Flavoured quantum Hall phase in graphene/CrOCl heterostructures

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Quantum Hall effect (QHE), the ground to construct modern conceptual electronic systems
with emerging physics [1–5], is often much influenced by the interplay between the host two-
dimensional electron gases and the substrate, sometimes predicted to exhibit exotic topological
states [6, 7]. Yet the understanding of the underlying physics and the controllable engineer-
ing of this paradigm of interaction remain challenging. Here we demonstrate the observation
of an unusual QHE, which differs markedly from the known picture, in graphene samples in con-
tact with an antiferromagnetic insulator CrOCl equipped with dual gates. Owing to the pecu-
uliar interfacial coupling, Landau levels in monolayer graphene remain intact at negative filling
fractions, but largely deviated for the positive gate-doping range. The latter QHE phase even
presents in the limit of zero magnetic field, with the consequential Landau quantization following a
parabolic relation between the displacement field D and the magnetic field B. This characteris-
tic prevails up to 100 K in a sufficiently wide doping range from 0 to 10^{13} cm^{−2}. Our findings thus
open up new routes for manipulating the quantum electronic states, which may find applications
in such as quantum metrology.

The QHE is found in a number of solid-state systems to demonstrate topologically protected dissipation-less edge
channels with their transversal conductance quantized by $\frac{e^2}{h}$, with $e$ and $h$ being the elementary charge and
the Planck constant, respectively [8,12]. This peculiar behaviour has been crucial for such as the implementa-
tion of quantum-based resistance standards with extremely high precision and reproducibility [13]. Among
the few known systems manifesting QHE, graphene received special attention for its distinct band structure and
the resulting $N^{th}$ Landau level (LL) at the energy of $\varepsilon_{LL}(N) = \text{sgn}(N)\sqrt{2\epsilon h B |N|}$ under a fixed magnetic
field $B$. Due to the linear transformation of energy and the square root of carrier density $n$ [14,16], the Landau
quantization of conventional graphene in the parameter space of $B$ and $n$ is defined as the famed Landau fan dia-
gram, with all LLs linearly extrapolated to the charge neutrality point [15,17].

Interfacial coupling is known to be an important fac-
tor to affect the QHE in graphene, usually by two different means: charge impurities that cause reduced mobility
leading to wider QH plateaux at some circumstances [13], and charge transfer that to some extent shifts the
effective doping [18,23]. Recent theorems predict that the interplay between an antiferromagnetic insulator sub-
strate and a graphene layer can give rise to topological quantum ground states such as quantum anomalous
Hall phases [3,7,24]. Experimentally, interfacial coupling from such as RuCl$_3$ to graphene is indeed spotted,
with strong charge transfer, which is sometimes possibly coupled to the magnetism [25] while sometimes not fully
evidenced so [26]. To date, understanding of the under-
lyring physics and controllable engineering of the interfa-
FIG. 1. Characterization of CrOCl supported graphene. (a) Field effect curves of graphene encapsulated with h-BN and/or CrOCl, with each configuration illustrated in the schematic cartoon in the inset. (b) Schematics of the crystallographic structure of CrOCl. (c) Optical micrograph image of a typical h-BN/graphene/CrOCl sample, with its cartoon illustration shown in (d). Scale bar is 5 µm. (e) Color map of a dual gate scan of field effect in a typical sample, measured at a temperature of $T = 3$ K and at magnetic field $B = 0$.

cial coupling between the specific insulator substrate and graphene, especially in the QHE regime remains largely unknown.

In this work, we investigate the case of monolayer graphene interfaced to CrOCl, an antiferromagnetic insulator with Cl atoms as the ending bonds at its surface. By examining multiple configurations of graphene encapsulated with h-BN and/or CrOCl, we mapped out the peculiar interfacial coupling between the carbon honeycomb lattice and CrOCl in the parameter space of temperature $T$, total gate doping $n_{\text{tot}}$, magnetic field $B$, and electrical field $D$. At low temperatures, where
the CrOCl bulk is totally insulating, strong interfacial coupling (SIC) is found at positive gate voltages. At finite magnetic fields, it leads to a gate-tunable crossover from fan-like to cascades-like Landau quantization. In the regime of positive total electron doping, a QHE phase with parabolic dependence between $B$ and $D$ is obtained, with the Landau quantization reaching the zero magnetic field limit. This way allows one to effectively engineer the QHE in this hybrid system, with a $\nu = \pm 2$ plateau starting from as low as sub 100 mT and prevails up to 100 K in a wide doping range from 0 to $10^{13}$ cm$^{-2}$. Our results open up new routes for manipulating the quantum electronic states via SIC, which may lead to experimental applications in such as quantum metrology in the Système International d’unités.

Characterizations of graphene/CrOCl heterostructures.

Monolayered graphene as well as thin CrOCl flakes and encapsulating hexagonal boron nitride (h-BN) flakes were exfoliated from high-quality bulk crystals and stacked in ambient condition using the dry transfer method. The vertically assembled van der Waals heterostructures were then patterned into Hall bars with their electrodes edge-contacted. As seen in Fig. 1a, the field-effect curve of h-BN/graphene/CrOCl samples (red curve) differs significantly from the conventional h-BN/graphene/h-BN ones (blue curve), with the resistive Dirac peak disappearing and the conductivity, in general, is enhanced compared with the former structure but with degraded gate tunability (art views of each configurations are illustrated in inset of Fig. 1a). Figure 1b shows the crystal structure of CrOCl, which consists of a Cr-O double-layer sandwiched between the Cl atom layers stacked along the $c$-axis. The extremely high electronegativity of Cl atoms may affect the carrier constructions of the 2D materials assembled on it. It is noticed that, once contacted with graphene, the few-layered CrOCl exhibits weak bulk conductivity at temperatures above $\sim 200$ K, as discussed in Extended Data Fig. 1-2. Therefore only top gate is allowed at room temperature, as bottom gate leakage currents will occur.

When cooled down, those CrOCl interfaced graphene samples equipped with one single bottom gate showed a restore of bipolar type of field effect curve, as if the sample was back to “normal” graphene (Extended Data Fig.3). However, at finite magnetic fields, Landau fans in them are found to be asymmetric with respect to the presumed charge neutrality point, with the transition from a fan-like diagram in the hole side to a cascades-like diagram in the electron side (Extended Data Fig.4). Low field Hall coefficient analysis indicates that hole carrier type dominates in the whole gate range, even though the sample is set to a positive gate voltage (Extended Data Fig.5a). Especially, there is excessive hole doping in response to a certain electron doping induced by the gate (Extended Data Fig.5b). This is a strong indication that a peculiar SIC is taking place at the interface of CrOCl and graphene. As shown in Extended Data Fig.6, scanning tunneling microscopy (STM) characterizations suggest that graphene on CrOCl samples show strong hole doping with the Fermi level shifted downwards by about 200 meV, while the energy of LLs on the hole side follows well the $N^{1/2}$ rule (Extended Data Fig.6c-d).

To further elucidate the specific state of the SIC in the current hybrid system, we fabricated devices equipped with dual gates so that a displacement field $D$ can be applied to decouple the total doping in the studied system from vertical electrical fields, as previously widely used in multiple-layered graphene devices [29][30]. Fig.1c shows the optical micrograph image of a typical h-BN/graphene/CrOCl heterostructure with top and bottom gates, with a cartoon illustrating the layered structure of such typical devices (Fig. 1d). An example of dual-gate mapping of resistance obtained at a temperature of $T = 3$ K is given in Fig.1e. Three notable regions are seen, each separated by a resistive peak and marked as either hole or electron doping, which is obtained via the analysis of the measurements at high magnetic fields, as will be discussed in the coming sections.

When a finite perpendicular magnetic field $B$ is applied, LLs develop in graphene, giving rise to a stripe-like features along constant filling fractions in the dual gated mapping of the field-effect curves, and usually symmetric with respect to the charge neutrality in conventional graphene samples [30][31]. However, in the h-BN/graphene/CrOCl heterostructures with dual gates, the LLs become much different when positive gate voltages are reached (Extended Data Fig.7a-b), yielding a new phase exhibiting cascades-like LLs in general, as illustrated in the schematic phase diagram in Extended Data Fig.7c. This phenomenon is reproducible in multiple monolayered graphene samples stacked on different thicknesses of CrOCl flakes, and is verified by sweeping along different directions within the reachable gate range (Extended Data Fig.8), which thus rules out significant distortions from possible gate-hysteresis in the system.

Gate tuned SIC in the QHE regime.

To analyse the dual gated doping, we first assume a simplified picture of our system, where the total doping $n_{\text{tot}}$ is defined by the two gates, while the real effective doping $n_{\text{eff}} = n_{\text{tot}} - \delta n$ in graphene can actually be different from $n_{\text{tot}}$, due to the adjustment by the graphene/CrOCl interface. Here, $\delta n$ is the partial doping trapped in CrOCl, as illustrated in Extended Data Fig. 9. This definition of $n_{\text{tot}}$ will be used to represent the “nominal” doping induced by top and bottom gates in the rest of this letter. Then we can plot the dual gate field-effect color map into the parameter space of $n_{\text{tot}}$ and $D$, with the total carrier density $n_{\text{tot}} = (C_{tg}V_{tg} + C_{bg}V_{bg})/e - n_0$, and the applied average electric displacement field $D =$
FIG. 2. Gate tunable SIC in the quantum Hall regime. (a) and (c) are color maps of $R_{xx}$ and $R_{xy}$ as a function of magnetic field $B$, respectively. Data recorded along dashed lines (fixed nominal total $n$ along the red and green dashed lines) in (b). (b) $R_{xx}$ in the parameter space of $D$ and $n$, measured at 14 T and 3 K. (d) Line profile of $R_{xx}$ and $R_{xy}$ at $B=12$ T in the color map in (c). (e) Line profile of $R_{xx}$ and $R_{xy}$ at $D=0.35$ V/nm (along the yellow dashed line) in the color map in (b), with the zoomed-in $\sigma_{xy}$ in the smaller range of hole doping shown in (f). (g) Gate tunable cross over from fan-like to cascades-like Landau quantization at $D=0.35$ V/nm.

$$(C_{tg} V_{tg} - C_{bg} V_{bg})/2\epsilon_0 - D_0, \text{ where } C_{TG} \text{ and } C_{BG} \text{ are the top and bottom gate capacitances per area, respectively}.$$
tively. $V_{TC}$ and $V_{BG}$ are the top and bottom gate voltages, respectively. $n_0$ and $D_0$ are residual doping and residual displacement field, respectively. The way we obtained $n_0$ and $D_0$ is given in Extended Data Fig. 10. With these two values, the sample resistance scanned in parameter space of $V_{tg}$ and $V_{bg}$ can be transformed into $n_{tot}$-$D$ coordinates, with $n_{tot}=0$ calibrated to $\nu=0$ Landau quantization in the conventional QHE phase, as given in the example in Extended Data Fig. 11. Using the same method, one can also plot such color-map at $B=0$, as shown in Extended Data Fig. 12. It is clear that Dirac-like peak still exist but in an unusual manner, with the charge neutrality indicated by the white dashed line in Extended Data Fig. 12. As shown in Fig. 2b (also Extended Data Fig. 11), measured from a typical h-BN/graphene/CrOCl heterostructure sample at $B=14$ T and $T=3$ K, longitudinal resistance $R_{xx}$ at hole and electron sides show distinctive features of Landau quantizations, with a crossover from straight stripes to cascades-like stripes as the total doping switches from hole to electron sides.

Figure 2a shows a magnetic field scan of $R_{xx}$ along fixed carrier density at the hole side with $n_{tot} \sim -3.8 \times 10^{12}$ cm$^{-2}$ (red dashed line in Fig. 2b). Little $D$-dependence of filling fraction (i.e., LLs) is seen. This is a typical behavior in monolayer graphene, as there is no $z$ dimension and thus the displacement field does not affect the LLs in it. Strikingly, shown in Fig. 2c, magnetic field scan of transverse resistance $R_{xy}$ along a fixed carrier density at the electron side, for $n_{tot} \sim +1.8 \times 10^{12}$ cm$^{-2}$ (green dashed line in Fig. 2b), has a drastically different pattern as compared to that in Fig. 2a. As discussed in the previous sections, with a single gate, only hole doping can be seen in graphene/CrOCl heterostructures (Extended Data Fig. 4-5). Here, at positive $n_{tot}$, dual gate sweep allows one to reach the electron doping at $D \sim 0.8$ V/nm, as clearly indicated by the line profiles of both $R_{xx}$ and $R_{xy}$ at 12 T in Fig. 2d, where a well-developed quantum Hall plateau of $\nu=+2$ can be seen. In this regime of $n$ and $D$ (we call it SIC-QHE phase), $R_{xy}$ is quantized in an extremely wide parameter space. For example, at $B=14$ T, filling fraction $\nu = \pm 2$ is found in the doping range of $n_{tot}$ from $0$ to $10^{13}$ cm$^{-2}$, and a displacement difference $\delta D$ of over $\sim 2$ V/nm, which converts into a very large variation of gate voltages. Notably, the quantized $R_{xy}$ can be found in very low $B$, reaching the zero magnetic field limit, which will be discussed in more details in the coming sections.

Fig. 2e shows the line profiles of $R_{xx}$ and $R_{xy}$ at a fixed displacement $D = 0.35$ V/nm (along the yellow dashed line in Fig. 2b) at $B=14$ T and $T=3$ K. It is seen that on the hole side (noted as conventional QHE phase) the curves show Landau quantizations, which is in agreement with that observed in conventional monolayered graphene [15,16]. Fig. 2f illustrates a zoomed view of transverse conductivity $\sigma_{xy}$ at $n_{tot}$ between $-0.5 \times 10^{12}$ cm$^{-2}$ and $-3.5 \times 10^{12}$ cm$^{-2}$, indicating full degeneracy lifting with quantized plateaux obtained in $R_{xy}$ at each integer filling fractions from $\nu=-2$ to $-10$. This speaks the high mobility of the graphene in the conventional QHE phase in the graphene/CrOCl heterostructure. Indeed, by fitting of the hole-side part of effect curve at zero magnetic field (Extended Data Fig. 13), hole carrier mobility is estimated to be about $10^4$ cm$^2$V$^{-1}$s$^{-1}$, which is comparable to the value in high-quality monolayer graphene reported elsewhere [27].

On the electron side of the line profiles of $R_{xx}$ and $R_{xy}$ in Fig. 2e, a SIC-QHE phase dominates, as the quantum Hall plateau of $\nu=-2$ extends through out the whole gate range, up to $+1.5 \times 10^{13}$ cm$^{-2}$. By varying the magnetic fields along fixed $D=0.35$ V/nm, one obtains a color map in the parameter space of $B$ and $n_{tot}$, shown in Fig. 2g. It can be seen that the gate tunable SIC leads to a change of Landau quantization from the well-known fan-like behavior to a cascades-like one, as the system undergoes a phase transition from conventional QHE to SIC-QHE. It is similar to the data obtained in single-gated samples (Extended Data Fig. 4), but with a definitively defined $n$ in the dual gated sample presented here. To verify the $n_{eff}$ as compared to $n_{tot}$ in the sample in the low field limit in Fig. 2g, we extracted $n_{eff}$ from Hall resistance at fixed $D$ (0.35 V/nm), which shows a slope of $\sim 1$ with $n_{tot}$ at the conventional phase, but strongly departures at positive $n_{tot}$, as shown in Extended Data Fig. 14. This behavior agrees well with the high field measurements. Moreover, to have a global picture of the major features described above, the color-maps shown in Fig.2 are re-plotted in a 3D presentation, as shown in Extended Data Fig. 15. All these observations are reproducible in multiple samples (Extended Data Fig.16-17), and also confirmed in samples fabricated in a glove box, ruling out possible strong moisture or air adsorption in the interface of graphene/CrOCl heterostructures (Extended Data Fig. 18-19).

The SIC-QHE phase.

Central result of this letter is the observation of a SIC-QHE phase, where Landau quantizations seem to be “pinned” at a fixed filling fraction at a certain hole doping and at a fixed $D$, such as shown in Fig. 2g. A trivial explanation for this would be that the coupling at the graphene-CrOCl interface has accumulated holes that screen and cancel out the positive gate voltages applied, leading to a failure of electron injection and thus the total effective doping remains pinned. Indeed, by a glance at the Hall coefficient at $B=0$ T (Extended Data Fig. 14), it looks that the net carrier density is pinned at an almost fixed value of hole charges, as long as a supposedly positive doping is reached. However, the most striking feature in this work, as shown in Fig. 2b-c, is that displacement field $D$ totally shuffles the Landau quantization (and hence the $n_{eff}$), which rules out the “charge
FIG. 3. Characteristics of the SIC-QHE phase in graphene/CrOCl heterostructures. (a) $R_{xx}$ and (b) $R_{xy}$ plotted in the parameter space of $\delta D$ and $B$. (c) Line profiles of $R_{xx}$ and $R_{xy}$ at $\delta D = -0.08$ V/nm. Inset shows temperature dependence of another typical sample at $\delta D = -0.2$ V/nm. (d) Line profile of $R_{xx}$ in Fig. 3a at $B = -1$ T (indicated by the vertical white dashed line). Red dots are resistive peaks picked by each maximum. (e) Dependence of $\delta D$ and $\sqrt{N}$. Black solid line is a linear fit. (f) Parabolic dependence of $\delta D = \alpha \sqrt{B \cdot |N|}$, plotted with $\alpha = 0.513$, and $|N| < 200$.

pinning" picture, as the latter should be $D$-independent just like in the conventional QHE phase (such as Fig. 2a). Moreover, the Landau quantization seems to be approaching the $B=0$ limit in the SIC-QHE phase, as shown in Fig. 2c.

To further clarify this perplexing scenario, we carried out a zoomed-in scan of the low magnetic field part of Fig. 2c. We define the displacement field where carrier type switches from holes to electrons as $D_{\text{Neutral}}$, thus the $D$ axis can be renormalized as $\delta D = D - D_{\text{Neutral}}$. As shown in Fig. 3a-b ($R_{xx}$ in Fig. 3a, and $R_{xy}$ in Fig. 3b), $\delta D$ was scanned in the range of -1 to +1 V/nm, while the magnetic field was scanned from -4 to +4 T. Wide Landau plateaux are seen in Fig. 3b, with the quantized regions indeed touching the $B=0$ T line, while a tiny width still exists at the vicinity of zero magnetic field, due to the signal swap from positive to negative sign. The $D$-$B$ relation of LLs observed here is distinct
from those found in other multilayered graphene systems \[32\] \[33\].

By bringing $\delta D$ infinitely close to $\delta D=0$, one expects to have a Landau quantization at $\nu=\pm 2$ at the very vicinity of zero magnetic field. Due to experimental noises, we take the $\delta D \approx -0.08$ V/nm here (indicated by the white dashed line in Fig. 3b), and plotted both $R_{xx}$ and $R_{xx}$ in Fig. 3c. The curves indeed show well quantized plateau at $R_{xx}=\pm 0.5$ h/e$^2$ starting from $B$ as low as sub 100 mT, with $R_{xx}$ showing near zero remisiscient values at each plateau, mimicking a quantum anomalous Hall effect (QAH), but without any magnetic hysteresis in the trace-retrace loop of magnetic scan (indicated by red and black arrows). Notably, this behavior is seen above 80 K, as shown in data recorded in a typical sample in the inset of Fig. 3c. This robust SIC-QHE phase in the graphene/CrOCl heterostructure prevails in much higher temperatures as compared to the QAHE systems so far reported, while the latter often requires temperatures of sub 2 K or even dilution fridge temperatures \[35\] \[37\]. It also requires very relaxed experimental conditions as compared to such as doped (Hg,Mn)/Te topological insulators quantum well, which is recently reported to show QH state below 50 mT at 20 mK \[38\]. It thus allows future applications in quantum metrology in the \textit{Système International d’unités} \[13\] \[39\].

Furthermore, by extracting a line profile of $R_{xx}$ in Fig. 3a at $B=-1$ T (indicated by the vertical white dashed line), resistive peaks can be found at each LLs, as indicated by the red dots in Fig. 3d. It is found that the $\delta D$ values at each resistive peak are in linear dependence with $\sqrt{N}$, with $N$ the $N^{th}$ LL, shown in Fig. 3e. This is a typical Landau quantization energy dependence in monolayered graphene. Indeed, $\delta D-B$ relation can be fitted using a parabolic curve as $\delta D = \alpha \sqrt{B} |N|$. Peaks of $R_{xx}$ of the first LL in Fig.3a are picked as red circles, and fitted with a white solid parabolic curve, with $\alpha=0.513$. The first 200 LLs are then plotted in Fig. 3f, well simulating the experimental $\delta D$ data.

Interestingly, the observed SIC-QHE phase seems to have no connection to the anti-ferromagnetic nature of CrOCl itself, as the Néel temperature of it is only $\sim 14$ K, way lower than the upper bound temperature for the SIC-QHE phase. Recently, charge transfer phenomena have been also found in such as RuCl$_3$/graphene systems, which give rise to a shifting of Raman signal or plasmonic characteristics in graphene, and seem to be often irrelevant to the magnetic order of the Cl-based substrates \[10\] \[31\]. Those other Cl-based compounds are reported to lead to inferior mobilities in the resulted hybrid systems, and are not suitable for QHE studies \[20\] \[26\]. In addition, we noticed that, a sister compound of CrOCl, FeOCl is far less stable, and could not be used to check the universality of the findings in this work (Extended Data Fig. 20).

To this stage, one could originate the observed emerging new QHE phase to mainly two possibilities. First, the $D$-tunable LLs in the SIC-QHE phase may come from the fact that there is a charge reservoir (such as defect bands that exist between the valence and conduction bands of CrOCl with large effective mass that do not contribute to transport), and thus electrons and holes can be balanced from graphene to the defect bands while keeping total nominal doping at constant. Indeed, this picture seems to be valid since the $D - B$ scan follows the $D \sim \sqrt{\left| B \right| N}$ relation, which is a consequence of LLs crossing the Fermi level at fixed $D$ and $n_{tot}$, assuming that the Dirac electrons are still dominating in transport in the system. By fitting the Shubnikov-de Haas oscillations from various temperatures at dopings in the conventional and the SIC-phase (Extended Data Fig. 21), the cyclotron mass $m_e$ in the CT-phase is estimated to be below 0.005 $m_e$, which is about one order of magnitude smaller than that of “ordinary” monolayer graphene, as also reported elsewhere \[15\] \[16\] \[42\]. It thus yields ten times the Fermi velocity, in the SIC-QHE phase, then in this regime the cyclotron gap of first LL $\Delta = v_F \sqrt{2eB}$ will be in the order of about 100 meV at 0.1 T. Further experiment of such as infra-red transmission may be needed to verify the cyclotron gap estimated in the current system.

Alternatively, the observed SIC-QHE phase that reaches a $B=0$ limit can be attributed to possible topologically protected symmetries in the electronic band structures. Nevertheless, no signature of topological physics is found in the current system, as indicated by density functional theory calculations. We investigated by modelling the system into a simplified monolayer CrOCl/graphene structure (see Methods), whose band structure is illustrated in Extended Data Fig. 22. A Dirac dispersion is seen with the Fermi level sitting at the Dirac point between $\Gamma$ and $X$ points. The band gap of CrOCl is estimated to be closing at a $D$ of about 5.5 V/nm, much higher than the experimental condition, indicating that there should not exist electric field driven insulator-to-metal phase transition in CrOCl in the tested samples. Moreover, even after the gap closing at a very high $D$, there is no signature of topological phase transition since the band crossing is trivial at $D=5.5$ V/nm, as simulated in Extended Data Fig. 23. In light of the above analysis, we could partially understand the observed behaviours, and an analytical solution of the SIC-QHE phase remains an open question, which requires further theoretical considerations. A complex band structure reconstruction could be an alternative origin of the observed exotic phases. However, at this stage, we regret that direct evidence of band reconstruction is absent, and further probes such as spectroscopic measurements would be helpful in understanding this system in this regard. The fact that, as shown in our results (Extended Data Fig. 24), a robust quantum Hall state with ultra-low magnetic fields at relaxed experimental conditions can be crucial for future constructions of topolog-
ical superconductivity as well as quantum information processing, which has long sought to be only possible in quantum anomalous Hall (QAHE) systems. It unambiguously tells that the interfacial coupling, in terms of tuning the quantum electronic states, is a powerful technique that we may have overlooked thus far.

In conclusion, we have demonstrated a hybrid system with graphene interfaced to an antiferromagnetic insulator CrOCl, in which quantum transport behaviours were examined with a dual gated device configuration. Thanks to the peculiar gate tunable interfacial coupling, a SIC-QHE phase was observed. At finite magnetic fields, and constant $D$, a crossover from fan-like to cascades-like Landau quantization was seen. And in the $D-B$ space, unlike the conventional $D$-independent ones, LLs in the SIC-QHE phase exhibit parabolic dependence between $B$ and $D$, with the Landau quantization reaching the zero magnetic field limit, mimicking a QAHE without coercivity at the vicinity of $D_{\text{Neutral}}$. It thus allows one to effectively engineer the QHE in this hybrid system, with for example a $\nu = \pm 2$ plateau starting from as low as sub 100 mT and prevails up to 100 K in a wide gate total doping range from 0 to $10^{13}$ cm$^{-2}$. Analysis including DFT calculations are adopted to qualitatively discuss the observed phenomena, the origin of which however remains shrouded in obscurity, and further theoretical modellings are needed. Our findings seem to open a new door of engineering the quantum Hall effect, and may shed light in the future studies of such as manipulation of quantum electronic states via interfacial coupling in order to construct novel topological superconductor, and to build quantum metrology standards.

METHODS

Sample fabrications and characterizations. The CrOCl/graphene/h-BN heterostructures were fabricated in ambient condition using the dry-transfer method, with the flakes exfoliated from high quality bulk crystals. CrOCl layers were etch patterned using an ion milling with Ar plasma, and dual gated samples are fabricated using standard e-beam lithography. A Bruker Dimension Icon atomic force microscope was used for thickness, morphology, and surface potential tests. The electrical performances of the devices were measured using a BlueFors LD250 at mK temperature, a Quantum Design PPMS system was used for temperature (3-300 K) and magnetic field ($\pm 14$ T) scannings, and a probe station (Cascade Microtech Inc. EPS150) for room temperature electrical tests.

Density functional theory calculations. In order to simulate the transport properties of graphene on CrOCl, we performed density functional theory plus U (DFT+U) calculations by using the Vienna ab initio simulation package (vasp) based on the projector augmented wave (PAW) potential\cite{43,44} and within the Perdew-Burke-Ernzerhof (PBE) type of GGA functional\cite{45}. The spin-orbit coupling is included in our calculation. Compared with other 2D materials, the relatively easy exfoliation of CrOCl layered structure implies the weak interlayer interaction in CrOCl. The plane-wave energy cutoff is set to be 600 eV and the $\Gamma$-centered $k$-grid for bulk CrOCl, monolayer CrOCl, and a hetero-structure of the graphene on monolayer CrOCl was $16 \times 13 \times 6$, $16 \times 13 \times 1$, and $5 \times 13 \times 1$, respectively. The “DFT+D3” type of vdW correction was adopted for bulk calculations to properly describe the interlayer interactions\cite{46,47}. The so-called fully localized limit of the spin-polarized GGA+U functional was adopted as suggested by Liechtenstein and co-workers\cite{48,49}. The crystal structure is fully relaxed until the residual forces on atoms are less than 0.01 eV/Å. For monolayers and a hetero-structure of the graphene on monolayer CrOCl, a 20 Å vacuum space was taken into account to prevent spurious self-interactions.

DATA AVAILABILITY

The data that support the findings of this study will be available at the open-access repository Zenodo with a doi link, when accepted for publishing.

CODE AVAILABILITY

The codes used in theoretical simulations and calculations are available from the corresponding authors on reasonable request.

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AUTHOR CONTRIBUTIONS

Z.H. and Y.Y. conceived the experiment. Z.H., Y.Y., J.Z., and Z.Z. supervised the overall project. W.L., J.K., and K.C. carried out theoretical modelings and simulations. Y.W., X.G., and K.Y. carried out device fabrications and electrical transport measurements; J.M. and Y.J. performed STM studies; K.W. and T.T. provided high quality h-BN bulk crystals. P.G. and Y.Y. carried out synthesis of CrOCl single crystals. Y.W., B.D., J.Z., and Z.H. analysed the data. The manuscript was written by Z.H. and Y.W., with discussion and inputs from all authors.

ADDITIONAL INFORMATION

Competing interests: The authors declare no competing financial interests.

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