Superconducting States in pseudo-Landau Levels of Strained Graphene

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(Dated: May 5, 2014)

We describe the formation of superconducting states in graphene in the presence of pseudo-Landau levels induced by strain, when time reversal symmetry is preserved. We show that superconductivity in strained graphene is quantum critical when the pseudo-Landau levels are completely filled, whereas at partial fillings superconductivity survives at weak coupling. In the weak coupling limit, the critical temperature scales linearly with the coupling strength and shows a sequence of quantum critical points as a function of the filling factor that can be accessed experimentally. We argue that superconductivity can be induced by electron-phonon coupling and that the transition temperature can be controlled with the amount of strain and with the filling fraction of the Landau levels.

PACS numbers: 71.27.+a, 73.20.Hb, 75.30.Hx

Graphene is a single atomic sheet of carbon with electronic excitations that behave as massless Dirac quasiparticles\textsuperscript{1, 2}. In general, graphene seems to be insensitive to electronic many body instabilities\textsuperscript{3}, except in the quantum Hall regime\textsuperscript{4, 5}, where fractional quantum Hall states\textsuperscript{6, 9} have been observed. We claim that one promising route to induce intrinsic superconductivity in graphene is to reconstruct the electronic density of states (DOS) into a discrete spectrum of Landau levels (LLs) with the application of strain fields. Current experiments observed the spontaneous formation of LLs on top of graphene nanobubbles\textsuperscript{2}, in deformed artificial graphene lattices, formed by a honeycomb grid of molecules sitting on a metallic surface\textsuperscript{10, 11} and in chemical vapor deposition grown graphene\textsuperscript{12, 13}. In specific engineered forms\textsuperscript{12, 13}, applied strain configurations in graphene mimic the application of strong uniform magnetic fields that can be as large as 300T\textsuperscript{12, 11}, but produce no net magnetic flux, preserving time reversal symmetry (TRS). Quantum Hall states induced by pseudomagnetic fields have been conjectured to give rise to topological order in strained graphene with spontaneously broken TRS\textsuperscript{14, 15}.

In this letter, we describe the formation of intrasublattice TRS spin singlet states, which occupy the LLs on top of graphene with spontaneously broken TRS\textsuperscript{16–19} and in deformed artificial graphene lattices, formed by a honeycomb grid of molecules sitting on a metallic surface\textsuperscript{10, 11} and in chemical vapor deposition grown graphene\textsuperscript{12, 13}. The in-plane components of the pseudo magnetic field are described by $A_x = u_{xy}$ and $A_y = \frac{1}{2}(u_{xx} - u_{yy})$, and correspond respectively to strain and shear, where $u_{ij} = \nabla_j u_i + \nabla_i u_j + \nabla_i u_z \nabla_j u_z$ is the strain tensor, with $u = (u_x, u_y, u_z)$ the deformation vector of the lattice normalized by the lattice constant\textsuperscript{12}. Although we assume strain configurations which produce approximately uniform pseudo-magnetic fields\textsuperscript{12, 13}, $B_s = \nabla \times A_s$, this restriction is not required for a macroscopic superconducting state to emerge\textsuperscript{20}.

In the Landau gauge, where $A_s = (−B_s y, 0)$, with $B_s$ the pseudo magnetic field, the electronic wavefunction takes the form $\Psi_{k,\sigma}(x, y) = \exp(ikx) \Theta_\sigma(y)$, where $\Theta_\sigma(y)$ is the eigenspinor of a 1D Hamiltonian. This Hamiltonian can be expressed in terms of ladder operators...
of the 1D harmonic oscillator, $a \equiv (\xi + \partial \xi) / \sqrt{2}$, $a^\dagger \equiv (\xi - \partial \xi) / \sqrt{2}$, where $\xi = \ell_B k - y / \ell_B$ is a dimensionless variable related to the valley dependent guiding center $X = -k \ell_B^2$, with $\ell_B = \sqrt{\hbar / e B_z}$ (restoring $\hbar$) the effective magnetic length, and $e$ the electron charge. In what follows, we define the valley dependent operator $\tilde{D}(\xi)$,

$$v(-i \nabla + A_\sigma) \cdot \sigma = \sqrt{\frac{2}{\ell_B}} \frac{v}{a \ d^a}{a^\dagger \ a} \equiv \tilde{D}(\xi),$$

which takes the form $-v(-i \nabla - A_\sigma) \cdot \sigma = -\tilde{D}(\xi)$ in the opposite valley, with $\xi = \ell_B k + y / \ell_B$.

In the presence of an effective attractive potential $U$ that stabilizes the superconducting state, the Bogoliubov-deGennes (BdG) Hamiltonian is

$$\hat{H}_{BG} = \int d^3 x \Phi^\dagger(x) \hat{H}_{BG} \Phi(x),$$

$$\hat{H}_{BG} = \left( \begin{array}{cc} \hat{H}_0(A_\sigma) & \tilde{\Delta} \\ \tilde{\Delta}^* & -T \hat{H}_0(A_\sigma) T^{-1} \end{array} \right),$$

where $\hat{H}(A_\sigma) = \tilde{D}(\xi) \otimes \nu^+ - \tilde{D}(\xi) \otimes \nu^- - \mu \sigma_0 \sigma_\alpha$ is the normal state Hamiltonian of strained graphene written in valley and sublattice spaces, $\nu^\pm = (\nu_0 \pm \nu z)/2$ are projectors in the $\pm$ valley spaces, with Pauli matrices $\nu_i (i = x, y, z)$, and $\Phi = (\Psi_{k,\sigma}, \Psi_{-k,\sigma}^\dagger)$ is the 8 component spinor in the Nambu space, with Pauli matrices $\tau_i (i = x, y, z)$. The off diagonal term, $\tilde{\Delta}$, is a pairing matrix that describes the formation of Cooper pairs $\Delta^2 = U \tr(\Psi_{k,\sigma}^\dagger \Delta \Psi_{-k,\sigma}^\dagger \sigma_\alpha)$. In strained graphene, the time reversal symmetry operation $T \hat{H}_0(A_\sigma) T^{-1}$ leaves the Hamiltonian invariant under an additional exchange between valleys, in contrast with the case of conventional magnetic fields, which explicitly break TRS [21].

In the intra-sublattice $s$-wave pairing state, which corresponds to the pairing matrix $\Delta = \Delta_0 \sigma_\alpha \sigma_\beta$, the eigenvalue problem $\hat{H}_{BG} \Phi(x, \xi) = E \Phi(x, \xi)$ can be solved by decomposing Hamiltonian [3] into two equivalent copies of $4 \times 4$ BdG Hamiltonians in pseudospin and Nambu spaces. In the reduced Nambu basis $\Phi = (\Psi_{1,\sigma}, \Psi_{1,-\sigma}^\dagger)$,

$$\hat{H}_{BG} = \left( \begin{array}{cc} \tilde{D}(\xi) - \mu \Delta & \tilde{\Delta} \\ \tilde{\Delta}^* & -\tilde{D}(\xi) + \mu \end{array} \right).$$

Fixing the gap of the $\Delta$ to be real, the eigenvalue problem $(E - \hat{H}_{BG}) \Phi = 0$ is equivalent to $M \Phi = (E + \hat{H}_{BG}) \Phi = 0$ where $\hat{H}_0 = \hat{C} \hat{H}_{BG} \hat{C}^{-1} = (\tilde{D} + \mu) \otimes \tau_3 + \Delta_1$, is the charge conjugated BdG Hamiltonian ($\mu \rightarrow -\mu$). When the matrix $\mathcal{M}$ is applied in a proper basis, $\mathcal{M}$ can be cast in the form $\mathcal{M} \Phi = (E + \hat{H}_0)(E - \hat{H}_0) \Phi$, with $\hat{H}_0 = (\pm \omega_c \sqrt{\nu} \Delta_{\pm} + \Delta_{\mp})$, where $\nu$ is the index of the Landau levels, $s(\nu) \equiv \pm 1$ accounts for the two branches of LLs in the conduction and valence bands, and $\omega_c = \sqrt{2e / \ell_B}$. $\hat{H}_0$ is equivalent to Hamiltonian [14] and gives the energy spectrum

$$\pm E_N = \pm [(s \omega_c \sqrt{\nu} \Delta_{\mp} + \Delta_{\pm})^2 + \Delta^2]^{1/2}.$$

Figure 1: Dependence of the chemical potential $\mu$ in units of $\omega_c$ with the filling factor $\nu$. Red (light) curve: normal state $\Delta = 0$. Solid black curves: superconducting state, with $\Delta / \omega_c$ ranging from 0 (red line) to 1. All curves are plotted at $T / \omega_c = 0.02$. Inset: detail of the zero LL $(n = 0)$. The red curve is analytically described by Eq. $\mu$ in the $T \rightarrow 0$ limit.

with eigenstates given by

$$\Psi^{(N)}_{\pm,\sigma,\alpha}(\xi) = \beta^{N}_{\pm,\sigma,\alpha}(\xi) \Phi_{N}(\xi) \left( \begin{array}{c} \phi_{N-1}(\xi) \\ s_0(\xi) \phi_N(\xi) \end{array} \right) \exp[i k x],$$

for the states $(\sigma, \alpha) = (\uparrow, -)$ and $(\downarrow, +)$, where $\beta^{N}_{\pm,\tau,\pm} = 1 / \sqrt{2} (\pm \omega_c \sqrt{\nu} \Delta_{\pm} + \Delta_{\mp})$ and $\beta^{N}_{\pm,\tau,\mp} = \mp 1 / \sqrt{2} (\pm \omega_c \sqrt{\nu} \Delta_{\mp} - \Delta_{\pm})$. $\phi_N(\xi)$ denotes conventional LL wavefunctions, with $\phi_{-1}(\xi) = 0$. In the zero LL, the Cooper pairs occupy only one sublattice, explicitly breaking the $\mathbb{Z}_2$ sublattice symmetry of graphene. As anticipated, the BdG quasiparticle spectrum is discrete and can be indexed by the LL index $N$. The superconducting ground state is given by [22]

$$|\Psi_0\rangle = \prod_{N,X} (\nu_N + \nu_N c^\dagger_{N,X,\uparrow} - c^\dagger_{N,X,\downarrow}) |0\rangle,$$

where $\nu_N = \beta^{N}_{\uparrow,\tau,\uparrow}$, $\nu_N = -\beta^{N}_{\downarrow,\tau,\uparrow}$, and $c^\dagger_{N,X,\sigma,\alpha}$ are fermionic creation operators of the relativistic LLs. This wavefunction describes intrasublattice pairing across opposite valleys, within the same LL. As in usual spin singlet superconductivity, the essence of Cooper phenomenon, that TRS states can pair up, is preserved by the discrete spectrum of LLs.

The discontinuous behavior of the chemical potential with the pseudomagnetic field and filling factor can be calculated by fixing the total number of particles in the system. Although the ground state wave function does not conserve the number of particles, the distribution is sharply peaked around the average $N$ in the thermodynamic limit [23], $N = g N_\phi \sum_{N = -\infty}^{\infty} \left[ a_N^\dagger f(E_N) + a_N^2 f(-E_N) \right]$, where $f(E) = (1 + e^{E/T})^{-1}$ is the Fermi distribution, $g = 4$ is the valley and spin degeneracy, and $N_\phi = A / (2 \pi \ell_B^2)$ is the number of flux quanta for a total area $A$, which sets the LLs degeneracy. In the low temperature and weak coupling regime $T, \Delta \ll v / \ell_B$, where the deep energy states $N < n$
are fully occupied, with $n$ the highest occupied LL, the constraint becomes

$$2(\nu/g - n) = -[(s\omega_c\sqrt{|n|} - \mu)/E_n] \tanh[E_n/(2T)], \quad (8)$$

where $\nu = N/N_\phi - g(N_A + 1/2)$ is the filling factor, and $N_A = (D/\omega_c)^2 > 0$ is an ultraviolet cutoff that regularizes the number of negative energy states, where $D \sim 6$eV is the bandwidth. In particular, at $T = 0$, the chemical potential

$$\mu(0, \nu) = s\omega_c\sqrt{|n|} + \Delta(n - gn) \sqrt{|g(n + 1/2) - \nu|}, \quad (9)$$

remains pinned to the $n$-th LL when half filled ($\nu = gn$) for small $\Delta$, and shows a power law divergence when the highest occupied LL is completely filled, at integer fillings $\nu = g(n \pm 1/2)$, indicating an incompressibility due to Pauli blocking. In the opposite regime, when $T, \Delta \gtrsim \nu/\nu_B$, the system crosses over to the usual Fermi liquid behavior when the electrons have multiple transitions between different LLs. In the normal state ($\Delta = 0$), as expected, the chemical potential

$$\mu(T, \nu) = s\omega_c\sqrt{|n|} + T \ln \left[ (\nu - gn)/(g(n + 1/2) - \nu) \right], \quad (10)$$

has a logarithmic divergence at integer filling. Eq. [9] and [10] describe analytically the numerical curves shown in the inset of Fig. 1 when $\Delta/\omega_c \ll 1$.

When all LLs are taken into account, these divergences are regularized, as shown in Fig. 1, leading to a sequence of jumps. At integer fillings $\nu(n) = g(n + 1/2)$, the chemical potential for the normal state does not diverge but sits half way between the LLs in the the zero temperature limit, $\hbar(n) \equiv \mu(0, \nu(n))/\omega_c = [s(n + 1)\sqrt{|n + 1|} + s(n)/\sqrt{|n|}]/2$. The red (light) curve in Fig. 1 describes the $\Delta = 0$ case at fixed temperature, while the black lines represent the superconducting case for fixed values of the gap. Unlike in conventional superconductors, in TRS LLs the chemical potential $\mu$ has a strong dependence with the gap, which must be accounted self consistently into the equation of state.

At the mean field level, the free energy of the superconducting state is $F = -TgN_\phi \sum_{}\gamma = \pm 1 \sum_{N = -\infty}^{\infty} \ln[1 + e^{-\gamma E_N(T)/T}] - A|\Delta|^2/U$, where $A$ is the total area normalized by the size of the unit cell. Minimization of the free energy gives the gap equation

$$1 = -(U/2)gN_\phi \sum_{N = \infty}^{\infty} \tanh[E_N(T, \nu)/(2T)]/E_N, \quad (11)$$

where $N_\phi = 3\sqrt{2}a^2/(4\pi\ell_2^2)$ is the number of flux quanta per unit cell, with $a = 1.42\text{Å}$ the lattice spacing. Defining $x \equiv |U|gN_\phi/\omega_c \propto |U|/\nu_B$ as the dimensionless coupling parameter that controls the strength of interactions and strain, at half filling ($\nu = 0$), the zero temperature gap in the weak coupling regime $T_c \ll \nu/\nu_B$ is

$$\Delta^{(0)}(0) = \frac{x}{(\sqrt{2}\nu_B)^3} \sqrt{1 - \zeta(3/2)/T_c}, \quad (12)$$

where $\zeta(3/2) = \sum_{N = 1}^{N_A} 1/\sqrt{N}$ is the zeta function regularized by an ultraviolet cut-off. The ratio between the critical temperature and the zero temperature gap at half filling is a universal number, $2T_c = \Delta^{(0)}(0)$. In the weak coupling limit, $T_c \sim \sqrt{2}x/\nu_B \propto B_N$ has a linear scaling with the coupling and with the amount of strain [2].

This scaling contrasts with the case of conventional weak coupling superconductors, where $T_c \propto \exp(-1/x)$ decreases exponentially with the effective coupling. As the coupling $x$ becomes larger, the system eventually crosses over to the strong coupling regime, when $T_c \gtrsim \nu/\nu_B$, as shown in Fig. 2. In the critical regime, when $\Delta/T_c \ll 1$, the gap at $\nu = 0$ is given by

$$\Delta^{(0)}(T) = \frac{x}{(\sqrt{2}\nu_B)^{3/2}} \sqrt{1 - T/T_c}/[\zeta(3/2)/T_c]^{1/2}, \quad (13)$$

at weak coupling, and scales with $\Delta^{(0)} \propto B_N^{3/4}$, where $\zeta(3/2) \approx 2.61$ is a zeta function. In Fig. 2a, we show the dependence of the critical temperature with the coupling $x$ for different filling factors. The red curve is the phase transition for $\nu = 2$, which is quantum critical.

At integer filling factors $\nu(n) = g(n + 1/2)$, when the highest LL is completely filled, the chemical potential of the normal state sits half way between the LLs at zero temperature. The normal state becomes incompressible, and the emergence of superconductivity requires a quantum critical coupling, which allows transitions between different LLs. At those integer fillings, the zero temperature gap is

$$\Delta^{(n)}(0) = 2\sqrt{2\pi n}(\nu_B)^{3/2} x^{1/2}/(nx_c - 1)^{1/2}, \quad (14)$$

Figure 2: Critical temperature $T_c/\omega_c$ vs. the dimensionless coupling strength $x \equiv |U|gN_\phi/\nu_B/\nu_B^0$. a) red (light) curve: $\nu = 2$, where the transition is quantum critical below $x_c^0 \approx 0.025$. Solid black curves: $\nu = 0$, 8, 24 and $\nu = 40$, from right to left. b) $\nu = 0$, 1.2, 1.6, 1.98, 1.9998 and 2, from left to right. c) $\nu = 4$, 5.2, 5.6595, 5.9998 and 6, from left to right. At partial filling of the LL, $T_c \propto x$ in the $x \to 0$ limit.

\[<\text{Figure 2: Critical temperature } T_c/\omega_c \text{ vs. the dimensionless coupling strength } x = |U|gN_\phi/\nu_B/\nu_B^0. \text{ a) red (light) curve: } \nu = 2, \text{ where the transition is quantum critical below } x_c^0 \approx 0.025. \text{ Solid black curves: } \nu = 0, 8, 24 \text{ and } \nu = 40, \text{ from right to left}. \text{ b) } \nu = 0, 1.2, 1.6, 1.98, 1.9998 \text{ and 2, from left to right}. \text{ c) } \nu = 4, 5.2, 5.6595, 5.9998 \text{ and 6, from left to right}. \text{ At partial filling of the LL, } T_c \propto x \text{ in the } x \to 0 \text{ limit.}>\]
limit \( x \ll x_n^c \). In two dimensions, the mean-field critical temperature \( T_c \) sets the onset of Cooper pair formation, while phase coherence is lost above the Kosterlitz-Thouless (KT) transition temperature \( T_{KT} < T_c \) [23], where pairs of vortices and anti-vortices unbind. Phase fluctuations will likely be relevant for transport and will be discussed elsewhere.

Besides transport measurements in long graphene junctions, where a supercurrent is expected to flow along the edges [20], one experimental signature of superconductivity in strained graphene is the specific heat at fixed volume, \( C_V = -T(\partial^2 F/\partial T^2) \). At low temperature, \( T \ll T_c \ll v/\ell_B \), the specific heat of the \( \nu = 0 \) state is \( C_V(T) = 2gN_0 \Delta^2(0)e^{-\Delta(0)/T}/T^2 \). In the quantum limit, \( \omega_c/T_c \gg 1 \), the specific heat jump at the phase transition normalized by the specific heat in the normal state for \( \nu = 0 \) is \( \Delta C/C_n = \omega_c e^{-\Delta(0)/T_c}/[16\zeta(3/2)T_c] \) which is non-universal. In the weak coupling regime \( x \ll x_0 \), where \( T_c \sim x\omega_c/4 \),

\[
(\Delta C/C_n)(x) = e^{4x}/[4x\zeta(3/2)]
\]

becomes exponentially large as \( T_c \) drops to zero. This feature is a signature of this state, and contrasts both with the specific heat jump expected for Dirac fermion superconductivity in unstrained graphene at half filling \( \Delta C/C_n \approx 0.35 \) [17] and in weak coupling superconductors in general \( \Delta C/C_n \approx 1.43 \) [23], which are universal constants.

Although strong Coulomb interactions inhibit superconductivity and can give rise to incompressible states at fractional filling factors [3], a condensate can be induced by phonons in the presence of substrates that screen the electronic repulsion at length scales larger than \( \ell_B \). The analysis of scanning tunneling spectroscopy experiments [27] in graphene on SiC for magnetic fields around 5T, when the LLs are well defined, indicate that the effective momentum independent electron phonon vertex in graphene is \( g_0 \sim 0.1eV \) for an Einstein phonon mode at the typical frequency \( \omega_{ph} \sim 0.2eV \). The analysis of scanning tunneling spectroscopy experiments [27] in graphene on SiC for magnetic fields around 5T, when the LLs are well defined, indicate that the effective momentum independent electron phonon vertex in graphene is \( g_0 \sim 0.1eV \) for an Einstein phonon mode at the typical frequency \( \omega_{ph} \sim 0.2eV \). This mode alone \( (E_2g \text{ phonon}) \) leads to an effective attraction \( U \sim -2g_0^2/\omega_{ph} \approx -0.1 \text{ eV} \). For a magnetic length of 20Å, which corresponds to \( 2 \times 10^{-3} \) flux quanta per unit cell, a net attractive coupling of that order results in a dimensionless coupling \( x \sim 0.003 \) and a critical temperature \( T_c \sim 8K \) at the \( \nu = 0 \) state.

In the current experiments where pseudomagnetic fields of 300 T were observed on the surface of graphene nanobubbles with 10nm in size each [4], a macroscopic superconducting state can emerge when the average spacing among the nanobubbles \( b \sim 40\text{nm} \) is shorter than the coherence length \( \xi \sim \nu/(x \Delta) \) [20]. For instance, a superconducting gap of \( \Delta \sim 1 \text{ meV} \) \( (T_c \sim 10K) \), corresponds to a coherence length \( \xi \sim 200\text{nm} \), which sets the length the Cooper pairs created on top of the bubbles can travel coherently in the normal unstrained regions.
A significant enhancement of the electron-phonon coupling, and as a result $T_c$, can be achieved for instance by coating graphene with ionic crystals and alkaline metals such as K, which is known to form a stable crystal on top of graphene [29–31]. This mechanism can lead to measurable transition temperatures in the regime where the broadening of the highest occupied LL due to disorder effects is small compared to the level spacing. Our analysis shows that the spin singlet states are robust, and present a sequence of quantum critical points, which can be experimentally accessed by tuning the filling factor of the LLs in the weak coupling limit of the problem.

The authors acknowledge M. Fogler, F. Guinea, K. Mullen, A. Jaefari, C. Bolech, N. Shah, C. Varma and V. Aji for discussions. BU acknowledges financial support from University of Oklahoma during the summer.

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