

Influence of Disorder on Electron-Hole Pair Condensation in Graphene Bilayers

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Graphene bilayers can condense into a state with spontaneous interlayer phase coherence that supports dissipationless counterflow supercurrents. Here, we address the influence of disorder on the graphene bilayer mean field and Kosterlitz-Thouless critical temperatures and report on a simple criteria for the survival of pair condensation.

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Introduction.—Superconductivity is due to electron pair condensation and is a spectacular and useful condensed matter phenomena. Unfortunately, its occurrence has so far been limited to relatively low temperatures, even in the case of high- T_c materials. The effective attractive interaction between two electrons that is imperative for superconductivity always relies on some artifice that diminishes the role of the bare strongly repulsive Coulomb interaction. It has recently been suggested that higher temperature superfluid behavior might be realized in graphene bilayers when conditions are favorable for Coulomb-driven electron-hole pair formation [1,2]. If realized, these systems will have spontaneous interlayer phase coherence and support a type of superflow in which opposing currents flow in opposite layers. Interesting novel transport phenomena [3,4] are expected in counterflow superfluids in which the two layers are contacted separately. One obstacle which stands in the way of realizing this state is the inevitable presence of unintended disorder. In this Letter, we derive a simple criterion for the survival of spontaneous interlayer coherence and counterflow superfluidity in graphene bilayer systems which is informed by current understanding [5] of the disorder present in these truly atomic [6] two-dimensional electron systems.

The system we consider [1,2] consists of two parallel graphene layers embedded in a dielectric. The thin barrier separating the layers suppresses interlayer tunneling [7]. External gates can shift the Dirac cones and transfer charge between the layers. Spontaneous interlayer coherence is most likely to occur [1,2] when the energetic Dirac-cone shifts are equal and opposite, so there is perfect nesting between the electron Fermi surface of the high-density layer and the hole Fermi surface of the low-density layer, i.e., when the bilayer is neutral. Nearly perfect nesting in neutral bilayers is guaranteed by the nearly perfect particle-hole symmetry of graphene's π -bands. At sufficiently low temperatures, the system is driven to condensation by the Cooper instability. Graphene is the ideal system for bilayer electron-hole condensation [8] because its bands are particle-hole symmetric, because the lack of a gap assists charge transfer, and because its linear dispersion and truly 2D character increase electron-hole pairing

energy scales at a given carrier density. The stiffness of the interlayer phase in the condensed state facilitates counterflow superfluidity and in separately contacted bilayers supports novel transport phenomena which are still relatively unexplored [3,4].

In this work, we study the influence of disorder on pair-condensation in graphene bilayers. We find that disorder suppresses both the mean-field-theory critical temperature T_c and the KT temperature T_{KT} , but that spontaneous coherence survives when

$$n_i^2 d^2 \pi \lesssim n \lesssim \frac{1}{d^2 \pi}. \quad (1)$$

Here, n_i is the density of the Coulomb scatterers that dominate [5,9] the disorder present in graphene sheets on dielectric substrates, $n = k_F^2/\pi$ is the external electric field induced carrier density in each layer, k_F is the Fermi momentum, and d is the separation between graphene layers. The right inequality in Eq. (1) expresses the requirement that interlayer interactions be comparable to intralayer interactions discussed in earlier work [1] while the left inequality places a limit on the allowable disorder strength. A window in carrier density for spontaneous coherence exists provided that $n_i d^2 \pi \lesssim 1$. Since n_i is typically in the range between 10^{11} cm^{-2} and 10^{12} cm^{-2} for graphene on SiO_2 substrates, layer separations less than about 10 nm are required for spontaneous interlayer phase coherence. In the body of this Letter, we explain the dependence of T_c and T_{KT} on the strength of disorder and derive condition (1).

Mean-field theory with disorder.—In order to focus on the role of disorder in spontaneous coherence, we simplify the mean-field theory by neglecting the inessential role of the full valence band of the high-density layer and the empty conduction band of the low-density layer. Furthermore, we incorporate the effects of the intralayer Coulomb interaction only through an implicitly renormalized Fermi energy ϵ_F . In the same spirit, the momentum dependence of the interlayer interaction is replaced by an energy cutoff v/d ($\hbar = 1$) where v is the velocity of the Dirac quasiparticles. The influence of disorder is incorpo-

rated here using the self-consistent Born approximation (SCBA).

The basic building block in this analysis is the retarded Green function matrix $G_{\sigma\sigma'}^r$ which satisfies the Dyson equation

$$(\omega\tau^{(0)} - \epsilon_k\tau^z - \Delta\tau^x - \Sigma)G^r = I. \quad (2)$$

In Eq. (2), $\epsilon_k = v_F(k - k_F)$ is the band energy, and the Pauli matrices act on layer labels $\sigma\sigma'$. The disorder and pairing self energies are, respectively,

$$\Sigma_{\sigma\sigma'}(\mathbf{k}, \omega) = n_i \sum_{\mathbf{p}} |S_{\mathbf{k}\mathbf{p}}|^2 \text{Re}\{U_{\mathbf{k}-\mathbf{p}\sigma}^* U_{\mathbf{k}-\mathbf{p}\sigma'}\} G_{\sigma\sigma'}^r(\mathbf{p}, \omega) \quad (3)$$

and

$$\Delta = -g \sum_{\mathbf{k}} |S_{\mathbf{k}\mathbf{p}}|^2 \overline{\langle c_{\mathbf{k}1}^\dagger c_{\mathbf{k}2} \rangle} \quad (4)$$

where $U_{q\sigma}$ is the disorder potential in layer σ and $S_{\mathbf{k}\mathbf{p}} = (1 + e^{i(\theta_{\mathbf{k}} - \theta_{\mathbf{p}})})/2$ is graphene's chiral form factor. The overbar in Eq. (4) denotes the average over disorder and g stands for the interlayer interaction coupling constant. Since elastic impurity scattering occurs in the vicinity of the Fermi surface, we can assume that $U = U(\theta)$, i.e., that the scattering rate depends only on the angle between the incoming and outgoing momenta. In a similar spirit, we also approximate the density of states by its value at the Fermi energy to obtain

$$G^r(\mathbf{k}, \omega) = \frac{\tilde{\omega}\tau^{(0)} + \epsilon_k\tau^z + \tilde{\Delta}\tau^x}{\tilde{\omega}^2 - \epsilon_k^2 - \tilde{\Delta}^2} \quad (5)$$

where

$$(\tilde{\omega}, \tilde{\Delta}) = (\omega, \Delta) + \left(\frac{u}{2\tau_S}, \frac{-1}{2\tau_D}\right) \frac{1}{\sqrt{1-u^2}}, \quad (6)$$

$u = \tilde{\omega}/\tilde{\Delta}$, and

$$\frac{1}{2\tau_{\sigma\sigma'}} = \frac{n_i\epsilon_F}{2v^2} \int \frac{d\theta}{2\pi} \text{Re}[U_\sigma^*(\theta)U_{\sigma'}(\theta)] \frac{1 + \cos\theta}{2}. \quad (7)$$

We have assumed here that the two layers are identical and set $\tau_{11} = \tau_{22} = \tau_S$ (S = same layer) and $\tau_{12} = \tau_{21} = \tau_D$ (D = different layer).

We now estimate τ_S and τ_D . The measured transport properties of graphene layers strongly suggest that disorder is dominated by Coulomb scatterers [5,9] near the interfaces between the dielectric and the graphene sheets. We therefore expect interlayer disorder potential correlations to play an inessential role and set $\tau_D \rightarrow \infty$ [10]. (Including a finite value of τ_D would not complicate our analysis in any way.) After accounting for the difference between transport and scattering lifetimes $\tau_{\text{tr}}/\tau_S \approx 2$ [12], we follow Ref. [5] in estimating that

$$\epsilon_F\tau_S \approx 10 \frac{n}{n_i}. \quad (8)$$

We use this equation below to express τ_S in terms of n_i .

Coherence criterion.—There is a complete analogy between the Green function (5) and that of an electron in a superconductor with magnetic impurities once the pair-breaking parameter is identified with $\delta = 1/2\tau_S + 1/2\tau_D$. The various ways in which disorder influences coherence in bilayers may therefore be understood by borrowing results from Abrikosov-Gorkov theory [13,14]. In the condensed state, the density of states (DOS) of a disordered bilayer graphene is $\nu(\omega) = \frac{\nu_0}{\zeta} \text{Im}u$ where $\zeta = \delta/\Delta(T)$ and $\nu_0 = k_F/\pi v$ is the DOS of the normal system. Disorder smooths the square root singularity of a clean system. Furthermore, it reduces the energy gap to $\Delta(T)[1 - \zeta^{2/3}]^{3/2}$ for $\zeta < 1$. For $\zeta > 1$, the spectrum is gapless. The mean-field critical temperature T_c in the presence of disorder is given by [13]

$$\log\left(\frac{T_{c0}}{T_c}\right) = \mathcal{H}(\alpha) \quad (9)$$

where $\mathcal{H}(\alpha) = \psi(\frac{1}{2} + \frac{\alpha}{\pi}) - \psi(\frac{1}{2})$ with ψ being the di-Gamma function, $\alpha = \delta/2T_c$, and T_{c0} is the mean-field critical temperature of the clean system. For weak disorder $T_c = T_{c0} - \pi\delta/4$; i.e., the critical temperature decreases linearly with n_i . For strong disorder, $T_c^2 = 6(\delta/\pi)^2 \times \log(\pi T_{c0}/2\delta\Gamma)$ where $\Gamma = 1.76$. When $\delta > 0.8T_{c0}$, the spectrum becomes gapless; however, interlayer phase coherence continues to exist. Eventually, disorder destroys superfluidity altogether when $\delta \geq 0.88T_{c0}$. This last result of Abrikosov-Gorkov theory [13] can be transformed into a more useful form by noting that for graphene bilayers in the strongly interacting regime ($k_B = 1$)

$$T_{c0} \sim \frac{0.1\epsilon_F}{k_F d}. \quad (10)$$

Combining Eq. (8) and (10) yields the left inequality in (1).

It is difficult to estimate T_{c0} in the weakly interacting regime when $k_F d \gg 1$. In that regime screening and other induced interaction effects are expected to significantly reduce the critical temperature. Precise estimates lie beyond the scope of mean-field theory and present an interesting theoretical challenge. Nevertheless, using Eqs. (8) and (9), we can rewrite the condition for the existence of interlayer coherence in terms of the physical measurable parameters:

$$n \geq \left(\frac{vn_i}{10T_c}\right)^2 e^{-2\mathcal{H}(\alpha)}. \quad (11)$$

For weak disorder, $\alpha \ll 1$, condition (11) is equivalent to $n \geq \left(\frac{vn_i}{10T_c}\right)^2 e^{-\pi\alpha}$. In the opposite limit ($\alpha \gg 1$), the system will condense for $n \geq \left(\frac{vn_i}{22\delta}\right)^2 e^{-\pi^2/12\alpha^2}$.

Kosterlitz-Thouless temperature.—In two dimensions, superfluidity is destroyed at the KT temperature by

vortex-antivortex proliferation. To estimate T_{KT} , we use the Kosterlitz-Thouless equation $\rho_s(T_{\text{KT}}) = (2/\pi)T_{\text{KT}}$ with $\rho_s(T)$ in the presence of disorder calculated from Abrikosov-Gorkov theory. The phase stiffness is determined from the counterflow current j_D generated by a uniform gradient of the relative phase between the two layers and from the relation $2\rho_s A = j_D(\Delta) - j_D(\Delta = 0)$. The phase gradient $2A$ is obtained by perturbing the two layers with a pair of constant vector potentials that are equal in magnitude but have opposite signs. The subtraction of $j_D(\Delta = 0)$ in the last relation is required [1] by a pathology of the Dirac model which implies that j_D does not vanish, as required by gauge invariance, in the normal ($\Delta = 0$) state. We find that

$$\begin{aligned} \rho_s(T) &= \rho_s^{(0)} - \frac{v^2}{4} \sum_{\mathbf{k}} \int \frac{d\epsilon}{\pi} n_F(\epsilon) \text{Im Tr}[G^r(\mathbf{k}, \omega)]^2 \\ &= \rho_s^{(0)} \left[1 + \int d\omega \frac{\partial n_F}{\partial \omega} \int d\xi \mathcal{F}(\omega, \xi, \Delta, \zeta) \right], \end{aligned} \quad (12)$$

where $\rho_s(0) = \epsilon_F/4\pi$ is the zero temperature phase stiffness of a clean system [1] and

$$\mathcal{F}(\omega, \xi, \Delta, \zeta) = -\frac{1}{\pi} \text{Im} \int_{-\infty}^{\omega} d\omega' \frac{\tilde{\omega}'^2 + \xi^2 + \tilde{\Delta}^2}{[\tilde{\omega}'^2 - \xi^2 - \tilde{\Delta}^2]^2}. \quad (13)$$

It is essential that the integration over ω' in Eq. (12) precedes the integration over ξ . Vertex corrections to $\rho_s(T)$ vanish when the disorder potential $U = U(\theta)$ is taken to be independent of the magnitudes of the incoming and outgoing momenta.

In the clean limit, \mathcal{F} is a sum of two delta functions centered at $\omega = \pm\sqrt{\xi^2 + \Delta^2}$. The phase stiffness is then

$$\rho_s^{(c)}(T) = \rho_s^{(0)} + \frac{v^2}{4} \int d\omega \frac{\partial n_F}{\partial \omega} \nu(\omega). \quad (14)$$

The second term in (14) captures the reduction in ρ_s at finite temperature due to the thermal excitation of quasiparticles. It can be understood as follows. A uniform phase gradient (induced here by a constant vector potential) generates a counterflow current in the superfluid. One contribution to the current $\sum_{\mathbf{k}} \mathbf{v} n_F(\epsilon_{\mathbf{k}} + \mathbf{v}\mathbf{A})$ originates from the change in the occupation of the quasiparticles due to the change of the energy dispersion. To linear order in \mathbf{A} the last expression becomes $\frac{v^2}{4} \sum_{\mathbf{k}} \partial_{\epsilon_{\mathbf{k}}} n_F 2A$ from which the second term in expression (14) readily follows. A similar expression holds for the phase stiffness of a BCS superconductor, but with two differences. First, the velocity of a particle in a parabolic band is energy dependent and must be left under the integral. Second, the zero temperature phase stiffness naturally arises in a parabolic band due to the change in the velocity to $\mathbf{v} + \mathbf{A}/m$ with m being the effective mass of the particle whereas in the graphene

spontaneous-coherence case, it is accumulated at the Dirac-model's cutoff wave vector, and conveniently captured [1] by subtracting $j_D(\Delta = 0)$.

The influence of disorder on $\rho_s(T)$ follows mainly from the finite lifetime of definite-momentum quasiparticles. The impurities reduce the energy gap, thus reducing the energy gain due to pair condensation and correspondingly its sensitivity to phase gradients. The reduction of the KT temperature due to disorder is readily obtained from Eq. (12) by setting the temperature to T_{KT} and using the relation $\rho_s(T_{\text{KT}})/T_{\text{KT}} = 2/\pi$. The pairing potential $\Delta(T_{\text{KT}})$ must be calculated self consistently using Eqs. (4) and (6).

Figure 1 shows the reduction of the KT critical temperature of counterflow superfluids due to disorder. In this figure, we plot the dependence of T_{KT} on the Fermi energy for various strengths of disorder. All energies are scaled with T_{c0} . The strength of disorder is parameterized by $1/2\tau_S T_{c0} = \delta/T_{c0}$ ($\sim n_i k_F d/n$ in the strongly interacting limit). In the weakly interacting regime, $\epsilon_F/T_{c0} \gg 1$ and the KT temperature is of order of T_c . The effect of disorder on T_{KT} can then be inferred from its effect on T_c . In that regime, $\Delta(T_{\text{KT}})$ is small; hence, the disorder will significantly reduce T_{KT} . As the interaction increases, so does T_{c0} ; however, the ratio T_{KT}/T_{c0} decreases; hence, $\Delta(T_{\text{KT}})$ is relatively large. Thus, in the strongly interacting regime the effect of disorder on the KT temperature is relatively weak.

Discussion.—Since the pioneering work of Abrikosov, Gorkov, and Anderson, it has been understood that both the critical temperature and the order parameter of s -wave superconductors are unaffected by a sufficiently low concentration of nonmagnetic impurities [15]. This result, known as Anderson's theorem [16], asserts that even in the presence of (nonmagnetic) impurities, two states related to one another by time reversal symmetry will pair

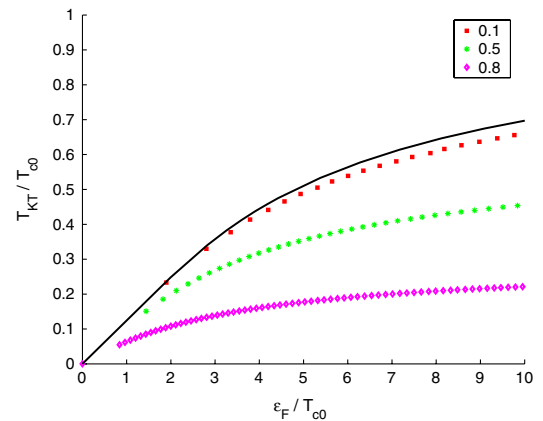


FIG. 1 (color online). KT temperature vs disorder strength. Disorder is parameterized by $\alpha = 1/2\tau_S T_{c0} = \delta/T_{c0}$. The solid line in this figure corresponds to the clean system and the broken lines correspond to (top to bottom) $\alpha = 0.1, 0.5, 0.8$. In graphene bilayers, $\epsilon_F/T_{c0} \sim 10k_F d$ in the strongly interacting regime.

and condense. On the other hand, as experimentally observed [17] and theoretically explained by Abrikosov-Gorkov theory, the values of T_c and Δ are suppressed by magnetic impurities that act to reduce the binding energy of Cooper pairs. Later work demonstrated that Abrikosov-Gorkov theory applies to other circumstances in which some perturbation breaks time reversal symmetry, for example, thin superconducting films subjected to a parallel magnetic field [18,19] or superconductors with an exchange field in the presence of strong spin-orbit interactions [20]. In this work, we extended the range of applicability of Abrikosov-Gorkov theory to dirty graphene bilayers [21].

The analogy between the effects of disorder on superconductivity in metals and on counterflow superfluidity in bilayer systems is evident if time reversal symmetry in the former is replaced by particle-hole symmetry in the latter. Kramer's degeneracy in a nonmagnetic superconductor translates to particle-hole symmetry in the spectrum of the bilayer system. Indeed, if we consider a fictitious scenario in which the disorder potentials of the two layers are perfectly anticorrelated, i.e., $U_1(\mathbf{r}) = -U_2(\mathbf{r})$, then particle-hole symmetry is preserved, $\tau_s = -\tau_D$, the pair-breaking parameter α vanishes, and both T_c and Δ retain their clean system values. Generic disorder potentials break particle-hole symmetry and therefore provide a particle-hole pair-breaking mechanism. The influence of disorder on the critical temperature is determined by the value of $\alpha = \delta/2T_c$. Disorder will have a negligible effect on T_c as long as $\alpha \ll 1$ reinforcing the plausibility of condensation in the strongly interacting regime.

In this work, we neglected the fully occupied valence band and empty conduction band. Accounting for these energy bands will increase interlayer coherence and diminish the influence of disorder on the critical temperatures. However, we expect these bands to have significant effect only in the regime of very strong interactions when $k_F d \ll 1$.

Ideas similar to the ones used here may be applied to study disorder effects on exciton condensation in quantum Hall bilayers. In current experiments [22], $1/2\tau_s T_c$ is of order of unity suggesting that incorporating disorder is imperative for a correct estimate of the mean-field critical temperature and of T_{KT} . It is also interesting to consider the influence of magnetic impurities on a bilayer graphene system. Because of the spin and valley degeneracy of the graphene sheets, the exciton pairing is SU(4) symmetric. Put differently, four identical superfluids coexist in the system. A scattering event by a magnetic impurity will thus scatter an exciton from one superfluid to another.

In summary, interlayer coherence is most likely to occur in the strongly interacting regime $k_F d < 1$. In this work, we showed that disorder destroys pair condensation unless $k_F d > \pi n_i d^2$. Furthermore, we numerically found the reduction in T_{KT} due to disorder.

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