Record-quality two-dimensional electron systems

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Abstract

Two-dimensional electrons confined to GaAs quantum wells are hallmark platforms for probing electron-electron interaction. Many key observations were made in these systems as sample quality improved over the years. However, progress in quality has been stagnant for over a decade. We present a major breakthrough via source-material purification and innovation in GaAs molecular beam epitaxy vacuum chamber design. Our new samples have a world-record mobility of $44 \times 10^6$ cm$^2$/Vs at an electron density of $2.0 \times 10^{11}$/cm$^2$; this is the highest mobility observed in any material. These results imply only $\sim 1$ residual impurity for every $10^{10}$ Ga/As atoms. The impact of such low impurity concentration is extraordinary; several new fractional quantum Hall states emerge, and exotic phases such as the $5/2$ state, which is widely believed to be non-Abelian and of potential use for topological quantum computing, and stripe/bubble phases are unprecedentedly robust.
Single crystal GaAs thin-film structures grown by molecular beam epitaxy (MBE) are often considered to be among the purest materials that can be made in the laboratory. Being nearly defect free, these structures provide an exceptional platform for exploring a diverse range of physical sciences, with extensive electronic and photonic applications. The highlight of ultra-high-quality GaAs films, however, is their utilization in the investigation of delicate electron-electron interaction phenomena. Typically this is achieved by studying the low-temperature magnetotransport of two-dimensional electron systems (2DESs) hosted in modulation-doped GaAs quantum wells, where the electrons are spatially separated from the dopants to reduce electron-impurity scattering. A magnetic field applied perpendicular to the 2DES enhances the relative scale of Coulomb energy in the system by quenching the Fermi energy via Landau quantization. A plethora of exotic many-body phases have materialized in GaAs 2DESs using this framework; the discovery of the odd- [1] and even-denominator [2] fractional quantum Hall (FQH) effects as well as the observation of Wigner solid [3–6], stripe/nematic [7, 8], and bubble phases [9] are some notable examples (for reviews, see, [10, 11]).

Naturally, these fascinating phases only started to emerge in experiments as sample quality increasingly improved. For example, after the first observation of the FQH effect [1], it took about two decades of growth condition improvements to realize stripe/nematic phases in GaAs 2DESs [7, 8]. The long history of success stories such as this continues to motivate the community to search for methods to produce ever-better-quality samples, as it is stimulating to anticipate what other rich physics is yet to be uncovered. It is worth noting that, while 2DESs in two-dimensional materials, such as graphene and transition-metal-dichalcogenides, have experienced tremendous improvement recently and reveal exquisite many-body phenomena [12], GaAs-based 2DESs continue to be a strong leader in the field; see, e.g., [13, 14].

A useful metric to quantify the quality of a 2DES is the electron mobility because it is simple to measure and is inversely proportional to the average electron scattering rate in the 2DES. High mobility values in 2DESs imply that electrons are less likely to scatter over prolonged trajectories in such samples, making them valuable platforms to study delicate many-body electron phases as well as ballistic or phase-coherent transport. After decades of development, electron mobility values as high as $\mu \simeq 35 \times 10^6$ cm$^2$/Vs have been observed in GaAs 2DESs with densities near $n \simeq 3 \times 10^{11}$ /cm$^2$ [15–18]. However,
despite constant subsequent efforts, the mobility of modern state-of-the-art GaAs 2DESs has been in a stalemate for more than a decade now. Here we present a major breakthrough in the MBE crystal growth of ultra-high-quality GaAs 2DESs that enhances the world record mobility to $\mu \simeq 44 \times 10^6$ cm$^2$/Vs at electron densities as low as $n \simeq 2 \times 10^{11}$/cm$^2$. This is the highest electron mobility value to ever be reported in any material. In the low-density ($n \lesssim 1.5 \times 10^{11}$/cm$^2$) regime, where scattering by residual (background) impurities is dominant, our samples show twice the mobility of the best previous samples. Low-temperature magnetotransport traces taken in a sample of this class show FQH states that have never been seen before in any 2DES, indicating great prospect for future studies of interaction-driven physics in the two-dimensional setting.

Numerical calculations have suggested that residual impurities are the limiting factor for mobility in the best available GaAs 2DESs [19, 20]. Recent efforts to systematically purify the Ga and Al source materials are in line with this understanding, as these metals are the most likely origin of unwanted impurities in a well-maintained, ultra-high-vacuum growth chamber [18, 21, 22]. Interestingly, despite the different history of the source materials loaded into the MBE chambers and the varying purification techniques, the best samples grown from the top groups in the world seem to be roughly tied in electron mobility [15–18]. These results hint that the impurities that are limiting the mobility in the highest-quality GaAs 2DESs come from somewhere other than the source material.

While the environment in ultra-high-vacuum MBE chambers is certainly very sparse in atomic/molecular density, it is not completely void of matter. Even in the best-equipped, conventional vacuum chambers, it is common that the mass spectrometer data of the growth space show traces of H$_2$O, N$_2$, O$_2$, CH$_4$, and their derivatives. If incorporated during growth, these species would act as impurities in the structure and cause degradation in the quality of the GaAs 2DES. Once the source materials become so pure that they are no longer the primary supplier of impurities to the growth environment, the background vacuum quality would determine the impurity concentration in the sample.

As shown in Fig. 1(a), we built an MBE chamber to verify this hypothesis. Our design includes conventional MBE chamber components such as a load-lock chamber and a liquid N$_2$ shroud, as well as three auxiliary cryo-cooled ($\sim 17$ K) cold plates that augment four large (3000 l/s) cryopumps to achieve extreme levels of vacuum during sample growth. The cold plates are made of Cu to maximize cooling power and are coated with Ni to prevent
FIG. 1. Improving vacuum quality and its assessment in a state-of-the-art MBE chamber. (a) Schematic diagram of the MBE chamber used in this study. In addition to four large (3000 l/s) cryopumps operating at ∼ 10 K, there are three auxiliary cryo-cooled, Cu cold plates (∼ 17 K on their back side) that pump the chamber. One ∼ 5 × 5 inch² cold plate is located in close proximity to the growth space while two ∼ 12 × 15 inch² cold plates are in the sump. (b) Sample structure used to analyze vacuum quality. If the source material is pure enough, the surface-segregation of impurities during the growth of the back AlGaAs layer can be exploited to evaluate the vacuum quality by analyzing the mobility measured in the GaAs quantum well. (c) Mobility of samples that have the structure shown in (b), with varying back AlGaAs thicknesses (d) and vacuum/source purity conditions. The 2DES density is \( n \approx 2.0 \times 10^{11} \) /cm². The data sets for different conditions are color coded, where the data shown in black, blue, and red were taken from samples grown in that chronological order; they correspond to samples grown with: (1) a fresh batch of sources, (2) purified sources, and (3) purified sources and the Cu cold plates turned on.

Possible corrosion when exposed to As and Ga that are ubiquitously present in the growth chamber. It is difficult to quantitatively assess how much the base pressure improved in our growth chamber with the cold plates operating because the ion extractor gauges installed in the chamber cannot reliably measure pressures below \( P \sim 2 - 3 \times 10^{-12} \) Torr, and the base
pressure already reaches this range even when only the four cryopumps are pumping on the chamber. However, when the cold plates are turned on, the mass spectrometer data shows a factor of 10 improvement in the partial pressures of $N_2$ and $O_2$ species and a factor of 2.5 improvement in the partial pressures of $H_2O$-related species.

Previously, we devised a scheme to systematically evaluate the cleanliness of the growth environment during the MBE of GaAs/AlGaAs heterostructures [22]. A brief summary of the concept is to use the mobility of a GaAs 2DES with the sample structure shown in Fig. 1(b) as a very sensitive probe to gauge impurity accumulation during growth. This method utilizes the fact that impurities surface-segregate on the growth front of the back AlGaAs layer and deposit at the AlGaAs/GaAs interface when the growing layer is changed from AlGaAs to GaAs. For a given back AlGaAs layer thickness, the mobility is lower when the growth environment is worse. The strength of this procedure is that the back AlGaAs layer thickness ($d$) can be made extremely large to detect even the most minute amounts of impurities incorporated in the crystal during the growth process.

Figure 1(c) shows the mobility of such structures grown in varying growth environments as a function of $d$. The much enhanced mobility of samples grown after the sources were sufficiently outgassed (blue symbols) compared to those grown with a fresh batch of source materials (black symbols) demonstrates the importance of preparing pure source materials when aiming for the cleanest growth conditions. Under these conditions, the source materials were clean enough that high-mobility GaAs 2DESs made from them displayed mobilities on par with the best samples in the literature. Despite further purification efforts, we could not obtain a significant improvement from the blue data set under the normal operating vacuum conditions of having four cryopumps operating. This implied that at this point, our source materials had been amply purified so that they were no longer the primary supplier of impurities for our samples. It is then plausible to assume that the background vacuum starts to play a more important role. Consistent with this assumption, samples grown with only one cryopump operating exhibited worse mobilities compared to those grown with all four pumps turned on. Following this test, we investigated a series of samples grown with all the cryopumps and the cold plates operating during growth (data shown in red in Fig. 1(c)). The data clearly reveal that there is a significant improvement in GaAs 2DES mobility at all values of $d$ compared to the case when the cold plates are off. Remarkably, the mobility of the samples grown with all cold plates operating sustains the high value of $\mu \simeq 8 \times 10^6$
cm$^2$/Vs even when $d = 10,000$ nm (data not shown).

These results clearly demonstrate that vacuum integrity plays a crucial role in determining the amount of unintentional impurities deposited on the sample during growth once the source material has been extensively purified. Consequently, we grew several GaAs samples with a wide range of 2D electron densities to investigate the impact of having unprecedented levels of ambient vacuum quality in the MBE chamber. Figure 2(a) compares the mobility of these samples with the best previous GaAs 2DESs from our group, which are similar to the best reported in the literature [15–18]. Figure 2(b) shows the layer structure of the samples used to obtain the data presented in Fig. 2(a). We used two types of structures, the standard modulation-doped structure and the doping-well structure (DWS) [23]. While the donor energy level, which determines the position of the Fermi level, is tied to the AlGaAs barrier in the standard modulation-doped structure, in the DWS it is tied to the narrow AlAs layers that flank a narrow GaAs doping quantum well [23]. The DWS is advantageous in comparison to the standard modulation-doped structure because the electrons confined to the AlAs layer in the doped region provide additional screening for the 2DES from both residual impurities and intentional dopant ions [20, 23]. It is striking that with the improvement in vacuum, even the standard modulation-doped samples (black circles in Fig. 2(a)) have significantly higher mobility values for all densities when compared to previous state-of-the-art DWSs (red open circles). This is particularly impressive considering that DWSs were necessary to achieve the best samples reported in the literature. It seems that the reduction in impurities from better vacuum conditions by implementing the cold plates is significant enough to overcome the lack of such screening in our standard modulation-doped structures.

Furthermore, when we grow DWSs with the cold plates operating, we see an even larger increase in the mobility (red solid circles in Fig. 2(a)). These record-quality samples display mobility values as high as $\mu \simeq 44 \times 10^6$ cm$^2$/Vs at the density of only $n \simeq 2.0 \times 10^{11}$/cm$^2$. Considering that, previously, the highest mobility of a GaAs 2DES was $\mu \simeq 35 \times 10^6$ cm$^2$/Vs at $n \simeq 3.0 \times 10^{11}$/cm$^2$, these results imply a tremendous improvement in sample quality. This is evident over a wide range of 2D electron densities in Fig. 2(a). When $n < 1.5 \times 10^{11}$/cm$^2$, our record-quality samples have mobility values that are roughly twice that of the best previous samples. For example, the mobility of our $n \simeq 1.0 \times 10^{11}$/cm$^2$ sample is $\mu \simeq 36 \times 10^6$ cm$^2$/Vs whereas the previously reported highest mobility values for
FIG. 2. Mobility vs. 2D electron density for the record-quality GaAs 2DESs. (a) Data from samples grown under the improved vacuum conditions are shown as solid symbols for the doping-well (red) and standard modulation-doped (black) structures. The mobility values from the best of our previous doping-well samples are shown as red open symbols for comparison. These previous samples have electron mobilities similar to the best samples reported in the literature [15–18]. The solid and dashed lines through each set of data points serve as guides to the eye, and their slopes are consistent with a power-law relation that is roughly $\mu \sim n^{0.7}$. (b) Layer structure of the samples used in (a). The spacer thicknesses and well widths of some representative samples can be found in the Supplementary Information.

GaAs 2DESs with a similar density are less than $\mu \simeq 18 \times 10^6$ cm$^2$/Vs [18, 24].

The power-law dependence observed for the mobility vs. 2D electron density profiles plotted in Fig. 2(a) is also noteworthy. Within the same category of samples, we observe a $\mu \propto n^{0.7}$ relation for all cases. A similar power-law dependence was reported in high-quality, low-density GaAs 2DESs with very large spacer-layer thicknesses, and it is usually interpreted as an indication that the mobility is limited by the residual impurities in the structure [25–27]; there is also theoretical justification for such an interpretation [28]. This understanding is certainly consistent with our results, as the primary improvement we have made to our samples compared to the previous data is the reduction of residual impurities in the structure. Based on models for the two types of structures used for our samples [20, 28],...
we estimate that the residual impurity concentration in our GaAs QWs is $\simeq 1 \times 10^{13} / \text{cm}^3$. Considering that there are $\sim 1 \times 10^{23}$ atoms/cm$^3$ in single-crystal GaAs, this means there is roughly one impurity for every 10 billion atoms in these samples.
As mentioned in the introduction, probing intricate many-body phenomena is a core application of ultra-high-quality GaAs 2DESs. In this context, we also studied the low-temperature ($T \simeq 30$ mK) magnetotransport of a representative record-quality sample with $n \simeq 1.0 \times 10^{11}$/cm$^2$. The specific layer structure for this sample is provided in Supplementary Information Table 1. Figure 3(a) shows a full-field longitudinal magnetoresistance ($R_{xx}$) trace of the sample, while Figs. 3(b) and (c) focus on specific regions near zero magnetic field and near $\nu = 1/2$, respectively; $\nu = h n/eB$ is the Landau level filling factor, where $h$ is the Planck constant, $e$ is the fundamental charge, and $B$ is the perpendicular magnetic field.

The sample reveals exceptional characteristics. For example, as seen in Fig. 3(b), there are prominent signatures of Shubnikov-de Hass oscillations up to $\nu = 106$ at $B < 0.04$ T. This implies that the Landau level broadening in this sample is smaller than the $B = 0.04$ T cyclotron energy gap of $h e B / m^* \simeq 68$ $\mu$eV ($m^* = 0.067$ is the effective mass of electrons in GaAs in units of the free-electron mass). In addition, the data plotted in Fig. 3(c) display high-order FQH states up to $\nu = 16/31$ and $\nu = 14/29$ on the left and right flanks of $\nu = 1/2$. The highest-order FQH states observed in the best previous GaAs samples with similar density are marked in black [24]. In total, nine extra FQH states are observed near $\nu = 1/2$ in our sample, whose lowest- and highest-order Landau level fillings are marked in red on each side of $\nu = 1/2$. For comparison, in the best quality monolayer graphene samples, the highest-order FQH states observed near $\nu = 1/2$ are $\nu = 8/15$ and $\nu = 7/15$ on the left and right flanks of $\nu = 1/2$ at similar temperatures but higher magnetic fields ($\sim 14$ T) [12]; the data presented in Fig. 3 exhibits 15 additional FQH states with respect to these samples.

The outstanding quality of our samples in this density range is also noticeable at higher Landau level fillings. Figures 4(a) and (b) show expanded $R_{xx}$ traces of Fig. 3 sample near $\nu = 3/2$ and $\nu = 5/2$, respectively. Remarkably, even at this relatively low density, FQH states up to $\nu = 20/13$ are observable in the vicinity of $\nu = 3/2$. Furthermore, the $\nu = 5/2$ and other FQH states in the second orbital Landau level ($N = 1$) are extraordinarily strong, considering that they occur at $B < 1.9$ T. In fact, the activation gap we measure for the $\nu = 5/2$ FQH state is $5/2 \Delta \simeq 820$ mK (see Supplementary Information Fig. S2), surpassing the previous record of $5/2 \Delta \simeq 625$ mK by a significant margin [29]. This is especially impressive considering the fact that the density in our sample is only $\sim 1/3$ of the sample with the previous record value. Given the potentially non-Abelian nature of the $\nu = 5/2$
FIG. 4. Low-temperature ($T \simeq 30$ mK) magnetotransport data of $n \simeq 1.0 \times 10^{11}$/cm$^2$ samples at higher Landau level fillings. (a) Magnetoresistance data near $\nu = 3/2$ ($N = 0$ orbital Landau level) and (b) $\nu = 5/2$ ($N = 1$). The number of observed FQH states as well as the strengths of each of the features are remarkable, considering the relatively low density of the sample. (c) Longitudinal (black and red) and Hall (green) resistance data for the high-index ($N = 2$ and 3) orbital Landau levels. The presence of reentrant integer quantum Hall states and stripe/nematic phases are evident at magnetic fields $B < 1$ T. The inset shows the configuration of current ($I$) and voltage ($V$) contacts used for the measurements.

FQH state [30, 31], the data presented here have exciting implications for the realization of fault-tolerant, topological quantum computing devices; using our samples, significantly more robust qubit operations could in principle be performed at much lower magnetic fields.

Figure 4(c) shows magnetotransport data of a different sample with a similar density of $n \simeq 1.1 \times 10^{11}$/cm$^2$ in magnetic field ranges that correspond to higher ($N = 2$ and 3) orbital Landau levels. Again, the exceptional sample quality is corroborated by the fact that well-quantized, reentrant integer quantum Hall states, as well as signatures of stripe/nematic phases, are observable at magnetic fields $B \lesssim 1.0$ T. These correlated states are extremely fragile, and are typically only observed in much higher density samples at larger magnetic fields and lower temperatures [7, 8, 13, 32].
Our results suggest a bright future for the investigation of interaction-driven physics in GaAs 2DESs. The emergence of several FQH states that have never been seen before, as well as the unprecedented robustness of fragile correlated phases such as the \( \nu = 5/2 \) FQH, reentrant integer quantum Hall, and stripe/nematic states, indicate that invigorating opportunities await us in fundamental studies and pragmatic applications alike. Moreover, we have experimentally demonstrated that vacuum integrity limits sample quality in current state-of-the-art MBE-grown GaAs. This gives a clear direction for further improvement in the quality of GaAs 2DESs. Some mysteries have also developed. When \( n \geq 1.5 \times 10^{11}/\text{cm}^2 \), the electron mobilities seem to deviate to lower values than the power-law relation \( \mu \propto n^{0.7} \) would predict. We are currently unsure of the origin of this behavior. It is possible that the remote ionized impurities from the intentional dopant atoms become relevant as a smaller spacer thickness is required to achieve higher 2D electron densities. If this is the case, in the future it may be useful to start from a low density sample with large spacer thickness and increase the density by applying gate voltages to circumvent this issue. Another option is to vary structural parameters in the doped region of the DWS so that the spacer thickness can be increased while maintaining the same 2DES density in the main quantum well [23].
Methods

Sample preparation

All of our samples are grown on 2-inch-diameter GaAs substrates in the vacuum chamber setup shown in Fig. 1(a). The substrates are outgassed for 30 minutes at $T \simeq 610 \, ^\circ C$ in an As beam flux of $P \sim 6.0 \times 10^{-6}$ Torr prior to growth. We always confirm clear single-crystalline features in the reflection-high-energy-electron-diffraction (RHEED) patterns of the substrate after this process. The substrate temperature is typically $T \simeq 640 \, ^\circ C$ during growth. The deposition rate of GaAs is calibrated to be $\simeq 2.83 \, \AA/s$ for all growths by tuning the temperature of the Ga oven based on RHEED oscillations. We tune the Al growth rate in a similar fashion to obtain the barrier alloy fraction of choice. The barrier alloy fraction is 32% for the samples whose data are shown in Fig. 1(c) while the record-quality samples use a stepped-barrier structure with alloy fractions 24% and 12%. The specifics of the sample structure of the record-quality samples can be found in Supplementary Information Section I.

Transport measurements

We performed all electronic measurements in the van der Pauw configuration using low-frequency lock-in amplifiers. Our samples have a square shape and a typical size of 4 mm×4 mm. The mobility values of the GaAs 2DESs are evaluated in a $^3$He cryostat with a base temperature of $T \simeq 0.3 \, K$. A simple Drude formula $\mu = 1/\rho ne$ is used to obtain the mobility, where $n$ is the 2DES density, $e$ is the fundamental electron charge, and $\rho$ is the resistivity of the 2DES. Quantum Hall features in the magnetoresistance data are used to deduce $n$. For $\rho$, we take the average value of the resistance ($R_{ave}$) measured between all the four-probe contact configurations in the sample, and use the standard van der Pauw geometry expression $\rho = \pi R_{ave}/ln(2)$. The low-temperature magnetotransport data presented in the main text are measured in a dilution refrigerator with a base temperature of $T \simeq 30 \, mK$. A red light-emitting diode is used to illuminate the samples briefly at $\sim 10 \, K$ for the samples in the $^3$He cryostat and $\sim 4 \, K$ for the samples in the dilution refrigerator before they are cooled down to base temperatures. For the magnetotransport measurements, a magnetic field sweep rate of 1 T/hour is used unless specified differently.

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

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Supplementary Information for “Record-quality two-dimensional electron systems”

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FIG. S1. Sample structure for the record-quality GaAs two-dimensional electron systems whose data are shown in the main text. The spacer layer thickness is varied to obtain the desired sample density, and the quantum well width is chosen accordingly to prevent second-subband occupation.

All record-quality samples follow the structure shown in Fig. S1. Standard modulation-doped structures are δ-doped in AlGaAs while the doping-well structures (DWSs) use the doping scheme described in the right panels of the figure. A 12%/24% stepped-barrier structure is implemented to reduce the Al composition of the barrier directly in contact with the main quantum well. The thickness of each of the barrier layers is controlled so that no parallel channel forms at the 12%/24% barrier interface, and the total spacer thickness is varied to attain the desired sample density. The well width of the quantum well is varied for each sample so that there is no second-subband occupation. Specific structural parameters are summarized in Table S1 for some representative samples.
| $n \times 10^{11} / \text{cm}^2$ | $\mu \times 10^6 \text{ cm}^2 / \text{Vs}$ | $s_1 \text{ (nm)}$ | $s_2 \text{ (nm)}$ | $w \text{ (nm)}$ |
|-------------------------------|---------------------------------|------------------|------------------|------------------|
| 2.06                          | 44.0                            | 68.2             | 60.0             | 34.0             |
| 1.15                          | 41.5                            | 114              | 100              | 45.3             |
| 1.00                          | 36.0                            | 136              | 120              | 50.0             |
| 0.71                          | 29.0                            | 195              | 171              | 58.5             |

TABLE S1. Structural parameters of some representative record-quality doping-well-structure samples whose data are shown in the main text; for definitions of $s_1$, $s_2$, and $w$, see the sample structure in Fig. S1.
II. MEASUREMENT OF ENERGY GAP FOR THE $\nu = 5/2$ FRACTIONAL QUANTUM HALL STATE

Magnetoresistance traces were taken at a sweep rate of 0.1 T/hour in the vicinity of the $\nu = 5/2$ fractional quantum Hall state to precisely determine the magnetic field position of the $R_{xx}$ minimum at base temperature. Then the energy gap $5/2 \Delta$ was measured by raising the temperature of the sample while monitoring the sample resistance at the magnetic field corresponding to $\nu = 5/2$. The data are shown in the Arrhenius plot of Fig. S2. A line corresponding to $R_{xx} \sim e^{-5/2 \Delta / 2k_BT}$ was fitted to the data plotted in Fig. S2 (shown in red), yielding an energy gap of $5/2 \Delta = 820$ mK. Here $k_B$ is the Boltzmann constant.

FIG. S2. Arrhenius plot of resistance at $\nu = 5/2$. The sample density is $n \simeq 1.0 \times 10^{11} / \text{cm}^2$. $5/2 \Delta = 820 \text{ mK}$