Surface superconductivity in multilayered rhombohedral graphene: Supercurrent

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\section{I. INTRODUCTION}

Fermionic systems with dispersionless branches of excitation spectrum (flat bands) have quite unusual properties; nowadays they attract lots of research interest. Flat bands were predicted in many condensed matter systems, see for example Refs.\textsuperscript{1–4}. In some cases the flat bands are protected by topology in momentum space; they emerge on the surfaces of gapless topological matter\textsuperscript{5,7}, such as surfaces of nodal superconductors\textsuperscript{6}, graphene edges\textsuperscript{8}, surfaces of multilayered graphene structures\textsuperscript{9–10}, and in the cores of quantized vortices in topological superfluids and superconductors\textsuperscript{11,12}.

The singular density of states (DOS) associated with the dispersionless spectrum may essentially enhance the transition temperature opening a new route to room-temperature superconductivity. The corresponding critical temperature depends linearly on the pairing interaction strength and can be thus considerably higher than the usual exponentially small critical temperature in the bulk\textsuperscript{13}. It was shown in Ref.\textsuperscript{5,3} that the flat band that appears on the surface of multilayered rhombohedral graphene is especially favorable for surface superconductivity. Formation of surface superconductivity is enhanced already for a system having $N \geq 3$ layers, where the normal-state spectrum has a slow power-law dispersion $\xi_p \propto |p|^N$ as a function of the in-plane momentum $p$. The DOS $\nu(\xi_p) \propto \xi_p^{(2–N)/N}$ has a singularity at zero energy which results in a drastic enhancement of the critical temperature.

Absence of dispersion in a flat band raises the questions of superconducting velocity and of the supercurrent: Can they be nonzero and, if they can, what is then the magnitude of the critical current? In this Letter we address the problem of supercurrent associated with the surface superconductivity in the flat-band multilayered rhombohedral graphene. Based on the model employed in Refs.\textsuperscript{13} for description of the surface superconductivity we calculate the supercurrent as a response to a small gradient of the order parameter phase using an approach similar to that used for calculations of the supercurrent in a single layer of graphene\textsuperscript{4}. We demonstrate that the supercurrent is finite; the critical current is proportional to the superconducting zero-temperature gap, i.e., to the critical temperature, and to the radius of the flat band in the momentum space. Being produced by the surface superconductivity, the total current through the sample is independent of the sample thickness.

\section{II. THE MODEL}

As in Ref.\textsuperscript{13} we consider multilayered graphene structure of $N$ layers in the discrete representation with respect to interlayer coupling. For simplicity we choose the rhombohedral stacking configuration considered in Refs.\textsuperscript{15–16} and assume that the most important are hoppings between the atoms belonging to different sublattices parameterized by a single hopping energy $t$. More general form of the multilayered Hamiltonian can be found in Refs.\textsuperscript{15–16}. In the superconducting case the Hamiltonian has the form of a matrix in the Nambu space. The Bogoliubov–de Gennes (BdG) equations are

$$\sum_{j=1}^{N} \left( \hat{H}_{ij} - \mu \delta_{ij} \right) \sigma_j \left( \begin{array}{c} \hat{u}_j \\ \hat{v}_j \end{array} \right) = E \left( \begin{array}{c} \hat{u}_i \\ \hat{v}_i \end{array} \right),$$

where the sum runs over the layers. The normal-state Hamiltonian\textsuperscript{16}

$$\hat{H}_{ij} = v_F (\hat{\sigma} \cdot \hat{p}) \delta_{ij} - t \hat{\sigma}_z \delta_{i,j+1} - t \hat{\sigma}_- \delta_{i,j-1},$$

is the the Hamiltonian acting on the envelope function of the in-plane momentum $p$ taken near one of the Dirac points, i.e., for $|p| \ll \hbar/a$ where $a$ is the interatomic distance within a layer; $v_F = 3t_\text{intr}/2\hbar$ where $t_\text{intr}$ is the hopping energy between nearest-neighbor atoms belonging to different sublattices on a layer. The particle-like, $\hat{u}_i$, and hole-like, $\hat{v}_i$, wave functions near the Dirac point are coupled via the superconducting order parameter $\Delta_i$ that can appear in the presence of a pairing interaction. Here we do not specify the nature of the pairing. It can be due to either electron-phonon interaction or other pairing interactions that have been suggested as a source for intrinsic superconductivity in graphene, see Refs.\textsuperscript{17}. The excitation energy for particles and holes is measured upwards or downwards, respectively, from the Fermi level.
which can be shifted with respect to the Dirac point due to doping. Here we assume that the shift is the same on all layers. The order parameter and the Fermi level shift \( \mu \) are scalars in the pseudo-spin space. We assume that \( \Delta \) and \( \mu \) are much smaller than the inter-layer coupling energy \( t > 0 \), which in turn is \( t \ll t_0 \). Usually, \( t \sim 0.1 t_0 \) where \( t_0 \sim 3 \text{ eV}^{16} \).

We decompose the wave function

\[
\left( \hat{u}_n \begin{pmatrix} \hat{v}_n \end{pmatrix} \right) = \left[ \left( \begin{pmatrix} \alpha_n^+ \\ \beta_n^+ \end{pmatrix} \right) \otimes \hat{\Psi}^+ + \left( \begin{pmatrix} \alpha_n^- \\ \beta_n^- \end{pmatrix} \right) \otimes \hat{\Psi}^- \right]
\]

(2)

into the spinor functions localized at each sublattice

\[
\hat{\Psi}^+ = \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \quad \hat{\Psi}^- = \begin{pmatrix} 0 \\ 1 \end{pmatrix}.
\]

We introduce matrices and vectors in the Nambu space

\[
\tau_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \quad \Delta_n = \begin{pmatrix} 0 & \Delta_n \\ \Delta_n & 0 \end{pmatrix}, \quad \alpha_n^+ = \begin{pmatrix} \alpha_n^x \\ \alpha_n^y \end{pmatrix}.
\]

The BdG equations take the form

\[
\tau_3 \left[ v_F (\hat{p}_x - i \hat{p}_y) \alpha_n^+ - t \alpha_{n-1}^- - \mu \alpha_n^+ \right] = E \alpha_n^+, \quad n \neq 1, \quad (3)
\]

\[
\tau_3 \left[ v_F (\hat{p}_x + i \hat{p}_y) \alpha_n^+ - t \alpha_{n+1}^- - \mu \alpha_n^+ \right] = E \alpha_n^-, \quad n \neq N, \quad (4)
\]

where \( \hat{p} \) is the momentum operator. In Eqs. (3) and (4) we assume that \( \Delta_n \neq 0 \) only at the outermost layers, while \( \Delta_n = 0 \) for \( n \neq 1, N \). The arguments supporting this assumption are given in Ref.\ref{1}. It was shown that the order parameter quickly decays as a function of the distance from the surface. We also neglect \( \Delta_n \) as compared to \( t \) in Eqs. (3) and (4) for \( n = N \) and \( n = 1 \), respectively, as they lead to higher-order corrections in \( \Delta/t \). The particle and hole channels are thus decoupled if \( n \neq 1, N \). Expanding the coefficients in plane waves \( \alpha, \beta \propto e^{i p \mathbf{r} + ipz} \) we find the energy in terms of in-plane \( p \) and transverse momentum \( p_z \) \((d \text{ is the interlayer distance})\)

\[
E^2 = v_F^2 p^2 - 2v_F p \cos(p_z d - \phi) + t^2
\]

(5)

where \( p = \sqrt{p_x^2 + p_y^2} \) and \( e^{i \phi} = (p_x + i p_y)/p \). Equations \ref{3} and \ref{4} determine the coefficients \ref{9,10}.

\[
\alpha_n^+ = \zeta_n^+ (p) \hat{A}^+ + \zeta_n^- (p) t^{-2}(\tau_3 \hat{E} + \hat{\mu}) v_F (p_x - i p_y) \hat{A}^- \quad (6)
\]

\[
\alpha_n^- = \zeta_n^+ (p) \hat{A}^- + \zeta_n^- (p) t^{-2}(\tau_3 \hat{E} + \hat{\mu}) v_F (p_x + i p_y) \hat{A}^+ \quad (7)
\]

where the basis functions are

\[
\zeta_n^+ (p) = [v_F (p_x + i p_y)/t]^{n-1},
\]

\[
\zeta_n^- (p) = [v_F (p_x - i p_y)/t]^{N-n}.
\]

Here we include the first-order corrections in energy. Having an imaginary momentum \( p_z \) for \( v_F p < t \), these solutions decay away from the surfaces and thus they describe the surface states. Normalization requires

\[
d \sum_{n=1}^{N} \left( (\alpha_n^+)^\dagger \alpha_n^- + (\alpha_n^-)^\dagger \alpha_n^+ \right) = 1.
\]

This gives

\[
d \left[ (\hat{A}^+)^\dagger \hat{A}^+ + (\hat{A}^-)^\dagger \hat{A}^- \right] = 1 - v_F^2 t^2 / t^2.
\]

(8)

A finite order parameter \( \Delta \) couples the particle and hole channels at the outermost layers, \( i = 1 \) and \( i = N \).

\[
\tau_3 v_F (\hat{p}_x - i \hat{p}_y) \alpha_1^- - \tau_3 \mu_1 \alpha_1^+ = E \alpha_1^+ - \Delta \alpha_1^+, \quad (9)
\]

\[
\tau_3 v_F (\hat{p}_x + i \hat{p}_y) \alpha_N^- - \tau_3 \mu N \alpha_N^+ = E \alpha_N^- - \Delta \alpha_N^- \quad (10).
\]

The boundary conditions \ref{9,10} select \( p_z \) and determine \( 2N \) particle and hole branches of the energy spectrum. Looking for the branches that belong to the surface states with energies of the order of \( \Delta \) and \( \mu \), we solve these equations for \( E \ll t \).

### III. SUPERCURRENT

The operator of current along a layer couples the states at different sublattices, \( \hat{u}_{\gamma\beta}(n) \sigma \hat{u}_{\gamma\beta}(n) + \hat{u}_{\gamma\beta}(n) \sigma \hat{u}_{\gamma\beta}(n) \). For example, the \( x \) component of current at layer \( n \) is

\[
J_x (n) = -e v_F \sum_{\gamma, \beta} \left[ \bar{\alpha}_{\gamma, n}(p) \alpha_{\gamma, n}(p) + \bar{\alpha}_{\gamma, n}(p) \alpha_{\gamma, n}(p) \right] \times \left[ 1 - 2f_{\gamma, p} \right].
\]

(11)

where \( \gamma \) labels different states for given \( p \), while \( f_{\gamma, p} \) is the distribution function.

To calculate the supercurrent we use the same approach as in Ref.\ref{1}. Consider \( \Delta = |\Delta| e^{i k \mathbf{r}} \). Separating the order-parameter phase, we put \( u_n = u_n(p) e^{i (p + k)/2} \), while \( v_n = v_n(p) e^{i (p - k)/2} \). For large \( N \approx 1 \) the most important corrections come from \( (p \pm k/2)^N \). (The exact condition for \( N \) will be established later.) We have

\[
\bar{\alpha}_{n} = \zeta_n^+ (p) \hat{A}^+ + \zeta_n^- (p) t^{-2}(\tau_3 \hat{E} + \hat{\mu}) v_F (p_x - i p_y) \hat{A}^- \quad (12)
\]

\[
\bar{\alpha}_{n} = \zeta_n^+ (p) \hat{A}^- + \zeta_n^- (p) t^{-2}(\tau_3 \hat{E} + \hat{\mu}) v_F (p_x + i p_y) \hat{A}^+ \quad (13)
\]

where \( \hat{p} = \hat{p} + \tau_3 k/2 \). Equations \ref{9,10} at the outermost layers give

\[
\tau_3 \xi_{p + \tau_3 k/2} \hat{A}^+ = (\hat{E} + \tau_3 \hat{\mu}) \hat{A}^+ - \tau_1 |\Delta| \hat{A}^+, \quad (14)
\]

\[
\tau_3 \xi_{p + \tau_3 k/2} \hat{A}^+ = (\hat{E} + \tau_3 \hat{\mu}) \hat{A}^- - \tau_1 |\Delta| \hat{A}^- \quad (15)
\]

Here

\[
\xi_{p} \equiv t [v_F (p \pm i p_y)/t]^{N} = e^{i N \phi_{\xi} p} \quad \xi_{p} = t(v_F p / t)^N
\]

Using the spinors in the sublattice space, Eqs. \ref{14}, \ref{15} can be written as

\[
\left[ \hat{H}_0 + \hat{H}_1 \right] \psi = \hat{E} \psi, \quad \psi = \left( \begin{pmatrix} \hat{A}^+ \\ \hat{A}^- \end{pmatrix} \right),
\]

(16)

where

\[
\hat{H}_0 = \tau_3 e^{-i \sigma_1 N \phi_{\xi}} \sigma \hat{\xi}_{p} - \tau_3 \hat{\mu} + \tau_1 |\Delta| \quad (17)
\]

\[
\hat{H}_1 = e^{-i \sigma_1 (N - 1) \phi_{\xi}} (\sigma \mathbf{k}) \frac{d \xi_{p}}{dp}.
\]

(18)
In the zero order in $k$ the coefficients $\tilde{\psi}^{(0)}$ satisfy
\[ \hat{H}_0 \tilde{\psi}^{(0)} = \tilde{E}^{(0)} \tilde{\psi}^{(0)}. \] (19)
The equation has four solutions
\[ \tilde{\psi}_{1,2} = \left( \tilde{A}_1 e^{-i N \phi/2} \tilde{A}_1 e^{+i N \phi/2} \right), \quad \tilde{E}_{1,2} = \pm \tilde{E}_0^+, \] (20)
\[ \tilde{\psi}_{3,4} = \left( \tilde{A}_3 e^{-i N \phi/2} \tilde{A}_3 e^{+i N \phi/2} \right), \quad \tilde{E}_{3,4} = \pm \tilde{E}_0^-. \] (21)

Here
\[ \tilde{E}_0^+ = \sqrt{(\xi_p - \mu)^2 + |\Delta|^2}, \quad \tilde{E}_0^- = \sqrt{(\xi_p + \mu)^2 + |\Delta|^2}, \]
and
\[ \tilde{A}_1 = \frac{C}{\sqrt{2}} \left( \frac{u_+}{v_+} \right), \quad \tilde{A}_2 = \frac{C}{\sqrt{2}} \left( \frac{v_+}{u_+} \right), \]
\[ \tilde{A}_3 = \frac{C}{\sqrt{2}} \left( \frac{v_-}{u_-} \right), \quad \tilde{A}_4 = \frac{C}{\sqrt{2}} \left( \frac{u_-}{v_-} \right). \]

Normalization is determined by Eq. (8), $|C|^2 = d^{-1}(1 - v^2_{pF}/T^2)$, the coherence factors are
\[ u_\pm = \frac{1}{\sqrt{2}} \left[ 1 + \xi_p \mp \mu \frac{E_0}{\tilde{E}_0^+} \right]^{1/2}, \quad v_\pm = \frac{1}{\sqrt{2}} \left[ 1 - \xi_p \mp \mu \frac{E_0}{\tilde{E}_0^-} \right]^{1/2}. \]

The different solutions are orthogonal,
\[ \left< \tilde{\psi}_i \right| \tilde{\psi}_k \right> = |C|^2 \delta_{ik}, \]

since $\tilde{A}_2 \tilde{A}_1 = \tilde{A}_4 \tilde{A}_3 = 0$. The trace is taken over pseudo-spin and Nambu indexes.

If the coefficients $\tilde{A}^\pm$ are taken in the zero order approximation in $k$, the product $\alpha_+^\dagger \alpha_-^\dagger$ in Eq. (12) contains the exponents $e^{-i \phi}$ and $(k_x + i k_y) e^{-2i \phi}$ and vanishes after integration over the momentum directions. Therefore, the basis functions $\tilde{c}_n^\pm$ can be taken in zero approximation in $k$ but the coefficients $\tilde{A}^\pm$ need to be calculated up to the first order terms in $k$.

The corrections due to the condensate momentum can be written as
\[ \tilde{\psi}_\alpha = \tilde{\psi}_\alpha^{(0)} + \sum_{\beta \neq \alpha} B_{\alpha \beta} \tilde{\psi}_\beta^{(0)}. \] (22)

Equations (10) - (18) give
\[ \delta \tilde{E}_\alpha = \frac{\left< \tilde{\psi}_\alpha \right| H_1 \tilde{\psi}_\alpha}{|C|^2}, \quad B_{\alpha \beta} = \frac{\left< \tilde{\psi}_\beta \right| \hat{H}_1 \tilde{\psi}_\alpha}{|C|^2 (E_\alpha - E_\beta)}. \] (23)

 Corrections to energies are
\[ \delta \tilde{E}_{1,2} = \frac{p k}{2p} \frac{d \xi_p}{d p} = E_D, \quad \delta \tilde{E}_{3,4} = -\frac{p k}{2p} \frac{d \xi_p}{d p} = -E_D \]

which is the usual normal-state Doppler shift. We have $B_{12} = B_{21} = B_{34} = B_{43} = 0$, while $B_{13} = B_{31} = -B_{24} = -B_{42}$ and $B_{23} = B_{32} = B_{14} = B_{41}$ where
\[ B_{13} = -\frac{i\left< \tilde{p} \cdot k \right| z \tilde{E}_0^+ (u_+ v_- + v_+ u_-)}{2p} \left( \tilde{E}_0^+ - \tilde{E}_0^- \right), \]
\[ B_{23} = -\frac{i\left< \tilde{p} \cdot k \right| z \tilde{E}_0^+ (u_+ v_- - v_+ u_-)}{2p} \left( \tilde{E}_0^+ + \tilde{E}_0^- \right). \]

The currents Eq. (11) at layer $n$ contains the product of $\tilde{c}_n^+ \tilde{c}_n^* = (\xi_p / v_{pF}) e^{i(N_n - 1)\phi}$ which is independent of the layer number, i.e., of the distance from the surface, and the products $\left< \tilde{E} \pm \mu / t \right| \tilde{c}_n^+ \tilde{c}_n^* \right\rangle \propto (v_{pF} / t)^{2(n-1)}$ and $\left< \tilde{E} \pm \mu / t \right| \tilde{c}_n^+ \tilde{c}_n^* \right\rangle \propto (v_{pF} / t)^{2(N-n)}$ which decay as functions of the distance from the surfaces. All these terms are of the order of $E/t$. We shall see, however, that it is the constant term that gives the main contribution to the total current through the sample, $I = \sum_{n=1}^{N} \tilde{j}(n)$. Using $\sum_p = \int (dp / dE_p) \tilde{v} p / 2\pi h$ we find for the current per unit sample width
\[ I = edNk \int \frac{\xi_p d \xi_p}{2\pi h} |C|^2 \times \left[ \frac{1}{\xi_p} - \left( \frac{\tanh E_0^+/2T}{2T} + \tanh E_0^-/2T \right) \frac{(u_+ v_- - v_+ u_-)^2}{(E_0^+ + E_0^-)} \right] \left( \frac{\tanh E_0^+/2T}{2T} - \tanh E_0^-/2T \right) \frac{(u_+ v_- + v_+ u_-)^2}{(E_0^- - E_0^+)} \right] \right] \right]. \] (24)

To obtain this expression we had to regularize Eq. (11) which diverges for large $\xi_p$. The regularization is described in detail in Ref. 14. In brief, we subtract the normal current which is obtained from the current operator taken at energies much higher than $\Delta$ and $T$. For $\xi_p \gg \Delta, T$ one has $E_0^+ = \xi_p - \mu$, $E_0^- = \xi_p + \mu$, $u = 1$, and $v = 0$. Therefore, the diverging part of Eq. (11) is
\[ I^{(\infty)} = -deNk \int \frac{\xi_p d \xi_p}{2\pi h} |C|^2 \frac{1}{\xi_p}. \] (25)

This contributes to the normal current which, of course, turns to zero in the end. Indeed, for $\Delta = 0$ when the particle and hole channels separate, the corrections to $\tilde{A}^\pm$ simply correspond to the full shift of the momentum $p \rightarrow p \pm k/2$ in the particle (hole) wave functions. As a result, the normal current vanishes after the momentum integration over the entire Brillouin zone. After subtracting the zero normal current, we arrive at Eq. (24).

For low temperature $T \ll |\Delta|$, the last two lines in Eq. (24) turn to zero. The total current thus becomes
\[ I = deNk \int \frac{\xi_p d \xi_p}{2\pi h} |C|^2 \left[ \frac{1}{\xi_p} - 2 \frac{(u_+ v_- - v_+ u_-)^2}{(E_0^+ + E_0^-)} \right] \right] \left( 1 - \frac{\xi^3_p}{E_0^3} \right) \right] \right]. \] (26)

which is similar to the result obtained in Ref. 14. For $\mu = 0$ we have
\[ I = deNk \int_0^{\infty} \frac{d \xi_p}{2\pi h} |C|^2 \left( 1 - \frac{\xi^3_p}{E_0^3} \right) \right] \right]. \]
For large $N$ one can consider $p$ as a slow function as compared to $\xi_p$. This is equivalent to the assumption that $d \left[ \xi_p (1 - v_F^2 p^2/t^2) \right]/dp = (1 - v_F^2 p^2/t^2) (d\xi_p/dp)$ i.e., that $(1 - v_F^2 p^2/t^2) \gg (2/N)(v_F^2 p^2/t^2)$. Since $\xi_p \sim \Delta$ we have

$$1 - v_F^2 p^2/t^2 = 1 - (\Delta/t)^{\frac{4}{3}} = (2/N)\ln(t/\Delta)$$

which holds for $N \gg \ln(t/\Delta)$. Therefore, the above condition is satisfied within the logarithmic approximation. Note that neglecting the terms $\zeta_n^* \zeta_n^*$ and $\zeta_n^* \zeta_n$ in Eq. (11) that decay away from the surfaces is also legitimate within the same logarithmic approximation $\ln(t/\Delta) \gg 1$. Integrating by parts and using that the integral is determined by $\xi_p \sim \Delta$ we find

$$I = \frac{eN\Delta^2k}{\pi h} \int_0^\infty \left(1 - \frac{v_F^2 p^2}{t^2}\right) \xi_p d\xi_p = \frac{2e\Delta \ln(t/\Delta)k_F}{\pi h}.$$

The total current does not depend on the sample thickness $N\delta$ as it should be for the surface superconductivity. The critical current is determined by $\max(k_F) \sim \xi_0^{-1}$ where the coherence length is $\xi_0 \sim h/p_{FB} = hv_F/t_F$,

$$I_c \sim e\Delta \ln(t/\Delta)p_{FB}.$$

For nonzero $\mu$ we find in the same way as in Ref. 14

$$I = \frac{e\ln(t/\Delta)k}{\pi h} \left[ \sqrt{|\mu|^2 + \Delta^2} + \frac{\Delta^2}{|\mu|} \ln \left( \frac{|\mu| + \sqrt{|\mu|^2 + \Delta^2}}{|\Delta|} \right) \right].$$

Recall that Eq. (27) holds for $T \ll |\Delta|$. As distinct from the case of intrinsic superconductivity in graphene considered in Refs. 14,18,19, the surface superconductivity gap $|\Delta|$ is suppressed by doping, such that both $|\Delta|$ and $T_c$ vanish as $\mu$ reaches the critical level $\mu_c = 2T_{c0}$.

To conclude, we have calculated the zero-temperature supercurrent for the surface superconductivity of a flat-band multilayered rhombohedral graphene. The supercurrent is finite despite the absence of dispersion of the excitation spectrum. The critical current is proportional to the zero-temperature gap, i.e., to the superconducting critical temperature and to the size of the flat band in the momentum space. Nonzero surface supercurrent can be responsible for the small Meissner effect and for the sharp drop in resistance seen in experiments on graphite. The enhanced superconducting density has been reported on twin boundaries in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$. This observation can also be considered as indications towards surface superconductivity described by our theory.

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