related to the exothermicity of the electron transfer, which in turn is dependent on the applied electric field. The exponent in the Marcus expression can be written as $\lambda_{tot} - 2\mu_z F_z$, where μ_z is the dipole moment component parallel to the field (which includes field induced contributions).

When the exponent is set to zero, one can obtain the relationship between F_{crit} and the dipole moment.

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When the exponent is set to zero, one can obtain the relationship between F_{crit} and the dipole moment.

$$F_{crit} = \frac{\lambda}{2\mu_z}$$

From a practical point of view, one would want the value of F_{crit} to be as small as possible. This can be obtained when molecules with large dipole moments are considered. The dependence of the switching time on the electric field can be calculated by the Golden Rule formalism, by calculating the electronic coupling integral.

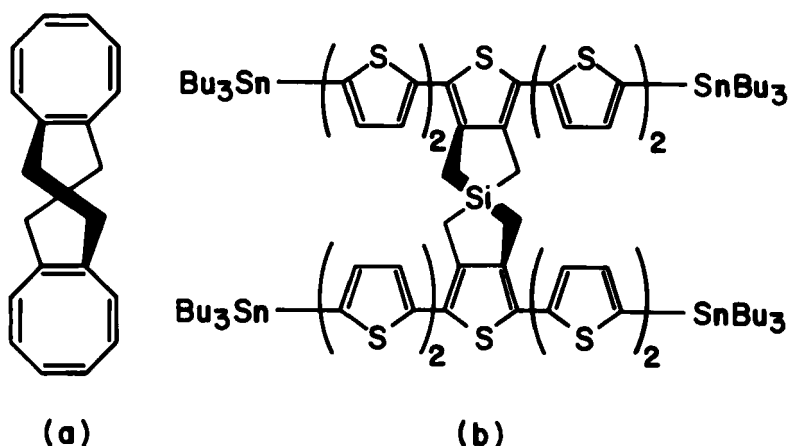


Figure 4

To this day no one has demonstrated switching of a single molecule as described by the theoretical model. However, there is ample experimental evidence for many parts of the model. The ability of the spiro bicyclo bridge to halt the process of electron transfer has been proved by the work of Mullen (20) who has studied the molecule 10,10'-spiro(bicyclo<6.3.0>undeca- 2,4,6,8-tetraene), shown in figure 4a. Measurements of the electron exchange in the related charged compound revealed no appreciable electron motion at room temperature (20). This is contrasted to other bi-cyclooctatetraene compounds that have no spiro bridge (22), which show high rates of electron oscillation. One should note that the references cited aren't ideal examples since the cyclooctatetraene is doubly charged and therefore two particles have to tunnel (in contrast to only one in the case of the molecular switch. The conformation of the rings have to substantially change. On the other hand, the molecule in reference 22, is conjugated. Therefore, the comparison is not conclusive.

In view of advances in the synthetic work of Tour, who reported the synthesis of the spiro compound shown in figure 4b, (14), and additional theoretical work that was published lately (13), additional experimental evidence is expected shortly.

The Synthesis of Logic

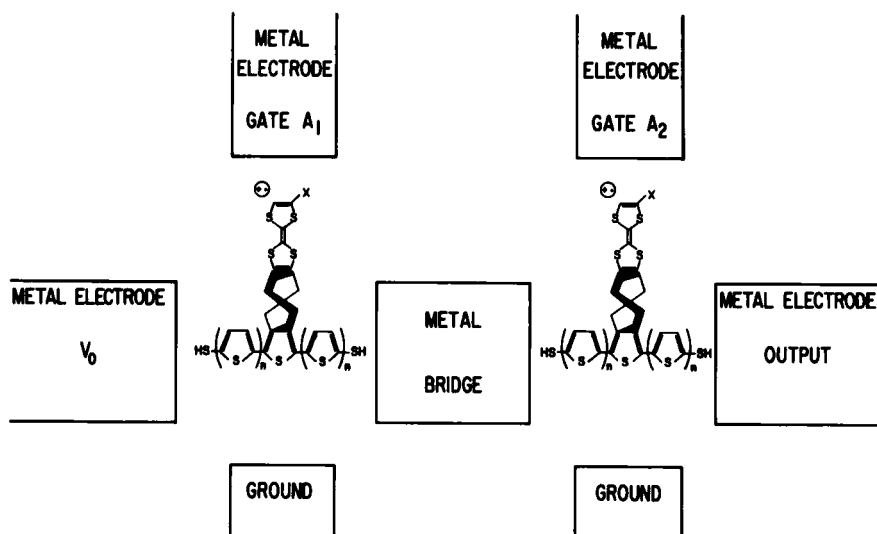


Figure 5

The class of molecules described is suitable for synthesis of all the logic operations needed for computation, and thus satisfies the third principle proposed. Figure 5 attempts to convey schematically the possible "wiring" of two identical molecules to obtain different electronic functions. These molecules have one unpaired electron in the highest occupied molecular orbital. When the thiophene segment contains the unpaired electron it becomes conductive. Thus, the "AND" gate function can be obtained when two molecules are connected in series by a metal bridge, as shown in figure 5. One molecule (shown on the left) is permanently connected to a voltage source V_0 . A_1 and A_2 are two control electrodes. Suppose that unpaired electrons (see figure 1) localize on the TTF segments that reside by the control electrodes. As a result, the other molecular segments are insulating. Therefore, no current can flow between the permanent voltage source and the output electrode. When a positive voltage is applied to one of the control electrodes, the molecular segment that is

perpendicular to that control electrode and connects the bridge and a corresponding electrode becomes conductive. This is expected to occur as a result of an electron transfer. Only when the control signals of both A_1 and A_2 are positive, will both oligomeric thiophene molecular segments become conductive and cause the output voltage to be the same as the source voltage (8). This is analogous to the operation of an "AND" gate (23).

The operation of various combinations that lead to the synthesis of other logic gates, such as "EXCLUSIVE OR" and computing cells, such as "HALF ADDER" have been presented elsewhere (8).

Such circuits could conceivably be implemented with available technology if the electrodes that lead the A_1 and A_2 signals were designed to be 150 Å wide and made of aluminum. (The proposed dimension is what is available by state-of-the-art nanolithography). The corresponding molecular segments that have to attach to them, could contain carboxylic acid groups for specificity (see Labinis et al. in reference 6). The other electrodes could be made of gold, with gaps of 250 Å between the bridge and the side electrodes. The molecular segments should be of corresponding length, with many more rings than shown in the figure, and have thiol end groups to attach specifically to the gold electrodes (6). The author wishes to emphasize that while the described structures could be made with currently available tools, the implementation would require considerable effort and team work.

Reliability and Redundancy.

From the fundamental point of view, it was demonstrated (24) that no physical limitations to molecular circuits can emerge due to quantum mechanics and the uncertainty principle. However, from the practical point of view, we may face considerable difficulties. An important point to consider is that the higher the density of circuits in a system, the higher the needed reliability of the individual components. Thus, the question arises whether molecules can be relied upon for the task of circuit components. To answer this question one has to resort to observations of nature, where individual molecules are used for information storage. Such a system exists, and of

course is the DNA molecule, where a combination of any three nucleic acids on the DNA chain encodes a bit of information. DNA is a static information recording system (memory mode) and a dynamic system (in replicating and transcribing mode). This shows that, in principle, molecules can be used in information processing devices and provides an existence theorem that legitimizes the concept of ME. However, the mere fact that DNA functions so well does not serve as a direct proof that other molecular systems would work with the same fidelity. Each individual approach to ME has to be examined on its own merit.

It is reasonable to assume that man-made molecular devices will not be as reliable as DNA. Thus, for the purpose of achieving reliability, one would have to resort to redundancies in the circuit and error correction codes. Just how redundant should the circuit be would depend on the reliability factor of particular molecules. Unfortunately there is no precise theoretical way to determine this reliability factor and the amount of redundancy needed. The only way to be correct in this context is to determine the reliability issue experimentally.

Conclusions

The article describes the backdrop of the microelectronics science and technology that influences the development of molecular electronics. It is pointed out that as the size of electronic devices approaches molecular dimensions, ME promises to have a distinct advantage over other technologies because the component function can be an outcome of the electronic structure of the molecule. The article proposes a strategy that can lead to the development of the ME concept with the scientific and technological knowledge of the present day.

There are a few groups that are currently developing parts of the model. These efforts may lead to demonstrations of the essential elements. On the other hand, we are all conscious that we do not fully understand the behavior of molecules in isolation and even less interactions with other molecules and structure. One can be confident that in addition to the technological promise, ME offers researchers in the field a handsome dividend, which is the inevitable enrichment of our knowledge

about the physics and chemistry of the molecular world, about surface science, and nanolithography.

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