

Nanoscale Science and Technology: Building a Big Future from Small Things

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Abstract

The following article is an edited transcript based on the MRS Medalist presentation given by Charles M. Lieber of Harvard University on December 4, 2002, at the Materials Research Society Fall Meeting in Boston. Lieber received the Medal "for controlled synthesis of nanowire and nanotube materials." This presentation begins with an introduction to the bottom-up paradigm of nanoscience and nanotechnology. The key concepts of this paradigm are explored through studies outlining progress toward meeting the challenge of nanocomputing through the assembly of functional nanowire elements. The richness of the bottom-up paradigm and nanowire building blocks is then illustrated with the development of chemical and biological nanosensors. Finally, the uniqueness of nanowires is exemplified through discussion of the assembly of nanophotonic devices, including the demonstration of multicolor and addressable nanoscale light-emitting diodes, nanowire injection lasers, and assembled arrays of these nanophotonic sources. Challenges and goals for realizing nanotechnologies in the future are discussed in the conclusion.

Keywords: assembly, nanoelectronics, nanotechnology, nanowires, photonics, sensors.

Introduction

"Nano" is a nearly ubiquitous prefix used today in science and technology; moreover, it is becoming widely recognized by the general public. But what is the science that will build nanotechnologies and that may make nanotechnology a unique field of endeavor that revolutionizes many areas in the future? I focus in this presentation on what I see as unique to this field of science and engineering and where nanoscale science and technology may lead us in the future.

Central to my vision for nanotechnology is the idea that by developing and following a common intellectual path—the bottom-up paradigm of nanoscale science and technology (Figure 1)^{1–10}—it will be possible in the future to build (or more correctly, assemble) virtually any kind of device

or functional system, ranging from ultrasensitive medical sensors to nanocomputers. Underpinning this bottom-up paradigm is the controlled growth of nanoscale materials—the building blocks of the bottom-up approach—pursued within the disciplines of materials science and chemistry. Specifically, our goal is to control with atomic precision the morphology (e.g., nanoscale clusters versus wires), structure, composition, and size of nanoscale materials, since these will define and enable our control over the physical properties of the resulting materials.

Next, understanding the physics of new nanoscale materials emerging from synthetic efforts is an important and fundamental part of the bottom-up paradigm, since such studies define properties that

may ultimately be exploited for nanotechnologies. Moreover, an intimate integration and interplay between materials growth and fundamental characterization enable us not only to expand our basic understanding, but also to make rational predictions and possibly define new device concepts unique to the nanoscale building blocks.

Finally, and to fully exploit the bottom-up paradigm, we must develop rational methods of organizing building blocks and device elements on multiple length scales, that is, hierarchical assembly, which takes us to chemistry and biology. We need to assemble building blocks not only in close-packed arrays, which give us one very fixed kind of interconnectivity, but we also need to be able to control the architecture, the spacing on multiple length scales. Through this hierarchical assembly, we are going to be able to take advantage of the interesting properties and create novel or functional systems. That has to be done within the context of architectural design.

As described, the bottom-up paradigm for nanotechnology is perhaps daunting in its highly interdisciplinary nature, although to me this makes it an exceedingly rich and exciting area to pursue. Moreover, I believe that this paradigm outlines a clear and universal pathway—from the controlled growth of nanoscale building blocks to understanding their properties and how to organize these building blocks in arbitrarily complex ways—for changing virtually any technology.

Growth of Nanowires

First, I will illustrate how we exploit the bottom-up paradigm to attack the problem of nanoscale computing, and later, in less detail, I will demonstrate how this common pathway can lead to advances in two other exciting areas—chemical/biological detection and photonics. The assembly of a universal nanoscale com-

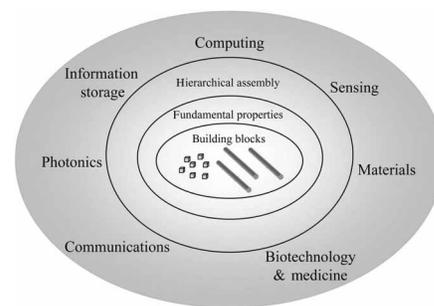


Figure 1. Overview of the bottom-up paradigm for nanotechnology.

putting machine encompasses all of the key elements described for implementing the bottom-up paradigm. We will first start at the very bottom or foundation—the building blocks.

For building blocks, I have focused primarily on nanoscale wires (nanowires) and, particularly, those made out of semiconductor materials.^{1,2} This emphasis on nanowires is driven by the realization that nanowires can function both as active devices and interconnects and thus have the potential to provide simultaneously two of the most critical functions in any integrated nanosystem.²

In general, the preparation of nanoscale wires requires that the addition of material during the growth process is constrained to occur along only one direction. This is conveniently accomplished by taking advantage of ideas from vapor-liquid-solid growth with a new twist: we produce a nanometer-scale catalyst that can form a liquid solution with the nanowire material we want to grow; the catalyst particle size controls the size of the initial nucleation event and the nanowire diameter. Specifically, when the liquid solution—a nanoscale droplet—becomes supersaturated, a nucleation event occurs, and then by adding reactant selectively to this nanoscale droplet, we grow a solid, crystalline nanowire (Figure 2).^{1,2}

An important feature of this approach to nanowire growth is that one can predict *a priori* essentially all of the conditions needed to make different materials; that is, phase diagrams can be used to choose a catalyst, composition (catalyst/nanowire material), and temperature for successful growth. An example of this powerful concept is our early studies of silicon (and germanium) nanowire growth using iron as a catalyst.¹ Examination of the silicon-rich end of the binary Fe-Si phase diagram shows a coexistence of FeSi liquid and Si solid phases typical of many binary and pseudobinary phase diagrams. By our approach, one needs only to produce a nanoscale catalyst cluster, or FeSi nanodroplet, and then continue to add silicon reactant in order to grow silicon nanowires. This concept was first implemented in work using laser ablation as a method to create both nanoclusters and additional silicon reactant, where transmission electron microscopy (TEM) analysis demonstrated that the resulting nanowire materials consisted of single-crystal silicon cores encased in an amorphous silicon oxide sheath. The sheath was formed by oxidation of the silicon core.

Using this general idea, we have shown that it is possible to grow in a controlled fashion essentially all of the periodic table's

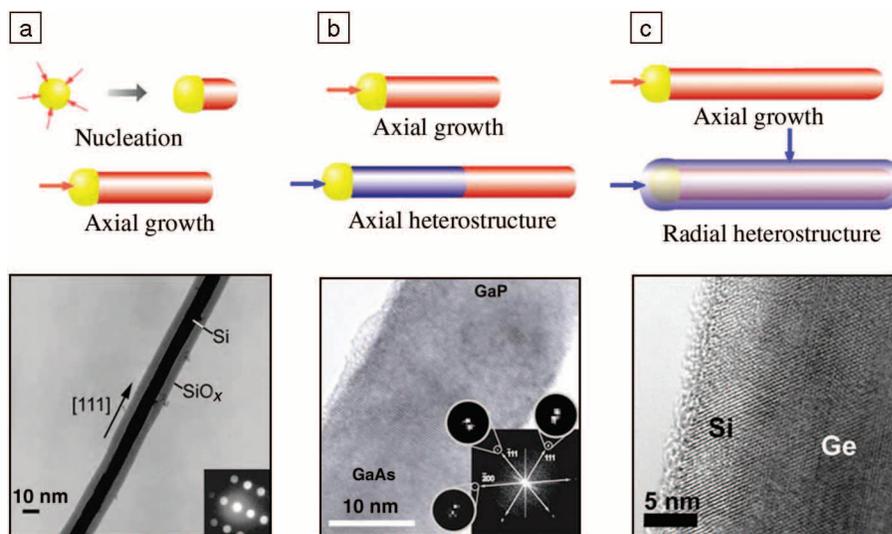


Figure 2. Growth and representative structures of (a) uniform single-crystal semiconductor nanowires, (b) axial nanowire heterostructures, and (c) radial nanowire heterostructures.

main group elemental, binary, and related alloy semiconductors as nanowires.^{2,3} This general approach also enables control of nanowire diameter and length. For example, small, medium, and large catalyst clusters should enable the growth of small, medium, and large nanowires, respectively. This simple concept was first demonstrated in the growth of gallium phosphide nanowires, where we further showed that the distribution of nanowire diameters was the same as that of the starting nanocluster catalyst particles. These results and subsequent studies of silicon and indium phosphide nanowires demonstrate the generality of our concept and the ability to push the diameters of these nanowires down to true molecular dimensions.

The control of composition and size for a wide range of nanowires provides a diverse set of building blocks from which to assemble nanodevices, yet I believe strongly that further control of the nanowire growth in order to modulate the structure on a length scale larger than atomic dimensions will open the potential for much richer function. It is within this context that my group has also focused on the growth of axial and radial nanowire heterostructures and superlattices (Figure 2).^{4,5}

In the case of axial heterostructures, we can conceptually carry out growth in the following way: initially (see Figure 2b), a reactant A is used to prepare a length of the first material (red), and then a reactant B is exchanged in the reactor for a fixed time to produce a length of the second material (blue).⁴ The exchange of reactants

can be repeated to produce superlattice structures in which composition and doping are modulated and for which there are one or more red-blue nanoscale intrawire junctions. As an example, we have demonstrated this new concept with the growth and characterization of single GaP-GaAs heterojunctions as well as superlattices of up to 21 controlled length periods (Figure 2b).

For radial heterostructures, we simply need to switch the growth mode to one dominated by homogeneous deposition on the existing nanowire core, for example, by changing growth temperature or reactants.⁵ By sequentially modulating the reactants to form shells around a nanowire core, it is in principle possible to create arbitrarily complex radial heterostructures in which the single-crystal nanowires serve a role analogous to planar substrates in two-dimensional heterostructures. We have recently demonstrated this new structural motif with the growth and characterization of different Si-Ge nanowire structures. For example, we have grown single-crystal germanium nanowire cores and produced epitaxial shells of doped silicon around these cores to create beautiful core-shell or radial heterostructures (Figure 2c).

I believe that our ability to control the growth of both the axial and radial nanowire heterostructures will open up many exciting opportunities in both nanoscience and nanotechnology for many years to come. As examples, we have shown that axial superlattices can be used to create new optical bar codes as well as nanoscale diodes and light-emitting diodes,⁴ while

studies of the Si-Ge radial heterostructures already suggest their potential for yielding one-dimensional hole and electron gases for fundamental physics studies.⁵

Fundamental Electronic Properties and Devices

Our approach to nanowire growth has enabled us to prepare an extremely diverse array of nanoscale building blocks with precisely controlled size, composition, and doping, including heterostructures. Moreover, because these materials can be isolated and manipulated in solution like molecules, it is interesting to consider how one might create unique function by assembling together distinct building blocks. To understand and predict how individual nanowires and combinations will behave, and thus define potential device functions, we must elucidate the fundamental properties of the different building blocks.

Within the context of nanocomputing, I will now focus on the electronic properties of several distinct types of nanowires. The flow of carriers through semiconductor nanowires can be readily assessed by fabricating devices like a field-effect transistor (FET), where lithographic processing is used to define source/drain contacts to the ends of a nanowire, and a third electrode coupled through a dielectric is used as a gate to vary the electrostatic potential.^{3,6}

Significantly, studies of silicon and other nanowire materials have shown that the nanowire devices can behave as excellent FETs, and moreover, the carrier mobilities in the silicon nanowire devices are comparable to or exceed the best achievable in planar silicon.^{6,7} The high mobilities in Si and other nanowires may be due to the perfection of these “synthesized” versus fabricated nanostructures, the low dimensionality of nanowires, or other effects, and this represents an important topic for future study. Synthesized nanowires are grown from an elemental source, while fabricated nanowires are formed by lithography or etching of a uniform silicon wafer. In terms of FETs, the nanowires have an on/off current ratio varying by many orders of magnitude for small changes in gate voltage, and thus they can be used to assemble devices and circuits with gain—that is, enable signals to be transferred across many devices as ultimately required in an integrated computing system.

These types of studies have been important in defining the basic properties of nanowire FETs, although our ability to integrate these structures is ultimately limited by the same lithographic constraints of conventional microelectronics. To exploit the potential of the bottom-up paradigm, we have thus conceived of a

scalable architecture consisting of crossed nanowires, where one nanowire consists of the active device and the other crossed nanowire serves as the gate, in the case of FETs. Central to this idea is that we define all nanoscale metrics during the synthesis of the wires and their assembly.^{7,8} This approach has been used to fabricate very good and reproducible crossed-nanowire FETs and, moreover, provides natural scaling and the potential for integration at the highest densities without additional complexity. For example, multiple crossed-nanowire gates can be assembled (see next discussion) over the active nanowire device to build a multi-input logic gate.

Nanowire transistors represent a known, albeit important, device needed for almost any nanocomputing architecture. A more distinctive device that we have also developed is a crossed-nanowire bistable switch, which could be used for non-volatile random-access memory and programmable logic. The concept underlying this work is that we can merge our advances in the growth of radial nanowire heterostructures—the foundation of the bottom-up approach—with our knowledge of semiconductor physics to design heterostructures with bistable on/off states. Such devices are stable in both the on and off states. This idea has been realized recently in a silicon nanowire radial heterostructure using a sequence of δ -doped *p*-type and *n*-type layers spaced by intrinsic silicon shells. In δ -doping, a single layer or a very small number of atomic layers of dopant are added, producing a nonuniform distribution of dopant that resembles a δ function. Notably, device characterization studies have shown that these crossed-nanowire structures exhibit bistable on/off states with large on/off ratios, reproducible switching, and retention times of at least a day. These characteristics suggest the use of these new nanodevices for both programmable logic and nonvolatile memory.

Assembling a Nanocomputer

I believe that revolutionary advances beyond existing technology will require systems in which all or nearly all function is derived from nanoscale elements. This challenge is where we are concentrating our efforts, although hybrid approaches that merge limited nanoscale function with conventional microelectronics circuitry may also play an important role in the future. At a very general level, it is possible to organize a universal computing architecture that is based upon hierarchically assembled and interconnected crossed-nanowire arrays of the bistable switch and FET elements described earlier.

Critical to this general architecture is the development of assembly strategies that enable testing of increasingly complex and integrated crossed-nanowire arrays. A powerful approach that we have developed to organize integrated nanowire structures is fluidic-directed assembly (Figure 3).⁸ In this method, the laminar flow within a microfluidic channel aligns a nanowire solution much like logs on a river to create a parallel array, where the spacing and the spatial position can be defined by the density of the solution and by chemical patterning of the substrate, respectively. Moreover, a second fluidic-directed assembly step carried out using a distinct nanowire solution at a right angle to the first step will yield a crossed-nanowire array with function determined by the nanowires used in the layer-by-layer assembly process.

We have used this approach to assemble NOR (not OR) logic gates by organizing different components in the two sequential steps: first, wires for FETs, and second, crossed nanowires for input gates. Significantly, we have shown that multi-input structures can be configured as NOR logic gates with both reproducible output and substantial gain (Figure 3).⁷ These results demonstrated for the first time that integrated nanodevices or circuits could be fabricated by assembly and not just lithography. More recently, we have used this same assembly approach with different nanowire building blocks to create bistable switch arrays. Electrical characterization studies of these arrays demonstrate that the individual elements are rewritable, point-addressable without crosstalk, maintain the large on/off ratios and retention times of the individual devices, and thus can be considered both nonvolatile memory and programmable logic arrays.

The small arrays produced using fluidic-directed assembly have served as critical tests to validate the bottom-up paradigm for building integrated electronics. Yet realizing a nanocomputing system will require integration and complexity to be increased significantly in the future. To this end, an exciting advance my group has made recently centers on the use of the Langmuir–Blodgett technique. In this approach, we disperse nanowires—again, thinking of them as large molecules—onto an aqueous subphase, increase the surface pressure to both align and control the inter-nanowire separation, and then transfer them to a substrate that could be arbitrarily large. Significantly, using this method we have produced uniform parallel and crossed-nanowire arrays with pitches controlled from microns down to as small as 30 nm, which is equivalent to a crosspoint

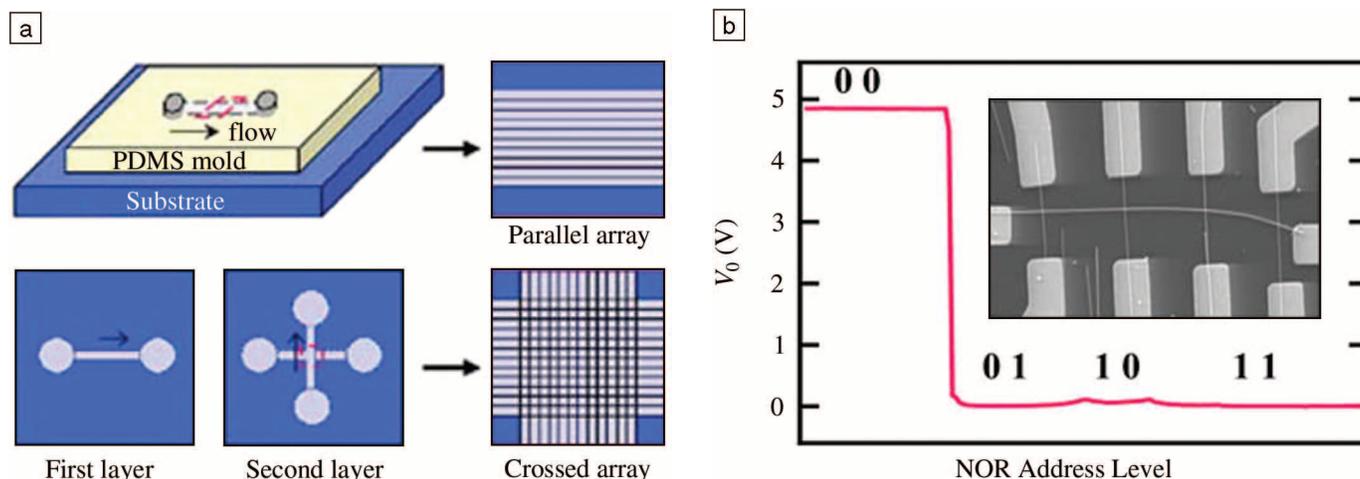


Figure 3. Integrated nanowire electronics. (a) Schematic overview of fluidic-directed assembly of parallel and crossed-nanowire structures. PDMS is poly(dimethylsiloxane). (b) Output from a crossed-nanowire NOR (not OR) logic gate. The inset shows a scanning electron micrograph of a NOR logic gate; thin horizontal and vertical lines are nanowires (spacing $\approx 1 \mu\text{m}$).

“device” density of $10^{11}/\text{cm}^2$ (each cross-point functions as a device). I also think this is significant because, within the bottom-up framework, we can carry out such integration with virtually any of the diverse nanowire building blocks now available.

I do want to stress that realistic architectures will require some hierarchy and cannot be implemented with a uniform transfer of nanowires on a substrate; that is, we must tile the substrate much like tiling a floor in a regular pattern with two or more colors. One approach we have explored to achieve this key goal has involved creating a chemical pattern on the substrate that is complementary to the hydrophobic nanowire surfaces prior to transfer from the Langmuir–Blodgett trough. Notably, this relatively simple approach has enabled us to transfer arrays of parallel nanowires in a regular pattern over large areas of substrate; in addition, by carrying out a second transfer step with the substrate rotated 90° , it is also possible to make crossed-nanowire arrays over a large substrate area. With our ability to hierarchically organize crossed nanowires *en masse*, together with our demonstration of function in smaller arrays, I believe we are in sight of being able to assemble high-density memory and, with more work, achieving the more challenging goal of a universal nanocomputing system.

Nanoscale Sensors

Another exciting venue in which to explore the bottom-up paradigm is at the interface between nanotechnology and biology.⁹ Many biological macromolecules, such as proteins and nucleic acids, are

comparable in size to nanoscale building blocks, and thus these structures can provide a natural bridge between biology and nano/microsystems.

My group has been aggressively exploring the potential of our very well-defined semiconductor nanowires to serve as sensors for the detection of chemical and biological species.⁹ The idea underlying our work is that the conductance of nanowires configured as FETs will change in response to the binding of chemical and biological species at the nanowire surface—that is, molecular or macromolecular species “gate” the FET and thereby change the conductance.

Our solid-state silicon nanowire FETs are transformed into nanosensors by modifying the silicon oxide surface with a receptor selective to a specific chemical or biological species (Figure 4a). A straightforward example of this concept is pH sensing, where we take a nanometer-scale FET that can be turned on and off by a back gate, functionalize the nanowire surface with molecules that respond to changes in hydrogen-ion concentration, and then measure the conductance as a function of solution hydrogen-ion concentration. Significantly, we have shown that as the solution pH is increased stepwise from pH 2 to pH 9, the nanowire conductance also increases in a stepwise manner (Figure 4b).⁹ The increase in conductance is due to an increasing negative surface charge density, as we demonstrated in independent studies of unmodified nanowires.

Our ability to measure pH in real time on the nanometer scale is interesting, although I believe the most important outcome of these studies was to demonstrate

that we can use chemistry to elaborate the surfaces of the silicon nanowire FET receptors for more complex molecules. For example, small-molecule drugs could be bound to the surface and used as receptors to study their interactions with proteins, where it is important to recognize that even small proteins can be similar in size to the nanowires (Figure 4c).⁹

As another example, we have been developing the nanowire sensors for early-stage detection of cancer marker proteins, such as prostate-specific antigen (PSA), using antibodies. We have recently demonstrated real-time, concentration-dependent measurements of PSA using antibody-modified nanowires and found that the binding constants extracted from these data were in agreement with bulk values and that the sensitivity could be increased to a level that exceeds the best clinical tests. In looking to the future, I would like to exploit not only the sensitivity of nanowires but also the potential to assemble them in highly integrated arrays with unique receptors. In the limit of ultrahigh density, one could create an almost universal health-care sensor that detects simultaneously in real time all serum proteins and disease marker proteins, viruses, and other pathogens, and screens genomic DNA for a large or complete set of single-nucleotide polymorphisms that are expected to provide a means for assessing disease risk.

Nanophotonics

The bottom-up paradigm implemented using optically active nanowire building blocks provides what I believe is a unique and highly flexible pathway for creating

multicolor nanophotonic structures and, moreover, for integrating photonics and planar silicon technology. To illustrate this idea, I will first focus on nanoscale light-emitting diodes (nanoLEDs), where we produce the critical active *p*-type/*n*-type

junction of the LED by sequential assembly of *p*-type and *n*-type nanowires in the crossed-nanowire device architecture described earlier. For example, we have shown that assembly of *p*-type and *n*-type indium phosphide crossed-nanowire structures behave as *p*-*n* diodes.³ Significantly, when a forward voltage bias is applied to an indium phosphide crossed-nanowire *p*-*n* diode, light characteristic of the bandgap of nanoscale indium phosphide is emitted from the single crosspoint; that is, we have assembled a true nanophotonic source (Figure 5a).³

The beauty of this bottom-up approach is that a wide range of existing building blocks can be assembled in a similar manner to create multicolor LEDs in a way not possible with conventional planar technology. For example, we have shown that nanoLEDs can be assembled using *n*-type direct-bandgap nanowires, including gallium nitride (ultraviolet), cadmium sulfide (green), and indium phosphide (near-

infrared), and silicon nanowires as a common *p*-type indirect-bandgap material. Notably, we have assembled multiple *n*-type nanowires across single *p*-type silicon nanowires to create nanoLEDs that can simultaneously produce ultraviolet, green, and red light from a very simple device.

This concept can be taken further to assemble nanowire LEDs with planar silicon technology. For example, silicon nanowires can be lithographically patterned on planar substrates; then, our direct-bandgap nanowires can be assembled over these "wires" to create LED arrays, as we have recently demonstrated using cadmium sulfide nanowires. I believe that this represents an exciting approach to integrating high-efficiency photonics with silicon through the bottom-up paradigm.

As a last example of the power of nanowires as photonic building blocks, let me describe recent efforts directed toward the assembly of single-nanowire electrical injection lasers.¹⁰ Nanowires with diame-

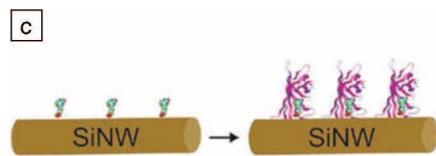
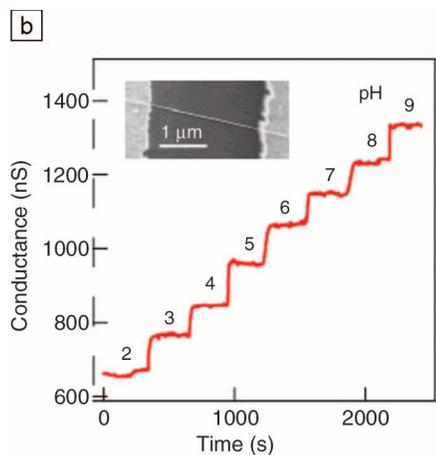
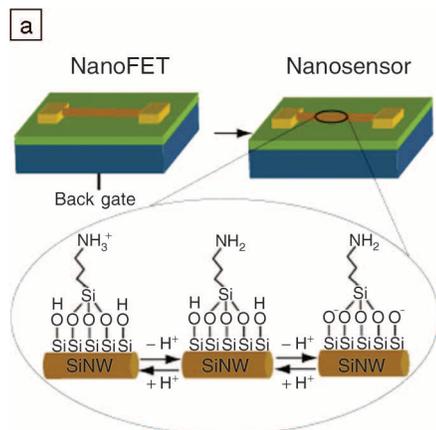


Figure 4. Silicon nanowire (SiNW) nanosensors. (a) Schematic illustration of the transformation of a nanowire field-effect transistor (FET) into a nanosensor. (b) Real-time sensing of solution pH with a functionalized nanowire device. Conductance is measured in nanosiemens. (c) Schematic illustration of the detection of proteins using a small-molecule receptor. The inset shows a scanning electron micrograph of a single-nanowire nanosensor; the thin line is the nanowire, connecting the source/drain metal contacts at the left and right edges of the image.

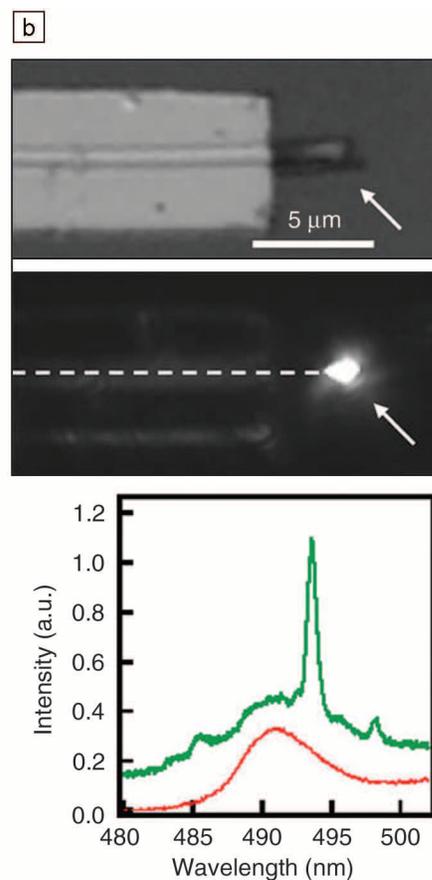
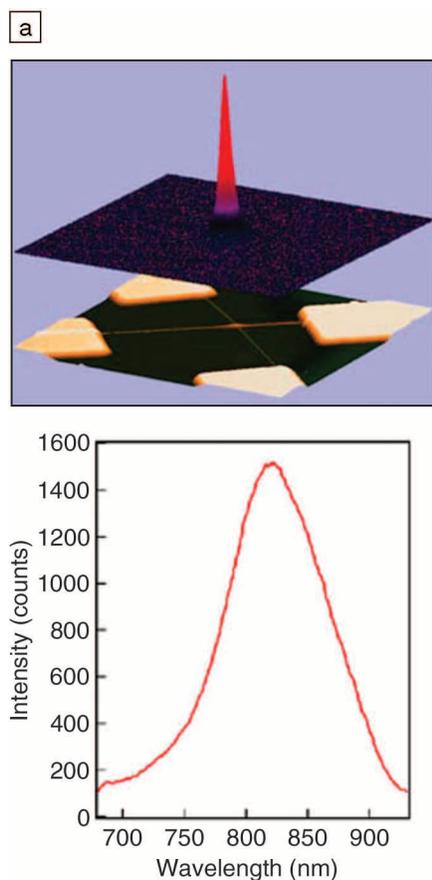


Figure 5. Nanowire photonic devices. (a) Image of the structure and light emission (top) and plot (bottom) of the emission spectrum from a crossed-nanowire light-emitting diode. (b) Images of the structure and light emission (top) and plots (bottom) of the emission spectrum from a single-nanowire injection laser below (red) and above (green) the lasing threshold.

ters on the order of 50–100 nm can function as single-mode waveguides, and when the ends are cleaved, serve as Fabry–Pérot cavities, that is, optical-resonator cavities. To create electrically driven lasers, we inject electrons and holes uniformly along the length of the nanowire cavity.

We have demonstrated this very important idea with cadmium sulfide nanowires assembled in a simple sandwich structure, where a *p*-silicon substrate on which the nanowire is assembled injects holes, and a metal electrode on top of the nanowire—leaving one end uncovered—injects electrons (Figure 5b).¹⁰ I believe it is very exciting and significant that when we apply a forward bias to these structures, light is emitted from the nanowire end with a highly nonlinear intensity–voltage relationship; moreover, we have shown that the emission corresponds to a single dominant mode with an instrumental limited linewidth. These features are characteristic of lasing. There are certainly many studies to envision for the future. In addition to fundamental studies of single-nanowire injection lasers, I believe it is worth recognizing the very real possibility of assembling from different nanowire building blocks integrated multicolor nanowire injection laser arrays that could be exploited in a host of technologies.

Conclusion

This overview has shown that semiconductor nanowires are attractive, versatile, and powerful building blocks for the assembly of functional nanometer-scale

systems ranging from nanoscale memory and computers to detectors with molecular sensitivity and novel photonic systems. These results suggest great promise for the bottom-up paradigm of nanotechnology, and by focusing on the fundamentals of nanoscience, I believe there is the potential to revolutionize some of the most important technologies we know today and most likely open unexpected areas in the future.

Acknowledgments

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Charles M. Lieber holds the Mark Hyman Jr. Chair of Chemistry at Harvard University. After receiving his PhD degree (1985) from Stanford University and serving as a post-doctoral research fellow at the California Institute of Technology (1985–1987), he joined the faculty of Columbia University. Lieber joined Harvard in 1991. Lieber has received numerous awards and honors, including most recently being elected a member of the American Academy of Arts and Sciences (2002) and receiving the 2003 American Physical Society James C. McGroddy Prize for New Materials, the 2002 Materials Research Society MRS Medal, and the 2001 Feynman Prize in Nanotechnology. He serves on several editorial and advisory boards and has over 200 publications and 18 patents. Lieber recently founded a nanotechnology company, NanoSys Inc., with the goal of revolutionizing commercial applications in chemical and biological sensing, computing, photonics, and information storage. He earlier received the Outstanding Young Investigator Award (1993) from MRS, and he is a fellow of the American Physical Society and of the American Association for the Advancement of Science.

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