

## Spectroscopy of electrons under extreme conditions: unexpected states in quantum hall bilayers

The properties of many electrons confined in low-dimensional materials can be very different from the ones than a single-particle, independent electron paradigm can predict. The observation of these unusual collective quantum fluids belongs to the frontiers of modern condensed-matter science. We used a resonant inelastic light scattering technique to study peculiar spin oscillations in an electron fluid embedded in artificial semiconductor structures; we demonstrate that these excitations contain the fingerprint of the exotic nature of the correlated state.

Electron bilayers in semiconductor quantum structures embedded in a quantizing perpendicular magnetic field ( $B_z$ ) are contemporary realizations of collective systems where bizarre quantum phases may appear [1-3].

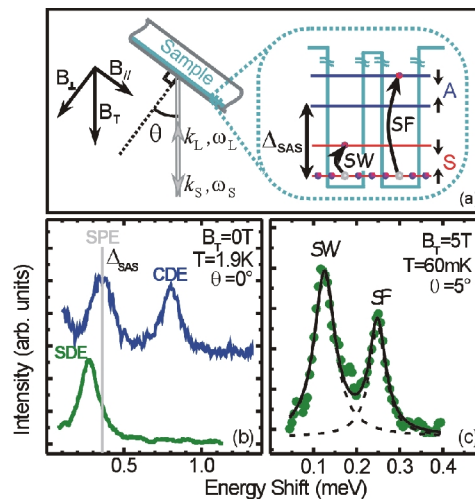
In particular, at Landau level filling factor  $n=1$  excitonic-superfluidity can occur when there is no tunneling between the layers [1]. This state is driven by unique interplays between intra- and inter-layer Coulomb interactions and have been the subject of large research efforts in last years [4].

A paradigmatic configuration occurs at  $n=1$  when a tunneling gap  $D_{SAS}$  splits the single-particle levels in their symmetric and anti-symmetric linear combinations. The configuration in which all the electrons precisely fill the lowest spin-up symmetric Landau level has full spin and pseudospin ferromagnetic order, where pseudospin ( $\mathbf{T}=\frac{1}{2}\mathbf{t}$ ) is a quantum operator describing layer occupation: electrons in the left (right) quantum well have pseudospin along the  $+z$  ( $-z$ ) direction normal to the plane, and electrons in the symmetric (anti-symmetric) states have pseudospin aligned along  $+x$  ( $x$ ). This is a mean-field state that has pseudospin order parameter  $\langle t_x \rangle$  equal to 1. It is the predicted ground state within the time-dependent Hartree-Fock approximation (TDHFA) that has been extensively employed to interpret bilayer experiments [5-7].

Here we provide direct evidence, from inelastic light scattering measurements of low-lying spin and pseudospin excitations, that the pseudospin order of this paradigm is lost. In the experiments reviewed here resonant inelastic light scattering is

employed to probe low-lying spin and pseudospin excitations. The suppression of the pseudospin order parameter manifests a new many-particle quantum state that can be interpreted in terms of formation of a highly correlated fluid of electron-hole pairs across the tunneling gap. The emergence of this fluid is evidence of unexpected major impact of electron correlations in the presence of finite tunneling.

The bilayers are confined in AlGaAs/GaAs double quantum wells, whose potential is schematically shown in Figure 1a with the geometry of the experiments and the single-particle-like electronic configuration at  $n=1$ . Figure 1b displays light scattering spectra of tunneling excitations that illustrate the determination of  $D_{SAS}$  at  $B=0$ . The spectra show collective modes in charge and spin, CDE and SDE, and also the single-particle transition SPE that measures  $D_{SAS}$  ( $D_{SAS}=0.36\text{meV}$  at  $B=0$  in the case of this sample) [9]. The spectrum of low-lying



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

excitations at the lowest tilt angle  $q=5^\circ$  and  $n=1$  is shown in Fig. 1c. The peak SW is due to the long wavelength spin-wave (SW) associated to excitations across the Zeeman gap  $E_z$  in which only the spin degree of freedom changes; this is centered at  $E_z$  as required by 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. We assign the SF feature to the long wavelength spin-flip (SF) excitation with simultaneous changes in pseudospin, due to transitions across the tunneling gap, and spin orientation. The identification is supported by the angular dependence displayed in Fig. 2a, showing that the SF energy approaches the SW energy (and thus  $E_z$ ) as  $D_{\text{SAS}}$  is reduced by the in-plane component of magnetic field  $B_{\parallel}$  [10]. The transitions that build SW and SF excitation modes are depicted in Fig. 1a.

To highlight the significance of these results it is necessary to recall that in TDHFA  $dE_t = D_{\text{SAS}}$  [6]. At the angle of  $q=5^\circ$ , where  $B_{\parallel}$  is small and finite angle corrections are negligible, we find that  $dE_t = 0.13 \pm 0.01 \text{ meV}$ , much smaller than the value of  $D_{\text{SAS}} = 0.36 \text{ meV}$ . This result uncovers a major breakdown of the TDHFA predictions that the state has full pseudospin polarization. It is extremely important that the bilayers at  $n=1$  continue to display well-defined magneto-transport signatures characteristics of a QH incompressible fluid [7], as also observed by us in both longitudinal and transverse resistivities. The implication is that the

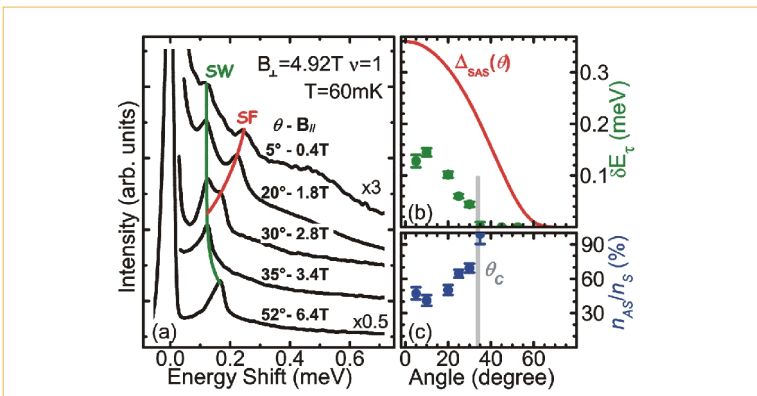
emergent highly-correlated fluid revealed by the light scattering measurements does not significantly change electrical conduction in the  $n=1$  QH state at low temperatures.

To gain more quantitative insights into the effect of correlations and pseudospin reduction on spin modes we used the pseudospin language introduced above and the coupled spin-pseudospin bilayer Hamiltonian derived in Ref. 11. By calculating, beyond TDHFA, the energies of the spin flip and the spin wave excitations on a ferromagnetic, fully spin polarized (at zero temperature) QH ground state with any degree of pseudospin polarization, [11] we found that in bilayers  $dE_t = \langle t_x \rangle \cdot D_{\text{SAS}}$  [12]. This result remarks that a reduced pseudospin order parameter determines the tunneling SF energy. This conclusion is likely to remain valid even when  $B_{\parallel} \neq 0$ , with a careful definition of the order parameter ( $\langle t(q) \rangle$ ) [12].

We stress that by reducing the pseudospin order electrons can efficiently optimize their inter- and intra-layer correlations by decreasing the charging energy associated to fluctuations in layer occupation (fluctuation of the pseudospin in the z-direction), increasing conversely in-plane pseudospin quantum fluctuations (pseudospin-squeezed state). The in-plane quantum fluctuations eventually lead to the incompressible-compressible phase transition associated to the disappearing of the QH state observed in this kind of systems [11-13]. For  $q=5^\circ$ ,  $\langle t_x \rangle = (n_s - n_{\text{AS}})/(n_s + n_{\text{AS}}) \gg 0.36$ , showing that the new state with reduced pseudospin order is characterized by an high expectation value  $n_{\text{AS}}/(n_s + n_{\text{AS}}) \gg 0.32$  (or 32%) for the fraction of the total electron density into the anti-symmetric 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 [14].

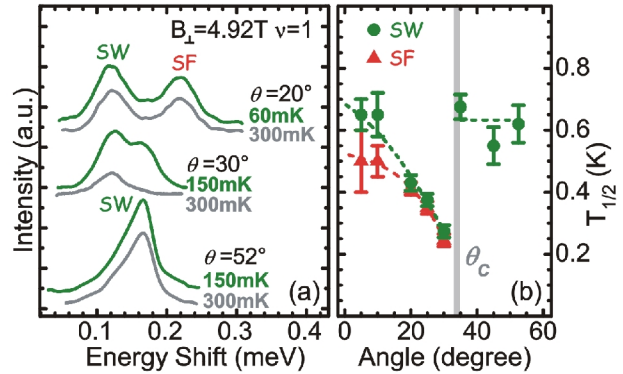
The suppression of  $\langle t_x \rangle$  is influenced by  $D_{\text{SAS}}$ . For a sample with larger  $D_{\text{SAS}} = 0.58 \text{ meV}$  the extrapolated value at

Fig. 2 (A) Resonant inelastic light scattering spectra showing spin wave (SW) and spin flip (SF) excitations as a function of tilt angle  $q$  at  $n=1$ . Values of the in-plane magnetic field  $B_{\parallel}$  are also shown. At  $q > q_c \sim 35^\circ$  only one peak identified as the SW is observed. (b) Angular dependence of the energy difference  $dE_t$  between the SW and SF peaks (dots). The red curve is the predicted angular dependence of the tunneling gap evaluated in Ref. 10. (c) Angular dependence of the emergent fraction of electrons in the excited state  $n_{\text{AS}}/n_s$ .



zero angle yields  $n_{AS}/(n_s+n_{AS})\sim 17\%$  ( $\langle t_x \rangle \sim 0.65$ ). We have also reduced  $D_{SAS}$  by increasing  $B_{\parallel}$  at larger tilt angles  $q$  [10]. Figures 2a and 2b show that, with increasing angles,  $dE_t$  shrinks until it collapses at a 'critical' angle  $q_c \sim 35^\circ$ . Points in Fig. 2b are the SF-SW splitting  $dE_t$  as a function of angle. We have not been able to observe SF modes for angles  $q > q_c$ . Figure 2c shows the ratio  $n_{AS}/n_s = (1 - \langle t(q) \rangle) / (1 + \langle t(q) \rangle)$  determined from the measured splitting  $dE_t(q)$  and from  $dE_t(q) = D_{SAS}(q) \cdot \langle t(q) \rangle$ , where  $D_{SAS}(q)$  includes the single-particle angular dependence derived in Ref. 10 and plotted in Fig. 2b. Within this framework  $n_{AS}/n_s \approx 1$  in a continuous way ( $\langle t(q) \rangle \approx 0$ ) when  $q \approx q_c$ . The value of  $D_{SAS}(q_c)$  is consistent with the phase transformation to the compressible phase [1,3,7].

Evidence of a phase transition at the collapse of pseudospin order at  $q_c$  is also seen in the marked temperature dependence of SF and SW modes, as shown in Fig. 3. For typical spectra with  $q < q_c$  the intensities of the peaks have a large temperature dependence. The temperature dependence is slower for the asymmetric SW modes measured at  $q > q_c$ . 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 3b displays  $T_{1/2}$  versus angle for both SW (green dots) and SF (red triangles). It can be seen that at  $q_c$  there is an abrupt change in  $T_{1/2}$  that is accompanied by the disappearance of the SF mode. The strong decrease of  $T_{1/2}$  for the SW for  $q < q_c$  suggests a rich spin dynamics in the ferromagnetic  $n=1$  state that may be linked to spin-pseudospin coupling.

#### Collaborations

This activity is carried out in collaboration with Aron Pinczuk (Columbia University, New York City, NY, USA) and Bell Laboratories, Murray Hill, NJ, USA) and Loren Pfeiffer (Bell Laboratories).

Fig. 3 (a) Resonant inelastic light scattering spectra of spin wave (SW) and spin flip (SF) excitations at  $n=1$  and different values of temperature and tilt angles  $q$ , 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 (green dots) and SF (red triangles) peaks is reduced to half of the lowest temperature value. The dashed lines are guides for the eyes.  $q_c$  is the angle at which the SW and SF peak merge.

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