Collapse of Spin Excitations in Quantum Hall States of Coupled Electron Double Layers

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Remarkable softenings of long wavelength intersubband spin excitations of dilute electron double layers are observed at even integer quantum Hall states. These excitations in coupled GaAs double quantum wells were probed by resonant inelastic light scattering. Their softening is attributed to enhanced exchange vertex corrections (excitonic binding) in the quantum Hall states. The collapse of the spin-density mode with $\delta S_z = 0$ to an energy close to the Zeeman splitting suggests the existence of unstable spin-flip intersubband excitations with $\delta S_z = 1$. [S0031-9007(96)02143-6]

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Studies of dispersive collective excitations in the integer and fractional quantum Hall regimes offer unique insights into fundamental effects and new phenomena in electron systems of reduced dimensionality. The presence of magnetoroton minima in the dispersions of charge-density excitations (CDE) is one of many characteristic manifestations of electron-electron interactions [1,2]. The minima occur at wave vectors $q \simeq 1/l_0$, where $l_0 = (\hbar c/eB)^{1/2}$ is the magnetic length and B is the magnetic field. The roton minima result from vertex corrections due to exchange Coulomb interactions in the neutral quasiparticle-quasihole pairs of the excitations. These interactions are excitonic effects that reduce the collective mode energies.

In electron double layers, the introduction of the additional degree of freedom associated with layer index creates remarkable new electron correlation effects. For example, at filling factor $\nu = 1$ electron bilayers show intriguing and rich phase diagrams determined by the magnitude of the symmetric-antisymmetric gap Δ_{SAS}, the distance d between the layers, and the in-plane magnetic field [3–5]. In magnetotransport experiments, these quantum phase transitions appear as the suppression of the $\nu = 1$ state [4,6]. Several theoretical works have linked the disappearance of the quantum Hall state to a vertex-correction-driven collapse of the symmetric to antisymmetric intersubband CDE energy at wave vectors $q \simeq 1/l_0$ [7]. Although not yet directly observed, the predicted CDE instability highlights the major role of excitonic effects on the dispersive collective excitations of the electron gas in the quantum Hall regime.

Inelastic light scattering experiments have previously shown that vertex corrections also have a major impact on the intersubband spin-density excitation (SDE) modes of electron bilayers in double quantum wells (DQW) [8]. For such excitations a different class of vertex-correction-driven quantum phase transitions has been predicted to occur at zero magnetic field [9]. Here the signature of the instability is the collapse of the energy of the long wavelength ($q = 0$) intersubband SDE mode of the symmetric to antisymmetric transitions. While this *zero-field* instability is not observed in GaAs DQW [10], inelastic light scattering measurements in *perpendicular magnetic field* have uncovered marked softenings of the intersubband SDE at even ν quantum Hall states [11]. A recent Hartree-Fock calculation links these softenings with a quantum phase transition to a state with antiferromagnetic spin correlations between the two layers [12].

This Letter reports inelastic light scattering experiments in coupled GaAs DQW subjected to perpendicular magnetic fields. The measurements reveal a dramatic softening of the long wavelength intersubband spin excitations of the dilute electron double layers in the DQW. The anomalies are observed in *spin-unpolarized* even integer quantum Hall states, where we find that the collective mode energies can be as low as *one-tenth* of the $B = 0$ values. This drastic drop in the energy of the sharp inelastic light scattering peak of intersubband SDE reveals a collapse of the mode to an energy close to that of the Zeeman splitting of the electron states.

These anomalous SDE modes are observed in double layers that have relatively small tunneling gaps ($\Delta_{SAS} \leq$ 1 meV), such that at $B = 0$ the electrons populate antisymmetric as well as symmetric states. Here the effective electron density participating in the intersubband excitations is reduced to (n_S-n_{AS}), the difference in the populations of symmetric and antisymmetric states. This available phase-space factor determines the strength of vertex corrections [8]. The softening of the intersubband spin excitations is thus caused by the enhancement in the vertex corrections associated with magnetic field induced changes in available phase space for the excitations. This is easily seen from the expression

$$n_S - n_{AS} = n \frac{\nu_S - \nu_{AS}}{\nu}, \tag{1}$$

where $n = n_S + n_{AS}$ is the total electron density. ν_S and ν_{AS} are the filling factors of the symmetric and antisymmetric states ($\nu = \nu_S + \nu_{AS}$). Equation (1) shows that a maximum in available phase space, and therefore in the vertex corrections, is attained at $\nu = 2$, when all the electrons participate in the intersubband excitations ($\nu_S = 2$ and $\nu_{AS} = 0$).

To further highlight the impact of our experiments, we recall that when the two orientations of spin are equally populated, spin excitations at even integer ν are triplets. These states are classified by changes in spin angular momenta along the magnetic field $\delta S_z = 0, \pm 1$, as shown in Fig. 1(a). The SDE mode has $\delta S_z = 0$ and energy ω_{SDE}. The other two excitations are spin-flip modes with energies $\omega_\pm \simeq \omega_{SDE} - E_z \delta S_z$, evaluated within the Hartree-Fock framework, where $E_z = g\mu_B B$ is the bare Zeeman energy [1]. Consequently, observations of intersubband SDE modes at energies close to E_z suggest the existence of lower lying intersubband spin-flip excitations ($\delta S_z = 1$) of vanishingly small energy. These results offer spectroscopic evidence that exchange vertex corrections drive the pronounced softenings that may trigger instabilities in 2D electron layers.

Inelastic light scattering spectra were obtained from several GaAs DQW samples in a backscattering geometry with light propagating parallel to the magnetic field, i.e., perpendicular to the layers. For these measurements the emission of a dye laser was tuned to the fundamental optical transitions of the GaAs DQW. Incident power densities were kept below 10^{-4} W/cm^{-2}, and spectra were recorded with multichannel detection at a spectral resolution of 0.02 meV. The samples were mounted in a ^{3}He/^{4}He dilution refrigerator with optical windows. Accessible temperatures were in the range $0.2 \leq T \leq 1.4$ K. We focus on results from two modulation doped coupled DQW grown by molecular beam epitaxy. They consist of two nominally identical 180-Å-thick GaAs quantum wells separated by a 79 Å wide $Al_{0.1}Ga_{0.9}As$ undoped barrier, with electron concentrations of $n = 6.2 \times 10^{10}$ cm^{-2} and $n = 9.4 \times 10^{10}$ cm^{-2} and low-temperature mobilities close to 10^6 cm^2/V s. At $B = 0$ these double layers have $\Delta_{SAS} \simeq 0.7$ meV and $\omega_{SDE} \simeq 0.45$ meV.

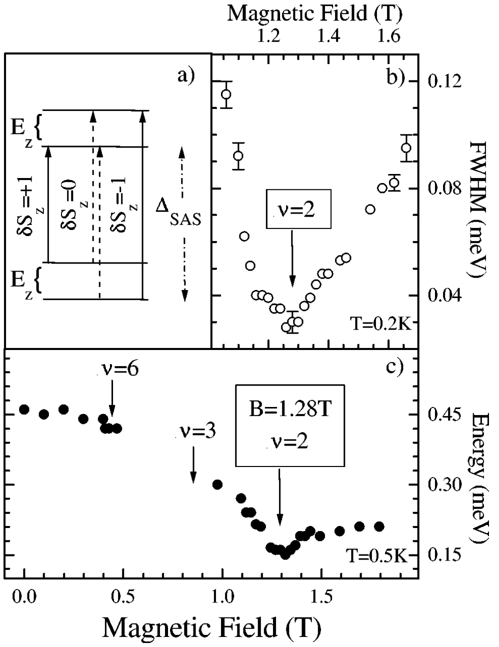

FIG. 1. (a) Schematic representation of the intersubband spin excitations at even integer quantum Hall states. The spin-density excitation (SDE) with $\delta S_z = 0$ results from the linear combination of the two transitions denoted by dashed lines. Solid lines correspond to spin-flip excitations ($\delta S_z = \pm 1$). E_z is the Zeeman splitting and Δ_{SAS} the symmetric to antisymmetric energy gap. (b) Plot of the full width half maximum (FWHM) of the SDE peak as a function of magnetic field for the lower density sample at $T = 0.2$ K. (c) Energy position of the $q = 0$ spin-density excitation (SDE) as a function of magnetic field for the lower electron density sample at $T = 0.5$ K.

In spin-unpolarized states at even values of ν the SDE collective modes are built from linear combinations of the two transitions with $\delta S_z = 0$ shown in Fig. 1(a). Light scattering polarization selection rules derived for bulk semiconductors also apply to 2D layers in semiconductor quantum structures [13,14]. They predict that SDE modes are active in the backscattering spectra with orthogonal incident and scattered light polarizations as measured in this experiment. For spin-flip transitions, also shown in Fig. 1(a), these selection rules require that one of the photons has to be polarized along the magnetic field. Spin-flip excitations are thus forbidden in the backscattering configuration employed here.

Figures 1(b) and 1(c) show results obtained in the lower electron density sample. Figure 1(b) reveals that the width of the SDE mode has a significant minimum at $\nu = 2$. At this filling factor the FWHM = 0.03 meV is 5 times smaller than that at $B = 0$ [10]. Similar behaviors are observed at the other even quantum Hall states that we investigated. Figure 1(c) shows the field dependence of ω_{SDE}. We find small changes for fields up to $\nu \simeq 6$, and the absence of the SDE mode near $\nu = 4$. These observations are consistent with the magnetic field dependence of the available phase space given by Eq. (1). At $\nu = 6$, $(n_S - n_{AS}) = n/3$ is close to the $B = 0$ value, implying that no significant changes are expected in collective interactions. At $\nu = 4$ the intersubband excitations do not exist because $(n_S - n_{AS}) = 0$.

In Fig. 1(c), the pronounced softening of ω_{SDE} at $\nu = 2$ is evidence of the enhancement of vertex corrections when the phase space available for the excitations includes all the electrons. An even larger softening is observed in the higher electron density sample. The results in Fig. 2 compare the SDE spectra measured in the two samples at $\nu \simeq 2$. The spectrum of the higher electron density sample displays a sharp peak at $\omega_{SDE} = 0.045$ meV. This energy is *one-tenth* of the value obtained at $B = 0$. This extremely low energy is close to the E_z calculated with $g = -0.4$ [15].

We have not been able to observe intersubband spin excitations at $\nu = 2$ in higher electron density systems where $\nu = 2$ lies at fields $B \geq 2.6$ T. Magnetotransport experiments have indicated that at such higher fields the $\nu = 2$ state is spin polarized with identical spin occupation of

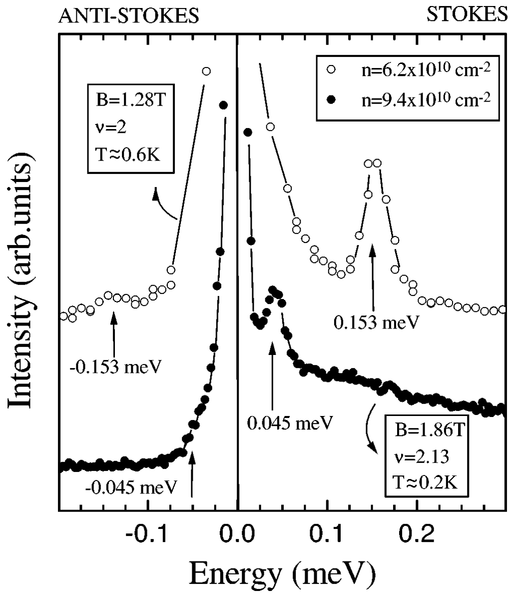

FIG. 2. Resonant inelastic light scattering spectra with orthogonal linearly polarized incident and scattered photons. The peaks correspond to the $q = 0$, $\delta S_z = 0$ spin-density excitations for the lower (open circles) and higher (filled circles) electron density samples. From the anti-Stokes peaks we deduce electron temperatures of $T \simeq 0.6$ K and $T \simeq 0.2$ K, respectively.

the two lowest symmetric and antisymmetric levels [6]. In such states intersubband excitations involve spin flips which are forbidden in our backscattering configuration. Further, we have also failed to observe the spin-flip intersubband excitations of the spin-polarized states at odd values of ν. These negative results are regarded as confirmation that the light scattering polarization selection rules apply to our experiments, and that the observed spin excitations are long wavelength intersubband SDE modes at energy ω_{SDE}.

Figure 3 displays measurements of the magnetic field and temperature dependence of ω_{SDE} in the higher electron density sample. Magnetic fields are in the range close to $\nu = 2$. The dashed line represents the Zeeman energy for $g = -0.4$. We find that for temperatures $T \geq 0.8$ K ω_{SDE} has a sharp minimum, like the ones at 1.2 and 1.4 K, at the magnetic field of $\nu \simeq 2$. At lower temperatures the minima are shallower. Figure 3 shows that at temperatures 0.6 and 0.25 K ω_{SDE} reaches its lowest values at $B = 1.92$ T ($\nu = 2.06$) and at $B = 1.86$ T ($\nu = 2.13$), respectively. Within the estimated experimental errors, these minima of ω_{SDE} come close to the Zeeman splitting indicated in Fig. 3.

The collapse of the intersubband SDE mode to energies close to Zeeman splitting implies the emergence of spin-flip excitations with $\delta S_z = 1$ at vanishingly small energies. While the observation of such excitations is beyond the reach of the present experiments, there is ample evidence of anomalous behaviors at the filling factors close to $\nu = 2$. Spectra at $T = 0.25$ K reveal a marked decrease of the SDE peak intensities for fields $B \geq 1.93$ T ($\nu \leq 2.05$), which prevents the determination of SDE energies in that range of filling factor. Surprisingly, the SDE peak recovers at higher temperatures. Figure 4 displays this behavior at $B = 1.98$ T, the field of $\nu = 2$. The inset to Fig. 4 summarizes this intriguing temperature dependence. Here the points represent the temperatures above which we recover a well-defined, sharp, ω_{SDE} peak in the spectra.

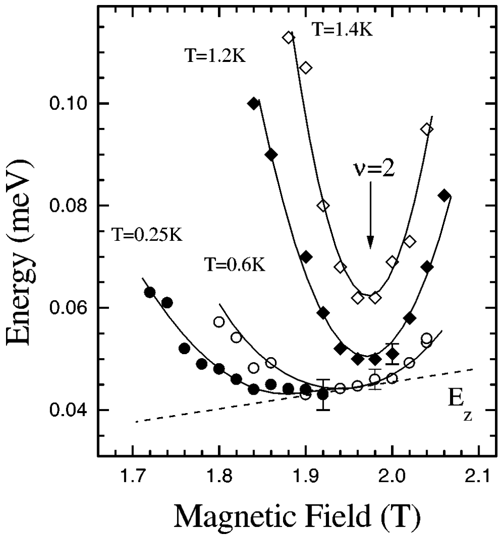

FIG. 3. Energy position of the $q = 0$, $\delta S_Z = 0$ spin-density excitations (SDE) for the higher electron density sample as a function of magnetic field and temperature. Solid lines are guides for the eyes. The dashed line represents the values of the Zeeman energy ($g = -0.4$) in the displayed range of magnetic field.

The anomalous temperature dependence of the light scattering peaks displayed in Fig. 4 is remarkable and unexpected. The striking disappearance of the SDE peak near $\nu = 2$ is due either to a reduction of its intensity or to a dramatic increase of its width or both. These results suggest that striking changes take place in the electron double layers at $\nu \simeq 2$ and $T \leq 0.5$ K. Given that the anomalies are observed when ω_{SDE} collapses to an energy close to the bare Zeeman splitting E_z, it is natural to assign them to instabilities associated with the emergence of $\delta S_z = 1$ spin-flip intersubband collective modes of vanishingly small energy. However, the reduction of the intensity of the SDE mode at low temperatures is presently not understood. It is conceivable that the collapse of the spin excitations leads to the spontaneous generation of large numbers of spin-flip excitonic pairs to form a highly correlated phase. The generation of such bound particle-hole pairs does not change the insulating behavior of the quantum Hall state and results in an emergence of a net spin polarization along the magnetic field axis. The observed reduction in the light scattering intensity of the SDE mode is

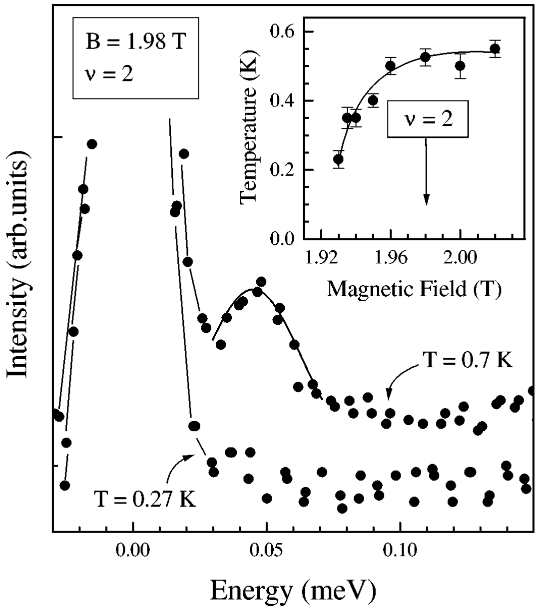

FIG. 4. Resonant inelastic light scattering spectra for the higher electron density sample at $\nu = 2$ and at two different temperatures. The peak corresponds to the $q = 0$ spin-density excitation. The inset shows the lowest value of the temperature at which the SDE peak first appears.

consistent with this hypothesis. The recovery of the light scattering intensity implies a phase transition that destroys the correlated state at higher temperature.

Another interpretation is suggested by the calculation of Zheng *et al.* [12]. The collapse of the spin-flip intersubband excitation is here associated with a quantum phase transition to a state with antiferromagnetic spin correlations between the layers. Such antiferromagnetic ordering should have in-plane spin polarization in each layer. This state of the electron double layers is expected to sustain new characteristic collective excitations. At the present time we have no evidence of such excitations, but further light scattering experiments currently in progress could shed some light on the possible existence of this novel highly correlated quantum state. Within the conceptual framework of this quantum phase transition, the striking temperature dependence displayed in Fig. 4 could be regarded as evidence of a finite temperature transition that destroys the perfect order in the canted antiferromagnetic phase [16].

In conclusion, inelastic light scattering experiments in even integer quantum Hall states of coupled electron double layers revealed that enhanced vertex corrections lead to a collapse of the energies of intersubband spin excitations. The light scattering intensities by low energy SDE modes display an anomalous temperature dependence as they soften to the Zeeman energy. Further studies are required to determine whether these anomalies are related to a quantum phase transition. We believe that these experiments reach beyond previous studies of instabilities in coupled electron double layers. Our work also demonstrates the success of inelastic light scattering methods to study Coulomb interactions and to determine the role of dispersive collective excitations in the intriguing physics of 2D electron systems in magnetic fields. Here, we have taken advantage of the fact that the relevant excitations occur at long wavelengths. Our light scattering studies will be extended to the larger wave vector regime to determine the impact of magnetoroton minima on quantum phase transitions of 2D electron systems in the quantum Hall regimes.

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