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MONTE CARLO STUDIES OF ELECTRONIC TRANSPORT IN HELICALLY COILED CARBON NANOTUBES

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Abstract: We studied the stationary electron transport of semiconduction single-wall straight and helically coiled carbon nanotubes in the presence of electron- phonon interaction. The electron and phonon bands as well as electron phonon coupling matrix elements are obtained from quantum mechanical calculations with the application of symmetry. Total scattering rate for all electronic states relevant for charge transport is obtained as a sum over independent processes. Transport simulation is realized by Monte Carlo algorithm, where free flight time and scattering mechanism are selected randomly. The obtained electron transport properties of helically coiled and straight carbon nanotubes are significantly different. The electron drift velocities in helically coiled nanotubes are several times lower than in straight carbon nanotubes.

Keywords: helically coiled carbon nanotubes, electron-phonon interaction, scattering rate, drift velocity.

1. INTRODUCTION

Straight single-wall carbon nanotubes (SWCNTs) are quasi one dimensional crystal structures, which could be simply modeled as rolled up graphene strip into the cylinder. Since the discovery, carbon nanotubes (CNTs) have been extensively studied due to their physical properties with the focus on electronic and mechanical properties. Due to remarkable physical properties, CNTs could be used as building blocks of electronic components, like one dimensional transistors, very sensitive to the change of electric field. There are predictions about their possible applications as electro-mechanical devices for detection of atomic masses or small forces, because CNTs have low mass density and low intrinsic energy dissipation and high electron mobility [1].

We have developed a model of helically coiled carbon nanotubes (HCCNTs), based on the method proposed by I. Laszlo [2]. Its helical geometrical parameters [3] are determined with regular spatial distributions of heptagons and pentagons along the helix. HCCNT can be built from the symmetry cell (*symcell*) by applying the full symmetry group, i.e. a group of the fifth line group family. The monomer itself is generated from the symcell, by rotating the symcell around the X-axis for π. The entire HCCNT is built up from the monomer, by arranging it regularly along the helix.

Monomer contains pairs of heptagons at negative and pentagons at positive Gaussian curvature. We labeled the model of HCCNTs with (n_6, n_r, n_7, n_5, (b_1, b_2)), where the given parameters describe a relative distance of heptagons and pentagons (n_6), between the pairs of hexagons in adjacent monomer (n_r), a separation of heptagons (n_7) or pentagons (n_5) in monomer, and a distribution as well as the orientation of polygons in monomer are determined with super-cell vectors (b_1, b_2) [4]. Calculation of relevant transport quantities begins with the evaluation of electron-phonon scattering rate for electrons in the conducted states, derived using quantum mechanical methods, since electron dynamics is treated classically via Mote Carlo (MC) simulations. Negative differential mobility (NDM) of zig-zag SWCNTs, earlier reported in [5] is confirmed here. Lower drift velocity of electrons in HCCNTs is obtained over the entire range of electric field (E_{el}), applied along the helix (tube) axis. Also NDM is found in HCCNTs at high electric field, but interpreted to be due to the different origin than in straight nanotubes.

2. ELECTRON-PHONON COUPLING AND SCATTERING RATE OF ELECTRONS

We studied the influence of electron-phonon interaction on the electron transport in helically

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coiled and straight-single wall carbon nanotubes. We considered the electronic dynamic in nanotubes, interrupted by emitting or absorbing phonons, specified by selection rules and energy conservation. Non-homogenous deformation of crystal lattice, caused by phonon excitation, leads to a change of electron potential energy. Matrix elements, that describe the electron transitions coupled with phonons, are derived from the variation of Kohn-Sham potential [6]

$$M = \left\langle \psi^{(k_{fin})}(r) \middle| \delta V^{(q)}(r) \middle| \psi^{(k_{in})}(r) \right\rangle,$$

where $\psi^{(k_{in}/k_{fin})}(r)$ are initial/final electrons wave functions, $\delta V^{(q)}(r)$ is a variation of the potential [14] caused by phonon vibrations. Electron-phonon matrix elements are calculated for phonons with arbitrary wave-vectors. The intensity of electron-phonon coupling depends on initial and final electronic states, phonon polarization vectors and atomic deformation potential. The deformation of bonds composed from orbitals (o=2s,2p_x,2p_y,2p_z), due to the change of relative position between carbon atoms is described with nine off-site inequivalent atomic deformation vectors (α_p) (i.e. projections along the carbon-carbon bond p = $ss,\pi\pi,\sigma\sigma,s\sigma,\sigma s$ and projections perpendicular to the carbon-carbon bond p = $\pi s,s\pi,\sigma\pi,\pi\sigma$), and six on-site ($\lambda_p$) inequivalent vectors (along the carbon-carbon bond p = $ss,\pi\pi,\sigma\sigma,s\sigma$ and orthogonal to the carbon-carbon bond p = $\sigma\pi,\pi\sigma$). All atomic deformation potential matrix elements are the function of interatomic distance, obtained from density functional theory [7]. Electron-phonon matrix elements have to be calculated for all allowed scattering channels independently, and they are 0 if the selection rules are not fulfilled. The probability of a transition per unit time, from an initial to a final electronic state via emitting or absorbing a phonon is given by Fermi golden rule. The total scattering rate (W) is a sum of transition probabilities over all possible electron and phonon states, where after application of energy conservation rule, summation over all allowed scattering channels (Ω) remains:

$$W_{m1}(k,T) = \frac{f}{\hbar n_{at}} \left(\sum_{qi\in\Omega} \left| M(k+q_i,k) \right|^2 \frac{f_B(\omega_v(q_i),T)}{\left| g'_{e,m2}(q_i) \right|} + \sum_{qj\in\Omega} \left| M(k-q_j,k) \right|^2 \frac{f_B(\omega_v(q_j),T)+1}{\left| g'_{a,m2}(q_j) \right|} \right),$$

$$g_{e/a,m2}(q) = \varepsilon_{m2}(k \pm q) - \varepsilon_{m1}(k) \pm \hbar\omega_v(q),$$

$$\Omega = \left\{ q \in (-\pi,\pi] \middle| g_{e/a,m2}(q) = 0 \wedge g'_{e=a,m2}(q) \neq 0 \right\},$$

where *e/a* corresponds to the emission/absorption of the phonon with angular frequency ω_v, and $\varepsilon_m(k)$ is energy of the electron with wave-vector k, from branch $m_{1(2)}$, while n_{at} and f are respectively the number of atoms in a monomer and fractional translation of the HCCNT [3,4], f_B is the phonon occupation number at temperature T in thermodynamic equilibrium.

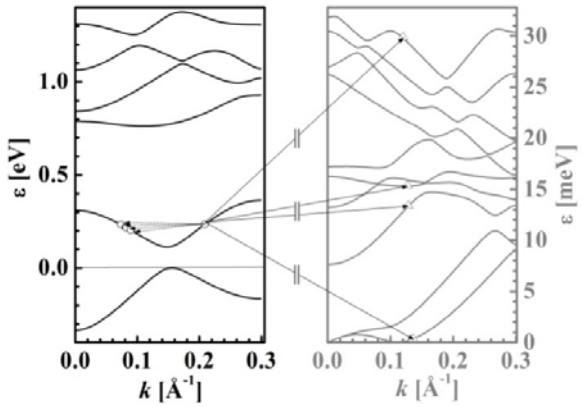

Figure 1. Electron (black) and phonon (gray) dispersion branches of (2,2,0,0,((1,0),(0,5))) HCCNT. Few allowed electron backward scattering from the state, labeled with a gray circle, into the states indicated with arrows by emitting phonons labeled with triangles, is shown.

The electronic bands and phonon dispersion, obtained by *POLSym* code [8] have been used in all segments of the calculations: the selection of allowed scattering channels (Figure 1a), derivation of electron-phonon matrix elements on the symcell, the evaluation of electron scattering rate and MC simulation. Harmonic constants, for all atoms in symmetric cell of helically coiled carbon nanotubes in relaxed configurations [9], used for the construction of the phonon Hamiltonians, are obtained numerically, using the Brenner potential [10] (as electronic bands are obtained by density functional tight binding method [11]).

3. ELECTRON MOBILITY

Accelerated conducting electrons in homogenous electric field are scattered on impurities, defects, electrons and phonons. Here we consider electron scattering in the presence of electron-phonon interaction, without any other contributions. Intrinsic characteristics, used for describing electronic transport, like relaxation time or drift velocity are obtained within the semi-classical approximation, where the scattering rate is evaluated using the quantum theory, while classical treatment of motion is applied in description of electrons dynamic . Due to the consideration of all allowed processes, the calculation of drift velocity

(v_d) generally requires multi-band transport model. Monte Carlo simulation of electron positions in the conduction band, in the presence of homogenous and time independent electric field, is applied. In order to achieve more efficient calculation, direct integration method, described in [12], is used to estimate the time of free flight (t_f)

$$-\ln r = \int_0^{t_f} W\left(k(t')\right)dt'$$

where r is uniformly distributed random number on [0,1]. The electron-free flight is interrupted by instantaneous electron-phonon interaction, since in the meantime, between two scattering events the wave vector is updated according to the equation $k\left(t+t_f\right)=k(t)+\frac{eE}{\hbar}t_f$. Scattering mechanisms do not contribute equally to the total scattering rate $W(k)$, and therefore it is selected by generating a random number, Figure 2 b), lying uniformly between 0 and 1, and n-th scattering event is chosen if is satisfied

$$\sum_{i=1}^{n}\frac{W_{i-1}(k)}{W(k)} < r \le \sum_{i=1}^{n}\frac{W_i(k)}{W(k)}.$$

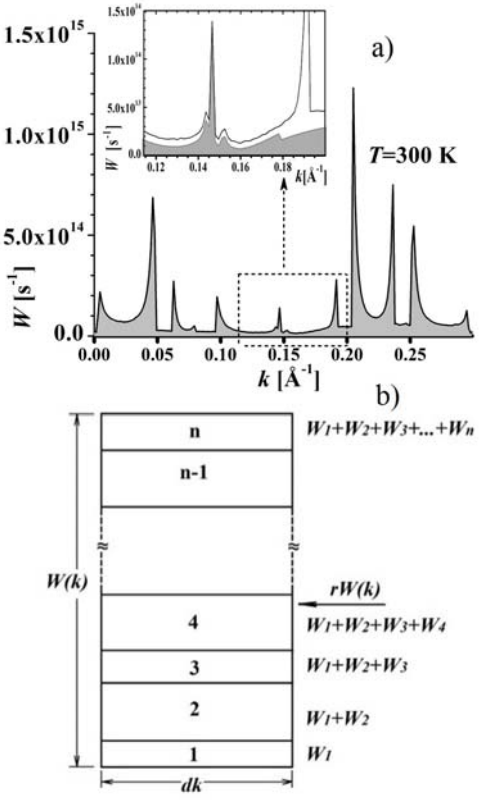

Figure 2. a) Total scattering rate as a function of wave-vector of electron states from conduction band of semiconducting HCCNT. Inset:The filled pattern field shows contribution of the emitted longitudinal and twisted acoustic phonons to the total scattering rate (black solid line). b) The scheme illustrates various contribution to the total scattering rate W(k) of a different scattering channels in the range of wave-vector [k-dk/2, k+dk/2]

At a low electric field, the backscatters of electrons from the vicinity of the states with high density in the conduction band of semiconducting (2, 2, 0, 0, ((1, 0), (0, 5))) HCCNT, shown in Figure 2 a), are mainly followed by the emission of longitudinal acoustic (LA) and twisting (TW) phonons mode. The scattered electron instantly changes its wave-vector and crosses into a new state, specified by the energy conservation and by the selection rules. This assumption is justified by the fact that electron-phonon interaction takes much shorter time interval than the free flight time period lasts. The usually used method for electron transport simulation in stationary homogeneous electric field is single particle MC, wherein the estimation of relevant quantities is performed by taking average over free flights. At the beginning of simulation, the distribution of particles in the inverse space is described by Fermi-Dirac statistics. Before starting to collect the data, relevant for statistical analysis, simulation must pass through many scattering events.

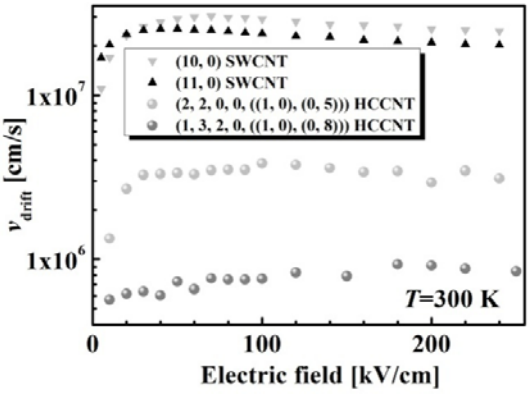

Figure 3. Electron drift velocity of semiconducting HCCNTs (circles) and zig-zag SWCNTs (triangles) at room temperature as a function of the applied homogenous electric field.

The dependence of the drift velocity of zig-zag SWCNTs on the electric field is shown in Figure 3. The presented results for SWCNTs are in full agreement with the previous predictions [5], also calculated using semi-classical technique. Electron distribution in conducting band, as well as drift velocity varies with the electric field intensity. At low fields, scattering is entirely intra-band, carried out by emitted acoustic phonons, and the drift velocity increases with the field intensity. With a further increasing of the electric field, scattering occurs via emitting optical phonons. At high electric field, the inter-band processes become more frequent. When the population of states in upper unoccupied, conduction band with lower group velocity becomes large enough, then the drift velocity begins to decrease. Negative differential mobility in SWCNTs is

obtained at high fields, identified by negative slope of drift velocity versus the electric field, Figure 3. The peak of drift velocity occurs at a critical field. The value of the maximal drift velocity increases with the radius of zig-zag semiconducting CNT, while the corresponding critical field decreases. This is in relation to the group velocity of the conduction band in zig-zag NTs, that weakly increases with tubular radius, since due to the decreasing of energy gap between bands, the occupation of upper band becomes facilitated.

In semiconducting helically coiled carbon nanotubes negative differential mobility is caused by frequently intra-band scattered of electrons into the states with lower group velocity. Considering energy conservation law, the electron transition from the conduction band of (2, 2, 0, 0, ((1, 0), (0, 5))) and (1, 3, 2, 0 ((1, 0), (0, 8))) HCCNTs, into the energetically closest unoccupied electron states from the adjacent bands, over emitting or absorbing phonon is forbidden, Figure 2 a). Thus, in the conduction regime of given HCNTs only intra-band scattering are possible. Though electron transition occurs inside a particular conduction band, the scattering rate has many singularities over Brillouin zone, indicating transitions between the electronic states of the same band with a high density of states. The electron distribution is shifted by the electric field, so that higher electron energy states are more filled, resulting in the higher conduction velocity of electron. From specific wavevector, group velocity in conduction band of (2, 2, 0, 0, ((1, 0), (0, 5))) HCCNT begins to decrease, so a further increase of E_{el} makes the occupation number of these states larger, thus causing the reduction of the drift velocity. All the bands of the electron and phonon spectra of HCCNTs have trivial angular momenta quantum number $\tilde{m} = 0$ as a result of the absence of rotational symmetry. Consequently, restriction in the selection of scattering channels is reduced and the number of allowed processes increases. Although, according to the non-crossing rule, there are no intersections of the bands. Electronic bands become densely packed in certain energy intervals [9,13] when the number of atoms in monomer is enlarged enough so that the group velocities of the conducted band electrons are substantially lowered. The group velocity of electrons and phonons states of HCCNTs is lower than in the conventional straight SWCNTs, while the number of scattering channels of conducted states is larger. The reduction of v_{d} occurs via the contribution of these two effects, which results in the lower drift velocity in HCCNTs than in SWCNTs over the entire range of the electric field. The population of phonons involved in the allowed processes increases with temperature, making scattering more frequent and inducing reduction of drift velocity.

4. CONCLUSION

We have modeled the electron transport of straight and helically coiled single-wall carbon nanotubes in the presence of electron-phonon interaction. Electron phonon matrix elements, which describe intensity of potential variation caused by non-homogeneous lattice deformation, are derived from extended tight binding method. Scattering probability of electrons per unit time of all independent processes, permitted by selection rules, for all conduction states are found.

The drift velocity of HCCNTs and SWCNTs versus applied electric field are calculated using the Monte Carlo method, based on the semi-classical approach. Distribution of conducted electrons in HCCNTs is spread in electric field, making significantly occupied electron state with low group velocity. Except low group velocity, many allowed scattering channels also affects to the reducing of electron drift velocity in HCCNTs.

5. ACKNOWLEDGEMENT

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7. LITERATURE

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МОНТЕ КАРЛО СТУДИЈА О ЕЛЕКТРОНСКОМ ТРАНСПОРТУ КОД ХЕЛИКАЛНИХ УГЉЕНИЧНИХ НАНОТУБА

Сажетак: Испитивали смо стационарни електрични транспорт полупроводних хеликалних (ХУНТа) и правих (ЈУНТа) једнослојних угљеничних нанотуба. Интензитет електрон-фонон интеракције рачунат је помоћу временски зависне теорије пертурбације уз примену линијских група симетрије, а у апроксимацији чврсте везе. Канали расејања су издвојени применом селекционих правила и закона одржања енергије. Укупна учесталост расејања рачуната је за свако стање електрона из проводних зона сумирањем независних доприноса по свим дозвољеним каналима расејања. Симулација транспорта наелектрисања рађена је применом вишечестичног Монте Карло алгоритма, при чему су време слободног лета и механизам расејања добијени стохастички. Електронске транспортне особине хеликалних и правих једнослојних угљеничних нанотуба се знатно разликују. Добијене дрифт брзине електрона код ХУНТа неколико су пута мање него код ЈУНТа.

Кључне речи: угљеничне нанотубе, електрон-фонон интеракција, учесталост расејања, дрифт брзина, транспорт наелектрисања.