Physics and applications of nanotubes

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ABSTRACT

Nanotubes have been pursued aggressively over the last three decades. Significant progress has been made in the selective growth and post-synthetic sorting of highly monodisperse carbon nanotubes, in understanding their physics, and in assembling and integrating them into high-performance devices. These discoveries have led to promising applications in areas such as high-performance CMOS, high-speed RF, thin-film transistors, flexible electronics, thermoelectrics, sensors, and optoelectronics. The rapid development of modern information technology depends on the exploitation of new and novel materials, and nanotubes have emerged as promising candidates for the post-Moore's Law era. This Special Topic on Physics and Applications of Nanotubes provides a valuable forum where researchers studying the fundamentals of nanotubes can share their most recent and novel findings.

I. INTRODUCTION

Nanotubes have been pursued aggressively over the last three decades since the commemorative paper[1] by Iijima in 1991. Particularly, single-walled carbon nanotubes (SWCNTs) with diverse optical and electronic properties (metallic or semiconducting) depending on chiral indices (n, m) have been the remarkable material for the challenge in synthesis, basic physics of one-dimensional material, and optoelectronic devices such as field-effect transistors (FETs) during 1990s.[2] During the 2000s, larger-scale synthesis and the separation of metallic and semiconducting, one-dimensional excitonic features are developed.[3] Later in the 2010s, chirality selective growth, practical application design of FET, and practical application of transparent conductive films for solar cells are proposed.[4] The diverse growth of the field was backed up by continuous progress in the physics of one-dimensional materials and optical spectroscopies such as Raman spectroscopy.[5] Selective growth and post-synthetic sorting of highly monodisperse carbon nanotubes led to promising applications in areas such as high-performance CMOS,[6,7] high-speed RF, thin-film transistors, flexible electronics, thermoelectric, sensors, optoelectronics, and photovoltaics.[8] The rapid development of modern information technology depends on the exploitation of new and novel materials, and nanotubes have emerged as promising candidates for the post-Moore's Law era. On the other hand, in response to the social needs of carbon neutrality, carbon nanotubes are playing important roles in energy harvesting devices such as solar cells[8] and thermoelectrics.[9]

The Special Topic "Physics and Applications of Nanotubes" in *Journal of Applied Physics* provides a valuable forum where researchers studying the fundamentals of nanotubes can share their most recent and novel findings for the rapid progress in the next decade.

II. SYNTHESIS AND PREPARATION

A. CVD growth and sorting of nanotubes

In the chemical vapor deposition (CVD) growth of carbon nanotubes (CNTs), the control of catalytic metal nanoparticles and chemical reaction pathways are a central research topic for decades. Present interests focus on chirality-specific growth[10] and the *in situ* studies for the growth mechanism. In general, supported catalyst CVD is preferred for chirality-specified and aligned growth, and floated catalyst CVD is preferred for mass production. For the floated catalyst CVD, first principles nonequilibrium molecular dynamics (MD) simulations are employed for the discussion of the decomposition of ferrocene, Fe metal formation, and carbon chain formation processes.[11] Optimized bimetallic catalysts are employed for chirality selective growth[10] and efficient mass production[12] of nanotubes.

DNA-wrapping, polymer-wrapping, density gradient ultracentrifugation (DGU), gel chromatography, and aqueous two-phase (ATP) techniques have been developed for sorting SWCNTs.[13] The focus of sorting shifts from the separation of semiconducting and metallic nanotubes to those of the chiralities and enantiomers. ATP sorted single chirality SWCNTs can be directly placed on the prepatterned electrode by dielectrophoresis.[14]

B. DWCNTs, heteronanotubes, and modified SWCNTs

The controlled modulation of nanotube properties can be possible either by employing the inner space of the SWCNTs to encapsulate various materials or by externally wrapping the SWCNT template with additional atomic layers.[15] For the latter case, concentric growth of BNNT and/or MoS_2 nanotubes on SWCNT are proposed.[16] The possibility of bulk synthesis and dispersion of SWCNT@BNNT, where @ denotes inner@outer, is demonstrated.[17] All these inner/outer modifications can be defined as one-dimensional (1D) van der Waals (vdW) heterostructures based on SWCNTs.[15]

Double-walled carbon nanotubes (DWCNTs) are a good example of a 1D vdW structure based on SWCNTs. Due to progress in inner- and outer-wall sorting,[18] DWCNTs are now available in the four possible combinations of metallic and semiconducting shells (M@M, M@S, S@M, S@S), which differ in their electrical, optical and chemical properties. In practice, an unsorted DWCNT film is demonstrated as an active electrode in an electro-optical modulator for the mid-infrared and terahertz regions[19] based on the concept that the vast majority of nanotubes in a DWCNT have at least a wall with the semiconducting character and that doping of large diameter outer tube with smaller bandgap requires a lower applied potential.

Linear carbon chains (LCCs) formed inside the carbon nanotubes by high-temperature annealing in vacuum or inert gas is an example of inner space modification of nanotubes. Vacuum condition is preferred for the high yield[20] formation of LCCs. The inner hollow core of a nanotube can accommodate many types of molecules,[15] such as fullerene, water, dye, DNA, etc. Dye-encapsulated SWCNTs (dye@SWCNT) can work as unique and efficient photocatalysts for H_2 evolution reaction.[21] Orientation or stacking of molecules inside a nanotube can be predicted by newly developed functional Lennard-Jones potential.[22]

Chemical modification of carbon nanotubes by defects (oxygen or sp^3 carbon) has been intensively studied because of the dramatic modification of photoluminescence with the localized exciton state at the defect site.[23] Electroluminescence from such defects or organic color-centers is demonstrated.[24] Tuning of spin–orbit coupling by introducing a heavy atom in the defect was examined by density functional theory (DFT).[25]

C. Assembled material of nanotubes

A film or a fiber of SWCNTs or composites are expected in various commercial applications.[26] The film can be prepared by dry deposition from floated catalyst CVD, spun from vertically aligned nanotubes, or filtration of suspended nanotubes. Particularly, a film by dry deposition attracts attention for the transparent conductive electrode. Densification and/or doping of films or fibers are the typical way to manipulate the electronic, optical, thermal, and mechanical properties. Modeling by the mesoscopic distinct element method of bundle network and experimental optical properties are compared for the densification process.[27] Another large-scale dynamics mesoscopic model is employed for the mechanical properties of a film with covalent cross-links.[28] On the other hand, the dependence of the electrical conductivity on the volume of the nanotube filler was discussed by the theory of continuum percolation.[29] The in-plane electromagnetic response and the exciton–plasmon interactions for parallel aligned semiconducting CNT arrays embedded in an ultrathin finite-thickness dielectric were theoretically predicted for the modification of optical properties.[30]

III. CHARACTERIZATION

A. Optical spectroscopy and optical characterization

Because of their unique one-dimensionality combined with structure-related electronic and optical properties, SWCNTs exhibit rich physics in optical spectroscopy. Typical spectroscopies, such as absorption/Rayleigh scattering, photoluminescence spectroscopy, and resonance Raman spectroscopy,[5] have been developed for probing and modulating excitonic features of SWCNTs.[31,32] To probe the unique features of individual SWCNTs, air-suspended, solution dispersed, or individually immobilized SWCNTs are employed. Temperature-dependent photoluminescence of suspended SWCNTs is employed to examine the structure of the water adsorption layer.[33] Single-molecule spectroscopy can probe individual sodium-dodecyl-sulfate (SDS) wrapped SWCNTs on a polymer-coated substrate. Photoluminescence brightening by the chemical reductant dithiothreitol (DTT) exhibits unexpected uniform brightening along a tube.[34] The diffusion trajectory of individual SWCNTs in a solution can be used to estimate the length distribution of nanotubes.[35] An alternative way is demonstrated with polyvinyl acetate (PVAc)-based SWCNT-polymer films for optical and far-infrared spectroscopy in cryogenic temperatures.[36] The optical absorption and circular dichroism (CD) spectra for doped SWCNTs as functions of the Fermi energy are predicted. The calculated CD values for the doped nanotubes are much larger than the CD for the undoped one because of the enhancement of the surface plasmon and the phase shift effect.[37]

B. Advanced characterizations

It is a challenging task to quantify intrinsic mechanical properties such as Young's modulus, tensile strength, and strain to failure of individual CNTs. Transverse loading is a common setup where the deflection curve can be measured to analyze the strain–stress relation. Assessments are proposed based on fully nonlinear, extensible Kirchhoff rod theory.[38] To increase the nominal tensile strength and Young's modulus of MWCNTs, carbon nanofibers (CNFs) are fabricated by epitaxial growth on MWCNTs.[39] Related to the heat conduction of SWCNT, localization of high-energy optical phonon by ^{13}C isotope is predicted.[40] Fast diffusion of gas molecules within CNTs predicted by MD simulations are examined in statistical self-diffusion of highly rarefied and only specular particle-wall reflections.[41]

IV. DEVICES AND APPLICATIONS

A. Field-effect transistors

The fabrication of logic integrated circuits (ICs) beyond Si or Ge has been the most expected and the most challenging application of CNTs for these two decades. It requires high-density arrays of exclusively semiconducting species. To fulfill the requirements for digital ICs,[42] significant progress has been made in chirality sorting in nanotube dispersions and the subsequent assembly,[6,43–45] as well as the selective removal of metallic species from aligned nanotubes grown on crystalline substrates.[46]

Methods to dope the CNTs need to be developed to achieve low parasitic resistance of the transistor. The doping mechanism of MoO_x on the CNT is proposed as bandgap modulation by charge transfer, which occurs due to the difference in work function between MoO_x and the CNT.[47] A clean and curved DWCNT structure with a definite curvature radius was employed to demonstrate that the curved structure will limit the on/off ratio by orders of magnitude but can maintain a high on-current.[48]

Single CNT FET configuration is indispensable for sensitive biosensors. By decorating the CNT surface with a peptide tag, a highly sensitive biosensor for protein analysis can be possible.[49]

B. Solar cells

CNTs are promising materials to be incorporated into thin-film solar cells owing to a wide range of properties ranging from conductors to semiconductors with different bandgaps based on their atomic structure. Films of SWCNTs or DWCNTs exhibit high transparency and conductivity. SWCNTs have already been exploited as electrodes in many electronic devices, replacing both brittle oxide transparent electrodes, for example, indium-doped tin oxide (ITO) and fluorine-doped tin oxide (FTO), and expensive metal electrodes.[50] CNT film has been employed in CNT-Si heterojunction solar cells as transparent conductive hole transport layers.[8,50] Recently, CNT as hole selective back contact of Si solar cells proves industrial level power conversion efficiency.[51] For an organic thin-film or metal-halide perovskite solar cells, CNT film is promising transparent hole transport electrodes.[8,50,51] Doping of CNT has been a critical and controversial issue for all those solar cells. For example, the doping mechanism of CNT-Si heterojunction solar cells is still being explored because of the interplay with the surface reaction on Si substrate.[52]

The most expected model solar cells with CNTs are semiconducting SWCNT-fullerene heterojunction solar cells.[53] The exciton dissociation at the donor (s-SWCNT)–acceptor (fullerene) interface drives solar energy conversion. The time-dependent tight-binding density functional theory (TB-DFT) combined with nonadiabatic MD can demonstrate photoinduced electron transfer and photoinduced hole transfer processes.[54]

C. Thermoelectric devices

Thermoelectric power generation, allowing recovery of part of the energy wasted as heat, is emerging as an important component of renewable energy and energy efficiency requirements. There are focused studies on thermoelectrics of SWCNT because very large Seebeck coefficients are expected for the 1D density of states (DOS) of SWCNTs.[55] However, the high thermal conductivity of SWCNT that is a direct disadvantage in the figure of merit of thermoelectric and requirements of chirality selected semiconducting SWCNT and adequate doping have decelerated the research activities. Recently, the thermoelectric application is intensively revisited with chirality sorted SWCNTs, doping technique, and the possible reduction of phonon transport of semiconducting SWCNT films.[5,53,56]

Electric double-layer capacitor structure with ionic liquid can be used as p-type and n-type pairs with controlled carrier density by gate voltage.[57] A very high power factor is predicted for fluorine-doped SWCNTs, which have a semiconducting nature by doping armchair SWCNT with fluorine.[58] Low-cost CNTs with lower thermal conductivity from oil fly ash are tested as thermoelectric material.[59]

V. CONCLUSIONS

The "Physics and Applications of Nanotubes" Special Topic in *Journal of Applied Physics* is a good landmark as the 30th anniversary of carbon nanotube research. The preparation of materials has been advanced, but still we have new challenges for chirality pure materials, sp^3 functionalized nanotubes, van der Waals heteronanotubes, etc. Carbon nanotubes are now a commodity in some applications. At the same time, emerging technologies in the era of information and global energy challenge are just in front of us.

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