Observation of Collapse of Pseudospin Order in Bilayer Quantum Hall Ferromagnets

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The Hartree-Fock paradigm of bilayer quantum Hall states with finite tunneling at filling factor $\nu = 1$ has full pseudospin ferromagnetic order with all the electrons in the lowest symmetric Landau level. Inelastic light scattering measurements of low energy spin excitations reveal major departures from the paradigm at relatively large tunneling gaps. The results indicate the emergence of a novel correlated quantum Hall state at $\nu = 1$ characterized by reduced pseudospin order. Marked anomalies occur in spin excitations when pseudospin polarization collapses by application of in-plane magnetic fields.

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Electron bilayers in semiconductor quantum structures embedded in a quantizing perpendicular magnetic field ($B_\perp$) are contemporary realizations of collective systems where bizarre quantum phases may appear [1–4]. In particular, at a Landau level filling factor $\nu = 1$ they may support excitonic superfluidity when there is no tunneling between the layers [4–6]. These phases are driven by unique interplays between intralayer and interlayer Coulomb interactions and have been the subject of large research efforts in recent years [7–12]. The bilayer system is also characterized by the "bare", or Hartree, tunneling gap Δ_{SAS} that represents the splitting between the symmetric and antisymmetric linear combinations of the lowest quantum well levels, as shown in Fig. 1(a). The ground state in the presence of tunneling displays the well known manifestations of the incompressible quantum Hall (QH) fluid. Interactions, however, create intriguing behaviors such as the disappearance at $\nu = 1$ of QH signatures when d/l_B (d is the interwell distance and l_B the magnetic length) is increased above a critical value [13,14].

The states of electron bilayers are efficiently described by introducing a pseudospin operator $\boldsymbol{\tau}$ [4]. Electrons in the left (right) quantum well have pseudospin along the $+z$ ($-z$) direction, normal to the plane. The symmetric (antisymmetric) states have pseudospin aligned along the $+x$ ($-x$) direction. The mean-field Hartree-Fock configuration of QH incompressible states with only the lowest symmetric level populated is shown in Fig. 1(a). This state has full pseudospin polarization along the x direction with an order parameter given by the average value of the normalized pseudospin polarization $\langle \tau^x \rangle = 1$.

Here we report direct evidence that the pseudospin order of the Hartree-Fock paradigm is lost. The unexpected QH states with reduced pseudospin polarization are probed by inelastic light scattering measurements of low-lying spin excitations. One of the excitations is the long wavelength ($q \to 0$) spin-flip (SF) mode built with transitions across Δ_{SAS} with simultaneous change in spin orientation. The other is the long wavelength spin-wave (SW) excitation built from transitions across the Zeeman gap E_Z of the lowest spin-split symmetric levels. The transitions that build SW and SF excitation modes are depicted in Fig. 1(a). The time-dependent Hartree-Fock approximation (TDHFA) that has been extensively employed to interpret bilayer experiments [14–17] dictates that in the mean-field $\nu = 1$ state with $\langle \tau^x \rangle = 1$, the splitting between long wavelength SF and SW modes is $\delta E_\tau = \Delta_{\mathrm{SAS}}$ [16]. We discuss below that the measured deviations of δE_τ from Δ_{SAS} provides direct evidence of the suppression of $\langle \tau^x \rangle$. The pseudospin order parameter can be written as $\langle \tau^x \rangle = \frac{n_S - n_{AS}}{n_S + n_{AS}}$, where n_S and n_{AS} are expectation values of electron densities in symmetric and antisymmetric levels, respectively. The reduced pseudospin order reported here thus implies that a new highly correlated incompressible fluid at $\nu = 1$ is formed by mixing states with both symmetric and antisymmetric electrons despite the large value of the tunneling gap.

Experiments were carried out in two nominally symmetric modulation-doped $Al_{0.1}Ga_{0.9}As$ double quantum wells grown by molecular beam epitaxy and having total electron densities $n \sim 1.1 - 1.2 \times 10^{11}$ cm^{-2}. Resonant inelastic light scattering was performed on samples in a dilution refrigerator with a base temperature of ~50 mK and with the tilted angle geometry shown in Fig. 1(a). Dye or titanium-sapphire lasers were tuned to the fundamental optical gap of the double quantum well. Figure 1(b) displays light scattering spectra of tunneling excitations that illustrate the determination of Δ_{SAS} at $B = 0$. The spectra show collective modes in charge and spin, and also the single-particle excitation (SPE) peak [18]. In the studied samples static exchange and correlation corrections at $B = 0$ are small due to similar populations and density probability profiles of the symmetric and antisymmetric subbands [19,20]. We find that local density approximation estimates of the tunneling gap differ from Hartree Δ_{SAS} values and from the measured SPE position by less than 10% [21]. We therefore use the measured SPE energy as the tunneling gap Δ_{SAS}. At the $\nu = 1$ incompressible quantum Hall states, the values of $\frac{\Delta_{SAS}}{E_c}$ for the two samples are ~ 0.036 and 0.06 ($\Delta_{SAS} = 0.36$ meV and 0.58 meV; $E_c = \frac{e^2}{\varepsilon l_B}$ is the average Coulomb interaction energy per electron, and ε is the static dielectric constant), with $\frac{d}{l_B} \sim$ 2.2 and 2, respectively.

The spectrum of low-lying excitations of the sample with $\Delta_{SAS} = 0.36$ meV at the lowest tilt angle $\theta = 5°$ and $\nu = 1$ is shown in Fig. 1(c). Two peaks labeled SW and SF are observed. The SW peak is the long wavelength spin wave that occurs at E_Z as required by the Larmor theorem. The higher energy peak, labeled SF, is also due to spin excitations because it displays light scattering selection rules identical to the SW peak. We assign the SF feature to the long wavelength spin-flip excitation across the tunneling gap. To highlight the identification of the two peaks, Fig. 2(a) reports the evolution of the SF and SW peaks as a function of angle. The data show that the SW energy follows the Zeeman gap E_z as the total magnetic field is increased and that the SF energy approaches E_z as Δ_{SAS} is reduced by the in-plane component of magnetic field $B_\parallel$ [22].

To evaluate the impact of these results we recall again that in TDHFA $\delta E_\tau = \Delta_{SAS}$ [16]. At $\theta = 5°$, where $B_\parallel$ is quite small and finite angle corrections are negligible, we find $\delta E_\tau = 0.13 \pm 0.01$ meV, much smaller than the value of $\Delta_{SAS} \sim 0.36$ meV determined from the spectra in Fig. 1(b). This result uncovers a major breakdown of the TDHFA predictions, in particular, that the state has full pseudospin polarization. It is extremely important that the bilayers at $\nu = 1$ continue to display well-defined magneto-transport signatures characteristic of a QH incompressible fluid [13,23]. The implication is that the emergent highly correlated fluid revealed by the light scattering measurements does not significantly change electrical conduction in the $\nu = 1$ QH state at low temperatures.

To gain more quantitative insights into the effect of correlations and pseudospin reduction on spin modes we can use the pseudospin language introduced above and the coupled spin-pseudospin Hamiltonian $\widehat{\mathcal{H}}$ derived in Ref. [24]. This provides a framework to analyze the dynamics of electron spins and pseudospins in incompressible bilayers at $\nu = 1$, where in-plane fluctuations of total charge density can be neglected. $\widehat{\mathcal{H}}$ is written in terms of

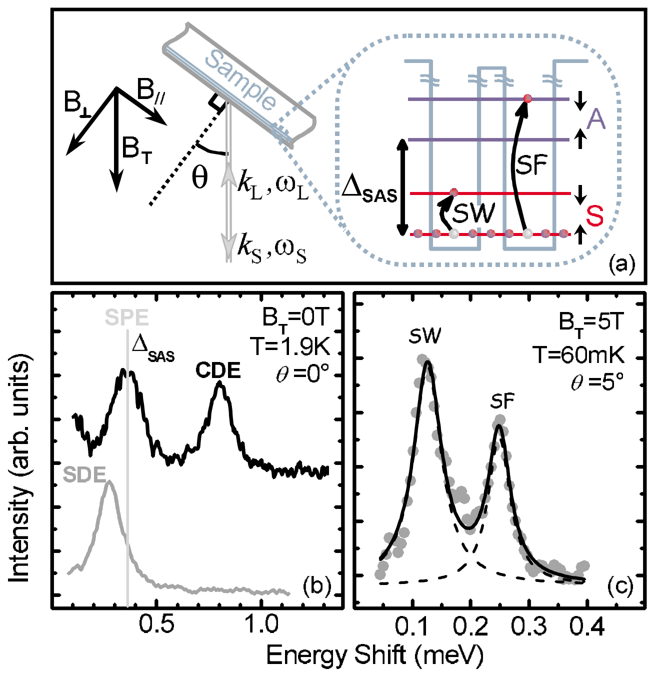

FIG. 1 (color). (a) Schematic representation of the backscattering geometry and of the double quantum well in the single-particle configuration at $\nu = 1$. Transitions in the SW and SF modes are indicated by curved vertical arrows; short vertical arrows indicate the orientations of spin, S and A label the symmetric and antisymmetric levels separated by the tunneling gap Δ_{SAS}. $k_{L(S)}$ and $\omega_{L(S)}$ indicate the wave vector and frequency of incident (scattered) light. B_T, $B_\perp$, and $B_\parallel$ are the total magnetic field and its components perpendicular and parallel to the plane of the sample. θ is the tilt angle. (b) Polarized (black curve) and cross-polarized (gray curve) inelastic light scattering spectra at $B_T = 0$ and normal incidence. SDE and CDE label the peak due to spin and charge density excitations, respectively, and SPE the single-particle excitation at $\sim\Delta_{SAS}$. (c) Resonant inelastic light scattering spectrum of SW and SF excitations at $\nu = 1$ after conventional subtraction of the background due to the laser and main luminescence. Solid and dashed lines show the fit with two Lorentzian functions.

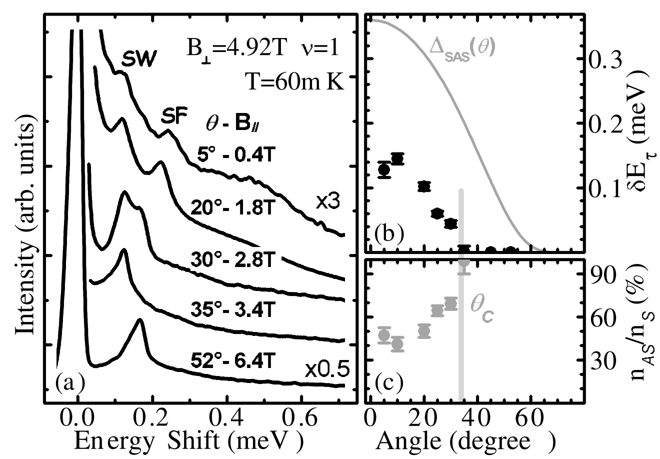

FIG. 2. (a) Resonant inelastic light scattering spectra showing SW and SF excitations as a function of tilt angle θ at $\nu = 1$. Values of the in-plane magnetic field $B_\parallel$ are also shown. At $\theta \geq \theta_c \approx 35°$ only one peak identified as the SW is observed. (b) Angular dependence of the energy difference δE_τ between the SW and SF peaks (dots). The gray curve is the predicted angular dependence of the tunneling gap evaluated in Ref. [22]. (c) Angular dependence of the emergent fraction of electrons in the excited state $\frac{n_{AS}}{n_S}$ (n_S and n_{AS} are the average electron densities in the lowest and excited levels, respectively).

spin and pseudospin operators ($\mathbf{S}_i$ and $\mathbf{T}_i = \frac{1}{2}\boldsymbol{\tau}_i$ respectively) acting on states belonging to a complete set of localized orbital wave functions $\{i\}$ in the lowest Landau level [24]. It includes all relevant interactions and coupling for spin and pseudospin channels.

In order to derive the energies of long-wavelength SW and SF we note that these excitations correspond to in-phase and out-of-phase spin modes in the two layers, respectively. The associated states can then be constructed by using projection operators into the two layers $\frac{1}{2} \pm T_i^z$ and the spin-lowering operator S_i^-. Following this procedure the associated states are written as

$$|\Psi_{\mathrm{SW}}\rangle = N^{-1/2}\sum_i S_i^- |\Psi_0\rangle, \tag{1}$$

$$|\Psi_{\mathrm{SF}}\rangle = N^{-1/2}\sum_i \tau_i^z S_i^- |\Psi_0\rangle, \tag{2}$$

where N is the total number of electrons, and $|\Psi_0\rangle$ is the ferromagnetic, fully spin polarized (at zero temperature) QH ground state with any degree of pseudospin polarization [24]. Using the Hamiltonian $\widehat{\mathcal{H}}$, it is possible to write the energy difference δE_τ between SF and SW as

$$\delta E_\tau = \langle \Psi_{\mathrm{SF}}|\widehat{\mathcal{H}}|\Psi_{\mathrm{SF}}\rangle - \langle \Psi_{\mathrm{SW}}|\widehat{\mathcal{H}}|\Psi_{\mathrm{SW}}\rangle = \Delta_{\mathrm{SAS}}\langle \tau^x\rangle, \tag{3}$$

where the last result follows a straightforward calculation. Equation (3) remarks that a reduced pseudospin order parameter determines the tunneling SF energy. This conclusion is likely to remain valid even when $B_\parallel \neq 0$. In this case phase differences are introduced between the wave functions in the two layers; their impact can be described in terms of a tumbling in the x-y plane of the pseudomagnetic field associated to the tunneling gap. In the commensurate phase expected at relatively low $B_\parallel$ [13] pseudospins are thus aligned in different directions as a function of in-plane position [3]. In Eq. (3), therefore, $\langle \tau^x\rangle$ must be replaced by the average of $\boldsymbol{\tau}_i$ in the direction of this pseudomagnetic field. We call this quantity $\langle \tau(\theta)\rangle$. In the mean-field configuration, neglecting correlation effects, we continue to have $\langle \tau(\theta)\rangle = 1$ and $\delta E_\tau = \Delta_{\mathrm{SAS}}$.

The prediction reported in Eq. (3) allows us to link the measured SF-SW splitting δE_τ with a reduced $\langle \tau^x\rangle$ beyond TDHFA. We stress that by reducing the pseudospin order electrons can efficiently optimize their interlayer and intralayer correlations by decreasing the charging energy associated to fluctuations in layer occupation (fluctuation of the pseudospin in the z direction). For $\theta = 5\,^\circ$, $\langle \tau^x\rangle = \frac{n_S - n_{AS}}{n_S + n_{AS}} \approx 0.36$, showing that the ground state is characterized by a high expectation value $\frac{n_{AS}}{n_S + n_{AS}} \approx 0.32$ (or 32%) for the fraction of electron density in the antisymmetric level. It is therefore tempting to describe this QH incompressible state in terms of bound electron-hole pairs across the tunneling gap making a particle-hole transformation in the lowest symmetric Landau level [25,26].

It is surprising to observe this major loss of pseudospin ferromagnetic order at sizable values of Δ_{SAS}. Correlations in electron bilayers at $\nu = 1$ were evaluated within models that consider the impact of quantum fluctuations from low-lying tunneling modes such as magnetorotons [24,27–29]. In these theories the order parameter is suppressed because of these in-plane pseudospin quantum fluctuations, leading eventually to the incompressible-compressible phase transition associated with the disappearing of the QH state. Because of the difference between intralayer and interlayer interactions, in fact, τ^x is not a good quantum number and it fluctuates in the true many-body ground state. In agreement with our findings, these calculations suggest that significant loss of pseudospin polarization leading to values of $\langle \tau^x\rangle \approx 0.2$ could occur at high $\frac{\Delta_{\mathrm{SAS}}}{E_c}$ values comparable to the ones of our sample.

The suppression of τ^x is influenced by Δ_{SAS}. For the sample with larger $\Delta_{\mathrm{SAS}} = 0.58$ meV the extrapolated value at zero angle yields $\frac{n_{AS}}{n_S + n_{AS}} \sim 17\%$ ($\langle \tau^x\rangle \sim 0.65$). We have also reduced Δ_{SAS} by increasing $B_\parallel$ at larger tilt angles θ [22]. Figures 2(a) and 2(b) shows that with increasing angles δE_τ shrinks until it collapses at a "critical" angle $\theta_c \sim 35°$. Points in Fig. 2(b) are the SF-SW splitting δE_τ as a function of angle. Figure 2(c) shows the ratio $\frac{n_{AS}}{n_S} = \frac{1 - \langle \tau(\theta)\rangle}{1 + \langle \tau(\theta)\rangle}$ determined from the measured splitting $\delta E_\tau(\theta)$ and from $\delta E_\tau(\theta) = \Delta_{\mathrm{SAS}}(\theta)\langle \tau(\theta)\rangle$, where $\Delta_{\mathrm{SAS}}(\theta)$ includes the single-particle angular dependence derived in Ref. [22] and plotted in Fig. 2(b). Within this framework $\frac{n_{AS}}{n_S} \rightarrow 1$ in a continuous way ($\langle \tau(\theta)\rangle \rightarrow 0$) when $\theta \rightarrow \theta_c$. It is possible, however, that close to the collapse of δE_τ, higher-order corrections to Eq. (3) will affect the precise determination of the pseudospin polarization.

The data, however, reveal a strong increase of correlations as $\Delta_{\mathrm{SAS}}(\theta)$ diminishes. The value of $\Delta_{\mathrm{SAS}}(\theta_c)$ is consistent with the phase transformation to the compressible phase [3,4,13]. Given the values in our samples of $\frac{\Delta_{\mathrm{SAS}}}{E_c}$ and $\frac{d}{l_B}$ the commensurate-incommensurate phase transition observed in Ref. [13] is not expected to occur here for $\theta \leq \theta_c$. Additionally, it would produce a nonobserved abrupt reduction of $\langle \tau_x(\theta)\rangle$. It is possible, however, that the decrease of exchange energy associated to the effect of $B_\parallel$ could contribute to increase quantum fluctuations, further reducing the order parameter.

The remaining peak at $\theta > \theta_c$ is assigned to the long wavelength SW since it follows the Zeeman gap. We have not been able to observe SF modes for angles $\theta > \theta_c$; the SF merges with the SW at $\theta = \theta_c$, as indicated by their gradual approaching without any reduction in intensity below θ_c. Additional features characterize the spectra shown in Fig. 2(a). A broad peak is observed below the critical angle (at $5\,^\circ$ is centered at ~0.45 meV) and its energy decreases at larger tilt angles. The SW modes dis-

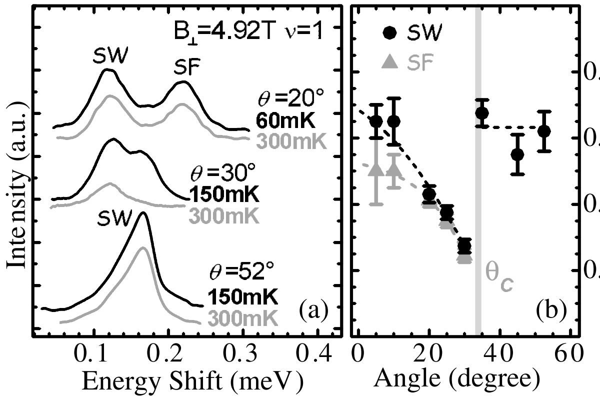

FIG. 3. (a) Resonant inelastic light scattering spectra of SW and SF excitations at $\nu = 1$ and different values of temperature and tilt angles θ, after conventional background subtraction. (b) Angular dependence of the temperature $T_{1/2}$. $T_{1/2}$ is defined as the temperature at which the intensity of the SW (black dots) and SF (gray triangles) peaks is reduced to half of the lowest temperature value. The dashed lines are guides for the eyes. θ_c is the angle at which the SW and SF peaks merge.

play markedly asymmetric spectral shapes above θ_c. These features, which seem related to a phase transition near the critical angle, are presently not understood and remain to be considered in further work.

Evidence of a phase transition at the collapse of pseudospin order at θ_c is also seen in the marked temperature dependence of SF and SW modes, as shown in Fig. 3(a). For typical spectra with $\theta < \theta_c$ the intensities of the peaks have a larger temperature dependence than the SW mode for $\theta > \theta_c$. To describe this behavior we introduce a temperature $T_{1/2}$ at which the peak intensity is half of its value at the lowest temperature. Figure 3(b) displays $T_{1/2}$ versus angle for both SW (black circles) and SF (gray triangles). It can be seen that at θ_c there is an abrupt change in $T_{1/2}$ that accompanies the disappearance of the SF mode. The strong decrease of $T_{1/2}$ for the SW for $\theta < \theta_c$ suggests a rich spin dynamics in the ferromagnetic $\nu = 1$ state that may be linked to spin-pseudospin coupling.

In conclusion, we determined major reductions of pseudospin ferromagnetic order due to correlations in the incompressible phase of coupled bilayers at $\nu = 1$ by inelastic light scattering measurements of spin excitations. The results are surprising by revealing large correlation effects in a range of $\frac{\Delta_{SAS}}{E_c}$ and $\frac{d}{l_B}$ values where magnetotransport results find well-defined QH signatures. Further studies, including absorption across the fundamental gap between valence and conduction bands [30,31], should clarify if the correlated state with reduction of pseudospin order displays the properties of an electron-hole excitonic quantum fluid.

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[1] S. Das Sarma, S. Sachdev, and L. Zheng, Phys. Rev. Lett. **79**, 917 (1997).
[2] V. Pellegrini *et al.*, Science **281**, 799 (1998).
[3] Kun Yang *et al.*, Phys. Rev. Lett. **72**, 732 (1994).
[4] S. M. Girvin and A. H. MacDonald, in *Perspectives in Quantum Hall Effect*, edited by S. Das Sarma and A. Pinczuk (Wiley, New York, 1997), Chap. 5, p. 161.
[5] H. A. Fertig, Phys. Rev. B **40**, 1087 (1989).
[6] X. G. Wen and A. Zee, Phys. Rev. Lett. **69**, 1811 (1992).
[7] M. Kellogg *et al.*, Phys. Rev. Lett. **93**, 036801 (2004).
[8] E. Tutuc, M. Shayegan, and D. A. Huse, Phys. Rev. Lett. **93**, 036802 (2004).
[9] J. Eisenstein, Science **305**, 950 (2004), and references therein.
[10] A. Stern *et al.*, Phys. Rev. Lett. **86**, 1829 (2001).
[11] M. M. Fogler and F. Wilczek, Phys. Rev. Lett. **86**, 1833 (2001).
[12] J. Schliemann, S. M. Girvin, and A. H. MacDonald, Phys. Rev. Lett. **86**, 1849 (2001).
[13] S. Q. Murphy *et al.*, Phys. Rev. Lett. **72**, 728 (1994).
[14] G. S. Boebinger *et al.*, Phys. Rev. Lett. **64**, 1793 (1990).
[15] S. Luin *et al.*, Phys. Rev. Lett. **90**, 236802 (2003).
[16] L. Brey, Phys. Rev. Lett. **65**, 903 (1990).
[17] A. H. MacDonald, P. M. Platzman, and G. S. Boebinger, Phys. Rev. Lett. **65**, 775 (1990).
[18] A. S. Plaut *et al.*, Phys. Rev. B **55**, 9282 (1997).
[19] P. I. Tamborenea and S. Das Sarma, Phys. Rev. B **49**, R16821 (1994).
[20] P. G. Bolcatto and C. R. Proetto, Phys. Rev. Lett. **85**, 1734 (2000).
[21] This uncertainty also includes different conduction-to-valence band-offset ratios ranging from 0.6 to 0.7.
[22] J. Hu and A. H. MacDonald, Phys. Rev. B **46**, 12554 (1992).
[23] Magneto-transport experiments down to 300 mK show the manifestations of a $\nu = 1$ quantum Hall state at angles below 30 °. Detailed temperature dependence studies are hindered by parallel conduction.
[24] A. A. Burkov and A. H. MacDonald, Phys. Rev. B **66**, 115320 (2002).
[25] S. M. Girvin, Phys. Rev. B **29**, R6012 (1984).
[26] A. H. MacDonald and E. H. Rezayi, Phys. Rev. B **42**, R3224 (1990).
[27] T. Nakajima and H. Aoki, Phys. Rev. B **56**, R15549 (1997).
[28] Y. N. Joglekar and A. H. MacDonald, Phys. Rev. B **64**, 155315 (2001).
[29] K. Moon, Phys. Rev. Lett. **78**, 3741 (1997).
[30] M. J. Manfra *et al.*, Physica E (Amsterdam) **6**, 590 (2000).
[31] R. Côté, Phys. Rev. B **64**, 205304 (2001).