Reviewers' comments:

Reviewer #1 (Remarks to the Author):

Experimental transport studies of monolayer graphene sandwiched between hBN are the focus of this work. The measurement of non-local resistance as a function of temperature, doping and magnetic field is used to infer information about valley transport. The main claim of the authors' is that there is large non-local signal at the primary Dirac point; while past studies have focused on secondary Dirac points and the associated Berry curvature. The Berry curvature gives rise to Valley current at secondary Dirac points. The authors interpret that the large non-local resistance at primary Dirac point is due to helical edge states that are ballistic. This is an interesting interpretation of the experimental data and the theoretical calculations suggest that this interpretation is reasonable.

The manuscript is clearly written and the data together with the analysis is interesting.

I recommend the publication of the manuscript.

Reviewer #2 (Remarks to the Author):

This manuscript reports strong evidences of topological valley current which are conveyed by edge modes. The experimental results are splendid. Although I am not sure that the interpretation is fully clear, I believe the work is ready for publication. It will bring an important contribution to the field.

Reviewer #3 (Remarks to the Author):

Authors describe transport measurements of graphene on hexagonal boron nitride superlattices (with various heterostructure configurations) revealing large nonlocal resistance that approach h/2e^2 at low temperatures. Interestingly, they find two regimes of nonlocal resistance, one at high temperature T> 60K, and one at low temperature T< 60K. While above T>60K, they have nonlocal resistance that scales in the usual for \rho_xx^3, when temperature is below T<60K, nonlocal resistance approaches a h/2e^2 leading them to speculate that a "quantum valley hall state" is being accessed. They provide systematic multi-terminal measurements to elucidate the origin of this unusual behavior and find that nonlocal resistance that come in the vicinity (no real plateaus but values spike close specific fractions of h/e^2) of the multi-terminal nonlocal resistance that is predicted from an edge state picture (Fig. 4d). They suggest that this may provide some evidence for counter-propagating edge states in G/hBN-type heterostructures.

I find the manuscript to be clear and interesting and definitely think these experimental results are deserving of being reported in your journal. I think the authors have made an interesting case that is suggestive of edge mediated nonlocal resistance; I particularly like the systematic multi-terminal investigations that the authors have embarked on that begin to address/discuss the nature of the valley hall state (quantum or not?, bulk or edge?) that may be present in G/hBN-type heterostructures. However, I have a couple of points/questions that may help to improve the readability of the manuscript as well as its exposition:

1. Authors use the fact that Rnl does not scale with rho_xx^3 below T<60K to imply that the bulk valley hall effect mechanism for Rnl (e.g., ref 8 and Nature Physics 11, 1027-1031 (2015), Nature Physics 11, 1032-1036 (2015) takes) does not dominate the behavior below T<60K in their devices. I say "imply" because while I appreciate the authors do not go out-right to rule it out, they go on in the very next paragraph to discuss an edge state picture. However, when valley hall

conductivity is large (and valley Hall angles approach 90 degrees) it has been argued that Rnl from the bulk valley hall effect mechanism becomes independent rho_xx [see e.g., Phys Rev B 94, 121408 (2016)]. I wonder if the authors have considered this. In this same regime of large valley hall angle, the bulk valley hall effect can also lead to low-dissipation channels [see e.g., Phys Rev B 99, 235405 (2019)]. Could these also help to account for change in sigma_xx as T is decreased below 60K in Fig. 2b?

2. Key to ruling out effects of edge states in the early experimental literature of valley hall effect in graphene heterostructures were multi-terminal nonlocal experiments in Nature Physics, 11, 1027-1031 (2015) (see Fig. 4) where they showed Rnl of very similar magnitudes for two nonlocal resistance geometries, both with the same bulk propagation distance but with very different possible paths along the edge. It seems that the authors measurements are uniquely capable of verifying this/showing a contrast to this experiment. I wonder if the authors can use their experiment (in a geometry similar to that of Fig. 4 Nature Physics, 11, 1027-1031 (2015)) to provide evidence or discuss the role that bulk effects play in the low-temperature regime.

3. Authors report a characteristic length of the non-local decay as around 2.0 microns. I wonder how this compares with the mean free path of the bulk (normal longitudinal conductivity) given they report very high mobilities for their samples?

4. Given that the authors argue for the presence of edge states in their system, could they comment on the origin of these edge states given they have encapsulated their samples in both bottom and top (hBN/G/hBN). How robust are these edge states to scattering? Are they guaranteed by any symmetry principle (i.e. symmetry protected)?

Reviewer #4 (Remarks to the Author): (***please also see attachment for more details***)

This paper discusses quantum transport of Graphene/hBN (Gr/hBN) superlattices,
in particular, possible emergence of the quantum valley Hall state.
The fabricated devices are of high quality and the non-local resistance in the quantum limit at the primary Dirac point (PDP) was first discovered in this paper so far as I know.
Furthermore, more detailed study on their devices imply the edge-meditated transport,
which is consistent with the quantum valley Hall state.
Compared with spin counterpart (‘quantum spin Hall effects’), these valley currents survive over distances of several microns and more progress in this line should pave a way toward possible applications.

Manuscript#: COMMSPHYS-20-0325
('Topological valley currents via ballistic edge modes in graphene superlattices near the primary Dirac point' by Yang Li et al.)

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The fabricated devices are of high quality and the non-local resistance in the quantum limit at the primary Dirac point (PDP) was first discovered in this paper so far as I know.
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Compared with spin counterpart ('quantum spin Hall effects'), these valley currents survive over distances of several microns and more progress in this line should pave a way toward possible applications.

In ref. [8] (R. V. Gorvachev et al. (2014)),
the non-local resistance is enhanced at PDP/SDP in Fig.1/Fig. S1 respectively.
In ref. [10] (K.Komatsu et al. (2018)), the quantum-limited non-local resistance was discovered at SDP.
I think that it would be better for the authors to give more comments on these differences (including their own results).

In the following, let me comment on some detailed points.

Concerning the non-local resistance under a magnetic field,
the authors discuss the spin-current contribution
('due to contributions from charge-neutral spin currents' in the main text).
I basically agree with this but one should care more about heat-current contribution.
Moreover, QHE edge current contribution can dominate with higher magnetic field.
I agree with authors' comments that these contributions are excluded for the case without a magnetic field.
Although careful assignment of these combined effects can be beyond the scope of this paper, I consider that the authors had better be careful about comments here.

In this paper, the crossover condition is fixed to be ~60K between bulk picture and edge-meditated conduction picture (Fig.4 a,b). This point should be discussed in more detail, for example, in relation to the energy band gaps at PDP/SDP.

In Supplementary Note.3, negative non-local resistance is discussed. On the other hand, I cannot find such a signature in the main text. Are these consistent with each other?

We thank referees 1 to 4 for their positive evaluation of our manuscript and for their helpful and constructive feedback. Referees 1 and 2 recommend publication without corrections or further work whilst referees 3 and 4 have provided helpful (positive) feedback, which has helped us to improve the paper.

Please find below a point-by-point response to all comment.

Reply to Reviewer #1

1. *"Experimental transport studies of monolayer graphene sandwiched between hBN are the focus of this work. The measurement of non-local resistance as a function of temperature, doping and magnetic field is used to infer information about valley transport. The main claim of the authors' is that there is large non-local signal at the primary Dirac point; while past studies have focused on secondary Dirac points and the associated Berry curvature. The Berry curvature gives rise to Valley current at secondary Dirac points. The authors interpret that the large non-local resistance at primary Dirac point is due to helical edge states that are ballistic. This is an interesting interpretation of the experimental data and the theoretical calculations suggest that this interpretation is reasonable."*

   "The manuscript is clearly written and the data together with the analysis is interesting. I recommend the publication of the manuscript."

   We thank the referee for their positive appraisal of our paper and for supporting publication.

Reply to Reviewer #2

1. *"This manuscript reports strong evidences of topological valley current which are conveyed by edge modes. The experimental results are splendid. Although I am not sure that the interpretation is fully clear, I believe the work is ready for publication. It will bring an important contribution to the field."*

   We thank the referee for their positive appraisal of our paper and for supporting publication.

Reply to Reviewer #3

1. *"Authors use the fact that R_{nl} does not scale with rho_xx^3 below T < 60 K to imply that the bulk valley Hall effect mechanism for R_{nl} (e.g., ref 8 and Nature Physics 11, 1027-1031 (2015), Nature Physics 11, 1032-1036 (2015) takes) does not dominate the behavior below T < 60 K in their devices. I say "imply" because while I appreciate the authors do not go out-right to rule it out, they go on in the very next paragraph to discuss an edge state picture. However, when valley Hall conductivity is large (and valley Hall angles approach 90 degrees) it has been argued that R_{nl} from the bulk valley Hall effect mechanism becomes independent rho_xx [see e.g., Phys Rev B 94, 121408 (2016)]. I wonder if the authors have considered this. In this same regime of large valley Hall angle, the bulk valley Hall effect can also lead to low-dissipation channels [see e.g., Phys Rev B 99, 235405 (2019)]. Could these also help to account for change in sigma_xx as T is decreased below 60 K in Fig. 2b?"*

   This is an interesting point and we thank the reviewer for pointing out this alternative explanation. We discuss these interesting results and cite the papers in the revised version of the manuscript in Lines 131-136, Page 8.

   The distinction between bulk valley Hall effect and edge states in the case of large valley Hall conductivity is expected to be difficult since, as Reviewer 3 points out, the bulk valley Hall effect can lead to a current distribution localized in the vicinity of the edge and in many respects resembles the edge transport [*Phys Rev B* **99**, 235405 (2019)]. In fact, even in the case of quantum

Hall effect, despite the accurately quantized Hall conductance, the distinction between the edge state picture and the bulk picture is ambiguous. Although the edge states can be directly observed [Patlatiuk, *Nat. Commun* **9**, 3692 (2018)] the current distribution does not often follow the simple edge state picture [J. Weis and K. von Klitzing, *Phil. Trans. R. Soc. A* **369**, 3954 (2011)]. However, in the case of the quantum Hall effect the edge and bulk pictures are two sides of the same coin due to the bulk-boundary correspondence, and thus the Landauer-Büttiker approach based on the edge state picture is always a valid phenomenological theory for the transport.

In the case of valley Hall effect the situation is even more complicated. On one hand the edge states can arise from the microscopic theory and the edge state transport can be calculated using quantum transport theory as discussed in Ref. [11]; on the other hand, however, the valley Hall bulk response can be calculated using local equilibrium approximation, these effects do not have the same robustness and protection as the quantum Hall effect, and neither theory yields a quantized response in general. For our devices, the nonlocal resistance is on the order of the quantum resistance suggesting that ballistic quantum mechanical channels are responsible for the transport. Moreover, the conductance matrix in the multiterminal geometry supports this conclusion. In the case of bulk response the nonlocal resistance could be on the order of resistance quantum only as a coincidence and by applying the equation (24) of nonlocal resistance given in [*Phys Rev B* **94**, 121408 (2016)], we obtain a significant deviation from the experimentally observed value.

Finally, we note that the observed dependence of R_{nl} on ρ_{xx} does not obey the predictions discussed in [*Phys Rev B* **94**, 121408 (2016)]. R_{nl} does not show a smooth and monotonic dependence and saturation as a function of ρ_{xx}, which is expected in the case of a bulk response but rather R_{nl} exhibits fluctuations, which resemble the mesoscopic conductance fluctuations in the quasiballistic regime of a quantum wire as expected in the case of unprotected edge modes.

2. *"Key to ruling out effects of edge states in the early experimental literature of valley Hall effect in graphene heterostructures were multi-terminal nonlocal experiments in Nature Physics, 11, 1027-1031 (2015) (see Fig. 4) where they showed R_{nl} of very similar magnitudes for two nonlocal resistance geometries, both with the same bulk propagation distance but with very different possible paths along the edge. It seems that the authors' measurements are uniquely capable of verifying this/showing a contrast to this experiment. I wonder if the authors can use their experiment (in a geometry similar to that of Fig. 4 Nature Physics, 11, 1027-1031 (2015)) to provide evidence or discuss the role that bulk effects play in the low-temperature regime."*

Another interesting point. We do not have a sample where we could perform exactly the same measurement undertaken in [*Nature Physics* **11**, 1027-1031 (2015)], highlighted by the reviewer; however, we have performed systematic multiterminal measurements in a ten-terminal device, and we have mapped the full conductance matrix that describes all the transmissions between the different terminals. The advantage of our setup is that even though the sample is in a quasi-ballistic operational regime meaning there are no universal signatures of the edge state transport, we obtain statistical evidence for edge modes. Our transmission matrix shows that there are altogether four connections between terminals, which are consistent with ballistic edge modes between these terminals as discussed in the main text. Additionally, we find one large connection through the bulk, which cannot be explained with edge states, but it might be due to one-dimensional conduction channel at a soliton-like domain wall. By more careful inspection of the rest of the transmission probabilities between the terminals, one notices that they also tend to be larger between terminals connected by the edge. Thus, although the agreement with the simple edge state picture is not perfect the edge states are the best qualitative explanation of our observations. This contribution is probably supplemented by a secondary contribution from a network of one-dimensional conducting channels appearing at the soliton-like domain walls.

We also point out that the statistical fluctuations in the transmissions are quite large. Even if we use terminal pairs, which have the same distance so that they should be equivalent, we can have quite different transmissions. Thus, we do not expect that sufficient evidence for identification of the transport mechanism could be obtained by preparing just two samples with different lengths of the edge. On would need to prepare a large number of samples and collect statistical evidence. Therefore, in some respects our multiterminal geometry with ten terminals is already better suited for the identification of the transport mechanism.

3. *"Authors report a characteristic length of the non-local decay as around 2.0 microns. I wonder how this compares with the mean free path of the bulk (normal longitudinal conductivity) given they report very high mobilities for their samples?"*

   In Supplementary Note 3, we calculate the mean free path of the device, which is 2 μm and confirm the ballistic character of device. In order to observe a coherent neutral long-ranged valley current through an all-electrical method requires nonlocal measurements from a geometry smaller than or comparable to the mean free path. For the nonlocal decay length, there does not need to be any connection between it and the mean free path; here both of them seem to coincide with the width of the device, one possible reason is that the disorder at the edge dominates both the scattering between the valleys as well as the scattering within the valley.

4. *"Given that the authors argue for the presence of edge states in their system, could they comment on the origin of these edge states given they have encapsulated their samples in both bottom and top (hBN/G/hBN). How robust are these edge states to scattering? Are they guaranteed by any symmetry principle (i.e. symmetry protected)?"*

   Our nonlocal measurements suggest that, below 60 K a spin-degenerate ballistic counter-propagating edge mode is dominant. These valley-helical edge modes exist at graphene zigzag edges or topological domain walls. This type of edge modes are discussed in Ref. [11] [*J. Phys. Mater.* **1**, 015006 (2018)]. These edge states can only exist when the intervalley scattering is suppressed, and which is the case in graphene. The edge modes are not protected by any symmetry. Their existence depends on the microscopic details as discussed in Ref. [11].

Reply to Reviewer #4

1. *"In ref. [8] (R. V. Gorvachev et al. (2014)), the non-local resistance is enhanced at PDP/SDP in Fig.1/Fig. S1 respectively. In ref. [10] (K.Komatsu et al. (2018)), the quantum-limited non-local resistance was discovered at SDP. I think that it would be better for the authors to give more comments on these differences (including their own results)."*

   We agree.

   In Ref. [8], nonlocal resistance was investigated in encapsulated hBN/graphene/hBN (top hBN layer is misaligned with respect to the graphene by 10°, bottom hBN layer is aligned to graphene) and non-encapsulated hBN/graphene/SiO_2 (hBN layer is aligned to graphene) devices. Both encapsulated and non-encapsulated devices show similar nonlocal response at the Dirac point (1 kΩ), which is explained as topological current. The activation behavior is different between encapsulated and non-encapsulated devices, only the latter (non-encapsulated one) shows activation behavior.

   In Ref. [10], nonlocal resistance was investigated in a ballistic encapsulated hBN/graphene/hBN (top and bottom hBN layers are aligned to graphene) device. At the Dirac point, the value of R_{nl} is similar to the reported value in Ref. [8], but at the secondary Dirac point, R_{nl} reaches a quantum-limited value.

Thus, the main difference of the experiments in Ref. [10] in comparison to Ref. [8] is that the layers are aligned, and presumably due to this reason R_{nl} reaches the quantum-limited value. The alignment seems to have a large effect on both the band gap and ballistic character of the nonlocal transport.

In our measurements, the nonlocal resistance was investigated in encapsulated hBN/graphene/hBN (top and bottom hBN layers are aligned to graphene) device. At the Dirac point, the value of R_{nl} is much larger than the reported values in Ref. [8, 10] and approaches a quantum-limited value.

The main difference between our results and the results reported in Ref. [10] is the observation of quantum-limited value of R_{nl} at the Dirac point instead of the secondary Dirac point. We have also made multiterminal measurements in a ten-terminal device, and mapped the full conductance matrix that describes all the transmissions between the different terminals. The advantage of our setup is that even though the sample is in a quasi-ballistic operation regime, so that there are no universal signatures of the edge state transport, we can obtain statistical evidence of edge modes. Our transmission matrix shows that there are altogether four connections between the terminals, which are consistent with the existence of spin-degenerate counter-propagating edge modes between these terminals.

We discuss these differences in detail in the manuscript in Lines 35-36, Page 2.

2. *"Concerning the nonlocal resistance under a magnetic field, the authors discuss the spin-current contribution ('due to contributions from charge-neutral spin currents' in the main text). I basically agree with this but one should care more about heat-current contribution. Moreover, QHE edge current contribution can dominate with higher magnetic field. I agree with authors' comments that these contributions are excluded for the case without a magnetic field. Although careful assignment of these combined effects can be beyond the scope of this paper, I consider that the authors had better be careful about comments here."*

   We thank the reviewer for pointing out other possible contributions to R_{nl} in the presence of a magnetic field. We have modified the manuscript according to the reviewer's suggestion in Lines 109-110, Page 6.

3. *"In this paper, the crossover condition is fixed to be ~60 K between bulk picture and edge meditated conduction picture (Fig.4 a,b). This point should be discussed in more detail, for example, in relation to the energy band gaps at PDP/SDP."*

   We agree.

   In our samples the temperature dependence of the longitudinal conductivity exhibits two distinct regimes of behavior, separated by a characteristic temperature (≈ 60 K). For $T > 60$ K the thermally activated transport starts to dominate, but for $T < 60$ K the longitudinal conductivity decreases slowly with T indicating that in this regime the effect of the thermally activated bulk carriers can be neglected. Interestingly, in Fig. 2c, the temperature dependence of the longitudinal conductivity at 2 T shows the appearance of $\nu = 0$ state below $T < 60$ K. The appearance of the quantum Hall state also requires that the effect of the thermally activated bulk carriers can be neglected. Finally, R_{nl} starts to show deviations from the semiclassical transport theory and approaches the quantum limited value below $T < 60$ K. If the quantum-limited value of R_{nl} is due to the edge states, we again have the requirement that the effect of the thermally activated bulk carriers can be neglected.

   Thus, all these observations are consistent with the corresponding crossover temperature being the temperature below which the effects of the thermally activated bulk carriers can be neglected. We point out that although the order of magnitude of the crossover temperature is the same in all these effects, the exact values of the crossover temperatures can of course be different, for example because the magnetic field influences the bulk spectrum and the effects of electron-electron interactions.

We have modified the manuscript according to the reviewer's suggestion in Lines 95-99, Page 6 and Lines 128-131, Page 8.

4. *"In Supplementary Note.3, negative non-local resistance is discussed. On the other hand, I cannot find such a signature in the main text. Are these consistent with each other?"*

In the main text, we plot the R_{nl} vs gate voltage or magnetic fields with the channel length of 3 um. The maximum positive value of R_{nl} is extremely large, so that it is difficult to see the small negative values of R_{nl} close to the Dirac point. In the Supplementary Note 3, in order to show the negative values of R_{nl} clearly, we plot the R_{nl} vs carrier density with the channel length of 5 um, where the positive and negative values of R_{nl} are similar.

We have added the channel length parameters in the captions of Figs. 3, 4 and Supplementary Fig. 5.

REVIEWERS' COMMENTS:

Reviewer #3 (Remarks to the Author):

I am satisfied with the authors reply and changes to the manuscript and think the manuscript is much improved with a clearer discussion of the results. I think the manuscript can now be published.

Reviewer #4 (Remarks to the Author):

In a private correspondence with the editor the reviewer recommends publication as is.