Surface transport and quantum Hall effect in ambipolar black phosphorus double quantum wells

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Quantum wells (QWs) constitute one of the most important classes of devices in the study of two-dimensional (2D) systems. In a double-layer QW, the additional “which-layer” degree of freedom gives rise to celebrated phenomena, such as Coulomb drag, Hall drag, and exciton condensation. We demonstrate facile formation of wide QWs in few-layer black phosphorus devices that host double layers of charge carriers. In contrast to traditional QWs, each 2D layer is ambipolar and can be tuned into n-doped, p-doped, or intrinsic regimes. Fully spin-polarized quantum Hall states are observed on each layer, with an enhanced Landé g factor that is attributed to exchange interactions. Our work opens the door for using 2D semiconductors as ambipolar single, double, or wide QWs with unusual properties, such as high anisotropy.

RESULTS

Here, we focus on a device with BP that is ~20 nm thick with Si/SiO₂ back gate and hBN/Al₂O₃/Au top gate at T = 0.3 K. The schematic of the device is shown in Fig. 1A, and an optical image is shown in Fig. 1B. The devices are measured in a pumped He⁴ or He³ cryostat. Similar data are observed in multiple devices. Figure 2A presents the four-terminal resistance R of the device (color) as a function of top gate V₉g (horizontal axis) and back gate Vbg voltages (vertical axis), where green and brown colors indicate conductive (R ~ 10⁷ to 10⁹ Ω) and highly resistive (R ~ 10¹⁰ to 10¹² Ω) states, respectively. Because of the thinner dielectric layers and higher dielectric constant of Al₂O₃, the coupling efficiency of the top gate is approximately four times that of the back gate. The white area in the center of the plot corresponds to an insulating regime (R > 10⁸ Ω) where the high resistance saturates the amplifier, indicating...
that the Fermi level is within the bandgap. Ambipolar transport can be attained by modulating either $V_{bg}$ or $V_{tg}$. The device is very conductive when highly hole-doped, with a high field effect mobility $\sim$6000 cm$^2$/Vs; because either gate is tuned close to 0, its resistance increases precipitously; when the gate voltages are highly positive, electron conduction is turned on, with a field effect mobility $\sim$1000 cm$^2$/Vs. This electron-hole asymmetry in mobility has been observed previously (53, 55, 59) and attributed to the Cr/Au electrodes that favor contacts to p-doped semiconductors. Another notable feature is the triangular shape of the insulating region: Its lower boundary at negative gate voltages ($-6.5 < V_{tg} < 3$ and $-40 < V_{bg} < -5$) moves with a negative slope, indicating that the “on”-state threshold voltage in the hole-doped regime is dependent on the total charge density induced by both gates; in contrast, the boundaries at positive gate voltages ($V_{bg} \sim 14$ V and $V_{tg} \sim 6$ V, respectively) remain constant, signifying that the on-state threshold voltage in the electron-doped regime is controlled by a single gate. We attribute the flat upper and right boundaries to bulk impurities that give rise to unoccupied localized states within the bandgap; because the Fermi level is located very close to the valence band, these states have minimal effect on the transport of BP in the hole-doped regime. Because an increasing gate voltage raises the Fermi level toward the conduction band, these unoccupied mid-gap states must first be filled. Thus, the back (top) gate cannot effectively switch on the top (bottom) surface state because of the large number of localized states within the bulk of the BP device. This effect, in combination with the Fermi level pinning that reduces the effect of gating, results in the flat boundaries of the insulating region.

Strikingly, at very large positive or negative $V_{tg}$ values, the insulating region almost completely disappears. For instance, at $V_{tg} = -8$ V, the resistance maximum near the charge neutrality point is $\sim$600 k$\Omega$, which is reduced from the global resistance maximum by more than two orders of magnitude. Such a disappearance of the insulating region may arise from the nontrivial closure of the bandgap by a large out-of-plane electric field (47); alternatively, it could also be a consequence of a nonuniform charge distribution, in which spatially separated surface states contribute to the transport even though the overall net charge is zero.

To determine the origin of the disappearing insulating region, we perform magnetotransport measurements. Figure 2C plots $R(V_{bg}, V_{tg})$ at a magnetic field $B = 18$ T, where quantum oscillations are visible. Several different patterns are observed, indicating distinct transport regimes. We first focus on the lower left quadrant: For negative $V_{bg}$ and $V_{tg}$, a checkerboard pattern is observed. The presence of horizontal and vertical sets of oscillations indicates the coexistence of two separate high-mobility 2D hole systems (2DHS), each independently tuned by the adjacent gate. The absence of diagonal features in this quadrant indicates that

**Fig. 1.** Device schematics and image. (A) Side view of device schematics. (B) Optical microscope image of an hBN/BP/hBN stack and a finished device without top gate (inset).

**Fig. 2.** Transport data. (A and B) $R(V_{bg}, V_{tg})$ and line traces $R(V_{bg})$ at different $V_{tg}$ at $T = 1.7$ K and $B = 0$. Note the logarithmic color scale (in Ω). (C) Right: $R(V_{bg}, V_{tg})$ at $T = 0.5$ K and $B = 18$ T, featuring a complicated quantum oscillations pattern. The color scale is in kΩ. Left: Schematics of the charge distributions that correspond to bipolar double-layer, single-layer, and unipolar double-layer regimes, respectively. Inset: Charge types for top and bottom surfaces at different combinations of gate voltages. p, hole-doped; n, electron-doped; i, intrinsic insulating state. (D) Band diagrams that correspond to the three regimes in (C), with dots illustrating mid-gap impurity states.
the effect of the farther gate on each layer is fully screened. From a simple Schrödinger-Poisson calculation, the gate-induced 2D hole wave function is expected to tightly confine to the outermost five or six atomic layers at the surface (60), thus limiting the screening length to <4 nm. Thus, the device forms a “naked” wide QW, with two distinct 2DHS residing at the top and bottom surfaces, separated by an intrinsic or insulating region. Similarly, the upper right quadrant of the figure corresponds to the formation of two distinct 2D electron systems (2DES) that, in principle, will display similar checkerboard patterns of oscillations at sufficiently high fields. However, because of the relatively low electron mobility, quantum oscillations are not observed at \( B = 18 \) T.

In the upper left (lower right) quadrants, that is, when both \( V_{tg} \) and \( V_{bg} \) have large magnitudes but different signs, only vertical (horizontal) oscillations are observed. Here, similar to the unipolar case, the device hosts top and bottom surface states; however, what distinguishes this case is that the states carry charges of opposite signs. Because the electron-doped regime has lower mobility, quantum oscillations are not observed; thus, a single set of oscillations emerges parallel to (that is, independent of) the axis that corresponds to the farther gate. Hence, these quadrants correspond to a wide QW with 2DHS and 2DES on opposite surfaces, which has not been realized in GaAs-based devices.

Last, when either \( V_{tg} \) or \( V_{bg} \) are tuned close to 0, we observe only a single set of diagonal oscillations, that is, the charge density is controlled by both \( V_{tg} \) and \( V_{bg} \). In this regime, one of the surfaces is tuned into the intrinsic regime and no longer screens the nearby gate; thus, the remaining surface state in this QW is subjected to field lines from both the back and top gates. The configurations of the top and bottom surface states that correspond to various regions of the \( R(V_{bg}, V_{tg}) \) map are summarized in the inset and left of Fig. 2C and the band diagrams in Fig. 2D, where the hole \((h)\)-doped, electron \((e)\)-doped, and intrinsic \((i)\) states are represented by red, blue, and white regions, respectively. These configurations also establish that the disappearance of the insulating region arises from the formation of 2DHS or 2DES on either surface at large doping.

**DISCUSSION**

To summarize our experimental observations thus far, we demonstrate that a thin BP device acts as a wide QW, which can host surface states on the top and bottom surfaces, whereas the interior is a gapped intrinsic semiconductor that acts as a soft tunnel barrier. Unlike conventional GaAs-based counterparts, these BP-based QWs support both single- and double-layer states that are exceedingly tunable, as each of the top and bottom surfaces may be independently tuned to intrinsic, 2D electron gas or 2D hole gas states. These novel wide QWs may be further optimized by improving mobility or reducing the BP flake thickness and, hence, the center barrier width so as to enhance Coulomb interactions between the surface states and tuning parameters, such as the symmetric-antisymmetric gap, thus allowing investigation of 2D correlated physics, such as two-component solid, Wigner crystals, interlayer coherence, and reentrant integer and fractional QH states (61–63) with charges of either or both polarities.

Furthermore, we can extract information about the 2D surface states by analyzing the temperature and density dependence of the quantum oscillations. Here, we focus on the \((p:p)\) region. Figure 3 (A and B) plots the background-subtracted resistance \( \Delta R(V_{bg}) \) at different temperatures and at constant \( V_{tg} = -6 \) and -4.4 V, respectively. The oscillation amplitudes decrease with temperature and can be fitted within the Lifshitz-Kosevich approach for 2D systems, yielding an effective mass \( m^* \approx 0.43 \pm 0.1 m_e \), where \( m_e \) is the electron rest mass. This value is in good agreement with that obtained from density functional theory calculations (53). Notably, we find no clear density dependence of \( m^* \) within error bars.

A close examination of Fig. 3 (A and B) reveals a salient feature: The oscillation amplitude is not monotonic in density; at some gate voltages, the peak height alternates between adjacent oscillations, as indicated by arrows. These nonmonotonic and/or alternating peak heights are not expected in conventional quantum oscillations, where the equally spaced Landau levels at constant \( B \) yield oscillation amplitudes that scale as \( \alpha^{-1/2} \). They have been observed in a number of systems, such as ZnO heterostructures (64), Si inversion layers (65, 66), SrTiO\(_3\) (67), and, more recently, thin BP sheets (53, 55), and are commonly attributed to the appearance of the Zeeman gap that is smaller than the single particle cyclotron gap, where the oscillation amplitude is given by (67)

\[
\frac{\Delta R}{R_0} = \frac{5}{2} \sum_{s=1}^{\infty} b_s \cos \left( \frac{2\pi \hbar}{2eB} - \frac{\pi}{4} \right)
\]

where \( h \) is the reduced Planck constant, \( g \) is the Lande \( g \) factor, \( n \) is the carrier density, \( \omega_c = eB/m^* \) is the cyclotron frequency, \( m^* \) is the effective mass, \( k_B \) is the Boltzmann constant, \( T_D \) is the Dingle temperature, and \( s = 1, 2 \). In these equations, the periodicity of the oscillations is controlled by \( n \), and our data yield a capacitive coupling \( \sim 6.5 \times 10^{10} \) cm\(^{-2}\) V\(^{-1}\) between

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**Fig. 3. Quantum oscillations at \( B = 18 \) T.** (A) Background-subtracted resistance \( \Delta R \) at \( V_{bg} = -6 \) V and \( T = 0.6, 1, 1.8, 4, 6, \) and 9 K, respectively. Arrows indicate nonmonotonic amplitude dependence on density. (B) Similar data set at \( V_{bg} = -4.4 \) V. (C) Oscillation amplitude as a function of temperature at \( V_{bg} = -3 \) V and different \( V_{tg} \) values (squares), fitted to Lifshitz-Kosevich formula (solid lines). The fits yield an effective mass \( m^* = 0.43 \pm 0.1 m_e \). (D) \( \Delta R(V_{bg}) \) at \( V_{tg} = 0 \) V and \( T = 1 \) K (solid lines) and fitted curve using Eq. 1 (dashed line), \( T_0 = 2 \) K and \( g m^* = 1.15 \).
the back gate and the bottom surface state. The ratio between the alternative peak heights is controlled by the combined product $g_n^n$. By fitting $AR(V_{bg})$ curves to Eq. 1, we obtain a good agreement using $T_H = 2 K$ and $g_{n,0} = 1.15 \pm 0.05$ (Fig. 3D). As we have determined $n^e \sim 0.43$ from the temperature dependence of the oscillations, the fitting results indicate that the Landé $g$ factor is $\sim 2.7$, which represents a $-33\%$ enhancement over the free hole value of $2.0$. This enhancement likely originates from the exchange interaction among electrons in spin-polarized Landau levels (68–70).

Finally, at sufficiently high magnetic field, the QH effect, which is a prototypical 2D phenomenon, can be observed on both 2DHIS. Figure 4A exhibits the Landau fan $R(V_{bg}, B)$ at $V_{bg} = 0$ V for $18 < B < 31$ T, and several line traces are shown in Fig. 4B. At $V_{bg} = 0$ V, the top layer is turned off, and only the bottom layer participates in electrical transport. Quantized plateaus at filling factor $\nu = 1, 2, 3, 4, 5, 6$ are observed, indicating full lifting of the spin degeneracy. On the other hand, at $V_{bg} = -8$ V, the $R(V_{bg}, B)$ data exhibit additional vertical strips superimposed on top of the Landau fan, signifying the presence of QH states on the highly hole-doped top layer (Fig. 4C), with an estimated hole density of $2.4 \times 10^{12}$ cm$^{-2}$. No quantized plateau is observed in the raw data (Fig. 4D, dashed lines) because of the coexistence of QH states on both the top and bottom surfaces. Because $B$ sweeps from 18 to 31 T, the filling factor of the top surface state is estimated to decrease from $\nu = -4.5$ to $-2$; thus, we model its conductance as stepwise quantized plateaus at appropriate filling factors (Fig. 4C, inset). By subtracting this calculated parallel conductance from the raw data, plateaus are recovered in $R(V_{bg})$ data, similar to those in Fig. 4 (A and B). Together, these results indicate that both the top and bottom 2DHS host QH states, with spin degeneracy fully lifted.

In conclusion, we have demonstrated the formation of ambipolar highly tunable wide QWs in hBN-encapsulated dual-gated BP devices, in which either or both surfaces may be tuned into intrinsic insulator or hole- or electron-doped surface states. At high magnetic fields, fully spin-resolved integer QH states are observed in both the top and bottom surface states. By further optimization, such as mobility improvement and thickness reduction, these BP-based wide QW devices may open the door to a wide range of novel physics in this highly anisotropic 2D system, ranging from Wigner crystallization and interlayer coherence to integer and fractional QH states with possible reentrant behavior.

**MATERIALS AND METHODS**

Bulk hBN and BP crystals were grown via high-temperature and high-pressure techniques (39) and were exfoliated into thin sheets onto Si/SiO$_2$ substrates. A dry transfer technique was used to assemble hBN/BP/hBN stacks (58) inside a Vacuum Technology Inc. glove box with moisture and oxygen concentration <0.1 parts per million. The top hBN layer was etched in SF$_6$ plasma to expose the BP layer, and Cr/Au electrodes were deposited thereafter by electron beam evaporation. To fabricate the top gate, a dielectric layer of 50-70 nm Al$_2$O$_3$ was deposited onto the entire stack. The devices were measured in an He$^+$ cryostat using standard direct current or lock-in techniques at the National High Magnetic Field Laboratory at magnetic fields ranging from 0 to 30 T.

**REFERENCES AND NOTES**

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