Carbon Nanotubes: Continued Innovations and Challenges

M.S. Dresselhaus and H. Dai, Guest Editors

Abstract

This article outlines the content of the April 2004 issue of *MRS Bulletin* on Advances in Carbon Nanotubes. Essentially, carbon nanotubes are self-assembling nanostructures constructed of sheets of hexagonal-shaped carbon atoms rolled up into cylinders. Carbon nanotubes have attracted a great deal of attention as model systems for nanoscience and for potential applications. The special interest in carbon nanotubes stems from their unique structure and properties: their very small size (down to ~0.42 nm in diameter); the possibility for carbon nanotubes to be metallic or semiconducting, depending on their geometrical structure; their exceptional properties of ballistic transport; their extremely high thermal conductivity and high optical polarizability; and the possibilities of high structural perfection. Research in the carbon nanotube field has now advanced to the stage where a good understanding of the structure and many of the basic properties are in place, together with much appreciation of their interrelation. On the other hand, major gaps in basic knowledge remain, with the major obstacles confronting the carbon nanotube field being the lack of a detailed understanding of the nanotube growth mechanism and control of the synthesis process to produce nanotubes with a desired diameter and chirality. The brief review of the carbon nanotube field by leading experts in this issue comes at an opportune time. Many exciting results on the structural, electronic, optical, and transport properties of these tiny well-ordered structures have already been achieved, and the research is well enough developed to assess present progress and identify new research directions waiting to be explored.

Keywords: *ballistic transport, carbon nanotubes, chemical functionalization, chirality, field-effect transistors, fluorescence, photophysics, Raman spectroscopy.*

Carbon nanotubes, as self-assembling nanostructures, have attracted a great deal of attention as model systems for nanoscience and for various potential applications, including composite materials, battery electrode materials, field emitters, nanoelectronics, and nanoscale sensors. The interest in carbon nanotubes stems from their unique structure and properties, including their very small size (down to ~0.42 nm in diameter); the possibility for carbon nanotubes to be metallic or semiconducting, depending on their geometrical structure; their exceptional properties of ballistic transport; their extremely high thermal conductivity and optical polarizability; and the possibilities of high structural perfection.

Single-walled carbon nanotubes (SWNTs) have only one atomic species (carbon) and a relatively simple structure—a sheet of regular hexagons rolled in a seamless way into a cylinder one atom thick. Because of this simplicity, detailed calculations of the unique properties of SWNTs can be carried out. In fact, theoretical predictions that SWNTs can be either semiconducting or metallic, depending on their geometrical structure, were made[1–3] before a SWNT was ever synthesized in the laboratory.[4,5]

From the inception of research on SWNTs, they have served as a model one-dimensional (1D) system for a variety of new phenomena, where theory and experiment have worked hand-in-hand in advancing the field. SWNTs thus provide a benchmark for new phenomena that might be possible, based on their unique molecular density of states, whereby every structurally distinct (n,m) nanotube (where n and m are the integers defining the wrapping vectors that describe the geometry of each tube) has its own unique and characteristic 1D density-of-states spectra. Now that nanotubes have been synthesized from many other chemical species including carbon, boron nitride, bismuth, metal chalcogenides, and many others, a wide range of novel properties may be discovered. Investigating how the properties of 1D nanotubes differ from 1D nanowires, 2D thin films, or 3D bulk structures of the same material should provide interesting and useful data.

Research in the carbon nanotube field has now advanced to the stage where a good understanding of the structure and of many of the basic properties is in place, together with an appreciation of their interrelation. Many unexpected phenomena that do not occur in the parent graphite material have been discovered in nanotubes, and these discoveries have energized not only nanotube research, but also nanoscience research in general. On the other hand, major gaps in our basic knowledge remain, with the major obstacle confronting the field being the lack of a detailed understanding of the nanotube growth mechanism. Such an understanding is needed because of the unusually close connection between nanotube properties (such as their metallicity) and their geometric structure; the knowledge will allow the diameter and chirality of nanotubes to be controlled by chemical synthesis methods.

The brief review by leading experts of the carbon nanotube field in this issue of the *MRS Bulletin* comes at an opportune time. Many exciting results regarding the structural, electronic, optical, and transport properties of these tiny well-ordered structures have already been achieved, and the research is well-enough developed to assess present progress and identify new research opportunities.

From the beginning, the main emphasis of carbon nanotube research has been in the synthesis area, and this remains the great challenge of the field. This viewpoint is strongly reflected in the organization of this issue. Rapid progress is being made to increase control of the synthesis process, steadily narrowing the diameter and chirality range of the nanotubes, decreasing defects and impurities, and in-

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creasing production efficiency and yield while expanding functionality. The main pursuits in controlling nanotube synthesis include the synthesis of molecular catalytic clusters with atomically well-defined size and shape, the development of mild, reduced-temperature catalytic synthesis conditions, the development of patterned growth with a high degree of control in nanotube location and orientation, and the synthesis of complex and organized networks or arrays of nanotubes on large substrates. Progress in the synthesis area and opportunities for new research are presented in the article by Liu et al.

Because SWNTs are usually grown in the presence of a variety of carbon species, catalyst particles, and other unwanted constituents, much attention has been given to nanotube purification. This has led to techniques for the characterization of nanotube purity, and the preparation/sorting of nanotubes by length, diameter, chirality, and metallic/semiconducting properties. These issues are discussed in detail in the article by Haddon et al.

The issues of synthesis and separation discussed in the articles by Liu et al. and Haddon et al. are closely related. Together, these articles reveal the approaches that eventually might lead to ultimate control of the nanotube synthesis process. With improved synthesis regarding purity and monodispersity in tube diameter and chirality, problems in purification and separation could be largely eliminated or avoided. On the other hand, if efficient and precise purification and separation techniques can be developed, challenges to growth can become much more tractable. It is highly likely that the convergence or combination of the two areas—that is, the controlled or preferential growth of a nanotube with a particular chirality, and purification/separation—could prove powerful in producing nanotubes with well-defined diameters and chiralities.

The filling of nanotubes with various fullerene species offers an entree into using nanotubes as a template for the synthesis of many new nanostructured species. Furthermore, the subsequent thermal conversion of the encapsulated fullerenes to an inner carbon nanotube gives rise to double-walled carbon nanotubes (DWNTs), which are a prototype for studying the structure and properties of multiwalled carbon nanotubes (MWNTs) in a quantitative way. Because of the increased stability and durability of DWNTs and MWNTs relative to SWNTs, these more robust nanotubes have greater potential for applications requiring the exceptional mechanical strength, stiffness, and high thermal conductivity of carbon nanotubes. An assessment of current knowledge about the two prototype materials, SWNTs filled with fullerenes (called peapods) and DWNTs, is presented in the article by Bandow et al.

The use of nanotubes as a laboratory for producing novel nanostructures, including nanowires, is discussed in the article by Sloan et al. The spatial and structural constraints of the hollow core and inner-shell "surface" of the nanotube can give rise to unusual structures. Great strides have been made in both the synthesis of the novel structures within the cores of nanotubes and their detailed structural characterization by high-resolution transmission electron microscopy. Much remains to be done in investigating the unique physical properties possessed by these nanowires, how these properties might relate to their novel nanowire structures, and how the structure and properties of these nanowires relate to those of bulk 3D materials. The wide range of materials that have been shown to form endohedral nanowires within SWNTs is impressive, including the case of peapods containing fullerenes that merge to form a second nanotube within SWNTs. These peapods have thus far received the most attention, with many publications on their novel structural, electronic, and vibrational properties, as described in a complementary way in the articles by Sloan et al. and by Bandow et al. These results are just harbingers of the richness of new phenomena that are expected to occur under the unusual nanolaboratory conditions found within the core region of nanotubes.

Regarding property measurements, the most attention thus far has been devoted to studies of the remarkable transport properties observed in individual field-effect transistors (FETs), including ballistic transport, the suppression of carrier backscattering, single-electron transistor effects, enormously high current densities, good FET device performance, and the demonstration of a variety of device functions (AND and NOR gates, etc.). While high mobility and ballistic transport are partly due to the structural perfection possible with nanotubes, the chemical stability and robustness of nanotubes and the lack of unsaturated surface dangling bonds make nanotubes unique among electronic materials. Sustaining ultrahigh current flows and retaining high mobility are possible when nanotubes are processed and integrated into realistic transistor structures. Recent achievements in advancing our understanding of the remarkable electron transport properties of both semiconducting and metallic single-walled nanotubes are described in the article by McEuen and Park.

The optical photophysics of nanotubes has likewise received considerable attention as a means for studying electron, phonon, and optoelectronic phenomena. Progress on this topic and future challenges are presented in the article by Jorio et al. Since Raman scattering and optical absorption and emission in SWNTs are resonant processes, depending on the singularities in the 1D density of electronic states, these techniques provide a convenient method for characterizing the diameter distribution and the metallicity distribution of the SWNTs in a sample. The relative sensitivities and resolution of the various techniques, such as Raman scattering, optical luminescence, and optical absorption are discussed, although the emphasis in this article is on the new physics uncovered through these studies. Fast optics provides unique information about lifetimes of excited states, while photophysics and transport properties merge in areas of electroluminescence and photoconductivity, two frontiers of nanotube photophysics at present.

A great amount of effort and emphasis in nanotube research has focused on the eventual use of carbon nanotubes in devices. The article by de Heer presents a critical evaluation of progress in the commercial exploitation of carbon nanotubes. Most researchers in the nanotube field at universities, industrial research laboratories, and government laboratories worldwide are optimistic about important and even large-scale applications eventually emerging from nanotube research, although at present the largest volume applications use MWNTs for increasing the strength and modulus of composite materials, and for improving the lifetime and performance of lithium-ion batteries. There are laboratory demonstrations of many interesting prototype devices, but much of the actual commercialization remains in the future. Almost all of the articles allude to applications of nanotubes and to their future commercial exploitation.

The future of nanotubes for electronics applications remains bright. The nearly unparalleled and unprecedented electrical properties and chemical stability of nanotubes provide a powerful driving force to fully utilize these properties. Nanotubes are just beginning to be exploited for building high-performance transistors, but their use has yet to be convincingly demonstrated for interconnect applications. Many device designers are eagerly awaiting breakthroughs to enable them to interconnect increasing numbers of devices in smaller spaces. Nanotubes offer great promise here. Applications of nanotubes as fluorescence-label-free electronic, chemi-

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cal, and biological sensors, especially in miniaturized, multiplexed, and arrayed forms, are novel and promising. For sensor applications, much less stringent requirements will be placed on the nanotube chirality, and just like other sensor technologies, the challenges lie in selectivity and strategies in multiplexing, arraying, and addressing. In the past several years, there has been a steady increase in activities that incorporate nanotubes into biological systems, including proteins, DNA, and living cells. This is an interesting and relatively new area that can be categorized as the "wet" side of nanotube science and technology. Exciting results and discoveries have already been achieved in nanotube research, and much more can be expected in the next few years.

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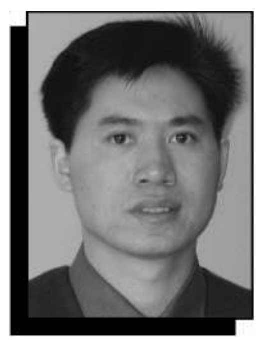
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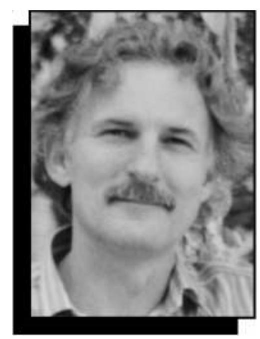
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Bandow received his BS degree in electric engineering from Meijo University in 1981 and his doctorate in science from the Tokyo Institute of Technology in 1991. Prior to joining the faculty of Meijo University in 2001, he was a technical associate at the Institute for Molecular Science, a research associate in the Instrument Center and the Center for Molecular Materials at the Institute for Molecular Science, and a research associate with the Nanotubulites Project of the Japan Science and Technology Corporation. He was also a visiting scientist at the University of Kentucky. Bandow has authored and co-authored more than 80 papers on a wide range of materials science topics.

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Chen received a BS degree (1994) in semiconductor physics and devices and a MS degree (1997) in condensed-matter physics from Wuhan University (China), working on the synthesis, purification, and optical properties of fullerene materials. Chen received his PhD degree from Penn State in 2003, where he focused on the optical properties of nanotubes.

Chen has authored more than 10 peer-reviewed research papers and 20 international conference presentations. He received the Duncan fellowship from Penn State in 2002 and 2003. Chen is a member of the American Physical Society (APS) and the Material Research Society (MRS).

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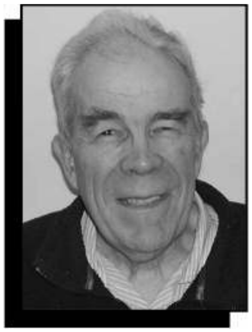
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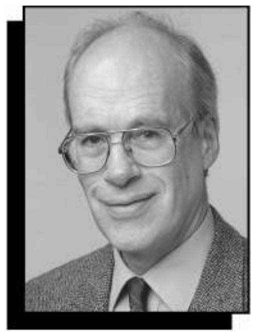
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the discovery of room-temperature ballistic conduction in carbon nanotubes in 1998. He developed a electron-microscopy-based resonance method to measure the elastic properties of nanotubes in 1999. Recently, he has found evidence for superconductivity in small niobium clusters. Currently, he is working on carbon nanotubes, ultrathin patterned graphite films, and the electronic and magnetic properties of cold metal clusters in molecular beams.

De Heer received his PhD degree at the University of California, Berkeley, in 1985. He investigated the properties of alkali clusters in molecular beams, resulting in the discovery of the electronic shell structure and plasma resonances. He moved to Lausanne, Switzerland, in 1987, where he worked at the EPFL until 1996. While there, he studied the magnetic properties of free transition-metal clusters. In 1992, he started his investigations of arc-produced multiwalled carbon nanotubes. Investigations of nanotube films cumulated in the discovery of their field-emission properties.

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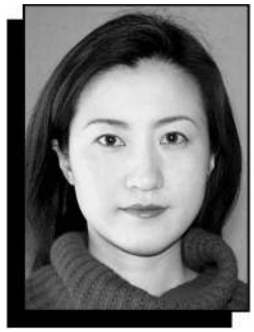
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Haddon is best known for the prediction and discovery of superconductivity in alkali-metal-doped carbon-60. His research group is currently working on the chemistry and applications of carbon nanotubes and on the synthesis and properties of neutral radical molecular conductors.

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Kirkland read natural sciences at Gonville and Caius College, Cambridge, graduating in 1986, prior to completing his PhD degree in high-resolution electron microscopy and image simulation at Cambridge in 1989. After his PhD, he worked in Cambridge as an SERC fellow (1989–1991), a British Ramsay Memorial research fellow (1991–1993), and a JEOL research associate (1993–1997) before being named a senior research fellow. He was appointed to his current position in 2002.

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Luzzi's research program has produced over 100 publications and patents on nanoscale phenomena, instrumentation, and the synthesis, structure, properties, and application of materials. Luzzi's research group discovered the "peapod" class of functional nanomaterials, developing efficient synthesis routes, solving structures, and demonstrating unique properties. For this work, Luzzi received the George Heilmeier Award for Research Innovation. He has conducted a total of three years of research in Japan, is an alumnus of the IDA Defense Science Study Group, and was a science and technology fellow with the CNO Strategic Study Group.

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tubes by length, type, and diameter. He has co-authored more than 70 publications, given more than 100 invited presentations, and co-organized five international conferences.

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