

Peer Review File

Manuscript Title: Quantum anomalous Hall octet driven by orbital magnetism in bilayer graphene

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referee #1 (Remarks to the Author):

The manuscript presents a state of the art experiment on suspended BLG with 2-terminal measurements, discussing a very peculiar phase, induced by a small vertical displacement field. The results are theoretically interpreted in terms of the "ALL" mean-field phase. The results are interesting, novel and the conclusions are supported by the actual results. I believe that the work is suitable for publication in Nature.

Still, I have several questions and comments that would hopefully help the authors to improve the paper.

- 1) It seems natural that the layer-imbalanced phase will be stabilized by the displacement field that would screen the electric field due to layer imbalance. Hence, naively, I would suspect that the maximal gap point in Fig 4 e would move to larger E fields with increasing B. Is that correct? Can the authors add the figure showing the evolution of the gap (or, of gap maximum) with B ?
- 2) In Fig 4d it is not clear to me, whether the expected $\nu = 2$ conductance quantum is subtracted when the Arrhenius plot is done
- 3) Fig 1a, it's better to place $1/\mu\text{m}$ directly over the scale bar.
- 4) On page 8 the authors state:
"This hysteretic behavior provides compelling evidence for the emergence of magnetism in pure bilayer graphene, which has not been observed previously in a moiréless purely carbon-based 2D system."
First, the hysteresis has already been observed in $\nu = 0$ phase in Ref. [18] .
Second, it is far from being obvious that hysteresis does obligatory signal a real magnetism and not, for example, the valley-ferromagnetic domains (that have no real magnetic field).

misprint on page 10:

"of a relative judgement" -> "for a relative judgement"

Referee #2 (Remarks to the Author):

Authors present several exciting experimental evidences that support quantum anomalous Hall octet ("ALL") states in suspended Bernal bilayer graphene. The key features discussed on favor of the "ALL" states are 1) the emergence of the filling factor ± 2 state only in an intermediate range of perpendicular electric fields; 2) that survives down to extremely low magnetic field (< 20 mT); and 3) the hysteretic magnetoresistance loops that seem to appear only in coincidence with the filling factor ± 2 states. This is in fact quite interesting, especially because it shows that the

intrinsic bilayer graphene can host such an extraordinary many-body quantum states in line with several previous observations of the spontaneous gap opening at zero density (refs. 24-26 and more recently Science 362, 324 (2018)). Once published, it can certainly draw extensive attention in various research area.

However, even though I believe that the analysis based on the "ALL" states does capture most of the findings, I have some reservations about supporting the publication of this manuscript in Nature as it is. My concerns and comments are listed below:

1) One of my main concerns is about the hysteresis. In magnetism, the hysteresis occurs when the states with an opposite magnetic moment (e.g., states I and IV in Fig. 1f) have different resistance values. However, since the device is measured in a two-terminal configuration, all "ALL" states are expected to show the same conductance $2e^2/h$. Thus, observing hysteresis is not something that comes out naturally.

In addition, the hysteresis does not cross zero magnetic field which means that it occurs within the same "ALL" state (e.g., state I in Fig. 1f). Then, why do we expect to see the hysteresis? Authors should discuss more why we expect to see hysteresis in the measurement and how it is related to the "ALL" states that they propose.

2) Moreover, the observed hysteresis is quite weak and in Fig. 3d, filling factor 3 state also shows quite some hysteresis similar to the others. To support the observation more, I wonder if authors have measured a so-called memory effect, that is, the resistance loop that appear when the magnetic field is swept only partially. If the resistance loop appears near filling factor ± 2 while it does not at other fillings, it could be more supportive evidence for the presence of hysteresis.

3) In Figs. 2 and S5, authors show the transconductance measurement to discuss the stability of the filling factor ± 2 state by counting the number of corresponding lines. Such a transconductance measurement, however, assumes that there exist puddles where Landau levels are confined such that as varying density n , the conductance exhibits a sort of resonance peaks along a constant filling factor in (n, B) space.

So, it is not a priori expected to use the number of lines as an indicator for the stability of the state at certain filling factor. In fact, if you have a perfect quantum Hall plateau, you will not expect the lines in a transconductance measurement. Can authors provide more relevant arguments or give us some references? (one minor comment related to Fig. 2: in the figure authors draw broken lines for some filling factors but there is no explanation about them in the caption.)

4) Several studies on fractional quantum-Hall states in high-quality bilayer graphene (e.g., Nano Lett. 14, 2135 (2014), Science 345, 55 (2014), Science 345, 61 (2014), Nature 49, 360 (2017), Science 358, 648 (2017) and more) have shown that the valley, spin and orbital degeneracy of zero energy Landau levels in bilayer graphene is spontaneously broken by interaction in such a way that the ground state at filling factor ± 2 have opposite valley or spin or their combined quantum number.

It does not seem to match with the diagrams shown in Figs. 1f, S2 and S3 where for $n > 0$ and $n < 0$, all K and K' valley and spin up and down quantum numbers are present. This can be from the fact that the filling factor ± 2 state discussed in this work appears near zero magnetic field, while in previous studies, higher magnetic fields were applied.

However, authors did also claim that this "ALL" state is further stabilized as increasing magnetic field and showed the data up to 3 T where the previous studies showed the filling factor ± 2 states as well. Therefore, authors should address the coexistence, or competition, or crossover of these (different) filling factor ± 2 states in detail.

5) Four-terminal suspended graphene devices were realized in some previous studies (e.g., Nano Lett. 13, 5165 (2013)) and used to investigate quantum Hall state in four-terminal geometry (e.g., Nano Lett. 14, 2135 (2014)). Especially, these studies showed quantum-Hall plateau at filling factor 4 down to 30 mT. The relevant citations seem missing in the current manuscript (e.g., page 11).

6) The curve in Fig. 1b inset shows about 2-3 times smaller conductance at finite charge density than at zero. Since it is taken at zero perpendicular electric field, I believe that the minimum conductance should appear at zero density. Authors should clarify this.

7) In supplementary page 8 line 15, authors state that “we see that electric field needed for the $\nu = +2$ to emerge decreases towards zero magnetic fields.”. However, in Fig. S4, it is hard to judge this statement and in fact, if I follow the dotted lines in Figs. S4e-h, the minimum electric field that is needed for the $\nu = +2$ state does not change. Can authors provide another plot, possibly the minimum electric field versus magnetic field, to support this statement?

Referee #3 (Remarks to the Author):

The manuscript reports the signatures of $\nu = 2$ quantum Hall states survived at quite low B-field. Their observation supports that the $\nu = +2$ QAH phases are potential ground states of bilayer graphene at zero B-field. I believe that the existence of $\nu = 2$ phase at such low B-field itself and its detailed behavior (tunability) as a function of multiple parameters (B-field, E-field, carrier density, temperature) are quite interesting. However, the following issues should be taken care of.

1. In Page 6, line 4, the authors say “the other family manifesting Hall conductance of $2e^2/h$ is termed “ALL” phase. The authors should elaborate the terminology “ALL” phase. Is “ALL” an abbreviation of a certain long word, or just a name the author created, or the same as “all”? I, probably many common readers as well, am not familiar with this. Besides, this word “ALL phase” was introduced as a singular but sometimes it is used as a plural. The authors’ intention is roughly understandable, but if possible, they should try to somehow refine the usage of this terminology not to make readers confused.

2. At what temperature were the Figure 1b, c, d data taken? Roughly $7 \text{ mK} < T < 10 \text{ mK}$?

3. At what temperature were the Figure 2 and Figure 3 data taken?

4. Figure 3a data are, according to the manuscript, conductance as a function of B-field at a fixed electric field and at a fixed filling factor. To measure those data, B-field and back-gate as well as top-gate voltages should be swept simultaneously or one-by-one step-by-step. More detailed measurement procedures for Figure 3a as well as Figure 2a should be provided. Did the authors acquire Figure 3a data directly, or extract from Figure 2a data? If there is a hysteresis depending on the sweep direction of B-field, how can we understand Figure 2 data? To obtain the Figure 2a data as well, it seems that B-field, top-gate and back-gate voltages should be swept simultaneously or one-by-one step-by-step. Anyway, in that case as well, B-field should be swept in certain direction and thus there should be some hysteresis. Doesn’t this hysteresis affect the transconductance fluctuation behavior? (Is it consistent regardless of the sweep direction of the B-field?).

5. Is there any of hysteresis depending on the direction of carrier density or top-gate or back-gate sweep?

6. In Page 9, line 21, the manuscript says “When setting the filling factor to higher or lower

nominal filling, e.g., $\nu = -1$ or $\nu = -3$, the hysteresis vanishes." Is this a valid argument? Well, there is some hysteresis in the data for $\nu = -3$ (Figure 3d). Do the authors mean the degree of hysteresis decreases as the filling factor increases? It is hard to tell clearly. There seems to be some hysteresis in all cases of $\nu = -2, -2.25, -2.5, -2.75, -3$ data.

7. This is not a critical issue, but can authors provide the conductivity as a function of carrier density at zero E-field and at a finite E-field? What is the roughly estimated electron and hole mobility?

8. Why is the conductivity at zero E-field and at carrier density corresponding to $\nu = \pm 2$ (Figure 1c and 1d) quite high ($\sim 6e^2/h$)?

9. Can the authors mark which ALL phase corresponds to each of the presented four $\nu = \pm 2$ phases (the green regions in Figure 1c and 1d) in Figure 1c and 1d? That can help readers understand more quickly.

Author Rebuttals to Initial Comments:

Referee #1 (Remarks to the Author):

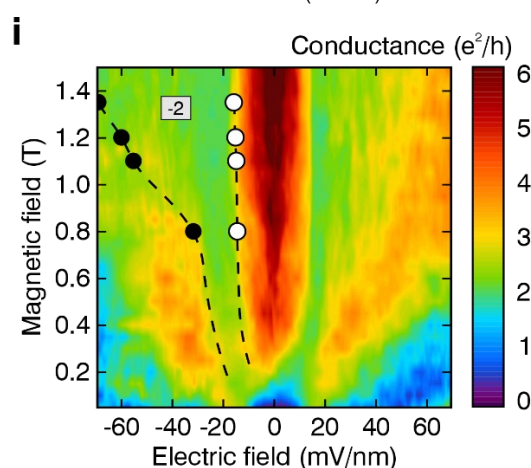
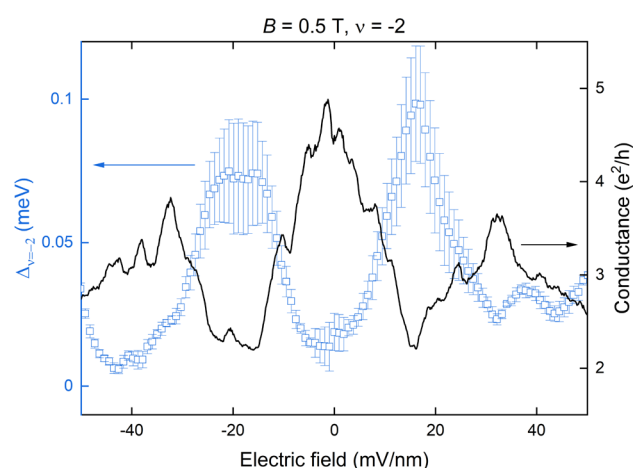
The manuscript presents a state of the art experiment on suspended BLG with 2-terminal measurements, discussing a very peculiar phase, induced by a small vertical displacement field. The results are theoretically interpreted in terms of the "ALL" mean-field phase. The results are interesting, novel and the conclusions are supported by the actual results. I believe that the work is suitable for publication in Nature.

Reply: We thank the referee for these encouraging comments about our work.

Still, I have several questions and comments that would hopefully help the authors to improve the paper.

1) It seems natural that the layer-imbalanced phase will be stabilized by the displacement field that would screen the electric field due to layer imbalance. Hence, naively, I would suspect that the maximal gap point in Fig 4 e would move to larger E fields with increasing B. Is that correct? Can the authors add the figure showing the evolution of the gap (or, of gap maximum) with B?

Reply: We thank the referee for this expert question. For our current set of measurements, we have included the temperature evolution of the gap as a function of electric field at constant magnetic field $B = 0.5$ T (Fig. 4e) and the conductance in the $\nu = -2$ phase as a function of electric and magnetic fields at base temperature (Fig. S6c). In Fig. S6c, one can identify that the maximal electric field that stabilizes the $\nu = -2$ QAH phase increases by increasing the magnetic field, while the minimal electric field only increases very slightly. Note that the two datasets in Fig. 4e and Fig. S6c had been taken on two different samples. In our revised manuscript, we have included a dataset like Fig. S6c but taken in the same sample used for Fig. 4e into the new Fig. S4 (i.e., Fig. S4i), as reproduced below. Clear, a similar conclusion can be drawn from Fig. S4i.



The two datasets above are consistent in the sense that the maximal gap point extracted from the temperature dependence taken at $B = 0.5$ T coincides with the electric field range in which the conductance in Fig. S4i is close to $2e^2/h$. We agree with the referee that adding additional temperature dependence (that we did not record) would make this point even clearer. But we just moved our lab from Munich to Göttingen and are in the setup phase. It could take many months to get to the new measurements. Given this situation and that our current datasets are quite elaborate, we hope that the referee will find our answer satisfactory even though not perfect.

The added section on page 10 in the Supplementary Information reads as follows:

“Furthermore, from the conductance map as a function of electric field and density at different magnetic fields (Fig. S4a-h) and the conductance map as a function of electric and magnetic fields at a fixed filling factor $\nu = -2.25$ (Fig. S4i), we can see the evolution of the ALL phase (e.g., the $\nu = -2$ state) in electric and magnetic fields. The required minimal and maximal electric fields and the electric field range for the $\nu = -2$ state to emerge increase slightly with increasing the magnetic field. At a very low magnetic field, the $\nu = -2$ state is only stable in a very limited electric field range, since at larger electric fields a fully layer polarized $\nu = 0$ state dominates over the partially layer polarized $\nu = -2$ state. However, as the magnetic field increases, the $\nu = -2$ state becomes more and more stabilized, i.e., the electric field range increases.”

The caption of the added figure panel Fig. S4i reads as follows:

"i, Conductance as a function of electric and magnetic fields at a fixed filling factor near $\nu = -2$ (at exactly $\nu = -2.25$). The black (white) dots indicating the maximum (minimum) electric field for the $\nu = -2$ to emerge are extracted from the data shown in (e – h). The dashed lines are guides to the eye, highlighting the region where the $\nu = -2$ state emerges at negative electric fields.

2) In Fig 4d it is not clear to me, whether the expected $\nu = 2$ conductance quantum is subtracted when the Arrhenius plot is done

Reply: We agree with the referee that we did not state clearly how our Fig. 4d had been plotted. First, we normalized the conductance at temperature T by dividing it by the conductance at $T = 10$ K. Then, we took the logarithm of the normalized conductance and plotted it as a function of temperature. This is a common technique to obtain activation gaps with quantum transport data and has been used previously, e.g., by G. Chen et al. Nature **579**, 56-61 (2020) in the study of correlated Chern insulator and ferromagnetism in ABC-trilayer graphene/h-BN. To make this process clearer to the readers, now we have added the aforementioned reference when we first mention Fig. 4d on page 10 and changed the relevant sentence in the caption of Fig. 4 to *"d, Arrhenius plots of conductance (normalized by its value at 10 K) measured at ..."*

3) Fig 1a, it's better to place $1/\mu_0$ directly over the scale bar.

Reply: We thank the referee for this good suggestion. Following it, in the revised manuscript we have improved Fig. 1a and its caption.

4) On page 8 the authors state: "This hysteretic behavior provides compelling evidence for the emergence of magnetism in pure bilayer graphene, which has not been observed previously in a moiréless purely carbon-based 2D system." First, the hysteresis has already been observed in $\nu = 0$ phase in Ref. [18] . Second, it is far from being obvious that hysteresis does obligatory signal a real magnetism and not, for example, the valley-ferromagnetic domains (that have no real magnetic field).

Reply: We thank the referee for this valuable comment and agree that the statement should be improved. First, we agree that ref. 18 did observe hysteresis in much thicker rhombohedral multi-layer graphene and have cited it explicitly now. Second, in the free energy of an ordered phase, it is magnetization (or polarization) that couples to the external magnetic field (or electric field). Thus, the presence of *magnetically* (or electrically) hysteretic behavior implies ferromagnetism (or ferroelectricity). To make these two points clearer, now we have rephrased the sentence on page 8 as follows:

"This magnetic hysteresis provides consistent evidence for the emergence of orbital magnetism in pure bilayer graphene; notably, such hysteretic behavior is rare for moiréless purely carbon-based 2D systems¹⁸."

More detailed comparison between our magnetic hysteresis and that reported for showing QAH effect and orbital magnetism in *twisted* BLG angle-aligned with h-BN has been shown below in our reply to the first question of referee # 2.

Misprint on page 10: ``of a relative judgement" -> ``for a relative judgement"

Reply: We thank the referee for pointing out this misprint. Now we have corrected it.

Referee #2 (Remarks to the Author):

Authors present several exciting experimental evidences that support quantum anomalous Hall octet ("ALL") states in suspended Bernal bilayer graphene. The key features discussed on favor of the "ALL" states are 1) the emergence of the filling factor ± 2 state only in an intermediate range of perpendicular electric fields; 2) that survives down to extremely low magnetic field (< 20 mT); and 3) the hysteretic magnetoresistance loops that seem to appear only in coincidence with the filling factor ± 2 states. This is in fact quite interesting, especially because it shows that the intrinsic bilayer graphene can host such an extraordinary many-body quantum states in line with several previous observations of the spontaneous gap opening at zero density (refs. 24-26 and more recently Science 362, 324 (2018)). Once published, it can certainly draw extensive attention in various research area.

Reply: We thank the referee for the appreciation of our work and for pointing out a relevant and important reference that we missed. To give the broad audience a better picture of the current state of the art, now we have included this reference on page 4 as follows:

"The observation of the latter and the location of the charge neutrality point at $V_t \approx V_b \approx 0$ demonstrates the high quality of the device (note that such a spontaneous gap is universal for rhombohedral few-layer and Bernal even-layer graphene^{9,18,27})." Ref. 27 is the added reference Science 362, 324 (2018).

However, even though I believe that the analysis based on the "ALL" states does capture most of the findings, I have some reservations about supporting the publication of this manuscript in Nature as it is. My concerns and comments are listed below:

1) One of my main concerns is about the hysteresis. In magnetism, the hysteresis occurs when the states with an opposite magnetic moment (e.g., states I and IV in Fig. 1f) have different resistance values. However, since the device is measured in a two-terminal configuration, all "ALL" states are expected to show the same conductance $2e^2/h$. Thus, observing hysteresis is not something that comes out naturally.

In addition, the hysteresis does not cross zero magnetic field which means that it occurs within the same "ALL" state (e.g., state I in Fig. 1f). Then, why do we expect to see the hysteresis? Authors should discuss more why we expect to see hysteresis in the measurement and how it is related to the "ALL" states that they propose.

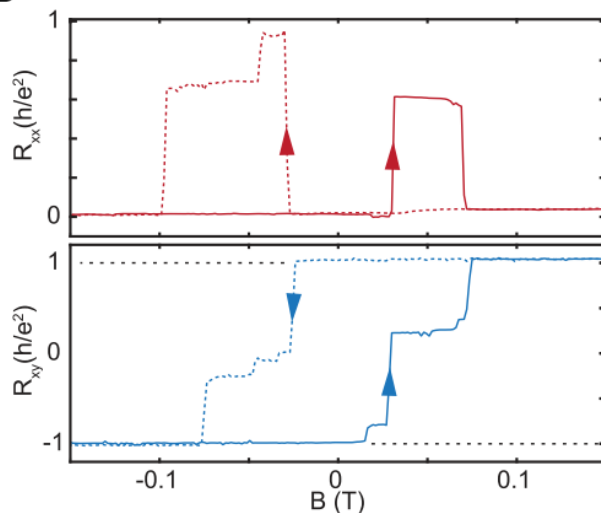
Reply: We thank the referee for these expert questions. We agree that measuring the hysteresis with two-terminal devices is not straightforward. First, the two-terminal conductance of ALL states is not exactly $2e^2/h$ but a mix of both σ_{xx} and σ_{xy} . Precisely, it has the form of

$$\sigma_{two-terminal} \propto \sqrt{\sigma_{xx}^2 + \sigma_{xy}^2} \quad \text{or} \quad \sigma_{two-terminal} \propto \frac{1}{\sqrt{\rho_{xx}^2 + \rho_{xy}^2}},$$

as demonstrated before (Abanin and Levitov, PRB 78, 035416 (2008)). This implies that both the two-terminal conductance and resistance have longitudinal and Hall contributions.

Then, consider the QAH state observed in twisted bilayer graphene (Serlin et al., Science 367, 900–903 (2020)). The Fig. 1B of this 2020 work (reproduced below) shows the evolution of both R_{xx} and R_{xy} in two separate measurements. Now imagine a two-terminal measurement of the same QAH state in the 2020 work. (i) The two-terminal conductance would not show any hysteresis around $B \approx 0$, since $R_{xy}(\text{forward}) = -R_{xy}(\text{backward}) \neq 0$ and $R_{xx}(\text{forward}) = R_{xx}(\text{backward}) = 0$ around $B \approx 0$. (ii) Hysteretic behaviour would show up when $R_{xx}(\text{forward})$ or $R_{xx}(\text{backward}) \neq 0$ and $R_{xy}(\text{forward})$ or $R_{xy}(\text{backward})$ changes sign. Consequently, the hysteresis would be most prominent at finite magnetic field and vanishing at zero magnetic field, which is in agreement with our observation.

B



From this analysis, we can safely conclude that even in two terminal measurements one can measure the magnetic hysteresis in resistance in transition between two distinct ‘ALL’ states. To clarify this issue, we have added a new section (“More details on the hysteresis”) in Supplementary Information (on page 17) as follows:

“At first sight, the observation of magnetic hysteresis with two-terminal measurements might be surprising, since only absolute values are measured

without resolving the two components, σ_{xx} and σ_{xy} . However, following the previous derivation of the two-terminal conductance⁹

$$\sigma_{two-terminal} \propto \sqrt{\sigma_{xx}^2 + \sigma_{xy}^2} \quad \text{or} \quad \sigma_{two-terminal} \propto \frac{1}{\sqrt{\rho_{xx}^2 + \rho_{xy}^2}},$$

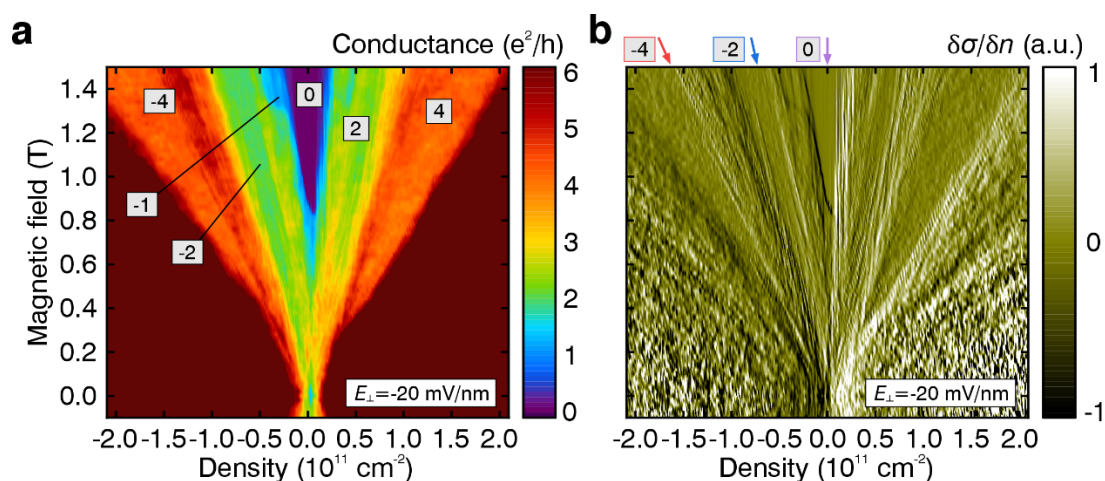
one can observe a hysteresis if $\sigma_{two-terminal}^{forward} \neq \sigma_{two-terminal}^{backward}$. This is true around the coercive field, where a transition between two different ALL phases occurs, i.e., $|\rho_{xx}^{forward}| \neq |\rho_{xx}^{backward}|$ and/or $|\rho_{xy}^{forward}| \neq |\rho_{xy}^{backward}|$. Around zero magnetic field, the hysteresis vanishes in two-terminal measurements, since $\rho_{xx}^{forward} = \rho_{xx}^{backward} = 0$ and $\rho_{xy}^{forward} = -\rho_{xy}^{backward}$, even though opposite orbital magnetizations (with $\rho_{xy} = \pm h/2e^2$) are present and distinguishable in four-terminal measurements¹⁰.”

Accordingly, in the revised main text, we have also added the following comment on page 8:

“Indeed, hysteretic behavior indicative of magnetism is observable in the samples, even though in our two-terminal measurements the absolute contributions of both longitudinal and Hall resistances are measured simultaneously²².” Ref. 22 cites the Supplementary Information.

2) Moreover, the observed hysteresis is quite weak and in Fig. 3d, filling factor 3 state also shows quite some hysteresis similar to the others. To support the observation more, I wonder if authors have measured a so-called memory effect, that is, the resistance loop that appear when the magnetic field is swept only partially. If the resistance loop appears near filling factor $\nu = -2$ while it does not at other fillings, it could be more supportive evidence for the presence of hysteresis.

Reply: We thank the referee for this comment. First, regarding the hysteresis for $\nu = -3$, we would like to point out that in Fig. 3d the $\nu = -3$ does not label any different state but rather the same $\nu = -2$ state at a density corresponding to the filling factor -3. At these low magnetic fields $|B| < 1$ T where the hysteresis was measured, the $\nu = -3$ and -1 states are yet to emerge. This fact can be seen in Fig. 2a,b that are reproduced below.



Clearly, the $\nu = -3$ state does not emerge at all for the low magnetic fields in Fig. 2a,b, while the $\nu = -1$ state emerges only for $B > 1.3$ T. Therefore, when the filling factor is varied in the range from -3 to -1, the phase remains the $\nu = -2$ state, and thus the hysteresis is indeed expected, though less prominent at larger $|-2.5 - \nu|$. To stress this point, in the original manuscript we have labelled “Density corresponding to $\nu = -1$ ” in the top panel of Fig. 3d. Additionally, we have stated the following on page 9 of the main text: “Since the $\nu = -1$ and -3 quantum Hall states do not emerge at $B < 1$ T, all nominal fillings in the range of $-1 < \nu < -3$ correspond to the $\nu = -2$ state.” To further stress this point to avoid any misunderstanding, now we have added the following sentence in the caption of Fig. 3: “Note that the varying ν in (d) labels the $\nu = -2$ state at slightly different densities rather than different quantum Hall states.”

Second, we have indeed traced the hysteresis for successively increasing the magnetic field range (i.e., performing partial hysteresis loops), as shown in the right figure. We agree with the referee that this is an important measurement to support our observation of the hysteresis. Therefore, we have added the results to the Supplementary Information as the new Fig. S7 with the following description:

“To further prove the presence of the magnetic hysteresis, we have measured it for different ranges of magnetic fields, as shown in Fig. S7. The degree of the hysteresis increases with the field range of the cycle. When the magnetic field is only swept in a small range ($-0.25\text{ T} \leq B \leq 0.25\text{ T}$), there is almost no hysteretic behavior. However, when the magnetic field is swept from a larger field toward zero ($-0.5\text{ T} \leq B \leq 0.5\text{ T}$), the hysteresis appears partially. The fact that a sufficiently large magnetic field is needed to observe a hysteresis has been seen in twisted bilayer graphene^{10,11}. The maximum degree of hysteresis is reached at $|B| \leq 1\text{ T}$ and then remains nearly the same with further increasing the field range.

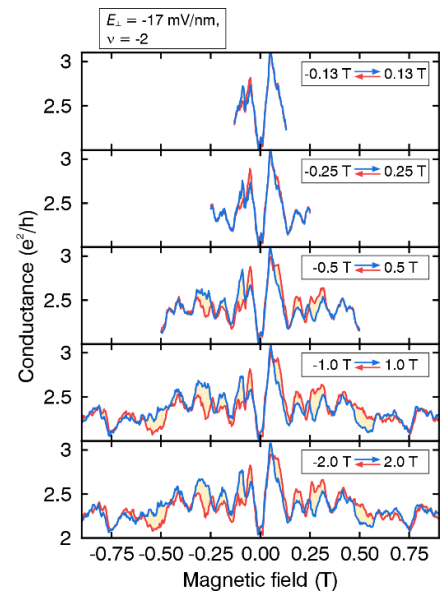


Fig. S7. Magnetic hysteresis loops for different field ranges. Two-terminal conductance hysteresis measured for different magnetic field ranges at $\nu = -2$ and $E_{\perp} = -17\text{ mV/nm}$. The hysteresis loop areas are shaded for clarity.”

In the manuscript, we have added the following sentence on page 8:

“Sweeping a smaller range than that between $B = \pm 650\text{ mT}$ reduces the hysteresis (see Fig. S7).”

3) In Figs. 2 and S5, authors show the transconductance measurement to discuss the stability of the filling factor ± 2 state by counting the number of corresponding lines. Such a transconductance measurement, however, assumes that there exist puddles where Landau levels are confined such that as varying density n , the conductance exhibits a sort of resonance peaks along a constant filling factor in (n, B) space. So, it is not a priori expected to use the number of lines as an indicator for the stability of the state at certain filling factor. In fact, if you have a perfect quantum Hall plateau, you will not expect the lines in a transconductance measurement. Can authors provide more relevant arguments or give us some references? (one minor comment related to Fig. 2: in the figure authors draw broken lines for some filling factors but there is no explanation about them in the caption.)

Reply: We thank the referee for pointing out that our reason to use conductance fluctuations is not self-explaining. First, we agree with the referee that in the case for a perfect quantum Hall plateau, one would not expect any transconductance fluctuations. In fact, in Fig. 2b for the $\nu = \pm 4$ states at magnetic fields $B > 1\text{ T}$, there are less fluctuations, and prominent plateaus emerges. In the center of the plateaus there are no signs of fluctuations, and they only appear at the edges of the filling factors. When the cyclotron gap is close to the disorder

potential, only transconductance fluctuations are expected to appear before the states completely disappear, and this is exactly the case for the $\nu = 0, \pm 2$ and ± 4 states toward the zero magnetic field. The idea of using transconductance fluctuations as an indicator of interaction-induced quantum Hall states has been exploited before. More details are explained in Ref. 36 (Lee et al., PRL 109, 056602 (2012)) that were cited in a non-ideal place in the original manuscript. Now we have cited it at the correct spot and added an additional reference (Kumar et al., Nat. Commun. 9, 2776 (2018)) that used the same technique of examining transconductance fluctuations. Now the relevant sentence on page 7 reads:

“Carefully examining the derivative of the conductance (Fig. 2b) to track fluctuations near incompressible quantum states provides more insight^{37,38}, since the traceable fluctuations are assignable to specific filling factors using their slopes and can appear even before the corresponding quantum Hall states emerge in conventional magneto-transport measurements.”

We also thank the referee for the comment on the caption of Fig. 2, and now we have added the following two sentences to the end of the caption:

“Solid (dashed) lines indicate the slopes of the respective states when they are present (absent). All measurements were taken at $T < 10$ mK.”

4) Several studies on fractional quantum-Hall states in high-quality bilayer graphene (e.g., Nano Lett. 14, 2135 (2014), Science 345, 55 (2014), Science 345, 61 (2014), Nature 549, 360 (2017), Science 358, 648 (2017) and more) have shown that the valley, spin and orbital degeneracy of zero energy Landau levels in bilayer graphene is spontaneously broken by interaction in such a way that the ground state at filling factor ± 2 have opposite valley or spin or their combined quantum number.

It does not seem to match with the diagrams shown in Figs. 1f, S2 and S3 where for $n > 0$ and $n < 0$, all K and K' valley and spin up and down quantum numbers are present. This can be from the fact that the filling factor ± 2 state discussed in this work appears near zero magnetic field, while in previous studies, higher magnetic fields were applied.

However, authors did also claim that this “ALL” state is further stabilized as increasing magnetic field and showed the data up to 3 T where the previous studies showed the filling factor ± 2 states as well. Therefore, authors should address the coexistence, or competition, or crossover of these (different) filling factor ± 2 states in detail.

Reply: We thank the referee for this comment. Exactly, the $\nu = \pm 2$ states discussed in this current work appears near zero magnetic field, while in previous studies higher magnetic fields were applied. In fact, there are two types of $\nu = \pm 2$ quantum Hall ferromagnetic states at large magnetic fields: one without layer polarization (layer XY-like) that appears near zero electric field and the other with layer polarization (layer Ising-like) that requires a finite electric field. This was mentioned in PRL 101, 097601 (2008) by MacDonald et al. and observed in dual-gated devices, e.g., in Nat. Comm. 5, 4550 (2014), Science 345, 61 (2014), and Nature 549, 360 (2017). The ALL states adiabatically evolve into the layer-polarized $\nu =$

± 2 quantum Hall ferromagnetic states with increasing the magnetic field. While there is no transition, the required electric field range does evolve with the magnetic field.

To address the referee's question, we have added a new paragraph in the section "*Evolution of the $\nu = \pm 2$ states in electric and magnetic fields*" in the SI which includes the aforementioned paragraph and papers.

5) Four-terminal suspended graphene devices were realized in some previous studies (e.g., Nano Lett. 13, 5165 (2013)) and used to investigate quantum Hall state in four-terminal geometry (e.g., Nano Lett. 14, 2135 (2014)). Especially, these studies showed quantum-Hall plateau at filling factor 4 down to 30 mT. The relevant citations seem missing in the current manuscript (e.g., page 11).

Reply: We thank the referee for pointing out the two missing references. In the revised main text, we have cited them in two places as refs. 36 and 40. On page 11, we have cited both in the conclusion: "*Future measurements using a four-terminal geometry^{36,40} could distinguish between longitudinal and Hall resistances and determine possible switching mechanism of the exotic ordering of such $\nu = \pm 2$ QAH phases by using both magnetic and electric fields.*" On page 7, we have cited the 2013 work for the observation of $\nu = \pm 4$ states at ultra-low magnetic fields as follows: "*From the fan diagram at $E_{\perp} = -20$ mV/nm (Fig. 2a,b) we can see that both the $\nu = \pm 2$ and ± 4 states emerge already at unusually small magnetic field. We focus here on the $\nu = \pm 2$ QAH phases, since they previously escaped observation at such low magnetic fields^{31,33,36}.*"

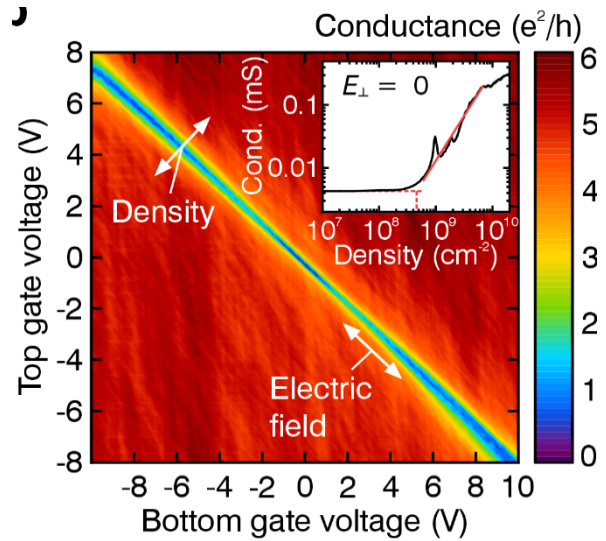
6) The curve in Fig. 1b inset shows about 2-3 times smaller conductance at finite charge density than at zero. Since it is taken at zero perpendicular electric field, I believe that the minimum conductance should appear at zero density. Authors should clarify this.

Reply: We thank the referee for pointing this issue out. Indeed, one would expect the minimal conductance to be directly at the charge neutrality point (CNP). However, during the calibration of the CNP and the offsets in top and bottom gate voltages, we performed a very precise density sweep at zero electric field (which has now been added in Fig. S1, as requested by referee #3). In this method, the CNP is not set to the point of minimal conductance. Instead, a Lorentzian fit to the conductance versus density sweep is performed, and the CNP is set to its apex. As a result, the CNP may not be at the point of minimal conductance.

If we set the CNP to the point of minimum conductance (only requiring a very minor adjustment in density), the calculated residual charge disorder is even lower ($< 5 \times 10^8 \text{ cm}^{-2}$). We think that the method of using a Lorentzian fit rather than the minimum conductance provides a more generic method for calibration, although for the current case the former gives an upper limit of the residual charge disorder while the latter might yield a more precise result. Thus, we decide to leave the inset as is and add the following in the new section (Measurement details) on page 2 of the Supplementary Information:

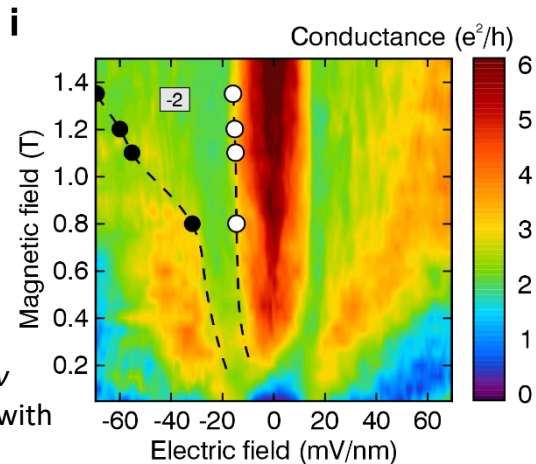
“A Lorentzian fit to a density sweep and a precise electric field sweep were used to find the exact charge neutrality point.”

Of course, if the referee prefers the alternative method, we can also provide the corresponding inset, which would look like the one on the right.



7) In supplementary page 8 line 15, authors state that “we see that electric field needed for the $\nu = +2$ to emerge decreases towards zero magnetic fields.”. However, in Fig. S4, it is hard to judge this statement and in fact, if I follow the dotted lines in Figs. S4e-h, the minimum electric field that is needed for the $\nu = +2$ state does not change. Can authors provide another plot, possibly the minimum electric field versus magnetic field, to support this statement?

Reply: We thank the referee for the comment and suggestion. We agree that the suggested new plot can make our statement better justified. In the revised manuscript, we have included a new panel Fig. S4i in Fig. S4, which shows the conductance map as a function of electric and magnetic fields at a fixed filling factor $\nu = -2$. Fig. S4i is reproduced on the right. Clearly, the required minimal and maximal electric fields and the electric field range for the $\nu = -2$ state to emerge all increase slightly with increasing the magnetic field.



The added section on page 10 in the Supplementary Information reads as follows:

“Furthermore, from the conductance map as a function of electric field and density at different magnetic fields (Fig. S4a-h) and the conductance map as a function of electric and magnetic fields at a fixed filling factor $\nu = -2.25$ (Fig. S4i), we can see the evolution of the ALL phase (e.g., the $\nu = -2$ state) in electric and magnetic fields. The required minimal and maximal electric fields and the electric field range for the $\nu = -2$ state to emerge increase slightly with increasing

the magnetic field. At a very low magnetic field, the $\nu = -2$ state is only stable in a very limited electric field range, since at larger electric fields a fully layer polarized $\nu = 0$ state dominates over the partially layer polarized $\nu = -2$ state. However, as the magnetic field increases, the $\nu = -2$ state becomes more and more stabilized, i.e., the electric field range increases."

The caption of the added figure panel Fig. S4i reads as follows:

"i, Conductance as a function of electric and magnetic fields at a fixed filling factor near $\nu = -2$ (at exactly $\nu = -2.25$). The black (white) dots indicating the maximum (minimum) electric field for the $\nu = -2$ to emerge are extracted from the data shown in (e – h). The dashed lines are guides to the eye, highlighting the region where the $\nu = -2$ state emerges at negative electric fields."

Referee #3 (Remarks to the Author):

The manuscript reports the signatures of $\nu = 2$ quantum Hall states survived at quite low B-field. Their observation supports that the $\nu = \pm 2$ QAH phases are potential ground states of bilayer graphene at zero B-field. I believe that the existence of $\nu = 2$ phase at such low B-field itself and its detailed behavior (tunability) as a function of multiple parameters (B-field, E-field, carrier density, temperature) are quite interesting. However, the following issues should be taken care of.

Reply: We thank the referee for the appreciation of our work.

1. In Page 6, line 4, the authors say "the other family manifesting Hall conductance of $2e^2/h$ is termed "ALL" phase. The authors should elaborate the terminology "ALL" phase. Is "ALL" an abbreviation of a certain long word, or just a name the author created, or the same as "all"? I, probably many common readers as well, am not familiar with this. Besides, this word "ALL phase" was introduced as a singular but sometimes it is used as a plural. The authors' intention is roughly understandable, but if possible, they should try to somehow refine the usage of this terminology not to make readers confused.

Reply: We thank the referee for this comment. We agree that the term 'ALL' was not explained well. The term 'ALL' comes from the fact that each ALL phase exhibits all types of quantized anomalous Hall effects: charge Hall, spin Hall, valley Hall, and spin-valley Hall. In this revision we have added the following explanation on page 6: "Markedly, each ALL phase exhibits quantized anomalous charge, spin, valley, and spin-valley Hall effects and hence the name^{9,10}." Moreover, we have gone through the manuscript and ensured that both the singular and plural forms of 'ALL phase(s)' are properly used without causing any confusion.

2. At what temperature were the Figure 1b, c, d data taken? Roughly $7 \text{ mK} < T < 10 \text{ mK}$?

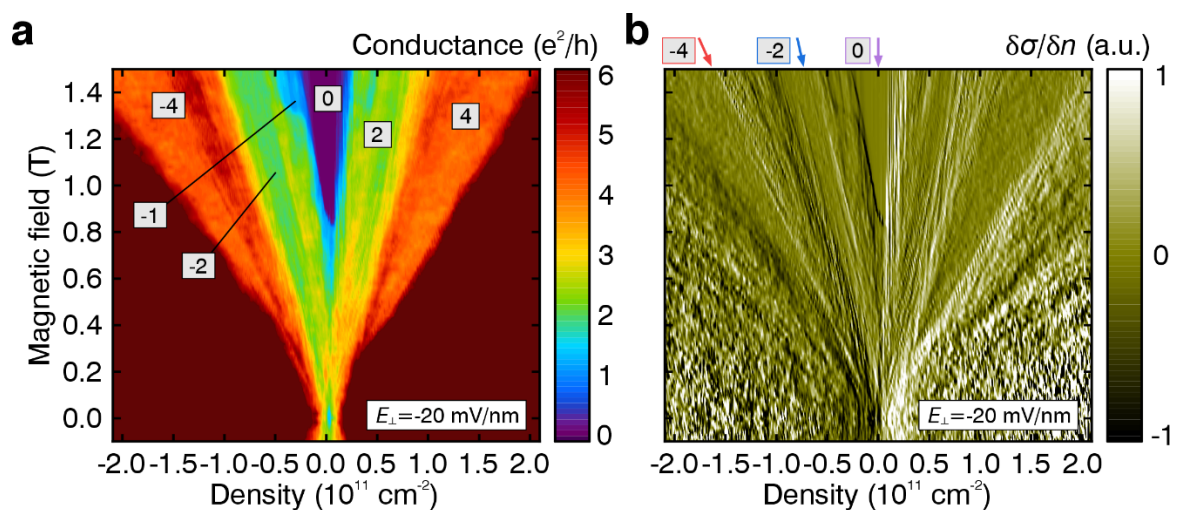
3. At what temperature were the Figure 2 and Figure 3 data taken?

Reply: We thank the referee for pointing out this missing data. In our original manuscript, we have included the information on page 4: "Unless stated otherwise, the transport data was taken using standard lock-in techniques and an ac bias current of 1 - 10 nA at a temperature

of $T < 10$ mK.” “ $T < 10$ mK” has also been given in the caption of Fig. 1. We agree that the temperature of the measurements is crucial, and now we have added the temperature information with the following description to the captions of Figs. 2 and 3: “All measurements were taken at $T < 10$ mK.”

4. Figure 3a data are, according to the manuscript, conductance as a function of B-field at a fixed electric field and at a fixed filling factor. To measure those data, B-field and back-gate as well as top-gate voltages should be swept simultaneously or one-by-one step-by-step. More detailed measurement procedures for Figure 3a as well as Figure 2a should be provided. Did the authors acquire Figure 3a data directly, or extract from Figure 2a data? If there is a hysteresis depending on the sweep direction of B-field, how can we understand Figure 2 data? To obtain the Figure 2a data as well, it seems that B-field, top-gate and back-gate voltages should be swept simultaneously or one-by-one step-by-step. Anyway, in that case as well, B-field should be swept in certain direction and thus there should be some hysteresis. Doesn't this hysteresis affect the transconductance fluctuation behavior? (Is it consistent regardless of the sweep direction of the B-field?).

Reply: We thank the referee for these questions. Indeed, we did not provide enough information on how the measurements were performed. In our experiments, we can apply both top and bottom gate voltages (V_t and V_b), apply a magnetic field (B), and set a temperature (T). The charge density (n) and electric field ($E_\perp$) are linear combinations of V_t and V_b . By changing V_t and V_b simultaneously, we can directly sweep n or $E_\perp$. In Figs. 2 and 3, the horizontal axes are always the fast-sweeping axes. In Fig. 2a, the density n is swept for a given B field, and afterwards the B field is lowered, and then the density n is swept again. In Fig. 3a, the B field is fast swept for a fixed $E_\perp$ field and a fixed filling factor ν . To sweep the B field while fixing $E_\perp$ and ν , V_t , V_b , and B need to be changed simultaneously, given the definition $\nu = nh/e|B|$.



Now we explain the hysteresis using Fig. 2a-b, as reproduced above. The fan diagram (Fig. 2a) shows the conductance as a function of B and n . Here the density n is swept for a given B field, and the transconductance fluctuations can be observed. Given that $\nu = nh/e|B|$, the filling factor ν appears as the inverse of the slope, indicated by the arrows on the top of Fig. 2b. Hence, when we sweep B for a fixed ν in Fig. 3, we follow such an arrow towards $B = 0$. Only

when (i) the sweeping direction follows the $\nu = -2$ state and (ii) the sweeping does not leave the state, the hysteresis can be observed. In the case in which the B field is swept not for a fixed ν but for a fixed n (Fig. 3b), no hysteresis can be observed; this corresponds to a vertical line in the fan diagram (Fig. 2b). To make this point clear, we have stated the following in the main text:

“First, cyclic B sweeps for fixed n (and consequently varying ν) do not show hysteretic behavior (Fig 3b). These measurements were performed at $n = -0.25, -0.5, -1.0 \times 10^{11} \text{ cm}^{-2}$ corresponding to the quantum Hall states of $\nu = -1, -2, -4$ at $B = 1 \text{ T}$, respectively. This implies that when the magnetic field is swept toward $B = 0$, the sample leaves the $\nu = -2$ QAH phase and reaches quantum Hall states with higher filling factors up to $\nu = -12$ for $n = -1.0 \times 10^{11} \text{ cm}^{-2}$, far away from the $\nu = -2$ QAH phase.”

To provide more details on our measurements, now we have rearranged the first part of the Supplementary Information and added a section II (“Measurement details”) as follows:

“The dual-gate structure allows independent tunability of the charge carrier density n and the perpendicular electric field $E_{\perp}$. We can derive n and $E_{\perp}$ as functions of top and bottom gate voltages V_t and V_b as follows¹:

$$n = \frac{C_b}{e}(\alpha V_t + V_b) \quad \text{and} \quad E_{\perp} = \frac{C_b}{2\varepsilon_0}(\alpha V_t - V_b),$$

where ε_0 is the vacuum permittivity and $\alpha = \frac{C_t}{C_b}$ is the ratio between the top and bottom gate capacitances C_t and C_b . Hence, by changing V_t and V_b simultaneously, we can directly sweep n or $E_{\perp}$. A Lorentzian fit to a density sweep and a precise electric field sweep are used to find the exact charge neutrality point at zero electric field. For the hysteresis measurements, the filling factor reads

$$\nu = \frac{nh}{e|B|} = \frac{C_b(\alpha V_t + V_b)}{e^2|B|},$$

with h the Planck constant and B the magnetic field. Hence, in order to sweep the magnetic field while fixing the filling factor, V_t , V_b , and B need to be varied simultaneously.”

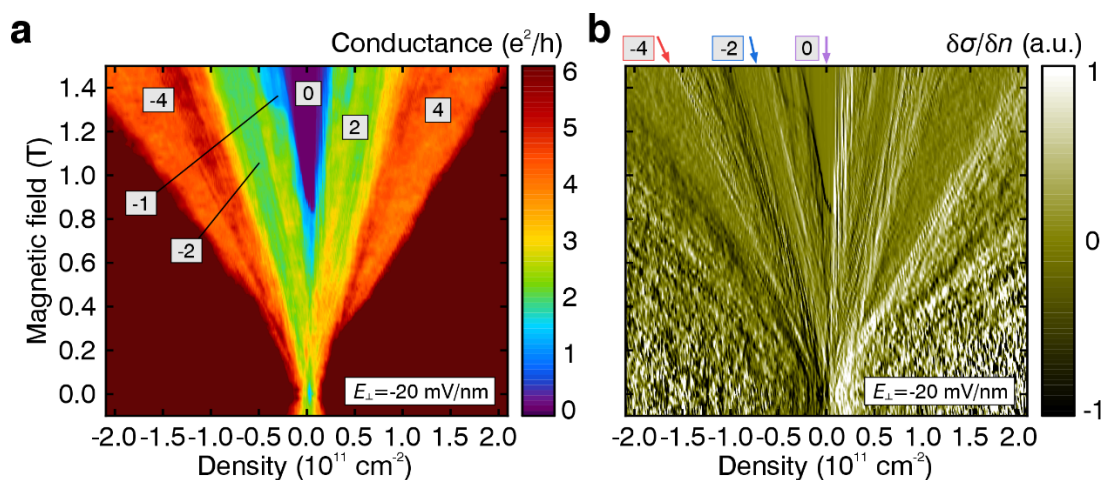
In the main text, now we have added a reference to the Supplementary Information on page 4 as follows: “The dual-gate structure allows independent tunability of the charge carrier density n and the perpendicular electric field $E_{\perp}$ ²².” Ref. 22 cites the Supplementary Information.

5. Is there any of hysteresis depending on the direction of carrier density or top-gate or back-gate sweep?

Reply: We thank the referee for this question. The answer is in the affirmative for the carrier density sweep. In Fig. 3c,d, each panel has a fixed filling factor ν , and its B field sweep amounts to a carrier density sweep, given that $n = e|B|\nu/h$; clearly, there are hystereses. On the other hand, there is no hysteresis for top- or back-gate sweep, because in such a sweep the sample leaves the $\nu = +2$ or -2 state eventually and enters into a series of states at different filling factors that have very different nature. Nevertheless, in the observation of hysteresis, we find that the most important are to fix the filling factor at $\nu = +2$ or -2 and to sweep the B field towards zero.

6. In Page 9, line 21, the manuscript says “When setting the filling factor to higher or lower nominal filling, e.g., $\nu = -1$ or $\nu = -3$, the hysteresis vanishes.” Is this a valid argument? Well, there is some hysteresis in the data for $\nu = -3$ (Figure 3d). Do the authors mean the degree of hysteresis decreases as the filling factor increases? It is hard to tell clearly. There seems to be some hysteresis in all cases of $\nu = -2, -2.25, -2.5, -2.75, -3$ data.

Reply: We thank the referee for this comment. We would like to point out that in Fig. 3d the $\nu = -1$ to -3 do not label any different states but rather the same $\nu = -2$ state at different densities. At low magnetic fields $|B| < 1$ T where the hysteresis was measured, the $\nu = -3$ and -1 states are yet to emerge. This fact can be seen in Fig. 2a,b that are reproduced below.



Clearly, the $\nu = -3$ state does not emerge at all for the low magnetic fields in Fig. 2a,b, while the $\nu = -1$ state emerges only for $B > 1.3$ T. Therefore, when the filling factor is varied in the range from -3 to -1 , the phase remains the $\nu = -2$ state, and thus the hysteresis is indeed expected, though less prominent at larger $|-2.5 - \nu|$. To stress this point, in the original manuscript we have labelled “Density corresponding to $\nu = -1$ ” in the top panel of Fig. 3d. Additionally, we have stated the following on page 9 of the main text: “Since the $\nu = -1$ and -3 quantum Hall states do not emerge at $B < 1$ T, all nominal fillings in the range of $-1 < \nu < -3$ correspond to the $\nu = -2$ state.” To further stress this point to avoid any misunderstanding, now we have added the following sentence in the caption of Fig. 3: “Note that the varying ν in (d) labels the $\nu = -2$ state at slightly different densities rather than different quantum Hall states.” Furthermore, we agree that the hysteresis does not completely vanish at $\nu = -3$ and -1 ; therefore, we have rewritten the relevant sentence in a more careful way as follows: “At higher or lower nominal filling, e.g. $\nu = -1$ or $\nu = -3$, the hysteresis almost vanishes.”

7. This is not a critical issue, but can authors provide the conductivity as a function of carrier density at zero E-field and at a finite E-field? What is the roughly estimated electron and hole mobility?

Reply: We thank the referee for this comment. We agree with the referee, to make the manuscript more accessible, the basic measurements such as the conductivity as a function of carrier density and the electron and hole mobilities should be included. Now we have

rearranged the first part of the Supplementary Information (SI) and added a section IV (Quality of the device) as follows:

“Fig. S1c shows the conductance of the device as a function of charge carrier density for zero and finite electric fields. The narrow width and low conductance of the device at $E_{\perp} = 0$ suggest a high quality of the device. Besides calculating the residual charge disorder (see Fig. 1b, inset), we have calculated the electron/hole mobility $\mu_{e/h} = 120,000/130,000 \text{ cm}^2/\text{Vs}$ at $n = \pm 5 \times 10^9 \text{ cm}^{-2}$, indicating the high quality of the device.”

Additionally, we have added a panel c to Fig. S1, which shows the conductance as a function of charge carrier density for zero and finite electric fields. Accordingly, in the main text, we have added a reference to the SI on page 5 as follows:

“Sweeping n at $E_{\perp} = 0$ reveals a residual charge density inhomogeneity of $< 10^9 \text{ cm}^{-2}$ (Fig. 1b, inset), underlining the high quality of the device^{22,28}.” Ref. 22 cites the SI.

8. Why is the conductivity at zero E-field and at carrier density corresponding to $\nu = \pm 2$ (Figure 1c and 1d) quite high ($\sim 6e^2/h$)?

Reply: We thank the referee for raising this question. For the $E = 0$ case, in short, near $B = 0$, BLG is metallic (i.e., no bulk energy gap) at finite densities corresponding to $\nu = \pm 2$. (i) First, near $B = 0$, normally a quantum Hall state is not expected. In fact, $\nu = \pm 2$ quantum Hall states only emerge at high B fields for the $E = 0$ case (see Nat. Comm. 5, 4550 (2014) for example). Note that our focus is the physics at low B fields. (ii) Second, at $B = E = n = 0$, the BLG ground state is a layer antiferromagnet (fully gapped) that evolves into a $\nu = 0$ canted antiferromagnet with the B field (see Nat. Nano. 7, 156 (2012) for example). (iii) Third, the ALL states (fully gapped), which are adiabatically connected to $\nu = \pm 2$ quantum Hall states, are competing ground states of BLG at $B = E = n = 0$. Given its layer polarization and orbital magnetization, an ALL state can be favoured in the presence of a small E field (supporting its layer polarization) and a tiny B field (coupling to its orbital magnetization), as revealed by our current work and emphasized on pages 6 and 10 of the main text.

9. Can the authors mark which ALL phase corresponds to each of the presented four $\nu = \pm 2$ phases (the green regions in Figure 1c and 1d) in Figure 1c and 1d? That can help readers understand more quickly.

Reply: We thank to referee for this valuable suggestion. In the revised manuscript, now we have included the corresponding roman numerals to the ‘ALL’ phases in Fig. 1 d. In the caption of Fig. 1, we have also added the following sentence: *“The roman numerals in (d) label the “ALL” phases labelled by the same numerals in (f).”*

Reviewer Reports on the First Revision:

Referee #1 (Remarks to the Author):

I am satisfied with the reply and improvements made to the text and recommend the work for publication.

Referee #2 (Remarks to the Author):

Authors have addressed all of my points well in the reply, and revised the manuscript and supplementary properly. Therefore, I support its publication in Nature.

Referee #3 (Remarks to the Author):

I believe that the manuscript is acceptable for Nature.