

## First-Order Quantum Phase Transition of Excitons in Quantum Hall Bilayers

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We show that the quantum phase transformation between compressible metallic and incompressible excitonic states in the coupled bilayers at total Landau level filling factor  $\nu_T = 1$  becomes discontinuous (first order) by impacts of different terms of the electron-electron interactions that prevail on weak residual disorder. The evidence is based on precise determinations of the excitonic order parameter by inelastic light scattering measurements close to the phase boundary. While there is marked softening of low-lying excitations, our experiments underpin the roles of competing order parameters linked to quasiparticle correlations in removing the divergence of quantum fluctuations.

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When dominated by quantum-mechanical fluctuations, ground state transformations display quantum criticality associated with collapse of the characteristic energy scale expressed by softening of low-lying excitation modes [1–3]. In the presence of competing interactions and weak disorder the nature of the quantum phase transition (QPT) is predicted to become discontinuous as the critical point is approached [4–7]. One example is offered by the metal-insulator Mott transition which, in the absence of disorder, is believed to be first order [8]. The observation of these elusive effects require a fine-tuning of the impact of the interactions that is difficult to achieve in real systems. We show in this Letter that quantum Hall (QH) bilayers display competing ground states stabilized by different terms of electron interactions that can be tuned with high precision. This competition drives this soft-mode system to have a first-order QPT.

In the bilayers the quantum well energy levels split into symmetric and antisymmetric combinations separated by the tunneling gap  $\Delta_{SAS}$ . At total Landau level filling factor  $\nu_T = n_T 2\pi l_B^2 = 1$  ( $n_T$  is the total electron density in the bilayer and  $l_B$  is the magnetic length), the electron bilayer in high-quality AlGaAs/GaAs double quantum wells display two phases that result from the interplay between interlayer and intralayer electron interaction [9,10]. These Coulomb interactions are parametrized by  $d/l_B$ , where  $d$  is the distance between the two layers and by  $\Delta_{SAS}/E_c$ , where  $E_c = e^2/\epsilon l_B$  ( $\epsilon$  is the dielectric constant) is the intralayer Coulomb energy [9,11]. At sufficiently low  $d/l_B$  or large  $\Delta_{SAS}/E_c$ , the ground states at  $\nu_T = 1$  are incompressible QH fluids [9,10]. Early theoretical approaches at  $\nu_T = 1$  based on a mean-field analysis described the QH physics of bilayers by assuming full occupation of the lowest symmetric Landau level [12,13]. These studies have linked the disappearing of the incompressible QH phase at low values of  $\Delta_{SAS}/E_c$  to the softening of the tunneling charge excitation at a magnetoroton

wave vector, thus suggesting a continuous second-order QPT.

However, evidence of softening of magnetoroton modes at  $\nu_T = 1$  is dwindling. Numerical studies and recent inelastic light scattering experiments demonstrated the breakdown of the mean-field picture close to the phase transition highlighting the role of electron correlations [14–16] at  $\nu_T = 1$ . The light scattering studies have shown that even at sizable  $\Delta_{SAS}$  interlayer correlations favor spontaneous occupation of the excited antisymmetric level [16]. Such correlated phases, in which antisymmetric wave functions are embedded in the ground state, retain the dissipationless transport characteristic of the QH regime. The retention of QH signatures suggests a description of the highly correlated phase at  $\nu_T = 1$  in terms of coherent electron-hole excitonic pairs [17] across  $\Delta_{SAS}$  (top-left drawing in Fig. 1). A different phase occurs for low  $\Delta_{SAS}/E_c$  (at sufficiently large  $d/l_B$ ), when intralayer correlations prevail on the interlayer correlations. The compressible (non-QH) ground state of this phase is currently described as a Fermi metal of composite fermions (CFs) (top-right drawing in Fig. 1) [18].

The keen interest in the interplays between excitonic and CF correlated phases in bilayers has prompted consideration of differing scenarios for the states close to the phase boundary, for the character of the transformations [14,19–22], and for possible roles of the electron spin [23,24]. Investigations of the QPTs in bilayers, however, have been hindered by the uncertain role of disorder, that could possibly result to quasicontinuous changes due to smearing of the free-energy landscape and presence of domains of the two phases close to the phase boundary [19,25,26].

To determine the character of the QPT in bilayers at  $\nu_T = 1$ , we employed resonant inelastic light scattering to study the spin excitations in the excitonic and CF phases, as shown in the middle panels of Fig. 1. The measurements were performed on the sample mounted on a mechanical rotator in a dilution refrigerator with base temperatures

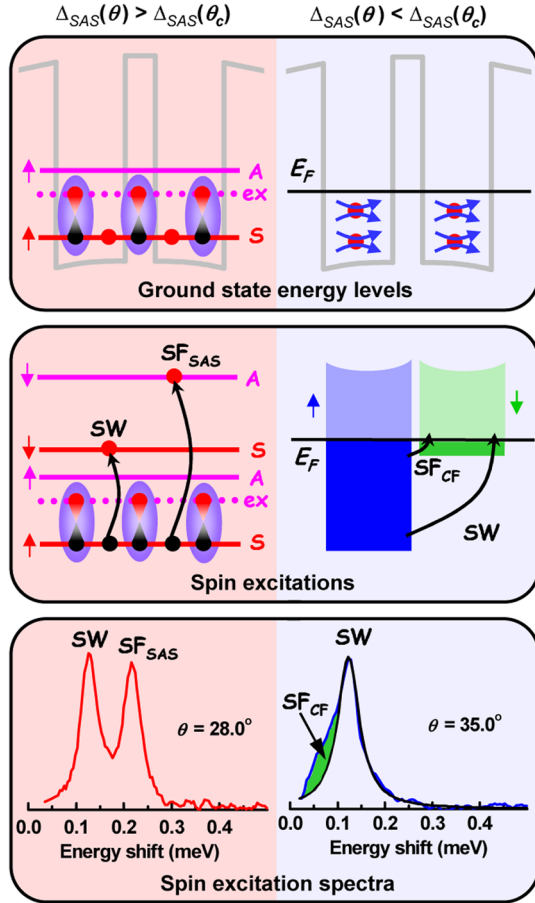


FIG. 1 (color online). Top panel: Representation of the correlated phases in bilayers at  $\nu_T = 1$ . The excitonic QH phase is shown on the left. Electron-hole pairs occur between symmetric ( $S$ ) and antisymmetric ( $A$ ) spin-up Landau levels. The CF phase is shown on the right. The Fermi energy indicates the occupied CF levels. Middle panel: Energy level diagrams and spin excitations.  $SW$  and  $SF_{SAS}$  refer to spin wave across the Zeeman gap and spin-flip mode across the tunneling gap, respectively. In the CF phase, a spin-flip ( $SF_{CF}$ ) continuum of collective excitations extends from the Zeeman gap down to an energy value determined by the relative position of the Fermi level within the spin-up and spin-down CF states. Bottom panel: Representative spin excitation spectra after background subtraction due to magnetoluminescence and stray light in the two phases at  $T = 50$  mK. A Lorentzian fit to the  $SW$  in the CF phase (black line) highlights the impact of the  $SF_{CF}$  continuum (shaded in green).

below 100 mK under light illumination. The in-plane magnetic field reduces the tunneling gap according to [27]  $\Delta_{SAS}(\theta) = \Delta_{SAS}(\theta = 0) \exp\{-[\tan(\theta)d/2l_B]^2\}$ . We reached a precision on angle of  $0.1^\circ$  [precision on  $\Delta_{SAS}(\theta)/E_c$  of  $1 \times 10^{-4}$ ] enabling the investigation of the evolution of the correlated phases with unprecedented accuracy. Long wavelength spin excitations were measured in a backscattering configuration using a Ti:sapphire laser finely tuned to reach the best resonance condition and detected by a triple-grating spectrometer equipped with a CCD detector. Laser power densities were kept at  $\sim 10^{-4}$  W/cm<sup>2</sup> and a crossed polarization condition,

where incident and scattered photons have opposite polarizations, was used [16]. The sample studied is a nominally symmetric modulation-doped  $Al_{0.1}Ga_{0.9}As/GaAs$  double quantum well grown by molecular beam epitaxy with total electron density of  $n_T \sim 1.1 \times 10^{11}$  cm<sup>-2</sup>, mobility above  $10^6$  cm<sup>2</sup>/Vs,  $d/l_B = 2.18$  ( $d = 25.5$  nm and the well width is 18 nm), and measured tunneling gap at zero magnetic field of  $\Delta_{SAS} = 0.36$  meV. Measurements of spin excitations across the Zeeman gap [spin wave ( $SW$ )] as a function of total magnetic field ( $B_T$ ) yields a  $g$  factor of  $g = -0.4$ .

The left-bottom panel of Fig. 1 reports an example of a spin excitation spectrum in the  $\nu_T = 1$  excitonic phase while spin spectra at different values of  $\theta$  (i.e., different values of  $\Delta_{SAS}/E_c$ ) are reported in Fig. 2. The spin-flip tunneling mode ( $SF_{SAS}$ ) [28] appears at energies above the  $SW$  at the “bare” Zeeman energy ( $E_z = g\mu_B B_T$ , where  $\mu_B$  is the Bohr magnetron) by the Larmor theorem. Signatures of the excitonic phase are manifested in the intensity and energy position of  $SF_{SAS}$ : the intensity at resonance is linked to the spatial extent of the excitonic domains and vanishes at the transition to the CF metallic state. The energy also carries key information on the excitonic phase. In fact, within Hartree-Fock, the  $SF_{SAS}$ - $SW$  energy splitting  $\delta(\theta)$  is equal to  $\Delta_{SAS}(\theta)$ . The experimental values that are reported in Fig. 3(a) in units of  $E_c$ , on the contrary, are significantly reduced from  $\Delta_{SAS}(\theta)/E_c$  (see horizontal scale) due to the impact of interlayer electron

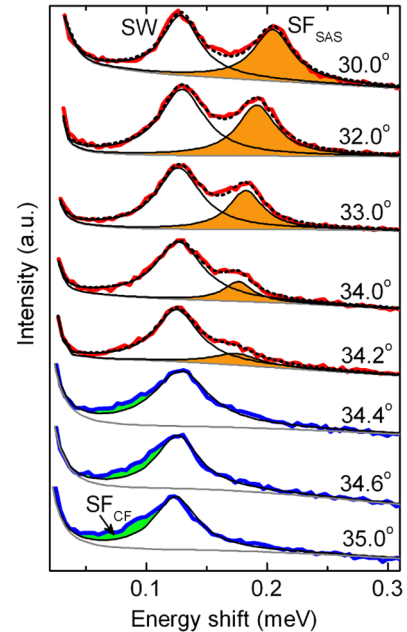


FIG. 2 (color online). Angular dependence of spin excitations around  $\theta_c = 34.2^\circ$ .  $SF_{SAS}$  excitation intensity in the excitonic phase is shaded in orange and the spin-flip continuum  $SF_{CF}$  in the composite-fermion metallic phase is shaded in light gray (green). Best fit results to the spectra (dotted lines) with two Lorentzians (one in the CF phase) and the magnetoluminescence continuum and laser stray light (solid gray lines) are shown.

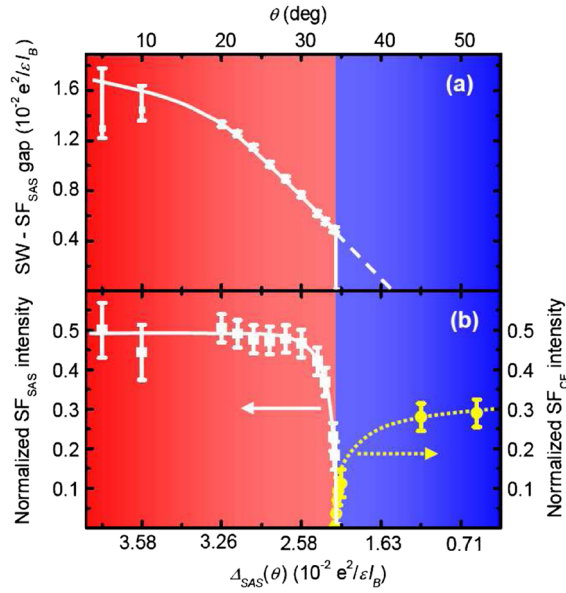


FIG. 3 (color online). (a) Evolution of the correlated gap of the excitonic phase, i.e., the energy splitting between SW and  $SF_{SAS}$  excitations, as a function of tunneling gap or tilt angle. The splitting remains finite at the phase boundary between the excitonic phase [red shaded (left-hand side)] and the CF metal [blue shaded (right-hand side)]. (b) Plot of integrated spin-flip excitation intensity. Data correspond to the  $SF_{SAS}$  (white) and  $SF_{CF}$  [light gray (yellow)] intensity (solid lines and dotted lines are guides for the eyes). The values are obtained after normalization with respect to the spin-wave SW intensity to take into account the angular dependence of oscillator strength of spin excitations.

correlation. Such correlation effects that yield a softening of the mode can be captured by a simple expression that links the splitting to the density of excitonic pairs  $n_{ex}$  through the relation [16]

$$\delta(\theta) = \Delta_{SAS}(\theta) \left[ \frac{n_S - n_{AS}}{n_T} \right] = \Delta_{SAS}(\theta) \left[ 1 - 2 \frac{n_{ex}}{n_T} \right], \quad (1)$$

where  $n_S(n_{AS})$  is the density of electrons in the symmetric (antisymmetric) level. The correlated excitonic state can also be described as an imperfect pseudospin ferromagnet (pseudospin  $\tau_x = \pm 1$  describes occupation of symmetric or antisymmetric Landau levels) with a pseudospin polarization (or order parameter)  $\langle \tau_x \rangle < 1$ . Within this framework [16]  $\delta(\theta) = \Delta_{SAS}(\theta) \langle \tau_x \rangle$ , and a continuous second-order QPT is expected when  $\langle \tau_x \rangle = 0$  [ $\delta(\theta) = 0$ ]. Current theories interpret the reduction of  $\langle \tau_x \rangle$  in terms of quantum fluctuations of the pseudospin ferromagnet [29].

In contradiction with this picture, the data in Figs. 2 and 3(a) demonstrate a QPT linked to the disappearance of the  $SF_{SAS}$  excitation at a finite value of  $\delta(\theta)$ , which is the correlated gap and order parameter of the excitonic phase. These results offer evidence of a first-order character of the transformation. Figure 3(a) is revealing. The evolution of  $\delta(\theta)$  in the excitonic phase suggests a continuous QPT at an angle slightly above  $40^\circ$ . There is, however, a marked

discontinuity in the value of  $\delta(\theta)$  at the critical angle ( $\theta_c = 34.2^\circ$ ) that points to a first-order QPT and to a subtle competition of the collective excitonic state with the CF metal phase.

Evidence of the  $\nu_T = 1$  CF phase is found in the spectral line shape of the SW mode that changes remarkably just above the critical angle as seen in the spectra at  $\theta = 34.4^\circ$ ,  $34.6^\circ$ , and  $35.0^\circ$  in the bottom panel of Fig. 1 and in Fig. 2. Here the CF quantum phase is characterized by a low-energy continuum of collective spin excitations [30] that appears below the SW mode. These  $SF_{CF}$  excitation modes are best identified as spin-flip transitions across the Fermi energy of the CF metal [18,31] as indicated in the schematic drawing of CF energy levels shown in the right-middle panel of Fig. 1.

The first-order character of the QPT is further supported by the abrupt collapse of the  $SF_{SAS}$  integrated intensities above  $\theta_c = 34.2^\circ$  that is shown in Fig. 3(b). The collapse links with the disappearing of the excitonic phase as the critical angle is approached. We note that the precision achieved in determining the value of the critical angle reflects the electron density (i.e., filling factor) uniformity within the quantum Hall excitonic phase. This stems from the observed large dependence of  $\theta_c$  on filling factor values ( $\delta\theta_c/\delta\nu_T = 160$  [32]). Taking this into account we can conclude that filling factor fluctuations ( $\delta\nu_T$ ) around  $\nu_T = 1$  within the excitonic phase are smaller than  $1.2 \times 10^{-4}$ .

The appearance of the  $SF_{CF}$  continuum of the CF metal is concomitant with the vanishing of the  $SF_{SAS}$  intensity and the collapse of the correlated gap of the excitonic phase to its lowest value as displayed in Figs. 2 and 3. This further highlights that the transition corresponds to the incompressible-compressible transition and not to a transition to an incommensurate phase [10]. The role of disorder in smearing the sharp QPT is manifested in the temperature dependence of the spectra shown in Fig. 4. The ‘‘critical’’ temperature of the disappearance of the  $SF_{SAS}$  mode decreases continuously as the critical angle is approached. This behavior, in agreement with transport data [26] performed in the limit of vanishing tunneling, suggests that phase coexistence at finite temperatures could be due to competition of the free energies of the two phases. It should be noted that despite this phase coexistence there is no clear overlap of the CF low-lying continuum and  $SF_{SAS}$  mode in the spectra at the lowest temperature. The minority compressible phase could be disordered or these puddles too small to support collective modes. At finite temperature instead (see the 200 mK spectrum in Fig. 4(g)) the CF low-lying continuum seems to appear. More studies are needed to clarify this issue. Figure 4 highlights that the collapsed value of the correlated gap  $\delta(\theta_c)$  is not affected by temperature in the range where the  $SF_{SAS}$  is visible ruling out that its measured finite value at the critical angle is due to the finite temperature of the system.

The topic of the impact of competing correlated phases on QPTs is the focus of considerable interest in studies of QH liquids [5,33]. Some theoretical works have described

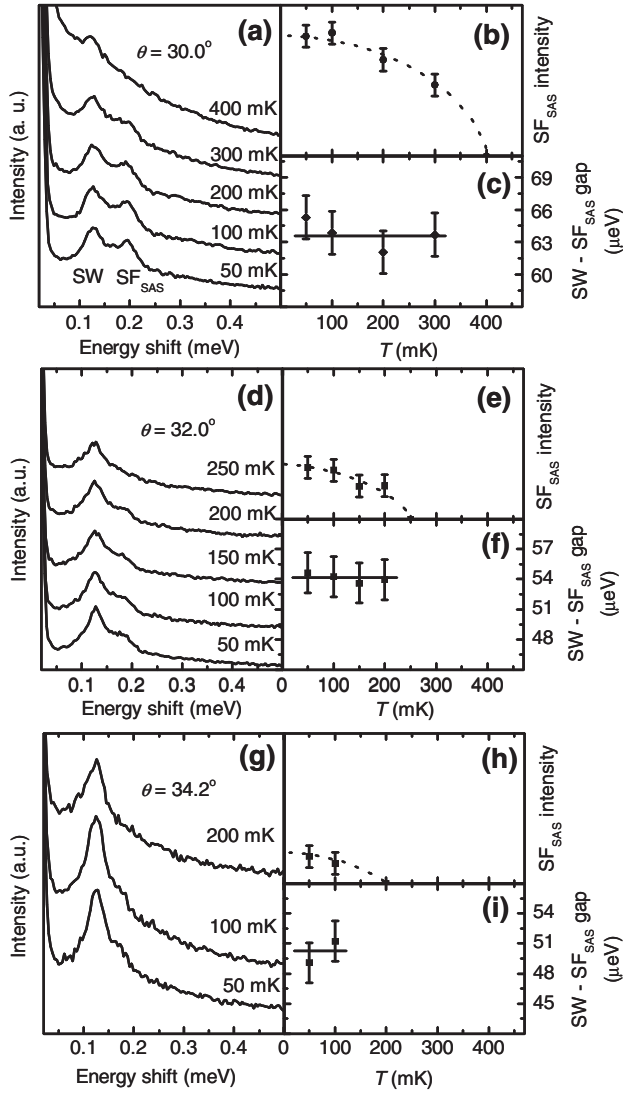


FIG. 4. (a),(d),(g) Temperature dependence of spin excitations in the excitonic phase at filling factor  $\nu_T = 1$  for three different tilt angles. (b),(e),(h) Normalized spin-flip  $SF_{SAS}$  intensity as a function of temperature and tilt angles. (c),(f),(i) Plot of  $SW - SF_{SAS}$  energy splitting versus temperature. The correlated gap of the excitonic phase remains constant within uncertainty as temperature increases.

competing interaction terms by gauge fields suggesting that gauge field fluctuations could lead to competing order parameters. First-order transitions in the absence of disorder are predicted in QH insulators [5] in other condensed matter systems [8] and in particle physics [7]. One key property of the electron bilayer system is that the roles of different electron correlation terms at the QPT can be tested by finely tuning their relative strength. Further studies in bilayers at  $\nu_T = 1$  should explore the nature of the transition in samples with zero tunneling gap and as a function of the spin polarization of the CF metallic state.

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