

Molecular-Scale Electronics

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Invited Paper

A review of nonequilibrium electronic transport in molecules and molecular-scale synthetic systems is given. Although the basic concepts and mechanisms for electronic conduction in polymers have been discussed for some time, recently developed experimental techniques provide systems for their validation. New fabrication techniques that create metallic contacts to a small number of conjugated organic molecules allow the study of the basic transport mechanism of these systems and will provide direction for the potential development of molecular-scale electronic systems.

Keywords—Molecular electronics, molecular-scale electronics, resonant tunneling, resonant tunneling diode (RTD), resonant tunneling transistor (RTT), self-assembled monolayers (SAM's), very large scale integration (VLSI).

I. INTRODUCTION

It is well recognized that conventional lithography-based very large scale integration (VLSI) technology is fast approaching the limits of its capabilities. The underlying issues responsible are numerous: ultrathin gate oxides; short channel effects; doping fluctuations; and last but not least, increasingly difficult and expensive lithography [1]. To surmount these problems, nanoscale quantum devices and circuits have been proposed for some time [2], [3]. Over the last two decades, demonstrations of many of these technologies have been accomplished [4]. These include resonant tunneling diode (RTD) and resonant tunneling transistor (RTT) devices and circuits that promise compact multivalued logic and memories, quantum dot and single electron devices, and others. Will these be viable technology alternatives for the post-VLSI era?

Present embodiments of these technologies demand tremendous control over the I(V) characteristics of the device, an undesirable property for downscaling. Dimensional control is a dominant obstacle, since nanodevices must operate by tunneling in some fashion. Since a barrier is needed for isolation in a three-terminal device with gain, tunneling will be exponentially sensitive to

atomic-layer fluctuations in barriers, resulting in device-specific variations that may be unacceptable for large-scale integration. A related problem is that devices using discrete electron charging (single-electron transistors) only work at reduced temperatures. Robust room temperature operation with leakage requirements similar to VLSI requirements requires junctions smaller than 1 nm, which will thus suffer severe tunneling fluctuations. Last, but clearly not least, is that these embodiments do not critically address the major limiting factors of a two-dimensional (2-D) lithography-based technology: accessible parallel fabrication; interconnection density; and alignment. Any successful new technology must: 1) solve the interconnect problem; 2) use self-aligned fabrication; and 3) operate at room temperature and at the atomic level. The last point is because scaling any technology to the 10-nm level may not be cost effective, as the performance increase is marginal compared to the development costs. An atomic- or molecular-scale technology may be the only approach worth the investment.

Recently, molecular electronics-based computation has attracted attention because it addresses the ultimate in a dimensionally scaled system: ultradense and molecular scale [5], [6]. The significant scaling factor gained from molecular-scale devices implies eye-opening comparisons: a contemporary computer utilizes $\sim 10^{10}$ silicon-based devices, whereas one could prepare $\sim 10^{23}$ devices in a single beaker using routine chemical syntheses. An additional driving factor is the potential to utilize thermodynamically driven directed self-assembly of components [7] such as chemically synthesized interconnects, active devices, and circuits. This is a novel technological approach for post-VLSI electronic systems and can conceivably lead to a new era in ultradense electronic systems. This approach for spontaneously assembling atomic scale electronics attacks the interconnection and critical dimension control problems in one step and is implicitly atomic scale. Concurrently, the approach utilizes inherently self-aligned batch processing techniques that address the fabrication limitations of conventional VLSI.

Molecular (i.e., organic) materials for electronic and optoelectronic applications have been realized for quite some

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time. In addition to uses such as liquid crystal displays, devices such light emitting diodes, lasers, transistors, and sensors have been demonstrated [5]. The distinction between these (essentially “bulk”) applications and molecular scale electronics is not just one of size, but of concept: the design of a molecule that itself is the active element.

Molecules were proposed as active electronic devices as early as 1973, when Aviram and Ratner [8] proposed that unimolecular rectification, or asymmetrical electrical conduction, should occur through the molecular orbitals of a single $D-\sigma-A$ molecule by “through-bond tunneling”. Here, D is an electron donor with low ionization potential, A is an electron acceptor with high electron affinity, and σ is a covalent “bridge.” The excited zwitterionic state $D^+-\sigma-A^-$ would be relatively accessible from the ground neutral state $D-\sigma-A$, while the opposite zwitterion $D^--\sigma-A^+$ would lie several eV higher and would be inaccessible. In solid-state language, the system is an asymmetric multilevel resonant tunneling structure. Indeed, molecular systems have good analogies to solid-state systems. Instead of the Fermi levels of the solid state, one deals with the highest occupied and lowest unoccupied molecular orbitals (HOMO and LUMO, respectively) of molecules. Instead of metal and interconnects and degenerate contacts, one uses conjugated linear polymeric systems. Instead of “doping” to modify Fermi levels, one modifies the electron affinity and ionization potential of molecules by the chemical substitution. By designing in the molecular orbitals, one has the equivalent of bandgap engineering [4].

For molecular-scale electronics to come of age, fabrication and measurement techniques had to reach the atomic scale. The advent of atomic imaging techniques, such as the scanning tunneling microscope (STM) and the atomic force microscope (AFM) have given us an atomic view of molecular placement, fabrication, and self assembly. Nanofabrication techniques have created interconnects small enough to reliably contact molecules. A better understanding of electronic transport at the atomic level has developed over the last decade, and theoretical models of conduction through such systems are beginning to develop. All these advances have led to the first electrical measurements of molecular systems. Among these are conductivity measurements of molecules by STM [9], the first measurement of electronic conduction through a single “molecular wire” [10], and the first molecular diodes [11], [12]. This paper reviews some of the initial work in this new, exciting field.

II. INTRODUCTION TO TRANSPORT IN MOLECULES

It is well documented that bulk conjugated organic materials can be semiconducting, or conducting, when doped [13]. However, the measurements of through-molecule long-distance electron transfer are now just emerging. Candidates for molecular wires and switches include polyporphyrins, polyphenylenes, polythiophenes, and other planar organic polymers with extended π -

conjugation, i.e., electron delocalization along the length of the molecule, which can be verified by optical measurements. A review of the synthesis of conjugated oligomers can be found elsewhere [5], [15], as well as a general review of candidate molecular conductors [6], [16]–[18]. Examples of some of these oligomers are shown in Fig. 1, which have a high degree of purity using a divergent-convergent method [19], [20], a vastly more efficient and rapid process that produces monodisperse, stable, and soluble conjugated oligomers.

The study of electronic conduction in molecular systems is just now being developed, with comparison to the first experiments. The theoretical approaches to understand these mechanisms can be found elsewhere [6] but can be summarized into five different regimes.

- 1) *Coherent electron motion*: Nonresonant. This is the most common situation for long-range electron transfer reactions and should hold for the case where the Fermi level of the contacts is in the middle of the HOMO/LUMO gap. In this case, the conductivity should exponentially decrease with length.
- 2) *Coherent electron motion*: Resonant. In this case, conductance is dominated by the contact scattering, is independent of length, and increases with the number of modes. The low temperature conductance of carbon nanotubes appears to be in this regime.
- 3) *Incoherent transfer*: Ohmic behavior. If the wire is allowed to couple to other modes, such as in the environment, then the transport can become familiar with the conductivity dependent on inverse length. For long chains, this may well be more efficient than exponential nonresonant transport.
- 4) *Quasi-particle formation and diffusion*: For degenerate electronic ground states (such as polyacetylene), the carriers can be charged solitons, corresponding to a structural defect with an extra electron sitting on a free radical site (a “polaron”). In nondegenerate conductors (such as poly p -phenylene), two carriers trap near each other associated with a structural defect, giving rise to the so-called “bipolaron.” Both mechanisms are postulated to be active in nonrigid polymers.
- 5) *Gated electron transfer*: Unlike solid-state analogies, molecular structures show different transfer rates dependent on geometrical modification. Transfer processes through molecules capable of undergoing dynamical stereochemical change, similar to ion channels in biological systems, might be a potentially useful molecular electronic gating mechanism.

Over the last two decades, scores of candidate molecules that might selectively enhance one of the above mechanisms have been synthesized, and numerous experiments to attempt to deduce the dominant transport mechanisms have been done. Only recently has advanced nanoscale fabrication and characterization allowed the clear elucidation of these experiments.

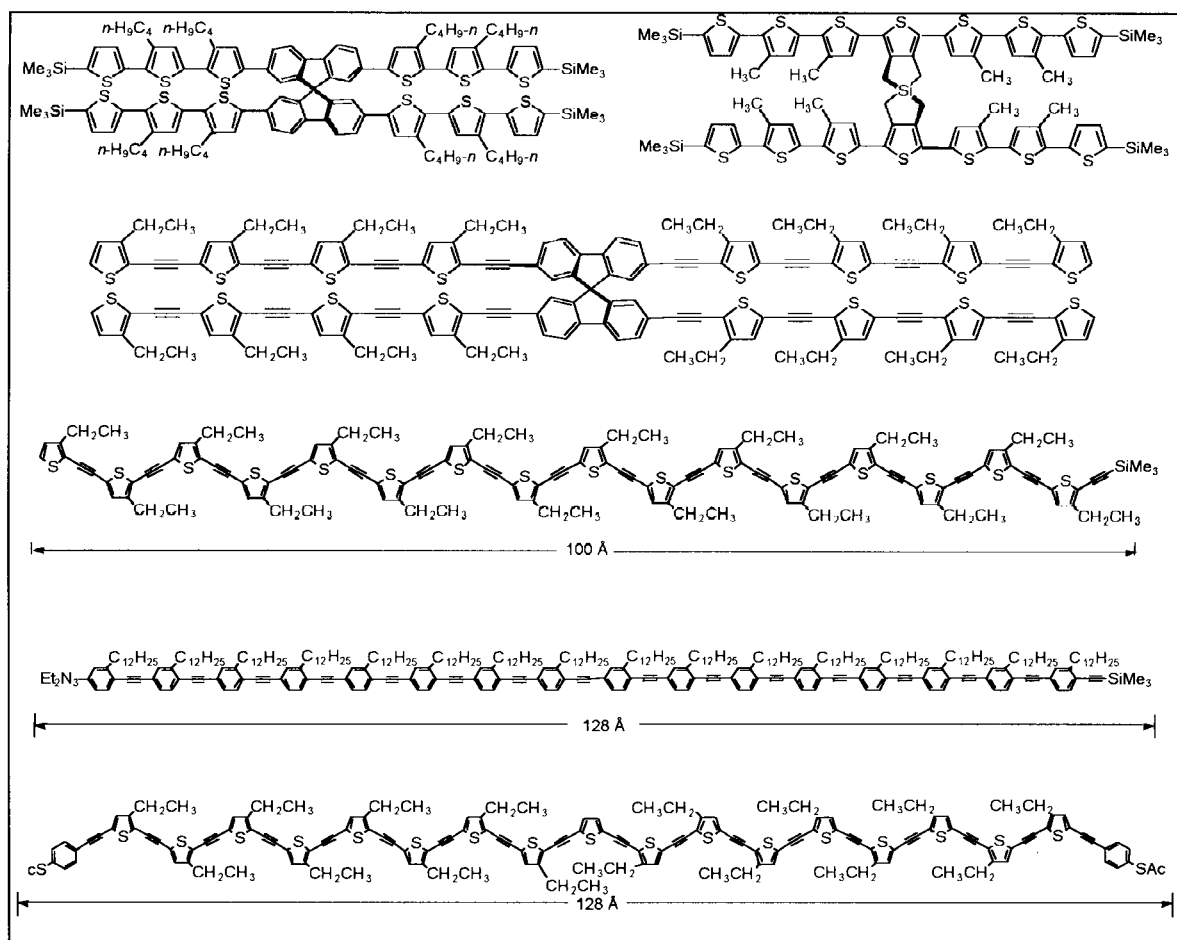


Fig. 1. A variety of molecular conductors. The top three are conductive oligomers connected to an orthogonally fused crossbar switch; the bottom two are long rigid-rod conjugated oligomers. Note the thiol termini on the bottom structure.

A very interesting subfield of molecular electronics has been the use of rigid-carbon structures, i.e., C₆₀ [31] and carbon nanotubes. Although distinctly different from the types of molecules discussed previously, since they do not have the desirable synthetic and functionalization properties that address the fabrication issues discussed previously, they nonetheless have quite interesting properties. Long coherence lengths and discrete electron states have been observed in single-wall nanotubes [39], and C₆₀ [6], [31] has even been used as an electromechanical amplifier. These structures may well become the best molecular-scale wires and interconnects, and development of this field will critically depend on the development of controlled synthesis and manipulation techniques.

III. SELF-ASSEMBLED CONJUGATED MOLECULES

The challenge that has stymied the entire field for more than two decades has been the elementary task of “soldering” one of these electrically active candidate molecules between contacts. With the development of atomic imaging, one can now imagine using STM or AFM tips to maneuver, by brute force, the molecules into the correct position. A much more attractive alternative is the spontaneous self-absorption of the molecular species between the contacts

[21]. Over the last decade, the ability to form very well-defined self-assembled monolayers (SAM's) of oligomers on metal has been demonstrated. The most widely studied system has been the Au-SR system (R=alkyl), which forms very well ordered single monolayers with a very aggressive ~2 eV bond. In fact, the bonding strength is so large that there is indication that the SAM actually “anneals out” defects.

Functional terminal moieties such as thiols now provide the key to self-assembled structures. By synthesizing these onto the oligomer endgroups [22], [23] such as the bottom oligomer example in Fig. 1, we have a self-absorbing and orienting species [the thioacetyl end groups (SAc=SCOCH₃) serve as a protected self-assembled chemisorbed termini with *in situ* generation of the free thiol]. This allows us to investigate conduction through conjugated molecules which are end bound onto surfaces, or placed between proximal probes (e.g., for gold probes one uses thiol-end groups). In the last few years, a variety of metal (or semiconductor) surface/functional end-group combinations have been investigated, giving design flexibility in both synthetic design and multiterminal interconnect orientation [7]. This technique has yielded the first electrical measurements of molecular systems.

IV. CONDUCTIVITY MEASUREMENTS OF SELF-ASSEMBLED MOLECULAR SYSTEMS

The measurement of charge transport in single organic molecules, and the determination of their conductance, has been a long sought goal. Such measurements are experimentally challenging and intriguing since one can test the validity of transport approximations at the molecular level. A conceptually simple configuration would be to connect a single molecule between metallic contacts. Such a metal-molecule-metal configuration would present the molecular embodiment of a system analogous to a quantum dot [24], [25], [26], [27] with the potential barriers of the semiconductor system replaced by the contact barrier of the molecule/metal interface. Although making nanofabricated planar contacts at the molecular scale is challenging, it has been done and candidate molecular conductors have been absorbed into the gaps [28]. An absence of observable conductivity in these systems may be due to inefficient electron transfer, undesirably large contact potentials, conjugation-breaking due to substrate interactions, or unfavorable absorption configurations. Thus, a number of alternative approaches have been attempted and realized.

The first obvious approach is to use STM techniques to locate and locally measure candidate oligomers absorbed on a metallic surface. However, the oligomers will not preferentially be near surface-normal in isolation. By using an alkanethiol (i.e., inert nonconductive) SAM as an inert host matrix, single conjugated molecules have been inserted randomly at defect sites and imaged [9] (Fig. 2). It was observed that the molecules had a significantly higher conductivity than the alkanethiol background; however, these experiments could not give the absolute value of the conductivity due to the involvement of a tunneling gap between the STM tip and the molecule. One variant of this approach was to use Au nanoclusters onto a doubly functionalized oligomer [29], [30] and by STM measurement deduce the molecule conductivity, although the uncertainty in cluster size resulted in large error bars.

The ideal embodiment is to create statically stable contacts, while concurrently restricting the number of active molecules to as few as one. This has been realized [10] (using benzene-1,4-dithiolate connected between stable proximal metallic gold contacts) using a mechanically controllable break junction (MCBJ) [32], [33] (Fig. 3). In this approach, a notched metal wire is glued onto a flexible substrate and is fractured by bending the substrate, after which an adjustable tunneling gap can be established. A large reduction factor between the piezo elongation and the electrode separation ensures an inherently stable contact or tunnel junction. The wire contacts are atomically sharp when broken, as demonstrated in the conductance quantization. The deposition onto the contacts resulted in formation of an SAM on the gold electrodes nearly perpendicular to the surface [34].

Current-voltage $I(V)$ and conductance $G(V)$ measurements showed reproducible characteristic features of stepped $G(V)$ —the first step of $22.4 \pm 0.3 \text{ M}\Omega$. This

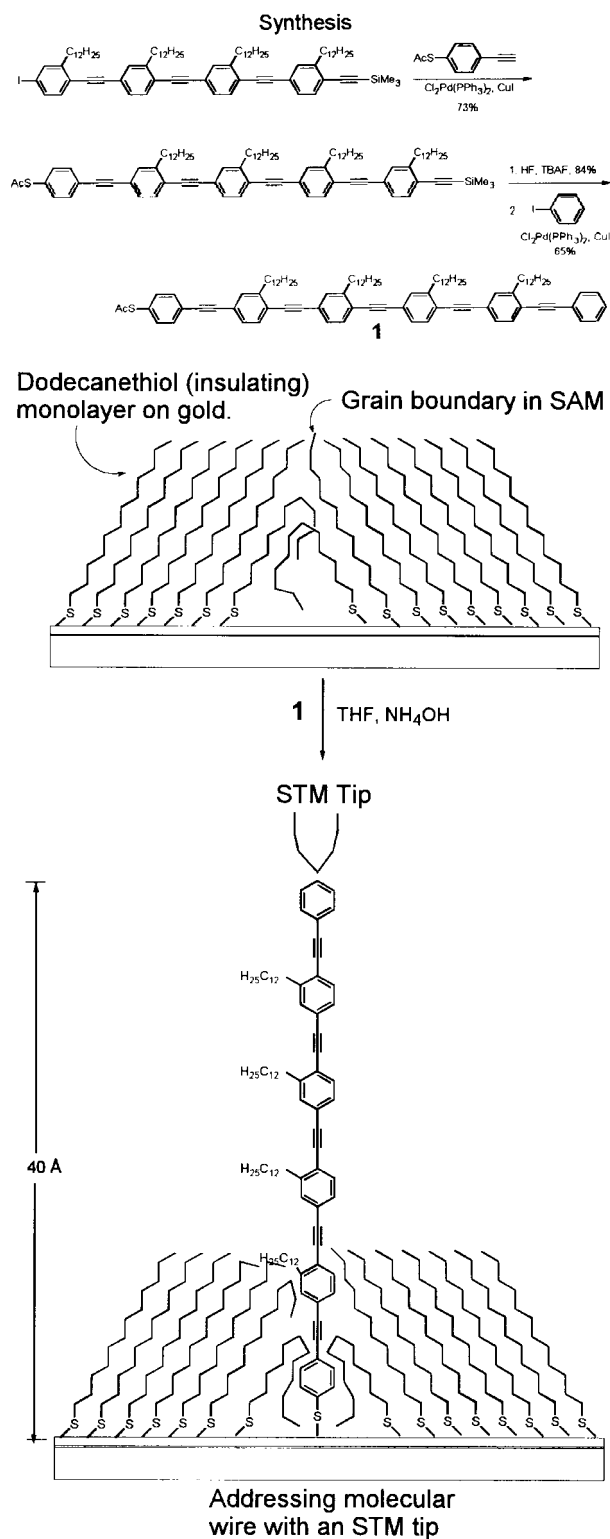


Fig. 2. Schematic representation of the STM experiments to observe enhanced conductivity in π -conjugated molecular wires. The candidate wires are absorbed into the grain boundaries of an alkanethiol host matrix.

is compared to a resistance of $\sim 9 \text{ M}\Omega$ and $18 \pm 12 \text{ M}\Omega$ deduced from the Au nanocluster experiments [29], [30]. An interpretation of the observed $\sim 0.7 \text{ V}$ gap is due to the mismatch between the contact Fermi level and the LUMO. Preliminary calculations using this interpretation

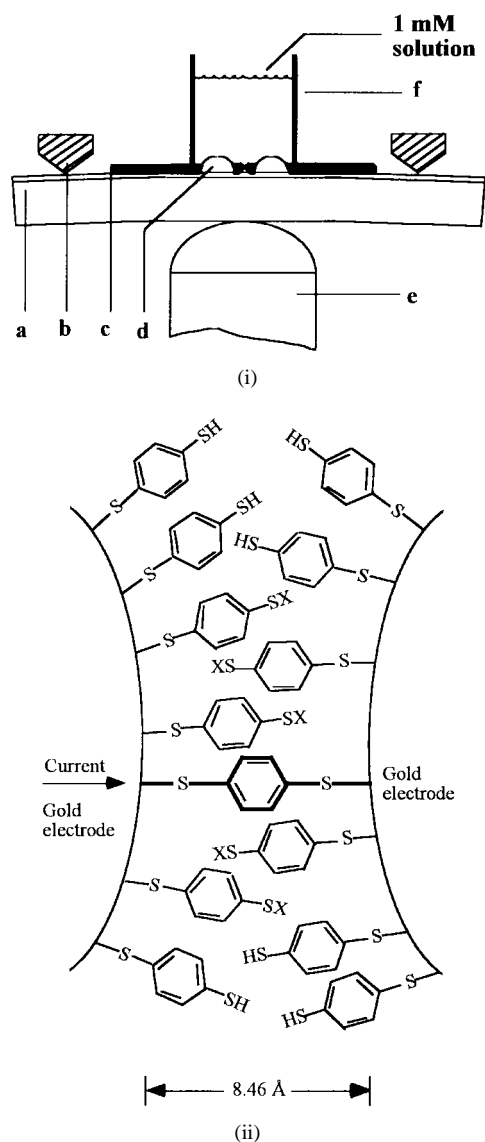


Fig. 3. (i) A schematic of the MCB junction with (a) the bending beam, (b) the counter supports, (c) the notched gold wire, (d) the glue contacts, (e) the piezoelement, and (f) the glass tube containing the solution. (ii) A schematic of a benzene-1,4-dithiolate SAM between proximal gold electrodes formed in a MCB. The thiolate is normally H-terminated after deposition; end groups denoted as X can be either H or Au, with the Au potentially arising from a previous contact/retraction event.

give similar characteristics to the experimentally observed data [35].

Direct contacting to organic thin films has been done. However, multilayers of the molecular wires and micron scale area of the devices containing a large number of molecules have been used, which complicates the analysis of the transport mechanism of single molecules. Experiments with an evaporated-metal-top-contact/molecules/metallic-bottom-contact configuration, which has ten of thousands of parallel active molecules, have been demonstrated [36]. One experiment on an organic system [37] reported evidence for Coulomb charging. Direct contacting to and electrical measurements of a single monolayer of a small number of molecules are needed for accurate spectroscopy.

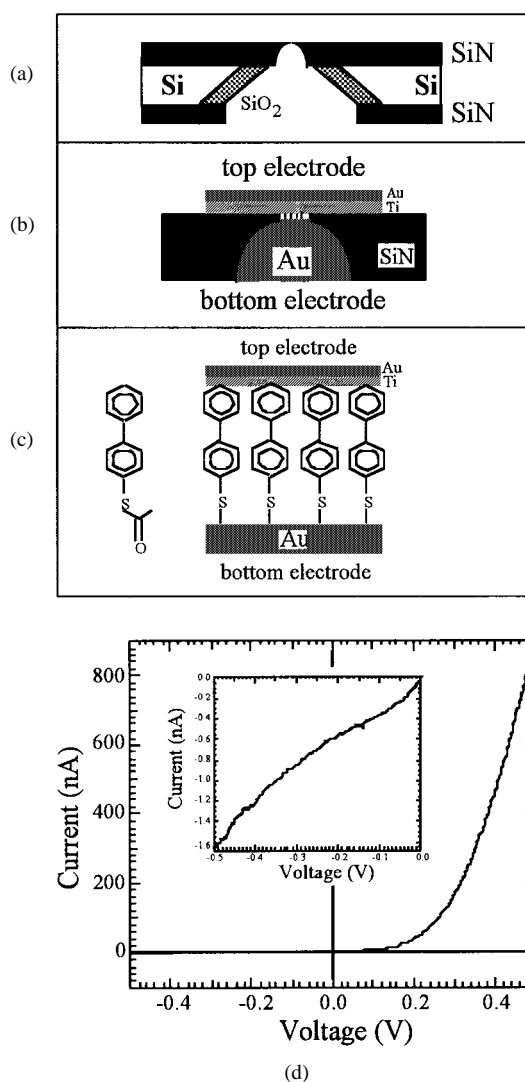


Fig. 4. Fabrication of nanopores. (a) Cross section of a silicon wafer showing the pore etched in a suspended SiN membrane; diameter about 300 Å. (b) Au-Ti top/SAM/Au bottom structure. (c) Specific SAM (4-thioacetyl biphenyl) and structure schematic. (d) Diode-like I-V characteristics at room temperature. Magnified view of the negative bias part in the inset.

A novel fabrication technique has been reported to measure directly the conduction through a small number of organic molecules [12]. These devices, called “nanopores,” consist of a self-assembled monolayer of conjugated molecular wires sandwiched between top and bottom metallic contacts. The employment of nanoscale device size insures that the adsorbed organic layer is highly ordered and defect free. A schematic of the device is shown in Fig. 4. This technique gives a yield of about 80% working and stable devices.

Prominent rectifying behavior is observed: the current at 1-V bias is about 500 times higher than the current at -1 V bias. This is distinctly different from the Aviram-Ratner unimolecular rectifier [8] (rectification due to an asymmetric ground-state/excited state zwitterion) in which experimental results in a hexadecylquinolinium tricyanoquinodimethanide Langmuir-Blodgett film have recently been reported [11]. In the present case, the asymmetry is simply

produced by the different contact barriers, loosely analogous to Schottky barriers. While the I-V curve at negative bias is rather linear,¹ the I-V curve at positive bias displays exponential behavior, with $\ln(I) \propto 1/T$ and the slope is a function of the bias voltage. Using standard thermal emission theory, a value of 0.26 eV for the direct metal-molecule barrier is found. This technique lends itself to rapid synthetic and metallic variation, and is expected to be a workhorse in uncovering a wealth of metal-molecule transport characteristics previously inaccessible by other techniques.

One of the most intriguing aspects of this field is to reproduce the ability to “bandgap engineer” (as is commonly done in the solid state) at the molecular level. Although the field is just at the stage of understanding Ohmic contacts and Schottky barriers, the first attempts at designing active devices have been done. By utilizing strong σ -bonds (such as CH_2 linkages in an oligomer structure), one can effectively insert tunnel barriers, analogous to heterojunction barriers (with the critical distinction that these are now truly orbitals). The transport barriers are CH_2 (methylene, the simplest alkylidene unit) since alkyl units pose a larger electronic transport barrier in π -conjugated moieties. The ethyl groups impart solubility to the larger molecules so that synthetic manipulations and depositions can be freely executed in solution, and the termini have self-assembling moieties (namely protected thiols). The breaking of conjugation by insertion of alkyl moieties has been demonstrated in optical spectroscopy [28], but not direct electronic transport. Fig. 5 shows the I(V) characteristics of a SAM deposited in a nanopore where the active species is designed to be a molecular RTD, with CH_2 groups cladding the central “quantum dot.” Synthesis of the oligomer is reported elsewhere [38]. The device exhibits a pronounced negative differential resistance at room temperature with a reasonably high peak current. The asymmetry is probably due to the asymmetric contact geometry and may well be the first demonstration of an electrically active and useful device utilizing molecular orbital engineering.

V. CONCLUSION

Discussed here are the basic concepts of self-assembled molecular-scale electronic devices. New fabrication techniques to study the transport mechanism of organic molecular wires, such as MCB's and nanoscale fabrication with special metal deposition techniques, are employed to provide metallic contacts to the self-assembled monolayer of a small number of conjugated molecular wires. We report the measurement of the conductivity of a single molecule, and the molecular equivalent of a RTD. These methods can be easily adapted to other molecular wire

¹For negative bias (electrons injected from the Au bottom electrode into the conjugated molecules through the thiolates), plots of $\ln(I/V)$ versus $1/T$ fall on one line indicating hopping, and a similar analysis yields a hopping barrier of 0.19 eV. At present it is unclear whether the hopping is related to defects in the SAM or hopping between neighboring molecular wires.

Current-Voltage Characteristics at Room Temperature

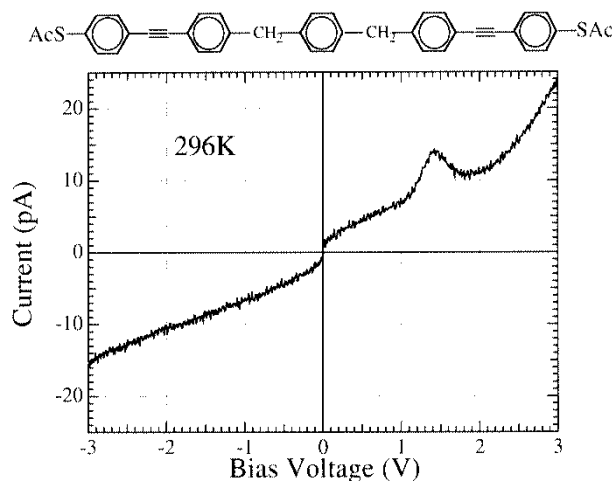


Fig. 5. Room temperature I(V) characteristics of a molecular RTD.

systems for determination of transport mechanism and band alignment. A clear and outstanding challenge is the synthesis and measurement of three-terminal structures, and the determination of gain mechanisms in such structures.

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