NT10: Recent Advances in Carbon Nanotube Science and Applications

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ABSTRACT A review of recent advances in carbon nanotube science and applications is presented in terms of what was learned at the NT10 11th International Conference on the Science and Application of Nanotubes held in Montreal, Canada, June 29—July 2, 2010.

Nanostructures have in recent years become a central theme in materials chemistry, while carbon nanotubes have become a model one-dimensional (1D) nanostructure of the nanoscale world. In this context, advances made in the science and applications of carbon nanotubes have been reported annually for more than a decade at the international carbon nanotube conference series, with NT10 being the 11th conference in the series, taking place in Montreal, Canada, during the week of June 29—July 2, 2010. As interest in carbon nanotubes has grown during the time frame of the conference series, so has worldwide attendance, reaching a level of 700 attendees in 2010, reflecting the growth of publications in the field (Figure 1). One tradition of the nanotube conference series has been the conference summary presented here, which attempts to capture some of the highlights at the conference, describe the notable trends occurring in the carbon nanotube research field, and provide an outlook toward the future.

One noteworthy trend is that the carbon nanotube field has not only grown over the past decade but has also impacted other areas of science and technology. As a result of this, four satellite conferences have been spawned, which in 2010 met prior to the main NT10 conference. These satellite conferences have led to cross-fertilization with the carbon nanotube research community as well as cross-fertilization among the four satellite conferences themselves. After more than a decade of the nanotube conferences, we now see further growth with four tutorial lectures preceding the start of NT10. The topics covered in the tutorial lectures were selected by the local organizing committee and served as an orientation for newcomers to these research fields. The four satellite workshops were called (1) the sixth International Symposium on Computational Challenges and Tools for Nanotubes (CCTN10); (2) the fourth International Workshop on Metrology, Standardization, and Industrial Quality of Nanotubes (MSIN10); (3) the third Carbon Nanotube Biology, Medicine, and Toxicology Satellite Symposium (CNBMT10); and (4) the first Graphene Satellite Symposium (GSS10). The four tutorials were (1) Nucleation and Growth of Carbon Nanotubes by David Geohegan, (2) Nanotube Structure Determination and Population Evaluation with Transmission Electron Microscopies and Spectroscopies by Annick Loiseau, (3) Fundamentals of the Optical Properties of Carbon Nanotubes and Current Status by Anna Swan, and (4) Graphene and Carbon Nanotube Electronics and Optoelectronics by Phaedon Avouris.

The long-term vitality of the carbon nanotube field has benefited from a fluidity of researchers moving in an interdisciplinary way from carbon nanotubes into other fields of science as well as into new types of applications through the interdisciplinarity and outreach of nanoscience in general. This outreach of the carbon nanotube field highlights the desirability of identifying future research directions for carbon nanotubes. Some insights and suggestions about new research directions come from studies such as the U.S. National Academy of Sciences Decadal Study on Condensed Matter Physics entitled "The Science of the World Around Us";[1] this outreach opportunity was noted by some NT10 speakers, such as Paul McEuen, who also participated in this decadal study. In this study, six research directions were identified for this decade,

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and each can be related in some way to the nanoscale world and to carbon nanotubes more specifically, as noted below.

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The Science of the World around Us: Research Directions. The research direction most directly related to NT10 is the question: "What new discoveries await us in the nanoworld?" Although the properties of materials are intimately connected with the structure of a unit cell, the physical and chemical properties of nanoscale materials differ in detail from their bulk counterparts. On the most fundamental level, the nanoworld is closely involved with the second focus of the decadal study:[2] "How do complex phenomena emerge from simple ingredients?" Clearly, it is at the nanoscale that collective phenomena such as the distinction between the metallic and semiconducting behavior of carbon nanotubes emerges.[3] Nanostructures and chemistry play important roles in providing the powerful devices that support the implementation of the information technology revolution, which is identified in the decadal study as the third research direction for this decade. As a fourth research direction, meeting the energy demands of future generations in a sustainable way will require major research and development activities over several decades, and much of this is expected to utilize nanostructures, chemistry, and the intersection between nanostructures and chemistry. Here, carbon nanostructures should play a significant role. Chemical reactions change the equilibrium conditions of pure reactants, and at the frontiers of chemistry, there are often reactions that take systems far from their equilibrium conditions. Understanding the frontiers of what happens far from equilibrium, which is identified as a fifth research direction for this decade, will surely lead to new research directions for nanotube chemistry. Finally, understanding the physical basis of life processes, identified as the sixth research direction of the decade, relates to the study of chemistry based on natural products and is a focal point of the field of biochemistry.

> What new discoveries await us in the nanoworld? How do complex phenomena emerge from simple ingredients?

Research Directions and Trends at NT10. Several speakers at the NT10 conference highlighted a growing trend of research in the nanoworld to use multiple techniques to characterize carbon nanotube samples. For example, two techniques might use two related scanning probe technologies, such as scanning tunneling spectroscopy (STS) and scanning tunneling microscopy (STM), but give different kinds of information (in this case, information on spectroscopy and visual information in the form of an image). Or one might use two techniques provided by a single instrument; for example, a high-resolution transmission electron microscope (HRTEM), might be used for imaging while also yielding related spectroscopic information about the sample through electron energy loss spectroscopy. Other joint studies, such as HRTEM and Raman spectroscopy, HRTEM and STM, and X-ray diffraction combined with Raman spectroscopy, were identified as fruitful directions for joint carbon nanotube studies. In fact, some speakers felt that the use of at least two different tools to characterize carbon nanotubes and other nanostructures was now becoming the trend for the reliable sample characterization of nanomaterials.

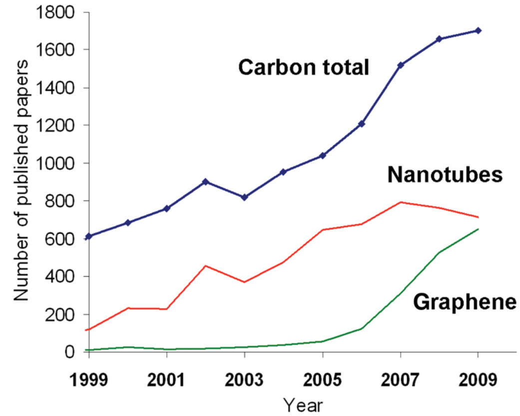

Figure 1. Attendance at international nanotube conferences. Plots of annual carbon nanotube and graphene published papers and their sum are included.

The most centrally recurring theme of the NT10 conference was nanotube growth, just as has occurred in the preceding 10 conferences in this series. Furthermore, major attention at NT10 was given to fundamental studies of growth mechanisms along with many more in-depth explorations into the specifics of various growth strategies. For example, Yoshikazu Homma[4] considered how a single-walled carbon nanotube grows from a nanoparticle and what role the catalyst plays in this growth process (Figure 2). Further reinforcement of the importance of the size of the catalytic particle was given

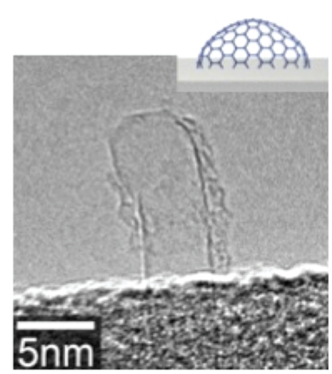

Figure 2. Carbon nanotube synthesis using giant fullerene cage as growth nucleus. Image courtesy of Yoshikazu Homma.

by Bilu Liu.[5] The fundamental issues focused on questions such as: What controls the nucleation of nanotube growth? What controls both the growth rate and the ultimate length of a nanotube? What controls the termination of nanotube growth? What is the role of the catalyst? How can nanotube nucleation and growth be optimized? And how can catalyst-free growth be promoted? There was increased emphasis at the conference on the use of *in situ* sampling, analysis, and characterization. The talk by Shigeo Maruyama emphasized the role of the carbon feedstocks, while the talk by Vincent Jourdain focused more attention on *in situ* characterization techniques.[6]

Challenges in Nanotube Chemistry. At each of the conferences in the series, the challenges of nanotube separation regarding metallic/semiconducting metallicity and (*n*,*m*) chirality have been highlighted. This year, the "light at the end of the tunnel" was emerging, with major progress being made in handling both of these significant and unique challenges for nanotube chemistry. Even though the density gradient nanotube sorting method has already been commercialized,[7] there continues to be a great deal of interest from an applications standpoint in developing new nanotube sorting schemes that work even more efficiently—faster and cheaper. However, Alan Windle stressed the importance of controlling chirality during the growth process itself by introducing specific chemical species, such as pyrozene, which could affect growth on a layer-by-layer basis in an interesting way.[8] At the same time, there remains great interest in the scientific community to address such fundamental issues as the role of catalytic particle size and catalyst faceting, and the more advanced analysis of nanotube cloning[9] and epitaxial growth approaches, some enabled by the present availability of both single species (*n*,*m*) samples, and others enabled by very heavily enriched metallic or semiconducting ensembles in nanotube samples.

Nanotubes and Graphene. Trends that we have seen at the NT conference series include the increasing interplay between nanotubes and graphene in the individual presentations and the increased presence of graphene-related talks. Efforts for promoting increased cross-fertilization between researchers working on graphene and carbon nanotubes, which have large intrinsic opportunities for overlap, need to be encouraged at future NTXX conferences.

At NT10, we heard a stimulating overview of recent advances in graphene physics from Andre Geim,[10] followed by reports on recent advances in the chemical vapor deposition (CVD) growth of graphene by Daiyu Kondo,[11,12] and a report on major advances in the photophysics of graphene by Tony Heinz.[13]

Several new topics attracted attention at the NT10 conference. One was a takeoff in the use of graphene as a substrate for studying nanotube photophysics. As an example, it was shown that the placement of carbon nanotubes on a graphene substrate resulted in a large enhancement of the Raman signal (Figure 3) through a so-called graphene-enhanced Raman spectroscopy (GERS) process,[14] related to surface-enhanced Raman scattering (SERS).[15] The SERS technique has been widely used in chemistry to enhance the spectroscopic signals of very small samples. For carbon nanotubes, SERS has had an especially long history, since the early SERS studies on carbon nanotubes stimulated the first work on single carbon nanotube spectroscopy,[16−18] and stimulated the development of resonance Raman spectroscopy as a powerful characterization tool for carbon nanotubes.[19] The increased sensitivity of GERS is likely to stimulate its future use as a sensitive spectroscopic tool.

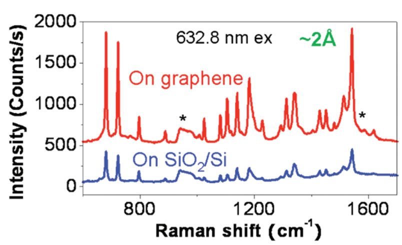

Figure 3. Comparison of Raman scattering in carbon nanotubes on a graphene substrate in comparison to a SiO_2/Si substrate. Reproduced from ref 14. Copyright 2010 American Chemical Society.

Carbon nanotubes on a graphene substrate result in a large enhancement of the Raman signal through a so-called graphene-enhanced Raman spectroscopy process. Synthesis will remain at the center stage for some time to come since the availability of the best possible materials leads to the best science.

Studying Specific Defects in Carbon Nanotubes. The detailed study of specific defects has for many decades played a central role in their exploitation in silicon and III−V compound semiconductor devices. For this reason, the theoretical report by Jean Christophe Charlier using *ab initio* techniques to study specific defects[20] was particularly welcome. This report distinguished specific defects from one another, such as (1) Stone−Wales defects, (2) tilted divacancies,[21] (3) defects with magnetic substitutional or interstitial atoms,[21] (4) decorated metallic clusters that modify the transport properties of carbon nanotubes, and (5) magnetic edge defects in carbon nanoribbons. The special effect of ozone as a defect for use in sensing was also highlighted in his talk. As another example, point defects and their photochemical effects were discussed by Philip Collins in his presentation. In the past, the Raman D band was often discussed in defect characterization studies, without reference to any particular defect type in the carbon nanotube. In this sense, the handling of defects in NT10 showed a significant advance relative to prior NTXX conferences.

Theory in Carbon Nanotube Research. Carbon nanotube research and nanoscience more generally have benefited over the years from the strong interaction between theorists and experimentalists. In this context, the talks by the theorists at the NT10 conference were especially informative.

Single-wall carbon nanotubes were predicted before they were observed experimentally.[22] An effort is needed to again promote increased interaction between theorists with experimentalists at nanotube conferences to advance the nanotube field more effectively for mutual benefit. The eventual exploitation of specific defects in nanotube- and graphene-based devices may be a future outcome of such joint research.

We have already seen at NT10 the use of strain and applied gate voltage to provide a forceful method for controlling the performance of electronic devices.[23] The advances made with carbon nanotube electronics and macroelectronics as reported by Chongwu Zhou were impressive, especially regarding wafer-scale fabrication of nanotube devices in operation.[24]

Paralleling these advances was the progress made with nanophotonics and optoelectronics, with the first demonstration of optical gain in carbon nanotubes.[25] This observation was made possible by the availability of samples with a high concentration of (8,6) large-chiral-angle nanotubes. The successful observation of optical gain in nanotubes can be interpreted as a precursor to the demonstration of a nanotube-based laser. In general, NT10 saw more activity in the involvement of fast optics to study relaxation processes in carbon nanotubes.

The NT10 conference showed several examples of new physical phenomena based on carbon nanotubes. Multiple-exciton generation, which recently has been widely discussed in the context of solar-energy harvesting, was discussed by Paul McEuen as a future direction for photophysics. He demonstrated this phenomenon in the context of carbon nanotubes.[26] Although Raman scattering by lattice vibrations has been well studied in carbon nanomaterials, Raman scattering by electronic excitations in carbon nanotubes was reported for the first time at NT10 prior to actual publication of this work. Previously unobserved, a broad Raman feature attributed to electronic Raman scattering (ERS) was found exclusively in Raman spectra of metallic carbon nanotubes but was absent in the spectra for semiconducting tubes. The position of the ERS peak depends on the laser energy as well as on the electronic structure of the nanotube; consequently, it can be used for identifying the optical resonances of metallic nanotubes. This technique also has the potential of being used to probe the low-energy electronic structure of metallic carbon nanotubes and is (n,m) dependent.[27]

Also reported at NT10 was a status report by Riichiro Saito on the exciton–phonon interaction and environmental effects on the Raman spectra of carbon nanotubes, which now explain the most recent experimental results by Araujo *et al.*[28] for excitonic transitions occurring in many carbon nanotubes.

Progress made in the knowledge about Raman resonance windows and resonance profiles in resonance Raman spectroscopy was nicely discussed by Stephen Doorn[29] and Anna Swan[30,31] in their talks. The use of Raman spectroscopy to identify armchair and zigzag edges in graphene ribbons and how to distinguish between them was also discussed by Gustavo Cancado.[32]

Nanotube Applications and Commercialization. NT10 showed a major increase in emphasis on applications, reflecting trends in both publications and funding initiatives. In fact, the NT10 conference opened with a session on applications, starting with a talk by Ray Baughman on artificial carbon-nanotube-based muscles. This was followed by several talks on composites and talks about large-scale production of carbon nanotubes in Japan. The talks by Daniel Resasco from the United States and Y. A. Kim from Japan at the end of the MSIN10 satellite conference on real commercial-scale products based on carbon nanotubes made many conference attendees more aware of the commercialization status of carbon nanotubes. Conference attendees of MSIN10 also heard reports of even larger scale efforts on multiwall carbon nanotube synthesis programs in Souzhou, China, for use in the production of polymer/nanotube composite materials.

Future of Nanotube Research. Several topics that are likely to receive more attention in the future can already be identified. Discussions of newly emerging topics are valuable to young people looking for new research directions for thesis work, postdoctoral studies, and starting independent careers. It should be emphasized that the specific topics given here were not suggested by any NT10 committee, and other people would have differing suggestions for open research opportunities in carbon nanotubes. Synthesis will remain at center stage for some time to come since the availability of the best possible materials leads to the best science. Most researchers believe that many opportunities to enhance nanotube synthesis outcomes still remain. New characterization tools and approaches will continue to be essential and should have significant impacts on progress in the field when applied to the advanced samples now becoming available. The commercial success of our science will surely incentivize researchers to work in the carbon nanotube field, in addition to the fascination with carbon nanotube science that brings new researchers to work in this field in the first place.

New characterization tools and approaches will continue to be essential.

To conclude this Nano Focus, I mention three topics that I find exciting as a principal investigator: double-wall nanotubes, graphene nanoribbons, and magnetic phenomena in carbon nanotubes. One reason why researchers should all have some interest in the topic of double-wall carbon nanotubes is that multiwall nanotubes (MWNTs)

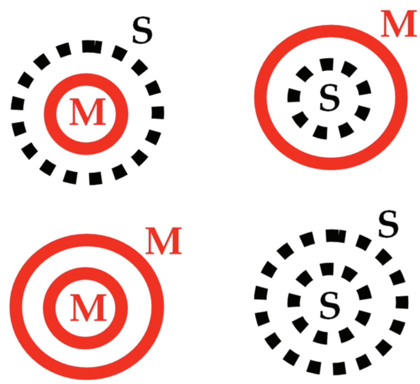

Figure 4. Four possible configurations of metallic and semiconducting tubes within a double-wall carbon nanotube.

are becoming a major commercial product today, and understanding the interactions that govern multiwall tubes can best be accessed through the study of double-wall nanotubes (DWNTs)—the simplest version of a MWNT. More explicitly, DWNTs come in four varieties of metallicity: S@S, S@M, M@S, and M@M (Figure 4). The intertube interactions are complex because of the lack of commensurability between the inner and outer tube walls, so that the property of any DWNT by itself varies upon rotation and translation of the inner tube relative to the outer tube. Two basic methods are presently used for the growth of DWNTs: CVD and peapods (a single-wall nanotube filled with fullerene molecules such as C_{60}). When researchers feel that the direction they are pursuing with SWNTs is getting saturated, they may find exciting research opportunities by extending their present research skills to DWNTs. Once researchers enter the field of DWNTs, they will soon see the complexity of both the structure and the properties of DWNTs.[33,34] When studying SWNTs, we have the notion that a given (*n*,*m*) nanotube is unique and has unique properties once environmental effects are accounted for (*e.g.*, freely suspended tubes under vacuum conditions). It was once thought that the placement of a nanotube inside another nanotube eliminated the effect of both supporting posts and the strain associated with free suspension, but Raman measurements on the same (6,5) nanotube inside a common outer tube[34] show spectral differences arising from the intertube rotation and translation effects within a DWNT with given (*n*,*m*) and (*n*′,*m*′) inner and outer tubes, respectively. These considerations complicate the study of fundamental issues, such as the dependence of intertube metallicity on the physical properties of DWNTs.

A second topic demanding further attention is the commonality between graphene nanoribbons and carbon nanotubes because both are 1D all-carbon nanostructures. A carbon nanotube can be cut to form a graphene nanoribbon, and the inverse operation can also be accomplished in some cases. Studies of this commonality may be interesting for providing a better understanding of curvature-related effects in nanostructures.

Magnetic phenomena occur at nanoribbon edges through divacancies of proper orientation and at magnetic impurities in graphene, as discussed above. Some interesting relations between the spin behaviors may occur for these different systems. In general, the spin and spin−orbit interaction in carbon systems have not been studied much in the past because of the low atomic number of six for carbon. The magnetic effects reported at NT10 bring new importance to spin in carbon systems. Raman scattering and transmission electron microscope (TEM) measurements have both successfully been used to distinguish zigzag and armchair edges from one another. Can anything special be learned from a joint Raman/TEM study of such edges?

Looking to the future, we see that carbon nanotube researchers are beginning to leave the field (Figure 1), looking for greener pastures in graphene or in the applications arena of nanotubes or elsewhere, while others (*e.g.*, new graduate students) are still entering the carbon nanotube field. Although the field is becoming mature, researchers are still finding topics of interest and new physical phenomena that are nicely revealed in the context of carbon nanotubes. Because of the interest in better synthesis methods regarding control of (*n*,*m*), metallicity, tube length, and tube quality, studies of the control of the synthesis process and studies of the mechanisms for carbon nanotube synthesis will remain at the forefront for some time to come. As always, the best materials often enable surprising scientific advances, and for this reason, we can expect continued emphasis on the growth area. Outreach of nanotubes to other fields of science remains at an early stage as mentioned above, with research opportunities for the future.

Joint technique studies such as TEM/Raman are sure to continue with emphasis given to measurements on advanced samples and joint *in situ* measurements. More time-resolved studies using shorter light pulses as a function of gate voltages and controlled strain are expected. The increased attention to nanoscience by policy regulators is likely to increase public interest in this topic, whereas increased concerns about health effects are likely to increase the testing of nanomaterials under environmental conditions outside the normal operating range. Close communication between researchers, health officials, regulators, and the public therefore needs to be promoted so that an appropriate balance between the advance of nanoscience can proceed while public interests are well protected.

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