Spontaneous Interlayer Charge Transfer near the Magnetic Quantum Limit

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Experiments reveal that a confined electron system with two equally-populated layers at zero magnetic field can spontaneously break this symmetry through an interlayer charge transfer near the magnetic quantum limit. New fractional quantum Hall states at unusual total filling factors such as \( \nu = \frac{1}{2} \) stabilize as signatures that the system deforms itself, at substantial electrostatic energy cost, in order to gain crucial correlation energy by “locking in” separate incompressible liquid phases at unequal fillings in the two layers (e.g., layered \( \frac{1}{2} \) and \( \frac{1}{2} \) states in the case of \( \nu = \frac{11}{12} \)).

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Imagine creating a high-quality two-dimensional (2D) electron system and then moving in from afar a second identical layer constrained to a parallel plane. As the separation between the two charged sheets shrinks, both Coulomb repulsion and electron tunneling between the layers increase. Now imagine, after establishing suitably close proximity between the two 2D systems, turning a knob that quenches the interlayer electron tunneling. What remains is a symmetric bilayer system in which charge transfer between layers, in analogy to a parallel-plate capacitor, can only occur by surmounting a sizable electrostatic energy barrier—the “out-of-plane” Hartree charging energy. For a system of sufficiently low density, this charging energy may be overcome by the exchange-correlation energy, leading to a spontaneous transfer of all charge into one of the layers. Such so-called “exchange-instabilities” have been predicted to occur in multilayers \(^1\) but have not yet been observed experimentally \(^2\). This may be due to the fact that samples of sufficiently low density and low disorder have not been available, or possibly because other more exotic bilayer phases \(^3\) intervene and maintain a lower energy than the spontaneously-generated monolayer phase.

Envision, on the other hand, approaching the diamagnetic regime—the extreme magnetic quantum limit. It is known that when a large magnetic field \( B_\perp \) is applied perpendicular to the plane of a 2D electron layer, electron-electron interactions can dominate and lead to new ground states such as the fractional quantum Hall (FQH) liquids at certain Landau-level filling factors \( \nu \). Because of the particular stability (cusp-like minima in energy \(^4\) ) of the FQH phases at these special fillings, one might expect a partial charge transfer between the two layers of an interacting bilayer electron system (ES) if the ensuing inequivalent layers can each support a strong FQH state. Here we report strong experimental evidence that such a charge transfer indeed occurs.

By modulation-doping a 750 Å-wide GaAs quantum well, we fabricated a special ES whose density \( n \) and charge distribution are controlled via a pair of metal front- and back-side gates \(^5\). Measurements were performed at base temperatures \( T \approx 30 \text{ mK} \). For symmetric (“balanced”) charge distributions, the ES in a wide well can, in general, be tuned from a single-layer to an interacting bilayer system by either increasing \( n \) or applying an in-plane magnetic field \( B_\parallel \). Increasing \( n \) results in an increase (decrease) of the charge density near the sides (center) of the well, and reduces the interlayer tunneling. For a fixed \( n \), increasing \( B_\parallel \) (by increasing the angle \( \theta \) between the normal to the sample plane and the magnetic field direction) has a qualitatively similar effect \(^6\). The evolution of the FQH states in this system with increasing \( n \) has been reported recently \(^7\). At the lowest \( n \) the sample exhibits the usual FQH effect at exclusively odd-denominator \( \nu \), while at the highest \( n \) the strongest FQH states are those with even numerators, as expected for a system of two 2D ESs in parallel. For intermediate \( n \), even-denominator FQH states at total fillings \( \nu = \frac{1}{2} \) and \( \frac{3}{2} \), which are stabilized by both interlayer and intralayer correlations, are observed. Also of relevance to this paper is the observed one-component (1C) to two-component (2C) transition of certain FQH states at total filling \( \nu = \frac{3}{2} \) and \( \frac{4}{2} \). Indeed the \( \frac{1}{2} \) and \( \frac{3}{2} \) FQH states in this ES exhibit a clear 1C to 2C phase transition with increasing \( n \) \(^8\) or \( \theta \), as evidenced by a pronounced minimum in each of their quasiparticle excitation gaps measured as the system is tuned from a monolayer to a bilayer.

Of central interest here is the appearance of FQH states at unusual \( \nu \) in this ES. We have observed such states near the \( \nu = \frac{2}{3} \) and \( \frac{4}{3} \) FQH states once these became of 2C origin. The data of Fig. 1, taken at fixed \( n = 11.2 \times 10^{10} \text{ cm}^{-2} \) with variable \( \theta \), provide an example. The top curve in Fig. 1(a) shows that at \( \theta = 0 \), the ES exhibits FQH states which are primarily single-layer-like. However, upon tilting the sample to \( \theta = 30^\circ \) and \( 48.4^\circ \) (bottom \( R_{xx} \) traces), several additional FQH states appear between \( \nu = \frac{2}{3} \) and \( \frac{4}{3} \) while the integral QH effect at \( \nu = 1 \) disappears. Considering that at these
large $\theta$ values the ES (for $\nu \leq 2$) has become bilayer-like, some of the additional FQH states—such as those at $\nu = \frac{4}{5}, \frac{6}{5}$, and $\frac{5}{3}$—can be understood as the descendents of the $\nu = \frac{2}{5}$ and $\frac{4}{3}$ states. For example, assuming that the 2C $\nu = \frac{2}{5}$ state arises from two $\frac{4}{5}$ FQH states in each layer, the FQH sequence at $\nu = \frac{2}{5}, \frac{4}{5}, \frac{5}{3}$ is simply the usual $\frac{1}{5}, \frac{2}{5}, \frac{3}{5}$ sequence for each layer. The well-developed FQH states at $\nu = \frac{11}{15}$ and $\frac{13}{21}$ (see vertical arrows), however, cannot be accounted for in any reasonable bilayer or single-layer scheme [11]. On the other hand, note that each of these unusual fractions can be expressed as the sum of two simpler fractions. In fact, this is not a mere coincidence, and our data indicate that at these special $\nu$, the system is unstable toward an interaction-induced interlayer charge transfer that “locks in” independent FQH states at unequal fillings in each layer. At $\nu = \frac{11}{15}$, for example, one layer has $\frac{1}{5}$ filling and the other $\frac{2}{5}$. Similarly, the FQH state at $\nu = \frac{13}{21}$ ($= \frac{2}{5} + \frac{2}{5}$) can arise from an interlayer charge transfer leading to FQH states at $\frac{2}{5}$ and $\frac{3}{5}$ fillings in two layers. In the remainder of the paper we elucidate the behavior of these unusual states as a function of $\theta$ (at fixed $n$), $n$ (at fixed $\theta$), and intentionally imposed charge transfer (at fixed $n$ and $\theta = 0$).

Figure 1(b) summarizes the evolution of the FQH states in this ES as a function of increasing $\theta$. We have condensed a large set of traces onto the $(B_{\perp}, \theta)$ plane by mapping $R_{xx}$ (normalized to its maximum value within the plotted parameter range) to a grayscale color between black and white. In such a plot, the QH and FQH phases show up as dark black regions, whose width along the $B_{\perp}$ axis is a reflection of the strength of the associated state, i.e. the magnitude of its energy gap. The traces in Fig. 1(a) can be interpreted as constant-$\theta$ slices through the image of Fig. 1(b). As $\theta$ is increased, the system is swept from the 1C through the 2C regime; a visible measure of this general evolution is the weakening and eventual collapse of the $\nu = 1$ QH state. The $\nu = \frac{4}{5}$ FQH state, another 1C state, is also destroyed by the increasing $B_{\parallel}$. For the states that can exist as both 1C and 2C phases, transitions between the two ground states are evident. For example, the $\nu = \frac{3}{2}$ and $\frac{5}{3}$ states undergo a 1C to 2C transition at $\theta \simeq 18^\circ$ and $27^\circ$, respectively. Nestled between these two states and in close proximity to their 1C+2C transitions, an $\frac{11}{15}$ FQH state develops and becomes quite strong. At the same time, $\rho_{xy}$ exhibits a quantized plateau at $\frac{11}{15}(h/e^2)$ [see lower right of Fig. 1(a)]. Very similar behavior is observed on the other side of $\nu = 1$ in Fig. 1(b). A $\frac{13}{21}$ state develops in the vicinity of the $\nu = \frac{3}{5}$ 1C+2C transition $(\theta \simeq 35^\circ)$ along with the appearance of the 2C $\frac{3}{5}$ state (at $\theta \simeq 38^\circ$).

We have observed a similar evolution of the $\nu = \frac{11}{15}$ state in the same sample at $\theta = 0$ as a function of increasing $n$. While at low $n$ the FQH states have single-layer origin, when the system is tuned to very high $n$, as shown in Fig. 2, the dominant states are of 2C origin, as evidenced by the preponderance of even-denominator FQH states. Of special note, however, are the states marked with vertical arrows that do not fit in the normal 1C or 2C hierarchy. Like the 2C case at high $\theta$ described above, states here at $\nu = \frac{13}{15}$ and $\frac{19}{15}$ are very strong and are accompanied by Hall plateaus as indicated. For $\theta = 0$, the $\nu = \frac{11}{15}$ FQH state starts to develop at $n \simeq 13 \times 10^{10}$ cm$^{-2}$, slightly above the $n$ at which the $\nu = \frac{6}{5}$ FQH state makes its 1C to 2C transition [9].

For conciseness, let us concentrate on the strongest of these “special” states, namely the one at $\nu = \frac{11}{15}$. To summarize the data presented so far, we observe a FQH state at $\nu = \frac{11}{15}$ in an ES in a wide quantum well under either of two conditions: (a) without an in-plane $B$ but at large $n$ so that the ES at and near $\nu = \frac{2}{3}$ has already made a 1C to 2C transition; (b) starting with a 1C state (i.e., low enough $n$) but applying a sufficiently large $B_{\parallel}$ so that again the ES at and near $\nu = \frac{2}{3}$ has made a 1C to 2C transition. We emphasize that in both cases, in the absence of any applied $B$, the ES has a symmetric (“balanced”) charge distribution. Since in both cases the $\frac{11}{15}$ FQH state appears when the system has become 2C,
it is reasonable to assume that tunneling is sufficiently reduced and can be ignored [12].

Thus, ignoring tunneling, we concentrate on the competition between two energies:

(1) **Energy cost** — The capacitive energy $\mathcal{E}_{\text{CAP}}$ which works against the formation of a $\left(\frac{4}{15} + \frac{2}{5}\right)$ state as it opposes interlayer charge transfer. A rough estimate of this energy, expressed per electron, for the system at $n = 11.2 \times 10^{10}$ cm$^{-2}$ is

$$\mathcal{E}_{\text{CAP}} \simeq \frac{Q^2}{2C} \frac{1}{n} = \frac{(en_l)^2}{2(\epsilon/d)n} \lesssim 1 \text{ K},$$

where $Q \equiv en_l = 5.1 \times 10^9$ e/cm$^2$ is the charge transfer per unit area from one layer to the other necessary to set up the $\left(\frac{4}{15} : \frac{2}{5}\right) = (5 : 6)$ layer density ratio, $C \equiv \epsilon/d$ is the interlayer capacitance (per unit area), $d \simeq 480$ Å is the interlayer spacing, and $\epsilon = 13.1$ is the GaAs dielectric constant. Equivalently, $\mathcal{E}_{\text{CAP}}$ is the electrostatic energy difference between the balanced and imbalanced charge configurations pictured in the insets to Fig. 3.

(2) **Energy savings** — The correlation energy $\mathcal{E}_{\text{COR}}$ gained by forming the incompressible $\frac{1}{3}$ and $\frac{5}{6}$ FQH liquid states in the two separate layers. To estimate this energy, we have calculated the total energy of a single layer (i.e. half of the bilayer system) as a function of the layer filling factor $\nu_l$. The results are plotted in Fig. 3 and are based on the best numerical calculations available to date [13] for the exchange-correlation energies and quasiparticle excitation gaps of principal FQH states in a realistic (i.e. of finite density and non-zero thickness) 2D system. Higher-order FQH states were added to the calculation by exploiting the energy gap scaling law [14,15] $
abla \nu \propto (\epsilon^2/\epsilon_B)/(2mp + 1)$, where $\nabla \nu$ is the quasiparticle excitation gap of the $\nu = p/(2mp + 1)$ FQH liquid ($p$ integer, $m = 1, 2, \ldots$), and $\epsilon_B \equiv \sqrt{\hbar/eB}$ is the magnetic length. Appropriate corrections for Landau level mixing and finite thickness were assumed: for a bilayer system of total $n = 11.2 \times 10^{10}$ cm$^{-2}$, a layer of density $n/2$ has $r_s \simeq 2.3$ (average interparticle spacing normalized to the effective Bohr radius) and half-width $\lambda \simeq 85$ Å. The spline procedure described in Ref. [13] was used to connect the various states, rendering the cusps evident in the energy per layer $\mathcal{E}_l(\nu_l)$ shown in Fig. 3. The liquid correlation energy gain is depicted by the peak-to-trough arrows in Fig. 3; its magnitude (again, per electron) is

$$\mathcal{E}_{\text{COR}} \simeq 2\mathcal{E}_l(\nu_l = \frac{1}{3}) - \mathcal{E}_l(\nu_l = \frac{5}{6}) \geq 1 \text{ K}. \tag{2}$$

The compressible ES at $\nu = \frac{11}{15}$, or equivalently the system of two layers each at $\frac{11}{15}$ filling, spontaneously “phase-separates” into two incompressible FQH liquids at unequal layer fillings of $\frac{1}{3}$ and $\frac{5}{6}$. The symmetry-breaking interlayer charge transfer that must occur [see Fig. 3(insets)] to produce the required density imbalance entails a significant electrostatic penalty $\mathcal{E}_{\text{CAP}}$, but proceeds only because this capacitive energy barrier is surmounted by the correlation energy advantage $\mathcal{E}_{\text{COR}}$ in forming two stable FQH liquids. That our best estimates indicate the competing energies $\mathcal{E}_{\text{CAP}}$ and $\mathcal{E}_{\text{COR}}$ are comparable attests to the quantitative plausibility of this explanation.

Finally, to verify our conjecture that the $\nu = \frac{11}{15}$ state is indeed stabilized by interlayer charge transfer, we did the following test at $\theta = 0$. Suppose we start with the ES at an $n$ where the $\nu = \frac{1}{3}$ FQH state has just become 2C so that the incompressible state at $\frac{11}{15}$ has barely developed [e.g., $n = 12.6 \times 10^{10}$ cm$^{-2}$; see Fig. 4(a)]. Now suppose we keep $n$ fixed but intentionally impose an interlayer charge transfer $n_l$ by applying a perpendicular electric field (physically generated via front- and back-gate biases of opposite sign). As we transfer charge, the $\frac{11}{15}$ FQH state should get stronger as $2n_l/n$ approaches the ratio $(\frac{3}{5} - \frac{1}{3})/(\frac{3}{5} + \frac{1}{3}) = \frac{1}{4}$, and then should become weaker once $2n_l/n$ exceeds $\frac{1}{3}$. The data shown in Fig. 4(a–c) demonstrate that this behavior is indeed observed in our experiment. In particular, the quasiparticle
excitation gap $\Delta_{11/15}$ measured for the $\frac{11}{15}$ FQH state is largest when the charge distribution is imbalanced close to the expected $(5:6)$ layer density ratio [dotted line in (d)].

Two additional features of the data in Figs. 1 and 2 are noteworthy. First, the $\nu = \frac{13}{15}$ state appears to become weaker with increasing $\theta \gtrapprox 40^\circ$. This is reasonable and stems from the fact that spontaneous charge transfer will only occur if $(\varepsilon_{\text{COR}} - \varepsilon_{\text{CAP}}) > 0$. At very large $B_{\parallel}$ (or $n$), the two layers become increasingly more isolated and the capacitive energy opposing charge transfer begins to dominate any correlation energy savings that would come from a $(\frac{1}{3} + \frac{2}{3})$ state. Thus, the system remains compressible, as expected for two distant and weakly-coupled parallel 2D ESs at $\nu = \frac{13}{15}$ ($\frac{11}{15}$ filling in each layer). Second, the $R_{xx}$ minimum near $\nu = \frac{29}{35} = (\frac{19}{35} + \frac{10}{35})$ suggests a developing FQH state at this filling [Figs. 1(a) and 2]. Such a state can be stabilized if, at $\nu = \frac{29}{35}$, there is an interlayer charge transfer so that one layer supports a FQH state at $\frac{2}{5}$ filling and the other at $\frac{3}{5}$. Similarly, the weak $R_{xx}$ minimum observed near $\nu = \frac{29}{37}$ [Fig. 1(a)] may hint at a developing FQH state stabilized by the formation of $\frac{1}{3}$ and $\frac{2}{5}$ FQH states in the separate layers.

To summarize, we present evidence that a bilayer ES near the magnetic quantum limit undergoes a correlation-driven interlayer charge transfer at $\nu = \frac{2}{5}$ and $\frac{3}{5}$, and possibly at $\nu = \frac{29}{37}$ and $\frac{30}{37}$. In closing, we note an elegant magnetic analogy for our observations. Our bilayer ES can be conveniently mapped onto a pseudospin-$\frac{1}{2}$ system by identifying the symmetric/antisymmetric subband states (front/back layer states) with the eigenstates of the $x$-component ($z$-component) of the pseudospin operator $\mathbf{\hat{z}}$. Within this framework, a layer imbalance is equivalent to a non-zero pseudomagnetization $m_z = 2n_t/n$ along the $z$-axis. As an increasing $B_{\parallel} \equiv B_2$ is applied, the layers remain balanced ($m_z = 0$) for all applied fields except at the special fillings $\nu = p/q$ listed above, when the system locks to a spontaneous fractional pseudomagnetization $m_z = 1/p$ (e.g., $m_z = \frac{1}{15}$ at $\nu = \frac{11}{15}$, $m_z = \frac{1}{15}$ at $\nu = \frac{19}{35}$, etc.) and behaves as a “quantized paramagnet” with susceptibility $\chi_{m_z} \equiv n\Phi_0 m_z / B_2 = 1/q$ ($\Phi_0 \equiv \hbar/e$ is the flux quantum).

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FIG. 4. Intentionally-imposed interlayer charge transfer $n_t$ will stabilize (b) an incompressible layered $(\frac{1}{3} + \frac{2}{3})$ FQH state at $\nu = \frac{11}{15}$ when the charge distribution is imbalanced close to the expected $(5:6)$ layer density ratio [dotted line in (d)].