# Physics Model of the Contact Resistivity of Metal-Graphene Junctions

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## ABSTRACT

While graphene-based technology shows great promise for a variety of electronic applications, including radio-frequency devices, the resistance of the metal-graphene contact is a technological bottleneck for the realization of viable graphene electronics. One of the most important factors in determining the resistance of a metal-graphene junction is the contact resistivity. Despite the large number of experimental works that exist in the literature measuring the contact resistivity, a simple model of it is still lacking. In this paper we present a comprehensive analytical model for the contact resistivity of these junctions, based on the Bardeen Transfer Hamiltonian method. This model unveils the role played by different electrical and physical parameters in determining the specific contact resistivity, such as the chemical potential of interaction, the work metal-graphene function difference, and the insulator thickness between the metal and graphene. In addition, our model reveals that the contact resistivity is strongly dependent on the bias voltage across the metal-graphene junction. This model is applicable to a wide variety of graphene-based electronic devices, and thus is useful for understanding how to optimize the contact resistance in these systems.

KEYWORDS: graphene, contacts, contact resistance

While graphene has emerged as a promising material for future electronic devices, it is often the metal-graphene (MG) contact resistance that dominates the performance of the device.[1] For example, although a high carrier mobility of ~10,000 $cm^2V^{-1}s^{-1}$ has been reported on $SiO_2$,[2-5] the small density of states (DOS) of graphene near the Dirac point can suppress current injection from the metal, resulting in high contact resistivity at the MG interface, which limits the total performance of a graphene transistor.[6,7] In radio-frequency circuits, the key figure of merit is the maximum frequency of oscillation $f_{max}$, which is the frequency at which the power gain drops to unity. This parameter turns out to be very sensitive to the contact resistance, especially in the absence of full current saturation.[6] Therefore, the metal-graphene contact resistance is a critical component of graphene-based devices, and controlling its properties is a prerequisite for device optimization.

In two- or three-terminal semiconductor devices, the contact resistance is commonly described with the transmission line model,[7-9] $R_c = \sqrt{\rho_c R_s}\coth\left(L/L_T\right)/W$, where $L$ ($W$) is the length (width) of the contact, $R_s$ is the semiconductor sheet resistance under the metal, $\rho_c$ is the specific contact resistivity of the metal-semiconductor junction, and $L_T$ is the transfer length, which is the characteristic length over which current injection occurs between the semiconductor and the metal. In the diffusive regime, this parameter is related to the specific contact resistivity through the relation $L_T = \sqrt{\rho_c / R_s}$, while a more detailed description of $L_T$ can be found in the work of Xia *et al.*[10] In any case, it is evident that $\rho_c$ plays a crucial role in the magnitude and length dependence of the metal-graphene contact resistance.

There have been many experimental studies of the MG junction in the scientific literature.[11-15] The relevant role played by graphene in metal-semiconductor junctions has also been evidenced by the recent experimental work of Byun *et al.*,[16] where a true Ohmic contact in *Ni-Si* junctions has been demonstrated by using an interfacial graphene layer to lower the Schottky barrier. In addition, the role of metallic leads in determining the transport properties of graphene-based junctions has been addressed by several theoretical studies.[10,17-19] However, a direct and comprehensive model of carrier transport between a two-dimensional graphene sheet and a three-dimensional metal is still lacking. Such a model is needed to understand the intrinsic factors that control the magnitude of the specific contact resistivity, and thus to understand how to optimize the graphene-metal contact resistance.

We address this problem by formulating an analytical model of the tunneling current and contact resistivity of the metal-insulator-graphene (MIG) heterostructure, from which the MG junction can be seen as a particular case. Our model is based on the Bardeen Transfer Hamiltonian (BTH) method,[20,21] which allows us to split the metal-graphene system into separate metal-dipole and dipole-graphene subsystems with known Hamiltonians. In the framework of the BTH method, the probability of elastic tunneling is calculated using Fermi's golden rule. This gives a quantitative estimate of the coupling between the metal and graphene states, allowing us to obtain analytical formulas for both the tunneling current and contact resistivity as a function of the applied voltage. This model allows us to identify the key parameters in determining the metal-graphene contact resistivity, and can also be used in larger-scale simulations of graphene-based electronic devices.

## RESULTS AND DISCUSSION

We start with a description of the electrostatics of the MIG heterostructure. Represented in Fig. 1a, it consists of a metal (M) electrode treated as an equipotential with voltage $V$ and work function $W_m$, and a grounded graphene layer (G) with work function $W_g$. These are separated by an intermediate insulator (I) layer with permittivity $\varepsilon = \varepsilon_r\varepsilon_0$ and thickness $d$, where $\varepsilon_r$ is the relative permittivity of the insulator and $\varepsilon_0$ is the permittivity of free space. In the MIG band diagram of Fig. 1b, $E_{Fg}$ is the graphene Fermi energy, $E_D$ is its Dirac point energy, and $E_{Fm}$ is the metal Fermi energy. An interfacial potential step $\Delta V$ is developed across the (I) layer due to charge transfer and the chemical interaction between the graphene and the metal.[22]

With respect to the Dirac point, the shift of the graphene Fermi level is defined as $\Delta E = E_D - E_{Fg}$ and a relationship between the bias voltage $V$ and $\Delta E$ can be obtained from a voltage (potential energy) loop around the MIG band diagram,

$$\Delta E = W_m - W_g - e\Delta V - eV. \tag{1}$$

The interfacial potential step $\Delta V$ can be expressed as $\Delta V = \Delta_{tr} + \Delta_{ch}$, where $\Delta_{tr}$ results from charge transfer between the (M) and the (G) and $\Delta_{ch}$ is the chemical potential describing the short-range interaction from the overlap of the (M) and (G) wavefunctions. According to the model of Khomyakov *et al.*,[23] the value of $\Delta_{tr}$ depends strongly on the separation distance $d$ (Fig. 1b) and becomes negligible for $d \geq 4$ nm.

To model the electron transfer contribution, $\Delta_{tr}$, we use a planar capacitor model such that $\Delta_{tr}(\Delta E) = z_d Q_{net}/e$, where $z_d$ represents the effective distance between the charge sheets of the (M) and (G) layers (Fig. 2b) and $Q_{net} = e(p - n)$ is the net charge sheet density within the graphene.[24] In addition, we have assumed charge neutrality for the structure (i.e., the electric field goes to zero outside the structure) and thus $Q_{net} = -Q_m$, where $Q_m$ is the surface charge density in the metal. Finally, although in MG junctions the electrodes permit transmission, the reflection probability is nearly unity because the electrons see a barrier height ~4 eV and thus the MG junction can be treated as a mesoscopic capacitor.[25] Substituting the conventional expression[23] for $p - n$ into Eq. 1 yields the relation

$$\alpha f(\Delta E/k_B T) + \Delta E + eV - eV_D = 0, \tag{2}$$

where we have defined

$$eV_D = W_m - W_g - \Delta_{ch}. \tag{3}$$

The physical meaning of $V_D$ is that of the voltage applied to the metal needed to align the graphene Fermi level to the Dirac point. Also, in Eq. 2, $\alpha = 2e^2 z_d / \left(e\pi\hbar^2 v_f^2\right)$ and $f(x) = (k_B T)^2 [F(-x) - F_1(x)]$, with $F_1(x)$ the Fermi-Dirac integral of order 1. Equation (2) has a closed-form analytical solution at zero temperature and should be solved

numerically for T > 0. As $T \rightarrow 0$ the $f$ function reduces to $f = \pm\Delta E^2/2$, where the upper (lower) sign applies for $\Delta E < 0$ ($\Delta E > 0$). Equation 2 then becomes a simple quadratic equation for $\Delta E$ whose solution is

$$\Delta E = \pm \frac{\sqrt{1 + 2\alpha\varepsilon|V - V_D|} - 1}{\alpha}. \tag{4}$$

In Eq. 4 the upper sign applies for $V \leq V_D$ and the lower sign for $V > V_D$. This result holds not only for MIG structures but also for the MG junction since, as was mentioned previously, a MG junction can be seen as a particular case of the MIG structure where the layer (I) represents a dipole layer with $d_{eq}$ the equilibrium separation between (M) and (G).[23,26,27] In this work, we model $z_d = d - d_0$ with $d_0 = 0.24$ nm. We have used $d = d_{eq}$, $\varepsilon_r = 1$ and $\Delta_{ch} > 0$ for MG junctions and $d > d_{eq}$, $\varepsilon_r = 4$ and $\Delta_{ch} = 0$ for MIG structures.

In Fig. 3 we show, for $T \rightarrow 0$, the behavior of $\Delta E$ as a function of the bias voltage $V$ in a MG junction for five different metals. At $V = 0$, metals such as Cu, Ag, and Al dope the graphene n-type while Pt and Au electrodes result in p-type graphene. For the sake of comparison, we have also shown $\Delta E$ for a hypothetical metal X with work function $W_m = W_g = 4.5$ eV and $\Delta_{tr} = 0$, so that the graphene is undoped at $V = 0$. Given the weak dependence of Eq. 2 on the temperature, at $T = 300$K the curves represented in Fig. 3 don't change significantly except for the slopes near $\Delta E = 0$. Therefore, Eq. 4 is a very good approximation to Eq. 2.

For the results shown in Fig. 3 we have not included metals such as Co, Ni, Pd, or Ti, typically used as metallic contacts, because those metals strongly disturb the graphene band structure. In particular, the characteristic linear dispersion of graphene at the K-point is destroyed[23] and our model fails. Furthermore, a model for the $\Delta_{ch}$ term has not been established with these metals. However, we can apply our model to these metals in MIG structures, as will be shown later. This is because we assume that the insulator layer doesn't modify the graphene linear dispersion relation[28] and the $\Delta_{ch}$ term is considered to be zero. The solutions for Eq. 2 ($T > 0$) and Eq. 4 ($T = 0$) will be used later for the calculation of the tunneling current.

The specific contact resistivity $\rho_c$ of the MG junction is defined as $\rho_c = \left(dJ/dV\right)^{-1}\Big|_{V=0}$, where $J$ is the tunneling current density between the metal (M) and the graphene (G) across the dipole layer and $V$ is the voltage applied to the metal.[7,29] In the Methods section we show how to calculate the tunneling current density from the BTH approach,[20,21] starting from the expression for the tunneling current

$$I = g_S g_V \sum_{g,m} \left\{ \Gamma_{gm} f_g\left(E_g\right)\left[1 - f_m\left(E_m\right)\right] - \Gamma_{mg} f_m\left(E_m\right)\left[1 - f_g\left(E_g\right)\right] \right\}, \tag{5}$$

where the subscripts $g$ and $m$ label the states in the (G) and (M) electrodes with energies $E_g$ and $E_m$, respectively, $g_S$ is the electron spin degeneracy, $g_V$ is the valley degeneracy, and $\Gamma_{gm}$ and $\Gamma_{mg}$ refer to the tunneling rates for electrons moving from $g \rightarrow m$ and $m \rightarrow g$, respectively. Finally, $f_g$ and $f_m$ are the Fermi occupation factors for the electrons. The tunneling rates are given by Fermi's golden rule as

$$\Gamma_{gm} = \Gamma_{mg} = \frac{2\pi}{\hbar}\left|M_{gm}\right|^2 \delta\left(E_g - E_m\right), \tag{6}$$

where

$$M_{gm} = \frac{\hbar^2}{2m_0}\iint\left(\Psi_g^* \frac{d\Psi_m}{dz} - \Psi_m^* \frac{d\Psi_g}{dz}\right)dS \tag{7}$$

is the matrix element for the transition, with $m_0$ the electron mass in the (I) layer. The terms $\Psi_g(\mathbf{r}, z)$ and $\Psi_m(\mathbf{r}, z)$ represent the (G) and (M) electron wavefunctions, respectively, and their explicit forms are shown in the Methods section. From the complex exponential dependence of the in-plane wavefunctions, part of the integral of Eq. 7 transforms into the delta function $\delta(\mathbf{k}_g - \mathbf{k}_m)$ when the contact area is large enough, implying conservation of the in-plane momentum **k**. On the other hand, the energy delta function in Eq. 6 guarantees that only energy-conserving tunneling processes are possible.

After some manipulation of Eq. 5, described in the Methods section, Fig. 4a shows, at $T$ = 300K, the magnitude of the tunneling current density of the MG junction as a function of $V$ for Pt, Au, Cu, Ag, and Al metal electrodes, with work functions, Fermi energies, and equilibrium separation distances given in Table 1. The current-voltage (*I*-*V*) relationship of a MG junction or a MIG structure can be understood by considering the possible locations of the metal and graphene Fermi levels around the graphene Dirac point, as illustrated in Fig. 2 assuming $\Delta E > 0$ at $V = 0$, as is the case for a Pt contact. The applied voltage $V$ changes the relative difference between the Fermi levels on each side of the junction according to $E_{Fg} - E_{Fm} = eV$. If $V > 0$ a positive current will flow from the graphene to the metal via tunneling across the (I) layer. A special situation arises when the applied voltage drives the graphene Fermi level to perfect alignment with the Dirac point, resulting in $\Delta E = 0$. We have labeled such a bias as $V_D$. Analogously, if $V < 0$ a negative tunneling current will flow across the (I) layer from the metal to the graphene, and there will be a bias that aligns the metal Fermi level with the graphene Dirac point. We have labeled this bias as $V_C$, which occurs when $eV = -\Delta E$. At these critical biases, the low graphene DOS at the Dirac point pinches off the junction and the current changes very little with changes in the applied bias. This can be seen, for example, in the *I*-*V* curve of Pt at a bias voltage of $V$ = 0.7 V. Although not shown in Fig. 4a, the tunneling current at $T = 0$ calculated by Eqs. 20 and 23-25 (see Methods) is barely distinguishable from the results at room temperature. The main difference is the slope of the *I*-*V* curve when the applied voltage is $V = V_D$.

Figure 4b shows the differential contact resistivity $(dJ/dV)^{-1}$ for a MG junction as a function of the bias voltage $V$. The thick lines are for $T$ = 300K and the thin lines are for $T = 0$. The differential contact resistivity (DCR) exhibits, for every metal, large peaks at voltages $V = V_D$ and $V = V_C$, which have been labeled for the Al curve. Metals such as Pt and Au show maximum values of the DCR at $V > 0$ while for Cu, Ag, and Al the DCR is maximized at $V < 0$. For the MG junction with the X metal, only one peak appears at $V$

= 0, similar to metal-insulator-metal (MIM) diodes.[32] In Fig. 4b, one can see that a metal such as Au leads to a high specific contact resistivity (DCR at $V = 0$) because its $V_D$ value is close to zero, resulting in a Fermi energy close to the Dirac point at zero bias. However, the DCR for Au is significantly lower than that of Pt for $V \sim 0.7$ V. Thus, the resistivity of the MG contact is strongly dependent on the voltage drop across the junction. A similar situation occurs for MIG structures, as will be shown later.

Taking advantage of the fact that the specific contact resistivity only weakly depends on the temperature (see Fig. 4b), it can be obtained, after some simple algebra, from Eq. 19 of the Methods section

$$\rho_c = \frac{\pi m_0^2 D v_f^2 \hbar^2 \exp(2\kappa d)}{8\sqrt{2} e^2 m^{3/2}} \left( \frac{E_{Fm} + E_\kappa}{E_{Fm}\sqrt{E_\kappa}} \right) \frac{1}{|\Delta E_0|}, \tag{8}$$

where $E_\kappa = (\hbar^2/2m)\kappa^2$ and $\Delta E_0$ is the shift of the graphene Fermi level with respect to the Dirac point at $V = 0$. Using the data reported in Table 1, Eq. 8 gives the following values for $\rho_c$: $4.59\times10^{-6}$, $22.44\times10^{-6}$, $8.53\times10^{-6}$, $7.9\times10^{-6}$, and $5.45\times10^{-6}$ Ohm-cm$^2$ for Pt, Au, Cu, Ag, and Al, respectively. These values are consistent with experimental results reported by Nagashio and Berdebes.[7,11] The traditional wisdom is that metals with high or low work functions with respect to pristine graphene make the best contacts because the resulting Fermi level sits far away from the graphene Dirac point.[6,33] However, our model shows that the contact resistivity depends not only on the work function difference but also on the voltage drop across the junction. Although Eq. 8 has been obtained for a MIG structure like that of the Fig. 1, it also describes the specific contact resistivity of a graphene-based three-terminal device such as the one studied by Xia *et al*. when a back gate voltage is applied.[16] However, in the latter case the value of $\Delta E_0$ must be calculated by means of an expression similar to Eq. 2, but taking into account the gate capacitance. Equation 8 permits, in a simple manner, an understanding of the intrinsic factors that control the value of the specific contact resistivity and therefore of the contact resistance.

Next, we show in Fig. 5 the behavior of the differential tunneling resistivity (DTR) as a function of the voltage $V$ for a MIG structure with $d = 0.6$ nm. The DTR is calculated in the same manner as the DCR for the MG junction, $(dJ/dV)^{-1}$. Here, we have assumed a relative permittivity $\varepsilon_r = 4$ for the (I) layer. In the inset, the expected exponential dependence of the resistivity with $d$ can be observed for a Pt contact at two different values of $V$. In these structures the DTR depends directly on the work function difference between the metal and graphene since $\Delta_{ch} = 0$. It is worth mentioning that, using Cu or Ag as the metal electrode, the graphene in the MG junction is n-doped (Fig. 4b) but in the MIG structure the graphene is p-doped (Fig. 5). This capability to change the doping type is mediated by the cancelation of chemical potential term $\Delta_{ch}$. In general, when considering a wide range of bias voltages and device geometries, Pt appears to offer the best performance of the contact metals studied here.

Figure 6a shows the asymmetry factor for different metals, which is defined as the magnitude of the ratio of the reverse current at $-V$ to the forward current at $+V$, and is a figure of merit for MIM diodes. The asymmetry factor of the MG junction with Au

reaches 2.3 at 0.2 V and in the X diode decreases monotonically, which can be explained by the differences between the dispersion relations of graphene and metals in the graphene conduction and valence bands. Also, Fig. 6b shows the variation of the DCR at $V = V_D$ as a function of temperature. In Fig. 4b it is observed that only at $V = V_D$ does the DCR depend strongly on the temperature, which is attributed to the reduced DOS in the graphene when $\Delta E = 0$, while at $V = 0$ that dependence is weak.

## CONCLUSIONS

In conclusion, we have developed models for both the tunneling current and the contact resistivity, as a function of the voltage applied to the metal electrode, for the MIG heterostructure based on the BTH method. We have considered the MG junction as a special case of the MIG structure by utilizing a dipole layer as the interfacial layer. From the model for the differential contact resistivity we have found a simple analytical expression for the specific contact resistivity, which elucidates the role played by the chemical potential of interaction $\Delta_{ch}$, the work function difference, and the insulator thickness between the metal and graphene. In general, the model reveals the role played by the electrical and physical parameters in determining the contact resistivity. Specifically, among the metals considered here, Pt exhibits the smallest specific contact resistivity. However, given the voltage dependence of the contact resistivity, metals like Au or Ti can show a smaller resistivity than that of the Pt depending on the operating regime. The obtained values of the specific contact resistivity are on the order of those given in previous reports. Also, our model can be useful to predict how the specific contact resistivity depends on the difference between the Dirac point and Fermi energies when a back gate voltage is applied in a graphene-based three-terminal FET, which makes it useful for understanding the metal-graphene contact resistance. Finally, the temperature dependence of the contact resistivity is generally weak at equilibrium for every metal, but is strong and decreases with increasing temperature at $V = V_D$, i.e. in the situation corresponding to undoped graphene. Overall, our model expounds on the role played by various parameters in determining the contact resistivity of the metal-insulator-graphene junction, which should be useful for optimizing the contact resistance of graphene-based electronic devices. Our model can also be used in larger-scale simulations of these devices.

Figure 1. (a) Physical structure and (b) band diagram of the metal-insulator-graphene structure examined in this paper.

Figure 2. (a) Band diagram of isolated graphene and metal. (b) Band diagram of a metal-insulator-graphene structure at equilibrium showing charge transfer. A voltage drop $\Delta V$ is developed across the interfacial layer, and $\Delta E$ represents the shift of the graphene Fermi level with respect to the Dirac point. (c) Non-equilibrium band diagram of the metal-insulator-graphene structure. As mentioned in the main text, the metal-graphene junction is obtained by letting $d \rightarrow d_{eq}$.

Figure 3. Graphene Fermi level shift with respect to the Dirac point as a function of the bias voltage $V$ in a metal-graphene junction for different contact metals. The equilibrium separation distances used here are taken from Table 1.

Figure 4. (a) Tunneling current density $J$ and (b) differential contact resistivity (thick line for $T$ = 300K, thin line for $T$ = 0) for the metal-graphene junction as a function of the bias voltage $V$ considering different metal electrodes at the equilibrium separation.

Table 1. $W_m$ and $d_{eq}$ represent the metal work function and equilibrium separation distance between graphene and the metal, respectively. $E_{Fm}$ is the metal Fermi energy at $T$ = 0, $\Delta_{ch}$ is the chemical potential of interaction, and $V_D$ is the voltage given by Eq. 3.

Figure 5. Differential tunneling resistivity (DTR) of the metal-insulator-graphene heterostructure and its dependence on the insulator thickness $d$, assuming $\varepsilon = 4\varepsilon_0$ and $T$ = 300K. The inset shows the exponential variation of the DTR with $d$ for two different bias voltages.

Figure 6. (a) Asymmetry factor of the metal-graphene junction for different contact metals. (b) Differential contact resistivity of the metal-graphene junction at $V = V_D$ for different contact metals as a function of the temperature.

Figure 7. Parabolic and linear dispersion relations corresponding to metal and graphene electrodes, respectively. At $T$ = 0 only states lying in the shaded region contribute to the tunneling current. For a given $k_z$ both the in-plane momentum $k$ and the total energy $E$ are conserved only for the states with in-plane momentum $k_1$ in the conduction band and $k_2$ in the valence band.

## METHODS

TBD

*Conflict of Interest*: The authors declare no competing financial interest.

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