Reviewers' comments:

Reviewer #1 (Remarks to the Author):

Chen et al. reported their numerical methods to simulate the transport properties of graphene superlattices, either formed with hBN or by patterning the underneath dielectric. The authors directly simulated both cases and compare them with experiments. The paper is very timely and well-written, with careful references to previous works. Therefore, I recommend its publication in Communications Physics. But I hope the authors can amend the paper to address the following points in the revised version:

In Fig. 4, it seems that the carrier densities needed to fill up the first hole miniband and the first electron miniband are not the same. Instead, it seems to me that the carrier density needed to fill up the first hole miniband at the positive electric field is equivalent to that needed to fill up the first electron miniband at the negative electric field. Can the authors give more explanations on such an interesting asymmetry (or symmetry of electric field * doping)?

Can the authors explain more what they have done, such that we can understand better why their calculation can show the quantized conductance in the magneto-transport measurement? Is it a 1D or 2D simulation? Is it in the longitudinal or transverse configuration?

In their scaled graphene lattice method, do they see the difference if they choose the scaling factors differently? For instance, in the superlattice of period 35 nm, what would be the best scaling factor? What is the guidance for choosing a good scaling factor for computation?

Reviewer #2 (Remarks to the Author):

The manuscript report new results where the scaled tight-binding model (STB) is used for large-scale quantum transport simulations on imposed superlattices (hBN-moire and electrostatic) in the graphene. The theory is compared directly to experiments – where the hBN-moire experiments are included in the work.
It is shown how the salient features of the experiments are well reproduced by the STB, in particular the minigaps and magneto transport close to the graphene chargeneutrality point.
The results are important and interesting since the STB provides a very effective way to model transport in systems of experimental relevance, and the effects and the superlattice/moire effects and their utilization are topics of high interest presently. The manuscript is very well written and I recommend publication after the authors have responded to my points below.

1) The twist angle is not known in the experiment? Is the theoretical twist angle (lambda) fitted to the experiment.

2) In the experiments the graphene is sandwiched between hBN and alined to one of these. Does the aligned hBN play a role on the transport? I guess it is assumed not to.

3) There is a pronounced electron-hole asymmetry. This can be reproduced in the simulation by shifting the Fermi energy of the electrodes. But it is suggested to come from higher-order terms (page 8 top). Why is this more likely compared to the first explanation? What is meant by "floating Fermi energies" (following the gate?)

4) The higher-order terms are again mentioned for the magneto-transport: What would it take to include these and why is it left out if this is important. This should be discussed a bit more.

5) It would be good to have the experiments of ref. 20 included in fig. 4 for comparison.

6) The STB reproduce the conductance properties. Does it also reproduce the correct current distribution? Is it clear if one should scale the potential or not – it seems to be taken from the non-scaled models (e.g. for the moire).

Minor:
Fig. 2 caption: "Scale bar" is mentioned for the inset in d: I see a box there and not a bar.

p. 9 english: "voltage range same as"

p. 19: U_G should be U(G)

Response to Referees

COMMSPHYS-19-0557-T

In the following, the texts colored in gray are questions and suggestions from the reviewers. We respond to them point by point and summarize the changes made at the end of this letter.

I. RESPONSE TO REVIEWER 1

1. In Fig. 4, it seems that the carrier densities needed to fill up the first hole miniband and the first electron miniband are not the same. Instead, it seems to me that the carrier density needed to fill up the first hole miniband at the positive electric field is equivalent to that needed to fill up the first electron miniband at the negative electric field. Can the authors give more explanations on such an interesting asymmetry (or symmetry of electric field * doping)?

   Our response: The difference between the carrier densities needed to fill up the first hole miniband and the first electron miniband comes from the asymmetric superlattice potentials (there is no symmetry between the upper and lower halves of superlattice potentials). We have performed a simple test calculation in order to answer this question of Reviewer 1. We consider a model function consisting of two cosine functions $(\cos(G_x x) + \cos(G_y y))$ to describe the back-gate-supplied carrier density profile $n_{\mathrm{bg}}(x, y)$ in units of nm^{-2}, as shown in the panel (a) below:
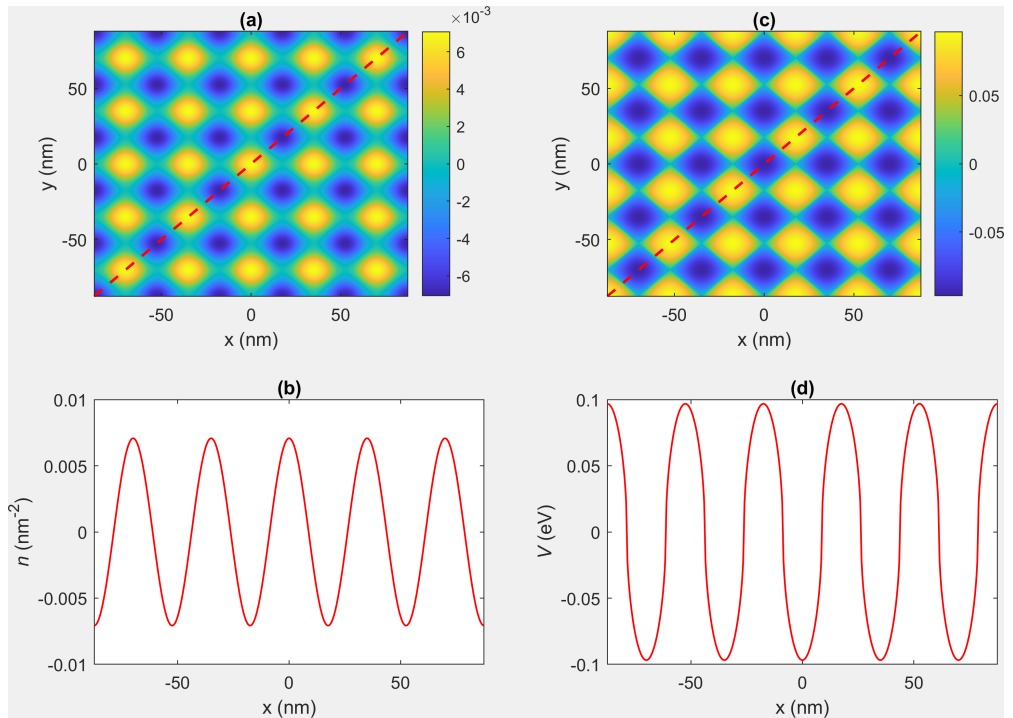

The resulting superlattice potential $U_s(x, y)$ is shown in (b). The symmetry of $n_{\mathrm{bg}}(x, y)$ and $U_s(x, y)$ can be better seen in their line cuts shown in (c) and (d) above, respectively. With such a symmetric superlattice potential, the inverse transmission as a function of top gate carrier density n_{tg} is shown below:
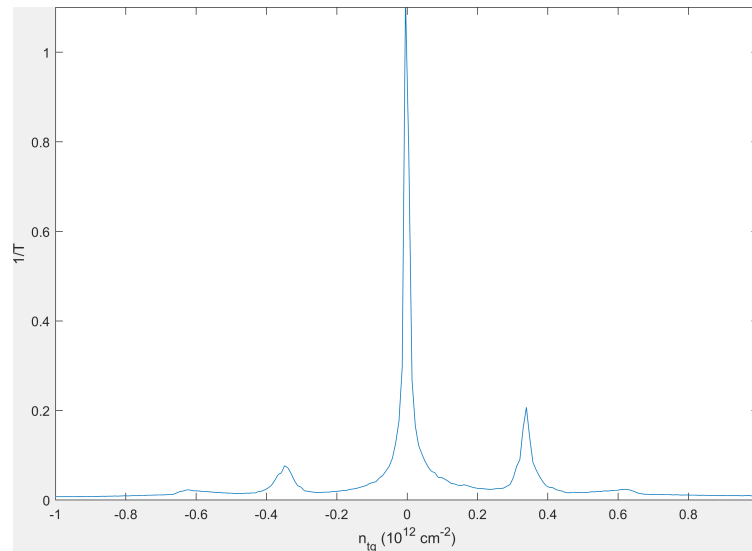
Clearly, the carrier densities needed to fill up the first hole and first electron minibands are the same. In our revised manuscript, we have added a few sentences (lines 185–190) to remark on this asymmetry.

2. Can the authors explain more what they have done, such that we can understand better why their calculation can show the quantized conductance in the magneto-transport measurement? Is it a 1D or 2D simulation? Is it in the longitudinal or transverse configuration?

Our response: Throughout our manuscript, we have considered two-terminal devices (Fig. 1**a** and the inset of Fig. 4**a** in the main text), for which the quantized conductance can be understood as the formation of the Landau levels. Because of the considered two-terminal geometry, there is no longitudinal and transverse configurations to distinguish. As a simple example, let us refer to Fig. 2 of Ref. 40 in our manuscript, reproduced here for convenience:
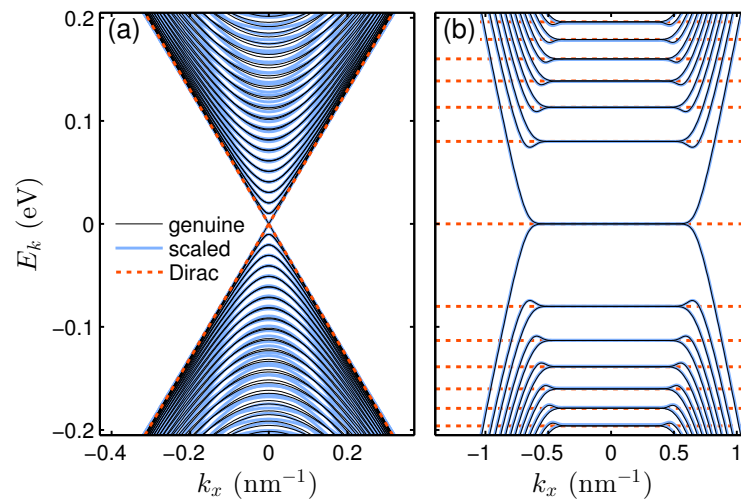

The above figure considers a spinless lattice of a 200-nm-wide armchair graphene ribbon without complication of superlattice potential. At zero magnetic field, its band structure sketched in panel (a) shows many subbands in addition to the Dirac cone due to the finite width of the considered ribbon. At magnetic field of $B = 5$ T (uniform and perpendicular to the graphene plane), the band structure evolves to panel (b), where the flat bands are the Landau levels in

the bulk, and the tails with finite slopes are quantum Hall edge states. In this example, if we consider Fermi energy of 0.1 eV, the conductance of the ribbon in the case of $B = 5$ T will be $3 \times 2 = 6$ (in units of e^2/h), where 3 is the number of bands intersected by $E_F = 0.1$ eV and 2 accounts for spin degeneracy because the model Hamiltonian is spin-independent. Typical quantum Hall conductance quantization in two-terminal structures can always be understood in this way. In the case of our graphene/hBN moiré superlattices, we do not have such a simple picture because of the presence of the periodic potential. However, at low energy below the secondary Dirac point, the physics of the system is mainly dominated by the main Dirac cone, and the above described picture can be regarded as a good approximation.

3. In their scaled graphene lattice method, do they see the difference if they choose the scaling factors differently? For instance, in the superlattice of period 35 nm, what would be the best scaling factor? What is the guidance for choosing a good scaling factor for computation?

Our response: The general criterion for the scaling to work well is to choose a scaling factor s_f such that the scaled lattice spacing is still much shorter than all important length scales of the system considered. This was stated in Ref. 40 and also emphasized in our manuscript (at the top of page 4). For the gate-controllable superlattice, we considered $s_f = 6$ as mentioned on page 14. To make it clearer, in the revised manuscript we have explicitly made a remark mentioning "implement it in the tight-binding... with $s_f = 6$ (such that $a = s_f a_0 \approx 0.85$ nm $\ll \lambda = 35$ nm) to perform quantum..." As for choosing different scaling factors, we did perform some test simulations and confirmed that the results are independent of s_f, as long as the scaling factor fulfills the general criterion (scaled lattice spacing much shorter than important physical length scales). The results significantly change only when the general criterion is not met. To show that the simulations results are insensitive to s_f as long as the general criterion is met, we show in in the following the conductance for the graphene/hBN moiré superlattice simulated using $s_f = 4$ and $s_f = 6$:
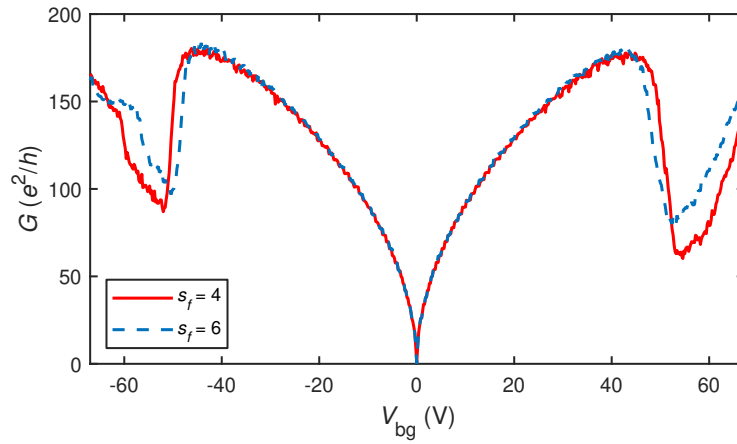
The $s_f = 4$ case corresponding to the red curve above has been shown in Fig. 2**e** in the main text. As it can be seen from the figure above, the difference between the two curves with $s_f = 4$ and $s_f = 6$ is negligible at low energies ($|V_{\mathrm{bg}}| \lesssim 40$ V) and moderate at higher energies ($|V_{\mathrm{bg}}| \gtrsim 40$ V). These two scaling factors $s_f = 4$ and $s_f = 6$ lead to scaled lattice spacing of $a \approx 0.57$ nm and $a \approx 0.85$ nm, respectively. The former is about $1/18$ of the moiré wavelength λ, while the latter is about $\lambda/12$. Whenever computationally affordable, we typically take the safer (smaller) scaling factor for transport simulations.

II.   RESPONSE TO REVIEWER 2

1. The twist angle is not known in the experiment? Is the theoretical twist angle (lambda) fitted to the experiment.

   Our response: Experimentally, there are two ways to extract the twist angle in transport measurements. First, via the position of satellite resistance peaks and just looking at the density value corresponding to 4 electrons per unit cell area. In our case, the peaks of hole and electron side are not at the same density, which is typically the case because in graphene/hBN moiré superlattices this asymmetry is expected. In the second way (which was done here), one can extract from Brown-Zak oscillations which are density-independent oscillations and periodic in $1/B$ and at rational values of the flux quantum. Thus, from the frequency one can extract magnetic flux and from that the unit cell area of the superlattice, and hence the moiré wavelength, which was around 10.9 nm, corresponding to a twist angle of $0.8°$. As described in the main text (page 7) and in the Supplemental Note 2, we found the best match between our simulation and experiment using $0.9°$, which corresponds to $\lambda = 10.4$ nm, slightly different from the value deduced from the experiment but within a reasonable range. In the revised manuscript, we have added a few sentences (lines 61–63 and 71–72) to make these clearer.

2. In the experiments the graphene is sandwiched between hBN and alined to one of these. Does the aligned hBN play a role on the transport? I guess it is assumed not to.

   Our response: In the simulations, our scattering region is composed of the (scaled) graphene lattice, in the presence of a periodic scalar potential. In the case of graphene/hBN moiré superlattices, the model superlattice potential is given by Eq. (1) which is a scalar potential. Therefore, the role of the aligned hBN is the induced periodic potential. In transport, electrons do not hop over sites within the hBN lattice, which is the case in the simulations, and presumably also the case in the experiment as hBN is a very good insulator.

3. There is a pronounced electron-hole asymmetry. This can be reproduced in the simulation by shifting the Fermi energy of the electrodes. But it is suggested to come from higher-order terms (page 8 top). Why is this more likely compared to the first explanation? What is meant by "floating Fermi energies" (following the gate?)

   Our response: This is because the adopted moiré model is only a scalar periodic potential and cannot significantly break the electron-hole symmetry. With higher-order terms, such as those models presented in Ref. 26 of our manuscript), the electron-hole symmetry can be significantly broken. The most obvious difference is that there are 6 secondary Dirac cones in the electron side, while in the hole side there can be only 3 or even 1, depending on the model. In our work, we tend to provide a straightforward simulation scheme that is not complicated by introducing too many tuneable parameters. The model potential based on Eq. (1), taken from one of the earliest studies on moiré superlattices, suits well this idea but always generate 6 secondary Dirac cones at both electron and hole sides (see figure 2 in our manuscript). On the other hand, shifting the Fermi energy of the electrodes can only capture the overall reduced conductance on the hole side compared to the electron side which is due to pn-junctions arising at the graphene/metal contact interfaces, but which can not account for the electron-hole asymmetry of the band structure modulation induced by the moiré perturbation (as observed by the asymmetric satellite conductance peak structures). The floating Fermi energies mean that the Fermi energy is the same as in the scattering region which is controlled by the back gate. We

apologize for being unclear about this. A sentence in the revised manuscript has been added accordingly (lines 76–78) to make this clearer.

4. The higher-order terms are again mentioned for the magneto-transport: What would it take to include these and why is it left out if this is important. This should be discussed a bit more.

Our response: We thank the reviewer for pointing this out. We have added a short paragraph (lines 136–141) in the revised manuscript to address this point. The paragraph is pasted here (with references suppressed): "At this stage, we comment that the periodic scalar potential enters the tight-binding model Hamiltonian through the on-site energy term (see Methods), so that it is readily compatible with the scaling method. Including higher-order terms of the graphene/hBN moiré superlattice should be possible but is beyond the scope of the present study. As we will see below, when the graphene superlattice potential arises solely from the electrostatic gating, our method becomes even more precise because in such systems the scalar potential is the only term comprising the superlattice."

5. It would be good to have the experiments of ref. 20 included in fig. 4 for comparison.

Our response: We agree that this would make the comparison even clearer. However, obtaining the data from Ref. 20 and the permission of using it may require some extra effort. We apologize for being unable to do so. However, we have now explicitly referred the figure label of Ref. 20 in the revised manuscript (line 185). We believe that this would be clear enough for readers to understand which figure of Ref. 20 to compare to.

6. The STB reproduce the conductance properties. Does it also reproduce the correct current distribution? Is it clear if one should scale the potential or not – it seems to be taken from the non-scaled models (e.g. for the moiré).

Our response: The basic idea of Ref. 40 about scaling the graphene lattice is to obtain band structure invariance within a reasonable energy range while using less dense lattice sites, which can be understood as simply the grid points for a continuous system. As long as the grid points are fine enough, important physics will not be lost using such scaled lattice. Because of the achieved band structure invariance, any potential energy terms appearing as local energy band offsets and applied to the model Hamiltonian do not need to be rescaled. On the other hand, when hoppings are involved (for example, the effective vector potential term mentioned in Ref. 27 of our manuscript), certain scaling (beyond the scope of the present work) is reasonably expected. The added paragraph mentioned in the response to Question 4 of Reviewer 2 (lines 136–141 of the revised manuscript) also addresses on this partly. As for current distribution, the scalable tight-binding model can be used to study current distributions indeed. This has been shown in, for example, Nat. Commun. (2015) and Nano Lett. (2015), both by P. Rickhaus et al., about ultraclean suspended graphene. In the presence of superlattice potential, the method is expected to reveal insightful current density distributions as well.

Minor:
- Fig. 2 caption: "Scale bar" is mentioned for the inset in d: I see a box there and not a bar.

    Our response: We have improved the corresponding figure accordingly. The scale bar should be clear enough now in the revised manuscript.

- p. 9 english: "voltage range same as"

    Our response: We thank the reviewer and have made corresponding correction.

- p. 19: U_G should be $U(G)$

    Our response: We thank the reviewer and have made corresponding correction.

III.   SUMMARY OF CHANGES MADE

1. Reference to Fig. 1**a** in the introduction paragraphs has been moved to line 31 of the revised manuscript in order to fulfill the format of the journal.

2. Lines 61–63 and 71–72: New sentences added to address Question 1 of Reviewer 2.

3. Lines 76–78: A new sentence added to address Question 3 of Reviewer 2.

4. Lines 82–84: Sentence slightly polished for better readability.

5. Lines 136–141: A short paragraph added to address Questions 4 and 6 of Reviewer 2.

6. Lines 175–176: A remark added to address Question 3 of Reviewer 1.

7. Lines 185–190: New sentences added to address Question 1 of Reviewer 1.

8. Typos in line 104 and 276 pointed out by Reviewer 2 are corrected.

9. Funding information elaborated (lines 430–432).

10. Data availability statement (lines 285–286) added.

11. Inset of Figure 2**d** improved to address the first minor question of Reviewer 2.

12. Supplementary information slightly modified: labels of the figures therein changed to "supplemental figure" to fulfill the formatting request.

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

I am satisfied with the answers the authors provided for my previous questions and therefore I recommend its publication in its present form.

Reviewer #2 (Remarks to the Author):

I am happy with all replies to my comments except for question 2 where I am not fully understanding the reply:

"2. In the experiments the graphene is sandwiched between hBN and alined to one of these. Does the
aligned hBN play a role on the transport? "

Here it seems that the theory and the reply address a single graphene on a single hBN: graphene/hBN.
However in the experiments they consider a stack: hBN/graphene/hBN where graphene is aligned to one of the two hBN layers. My question was related to the effect of the 2nd (aligned hBN) and where as this will play a role -- is it clear that the theory also covers hBN/graphene/hBN and not just graphene/hBN?

Response to Referee(s)

COMMSPHYS-19-0557-A

In the following, the text colored in gray is from Reviewer 2. We respond to it in detail and summarize the changes made to the revised manuscript at the end of this letter.

I.   RESPONSE TO REVIEWER 2

- I am happy with all replies to my comments except for question 2 where I am not fully understanding the reply:

  "2. In the experiments the graphene is sandwiched between hBN and alined to one of these. Does the aligned hBN play a role on the transport? "

  Here it seems that the theory and the reply address a single graphene on a single hBN: graphene/hBN. However in the experiments they consider a stack: hBN/graphene/hBN where graphene is aligned to one of the two hBN layers. My question was related to the effect of the 2nd (aligned hBN) and where as this will play a role – is it clear that the theory also covers hBN/graphene/hBN and not just graphene/hBN?

  Our response: We regret that our previous response was not clear enough for Reviewer 2, as we focused on explaining the role of the "aligned hBN" but not the issue of graphene/hBN vs hBN/graphene/hBN. The "aligned hBN" means that the twist angle ϕ of the hBN lattice with respect to the graphene lattice is small enough such that the resulting moiré wavelength is long enough to induce the secondary Dirac point at an energy reachable in experiment. Although there is another hBN layer in contact with graphene, its twist angle is very likely too large such that the resulting moiré wavelength is very short, and hence the secondary Dirac point is far from reachable in experiment. In this case, the hBN layer resulting in a short wavelength moiré is practically not doing anything to the graphene sample, playing just a plain role of a good insulating substrate. Note that encapsulation of graphene with two hBN layers is very common in device fabrications nowadays, and our sample was also the case. However, the chance of both hBN layers being aligned (small twist angles) is very low. A recent work (Nano Lett. **19**, 2371) has reported this rare case, but in our experiment, we did not observe the effect of two different moiré periodicities. Therefore, we conclude that modeling with graphene/hBN is sufficiently accurate, even though the device was fabricated with hBN/graphene/hBN. To clarify these details, we have added a few sentences (lines 63–68) and one new reference (Ref. 35) in the revised manuscript. We thank Reviewer 2 for pointing out this ambiguity.

II.   SUMMARY OF CHANGES MADE

- Two sentences (lines 63–68) and reference 35 newly added to address the ambiguity pointed out by Reviewer 2.

REVIEWERS' COMMENTS:

Reviewer #2 (Remarks to the Author):

The authors has addressed my point and I recommend publication in its present form.