Observation of Collective Excitations of the Dilute 2D Electron System

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We report inelastic light scattering measurements of dispersive spin and charge density excitations in dilute 2D electron systems reaching densities less than $10^{10}$ cm$^{-2}$. In the quantum Hall state at $\nu = 2$, roton critical points in the spin inter–Landau level mode show a pronounced softening as $r_s$ is increased. Instead of a soft mode instability predicted by Hartree–Fock calculations for $r_s \sim 3.3$, we find evidence of multiple rotons in the dispersion of the softening spin excitations. Extrapolation of the data indicates the possibility of an instability for $r_s \gtrsim 11$.

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As the density of a two-dimensional electron system decreases, unusual behavior is expected due to the increasing importance of the electron–electron interaction. Wigner originally predicted that at sufficiently low density an interaction driven phase transition to a crystal state would occur \cite{Wigner59}. It is possible, however, that quantum phase transitions to other broken symmetry states may occur before the onset of crystallization. In the quantum Hall regimes, low energy rotons in collective excitations have been identified as possible precursors of instabilities at low electron densities \cite{Burkov02, Balatsky02, Galitski03}. Such collective excitations of quantum Hall systems have been observed using inelastic light scattering \cite{Burkov02, Balatsky02, Galitski03}. Until now, however, measurements have been restricted to high density systems, relatively far from predicted instabilities. In this letter we report inelastic light scattering measurements of high mobility dilute 2D electron systems with densities less than $10^{10}$ cm$^{-2}$, in the range where instabilities have been predicted to occur \cite{Burkov02, Balatsky02, Galitski03}.

In the quantum Hall regime breakdown of wavevector conservation due to weak residual disorder activates light scattering by critical points in the dispersion of collective modes, allowing the observation of roton minima. We find that the roton of spin–density inter–Landau level (ILL) excitations at $\nu = 2$ displays a significant softening at low density. At densities similar to those where Hartree-Fock calculations predict a zero–energy spin density mode \cite{Burkov02, Balatsky02, Galitski03}, additional peaks appear in the light scattering spectra. These sharp peaks can be understood as the appearance of multiple rotons in the dispersion of the spin density excitation. We propose that the emergence of multiple rotons at very low density arises from a mixing of higher Landau levels into the ground state wavefunction. The roton instability of Hartree-Fock calculations seems to be removed or pushed to lower density by Landau level mixing in the ground state.

We also measure dispersive 2D plasmons (at magnetic field $B = 0$) and charge density ILL excitations at $B \neq 0$ (magnetoplasmons). Well defined dispersive modes are observed at densities $n < 10^{10}$ cm$^{-2}$. Because of the low electron density, we are able to measure the dispersion curves up to wavevectors $q$ approaching $1/l_0$, where $l_0 = \sqrt{\hbar c/eB_\perp}$ is the magnetic length and $B_\perp$ is the magnetic field normal to the sample. At low densities disorder may easily dominate the physics of the electron system. The observation of sharp dispersive modes is evidence that disorder is not important in the very low density samples of this study.

\begin{figure}[h]
\centering
\includegraphics[width=0.8\linewidth]{figure1}
\caption{(a) Experimental geometry. Changing the angle $\theta$ changes the scattering wavevector $q = \frac{\pi}{L} \sin \theta$. (b) Depolarized inelastic light scattering spectrum of a sample with $r_s = 1.8$ at $\nu = 2$. (c) Charge density and spin density excitations as a function of wavevector $q$ in a Hartree-Fock approximation.}
\end{figure}

The samples in this work are GaAs/Al$_x$Ga$_{1-x}$As single quantum wells containing high quality two-dimensional electron systems. In order to achieve low densities without gating the samples, we use a heterostructure design with a large setback of the quantum well from the Si delta doping and very low aluminum concentration ($x \sim 0.03$) in the barriers. The transport mobility of our samples exceeds $1.5 \times 10^6$ cm$^2$/Vs at a density of $1.3 \times 10^{10}$ cm$^{-2}$. For more dilute samples, the dispersive plasmon is used to determine the electron density \cite{Burkov02, Balatsky02, Galitski03}. As shown
in Fig. 1(a), light scattering spectra are acquired using a backscattering geometry at sample temperatures less than 200 mK. The angle $\theta$ between the normal to the sample and the incident/scattered light is continuously tunable between $\theta = \pm 60^\circ$ at low temperature, allowing the scattering wavevector $q = \frac{2\pi}{\lambda} \sin \theta$ to vary between $q_{\text{min}} \approx 0$ and $q_{\text{max}} = 1.3 \times 10^5$ cm$^{-1}$. Magnetic fields as high as 17 T are available, and incident power densities are always lower than $3 \times 10^{-4}$ W/cm$^2$.

It is convenient to define the electron density in terms of the usual dimensionless parameter $r_s \equiv \frac{1}{\sqrt{\pi n e^2 \epsilon_0}}$, where $m_0 = 0.067$ is the band effective mass, and $c = 13$ is the dielectric constant [8]. Large $r_s$ corresponds to low density and strong interactions. At $\nu = 2$, $r_s = E_c/\hbar \omega_c$, where $E_c \equiv e^2/\epsilon l_0$ is the interaction energy, $\hbar \omega_c = \hbar e B_\perp/m_0$ is the cyclotron energy, and $B_\perp = B \cos(\theta)$.

Figure 1(b) shows a light scattering spectrum of ILL excitations in a sample with density $n = 9.6 \times 10^{10}$ cm$^{-2}$, corresponding to $r_s = 1.8$. The incident and scattered light have perpendicular polarizations (depolarized spectra), indicating that the light scattering peak is due to spin–density excitations [10]. Because of the spin symmetry at $\nu = 2$, the spin density inter–Landau level excitation is expected to be a triplet mode [2,3]. The sharp triplet of Fig. 1(b), whose components have a full–width at half–maximum less than 20 $\mu$eV, is a direct observation of this degeneracy. The peaks in the triplet are labeled by the change in angular momentum $\delta m_z$ parallel to the magnetic field $B$, and their splitting is the Zeeman energy. We observe triplet excitations in all measured samples up to $r_s = 3.3$. At larger $r_s$, the Zeeman splitting is too small at $\nu = 2$ to resolve the triplet.

The triplet in the depolarized spectra of Fig. 1(b) is shifted below the cyclotron energy $\hbar \omega_c$ by $12\%$. The light scattering we observe in all depolarized spectra is independent of angle $\theta$ and wavevector $q$, so we identify the triplet as the roton minimum of the SDE, as shown in Fig. 1(c). Roton minima of quantum Hall collective modes typically occur at wavevectors larger than those available in light scattering experiments. Rotons have been observed, however, because residual disorder relaxes the requirement of wavevector conservation, allowing the observation of critical points in the density of states [3]. The intensity of Raman scattering with weak breakdown of wavevector conservation can be written as [11]

$$I(q, \omega) \sim \int_0^\infty f_q(k)S_a(k, \omega)dk,$$

where $S_a(k, \omega)$ is the dynamical structure factor. For the charge density response $a = \rho$ and $S_\rho$ is a density–density correlator; for the spin density response $a = \sigma$ and $S_\sigma$ is a spin–spin correlator. In the absence of disorder wavevector conservation is maintained: $f_q(k)$ will be sharply peaked at $k = q$, and $I(q, \omega)$ will map out the dispersion curves of collective excitations. In the presence of disorder, $f_q(k)$ is broad, and light scattering is independent of $q$. Then $I(\omega)$ will be peaked at the energies of critical points in the density of states for collective excitations, such as the roton of the SDE in Fig. 1(b). In the intermediate case of very weak disorder, both critical points in the density of states and the wavevector conserving dispersion may be visible.

![Figure 2](image)

**FIG. 2.** (a) Two SDE peaks in a depolarized spectrum of a sample with $r_s = 4.9$. The peaks are split by much more than the Zeeman energy. (b) Polarized spectra from the same sample at four angles $\theta$. Stars indicate critical points activated by residual disorder, and downward arrows indicate the dispersive magnetoplasmon.

Figure 2(a) shows light scattering spectra of spin excitations at $\nu = 2$ in a sample with $r_s = 4.9$. The two sharp peaks do not depend on angle $\theta$, indicating that they correspond to densities of states in the dispersion curves. Both peaks are shifted well below $\hbar \omega_c$. The two peaks do not correspond to $\delta m_z$ components of the spin excitations, because the Zeeman energy is less than 10 $\mu$eV at this field. The appearance of two such distinct peaks in light scattering spectra of the spin excitations at large $r_s$ is surprising, because only a single critical point is expected in the dispersion of spin excitations [2,3].

The solid circles in Fig. 2(a) are the measured energies of spin density excitations at $\nu = 2$ in 8 samples covering a wide range in $r_s$. We can compare these energies with a calculation in the Hartree–Fock approximation (HFA) of the energy of the spin–density roton minimum, shown as the dotted line in Fig. 2(a). The calculation includes both
the effects of the finite thickness of the quantum well and the self-consistent coupling between the excited states. Although the fit is excellent for \( r_s < 2 \), the HFA predicts a collapse of the roton energy at \( r_s \approx 3.3 \) which is not observed in our data. The HFA is perturbatively exact when \( r_s \ll 1 \), but is of questionable validity for \( r_s \gtrsim 1 \), because the Coulomb interaction becomes too strong to treat perturbatively.

It is remarkable that the onset of multiple peaks in spin–excitations, like the two peaks in Fig. 2(a), occurs at approximately the same value of \( r_s \) as the predicted HFA spin instability. Fig. 3 shows that not only the sample with \( r_s = 4.9 \), but also the samples with \( r_s = 3.3 \) and \( r_s = 5.9 \), have more than one peak in depolarized spectra. The multiple peaks indicate that the spin–excitation dispersion has become more complicated at large \( r_s \). Multiple peaks can be explained by the emergence of multiple rotons in the dispersion of the SDE at large \( r_s \), which increase the number of critical points in the collective mode density of states. Multiple critical points produce multiple light scattering peaks, as can be deduced from Eq. (1). A naive extrapolation of the measured roton excitation energies to larger \( r_s \), as shown by the dashed lines in Fig. 3 reveals potential instabilities in the range \( 11 \lesssim r_s \lesssim 14 \); although the HFA seems to overestimate the tendency towards instability, a spin instability may still occur before the anticipated transition to a Wigner crystal at even larger \( r_s \).

Figure 4 shows how the concept of multiple rotons explains the depolarized (SDE) light scattering peaks we observe at large \( r_s \). As shown in Fig. 4(c), however, the HFA predicts only a single roton minimum in the dispersion of spin excitations. The disagreement with Hartree-Fock calculations of the collective modes may arise because the HFA assumes a priori that the ground state at \( \nu = 2 \) fills the lowest spin–split Landau level, without Landau level mixing. The origin of the single roton minimum in the HFA is essentially the simple structure of the 0th and 1st (orbital) Landau level single particle wavefunctions, which have zero and one node respectively. We suggest that the incorporation of higher Landau levels into the ground state at large \( r_s \) increases the complexity of the ground state wavefunction by including single particle states with a finite number of nodes. The finite number of nodes can lead to multiple rotons in the collective excitations. This is analogous to collective excitations at higher filling factors, which are predicted to have multiple rotons.

Figure 2(b) shows polarized spectra at four angles in the sample with \( r_s = 4.9 \). The light scattering peaks are due to the \( \nu = 2 \) charge density excitation (CDE). Two peaks, marked by the \( \star \) under the peaks, are independent of \( \theta \) and are due to critical points in the density of states (DOS) for the CDE. Unlike the all the spectra we have discussed so far, there is one peak in Fig. 2(b) which does depend on angle \( \theta \) and wavevector \( q \). It is marked by the vertical arrows in Fig. 2(b). At 25° this peak overlaps the DOS peak at 0.9 meV. Previous experiments using inelastic light scattering to measure the CDE in the quantum Hall regime have utilized wavevector breakdown to measure densities of states, or have used gratings to determine a scattering wavevector. At this density we observe both wavevector conserving and wavevector nonconserving scattering in the same spectrum without artificial gratings. In polarized spectra at higher densities only the DOS peaks are visible, and at lower densities we observe only the dispersive mode.

![FIG. 3. Solid circles: Summary of peaks in depolarized spectra for eight samples, each with a different value of \( r_s \). Peaks at the same value of \( r_s \) appear in the same sample. Note the onset of multiple peaks at \( r_s = 3.3 \). Dotted line: Calculated energy of the roton minimum in a Hartree-Fock approximation. Dashed lines: Linear extrapolation of the data.](image-url)
push Fermi liquid theory out of its strict range of validity to large $B$, into the quantum Hall regime, in order to compare with our results.

![Graph showing dispersion curves](image)

**FIG. 4.** Circles: Experimentally measured dispersion. Asterisks: Energies of observed critical points. Lines: SDE and CDE dispersions in a Fermi liquid theory approximation. The solid lines indicate the region in the dispersions where the collective mode has a large weight in $S_a(q, \omega)$. The lines in Fig. 4 represent the calculated dispersions of spin and charge modes in a semiclassical FLT approximation using only the first significant Fermi liquid parameters for charge, $F_1^c$, and spin, $F_0^a$. We recall that $F_1^c$ and $F_0^a$ determine the effective mass and the spin susceptibility respectively. The best fit to our data is obtained with $F_1^c = -0.1$ and $F_0^a = -0.87$, which are not unreasonable values for these parameters at this density. We mark with thick lines the regions where the weight of the collective mode is large in the dynamical structure factor $S_a(q, \omega)$. The weight in $S_a(q, \omega)$ oscillates because of variations in the commensurability of the cyclotron orbits and the wavevector of the collective oscillation; this is analogous to the variation in the matrix elements for the overlap of particle–hole pairs in HFA calculations.

The dispersive peak represented by the solid circles in Fig. 4 is revealed to be the dispersive magneto-plasmon. The weight in $S_a(q, \omega)$ continues through the avoided crossing at $\sim 2.1 \text{ meV}$, as observed in the experiment. The two critical points in the spin (charge) mode are explained as arising from two minima (maxima) in the dispersions. The variation in the weight of $S_a(q, \omega)$ explains why peaks are observed at only specific flat regions of the dispersion curves.

The surprisingly good numerical agreement (only two fitting parameters were used, $F_1^c$ and $F_0^a$) between our data and calculations based on Landau Fermi liquid theory must at this point be regarded as fortuitous. Neither FLT nor the HFA are fully valid in our experimental regime. While the HFA provides an excellent treatment of the $\nu = 2$ quantum Hall system with weak interactions, it is expected to fail in the strong interaction limit. Conversely, although FLT is an excellent paradigm in which to treat strong interactions in weak magnetic fields, it is not well suited to handling the fully quantum limit at $\nu = 2$. These two complimentary approaches have allowed us to suggest mixing of higher Landau levels in the ground state wavefunction to explain our data.

In conclusion, we studied dilute 2D electron systems by means of inelastic light scattering. The magnetoplasmon shows a well defined dispersion in the quantum Hall regime down to $n < 10^{10} \text{ cm}^{-2}$. At $\nu = 2$ the roton of the spin–density mode softens at large $r_s$. The spin instability predicted by HFA calculations at $r_s \sim 3.3$ is not observed, but we find the emergence of multiple rotons in the spin–density mode. We suggest a mixing of higher Landau levels in the ground state wavefunction as an explanation for the appearance of multiple rotons at large $r_s$. These results are further indication that large $r_s$ integer quantum Hall states will involve interaction–driven correlation. The collapsing energy of the roton of spin excitations suggests the possibility of instabilities at large $r_s$ due to spin correlations, in addition to the anticipated Wigner crystal.

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