High Temperature Superfluidity in Double Bilayer Graphene

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Exciton bound states in solids between electrons and holes are predicted to form a superfluid at high temperatures. We show that by employing atomically thin crystals such as a pair of adjacent bilayer graphene sheets, equilibrium superfluidity of electron-hole pairs should be achievable for the first time. The transition temperatures are well above liquid helium temperatures. Because the sample parameters needed for the device have already been attained in similar graphene devices, our work suggests a new route towards realizing high-temperature superfluidity in existing quality graphene samples.

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It is proving a challenging task to observe superfluidity in a semiconductor electron-hole double quantum well system. Despite long-standing theoretical predictions[1, 2] and significant experimental efforts[3–5], it is only very recently that superfluid behavior has been demonstrated with a hexagonal boron nitride (hBN) dielectric.[11] This makes it difficult to access the region of strong interactions. Also, bound excitons do not form because of the massless carriers.[12] The ratio \( r_s = (V_{\text{Coul}})/E_F \) is a useful measure of the importance of interactions relative to kinetic energy. \( V_{\text{Coul}} = (e^2/\kappa)\sqrt{\pi n} \) is the average Coulomb interaction energy, where \( n \) is the charge carrier density and \( \kappa = \pi n \). The Fermi velocity \( v_F \sim 10^6 \text{ m s}^{-1} \) is independent of \( n \) and the Fermi momentum for the spin-up carriers in 2D is \( k_F = \sqrt{2\pi n/g_s} \), where \( g_s = 2 \) is the pseudospin factor. Fermi velocity is then \( E_F = h v_F \sqrt{2\pi n/g_s} \). This makes \( r_s = e^2/(\kappa h v_F) \lesssim 1 \) for monolayer graphene, a constant of order unity, while recent theory suggests that an electron-hole superfluid can only occur at measurable temperatures for \( r_s > 2.3 \) in the 2xMLG system.[13] Very recent experiments have shown no evidence of superfluidity in this system despite achieving barrier thicknesses as low as \( D_B = 1 \text{ nm} \).[11] This poses an exciting challenge: can new experimentally realistic structures be designed using atomically thin crystals that allow the transition to a superfluid state?

We concentrate here on bilayer graphene since it has been well characterized and exhibits extremely low levels of disorder, but a number of other such crystals are possible.[14] Our proposed system (2xBLG) consists of a pair of parallel bilayer graphene sheets (Fig.1). The lower bilayer sheet is an electron bilayer comprising two parallel, \( A-B \) stacked, closely coupled electron layers of graphene with layer separation \( D_e = 0.37 \text{ nm} \).[15] There is strong electron hopping between the two layers. The upper bilayer sheet is a hole bilayer consisting of two \( A-B \) stacked hole layers, but is otherwise analogous to the lower bilayer sheet with an identical layer separation \( D_h = D_e \). The two bilayer sheets are separated by a hBN insulating barrier of width \( D_B \) to prevent tunneling between the sheets. There are separate electrical contacts to the two layers and a bias \( V_{BB} \) can be applied between them. The bias \( V_{BB} \) and biases \( V_{TG} \) and \( V_{BG} \) on top and bottom metal gates allow independent control over the carrier density.
in each layer and can adjust the symmetry of the electric field across the two sheets. By tuning the three biases, a wide range of electron and hole densities can be achieved.

Bilayer graphene eliminates the problems caused by the linear dispersion of monolayer graphene since over a wide range of electron or hole densities, $1 \times 10^{11} < n < 4 \times 10^{12}$ cm$^{-2}$, symmetrically biased graphene bilayers behave as a zero gap semiconductor with a quadratic dispersion around the Fermi level $E_\pm(k) \approx \pm \hbar^2 k^2/2m^*$. The effective mass is $m^* \approx (0.03$ to $0.05)m_e$, depending on the carrier density. At very high densities $n > 4 \times 10^{12}$ cm$^{-2}$, $E_\pm(k)$ crosses over to linear behavior, but this density region lies in the weakly interacting regime and is not of interest. At very low densities $n \lesssim n_{\text{min}} = 1 \times 10^{11}$ cm$^{-2}$ a trigonal warping of the bands transforms the two quadratic bands into three sets of Dirac-like linear bands. This warping can be reduced if necessary by applying an asymmetric bias across each of the two bilayers which will also open up a gap separating the conduction and valence bands.

With quadratic energy dispersion, the Fermi energy in the bilayer sheet depends linearly on density, $E_F = \pi \hbar^2 n/g_e m^*$, so the parameter $r_s$ now has a density dependence, $r_s = (e^2/\kappa)(g_e m^*/\hbar^2)(1/\sqrt{n})$. To experimentally reach large $r_s$ and the strongly interacting regime in the bilayer sheet, it is enough to decrease the carrier density $n$ with suitable gate voltages. In graphene sheets the lowest density is restricted by onset of electron puddle formation, which occurs at densities below $10^{11}$ cm$^{-2}$ in high quality graphene bilayers. This corresponds to $r_s = 9$ in the strongly interacting regime.

We know that $D_{\text{eh}} \ll a_0^*$ ensures strong electron-hole pairing. For a 2xBLG system embedded in a hBN dielectric with $\kappa = 3$, setting $m^* = m_e^* = m^*_h = 0.04m_e$ and using the reduced mass yields an effective Bohr radius $a_0^* = 8$ nm. This is large compared with the thickness of barriers already fabricated, $D_B \approx 1$ nm $\approx D_{\text{eh}}$. Table I summarizes key physical parameters for three systems in which superfluidity of spatially separated electron-hole pairs has been predicted. For GaAs DQW, although the $E(k)$ is quadratic, the following experimental restrictions make it difficult to access the region with strong pairing effects. (a) The barriers are wide, $D_{\text{eh}} \gtrsim 2a_0^*$, (b) the largest value of $r_s$ attained is $r_s = 2$, and (c) the effective Rydberg ($Ry^*$) binding energy is small. For 2xMLG, the barriers can be very thin, $D_{\text{eh}} \ll a_0^*$, but the linear $E_\pm(k)$ keeps the system in the weakly interacting regime with $r_s < 1$. What decisively favors the 2xBLG system is that the strong pairing regime is accessible in current samples because: (a) the extremely thin barriers $D_B \gtrsim 1$ nm and thin bilayer sheets $D_e = 0.37$ nm, (b) the ability to tune $r_s$ to large values in order to access strong pairing, (c) the almost perfectly matched electron and hole bands resulting from their near equal effective masses, leading to almost perfect particle hole symmetry and nesting between circular Fermi surfaces, and (d) the larger $Ry^*$ than for GaAs.

We now calculate the superfluid energy gap and transition temperature to see whether a superfluid state can form in the 2xBLG system with realistic sample parameters and at experimental attainable temperatures. For simplicity we restrict $D_B > D_e$ and approximate the system by a single layer of electrons $\ell = e$ interacting with a single layer of holes $\ell = h$. The quadratic energy bands are $E_\ell(k) = \gamma e k^2/(2m^*_e) - \mu^\ell$, where the band index $\gamma = 1$ and $-1$ for the upper and lower bands and $\mu^\ell$ is the chemical potential. We make a standard transformation to positively charged particles for the holes. The effective Hamiltonian is

$$H = \sum_{\ell\gamma k} c^\ell\gamma_{k} c^{\ell\gamma\dagger}_{k} + \sum_{qk\gamma k'} V^{\ell\dagger}_{k-k'} c^{\ell\gamma\dagger}_{k+q} c^{\ell\gamma}_{k'+q} + \frac{1}{2} \sum_{qk\gamma} \gamma e q^2 k^2 + \frac{1}{2} \sum_{qk\gamma} \gamma' e q^2 k'^2$$

$c^\ell\gamma_{k} \dagger$ and $c^{\ell\gamma}_{k}$ are creation and destruction operators for charge carriers in layer $\ell$ and band $\gamma$. Spin indices are implicit. $V^{\ell\dagger}_{k-k'}$ is the static screened electron-hole interaction.

A mean-field description is applicable on both the weak-coupling and strong-coupling sides of the BCS-BEC crossover for conventional pairing systems. The mean-field equations at temperature $T$ for $\mu^\ell$ and the momentum-
We calculated the static screened effective mass.

\[ \Delta_k^\gamma(T) = -\sum_{k'\gamma'} \frac{1}{2} V_{k-k'} \frac{\Delta_{k'}}{2\varepsilon_{k'}} [1 - f(E_{k'}^{\gamma^+}) - f(E_{k'}^{\gamma^-})] \]

\[ n_j(T) = 2 g_{\gamma} \sum_{k\gamma} [(\dot{u}_k^j)^2 f(E_k^{\gamma^\pm}) + (v_k^j)^2 (1 - f(E_k^{\gamma^\pm}))] \]

\[ E_k^{\gamma^\pm} = E_F^\gamma \pm \frac{1}{2} (\varepsilon_k^e - \varepsilon_k^h), \quad F_\gamma = \sqrt{\frac{1}{2} (\varepsilon_k^e + \varepsilon_k^h)^2 + \Delta_k^{\gamma^2}} \]

\[ V_{q}^{eh}(T) = \frac{v_q e^{-qD_{eh}}}{1 - 2v_q \Pi(0,q,T) + v_q^2 \Pi(0,q,T)}[1 - e^{-2qD_{eh}}] \]

The nature of the superfluidity can be understood by looking at the dependence of the gap \( \Delta_k \) on \( n \). In the weak-coupled BCS limit of Cooper pairs there would be a peak in \( \Delta_k \) centered at \( k = k_F \), while in the BEC region the peak in \( \Delta_k \) would be centered at \( k = 0 \) with a long tail falling off as \( 1/k \), indicating pairs localized in real space. Figure 2(a) shows \( \Delta_k \) at different densities \( D_{eh} \) at \( T = 0 \). Figure 2(b) shows \( \Delta_k \) at different densities \( D_{eh} \) at \( T = 0 \). The electron and hole densities are set equal \( n_e = n_h = n \) only, and this cannot be achieved in the 2xMLG system. We neglect the small contributions from the negative branches of the bands.

Figure 2(a) shows for each \( D_{eh} \) there is a critical density \( n_c \) above which the gap \( \Delta_{min} \) is, at most, in the sub-mK energy range. In realistic disordered systems it is unlikely there would be pairing in this case. At \( n = n_c \), there is a sudden discontinuous jump in \( \Delta_{max} \) to much higher energies. Reference [13] reported a similar effect for the 2xMLG system but for \( r_s > 2.35 \) only, and this cannot be achieved in the 2xMLG system. As \( D_{eh} \) decreases, the pairing interactions become stronger and the superfluidity persists up to a higher \( n_c \).

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form but the gap collapses before that happens so the superfluid does not reach the BCS limit before the effectiveness of the screening in Eq. 6 discontinuously jumps and superfluidity is suppressed.

The very large gaps shown in Fig. 2(a), in some cases \( \Delta_{\text{max}} > 500 \) K, are particularly interesting as, in contrast with high-\( T_c \) superconductors, these values of \( \Delta_{\text{max}} \) are maintained across a wide density range. For reference, at \( n = 5 \times 10^{11} \) cm\(^{-2} \) the Fermi temperature \( T_F = 175 \) K.

To determine the superfluid transition temperatures \( T_c \), we first recall that for superfluids in 2D the mean-field critical temperature \( T_{c,\text{MF}} \), the temperature at which the mean-field superfluid gap goes to zero, can overestimate the true \( T_c \). The superfluid transition in 2D systems has a topological Kosterlitz-Thouless (KT) character[23], and \( T_c \) is determined by the KT temperature,

\[
T_c = T_{KT} = \left( \frac{\pi}{2} \right) \rho_s(T_{KT}).
\]

\( \rho_s(T) \) is the superfluid stiffness. Mean-field theory gives a good estimate of \( \rho_s(T) \) for quadratic bands in both the BCS and BEC limits. \[6 \] \( \rho_s(0) = E_F / \pi \tau \) at \( T = 0 \), and \( \rho_s(T) \) falls off slowly up to \( T \sim \Delta_{\text{max}} \) if \( k_F D_{\text{eh}} \) is small, generally the case for our parameters. For \( n < n_c \) we can take \( \rho_s(T) \simeq \rho_s(0) \), Eq. 5 then giving \( T_{KT} = E_F / \pi \). This is because even at a density as high as \( n = 6 \times 10^{11} \) cm\(^{-2} \) \( E_F / \pi \sim 26 \) K, still much less than the \( \Delta_{\text{max}} \) shown in Fig. 2(a). For \( n > n_c \), \( \Delta_{\text{max}} \) is extremely small. Since \( \rho_s(T) \) collapses to zero as \( T \) becomes larger than \( \Delta_{\text{max}} \), \( T_c = T_{KT} \) is similarly small for \( n > n_c \).

Figure 5 shows the \( T-n \) phase diagram. For densities \( n \) greater than a critical density \( n_c \), the system is a Fermi liquid at all practicable non-zero temperatures. For \( n < n_c \) and \( T \) below the transition temperature \( T_c \), the system is a superfluid. \( T_c = T_{KT} \) grows linearly with density so the maximum transition temperature \( T_{c,\text{max}} = E_F / (\pi n_c) / 8 \). For \( n < n_c \) and \( T > T_c \) there will still be strong signatures of the underlying superfluid state through the pseudogap. The pseudogap is a normal state precursor of the superconducting gap due to local dynamic pairing correlations and is produced by non-coherent fluctuations of the pairing field. \[24 \] The deviations from Fermi liquid behavior due to the pseudogap can persist up to room temperature, \( T \sim \Delta_{\text{max}} \). \[25 \]

Table II gives values of \( T_{c,\text{max}} \) and \( n_c \) which increase with decreasing \( D_{\text{eh}} \) because of the stronger electron-hole coupling. Using hBN barriers between the gates and between the graphene sheets (System (A)) gives \( \kappa = 3 \), and \( T_{c,\text{max}} \) is well above liquid helium temperatures. If the two graphene bilayer sheets are separated by a hBN dielectric barrier and suspended in air between upper and lower gates (System (B)), the effective \( \kappa = 1.5 \) and \( T_{c,\text{max}} \) is greatly increased. Devices similar to System (B) have recently been fabricated with a single bilayer graphene sheet suspended between two gates. \[27 \]

We now discuss the experimental consequences of our phase diagram. Even at the lowest density \( n_{min} = 10^{11} \) cm\(^{-2} \), the transition temperatures in systems (A) and (B) remain above liquid helium temperatures. There will be strong experimental signatures of the superfluid below \( T_c \). Although the pairs are neutral, the ability to make separate electrical contacts to the electron and hole layers allows for spectacular electrical effects. \[28 \] Coulomb drag and inter-layer tunneling measurements will show significant enhancements as \( T \) is decreased below \( T_c \) while counterflow measurements can directly probe the superflow. \[28 \] Even up to room temperature there will be strong signatures of the pseudogap in phenomena such as compressibility, specific heat capacity and spin susceptibility. The wide density range over which superfluidity can be observed in a single device is in marked contrast with high-\( T_c \) superconductors, where superconductivity occurs only in a narrow 30% band of doping centered at optimal doping.

The 2xBLG system is the first multiband system with just one condensate. It is an opposite case to multiband superconductors such as magnesium-diboride where pairing is only within the bands and multiple coupled condensates appear, one condensate and one gap for each band. \[30 \] In contrast, in the 2xBLG system, pairing is only possible between the different hole and electron bands, leading to a single condensate and a single gap.

The combination of extremely thin barriers, large \( r_s \), near-equivalent masses, and strong pairing attraction makes the 2xBLG system ideal for observing high-\( T_c \) superfluidity. In existing quality samples it should be possible to study the phase transition from Fermi-liquid to superfluid as well as the

![FIG. 3: Density n – temperature T phase diagram (see text).](image318x629 to 561x740)

### Table II: Maximum transition temperatures \( T_{c,\text{max}} \) and critical densities \( n_c \) for barrier thicknesses \( D_{\text{eh}} \). System (A) is the two graphene bilayer sheets embedded in a hBN dielectric. System (B) is for the two graphene bilayer sheets separated by a hBN barrier and suspended in air between the gates.

| \( D_{\text{eh}} \) (nm) | System (A) | System (B) |
|----------------|------------|------------|
| 0.5 | \( T_{c,\text{max}} \) (K) | \( n_c \) (\( 10^{11} \) cm\(^{-2} \)) | \( T_{c,\text{max}} \) (K) | \( n_c \) (\( 10^{11} \) cm\(^{-2} \)) |
| 1 | 27 | 6.2 | 72 | 17 |
| 2 | 14 | 3.2 | 34 | 7.9 |
pseudogap physics. Our approach would also apply more generally to other small gap semiconductors that can be made into atomically thin flakes and used with dielectrics such as hBN.

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FIG. 4: $\Pi_0^{(n)}(q)$ and $\Pi_0^{(a)}(q)$ with two normal and two anomalous Green functions, respectively.

In RPA the polarization bubbles are constructed with the normal and anomalous Green’s functions of BCS theory. Assuming the Green’s functions are diagonal in the band indices, the normal and anomalous Green’s functions are given by,

$$G_{\gamma\gamma}(k, i\epsilon_n) = \frac{(u_{k\gamma})^2}{i\epsilon_n - E_k} + \frac{(v_{k\gamma})^2}{i\epsilon_n + E_k},$$

$$F_{\gamma\gamma}(k, i\epsilon_n) = \frac{u_{k\gamma}v_{k\gamma}^*}{i\epsilon_n - E_k} - \frac{u_{k\gamma}v_{k\gamma}^*}{i\epsilon_n + E_k},$$

respectively, where $u_{k\gamma}$ and $v_{k\gamma}$ are the Bogoliubov factors

$$(u_{k\gamma})^2 = \frac{1}{2} \left( 1 + \frac{\xi_{k\gamma}}{E_k} \right),$$

$$(v_{k\gamma})^2 = \frac{1}{2} \left( 1 - \frac{\xi_{k\gamma}}{E_k} \right).$$

The summation of the fermionic Matsubara frequencies $\epsilon_n$ of the particle-hole loop can then be done analytically, and taking the static limit the expressions for the bubbles are given by,

$$\Pi_0^{(n)}(q, T) = -2g_v \sum_{k, \gamma\gamma'} \frac{1}{2} \frac{(u_{k\gamma})^2(v_{k-q, \gamma'}^*)^2 + (v_{k\gamma})^2(u_{k-q, \gamma'}^*)^2}{E_k + E_{k-q}}.$$ (11)

$$\Pi_0^{(a)}(q, T) = 2g_v \sum_{k, \gamma\gamma'} \frac{1}{2} \frac{2u_{k\gamma}v_{k\gamma}u_{k-q, \gamma'}^*v_{k-q, \gamma'}^*}{E_k + E_{k-q}}.$$ (12)

When the chemical potential $\mu$ is positive, the contributions of the lower band ($\gamma$ or $\gamma' = -1$) to the total polarization bubbles are small because the corresponding $k$-states are filled and far in energy from $\mu$. Hence they contribute little to the particle-hole processes. We considered only the $\gamma = \gamma' = 1$ contribution and, as in the gap equation, only the s-wave harmonic of the geometrical form factor of graphene $F_{\gamma\gamma'}^{\gamma^\prime}$.

Physically, the effect of screening is suppressed because of the opening of the energy gap $\Delta$ at the Fermi surface. This exponentially suppresses particle-hole processes with energies less than the gap energy $\Delta$ and it is precisely these low-energy processes that are needed to screen the long-range Coulomb interaction. Once a gap appears in the excitation spectrum, screening exactly vanishes in the $q = 0$ limit. From a diagrammatic point of view, in addition to a suppression of the bubble with diagonal Green functions at low momentum, there is an additional canceling contribution from the anomalous bubble with off-diagonal Green functions. At $q = 0$ the cancelation is exact for any non-zero $\Delta$. 