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GaN/AlGaN 2DEGs in the quantum regime: Magneto-transport and photoluminescence to 60 tesla

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ABSTRACT

Using high magnetic fields up to 60 T, we report magneto-transport and photoluminescence (PL) studies of a two-dimensional electron gas (2DEG) in a GaN/AlGaN heterojunction grown by molecular-beam epitaxy. Transport measurements demonstrate that the quantum limit can be exceeded (Landau level filling factor $\nu < 1$) and show evidence for the $\nu = 2/3$ fractional quantum Hall state. Simultaneous optical and transport measurements reveal synchronous quantum oscillations of both the PL intensity and the longitudinal resistivity in the integer quantum Hall regime. PL spectra directly reveal the dispersion of occupied Landau levels in the 2DEG and, therefore, the electron mass. These results demonstrate the utility of high (pulsed) magnetic fields for detailed measurements of quantum phenomena in high-density 2DEGs.

The wide-bandgap semiconductor GaN is a foundational material for solid-state lighting applications and high-power electronics. Furthermore, the two-dimensional electron gas (2DEG) that forms naturally at GaN/AlGaN heterointerfaces[1–3] is of considerable interest for high-electron-mobility transistors. 2DEG structures grown by molecular-beam epitaxy (MBE) have exhibited low-temperature electron mobilities exceeding 10^5 cm^2/V s,[4,5] galvanizing interest in quantum phenomena and novel electron correlations in GaN-based materials. Indeed, transport measurements have shown a robust integer quantum Hall effect (IQHE) in GaN/AlGaN heterojunctions,[2–9] and an indication of a fractional quantum Hall state (Landau level filling factor $\nu = 5/3$) was reported by Manfra *et al.* nearly two decades ago.[10] In comparison with the more widely studied GaAs-based 2DEGs, electrons in GaN-based 2DEGs have significantly heavier effective masses ($\approx$0.24 m_0 vs $\approx$0.07 m_0 in GaAs, where m_0 is the bare electron mass), and the dielectric constant is smaller ($\epsilon \approx 9.5$ in GaN vs $\approx$13 in GaAs), so that enhanced electron-electron interactions are expected. In this regard, 2DEGs in GaN more closely resemble those found in other wide-bandgap semiconductors such as ZnO, where significant progress has recently been made.[11]

However, peak mobilities in GaN-based 2DEGs are, to date, typically achieved at relatively large electron densities of $n_e \sim 10^{12}$/cm^2,[12,13] so that high magnetic fields $B \gtrsim 40$ T are required to reach the so-called "quantum limit" wherein all the electrons reside in the lowest spin-polarized Landau level (i.e., $\nu \leq 1$). Such large B values are (just) within reach of modern superconducting-resistive hybrid magnet technologies but are routinely exceeded by pulsed magnets.[14] Pulsed fields can, therefore, enable detailed studies of high-density 2DEGs, including not only transport but also optical measurements that probe the response of the 2DEG to a photogenerated hole, which have historically proven to be a very powerful tool to measure screening and many-body effects in GaAs- and ZnO-based systems.[15–24]

To this end, we report both transport and optical studies of a high-mobility 2DEG in a GaN/AlGaN heterojunction in pulsed magnetic fields to 60 T. We demonstrate that beyond the quantum limit, transport measurements show clear evidence for the $\nu = 2/3$

fractional quantum Hall effect (FQHE) state. Moreover, simultaneous optical and transport studies reveal nearly synchronous quantum oscillations of both the photoluminescence (PL) intensity and the longitudinal resistivity; however, the optical illumination required to perform PL significantly (and persistently) increases n_e to the point where only $\nu \geq 3$ can be reached at 60 T in the same heterostructure.

The GaN/AlGaN structure [see the inset in Fig. 1(a)] was grown by MBE on a semi-insulating single-crystal GaN substrate with low dislocation density ($\sim 5 \times 10^4/\mathrm{cm}^2$), following Ref. 13. After the initial growth of a 300 nm GaN buffer layer, a thin 21 nm $Al_{0.07}Ga_{0.93}N$ barrier layer was grown. A high-mobility 2DEG formed naturally at the interface due to the spontaneous polarization discontinuity across the junction.[1] The structure was capped by a final 3 nm GaN layer. For transport studies, Ti/Au contacts were deposited and annealed at the corners of 3 mm × 3 mm squares in a van der Pauw geometry. The sample was mounted in a ^{3}He cryostat in a 60 T capacitor-driven pulsed magnet. The magnet pulse has a rise time of 9 ms and a total duration of ≈90 ms. Resistivity was measured using dc current, which avoided measurement-phase and RC time constant issues associated with high-frequency lock-in detection of high-resistance samples. Appropriate combinations of current and magnetic field direction were used to symmetrize the data and subtract off any induced voltages due to the rapidly-changing field, so that both the longitudinal (R_{xx}) and transverse (R_{xy}) magnetoresistance could be measured accurately.

Separately, photoluminescence (PL) measurements up to 60 T were performed on the same structure, which was mounted on a fiber-coupled probe and immersed in superfluid ^{4}He at 1.5 K for optimal heat sinking. Unpolarized excitation light from the 325 nm (3.82 eV) line of an HeCd laser was directed to the sample, and PL was collected from the sample, using a multimode optical fiber. The circular polarization of the PL was not resolved, due to a lack of cryogenic-compatible thin-film ultraviolet polarization optics. The PL was dispersed in a 300 mm spectrometer and measured using a fast charge-coupled device (CCD). Full spectra were continuously acquired every 0.6 ms throughout the pulse.[25]

In a final set of measurements, resistivity and PL were simultaneously measured in a 18 T superconducting magnet, using a fiber-coupled transport probe with the sample in superfluid ^{4}He at 1.5 K. AC excitation at 17 Hz and lock-in detection provided a measure of the longitudinal resistance, while high-resolution PL spectra were detected using a 500 mm spectrometer and a liquid nitrogen-cooled CCD.

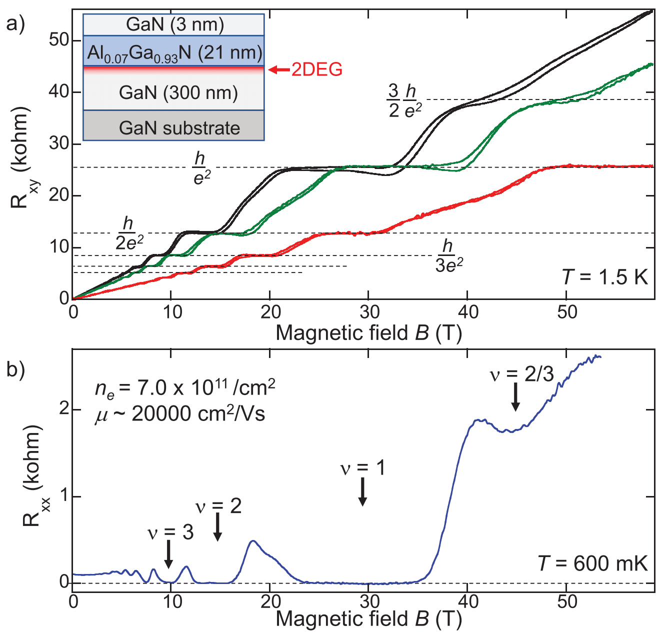

FIG. 1. (a) Inset: schematic of the GaN/AlGaN 2DEG structure. The plot shows the transverse resistance (R_{xy}) at 1.5 K up to 60 T, showing quantized Hall resistance $h/\nu e^2$ at integer filling factors ν. Data acquired during both the upsweep and downsweep of the pulsed field are shown. The black trace was acquired after three days in the dark, and the quantum limit ($\nu = 1$) is reached at ≈27 T. The red trace was then acquired after briefly illuminating the structure *in situ* with white light—note that n_e approximately doubled. The green trace was then acquired after cycling the temperature up to 300 K and back to 1.5 K in the dark. (b) Longitudinal resistance R_{xx} of this structure (separate cooldown; $\nu = 1$ is reached at ≈29 T). The DC current was 12 μA. Both R_{xx} and R_{xy} show evidence for the $\nu = 2/3$ FQHE state. Additional magneto-transport data acquired at different temperatures are shown in the supplemental material.

Figure 1(a) shows the transverse resistance R_{xy} up to 60 T, from which can be seen the overall signal-to-noise, drifts, and data quality that are achievable using dc transport methods to measure 2D electron systems in pulsed magnets. During these studies, it was observed that the 2DEG's conductivity varied from cooldown to cooldown, suggesting a history-dependent carrier density n_e. As demonstrated in early work,[3] these variations arise from the history of optical illumination on the sample, as shown below.

We focus first on the black curve in Fig. 1(a), which was acquired after the sample had been three days in the dark at 300 K. Well-defined IQHE plateaus were observed, with the quantum limit ($\nu = 1$) achieved at $B \simeq 27$ T, indicating 2DEG electron density $n_e = \nu eB/h \simeq 6.5 \times 10^{11}/\mathrm{cm}^2$. We note that while the IQHE has been reported many times in GaN-based 2DEGs,[4–9] these measurements are the first to explore transport in the FQHE regime beyond the quantum limit. Crucially, R_{xy} also shows an *additional* plateau forming at approximately 41 T, which coincides with the expected position (and quantized resistance $R_{xy} = 3h/2e^2$) of the $\nu = 2/3$ FQHE state.

The longitudinal resistance R_{xx} is shown in Fig. 1(b), which was acquired during a separate cooldown, again after several days in the dark. Zero-resistance minima confirm the IQHE and a slightly larger n_e ($\nu = 1$ occurs at 29 T; thus, $n_e \simeq 7.0 \times 10^{11}/\mathrm{cm}^2$). Most importantly, the pronounced dip at 44 T again strongly supports an interpretation in terms of the $\nu = 2/3$ FQHE state. While evidence for the fractional $\nu = 5/3$ state in a GaN/AlGaN 2DEG was observed previously,[10] we emphasize that Fig. 1 demonstrates for the first time that phenomena beyond the quantum limit ($\nu < 1$) are indeed accessible in GaN-based 2DEGs, here through the use of pulsed magnetic fields.

We now turn to the other R_{xy} curves in Fig. 1(a). After measuring the black curve, the sample was weakly illuminated at 1.5 K by an *in situ* white-light LED for several seconds. Then, the red curve was measured in the dark. IQHE plateaus were again observed, but significantly shifted in field: $\nu = 1$ occurred at ≈57 T, indicating over a twofold increase in n_e. Then, the sample was thermally cycled (in the dark) up to 300 K for 6 h and back to 1.5 K, and the green curve was measured.

IQHE plateaus were again observed, but $\nu = 1$ occurred at ≈ 32 T, indicating that n_e had (mostly) recovered back down to its initial value. IQHE plateaus in all curves show no obvious signs of inhomogeneity or disorder changes resulting from illumination.

As described in early[3] and more recent[9] studies, illumination has a significant and persistent effect on n_e in GaN/AlGaN 2DEGs. Optical excitation of electrons out of traps and impurity states leads to an immediate and persistent increase in n_e (changes occur on the timescale of 1 s[9]) that can be reset by thermal cycling in the dark. These data point to the fact that—at least in this particular structure—FQHE states with $\nu < 1$ are not accessible with 60 T fields if the sample has been recently illuminated. This limits the use of optical spectroscopy to study FQHE states (in this structure), which, in GaAs-based 2DEGs, have historically proven to be very powerful for probing electron correlations and novel phases.[15–23]

Optical phenomena related to the IQHE can, however, be studied quite well in GaN/AlGaN 2DEGs, even in low-field (<20 T) superconducting magnets. The PL spectrum from this GaN/AlGaN structure is shown in Fig. 2(a). The two bright peaks at 3.481 eV and 3.474 eV correspond to free "A"-excitons and to donor-bound excitons in the GaN buffer layer, respectively.[26] At lower energy, the broad and weak emission band in the 3.44 – 3.46 eV range corresponds to radiative recombination of photogenerated holes with electrons in the 2DEG, as depicted in the inset and as confirmed below.

The PL intensity map of Fig. 2(b) shows how this low-energy emission band evolves with B up to 18 T. Pronounced oscillations of the net PL intensity are observed, along with the formation of discrete peaks that shift linearly with B. These peaks reveal the formation and dispersion of discrete Landau levels in the 2DEG, separated by the electron cyclotron energy eB/m_e, from which an effective electron mass $m_e \simeq 0.24 m_0$ can be inferred. These data corroborate and extend recent results from the study by Schmult *et al.*,[8] who measured PL from a similar GaN/AlGaN 2DEG up to 15 T.

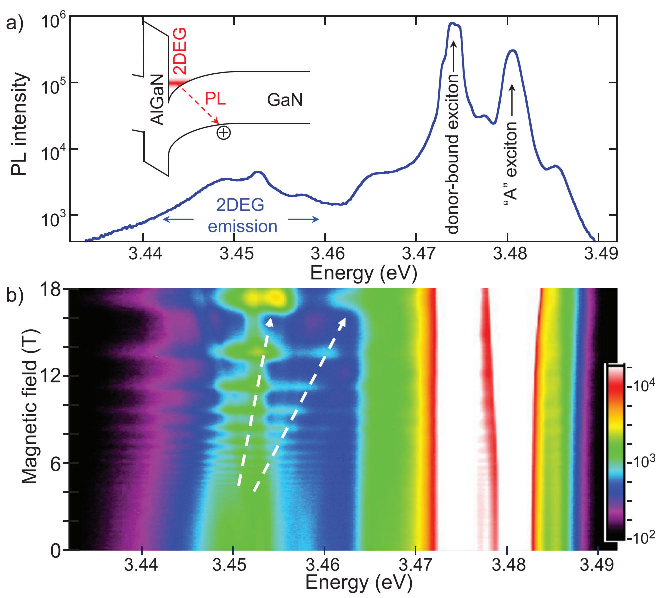

FIG. 2. (a) Photoluminescence (PL) spectrum of the sample at 1.5 K, on a log scale. A broad PL emission from radiative recombination of 2DEG electrons is located at low energies (~3.45 eV), below the sharp exciton-related PL peaks that originate from the GaN buffer layer. (b) An intensity map showing the PL spectra up to 18 T. SdH-like intensity oscillations are observed in the low-energy 2DEG region; discrete peaks reveal the formation and dispersion of 2DEG LLs (white dashed arrows), separated by the cyclotron energy eB/m_e.

Figure 3(a) shows how the PL intensity oscillations (red trace) compare with the simultaneously measured longitudinal resistivity R_{xx} (blue trace). Figure 3(b) shows the same data plotted against $1/B$: both curves have the same periodicity in $1/B$, as expected for Shubnikov-de Haas (SdH) quantum oscillations. However, the minima in R_{xx} – which accurately indicate integer filling factors – align only approximately with the maxima in the PL intensity. The latter exhibit a relative phase shift that is most noticeable at large B. Moreover, intensity oscillations at odd-integer ν, which manifest clearly in R_{xx} for $B > 7$ T, are not observed in the PL data up to 18 T.

Studies of SdH-like PL oscillations in 2DEGs have a long and rich history.[15–17,24,27–29] Typically, they arise from electron-hole correlations and the efficacy with which 2D electrons screen the Coulomb potential of a nearby photogenerated hole, which, in turn, modifies the spatial overlap of their respective wavefunctions and, therefore, changes the radiative recombination rate. In many models, screening is less effective when the 2DEG Fermi level lies between Landau levels

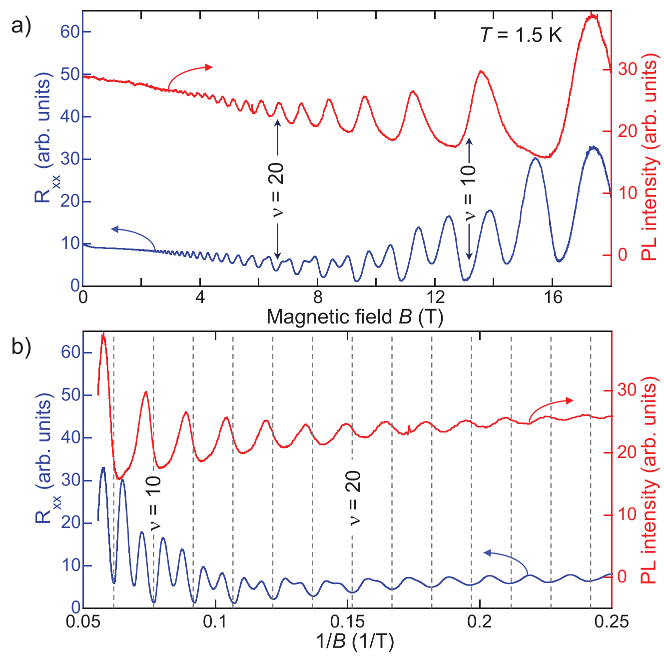

FIG. 3. (a) Correlating the B-dependent oscillations of the total 2DEG PL intensity (integrated from 3.440 – 3.464 eV; red trace) with the simultaneously measured resistance R_{xx} (blue trace) up to 18 T in a superconducting magnet, at 1.5 K. Due to the excitation laser, the steady-state 2DEG density is much higher than that in the dark (*cf.* Fig. 1); $n_e \simeq 3.1 \times 10^{12}$/cm^2, and $\nu = 1$ is not expected until ~130 T. (b) The same data vs $1/B$. Dashed vertical lines are equally spaced and aligned with R_{xx} minima and indicate even-numbered ν. Both the PL intensity and R_{xx} are periodic in $1/B$, but only the latter shows odd filling factors. Note that PL maxima are slightly shifted from even-numbered ν. A Dingle analysis of the low-field R_{xx} oscillations yields a quantum scattering time of $\tau_q \simeq 0.45$ ps (*cf.* a transport scattering time of $\tau_t \simeq 2.8$ ps given by the mobility).

(i.e., in localized states, at integer ν). On the one hand, this can lead to a reduced wavefunction overlap and smaller PL intensity; however, reduced screening can also increase electron-hole binding (exciton formation), increasing PL intensity.[15,17] The phase shift that is observed between the quantum oscillations of R_{xx} and PL (see Fig. 3)—which increases at large B—suggests that the interplay between these competing effects likely plays an important (and B-dependent) role. Additionally, the fact that R_{xx} clearly exhibits spin-resolved SdH oscillations (odd-integer ν) for $B > 7$ T, while the PL does not, also presents an unresolved puzzle (although $\nu = 3$ appears in PL studies to 60 T, shown below). Similar discrepancies between transport and PL were also observed in early GaAs 2DEG studies[27] as well as in recent GaN work.[8]

Importantly, Fig. 3 also shows that due to the weak ($\sim$100 μW) above-gap optical excitation that is used to enable PL measurements, the steady-state electron density is about 5× larger than when measured in the dark (*cf.* Fig. 1). In Fig. 3, $\nu = 10$ occurs at 13 T, indicating that $n_e \simeq 3.1 \times 10^{12}/\text{cm}^2$. A consequence of this photo-doping effect is that future optical studies of GaN-based 2DEGs in the $\nu \leq 1$ regime will require structures specifically designed to host lower carrier densities.

Nonetheless, to explore the limits of high-B magneto-optics in GaN-based 2DEGs, Fig. 4 shows PL studies of the same structure in a 60 T pulsed magnet. Due to the fast CCD exposure times that are used, fewer photons are collected, limiting signal:noise. Regardless, Fig. 4 clearly shows that the 2DEG emission continues to exhibit pronounced intensity oscillations. A maximum is observed at 17.5 T (the same as observed in Figs. 2 and 3) and then also at $B \approx 24$ T, 39 T, and 46 T. With the exception of the 39 T peak, these values are in reasonable agreement with the expected filling factors $\nu = 8$, 6, 4 and also $\nu = 3$, as indicated. The appearance of a PL maximum at the expected position of $\nu = 3$ suggests that detailed optical studies of *spin-resolved* many-body screening in GaN-based 2DEGs are possible. Future measurements incorporating *in situ* polarizers to resolve both right- and left-handed circular PL polarizations, ideally in conjunction with transport measurements performed on intrinsically lower-density 2DEGs, are expected to address this interesting regime.

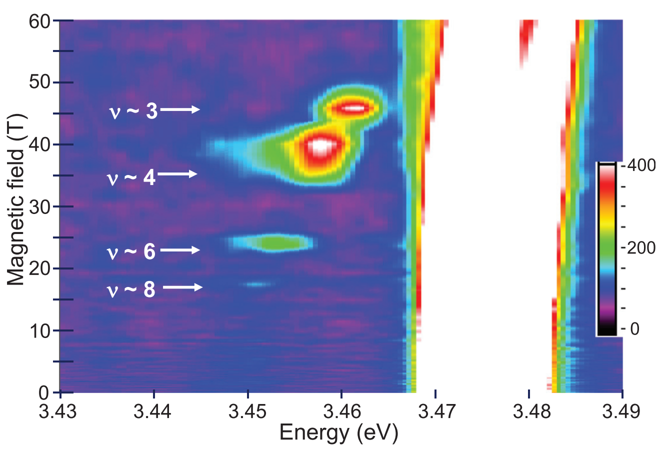

FIG. 4. PL measured up to 60 T. PL from the 2DEG brightens considerably at 17.5 T (as also shown in Figs. 2 and 3), 24 T, 39 T, and 46 T, which corresponds approximately to $\nu \approx 8$, 6, 4, and 3. Limited signal:noise precludes resolving the smaller intensity oscillations for $B < 15$ T.

Through the use of pulsed magnetic fields, these studies demonstrate that the $\nu = 1$ quantum limit can be exceeded in high-density GaN-based 2DEGs. First evidence for the $\nu = 2/3$ FQHE is shown by magneto-transport. PL measurements also reveal SdH-like oscillations of the intensity, from which n_e and the electron mass are inferred in a contactless approach. However, the increase in n_e associated with optical excitation currently limits PL studies to the IQHE regime although strong indications of 2DEG screening in the spin-resolved $\nu = 3$ state is observed at large fields.

See the supplementary material for figures that show additional pulsed-field magneto-transport data acquired at other temperatures.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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