Seebeck effect in the graphene-superconductor junction

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Thermopower of graphene-superconductor (GS) junction is analyzed within the extended Blonder-Tinkham-Klapwijk formalism. Within this approach we have also calculated the temperature dependence of the zero-bias conductance for GS junction. Both quantities reflect quasi-relativistic nature of massless Dirac fermions in graphene. Both, the linear and the non-linear regimes are considered. For the sake of completeness, in Appendix we discuss limitations and consequences resulting from different approximations in the transverse-mode counting.

I. INTRODUCTION

Graphene is one of the most remarkable new materials. Not only has its discovery\textsuperscript{1} violated in same sense Landau’s theory of the thermodynamical instability of a two-dimensional structure\textsuperscript{2}, but also due to the peculiar band structure it has provided us with an invaluable opportunity to test relativistic quantum electrodynamics in the desktop laboratory\textsuperscript{3}. For that reason, much effort has been put into understanding of the phenomena associated with this material.

Since the graphene-based devices are usually considered as a mesoscopic systems, the Landauer approach is widely utilized to study the ballistic transport in them\textsuperscript{4,5}. Even though this approach does not account in the simplest form for all the features of the material, it provides a good overall description of the electric transport. This approach was extended by Blonder, Tinkham, and Klapwijk\textsuperscript{6} (BTK) to the case of standard normal metal-superconductor junction (NS), and in this manner yielded a very good description of experimental data\textsuperscript{7}. Their method has been widely used for different specific situations\textsuperscript{8,9}, and finally adapted for graphene-superconductor hybrid systems\textsuperscript{10–12}. One of the most peculiar properties predicted in such systems is the peculiar Andreev reflection and deviations of the conductance spectra\textsuperscript{13} from those predicted by BTK for normal metals\textsuperscript{14}.

Landauer formalism has also been successfully adapted for thermoelectrical transport in mesoscopic devices\textsuperscript{15–17}. In the case of standard NS junctions BTK formula, also turned out to be useful technique for predicting effects concerning thermal properties of electric and heat currents\textsuperscript{18–20}. This method has also been used for the graphene-based superconducting hybrid structures for obtaining the thermal conductance\textsuperscript{21–23}. However, the thermopower has not been studied so far. This topic is addressed in this article.

In this work we provide systematic study of the effect of the temperature on the charge current in the superconducting graphene junction (GS) using a generalized BTK formalism for the specific case of graphene. We present results concerning the temperature dependence of the zero-bias conductance and the Seebeck coefficient in the linear regime. For the sake of completeness, we also discuss the non-linear thermopower.

The paper is organized as follows. In Sec. II (and in Appendix A), we present briefly a generalized BTK approach for the charge current through the GS junction. The linear transport coefficients are discussed, in particular the zero-bias conductance and the thermopower. We also briefly comment on the effect of non-linear corrections on the Seebeck coefficient. Finally, we conclude in the Sec. IV. As a supplement, in Appendix B we add a brief discussion on the transverse-mode counting.

II. MODEL

We consider a ballistic limit for graphene based junction composed of the normal region and induced by means of the proximity effect superconducting region (cf. Fig.1). For the description of the unconventional quasiparticle states we utilize Dirac - Bogoliubov - de Gennes equations for the two-dimensional (2D) sheet of graphene in the form\textsuperscript{18,30}

\[
\left( H_j - E_F 1 \right) \begin{pmatrix} u \\ v \end{pmatrix} = \epsilon \begin{pmatrix} u \\ v \end{pmatrix},
\]

where the index $j$ can be either $+$ or $-$ what refers to the two inequivalent valleys K and K’ in the Brillouin zone.

[Image: Fig. 1. (Color online) Proposed schematic, experimental setup considered in our modeling.]
The single particle Hamiltonian is given by
\[ H_x = -\hbar v_F (\sigma_x \partial_x + \sigma_y \partial_y) + U, \]
where \( v_F \) is the energy independent Fermi velocity for the graphene, and \( \{ \sigma_i \} \) denote respective Pauli matrices. Because of the valley degeneracy one can effectively do the calculation for the one valley only. We assume that in the geometry, where the interface is determined by the \( y \)-axis, the pair potential with the \( s \)-wave symmetry changes step-like in the \( x \)-axis direction,
\[ \Delta(r, T) = \begin{cases} 0 & x < 0, \\ \Delta(T)e^{i\phi} & x > 0, \end{cases} \]
where the temperature dependence of the gap function can be deduced from the usual BCS theory and is taken in the following form
\[ \frac{\Delta(T)}{\Delta_0} = \tanh \left( \sqrt{1.76 \cdot \frac{T_c}{T} - 1} \right). \]

The BCS theory of the superconductivity based on the requirement that the coherence length is large when compared to the Fermi wavelength. Under that condition we can assume that the additional potential \( U \) has the form,
\[ U(r) = \begin{cases} 0 & x < 0, \\ -U_0 & x > 0, \end{cases} \]
with large \( U_0 \) (\( E_F + U_0 \gg \Delta_0 \)) and for the simplicity we define \( E'_F = E_F + U_0 \). In numerical calculations we set \( E'_F = 1000\Delta_0 \).

In the spirit of the BTK scheme (by matching the wave functions at the boundary \( x = 0 \)), we obtain expressions for the amplitudes of the Andreev hole reflection (AR) \( (a(\epsilon, \theta)) \) and the normal reflection \( (b(\epsilon, \theta)) \) of the incident electron - see Appendix A for details of the method. Note that there is no intrinsic barrier at the GS junction, and thus the Fermi vector mismatch is the source of the normal reflection. The transmission probability, averaged over the angles, takes the following form\(^{(7,14)}\),
\[ T(\epsilon) = \int_{\pi/2}^{\pi/2} d\theta \cos^2 \theta \left( 1 - |b(\epsilon, \theta)|^2 + \frac{\text{Re}[e^{i\theta A}]}{\cos \theta}|a(\epsilon, \theta)|^2 \right). \]

The BTK formalism combined with the specific transmission probability derived for graphene, defines charge current through the GS interface as\(^{(7,18)}\),
\[ I_c = \frac{4e^2}{h} \int_{-\infty}^{\infty} d\epsilon N(\epsilon) T(\epsilon) \left( f^G(\epsilon - eV) - f^S(\epsilon) \right), \]
where \( f^G \), \( f^S \) are the Fermi distribution functions for the normal (G) and the superconducting (S) region of GS junction respectively, and
\[ N(\epsilon) = \frac{|E_F + \epsilon| W}{\pi \hbar v_F}, \]
is the energy dependent number of transverse modes in the graphene sheet of width \( W \). However, formula \(^{(7)}\) is not always accurate. The additional assumption is needed, that for each mode carrying incident electron, having energy \( E_F + \epsilon \) and being Andreev reflected there is always enough modes at the level \( E_F - \epsilon \) for this process to happen. For a more detailed discussion of this topic see Appendix B.

The quantity describing thermoelectric properties of the system is thermopower, or Seebeck coefficient \( (S) \) measuring the voltage driving to zero the current flowing in response to the temperature difference, namely
\[ S = \frac{V}{eT} I_c = 0. \]

**III. CHARGE TRANSPORT**

**A. Linear regime**

Expansion of the Fermi functions in the normal and superconducting regions to the first (linear) order in both the bias and the temperature difference, with the average temperature \( T \), i.e.,
\[ f^G \equiv f_{T-\delta T/2}(\epsilon - eV) \simeq f_T(\epsilon) - eV \frac{\partial f}{\partial \epsilon} + \frac{\delta T}{2T} \epsilon \frac{\partial f}{\partial \epsilon}, \]
\[ f^S \equiv f_{T+\delta T/2}(\epsilon) \simeq f_T(\epsilon) - \frac{\delta T}{2} \frac{\partial f}{\partial \epsilon}, \]

enables to decouple Eq.\(^{(7)}\) in the form,
\[ I_c = GV + I_c^T \delta T. \]

The resulting from it the linear transport coefficients can be thus rewritten in the following closed forms,
\[ G = \frac{4e^2}{h} \int_{-\infty}^{\infty} d\epsilon \frac{\partial f}{\partial \epsilon} N(\epsilon) T(\epsilon), \]
\[ I_c^T = \frac{4e}{hT} \int_{-\infty}^{\infty} d\epsilon \frac{\partial f}{\partial \epsilon} \epsilon N(\epsilon) T(\epsilon). \]

The temperature gradient and the bias are set as positive with respect to \( x \) coordinate. For the temperature in the system approaching zero, the expression for the electric conductance \( (G) \) reduces to the well-known BTK zero-bias conductance formula\(^{(7)}\),
\[ G_{T \rightarrow 0} = \frac{4e^2}{h} N(0) T(0). \]

In Fig\(^{(2)}\) we have plotted the zero-bias differential conductance (calculated from Eq.\(^{(12)}\)) as a function of temperature, normalized by the ballistic conductance \( g_0 \) with having \( N \) transverse modes in a sheet of graphene of width \( W \) and given by
\[ g_0 = \frac{4e^2}{h} \int_{-\infty}^{\infty} d\epsilon \frac{\partial f}{\partial \epsilon} N(\epsilon). \]
For relatively high position of the Fermi level in graphene (roughly \(E_F \gtrsim 5\Delta_0\)), the influence of the increasing temperature (up to the approximately \(T/T_c = 0.5\)) on the conductance is almost negligible (cf. Fig. 2). The origin of this behavior is strictly connected with the relatively slow variation of the transmission probability \(T(\epsilon)\) for the subgap energies in this temperature regime, and thus does not differ qualitatively from the standard NS case. In the low doping regime \((E_F \lesssim 5\Delta_0)\) the linear differential conductance as a function of the temperature vastly differs from the standard NS case and reflects the specific electronic nature of graphene.

The non-trivial behavior of the Dirac fermions in graphene has also its influence on the linear thermopower. From Eq. \(9\) and \(11\) formula for this quantity reads

\[
S = \frac{T}{G} = -\frac{1}{k_B T} \int_{-\infty}^{\infty} \frac{d\epsilon}{e} e^{\frac{\epsilon}{k_B T}} \int_{-\infty}^{\infty} \frac{d\epsilon}{e} + E_F \frac{\partial f}{\partial \epsilon} T(\epsilon) \frac{k_B}{e} \epsilon.
\]

(15)

Results obtained by the numerical integration are presented in the Fig. 3. The low temperature regime differs from the one obtained for the NS junction. Contrary to the NS case, for the graphene-based structure the thermopower does not vanish for non-zero temperatures. This is due to the relativistic nature of charge carriers in graphene, where AR does not vanish even for the high effective barrier (Fermi velocity mismatch in our case) for the subgap energies (for the normal incidence AR happens always with certainty). For the low doping regime \((E_F \lesssim \Delta_0\)) we observe a clear maximum of the thermopower in the temperature range where the superconductivity exists. In the linear regime, the thermopower for the selected range of parameters near \(T_C\) is large and reaches values up to \(1k_B/e\). This suggests that there is a potential for application of this setup for cooling of various nanostructures.

### B. Effect of the non-linearity

We have also studied numerically the effect of the non-linearity in our system. The thermopower in the non-linear regime can be calculated as a ratio between the bias voltage and the temperature gradient, when no charge current is flowing (c.f. Eq.\(10\)). The results in the non-linear regime are presented in Fig. 3. We have found that the non-linearity influences, the thermopower in not systematical manner with changing Fermi level position and the average temperature of the system. However the change is not as dramatic as in the NS case and is almost unnoticeable in the doped regime \(E_F \gtrsim 5\Delta_0\).

### IV. CONCLUSIONS

In this work we have analyzed the thermoelectric charge transport in the graphene junction consisting of the normal and the superconducting parts. In the linear regime we have calculated the temperature dependence of the zero-bias conductance and the thermopower. We have found deviations of these quantities from the standard normal metal-superconductor junction case that are caused by the relativistic nature of electrons in graphene.

We have also studied the effect of non-linearity on the thermopower and we have found that for a high Fermi level positions \((E_F \gtrsim \Delta_0)\) it stays almost unaffected and in the low Fermi level regime is noticeably enhanced with the increase of the temperature gradient.

As a supplemental discussion in the Appendix B we discuss some consequences of including into the standard BTK model the energy dependent number of transverse modes. We point out that in graphene with the Fermi level near the Dirac point, where the energy dependence of the number of transverse modes is not negligible, one needs to be cautious in using the BTK formula as applied to the discussion of the effects associated with an
interplay of the heat and the electric currents, since one of the Onsager reciprocity relation can be violated.

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**Appendix A: BTK for graphene**

The wave function in the normal part of graphene (NG) $\psi_N$ and in the superconducting region (SG) $\psi_S$, look respectively as follows

$$\psi_N^{e, h} = [1, ± e^{± i θ}, 0, 0]^T e^{± i k x} \cos θ,$$

$$\psi_S^{e, h} = [0, 0, 1, e^{−i θ_A}]^T e^{−i k x} \cos θ_A,$$

where the superscripts $e, h$ refers to electron and hole in NG and electronlike and holelike excitation in SG, and the superscripts + and - to right and left moving particle respectively.

Spinors resulting from Eq. (A1) are expressed in the similar manner as in the Ref. [19], i.e., in the form

$$\psi^{e, h}_N = \begin{bmatrix} 1, ± e^{± i θ}, 0, 0 \end{bmatrix}^T e^{± i k x} \cos θ,$$

$$\psi^{e, h}_S = \begin{bmatrix} 0, 0, 1, e^{−i θ_A} \end{bmatrix}^T e^{−i k x} \cos θ_A,$$

where for the sake of clarity we do not include phase factor $e^{i k y}$ since it corresponds to conservation of momentum in $\hat{y}$ direction. The corresponding wave vectors are defined as follows,

$$k^{e, h} = \frac{e + (-)E_F}{\hbar v_F}, \quad q^{e, h} = \frac{E_F’ + (-)\sqrt{\epsilon^2 - \Delta^2}}{\hbar v_F},$$

and the coherence factors are given by

$$u = \sqrt{\frac{1}{2} \left( 1 + \frac{\sqrt{\epsilon^2 - \Delta^2}}{\epsilon} \right)}; \quad v = \sqrt{\frac{1}{2} \left( 1 - \frac{\sqrt{\epsilon^2 - \Delta^2}}{\epsilon} \right)}.$$

The conservation of momentum at the interface and along $\hat{y}$ direction enables us to obtain mutual relations for the specific angles, namely

$$k^e \sin θ = k^h \sin θ_A = q^e \sin θ^e_S = q^h \sin θ^h_S.$$

The system must also satisfy the continuity condition at the interface, $ψ_{σL}(0) = ψ_{σR}(0)$. The Hamiltonian is linear therefore is no need in matching derivatives. The resulting wave function amplitudes take the form,

$$a(ε, θ) = \frac{2\cos θ(e^{−i θ_A} + e^{i θ_S})uv}{(e^{−i θ_A} + e^{i θ_S})(e^{−i θ_A} + e^{i θ_S})u^2 - (e^{−i θ_A} - e^{i θ_S})(e^{−i θ_A} - e^{i θ_S})v^2},$$

$$b(ε, θ) = \frac{2\cos θ(e^{i θ_S} - e^{−i θ_A})u^2 + (e^{i θ_S} + e^{−i θ_A})u^2}{(e^{−i θ_A} + e^{i θ_S})(e^{−i θ_A} + e^{i θ_S})u^2 - (e^{−i θ_A} - e^{i θ_S})(e^{−i θ_A} - e^{i θ_S})v^2} - 1.$$
Appendix B: On transverse-mode counting and fulfilling the Onsager relations

In this Appendix, which has rather a Supplementary character, we raise the question, whether standard ways of including the number of transverse modes in the BTK formalism is applicable in the situation, when one considers not only electric current, but also the heat transport as well. In particular, we address the question of the validity of the Onsager relations which in our system should be obeyed.

B.1. Two different attempts

One encounters some difficulties when tries correctly considering number of transverse modes in BTK model for graphene. In the BTK scheme in order to obtain transmission probability one has to take the difference between distributions of the populations of particles moving towards and away the interface,

\[ T'(\epsilon) = f_+ - f_- = (1 - B) f_- - A f_+ - (1 - A - B) f, \]

(B1)

where for convenience and clarity we omit angle dependence and used generalized notation - A stands for probability of the Andreev reflection and B for the normal reflection, and we define \( f^\pm \equiv f(\epsilon \pm eV) \), whereas \( f \equiv f(\epsilon) \). The above formula (B1) usually is used in the more compact form as

\[ T(\epsilon) = (1 - B + A)(f_- - f). \]

(B2)

However, this is not a true transmission amplitude across the interface. Nevertheless, when one takes the integral over whole range of energies, it provides the same result as the true transmission \( T'(\epsilon) \),

\[ \int_{-\infty}^{\infty} d\epsilon T(\epsilon) = \int_{-\infty}^{\infty} d\epsilon T'(\epsilon). \]

(B3)

The above relation becomes not valid, when one includes directly the number of transverse modes (which would not be an even function of \( \epsilon \)) under the integral. It is clearly seen when one calculates the resulting differential conductance in the zero temperature for the transmission \( T'(\epsilon) \),

\[ G = \frac{4e^2}{h} \frac{d}{d eV} \int_{-\infty}^{\infty} d\epsilon T(\epsilon) N(\epsilon) = \frac{4e^2}{h} (1 + A(eV) - B(eV)) N(eV), \]

and analogically for the transmission \( T'(\epsilon) \),

\[ G' = \frac{4e^2}{h} \frac{d}{d eV} \int_{-\infty}^{\infty} d\epsilon T'(\epsilon) N(\epsilon) = \frac{4e^2}{h} [(1 - B(eV)) N(eV) + A(eV) N(-eV)]. \]

(B4)

(B5)

If \( N(\epsilon) \) is not an even function of \( \epsilon \), \( G \neq G' \).

B.2. Standard approximation

Usual way to include the number of transverse modes and utilized in majority of papers studying differential conductance of graphene\(^{18,33,34}\) is to take into account the transmission \( T(\epsilon) \) (Eq. (B2)) what leads to the formula (B3). Physically it means that there is always enough modes for propagation of created in the AR hole beneath the Fermi level (for convenience, we call it Assumption 1). The need of this assumption is clearly visible in the perfect AR regime. For ideal transparent interface and for subgap energies, AR happens with certainty. It means that in the normal region every incident electron should be converted into the reflected hole. However, due to mismatch in the number of available modes above and beneath the Fermi level, a part of incident electrons cannot Andreev reflect. Since the transmission to the superconductor is also forbidden, the excess electrons have to scatter via the normal-reflection process.

In systems where the Fermi energy is quite high it does not matter for studying the electric current since the normal reflection due to the mismatch in modes number above and beneath Fermi level would be negligible small. However, in the case of graphene in the low doping regime, near the perfect AR, it can make a difference.

The Assumption 1 is also used in many papers to study the thermal conductance\(^{20,27,29}\). Therefore, a natural question arises, whether the Onsager reciprocity relations are satisfied (they should be obeyed in our system\(^{32}\)).

For the ballistic transport and with the use of Assumption 1, the heat current through the barrier is defined as follows,

\[ I_Q = \frac{4}{h} \int_{-\infty}^{\infty} d\epsilon N(\epsilon)(-eV) T''(\epsilon) (f^G(\epsilon - eV) - f^S(\epsilon)) + \epsilon \frac{\partial f}{\partial \epsilon} \frac{\partial f}{\partial \epsilon} N(\epsilon) T''(\epsilon). \]

(B6)

Note that here the transmission probability is different than in the electric current case, i.e.,

\[ T''(\epsilon) = (1 - B - A)(f_- - f). \]

(B7)

Physically, it means that there is no heat current into the superconducting region for the subgap energies. The same form of the transmission (Eq. (B7)) has been used for instance in Refs. 24, 27, 29.

In general we can write that the electric and the heat currents are driven by the bias voltage or the temperature gradient, so that

\[ I_e = G V + I_e^T \frac{\partial V}{\partial T}, \]

\[ I_Q = I_Q^T V + K \frac{\partial T}{\partial T}. \]

(B8)

In that situation, the selected linear coefficients read,

\[ I_Q^T = \frac{4e}{hT} \int_{-\infty}^{\infty} d\epsilon \frac{\partial f}{\partial \epsilon} \frac{\partial f}{\partial \epsilon} N(\epsilon) T''(\epsilon), \]

\[ I_e^T = \frac{4e}{h T} \int_{-\infty}^{\infty} d\epsilon \frac{\partial f}{\partial \epsilon} \frac{\partial f}{\partial \epsilon} N(\epsilon) T(\epsilon). \]

(B9)
One can see clearly that within the Assumption 1 $I_Q \neq -T^I_{\mathcal{E}}$, and thus the corresponding Onsager relation is broken. This is due to the difference between $\mathcal{T}''$ and $\mathcal{T}$. Therefore, one should be careful in using this formalism for calculating for instance the thermal conductance, which from definition should results from an interplay between electric and heat currents, where the Onsager relations should be satisfied.

**B.3. Restoration of Onsager relations**

Contrary to the Assumption 1 one can take into account full form of transmission (Eq. (151)) and directly add the number of transverse modes under the integral to formulas for the electric and the heat currents. This attempt was used before in Ref. [1] and [21]. Within this assumption one restores the Onsager reciprocity relation. Despite an apparent success, one pays a price of ‘decoupling’ the coherent process of AR. More precisely, AR is a process in which incident electron in some sense borrows a partner electron from beneath the Fermi surface, and as a Cooper pair is absorbed into a superconductor at the Fermi level, and simultaneously the hole is reflected back into graphene. Therefore, it should be clear that there is the same number of incident electrons scattered in AR as holes reflected. In this attempt of including number of transverse mode, the reflection back is not guarntied. It is clearly visible if one considers the heat current through the interface in the subgap regime. Any energy transported towards the interface is taken back by reflected hole or electron, and therefore, the heat current should be zero. However, if we calculate within this attempt (with $\mathcal{T}''$) heat current, we obtain a nonzero energy transported through the interface, what is clearly wrong. Note that still this way of including number of transverse modes is reasonable for the electric current for the high Fermi level position as in Ref.[1].

**B.4. Remarks**

The Assumption 1 represents a reasonable approximation for the electric-current calculation. Therefore, we use it for calculating the thermopower and the zero-bias conductance of the GS junction in this article. Furthermore, the resulting formula for the charge flux is rigorous away from the perfect AR limit (where AR happen with certainty).

However, since within this assumption the Onsager reciprocity relations in the linear regime are not fulfilled, for investigating the effects of interplaying the electric and the heat fluxes one should be careful to construct a model which would also obey Onsager relations.

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