Observation of a new phase transition between fully and partially polarized quantum Hall states with charge and spin gaps at $\nu = \frac{2}{3}$

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(Dated: March 22, 2022)

The average electron spin-polarization $\mathcal{P}$ of two-dimensional electron gas confined in GaAs/GaAlAs multiple quantum-wells was measured by nuclear magnetic resonance (NMR) near the fractional quantum Hall state with filling factor $\nu = \frac{2}{3}$. Above this filling factor ($\frac{2}{3} < \nu < 0.85$), a strong depolarization is observed corresponding to two spin flips per additional flux quantum. The most remarkable behavior of the polarization is observed at $\nu = \frac{2}{3}$, where a quantum phase transition from a partially polarized ($\mathcal{P} \approx \frac{2}{3}$) to a fully polarized ($\mathcal{P} = 1$) state can be driven by increasing the ratio between the Zeeman and the Coulomb energy above a critical value $\eta_c = \frac{\Delta Z}{\Delta C} = 0.0185$.

The integer quantum Hall effect (QHE) occurs when the Landau level (LL) filling factor $\nu$ is an integer $p$ and the lowest $p$ LL’s are filled. However, if the Zeeman splitting $\Delta Z = |\mu| B$ is increased with respect to the LL spacing $\hbar \omega_c$ ($\omega_c = 0.44$ is the electronic Landé factor in GaAs, $\mu_B$ the Bohr magneton, $B$ the magnetic field and $\omega_c$ the cyclotron frequency), crossings of LL’s with different spin states occur when $k_\perp \hbar \omega_c = \Delta Z$. If the level crossing occurs for LL’s at the Fermi energy, the spin configuration of the electrons changes. The fractional QHE (FQHE) occurs when $\nu = \frac{\mathcal{P} \cdot \mathcal{C}}{c}$, where $m = 1, 2$ and can be visualized with the composite fermion (CF) model. In this picture FQHE corresponds to integer QHE of CF’s, where the CF-LL spacing is now determined by the Coulomb energy $\Delta_C = \frac{1}{4e^2 \epsilon \epsilon_r} \frac{\epsilon_r}{\epsilon_B} \sqrt{\frac{1}{B}}$ ($\epsilon \epsilon_r$ is the permittivity GaAs, $\epsilon_B = \sqrt{\hbar/e B}$ is the magnetic length, the average distance of two electrons). Hence similar spin transitions do also occur in the FQHE. Since the relevant energy scales for CF’s are the Zeeman and the Coulomb energies, the spin configuration of a 2DEG is mainly driven by the exchange part of the Coulomb energy ($\propto \sqrt{B}$) at low $B$, whereas at large fields, the Zeeman energy ($\propto B$) favors spin-polarized states. For example, $\nu = \frac{2}{3}$ corresponds to two filled CF-LL’s: only two states are possible, $\mathcal{P} = 0$ at low $B$ and $\mathcal{P} = 1$ in the opposite limit. The transition between these states are controlled by the ratio $\eta = \frac{\Delta C}{\Delta Z}$ at a given $\nu$. This ratio can be changed by varying the electron density ($n$) or by rotating the angle ($\theta$) between the normal to the 2DEG and the applied magnetic field, keeping the perpendicular field $B_\perp$ constant (tilted field technique).

In this Letter, we report an NMR observation of a phase transition between a new, partially polarized QH phase and the fully polarized high field phase at $\nu = \frac{2}{3}$. The partially polarized phase has charge and spin gaps and is completely unexpected by theory. Furthermore, this is the first observation of a phase transition between QH states without the gap going through a minimum at the transition point.

In this study, two GaAs/GaAlAs multiple (100) quantum well (QW) samples M280 (M242) with electron densities $n = 8.5 \cdot 10^{10} (1.4 \cdot 10^{11})$ cm$^{-2}$ and mobilities $\mu_0 = 7 \cdot 10^5 (3 \cdot 10^5)$ cm$^2$/Vs have been used. The $w = 30$ (25) nm wide GaAs QWs are separated by 250 (185) nm thick Al$_{0.1}$Ga$_{0.9}$As (Al$_{0.3}$Ga$_{0.7}$As) barriers with Si doping near their centers. Two pieces of each sample, $\approx 25$ mm$^2$ in size, were placed in the center of a radiofrequency (RF) coil, so that 200 QWs effectively contributed to the nuclear magnetic resonance (NMR) signal. At higher temperatures ($1.5 \text{ K} \leq T \leq 10 \text{ K}$), the NMR signal from barriers (without electrons) remained unshifted and is used as a reference. If the electron motion is rapid with respect to the NMR timescale ($\approx 1-10 \mu$s), the nuclei in QWs by a magnetic hyperfine shift $K_S$ proportional to $\mathcal{P}$ contribute to the nuclear magnetic resonance (NMR) signal. At lower temperatures ($40 \text{ mK} \leq T \leq 1.5 \text{ K}$), the RF-coils were mounted at fixed $\theta$ into the mixing chamber of a dilution refrigerator.

NMR is a very sensitive direct measurement of the spin polarization of 2D-electrons in the QWs, since the Fermi contact interaction $H = \frac{\hbar}{2 \pi} [\gamma_e \gamma_n h^2 \sum_{ij} S_i \cdot J_\perp (r_i - R_j)]$ ($\gamma$ is the gyromagnetic ratio) between itinerant electron spins $S_i$ at position $r_i$ and nuclear spins $J_\perp$ at $R_j$ shifts the resonance frequency of $^{71}$Ga nuclei in the QWs by a magnetic hyperfine shift $K_S$ proportional to $\mathcal{P}$. The NMR signal from barriers (without electrons) remains unshifted and is used as a reference. If the electronic motion is rapid with respect to the NMR timescale ($\approx 1-10 \mu$s), the nuclei in QWs see an average value $\langle S \rangle$. Hence, the average spin-polarization is inferred from the hyperfine shift from $\mathcal{P} = 0$ to $\mathcal{P} = 1$ by $K_S = \frac{\langle S \rangle}{K_S(\mathcal{P} = 1)}$, where $K_S(\mathcal{P} = 1)$ is the maximum shift measured in each sample for high Zeeman energy. Values of $K_S(\mathcal{P} = 1)$ for our two samples obey the empirical definition of the “coupling constant” $A_c = \frac{\hbar}{n} K_S(\mathcal{P} = 1) \approx 4.5 \cdot 10^{-13}$ cm$^3$/s, determined previously by optically pumped NMR. In contrast to other quantitative measurements of $\mathcal{P}$ which use optical techniques, standard NMR is a quasi-equilibrium probe of the 2DEG polarization and does
not affect the electron system.

At high \( T \), where \( K_S \) is small, we take advantage of the much shorter nuclear spin-lattice relaxation times \( T_1 \) of Ga nuclei in the QWs compared to those in the barriers in order to distinguish their contributions to the signal \( ^5 \). An NMR pulse-sequence first destroys the nuclear magnetization \( M_0 \) in the whole sample. After a recovery time \( t_R \) of the order of \( T_1 \) of the QWs nuclei, few nuclei in the barriers have recovered and the NMR signal is dominated by the nuclei in the wells. The spectrum is obtained either by free induction decay after a \( \frac{\pi}{2} \) pulse or by a \( \frac{\pi}{2} \) – \( \pi \) spin-echo technique.

At low temperature, where \( K_S \) is sufficiently large to distinguish both QWs and barriers’ line positions in a single spectrum (see inset Fig. 4), a simple read-out sequence with small “tip-angle” pulses spaced by a long waiting time \( ^5 \) is preferred, in order to avoid heating the sample and spurious effects on the electron system.

The dependence of the electron spin polarization on \( \eta = \frac{\Delta_{\nu}}{\Delta} \) at the exact filling factor \( \nu = \frac{2}{3} \) is shown in Fig. 4 for both samples M280 and M242, as obtained from the low temperature saturation values of \( P \) at various tilt angles \( \theta \) (see Fig. 3). We observe a Zeeman energy induced phase transition from a partially polarized ground-state (GS) with an average \( P \approx \frac{1}{2} \) to a fully polarized GS above the same critical value \( \eta_c \approx 0.018 \) for both samples (the total magnetic fields at this critical point are \( B_{M280}^{M280} = 6.7 \) T and \( B_{M242}^{M242} = 8.9 \) T). The transition from one GS to the other is very sharp, within a change of \( \eta \) of about 3\%. Such a sharp jump in spin-polarization suggests a first-order phase transition. The precision of this measurement can be seen from the inset to Fig. 4 where the spectra for M280 are plotted. The error on the line’s position is less than 0.1 kHz.

A full polarization at large \( \eta \) qualitatively agrees with numerical calculations on small systems \(^{11} \) where the fully spin-polarized GS at \( \nu = \frac{2}{3} \) has been found to have the lowest energy for large Zeeman energy. In this limit, there is a one to one correspondence between \( \nu = \frac{1}{3} \) and \( \frac{2}{3} \). The \( \frac{2}{3} \) state can be regarded as the particle-hole conjugated state of \( \frac{1}{3} \) within the lowest LL. When Zeeman energy is reduced the symmetry between these states is broken. Whereas the \( \nu = \frac{1}{3} \) state is still fully polarized, \( \frac{2}{3} \) is an unpolarized state. Also within the composite fermion (CF) model at \( \nu = \frac{2}{3} \) only \( P = 0 \) and \( P = 1 \) are possible. The \( \nu = \frac{2}{3} \) state is built out of the fully polarized or unpolarized \( \nu = 2 \) state by attaching two flux quanta, pointing anti-parallel to the external magnetic field, to each electron and projecting the wavefunction on the lowest LL \(^{11} \). Within this picture a partial polarization \( P \approx \frac{1}{2} \) cannot be constructed from filled CF-LL’s, but have to be considered as an inhomogeneous mixture of \( P = 1 \) and \( P = 0 \) states. Similarly, this value of the polarization cannot be reached by exact diagonalization on a small system unless at least \( 8 \cdot k \) electrons are included. This exceeds the limit of existing state-of-the-art calculations already for \( k > 2 \). Recently Apalkov et al. found a transition upon increasing \( \eta \) from an unpolarized to half polarized, and to fully polarized GS within a 12 electron calculation \(^{13} \).

There are other pieces of evidence for QH phases at \( \nu = \frac{2}{3} \) with \( P \neq 1 \). Using optical methods, Kukushkin et al. \(^{9} \) have observed partially polarized GS at \( \nu = \frac{2}{3} \) with \( P = \frac{1}{2} \) at \( \eta \approx 0.005 \) and to \( P = 1 \) for \( \eta > 0.01 \) with a narrow plateau at half polarization. Nevertheless, transport measurements on electron doped samples at \( \nu = \frac{2}{3} \) are displaying a deep minimum of the thermal activation gap at \( \eta \approx 0.01 \). This indirect measurement was interpreted as a transition from unpolarized to fully polarized states \( P = 0 \rightarrow P = 1 \) \(^{14} \). In our samples, where \( \eta(\theta = 0^\circ) \geq 0.0138 \), the transport gap at \( \nu = \frac{2}{3} \) monotonically increases with increasing Zeeman energy, even across the transition at \( \eta_c \) \(^{15} \).

The temperature dependence of the polarization at \( \nu = \frac{2}{3} \) for both samples at various tilt-angles is presented in Fig. 3. The lines are fits to the data using a two-level model: assuming the partition function factorizes into a spin-independent and a spin-dependent part, the temperature dependence of the polarization is \( P(B, T) = P_{T=0}(B) \cdot \tanh \left( \frac{\Delta}{4k_B T} \right) \), where the gap \( \Delta = g^*\mu_B B \) (\( g^* \) is the enhanced \( g \)-factor) is the splitting between the spin-up and spin-down components. Away from the transition (\( \eta \neq \eta_c \)), this fit describes well the data (M280: \( \eta = 0.0138 \), 0.0276 and M242: \( \eta = 0.0229 \)). But close to the transition (\( \eta \approx \eta_c \)), a two-level model fails to describe the temperature dependence of \( P \): the saturation value \( P_{T=0} \) of \( P \) is only reached at

![FIG. 1: The phase transition of the \( \nu = \frac{2}{3} \) state as revealed by the dependence of the spin polarization on the ratio of Zeeman and Coulomb energies \( \eta = \frac{\Delta_{\nu}}{\Delta} \) for Sample M280 (•) and M242 (○) at temperatures below 100 mK. \( \eta \) was varied by changing the angle \( \theta \) between the magnetic field direction and the normal to the 2D plane. The inset shows the NMR spectra corresponding to the points for M280, where the QWs signal amplitude is renormalized.](image-url)
very low $T$, while $P$ is reduced at intermediate temperatures. This may be a consequence of a coexistence region between two phases with different polarizations. This model defines a thermally activated spin-gap $\Delta$ which varies smoothly across the transition at $\eta_c$. Such a dependence rules out a simple “level crossing transition” between two magnetic GSs, where $\Delta$ should collapse at the transition, and suggest a first-order transition. We extracted $\Delta$ even in the case where the two level model fails by determining the temperature where half of the saturation value is reached: $\Delta \approx 2.2 \, T(\mathcal{P} = \mathcal{P}_{T=0}/2)$. These gaps are plotted in Fig. 3. They correspond to $\Delta = (1.54 \pm 0.04) \Delta_S$ and yield a 50% exchange enhancement of the Zeeman splitting $\Delta_S$.

In Fig. 4 the filling factor dependence of the spin polarization is shown for both samples and various tilt angles. The most surprising observation is the strong depolarization measured above filling factor $\nu = \frac{3}{5}$, whether the polarization at $\nu = \frac{3}{5}$ is full or partial. The data below and above the transition represented in Fig. 3 as open and filled symbols respectively, superpose one line for both samples, M280 and M242. This spin depolarization can be well reproduced over a wide range of $\nu$ if we assume two spin flips per removed flux quantum.

This is shown by solid red lines in Fig. 4. For comparison, a depolarization corresponding to a single spin flip per flux quantum is also shown in dotted red lines, clearly out of the observed behavior. The two experimental depolarization lines converge around $\nu \approx 0.85$ towards a polarization close to $\frac{1}{2}$.

For filling factors lower than $\nu = \frac{2}{3}$, the behavior of $\mathcal{P}(\nu)$ is radically different; the polarization remains constant for $\nu < \frac{2}{3}$, for both full and partial polarization. The exception to this behavior are only the data sets (M280: $\theta = 37^\circ$, M242: $\theta = 40^\circ$) at the transition $\eta \approx \eta_c$, where polarization increases from the partial towards the full polarization when $\nu$ is reduced below $\frac{2}{3}$.

Before discussing these data, we recall that the number of orbital states available in one LL is equal to the
number of magnetic field flux quanta \(D = \frac{\phi}{\phi_0} = \frac{A}{\phi_0} \cdot B\) (\(A\) is the sample area, \(\phi\) the flux through the sample, and \(\phi_0\) the flux quantum). When \(\frac{2}{3}\) of the available states are occupied by electrons (\(\nu = n_A/D = \frac{2}{3}\)), the system condenses in the \(\frac{2}{3}\)-FQH state, a state with precisely 3 flux quanta per 2 electrons. If the magnetic field is slightly decreased or increased, the filling factor will be slightly higher or lower than \(\nu = \frac{2}{3}\), but the lowest energy GS will remain to be the same \(\frac{2}{3}\)-FQH state, now containing the quasi-holes or quasi-particles (QP) corresponding to “extra” or “missing” flux quanta \([14]\). The QP excitations at \(\nu = \frac{2}{3}\) carry a charge of \(\frac{1}{3}\) and there are \(\frac{3}{2}\phi_0\) per electron. Hence the QP corresponds to \(\frac{1}{3} + \frac{3}{2}\phi_0 = \frac{1}{2}\phi_0\), meaning that per added \(\phi_0\) two QPs are added. This can easily provide spin \(S = 1\) or 0 (and never \(S = \frac{1}{2}\)).

The same conclusion also holds in a naive CF picture, where the \(\nu = \frac{2}{3}\) state is obtained by a mapping from the \(\nu = 2\) state. Increasing the field, i.e. injecting additional flux quanta, increases the degeneracy of the LLs. The quasi-holes added in this way are placed into the same, spin-aligned LLs as the CFs which does not modify the polarization of the system of CFs, as is experimentally observed below \(\nu = \frac{2}{3}\). Decreasing the field is equivalent to adding QPs to the system of CFs. Per missing \(\phi_0\) there is one QP per CF-LL and at \(\nu = \frac{2}{3}\) there are two occupied CF-LLs. If both QPs are occupying spin-reversed CF-LLs, this provides precisely the depolarization with two reversed spins per additional \(\phi_0\), as is observed above \(\nu = \frac{2}{3}\).

There is no satisfactory understanding of the FQHE in the low magnetic field limit. In particular, since filled CF-Landau levels can only produce fully polarized or unpolarized states at \(\nu = \frac{2}{3}\), the nature of a partially polarized state at this filling factor is open. Among the possible states, it is natural to consider some of the other known spin-liquid states which have been considered in the literature such as the valence-bond GS of the 2D Shastry-Sutherland model \([17]\). Other well-known spin-gapped states are commensurate spin-density waves.

An alternative viewpoint is to consider partially polarized states as inhomogeneous mixture of fully polarized and unpolarized regions \([13]\). The GS could also be described as isolated singlets forming a commensurate modulated structure through the sample, in many ways similar to valence-bond commensurate spin-liquid GSs. Naturally, the real challenge is to determine among all the possible GSs, which one has the lowest energy.

In conclusion, this NMR study has revealed a new phase transition between FQH ground-states at \(\nu = \frac{2}{3}\) with different spin-polarizations. Considering the sharpness of the jump in polarization as well as the thermal activation close to \(k_B\), the transition appears to be first order. Whereas the quasi-hole excitations for \(\nu < \frac{2}{3}\) are spin-aligned, the quasi-particle excitations for \(\nu > \frac{2}{3}\) are found to be spin-reversed, independent from the transition, leading to a rapid depolarization above \(\nu = \frac{2}{3}\).

We gratefully acknowledge enlightening discussions with R. Morf.

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