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One-dimensional Crystals inside Single-walled Carbon Nanotubes: Growth, Structure and Electronic Properties

Andrei Eliseev1, Lada Yashina2,3, Marianna Kharlamova1 and Nikolay Kiselev4

1Moscow State University, Department of Materials Science
2Moscow State University, Department of Chemistry
3Rare Metals Institute “GIREDMET”
4Institute of Crystallography RAS

Russia

1. Introduction

Single-walled carbon nanotubes (SWNTs) discovered in 1993 are currently among the most exciting and promising nanostructures (Bethune et al., 1993; Iijima & Ichihashi, 1993). They arouse huge interest due to their unique atomic structure, outstanding chemical and electronic properties (thermal and electric conductivity), as well as mechanical characteristics (high values of Young’s modulus, tensile and compressive strengths, high cracking resistance etc.). SWNTs possess the maximum geometric anisotropy factors among the nanostructures known so far. The unique properties of carbon nanotubes (CNTs) are governed not only by their unusual tubular structure, but also by the fact that they are virtually devoid of any structural defects (Dresselhaus et al., 1995; Iijima, 1991; Saito et al., 1992). As a result, CNTs are of a great importance for development of nanoelectronics elements (logical gates, memory devices, emitters, and nanowires), nanoelectromechanical systems, nanocomposite fillers (aimed at increasing strength and functionality of bulk materials), probe tips for scanning probe microscopy etc. One of the major areas of SWNTs technological application has been the development of a new generation of field-effect transistors (Tans et al., 1998).

The electronic properties of defect-free SWNTs are extremely sensitive to the nanotube’s geometric structure (Avouris et al., 2007; Saito et al., 1992), which depends to a great extent on the chiral vector; this may be regarded both as an advantage and a serious drawback of this material. So far, no efficient methods have been developed for the preparation and isolation of SWNTs with a desired chirality (Hou et al., 2008; Odom et al., 2000). For this reason, many attempts have been undertaken to develop methods that would allow separating the array of SWNTs into semiconducting and metallic nanotubes and/or modify the electronic properties of SWNTs without their separation by chirality (Chaturvedi et al., 2008; Eliseev et al., 2009a; Monthioux et al., 2006).

Modification of nanotubes allows direct adjustment of their electronic properties. One of the simplest ways to controlled modification of the SWNTs is filling of the nanotube channels...
with appropriate substances (Brown et al., 2001). Encapsulation of a substance into a nanotube can either lead to a complete amendment of the nanotube’s band structure (in case the encapsulated substance interacts intensively with the nanotube walls, e.g. in fluorinated SWNTs), or only to a shift of the electron density within the rigid band structure approximation (Sceats et al., 2006; Sloan et al., 2002a). In the simplest case, if an electron donor with the Fermi level located higher than that of the SWNT is encapsulated into metallic nanotubes, the electron density at the nanotube walls, as well as the nanomcomposite conductivity increase, whereas an electron acceptor with the Fermi level located lower than that of the SWNT would cause the nanocomposite transition into the semiconducting state (Chaturvedi et al., 2008; Rahman et al., 2005; Weissmann et al., 2006). Therefore, this approach based on electron transfer upon the introduction of electron-donor or electron-acceptor compounds (metals, semiconductors, dielectrics) into the channels of single-walled nanotubes allows controlling the electronic structure of the SWNTs, as well as creating the $p-n$-junctions inside a single nanotube if the channels are partially filled (e.g. if a nanotube is half-filled).

The synthesis of filled nanotubes was first reported by Ajayan and Iijima in 1993; they used multi-walled nanotubes as “molecular containers” for lead (Ajayan & Iijima, 1993). These experimental results confirmed the theory-based conclusions on the existence of sufficiently strong capillary forces inside carbon nanotubes, which may retain gases and liquids inside the channels (Pederson & Broughton, 1992). Later on, other researchers developed and employed this approach for filling carbon nanotubes with a variety of metal halides \( M \text{Cl}_x \) (\( M = Li, Na, K, Cs, Rb, Ag \)), \( \text{M}_2 \text{Cl}_2 \) (\( M = Ca, Cd, Co, Sr, Ba, Fe, Pb, Hg \)), \( \text{M}_3 \text{Cl}_3 \) (\( M = La, Ce, Pr, Nd, Gd, \) (Te/Sn))\( _8 \), \( \text{Al}_2 \text{Cl}_6 \), \( \text{AgCl}_3 \text{Br}_7 \text{I}_9 \) or \( \text{M}_4 \text{Cl}_6 \) (\( M = Na, Cs, Ti \)), \( \text{MoO}_4 \text{Cl}_2 \) (\( M = Cd, Fe, Co, Pd \)), \( \text{M}_3 \text{Cl}_3 \) (\( M = Na, Sm, Eu, Gd, Tb, \) \( \text{MoCl}_4 \) (\( M = Hf, Th, Zr, Pt, \) \( \text{Al}_2 \text{Cl}_6 \) (\( \text{Th} / \text{V} \text{Cl}_4 \)), elemental forms \( (S, Se, \text{Te}, \text{I}_2, \text{Cs}, \text{Re}, \text{Bi}, \text{Pt}, \text{Au}, \text{Ru}, \text{Fe}, \text{Ag}) \), fullerences \( (\text{C}_{20}, \text{C}_{70}, \text{C}_{80}) \), endofullerenes \( (\text{Gd} @ \text{C}_{25}) \), a \( (\text{KCl})_x(\text{UCl}_4)_y \), oxides \( (\text{ReO}_3, \text{V}_2\text{O}_5, \text{Sb}_2\text{O}_5, \text{CrO}_3, \text{PbO}, \text{UO}_2, \text{ZrO}_2, \text{MoO}_2, \text{NiO}, \text{C}O, \text{La}_2\text{O}_3) \), metals \( (\text{Pd}, \text{Pt}, \text{Cu}, \text{Ag}, \text{Au}) \), hydroxides \( (\text{KOH}, \text{CsOH}) \), and chalcogenides \( (\text{SnSe}, \text{HgTe} \text{and C}_x\text{Br}_y\text{Te}_z) \) (Chaturvedi et al., 2008; Cohen, 2001; Corio et al., 2004; Eliseev et al., 2009a; Fagan et al., 2005; Govindaraj et al., 2000a; Kataura et al., 2002; Monthioux, 2002; Monthioux et al., 2006; Sceats et al., 2006; Sloan et al., 2000a).

At present, several methods are used for filling carbon nanotubes with various substances, which fall into two large groups: filling of nanotubes during their growth (i.e. the \textit{in situ} methods) and encapsulation from the gas or liquid phases into cavities of pre-formed carbon nanotubes (i.e. the \textit{ex situ} methods) (Monthioux et al., 2006).

2. Filling of single-walled carbon nanotubes during their growth (\textit{in situ} methods)

The simplest of all the approaches that have been proposed to date for the nanotubes encapsulation is filling of SWNTs in the course of their catalytic growth (\textit{in situ}). Currently two methods are applied that employ the \textit{in situ} strategy for the encapsulation of inorganic compounds into the nanotubes: catalytic chemical vapour deposition (CCVD) of hydrocarbons and arc-discharge synthesis (Monthioux et al., 2006).

Arc-discharge synthesis of carbon nanotubes filled with various compounds is performed using graphite rods electrodes, a compound-containing anode (usually metals are encapsulated using this approach), and a catalyst. This approach was used to prepare single-walled carbon nanotubes for the first time (Bethune et al., 1993; Iijima & Ichihashi, 2001).
Electronic Properties of Carbon Nanotubes


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Carbon nanotubes (CNTs), discovered in 1991, have been a subject of intensive research for a wide range of applications. These one-dimensional (1D) graphene sheets rolled into a tubular form have been the target of many researchers around the world. This book concentrates on the semiconductor physics of carbon nanotubes, it brings unique insight into the phenomena encountered in the electronic structure when operating with carbon nanotubes. This book also presents to reader useful information on the fabrication and applications of these outstanding materials. The main objective of this book is to give in-depth understanding of the physics and electronic structure of carbon nanotubes. Readers of this book should have a strong background on physical electronics and semiconductor device physics. This book first discusses fabrication techniques followed by an analysis on the physical properties of carbon nanotubes, including density of states and electronic structures. Ultimately, the book pursues a significant amount of work in the industry applications of carbon nanotubes.

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