Excitonic condensation of massless fermions in graphene bilayers

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Graphene, a single sheet of graphite with honeycomb lattice structure, has massless carriers with tunable density and polarity. We investigate the ground-state phase diagram of two graphene sheets (embedded in a dielectric), separated by distance $d$ where the top layer has electrons and the bottom layer has holes, using mean-field theory. We find that a uniform excitonic condensate occurs over a large range of carrier densities and is weakly dependent on the relative orientation of the two sheets. We obtain the excitonic gap, quasiparticle energy, and the density of states. We show that both the condensate phase stiffness and the mass of the excitons with massless particles as constituents, vary as the square root of the carrier density, and predict that the condensate will not undergo Wigner crystallization.

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I. INTRODUCTION

Over the past three years, graphene has emerged as the unique candidate that provides a realization of two-dimensional massless fermions whose carrier density and polarity are tunable by an external gate voltage. Subsequent experimental and theoretical investigations have led to a thorough re-examination of some of the properties of linearly dispersing massless fermions. The truly two-dimensional (2D) nature of graphene permits control and observation of local carrier density and properties. In graphene bilayers, the ability to change the carrier polarity of an individual layer implies that the interlayer Coulomb interaction can be tuned from repulsive to attractive. This raises the possibility of formation of electron-hole bound states or indirect excitons, albeit with massless fermions as its constituents. Properties of such bound states of massless particles are an open question; the only other example, to our knowledge, is the proposed color superconductivity in dense quark matter.

Graphene bilayers provide an ideal and unique candidate for straightforward experimental investigations of such phenomena. A uniform Bose-Einstein condensate of excitons in electron-hole bilayers occurs when the interlayer distance is comparable to the distance between the particles within each layer. These excitons have mass $m_e = m_e + m_h$, where $m_e$ ($m_h$) is the band mass of the electron (hole). At high densities, dipolar repulsion between the excitons can lead to a condensate ground state with broken translational symmetry: a supersolid. Biased bilayer quantum Hall systems near total filling factor $\nu_f = 1$ have shown uniform excitonic condensation. In this case, the exciton mass is determined solely by interlayer Coulomb interaction and is independent of the bias voltage. These observations raise the questions: What is the mass of an exciton with massless constituents? Will such an excitonic condensate lead to a supersolid if the dipolar repulsion between such excitons (with a nonzero mass) is increased?

In this Brief Report, we investigate the excitonic condensation in two graphene sheets embedded in a dielectric and separated by a distance $d\gg a$ ($a = 1.4$ Å is the honeycomb lattice size) so that the tunneling between the layers is negligible, but interlayer Coulomb interaction is not. The layers have opposite polarity and equal density of carriers $n_{2D}$. We remind the reader that in graphene, in the continuum limit, the length scale $1/k_F$ and the energy scale $E_F$ are both set by the density of carriers $n_{2D}$ ($k_F = \sqrt{m_{2D}}$ is the Fermi momentum, $E_F = \hbar v_F k_F$ is the Fermi energy, and $v_F \sim c/300$ is the speed of massless carriers). Therefore, the ground-state phase diagram depends only on one dimensionless parameter $k_Fd$. This is markedly different from conventional bilayer systems parameterized by $(d/aB, r_e)$, where $a_B$ is the band Bohr radius and $r_e = 1/\sqrt{4\pi a_B^2 \hbar^2}$ as well as biased bilayer quantum Hall systems, parameterized by $(d/l_B, \Delta \nu)$, where $l_B$ is the magnetic length and $\Delta \nu$ is the filling factor imbalance.

We use the mean-field theory to obtain the ground-state phase diagram as a function of $k_Fd$. We find that (a) excitonic condensation occurs at all densities as long as $k_Fd \gg 1$, (b) the condensate properties are weakly sensitive to the relative orientation of the two sheets (stacking), (c) the superfluid phase stiffness $\rho_s$ and the exciton mass have a $\sqrt{n_{2D}}$ dependence, and (d) the excitonic condensate does not undergo Wigner crystallization in spite of dipolar repulsion between excitons with a nonzero mass.

The plan of this Brief Report is as follows: In Sec. II, we present the mean-field Hamiltonian and briefly sketch the outline of our calculations. In Sec. III, we show the results for the excitonic gap $\Delta_k$, the quasiparticle energy $E_k$, and the quasiparticle density of states $D(E)$. We discuss the density dependence of the superfluid stiffness $\rho_s$ and the mass of the excitons. In Sec. IV, we show that these results are equivalent to the absence of Wigner crystallization and mention the implications of our results to experiments.

II. MEAN-FIELD MODEL

We consider two graphene sheets embedded in a dielectric and separated by distance $d$ with chemical potentials in the two layers that are adjusted so that the top layer (denoted by pseudospin $\tau = +1$) has electrons and the bottom layer (denoted by pseudospin $\tau = -1$) has holes with the same density. We consider two stackings: the Bernal stacking that occurs naturally in graphite and the hexagonal stacking in which
each sublattice (A and B) in one layer is on top of the corresponding sublattice in the other layer. Since the Hamiltonian in the continuum description is SU(4) symmetric in the spin and valley indices, we ignore those indices for simplicity. In the continuum limit, the single-particle Hamiltonian for carriers in layer $\tau$ is:

$$\hat{h}_{0} = \Sigma_{\alpha^{\prime}}(\alpha \hbar \omega_{k} g) \hat{c}_{k\alpha \tau}^{\dagger} \hat{c}_{k\alpha \tau},$$  \hspace{1cm} (1)$$

where $k$ is the momentum measured from the K point and $\alpha = \pm$ denote the conduction and valence bands that result from diagonalizing the Hamiltonian in the sublattice basis. $c_{k\alpha \tau}$ is creation (annihilation) operator for an electron in band $\alpha$ in layer $\tau$ with momentum $k$. We point out that for the hexagonal stacking, $c_{k\alpha \tau}$ is independent of the layer index $\tau$. For the Bernal stacking, the creation operators in the two layers are related by complex conjugation, $c_{k\alpha \tau}^{\dagger} = \frac{1}{\sqrt{2}} c_{k\alpha \tau}^{\dagger} + \frac{1}{\sqrt{2}} c_{k\alpha \tau}^{\dagger}$. The interaction Hamiltonian consists of intralayer Coulomb repulsion $V_{I}(q) = 2 \pi\epsilon^{2}/\epsilon_{q}$ and interlayer Coulomb attraction $V_{I}(q) = -V_{q}(q) \exp(-qd)$, where $q$ is the dielectric constant. Using standard mean-field techniques, we obtain the following mean-field Hamiltonian:

$$\hat{H} = \sum_{k} \left[ e_{k}^{A} h_{-k}^{A} + e_{k}^{B} h_{-k}^{B} \right]$$

Here, $e_{k}^{A} = e_{k} + \mu$ creates an electron in the conduction band ($\alpha = +$) in the top layer $(\tau = +1)$ and $h_{k}^{A} = e_{k} + \mu$ creates a hole in the valance band $\alpha = -$ in the bottom layer $(\tau = -1)$. The term $e_{k}^{A}$ contains single-particle energy, capacitive Hartree self-energy and the intralayer exchange self-energy. The off-diagonal term $\Delta_{k}$ is proportional to the excitonic condensate order parameter $\langle \hat{n}_{-k} \rangle$. The eigenvalues of the mean-field Hamiltonian are given by $\pm \Delta_{k} = \pm \sqrt{(e_{k}^{A} - \mu)^{2} + \Delta_{k}^{2}}$. We consider mean-field states with a real $\Delta_{k}$ and spatially uniform density. It is straightforward to diagonalize the Hamiltonian and obtain the mean-field equations:

$$e_{k} = \hbar u_{G} k + \frac{e^{2} n_{2D}}{C} - \frac{1}{2} \int_{k'} V_{A}(k - k') \left[ 1 - \frac{e_{k'}^{B}}{E_{k'}} \right].$$  \hspace{1cm} (3)$$

$$\Delta_{k} = -\frac{1}{2} \int_{k'} V_{e}(k - k') / (\theta_{k,k'} \Delta_{k}^{B} / E_{k'}).$$  \hspace{1cm} (4)$$

where $e_{k}^{B} = e_{k} - \mu$, $C = e^{2}/2 \pi d$ is the capacitance per unit area, and $\theta_{k,k'} = \theta_{k} - \theta_{k'}$. The form factor for the two stackings is:

$$f(\theta_{k,k'}) = \begin{cases} 1 + \cos \theta_{k,k'} & \text{Hexagonal} \\ \cos \theta_{k,k'} / (1 + \cos \theta_{k,k'}) & \text{Bernal} \end{cases}.$$  \hspace{1cm} (5)$$

We point out that the self-energy in Eq. (3) takes into account both intrinsic and extrinsic contributions that cancel the cos $\theta_{k,k'}$ dependent terms in the form factor and make the results independent of the ultraviolet cutoff. Therefore, the intralayer self-energy in Eq. (3) is the same as that for a conventional system. The chemical potential $\mu$ is determined by the carrier density that takes into account the four-fold spin and valley degeneracy.

It is straightforward to derive similar equations for a conventional electron-hole system. They are obtained by changing the single-particle dispersion to a quadratic and replacing the form factor $f(\theta_{k,k'})$ by a constant $f = 2$. We solve Eqs. (3), (4), and (6) iteratively to obtain self-consistent results.

### III. RESULTS

Figure 1 shows the excitonic gap $\Delta_{k}$ and the quasiparticle energy $E_{k}$ for the Bernal (green dashed) and the hexagonal (red solid) stacking. The excitonic gap $\Delta_{k}$ is maximum at the Fermi momentum $k_{F}$ where the quasiparticle energy $E_{k}$ is minimum. Since the electron-hole Coulomb interaction is always attractive, the excitonic condensate order parameter is nonzero down to the bottom of the Fermi sea $\Delta_{k=0} \neq 0$. Our results predict that the hexagonal-stacked system will have a larger excitonic gap than the Bernal-stacked system. The quasiparticle energy $E_{k}$ becomes linear at large $k > k_{F}$, since the constituent particles of the exciton have a linear dispersion. The speed of these quasiparticles is increased due to intralayer exchange self-energy, although the increase is modest, $\sim 10\%$. Corresponding results for a conventional electron-hole system (blue dotted) are also shown in Fig. 1.

The chemical potential $\mu$ is determined by the carrier density that takes into account the four-fold spin and valley degeneracy.

With typical graphene carrier densities $n_{2D} \sim 10^{12}/\text{cm}^{2}$ and $d \sim 100 \text{ Å}$ or $k_{F}d \sim 1$, the excitonic gap is appreciable, $\Delta_{m} \sim 30 \text{ meV}$. 

![FIG. 1. (Color online) The excitonic gap $\Delta_{h}$ and the quasiparticle energy $E_{k}$ in graphene bilayer for Bernal (green/light gray dashed) and hexagonal (red-gray solid) stacking with $k_{F}=1$. The quasiparticle spectrum $E_{k}$ becomes linear with a renormalized velocity $v_{G}>v_{F}$ for large $k > k_{F}$. The dotted blue curves show corresponding results for an electron-hole system at $r_{s}=2.7$ and $k_{F}=1$ when plotted using relevant (unit) area for energy.](image-url)
there are no states for condensate phase, for intermediate energies corresponding result for a conventional system at $r_s$=2.7 is shown in dotted blue/dark gray.

A direct probe of the excitonic gap is the quasiparticle density of states, for graphene with no interactions, the density of states is linear, $D_0(E)=2E/\pi\hbar^2v^2_G$. In the excitonic condensate phase, for intermediate energies $\Delta_m=0\leq E\leq E_{k_F0}$ there are two rings in the phase space consistent with that energy: one with $k<k_F$ and the other with $k>k_F$. Therefore, the quasiparticle density of states is given by $D(E)=D_s(E)+D_e(E)$, where $D_s$ ($D_e$) denotes the density of states from respective rings. Figure 3 shows $D_s(E)$ (green dashed) and $D_e(E)$ (red solid); they are both zero for $E<\Delta_m$ and diverge at $\Delta_m$ as is expected. Note that $D_s(E)=0$ for $E>E_{k_F0}$ since there are no states for $k<k_F$ with energies higher than $E_{k_F0}$. The asymmetry in $D_s$ and $D_e$ for $E>\Delta_m$ is due to the linear dispersion of carriers and the nonzero electron-hole pairing that extends to the bottom of the Fermi sea $\Delta_{k\neq0} \neq 0$. The inset shows corresponding results for an electron-hole system at $r_s=2.7$ and $k_Fd=1$. All results are expressed in their respective units.

In this Brief Report, we have investigated the properties of excitonic condensates in graphene bilayers. Our calculations predict that excitonic condensation will occur at all carrier densities as long as $(k_Fd)\sim 1$ and that the strength of the condensate, as measured by the excitonic gap $\Delta_m$, is relatively insensitive to the stacking.

The mean-field results presented in this paper are obtained at zero temperature $T=0$. (Finite temperature analysis gives a critical temperature $T_{MF}/E_F\sim 0.2$ or $T_{MF}\sim 20$ meV. This is an artifact of the mean-field approximation.) In two dimensions, the critical temperature $T_c$ for Bose-Einstein condensation is zero but the superfluid properties survive for $T\approx T_{KT}$, where $T_{KT}$ is the Kosterlitz-Thouless transition temperature. Therefore, our results will be valid at nonzero temperature $T\approx T_{KT}$. A weak disorder will suppress the excitonic condensate order parameter and reduce the excitonic gap, an effect equivalent to increasing the value of $k_Fd$. Therefore, we have ignored the effects of a weak disorder potential.

In our analysis, we have only considered excitonic condensation with uniform density. In conventional (quantum Hall electron-hole) bilayers, varying $d$ and $r_s$ ($\nu$) leads to excitonic condensates with lattice structure. The origin of the lattice structure is Wigner crystallization of carriers in an isolated layer at large $r_s$ (small $\nu$). Graphene does not undergo Wigner crystallization as its carrier density is changed. Therefore, we expect that the excitonic condensate in graphene bilayers remains uniform. Now we show that this result is equivalent to our predictions for density dependence of $\rho_s$ and $m_{\nu\nu}$. The quantum kinetic energy of an exciton, associated with localizing it within a distance $1/k_F$,
is $K = \hbar^2 k_F^2 / 2m_e$. The potential energy due to the dipolar repulsion between them is $P = e^2 d^2 k_F / \epsilon$. Hence, their ratio is given by $P / K = e^2 d^2 k_F m_e / \epsilon \hbar^2$. Wigner crystallization occurs when the ratio $P / K > 1$. This ratio will solely be a function of $k_F d$—no matter what the value of $d$ is—if and only if $m_e = \hbar^2 / \pi k_F^2$. Therefore, results in Sec. IV show that the excitonic condensate in graphene will not undergo Wigner crystallization in spite of the dipolar repulsion between excitons with a quadratic dispersion. This result, too, is unique in graphene and is markedly different from the behavior of dipolar excitonic condensates in conventional bilayers. It is interesting that the mass of these effective bosons has the same density dependence and order of magnitude as the cyclotron mass of fermionic carriers in graphene.

The onset of excitonic condensation can be detected by a divergent interlayer drag. A uniform in-plane magnetic field $B_I$ between the two graphene sheets is expected to induce a (counterflow) supercurrent $J_d$ in such a condensate. This phase stiffness $\rho_d$ and its density dependence can be directly obtained from experimental measurements of the counterflow supercurrent. The verification (or falsification) of our predictions, including the density dependence of $\rho_d$ and $m_e$, will deepen our understanding of properties and condensation of excitons with massless fermions as constituent particles. Recently, we have also become aware of related works.

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