ATOMISTIC MODELS FOR NANOTUBE DEVICE ELECTROSTATICS

Slava V. Rotkin*, Kirill A. Bulashevich†, Narayan R. Aluru
Beckman Institute, UIUC, 405 N.Mathews, Urbana, IL 61801, USA
E–mail: rotkin@uiuc.edu

A quantum capacitance is calculated for a single–wall carbon nanotube in a field effect device. The calculation is performed on a base of a continuum model taking into account full microscopics. A general expression for the atomistic capacitance of a nanotube of arbitrary shape is derived. The result is useful for modeling of electromechanical action of a nanotube device and for nanotube electronics simulation.

I. INTRODUCTION

Carbon nanotubes are natural nanoscale objects with sizes $\sim$ 1–50 nm which fit anticipations for the next generation of electronics. Their potential promise for electromechanical engineering, molecular and bio–electronics makes a fundamental research in this field important for applications. The single–wall nanotubes (SWNTs) are known to have various electronic properties. The metallic nanotubes, degeneratively doped and intrinsic semiconductor nanotubes were observed experimentally [1]. Fundamentals of an electronic structure of an ideal SWNT were understood but no developed device theory exists, similar to what does exist for standard semiconductor materials, *e.g.*, the silicon.

We aim our theory to calculating basic electrostatics and electronic properties of nanotubes. The microscopic model for that is proposed and successfully applied to calculate the electrostatic forces. Recent success in a fabrication of nanotube based nanoelectromechanical devices [2,3] confirms that our theoretical research is very topical. First demonstrations of carbon nanotube electronics were recently given by Avouris et.al. [4] and Dekker et.al. [5]. As the field of applied physics grows, the basic theory and compact modeling of nanotube devices becomes needed. We consider this paper as a first step towards complete nanotube device theory.

II. BASIC MODEL OF SWNT ELECTROSTATICS

In the paper we compute the electronic structure of a realistic device: a carbon nanotube of a finite length in a conductor environment (gates and contacts). The selfconsistent calculation of the equilibrium charge density for a SWNT with a moderate mechanical deformation has been performed in relation to recent modeling of nanotube electromechanical systems [6–8]. A knowledge of the induced charge allows

---

*On leave from Ioffe Institute, 26 Politekhnicheskaya st., St.Petersburg 194021, Russia

†Permanent address: St. Petersburg State Technical University, 21 Politekhnicheskaya, St.Petersburg 195251, Russia

us calculating an electrostatic energy of the system, which can be rewritten in terms of a distributed capacitance. We demonstrated that this capacitance has two contributions: purely geometrical term and another one, specific for the nanotube. It is very natural to call the second term "a quantum capacitance" as a similar definition was proposed for a two–dimensional electron gas system in [9].

The quantum mechanical modeling of the SWNT electronic structure may be readily obtained within a tight–binding approximation (TBA). In principle, the full quantum mechanical result provides a single nanotube polarizability, which is an atomistic analog of the bulk dielectric function. It contains complete information on the electronic structure and charge distribution. However, a selfconsistent calculation of the polarizability is tricky and we prove here that a classical continuum approach works well for calculating the charge distribution when the atomistic capacitance is used instead of a classical expression. This brings the quantum correction term into the solution.

The results of our compact model, for example, the analytical approximation for atomistic capacitance, are valid for a moderate device operation voltage. The criterion for the applicability is a small perturbation of the original electronic structure of the SWNT (rigid band approximation). Our analytical expression can be used for a moderately mechanically deformed nanotube. In this case, the geometric curvature of the nanotube axis has to be small as discussed in [10]. These conditions are not too restrictive and allow a wide use of our compact model for many of existing nanotube devices. The calculation of the nanotube charge distribution allows us to find an electrostatic energy for the electromechanical analysis, a Drude conductivity (to be discussed elsewhere), and other device properties requiring knowledge of Coulomb forces.

A. Principles of Compact Model for Nanotube Devices

A nanotube capacitance defines a relation between the applied voltage and induced charge density. The capacitance will be shown to contain two contributions: a purely geometrical capacitance (of the metal cylinder of the same size and shape) and a microscopic correction. When only one "metallic" subband of SWNT is populated, the quantum term is given by a dimensionless parameter: the nanotube density of states times the elementary charge squared. For a metallic SWNT in the low voltage regime, it corresponds also to a ratio of the potential energy to the kinetic energy of a nanotube valence band electron, which is an analog of the electron gas parameter, widely used in condensed matter theory.

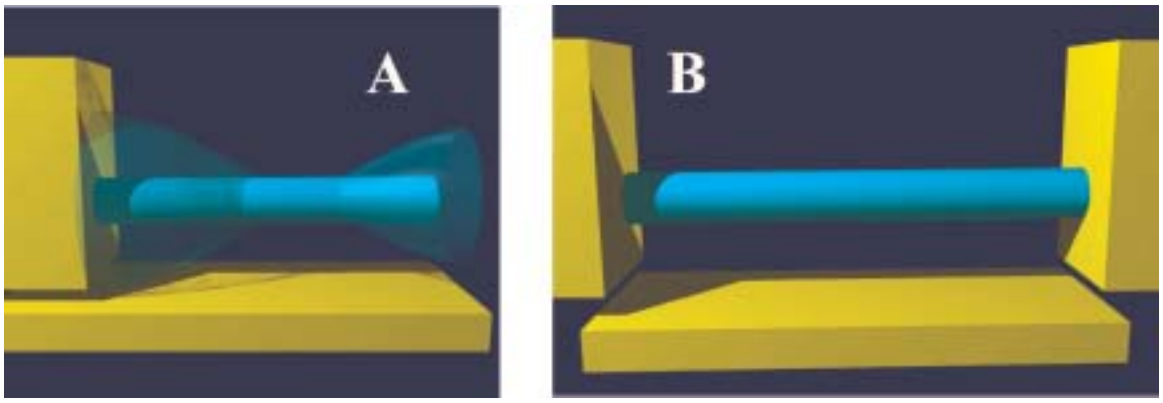

FIG. 1. Geometry of two single–wall nanotube devices studied in the paper: (A) cantilever NEMS, and (B) fixed–fixed NEMS.

It was found that the statistical description (similar to what was used first in Ref. [11]) is valid and gives a fairly good estimate as compared to full quantum mechanics. The reason for this "classical" behavior of so small quantum object is that the selfconsistent electrostatic energy level shift is the same for every subband of the SWNT in a first approximation [12]. The SWNT has a very high depolarization factor as it was first noted by Louie [13]. Hence, one is allowed to apply a "classical" theory, taking into account the quantum mechanical modification of the electronic structure in a selfconsistent way.

We studied a SWNT device which comprises the straight nanotube fixed (suspended without a slack) between two metal side electrodes over a back–gate electrode (Fig.1A). The side electrodes are kept at the same potential with respect to the back–gate. This design is standard for electromechanical systems, and the first experimental realization of a single SWNT NEMS appeared recently [3]. We used this and a cantilever geometry (Fig.1B) for theoretical study of the nanotube electromechanical switch in [6].

The aim of this paper is to find a selfconsistent solution for a nanotube charge density as a function of applied voltage. For this purpose, one may use a quasi–static (low frequency) limit of an electrodynamical response function which relates the induced charge density to an a.c. external potential. However, we are not aware about any theoretical calculation of that type which included both nanotube electronic structure and nanotube environment selfconsistently. We propose to use rather simple phenomenological continuum model instead of full quantum mechanics. The latter is more accurate but also computationaly demanding. We performed full quantum mechanical computation and conclude that in case of electromechanical device our compact model gives an adequate solution. We already discussed the applicability of the continuum electrostatics modeling in [10] with respect to an equilibrium charge distribution. Same arguments may hold for a system shifted from an equilibrium slightly, *i.e.*, for current carrying device (which description is beyond the article limits).

The continual compact modelling of the nanotube device bases on three elements: (i) local statistical description of the charge density, (ii) perturbation theory for changes in the charge density due to nanotube deformations and(or) external fields, and (iii) external screening which results in a short range Coulomb potential and allows obtaining analytical expressions. Hypothesis (i) has been proved by comparing

the result of full quantum mechanics (TBA for pi–electrons using a scheme similar to what was used in [14]) with the selfconsistent solution of the Poisson–Boltzmann equations (see also [10] and [12]). Second supposition is valid until the deformation or(and) axial component of the external field is not too large, which is true for SWNT applications in nanoelectronics but may not fulfill for nano–actuators. We consider third element of the compact model to be most important. We demonstrated that for the nanotube, which is a one dimensional nanoscale system, it is not possible to separate pure material properties and effects due to geometry/design of the device. The electronic properties of nanotube–in–device differ from what one obtains for a free tube in vacuum.

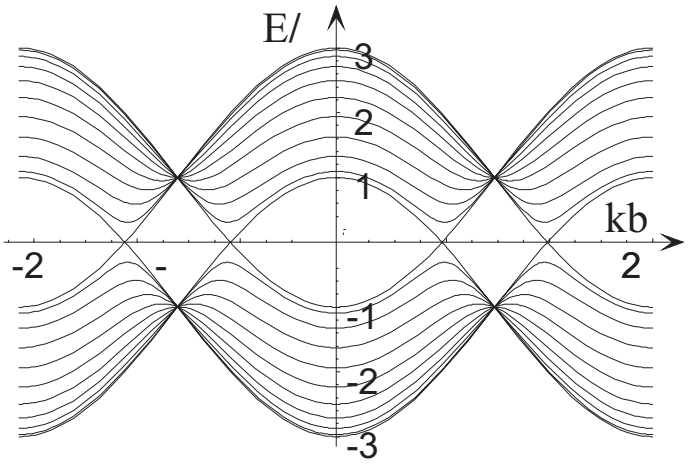

FIG. 2. Dispersion curves for electronic structure of an armchair SWNT [10,10] within first Brillouin zone.

In the compact modeling of the armchair SWNT we used the knowledge of the quantum mechanical electronic structure. This one–dimensional electronic system is characterized by energy subbands labeled with two quantum numbers k and m, which are the wavevector of the electron along the nanotube axis and its angular momentum. Each subband has its own discrete value of m, while k varies in each subband within the Brillouin zone (fig.2). Interestingly, for the lowest subband of the armchair nanotube the electron energy is linear in k and, hence, the number of states per unit energy interval below the Fermy energy is constant (before entering the next subband, see Fig.3). We will use the approximation of one filling subband in this paper. This is not a principal limitation of the model but it allows to obtain very compact expressions for the SWNT electrostatics. This approximation breaks when the transverse component of the electric field across the nanotube becomes very large (see [10,12] for details). This may happen for nanotube cathode devices but it is unlikely for NEMS or electronics.

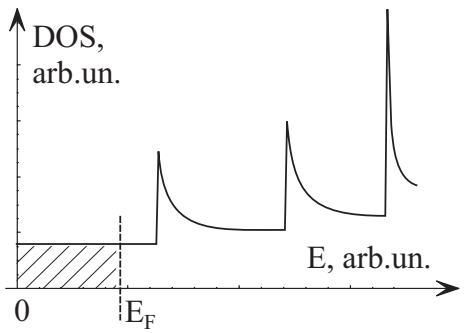

FIG. 3. Density of states of the metallic SWNT near the Fermi level: shaded area represents an extra charge induced in the SWNT by shifting the Fermi level away from the charge neutrality level.

B. Calculation of Atomistic Capacitance

In order to calculate the charge distribution of the straight SWNT as a function of total acting potential we represent the latter as a sum of the external and induced potentials:

$$\varphi^{\rm act} = \varphi^{\rm xt} + \varphi^{\rm ind}. \tag{1}$$

The statistical model supposes that the induced charge is an integral over the nanotube density of states (DOS) from a local charge neutrality level to a local chemical potential which becomes a Fermi level at zero temperature (see Fig.3). The local chemical potential is supposed to follow the local acting potential. Great simplification is achieved in case of metallic nanotube operating at low voltage when the Fermi level shifts within the first sub–band. Then, the electron dispersion is linear and the DOS is constant and equals $\nu_M = 8/(3b\gamma)$. Here $b \simeq 1.4$ Å is the interatomic distance and $\gamma \simeq 2.7$ eV is the hopping integral (we use standard definitions for TBA calculation of DOS). Within this approximation of the linear energy dispersion in the lowest subband, the induced charge density reads as:

$$\rho(z) = -e^2 \nu_M \phi^{\rm act}(z). \tag{2}$$

We note that Eq.(2) holds in one–dimensional (1D) case; while in 2D the charge is proportional to the electric field (first derivative of the potential) and in 3D the charge is proportional to the Laplasian of the potential (the Poisson equation).

In order to obtain a selfconsistent solution for the charge density we calculate the induced potential with use of a Coulomb operator Green function, $G(\mathbf{r}, \mathbf{r}')$:

$$\phi^{\rm ind}(\mathbf{r}) = 4\pi \int G(\mathbf{r}, \mathbf{r}')\rho(\mathbf{r}')\mathbf{dr}'. \tag{3}$$

The Green function of a 1D system is known to diverge logarifmically until some external screening is considered. In case of a nanotube device, this screening is due to the closest gates/contacts. An equation, giving the nanotube charge density implicitly, follows from Eqs.(2,3) and reads as:

$$-\frac{\rho(\mathbf{r})}{e^2\nu_M} - 4\pi \int G(\mathbf{r}, \mathbf{r}')\rho(\mathbf{r}')\mathbf{dr}' = \varphi^{\rm xt}(\mathbf{r}). \tag{4}$$

The equation can be inverted analytically in simple case. In general, it allows only numerical solution or may be expressed as a series.

An interesting result of our study is that the nanotube may be divided in three parts: two contact regions and a "central" region. The side parts are the regions near the contacts of a length about several h long, where h is the distance to the gate. Aspect ratio of devices of the state–of–art of nanotube technology is very high, which means that the length of the nanotube, L, is much larger than the h. Then, the central region of the nanotube covers almost all the device length.

The electrostatics of the central region is elementary and allows an analytical solution for Eq.(4). Because of the screening of the Coulomb interaction by the back–gate and the valence electrons of the nanotube, the corresponding Green function is short–ranged. Therefore, (at the distance about $2-3h$ from the contact) the self–consistent charge density is given by a simple expression:

$$\rho \simeq \rho_\infty = -\frac{\varphi^{\rm xt}}{C_g^{-1}+C_Q^{-1}} \simeq -\varphi^{\rm xt}\, C_g \left(1-\frac{C_g}{C_Q}\right), \qquad (5)$$

here we used notations $C_g^{-1} = 2\log\left(\frac{2h}{R}\right)$ and $C_Q^{-1} = 1/(e^2\nu_M)$ for the inverse capacitance (potential coefficient) of a straight metal cylinder and the atomistic correction, respectively. ρ_∞ stands for an equilibrium charge density of the SWNT, calculated at the distance from the side electrode much larger than the distance to the back–gate, h.

III. MODEL EXTENSIONS

A. Quantum Mechanical calculation

We derived the Green functions for several realistic device geometries and calculated the selfconsistent charge densities. These charge densities were compared with the results of the quantum mechanical computation. We solved joint Shroedinger and Poisson equations for the valence pi–electrons of a metallic armchair [10,10] SWNT in one subband approximation (in full neglecting the intersubband or sigma–pi mixing which has been estimated and is of minor importance for our problem). Aside such purely quantum effects as quantum beatings at the ends of the finite length nanotube, the statistical, semi–classical and full quantum mechanical charge distributions are almost identical (a cross check has been done with use of periodic boundary conditions to exclude the finite length effects). Fig.4A shows typical charge density distributions calculated with use of the TBA and the Boltzmann equation for a cantilever SWNT of 50 nm long. We must conclude that a simple statistical description works fairly well for the case of straight ideal single wall nanotube. Similar result is obtained for a SWNT with two side contacts (Fig.4B).

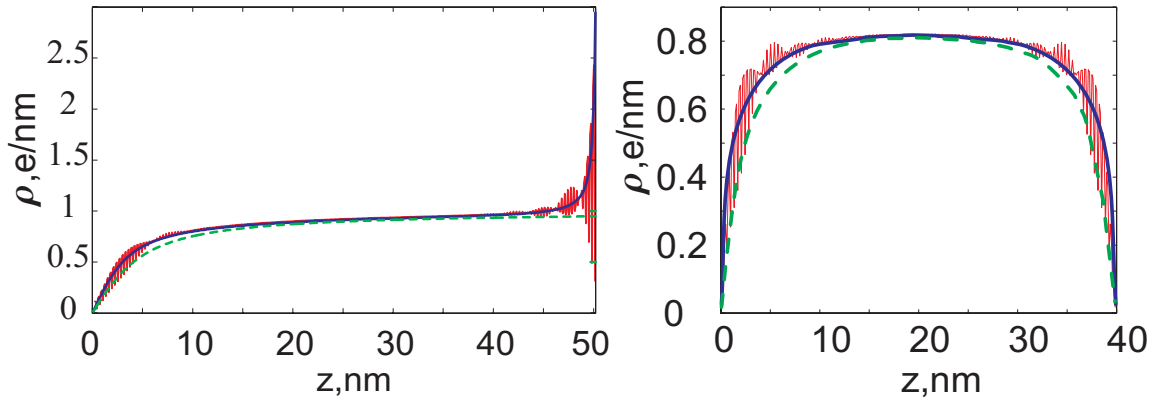

FIG. 4. Specific charge density for two devices with geometry given in Fig.1. The solid oscillating curve is a result of full quantum mechanical calculation. The solid line is a solution of joint Poisson and Boltzmann eqs. The dotted line is our analytical approximation.

B. Capacitance of Distorted SWNT

The equation for the equilibrium charge density is valid for a distorted nanotube as well as for an ideal straight nanotube. In case of slightly bent SWNT, one has to use in Eq.(5) the capacitance of the bent metallic cylinder, $C_g^{-1}(h(z))$, instead of the logarithmic capacitance which is valid only for a straight one. Thus, the atomistic capacitance of the distorted SWNT depends on its shape:

$$C(z) \simeq \frac{1}{C_g^{-1}(h(z)) + C_Q^{-1}} \simeq C_g(z)\left(1 - \frac{C_g(z)}{C_Q}\right). \tag{6}$$

This analytical form of solution for the device electrostatics is a great simplification for calculating electrostatic forces in various NEMS devices.

CONCLUSIONS

In summary, we have developed a continuum device theory for nanotube electromechanical systems and nanotube electronics. The theory gives a fast and accurate method for a simulation of a charge density for a nanotube of an arbitrary shape displaced by a voltage applied to the nanotube end(s). The charge density is given by an atomistic capacitance of the nanotube. The atomistic capacitance is not defined solely by material properties of the nanotube itself. It depends also on the environment because of the charge interaction in low–dimensional electronic system of the nanotube is screened by near placed electrodes. An analytical expression for the atomistic capacitance of the nanotube subjected to moderate distortions is found as well as for an ideal nanotube. The role of quantum effects is evinced. The model gives an estimation of the charge density profile in the gated nanotube device which is useful for simulating SWNT electronics.

ACKNOWLEDGMENTS

Authors are indebted to Professor K. Hess and Dr. A.G. Petrov for valuable discussions. Authors acknowledge support through a CRI grant of UIUC. S.V.R. ac-

knowledges DoE grant DE-FG02-01ER45932, RFBR grant 00–15–96812 and Beckman Fellowship from the Arnold and Mabel Beckman Foundation. K.A.B. is grateful to Beckman Institute for hospitality during his work in Urbana.

---

[1] R. Saito, G. Dresselhaus, M. S. Dresselhaus, *Physical Properties Of Carbon Nanotubes*, Imperial College Press, London, 1998.

[2] S. Akita et. al., "Nanotweezers consisting of carbon nanotubes operating in an atomic force microscope", Appl. Phys. Lett. **79** (11), 1691–1693, 2001.

[3] E. Minot, V. Sazonova, Y. Yaish, J-Y. Park, M. Brink, P. McEuen, unpublished.

[4] V. Derycke, R. Martel, J. Appenzeller, and P. Avouris, "Carbon Nanotube Inter- and Intramolecular Logic Gates," Nano Letters, vol. **1**, pp. 453–456, 2001.

[5] A. Bachtold, P. Hadley, T. Nakanishi, and C. Dekker, "Logic Circuits with Carbon Nanotube Transistors," Science, **294**, pp. 1317–1320, 2001.

[6] M. Dequesnes, S. V. Rotkin, N. R. Aluru, "Calculation of pull-in voltages for carbon nanotube-based nanoelectromechanical switches," Nanotechnology **13**, 120–131, 2002.

[7] M. Dequesnes, S. V. Rotkin, N. R. Aluru, Int. Journal of Computational Electronics (to appear in April 2002).

[8] S. V. Rotkin, "Analytical Calculations For Nanoscale Electromechanical Systems," in Microfabricated Systems and MEMS – VI, vol. PV 2002–6, Symposium – the Electrochemical Society Proceedings, P. J. Hesketh, S. S. Ang, J. L. Davidson, H. G. Hughes, and D. Misra, Eds.: ECS Inc., Pennington, NJ, USA., 2002, pp. 90–97.

[9] S. Luryi, "Quantum capacitance devices", Appl. Phys. Lett. **52** (6), pp. 501–503, 1988.

[10] K.A. Bulashevich, S.V. Rotkin, "Nanotube Devices: Microscopic Model," JETP Letters (Pis'ma v ZhETF) **75**(4), pp. 205–209, 2002.

[11] A. A. Odintsov, Y. Tokura, Journal of Low Temperature Physics, **118**, 509 (2000).

[12] A.G. Petrov, S.V. Rotkin, unpublished.

[13] L.X. Benedict, S.G. Louie, M.L. Cohen, "Static Polarizabilities of Single Wall Carbon Nanotubes," Phys. Rev. B. **52**, pp. 8541–8549, 1995.

[14] M. S. Dresselhaus, G. Dresselhaus, and P. C. Eklund, *Science of Fullerenes and Carbon Nanotubes*. San Diego, CA: Academic Press, 1996.