A Phase Diagram for Quantum Hall Bilayers with Strong Inter-layer Correlation

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Inter-layer excitonic coherence in a quantum Hall bilayer with negligible tunneling is monitored by measurements of low-lying spin excitations. At \( \nu_T = 1 \) new quasiparticle excitations are observed above a transition temperature revealing a competing metallic phase. For magnetic fields above an onset Zeeman energy this metallic phase has full spin polarization. A phase diagram in the parameter space of temperature and Zeeman energy reveals that the transition temperature increases at higher fields. This unexpected result suggests intriguing impacts of spin polarization in the highly correlated phases.

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Two-dimensional electron systems in double quantum wells in the limit of vanishingly-small tunneling gap \((\Delta_{SAS} \to 0)\) display inter-layer coherence in the quantum Hall (QH) states at total filling factor \( \nu_T = 1 \) (just one Landau level fully occupied) \([1,2]\). This inter-layer correlated quantum state underlying the QH effect is now understood as an easy-plane pseudospin ferromagnet (up/down pseudospin labels the electron occupation in the two layers). Equivalently, the state can be viewed as a Bose-Einstein condensate of inter-layer excitons by making a particle-hole transformation (see Fig.1A) \([3]\). The strongly correlated excitonic phase is incompressible with gapped low-lying collective excitations that are linked to superfluid-like signature for the excitonic transport such as seen in counterflow current experiments \([3,4,5,6,7,8]\). In counterflow the onset of inter-layer coherence occurs when the parameter \( d/l_B \) is made smaller than \( \approx 1.9 \) \((d \) is the interlayer distance and \( l_B \) the magnetic length) \([2,4,5,7,8]\). Further, recent magneto-transport experiments have stressed the role of spin \([10]\) and focussed on the finite temperature properties of the broken-symmetry excitonic phase \([11]\). Recent theory of inter-layer coherence in bilayer graphene suggests that such superfluid-like behavior might be observable up to room temperature \([12,13]\).

Theoretical formulations have raised the possibility that the excitonic fluid competes with compressible (metallic) states of composite-fermions (CF) \([14,15]\). In these interpretations a CF phase, metallic or BCS-like, occurs in the limit of large \( d/l_B \) when the system is regarded as two single layers each of them at \( \nu = 1/2 \). The emergence of a Fermi sea of CFs has key manifestations in excitations above the quantum ground state. In the low-lying sector of spin modes there is a continuum of spin-flip excitations from transitions between spin-up and spin down states across the CF Fermi energy \([16]\). The quasiparticle transitions in this scenario are shown in Fig.1B for a bilayer structure. The presence of such spin excitations in spectra of bilayers are signatures of CF phases. Studies of this class of low-lying spin modes thus offer direct insights on the interplay between excitonic and CF phases in bilayers with strong inter-layer correlation.

We report here an experimental study of low-lying spin excitations in QH bilayers at \( \nu_T = 1 \) and with \( \Delta_{SAS} \to 0 \). The modes are observed by resonant inelastic light scattering. The experiments probe the regime where inter-layer coherence and excitonic superfluidity have been reported. At the higher temperatures the spectra show low-lying spin-flip modes below the Zeeman energy at \( E_Z \) similar to the CF excitations reported in single electron layers at \( \nu \to 1/2 \) \([16]\). We surmise that at the higher temperatures the two layers are largely decoupled and that each layer is in a metallic state with characteristic low-lying spin-flip excitations such as those of a CF Fermi sea in a single electron layer at \( \nu \to 1/2 \). At lowest temperatures and \( \nu_T = 1 \) there is near-absence of low-lying spin-flip modes. This demonstrates the emergence of inter-layer coherence of an excitonic phase. Slight filling factor deviations from \( \nu_T = 1 \) restore the signatures of low-lying spin-flip modes.

We have carefully monitored the evolution of low-lying spin excitations as a function of temperature and Zeeman energy. With these results we construct a finite-temperature phase diagram for the interplay between excitonic and metallic phases of bilayers at \( \nu_T = 1 \). We find that full spin polarization of the metallic phase with CF signatures occurs at an onset Zeeman energy \( E_Z \) \( \approx 0.12 \) meV that is reached at fields \( B_T \) \( \approx 5 \)T. Surprisingly, we find that the transition temperature for the transformation to the CF phase significantly increases for Zeeman energies \( E_Z \geq E_T \). The unexpected behavior of the critical temperature could be linked to the emergence of

\[\Delta_{SAS} \to 0.\]
a novel correlated phase at intermediate temperatures. Our results therefore highlight a rich phase diagram with multiple phases dictated by competition between quantum and thermal effects.

Measurements were performed on the sample mounted on a mechanical rotator in a dilution refrigerator with base temperature of 50 mK under light illumination. The sample is a nominally symmetric modulation-doped AlO.1Ga0.9As/GaAs double quantum well structure with AlAs barrier in between the wells, having well width of 18nm and barrier width of 7nm. The large barrier ensures that the tunneling gap is vanishingly small ($\Delta_{SAS} \rightarrow 0$). The total electron density is $n_T \sim 6.9 \times 10^{10}$ cm$^{-2}$ and electron mobility above $10^6$ cm$^2$/Vs. A perpendicular magnetic field of 2.85T was applied to bring the electron bilayer in the quantum Hall (QH) state corresponding to $\nu_T = 1$. The sample has $d/l_B = 1.65$. Therefore electrons at sufficiently low temperatures are in the correlated phase where exciton condensation and counterflow superfluidity take place. Spin excitations were measured by resonant inelastic light scattering in a backscattering configuration at different tilt angles $\theta$ between the magnetic field direction and the plane of the sample (see left part of Fig.2). To this end a single-mode tunable Ti:Sapphire laser at around 810 nm and a triple-grating spectrometer equipped with a CCD detector were used. Laser power densities were kept at $\sim 10^{-4}$ W/cm$^2$ to avoid electron heating effects and a crossed polarization configuration with perpendicular incident and scattered photon polarizations was exploited to have access to spin modes [15] (see supplementary materials).

Figure 1 describes the energy level structure and electronic spin-flip excitations of the excitonic QH phase (A) and of the Fermi seas of CFs (B) that occur when inter-layer coherence is lost. We recall that a CF quasiparticle can be viewed as an electron with an even-integer number of magnetic flux quanta attached to it. As a consequence, the CFs sense an effective magnetic field that is zero when the electron filling factor is $\nu = 1/2$. In this limit the spacing between the CF Landau levels vanishes and the CFs coalesce into a Fermi sea as shown in Fig.1 right panel in the case of double layers with $\nu_T = (\nu=1/2) + (\nu=1/2) = 1$.

In the excitation spectra, the formation of a Fermi sea of CFs manifests into a continuum of spin-flip modes between spin-up and spin-down states across the CF Fermi energy (see the diagram in Fig.1B) [16, 21, 22]. Such excitations, that are accessible by inelastic light scattering methods, represent the hallmark of the CF phase in bilayers. As shown below these collective modes offer direct probes of the manifestation of phases with strong inter-layer excitonic correlation. The collective electronic spin-flip excitation in the excitonic phase, instead, can be understood by considering the excess spin-up electrons that remain after the particle-hole transformation. These electrons fill exactly one Landau level and support a well-defined long wavelength SW mode at the Zeeman energy as shown in Fig.1A.

Figure 2 reports representative low-lying spin excitations measured as function of small changes in filling factor near $\nu_T = 1$. The spectra reveal a well-defined SW mode with Lorentzian lineshape and the continuum of CF spin excitations with a pronounced minimum when the bilayer is in the QH state at $\nu_T = 1$. This significant result indicates a trend for the compressible CF metal to transform into a new phase with inter-layer coherence when the total filling factor approaches $\nu_T = 1$. Observation of residual (SF$_{CF}$) modes seen at $\nu_T = 1$ at the lowest temperatures (see Fig.3 also) is consistent with co-existence of compressible-incompressible puddles due to disorder in agreement with previous experimental and theoretical analysis [23, 24, 25, 26]. No spin-flip mode is seen above the SW. This mode is indeed observed in samples with finite values of $\Delta_{SAS}$ [18, 22]. Its absence supports the conclusion of negligible interlayer tunneling in the sample studied here.

The observation of SF$_{CF}$ excitations demonstrates the emergence of a CF metallic phase and illustrates how the inter-layer coherence of the QH fluid is lost by varying the filling factor. As inter-layer coherence disappears the physics becomes dominated by the intra-layer correlations that build up the CF quasiparticles. Particularly revealing of the competition between the inter-layer excitons and the CFs is the behavior as a function of temperature shown in Fig.3. The integrated intensity of the spin
FIG. 2: Left panel: Geometry of the experimental set-up for inelastic light scattering. $\omega_L$ and $\omega_S$ refer to incident and scattered photon frequencies (see supplementary materials for a description of the inelastic light scattering mechanism). Representative spin excitation spectra at $T = 50$ mK and three values of filling factor $\nu_T$ at a tilt angle of $\theta = 67.5^\circ$. A lorentzian fit to the spin wave (SW) is shown (red line) to highlight the impact of the continuum of spin-flip excitations SF$_{CF}$ (shaded in green). The gray line is the background due to the magneto-luminescence.

continuum remains small and constant up to a critical value of temperature $T_c$ and then it increases markedly. This anomalous behavior also seen in magneto-transport characteristics of the bilayer QH state $[11, 27]$ suggests a finite-temperature phase transformation of the coherent QH state to the CF metal at $T_c$. Weather the transition exploits a Kosterlitz-Thouless mechanisms as expected for the broken-symmetry bilayer state remains an open issue that deserves additional experimental investigation.

It can be noted that the lowest energy value $E_{CF}^{opp}$ of the SF$_{CF}$ continuum remains finite at the lowest temperature in the data displayed in Figs. 2 and 3, measured at the relatively large angle $\theta = 67.5^\circ$ ($B_T = 7.46$T). This shows that the CF metal is fully spin-polarized. The lowest-energy value of the spin-flip continuum, however, lowers to values below the experimental resolution (30µeV) as temperature increases (see the lowest spectrum in Fig.3 at $T = 1$K). The same happens as the angle decreases (data not shown). The collapse of $E_{CF}^{opp}$ signals a spin transition to a CF state with partial spin polarization. Further evidence of the spin transition of the CF metal as a function of angle is also found in the behavior of the SW intensity versus temperature (see supplementary materials).

The phase diagram shown in Fig. 4 is obtained by reporting the measured values $T_c$ at different values of the tilt angle while keeping the filling factor at $\nu_T = 1$. This phase diagram highlights an intriguing competition between a low-temperature region where inter-layer correlation dominates with the high-temperature CF phase. It also shows that a spin transition to a fully spin-polarized CF state occurs at a critical magnetic field of $B_T^* \approx 5$T.

The increase of $T_c$ for magnetic fields above $B_T^* = 5$T is unexpected. The reason is that no further change in the boundary between the two phases is expected for $B_T \geq 5$T when also the CF state becomes fully spin-polarized (see the horizontal dotted line in Fig.4) $[10, 11]$. Additionally, at large $\theta$ values, the in-plane magnetic field leads to the compression of the electron wavefunction along the z-direction. Although this orbital effect is expected to be rather small in our quantum wells $[10]$, it increases the intra-layer interactions responsible for the formation of CF quasiparticles $[28]$ and should favor the CF phase. Again, this scenario is in contrast to the behavior shown in Fig.4.

We conjecture that the increase in $T_c$ for $B_T \geq B_T^* = 5$T could be evidence for the formation of an intermediate non-CF phase. Unlike the coherent phase of inter-layer excitons that occurs at the lowest temperatures below the dotted line in Fig.4, this intermediate phase could have residual but non negligible inter-layer correlation. It is tempting to associate this additional phase to the formation of a new ground state due to coupling between CFs in the two layers. Recent predictions have indeed shown that a QH state due to CF pairing could lead to an
inter-layer excitonic and intermediate phases. The orange region extends down to below 30
points define the temperature at which the spin-flip transition temperature is given as function of the total magnetic
field \( B_T \) and Zeeman energy \( E_Z \). Green (blue) regions indicate spin-unpolarized (spin-polarized) CF metals. The red
region at large total fields defines a possible intermediate phase. The dotted line is the hypothetical boundary between the inter-layer excitonic and intermediate phases. \( E_Z^* \) represents the critical value of the Zeeman energy that marks the transition from the spin-unpolarized to spin-polarized CF metal.

FIG. 4: Phase diagram for the competition between the incompressible inter-layer coherent state and the compressible bilayer composite-fermion metal (CF) at \( \nu_T = 1 \). The transition temperature is given as function of the total magnetic field \( B_T \) and Zeeman energy \( E_Z \). Green (blue) regions indicate spin-unpolarized (spin-polarized) CF metals. The red
region at large total fields defines a possible intermediate phase. The orange region at large total fields defines a possible intermediate phase. The dotted line is the hypothetical boundary between the inter-layer excitonic and intermediate phases. \( E_Z^* \) represents the critical value of the Zeeman energy that marks the transition from the spin-unpolarized to spin-polarized CF metal.

Intermediate phase between the coherent excitonic and CF states with a BCS order parameter \[15, 17\]. As the inter-layer correlation further increases at lower temperatures this intermediate phase develops eventually into the excitonic state. Further experiments are needed in this regime to test this interpretation. We would like to remark, however, that in transport experiments at large tilt angles, the disappearing of the excitonic phase is monitored by the collapse of the QH effect. This observation seems to suggest that the intermediate phase is compressible.

**Supplementary materials**

**Mechanism for resonant inelastic light scattering** Within time-dependent perturbation theory the resonant inelastic light scattering cross section includes two virtual intermediate valence-to-conduction band optical transitions. The resonance enhancement occurs when the incoming and/or scattered photon overlaps in energy a real interband transition across the GaAs band gap. Conservation of energy implies that \( \hbar \omega_l - \hbar \omega_S = \pm \hbar \omega(q) \) where \( \hbar \omega_l,S \) represents the energy of the incoming or scattered photon and \( \hbar \omega(q) \) the energy of the collective mode with in-plane wavevector \( q \). When the two-dimensional electron gas has full translational invariance there is conservation of in-plane wavevector in the light scattering process. In this case the transferred component of the wavevector \( q \) is related to the photon momentum in the crystal and with infrared source is small and typically in the range \( q \cdot l_B \leq 0.2 \) where \( l_B \) is the magnetic length \[29\].

Inelastic light scattering by excitations with reversal of the spin such as the spin-wave (SW) exploits p-like states at the top of the valence band such as light-hole states with mixed spin and orbital character due to spin-orbit coupling. In addition the incoming or scattered light must have a component of the polarization along the z-axis perpendicular to the QW plane which requires a tilted field geometry with a non-zero angle \( \theta \). The cross section for spin excitations is proportional to \( e_l \times e_S \) where \( e_l(S) \) are the incoming (scattered) photon polarizations \[30\].

**Activation energy of the spin wave** Further evidence of the spin transition of the CF metal is also found in the behavior of the SW intensity versus temperature. The integrated intensity of the Lorentzian fit to the SW reported in Fig.1 as a function of the inverse of temperature for \( \theta = 67.5^\circ \) remains constant up to \( \approx 150 \) mK and then displays an activated behavior. The resulting activation gap \( \Delta \) depends on \( B_T \) as shown in the inset to Fig.1 with a minimum at 5T that signals a change in the spin properties of the CF phase. For comparison the val-

FIG. 5: Integrated intensity \( I \) of the SW peak as a function of temperature. The red line represents the best-fit result with the function \( I(T) = I_o(1 - e^{\Delta/kT}) \). The inset shows the behavior of \( \Delta \) as a function of total magnetic field \( B_T \). The peak energy of the SW peak that corresponds to the Zeeman energy \( E_Z \) is also shown. The black line is a fit to the equation \( E_Z = g \mu_B B_T \) where \( \mu_B \) is the Bohr magneton and \( g = -0.4 \) the gyromagnetic factor.
ues of $E_Z$, identified as the peak energy of the SW mode as predicted by the Larmour theorem, is also plotted in the inset to Fig.1. Further work is required to develop the microscopic mechanism underlying the behavior shown in Fig.1

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