Carbon nanotubes are cylindrical molecules with a diameter of as little as 1 nanometer and a length up to many micrometers. They consist of only carbon atoms, and can essentially be thought of as a single layer of graphite that has been wrapped into a cylinder. (See figure 1 and the article by Thomas Ebbesen in PHYSICS TODAY, June 1996, page 26.)

Nanotubes have aroused great excitement recently. What is so special about them? The answer lies in their unique physical properties, which span a wide range—from structural to electronic. For example, nanotubes have a light weight and a record-high elastic modulus, and they are predicted to be by far the strongest fibers that can be made. Their high strength is accompanied by their ability to buckle in a reversible manner: When a tube is bent, it does not directly fracture like most materials but buckles like a drinking straw. When the bending strain is released, the tube straightens out again. Such remarkable mechanical properties are relevant to a broad range of potential applications. (Also see page 9.) Similarly, the capillary properties of nanotubes hold promise in applications such as catalysis and energy storage. Then there are the exceptional electrical properties of nanotubes—the subject of this article. As described below, nanotubes can be semiconductors or metals, and they display exciting quantum wire properties.

**Growth of carbon nanotubes**

Nanotubes were discovered in 1991 by Sumio Iijima at the NEC Fundamental Research Laboratory in Tsukuba, Japan. He observed tubular features in electron microscopy images of fullerene soot produced in an arc discharge. They were identified as fullerene tubes consisting of multiple shells, in which many tubes were arranged in a coaxial fashion. In retrospect, it is quite likely that such multiwall nanotubes were produced as early as the 1970s during research on carbon fibers. At that time, however, they were not recognized as interesting new nanoparticles and did not receive much attention. In 1993, Iijima’s group, as well as Donald Bethune and his colleagues at IBM’s Almaden Research Center in San Jose, California, found that the use of transition-metal catalysts leads to nanotubes with only a single shell. Because of their simple and well-defined structure, such single-wall nanotubes serve as model systems both for theoretical calculations and for key experiments.

Research on the physical properties of tubes really took off after 1995, when Richard Smalley and his coworkers at Rice University found a laser ablation technique that could produce single-wall nanotubes at yields of up to 80% instead of the few percent yields of early experiments. The tubes are formed with a narrow distribution of diameters and generally assemble into “ropes”—bundles of parallel nanotubes (see figure 2). Later, Catherine Journet and her colleagues in Patrick Bernier’s group at the University of Montpellier II in France showed that high yields of single-wall nanotubes can also be obtained with the arc-discharge method. A recent development is the use of chemical vapor deposition (CVD), in which nanotubes are grown by decomposing an organic gas over a substrate covered with metal catalyst particles. Either multiwall or single-wall tubes can be grown with this technique, as has recently been shown by Jing Kong and her coworkers in Hongjie Dai’s group at Stanford University. The CVD technique has the potential for making possible the large-scale production of nanotubes as well as the growth of nanotubes at specific sites on microfabricated chips or at the tips of scanning probe microscopes.

A nanotube can be characterized by its diameter and its chiral angle, as illustrated by the wrapping process of figure 1. A major challenge in nanotube growth remains to control the tube diameter and in particular the tube chirality.

**Metals or semiconductors**

The conductive properties of nanotubes depend drastically on both the diameter and the chirality of the hexagonal carbon lattice along the tube. A slight change in the winding of hexagons along the tube can transform the tube from a metal into a large-gap semiconductor! It turns out that about two-thirds of tubes are semiconducting and one-third are metallic (see figure 1). These remarkable electronic properties of nanotubes were first predicted by the Naval Research Laboratory’s John Mintmire, Brett Duncan, and Carter White, who had already performed electronic band-structure calculations before the experimental discovery of nanotubes was even disclosed. The same results were independently obtained by Noriaki Hama and his colleagues at NEC, as well as by Riichiro Saito and his coworkers in the group of Mildred Dresselhaus at MIT.

Although these theoretical predictions were reported as far back as 1992, only recently has experimental verification become possible, using the tip of a scanning tunneling microscope (STM) as a spectroscopic probe. In such a mode, the position of the STM tip is fixed above a nanotube, the voltage on the sample is swept, and the tunneling current between tip and nanotube is recorded. The observed tunneling conductance is a direct measure of the local electronic density of states of the nanotube. Since the STM has the additional power to obtain atomically re-

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solved images of the tube’s hexagon lattice, the electronic structure can be correlated with the chiral structure of the tube. Both Jeroen Wildöer and his coworkers in our Delft group and Teri Odom and her colleagues with Charles Lieber at Harvard University independently showed that the STM spectra for various nanotubes appear to exhibit only two characteristic types of behavior (see figure 3). The measured density of states is remarkably similar to the results of tight-binding calculations, verifying that nanotubes can indeed be either semiconducting or metallic.

Molecular quantum wires

Nanotubes exhibit unique quantum wire properties that derive from the tubes’ nanometer diameters in combination with the special electronic structure of graphite. The electron wavelength around the circumference of a nanotube is quantized due to periodic boundary conditions—only a discrete number of wavelengths can fit around the tube. Along the tube the electrons are not confined; because of the quantization of circumferential modes, however, the tube’s electronic states do not form one wide electronic energy band but instead split into one-dimensional subbands with band onsets at different energies. For single-wall nanotubes, these subbands are widely separated in energy, on the scale of 1 eV (see figure 3)—much larger than the room-temperature thermal energy $k_B T$, about 0.025 eV. Only two of the one-dimensional subbands cross the Fermi energy in metallic nanotubes; all the current through such tubes is therefore predicted to be carried by only this pair of subbands. Because each subband can in principle support a conductance of $G_0 = 2e^2/h$ (the conductance quantum), one expects a conductance of $2G_0$ for metallic nanotubes. Nanotubes are thus predicted to be prototype one-dimensional quantum wires.

The one-dimensional nature of the energy bands is illustrated in the Van Hove singularities in the STM spectra (figure 3). They are the sharp peaks that appear in the density of states at energies where the bottom or the top of the one-dimensional subbands are located. Resonances in Raman scattering experiments by Peter Eklund’s group at the University of Kentucky have provided additional evidence for such sharp peaks in the density of states of nanotubes.

That it is possible at all to have a metallic wire formed from a single molecule is very unusual. In 1930, Rudolf Peierls showed that one-dimensional metallic wires are essentially unstable and will turn semiconducting. For instance, imagine a metallic wire consisting of a regularly spaced row of atoms with a number of electrons on it. It is energetically favorable to rearrange the atomic positions along the row a tiny bit so that the electrons set up a charge density wave and gain electronic energy at the price of the elastic energy associated with the lattice distortion. In doing so, an energy gap opens up right at the Fermi energy, rendering the row semiconducting. That is what happens to polyacetylene and other so-called con-
ducting polymers, which have gaps of a few eV. (See the article on charge density waves in PHYSICS TODAY, May 1996, page 42.)

Nanotubes are an exception to this general rule. Because of the tubular structure, the energy change of setting up a Peierls distortion is very unfavorable. The lattice cost of simultaneously rearranging the positions of all the carbon atoms is large, and the gain in electronic energy is low since there are only two subbands near the Fermi energy. The fact that nanotubes can be metallic at the level of a single molecule is therefore unique.

Electron transport studies

For careful studies of the electrical transport properties, experiments on individual tubes are highly preferred over the spaghetti-like structure of pristinely grown tubes. The first measurements on individual nanotubes were carried out on multiwall nanotubes by a number of groups. Using STM lithography to contact a tube, Luc Langer and his coworkers, working with Jean-Paul Issi and Chris van Haesendonck at the Catholic University of Louvain/Leuven in Belgium, observed weak localization and universal conductance fluctuations at low temperatures. At Harvard, Dai, Eric Wong, and Lieber used a conductive tip of an atomic force microscope to measure the resistance of a tube along its length. Ebbesen and his coworkers at the NEC Research Institute in Princeton, New Jersey, put multiple metallic probes on individual tubes and found highly varying resistivities. These early studies on multiwall nanotubes pointed to defect scattering, diffusive electron motion, and localization with a characteristic length scale of only a few nanometers.

It therefore came somewhat as a surprise when the first experiments on individual single-wall tubes showed that nanotubes can have delocalized wavefunctions and behave as true quantum wires. These properties were discovered independently by Sander Tans and his coworkers in our group at Delft when studying individual single-wall nanotubes, and by Marc Bockrath and his colleagues working with Paul McEuen at the University of California, Berkeley in their experiments on a bundle of tubes. Both teams obtained their results in collaboration with Smalley’s group, which provided the high-quality single-wall nanotube material.

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**Figure 3.** CARBON NANOTUBES CAN BE metallic or semiconducting, depending on their chirality. The panels on the left show the atomic and electronic structure of a metallic (9,9) “armchair” nanotube shown in the inset; the panels on the right show the same for a semiconducting (11,7) nanotube. In both sets of panels, the upper portion denotes the calculated density of electronic states. The density of states at the Fermi energy ($E = 0$) is finite for a metallic tube, but zero for a semiconducting tube. The gap for the semiconducting nanotube is about 0.7 eV. At higher energies, sharp Van Hove singularities show up in the density of states for both the semiconducting and metallic nanotubes. Scanning tunneling microscope spectroscopy on single-wall nanotubes confirms all these features, as shown in the experimental curves in the two lower panels. The normalized differential conductance ($dI/dV)/(dI/dV)_0$ is a measure of the density of states. (Upper images are courtesy of Philippe Lambin, University of Namur; lower images are from Liesbeth Venema, Delft University of Technology.)

**Figure 2.** GROWTH OF CARBON NANOTUBES. (a) This entangled web of ropes of single-wall nanotubes, observed with an electron microscope, was grown by the laser ablation technique developed by Andreas Thess and his colleagues in Richard Smalley’s group at Rice University. This technique provided high-quality material in amounts that enabled experiments to be conducted on the physical properties of single-wall nanotubes. Scale bar is 100 nm. (b) Cross section of a single bundle (a “rope”) with many parallel nanotubes. Each circle is the cross section of an individual single-wall nanotube. Scale bar is 10 nm. (Adapted from ref. 4.)
As shown in figure 4, measurements at millikelvin temperatures on single metallic nanotubes lying across two metal electrodes reveal steplike features in the current–voltage curves, whose positions depend on the voltage applied to a third electrode, a gate electrode that is electrostatically coupled to the nanotube. These features are due to single-electron tunneling and resonant tunneling through single molecular orbitals. Such concepts are well studied in the field of mesoscopic physics. Single-electron tunneling occurs if the capacitance of a conducting nanostructure—the nanotube in this case—is so small that adding even a single electron requires an electrostatic charging energy that is larger than the thermal energy $k_BT$. Transport at low bias will then be blocked (the so-called Coulomb blockade), but it can be restored by tuning the electrostatic potential by means of the gate voltage. With increasing bias voltage, the current increases in steps, indicating resonant tunneling through single molecular orbitals in the nanotube as shown in the electronic energy diagram in the inset. At each step, an additional electron level on the tube becomes available for carrying current. The width of these current steps thus reflects the splitting $\Delta E$ between the quantized energy levels, about 1 meV for a metallic nanotube 1 μm long. Note that these data exemplify the physics of transport at the meV energy scale rather than the eV-scale band-structure physics of figure 3. (Adapted from Tans et al., ref. 9.)

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FIGURE 4. ELECTRICAL TRANSPORT through metallic carbon nanotubes. (a) An individual single-wall carbon nanotube (blue) lying on two platinum electrodes (brown) on an insulating silicon dioxide substrate (green). Electrical transport through the nanotube is measured by applying a bias voltage to the electrodes and measuring the current. A gate voltage, applied to the third electrode in the upper-left corner of the image, can be used to modulate the transport through the tube. (b) Current–voltage curves measured for the sample of (a) at about 50 mK for three different gate voltages. At low bias voltage, no current can flow because of Coulomb charging of the nanotube. The size of the conductance gap is modulated by the gate voltage. With increasing bias voltage, the current increases in steps, indicating resonant tunneling through single molecular orbitals in the nanotube as shown in the electronic energy diagram in the inset. At each step, an additional electron level on the tube becomes available for carrying current. The width of these current steps thus reflects the splitting $\Delta E$ between the quantized energy levels, about 1 meV for a metallic nanotube 1 μm long. Note that these data exemplify the physics of transport at the meV energy scale rather than the eV-scale band-structure physics of figure 3. (Adapted from Tans et al., ref. 9.)

FIGURE 5. QUANTIZED CONDUCTANCE through a metallic nanotube. (a) In situ electron-microscope image of a multiwall carbon nanotube fiber above a liquid metal contact (mercury), before and after (inset) contact is made. (b) Conductance versus position of the nanotube fiber at room temperature. As the nanotube fiber is dipped into the liquid metal contact (gallium in this case), the conductance increases in steps of $G_0 = 2e^2/h$. The steps correspond to different nanotubes coming into contact with the liquid metal as the fiber is lowered. (Images from Walt de Heer, Georgia Institute of Technology.)

![Figure 4](image1.png)

![Figure 5](image2.png)
two-dimensional regime. Although this pure one-dimensional behavior to the doughnut-shaped electron wavefunctions will appear to be only weakly sensitive to disorder. Whereas a local defect will have a drastic effect in a single row of atoms, leading to localization, the same defect will be much less severe in a nanotube because its effect will be averaged out over the whole tube circumference due to the doughnut-shaped electron wavefunctions.

Such a large spatial extent for the electron wavefunctions is remarkable because it has been known since the work of Philip Anderson in the 1950s that states in a one-dimensional system with a finite amount of disorder cannot be extended infinitely but will inevitably be spatially localized. The experimental findings of micron-sized localization lengths in nanotubes have recently received theoretical support. The University of Oxford’s Tchavdar Todorov and White have calculated that the localization length can be extremely large in metallic nanotubes—on the order of 10 μm—because of their cylindrical physical structure and the associated electronic structure with only two relevant subbands. Nanotubes appear to be only weakly sensitive to disorder. Whereas a local defect will have a drastic effect in a single row of atoms, leading to localization, the same defect will be much less severe in a nanotube because its effect will be averaged out over the whole tube circumference due to the doughnut-shaped electron wavefunctions.

For very large diameters, there is a crossover from this pure one-dimensional behavior to the two-dimensional character of a sheet of graphite. Although thin single-wall nanotubes are within the one-dimensional limit, thick multi-wall nanotubes may get into the two-dimensional regime.

Recent surprises

Research on carbon nanotubes is developing at a very fast pace. All of the above experimental work was done in 1996–97. Although a number of basic features in the electron transport through nanotubes were discovered, many questions remain. In the past year, many additional interesting features have been found.

One fact being explored is the relevance of the electron spin to transport through a nanotube. Experiments on nanotube ropes by David Cobden and his colleagues in McEuen’s group at Berkeley indicate that electron transport depends on whether the electron number is even, with spins paired in singlet states, or odd, with one unpaired electron spin. Experiments on single nanotubes by Tans in our Delft group show that such a simple spin alternation may not be the complete story, because many electrons that subsequently enter the nanotube upon increasing the gate voltage are found to have the same spin direction. In a collaboration with Michel Devoret from CEA/Saclay (the French Atomic Energy Commission’s Saclay research center), our group has developed a model for this behavior, in which spin-polarized states can result from spin flips induced by the external gate voltage. Interestingly, the capacitance between the nanotube and the gate electrode is experimentally found to depend on the many-body quantum state of the nanotube. These experiments indicate the relevance of electron–electron correlations.

The effects of correlations in a one-dimensional electron liquid is a subject with a long history. In the years after World War II, Sin-itiro Tomonaga—and later, Joaquín Luttinger—found that electrons in a one-dimensional system no longer form the Fermi liquid that is the common state for metals in the three-dimensional world. Instead, a correlated electron liquid, commonly called a Luttinger liquid, is formed and exhibits various peculiar properties such as a separation of spin and charge as well as power-law correlations. (See the reports in PHYSICS TODAY, September 1996, page 19 and June 1994, page 21.) The theory for correlated electrons in carbon nanotubes—which are, after all, prototype one-dimensional conductors—has been worked out independently by Leon Balents and Matthew Fisher at the University of California, Santa Barbara with Charles Kane at University of Pennsylvania, and by Reinhold Egger at the University of Freiburg in Germany with Alexander Gogolin at the University of London’s Imperial College of Science, Technology and Medicine. Recent work by Bockrath and his coworkers at Berkeley suggests that Luttinger signatures can indeed be observed in a power-law dependence of the resistance on bias voltage ($R \propto 1/V^n$) and temperature ($R \propto 1/T^\nu$). The resistance of a metallic nanotube thus decreases with increasing temperature. Some other groups, however, have found the reverse trend above 100 K or so. An understanding of such a basic measurement as the temperature dependence of the resistance is not yet available, and more research needs to be done to fully clarify the electron transport properties of nanotubes.

It appears possible to improve the conductance of carbon nanotubes by use of doping. Roland Lee and his coworkers in the group of Jack Fisher at the University of Pennsylvania have found that the room-temperature resistance of bulk nanotube material can be decreased by up to a factor of 30 by adding electrons through doping with potassium or adding holes using bromine. The origin of the large increase of conductance is still unclear. One of the motivations for the doping experiments has been a search for superconductivity in carbon nanotubes. Attempts to find it have been unsuccessful until now. Very recent work done by Alekber Kazumov and his collaborators with Helène Bouchiat at the University of Paris XI in Orsay, France, suggests the occurrence of proximity-induced supercurrents running through undoped carbon nanotubes mounted on superconducting electrodes. High

$\nu_p = 8 \times 10^4$ m/s is the Fermi velocity. This simple estimate fits well with the observation, indicating that the wavefunction of electron states is extended over the full length of the nanotube. These experiments thus show that nanotubes indeed behave as beautiful quantum wires.
values of the critical current were found, especially for individual single-wall nanotubes.

Last January, Liesbeth Venema and her coworkers in our group at Delft reported that it is possible to image the wavefunctions of the quantized “particle in a box” states with an STM. An STM was used to cut a nanotube down to lengths of only 10–100 nm. In such short nanotubes, the energy level splitting $\Delta E$ is large, about 0.1 eV, because it scales inversely with length in a one-dimensional system. With an STM we could therefore image the wavefunctions of the well-separated quantized energy levels. Experiments on a short metallic nanotube indeed showed standing electron waves with a wavelength of about 0.75 nm, which agrees with the expected Fermi wavelength.

A recent development in the field is the renewed interest in multiwall carbon nanotubes. One advantage for certain experiments is that such tubes have a much larger diameter, say 20 nm. This size allows, for example, Aharonov–Bohm studies, in which the nanotube resistance is modulated by a magnetic field along the tube axis. This effect was reported last February by Adrian Bachtold and his coworkers in the group of Christian Schönenberger at the University of Basel in Switzerland. They observed a periodic variation of the resistance with magnetic field with a flux period that suggested that only the outer shell of the multiwall nanotube contributed to transport.

Conduction through only the outer shell was also suggested by the results of experiments done by Stefan Frank and his colleagues in Walt de Heer’s group at the Georgia Institute of Technology. They dipped a multiwall nanotube into liquid mercury or gallium and simultaneously measured the conductance of the tube (see figure 5). Remarkably, they found that the conductance was nearly equal to the conductance quantum $G_0$. Note that even for a single shell contributing to transport, one would expect twice that value for the conductance because of the two current-carrying subbands. This difference is not yet understood.

The Georgia Tech experimenters found that the measured conductance appeared to be independent of the depth to which the tube was inserted into the metal, suggesting that electronic transport occurs ballistically in nanotubes—even at room temperature! As a result, no heat is dissipated in the tubes, and very high current densities can be obtained without destroying the tube. This behavior was recently confirmed by Zhen Yao in our Delft group. He was able to run currents up to 30 $\mu$A (an extraordinary current density of about $10^9$ A/cm$^2$—many orders of magnitude larger than the fusing current density for a regular copper wire) through an individual single-wall nanotube between two electrodes at room temperature.

Applications

The exceptional properties of nanotubes can be exploited in numerous applications. Proposals span a very wide
spectrum, from molecular electronics to even bulletproof vests!

Using nanotubes as field-emitter tips is one promising possibility. Yahachi Saito and his coworkers at Mie University in Japan recently developed cathode-ray tubes equipped with nanotube field emitters14 (see figure 6), which can be used as commercial lighting elements for giant outdoor displays. Replacement of the conventional metallic emitter tip by nanotubes led to favorable device characteristics such as a stable electron emission, adequate lumiance, and long lifetime. The use of nanotubes removes the need for ultrahigh vacuum in these devices, and saves energy because nanotubes field emit at room temperature—no heating is required. The demonstration of this device is an important first step towards real commercial products based on carbon nanotubes.

Another interesting application is the use of nanotubes as tips for scanning probe microscopy (SPM). Ever since the discovery of SPM, the tip has been a "black box." Nanotube tips offer a number of advantages. Nanotubes are chemically inert and mechanically robust, and they have a large aspect ratio and a tip end that is, in principle, well defined. Furthermore, they are "crash-proof": Pressing the nanotube tip onto a surface will buckle the tube rather than induce tip damage. Subsequent withdrawal will relieve the buckle and recover the original tip. Dai pioneered the use of nanotube for SPM tips as a postdoc with Smalley at Rice.15 Stanislauus Wong and his coworkers in Lieber's group at Harvard have recently developed methods to chemically functionalize the very end of the nanotube. Attaching a molecular group that can bind to specific chemical or biological sites makes possible SPM imaging with chemical or biological discrimination. Similar tube-end functionalization has recently been developed by Jie Liu and his colleagues in Smalley's group at Rice.

**Molecular electronics?**

One possible future direction of the field is the exploration of so-called molecular electronics. Dreams about the use of single molecules as active electronic elements have been around for decades. One naturally attractive feature is the ultimate scale of device integration because of the small size of molecular elements. Experimental realization of such dreams has proved hard, but interest in these ideas has been revived with the advent of nanotube molecules. Recently, Tans in our Delft group demonstrated the first prototype device based on a single molecule: a field-effect transistor with only one single-wall nanotube as the active element.16 The current through a semiconducting nanotube lying over two electrodes could be strongly modulated by an external gate voltage, yielding transistor action. Importantly, the device functioned at room temperature. Although it is clear that commercial applications remain far away, this device demonstrates that it is indeed possible to create a device based on a single molecule. Richard Martel in the group of Phaedon Avouris at IBM's Thomas J. Watson Research Center in Yorktown Heights, New York, has recently confirmed the field effect in individual nanotubes.

There is much more to explore in the arena of molecular electronics. An exciting possibility is the creation of on-tube junctions. One option for realizing such a junction is to connect two different tubes through a topological defect containing a 5-atom ring and 7-atom ring pair (figure 7a). If one tube is semiconducting and the other metallic, a heterojunction is formed that possibly will act like a rectifying diode. Note that a device is then created at a scale within a single molecule. The transport properties of such junctions have already been considered theoretically—independently by Leonor Chico and his coworkers in the group of Steven Louie at the University of California, Berkeley, by Riichiro Saito in the Dresselhaus group at MIT, and by Philippe Lambin and his collaborators at the University of Namur in Belgium.17 Nanotubes with kinks as in figure 7a are indeed observed experimentally. Another route toward on-tube devices is to take advantage of the coupling between mechanical and electrical properties. Junctions can be induced by local mechanical deformation—bending the tube with an SPM tip, for example (figure 7b). Alexey Bezyardin in our Delft group found experimental evidence that local bending of a nanotube causes local barriers that act as scattering defects for electrons. This has recently been confirmed by calculations made by Alain Rochefort and Dennis Salahub at the Centre for Research on Computation and its Applications in Montreal, Quebec, with Avouris at IBM, who found a change in electronic structure near local bends.

A strategy for developing integrated devices with many individual elements has yet to be formulated. One promising route is the self-assembly of functionalized nanotubes into single-molecule devices. The chemistry of nanotubes—only now beginning to be explored—will be particularly interesting at the tube ends and also near buckles (recently called “kinky chemistry”).

The future for nanotubes looks very bright. They are extremely versatile: They can, for instance, be functionalized at their ends and along their bodies, filled with superconducting or magnetic metals, coated on the outside, and joined together seamlessly. There appears to be no material in the world that is as strong, conducting, inert, and so forth, all at the same time! The rapid progress with nanotubes is largely driven by this unique combination of properties. Nanotubes provide a showcase in which fundamental science and applications go side by side. To quote Richard Smalley, “These nanotubes are so beautiful that they must be useful for something.”

**References**


