Modeling of electrotransport properties of Li-intercalated graphene film

D Sergeyev\textsuperscript{1,2}, A Duisenova\textsuperscript{1} and Zh Embergenov\textsuperscript{1}

\textsuperscript{1}K. Zhubanov Aktobe Regional University, Aktobe, Republic of Kazakhstan
\textsuperscript{2}T. Begeldinov Aktobe Aviation Institute, Aktobe, Republic of Kazakhstan

E-mail: serdau@mail.ru

Abstract. In this work, within the framework of density functional theory combined with the method of nonequilibrium Green’s functions the density of states, transmission spectrum, current-voltage characteristics, and differential conductivity of Li-intercalated graphene (LiC\textsubscript{6}) have been determined. It is shown that in the energy range of -1.3\textperthousand -1.05 eV the quasiparticle transport through the nanostructure is disable. The features of IV- and dl/dV-characteristics of LiC\textsubscript{6} in the form of decreasing of resistance in the range of -0.4\textperthousand -0.4 V were revealed, and in the interval of 0.4\textperthousand 1.4 V formation of negative differential resistance area, related to scattering of quasiparticles. It is established, that LiC\textsubscript{6} nanodevice has 12\textperthousand 13 ballistic channels and has the maximum amount of conductance \textasciitilde12\textperthousand 13\textsubscript{G}, where G\textsubscript{0} is the conductance quantum.

1. Introduction
Recently, to reduce the size, mass, and energy consumption of electronic techniques, electrophysical properties of different nanoscale contacts are being investigated to be used as active and passive elements in electric circuits and to replace traditional elements (see, for example, [1, 2]). New directions of electronics based on exotic materials are being developed to create a new generation of electronic devices and appliances, such as superconductor [3\textperthousand 5], organic [6], single-electronics [7, 8], spintronics [9] and others. In this respect, one of the most promising nanomaterials for such nanocontacts is graphene, which is an allotropic modification of carbon in the form of a two-dimensional monatomic layer [10,11]. After the discovery of graphene, the search for new two-dimensional materials was started, and some quasi-two-dimensional materials with unusual electronic properties were created [12\textperthousand 14]. In theoretical works [15\textperthousand 17], the possible superconducting properties of the calcium- and lithium-doped graphene film have been considered. However, there is considerable disagreement in explaining the electronic properties of these two-dimensional materials. In work [18] it is noted that for the appearance of superconductivity is responsible non-phonon mechanism of formation of Coopers pairs of electrons due to the weak repulsion between them, leading to spiral type d-pairing, and in work [19] usual phonon mechanism of electron pairing in s-channel is considered, and within the framework of Eliashberg theory is estimated the critical temperature of LiC\textsubscript{6} nanostructure, equal to \textasciitilde8 K.

Even though the superconductivity in the LiC\textsubscript{6} monolayer has not been confirmed experimentally yet, a significant change in the electronic properties of the Li-intercalated graphene compared to the “pure” graphene are observed. In this work, the electrotransport characteristics of Li-intercalated graphene are simulated and analyzed within the framework of the electron density functional theory (DFT) in combination with the non-equilibrium Green’s function (NEGF) method.
2. Geometry

The geometry of the Li-intercalated graphene nanodevice is shown in Figure 1. The length of the nanodevice is ~51.15 Å. The electrodes are an extension of the Li-intercalated graphene with a length of ~8.525 Å. The Li-graphene surface with a size of ~34.1×17.23 Å consists of 255 carbon atoms and 40 lithium atoms. To describe the interatomic interaction and optimize the nanostructure, Brenner [20] and ReaxFF_CHONSSiLi_2013 [21] potentials have been used. During the nanodevice geometry optimization, the parameters of the atomic configuration were relaxed until the forces at all atoms became lower than a given threshold value of 0.05 eV/Å.

![Figure 1. The geometry of a Li intercalated graphene nanodevice.](image)

3. Fundamental equations

Computer simulation of electrotransport characteristics of Li-graphene nanodevice was carried out in the framework of DFT + NEGF in local density approximation (LDA). Modeling of electrotransport characteristics of the nanodevice was realized in program Atomistix ToolKit with Virtual NanoLab [21]. (Fundamental equations of this method are described in detail in our previous works [22, 23]). The current-voltage-characteristic (CVC) of the nanostructure was determined by solving the Landauer equation which indicates the fundamental coupling of the electric current $I$ with the transmittance spectrum $T(E)$:

$$I(V_L, V_R, T_L, T_R) = \frac{2e}{h} \int_{-\infty}^{\infty} T(E) f\left(\frac{E - \mu_R}{k_BT_R}\right) - f\left(\frac{E - \mu_L}{k_BT_L}\right) dE,$$

(1)

where $e$ is electron charge, $h$ is Planck’s constant, $E$ is energy, $f(\varepsilon)$ is Fermi energy distribution function of quasiparticles, $k_B$ is the Boltzmann’s constant, $T_L$, $T_R$ are current temperatures and $\mu_R$, $\mu_L$ are electrochemical potentials of the right and left electrodes. $T(E) = \text{tr} \left[ \Gamma_T G\Gamma_T G^+ \right] = \text{tr} \left[ \Gamma_R G\Gamma_T G^+ \right]$ is transmission spectrum (function), $\Gamma_T$ is broadening matrix, $G$, $G^+$ is lagged and leading Green’s functions. The differential conductivity of the nanostructure was obtained by calculating the self-consistent current at a number of applied biases and performing numerical differentiation.

4. Results

The density of states (DOS) of the LiC$_6$ nanodevice is shown in Figure 2. The DOS of the LiC$_6$ nanostructure near the Fermi energy ($\varepsilon_F \approx 0$) is 35–52 eV$^{-1}$, when in “pure” graphene it is 20 eV$^{-1}$, i.e., the DOS of LiC$_6$ increases in 1.75–2.6 times relative from graphene. This means that the number
of energy levels in the LiC₆ nanodevice near the Fermi energy increases, which should lead to the strengthening of the electron transport of quasiparticles in low bias voltage.

![Figure 2](image1.png)

**Figure 2.** The density of states of LiC₆ nanodevices: 1 - LiC₆, 2 - graphene (for comparison).

The evolution of the transmission function (spectrum) $T(E)$ of the LiC₆ nanodevice with increasing bias voltage from -2 V to 2 V in 0.2 V steps is shown in Figure 3a,b. As the bias voltage increases, the intensity of the transmission spectrum increases. At positive energy, the transmission capacity of quasiparticles through the nanostructure in consideration is higher than at negative energy. In the energy range -1.3÷-1.05 eV the probability of quasiparticles passing through the nanostucture is small. As can be seen, the features of DOS appear in the transmission spectrum of the considered nanostucture at the same energy values, as these values are directly proportional $T(\varepsilon) = D(\varepsilon - U) \frac{2 \pi \gamma \Lambda}{\gamma}$ (here $U$ is self-consistent potential, $\gamma$ is Luttinger parameter) [24].

![Figure 3](image2.png)

**Figure 3.** The evolution of the transmission function: a) when the bias voltage is negative (0 to -2 V); b) when the bias voltage is positive (0 to 2 V).

The current voltage characteristic (CVC) and the differential conductivity of the LiC₆ nanodevice are shown in Figure 4a,b. In the voltage range of -0.4÷0.4 V in LiC₆ nanodevice a rapid linear increase of current to 0.11 mA is observed. It is supposed that the decreasing of the film resistance in the stated interval of voltage appears because of the increasing of number of energy levels near to the Fermi energy. Further increasing of bias voltage from 0.4 to 1.4 V will lead to quasi-linear decrease of current due to scattering of quasiparticles. Notice that a significant decrease in current in this voltage range forms a negative differential resistance (NDR) region on the CVC. These changes are reflected
in the differential conductivity of the LiC$_6$ nanodevice (Figure 4b). The LiC$_6$ nanodevice demonstrates 12÷13 ballistic channels and has a maximum conductance value of $\sim$12÷13$G_0$, where $G_0=e^2/h$ is the conductance quantum.

![Figure 4. The current-voltage - (a) and dI/dV-characteristics (b) of LiC$_6$ nanodevices: 1 – LiC$_6$, 2 – graphene (for comparison).](image)

5. Conclusion
In summary, the main electrotransport characteristics of Li-intercalated graphene (DOS, transmission spectrum, CVC) and differential conductivity have been determined within the framework of DFT + NEGF. It is found that in the energy interval $-1.3÷-1.05$ eV the transport of quasiparticles through the nanostructure is disable. The features of IV- and dI/dV-characteristics are determined: decreasing of resistance in the range of $-0.4÷0.4$V leading to the linear increase of current, appearance of NDR region in the range of $0.4÷1.4$V because of quasiparticles scattering. It is established that the electronic conductivity of LiC$_6$ near the Fermi energy is higher in comparison with graphene.

Acknowledgments
The research has been supported by the grant of the Ministry of Education and Science of the Republic of Kazakhstan AP08052562.

References
[1] Cuevas J C, Scheer E 2017 Molecular Electronics: an Introduction to Theory and Experiment (World Scientific Publishers) p 846
[2] Dragoman M, Dragoman D 2017 2D Nanoelectronics: Physics and Devices of Atomically Thin Materials (Switzerland: Springer International Publishing,) p 199
[3] Likharev K K 2012 Physica C 482 6–18 doi: 10.1016/j.physc.2012.05.016
[4] Hinken J H 1989 Superconductor Electronics: Fundamentals and Microwave Applications (Berlin: Springer-Verlag Heidelberg) p 158
[5] Sergeyev D M 2012 Russian Physics Journal 55 84–91 DOI: 10.1007/s11182-012-9779-4
[6] Kanbur Y, Irimia-Vladu M, Glowacki E D , Voss G, Baumgartner M , Schwabegger G et al 2012 Organic Electronics 13(5) 919–924 doi: 10.1016/j.orgel.2012.02.006
[7] Averin D V, Likharev K K 1986 Journal of Low Temperature Physics 62 345–373 doi: 10.1007/BF00683469
[8] Patel R, Agrawal Y, Parekh R 2021 Microsystem Technologies-Micro-And Nanosystems-Information Storage and Processing Systems 27(5) 1863–1875 doi: 10.1007/s00542-020-05002-5
[9] Baranov P G, Kalashnikova A M, Kozub V I, Korenev V L et al 2019 Phys. Usp. 62 795–822 doi: 10.3367/UFNe.2018.11.038486
[10] Novoselov K S, Geim A K, Morozov S V, Jiang D et al 2004 Science 306(5696) 666–669 doi: 10.1126/science.1102896
[11] Ratnikov P V, Silin A P 2018 Phys. Usp. 61 795–822 doi: 10.3367/UFNe.2017.11.038231
[12] Liu N, Bo G, Liu Y, Xu X et al 2019 Small 15(32) 1805147 doi: 10.1002/smll.201805147
[13] Kiraly B, Liu X, Wang L, Zhang Zh, Mannix A J et al 2019 ACS Nano 13(4) 3816–3822 doi: 10.1021/acs.nano.8b09339
[14] Sahoo S K, Wei K H 2019 Advanced materials interfaces 6(18) 1900752 doi: 10.1002/admi.201900752
[15] Emery N, Herold C, d’Astuto M, Garcia V et al 2005 Phys Rev Lett 95(8) 087003 doi: 10.1103/PhysRevLett.95.087003
[16] Guzman D M, Alyahyaei H M, Jishi R A 2014 2D Materials 1(2) 021005 doi: 10.1088/2053-1583/1/2/021005
[17] Gholami R, Moradian R, Moradian S, Pickett W E 2018 Scientific Reports 8 13795 doi: 10.1038/s41598-018-32050-9
[18] Nandkishore R, Levitov L, Chubukov A 2012 Nature Phys 8 158–163 doi: 10.1038/nphys2208
[19] Profeta G, Calandra M, Mauri F 2012 Nature Phys 8 131-134 doi: 10.1038/nphys2181
[20] Brenner D W, Shenderova O A, Harrison J A et al 2002 J. Phys.: Condens. Matter. 14 783-802 doi: 10.1088/0953-8984/14/4/312
[21] Brandbyge M, Mozos J L, Ordejon P, Taylor J, Stokbro K 2002 Phys. Rev. B 65 165401 DOI: 10.1103/PhysRevB.65.165401
[22] Sergeyev D, Zhanturina N 2019 Radioengineering 28(4) 714-720 doi: 10.13164/re.2019.0714
[23] Sergeyev D M 2018 Journal of Nano- and Electronic Physics 10(3) 03018 doi: 10.21272/jnep.10(3).03018
[24] Datta S 2005 Quantum Transport: Atom to Transistor (Cambridge: Univ. Press) p 404