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Importance of Nanosensors: Feynman's Vision and the Birth of Nanotechnology

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ABSTRACT

In his visionary 1959 lecture at Caltech, Feynman envisaged the potential of the ability to manipulate matter at the atomic scale. I discuss implementations of Feynman's vision in the field of nanosensors and perspectives of its further development and applications.

INTRODUCTION

As the theme of this presentation, I have chosen the words by Richard Feynman: "I will not now discuss how we are going to do it, but only what is possible in principle – in other words, what is possible according to the laws of physics" [1]. Just after Christmas 1959, he delivered a now-famous talk – titled "There's Plenty of Room at the Bottom" – at the California Institute of Technology. It is possible, he proposed, for scientists to assemble new materials at the level of single atoms and molecules, where there are "new kinds of forces and new kinds of possibilities, new kinds of effects". It is generally accepted that Feynman's visionary discussion of the problems and promise of miniaturization constituted the starting point for the new field that today is called *nanotechnology*.

Feynman concluded his talk, i.a., with a challenge to build a working electric motor no larger than 0.04 cm on a side, expecting that making such a device would take an entirely new approach to engineering. Amazingly, Feynman's micromotor challenge was quickly met by a CIT graduate, William McLellan, who designed a motor only 0.038 cm in diameter, using conventional tools. Feynman's disappointment was that while the motor met the conditions it did not advance the art. What could Feynman say about the implementation of his predictions at present?

NANOTECHNOLOGY: VISIONARY'S DREAMS COME TRUE

First of all, I give some examples, how nanotechnology has made it possible to realize some of Feynman's dreams. One of the ambitions of nanotechnology, building motors on a molecular scale, has been realized in 2003. The key element of the motor is a multiwall nanotube (Figure 1). The inner tube is used as an axle, and the outer tube as the outside bearing. The axle element is about 20-40 nanometers in diameter [3].

In his lecture, Feynman suggested that it would be possible to use atomic-size elements to store bits of data: "Suppose, to be conservative, that a bit of information is going to require a little cube of atoms 5 times 5 times 5 – that is 125 atoms… And it turns out that all of the information that man has carefully accumulated in all the books in the world can be written in this form in a cube of material one two-hundredth of an inch wide – which is the barest piece of dust that can be made out by the human eye" [1]. The ultimate storage medium would store a bit in a single atom, with a few atomic spacings between bits in order to prevent coupling between them. Today, the highest commercial storage density – more than 30 Gigabits per square cm – is achieved with magnetic hard disks. As described in [4,5], a much higher density is achieved for a two-dimensional version of Feynman's atomic memory, formed on the surface of silicon by a small amount of gold (see Figure 2). It looks similar to the CD-ROM, but the scale is nanometers instead of micrometers. Therefore, the storage density is much higher. "The gold triggers the formation of self-assembled tracks, which are exactly five atoms wide. Extra silicon atoms sit on top of the tracks in well-defined positions. It is suggestive to assign an extra silicon atom to a "1" and a vacancy to a "0". The minimum empty area required around each bit is 5×4=20 atoms, 4 atoms along the track and 5 atoms from one track to the next. Feynman's 1959 suggestion of spacing the bits 5 atoms apart was right on the mark." [5] Reading the memory consists of a line scan with a scanning tunneling microscope along the self-assembled tracks. By investigating a storage device at the single atom limit one can learn something about how contemporary data storage might evolve in the future. The key property of a memory is: readout speed versus storage density. Compared to traditional data storage in hard disks, the silicon atom memory has a very high density (40 Terabits per square cm), but its readout data rate is extremely low. "Even the theoretical limit of the readout data

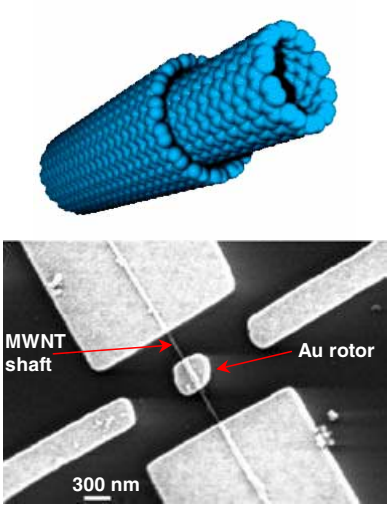

Figure 1. Upper panel: A computer-rendered model of a rotational nanotube bearing (From [2]. Courtesy Zettl Research Group, University of California and Lawrence Berkeley National Laboratory). Lower panel: Scanning electron microscopy (SEM) image of the nanomotor. A 300 nm Au plate rotor is attached to a multi-walled carbon nanotube (MWNT) which acts as a support shaft and is the source of rotational freedom. Electrical contact to the rotor plate is made via the MWNT and its anchor pads. Three stator electrodes, two on the SiO_2 surface and one buried beneath the surface, provide the control elements. (Adapted by permission from Macmillan Publishers Ltd: Nature [3], © 2003)

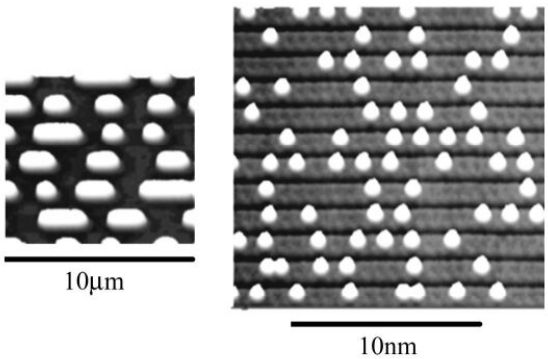

Figure 2. Comparison of the atomic memory on silicon (rhs) with a CD-ROM (lhs). (Reprinted by permission from IOP Publishing: Nanotechnology [4], © 2002. Courtesy of F. J. Himpsel.)

rate with the best possible readout electronics… is still lower than the readout data rate achieved with hard disks today… The data rate will have to be recovered by a high degree of parallelism, including e.g. many reading heads and multiple disks." [5] It is interesting to compare data storage in a silicon atom memory to data storage in nature. DNA needs 32 atoms to store one bit, which is comparable to using 20 atoms around each bit at the silicon surface. The readout data rates are similar as well [4].

NANOSENSORS: A PROMISING REALITY

In this Section, I turn to our central theme – nanosensors.

"When we get to the very, very small world – say circuits of seven atoms – we have a lot of new things that would happen that represent completely new opportunities for design. Atoms on a small scale behave like *nothing* on a large scale, for they satisfy the laws of quantum mechanics" [1].

Nanoparticles are unique tools as sensors. First, they are larger than typical molecules yet smaller than viruses. They are similar in size to many proteins. This is part of the reason they can operate well inside cells.

Second, nanosensors possess unique physical characteristics. They deliver sensitivity orders of magnitude better than conventional devices and provide such performance advantages as fast response and portability. For example, nanoshells [6] and nanorice [7] are made of a non-conducting core that is covered by a metallic shell. Nanoshells are about 10,000 times more effective at Surface-Enhanced Raman Scattering (SERS) than traditional systems. Nanoshells provide an opportunity to design all-optical nanoscale sensors – essentially new molecular-level diagnostic instruments – that could detect as little as a few molecules of a target substance [8].

Third, nanoparticles reveal unique physical properties, which do not exist in bulk materials. For example, the optical response of Au colloidal nanoparticles (from 5 to 20 nm size) is characterized by a localized surface plasmon resonance around 520 nm, which is absent from the spectrum of bulk metals (see Figure 3).

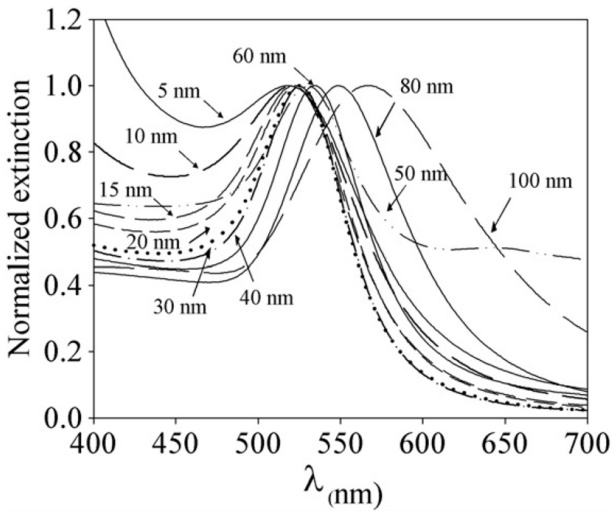

Figure 3. Extinction coefficient versus wavelength of gold colloidal nanoparticles in aqueous medium. The different spectra correspond to different sizes of the gold nanoparticles. (Reprinted by permission from IOP Publishing: Nanotechnology [9], © 2005. Courtesy of C. Rinaldi.)

Fourth, nanosensors allow for building integrated devices, providing an elemental base for intelligent sensors. Intelligent sensors are characterized as having significant data storing-, processing-, and analyzing power. Intelligent sensors can be utilized as autonomous systems or they can be spread out in large numbers to form networks. The concept of intelligent sensors, which is being presently developed, is an implementation of another of Feynman's visions. Among

the most central and fundamental problems of biology in 1959, Feynman mentioned the question: "What is the sequence of bases in the DNA"? It took more than 40 years to find an answer to this question on the basis of many technological innovations: a draft human genome sequence was presented [10] as recently as in 2001. This sequence is an instance of "the biological example of writing information on a small scale" which inspired Feynman "to think of something that should be possible for technical applications. Biology is not simply writing information; it is *doing something* about it" [1].

"What would the properties of materials be if we could really arrange the atoms the way we want them? … I can't see exactly what would happen, but I can hardly doubt that when we have some *control* of the arrangement of things on a small scale we will get an enormously greater range of possible properties that substances can have, and of different things that we can do" [1]. Quantum confinement effects in semiconductor nanostructures have made possible many approaches to design novel sensing materials. An attractive feature of nanostructured materials is the ability to tailor optical and electrical properties by changing simple parameters like layer thickness, materials composition, etc. For example, infrared photodetector applications have been developed on the basis of semiconductor Quantum Dots [11]. An InGaAs/InGaP Quantum-Dot Infrared Photodetector (QDIP) is characterized by a high sensitivity in the mid-infrared/ infrared range. Peak responsivity as high as 3.1 A/W and record high detectivity (3.623×10^{10} cm $Hz^{1/2}$/W) were achieved for this QDIP [11].

One of the exciting prospects of nanotechnology is the use of Quantum Dots in biology. "The unique optical properties of Quantum Dots make them appealing as *in vivo* and *in vitro* fluorophores in a variety of biological investigations. The ability to make Quantum Dots water soluble and target them to specific biomolecules has led to promising applications in cellular labeling, deep-tissue imaging, assay labeling and as efficient fluorescence resonance energy transfer donors." [12] The scheme in Figure 4 compares the emission range for selected Quantum Dot core materials with representative spectral ranges of biological interest [12].

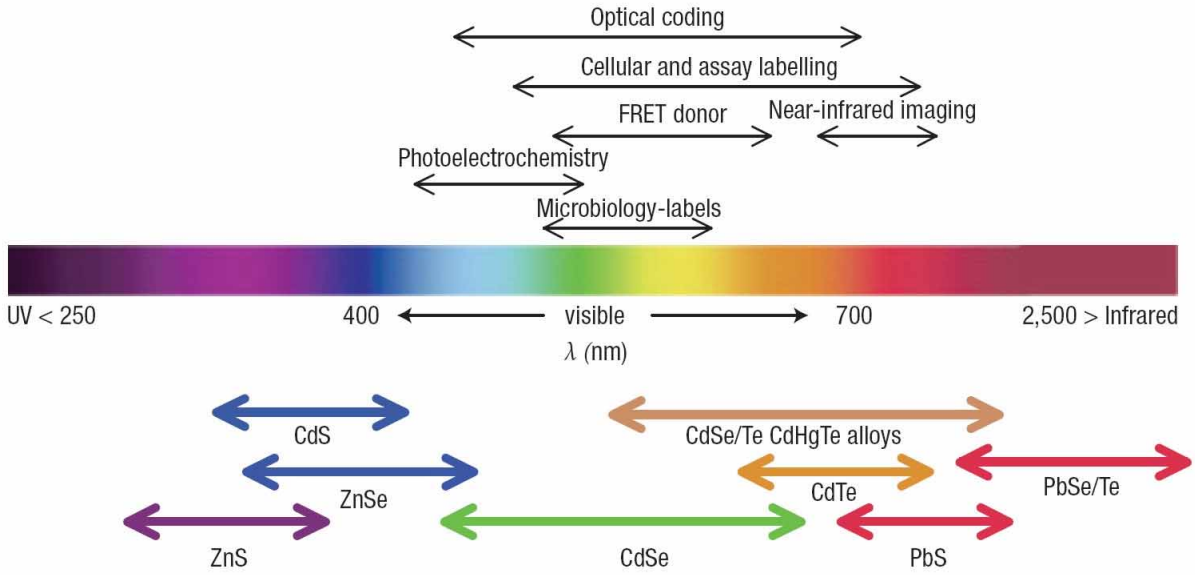

Figure 4. Selected Quantum Dot core materials scaled as a function of their emission wavelength superimposed over the spectrum. Representative areas of biological interest are also presented. (Reprinted by permission from Macmillan Publishers Ltd: Nature Materials [12], © 2005)

Optical properties of interest to biologists include high quantum yield, broad absorption with narrow photoluminescence spectra (full-width at half-maximum ~25–40 nm), spanning the UV to near-infrared. As shown in [12], in comparison with molecular dyes, two properties of Quantum Dots stand out: (i) the ability to size-tune fluorescent emission as a function of core size and (ii) the broad excitation spectra, which allow for excitation of mixed Quantum Dot populations at a wavelength far removed (>100 nm) from their emissions. Optical properties of semiconductor Quantum Dots constitute one of the research topics of our laboratory TFVS at the Universiteit Anwerpen. Numerous experiments on photoluminescence and Raman scattering reveal a surprisingly high efficiency of the exciton-phonon interaction in Quantum Dots. For some cases, attempts to interpret these experiments on the basis of the widely used adiabatic theory meet with difficulties. In this context, using the Feynman ordered operator calculus, we have developed a theory of phonon-assisted optical transitions in semiconductor Quantum Dots, which takes into account the non-adiabaticity of the exciton-phonon system. The proposed non-adiabatic treatment of phonon-assisted optical transitions is shown to provide an explanation for the remarkably high intensities of the phonon satellites observed in the optical spectra of various Quantum-Dot structures [13,14].

Detection of minute concentrations of nucleic acid sequences is important for medical diagnosis and for the understanding of biomolecular mechanisms. The use of Quantum Dots as DNA nanosensors promises to significantly enhance the sensitivity of fluorescence-based DNA detection. Following [15], the DNA nanosensor, schematically shown in Figure 5, includes a Quantum Dot (CdSe–ZnS core–shell nanocrystal) conjugated with several streptavidins and two target-specific probes: a reporter probe labeled with a fluorophore (Cy5 - Cyanine 5) and a capture probe labeled with biotin. In the absence of the DNA target sequence, the Quantum Dot and the reporter probe are unlinked and only the Quantum Dot fluorescence is detectable. “When a target DNA is present in solution, it is sandwiched by the two probes. Several sandwiched hybrids are then captured by the Quantum Dot through biotin–streptavidin binding… The resulting assembly brings the fluorophore and the Quantum Dot into close proximity. This leads to fluorescence emission from the acceptors by means of FRET [Fluorescence Resonance Energy Transfer] on illumination of the donor. As a result, detection of the acceptor emission indicates the presence of targets.” [15]

Single nanometer-sized pores (nanopores) embedded in an insulating membrane are a new class of nanosensors for rapid electrical detection and characterization of biomolecules. Recently, a new technique was reported [16] for fabricating silicon oxide nanopores with single-nanometer precision. In a translocation experiment, particles suspended in an electrolyte solution are electrophoretically driven through a nanometer-sized aperture, located between two reservoirs kept at a potential difference. “The presence of a molecule inside the pore lowers the amount of conducting solution inside the pore. Passing molecules can thus

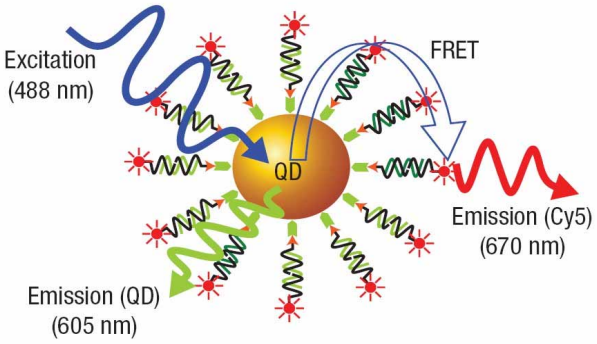

Figure 5. Fluorescence emission from Cy5 due to FRET between Cy5 acceptors and a Quantum Dot donor in a nanosensor assembly. (Reprinted by permission from Macmillan Publishers Ltd: Nature Materials [15], © 2005)

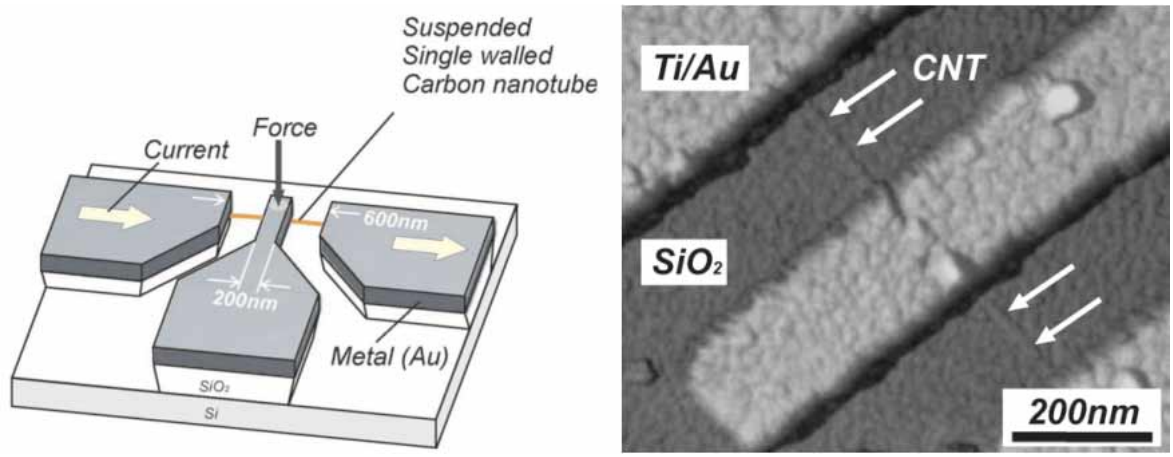

Figure 6. Lhs: Schematic illustration of the fabricated CNT-based nanoelectromechanical force sensor. Rhs: AFM image of a contacted SWNT (diameter approximately 1.2 nm). (Reprinted by permission from IEEE: IEEE Sensors Journal [18], © 2006 IEEE)

be detected as dips in the ionic current through the pore." [17] It is then possible to detect single-molecule DNA translocation events through a nanopore. A detailed analysis of the precise shape of the events allows one, for instance, to characterize the lengths of DNA fragments present in a mixture [17]. The goal is to reveal the DNA sequencing (cp. [1]) by analyzing the DNA translocation events through a nanopore.

Following [18], as quasi-one-dimensional nanostructures, Single-Walled Carbon NanoTubes (SWNTs) show a strong interdependence between structural symmetry and physical properties, in particular, electronic properties. It has been shown by several experiments that the mechanical deformation of a suspended SWNT leads to a noticeable change of conductance. The force sensor (Figure 6) contains "a contacted, suspended SWNT attached underneath a freestanding cantilever. An external out-of-plane force acting on the cantilever deflects the cantilever, resulting in a mechanical deformation of the underlying SWNT." [18] The structural change of the SWNT yields a measurable change of the conductance. In test measurements of the force sensor, an AFM tip in contact force mode is used to apply a force on the cantilever. The cantilever is deflected, which leads to a significant change in conductance (a factor of 3 for a force of approximately 120 nN). The key result of Ref. [18] is that the resistance change, due to the force induced deflection and subsequent deformation of the SWNT, is reversible. This makes the proposed system suitable for force sensing at nanoscale.

Nanostructures are used within the EU Integration Project SENSATION, which is aimed at exploring a wide range of micro- and nanosensor technologies "for unobtrusive, cost-effective, real-time monitoring, detection and prediction of the human physiological state in relation to wakefulness, fatigue and stress." [19] The electroencephalography electrodes based on Carbon NanoTubes (CNT) are developed within SENSATION for the transduction of biopotentials to an electronic signal. Traditionally, AgCl-coated electrodes are used for this transduction. Within SENSATION, CNT arrays, homogeneously decorated by Ag particles, have been produced [19]. The obtained coated CNTs can be used to transduce ionic flows in the human body.

In a recent paper on the prospects of nanotechnology in cardiology it is stated: "In vivo sensors [which] could constantly monitor O_2 blood concentrations and cardiac function to detect problems during sleep. In addition, heart-specific antibodies tagged with nanoparticles may

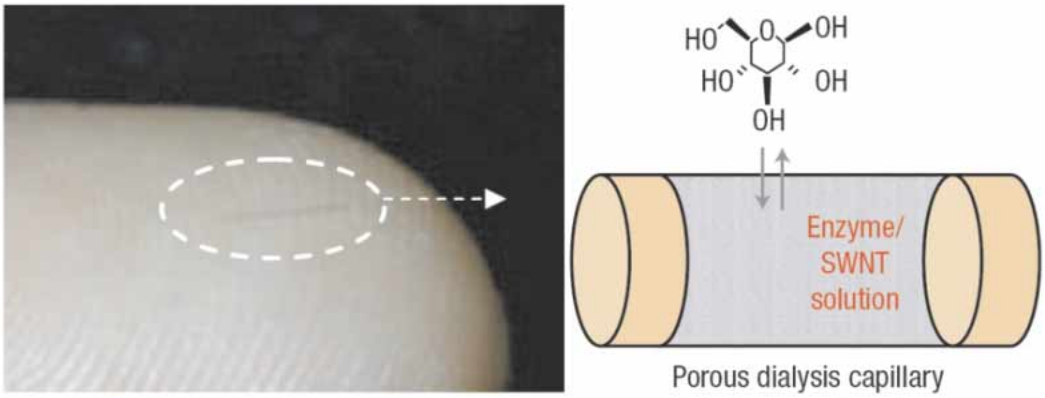

Figure 7. A permeable capillary, shown to size on a human finger, is loaded with the nanotube solution allowing glucose to diffuse through the membrane with the containment of the sensing medium. On placing the capillary beneath a human epidermal tissue sample, one can map the nanotube fluorescence from the capillary. (Adapted by permission from Macmillan Publishers Ltd: Nature Materials [21], © 2005)

allow doctors to visualize heart movement while a patient experiences sleep apnea to determine both short- and long-term effects of apnea on cardiac function" [20]. Carbon nanotubes can be used for optical nanosensors, because they typically fluoresce in the NIR spectral region, where human tissue and biological fluids are characteristically transparent. An example is the application of Single-Walled Carbon Nanotubes (SWNTs) to monitor blood glucose [21]. "The tubes are first suspended in water and then coated with a monolayer of glucose oxidase, an enzyme that prevents the tubes from cohering to one another and also forms a selective site on which glucose can bind and generate hydrogen peroxide. The tubes are then exposed to ferricyanide, an ion sensitive to hydrogen peroxide, which passes through the porous monolayer and attaches to the tube surface. In the presence of glucose, the glucose oxidase produces hydrogen peroxide that quickly reacts with the ferricyanide. The resulting changes in the nanotubes' electron density and optical properties affect their fluorescent qualities – the more glucose, the greater the fluorescence. To demonstrate the practicality of using the treated nanotubes as biomedical indicators, the investigators secured some of them in a capillary permeable to glucose and inserted it into human tissue. The sensor's fluorescent behavior corresponded to the local glucose concentration" [21] (Figure 7). As shown in [22], single-walled carbon nanotubes wrapped with DNA can be placed inside living cells in order to detect trace amounts of harmful contaminants at the subcellular level. When the DNA is exposed to ions of certain atoms (e.g., calcium, mercury and sodium) the DNA changes shape, perturbing the electronic structure of SWNT and shifting the nanotube's fluorescence to lower energy. "The change in emission energy indicates how many ions bind to the DNA. Removing the ions will return the emission energy to its initial value and flip the DNA back to the starting form, making the process reversible and re-usable." [23]

Following [24], "SWNTs can be viewed as a graphene layer that is rolled up into a tubular structure. However, the graphene layer can be rolled up at different vectors around the circumference of the nanotube designated by the indices (n,m). This has important implications for the electronic structure of the SWNTs. Differences in the electronic states result in some SWNTs being semiconductors with different band gaps." Fluorescence spectra of a SWNT strongly depend on its (n,m) type. "The high fidelity of the crystalline structure of each SWNT also results in narrow band emission, allowing fluorescence to determine analytically the

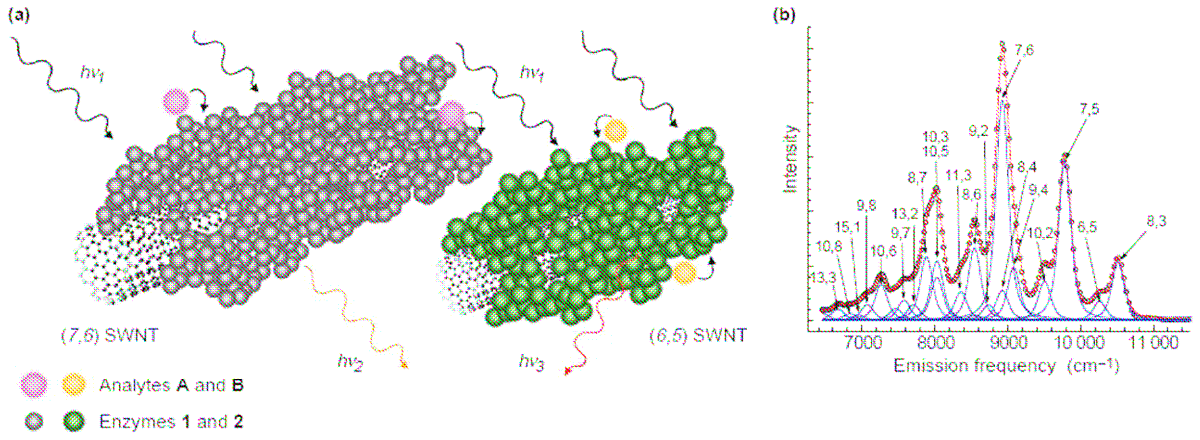

Figure 8. (a) Nanotubes of a given (*n*,*m*) type are surrounded by a specific enzyme designed to interact with a specific analyte. These enzymes modulate the transfer of electrons in and out of the nanotube, thereby affecting the fluorescence intensity. Each (*n*,*m*) type will give a characteristic NIR fluorescence with narrow band emission that can be collected with a multichannel detector. (b) Deconvolution of the emission spectrum by (*n*,*m*) type will then enable simultaneous measurement of the concentration of each analyte. (Reprinted from [24], © 2005, with permission from Elsevier)

presence of each (*n*,*m*) type." [24] The ultimate goal is "to develop a multi-analyte sensor that can simultaneously monitor each target analyte" [24] (see Figure 8). It has been demonstrated "that the emission from SWNTs red shifts when surrounded by enzymes by as much as 20 nm. Therefore, if different enzymes result in distinguishable shifts in fluorescence", it may "be possible to monitor each analyte by monitoring the intensity of the shifted fluorescence spectra. However, to achieve higher fidelity signals from a particular analyte, the SWNTs should be separated by their… (*n*,*m*) type and then selectively fictionalized with the appropriate enzyme." Although a pure sample of a particular (*n*,*m*) type has not yet been obtained, "several groups have demonstrated some promising initial results. Once semiconducting SWNTs of a specific (*n*,*m*) type can be synthesized or separated, multi-analyte biosensors can be developed." [24]

"At the atomic level, we have new kinds of forces and new kinds of possibilities, new kinds of effects. The problems of manufacture and reproduction of materials will be quite different" [1]. The "top-down approach to nanotechnology, whereby nanostructures are created, manipulated, and modified by machine, is sometimes incapable of offering the complexity and economy of scale that Molecular Self-Assembly (MSA) demonstrates in nature… Solution processing and manufacturing of MSAs offer" the prospect "of mass production with the possibility of error correction at any stage of assembly" [25] (see Figure 9). "It is well recognized that this method could prove to be

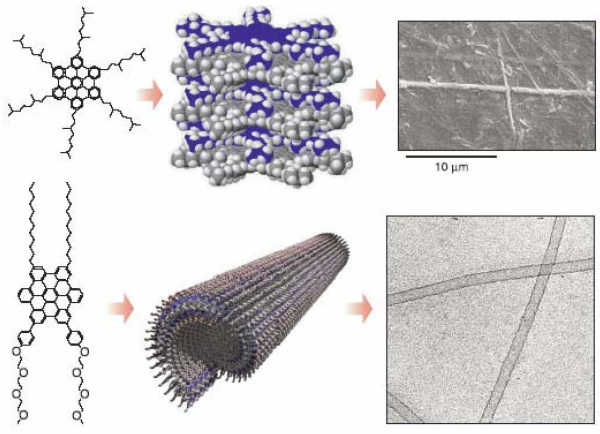

Figure 9. Examples of Molecular Self-Assemblies formed by hexabenzocoronenes: (left) molecular structure; (center) MSA simulation; (right) scanning and transmission electron microscope images of the respective MSAs. (From [25]. Reprinted with permission from AAAS, © 2004)

the most cost-effective way for the semiconductor electronics industry to produce functional nanodevices such as nanowires, nanotransistors, and nanosensors in large numbers" [25] and at a high rate.

UNPRECEDENTED PERSPECTIVES

In this Section I touch a topic, which resonates with Feynman's 1959 talk: the perspectives of nanosensors. "The problems of chemistry and biology can be greatly helped if our ability to see what we are doing, and to do things on an atomic level, is ultimately developed – a development which I think cannot be avoided" [1].

Sensors with sizes at the nanoscale possess unprecedented physical properties. Nanosensors are expected to be closer to the theoretical limit of sensitivity (for their size), than macrosensors are (for their size). For example, "both blood pressure and pulse rate can be reliably monitored by a medical nanodevice virtually anywhere in the vascular system, using a $(68\ \text{nm})^3$ pressure sensor with ~0.001 atm sensitivity" [26]. The estimated power budget for the data transfer in typical in vivo medical nanodevices is as follows. "The maximum [of the data transfer rate] $I_{max} \sim 10^9$ bits/sec bandwidth requirement must draw ≥3 picowatts (pW), well within the anticipated 1-1000 pW power budget of typical in vivo medical nanodevices. Slower bit rates can draw even less power. The design challenge is to closely approach this minimum theoretical limit" [26].

The underlying physics at the nanoscale differs from the physics at the microscale. "The atomistic structure of matter appears clearly and quantum mechanical effects… dominate the physical behavior that determines the functionality of nanosystems" [18].

"I do know that computing machines are very large; they fill rooms. Why can't we make them very small, make them of little wires, little elements – and by little, I mean *little*. For instance, the wires should be 10 or 100 atoms in diameter, and the circuits should be a few thousand angstroms across" [1]. The progress in this direction has been tremendous: the development of semiconductor microchips has allowed for minituarization of computers. The present efforts are related to the building up of a new elemental base at the nanoscale (semiconductor Quantum Dots and Quantum Rings, superconductor nanostructures) and to the implementation of the novel principles of quantum computing.

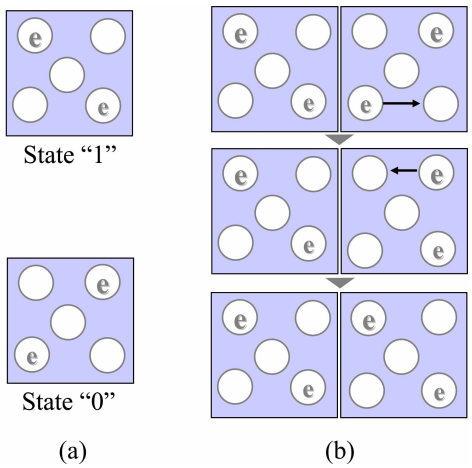

Figure 10. Quantum-Dot cell [27]. (a) Two states of equal energy in a Quantum-Dot cell. (b) Transfer of a "1" to the next cell. (See details in the text.)

As an example of communication with nanodevices, I refer to a proposal of a wireless two-state Quantum Dot device called a "cell" [27,28]. A cell consists of 5 Quantum Dots and two electrons. Electrons can tunnel from one Quantum Dot to the next. The Coulomb repulsion between the two electrons causes them to move to opposite corners of the cell. This leads to two states of equal energy in the cell (see Figure 10). By placing two cells adjoining to each other and compelling the first cell into a certain state, the second cell will assume the same state, because this lowers its energy. The

result is that a "1" has moved on to the next cell. By stringing cells together, a "pseudo-wire" at the nanoscale can be produced in order to transport a signal [27,28].

In future applications of nanomedicine [26], "billions of [nanorobots] would have to be stationed in tissues, bones, and blood throughout the body, where they would monitor various physiological parameters such as blood pressure and periodically communicate their findings. The actual communication network would also be made up of the tiny robots." [29]

A recent publication in Nature projects a picture of computing in 2020: "Computers could go from being back-office number-crunchers to field operatives. Twenty-four hours a day, year-in, year-out, they could measure every conceivable variable of… a human body, at whatever scale might be appropriate… These new computers would take the form of networks of sensors with data-processing and transmission facilities built in" [30]. This futuristic picture represents one of the ways in which Feynman's vision may come true: "A biological system can be exceedingly small. Many of the cells are very tiny, but they are very active; they manufacture various substances; they walk around; they wiggle; and they do all kinds of marvelous things – all on a very small scale. Also, they store information. Consider the possibility that we too can make a thing very small which does what we want – that we can manufacture an object that maneuvers at that level!" [1]

CONCLUSION

Feynman's 1959 lecture was visionary. He considered the potential ability to manipulate matter at the atomic scale. Nanotechnology has made it possible to realize some of Feynman's dreams.

Feynman's vision and his predictions have been implemented in the field of nanosensors. State-of-the-art nanosensors are based on various advanced materials, e. g. Quantum-Dot Infrared Photodetectors, Quantum-Dot based DNA nanosensors, nanopores – nanosensors for biomolecules, carbon nanotube based optical and electromechanical force sensors, transducers of biopotentials, multi-analyte biosensors… Applications of nanosensors open great perspectives, e.g. for monitoring the human body, due to their unprecedented sensitivity.

"This field is not quite the same as the others in that it will not tell us much of fundamental physics (in the sense of, "What are the strange particles?") but it is more like solid-state physics in the sense that it might tell us much of great interest about the strange phenomena that occur in complex situations. Furthermore, a point that is most important is that it would have an enormous number of technical applications." [1]

ACKNOWLEDGMENTS

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