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TRANSPORT IN HELICALLY COILED CARBON NANOTUBES: SEMICLASSICAL APPROACH

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Abstract: Semiconducting single wall carbon nanotubes (SWCNTs) exhibit high electron mobility in low electric field. Tube diameter and temperature have been found to strongly affect transport properties of SWCNTs. We have investigated electron mobility of helically coiled carbon nanotubes (HCCNTs). Electron and phonon band structures of HCCNTs are used in calculation of electron-phonon matrix elements. Scattering rates are calculated using the first order perturbation theory while taking care of energy and momentum conservation law. In order to obtain electron drift velocities, steady state simulation of charge transport is performed using Monte Carlo method.

Keywords: carbon nanotubes, electron-phonon scattering, drift velocity, electron mobility, charge transport.

1. INTRODUCTION

In semiconducting single-wall carbon nanotubes (SWCNTs), low intensity of electron scattering on acoustic phonons causes their high electron mobility. The theoretically predicted charge mobility in semi-conducting zig-zag carbon nanotubes (CNTs) is in the range $10^3 - 10^5$ cm²/Vs [1]. Electrons accelerated by high bias voltage are scattered by optical phonons, mostly through the interband relaxation processes, while when moving in a low electric field, they emit or absorb acoustic phonons.

The most demanding task in the charge carrier mobility calculations is to obtain the scattering rate of an electron in a specified state. The electron scattering rate increases with the concentration of impurities or defects in a crystal lattice. Besides, electron-electron interaction impacts the electron relaxation time, as well. However, electron-phonon (el-ph) interaction gives the largest contribution to the electron scattering rate and plays an important role in processes like charge transport, Raman scattering and also in fast optical spectroscopy [2].

Here, we calculate the scattering rates of conduction band electrons in semiconducting infinitely long single-walled HCCNTs without impurities, caused only by electron-phonon interaction, neglecting all other contributions. Given are also the results obtained from the single-particle Monte Carlo simu-

lations of electron free flight interrupted by a stochastic scattering event. Drift velocity is obtained as a time averaged velocity of free flight in the k -space during the total simulation time [3,4]. All the calculations are simultaneously derived for semiconducting straight single-wall carbon nanotubes, in order to make a comparison based on the results obtained by the same method of calculation.

2. MODEL

Models of helically coiled carbon nanotubes (HCCNTs) are generated from symcell atoms applying line group symmetry from the fifth family. Helically coiled nanotubes are multiple orbit systems, the symcell of which is a half of the monomer. Generators of the symmetry group of HCCNTs are screw axis, rotation followed by a fractional translation, U axis (rotation around X axis for π) [5,6]. Generally, HCCNTs are not translationally periodic and do not have pure rotational symmetry. Group parameters are simple functions of geometrical parameters: monomer length (a) (distance between two successive knees along the helix), helical radius (R) and inclination angle χ (Figure 1). Except hexagons, carbon network of HCCNTs contains also pentagons and heptagons. Every monomer has pairs of heptagons at negative and pentagons placed at positive

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Gaussian curvature. Geometry and physical properties depend on spatial distribution of these 5–7 carbon rings. Topological coordinate method, based on graph theory, is used to construct a monomer of HCCNTs, which is explained in detail in Ref. [7]. In our calculations we use the extended model of helically coiled carbon nanotubes, obtained by changing the concentration and position of pentagons and heptagons in a monomer [6]. Therefore, the indices of our model of HCCNT are $(n_6, n_r, n_7, n_5, (b_1, b_2))$. Namely, the model is labeled by 4 graph parameters, the number of hexagons between pentagons and heptagons (n_6), the number of hexagons added between two neighboring monomers (n_r), the number of hexagons added between pairs of heptagons and pentagons respectively, (n_7, n_5), and finally (b_1, b_2) are the supercell vectors.

3. METHOD

Calculations of electron-phonon matrix elements and determining allowed scattering channels in relaxed helically coiled and straight carbon nanotubes were performed by using electron and phonon dispersion branches, atomic deformation potential vectors, symmetry adapted electronic wave functions and phonons polarizations [9, 12]. Optimized configuration of HCCNTs is obtained by minimi-

$$W_{k,k'}^{\nu} = \frac{2\pi}{\hbar} |M^{(\nu q)}(k'm'l_f, kml_i)|^2 \delta(\varepsilon_m(k') - \varepsilon_m(k) \pm \hbar\omega_\nu(q)) \left(f_T(\omega_\nu) + \frac{1}{2} \pm \frac{1}{2} \right),$$

where $f_T(\omega_\nu)$ is Bose-Einstein occupation number for the phonon (q, ν) , and $M^{(\nu q)}(k'm'l_f, kml_i)$ is an electron-phonon matrix element. Here, in the formula of scattering rate, \pm applies for different types of the events. Namely, (+) describes emission while (–) describes absorption of a phonon. Generally, the scattering probability from the initial state to all possible final states is obtained by integrating $W_{k,k'}^{\nu}$ over all phonon branches over the entire Brillouin zone. The integration, in the case of CNTs (and 1D systems in general), is reduced to the summation over the individual contributions of the allowed scattering channels, due to the one-dimensionality of the Brillouin zone and the wave-vector and energy conservation rules [11].

First order perturbation theory was used to describe el-ph coupling for a general q point phonon mode. Electron-phonon matrix elements are defined

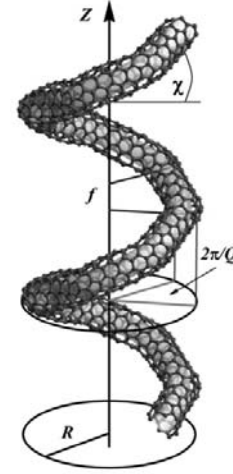


Figure 1. Totally relaxed helically coiled carbon nanotube $(1,3,2,0,((1,0),(0,8)))$. Screw axis parameters, fractional translation and rotation angle are shown as projections of monomer on helical axis (Z) and on contour in plane orthogonal to Z respectively. Inclination angle (χ) and helical radius (R) of HCCNT are also shown.

zation of Brenner interatomic potential for carbon in solids [10]. All allowed scattering channels of an electron in a specified state (k, m) are obtained by applying the selection rules for angular momentum $m \mp \nu = m'$ and for wave-vector $k \pm q = k'$ together with energy conservation relations $\varepsilon_{m,k} \mp \hbar\omega_{\nu,q} = \varepsilon_{m',k'}$. The scattering probability from the initial (k, m) to the final (k', m') electronic state is given by the Fermi golden rule:

by electron potential variation $\delta V^{(\nu q)}$ due to the lattice vibration

$$M^{(\nu q)}(k'm'l_f, kml_i) = \langle \psi^{(k'm)l_f}(r) | \delta V^{(\nu q)} | \psi^{(k'm)l_i}(r) \rangle \quad [2].$$

Extended tight binding model is used in calculations of el-ph matrix elements which includes $2s$, $2p_x$, $2p_y$ and $2p_z$ orbitals, wherein, the atomic deformation potential (ADP) of σ and π bonds for neighboring carbon atoms is taken into account. Two independent atomic deformation potential vectors are defined by keeping only two-center integrals for the on-site (α_p) and off-site (γ_p) ADP. For both of these deformation potentials, the considered projections along the bond direction ($p = ss, s\sigma, \sigma\sigma$, or $\pi\pi$) and perpendicularly to the bond direction ($p = \pi s, s\pi, \sigma\pi$ or $\pi\sigma$), depend only on a distance between the carbon atoms. Electron-phonon matrix elements are calculated by using symmetry adapted formalism, where electron and phonon bands are used, and the corresponding electronic wave functions and phonon

displacement vectors obtained by using *POLSym* code [8] and ADP [2].

4. RESULTS

Due to the absence of the rotational symmetry of HCCNTs, the angular quantum number of all electron and phonon bands is zero. Hence, unlike the case of straight CNTs, the number of scattering channels in HCCNTs is not reduced due to the angular quantum number selection rules. The restrictions for emission or absorption of phonons are imposed by conservation of energy and quasi-momenta only. In accordance with the non-crossing rule, as the number of atoms within a monomer increases, the electronic bands flatten and the corresponding group velocities decrease.

Electronic states in the conduction band of HCCNTs have a large number of allowed scattering channels and typically have a high density of the final electron states. This results in higher frequency of electron scatterings in HCCNTs than in SWCNTs. One allowed process of electron back-scattering which is followed by phonon emission in (1, 3, 2, 0, ((1, 0), (0, 5))) HCCNT is shown in Fig-

ure 2a). A very high scattering rate is obtained in the vicinity of local minima of the conduction band of a HCCNT due to intense emission of the long-wavelength longitudinal acoustic (LA) phonons at 300 K, Figure 2b). Phonon occupation number increases with temperature and electron scattering frequency rises, Figure 2c). According to the energy conservation law, due to the large energy gap between the conduction and other bands of HCCNT, Figure 2a), only intra-band electron transitions were found. As the number of atoms in a monomer increases, increases the number of electronic bands over the Brillouin zone, they necessarily become narrower, and inter-band electron scattering emerges.

A semi-classical approach is applied to calculate electron drift velocity of semiconducting SWCNTs and HCCNTs in homogeneous external electric field [3], by means of standard Monte Carlo techniques. Band structures and scattering rates are obtained from quantum theory, while electron motion is simulated by Newton classical equations. Direct integration method [4] is used to increase the computing efficiency of the free flight time calculations.

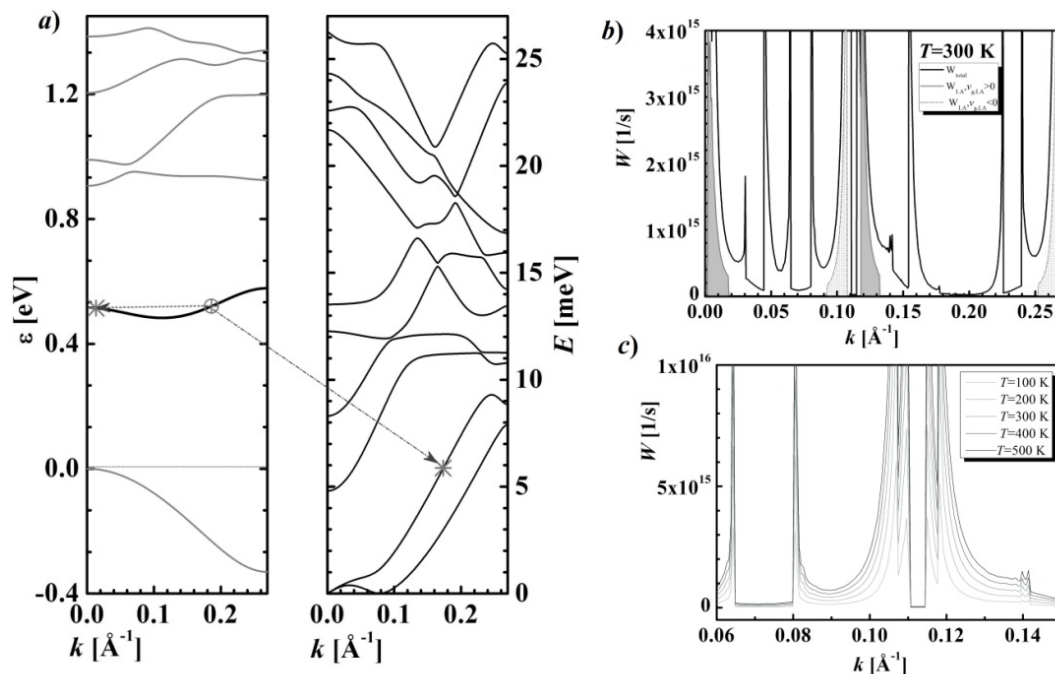


Figure 2. a) Electronic bands (left) in the range from -0.4 eV to 1.4 eV and phonon bands (right) up to 26 meV of (1, 3, 2, 0, ((1, 0), (0, 8))) HCCNT. Dashed arrows indicate transition from the initial electron state (gray circle) into the final electron state followed by the phonon creation (gray stars). b) Electron scattering rate as a function of the wave-vector at room temperature (thick black line) and contribution of LA phonons in the vicinity of Γ point (pattern and gray area). c) Dependence of the scattering rate of the conduction band electrons in the vicinity of local minimum at different temperatures (from 100 to 500 K).

The number of inter-band transitions as well as population of electrons in energetically higher band of zig-zag SWCNTs increases with the electric field (E_{el}). Electron distribution function shifts with E_{el} and from a certain value, when the concentration of electrons in the upper band becomes significant, drift velocity begins to increase and negative differential mobility (NDM) occurs [3]. In comparison with SWCNT, drift velocity of the given semiconducting helical nanotube is lower by an order of magnitude, Figure 3. Low group velocity and frequent backscattering of electrons from the conduction band of HCCNTs results in low electron drift velocity. The states with low group velocity are more occupied in high electric field due to the shift of the electronic distribution function, which results in the negative differential mobility. Over the whole range of the applied homogeneous electric field, the drift velocity decreases with the temperature, for all types of carbon nanotubes Figure 3.

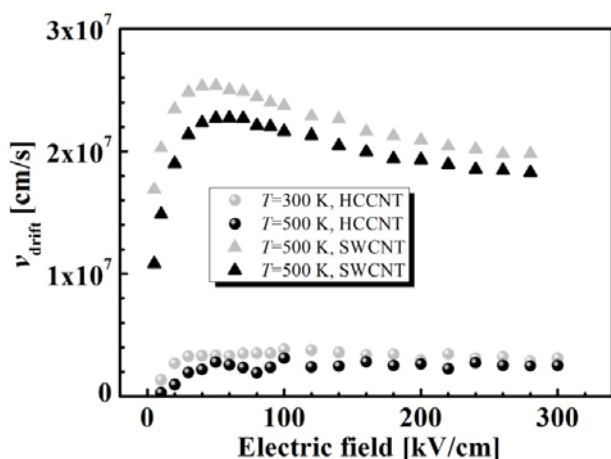


Figure 3. Drift velocity dependence on electric field of $(11, 0)$ SWCNT (triangles) and $(2, 2, 0, 0)$, $((1, 0), (0, 5))$ HCCNT (circles) at 300 K (gray) and 500 K (black).

5. CONCLUSION

Electron and phonon dispersion branches of helically coiled carbon nanotubes are calculated. Scattering rate for electrons in the conduction band is obtained assuming that the scattering is only due to the electron-phonon coupling. In the calculation of the latter all phonon branches are taken into account. We find that electrons from conduction band in helically coiled nanotubes have lower group velocity, more scattering channels and consequently, higher scattering probability than their straight CNT counterparts. We have performed charge transport simulations in homogeneous electric field using Monte Carlo method. Drift velocity dependence on electric field and temperature is obtained both for

HCCNTs and SWCNTs. At certain values of electric field intensity and temperature the electron drift velocity of HCCNTs is by one order of magnitude lower than the drift velocity of the corresponding SWCNTs.

6. ACKNOWLEDGEMENT

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ТРАНСПОРТ У ХЕЛИКАЛНИМ УГЉЕНИЧНИМ НАНОТУБАМА: ПОЛУКЛАСИЧНИ ПРИСТУП

Сажетак: Полупроводне једнослојне угљеничне нанотубе (ЈУНТе) имају велику мобилност електрона на ниским електричним пољима. Транспортне особине ЈУНТа у великој мери зависе од њеног пречника и температуре. Испитивали смо мобилност електрона хеликалних угљеничних нанотуба (ХУНТа). За налажење електрон–фонон матричних елемената коришћени су атомски деформациони потенцијали за суседне атоме, електронске и фононске дисперзионе гране ХУНТа. Применом теорије временски зависне пертурбације добијена је учесталост расејања електрона на фононима у зависности од температуре, водећи рачуна о закону одржања енергије и селекционим правилима. Симулацијом транспорта наелектрисања у хомогеном и временски непроменљивом електричном пољу Монте Карло методом добијене су дрефт брзине електрона угљеничних нанотуба у зависности од јачине поља и температуре.

Кључне речи: угљеничне нанубе, електрон–фонон расејање, дрефт брзина
мобилност електрона, транспорт наелектрисања.

