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Proximity-induced superconductivity in Landau-quantized graphene monolayers

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We consider massless Dirac fermions in a graphene monolayer in the ballistic limit, subject to both a perpendicular magnetic field B and a proximity-induced pairing gap Δ . When the chemical potential is at the Dirac point, our exact solution of the Bogoliubov–de Gennes equation yields Δ -independent relativistic Landau levels. Since eigenstates depend on Δ , many observables nevertheless are sensitive to pairing, e.g., the local density of states or the edge state spectrum. By solving the problem with an additional in-plane electric field, we also discuss how snake states are influenced by a pairing gap.

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Introduction. It is well known that at energies close to the neutrality point, the electronic properties of graphene monolayers are accurately described in terms of two-dimensional (2D) massless Dirac fermions [1–7]. Recent advances in fabrication and preparation technology [6,8] allow experimentalists to routinely reach the ballistic (disorder-free) transport regime. Our theoretical work reported below is largely motivated by spectacular recent progress on Josephson transport in ballistic graphene flakes contacted by conventional superconductors [9–19], demonstrating in particular that proximity-induced superconductivity can coexist with rather high (Landauquantizing) magnetic fields [12,15,18]. This raises the question of how a proximity-induced bulk pairing gap will affect the electronic properties of graphene in an orbital magnetic field. In contrast to lateral graphene-superconductor interfaces, where theory is well developed [3,20-22], we therefore investigate vertical hybrid structures, as shown schematically in Fig. 1. Superconductivity can be proximity induced in the graphene sample from a 2D van der Waals superconductor [23], e.g., using a NbSe₂ film supported on a standard hexagonal boron nitride (h-BN) substrate [8]. NbSe₂ is a good superconductor with a high critical field ($B_{c2} \approx 5$ T at T =1 K), remaining superconducting down to a few monolayers, and exhibiting high-quality interfaces with graphene [13]. For gating the device, another h-BN monolayer may be inserted, as indicated in Fig. 1, at the expense of reducing the proximity gap. The proximitized graphene flake can be probed by a scanning tunneling microscope (STM), e.g., using a graphite finger tip for ultrahigh-energy resolution [19].

Before turning to derivations, we briefly summarize our main results which can be tested by established STM techniques [24], transport experiments, and/or the local manipulation of defect charges in the substrate [25]: (i) By means of an exact solution of the Bogoliubov–de Gennes (BdG) equation, we show that at the Dirac point, i.e., for a chemical potential $\mu=0$, the energy spectrum of a proximitized graphene layer in a homogeneous magnetic field B is independent of the proximity gap Δ . The BdG spectrum thus reduces to the familiar relativistic Landau level spectrum [4], in marked difference to the time-reversal-symmetric case with a strain-induced pseudomagnetic field where the spectrum depends on Δ in a conventional manner [26–28]. (ii) Even though

the energy spectrum is independent of Δ at the Dirac point, the corresponding eigenstates are sensitive to the pairing gap. Clear experimental signatures of proximity-induced superconductivity in Landau-quantized graphene are predicted for the energy-resolved local density of states (DOS) as well as for the edge states present near the sample boundaries. Away from the Dirac point, also the spectrum itself depends on Δ . (iii) Chiral snakelike states are expected in graphene for $\Delta=0$ in the presence of a weak electric field $\mathcal E$ perpendicular to B [29–31]; see Refs. [32,33] for recent experimental reports. We solve the corresponding BdG equation for arbitrary Δ through a Lorentz transformation of our solution for case (i), and thereby discuss how snake states are affected by a pairing gap.

Model. We start from the BdG equation, $H\Psi = E\Psi$, for proximitized graphene samples, as in Fig. 1. The BdG Hamiltonian is represented by the matrix [3,20],

$$H = \begin{pmatrix} v_F(\hat{\boldsymbol{p}} + \frac{e}{c}\boldsymbol{A}) \cdot \boldsymbol{\sigma} + V & \Delta \\ \Delta^* & -v_F(\hat{\boldsymbol{p}} - \frac{e}{c}\boldsymbol{A}) \cdot \boldsymbol{\sigma} - V \end{pmatrix}, \tag{1}$$

with canonical momentum $\hat{\boldsymbol{p}} = (\hat{p}_x, \hat{p}_y) = -i\hbar\nabla$ and Fermi velocity $v_F \approx 10^6$ m/s. Pauli matrices $\sigma_{x,y}$ act in sublattice space, while explicitly written 2 × 2 matrices refer to Nambu (particle-hole) space throughout. In particular, H in Eq. (1)acts on Nambu spinors $\Psi(\mathbf{r}) = (u, v)^T$ containing the spin-up electronlike (spin-down holelike) wave function u(v) near the K(K') valley, where u and v are spinors in sublattice space and $\mathbf{r} = (x, y)$. A decoupled identical copy of H with opposite spin is kept implicit [20]. The vector potential $\mathbf{A} = (0, Bx)$ describes a perpendicular homogeneous magnetic field B in Landau gauge, where we neglect the typically small Zeeman splitting. The potential term in Eq. (1) also accounts for the chemical potential μ through the shift $V - \mu \rightarrow V$, and the homogeneous spin-singlet pairing amplitude Δ (taken real positive below) comes from the proximity effect. Note that intrinsic superconductivity in graphene [34,35] has not been found experimentally. Finally, we neglect Coulomb interactions, which are largely screened off by the proximityinducing superconductor. In what follows, we measure lengths (wave numbers) in units of the magnetic length l_B (1/ l_B), and

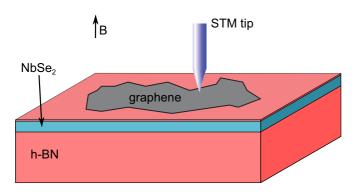


FIG. 1. Sketch of a vertical hybrid structure in a perpendicular magnetic field *B*, where the graphene flake is deposited on a superconducting film (e.g., a few monolayers of NbSe₂) supported by an h-BN substrate. Inserting an h-BN monolayer between the superconductor and the graphene sample allows one to gate the device (gates not shown). The graphene layer may be probed by an STM tip, as indicated. Alternatively, the stack could be closed by a top h-BN monolayer.

energies in units of the cyclotron scale E_B , where

$$l_B = \sqrt{\hbar c/eB}, \quad E_B = \hbar v_F/l_B. \tag{2}$$

Equation (1) tacitly assumes applied magnetic fields below the critical field of the proximity-inducing superconductor and that the Meissner effect is too weak to completely expel the magnetic field from the proximitized graphene layer. In principle, renormalized values of B and Δ entering Eq. (1) can be obtained from self-consistency equations (cf. Refs. [36,37]). However, since the coexistence of B and Δ has already been observed in graphene [12,15,18] and other 2D electron gases [38], we here take them as effective parameters and focus on the physics caused by their interplay.

Chiral representation. It is convenient to reformulate Eq. (1) using 4×4 Dirac matrices in the chiral representation, $\beta = \begin{pmatrix} 0 & -\sigma_0 \\ -\sigma_0 & 0 \end{pmatrix}$ and $\alpha^j = \begin{pmatrix} \sigma_j & 0 \\ 0 & -\sigma_j \end{pmatrix}$, with j=1,2,3 and identity σ_0 in sublattice space. Anticommuting γ^{ν} matrices are then given by $\gamma^0 = \beta$ and $\gamma^j = \beta \alpha^j$, where we also define $\gamma^5 = \text{diag}(\sigma_0, -\sigma_0)$. In Landau gauge, Eq. (1) is equivalently expressed as

$$H = \alpha^1 \hat{p}_x + \alpha^2 (\hat{p}_y + x\gamma^5) + \gamma^5 V - \beta \Delta. \tag{3}$$

Formally, Eq. (3) describes 2D Dirac fermions with mass $-\Delta$ subject to pseudovector and pseudoscalar potentials: The A and V terms involve γ^5 . Given a BdG eigenstate $\Psi_E = (u_E, v_E)^T$ with energy $E \geqslant 0$, a particle-hole transformation yields a solution with energy -E,

$$\Psi_{-E}(\mathbf{r}) = -\gamma^2 \Psi_E^*(\mathbf{r}) = \begin{pmatrix} -\sigma_y v_E^*(\mathbf{r}) \\ \sigma_y u_E^*(\mathbf{r}) \end{pmatrix}. \tag{4}$$

Therefore it is sufficient to find solutions with $E \geqslant 0$, and Eq. (4) is a self-conjugation relation for E=0. For a complete set $(u_{\lambda}, v_{\lambda})^T$ with energies $E_{\lambda} \geqslant 0$, the local DOS $\rho(E)$ is defined in a standard way [39] and can be measured by STM techniques (see Fig. 1). Furthermore, the charge current density $J = (J_x, J_y)^T$ corresponding to a given eigenstate is

$$\boldsymbol{J}_{\lambda}(\boldsymbol{r}) = -ev_F(u_{\lambda}^{\dagger}\boldsymbol{\sigma}u_{\lambda} + v_{\lambda}^{\dagger}\boldsymbol{\sigma}v_{\lambda}). \tag{5}$$

In what follows, we assume V = V(x) such that Eq. (3) enjoys translation invariance along the y direction. BdG solutions are given by $\Psi_k(\mathbf{r}) = e^{iky}\psi_k(x)$, where $\psi_k(x)$ is an eigenstate to H_k obtained from H in Eq. (3) with $\hat{p}_y \to k$. We now perform a partial (involving only the momentum in the y direction) Bogoliubov transformation $\psi_k(x) = M_k \phi_k(x)$, with the unitary 4×4 matrix

$$M_{k} = a_{k,+} - a_{k,-} \gamma^{2} = \begin{pmatrix} a_{k,+} & -\sigma_{y} a_{k,-} \\ \sigma_{y} a_{k,-} & a_{k,+} \end{pmatrix},$$

$$a_{k,\pm} = \sqrt{\frac{X_{k} \pm k}{2X_{k}}}, \quad X_{k} = \sqrt{k^{2} + \Delta^{2}}.$$
(6)

The BdG equation, $\tilde{H}_k \phi_k(x) = E \phi_k(x)$ with $\tilde{H}_k = M_k^{-1} H_k M_k$, then involves the transformed Hamiltonian

$$\tilde{H}_k = \alpha^1 \hat{p}_x + \alpha^2 (X_k + x\gamma^5) + \frac{k + \gamma^2 \Delta}{X_k} \gamma^5 V(x).$$
 (7)

For B=0 and constant V, one has plane waves with $\mathbf{k}=(k_x,k)$ and energy $E_{\mathbf{k},\pm}=\sqrt{(\pm\hbar v_F|\mathbf{k}|+V)^2+\Delta^2}$ [20], where the DOS for $E\geqslant 0$ and $V\geqslant 0$ is given by

$$\rho(E) = \frac{1}{\pi (\hbar v_F)^2} \times \begin{cases} 0, & E < \Delta, \\ \frac{EV - (E^2 - \Delta^2)}{\sqrt{E^2 - \Delta^2}}, & \Delta < E < \sqrt{V^2 + \Delta^2}, \\ E - V, & E > \sqrt{V^2 + \Delta^2}. \end{cases}$$
(8)

Note that at the Dirac point, i.e., for V=0, the usual BCS square-root singularity is replaced by a finite jump at $E=\Delta$, with $\rho(E)\sim E$ for $E>\Delta$.

Exact solution at the Dirac point. For V=0, we next observe that \tilde{H}_k in Eq. (7) coincides with the original Hamiltonian in Eq. (3) for $\Delta=0$ and $\hat{p}_y\to X_k$. As a consequence, the entire spectrum coincides with the (k,Δ) -independent relativistic Landau energies, $E_{k,n,s}=E_n=\sqrt{2n}E_B$ with $n=0,1,2,\ldots$ [4]. On top of the k degeneracy, we have an additional double degeneracy with $s=\pm$ (see below). Eigenstates follow by the above M_k transformation from relativistic Landau states. The latter are given by the Nambu spinors $\phi_{k,n,+}(x)=[\mathcal{F}_n(x+X_k),0]^T$ and $\phi_{k,n,-}(x)=[0,\sigma_y\mathcal{F}_n(x-X_k)]^T$, where sublattice spinors $\mathcal{F}_n(x)=(\frac{1}{\sqrt{2}})^{1-\delta_{n,0}}[\mathrm{sgn}(n)\varphi_{|n|-1},i\varphi_{|n|}]^T$ are expressed in terms of normalized oscillator eigenfunctions [40]. Note that the usual center-of-mass coordinate k is replaced by $X_k(-X_k)$ for the electron (hole) spinor component [cf. Eq. (7)]. Using Eq. (6), eigenstates follow as

$$\Psi_{k,n,s=\pm}(\mathbf{r}) = e^{iky} \begin{pmatrix} \pm a_{k,\pm} \mathcal{F}_n(x \pm X_k) \\ a_{k,\mp} \sigma_y \mathcal{F}_n(x \pm X_k) \end{pmatrix}. \tag{9}$$

In contrast to the spectrum, these states depend on Δ and thus most observables will be sensitive to pairing. For given $\Psi_{k,n,s}$, Eq. (4) yields a mirror state $\Psi_{-k,-n,\pm}(\mathbf{r}) = \pm \gamma^2 \Psi_{k,n,\pm}^*(\mathbf{r})$ with $E = -E_n$. For n = 0, this relation connects +k and -k states, and one can construct two $(s = \pm)$ 1D zero-energy Majorana fields.

Density of states at the Dirac point. By using the states in Eq. (9) and restoring units, we obtain an exact integral

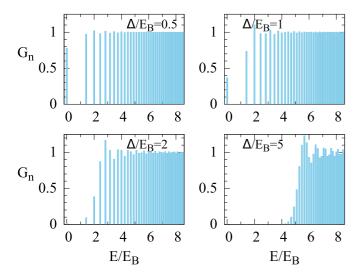


FIG. 2. Bar plots of the DOS weights G_n vs Landau energy E_n for different Δ/E_B [see Eqs. (10) and (11)].

representation for the DOS [41],

$$\rho(E) = \frac{e^{-(\Delta/E_B)^2}}{\pi l_B^2} \delta(E) + \frac{|E|}{\pi (\hbar v_F)^2} \int_{-\infty - i0^+}^{+\infty - i0^+} \frac{d\lambda}{2\pi i} \times e^{i(E^2\lambda - \Delta^2 \tan \lambda)/E_B^2} \cot \lambda, \tag{10}$$

which is singular and applies in the distribution sense. For $B \to 0$, the asymptotic approximation of Eq. (10) reproduces Eq. (8) with V = 0. The bar plots in Fig. 2 show the dimensionless DOS weights

$$G_n = \pi l_B^2 \int_{E_n - 0^+}^{E_n + 0^+} dE \rho(E), \quad E_n = \sqrt{2n} E_B,$$
 (11)

characterizing the $\delta(|E|-E_n)$ peaks in the DOS and hence also the degeneracy per unit area of the energy levels E_n . For $\Delta \to 0$, Eq. (10) yields the standard Landau comb with $G_n=1$. Figure 2 illustrates the crossover between the analytically accessible limits $\Delta/E_B \to 0$ and $\Delta/E_B \to \infty$, where low-energy states with $|E| < \Delta$ become gradually depleted as Δ/E_B increases. The DOS in Fig. 2 also exhibits oscillatory features in the energy dependence.

Edge states. Next, we consider a semi-infinite graphene sheet (x < 0) with V = 0. The boundary is modeled by imposing armchair conditions [3,4] along the line x = 0. Solutions to the BdG equation are then given in terms of parabolic cylinder functions $D_p(z)$ [42]. The spectrum is obtained by numerically solving $\det[W(E)] = 0$, where the matrix W follows with $\epsilon = E/\sqrt{2}$, $a_{\pm} = a_{k,\pm}$ [cf. Eq. (6)], and $\tilde{D}_p^{(\pm)} = D_p[\pm\sqrt{2(k^2+\Delta^2)}]$ in the form [41]

$$\begin{pmatrix}
-a_{+}\epsilon \tilde{D}_{\epsilon^{2}-1}^{(-)} & a_{+}\tilde{D}_{\epsilon^{2}}^{(-)} & a_{-}\epsilon \tilde{D}_{\epsilon^{2}-1}^{(+)} & a_{-}\tilde{D}_{\epsilon^{2}}^{(+)} \\
a_{+}\tilde{D}_{\epsilon^{2}}^{(-)} & -a_{+}\epsilon \tilde{D}_{\epsilon^{2}-1}^{(-)} & -a_{-}\tilde{D}_{\epsilon^{2}}^{(+)} & -a_{-}\epsilon \tilde{D}_{\epsilon^{2}-1}^{(+)} \\
a_{-}\tilde{D}_{\epsilon^{2}}^{(-)} & -a_{-}\epsilon \tilde{D}_{\epsilon^{2}-1}^{(-)} & a_{+}\tilde{D}_{\epsilon^{2}}^{(+)} & a_{+}\epsilon \tilde{D}_{\epsilon^{2}-1}^{(+)} \\
a_{-}\epsilon \tilde{D}_{\epsilon^{2}-1}^{(-)} & -a_{-}\tilde{D}_{\epsilon^{2}}^{(-)} & a_{+}\epsilon \tilde{D}_{\epsilon^{2}-1}^{(+)} & a_{+}\tilde{D}_{\epsilon^{2}}^{(+)}
\end{pmatrix}.$$
(12)

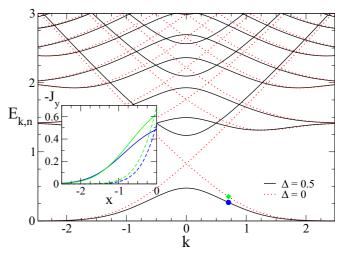


FIG. 3. Edge states for a semi-infinite (x < 0) graphene sheet with V = 0 and armchair conditions at x = 0. Main panel: Dispersion relation for $\Delta = 0.5E_B$ (solid black) and for $\Delta = 0$ (red dotted curves). Inset: Current density $J_y(x)$ (in units of $-ev_F$) vs position x/l_B for the two degenerate eigenstates (solid and dashed curves for s = + and s = -, respectively) with $kl_B = 0.705$. Blue (green) curves are for $\Delta/E_B = 0.5$ ($\Delta = 0$) with $E_{k,n,s}/E_B \simeq 0.2683$ ($\simeq 0.3520$) [cf. the blue circle (green diamond) in the main panel].

The spectrum is shown in Fig. 3. For $\Delta=0$, we recover earlier results [43–45] reporting chiral edge states. For $\Delta>0$, electron- and hole-type edge states become mixed and the edge state dispersion exhibits gaps near k=0. Turning to the current density (5), the current flows along the y direction only, $J_x=0$. The respective profile $J_y(x)$ is illustrated for the two degenerate states with k=0.705 and lowest energy in the inset of Fig. 3. Since the current density has a pronounced peak near x=0 and a specific sign, we have unidirectional edge states also for $\Delta>0$. However, the overall current becomes smaller with increasing Δ (cf. Fig. 3).

Going away from the Dirac point. Let us briefly address the case $V \neq 0$, where numerical diagonalization of the BdG equation using Landau states as a basis shows that a (chemical) potential shift causes dispersion (see Fig. 4). Notably, most features in Fig. 4 can be understood by expanding around the V=0 solution (9) using the term $\sim V$ in Eq. (7) as a small perturbation. Writing $E_{k,n,s}=E_n+\delta E_{k,n,s}$, first-order degenerate perturbation theory yields the correction

$$\delta E_{k,n,\pm} = \pm \frac{|V|}{X_k} \sqrt{k^2 + \Delta^2 S_{k,n}^2},\tag{13}$$

where the overlap between Landau states \mathcal{F}_n centered at $+X_k$ and $-X_k$ is encoded by $S_{k,n}$. Explicitly, we find $S_{k,0}=e^{-X_k^2}$ and $S_{k,n>0}=\frac{1}{2}e^{-X_k^2}[L_{n-1}(2X_k^2)+L_n(2X_k^2)]$ with the Laguerre polynomials L_n [42]. For $|k|\gg \Delta$, Eq. (13) yields a uniform shift $\pm |V|$ of all Landau energies, while for k=0, the correction simplifies to $\pm |V| S_{0,n}$, where $S_{0,n}$ oscillates when changing n.

Crossed electric and magnetic fields. We finally also include an in-plane electric field \mathcal{E} by putting $V = e\mathcal{E}x$. With the dimensionless parameter $\varepsilon = (c/v_F)\mathcal{E}/B$, we consider the regime $|\varepsilon| < 1$. The corresponding $\Delta = 0$ problem has been

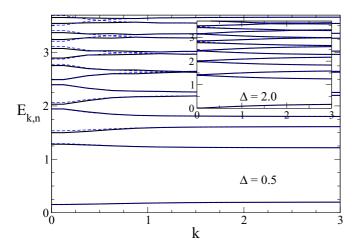


FIG. 4. Dispersion relation for an infinite graphene sheet with potential $V = 0.2E_B$ for $\Delta = 0.5E_B$ (main panel) and $\Delta = 2E_B$ (inset). Since $E_{-k,n} = E_{k,n}$, only $k \ge 0$ is shown. Solid black and dashed blue curves refer to numerical diagonalization and perturbative results [Eq. (13)], respectively.

solved analytically by a Lorentz boost into the reference frame with vanishing electric field $(\mathcal{E}'=0)$ [29]. Remarkably, such a strategy also admits an exact solution for $\Delta \neq 0$: First, we write down the spinor transformation law, $\psi = S\psi'$ with $S = \cosh(\eta/2) - \sinh(\eta/2)\gamma^0\gamma^2$, where the Lorentz angle $\eta = \tanh^{-1}\varepsilon$ defines the frame with $\mathcal{E}'=0$. Next, using the parameter $\zeta \equiv (1-\varepsilon^2)^{1/4}$, we rescale (i) the x coordinate $x' = \zeta x$, (ii) the wave number $k' = (k+\varepsilon E)/\zeta^3$, (iii) energy $E' = (E+\varepsilon k)/\zeta^3$, and (iv) the proximity gap $\Delta' = \Delta/\zeta$. With these rescalings and $X_k' = \sqrt{k'^2 + \Delta'^2}$ [cf. Eq. (6)], the BdG equation in the new frame coincides with the V=0 problem solved above. Transforming the solution, Eq. (9), back to the laboratory frame and restoring units, we obtain the Δ -independent spectrum

$$E_{k,n,s} = -\hbar \varepsilon v_F k + \operatorname{sgn}(n) \sqrt{2|n|} \zeta^3 E_B, \tag{14}$$

where n runs over all integers and k is restricted to those values with $E_{k,n,s} \ge 0$. Each level is twofold degenerate ($s = \pm$), and the corresponding eigenstates are

$$\Psi_{k,n,\pm}(\mathbf{r}) = e^{iky} \zeta^{3/2} \left[\cosh(\eta/2) \begin{pmatrix} \pm a_{k',\pm} \mathcal{F}_n(x' \pm X_k') \\ a_{k',\mp} \sigma_y \mathcal{F}_n(x' \pm X_k') \end{pmatrix} + \sinh(\eta/2) \begin{pmatrix} \mp a_{k',\pm} \sigma_y \mathcal{F}_n(x' \pm X_k') \\ a_{k',\mp} \mathcal{F}_n(x' \pm X_k') \end{pmatrix} \right].$$
(15)

States with negative energy follow from Eq. (4), and for $\varepsilon = 0$, Eq. (15) reduces to Eq. (9).

In the normal ($\Delta=0$) case, so-called snake states exist near the interface between the V>0 and V<0 regions [30–33], which are semiclassically described by snakelike orbits propagating along the interface (here the y direction) with velocity $c\mathcal{E}/B=\varepsilon v_F$. In the superconducting case ($\Delta>0$), the spectrum in Eq. (14) suggests that unidirectional snake states remain well defined and propagate with the same snake velocity as for $\Delta=0$. In particular, for n=0, these states are localized near the line x=0. Computing the total charge current carried by a given state along the y direction, $I=\int dx J_y(x)$, Eqs. (5) and (15) yield the analytical result $I(\Delta)/I(0)=1/\sqrt{1+(\Delta'/k')^2}$. Similar to the above edge state case, we thus find that the magnitude of the current becomes gradually suppressed with increasing Δ .

Conclusions. We have studied the electronic properties of graphene monolayers in an orbital magnetic field when also proximity-induced pairing correlations are present. Remarkably, at the Dirac point, the energy spectrum is independent of Δ , but observables may still show pronounced pairing effects since eigenstates depend on Δ . We hope that our work will stimulate experimental and further theoretical work on the coexistence of magnetism and superconductivity in graphene.

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