Tunneling conductance of telescopic contacts between graphene layers with and without dielectric spacer

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Abstract

The telescopic contact between graphene layers with a dielectric spacer is considered as a new type of graphene-based nanoelectronic devices. The tunneling current through the contacts with and without an argon spacer is calculated as a function of the overlap length, stacking of the graphene layers and voltage applied using non-equilibrium Green function formalism. A negative differential resistance (similar to semiconductor tunnel diode) is found with the peak to valley ratio up to 10 and up to 2 for the contacts without any spacer and with the argon spacer, respectively. The capacitance of the contacts between the graphene layers with the argon spacer is calculated as a function of temperature taking into account the quantum contribution. The related $RC$ time constant is estimated to be about 3 ps, which allows elaboration of fast-response nanoelectronic devices. The possibility of application of the contacts as memory cells is discussed.

1. Introduction

Significant progress has been achieved recently in design of novel graphene-based heterostructures that offer great promise for application in nanoelectromechanical systems (NEMS) and nanoelectronic devices. Among such heterostructures there is double-layer graphene [1–4], a system comprising two graphene layers separated by a dielectric spacer. While in bilayer graphene the layers are located at the distance of about 3.4 Å, similar to graphite, in double-layer graphene the distance between the layers is controlled by the thickness of the dielectric spacer. Graphene-based field-effect transistors consisting of two graphene layers separated by a layer of adsorbed molecules [1] and by one or several layers of hexagonal boron nitride (h-BN) [2] have been realized recently and transport properties of such heterostructures have been measured [3,4]. The characteristics of the pressure sensor based on changes of interlayer tunneling conductance of the graphene/h-BN heterostructure have been calculated [5]. An idea of a high-speed memory cell based on relative rotation of layers of double-layer graphene with an argon spacer has been proposed [6]. In the present paper we consider the tunneling conductance of a telescopic contact between graphene layers with and without a monolayer argon spacer showing effects relevant both for traditional electronic devices and NEMS. We also analyze the capacitance of double-layer graphene with the argon spacer to estimate its characteristic response time in electronic devices based on this heterostructure.

The conductance between neighbor graphene layers has been addressed in a number of works [7–10]. The calculations show that the electronic structure of twisted bilayer graphene strongly depends on the twist angle [8]. The tunneling conductance of bilayer graphene can be modified by an order of magnitude upon atomic-scale in-plane relative displacement [7,9] or rotation [9] of the layers, similar to $c$-axis conductivity of graphite [10]. Significant variations in the band gap of bilayer graphene nanoribbons upon atomic-scale in-plane relative displacement of their layers are found using density functional theory (DFT) calculations [11,12]. Quasi one-dimensional transport in systems consisting of...
two overlapping layers [13–17] or a layer with an adsorbed graphene flake [16,18,19], both with a central bilayer region, has been recently a subject of extensive research. The tunneling conductance of such systems based on nanoribbons is found to change by an order of magnitude upon atomic-scale in-plane relative displacement of the layers [13]. Resonances and anti-resonances in the transmission as a function of the carrier energy and overlap length have been explained [16,17,19]. The case of crossed graphene nanoribbons has also been addressed [20,21]. Here we use the non-equilibrium Green function (NEGF) formalism on top of DFT calculations to investigate the influence of stacking and overlap length of graphene layers on the $k_z = 0$ component of the tunneling current between them. Such an approach also allows us to evaluate the effect of the argon spacer.

The effect of negative differential resistance is widely used in traditional electronic devices such as amplifiers, oscillators, frequency mixers and memory cells [22]. As for carbon-based nanoscale systems, up to now the negative differential resistance have been predicted for very narrow graphene nanoribbons [23], chemically doped [24] and field-effect doped [25] graphene nanoribbons, doped graphene monolayer with $p-n$ junction [26], parallel graphene nanoribbons contacts [14,15] and parallel single-walled carbon nanotube contacts [27]. Here the negative differential resistance is found for contacts between graphene layers both with and without the argon spacer.

Extensive studies have been performed to demonstrate the possibility of using graphene layers as movable elements of NEMS. Fast mechanical response of such devices can be predicted from low Q-factor values obtained for relative vibrations [28,29] and self-retracting motion of graphene layers [29,30] observed experimentally [31]. Schemes and operational principles of a set of NEMS based on relative motion and interaction of graphene layers have been considered [7,11–13,29,32–35]. It is worth mentioning a nanorelay [29] and inertial sensor [32] based on telescopic motion of graphene layers [29], memory cells based on motion of a graphene flake on a graphene layer [33,34], nanoelectromechanical switches based on changes of the distance between graphene layers [35] and in-plane relative displacement of layers [11], various nanosensors based on measurements of conductance changes at the in-plane relative displacement of layers of bilayer graphene [7,11–13,35]. Operation of all these NEMS involves application of a bias and conductance measurements. Therefore, understanding of tunneling conductance between graphene layers, which can be gained through theoretical studies, holds the key to success of NEMS developments.

Nearly frictionless relative motion of graphene layers separated by an incommensurate spacer can be also used in fast-response nanoelectromechanical devices [6,36]. However, analysis of capacitance of such systems determining the characteristic time of their electronic response is still lacking. Experimental measurements [37] have shown that an important contribution to the capacitance of single-layer graphene [38] is provided by the two-dimensional electronic gas [39]. The influence of this quantum capacitance on the operation frequency of a radio-frequency device based on single-layer graphene has been studied [40]. Here we apply the approach [38] to obtain the total capacitance of double-layer graphene with the dielectric spacer including the quantum contribution. On the basis of these results and calculated tunneling conductance we estimate the $RC$ time constant of double-layer graphene with the argon spacer.

The paper is organized in the following way. Methods of calculation of tunneling conductance are presented in Section 2. Section 3 is devoted to calculations of the tunneling conductance between graphene layers for telescopic contacts with and without the argon spacer and estimates of the capacitance and related $RC$ time constant of the contact between the graphene layers with the argon spacer. Our conclusions are summarized in Section 4.

2. Methods

The tunneling conductance of telescopic contacts between graphene layers with and without the spacer has been obtained using the non-perturbative approach based on the Green function formalism for systems with semi-infinite periodic electrodes [16,41]. We use the localized basis set to divide the system along the current direction into segments that can only interact with the neighbor ones. In this way, the Hamiltonian of the system includes only Hamiltonians of separate segments and coupling matrices between adjacent segments. Thus, instead of considering the whole infinite system only electronic structures of several segments surrounded by their neighbors have to be calculated. As such segments, we take the contact area between the graphene layers and sufficiently long periodic segments of the electrodes (the size of the segments should be greater than the doubled cutoff radius of the orbitals). Therefore, we perform first-principles calculations for three relatively small structures. The periodic segments of the electrodes are considered under the periodic boundary conditions along the current direction. The third structure is the model of the tunneling contact between the graphene layers of a finite

![Fig. 1. The finite-length structures used for calculation of the electronic structure of the contact area between graphene layers with different stacking separated by the quasi-incommensurate argon layer (in position I) and coupling of the contact area to the graphene electrodes. Carbon, hydrogen and argon atoms are colored in gray, white and blue, respectively. The horizontal black dotted lines indicate boundaries of the simulation boxes with periodic conditions. The vertical red dashed lines denote the contact area and segments of the electrodes. The calculations of Green functions of the electrodes are performed for these segments with periodic boundary conditions (PBC) at the vertical dashed lines. The elementary unit cell width $w = \sqrt{3}a_0$, overlap length $L$ of the graphene layers and distance $d$ between them are shown, where $a_0 = \sqrt{3}a_C = 2.466$ Å, and $a_C = 1.424$ Å is the carbon-carbon bond length. Inequivalent AB and MP stackings for the systems with the argon spacer are referred to in the text as AB1, MP1, AB2 and MP2. The AB and MP stackings shown in the figure correspond to AB1 and MP1. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)](image)
length in the current direction (Fig. 1). The model consists of the overlap area and one segment of each electrode.

Based on the Landauer formalism [16,41] the net current $I$ through the contact of the electrodes at voltage $U$ is given by

$$I(U) = \frac{e}{\hbar} \int_{-\infty}^{+\infty} T_{R}(U,E) \times [\mathbf{f}_{R}(E-E_{F} - eU/2)$$

$$- \mathbf{f}_{R}(E-E_{F} + eU/2)] dE,$$

(1)

where $T_{R}(E,U)$ is the transmission coefficient as a function of the electron energy $E$ and voltage $U$, $E_{F}$ is the Fermi level (chemical potential) of unbiased electrodes, $f_{R}(E-E_{F}) = (1 + \exp((E-E_{F})/k_{B}T))^{-1}$ is the Fermi–Dirac distribution function in the equilibrium state ($U=0$ and $I=0$), $k_{B}$ is the Boltzmann constant, $T$ is the absolute temperature, $e$ is the elementary charge and $h$ is the Planck constant. The electronic structure needed as an input for Eq. (1) is obtained in the present paper by DFT calculations.

In terms of Green functions, the transmission coefficient can be written as [16]

$$T_{R}(E,U) = \text{Tr}[[\Sigma_{V} - \Sigma]^{-1}]$$

(2)

where indices $L$, $R$ and $C$ refer to the left and right electrodes and the central area, respectively, $\Gamma_{C} = i[\Sigma_{V} - \Sigma]$ is the coupling between the central area and the electrode $\ell = L, R$ determined by the electrode self-energy $\Sigma_{V} = V_{C}G_{C}/V_{C}$, which is in turn expressed through the coupling matrix $V_{C}$ between the central area and electrode and the electrode Green function $g_{\ell} = (\mathbf{S} - H_{\ell})^{-1}$ ($\mathbf{S} = E + i\delta$ and $S$ is the overlap matrix, which should be included as the non-orthogonal basis is used), and $G_{C} = (\mathbf{S} - H_{C} - \Sigma_{R} - \Sigma^{-1})^{-1}$ is the Green function of the central area. The potential energies of all carbon atoms within the graphene layers are assumed to be shifted by $\pm U/2$ [14,15].

The current along the zigzag direction is studied and all the structures are considered under periodic boundary conditions along the perpendicular armchair direction to model the case of wide ribbons with negligibly small edge effects [42]. The width of the simulation box along the armchair direction is $w = 3\sqrt{3}d_{0} = 4.27$ Å (one rectangular elementary unit cell), $d_{0} = 2.466$ Å is the lattice constant of graphene obtained in our previous calculations [6] for pristine graphene and $3w = 3\sqrt{3}d_{0} = 12.81$ Å (three rectangular elementary unit cells) for the system with the argon spacer. The size of the simulation box perpendicular to the plane is 27 Å. The periodic electrodes are modeled by segments of $4a_{0} = 9.86$ Å length along the zigzag direction to ensure that non-adjacent segments of the electrodes do not interact. The finite-length models of the telescopic contacts between the graphene layers include the central contact area consisting of the layer overlap of variable length $L$ surrounded by three unit cells of each electrodes, one segment of each electrode and one more edge elementary unit cell of each graphene layer to set properly boundary conditions for the periodic segments. The edges of the graphene layers are terminated by hydrogen atoms. The model structures of the tunneling contact are considered in the simulation box of 90–135 Å length along the zigzag direction.

The AA, MP (middle-point), AB and SP (saddle-point) stackings of the graphene layers are considered, which can be generated one from another by shifting one of the layers by a half of the bond length in the armchair direction (Fig. 1). In the pristine system, the graphene layers are placed at the experimental interlayer distance of $d = 3.34$ Å. As shown by our previous calculations, an argon spacer between the graphene layers prefers to be in an incommensurate phase [6]. To use periodic boundary conditions a quasi-incommensurate layer of argon is constructed with the argon–argon distance of 3.7 Å and argon to carbon ratio in the double-layer graphene of $Ar:C = 1.9$ (Fig. 1). The distance between the graphene layers for this heterostructure was calculated to be $d = 6.82$ Å [6]. Two types of structures with different in-plane position of the argon layer are considered. In position I, the argon layer is placed so that some of the argon atoms are directly on top of the carbon atoms (Fig. 1). In position II, the argon layer is shifted by 0.43 Å in the armchair direction, so that no argon atom turns out to be on top of a carbon atom for any considered stacking of the graphene layers.

We find considerable changes in the $I$–$U$ curves upon increasing or decreasing the overlap area of the graphene layers by just one row of atoms. This is related to modul 3 parity of the quasi-localized states of the bilayer with armchair edges in the central contact area [19], the same as for armchair nanoribbons [42,43]. The most pronounced current oscillations and negative differential resistance effects are observed for the systems with the overlap length of $L = 3na_{0}/2$ (number of atomic rows $N = 3n + 1$, where $n$ is integer), making them especially promising for nanoelectronics. Therefore, we focus on such systems in the present paper.

Density functional theory calculations are performed within the local density approximation (LDA) [44] using OpenMX3.7 [45]. The basis sets of s2p2d1 pseudo-atomic orbitals (PAOs) [46,47] with the cutoff distance of 7 Bohr radii for carbon and hydrogen and s3p3d2 PAOs with the cutoff distance of 11 Bohr radii for argon (C7.0-s2p2d1, H7.0-s2p2d1 and Ar11.0-s3p3d2) as well as fully relativistic pseudopotentials are taken from the 2011 database. The $1 \times 24 \times 1$ and $8 \times 24 \times 1$ $k$-point grids are considered for the finite-length contact model and periodic electrodes, respectively, for the pristine bilayer graphene. The $1 \times 8 \times 1$ and $8 \times 8 \times 1$ $k$-point grids, respectively, are used for the double-layer graphene with the argon spacer. The energy cutoff is 50 Ry. The convergence with respect to these parameters was tested by us previously by the example of carbon nanotubes [48]. The tunneling current between the graphene layers is calculated at temperature 300 K using in-house transport code [41]. One $k_{x} = 0$ $k$-point is considered in the direction perpendicular to the current to access large overlap lengths of the graphene layers and systems with the spacer.

3. Results

3.1. Conductance between pristine graphene layers

The calculated dependences of $k_{x} = 0$ component $I$ of the current on voltage $U$ ($I$–$U$ curves) for pristine graphene layers with the AA and AB stackings have the following common features. (i) The $I$–$U$ curves exhibit significant oscillations with a period on the scale of 1 V and amplitudes on the order of 30–40 µA (Fig. 2). These oscillations imply that there are voltage intervals with a strongly pronounced negative differential resistance, the same as in previous calculations for graphene nanoribbons [14,15,17]. Such a behavior is known to originate from the interference of carrier paths through the quasi-localized states in the bilayer region [16,17] or from Fabry–Pérot-like interference of one propagating channel with itself [16]. (ii) The period of the current oscillations and the amplitude of the first current peak decrease with increasing the overlap length, accompanied by an increase of peak currents of the second and third peaks. (iii) The peak-to-valley ratio $I_{p}/I_{v}$ for the first peak is nearly stationary when it is observed at sufficiently high voltages (or equivalently small overlap lengths) but decreases drastically when the peak position $U_{p}$ approaches 0.2–0.4 V (Table 1). The similar trends were observed previously for metallic armchair nanoribbons [14,15]. (iv) The initial slopes of all the $I$–$U$ curves at low voltage ($U \rightarrow 0$) are finite and depend non-monotonically on the overlap length. The linear piece of the
systems with the AA stacking at the overlap lengths of $L = 18\alpha_0$ when the position of the first peak approaches 0.2 V (Table 1). These results mean that the graphene layers with the AB stacking, which corresponds to the ground-state configuration, produces a more remarkable negative differential resistance effect. An opposite result was observed in the paper [14] for metallic armchair nanoribbons, where the peak-to-valley ratio for the AA stacking was much higher than that for the AB stacking. Comparable values of the peak-to-valley ratios on the order of 10–20 were obtained [14].

The drastic changes in the $I$–$U$ curves with changing the overlap length of the graphene layers allow to design graphene-based nanoelectromechanical memory cells. The graphene layers with slightly different overlap lengths can be considered as “0” and “1” states of the memory cell with very different conductances. For example, the ratio of conductances $G(L - \alpha_0)/G(L)$ for the systems with the AB stacking and difference in the overlap length of one unit cell $(L - \alpha_0 = \alpha_0(3n/2 - 1)$ and $L = 3n\alpha_0/2$ with integer $n = 4–18)$ exceeds 10 for $L = 9\alpha_0–21\alpha_0$ (Fig. 3). The positions of these peaks are virtually the same as of the first minima in the $I$–$U$ curves for $L = 3n\alpha_0/2$, which explains their origin. It is seen that the choice of voltages corresponding to peaks in Fig. 3 allows to build nanoelectromechanical memory cells with quite different conductances in the “0” and “1” states on the basis of graphene layers with the AB stacking and overlap lengths different only by one unit cell.

### 3.2. Conductance between graphene layers with argon spacer

The calculations of the $k_z = 0$ component of the tunneling current for the contact between graphene layers separated by the monolayer argon spacer revealed that below 1 V the current is reduced by three orders of magnitude as compared to the spacer-free case (Figs. 4 and 5). The same as for pristine graphene layers, a strong dependence of the $I$–$U$ curves on stacking of the graphene layers and overlap length is observed. Another factor that affects the $I$–$U$ curves in this case is the in-plane position of the argon layer with respect to the graphene layers (compare upper and lower panels of Figs. 4 and 5). This strong dependence of the conductance on atomic-scale displacements of any of the graphene and argon layers, different from the very weak dependence of the binding energy related to incommensurability of the layers [6], is a manifestation of the strong dependence of tunneling probability on the distance between atoms.

The similarity between the AA, MP1, AB1 and SP stackings and their strong difference from those for the AB2 and MP2 stackings extends to non-linear regions of the $I$–$U$ curves (Figs. 4 and 5). For position I of the argon layer, the curves for the AA, MP1, AB1 and SP stackings are no longer the same. Nevertheless, the pairs...
Table 1
Calculated low-voltage conductance $G(U \rightarrow 0)$, peak $I_p$, and valley $I_v$ currents, corresponding voltages $U_p$ and $U_v$, and peak-to-valley ratios $I_p/I_v$ for telescopic contacts between graphene layers with and without the monolayer argon spacer for different graphene layer stackings (SGL), overlap lengths $L$ and in-plane position of the argon spacer (ASP).

<table>
<thead>
<tr>
<th>SGL</th>
<th>ASP</th>
<th>$G(U \rightarrow 0)/G_0$</th>
<th>$U_p$, V</th>
<th>$I_p$, μA</th>
<th>$U_v$, V</th>
<th>$I_v$, μA</th>
<th>$I_p/I_v$</th>
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<tbody>
<tr>
<td>AA</td>
<td>I</td>
<td>6.029</td>
<td>1.071</td>
<td>37.096</td>
<td>1.857</td>
<td>3.751</td>
<td>9.891</td>
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<td></td>
<td></td>
<td>7.5</td>
<td>0.343</td>
<td>0.886</td>
<td>34.405</td>
<td>1.533</td>
<td>4.087</td>
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<tr>
<td></td>
<td></td>
<td>9.0</td>
<td>0.417</td>
<td>0.747</td>
<td>30.302</td>
<td>1.302</td>
<td>3.317</td>
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<td></td>
<td></td>
<td>12.0</td>
<td>0.488</td>
<td>0.562</td>
<td>22.724</td>
<td>0.979</td>
<td>2.334</td>
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<td></td>
<td></td>
<td>15.0</td>
<td>0.518</td>
<td>0.424</td>
<td>15.911</td>
<td>0.794</td>
<td>1.935</td>
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<tr>
<td></td>
<td></td>
<td>18.0</td>
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<td>0.377</td>
<td>10.919</td>
<td>0.655</td>
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<td>0.239</td>
<td>5.051</td>
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<td>27.0</td>
<td>0.440</td>
<td>0.192</td>
<td>3.549</td>
<td>0.424</td>
<td>1.198</td>
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<td>AA</td>
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<td>1.857</td>
<td>6.968</td>
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<td></td>
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<td>0.562</td>
<td>23.050</td>
<td>1.117</td>
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<td></td>
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<td>12.0</td>
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<td>11.500</td>
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<td>3.424</td>
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<td>0.239</td>
<td>2.370</td>
<td>0.424</td>
<td>1.345</td>
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<tr>
<td></td>
<td></td>
<td>18.0</td>
<td>0.054</td>
<td>0.121</td>
<td>0.901</td>
<td>0.218</td>
<td>0.698</td>
</tr>
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</table>

Graphene layers separated by argon spacer

| AA   | I     | 9.0  | 3.59 $10^{-4}$ | 0.724 | 25.540 |
|      |       |      | 4.04 $10^{-4}$ |       |        |
| MP1  |       |      | 2.99 $10^{-4}$ |       |        |
| AB1  |       |      | 3.14 $10^{-4}$ | 0.655 | 24.161 |
| SP   |       |      | 1.44 $10^{-4}$ | 0.308 | 2.177  | 0.539   | 1.699   | 1.282   |
| MP2  |       |      | 7.57 $10^{-5}$ | 0.401 | 1.650  |
| AA   | II    | 9.0  | 4.29 $10^{-4}$ | 0.609 | 27.559 | 0.955   | 21.289  | 1.294   |
|      |       |      | 3.80 $10^{-4}$ | 0.609 | 26.774 | 0.909   | 20.940  | 1.279   |
| MP1  |       |      | 3.64 $10^{-4}$ |       |        |
| AB1  |       |      | 2.98 $10^{-4}$ |       |        |
| SP   |       |      | 1.08 $10^{-4}$ | 0.447 | 2.460  |
| MP2  |       |      | 2.49 $10^{-4}$ | 0.308 | 4.124  | 0.493   | 3.699   | 1.115   |
| AA   | I     | 7.5  | 1.01 $10^{-3}$ | 0.516 | 18.545 | 1.025   | 9.669   | 1.918   |
|      |       |      | 9.53 $10^{-4}$ | 0.470 | 18.090 | 0.979   | 10.267  | 1.762   |
| MP1  |       |      | 9.70 $10^{-4}$ | 0.447 | 17.848 | 1.002   | 9.571   | 1.865   |
| AB1  |       |      | 1.08 $10^{-3}$ | 0.377 | 19.688 | 0.955   | 9.829   | 2.003   |
| SP   |       |      | 3.84 $10^{-5}$ |       |        |
| MP2  |       |      | 2.91 $10^{-5}$ |       |        |
| AA   | II    | 7.5  | 7.64 $10^{-4}$ | 0.516 | 15.938 | 0.955   | 9.011   | 1.769   |
|      |       |      | 1.13 $10^{-3}$ | 0.424 | 22.326 | 0.886   | 13.529  | 1.650   |
| MP1  |       |      | 1.14 $10^{-3}$ | 0.424 | 22.248 | 0.886   | 13.413  | 1.659   |
| AB1  |       |      | 8.01 $10^{-4}$ | 0.447 | 15.645 | 0.955   | 7.962   | 1.965   |
| SP   |       |      | 1.17 $10^{-4}$ | 0.470 | 3.154  | 0.770   | 2.209   | 1.428   |
| MP2  |       |      | 1.31 $10^{-4}$ | 0.539 | 4.659  | 0.840   | 3.286   | 1.418   |

Fig. 3. Calculated ratio of conductances $G(L – a_0)/G(L)$ for spacer-free telescopic contacts between graphene layers with the AB stacking and overlap lengths of $L – a_0 – a_0(3n/2 – 1)$ and $L – 3na_0/2$ with integer $n = 4–16$ as a function of voltage $U$ (in V).
the peak-to-valley ratio up to 2 (Fig. 5, Table 1). The peak-to-valley ratio slightly decreases upon shifting the argon layer to position II.

Therefore, the atomistic simulations demonstrate the tunneling current through overlapping graphene layers is substantially suppressed even when the argon spacer is just one atomic layer thick. Much stronger suppression should be expected upon incorporation of two and more atomic layers of argon between the graphene layers. The tunneling current through telescopic contacts between graphene layers both with and without the argon spacer is found to be very sensitive to the relative position of the layers at the atomic scale. The both contacts with and without the argon spacer exhibit negative differential resistance, though the effect is much more pronounced in the pristine system.

3.3. Capacitance of graphene layers with argon spacer

Let us analyze the capacitance of the telescopic contact between graphene layers with a dielectric spacer taking into account the quantum contribution \( U_d \) into the potential difference \( U \) between the electrodes corresponding to the relative shift in the chemical potentials of the graphene layers in the bilayer region. Application of a negative bias \(-U/2\) to the top graphene layer and a positive bias \(+U/2\) to the bottom graphene layer leads to accumulation of electrons and holes, respectively. This results in shifts of the chemical potentials \( E_F \) of the graphene layers in the bilayer region by \( \pm U_0/2 \) (analogously to \( p-n \) junctions in semiconductor devices [49]). The total potential difference \( U \) between the electrodes is, therefore, represented as

\[
U = U_b + U_q,
\]

where \( U_b = Qd/A\varepsilon_0 \) is the classical potential difference related to the electric field between the graphene layers separated by the dielectric spacer with the relative permittivity \( \varepsilon_r \). \( Q \) is the absolute value of the charge accumulated at each graphene layer, \( \varepsilon_0 \) is the electric constant and \( A \) is the overlap area of the graphene layers \( (A = LW, \text{where } W \text{ is the width of graphene layers and } L \text{ is the overlap length shown in Fig. } 1) \). Such an approach is analogous to the one used by Luryi for consideration of quantum capacitance devices comprising the two-dimensional electron gas in a quantum well [39] and is applicable to the contact of any two-dimensional conductors separated by a dielectric spacer.

In double-layer graphene with the dielectric spacer the 2D densities of electrons \( (n_{2d}) \) in the top graphene layer and holes \( (p_{2d}) \) in the bottom graphene layer are expressed via their chemical potentials \( (E_F + eU_q/2) \) and \( (E_F - eU_q/2) \) as [38]

\[
n_{2d}(U_q) = \frac{2}{\pi \hbar^2 v_F^2} \int_{-\infty}^{\infty} |E| f_{1d}(E - E_F - eU_q/2) \, dE,
\]

\[
p_{2d}(U_q) = \frac{2}{\pi \hbar^2 v_F^2} \int_{-\infty}^{0} |E| (1 - f_{1d}(E - E_F + eU_q/2)) \, dE,
\]

where \( v_F = 8.4 \cdot 10^5 \, \text{cm/s} \) is the Fermi velocity of carriers in graphene. These equations, Eq. (3) and the condition of electrical neutrality with account of charged impurities implicitly relate the charge \( Q \) and applied voltage \( U \) and allow to calculate the total capacitance \( C \) of double-layer graphene in the general case.

Pure graphene is a zero-gap semiconductor [50], thus in the absence of the external electric fields and charged impurities the Fermi level goes through Dirac points, which corresponds to \( E_F = 0 \) in Eq. (4). In this case, the charge \( Q \) as a function of applied voltage \( U \) is found from equality \( n_{2d} = p_{2d} = Q/eA \) of 2D densities [38] of electrons and holes in the negatively and positively biased layers, respectively.

The total reciprocal capacitance is, therefore,

\[
C^{-1} = \frac{d(U_b + U_q)}{dQ} = C_b^{-1} + C_q^{-1} = \frac{d}{\varepsilon_s e_0 A} + \frac{\pi e^2 h^2}{2 e k_B T A} \ln^{-1} \left( 1 + \exp \left( \frac{eU_q}{2k_BT} \right) \right),
\]

Fig. 4. Calculated current \( I \) (in nA) as a function of voltage \( U \) (in V) for telescopic contacts of graphene layers of different stacking separated by the monolayer argon spacer with in-plane positions I and II (upper and lower panels, respectively) for the overlap length \( L = 9.5a_0 \).

Fig. 5. Calculated current \( I \) (in nA) as a function of voltage \( U \) (in V) for telescopic contacts of graphene layers of different stacking separated by the monolayer argon spacer with in-plane positions I and II (upper and lower panels, respectively) for the overlap length \( L = 7.5a_0 \).
where $C_b = \varepsilon \varepsilon_0 A/d$ is the geometric capacitance and $C_q$ is the quantum capacitance; $U_{ab} = U - U_b = -en_2d(U_0 d)/\varepsilon \varepsilon_0$, which taking into account (4) gives explicit dependence $U(U_0)$. It should be noted that the quantum capacitance term $C_q$ for double-layer graphene in Eq. (5) differs from quantum capacitance of single-layer graphene [37,38] only by the factor of 1/2 in the exponent.

On condition that $T \ll eU/k_B$ Eq. (4) takes the form

$$n_{2d} = p_{2d} = \frac{(eU_0/2)^2}{\pi \hbar^2 \tau_f^2} = \frac{Q}{eA},$$

and the total capacitance from (5) can be expressed via

$$C = \frac{dQ}{dU} = \frac{\varepsilon \varepsilon_0 A}{d} \left( 1 - \left( 1 + \frac{e^2 d}{\pi \hbar^2 \tau_f \varepsilon \varepsilon_0} U \right)^{-1/2} \right).$$

The permittivity $\varepsilon_r$ of the argon spacer between graphene layers is estimated by Clausius–Mossotti formula [51]

$$\varepsilon_r = 1 + \frac{N_{Ar} \tau_{Ar}}{1 - N_{Ar} \tau_{Ar}/3},$$

where $N_{Ar} = 1.86 \cdot 10^{22} \text{ cm}^{-3}$ is the argon concentration for the spacer of thickness $d = 6.82 \text{Å}$ shown in Fig. 1 and $\tau_{Ar}$ is the argon polarizability ($\tau_{Ar} = 11.1$ in atomic units [52]). The calculated values of the permittivity of the argon spacer and geometric capacitance per unit area are $\varepsilon_r = 1.44$ and $C_b/A = 1.87 \mu \text{F/cm}^2$.

The voltage dependence of the total capacitance per unit area for the telescopic contact between graphene layers separated by the monolayer argon spacer calculated using Eq. (5) is shown in Fig. 6. Three regions can be distinguished in this dependence. At high voltages, the total capacitance per unit area tends to its classical value $C_b/A$ (for example, at voltage 3 V, the decrease of the total capacitance due to the quantum contribution is less than 20%). For intermediate voltages 0.1–3 V, the quantum contribution leads to the considerable decrease in the total capacitance upon decreasing voltage, however the temperature dependence of the total capacitance is not essential, i.e. Eq. (6) is adequate. For small voltages less than 0.1 V, the total capacitance considerably depends on temperature. For instance, at voltage 0.01 V the total capacitance per unit area $C/A = 0.23 \mu \text{F/cm}^2$ and $C/A = 0.0082 \mu \text{F/cm}^2$ at temperature 300 and 0 K, respectively.

Let us estimate the $RC$ time constant for the telescopic contact between graphene layers with the dielectric spacer. In our atomistic simulations we neglected the effect of graphene edges, that is realistic for nanoribbon widths above $W > 100 \text{ Å}$ [42]. At input voltage $U = 0.1 \text{ V}$ the assumption that only one conductance channel contributes to the conductance of a single graphene layer is valid for nanoribbon widths $W < 2\pi \hbar v_F/(eU) = 400 \text{ Å}$. Therefore, for an argon spacer in position II, $T = 300 \text{ K}$, input voltage $U = 0.1 \text{ V}$, nanoribbon width $W = 100 \text{ Å}$ and overlap length $L = 22.19 \text{ Å}$ the conductance is $G = 4.29 \cdot 10^{-4} \text{ G}_0 = 3.32 \cdot 10^{-8} \text{ Ohm}^{-1}$ and the capacitance is $C = LW \cdot 0.51 \mu \text{F/cm}^2 = 1.13 \cdot 10^{-15} \text{ F}$. Correspondingly, we estimate the time constant to be as small as $RC = C/G = 3 \text{ ps}$.

4. Conclusions

The non-equilibrium Green function formalism on top of density functional theory calculations has been used to analyze the $k_x = 0$ component of the tunneling conductance of telescopic contacts between pristine graphene layers with the AA and AB stackings and between graphene layers with the argon spacer for a set of stackings of the graphene layers and relative positions of the spacer. We have found that for the contacts between pristine graphene layers, the $I$–$U$ curves exhibit considerable oscillations for any overlap length $L$ between the layers in the considered range between $L = 6a_0$ and $L = 27a_0$. The most pronounced negative differential resistance effects are observed for the contacts with the overlap length of $L = 3na_0/2$ with integer $n$. The following main results have been obtained for the contacts with these overlap lengths: (i) the maximal values of the peak-to-valley ratio of 8–10 are found for the graphene layers with the AB stacking and the overlap lengths between $L = 6a_0$ and $L = 15a_0$, whereas for the AA stacking, the values of the peak-to-valley ratio are 3–5 for these overlap lengths, (ii) the initial slopes of the $I$–$U$ curves depend non-monotonically on the overlap length and are within 0.25–0.52$G_0$ for the AB stacking, whereas for the AA stacking, the low-voltage conductance (at $U \to 0$) reaches $G_0$ at the overlap lengths $L = 9a_0$ and $21a_0$ and virtually vanishes for the overlap length of $L = 18a_0$.

The high values of the peak-to-valley ratio offer promise for application of telescopic contacts between pristine graphene layers in various nanoelectronic devices. Moreover, the performed calculations show that the ratio of conductances for overlap lengths different only by one lattice constant can exceed 10. It is proposed that these changes of the conductance at atomic scale changes of the overlap length can be applied for development of graphene-based nanoelectromechanical memory cells and sensors.

For the contacts between graphene layers separated by the monolayer argon spacer, the current at voltage below 1 V is found to be three orders of magnitude less in comparison with the case without the spacer. The calculated values of low-voltage conductance range from $2.9 \cdot 10^{-9} G_0$ to $1.1 \cdot 10^{-7} G_0$. Both stacking of the graphene layers and relative position of the argon spacer are found to influence considerably the $I$–$U$ curves. The negative differential resistance effect is found only for some of the stackings of the graphene layers and relative positions of the argon spacer with the maximal values of the peak-to-valley ratio about 2. Much stronger suppression of the current should be expected for the spacers consisting of two and more atomic layers of argon and such contacts can be used for elaboration of nanocapacitors.

The expression for the capacitance has been derived for the telescopic contact between graphene layers separated by the monolayer argon spacer as a function of voltage taking into account the quantum contribution. It is found that the quantum contribution leads to the considerable decrease in the total capacitance upon decreasing the voltage below 3 V, while the temperature dependence of the total capacitance becomes important at small voltages below 0.1 V. Based on the conductance and

![Fig. 6. Calculated by (5) capacitance per unit area $C/A$ for the telescopic contact between graphene layers with the argon spacer as a function of applied voltage $U$ at temperatures $T = 300$ K (1), 100 K (2), 30 K (3), and 0 K (4); by (6). The dashed line corresponds to the geometric capacitance $C_b/A = \varepsilon \varepsilon_0 /d = 1.87 \mu \text{F/cm}^2$ taking into account (7).](image)
capacitance calculations, the RC time constant for this contact is estimated to be about 3 ps. Such a small time constant of double-layer graphene with the monolayer argon spacer makes possible elaboration of fast-response nanoelectronic and nanoelectromechanical devices.

Acknowledgments


References


