Anomalous Exciton-Condensation in Graphene Bilayers

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In ordinary semiconductor bilayers, exciton condensates appear at total Landau level filling factor $\nu_T = 1$. We predict that similar states will occur in Bernal stacked graphene bilayers at many non-zero integer filling factors. For $\nu_T = -3,1$ we find that the superfluid density of the exciton condensate vanishes and that a finite-temperature fluctuation induced first order isotropic-smectic phase transition occurs when the layer densities are not balanced. These anomalous properties of bilayer graphene exciton condensates are due to the degeneracy of Landau levels with $n = 0$ and $n = 1$ orbital character.

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Introduction— When two semiconductor quantum wells occupied by half-filled Landau levels are narrowly separated, the bilayer system ground state spontaneously establishes inter-layer coherence [1]. These broken symmetry states possess a condensate of pairs (each composed of an electron in one layer and a hole in the other) which opens up an energy gap responsible for a total filling factor $\nu_T = 1$ quantum Hall effect, supports a dissipationless counter-flow excitonic supercurrent, and is responsible for a wide variety of incompletely understood transport anomalies [2, 3]. Recently an interesting new type of bilayer two-dimensional electron system has become available [4, 5] which consists of two carbon-atom honeycomb-lattice (graphene) layers separated by a fraction of a nanometer. The electronic structure of graphene bilayers is full of surprises [6] because of an interplay between the sublattice pseudospin chirality of each layer [7] and the Bernal stacking arrangement, particularly so in the presence of an external magnetic field. In this structure, one of the two-carbon atom sites in each layer has a near-neighbor in the other layer and one does not. Inter-layer hopping drives electrons on the closely spaced sites away from the Fermi level, leaving one low-energy site for carbon $\pi$-orbitals in each layer. Because the hopping process between low-energy sites occurs in two steps, via inter-layer degeneracy. The octet is revealed by a jump in the quantized Hall conductivity [4] from $-4(e^2/h)$ to $4(e^2/h)$ when the charge density is tuned across neutrality in moderately disordered samples. When the sample quality is sufficient for interactions to dominate over disorder, quantum Hall effects occur [8, 9, 10] at all intermediate integers. Spontaneous valley-symmetry breaking is expected [9] in all but the $\nu_T = 0$ (half-filled octet) case. Interactions favor states with spontaneous coherence between valleys, and hence between layers, because this broken symmetry does not require charge transfer between layers. In this Letter we illustrate the unique and rich properties of bilayer graphene exciton condensates by concentrating on the simplest case in which the octet is occupied either by single majority-spin ($\nu_T = -3$) or a single minority-spin ($\nu_T = 1$) Landau level, and allowing for an external potential difference $\Delta V$ between the layers. We demonstrate that the superfluid density vanishes at these filling factors, and that a finite-temperature fluctuation induced first order isotropic-smectic phase transition occurs when the layer densities are not balanced. These anomalous properties of bilayer graphene exciton condensates are due to the degeneracy of Landau levels with $n = 0$ and $n = 1$ orbital character. Below we first discuss the mean-field ground state, demonstrating that it exhibits spontaneous inter-layer phase coherence for small $\Delta V$ at the filling factors of interest. We then explain why the superfluid density vanishes and discuss two important consequences, namely that the phonon collective mode has quadratic rather than the expected linear dispersion and that long-wavelength instabilities appear when $\Delta V \neq 0$. We conclude with some speculations on the experimental implications of our findings.

Octet mean-field theory for unbalanced bilayers— Bilayer graphene’s $N = 0$ Landau-level octet is the direct product of three $S = 1/2$ doublets: real spin and which-layer pseudospin as in a semiconductor bilayer, and the Landau-level ($n = 0,1$) pseudospin which is responsible for the new physics we discuss in this paper. The octet degeneracy is lifted by the Zeeman coupling $\Delta Z$, assumed here to maximize spin-polarization, and by $\Delta V$ which...
is defined so that it favors top layer occupation when positive. In graphene bilayers, $\Delta V$ also drives a small splitting of the Landau-level pseudospin that plays a key role in this paper: $\Delta_{LL} = \alpha \Delta V/h\omega_c/\gamma_1$ where $h\omega_c = 2.14B[\text{Tesla}]$ meV, $\gamma_1 \sim 400$ meV is the interlayer hopping energy, and $\alpha = +1(-1)$ for top(bottom) layers. The fact that $\Delta_{LL}$ 1 has a higher energy than $\Delta_{LL}$ 0 in the top layer and a lower energy in the bottom layer will prove to be important.

The octet mean-field Hamiltonian can be separated into single-particle and exchange contributions:

$$H_{HF} = E_{\alpha,n,s} \rho_{\alpha,n,s;\alpha,n,s}$$

(1)

$$-\frac{1}{g} X^{(\alpha,\alpha)}_{n_1,n_2,n_3,n_4}(0) \langle \rho^{\alpha,\alpha}_{n_1,n_2} \rangle \rho^{\alpha,\alpha}_{n_3,n_4}$$

$$-\frac{1}{g} X^{(\alpha,\pi)}_{n_1,n_2,n_3,n_4}(0) \langle \rho^{\alpha,\pi}_{n_1,n_2} \rangle \rho^{\pi,\alpha}_{n_3,n_4},$$

(repeated indices are summed over) where the single particle energy (which includes the Hartree capacitive term) is

$$E_{\alpha,n,s} = -\frac{s}{g} \frac{\Delta_{LL}}{2} + \alpha \Delta_{LL} + \frac{d}{l_B} \left( \frac{\nu}{2} - \nu \right),$$

(2)

and the exchange interactions

$$X^{(\alpha,\beta)}_{n_1,n_2,n_3,n_4}(q) = \int \frac{d^2p^2_B}{2\pi} e^{-(1-\delta_{\alpha,\beta})}$$

$$\times F_{n_1,n_2}(p) F_{n_3,n_4}(-p) e^{iq \times p^2_B/2}. $$

(3)

In these equations, $g$ is the Landau level degeneracy, $d = 3.337\AA$ is the interlayer separation, $l_B$ is the magnetic length, $\Delta_{LL} = g\mu_B B$ is the Zeeman coupling, $n = 0, 1$ are LL indices, $\alpha, \beta = t, b$ for top(bottom) layers, $r, s = 1(-1)$ for up(down) spins and $t = b, \bar{t} = t$. All energies are in units of $e^2/\varepsilon_{1B}$. The average value of the operators $\rho^{\alpha,\beta}_{n_1,n_2,n_3,n_4} = \sum_X c^{\alpha}_{n_1,n_2} X c^{\beta}_{n_3,n_4}$ (where $c^{\alpha}_{n_1,n_2}$ creates an electron with guiding-center $X$ in the Landau gauge and layer $\alpha$, spin $s$ and Landau level character $n$) must be determined self consistently by occupying the lowest energy eigenvectors of $H_{HF}$. The form factors ($F_{00}(q) = e^{-\nu(q^2l_B^2)/4}$, $F_{10}(q) = (i \nu x + q_y)l_B e^{-\nu(q_z^2l_B^2)/4}/\sqrt{2} = [F_{01}(-q)]^*$ and $F_{11}(q) = (1 - (q_B l_B^2/2)e^{-\nu(q_z^2l_B^2)/4})$ reflect the character of the two different quantum cyclotron orbits.

Although an infinitesimal $\Delta V$ would be sufficient to produce complete layer polarization in a non-interacting system, a finite value is required once interactions are accounted for. Spontaneous interlayer phase coherence arises because it is able to lower energy by inducing a gap at the Fermi level even when both layers are partially occupied. When the layer index is viewed as a pseudospin state, the phase-coherent state corresponds to an $\hat{x} \sim \hat{y}$ easy-plane ferromagnet and the interlayer phase difference corresponds to the azimuthal magnetization orientation. In this language $\Delta V$ is a hard-direction external field which gradually tilts the magnetization direction toward the $\hat{z}$ direction. We find that for $\Delta V \geq \Delta V^* (B)$, with $\Delta V^* (B) / (e^2/\varepsilon_{1B}) \approx 0.001$ at $B = 10T$, the system exhibits full charge imbalance with all electrons (all minority spin electrons in the $\nu_T = 1$ case) occupying one layer. The critical $\Delta V$ is given by

$$\Delta V = \frac{e^2}{\varepsilon_{1B}} \left( \frac{d}{l_B} - \frac{\pi}{2} \left[ 1 - e^{d^2/2\nu^2} \text{Erfc} \left( \frac{d}{\sqrt{2l_B}} \right) \right] \right).$$

(4)

In the graphene bilayer case, the $\Delta V$ sufficient to achieve full pseudospin polarization is reduced compared to the semiconductor case because the layers are close together. For $\Delta V \leq \Delta V^*_r$, the charge-unbalanced mean-field ground state consists of a full Landau level of states which share partially polarized layer and $n = 0$ Landau-level pseudospinors:

$$|\Psi_{GS} \rangle = \prod_X \left[ \cos \left( \frac{\theta_V}{2} \right) c_{1,0,X} \right] e^{i\nu \sin \left( \frac{\theta_V}{2} \right) c_{0,0,X}^\dagger} |0\rangle,$$

(5)

where we have dropped the irrelevant spin degree-of-freedom. In Eq. (5), $\varphi$ is the spontaneous interlayer phase and $\cos (\theta_V) = \Delta V / \Delta V^* (B)$.

Vanishing superfluid density at zero bias—The energy cost of phase gradients is the key property of any superfluid. In normal superfluids, including semiconductor bilayer exciton superfluids, the leading term in a phase-gradient expansion of the energy-density is proportional to $|\nabla \varphi|^2$. We now show that the coefficient which specifies the size of this term, often referred to as the superfluid density, is zero in bilayer graphene exciton condensates at zero bias. We proceed by explicitly constructing a pseudospin wave state in which $\nabla \varphi = q x$ is constant:

$$|\Psi_{PSW} \rangle = \prod_X \left[ \cos \left( \frac{\theta_V}{2} \right) c_{1,0,X} \right] e^{i\nu \sin \left( \frac{\theta_V}{2} \right) \Lambda_{q,X}^\dagger} |0\rangle,$$

(6)

where $\Lambda_{q,X}^\dagger = u q c_{0,1,X}^\dagger + v q c_{1,1,X}^\dagger$ and $u^2 + v^2 = 1$. In this wavefunction the factor exp$(i q X)$ is responsible for the phase gradient. The term proportional to $v_q$ in Eq. (6) allows for the crucial possibility, unique to bilayer graphene, of combining the phase gradient with an admixture of the $n = 1$ wavefunction.

In mean-field theory the energy is proportional to the square of the density-matrix. We now show that it is possible to choose $v_q$ so that the density matrix is unchanged to first order in $q$ and that the superfluid density vanishes as a consequence. Since $v_q$ must vanish for $q \rightarrow 0$ to reproduce $|\Psi_0 \rangle$, we have $v_q \sim q$ and $u_q \sim 1$ to this order. It is then easy to see that the density-matrix component within each layer is unchanged to leading order in $q$. In quantum Hall bilayer exciton condensates, the superfluid density is due to reduced interlayer exchange energy in the presence of a phase gradient[13, 14], and hence to
changes in the interlayer density matrix, $\rho_{tb}(r,r') = \langle \Psi_t^\dagger(r) \Psi_b(r') \rangle$ where the field operator $\Psi_t(b)(r) = 1/\sqrt{\mathcal{V}} \sum_{n=0,1} \sum_X \phi_n(x-X) e^{-iXy/\mathcal{V}} \tilde{\phi}^\dagger(\lambda_{n,b,n,X}$ with $\phi_n(x)$ the harmonic oscillator state with orbital Landau character $n$. At zero bias, Eq. [5] with $\varphi = 0$ gives $\rho_{tb}^{GS}(r,r') = 1/(2\mathcal{V}) \sum_{X} \phi_n^* (x-X) \phi_0 (x'-X) e^{-iX(y-y')/\mathcal{V}}$ while Eq. [6] implies

$$\rho_{tb}^{PSW}(r,r') = \frac{1}{2\mathcal{V}} \sum_{X} \phi_n^* (x-X) e^{-iX(y-y')/\mathcal{V}} \tilde{\phi}^\dagger \tilde{\phi}^\dagger \phi_0 (x'-X) \left[ u_q \phi_0 (x'-X) + v_q \phi_1 (x'-X) \right]$$

(7)

Since $\phi_1(x) = \sqrt(2)(x/l_B)\phi_0(x)$ and $q(x'-X)$ is small because of the localized oscillator wavefunctions, it follows that $\rho_{tb}^{GS}(r,r')$ is altered only by an irrelevant phase factor to first order in $q$ if we choose $u_q = 1, v_q = iql_B/\mathcal{V}$.

The leading change in $|\rho_{tb}|$ is therefore proportional to $q^2$ and the leading energy change proportional to $q^4$.

**Unusual collective mode dispersion**—We now consider collective excitations of the bilayer quantum Hall exciton condensate at $\nu_T = -3,1$, first for balanced ($\Delta V = 0$) bilayers. As discussed above, the ground state is a full Landau level with shared layer symmetric $n = 0$ pseudospin orbitals $|+,0\rangle = (|t,0\rangle + |b,0\rangle)/\sqrt{2}$. The pseudospinor fluctuations that are quantized in the system’s collective modes mix in components from the three orthogonal pseudospin states: $|+,1\rangle$ also symmetric in layer indices and $|-,0\rangle$ and $|-,1\rangle$ antisymmetric in layer indices. Fig. 4 shows the collective mode spectrum calculated in the time-dependent mean-field theory [15]. The inversion symmetric mode (transition $|+,0\rangle \rightarrow |+,1\rangle$) is unrelated to superfluidity and has been discussed previously [9]. For $q = 0$ the two asymmetric modes correspond respectively to global rotations of the $|+,0\rangle$ pseudospinor toward the states $|-,0\rangle$ and $|-,1\rangle$ respectively. The later rotation costs a finite energy because the exchange energy in $n = 1$ is smaller than in $n = 0$.

One physically transparent way of performing these collective mode calculations is to construct a fluctuation action in which each transition has canonically conjugate density $\rho$ and phase $\varphi$ components corresponding to the real and imaginary parts of the final state component in the fluctuating spinor. The fluctuation action

$$S[\rho, \varphi] = S_B[\rho, \varphi] - \int d\omega \int d^2q E[\rho, \varphi],$$

(8)

contains a Berry phase term $S_B$ [16]

$$S_B[\rho, \varphi] = \int d\omega \int d^2q \left[ \frac{1}{2} \rho^\dagger \cdot \rho \partial_q \rho^\dagger - \varphi^\dagger D^\dagger \varphi_q \right],$$

(9)

where $\rho_q = (\rho_{1,q}, \rho_{2,q}, \rho_{3,q})$ and $D = -i\omega_{3,3}$, and an energy functional $E[\rho, \varphi]$ closely related to the discussion in the preceding section

$$E[\rho, \varphi] = \frac{1}{2} \left[ \rho^\dagger \cdot \rho \Lambda_\rho(q) \rho^\dagger + \varphi^\dagger \cdot \varphi \Lambda_\varphi(q) \varphi^\dagger \right].$$

(10)

Here, $\Lambda_\rho(q)$ and $\Lambda_\varphi(q)$ capture the energy cost of small pseudospinor fluctuations and can be evaluated explicitly. Because $\rho$ fluctuations change the charge distribution in the system they remain finite for $q \rightarrow 0$ and do not play an essential role in our discussion. At long wavelengths, we find that the inversion asymmetric block in $\Lambda_\varphi$ has the form

$$\left( \begin{array}{c} X_{1001}^{(0)}(q) \sqrt{2} + \cdots \left( -\frac{i}{2} \frac{X_{1001}^{(-)}(q)}{\sqrt{2}} q l_B + \cdots \right) \\ -\frac{i}{2} X_{1001}^{(-)}(q) \sqrt{2} q l_B + \cdots \end{array} \right) \right.$$  

(11)

with "\cdots" representing terms higher order in $ql_B$.

In a semiconductor bilayer only $n = 0$ phase fluctuations are possible without paying a kinetic energy penalty. In the strong magnetic field limit the superfluid density is therefore proportional to the interlayer exchange-interaction constant $X_{1001}^{-}$. For the bilayer graphene octet, however, the energy cost of phase variation is proportional to the smallest eigenvalue of the matrix in Eq. [11] and this has the $q^4$-long wavelength dependence anticipated above. Indeed the eigenvector of this quadratic form captures the orbital character of the Goldstone mode

$$|GM \rangle = |-,0\rangle + \frac{q l_B}{\sqrt{2}} |-,1\rangle,$$  

(12)
corresponding to the \( v_q/u_q \) ratio in that analysis. We find that the leading small \( q \) behavior for the energy functional is

\[
E_-[\varphi_-] = \beta(qLB)^4 \varphi_-^2 + \cdots ,
\]

where \( \beta = 0.093 (e^2/\varepsilon BL) \) at 10T and \( \varphi_- \) is the amplitude of the lowest-energy eigenmode of the phase matrix \( \Lambda_\varphi(q) \). Because of the conjugate relationship between \( \rho \) and \( \varphi \) fluctuations, the collective mode energy is proportional to the square root of the eigenvalues of \( \Lambda_\rho \) and \( \Lambda_\varphi \) so that the quadratic Goldstone mode dispersion simply signals the vanishing superfluid density.

**Long-wavelength instability at finite bias**—The \( q^4 \) interaction energy cost of inter-layer phase gradients holds for balanced and unbalanced bilayers. In unbalanced layers, however, the Landau level splitting \( \Delta_{LL} \) means that the single-particle energy is lowered by transitions to the \( n = 1 \) Landau level in the bottom layer. For unbalanced layers we find that the phase fluctuation action goes to

\[
\begin{pmatrix}
\frac{X_{1001}^+(0)}{2} qLB^2 + \cdots + i \frac{X_{1001}^-(0)}{\sqrt{2}} qLB + \cdots \\
- i \frac{X_{1001}^-(0)}{\sqrt{1001}} qLB + \cdots X_{1001}^-(0) - \cos(\theta_V) \Delta_{LL} + \cdots
\end{pmatrix}.
\]

The energy cost of phase fluctuations is again proportional to the smallest eigenvalue of the phase matrix,

\[
E_-[\varphi_-] = \frac{1}{2} \cos(\theta_V) \Delta_{LL}(qLB)^2 \varphi_-^2 + \beta(qLB)^4 \varphi_-^2 + \cdots ,
\]

which is negative at small \( q \), indicating that a uniform condensate is unstable when \( \Delta_{LL} \neq 0 \). The energy functional in Eq. (13) reduces to the classical Swift-Hohenberg (SH) model Hamiltonian:

\[
E[\varphi_-] = \left[ \frac{\Delta_0}{2} + \frac{\xi_0^2}{2} (\nabla^2 + q_0^2) \right] \varphi_-^2 + \frac{\lambda}{4} \varphi_-^4 ,
\]

when we set \( \Delta_0 = -\frac{1}{2} (\cos(\theta_V) \Delta_{LL})^2 / 2 \beta, \xi_0^2 = \sqrt{2} \beta q_0^2 \) and \( (q_0LB)^2 = \cos(\theta_V) \Delta_{LL} / 4 \beta \). Using the detailed microscopic form of the mean-field energy functional, we estimate that \( \lambda \sim (q_0LB)^2 (e^2/\varepsilon BL) \). The SH model exhibits a fluctuation-induced first order smectic-isotropic phase transition from a stripe ordered phase with \( \langle \varphi_- \rangle = A \cos(q_0 \cdot r) \) to a disordered phase with \( \langle \varphi_- \rangle = 0 \). Following the self-consistent Hartree approximation analysis of the Swift-Hohenberg model, for \( \Delta_V < \Delta_V^* \) we estimate the transition temperature to be

\[
k_BT_c = \frac{4\xi_0^2}{2.03} \lambda^{3/2} = 1.97 \frac{\beta}{e^2/\varepsilon BL} \left( \frac{\hbar \omega_c}{\gamma_1} \right) \left( \frac{\Delta_V}{\Delta_V^*} \right)^2 .
\]

which is typically below 10mK.

**Discussion**—We identify the \( T > T_c \) phase with a normal quantum Hall ferromagnet and the \( T < T_c \) phase with a quantum Hall smectic state which should exhibit anisotropic transport properties. For \( T > T_c \) we expect properties similar to those observed in semiconductor bilayers except that the coherent interlayer tunneling processes, which plays a key role in tunneling experiments, should be essentially absent. In graphene bilayers therefore, spontaneous coherence is likely to be most conveniently manifested by strongly enhanced Hall drag. Finally we note that trigonal warping terms, which we have neglected, will reduce \( \Delta_{LL} \) by at most 5% for magnetic fields of interest, that correlation effects we have not considered could contribute positively or negatively to the superfluid density, and that the small superfluid densities in this system might lead to important thermal fluctuation effects beyond those considered here. This work was supported by the NSF under grant DMR-0606489 (AHM) a grant by NSERC (RC) and the State of Florida (YB).