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Towards molecular-scale electronics and biomolecular self-assembly

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Abstract

This paper provides an overview of recent research developments in the field of nanoelectronics with organic materials such as carbon nanotubes and DNA-templated nanowires. Carbon nanotubes and gold electrodes are chemically functionalized in order to contact carbon nanotubes by self-assembly. The transport properties of these nanotubes are dominated by charging effects and display clear Coulomb blockade behaviour. A different approach towards nanoscale electronics is based on the molecular recognition properties of biomolecules such as DNA. As an example, DNA is stretched between electrodes using a molecular combing technique. A two-step metallization procedure leads to the formation of highly conductive gold nanowires.

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1. Introduction

The enormous achievements of semiconductor science and technology have driven device miniaturization almost to the level of single atoms and molecules. For many researchers coming from the field of semiconductor nanostructures it therefore seems quite natural to integrate molecular materials into traditionally fabricated devices. However, with these new materials there are new methodologies to be developed and new experimental challenges to be met. Knowledge compiled in the neighbouring disciplines of chemistry, biology and materials science have to be integrated into nanoscale research—which is challenging and fascinating at the same time. On the other hand, many of the concepts

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known from nanoscale semiconductor physics can be easily adapted for molecular systems, in particular in the context of electronics and charge transport. We here give a short survey on recent developments in molecular electronics with carbon nanotubes and biomolecular self-assembly and present results of our own efforts in these fields.

2. Miniaturization of semiconductor devices

Since the invention of the integrated circuit by Noyce and Kilby, the performance of microchips has steadily improved. This development was foreseen by G.E. Moore in 1965, who predicted that the number of transistors on a computer chip would double every couple of years, known as “Moore’s law” [1]. Currently, state-of-the-art computer chips, such as Intel’s Pentium 4 processor, contain 55 billion transistors which corresponds to feature sizes of 0.13 μm . Gate oxide thicknesses in CMOS devices have shrunk to about 1.5 nm. Although the International Technology Roadmap for Semiconductors [2] projects that Moore’s law could hold during the next 10 to 15 years, the devices will become more difficult and costly to fabricate. The obstacles to scaling down field-effect transistors (FET) are numerous, such as damages induced by high electric fields and heat dissipation, non-uniformity of doping, quantum mechanical tunnelling of electrons from source to drain and leaking of electrons through the gate oxide [3]. Moreover, economic considerations will play an important role in the development of future microchips, taking into account that the fabrication plant costs double for each new generation. No one expects conventional silicon-based microelectronics to continue following Moore’s Law forever. If miniaturization of electronic circuits is continued down to the nanometer scale, alternative concepts will be needed. One idea which could pave the way towards nanometer sized devices is the use of organic molecules. These are highly organized structures with well-defined electronic states which can be produced at low cost in high quantities. Another advantage of using molecules as basic components for electronic devices is that they can be designed to self-assemble on specific electrodes, leading to organic–inorganic hybrid structures.

Thus the miniaturization trend in semiconductor industry naturally leads research into the realm of organic chemistry, and even biology. However, in nanoscience, molecular systems traditionally studied in these fields are seen from a different perspective and quite different goals are envisioned. Nonetheless, before organic molecules may be used in electronic circuits or to build other artificial molecular structures, it is very likely that applications in other, more traditional fields become feasible, e.g. in medicine or in sensor devices.

3. Molecular electronics with carbon nanotubes

3.1. *On route to carbon nanotubes based electronics*

One of the most promising organic materials for molecular electronics are undoubtedly carbon nanotubes (CNT). Since their discovery in 1991 by Iijima [4], scientists have revealed a wide range of unique properties that make them excellent candidates for

nanoelectronic and other applications. CNTs can be metals or semiconductors, depending on their chirality. Because of their strong chemical bonds and satisfied valencies, the materials boast high thermal, mechanical, and chemical stability. In addition, CNTs can be efficient conductors as a result of their tiny diameters, long lengths, and defect-free structures that make them ideal one-dimensional systems [5].

It was first shown by C. Dekker's group [6] that it is possible to build field-effect transistors using semiconducting CNTs. On the other hand, metallic CNTs are interesting candidates to replace interconnects and vias (vertical interconnects) in microchips due to the extremely high current densities that they can carry. These high current densities are possible because electronic transport in CNTs can be ballistic [7]. One of the issues in this context is the contact resistance between the CNT and the metallic leads. In the ideal case, transparent electrical contacts made to a ballistic conductor exhibit two units of quantum conductance $4e^2/h$ [8]. However, in most cases contact resistances in the range of some hundred $k\Omega$ to $M\Omega$ are observed. This is most likely caused by Schottky barriers at the CNT-metal interface [9]. In most transport studies, contacts are produced by electron beam or optical lithography and evaporation of metal on top of the CNTs. Alternatively, CNTs can be deposited on the electrodes which in most cases yields higher contact resistances. Chemical vapour deposition (CVD) offers the possibility to directly grow in situ contacted CNTs on electrodes [10]. Contact resistances can be improved by annealing or by electroless metal deposition [11]. Recently, researchers from H. Dai's group [12] found a way to greatly suppress the Schottky barrier at the nanotube-metal contacts by using palladium as the contact material. Palladium has a high work function and good wetting interactions with the nanotubes. This resulted in current densities of the order of 10^9 A cm^{-2} and ratios of 10^6 between the on and the off state of CNT-transistors.

There remains the problem of the placement of CNTs. Some placement can be achieved by lithographically defining areas where a catalyst is deposited, followed by CVD growth of nanotubes [13]. Other techniques aiming at aligning or trapping of CNTs from solution use an ac bias between two electrodes [14–16]. This way, even CNT crossbar circuits could be produced [17]. Another technique allowing the fabrication of nanowire crossbars that employs microfluidics was reported by C. Lieber's group [18]. Probably, the simplest and cheapest way of placing nanowires would be by self-assembly. This could be achieved by an appropriate functionalization of the CNTs and the surface. First steps in this direction were carried out by Rao et al. [19] who patterned distinct regions on gold surfaces by coating with different organic molecules, either with polar or with non-polar chemical groups. Since the nanotubes are attracted towards the polar regions, self-assembly of CNTs on predefined structures was accomplished.

3.2. Contacting carbon nanotubes by self-assembly

We use a similar procedure to produce contacted CNTs by self-assembly on metallic electrodes. The single-wall CNTs used in this study were produced by laser-ablation, the multi-wall CNTs by arc-evaporation. In a first step, contact structures are defined by optical or electron beam lithography on a highly doped Si wafer covered with 150 nm of SiO_2 which can be used as a back-gate. This is followed by an evaporation of 4 nm of NiCr and 70 nm of Au. After lift-off, positively charged gold contacts are formed by immersing

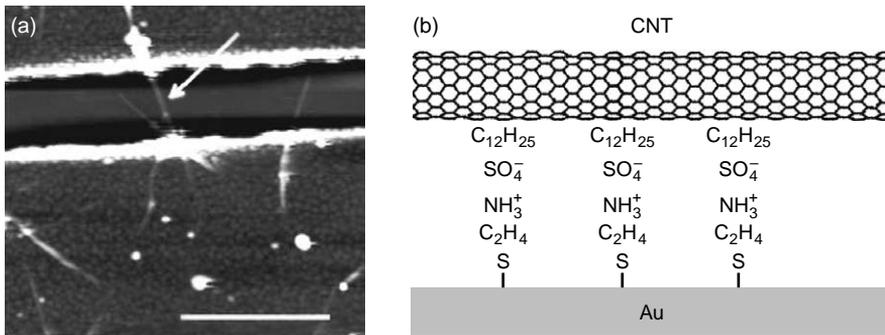


Fig. 1. (a) AFM image of two Au electrodes covered with multi-wall CNTs. The scale bar corresponds to 1 μm . (b) Schematic representation of a CNT attached to a Au contact via cysteamine and SDS.

the chip in a 1 mM aqueous solution of cysteamine for 45 min. Cysteamine is a polar molecule with a thiol endgroup which forms a self-assembled monolayer on gold surfaces. Meanwhile, CNT in suspension with the surfactant sodium dodecyl sulphate (SDS) are purified by centrifugation and treated in a sonicator in order to separate the CNTs [20]. This produces single negatively charged CNTs in H_2O . When this suspension is dropped on a functionalized chip, CNTs are attracted towards the positively charged electrodes. The remaining suspension is blown away with N_2 after 15 min, followed by cleaning the chip in pure H_2O . Fig. 1 shows an AFM image of multi-wall CNTs on two Au electrodes with a separation of approximately 0.4 μm . One of the CNTs is contacted by both electrodes (white arrow in Fig. 1), two of the other CNTs are only in contact with the lower electrode. Similar experiments were carried out using single-wall CNTs. In both cases, about 80% of the electrode pairs are connected by at least one CNT.

Conductance measurements on 39 multi-wall and on 19 single-wall CNT samples were carried out at 4 K. About half of the samples show Coulomb blockade oscillations as a function of the voltage V_G applied to the back-gate. Fig. 2 shows the conductance of a single multi-wall CNT as a function of V_G and source–drain voltage V_{SD} . White areas correspond to low conductance, black regions correspond to high conductance. Coulomb blockade diamonds can clearly be seen. The resistance of the measured samples range between tens of $\text{k}\Omega$ and a few $\text{M}\Omega$.

Coulomb blockade is most likely caused by tunnelling barriers between the CNT and the electrodes. This would mean that the CNT acts as a single quantum dot weakly coupled to source and drain electrodes. The tunnelling barriers probably consist of cysteamine and SDS which were previously used as coatings of the CNTs and the electrodes. In most applications, such tunnelling barriers are highly undesirable. The relatively high contact resistances could be reduced by using shorter and more conducting organic molecules for the coatings [19]. Electroless metal deposition [11] or chemical removal of the coatings could also improve the coupling between the CNTs and the electrodes. Connecting CNTs with electronic components or interconnecting CNTs could lead to the development of nanoscale networks. First steps in this direction have already been undertaken, e.g. by coupling chemically functionalized CNTs with molecular linkers [21].

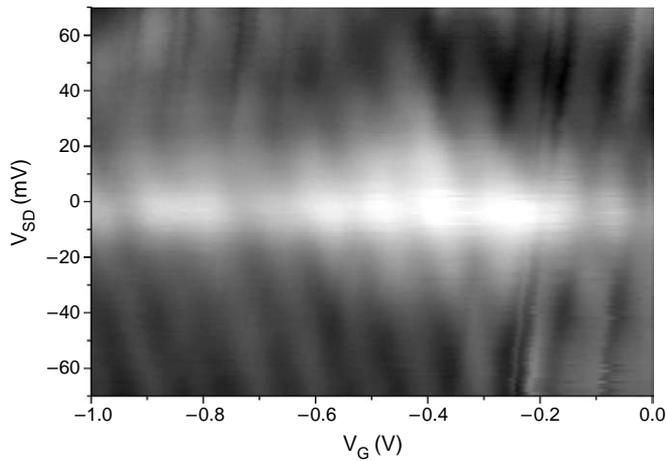


Fig. 2. Grey-scale plot of the conductance of a single multi-wall CNT as a function of back-gate voltage V_G and source–drain voltage V_{SD} at 4 K. White areas correspond to low conductance, black areas correspond to high conductance.

4. At the interface with biology

4.1. Biology as a guide towards nanotechnology

Many researchers in the field of nanoscience have come to realize that most of the goals of nanotechnology have already been achieved by biological systems [22]. In biology, the organization of matter on a molecular scale is generated by an intricate molecular machinery connected in complicated regulatory networks reminiscent of engineered control circuits. Quite naturally, the nanoscientist dreams of similar machinery to “self-assemble” artificial structures with a possibly non-biological function. However, the sheer complexity of biological systems makes this a formidable task. Most of the current research on bio-inspired nanosystems is concerned with rather simple processes and is far less impressive than the elegant—albeit complicated—molecular engineering solutions found in nature.

One of the simplest aspects of biological systems which might be exploited in nanoscience in the near future is the molecular recognition property of many biomolecules. Here, the most prominent example, of course, is the formation of a duplex structure by two complementary strands of deoxyribonucleic acid (DNA). DNA is a polymer of nucleotides, building blocks consisting of one of the four DNA bases adenine (A), guanine (G), cytosine (C), or thymine (T), a ribose sugar unit and a phosphate. The nucleotides are linked together via phosphodiester bonds. The bases adenine and thymine can associate with each other via two hydrogen bonds, whereas guanine binds cytosine via three of these bonds (this is called Watson–Crick pairing; in fact, there are many more possible pairings between the bases, but only these two are relevant in the context of a standard double helix [23]). Two single strands of DNA can form a duplex if the sequence of the bases on one strand is exactly complementary to the sequence of the other strand, i.e. when each A finds a T and each

G finds a C on the other strand, where the strands have to be aligned in an antiparallel fashion. In its native form (“B DNA”), a DNA duplex has a double-helical structure with a helix pitch of 3.4 nm (10 base pairs (bp)/turn) and a diameter of approximately 2 nm. If the sequences of two strands are sufficiently non-complementary (i.e. if there are many “defects”), no duplex will be formed. Thus, the binding properties of two single-strands are determined by their base sequences. This unique property already inspired a number of nanoscientists to make use of DNA for “programmable self-assembly” of nanostructures. DNA could already be used to build supramolecular structures like cubes or other polyhedra [24, 25] as well as two-dimensional molecular sheets [26, 27]. Another recent development is the construction of nanomechanical devices made from and powered by DNA [28–35].

4.2. DNA-based nanocircuits

In the context of nanoelectronics, DNA might be used to assemble the basic layout of an electronic circuit (often referred to as a molecular “scaffold”) which can be subsequently modified by template-directed deposition of materials or the specific binding of DNA-labelled nanoparticles or molecules. For DNA-based assembly of electronic circuits, several issues have to be addressed: (i) The chemical modification of DNA to enhance its electronic properties; (ii) the connection of DNA-based structures to the outside world; (iii) the construction of supramolecular templates for electronic circuits.

The first point is of crucial importance as DNA itself does not seem to be a good conductor, at least for electronics applications [36, 37]. Although this issue is still somewhat controversial, it is clear that even the best reported values for the conductivity of pure DNA would not be sufficient for any real application. Therefore, in recent years many groups have sought for a way to enhance DNA’s electronic properties. One of the most promising approaches seems to be DNA-templated synthesis of metallic nanowires [39–42] (see also the next section). Of course, an electronic component is comprised of more than metal wires, so other materials—like semiconductors, conducting polymers or smaller molecules—are needed for DNA modification. Also, it has to be elucidated how to deposit different materials on DNA in an orderly fashion. One possible route towards a sequence dependent materials deposition technique is the use of DNA binding proteins like RecA as a “deposition mask” [43].

Apart from this issue, the controlled orientation and placement of biomolecules or biomolecular nanostructures within lithographically defined microstructures is an experimentally very challenging task by itself. One possible approach is to use electrostatic manipulation of charged biomolecules in combination with their molecular recognition properties. For example, one can define specific binding sites for DNA strands by covalently linking short DNA strands to metal electrodes (Fig. 3(a)). These “sticky labels” can be used to address the electrodes with different DNA sequences which can be recognized by DNA strands containing the complementary sequences (Fig. 3(b)). The distribution of the labels as well as the guidance of complementary strands towards their binding sites can be assisted by electric fields. The feasibility of such an approach has already been demonstrated for biochips designed for DNA hybridization studies [44].

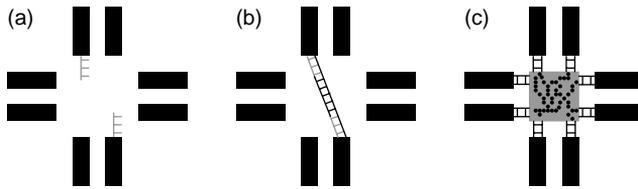


Fig. 3. Schematic representation of a DNA-based assembly strategy for nanoelectronic circuits. (a) Metal electrodes defined by conventional lithography can be “labelled” with DNA tags which can be recognized by DNA molecules containing their complementary sequences. (b) This may be used to interconnect specific electrodes utilizing DNA’s molecular recognition property. (c) To actually make use of the smaller size of molecular-scale components, a more sophisticated approach has to be taken: we envision the assembly of whole circuits composed of nanoscale components (grey box) with defined input/output ports which are connected to the bulky pre-defined metal contacts. In such an approach, the size of the circuit is not limited by the lithographic dimensions of the electrodes.

However, with such a technique one is still limited by the lithographic resolution of the metal electrodes to which the DNA labels can bind. In order to make full use of the reduced size of molecular-scale electronic components it is not sufficient just to make electric contact to an individual of such a component. Rather, it will be necessary to construct complete circuits with internal wiring with defined functions and defined input/output lines to which bulky metal electrodes can make contact (Fig. 3(c)). To this end, supramolecular templates for molecular-scale circuits have to be built by self-assembly. Here, the concepts developed in the field of algorithmic self-assembly [26] might prove particularly useful. A first example of an artificial supramolecular DNA structure connected to metal electrodes has been demonstrated just recently [45]. In the next section, we concentrate on the first of the problems stated above: the modification of DNA to enhance its electronic properties.

4.3. Experiment: DNA-templated nanowires

For the fabrication of DNA-based Au nanowires stretched between lithographically defined electrodes, a mixed strategy of nanocluster growth, molecular combing and electroless plating was adopted. As a template λ -DNA was used.¹ λ -DNA is the 48 502 base pair (bp) long genome of bacteriophage λ with a native length of about 16.5 μm . In a first step (adapted from [41]), DNA was modified with platinum nanoclusters to serve as seeds for a subsequent development reaction: 20 μl of a solution of λ -DNA in TE buffer (10 mM Tris (Tris(hydroxymethyl)-aminomethane), pH 8.0, 1 mM EDTA (ethylene diamine tetraacetic acid)) at a concentration of 50 $\mu\text{g}/\text{ml}$ was added to 100 μl of a solution of K_2PtCl_4 (1 mM) in deionized water and incubated for up to one week at room temperature. During this incubation time, platinum clusters grow on the DNA duplex. After incubation, further deposition is achieved by the addition of 16 μl of a 10 mM solution of the reducing agent dimethylaminoborane in H_2O . This results in the

¹ λ -DNA has been obtained from New England Biolabs, Inc. All other chemicals have been obtained from Sigma-Aldrich Europe and used without further purification.

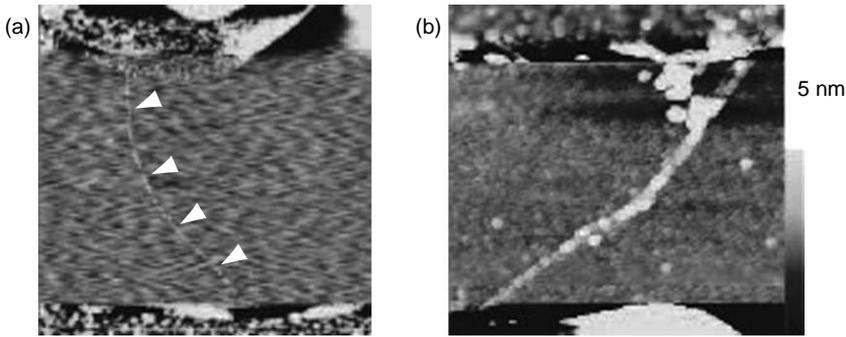


Fig. 4. Topographic AFM images of DNA stretched between two metal electrodes: (a) without and (b) with modification with Pt nanoparticles. The spacing between the electrodes is 2 μm . In (a) the position of the DNA is indicated by white arrows.

formation of closely spaced platinum particles along the DNA with sizes between 3 and 5 nm. In the next step, the platinum modified λ -DNA (Pt-DNA) is stretched between two lithographically defined gold electrodes on a silicon substrate. The stretching of the DNA is accomplished using a modified molecular combing technique [46, 47]. To this end, the silicon substrate is silanized with trimethyl chlorosilane to render the surface hydrophobic. Then a small droplet ($\approx 1 \mu\text{l}$) containing Pt-DNA is applied to the surface and removed using filter paper. The receding meniscus of the droplet stretches DNA molecules between the (hydrophilic) gold electrodes. Fig. 4(a) is an AFM micrograph of a single molecule of λ -DNA, Fig. 4(b) shows platinized DNA stretched between electrodes. After stretching, the sample is washed several times with deionized water and blown dry in a gentle flow of nitrogen. At this stage, the modified DNA wire is not yet conductive as the Pt clusters are not spaced closely enough to permit an electric current to flow. To render the Pt-DNA structure conductive, an electroless plating procedure is applied (closely following [43]). The sample is incubated in a solution of KAuCl_4 , KSCN and hydroquinone which results in the deposition of gold on the platinum nanoparticle seeds. Depending on the reaction time (a few minutes), a continuous gold wire along the DNA with a diameter of 20–50 nm is formed. The result of an I – V measurement on several metallized DNA molecules in parallel is shown in Fig. 5. As expected, the metal wires display Ohmic behaviour. Depending on the number of DNA molecules connecting the two electrodes, we obtained resistances between $\approx 100 \Omega$ (for many molecules) and $\approx 400 \text{ k}\Omega$ (for a few molecules). From these values, the resistivity of individual metal coated DNA wires can be estimated to be on the order of 2×10^{-3} – $5 \times 10^{-2} \Omega \text{ cm}$ which is three to four orders of magnitude higher than the resistivity of bulk gold ($\rho = 2 \times 10^{-6} \Omega \text{ cm}$). The discrepancy may be due to bad electrical contact to the electrodes or due to additional resistances introduced by gold grain boundaries within the wire. In the inset of Fig. 5 we also present a current–voltage measurement on unmodified DNA. The measured current of $I = 1.36 \pm 0.02 \text{ pA}$ is the input offset current of the current amplifier used. From the differential conductance one can deduce a lower bound for the resistance of λ -DNA in this setup of 10 T Ω which is consistent with the findings of other groups [37–39].

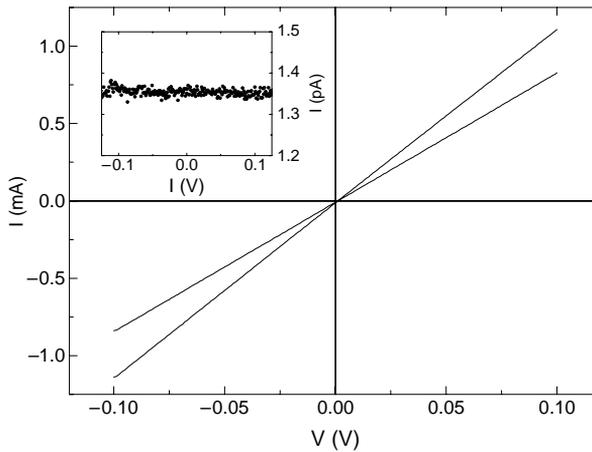


Fig. 5. I - V -curves obtained from gold coated DNA molecules. The Ohmic I - V -characteristics correspond to resistances of 90 and 120 Ω , respectively. In the inset, the result of a typical current-voltage measurement for unmodified DNA is displayed. From this measurement, a lower bound of 10 T Ω can be deduced for the resistance of pure λ -DNA.

4.4. Further steps. Other applications

As already mentioned above, a metallic nanowire can only serve as a nanoscale interconnect for electronic components, but does not have any active role itself. For this reason, we currently follow several routes to functionalize DNA-templated networks with conductive polymers or semiconductors. As an example, an efficient procedure to synthesize continuous semiconductor nanowires along DNA in an aqueous environment has been developed in our group recently [48]. With these new materials, a variety of electronic components can be fabricated, assisted by the self-assembling properties of biomolecules. We are also actively working on the manipulation and orientation of biomolecular structures in micro- and nanofabricated environments—in combination with DNA-based materials synthesis we hope to soon be able to build the first nanoscale electronic circuits following the general strategy outlined above. Apart from nanoelectronics, there are many other possible applications of biomolecules linked to microstructures with electronic functionality. The techniques developed for biotemplated nanocircuits should also prove useful for the fabrication of novel biosensors and bioelectronic devices. Furthermore, controlled on-chip manipulation of biomolecules can be expected to result in improved and simplified miniaturized separation systems. In this way, both bio- and nanoscience will profit from their convergence.

5. Summary and conclusion

We have shown that by the use of conventional lithographic techniques combined with the self-assembly properties of organic or biological molecules, hybrid electronic structures composed of both traditional materials as well as molecular components can

be fabricated. Carbon nanotubes can be guided to bind to electrodes using a negatively charged surfactant (SDS) and a positively charged electrode modification (cysteamine). Their electronic transport characteristics display Coulomb blockade behaviour analogous to that observed for semiconductor-based mesoscopic systems. In a different approach towards nanoscale electronics, single DNA molecules are stretched between metal electrodes using a molecular combing technique. Our I – V measurements on pure DNA support the view that DNA itself is not conductive. However, DNA molecules can serve as a template for the synthesis of materials such as metal wires. Nanoscale gold wires obtained in this way are conductive and display perfect Ohmic behaviour. These results indicate that future nanoscale electronic circuits might be built on the basis of molecular self-assembly rather than lithographic techniques. At the same time, they show that many of the concepts developed in semiconductor science are still applicable in the realm of molecular-scale electronic systems.

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