Electric Contact Resistance in Graphite-Graphene contacts from ab initio methods

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Abstract. We study the ballistic transmission and the contact resistance ($R_c$) of a graphite-graphene contact in a top contact geometry from first principles. We find that the calculated $R_c$’s depend on the amount of graphene-graphite overlap, but quickly saturate for transfer lengths of the order of 20 Å. For contacts overlapping more than this transfer length, the $R_c$ can be lower than the 100 Ω·µm mark. On the other hand, edge graphite-graphene contacts are expected to have exceptionally low contact resistance.

Keywords: DFT, graphene, graphite, contact resistance, ballistic injection
1. Introduction

Throughout the last fifteen years, graphene has demonstrated its capabilities as a new material with its extraordinary properties [1]. Although the lack of bandgap forbids the use of this material for digital applications, its properties are very well suited for analog radiofrequency devices [2]. However, before graphene can be widely adopted, several difficulties must be overcome. In particular, one of the limitations for the use of graphene in analog electronics is the high contact resistance when metal-graphene contacts are fabricated, while an upper bound of $100 \ \Omega \cdot \mu m$ would be desirable [2, 3].

Theoretical work has been carried out for contact resistance between graphene and other metals. For instance, Chaves et al. [4] created a model for contact resistance between metal and graphene in a top contact-like geometry. The metal-graphene edge contact geometry, in spite of its vanishing contact overlap, has also been proven to be at least as good as some of the top contact geometries [5].

Despite their obvious similar structural properties, the use of graphite as an electrode for contacting graphene has received much less attention. The Lieber group has synthesized monolithic graphene-graphite structures, obtaining specific contact resistivities in the range of 700-900 $\Omega \cdot \mu m$, better than similarly fabricated Cr/Au junctions [6]. Also, Chari et al. measured the resistivity of rotated graphite-graphene contacts, obtaining specific contact resistivities as low as $133 \ \Omega \cdot \mu m$ for holes and $200 \ \Omega \cdot \mu m$ for electrons [7].

The objective of this paper is to demonstrate the viability of graphite-graphene contacts and show their fundamental limits. To this purpose, we describe in section 2.1 the used geometry, followed by the computational methodology in section 2.2. We show in section 3 that this yields results well below the upper contact resistance limit for certain values of the overlap area and doping level. Then, an eigenchannel analysis give us more insight about the scattering processes in the interface between the graphite substrate and the graphene. This analysis brings us to the conclusions, in section 4, that the graphite-graphene interface presents more of an area effect than metal-graphene contacts [8], but still with very small transfer lengths of $\sim 20$ Å.

2. Methodology

2.1. Geometry Description

We will focus on top contact geometries because they are the most easily fabricated. In Figure 1, a ball-and-stick representation of the structure of the graphite-graphene contact is displayed. As usual in ballistic transport calculations, there are three differentiated zones: a left electrode—where the electrons are injected—, a scattering zone through which electrons will pass or reflect, and a right electrode into which electrons that were not backscattered will arrive. The electrodes are semi-infinite and
we will be studying a single graphite-graphene contact.

In this work we studied the effect of overlap length of graphene over the graphite bulk. The numbers in the scattering zone indicate the different number of overlapping C-pairs providing the contact between graphene and the graphite substrate. We also studied the case where the last graphite layer turns into the graphene sheet, which we interpret as an edge graphite-graphene contact [5].

Structures were relaxed from first-principles using the SIESTA code [9], an efficient implementation of the Density Functional Theory (DFT) using localized pseudo-atomic orbitals. Transport calculations were carried out using TRAnSiESTA [10, 11], which implements the Non-Equilibrium Green’s Function formalism under the DFT as well.

2.2. Computational Details

Calculations were performed with a double-\(\zeta\) polarized basis set, using norm-conserving pseudopotentials of the Troullier-Martins type [12]. Numerical integrals were carried out on a discretization mesh equivalent to a cutoff of 250 Ry, which provides total energies for graphene and graphite converged to the few meV range.

The Generalized Gradient Approximation (GGA) in the parametrization of Perdew-Burke-Ernzerhof (PBE) [13] was used to describe exchange-correlation effects. GGA accurately describes the lattice parameter of graphene, but underestimates the interlayer distance [14]. This, of course, can be corrected with the use of van der Waals (vdW) type functionals. It has been demonstrated that the parametrization of Dion-Rydberg-Schröder-Langreth-Lundqvist (DRSLL) of the vdW interaction provides a good description of the interlayer distance while slightly overestimating the in-plane lattice constant [14, 15]. Thus, we have carried all structural relaxations with the vdW-DRSLL functional until residual forces were less than 0.04 eV/Å. However, since the GGA is more computationally efficient, we have used the PBE functional for
transport calculations. For a fixed geometry, the two functionals yield very similar energy dispersions. In Figure 2 the energy bands for bulk graphite are shown, comparing the two functionals. Around the Fermi level, the energy difference between the two curves is negligible, and therefore it is concluded that both functionals provide a good description of the energy of the system. In particular, it must be stressed that the dispersion along $z$, which is closely related to the interlayer coupling (i.e. transport) of the electronic states, is not affected by the passage from the vdW-DRSLL functional to PBE.

3. Results

From the TransSiesta calculations we obtain the energy-resolved specific conductance (i.e. the conductance per unit of tranverse length) for the different structures.

In Figure 3 the specific conductance, in units of $G_0 = e^2/h$ over the transverse length of the calculation cell ($a_t = 2.484 \text{ Å}$), is shown for the graphite-graphene top contact for different values of the overlap (cf. Figure 1) and the edge contact. The pristine graphene case—which provides the quantum limit for the conductance of the whole structure—is shown as well for reference. We note that the difference between the
edge contact and the pristine graphene limit is very small, suggesting that an edge, or large overlap, contact between graphite and graphene would provide a very low contact resistance.

Regarding the varying amount of overlap, we observe that, contrary to the metal-graphene case [8], there is a noticeable monotonic dependence of the conductance on the overlap width. This is a reflection of the weaker substrate-graphene $p_z - p_z$ coupling compared to the stronger $d - p_z$ coupling in metal substrates. Despite the weaker coupling, a relatively narrow overlap of $\sim 20$ Å suffices to achieve a conductance similar to metallic substrates (see Ref. [8]).

We now turn our attention to the (specific) contact resistance. It can be extracted from the calculated conductances of the whole graphite-graphene structure, $G_{gg}^{-1}(E_{FL}, V)$ and the pristine graphene layer, $G_g^{-1}(E_{FL}, V)$:

$$R_c(E_{FL}, V) = G_{gg}^{-1}(E_{FL}, V) - G_g^{-1}(E_{FL}, V).$$

Now, in order to calculate the zero bias contact resistance considering a thermal and gaussian electron-hole (e-h) puddle [16] broadening, we used:

$$R_c(E_F) =$$
Figure 4. Specific Contact Resistance. (a) Thermal broadening at 300K plus electron-hole puddle of 5 meV. (b) Thermal broadening at 300K plus electron-hole puddle at 50 meV.

\[
+ k_B T \left( \int \int \frac{\exp \left[ \frac{(E - E')}{k_B T} \right]}{1 + \exp \left[ \frac{(E - E')}{k_B T} \right]^2} G_{gg}(E) \ w(E' - E_F; \eta) \ dE \ dE' \right)^{-1}
\]

\[
- k_B T \left( \int \int \frac{\exp \left[ \frac{(E - E')}{k_B T} \right]}{1 + \exp \left[ \frac{(E - E')}{k_B T} \right]^2} G_g(E) \ w(E' - E_F; \eta) \ dE \ dE' \right)^{-1}
\]

where \( w(E' - E_F; \eta) \) is the gaussian broadening function and \( \eta \) the broadening parameter, taken to be 50 meV for SiO₂ substrates \[16\].
The obtained specific contact resistance results are shown in Figure 4. The contact resistance at the Dirac point (undoped graphene) strongly depends on the amount of overlap, with the widest overlaps getting close to the 100 $\Omega \cdot \mu m$ value, especially in the case of high e-h puddle broadening. The values of the contact resistance at higher/lower values of the Fermi energy (i.e. doped samples) rapidly decrease below the landmark value of 100 $\Omega \cdot \mu m$. These values are represented, for carrier concentrations, as a function of the graphene-graphite overlap, $L_c$, in Figure 5, where we see that the $R_c$ values effectively saturate for $L_c > \sim 20$ Å.

### 3.1. Current path analysis

In order to assess our conclusions and gain insight into the graphite-graphene coupling, an eigenchannel analysis has been carried out using the Inelastica package [17]. In Figure 6, current is represented by arrows for each atom in the geometry, represented by translucent balls, and with the arrow thickness proportional to the magnitude of the current. Each plot corresponds to a wave vector $k_{\perp}$, perpendicular to the plane of the representation, different energy of the incoming particle and/or different overlap, resulting in a transmission coefficient, $T$.

Figures 6.(a)-(d) show the current lines for electrons in the cases with overlap 2, 5 and 9, with $k_{\perp}$ and $E$ chosen in such a way that high $T$’s are obtained. We can see that, as the overlap increases, injection becomes more distributed across the overlapping area, in opposition to the case of metal-graphene contacts [8], where only 1-2 metal-carbon bonds contributed to injection. Notwithstanding that, when only a very small area is
Figure 6. Current paths for different contact lengths for both carriers at fixed $k_\perp = 0.660 \pi / a_\perp$.
(a) Overlap 2 for $e$: $E = 0.105 \text{ eV}, T = 0.99016$
(b) Overlap 5 for $e$: $E = 0.105 \text{ eV}, T = 0.48904$
(c) Overlap 5 for $h$: $E = -0.105 \text{ eV}, T = 0.51762$
(d) Overlap 9 for $e$: $E = 0.105 \text{ eV}, T = 0.15161$

available for injection (e.g. overlap 2), high transmission is still achievable, with nearly complete injection to graphene taking place through the last two pairs.
4. Discussion and summary

Of course, any graphite-graphene contact will eventually need to be contacted to metal leads. One set of measurements of the contact resistance of metal-multilayer graphene (1,3,4,~50,~100 layers) did not find any strong dependence on the number of layers, which was attributed to only the top layer or two of a graphene stack playing a role in the contact formation [18]. It is expected that a different fabrication procedure promoting the formation of edge metal-C bonds, such as demonstrated in Ref. [19], would significantly decrease the metal-graphite contact resistance.

In conclusion, we have shown that graphite-graphene contacts provide a promising route towards the reduction of the contact resistance in graphene FET channels. Although transfer lengths are significantly higher than in metal-graphene contacts, their magnitudes are still quite small, at ~20 Å. In addition, edge graphite-graphene contacts are expected to have exceptionally low contact resistance.

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